Writing lines in turbulent air
using Air Photolysis And Recombination Tracking

PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de
Technische Universiteit Eindhoven, op gezag van de
Rector Magnificus, prof.dr.ir. C.J. van Duijn, voor een
commissie aangewezen door het College voor
Promoties in het openbaar te verdedigen op
maandag 16 januari 2006 om 16.00 uur

doors

Thijs Elenbaas

geboren te Amsterdam
Dit proefschrift is goedgekeurd door de promotoren:

prof.dr.ir. W. van de Water

en

prof.dr. J.J. ter Meulen

Copromotor:

dr. N.J. Dam

Dit werk maakt deel uit van het onderzoekprogramma van de Stichting voor Fundamenteel Onderzoek der Materie (FOM), die financieel wordt gesteund door de Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).

CIP-DATA LIBRARY TECHNISCHE UNIVERSITEIT EINDHOVEN

Elenbaas, Thijs


ISBN-10 90-386-2401-8
NUR 926
Trefw.: turbulentie / turbulente stroming / patroonvervorming / stromingsvisualisatie / anemometrie / lasersnelheidsmetingen
Subject headings: turbulence / turbulent flow / Air Photolysis and Recombination Tracking / Molecular Tagging Velocimetry / pattern deformation
# Contents

1 Introduction
   1.1 Turbulence ........................................... 1
   1.2 Overview ............................................ 3
   1.3 RELIEF ............................................. 8
   1.4 APART .............................................. 10
   1.5 Outline of the thesis ............................... 11

2 Molecular scale tagging .................................. 13
   2.1 Introduction ........................................ 13
   2.2 What is a fluid? .................................... 13
   2.3 Competition between diffusion and turbulence ....... 15
   2.4 Non-seeded velocimetry: those who sow, will harvest . 17

3 Experimental setup & jet characterisation ............... 21
   3.1 Introduction ......................................... 21
   3.2 Experimental set up .................................. 22
   3.3 Examples of written lines ............................ 24
   3.4 Line center determination ........................... 25
   3.5 Jet characterisation .................................. 28
       3.5.1 Jet ............................................... 28
       3.5.2 Mean flow properties ........................... 29
       3.5.3 Velocity fluctuation properties ................. 30
   3.6 Setup parameters ..................................... 36
   3.7 Measurements ........................................ 40

4 Chemistry and physics of APART .......................... 43
   4.1 Introduction .......................................... 43
   4.2 Photochemical creation of NO ........................ 43
       4.2.1 Spectroscopic investigations ................. 43
       4.2.2 NO formation chemistry ........................ 47
   4.3 Line deformation during tagging ...................... 49
       4.3.1 Introduction .................................... 49
       4.3.2 The shape of a written line ................. 49
Contents

4.3.3 Molecular diffusion ........................................... 52
4.3.4 Mass and heat transfer ........................................ 52
4.3.5 Lagrangian coordinates ..................................... 53
4.4 Line width experiments ....................................... 57
  4.4.1 Enhanced diffusion ..................................... 57
  4.4.2 Pressure dependency ..................................... 58
4.5 Line temperature ............................................... 63
  4.5.1 Energy absorption ....................................... 63
  4.5.2 Laser-Induced Fluorescence Thermometry .............. 67
4.6 Conclusion ....................................................... 68

5 Two-point correlation statistics ............................... 71
  5.1 Introduction ................................................ 71
    5.1.1 Longitudinal and transverse two-point correlations ... 73
  5.2 Spectra ................................................... 74
    5.2.1 Theory ................................................ 74
    5.2.2 Experimental results ................................ 76
  5.3 Image pre-processing ....................................... 80
  5.4 Kinematic simulations ....................................... 86
    5.4.1 Introduction ......................................... 86
    5.4.2 Velocity field ........................................ 87
    5.4.3 Line simulation ...................................... 89
  5.5 Conclusions ................................................ 96

6 Intermittency ..................................................... 99
  6.1 Introduction ................................................ 99
  6.2 Probability distribution functions ......................... 99
    6.2.1 Theory ................................................ 99
    6.2.2 Results .............................................. 100
  6.3 Structure functions ......................................... 102
    6.3.1 Theory ................................................ 102
    6.3.2 Results .............................................. 105
  6.4 Violent events .............................................. 107
  6.5 Conclusions ................................................ 108

7 Line stretching .................................................. 111
  7.1 Introduction ................................................ 111
    7.1.1 Stretching rate ...................................... 112
    7.1.2 Fractal scaling ...................................... 112
    7.1.3 The stretching rate in a 2D projection within a bounded region 114
    7.1.4 Transient behaviour ................................... 114
  7.2 Experimental considerations ................................ 115
    7.2.1 Fitting ............................................... 115
1 Introduction

1.1 Turbulence

An apocryphal story, attributed to several great physicists such as Albert Einstein, Richard Feynman, and Horace Lamb, goes as follows. As he lays dying the modern physicist plans to ask God two questions: Why relativity (or quantum mechanics, depending on who is departing), and why turbulence? “I really think”, says the famed physicist, “God may have an answer to the first question”.

Even though fluid motion can be described using a relatively simple set of equations –namely, the Navier-Stokes equations– our understanding of the described flow fields is certainly not complete. For low velocities of the flow we can easily work out what the flow parameters are, but when the flow velocity (or rather, the so-called Reynolds number) exceeds a certain value, the solution of the Navier-Stokes equations becomes unstable, which means that the described flow becomes turbulent. Although the governing equations still describe the flow field, the outcome will completely change for infinitesimal changes in initial and boundary conditions. This leaves us with a continuously changing flow field, and determining the nature of this turbulence is not simple.

Irrespective of whether one believes the story about the physicist and irrespective of whether one perceives turbulence as “the last conundrum” [1] in physics, it is clear that turbulence represents a not yet fully solved problem, not only theoretically, but also experimentally as well as numerically. This is not due to any lack of interest. From the first detailed observations of turbulent eddies by Leonardo da Vinci in the 16th century, turbulence has been unceasingly studied.

This interest in turbulence has been fueled by academic curiosity but also by industry. Where in some fields of physics it is hard to imagine direct applications, in turbulence they are obvious and ubiquitous. Since energy is a sparse resource, efficiency in energy conversion and consumption is a necessity. At the side of energy conversion one can easily see the applicability of turbulence knowledge in the form of more efficient rotors of windmills and turbines. On the consumer side one can think of efficient combustion in an internal combustion engine and aerodynamically shaped means of transportation such as trains, planes and automobiles. Another example that demonstrates the applicability of turbulence is its ability to mix two materials efficiently. Think of how easily milk is spread when stirred through a cup of coffee, compared to just letting the drop of milk diffuse by itself. In fact, turbulent stirring is used in many industrial applications.
1 Introduction

In this thesis, we will present a new technique, dubbed APART (an acronym for Air Photolysis And Recombination Tracking), for measuring turbulence. A new technique often introduces new possibilities (as well as new problems) and we will test-drive it over some more and some less trodden fields of turbulence experiments, to see if we can exploit the unique properties of this technique to measure turbulence properties that are not readily accessible using other techniques.

The technique has already been referred to in the title of this thesis as “writing in air”. We will now discuss what we mean by this (somewhat popular) denomination.

Molecular Tagging Velocimetry

If we perceive our technique as writing in air, then our pencil is a laser: A pulsed laser is used to ‘write’ a pattern in a flow field (Fig. 1.1). After a certain time delay the pattern, that has been altered by the flow-field, is read back. The velocity field follows directly from the displacement and the deformation of the pattern together with the time delay. This form of velocimetry is called Molecular Tagging Velocimetry (MTV). In its simplest form, a single “write” laser beam is shot once through the sample space. Along its path an optically induced chemical process is initiated, resulting in the creation of a new chemical species or in changing the internal energy state of an existing one.

This line of tagged molecules is now transported by the fluid flow. If the flow is turbulent, as we are interested in, the differences in velocity will result in a line that is not only displaced, but also deformed.

There are three optical ways via which these tagged molecules can be visualized: fluorescence, phosphorescence and laser induced fluorescence (LIF). In all three cases molecules relax to a lower state and their excess energy is released as photons.

Figure 1.1: A schematic drawing of Molecular Tagging Velocimetry showing 1) creation of pattern of tagged molecules, 2) position of tagged molecules after a delay $\Delta t$ in which the pattern deforms and moves, and 3) the readout beam for the visualisation.
fluorescence this energy decay occurs rapidly (within $10^{-7}$ s to $10^{-9}$ s at atmospheric pressure), thus making “direct” fluorescence impractical for tagging. In phosphorescence the decay is slower, because the transition is (quantum-mechanically) unfavorable.

In some writing schemes, the tagged molecule ends up in an excited state, as shown in Fig. 1.2a (1). If the molecule relaxes through phosphorescence (2), lasting long enough to see line displacement, this can be used to track the written line and no additional visualisation step is needed. If during tagging the molecule did not reach a phosphorescing state, or relaxed before the molecule was “read”, a second step is needed. This is shown in Fig. 1.2b. The tagged molecule is then excited using a second laser beam (2), employing a wavelength such that it specifically excites the tagged molecule. The molecule will fluoresce and this fluorescence is captured by means of a camera. This manner of visualisation is called Laser-Induced Fluorescence.

1.2 Overview

Although optical techniques are frequently used in modern fluid velocimetry, most are opto-mechanical in nature. Opto-mechanical techniques do not rely on photonics alone for flow measurements but require macro-size seeding. The best known and often used examples are Particle Image Velocimetry (PIV) and Laser Doppler Anemometry (LDA). Within the field of all-optical techniques we can distinguish analogous techniques but using molecular tracers. In Doppler schemes, light quasi-elastically scatters off molecules and the velocity of the molecules convey a Doppler shift to the frequency of the scattered light. In molecular tagging techniques, like in PIV, velocimetry is based on visualizing the tracer displacements.

Let us first make some remarks concerning some advantages that MTV may have compared to traditional techniques like PIV, LDA and hotwire velocimetry. In fluid
mechanics there are several very accurate and proven methods for velocimetry, so there should be good reasons to explore a new technique.

First of all, MTV techniques have proven to allow measurements of velocities in inhospitable environments, like jet engines, flames, high pressure vessels. Techniques like Pitot and hotwire velocimetry and PIV will not, or barely, work under these circumstances.

Most other possible advantages arise from the freedom of being able to create tracers at the desired location and the desired moment. For one, traditional optical velocimetry suffers from the problem of velocity bias: as more fast particles are traversing the scattering volume than slow ones, measured statistics are biased toward the fast particles. Molecular tagging velocimetry techniques allow us, in theory, to write arbitrary patterns at will. This means that we are not dependent on particles that traverse the measurement area, but instead we have full control on the position and time at which tracers are introduced to the flow. This makes MTV techniques, other than Laser Doppler anemometry (LDA) and particle imaging velocimetry (PIV) [2], insensitive to velocity bias errors.

Furthermore, standard techniques like hotwire velocimetry and LDA are point-wise measuring schemes. In order to measure space-dependent spatial structures, use is made of Taylor’s hypothesis of frozen turbulence. In this hypothesis it is assumed that advection by turbulence is small and therefore the advection of turbulence past a fixed point may be taken to be entirely caused by the mean flow. Spatial structures can thus be derived from time-dependent signals. However, fluctuations of the convecting velocity and several other phenomena can cause distortion of statistics that result from use of Taylor’s frozen turbulence. These distorting phenomena have been discussed by Fisher et al. [3], Lumley [4] and others. With MTV we are capable of measuring true spatially separated velocities.

Because MTV allows us to create patterns at will, we are able to measure turbulence properties that have been very difficult to study otherwise. For example, in Chapter 7 we will discuss the stretching and deformation of material lines in turbulence. As another example, we plan to create sets of dots and crosses by crossing laser beams. This may allow us to study Lagrangian properties but this falls outside of the scope of this thesis.

In this overview we will ignore techniques that are not all-optical and ignore Doppler-based schemes. We will only focus on the optical, molecular, tagging techniques. The field of MTV is fairly young; the first demonstration of implementation emerged approximately 20 years ago, and the number of schemes developed and investigated for use in air is still fairly small. We have aimed to give a complete overview of the schemes, including APART, that have currently been published. This data is shown in Tables 1.1, 1.2 and 1.3.

It is worth noting that, although some of the experiments in which these techniques were demonstrated seem very promising, not many have grown far beyond a proof of principle. Although MTV techniques have been demonstrated in a mul-
<table>
<thead>
<tr>
<th>Name</th>
<th>Tracer</th>
<th>Precursor</th>
<th>write</th>
<th>read</th>
<th>Seeding</th>
<th>Width [μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>APART</td>
<td>NO</td>
<td>air</td>
<td>ArF excimer (193 nm)</td>
<td>LIF (226 nm)</td>
<td>no</td>
<td>50 − 80</td>
</tr>
<tr>
<td>HTV</td>
<td>OH</td>
<td>H₂O</td>
<td>ArF/KrF excimer (193 nm/248 nm)</td>
<td>LIF (308 nm)</td>
<td>no²</td>
<td>300</td>
</tr>
<tr>
<td>LEI</td>
<td>Na⁺</td>
<td>Na</td>
<td>PDL (589+449 nm)</td>
<td>LIF (589 nm)</td>
<td>yes</td>
<td>≃ 500</td>
</tr>
<tr>
<td>OTV</td>
<td>O₃</td>
<td>O₂</td>
<td>ArF excimer (193 nm)</td>
<td>LIF (248 nm)</td>
<td>no</td>
<td>≃ 500</td>
</tr>
<tr>
<td>RELIEF</td>
<td>O₂(ν = 1)</td>
<td>O₂</td>
<td>Nd:YAG + SRS (532+580 nm)</td>
<td>LIF (193 nm)</td>
<td>no</td>
<td>50 − 100</td>
</tr>
<tr>
<td></td>
<td>acetone*</td>
<td>acetone</td>
<td>Nd:YAG (266 nm)</td>
<td>phos</td>
<td>yes</td>
<td>≃ 200</td>
</tr>
<tr>
<td></td>
<td>biacetyl*</td>
<td>biacetyl</td>
<td>XeCl excimer (308 nm)</td>
<td>phos</td>
<td>5% mf</td>
<td>≃ 200</td>
</tr>
<tr>
<td></td>
<td>NO(A)</td>
<td>NO</td>
<td>Dye (225 nm)</td>
<td>fluo</td>
<td>1% mf</td>
<td>200</td>
</tr>
<tr>
<td></td>
<td>NO</td>
<td>NO₂</td>
<td>XeCl excimer (308 nm)</td>
<td>LIF (226 nm)</td>
<td>600 ppm</td>
<td>750</td>
</tr>
<tr>
<td></td>
<td>NO</td>
<td>tBN</td>
<td>ArF/KrF excimer (193/248 nm)</td>
<td>LIF (226 nm)</td>
<td>1% mf</td>
<td>1000</td>
</tr>
</tbody>
</table>

Table 1.1: Overview of published MTV schemes in air. LEI = Laser Enhanced Ionisation, OTV = Ozone Tagging Velocimetry, LIF = Laser induced Fluorescence, PDL = Pulsed Dye Laser, SRS = Stimulated Raman Scattering, fluo = fluorescence, phos = phosphorescence, mf = mol fraction, ppm = parts per million.
### Table 1.2: Continuation of table 1.1: overview of different MTV schemes and their properties.

<table>
<thead>
<tr>
<th>Name</th>
<th>Application</th>
<th>Measured properties</th>
<th>Remarks</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>APART</td>
<td>supersonic jet ( (U = 300 - 600 \text{ms}^{-1}) ), laminar dry flow ( (U = 1 \text{ms}^{-1}) ), pulsed valve, free turbulent jet ( (U = 5 - 50 \text{ms}^{-1}) ), laminar flame</td>
<td>mean &amp; rms velocity profiles, PDF, higher order statistics</td>
<td>see sect. 1.4</td>
<td>[5, 6, 7, 8]</td>
</tr>
<tr>
<td>HTV</td>
<td>Space shuttle engine, supersonic nozzle ( (U = 1,300 - 1,800 \text{ms}^{-1}) ), wall cavity in a supersonic flow</td>
<td>mean &amp; rms velocity profiles, flow field</td>
<td>Promising technique. Tagging is a single photon process and efficient.</td>
<td>[9, 10, 11, 12, 13, 14]</td>
</tr>
<tr>
<td>LEI</td>
<td>acetylene flame ( (U = 1 - 2 \text{ms}^{-1}) ), shock tube, supersonic flow around cylindrical body ( (U = 500 \text{ms}^{-1}) )</td>
<td>mean velocity profile</td>
<td>high pressure and temperature required</td>
<td>[16, 17, 18]</td>
</tr>
<tr>
<td>OTV</td>
<td>laminar jet ( (U = 2 \text{ms}^{-1}) )</td>
<td>mean velocity profile</td>
<td>inefficient visualisation</td>
<td>[19, 20, 13]</td>
</tr>
<tr>
<td>RELIEF</td>
<td>aircraft inlet ( (U \approx 1,200 \text{ms}^{-1}) ), under-expanded supersonic jet ( (U \approx 600 \text{ms}^{-1}) ), free turbulent jet ( (U \approx 40 \text{ms}^{-1}) )</td>
<td>mean &amp; rms velocity profiles, PDF, higher order statistics</td>
<td>see Sect. 1.3</td>
<td>[21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31]</td>
</tr>
</tbody>
</table>
### Table 1.3: Continuation of table 1.1: overview of different MTV schemes and their properties.

<table>
<thead>
<tr>
<th>Name</th>
<th>Application</th>
<th>Measured properties</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>(6)</td>
<td>high subsonic-supersonic micro-jet (exit Ø 1 mm) ($U = 200 - 600 \text{ms}^{-1}$)</td>
<td>mean velocity profile lifetime $50 - 200 \text{ns}$</td>
<td>high efficiency, lifetime $\approx 100 \mu\text{s}$, mean velocity profile in a $50 \times 50$ grid</td>
</tr>
<tr>
<td>(7)</td>
<td>internal combustion engine model ($U \approx 5 \text{ms}^{-1}$)</td>
<td>mean velocity field in a $50 \times 50$ grid</td>
<td>mean and rms velocity profiles, high efficiency</td>
</tr>
<tr>
<td>(8)</td>
<td>laminar, hypersonic boundary layer, ($U \approx 3000 \text{ms}^{-1}$)</td>
<td>mean and rms velocity profiles</td>
<td>NO created in shock tube, lifetime $\approx 200 \mu\text{s}$, quantitative observations on turbulence</td>
</tr>
<tr>
<td>(9)</td>
<td>flow through optical cell</td>
<td>velocity profiles</td>
<td>high efficiency</td>
</tr>
<tr>
<td>(10)</td>
<td>laminar jet and dense spray.</td>
<td>mean and rms field, in a $15 \times 15$ line grid, stereoscopic</td>
<td>high efficiency</td>
</tr>
</tbody>
</table>
1 Introduction

titude of environments, ranging from simple pulsed valve flows [5] to space shuttle engines [9], actual flow measurements have remained restricted to a few patterns from which mean velocities or at most root mean square (RMS) velocities are calculated. To our knowledge, except for the work presented in this thesis, the only rigorous fluid mechanics study has been performed by Noullez, Miles and Frisch [29] using the RELIEF scheme, where many thousands of lines were measured in a free turbulent jet and thorough statistical analyses on this data have been performed. It seems that otherwise the MTV techniques that have been developed by molecular physicists have not been taken up by fluid mechanics, and have not yielded much more than semi-quantitative data. We believe that the potential strengths (but also the limitations) of the MTV measurement techniques have not really been explored, and warrant more research. This is the reason that we will pay extra attention to the RELIEF technique in Sect. 1.3, and compare our findings with the turbulence data published in [29]. But first, let us quickly glance over some other techniques, before turning our attention to RELIEF and ultimately our own measurements.

Molecular tagging velocimetry by means of phosphorescent tracer molecules is an interesting concept since only a single laser system is required for the whole experiment. In many respects, biacetyl is a good phosphorescent tracer molecule. It has a long phosphorescence lifetime and is efficiently excited. For experiments this means that it is easy to create a multi-line grid and easy to capture an image at one or more moments in time with a single laser. The drawbacks are that biacetyl is not a native constituent of air and therefore has to be seeded. Although non-toxic, its smell is severely unpleasant and persistent such that in practice flows can only be measured in a closed circuit. Finally, the lifetime of the phosphorescence is strongly reduced in the presence of molecular oxygen, so that experiments must be performed in an oxygen-free environment (e.g. in pure nitrogen).

Another technique that holds promise for the future is Hydroxyl Tagging Velocimetry (HTV). It is based on photo-dissociation of water vapor followed by visualisation of the resulting OH radical using LIF. Since dissociation can be achieved using a single 193 nm photon, the tagging molecule density is linearly dependent on the laser intensity. This means that, at worst, the line intensity decreases quadratically with the distance to the focus of the write laser beam, allowing for long lines. Additionally, the write- and read stage are both efficient, making it possible to split the write laser-beam into multiple beams that are made to cross, thereby forming a grid. The read laser-beam can be widened sufficiently to illuminate the full grid [12, 13, 14, 15].

1.3 RELIEF

The basis for RELIEF (Raman Excitation plus Laser-Induced Electronic Fluorescence) was laid almost two decades ago by Miles and Cohen in [21]. Here, the technique of using excited oxygen as tracer particles was first introduced, although
the technique was not yet dubbed RELIEF. The method takes advantage of the fact that in homonuclear diatomic molecules transitions between vibrational states are prohibited by the electric dipole selection rules, so that excited vibrational states have a relatively long lifetime: a few microseconds in atmospheric air. A Nd:YAG laser was used to create two 532 nm beams. One of these is used to pump a dye laser, that produces radiation in the vicinity of 580 nm. The frequency difference of the two beams matches the vibrational energy of the first vibrationally excited state of oxygen. The two colors of laser light are overlapped and focused in the tagging region where oxygen molecules are now driven into the vibrationally excited state by means of Stimulated Raman Scattering (SRS).

Raman scattering is –virtually instantaneous– inelastic scattering of light off molecules. In the case of RELIEF, the energy exchange is vibrational in nature. In the Raman effect a photon is absorbed by the molecule and re-emitted again via an intermediate (virtual) state. The energy difference between the emitted and pump radiation is determined by the energy difference between the initial and final states of the molecule. Under certain conditions the contributions from all molecules can add up to a coherent wave which gains in intensity along its path. This is called Stimulated Raman Scattering. The interrogation step is accomplished using a narrow linewidth ArF laser. This laser further excites the initially excited \( O_2 \) molecules, up to the \( B(\nu' = 7) \) vibronic state of oxygen and the subsequent fluorescence back to high vibrational levels of the ground electronic state is recorded by an intensified CCD camera.

The technique has been shown to work in supersonic flows [22, 23], albeit with very low image quality. This low quality was at least partly attributable to the limited lifetime of \( O_2(v = 1) \) as a result of molecular collisions. In subsequent research, the first steps towards velocimetry in turbulence were taken. In a high speed windtunnel behind a screen grid mesh, velocity fluctuations were measured by writing lines [25]. From these velocity fluctuations a histogram of the mean square velocity was calculated. It should be noted that this histogram was neither smooth nor symmetrical, indicating that the authors did not yet succeed in obtaining high quality turbulence statistics.

The setup was simplified by using a pressurized oxygen cell of 1 meter length in which a part of the 532 nm light was up-converted by Stimulated Raman Scattering [24]. Although now only two instead of three lasers were needed, the 580 nm beam was of quite low power (7% of the pump beam). By seeding [43] and inducing convection in the Raman cell [26] this was increased up to 35%.

In 1997 a landmark article on molecular tagging velocimetry in turbulence was written by Noullez, Wallace, Miles, Lempert and Frisch [29]. Here, lines were written in a free jet of fully developed turbulent air. Large sets of lines (5000 to 10000) were recorded and analysed off-line. From each deformed line, 512 velocity components \( u(y) \) were calculated and subjected to statistical analysis. The accuracy of this data was (nearly) comparable to that of conventional velocimetry: Smooth and
symmetrical histograms of velocity increments were found as well as correct higher order structure functions. We will not discuss the result here nor explain the nature of the measurements. Rather, in Chapters 5 and 6 we will repeat parts of these experiments with our own setup, and link these to the experiments described in [29].

1.4 APART

Whereas in RELIEF the tagged metastable states of oxygen molecules were rather short-lived (< 10 µs), in APART tracer molecules enjoy much longer lives. APART is based on the “photosynthesis” of nitric oxide and since NO is a stable molecule, patterns written with it can, in principle, be followed almost indefinitely. Additionally, it is significantly easier to set up, since it does not require a double wavelength beam for tagging. There is no need for a Raman-shifting cell and there is no problem of overlapping two beams. However, APART shares the main advantage of RELIEF of only requiring constituents that are natively present in air.

The beam of an ArF excimer laser operated at \( \lambda = 193 \text{ nm} \) with a pulse energy of about 40 mJ/pulse is focused in air using an optimized lens. We succeeded in creating a tagged line with a waist diameter of about 50 µm (Full Width at Half Maximum). Along this focus NO is formed over a length of approximately 1 cm.

The probe laser is fired with delays ranging from 3 µs to as much as 40 µs with respect to the tagging laser. This Nd:YAG pumped dye laser is operated at a wavelength of 226 nm to visualize the created NO tracer molecules. The probe laser excites the \( \gamma \)-bands of NO, and the resulting LIF emission from the excited (A) state was detected with a camera system. In the experiments the dye laser beam is aligned anti-collinearly to the excimer beam and both travel perpendicular to the mean flow direction. The beam diameter was regulated to result in maximal intensity while still encompassing the written line, resulting in a beam waist at the observation volume between 2 mm and 5 mm in diameter.

A UV-sensitive, intensified CCD camera is then used to detect the fluorescence. The light incident on the camera is filtered using a high-pass spectral filter, removing Rayleigh scattered light from both lasers while transmitting NO fluorescence. The camera images are captured using a high speed frame grabber and then stored for off-line processing.

The success of the technique is illustrated in Fig. 1.3 which shows an example of single APART lines written in a strongly turbulent flow. We notice that in these images both lines have a good signal to noise ratio and the displacement and deformation by turbulence increases with delay.

Our knowledge of the photochemical pathway in which NO is created is still incomplete. Investigations show that photo-excitation by the ArF excimer laser results in excited \( \text{O}_2 \) being created in the \( B \)-state, which easily dissociates into atomic oxygen, or in some cases, ionizes to \( \text{O}_2^+ \) by absorption of additional photons.
The same tagging wavelength causes nitrogen to end up in certain ionized and excited states. Besides these species that are assumed to be critical in NO production, many other products are reported to be created during photolysis, such as ozone (O$_3$). These and other species that are present in air after the tagging process will now chemically interact to ultimately create ground-state nitric oxide.

Directly after the tagging laser pulse, the NO production is still in progress and, under ambient condition, the nitric-oxide intensity will find its maximum between 3 and 5 µs after the write laser pulse, slowly decreasing afterwards. During this whole time several chemical processes take place. In order to study the chemistry of the NO formation, a detailed chemical kinetics model was developed by Nandula and Pitz [8]. The analysis of the simulations indicates that the amount of N present, is the most important factor in NO production. The N atoms can be produced via direct photo-dissociation or via dissociative recombination $N_2^+ + e^- \rightarrow N + N$. This model, however, remains to be investigated.

1.5 Outline of the thesis

The main theme of this thesis is the introduction of the new APART technique and an exploration of its possibilities as well as its limitations in measuring turbulence statistics. We will investigate how far the technique can be extended in terms of accuracy. We shall also discuss whether the unique features of this technique can be used to measure a flow property that has been difficult or impossible to measure using conventional techniques, such as line stretching.

In Chapter 2 we will discuss the question: are molecules the ultimate tracer particles? On first sight, they appear to be so. If tracer particles become lighter they will be better able to follow acceleration even at the smallest scales in flows. We will however see that thermal diffusion will impose an intrinsic limitation on tracing fluids.
1 Introduction

In Chapter 3 the setup of the experiments will be described in detail. Here, the different elements of the experiments, turbulence, writing of the pattern and reading of the pattern, will be discussed in terms of experimental equipment used. This is finally illustrated by some exemplary written lines. Subsequently, the basic technique used to extract velocity information from such lines is described. In order to validate the APART tagging velocimetry data, we will quantify the turbulent jet used in all following experiments using hotwire anemometry. Finally we show measurements of the mean- and root mean square velocity using APART, and compare those to values extrapolated from the hotwire results.

In Chapter 4 the chemistry underlying the creation of the NO molecular tracer particles is investigated. In this chapter we will describe a spectroscopic investigation of nitric oxide and oxygen. We shall discuss a chemical reaction model describing NO creation and its results. Subsequently, the influence of local heating by the laser-induced creation process on the shape of the written lines will be considered and (indirect) measurements of local heating will be investigated. We will show to what extent heating will influence the width of the written lines.

In Chapter 5 we will investigate well-known flow properties. We describe second-order statistics in the form of spectra and second-order structure functions. Here, we note significant differences between the APART measurements and extrapolated data derived from the turbulence characterisation done in Chapter 3. We investigate several reasons for this difference and show that different forms of ambiguity in line center determination affect small- and inertial scale properties. For this, different forms of image pre-processing are tested and deforming lines are simulated by means of numerical experiments (kinematic simulation).

Next, the measurements are extended to higher-order correlations. Chapter 6 explores probability density functions at different separations, allowing us to observe intermittency in transverse structure functions. Finally, the measurements are compared with experiments from literature and the She-Leveque intermittency model.

The technique of writing lines allows one to experimentally investigate the stretching of material lines, a subject that was previously almost exclusively explored in numerical simulations. Chapter 7 discusses the caveats of measuring line stretching parameters using the (single camera) line setup. Large sets of line-data are acquired for increasingly longer delays and stretching rates are calculated from them. The results will be interpreted in terms of the fractal nature of wrinkled lines in turbulence and compared with literature.
2 Molecular scale tagging

2.1 Introduction

A very specific feature of the APART technique is that it employs molecules as tracers. The advantage seems obvious: if tracer particles become smaller (and thus lighter) they will be better able to follow acceleration even at the smallest scales in flows. Additionally, (almost) the only manner to create tracer particles at will, is to use a photo-chemical tagging technique that tags or creates molecules.

However, we will see that molecules and turbulence (even at the smallest scales) reside at such different scales that they are governed by different mechanics. The result of this is diffusion, which imposes an intrinsic limitation on tracing fluids using molecules.

2.2 What is a fluid?

Naively, one could say that there is nothing which is more appropriate than using the molecules of a fluid for velocimetry. And, when such velocimetry appears thwarted by molecular diffusion, the erroneous idea may rise that, since there is nothing beyond molecules, such velocimetry is all there is. This is not true and there is a clear distinction between molecular motion and the motion of a fluid, a distinction that is rooted in the smallness of the molecular scales in comparison to the scales of the fluid motion.

First, we must realize that fluid flow is not the average motion of marked molecules. Fluid flow is the volume averaged motion of many (Avogadro’s number) molecules that at a certain instant live in a small parcel of fluid. This parcel is so small that the mean motion is independent of its size, but it is also much larger than the mean free path between collisions. In time, marked molecules straggle away from that parcel and move into other parcels, where they will almost instantly adapt to the local mean velocity. The force needed for this adaptation is related to the molecular viscosity.

Therefore, in tagging velocimetry, the fluid velocity in a point is not the average motion of a cloud of marked molecules that have straggled away from that point. Instead, the found fluid velocity is an average over the different fluid velocities that are sampled by the molecules of the spreading cloud.

The situation is sketched in Fig. 2.1. The measured velocity is an average over the molecular trajectories that are determined both by the flow field and the Brownian
forces. In tagging velocimetry we infer the (Lagrangian) velocity from the displacement of the center of gravity of clouds of marked molecules. One might think that this center of gravity follows the (average) Lagrangian fluid parcel. However, as Saffman points out [44] the interaction between Brownian motion and turbulence is subtle.

Let us call $D$ the diffusion coefficient of the tagged molecules. Then, as we observe displacements in the, say, $y-$direction, we do a one-dimensional analysis in which we call $Y(t)$ the displacement of a fluid parcel, and $\Delta(t)$ the displacement of a molecule. Since the flow is turbulent, $Y(t)$ is a fluctuating quantity, which is related to the Lagrangian velocity $v(t)$

$$Y(t) = \int_{t_0}^{t} v(t') \, dt',$$  \hspace{1cm} (2.1)

for fluid parcels which started at $Y = 0$ at $t = t_0$. Then, the mean square displacement is

$$\langle Y^2(t - t_0) \rangle_f = \int_{t_0}^{t} \, dt_1 \int_{t_0}^{t} \, dt_2 \langle v(t_1) \, v(t_2) \rangle_f,$$ \hspace{1cm} (2.2)

where $\langle \ldots \rangle_f$ is the average over turbulence realisations. Due to stationarity, the correlation function depends only on the time difference. For very small times, Brownian motion and turbulence do not interact and the mean square displacement of a marked molecule becomes [44],

$$\Delta^2(t - t_0) = \langle Y^2(t - t_0) \rangle_f + 2D(t - t_0).$$  \hspace{1cm} (2.3)

For longer times, however, these two are intertwined. Saffman shows that the position of the centroid of the Gaussian molecular cloud increases with time as

$$\Delta(t - t_0) = v(t_0) \, (t - t_0) + \frac{1}{2} D(t - t_0)^2 (\nabla^2 u_2)_p,$$ \hspace{1cm} (2.4)

where the first term is the position of the fluid parcel and $u_2$ is the Eulerian velocity which can, for short times, be related to the Lagrangian velocity. This equation
expresses that for a particular realization of the velocity field the centroid of a molecular cloud lags behind, or advances before the fluid parcel (depending on the sign of $(\nabla^2 u_2)_p$). When averaging over realizations of the velocity field, the extra term disappears as $\langle (\nabla^2 u_2)_p \rangle_f = 0$. However, in the correlations the term is still present

\[
\langle \Delta^2(\tau) \rangle_f = \tau^2 \langle \nu^2 \rangle_f + \frac{1}{2} D\tau^3 \langle v(\nabla^2 u_2)_p \rangle_f
\]  

(2.5)

with $\tau = t - t_0$. In our experiment we measure velocities from the displacement of the centroid of molecular clouds. The consequence of Eq. 2.5 is that the measured rms velocity now increases with the delay time as

\[
\frac{1}{\tau} \langle \Delta^2(\tau) \rangle_f^{1/2} = \left( u_2^2 + \frac{1}{2} D\tau \langle v(\nabla^2 u_2)_p \rangle_f \right)^{1/2}.
\]  

(2.6)

Following Saffman, we now make the estimation for short times:

\[
\langle v(\nabla^2 u_2)_p \rangle_f \approx \langle u_2(\nabla^2 u_2)_p \rangle_f = -\frac{1}{3} \omega^2 = -\frac{4}{15} \frac{\epsilon}{\nu},
\]

where $\omega$ is vorticity (dimension $s^{-1}$), $\epsilon$ the turbulent dissipation per unit mass (dimension $m^2 s^{-3}$) and $\nu$ the kinematic viscosity (dimension $m^2 s^{-1}$). Also, we have assumed homogeneity and isotropy. Finally, we can write Eq. 2.6 as

\[
\frac{1}{\tau} \langle \Delta^2(\tau) \rangle_f^{1/2} \approx u_2 \left( 1 - \frac{1}{15} \frac{D\epsilon}{\nu u_2^2} \tau \right).
\]  

(2.7)

We thus have an estimation of the rms velocity as measured from the displacement centroid of a molecular clouds rather than a fluid parcel. The additional term in Eq. 2.7 is determined by a combination of the diffusion constant and turbulence properties $\nu$ and $\epsilon$. By sticking in the values that are typical for our experiments, it is seen that the $\tau$ dependence in Eq. 2.7 is approximately $1 - \tau/20$, with $\tau$ in s. Such a dependence is unobservable in the experiment, and will be overwhelmed by other effects, such as the translation from Lagrangian to Eulerian velocities.

### 2.3 Competition between diffusion and turbulence

In the previous section we discussed how the centroid of a molecular cloud is related to the position of a fluid parcel when the cloud is subject to both thermal diffusion and turbulence. Now we will consider how the size of the particle cloud will change during its walk through the turbulence field. In our experiments we have written lines that have a width of the same size as the Kolmogorov-scale structures. If the lines would have been significantly thinner, the small scale turbulence would have deformed the lines. In our case however the lines will also widen due to turbulent diffusion, and let us consider how.
A pair of marked points in turbulence that have a dissipation-range distance, $\Delta_s(t=0) < \eta$, (where $\eta$ is the Kolmogorov distance) will spread exponentially over dissipation-range times, $t < \tau_\eta$ (where $\tau_\eta$ is the Kolmogorov time) [45],

$$\Delta_s^2(t) = \Delta_s^2(0) e^{\gamma t / \tau_\eta}. \quad (2.8)$$

This (ensemble averaged) deterministic spreading must be compared to diffusive spreading. If we consider the width (at height $e^{-1}$) of a Gaussian distribution, diffusive spreading will occur as

$$\Delta_d^2(t) = \Delta_d^2(0) + 4 D t. \quad (2.9)$$

For small times, $e^{\gamma t / \tau_\eta} \approx 1 + \gamma t / \tau_\eta$, and the deterministic spreading looks also diffusive,

$$\Delta_s^2(t) = \Delta_s^2(0) + 4 D_s t. \quad (2.10)$$

but with an apparent diffusion coefficient $D_s$ proportional to the squared initial width, $D_s = \Delta_s^2(0) / 4 \gamma / \tau_\eta$. Substituting the typical values, $\gamma \approx 0.2$, $\tau_\eta \approx 2 \times 10^{-5}$ s, and $\Delta_s(0) \approx 8 \times 10^{-5}$ m, we find $D_s \approx 1.6 \times 10^{-5}$ m$^2$s$^{-1}$, which is comparable to the molecular diffusion constant ($D \approx 2 \times 10^{-5}$ m$^2$s$^{-1}$). Therefore, turbulent broadening is an important ingredient of the blurring of lines. It remains a question how both processes interact. If we can indeed add these two processes, the effective diffusion coefficient in a turbulent flow will be proportional to the squared initial width of the line. This dependence can be shown by measurements done using the MTV technique. We will discuss the precise setup more thoroughly in Chapter 3, but for now we will only give a short description. A first laser beam is focused such that a laser beam is created with a minimal waist of approximately $60 \mu$m. By means of this ‘write’ laser a line of NO molecules is created. This line will widen by diffusion for a fixed time, after which a second laser beam induces fluorescence that is captured using a camera. This procedure is repeated until a 1000 images have been acquired and results have been plotted in Fig. 2.2. Here we calculated the diffusion for every position on the line, by fitting the width increase with increasing write-read delay. This position dependent diffusion rate was averaged over all lines. As the line width decreases from the left side of the line (position 1) towards the focus (in the center of the line, position 2) and increases again along the other side of the line (position 3), we have a distribution of initial line widths. Their corresponding diffusion coefficients were now plotted against this width.

The figure demonstrates that such a relation may indeed hold. Along sides 1 and 3 there is indeed a nearly linear relationship between diffusion and the initial width squared. However, it is also seen that the left and right side of the line do not collapse. This is attributable to laser alignment. Because the NO line does not lie perfectly in the focal plane, the perceived line is slightly widened on the camera. The corresponding diffusion rate is, however, related to the true initial line width, hence resulting in an offset.
Figure 2.2: The effective diffusion constant $D$ of a broadening line in turbulence as a function of its squared initial width, as derived from experimental line widening data. The line width, averaged over a 1000 lines, decreases from the left side of the line (position 1) towards the focus (in the center of the line, position 2) and increases along the other side (position 3). The corresponding local diffusion coefficients were plotted against this width.

However, it should still be verified that lines in turbulence expand in a different way than lines in still air. This is shown in Fig. 2.3. Here, Fig. 2.3a shows line widening without turbulence and Fig. 2.3b shows line widening with turbulence present. Clearly, the combined molecular and turbulent diffusion is more than a factor of 3 larger than that of molecular diffusion alone. This is more than was expected from our simple estimation of $D$ and $D_s$ and this may be due to interaction between turbulent and molecular widening. The bumps that are present in both images are most likely a measuring artifact.

### 2.4 Non-seeded velocimetry: those who sow, will harvest

Non-seeded velocimetry in gases using molecular tagging suffers from the loss of accuracy due to diffusion of the marked molecules. This is because in gases momentum and mass diffuse at approximately the same rate. This is expressed by the ratio

$$\frac{\nu}{D} = Sc, \text{ the Schmidt number.} \quad (2.11)$$
Figure 2.3: Diffusion difference between turbulent and still air. Plot a) shows the width squared against time in still air and b) shows the same data in turbulence. The combined molecular and turbulent diffusion is more than a factor of 3 larger than molecular diffusion.

In “normal” gases like air, the Schmidt number is of order unity. This has a remarkable consequence for the observability of the smallest eddies in turbulence. We will show that the wrinkles in material lines that are due to the smallest eddies will always be blurred by diffusion.

The small-scale properties of turbulence are determined by the turbulent dissipation $\varepsilon$ per unit mass and kinematic viscosity $\nu$. By a dimensional argument, the smallest length scale, the Kolmogorov length $\eta$ [46], is

$$\eta = \left( \frac{\nu^3}{\varepsilon} \right)^{\frac{1}{4}},$$

(2.12)

the smallest velocity difference, the Kolmogorov velocity, is

$$v_\eta = (\nu \varepsilon)^{\frac{1}{4}},$$

(2.13)

which can be viewed as the velocity across an eddy of size $\eta$. The turnover time of that eddy, the Kolmogorov time, is

$$\tau_\eta = \frac{\eta}{v_\eta} = (\nu / \varepsilon)^{\frac{1}{2}}.$$

(2.14)

During one turnover time, a physical line will be blurred due to diffusive spreading resulting in a width increase $\Delta_d$. Then

$$\Delta_d(\tau_\eta) = \left(4 D \tau_\eta\right)^{1/2} = \left(4 \nu S c^{-1} \tau_\eta\right)^{1/2} = \left(4 \nu S c^{-1} (\nu / \varepsilon)^{1/2}\right)^{1/2} = 2 \eta S c^{-1/2}.$$

(2.15)
Therefore, if $Sc \leq 1$, after one smallest eddy turnover time ($\tau_\eta$) the width of a line will always be at least twice the Kolmogorov length, which irons out wrinkles of that size. Clearly, in order to resolve the Kolmogorov scale, we need tracers that have a much larger Schmidt number. At the same time, these tracers must still be small enough so as to be able to follow the largest accelerations of the turbulent velocity field. We will now derive an expression for the largest size such a tracer may have.

First assume that the tracer is a sphere, which according to Stokes-Einstein has a diffusion coefficient

$$D = \frac{kT}{\mu},$$

with the Stokes mobility $\mu = 6\pi \rho_p \nu r$, where $r$ is the radius of the sphere and $\rho_g$ the density of the gas. Now, we must equate the drag force on the sphere to the force needed to accelerate it in turbulence,

$$6\pi \rho_g \nu u \cdot u = \frac{4}{3} \pi r^3 \rho_p a, \quad \text{or} \quad \frac{u}{a} = \left(\frac{2}{9} \frac{\rho_p}{\nu \rho_g}\right) r^2,$$

where $\rho_p$ is the density of the particle. Clearly, the velocity $u$ and acceleration $a$ need to be understood in terms of rms quantities.

Much is known about the ratio $\langle u^2 \rangle^{1/2}/\langle a^2 \rangle^{1/2}$ in turbulence. First, the dimensional Kolmogorov estimate for the acceleration variance is [47]

$$\langle a^2 \rangle = a_0 \epsilon^{3/2} \nu^{-1/2},$$

which follows from the observation that the acceleration is a dissipation scale quantity and is determined by $\epsilon$ and $\nu$ only. Here $a_0$ is a universal constant. We will use the constant $C_\epsilon$ to link micro-scale parameter $\epsilon$ to macro-scale parameters according to $\epsilon = C_\epsilon u^3/L$, where $L$ is the macroscopic length scale of the experiment. This relation will be discussed in more detail in Sect. 3.5.3. it then follows that

$$\langle a^2 \rangle = a_0 C_\epsilon^{3/4} u^{5/4} L^{-3/4} \nu^{-1/4}.$$  \hspace{1cm} (2.19)

It is found experimentally [48] that the scaling Eq. 2.19 is excellent, whilst $a_0 \approx 5$ for $Re_\lambda > 500$. Taking the ensemble averages for granted and omitting the $\langle \rangle$, we now have

$$\frac{a}{u} = C_I a_0^{1/2} C_\epsilon^{3/4} u^{5/4} L^{-3/4} \nu^{-1/4}.$$  \hspace{1cm} (2.20)

It turns out that the acceleration is an extremely intermittent quantity, with extremely non-Gaussian probability density functions. Therefore, the acceleration variance is a poor measure for the actual accelerations that occur in strongly turbulent flows. This was accounted for in Eq. 2.20 through the introduction of an intermittency factor $C_I$ which is $\mathcal{O}(10 - 100)$. Combining equations 2.17 and 2.20 then results in

$$r^2 \leq L^2 \frac{\frac{9 \rho_p}{2 \rho_g}}{C_I a_0^{1/2} C_\epsilon^{3/4}} Re^{-5/4}$$

(2.21)
2 Molecular scale tagging

with $Re$ the Reynolds number $Re = UL/\nu$ or, expressed in the Taylor-scale Reynolds number $Re_\lambda = \sqrt{15Cf Re}$:

$$r^2 \leq L^2 \frac{\frac{9 \rho_w}{2 \rho_p}}{C_f \frac{a_0}{\epsilon} \frac{1}{C_c}} Re_\lambda^{-10/4}.$$  \hspace{1cm} (2.22)

Substitution of the typical values, $Re_\lambda = 10^3$, $L = 10^{-2}$ m, $\rho_p = 10^3$ kg m$^{-3}$, $\rho_g = 1$ kg m$^{-3}$ and $C_f = 100$ leads to $r \leq 10^{-8}$ m. These particles have a diffusion coefficient $D = kT/6\pi \rho_g \nu r \approx 10^{-8}$ m$^2$s$^{-1}$, which is 3 orders of magnitude less than that of our molecules.

In conclusion, we can afford fairly large particles (compared to NO) which will be accelerated along with the flow, but which will diffuse much less than molecules and will therefore be more indicative of the path of fluid parcels. In Chapter 8 we will briefly discuss a candidate molecule.
3 Experimental setup & jet characterisation

3.1 Introduction

Molecular tagging velocimetry uses the molecules of a fluid to do velocimetry. By use of a strong laser, molecules are marked by altering their internal state, or new molecules are created in a specific pattern that is governed by the intensity profile of the laser beam. Fluid velocimetry is then achieved by following the displacement of the pattern in time, by visualisation of this pattern at a known time ($\Delta t$) after creation. Generally, a second laser is used for visualisation, in particular by means of laser-induced fluorescence.

A first successful attempt to use molecular tagging velocimetry for the measurement of turbulence has been reported by Miles et al. [29]. Metastable excited oxygen molecules were used as tracers and they coined the acronym RELIEF (Raman Excitation & Laser Induced Electronic Fluorescence). Because these metastable states have a very short lifetime due to collisional de-excitation, we have sought another MTV technique that involves a stable tagged molecule. We have used the newly developed technique called APART [5]: Air Photolysis And Recombination Tracking. In this scheme molecular tagging is done by photosynthesis of NO molecules out of $N_2$ and $O_2$ molecules in air: a pulsed laser beam is used to write, and another laser beam is used to read, both on single shot basis, see Fig. 3.1.

In this chapter we discuss the experimental setup of the APART MTV method and characterize the flow using conventional techniques, so that in Chapter 5 we can compare the APART measurements with this characterisation. In section 3.2 we will describe the main components of the setup and in section 3.3 we will show examples of tagged lines. In section 3.5 more emphasis is placed on the source of our turbulence: the free jet. In experiments, our interest is in strong turbulence with a well-developed inertial range. Since the turbulent flow needed to be small enough to fit in our optics lab, this resulted in very small scales that needed to be resolved. These scales are so small that conventional techniques (such as hot-wire anemometry) fall short in resolution. In section 3.5.2 we start by characterizing the mean flow properties and in section 3.5.3 we will discuss extensively how to bridge the resolution gap such that the results of the molecular tagging experiments can be compared to those of more conventional techniques. In section 3.6 we investigate the conditions that at least have to be met in order to use the described setup for...
Figure 3.1: The principle of Molecular Tagging Velocimetry. At $t = 0$ a line is written in the flow with a laser beam (“write”) and after a delay time $\Delta t$, the deformed and displaced line is read out by LIF (“read”).

quantitatively accurate measurements. Finally, we apply the APART method to the turbulent jet flow and measure the basic flow properties in section 3.7. We will see that the measured basic flow properties are comparable to the hotwire results.

3.2 Experimental set up

Let us discuss the main elements that make up the experimental setup. This setup will be used in (almost) all measurements in the subsequent chapters in this thesis. A schematic overview is given in Fig. 3.2.

Turbulence source

Our goal is to fully characterize the turbulence that will be used in the experiments that will follow. This enables us to compare APART molecular tagging velocimetry measurements with the known properties of the flow and assess the quality of the APART technique. In order to do this, one of the most commonly studied free shear flows [49] was used: the axi-symmetrical free turbulent jet. The jet, whose construction will be discussed in more detail in section 3.5.1, consists of a diffuser that smoothly goes over into a 1 cm diameter orifice. The exit velocity of the air is near sonic ($\sim 200$ ms$^{-1}$), which creates a strongly turbulent flow further downstream. The experiments are performed about 40 nozzle diameters downstream (along the $y$-
3.2 Experimental set up

Figure 3.2: Experimental set-up in detail. The pulsed ArF excimer laser creates a line of NO particles, the flow displaces and wrinkles the line. The pulsed dye laser allows the NO particles to fluorescence. The camera displaces the fluorescence signal within the readout area.

axis) of the orifice where the mean velocity has dropped to $\sim 45\text{ms}^{-1}$, with Reynolds numbers of $Re_{\lambda} = 460$.

Creation

APART is based on the “photosynthesis” of NO. The beam of a broad-band ArF excimer laser ($\Lambda$ Physik, CompeX 350T) operated at $\lambda = 193\text{nm}$ with a pulse energy of about $40\text{mJ/pulse}$ and a pulse duration of $18\text{ns}$ was tightly focused in air using a composite lens, specifically optimized for excimer laser beams, with focal length $f = 250\text{mm}$. A minimum waist diameter of about $50\mu\text{m}$ (Full Width at Half Maximum) was achieved. The resulting Rayleigh range (the distance from the focus where the beam waist has increased from $w_0$ to $\sqrt{2}w_0$) is approximately $5\text{mm}$. Along this line focus NO is formed in a process, the details of which are discussed in Chapter 4. For now, it is important to know that the creation of NO is non-linearly dependent on laser power. Optical breakdown is avoided as much as possible, since it spoils the non-intrusive nature of the technique.

Illumination

A Nd:YAG pumped dye laser (Spectron or Radiant Dye 'Jaguar') was frequency doubled and mixed with the fundamental wavelength of the Nd:YAG laser to obtain radiation with a wavelength of $226\text{nm}$. The $226\text{nm}$ beam was used to excite the $R_{21}(17.5)$ line in the $A \leftarrow X(0,0)$ system ($\gamma$-bands) of NO and the resulting LIF emission from the A-state was detected with a camera system. In the tagging
3 Experimental setup & jet characterisation

experiments the dye laser beam was aligned anti-collinearly to the excimer beam, perpendicular to the flow direction. In some experiments that required high intensity the dye laser beam was collimated. This was done using a lens with 1000 mm focal length where the focal point was located outside of the readout area. By changing the position of the focus nearer to, or farther from, the readout area, the beam diameter can be regulated to result in maximal intensity while still encompassing the written pattern. The readout beam diameter in the region of interest could be changed by shifting the location of the focus relative to that of the writing beam, resulting in a beam waist at the camera between 2 mm and 5 mm diameter.

**Visualization**

An intensified CCD camera (*Photonic Science Coolview*) was used to detect the fluorescence. This camera was selected for what was considered high speed at the time of purchase (max. 8 Hz) and high resolution (1Kpixel × 1 Kpixel) combined with UV-sensitivity and a fairly high image depth (12 bits). A custom lens (*Bernard Halle Nachfl.*) was used with a fixed focus of 250 mm and f/2.5. When the objective and image distances are set for minimal aberrations, a region of 65 × 65 mm is imaged onto the intensifier, that is optically linked to the CCD chip. The resolution of the chip in the imaging system is 1024 × 1024 pixels. The light incident on the camera was filtered using a custom long-pass filter (*Laser Optik*) with low transmittance (nearly 0%) at 226 nm and high transmittance (> 95% or above), thereby effectively removing Rayleigh scattered light from both lasers while transmitting NO fluorescence. The camera images are captured using a high speed frame grabber and stored on a large disk array. In processing, the images are loaded into a computer where the line center is determined by fitting Gaussian intensity profiles to local cross-sections. The algorithm will be discussed in more detail later in this chapter.

3.3 Examples of written lines

In Figure 3.3 some examples of APART measurements are shown under the conditions as described above. Each image was measured using single pulses of both excimer and dye lasers. The spatial resolution of the images is 6.4 µm/pixel. The images show that for short delays, between Δt = 0 µs and 3 µs the NO production is still in progress and the density is not yet maximal. The line intensity finds its maximum between 3 and 5 microseconds and slowly decreases afterwards. For longer times we can notice that the line deformations grow. Differences in intensity along the lines become more prominent: there are line elongations where the line intensity decreases, and there are patches where the intensity is relatively high. Also the line broadening becomes noticeable by eye. The major contributors to the width increase are thermal diffusion and turbulence at the smallest scale, but there are other contributions, such as convection and perhaps a chemistry based effect. If
the line is sufficiently displaced perpendicular to the object plane of the detection system, the line will widen due to defocussing.

![Line center determination](image)

**Figure 3.3:** APART measurements in a strong turbulence. The images are measured at different write-read delays, as indicated in the individual images. The lines are independent of each other, each image being recorded after a separate write laser shot.

### 3.4 Line center determination

In order to extract velocity information from the lines, the line center has to be determined with a high accuracy. The simplest approach would be to divide the
image in vertical cross-sections of one pixel width, and to find the maximum intensity value in this slice. However, this will give a 1-pixel accuracy at best and ignores all profile data that may help in finding a more accurate line center. Fitting the data with a profile that matches the distribution can yield sub-pixel accuracy [29]. To this aim, the intensity profile of the line cross section was represented by a Gaussian profile whose height, width, and center were adjustable parameters. If the signal-to-noise ratio is sufficient, the line center can now be determined with sub-pixel resolution.

Since the focal point of the tagging beam lies in the center of the image, the beam diverges toward both sides of the field of view and decreases in intensity. Especially because NO formation is a non-linear process, this results in a lower signal and signal-to-noise ratio at the image edges. For this reason, fitting is always started in the image center, and moves toward the edges, while keeping each fit result as starting values for the next fit. This way, the first line profile has the highest signal-to-noise ratio, and the following ones will start with already fairly accurate initial values. Let us now discuss the algorithm used. If $x$ is the horizontal coordinate and $y$ the vertical coordinate in an image, we fit for each vertical cross-section at $x_i$.

$$T_i(x_i, y) = a(x_i) + b(x_i) e^{(y - y_c(x_i))^2/\sigma^2(x_i)}, \quad (3.1)$$

we then form the normalized squared distance to the actually measured intensity $I(x, y)$ by

$$\chi^2 = k \sum_j \left( \frac{(I(x, y_j) - I(x, y_j))}{I(x, y_j)} \right)^2. \quad (3.2)$$

where the column number $i$ is omitted.

The normalization is based on the assumption that the number of registered photons is a Poisson process with variance $(kI)^{1/2}$. If the calibration factor $k$ which relates the number of photons to the registered intensity were known, $\chi^2 \leq 1$ would indicate a perfect fit. Later on we will further refine Eq. 3.2, to take the variation of the sensitivity of the CCD chip into account. By binning several central columns, the signal to noise ratio is increased and the initial values of the fit parameters can “guessed”. This is done by first finding the peak intensity pixel and subsequently the pixels on both sides of the maximum at half peak intensity. The Gaussian crossing these three points, delivers the initial fit values. Then, working outwards, all columns along the line are fit, using the fit parameters of the current column as initial parameters for the next one.

For lines that are only slightly deformed by turbulence, our results indicate that a line center can be determined with an accuracy of 0.1 pixel ($0.6 \mu m$). The Gaussian distribution matches the NO profile well and typically yields $\chi^2 < 1$ when using an accurate error model. However, it has to be noted that the accuracy of the line center displacement alone does not determine the accuracy of the measured flow speed. In
3.4 Line center determination

Figure 3.4: Fits are performed for cross-sections through the image. Plane 1 illustrates a vertical fit, plane 2 displays a fit normal to the local line slope. The right hand graph shows the cross-section of the line in plane 1.

Chapter 4 we will discuss further uncertainties which are due to small-scale motion within a line, in combination with the diffusion of the tagged molecules.

It follows that, not considering other sources of error, the minimum velocity difference observable is of the order of $0.1 \text{ m s}^{-1}$ for delays $\mathcal{O}(10 \mu\text{s})$. Although line tagging using similar geometries has been studied before [23], it remains to be investigated what the influence of turbulence structures smaller than the line width exactly is, considering the distortion of the Gaussian profile of the lines.

Fits over a vertical column only allow for accurate parameter determination if the written line is nearly horizontal. For longer times between tagging and probing line bending can become quite significant and a new method of line center determination that is always perpendicular to the local line orientation can be advantageous. Fig. 3.5 demonstrates this. Point 1 shows the line center determined by a vertical fit and point 2 shows the line-center as determined by a fit in the perpendicular slice. For a straight line that makes an angle with the horizontal no difference will be seen between the two fit orientations, but this can change when the line is curved.

We have implemented a relatively straightforward extension on the per-column fit routine that applies fits perpendicular to the local line direction: First, vertical fits are performed along the line as an initial guess. These data are then used to determine the local slope of the line. Since the point-to-point local slope is often fairly erratic, the slope is smoothed using a second-order Savitzky-Golay filter over 25 pixels. The new cross-sections are extracted normal to this smoothed slope, interpolating neighbouring pixels on the normal line (as shown in Fig. 3.5). The vertical fit parameters are used as initial parameters for the normal fit. If necessary, the perpendicular fit can be iterated until the line center is found with sufficient
3 Experimental setup & jet characterisation

Figure 3.5: Difference between a perpendicular and vertical fit. The intensities in the perpendicular slice are determined by using bilinear interpolation of neighbouring pixels along the normal line. Point 1 is the line center determined by a vertical fit and point 2 is the line-center as determined by a fit in the perpendicular slice.

accuracy, although for present measurements no substantial improvements have been observed.

It should be noted that since the initial fit is still done vertically, strong deformations that result in parts of the line that fold back or even intersect will cause problems during fitting.

3.5 Jet characterisation

3.5.1 Jet

The turbulence from a standard jet has well-documented scaling properties (for example Hussein et al. [50]): the mean flow depends in a characteristic way on the distance to the orifice; at a particular location there is a fixed ratio between mean and fluctuating velocities, and there is a well-defined relation between the turbulent dissipation and the fluctuating velocity. These relations are governed by parameters which we will determine experimentally. Finally, the jet’s self-similarity is condensed in its energy spectrum, which we will also measure. At low values of the mean velocity we can still measure the turbulence properties of the jet using hot-wire. We will then use the measured scaling relations to extrapolate these properties to mean velocities that are used in the tagging experiments.

In order to ultimately create homogeneous and isotropic turbulence the flow is
3.5 Jet characterisation

Figure 3.6: Jet setup. pressure regulator (2,4), filters for water, oil, etc (3), stagnation chamber (7) with honeycomb rectifier (8) and smooth nozzle (9).

first made laminar. In order to do this, filtered and compressed air is first fed into a stagnation chamber and subsequently through a honeycomb rectifying grid, Fig. 3.6. Any flow structures that are still present afterwards are removed by a smooth (3rd order polynomial) contraction in the circular nozzle from 8 cm to 1 cm. The jet emanates vertically upwards into ambient air with an initially flat-topped velocity profile of maximally 200 ms$^{-1}$. Strong turbulence is now generated in the boundary of the jet with the ambient that is nominally at rest. APART measurements are performed at a distance of 38 nozzle diameters above the exit, where the turbulence is fully developed and the maximum mean flow speed is 50 ms$^{-1}$. This setup is similar to the one described in [29].

3.5.2 Mean flow properties

To determine the outflow speed, the pressure is measured close to the exit of the nozzle. Assuming an incompressible flow, the outflow velocity can be calculated from the nozzle pressure with respect to the ambient pressure ($\Delta P$) using Bernoulli’s law:

$$\Delta P = \frac{1}{2} \rho v^2. \tag{3.3}$$

The macroscopic flow is measured at other positions in the same way using a Pitot tube. Measuring pressures using a Pitot tube is slow compared to velocity fluctuations and effectively averages the velocity. Moreover, the Pitot tube is a quadratic device so the measured average $v$ will be determined both by the mean velocity $U$ and the rms velocity $u$. If we write the instantaneous velocity as $U + u(t)$, $\langle u^2(t) \rangle = u^2$, then the velocity measured is related to $\Delta P$ by:

$$\langle \Delta P \rangle = \frac{1}{2} \rho \langle (U + u)^2 \rangle = \frac{1}{2} \rho \left( U^2 + u^2 \right). \tag{3.4}$$
3 Experimental setup & jet characterisation

We will argue in Section 3.5.3 that, due to the scaling properties of the jet, there is a constant ratio between $u$ and $U$, $C_u = \frac{u}{U}$. Then, if $\tilde{U}$ is the apparent mean velocity,

$$\tilde{U} = \left( \frac{2}{\rho} \langle \Delta P \rangle \right)^{\frac{1}{2}} = U \left( 1 + C_u^2 \right)^{\frac{1}{2}}. \quad (3.5)$$

On the jet axis the mean velocity is determined by the outflow pressure $p$ and height $z$ as [50]

$$U(p, z) = f(p) g(z). \quad (3.6)$$

Close to the jet (where $z$ is within the first several nozzle diameters) the radial mean velocity profile changes from approximately block shaped to the shape depicted Fig. 3.7c. When the flow is fully developed, the velocity scales with pressure and height, according to the non-compressible model as

$$f(p) = b \cdot p^{\frac{1}{2}} \quad (3.7)$$
$$g(z) = \frac{1}{(z - c)}. \quad (3.8)$$

Measurements have been performed relating the mean flow velocity to pressure (Fig. 3.7a) and the velocity dependence $g$ as a function of height (Fig. 3.7b). The flow velocities have been measured with a Pitot tube centered above the nozzle opening and have been corrected to produce the real mean velocity. For low velocities hot-wire anemometry has been used. It can be seen in Fig. 3.7a that the two techniques yield corroborating data. Fits of $f(p)$ and $g(z)$ yield $b = 59 \text{ kg}^{-1/2} \text{m}^{3/2}$ and $c = 2.4 \cdot 10^{-2} \text{ m}$. Note that the virtual origin of the jet lies outside of the jet.

The self-similarity is demonstrated in Fig. 3.7c. Here the radial velocity profile is shown for different heights and pressures, with velocity normalised by the mean velocity from Eq. 3.6 and width by the full width at half maximum (FWHM). The profiles collapse perfectly and display a normal distribution. At height $z = 38 \text{ cm}$, where most measurements will be done, the FWHM is 10 cm.

3.5.3 Velocity fluctuation properties

In order to assess the quality of the molecular tagging velocimetry, it is necessary to compare the MTV results with those using conventional hot-wire anemometry. Since the resolution of hot-wire velocimetry is much coarser than that of APART, we must employ a careful extrapolation procedure which allows to determine the small-scale jet properties from a measurement at much lower turbulence intensities where the flow can still be resolved using the hot wire.

The small-scale turbulence properties $\eta$, $v_\eta$ and $\tau_\eta$ are determined by the turbulent dissipation $\varepsilon$ per unit mass (which has dimension $\text{m}^2\text{s}^{-3}$). The turbulent dissipation
3.5 Jet characterisation

Figure 3.7: Jet characteristics a) Mean velocity versus pressure at 38 cm height above the jet center b) measurements of function $1/g(z)$ for pressures 1.5, 1.6, 1.7, 1.8 above the jet center c) radial velocity profiles normalised for different heights and flow speeds, with the velocity normalised by the mean velocity found by Eq. 3.6 and width by the measured full width at half maximum (FWHM).

can be measured if all velocity derivatives at the smallest scale are available. For isotropic turbulence there exist simple relations between those derivatives such that measurement of a single derivative suffices, and [51]

$$\epsilon = 15\nu \left\langle \left( \frac{\delta u}{\delta x} \right)^2 \right\rangle. \quad (3.9)$$

Using Taylor’s frozen turbulence hypothesis, the spatial derivative in Eq. 3.9 can be
approximated by the time derivative:

$$\frac{\delta u(t)}{\delta x} = \frac{1}{U} \frac{\delta u(t)}{\delta t}. \quad (3.10)$$

The problem is that this derivative can no longer be resolved when \( \eta \) becomes smaller than the probe resolution. In order to solve this problem we employ a fundamental relation of small-scale turbulence, namely that the dimensionless dissipation constant \( C_\epsilon \) is independent of the Reynolds number [52],

$$\epsilon = C_\epsilon \frac{u^3}{L} \quad (3.11)$$

where \( L \) is the integral (length) scale, the longest length scale over which velocities in the flow are still correlated. This value can be approximated by the diameter of the jet. We will verify below that the jet is self-preserving with a constant relative turbulence intensity:

$$C_u = \frac{u}{U}. \quad (3.12)$$

Combining Eq. 3.11 and 3.12 gives

$$\epsilon = C_\epsilon C_u^3 \frac{U^3}{L}. \quad (3.13)$$

We see that \( \epsilon \) can be measured from the mean velocity alone, provided that the constants \( C_\epsilon \) and \( C_u \) are known. When values for \( C_\epsilon \) and \( C_u \) have been determined, they can then be used to infer the small-scale velocities and so we can estimate \( \epsilon \) (and thus \( \eta \)) from a measurement of \( U \) only.

The hot-wire anemometry experiments cannot be executed at the same high turbulence conditions as the APART measurements. One of the limiting properties is the finite sampling speed of the hot wire. This sampling frequency \( f_s \) limits the temporal resolution. Of relevance is the ratio

$$R_s = \frac{\frac{1}{2} f_s}{U/\eta} \quad (3.14)$$

where the factor \( \frac{1}{2} \) represents anti-aliasing filtering at half the sampling frequency \( f_s \). In order to successfully sample the flow, this ratio should be higher than 1. The probe size is another factor which determines the resolution. Similar to Eq. 3.14 we can define the probe length ratio:

$$R_l = \frac{\eta}{l_p}. \quad (3.15)$$

For decreasing turbulence intensity, \( R_s \) and \( R_l \) will decrease as well and when they approximate 1 the true \( \epsilon \) will be measured. This leads to a reliable estimate of
3.5 Jet characterisation

Figure 3.8: Calculation of $C_u$ (solid squares) and $C_\epsilon$ (solid circles) for different turbulence levels at 38 nozzle diameters above the nozzle.

$C_\epsilon$ and $C_u$. The values found for $C_\epsilon$ and $C_u$ can then be used to infer the small-scale velocities. The required derivatives were computed from time series of velocity measurements. The resulting $C_\epsilon$ and $C_u$ for a range of mean velocities (and therefore Reynolds numbers) are shown in Fig. 3.8.

Alternatively, all averages required for jet characterization can be computed from the energy spectrum $E(k)$ of the velocity field as in Fig. 3.9. We will discuss turbulence spectra in more detail in Chapter 5 but for now we will summarize that (one-dimensional) energy spectra are defined as the Fourier transforms of a function $R_{ij}(\vec{r}, t)$:

$$E_{ij}(k_1) = \frac{1}{\pi} \int_{-\infty}^{\infty} R_{ij}(r_1, t) e^{-ik_1 r_1} dr_1,$$

(3.16)

where $R_{ij}(\vec{r}, t)$ is the autocorrelation function of the velocity

$$R_{ij}(\vec{r}) = \langle u_i(\vec{x} + \vec{r}) u_j(\vec{x}) \rangle.$$

(3.17)

with $k_1$ the wavenumber, $r_1$ the separation and $i, j$ indicating the respective components of the two velocity vectors.

In hotwire measurements, structures of size $l = 2\pi/k$ are swept along the probe with mean velocity $U$ resulting in a frequency $f$. For all following spectra we will use wavenumbers $k$ rather than the frequency using the relation $k = \frac{2\pi f}{U}$.

For now it is enough to realize that turbulence spectra have a very steep fall off according to $E(k) \approx k^{-5/3}$. The rms velocity is the integral over the spectrum

33
3 Experimental setup & jet characterisation

Figure 3.9: Energy spectra $E(f)$ of the jet turbulence, measured by means of hotwire velocimetry. The spectra are measured for increasing Reynolds number, $Re_\lambda = 135$, 162, 212, 257 and 283 for spectra 1 to 5, respectively.

Figure 3.9: Energy spectra $E(f)$ of the jet turbulence, measured by means of hotwire velocimetry. The spectra are measured for increasing Reynolds number, $Re_\lambda = 135$, 162, 212, 257 and 283 for spectra 1 to 5, respectively.
and as most energy is contained in the lower wavenumbers (or frequencies), it is not very sensitive to high frequency cut-off due to probe resolution. Therefore, the measured \( C_u = 0.234 \pm 0.005 \) does not strongly depend on \( U \) and reaches its asymptote \( C_u = 0.234 \) at 15 ms\(^{-1} \). On the other hand, the dissipation is proportional to \( \int E(k) k^2 dk \), which does depend strongly on the high frequency resolution. Consequently, the measured \( C_\epsilon \) is susceptible to errors due to the inability to resolve the highest frequencies. This is seen in the measured \( C_\epsilon \), which decreases strongly with increasing velocity for \( U > 8 \) ms\(^{-1} \). In this dataset the plateau reached at small \( U \) is not well defined, so that the estimated true \( C_\epsilon = 0.47 \pm 0.04 \) has a large uncertainty.

From these numbers we now can compute \( \epsilon \) from the measured mean velocity as

\[
\epsilon = C_\epsilon \frac{u^3}{L} = C_\epsilon C_u^3 \frac{U^3}{L} \approx 6.0 \cdot 10^{-3} \frac{U^3}{L}.
\]

(3.18)

To check the found scaling parameters we will now collapse the measured spectra of Fig. 3.9 on a single curve using \( \epsilon \) as found through Eq. 3.18. Clearly, the highest frequencies are unresolved at the largest mean velocities, so that the normalization of the spectra depends on the extrapolated dissipation \( \epsilon \). However, since \( u \) is not very sensitive to resolution, normalisation based on directly measured values of \( u \) would yield spectra that are only slightly different. The wavenumbers are normalized to 1 at the Kolmogorov length scale \( \eta \), resulting in

\[
k^* = \frac{2\pi f}{U},
\]

where the \( 2\pi/U \) translates the frequency with which structures are swept by the hotwire into wave numbers.

The second similarity hypothesis predicts that in the inertial subrange (the range of scales where the geometry of large eddies is lost and dissipation is not yet felt), the ideal energy-spectrum function is given by \( E(k) = c_k \epsilon^{2/3} k^{-5/3} \). Thus, all spectra can be collapsed at the Kolmogorov scale as \( E(k = \eta) = c_k \) by normalisation

\[
E^*(k^*) = E_f(f(k^*)) \frac{U}{2\pi \epsilon^{2/3} \eta^{5/3}}.
\]

(3.19)

As is demonstrated in Fig. 3.10, all spectra now collapse reasonably. It must be noted that in Fig. 3.10 the smallest and largest frequencies that are just resolved differ by an order of magnitude. As a last check of our measured spectra, we compare them to an analytical form which is known to accurately reproduce experimental turbulence spectra [49]. In three dimensions this spectrum takes the form

\[
E(k) = c_k \epsilon^{2/3} k^{-5/3} f_L(kL) f_\eta(k\eta),
\]

(3.20)

where \( f_L(kL) \)

\[
f_L(kL) = \left( \frac{kL}{((kL)^2 + C_L)^{1/2}} \right)^{5/3+2}
\]

(3.21)
models the small wavenumber behaviour of the spectrum so that \( E(k \ll 1/L) \propto k^2 \) and
\[
f_\eta(k\eta) = \exp \left[ -\beta \left( \left( (k\eta)^4 + c_\eta^4 \right)^{1/4} - c_\eta \right) \right]
\]
models the large wavenumber dissipative behaviour. The accepted values of the constants are \( C_L = 6.78 \), \( \beta = 5.2 \) and \( c_\eta = 0.40 \), with the Kolmogorov constant \( c_k = 1.62 \). The normalized spectrum then becomes
\[
E^*(k^*) = c_k \epsilon^{2/3} k^{*-5/3} f_L(kL/\eta) f_\eta(k^*)
\]
and only depends on the Taylor scale Reynolds number \( Re_\lambda^1 \).
\[
\frac{L}{\eta} = \left( \frac{3}{20} \right)^{3/4} Re_\lambda^{3/2}.
\]
From the 3D spectrum the one-dimensional spectrum follows through isotropy
\[
E_{11}(k_1) = \int_{k_1}^{\infty} \frac{E(k)}{k} \left( 1 - \frac{k_1^2}{k^2} \right) dk.
\]
These spectra are compared to our measured spectra in Fig. 3.10 where we only show the measured spectra at the smallest, the largest and three intermediate Reynolds numbers. It is seen that, although there is quite a large uncertainty in scaling constant \( C_\epsilon \), the model agrees very well with the measured spectra.

### 3.6 Setup parameters

We can adjust the turbulence by setting the control parameter \( U \). In Table 3.1 the flow properties derived from this parameter are calculated. Let us now consider the conditions (geometrical, temporal, optical) that have to be met in order to successfully use the setup for quantitatively accurate measurements.

**Length scales**

The jet was designed to fit in an optics laboratory, so it is a relatively small setup (with a nozzle size of 1 cm). To achieve isotropic homogeneous turbulence, a range of scales as large as possible is aspired. On the 1000 \( \times \) 1000 pixel camera we can just resolve 3 decades of scales. When taking the nozzle size as an estimation of the integral length scale, this sets the Kolmogorov scale to \( O(10 \mu m) \) corresponding to a turbulence level \( Re_\lambda \approx 600 \). In our experiment maximum turbulence is ultimately limited by the maximum flow of compressed air that is available in our lab, resulting

\[1\text{The factor } \left( \frac{3}{20} \right)^{3/4} \text{ is a good approximation, but not entirely correct. However, it was adopted to compare our spectra to those in [49].}\]
Figure 3.10: The normalized energy spectra for $Re_\lambda = 135, 162, 212, 257$ and $283$. Normalisation was done using Eq. 3.18. The dashed lines indicate the corresponding synthetic spectra as described in Eq. 3.23 based on the jet characterisation parameters, where 1) represents the spectrum for $Re_\lambda = 135$ and $Re_\lambda = 283$.

<table>
<thead>
<tr>
<th>$U$(ms$^{-1}$)</th>
<th>$u$(ms$^{-1}$)</th>
<th>$\epsilon$($\cdot10^3$)(m$^2$s$^{-3}$)</th>
<th>$\eta$(µm)</th>
<th>$v_\eta$(ms$^{-1}$)</th>
<th>$\tau_\eta$(µs)</th>
<th>$R_\lambda$</th>
<th>$R_s$</th>
<th>$R_l$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.52</td>
<td>0.36</td>
<td>2.1 $\cdot$10$^{-3}$</td>
<td>200</td>
<td>0.075</td>
<td>2670</td>
<td>84</td>
<td>6.58</td>
<td>1.00</td>
</tr>
<tr>
<td>4.45</td>
<td>1.04</td>
<td>0.053</td>
<td>89</td>
<td>0.168</td>
<td>533</td>
<td>145</td>
<td>1.00</td>
<td>0.45</td>
</tr>
<tr>
<td>10</td>
<td>2.3</td>
<td>0.60</td>
<td>49</td>
<td>0.30</td>
<td>158</td>
<td>217</td>
<td>0.25</td>
<td>0.32</td>
</tr>
<tr>
<td>20</td>
<td>4.7</td>
<td>4.8</td>
<td>29</td>
<td>0.51</td>
<td>56</td>
<td>307</td>
<td>$\ll$ 1</td>
<td>0.14</td>
</tr>
<tr>
<td>30</td>
<td>7.0</td>
<td>16</td>
<td>21</td>
<td>0.70</td>
<td>30</td>
<td>375</td>
<td>$\ll$ 1</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>9.3</td>
<td>38</td>
<td>17</td>
<td>0.97</td>
<td>19</td>
<td>434</td>
<td></td>
<td></td>
</tr>
<tr>
<td>50</td>
<td>11.7</td>
<td>75</td>
<td>15</td>
<td>1.0</td>
<td>14</td>
<td>485</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3.1: Flow properties based on jet characterisation parameters. As a function of mean velocity $U$ the following properties are directly calculated: rms velocity $u$, dissipation rate $\epsilon$, Kolmogorov scale $\eta$, Kolmogorov velocity $v_\eta$, Kolmogorov time $\tau_\eta$, the Taylor scale Reynolds number $R_\lambda$, the sample frequency ratio $R_s$ as defined in Eq. 3.14 and the probe length ratio as defined in Eq. 3.15. The latter two ratios are only calculated for values near 1.
in the maximum reachable turbulence level $Re_\lambda \approx 450$ and Kolmogorov scale $\eta = 16\ \mu m$. The objective that is used to image the tagged lines has the object and image distances set for minimal aberrations resulting in a visualized region of $6.5 \times 6.5 \text{mm}^2$. This results in an effective pixel size of $6.5 \times 6.5 \mu \text{m}^2$. This is approximately enough to resolve the full spectrum.

**Time scales**

Depending on the goal of an experiment, we need to change the tag-probe time. The delay should be very short (near-instantaneous) for 2-point correlation measurements, and should be as long as possible when looking at other (Lagrangian) properties such as material line deformations. For short times there are two limitations:

- NO creation is a relatively slow process that occurs on the time scale of microseconds. After $1\ \mu s$ there is barely enough NO made to produce a visible LIF signal and only after $3\ \mu s$ to $5\ \mu s$ the NO concentration is at a maximum.

- The object magnification in the stream-wise and transverse direction is equal. This means that, even for the highest turbulence level, the line displacement based on the rms velocity after $1\ \mu s$ is less than 2 pixels (in stream-wise direction). This small displacement results in poor resolution and measurements show that the delay should indeed be greater than $1\ \mu s$ in order to find displacements with sufficiently small relative errors.

When the goal is to observe line deformations over longer times the following constraints will have to be met:

- The interpretation of a velocity field can only be done when the displacement of an element on the line is unambiguous. This means that the line should not fold back or even self-intersect. The maximum interrogation delay is therefore set by the shortest eddy turn-over time. It is readily appreciated that the shortest turnover times occur on the smallest scale (although the turnover velocities are smallest there). This follows from the simple relation:

$$\frac{u(l)}{u(L)} = \left(\frac{l}{L}\right)^{1/3}$$

(3.25)

where $u(L)$ is the large scale velocity, for which we take the rms value of the velocity field. Therefore:

$$\frac{t(l)}{t(L)} = \frac{l}{u(l)} = \frac{l}{L/u(L)} = \left(\frac{l}{L}\right)^{2/3}.$$  

(3.26)

So, the probe time delay is set by the Kolmogorov time. At $Re_\lambda \approx 450$ the Kolmogorov time is $\tau_\eta \approx 18\ \mu s$. 

---

3 Experimental setup & jet characterisation
3.6 Setup parameters

- The probe beam diameter has to be sufficiently large to encompass the deformed line. The maximum displacement difference along the line can be conservatively estimated as $u \Delta t \approx 450 \mu$m so that a cross-section of at least $450 \times 450 \mu m^2$ perpendicular to the line has to be illuminated by the read laser. In practice it was found that an area of at least $2 \times 2 mm^2$ is needed to illuminate all NO. The difference can be accounted for by the non-uniform (normal) intensity distribution in the beam and imperfect alignment of the beam traversing the readout area. The dye laser produces an intensity of approximately $1.25 mJ/mm^2/pulse$ in the probe region which is found to be just sufficient to induce LIF such that the signal to noise ratio is adequate for line-fitting.

Line width

We aim to write a line as thin as possible so that the width of the line is small compared to the eddies ($\eta = 16 \mu$m) that the line must resolve. In principle this is set by the diffraction limit. For the rectangularly shaped excimer laser beam with the $f = 250$ mm composite focusing lens a waist of $w_0 = 50 \mu$m is found. This might be further reduced by decreasing the focal length

$$w_0 \approx f \theta$$

(3.27)

where $\theta$ is the diffraction angle of the beam exiting the laser. However, decreasing the focal length will also increase the beam divergence. We will assume Gaussian optics as an approximation

$$w(z) = w_0 \left(1 + \left(\frac{\lambda \cdot z}{\pi w_0^2}\right)^2\right)^{1/2}$$

(3.28)

which is characterized by the Rayleigh range $z_R = \frac{\pi w_0^2}{\lambda}$ (the distance from the waist over which the beam expands by a factor of $\sqrt{2}$). Due to the non-linear nature of the NO creation process, shorter focal lengths will rapidly decrease signal intensity at the line edges. Minimizing the waist size by decreasing the focal length also has the adverse effect that the Rayleigh range $z_R$ is decreased. Going to even smaller beam widths will not significantly help decreasing the tagging line widths, since diffusive broadening will play a significant role, even on short timescales. With a diffusion constant of $D \approx 2 \cdot 10^{-5} m^2s^{-1}$, the diffusive spreading in an interrogation time of $20 \mu$s is $2 \cdot 10^{-5} m$. This is only a lower limit because the diffusion is enhanced due to laser light absorption. This point will be discussed in more detail in Chapter 4.
3 Experimental setup & jet characterisation

3.7 Measurements

Let us now look at a set of 4000 deformed lines, where the jet was set so that a mean velocity of \( \approx 45 \text{ ms}^{-1} \) was expected and the tag-probe delay was set to 8 \( \mu \text{s} \). The histograms versus the position along the line are plotted in Fig. 3.11a. The histograms of 4 neighbouring line positions are binned. The histogram of the instantaneous velocity has a Gaussian shape whose width and mean do not vary significantly along the line. This is better seen in Fig. 3.11b. Here the averaged mean and rms velocity are plotted for all line positions. It should be noted that the measurements match the calculated values \( U = 45 \text{ ms}^{-1} \) and relative rms velocity of 25% of mean.

![Figure 3.11: a) Velocity histogram versus position along the axis transverse to the flow. b) Mean and rms values of the velocity versus position based on the histogram.](image)

If the line intensity \( I \) and width \( \sigma \) are examined, we do see a strong position dependence. In Fig. 3.12a the histograms of the line intensity from the same dataset as the previous graph are plotted. There is a striking increase in intensity toward the line center. This is to be expected since this position corresponds with the beam waist of the write laser.

It is also noted that in the line center the maximally resolvable intensity of the 12 bits resolution is reached. One can see the peak at maximum intensity \( I = 4096 \) and the immediate drop-off afterwards, suggesting that the Gaussian-like shape was cut off prematurely. It is important that the image intensity is attenuated such that the camera does not saturate in the line focus, since this can result in a flattened top of the velocity distribution, resulting in a less than optimal fit of the line center. At the same time care must be taken that the signal intensity at the line edges remains high enough with respect to the Poisson noise. It should also be considered that there is a significant fluctuation in the write laser power in time. Together with the non-linear manner in which NO is created, this accounts for the spread in the beam intensity.
Although it is hard to discern from Fig. 3.12a, it has been noted that, contrary to the beam profile of the tagging laser, close to the line center the width of the line is actually slightly increasing. This can be accounted for by effects due to increased energy in the line center: heightened diffusivity and convection. It is not clear if there is also a photochemical reaction based contribution. This topic will be further explored in Chapter 4.
3 Experimental setup & jet characterisation
4 Chemistry and physics of APART

4.1 Introduction

In order to gain understanding in the manner in which the technique of APART can be used, it is essential that the chemical mechanisms that allow us to create and visualize particles are well understood. Important parameters that should be considered, are, among others, heat production, creation time, production efficiency, lifetime and visualisation efficiency. These data will help us to investigate the possibilities and limitations of our experiments, in terms of temporal and spatial scales, suitable environments and accuracy. Furthermore, understanding the underlying mechanisms will help us to optimally tune the technique to our experiments.

In this chapter we will investigate to what extent the light-induced creation of tracers perturbs the flow. We will make an estimation of the amount of energy that is deposited in the flow by measuring energy absorption and discussing measurements done with Laser-Induced Fluorescence Thermometry. This absorption of energy along the beam path will result in local heating up and radial advection. We will investigate these heating effects on the transient line shape and show that line widening indeed corresponds with the estimated temperature increase.

4.2 Photochemical creation of NO

This paragraph will be a summary of the investigations that have been undertaken to understand the NO creation process. Most of the results that will be discussed have been obtained by Schoemaeker et al. [53]. Work done by other authors will be referenced as such.

4.2.1 Spectroscopic investigations

In order to find the optimal wavelength at which to create NO, a standard APART setup as introduced in Chapter 3 was used to create nitric oxide. An ArF excimer laser was operated at 193 nm for tagging and a Nd:YAG pumped dye laser was operated at 226 nm for probing. The probe laser was fired with a delay of 3 μs with respect to the tagging laser. During the experiment the tagging wavelength was
scanned over its tuning range (193.0 nm to 193.8 nm) and the light intensity of the stimulated emission within a wavelength range from 236 nm to 248 nm was recorded at each excitation wavelength. This data is plotted in the lower panel of Fig. 4.1. Since the visualisation of NO is done by 1-photon fluorescence, the fluorescent light intensity is directly proportional to the product of the probe laser intensity and the amount of NO present if the excitation level is not yet saturated. Because we do not alter the wavelength and intensity of the probe beam, the fluorescent light is only dependent on the NO density. The amount of NO created is, however, dependent on the wavelength of the write laser. The spectrum shows strong resonance peaks at 193.26, 193.29, 193.40 and 193.44 nm and low intensity regions centered around respectively 193.22 nm and 193.42 nm.

These low intensity regions are the result of strong absorption of the tagging beam by oxygen. This effect is present in the focus region, but mainly along the total path the beam has travelled. This results in a considerable reduction of the laser power at the measurement region. The upper panel of Fig. 4.1 shows the Raman scattering spectrum of the excimer laser beam by O\textsubscript{2}. Raman scattering is an inelastic, linear scattering process and the Raman scattered light by O\textsubscript{2} can thus be used as an indicator of the beam intensity. It has been found that the absorption of laser light is due to excitation of oxygen\textsuperscript{1}. This 1-photon absorption process occurs in the B(v'' = 4) \rightarrow X(v' = 0) band of the Schumann-Runge system [54]. The extends of these absorption bands are indicated by the dashed lines in both panels of Fig. 4.1. The B(v'' = 4) state is strongly predissociated due to couplings to repulsive states that converge to O(\textsuperscript{3}P) atoms, marked 6 in Fig. 4.2a. In tagging experiments it is important to tune the laser carefully, such that the wavelength lies outside the Schumann-Runge bands. This will be emphasized by the absorption measurements in Section 4.5.1.

Outside these bands we can distinguish strong peaks in the NO spectrum. It is speculated that these peaks are due to 2+1-photon transitions in oxygen, marked as 1,2,3 in Fig. 4.2a. The first two photons are in resonance with a Rydberg state converging to X\textsuperscript{2}Π\textsubscript{g} of O\textsubscript{2}\textsuperscript{+}. A Rydberg state is a state of an atom or molecule in which one of the electrons has been excited to a high principal quantum number orbital. Classically, such a state corresponds to putting one electron into an orbit whose dimensions are very large compared to the size of the ion core. Subsequently, these high Rydberg states may auto-ionize into O\textsubscript{2}\textsuperscript{+} (X\textsuperscript{2}Π\textsubscript{g}), or a third photon may be absorbed. This third photon is in resonance with a super-excited Rydberg state [55]. Laser-induced fluorescence has been recorded in the 200 – 260 nm range (similar to the results of Vershlis [56]) resulting from ionisation from the super-excited neutral O\textsubscript{2} to the fluorescent levels of A\textsuperscript{2}Π\textsubscript{u} marked as 5 in Fig. 4.2a. If one examines the emission spectra while changing the excitation wavelength, it is observed that new lines appear for the 2+1 photon resonance wavelengths. Additionally, fluorescence

\textsuperscript{1}Although the absorption occurs in oxygen, one does not need to use oxygen as an indicator of light attenuation. For example, the N\textsubscript{2} Raman spectrum exhibits the same bands.
4.2 Photochemical creation of NO

Figure 4.1: Excitation spectra. Upper spectrum: The 1st Stokes of Raman scattered O$_2$ as indicator of the local laser intensity as a function of photolysis wavelength. Lower spectrum: laser-induced fluorescence at 236 nm of the created NO as a function of photolysis wavelength.

from $B^3\Sigma_u^-$ to $X^3\Sigma_g^-$ can be observed (marked as 7).

During the laser-induced photolysis process in which oxygen is excited, molecular nitrogen may also be excited. With a single 6.42 eV (= 193 nm) photon only the $A^3\Sigma_u^+$ state can be reached, but this 1-photon process is unlikely, since a singlet-triplet transition is electronically forbidden [57]. However, experiments by Bominaar et al. [58] have shown that a 3-photon transition (marked as 1,2,3 in Fig. 4.2b) is in resonance with the Rydberg states that converge to the $B^2\Sigma_u^+(v' = 2)$ state of N$_2^+$. Subsequently, these high Rydberg states may now auto-ionize. Indeed, experiments have been performed that show fluorescence from $B^2\Sigma_u^+$ to the $X^2\Sigma_g^+$ ground state of the ion (marked as 4).

The excitation pathways just described do not seem to be enough to explain the nitrogen photolysis during APART tagging. It was observed by Bominaar et al. [58] that an excitation spectrum of nitrogen in air exhibits resonances at the same wavelengths as are observed in the excitation spectrum of oxygen. Furthermore, the
Figure 4.2: Potential energy diagram of the electronic and vibrational states of a) oxygen and b) nitrogen, as a function of the internuclear distance. The dotted lines indicate the ground state energies of the different electronic states and the arrows indicate the excitation and relaxation during APART "writing".

The spectrum of nitrogen in air is significantly different from that of nitrogen in a pure nitrogen environment. That has led to the conclusion that there is a mechanism that allows for energy transfer between oxygen and nitrogen, the exact pathways of which are not yet known.

To summarize, 1-photon excitation results in \( \text{O}_2(\text{B}^3\Sigma^-) \) being created, which easily dissociates in atomic oxygen. By absorption of 2 more photons highly excited oxygen is created which can easily lead to \( \text{O}_2^+ \). At the same excitation wavelengths nitrogen is excited to end up in ionized and excited states.

Besides these species that are assumed to be critical in NO production, many other products are reported to be created during photolysis, such as ozone (\( \text{O}_3 \)). A subset of the species that will be present in air after the photochemical processes that have been induced by the "write" laser pulse will chemically interact to eventually create ground-state nitric oxide. The created NO is easily visualized by applying a 226 nm laser to induce fluorescence. The fluorescence signal is observed by
4.2 Photochemical creation of NO

Figure 4.3: Calculated net NO production as a function of the time after the write laser pulse. The simulated values are compared with measurements from the integrated NO LIF signal in atmospheric air. Each point is the accumulation of 100 tagged lines. The signal intensity is rescaled to the simulated values.

integrating NO emission bands at 237 nm \([A^2\Sigma^+(v' = 0) \rightarrow X^2\Pi(v'' = 0)]\), 248 nm \([A^2\Sigma^+(v' = 0) \rightarrow X^2\Pi(v'' = 1)]\) and 259 nm \([A^2\Sigma^+(v' = 0) \rightarrow X^2\Pi(v'' = 2)]\).

4.2.2 NO formation chemistry

In the previous paragraph an overview has been given of the spectroscopic experiments that have been performed to gain insight in the air photolysis. What remains is to obtain a better understanding of the chemical reactions that eventually lead to the creation of NO. Here, some of the work that has been performed by Schoe maeccker \textit{et al.} [53] in collaboration with Nandula and Pitz [8] will be discussed.

In order to study the chemical NO tag formation, a detailed chemical kinetics model was developed [20, 8]. The model consists of chemical formation and destruction reactions and is solved by means of the Chemkin and Senkin chemical kinetics simulation suite. The package allows modelling of the evolution in time of the species concentrations, given an initial composition. This will help to isolate the main reactions that are of importance for NO creation. The suite can also calculate the thermal energy released during the reaction processes. The current model
4 Chemistry and physics of APART

<table>
<thead>
<tr>
<th>Reaction</th>
<th>No</th>
<th>Reaction rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{N}_2 = \text{N} + \text{N}$</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td>$\text{N}_2^+ + e^- = \text{N} + \text{N}$</td>
<td>2</td>
<td>$2.55 \cdot 10^{-7}$</td>
</tr>
<tr>
<td>$\text{N} + \text{O}_2 = \text{NO} + \text{O}$</td>
<td>3</td>
<td>$8.5 \cdot 10^{-17}$</td>
</tr>
<tr>
<td>$\text{N} + \text{O} + \text{M} = \text{NO} + \text{M}$</td>
<td>4</td>
<td>-</td>
</tr>
<tr>
<td>$\text{NO} + \text{N} = \text{N}_2 + \text{O}$</td>
<td>5</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 4.2: Main processes leading to production and destruction of NO.

involves 119 reactions and 30 species including ions.

Since for many of the species the volume fractions in air after the photolysis process are not known, there is a large number of non-fixed parameters. For example assuming additional (dissociated) water vapor will significantly increase the predicted NO creation. Current research is performed to obtain quantitative data on the concentrations of species that are present by means of cavity ring down spectroscopy (CRDS) [59]. Recent comparisons between the CRDS experiments and the simulations show good agreement of the amount of NO (42 ppm) at 3 $\mu$s after photolysis but an overestimation of ozone in the simulations (140 ppm vs 75 ppm).

Another property of the gas mixture that the model should reproduce is the species evolution in time, in particular nitric oxide. It can be seen in Fig. 4.3 that the experiment and simulations display roughly the same trend, but the rise in NO concentration occurs earlier than is estimated by the model. Also the destruction of NO is more significant than predicted. Different reasons for the discrepancies between the model and the experiment can be found. The model is limited in several aspects. Both flow dynamics and thermodynamics are not included in the model. Also, species created in excited electronic states after photolysis may follow different reaction dynamics than the ground state species, which is not yet taken into account.

Another reason for the discrepancy may lie within the experiment. Diffusive broadening of the written line may cause it to extend out of the probe laser beam. This would give an underestimation of the amount of nitric oxide present at longer times.

The analysis of the chemical simulations shows that the reactions given in Table 4.2 give the most significant pathways in the creation and destruction of nitric oxide. The model demonstrates that the amount of N present is the most important factor in NO production. Atomic nitrogen can be produced via direct photo-dissociation (Table 4.2 process 1) or via dissociative recombination of $\text{N}_2^+ + e^-$ (Table 4.2 process 2).

Currently, the link between spectroscopic data and chemical data is still weak. An important step in understanding the creation of NO would be to understand how the creation efficiency of nitric oxide depends on the photolysis of the air con-
stituents. Or, more specifically: how can the resonances in the oxygen spectrum be linked to peaks in the NO LIF spectrum. A possible mechanism could involve the $N_2 + O_2^+ \rightarrow N_2^+ + O_2$ electron transfer. It is known that, on resonance, additional oxygen is ionized to $O_2^+$. Then, due to the electron transfer excess $N_2^+$ could be produced resulting in more NO created through reactions (2) and (3) in Table 4.2.

4.3 Line deformation during tagging

4.3.1 Introduction

A feature of MTV techniques that may prove problematic, is the (anomalously large) diffusion of nitric oxide molecules. This will make the measurement of turbulence properties substantially more difficult. Diffusion interferes with the small-scale motion of the fluid, whilst large-scale (inertial range scale) motion is seen filtered through molecular diffusion. In this section we will discuss in detail the relation between molecular and turbulent diffusion.

We will describe a model that explains the effect of heat absorption on the diffusion of the written NO lines. As the diffusive broadening of these lines sets a limit on the accuracy of APART when it is used for velocimetry, it is important to understand the essential mechanism that leads to (i) faster broadening of the line than given by the molecular diffusion constant of NO and (ii) a non-vanishing diffusion at infinite pressure. These effects have been encountered in experiments and we will now discuss the framework to give qualitative explanations of both aspects.

4.3.2 The shape of a written line

The key point in understanding line broadening is the temperature rise of the gas in the focus of the writing laser beam. This temperature increase affects the values of the mass and heat diffusion coefficients. It also changes the density of the gas, which results in a convection velocity and an enhanced flow of molecules out of the laser beam focus. Finally, the enhanced temperature will also affect the detection of NO fluorescence, due to the population change of the probed rotational state.

First, we must realize that mass and heat diffuse at approximately the same rate. In a simple (hard sphere) kinetic view, the mass diffusion coefficient $D$ is proportional to the product of the thermal velocity $v_{th}$ and the mean free path between collisions $\lambda_{mf}$,

$$D \sim v_{th} \lambda_{mf},$$

while heat diffuses at a rate

$$\kappa = \frac{\lambda}{\rho c_v} \sim v_{th} \lambda_{mf}$$

where $\lambda$ is the heat conduction coefficient, and $c_v$ the specific heat at constant volume. The thermal velocity is proportional to the absolute temperature $v_{th} \sim T^{1/2}$,
4 Chemistry and physics of APART

<table>
<thead>
<tr>
<th></th>
<th>M</th>
<th>σ [Å]</th>
<th>ϵ/k [K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air*</td>
<td>28.97</td>
<td>3.617</td>
<td>97.0</td>
</tr>
<tr>
<td>NO</td>
<td>30.01</td>
<td>3.470</td>
<td>119</td>
</tr>
<tr>
<td>N₂</td>
<td>28.02</td>
<td>3.681</td>
<td>91.5</td>
</tr>
<tr>
<td>O₂</td>
<td>32.00</td>
<td>3.433</td>
<td>113</td>
</tr>
</tbody>
</table>

Table 4.3: Properties of relevant gases. \( M \) is the molecular mass, Lennard-Jones parameter \( σ \) determines the collision diameter and Lennard-Jones parameter \( ϵ/k \) is used to scale the interaction potential in Eqn. 4.4. * The molecular mass and Lennard-Jones parameters of air are averages based on its constituents.

while the mean free path is inversely proportional to the density, \( λ_{mf} \sim ρ^{-1} \). The ideal gas law is \( p = ρRT \), so that at constant pressure, \( D,κ \sim T^{3/2} \).

To describe real gas effects we will use the Chapman-Enskog kinetic theory. This gives expressions for the transport coefficients in terms of the potential energy of interaction between a pair of molecules. This potential energy is calculated by means of the empirically determined Lennard-Jones potential:

\[
\varphi(r) = 4\epsilon_{AB} \left[ \left( \frac{\sigma_{AB}}{r} \right)^{12} - \left( \frac{\sigma_{AB}}{r} \right)^{6} \right],
\]

(4.3)

Where \( σ_{AB} \) is the collision diameter for hard sphere collisions between species A and B, \( r \) the distance between a pair of molecules and \( ϵ \) a characteristic interaction energy. This results in the following expression for the diffusion rate \( D_{AB} \left[ m^2 s^{-1} \right] \) of a species A in a gas of species B

\[
D_{AB} = 1.8583 \cdot 10^{-7} \left( \frac{1}{M_A} + \frac{1}{M_B} \right)^{1/2} \frac{T^{3/2}}{ρ\sigma_{AB}^2 \Omega_{AB}},
\]

(4.4)

where \( M \) is the molecular mass, \( k \) the Boltzmann constant and \( Ω_{AB} = Ω(kT/ϵ_{AB}) \) is a dimensionless function of the temperature and the intermolecular potential field. For rigid spheres \( Ω_{AB} \) would be unity at all temperatures. Instead we assume a Lennard-Jones potential field and use values of \( Ω(kT/ϵ_{AB}) \) as known from literature [60]. The following empirical relations are used to combine the Lennard-Jones parameters of species A and B to find the parameters that will describe the diffusion of species A in B, and vice versa.

\[
σ_{AB} = \frac{1}{2} (σ_A + σ_B),
\]

(4.5)

\[
ϵ = \sqrt{ϵ_A ϵ_B}.
\]

(4.6)

Some values of the gas-specific parameters for gas as found by [60] are shown in Table 4.3.
4.3 Line deformation during tagging

Figure 4.4: Molecular diffusion rate of NO in air as function of the temperature. The lower line indicates the rigid sphere approximation and the upper line accounts for a Lennard-Jones interaction potential field between molecules.

The diffusion of nitric oxide in air can be calculated by using $\sigma_{\text{NO, air}} = 3.544 \text{ Å}$ and $\epsilon_{\text{NO, air}}/k = 107.4 \text{ K}$. The resulting diffusion rate is plotted in Fig. 4.4. Although a single power law does not fully describe $\Omega$, it gives a reasonable approximation for temperatures around ambient. Notice the significant difference when compared with the rigid sphere approximation (lower plot). If the Lennard-Jones potential energy is taken into account, the diffusion constant $D$ increases roughly with $T^{2.0}$ for low temperatures ($< 200 \text{ K}$) and $T^{1.65}$ at high temperatures ($> 700 \text{ K}$). At ambient pressure, the temperature dependency for the region between 200 K and 700 K can be well described by

$$D_{\text{NO, air}} = 1.16 \cdot 10^{-9} \cdot T^{1.7}. \tag{4.7}$$

Also, when applying the Lennard-Jones interaction potential in the heat conduction model we find that the heat conduction scales as $\lambda \sim T^{0.7}$, rather than $T^{0.5}$. We will use these circumstances in our model for the time-dependent line width, since now both $\rho \lambda$ and $\rho^2 D$ are approximately constant at constant pressure. The ratio $D/\kappa$ is called the Lewis number, $Le$, which is clearly of order unity.

We will base our model on a simple low Mach number approach, which applies at relatively long time scales $O(\mu s)$ after the NO was formed. Shorter time scales, which may involve shocks and sound, determine the initial line shape. We will describe the temporal evolution after these events. A key assumption is that on the time scales of interest, the pressure has already equilibrated through sound waves,
so that we can assume a constant pressure throughout.

### 4.3.3 Molecular diffusion

Let us first consider simple molecular diffusion. If the diffusion constants for mass and heat are truly constant, the diffusion equation in 1 dimension reads:

\[
\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2},
\]

with the fundamental solution

\[
C(x, t) = \frac{1}{(4\pi D t)^{1/2}} e^{-x^2/4Dt}.
\]

The full width at half maximum \(w\) of this distribution is given by \(w^2(t) = 16 \ln 2Dt\).

### 4.3.4 Mass and heat transfer

Assume that at \(t = 0\) a (Gaussian) initial temperature and \(NO\) concentration profile are established. Also assume that the air is an ideal gas, with the equation of state

\[
p = \rho R_{gas} T.
\]

Then the temperature satisfies

\[
\rho c_p \frac{dT}{dt} - \frac{dp}{dt} = \nabla \cdot (\lambda \nabla T), \quad \text{with} \quad \frac{d}{dt} = \left( \frac{\partial}{\partial t} + \vec{u} \cdot \nabla \right),
\]

where \(\vec{u}(x, t)\) is the velocity of the air. At the time scales of interest we can ignore the dynamics of the pressure, so that we drop the term \(\frac{dp}{dt}\). The tagged \(NO\) density \(y_t\) is described by the advection-diffusion equation

\[
\frac{\partial y_t}{\partial t} + \nabla \cdot (y_t \vec{u}) = -\nabla \cdot \vec{J}_t, \quad \text{with} \quad \vec{J}_t = -\rho D \nabla \left( y_t/\rho \right),
\]

while the total density \(\rho\) of the air satisfies the equation of mass balance

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = 0.
\]

The system of equations Eqs. 4.9-4.12 embodies the following physics: upon writing, the absorbed heat induces a temperature rise, which results in a lower density (through Eq. 4.9), which induces a convection velocity \(\vec{u}\) through Eq. 4.12, which

\[\text{Note that we cannot use the constant entropy equation of state, } p\rho^{-\gamma} = C, \text{ with } \gamma = c_p/c_v, \text{ because we have diffusion of heat.}\]

\[\text{This low Mach-number approach is rationalised in [61].}\]
then enhances the transport of the tagged molecules through Eq. 4.11. It will appear that this complicated set of partial differential equations can miraculously be solved analytically. We will restrict ourselves to one dimension, so that we can summarize the equations as

\[
\begin{align*}
\rho c_p \left\{ \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} \right\} &= \frac{\partial}{\partial x} \left( \frac{\lambda}{\partial x} \frac{\partial T}{\partial x} \right), \\
\rho \frac{\partial y_t}{\partial t} + \rho u \frac{\partial y_t}{\partial x} &= \frac{\partial}{\partial x} \left\{ D \rho \frac{\partial y_t}{\partial x} \right\}, \\
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x} (\rho u) &= 0, \\
\rho R_{gas} T &= p.
\end{align*}
\]

(4.13)  (4.14)  (4.15)  (4.16)

Let us now introduce non-dimensional variables through

\[
\begin{align*}
\tilde{\rho} &= \frac{\rho}{\rho_0}, \quad \tilde{T} = \frac{T}{T_0}, \quad \lambda = \frac{\lambda}{\lambda_0}, \quad \tilde{D} = \frac{D}{D_0}, \quad \tilde{x} = \frac{x}{R}, \quad \tilde{t} = \frac{t}{\tau}, \quad \tilde{u} = \frac{u}{U},
\end{align*}
\]

(4.17)

with the time scale \( \tau = R^2/D_0 \), and consequently the velocity scale \( U = R/\tau \), and where \( \rho_0, T_0 \) and \( D_0 \) refer to the standard condition \( (T_0 = 293 \text{ K}, p_0 = 1 \times 10^5 \text{ Nm}^{-2}) \), and \( R \) is the width of the initially written line. Omitting the \( \sim \) decoration, we can then write

\[
\begin{align*}
\rho \left\{ \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} \right\} &= \frac{1}{Le} \frac{\partial}{\partial x} \left( \frac{\lambda}{\partial x} \frac{\partial T}{\partial x} \right), \\
\rho \frac{\partial y_t}{\partial t} + \rho u \frac{\partial y_t}{\partial x} &= \frac{\partial}{\partial x} \left\{ D \rho \frac{\partial y_t}{\partial x} \right\}, \\
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x} (\rho u) &= 0, \\
\rho T &= 1.
\end{align*}
\]

(4.18)  (4.19)  (4.20)  (4.21)

4.3.5 Lagrangian coordinates

The convection velocity \( u(x, t) \) can be removed from our equations by moving to so-called Lagrangian (or Von Mises-) coordinates. They are defined by

\[
(x, t) \rightarrow (\Psi, s), \quad \text{with} \quad \Psi = \int_0^x \rho(x', t) \, dx', \quad \text{and} \quad s = t.
\]

(4.22)

In this new coordinate system the material derivative becomes

\[
\frac{\partial}{\partial t} + u \frac{\partial}{\partial x} = \frac{\partial}{\partial s} + (\rho u_{|x=0}) \frac{\partial}{\partial \Psi} = \frac{\partial}{\partial s}.
\]

(4.23)
if we take \( \rho u|_{x=0} = 0 \) at the lower bound of the integral (which could also be taken at \(-\infty\)). The inverse transform is
\[
x(\Psi, s) = \int_0^\Psi \frac{1}{\rho(\Psi', s)} \, d\Psi', \quad \text{and} \quad t(\Psi, s) = s.
\] (4.24)

The convection velocity \( u(x, t) \) can be recovered from the mass balance Eq. 4.20,
\[
\frac{\partial \rho}{\partial s} + \rho^2 \frac{\partial u}{\partial \Psi} = 0,
\]
so that
\[
u(\Psi, s) = -\int_0^\Psi \frac{1}{\rho^2(\Psi', s)} \frac{\partial \rho}{\partial s} \, d\Psi'.
\]

We now make the drastic assumption that for constant pressure the dimensionless heat conductivity and diffusion coefficient are given by
\[
\rho \lambda = 1, \quad \text{and} \quad \rho^2 D = 1.
\] (4.25)

This assumption was argued before on page 51. It has the pleasant consequence that the diffusive terms on the right-hand side of Eqs. 4.18 and 4.19 now have a constant (unit) diffusion coefficient in Lagrangian coordinates:
\[
\frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right) \rightarrow \left( \rho \frac{\partial}{\partial \Psi} \frac{1}{\rho} \rho \frac{\partial}{\partial \Psi} \right) T = \rho \frac{\partial^2 T}{\partial \Psi^2},
\] (4.26)

so that we are left with the equations
\[
\frac{\partial T}{\partial s} = \frac{1}{Le} \frac{\partial^2 T}{\partial \Psi^2},
\]
\[
\frac{\partial y_t}{\partial s} = \frac{\partial^2 y_t}{\partial \Psi^2},
\]
\[
\rho T = 1.
\] (4.27)

These equations can be solved analytically using the fundamental solution of the diffusion equation. Such a solution then proceeds as follows:

First we assume an initial Gaussian temperature profile where the temperature increase in the written line is a factor \( T_0 \) above ambient temperature at \( x = 0 \).
\[
T(x, t = 0) = 1 + T_0 e^{-x^2}.
\] (4.28)

This allows us to compute the Lagrangian coordinate \( \Psi \) at the initial time,
\[
\Psi = \int_0^x T^{-1}(x', 0) \, dx',
\]
and allows us to express the initial temperature in Lagrangian coordinates \( T(\Psi, 0) \). The temperature now satisfies the ordinary diffusion equation Eq. 4.27, and from the initial temperature profile \( T(\Psi, 0) \) we can compute the temperature at later times \( T(\Psi, s) \) through the fundamental solution,
\[
T(\Psi, s) = \frac{1}{(4\pi s)^{1/2}} \int_{-\infty}^{\infty} T(\Psi', 0) \, e^{-(\Psi-\Psi')^2/4s} \, d\Psi',
\] (4.29)
where we have taken the Lewis number $Le = 1$. A similar procedure can be followed to compute the time-dependent fraction of tagged molecules $y_t(\Psi, s)$. Finally, we will use Eq. 4.29 to revert back to physical variables,

$$
x = \int_0^\Psi \frac{1}{\rho(\Psi', s)} d\Psi = \int_0^\Psi T(\Psi', s) d\Psi'.
$$

(4.30)

Some of the integrals must be computed numerically, but this is a negligible effort compared to the numerical solution of partial differential equations.

The computed line profile $\rho_t(x, t)$ for $T_0 = 2$ and $t = 1$ is shown in Fig. 4.5. In the simple diffusion model without convection, the initial Gaussian density distribution with maximum $\tilde{\rho} = 1$ and width $\tilde{x} = 1$ evolves as

$$
\rho_t = \frac{1}{\pi(4t + 1)} e^{-x^2/(4t+1)}.
$$

(4.31)

It is seen that at $t = 1$ this solution differs from the complex model. Since we will have non-Gaussian line profiles, we will characterize line width always by the full width at half maximum $w$. Although the shape of the profile differs from Gaussian, it is difficult to see the difference in the experiment. It is most interesting to see how the width $w$ depends on time. Let us recall that for ordinary diffusion, the width increases with time as $w^2(t) = (16 \ln 2) D t$, with the factor $(4 \ln 2)$ accounting for...
for the ratio between squared half width and squared standard deviation. For the (dimensionless) temperature elevations $T_0 = 0, 1, 2$, the result is shown in Fig. 4.6.

A first observation is that the width of the tagged line in heated air increases more rapidly than in case of ordinary diffusion. Second, for short (scaled) times, the increase of the width is super-diffusive, that is, line widening does not scale as $w^2 \propto t$.

In order to further see the implications of this description, we must realize that the figure is in scaled variables. In particular, the time scale is $\tau = R^2/D_0$, which for our experiments, with $R = 50 \mu m$ and $D_0 = 2 \times 10^{-5} m^2 s^{-1}$, amounts to $\tau = 100 \mu s$ at atmospheric pressure. However, at a higher pressure of, let us say, $p = 10$ bars, it increases to $1000 \mu s$. In high pressure experiments, therefore, we are sensitive to small scaled times where the spreading curves start to bend non-diffusively. This will be shown in Section 4.4.2.

Incidentally, we must also realize that the diffusion exponent has dropped out from our theory and that the only way in which the physical parameters come in is through the rescaling of the axes of the figures. In these scalings there is an apparent contradiction since we have assumed that the rescaled $\rho^2 D = 1$, whereas $D_0 \sim 1/p$, or $\tau \sim p$. However, we must realize that the first scaling applies to the constant
pressure situation only, whereas the second relation serves to compare experiments at different pressures.

4.4 Line width experiments

Let us now test the convection model proposed in the previous section. The model yields interesting predictions for enhanced diffusion due to heating. Additionally anomalous scaling was expected for the diffusion as a function of the pressure.

4.4.1 Enhanced diffusion

Fig. 4.7 illustrates the widening of the lines in time. Each of the depicted lines is an average of 800 measured lines, written using the standard APART setup. These lines show that for times up to approximately 50 µs the width increase is nearly constant for the whole line, but with a slight bump in the center. For longer times, the edges of the line display a higher diffusion rate, but this can be accredited to the decreased line intensity resulting in a lower signal to noise ratio.

![Figure 4.7: Line width evolution of the experimental lines as a function of time. Each line is an average of 800 measured lines.](image)

Let us compare this data with the predictions shown in Fig. 4.6. We calculated the squared average line width $\langle w(x)^2 \rangle$ for each delay. The obtained line widths
were plotted versus time in Fig. 4.8a and zoomed in on shorter times in Fig. 4.8b. Both width and time have been normalized to non-dimensional variables, through Eq. 4.17. The standard deviation is always below 0.5% of the measured value.

It can be seen that for longer times the relation between \( w^2 \) and \( t \) seems linear. For short times the line slope is comparable with that of the model prediction for \( T_0 = 1 \) (solid line). Furthermore, we see that in this region the model predicts the bending behaviour in the experiments very nicely (see Fig. 4.8b). However, for longer times the line widening is faster than predicted and does not yet seem to asymptote to the rate associated with ambient temperature (that is, parallel to the dashed line). It is not clear why this is so. It may be that heating of oxygen plays an important role. In these measurements a write wavelength was chosen such that NO creation is optimal. However, this optimal peak is very close to a strong oxygen absorption band, as is shown in Fig. 4.1.

4.4.2 Pressure dependency

When only considering thermal diffusion at ambient temperature, the diffusion constant is inversely proportional to the density (pressure) so key information about the line spreading process can be learned from APART measurements at various pressures. Measurements where done by Bominaar et al. [58] at various pressures and gas compositions averaging over 10 images at each time delay. The lines were written in a closed pressure vessel with optical access using a slightly lower powered excimer laser (60 mJ/pulse) than in the APART setup. The narrow-band laser was not tuned to a wavelength for optimal NO production. Significant results were obtained by summing these 10 images, which partially masks fluctuations in the data.

The data was obtained by fitting the line cross section \( I(x, t) \) following the procedure described in Sect. 3.4 and averaging the line width over the line. The squared (dimensionless) width is plotted versus (dimensionless) time for different pressures in Fig. 4.9. The relation is quite linear and the slope increases with decreasing pressure as is expected from theory. In fact, contrary to the measurements discussed in Section 4.4.1, the line width data fits very nicely to the model prediction for \( T_0 = 1 \) (the dashed line). This may be because in these measurements a significantly lower percentage of oxygen was present. It must also be considered that now the write laser was not tuned on an optimal NO production wavelength, nor near to a strong oxygen absorption band.

In Fig. 4.10 we plot the diffusion rate as calculated from the slopes found in Fig. 4.9, plotted against the inverse pressure. The same trend is seen for lines written in environments with different amounts of oxygen. The plot contains both data measured at 0.24% \( \text{O}_2 \) and 0.49% \( \text{O}_2 \).

As expected there is a linear relation between diffusion and the inverse pressure.
Figure 4.8: Measured and calculated squared line width as function of time. Upper graph: The solid squares are measured averaged widths. The dashed line indicates the rate of diffusion based widening when $T_0 = 0$. The lower solid line shows the prediction for $T_0 = 1$ and the upper for $T_0 = 2$. Lower graph: Zoomed in on the area indicated in the upper graph.
Figure 4.9: Line width in still gas. For pressures of 1, 3, 6, and 11 bar (upper to lower curves), the width $w^2$ increases linearly with delay time. The dashed line is the model prediction at $p = 1$ and $T_0 = 1$. All lines are written in a 0.49% O$_2$ in N$_2$ environment.

From Fig. 4.10 we find a pressure dependency of the diffusion rate of

$$D = 0.37 \times 10^{-5} + 2.04 \times 10^{-5} \frac{1}{p}.$$  \hspace{1cm} (4.32)

This result for the diffusion coefficient is close to the value of the NO diffusion constant in plain air which is $D_{\text{NO,air}} = 1.8 \times 10^{-5} \frac{1}{p}$ as predicted by Eq. 4.4. However, the striking result that we observe is not that the diffusion constant $D$ indeed grows linearly with the inverse pressure, but that it also has an offset.

Using the theory of Sect. 4.3 we can now explain the behavior of the apparent diffusion rate that we have found experimentally. The argument is the following:

Let us first look at the normal diffusion, as indicated by the dashed line in Fig. 4.6. In order to compare this line to the measured pressure dependence of the diffusion coefficient as shown in Fig. 4.10, we must realize that in our analysis a changing pressure is only expressed in the rescaled time axis. Therefore, if we plot $t \frac{dw^2(t)}{dt} = D_0 t$ versus $t$, we find the dependence of the ordinary diffusion constant as a function of the pressure when we equate $p^{-1}$ to $t$. If $t \frac{dw^2(t)}{dt}$ would be fully linear this would give us for the pressure dependent diffusion $D_0/p$.

In other words, we have $D(p)$ versus $1/p$, which passes through the origin and the point $(1, D_0)$ when $p$ is measured in bars and $D_0$ is measured at a reference pressure of 1 bar.

For our convection model we see that the local slope of $w^2(t)$ is not constant
Figure 4.10: The diffusion constant as a function of inverse pressure. (for both 0.24% $O_2$ and 0.49% $O_2$). A linear relation is found, as shown by the dashed line.
anymore, but increases as \( t \) becomes smaller. It is on these times that we zoom in when increasing the pressure. Indeed, when re-examining Fig. 4.9 we can see the start of bending of the spreading rate at the highest pressures in the experiment.

Again, the pressure dependence of the convection model can be found by plotting \( t \, dw^2(t)/dt \) (\( D(p) \)) as a function of \( t \) (\( 1/p \)). This quantity is plotted in Fig. 4.11 for two values of the initial temperature increase \( T_0 \). We again find approximately straight lines, but now with an offset, and we notice the striking similarity to the measured pressure dependent diffusion in Fig. 4.10. In fact, with the choice of \( T_0 = 1 \), the agreement is good, even quantitatively. Therefore, we believe that we have satisfactorily explained the offset in Fig. 4.10.

The spreading of tagged lines is indeed determined by the heat deposited in the writing process. For our model to fit the data, the temperature increase must be of the order of the ambient temperature.

Remember that our problem has cylindrical symmetry, whereas our model assumes a slab geometry. It turns out that the mathematical trick that rendered our model exactly solvable does not work in cylindrical coordinates. However, we believe that our conclusions will be qualitatively the same in the cylinder symmetry.
4.5 Line temperature

In the experiments shown in Sect. 4.4 we compared the data with our model of anomalous diffusion due to local heating. Here we made an estimation of the maximum temperature increase in the line-center of $T_0 = 1$. We will now see if this heating correlates with other (indirect) measurements of the temperature along the focus of the written line. Different techniques can be employed to measure this property.

4.5.1 Energy absorption

One way to determine the heating of the tagged air is by measuring the amount of deposited energy per unit volume. The absorption of energy will result in local a temperature increase, and for high energy density it may also lead to breakdown (although much care is taken to prevent this). Let us consider the interaction of a coherent laser beam propagating along the $x$-axis with ambient air. The absorption length $\zeta$ is dependent on local gas properties, such as density $\rho$, temperature $T$ and wavelength, etc. According to Lambert-Beer

$$dE(x) = -\zeta(\rho, T, x, \ldots) \cdot E(x) dx,$$

where $E(x)$ is the energy present in the laser beam at position $x$ in the medium. If all gas properties are considered constant, the beam energy is

$$E(x) = E_0 e^{-x/\zeta},$$

where $E_0$ is the initial laser energy and $x$ the propagated distance. Notice that the absorption length $\zeta$ has the form of a characteristic length scale.

In this experiment the same ArF excimer laser that is used in APART measurements, is employed to write lines. However, initially, the laser is used unfocused and in broad-band mode. An Ophir L150 excimer power head is used to measure the laser power along the laser path. The mean laser power is measured, and the mean energy per pulse is calculated from it (repetition rate of 10 Hz). The interval between laser pulses is long enough for the heat to be dissipated.

A broad-band parallel laser beam with cross-section of $4 \times 1$ cm, a center frequency of 193 nm and a bandwidth of about $100 \text{ cm}^{-1}$ propagated through ambient air. The energy was averaged for 50 s per position and the probe was shifted along the laser beam over a distance of 2 m with samples taken at every 0.10 m. This data is shown in Fig. 4.12a. The mean energy decay trend was fitted using Eq. 4.34 resulting in an absorption length $\zeta$ of $5.1 \pm 0.1$ m.

In the light of the discussion in Section 4.2.1 it is easily understood that the absorption length is dependent on the wavelength of the laser light. If the wavelength is resonant with an oxygen state as shown in Fig. 4.1, energy absorption will occur more efficiently than when off-resonant. Fig. 4.12b shows the energy absorption versus distance for a narrow-band laser with bandwidth range of $0.5 \text{ cm}^{-1}$ FWHM,
tuned to a wavelength off-resonant with the strong absorption bands of oxygen. It can be seen that the absorption length is significantly longer than $\zeta$ for the broad-band laser beam. This is because the broad-band beam contains frequencies in the oxygen resonance range. We should realize that we are measuring a wavelength integrated absorption rate, especially for the broad-band beam, which could have spoiled our mono-exponential decay. Nevertheless, as Fig. 4.12 showed, both broad-band and narrow-band light yield a fairly good mono-exponential decay.

If the laser is used in a narrow-band setup, one can observe the absorption length dependence on wavelength. Since it is known that the attenuation of the laser beam is exponential it suffices to measure the laser intensity at two locations. $\zeta$ was calculated from the difference in the energy spectra measured at both positions. This is shown in Fig. 4.13. From this spectrum we find $\zeta$ values ranging from 8 m down to 3 m in the areas associated with oxygen bands. As expected, this graph is directly comparable with the Raman scattering of O$_2$ graph in Fig. 4.1.

The temperature rise in the probe volume is determined by the energy which is transformed into translational energy. O$_2$ molecules that dissociate contribute less than those that are quenched. With $E_{\text{diss}}$ the dissociation energy of oxygen and $h\nu$ the photon energy in the excimer laser, we find that the contribution to translational energy after dissociation is

$$E_{\text{diss}} = 41260 \text{ cm}^{-1} \quad h\nu = 51810 \text{ cm}^{-1} \quad \Delta E_\nu = 10550 \text{ cm}^{-1} = 0.2 h\nu$$

This means that the fraction of O$_2$ that dissociates contributes only 20% of its energy
Figure 4.13: Absorption length spectrum. The data is derived from two spectra at \( x = 0 \text{ m} \) and \( x = 2 \text{ m} \) and has been smoothed by a 50 points FFT low-pass filter.

to translation. The translation energy now becomes

\[
\Delta E_T = (1 - \delta) \cdot \Delta E + 0.2 \delta \cdot \Delta E
\]  
(4.36)

where \( \delta \) is the fraction of \( \text{O}_2 \) that dissociates and \( dE \) the input energy. If we neglect fluorescence, we find for \( \delta \) the following relation:

\[
\delta = \frac{k_s}{k_s + k_q}
\]  
(4.37)

in which \( k_s \) and \( k_q \) are, respectively, the dissociation and quenching rate. For the B(4) state \( k_s = 3 \text{ cm}^{-1} = 9 \cdot 10^{10} \text{ Hz} \). We can assume \( k_q = 1 \cdot 10^7 \text{ Hz} \) so \( \delta \) can be approximated as \( \delta \approx 1 \) (that is, all \( \text{O}_2 \) dissociates). This results in a translation energy:

\[
\Delta E_T = 0.2 \Delta E
\]  
(4.38)

And the corresponding temperature rise can be derived from:

\[
\Delta T = \frac{dE_T}{dm \, c_v} = \frac{0.2dE}{\rho \gamma c_p (dl \times w \times h)} = \frac{0.2}{\rho \gamma c_p A} \frac{dE}{dl}
\]  
(4.39)

Where \( c_p \) is the heat capacity at constant pressure, \( c_V \) is the heat capacity at constant volume, \( dl \times w \times h \) indicates the dimensions of the heated volume. In order to make a reasonable calculation of the local heating, we need to know the laser beam
profile at the tag-probe position. This is obtained by recording the Rayleigh signal in the $x - y$ plane and assuming the same distribution in the $x - z$ plane. The parameters for measurements performed with the APART setup are: $E_0 = 40 \text{ mJ}$, $\rho = 1.2 \text{ kg/m}^3$, $\gamma = 1.4$ and $c_p = 10^3 \text{ J/kgK}$. Using these data, Eq. 4.39 and the spatial energy distribution of the laser as derived from Fig. 4.14, we can calculate the local heating. We find a temperature distribution in the $x - z$ cross-section which is plotted in Fig. 4.15. The maximum temperature increase turns out to be only $\sim 20 \text{ K}$, much lower than expected.

However, when the laser beam is focused, the laser intensity will increase significantly. In Sect. 4.2.1 we have seen that NO creation is a multi-photon process, so that energy absorption will increase non-linearly with intensity. In order to investigate this, the energy absorption over a fixed distance of approximately 10 cm was compared for an unfocused and a focused beam. Within this range, the focused beam exceeded the damage threshold of the powerhead and could not be measured. The mean difference (averaged over the spectral range) between the absorbed energy in a focused and an unfocused beam is $1.7 \text{ mJ/pulse}$. It should be noted that this is a significant amount of energy: it is approximately the energy absorbed from an unfocused beam in 20 cm in air.

We assume that all energy is equally absorbed in the focus of the laser beam according to the beam profile over a certain distance around the focus, too small to be resolved with the powerhead. On the basis of plots of the line-width, as shown in Fig. 4.14, we assumed the absorption region to have a length of the Rayleigh range, approximately 6 mm. When the additional energy increase is calculated on the basis of this volume an additional heat-up of $\Delta T = 500 \text{ K}$ is found. The unknown size of the region makes the determination of the amount of heating very uncertain and
4.5 Line temperature

Figure 4.15: Temperature increase plot at the $x-y$ cross-section of the focused laser beam. Scale is in mm. The maximum temperature increase is approximately $\sim 20$ K.

makes it hard to make a temperature increase estimation with a high degree of accuracy.

4.5.2 Laser-Induced Fluorescence Thermometry

Another technique that has been employed for determining the heating along the line focus is Laser-Induced Fluorescence. In these measurements by Bominaar et al. [58] the nitric oxide molecules were created using an excimer laser, following the APART scheme. Also, like in APART, the NO is made to fluoresce using a Nd:YAG pumped dye laser and the fluorescence signal is recorded using a camera. However, the camera was zoomed in on a small part of the line around the focus ($150 \mu m \times 150 \mu m$) and the dye laser was scanned over a wavelength range of $\lambda = 225.45 \ldots 225.75$ nm. The resulting spectrum was compared to a simulated spectrum. The relative peak intensities in the spectrum depend on the local temperature and when compared to the simulation of the excitation spectrum, calculated using LIFBASE [62], an indication of this temperature can be found. These measurements were repeated for several delays after NO creation and plotted in Fig. 4.16. It was found that at $t = 0.02 \mu s$, the temperature increase is approximately 260 K, after which the temperature decreases nearly mono-exponentially until $t = 3 \mu s$, with a slight jump down to ambient temperature at $t = 5 \mu s$. 
4.6 Conclusion

We have summarized the current knowledge of the photochemical processes that are set into motion during the writing stage of APART scheme. This knowledge is by no means complete. It was observed that an excitation spectrum of nitrogen in air exhibits resonances at the same wavelengths as observed in the excitation spectrum of oxygen. We do have an idea of some of the chemical processes that play a role in the creation of NO, but how these are linked to the photon-induced initiation is not yet fully understood.

Next, we have described a model that explains the effect of heat absorption on the diffusion of the resulting NO lines in non-turbulent air, in which mass and heat diffusion are taken into account. This model explains super-diffusive line widening in time as well as the non-zero diffusion for infinite pressures. In fact, the theory can qualitatively describe both aspects when a local heating of approximately $\Delta T = 300−600\text{ K}$ is assumed. The data of Bominaar et al. [58] are in reasonable agreement with the model for $\Delta T = 300\text{K}$. However, line-widening measurements performed in air with the excimer laser tuned for optimal NO creation showed a diffusion rate that exceeded the model prediction for $\Delta T = 300\text{K}$, even for long delays. The increased line widening rate for longer times may be related to a different ‘write’ wavelength, which causes increased heating of oxygen.

The temperature increase was also checked by means of LIF measurements, which yield an approximate temperature rise shortly after writing of nearly 550 K. We have also considered the absorption of energy along the laser path, but the results

Figure 4.16: Line temperature as a function of time, as determined from LIF spectra. The standard deviation is approximately 100 K.
indicated that a large part of the absorption occurs within a very small region around the line focus, due to non-linear effects. Since the absorption volume could only be roughly estimated, the result suffers from a large error. However, the data do not contradict the model and LIF results.
4 Chemistry and physics of APART
5 Two-point correlation statistics

5.1 Introduction

In Chapter 3 we discussed the principle of our implementation of molecular tagging velocimetry, and demonstrated that it can be used to measure the mean and fluctuating velocities in turbulence. Now, we will address the question whether it can also provide a quantitatively correct measurement of scale-dependent turbulence parameters, such as its energy spectrum. The energy spectrum \( E(k) \) is a second-order property that quantifies the energy \( E(k) \, dk \) residing in the wavenumber interval \([k, k + dk]\). Related to the energy spectrum are the correlation function and the second-order structure function. Other turbulence quantities, such as the energy dissipation \( \epsilon \), the Reynolds number \( R_\lambda \) and the Taylor microscale \( \lambda \) simply derive from the energy spectrum. In this chapter we will consider second-order quantities, whereas higher order quantities will be studied in the next chapter. We will see that the measured spectra differ from the synthesized spectra, and we will discuss the reasons for this. A background contribution to the spectra is believed to be caused by photon noise. The inertial range is affected by different forms of ambiguity in the velocity determination.

In Chapter 4 we already have identified two main sources of ambiguity in molecular tagging velocimetry of turbulent air. The first one is that we are using the Lagrangian displacement of marked molecules for a measurement of the Eulerian velocity field. The second source of ambiguity comes from the diffusion of the molecules, which affects the resolution of the velocity measurement. The two sources of ambiguity are illustrated in Fig. 5.1.

First, we must realize that following a marked fluid element provides a Lagrangian measurement of the velocity field. Let us suppose that the fluid particle starts at \( \vec{x}(0) \), then the averaged Lagrangian velocity is

\[
\overline{v}_{\Delta t}(\vec{x}(0), 0) = \frac{1}{\Delta t} \int_0^{\Delta t} \vec{u}(\vec{x}(t), t) \, dt = \frac{1}{\Delta t} (\vec{x}(\Delta t) - \vec{x}(0)),
\]

(5.1)

where \( \vec{x}(t) \) is the trajectory that starts at \( \vec{x}(0) \) at \( t = 0 \). Clearly (5.1) gives only a correct estimation for the Eulerian velocity \( \vec{u}(x) \) if \( \Delta t \) is very small. Further, in our method we do not take \( \vec{x}(\Delta t) \) but the point \( \vec{x}'(\Delta t) \) of Fig. 5.1, since the true
location of $\vec{x}(\Delta t)$ is also dependent on an unknown velocity component in the $y$-direction. Then, on the line the point $\vec{x}'$ cannot be identified. In our experiments we use a single 2D projection of a line, which may display apparent self-intersections for time delays between writing and reading that are several Kolmogorov times long. However, we argue in Chapter 4 that in air, these self-intersections are always blurred by molecular diffusion. Finally, the most important effect of molecular diffusion is the filtering of the velocity field. In fact, there are a few subtleties related to this filtering, which have been dealt with in Chapter 2.

Of course, these ambiguities become less important if the delay $\Delta t$ between writing and reading is made smaller. However, the line displacement then decreases accordingly, so that the finite pixel size and other sources of error become significant and influence the measured velocity. Additionally, minimizing $\Delta t$ will require a high sensitivity of the camera, since NO is gradually created within the first $2\mu s$.

The present chapter is organised as follows. First, we will relate the geometrical arrangement of our experiment to the turbulence properties that are measured. Then we will briefly describe the various energy spectra that can be measured. In Section 5.2.2 we compare measured energy spectra and their theoretical counterparts at the set Reynolds number. It will turn out that especially the measured energy spectra are very different from the theoretical spectra.

Since the precise shape of the measured lines is also determined by the noise on the image, together with the variation of the pixel sensitivity across the image, we study the influence of these two effects on the appearance of measured spectra in Section 5.3. Because individual molecular tracers cannot be distinguished in a line, this will lead to some ambiguity in determining the actual motion of such particles. This point will be discussed in more detail in paragraph 5.4.3.

For a better understanding of MTV, we will turn to simulations in Section 5.4.3. In this approach we simulate a turbulent velocity field with known second-order properties, write a line in this field and follow its deformation in time. In a further
refinement, we will model diffusion by sprinkling material particles on a line which undergoes Brownian motion. This allows us to isolate the ambiguities of MTV that are illustrated in Fig. 5.1.

In these so-called kinematic simulations, the velocity field is not a true velocity field that satisfies the Navier-Stokes equation. Instead, it is a random model field that is isotropic and incompressible and that has a prescribed spectrum. It is thus only suited to study second order properties, and lacks true turbulence phenomena such as intermittency. These effects will be studied in the next chapter.

### 5.1.1 Longitudinal and transverse two-point correlations

In APART velocimetry (and in most other instances of molecular tagging velocimetry) the visualization of tagged molecules is done by pulsed lasers. The pulse repetition frequency of these lasers is in general much smaller than the largest frequency in the flow. Therefore, subsequent images of the displaced tagged molecules are uncorrelated snapshots of the velocity field. When writing simple lines, their deformation gives access to transverse correlations only. Other correlations would become accessible when writing more complicated patterns. Briefly, the deformation of a line gives information on the component of the velocity perpendicular to the line. In order to see the correlations of these velocities in this direction (the longitudinal correlations) we would need at least two closely spaced lines.

In most of turbulence research, the relations between velocities at two different points in space (or in time) are of interest. The most common way to look at 2-point velocity data is by means of correlations between velocity components \( \langle \vec{u}'(\vec{x}') \vec{u}(\vec{x}) \rangle \), or by related properties, such as the energy spectrum. In homogeneous turbulence, the 2-point statistical quantities depend only on the difference vector \( \vec{r} = \vec{x} - \vec{x}' \), but the directions of the velocity components \( \vec{u} \) and \( \vec{u}' \) can still be arbitrary. In isotropic turbulence, however there are only two distinct possibilities, see Fig. 5.2. In the longitudinal one, the vectors \( \vec{r}, \vec{u} \) and \( \vec{u}' \) all point in the same direction whilst in the transverse setup the vectors \( \vec{u} \) and \( \vec{u}' \) are perpendicular to the separation vector \( \vec{r} \). In the currently used MTV technique, we have only access to the transverse statistics.

![Figure 5.2: Transverse (left) and longitudinal (right) velocity increments. Longitudinal measurements can be performed from a single point by usage of Taylor’s hypothesis.](image)
5 Two-point correlation statistics

In traditional turbulence velocimetry, the velocity is measured in a point as a function of time. In order to measure space-dependent spatial structures, use is made of the Taylor’s hypothesis of frozen turbulence. In this hypothesis it is assumed that the advection of turbulence past a fixed point may be taken to be entirely caused by the mean flow. Spatial structures can thus be derived by using the substitution

\[ \frac{\partial}{\partial x} = \frac{1}{U} \frac{\partial}{\partial t} \] (5.2)

This approximation holds for small velocity fluctuations on the mean flow, \( u_{rms}/U < 0.1 \). In the case of the strong turbulence in our setup this ratio becomes \( u_{rms}/U = 0.24 \), and the frozen turbulence approximation will be poor. Furthermore, fluctuation of the convecting velocity is only one of several phenomena that cause distortion of statistics that result from use of Taylor’s frozen turbulence approximation. These distorting phenomena have been discussed by Fisher et al. [3], Lumley [4] and others.

In point measurements, the vector \( \vec{r} \) is, on average, in the mean flow direction. Transverse velocity increments can therefore only be measured if the velocity component perpendicular to the mean flow can be measured.

5.2 Spectra

5.2.1 Theory

Let us consider the autocorrelation function \( R_{ij} \) of the velocity in statistically stationary and homogeneous turbulence.

\[ R_{ij} (\vec{r}) = \langle u_i (\vec{x} + \vec{r}) u_j (\vec{x}) \rangle \] (5.3)

Stationarity implies that the result of the average does not depend on time \( t \), whereas homogeneity implies that it only depends on \( \vec{r} \) but not on \( \vec{x} \). At the origin it is \( R_{ij} (0) = \langle u_i u_j \rangle = u^2 \delta_{ij} \) where \( u \) is the rms velocity. As was argued in the introduction, in isotropic turbulence there are two distinct arrangements of \( \vec{u} \) and \( \vec{r} \). It can be shown [49] that \( R_{ij} \) can thus be expressed in terms of two scalar functions as

\[ R_{ij} (\vec{r}) = u^2 \left( g (\vec{r}) \delta_{ij} + [f (\vec{r}) - g (\vec{r})] \frac{\vec{r}_i \vec{r}_j}{r^2} \right). \] (5.4)

With \( \vec{r} = \vec{e}_i r \), these longitudinal and transversal autocorrelation functions \( f(r) \) and \( g(r) \) become, respectively,

\[ f (r) = \langle u_1 (\vec{x} + \vec{e}_1 r) u_1 (\vec{x}) \rangle / \langle u_1^2 \rangle, \] (5.5)

\[ g (r) = \langle u_2 (\vec{x} + \vec{e}_1 r) u_2 (\vec{x}) \rangle / \langle u_2^2 \rangle. \]

as sketched in Fig. 5.3
5.2 Spectra

Figure 5.3: A sketch of the velocity components in the longitudinal and transverse structure functions for separation $\vec{r} = \vec{e}_1 r$.

Incompressibility of the velocity field implies $\frac{\partial R_{ij}}{\partial r_j} = 0$ and together with Eq. 5.4 this can be used to find a relation between $f$ and $g$ [49],

$$g(r) = f(r) + \frac{r}{2} \frac{d}{dr} f(r).$$  \hfill (5.6)

Thus, in incompressible, isotropic turbulence the two-point correlation tensor is completely determined by the longitudinal autocorrelation function $f$. The one-dimensional energy spectra are defined as the Fourier transform of the correlation function $R_{ij} (\vec{r}, t)$:

$$E_{ij} (k_1) = \frac{1}{\pi} \int_{-\infty}^{\infty} R_{ij} (r_1, t) e^{-ik_1 r_1} dr_1,$$  \hfill (5.7)

Here, $k_1 = \vec{e}_1 \vec{k}$. Since $R_{ij}$ is real and even for the diagonal elements, $E_{ij}$ will also be real and even. A measure for the large scale eddy size is the longitudinal integral length scale, calculated as the integral over the correlation function $f$:

$$L = \int_0^\infty f(r) dr.$$  \hfill (5.8)

The energy spectrum is defined according to Kolmogorov’s 1941 theory (K41). According to Kolmogorov’s first similarity hypothesis [46], for sufficiently high Reynolds numbers, the statistics of the small-scale motions are uniquely defined by $\nu$ and $\varepsilon$. This implies that the energy spectrum $E(k)$ is a universal function of $\nu$, $\varepsilon$ and $k$. From simple dimensional analysis we find that this function can be written as

$$E(k) = C (\varepsilon \nu^5)^{1/4} \phi(k \eta),$$  \hfill (5.9)

where $k \eta$ is the non-dimensionalised wavenumber, with $\eta$ the Kolmogorov length scale, and $\phi$ a universal (dimensionless) function. According to the second similarity hypothesis [46] the spectrum is uniquely dependent on $\varepsilon$ and $k$ (and thus self-similar) in the inertial subrange. From this, again using only dimensional arguments, we can find the ubiquitous Kolmogorov $-5/3$ spectrum

$$E(k) = C_k \varepsilon^{2/3} k^{-5/3},$$  \hfill (5.10)
relating Eq. 5.9 with Eq. 5.10 using the function \( \phi(k\eta) = (k\eta)^{-5/3} \). As was already explained in Chapter 3, a convenient parameterization of the energy spectrum \( E(k) \) that encompasses large, inertial-range, and dissipative scales is used. This spectrum is parametrized by the energy dissipation and thereby the Reynolds number, and was shown to agree well with the spectra that were measured at low Reynolds numbers in the turbulent jet. For the Kolmogorov constant \( C \) we take the most recent \( C = 1.619 \) [52].

In Ch. 3 it was already shown how to derive the longitudinal spectrum \( E_{11}(k) \) from \( E(k) \). In our tagging velocimetry experiments we need the transverse \( E_{22}(k) \), which has a similar relation to the 3D spectrum [49]:

\[
E_{22}(k_1) = \frac{1}{2} \int_{k_1}^{\infty} \frac{E(k)}{k} \left( 1 - \frac{k_1^2}{k^2} \right) dk.
\] (5.11)

The spectrum describes the behavior in Fourier space, whereas we are often interested in real space. Consider the second-order structure function,

\[
G_{ij} = \left\langle (u_i(\vec{x} + \vec{r}) - u_j(\vec{x}))^2 \right\rangle,
\] (5.12)

which is directly related to the spectrum \( E_{ij} \) through a Fourier transformation. Specifically, the transverse second order structure function and the (one-dimensional) transverse spectrum are related as [49]:

\[
G_{22}(r_1) = \int_0^\infty E_{22}(k_1) (1 - \cos(k_1 r_1)) dk_1.
\] (5.13)

Also, the second order structure function has a scaling behaviour [49]

\[
G_{22}(r) = \frac{4}{3} G_{11}(r) \sim \frac{4}{3} C_2 (\varepsilon r)^{2/3},
\] (5.14)

with \( C_2 \approx 2 \). For large \( r \), the two velocities \( u_i \) and \( u_j \) become uncorrelated and \( G_{22} \to 2u^2 \).

While \( E(k) \) gives access to scaling in the wavenumber domain, \( G_{ij}(r) \) works in the spatial domain. Although \( E(k) \) and \( G_2(r) \) (the second order structure function) are (trivially) each other’s Fourier companions they can give different information if the largest scales are truncated, for example because of a restricted field of view of the camera.

5.2.2 Experimental results

In Chapter 3 hotwire measurements have been described to characterize the turbulent flow at low Reynolds numbers. However, the used hotwire setup resolution is bounded by the maximum sampling rate of 100 kHz. This means that in our small scale setup only low Reynolds numbers are accessible \( (R_\lambda = 160) \). This is why the
small scale features of the flow have been characterized so that by extrapolation the flow properties can be predicted for higher Reynolds numbers. In Chapter 3 only the mean velocity $U$ and rms velocity $u$ of the APART measurements where checked against extrapolated values. In this chapter we will have a look at spectral data measured with APART MTV. In order to do this we will use synthetic (Pao) spectra as described in Eq. 3.20-3.23 characterized by $L$ and the parameters $U$ and $\epsilon$. These parameters $U$ and $\epsilon$ are known through the characterisation of the jet. The measured spectra determined from APART, can now be readily compared with the synthetic spectrum by collapsing the spectra onto each other. This scaling is done using the small-scale length $\eta$ and velocity scale $v_\eta$. Since the APART line data consists of truly spatial measurements this spectrum is normalised by

$$k^* = k\eta \quad (5.15)$$

and the reduced energy $E^*$:

$$E^*(k^*) = E(k) \frac{1}{\epsilon^{\frac{5}{3}}\eta^{\frac{11}{3}}} \quad (5.16)$$

Molecular tagging spectra were obtained by taking averages over $5 \cdot 10^3$ written lines, whose centers were determined with help of the Gaussian fits described in
Figure 5.5: Spectra of lines in still air at $\Delta t = 30 \mu s$ (lower) and $\Delta t = 10 \mu s$ (upper). Note that the displacements in the line due to noise have been rescaled with the respective delay, leading to a decrease of the background spectrum energy for increasing delays.

Chapter 3. Using the measured $u$ and the scaling constant $C_\epsilon$, whose measurement was described in Ch. 3, the MTV spectrum could be normalised.

In Fig. 5.4 this MTV spectrum is compared to the corresponding synthetic spectrum. The two spectra are different: first, at the large wave numbers the measured spectrum has a huge bump. We believe this bump is due to photon noise in the images. This is demonstrated in Fig. 5.5, where the spectra of sets of 800 lines in still air at different delays are plotted. The spectra show exactly the same bump as observed in the spectra of turbulence, and is shown to be part of a broad background spectrum. This rules out that the bump is caused by turbulence. Note that in Fig. 5.5 the line displacements have been rescaled with the delay as is done for velocity spectra, leading to a decrease of the background spectrum energy for increasing delays. When the displacement spectra rather than the velocity spectra for the 2 different delays ($\Delta t = 30 \mu s$ and $\Delta t = 10 \mu s$) are compared, it was seen that they exactly overlap, indicating that this spectrum is not time dependent, thereby ruling out diffusion. Besides, without interaction with turbulence, the center of mass of the line will not move due to diffusion. Therefore, by diffusion alone, no such spectrum could be created. The remaining option is that this background is inherent to the way in which lines lines are imaged and converted to velocity data.
It is believed that photon noise influences the pixel values, which is reflected in the fits. Although the energy contained in this 'fit-noise' is very little, it is surprising that it covers all length scales.

At the low wavenumbers in Fig. 5.4 the effect of the finite extent of the images is felt. Compared to the synthetic spectrum, the integral length scale is still an order of magnitude away. These two effects make that the experimental spectrum does not display a \( k^{-5/3} \) behaviour anywhere in the spectrum. In Section 5.4 we will try to understand the MTV spectra through numerical simulations.

The measured second order structure function shown in Fig. 5.6 is in much better shape. Although it does not increase rapidly enough at the smallest scales (in agreement with the large \( k \) behaviour of the spectrum), it shows a correct scaling behaviour over one decade of large scales. The scaling exponent \( G_2(r > 50\eta) \sim r^{\zeta_2} \), \( \zeta_2 \approx 0.77 \) is significantly larger than expected from Eq. 5.14, \( \zeta_2 = \frac{2}{3} \), a fact we will comment on in the next chapter. In Fig. 5.6 it is also seen that at the largest separations, the measured structure function has not yet reached the asymptotic value, \( 2u^2/v_\eta^2 \) (the horizontal line in Fig. 5.6). Clearly, the field of view in our images is too small.

Because of the limited spatial extent of our measurements, Fourier transforms will always suffer from edge effects. This is illustrated in Fig. 5.7 where we compare

---

Figure 5.6: Second order structure function derived from APART velocimetry data. The structure function is normalised so that it ideally crosses (1,1) at the Kolmogorov scale. The dotted line indicates the fit of the scaling exponent for large scales.
the directly measured $G_2(r)$ with the one that follows from Fourier transforming the spectrum through Eq. 5.4. While $G_2$ has a clear scaling behaviour, no $\sim k^{5/3}$ scaling can be observed in the Fourier transform of $E(k)$.

In the following sections, we will first study the influence of camera imperfections on the shape of the measured spectrum. Then, in Section 5.4, we will turn to numerical simulations in order to study the effect of the finite delay time between reading and writing, and the effect of molecular diffusion on measured spectra.

### 5.3 Image pre-processing

In view of the apparently deviating form of the recorded APART spectra, we will investigate to which extent the shape of the spectrum is the result of artifacts in the unprocessed images. The centroid of written lines is determined by fitting a Gaussian profile to (perpendicular) line cross-sections. These fits depend on variations in sensitivity, offset and noise level of individual pixels. For example, in a least squares procedure the weight of individual pixels is inversely proportional to their (squared) noise level, and also the sensitivity of the pixels may vary across the camera chip. Since the method employed for velocity extraction aims to resolve the line centre
5.3 Image pre-processing

with high (sub-pixel) resolution and the line width is usually a mere decade of pixels wide, the sensitivity to noise is fairly large.

In this section we will map out these variations, remove the image offset and include this map in our least squares procedure and study the effect on measured turbulence. As the error introduced after digitisation in the all-digital stages of the image recording (frame grabber and data storage) is negligible, we will only consider errors produced in the intensified camera [63].

Dark current arises from thermal energy within the silicon lattice of the CCD chip. The amount of electrons that are created over time by dark current is not dependent on the amount of incident light. Although this effect is reduced through deep cooling, a contribution remains. The accumulated dark current noise will have a Poisson distribution.

A related effect is smearing during camera readout. During readout, the image is digitized line by line through shift registers located at one edge of the CCD chip. While the pixels are waiting to be digitized, thermal noise is accumulated. This effect is dependent on the readout time, making the last portion of the CCD to be read appear brighter than the first. Because the readout time is fixed and not dependent on the shutter time, this effect adds to the position-dependent but time-independent offset.

If \( F \) is the photon flux, \( q \) the quantum efficiency, \( t \) the integration time, \( F_d \) the dark current and \( S_o \) the pixel offset, then the ensemble-averaged signal of an individual pixel is

\[
S = A [F q t + F_d t] + S_o,
\]

(5.17)

where \( A \) is the constant that relates the (binary) signal value to the number of stored photo-events. The readout noise for a Poisson process where uncorrelated photo events are accumulated is then given by

\[
\sigma_s^2 = A^2 [F q t + F_d t] + \sigma_d^2
\]

(5.18)

where \( \sigma_d \) is the noise contribution due to the digitisation of the accumulated photo-electron charge.

As a first step, the systematic error \( S_o \) is removed per pixel. This is trivially done by recording an image for a short time with closed shutter. This offset frame is now subtracted from each image resulting in a reduced signal \( \tilde{S} = A [F q t + F_d t] \).

The noise per pixel in the image signal can be expressed in the pixel value (intensity), thereby removing the explicit time dependence.

\[
\sigma_s = \sqrt{A \cdot \tilde{S} + \sigma_d^2}.
\]

(5.19)

Although optimally for each pixel a full noise model should be defined, we will first assume that a global noise model will suffice. To obtain this noise model a filter with a set of (approximately linear) transmission gradients was designed as shown in Fig. 5.8.
The filter was placed in the focal plane of the camera and was illuminated using ambient light. The filter was imaged on the CCD, choosing a shutter time such that the highest pixel value present is near the maximum value of 4096. Due to the gradient filter, pixel values ranging from 0 to 4096 were present. For each pixel position the mean value and standard deviation were calculated over a set of 1000 images.

In Fig. 5.9 the squared standard deviation $\sigma_s^2$ of the pixel value is plotted against the mean value $\tilde{S}$, where the squared standard deviation is smoothed in a 1 count range. This procedure is repeated for different gain settings of the image intensifier. The plots with increasing slopes indicate increasing intensifier gain settings. All curves exhibit a linear dependence of $\sigma_s^2$ on $\tilde{S}$. Furthermore, we find $\sigma(S = 0) = 0$, indicating that in all intensifier settings the error $\sigma_d$ is negligible. This means that for all amplification settings the signal-to-noise ratio can be accurately expressed by

$$
\sigma_S = \sqrt{A \cdot \tilde{S}},
$$

where $A$ is a constant dependent on the image intensifier gain. We do observe a slight deviation of this trend for large values of $\tilde{S}$, but this is due to saturation of the camera. For increasing gains (and thus intensity distributions with higher values of $\sigma$) the saturation will occur for a larger part of the velocity distribution. The procedure that has been described above results in a global noise-model. However, in reality, each pixel has its own error model and sensitivity. Rather than constructing a complete model for each pixel, we will only determine a per-pixel quantum efficiency $q$. This is achieved by uniformly illuminating the CCD chip at different lighting settings and calculating the overall mean image intensity. For each pixel a linear correlation is calculated between the pixel intensity and the “true” intensity:

$$
S(x, y) = S_o(x, y) + q(x, y) \cdot I_{\text{mean}}.
$$

Here $S(x, y)$ is the value of the pixel at position $(x, y)$, $I_{\text{mean}}$ the mean intensity and $S_o(x, y)$ is the pixel offset. In this way we can allow for the differing sensitivity of
5.3 Image pre-processing

Figure 5.9: Noise model for different intensifier gains. Plotted are the square of the standard deviation versus intensity for all pixels. To reduce errors, for pixels with equal averaged intensity an averaged standard deviation is used. From bottom to top: signal intensifier gain $g = 0, 2.5, 5, 7.5$ and $10.$

Individual pixels. We should note that, even though an image offset was already subtracted, we still find non-zero offsets for the illuminated pixels.

In Fig. 5.10 the different steps are shown for a sample image. In Fig. 5.10a the unprocessed line is shown. First, the offset image is subtracted (Fig. 5.10b), which slightly reduces the background as can be seen in the inset. Secondly, the per-pixel calibration is applied. Although the algorithm is mainly intended to amplify pixels with low sensitivity, it also corrects for the global intensity gradient that was present in the image. After this correction the whole image is of comparable intensity. The inset shows that the noise has increased for some pixel positions. However, it should be noted that the (in) sensitivity of pixels is now taken into account during fitting.

In Fig. 5.11 the result of the image pre-processing on the energy spectrum is shown for the unprocessed data and for the data processed with the corrections described above.
Figure 5.10: Recorded images in different stages of processing before fitting. The insets show a magnification of a part of the image, where the intensity is increased, so that noise is better visible. a) Unprocessed image, b) image after subtracted offset, c) previous image with added pixel sensitivity calibration.
5.3 Image pre-processing

It can be seen that, although effects are visible, they do not change the overall trends in the spectrum. The background spectrum, as shown in Fig. 5.5, is reduced somewhat, except for the very smallest scales where the amount of energy present has become slightly higher. The photon noise remains present in all images and since we need to work with single shot images, this effect cannot be averaged out.

The image processing does not have any effect on the slope in the integral range and in this region the distribution remains far from the $k^{-5/3}$ Kolmogorov spectrum. It enforces the notion that at the integral range, the measured spectrum does not differ from the Kolmogorov spectrum due to equipment imperfections or due to photon noise, but that other effects play a role. We will show that diffusion is the main reason of the deviating spectrum.

Figure 5.11: 2D normalised energy spectra after multiple forms of image pre-processing. Curve 1 shows the spectrum without image processing, curve 2 shows the effect of offset subtraction, pixel calibration and a pixel-dependent noise model.
5.4 Kinematic simulations

5.4.1 Introduction

When the deformation of a line is measured between two instants in time, one is essentially observing Lagrangian flow properties. From these properties we will extract Eulerian quantities such as spectra and structure functions. Clearly, for short times the instantaneous velocity field can be inferred from the displacements of a material point or line. However, this becomes more and more problematic if the length of the time interval increases. On the other hand, displacements can be measured more accurately if the time interval is larger. The question that needs to be answered, is how the error in the measured velocity field grows with delay time. Since the smallest eddies turn over on the shortest time scales, we expect that the error affects the small scales first. Therefore, in measured energy spectra the large wavenumbers will be affected first by this problem. In order to quantify the effect we will use kinematic simulation software generously provided by Vassilicos and the department of Aeronautics at the Imperial College London.

In kinematic simulations the turbulent velocity field is simulated by a large number of random Fourier modes [64, 65, 66]. The orientations of these waves are drawn from a random distribution, but with amplitudes such that the overall energy spectrum takes on a prescribed form. Their direction in space and their phase are uniformly random with the only requirement that the resulting velocity field is incompressible. Kinematic simulation is computationally cheap, because it does not solve the Navier-Stokes equation. By including a large dynamical range of wavenumbers, arbitrarily large Reynolds numbers can be reached. In contrast, for direct numerical simulations the cost would rise with Reynolds number as $Re_\lambda^6$ [49]. Although one may object that a kinematic field is not real turbulence, it has all proper second-order statistics. We will therefore use it to study properties of molecular tagging velocimetry.

This section will be organised as follows: first we will introduce the technique of kinematic simulation. We will perform two different forms of kinematic simulations. In the first we will address the question what error we make when using the finite-time Lagrangian displacement to find the Eulerian velocity field. To this aim we will study the displacement of material lines in the simulated velocity field. Next, we will study the dynamics of “sausages” (thick lines) of molecules that constitute the lines of our experiments. These molecules are advected by the velocity field, but at the same time undergo thermal Brownian random displacements: the sausages diffuse. In this way we can simulate the images that are obtained in the experiment. In these more costly simulations we can study the combined effect of finite displacements and diffusion.
5.4.2 Velocity field

The simulated velocity field is incompressible, statistically stationary, homogeneous and isotropic. It features many of the important statistical properties of a real turbulent velocity field. In particular, it has the same energy spectrum and the same correlation length. The velocity field is realized by a sum of Fourier components. To construct it we generate randomly chosen vector components and use these to synthesize the field.

$$\vec{u}(\vec{x}, t) = \sum_{n=1}^{N} \left[ \vec{v}_n \cos(\vec{k}_n \cdot \vec{x} + \omega_n t) + \vec{w}_n \sin(\vec{k}_n \cdot \vec{x} + \omega_n t) \right].$$ (5.22)

The wavenumbers $\vec{k}_n$ and the Fourier amplitudes $\vec{v}_n = \vec{v}(\vec{k}_n)$ and $\vec{w}_n = \vec{w}(\vec{k}_n)$ will be picked to represent a realistic turbulent flow. An unsteadiness frequency $\omega_n$ has been added to introduce time dependence in our model. It is possible to devise a time-dependent kinematic model [64] which has realistic sweeping effects in which the small scales are swept by the velocity field at large scales. However, such a calculation needs much more computational resources. We have chosen to use a more simple model [64, 67] in which the unsteadiness frequency $\omega_n$ of mode $n$ is proportional to the eddy turnover time associated with wavevector $\vec{k}_n$ as

$$\omega_n = \lambda \sqrt{k_n^3 E(k_n)},$$ (5.23)

with an adjustable weighting factor $\lambda$.

To obtain incompressibility, the velocity field has to comply with $\nabla \cdot \vec{u} = 0$. This can be ensured by taking the vector Fourier components perpendicular to $\vec{k}_n$ by $\vec{v}_n = a_n \times \vec{k}_n$ and $\vec{w}_n = b_n \times \vec{k}_n$ with unit vectors $\vec{k}_n$ defined as $\vec{k}_n/k_n$.

To obtain a physically realistic flow field the vectors $\vec{k}_n$, $\vec{v}_n$, and $\vec{w}_n$ will be taken such that the simulation is in accord with the Kolmogorov (1941) spectrum $E(k)$ in the inertial range, Eq. 5.10:

$$E(k) = C\varepsilon^{2/3}k^{-5/3},$$ (5.24)

where the Kolmogorov constant $C$ is taken to be 1.5 [64]. The energy is set to zero for $k > k_\eta$ and $k < k_c$. The small scale cutoff $k_\eta = 2\pi/\eta$ is determined by the Kolmogorov scale. Together with the integral scale $k_c = 2\pi/L$ this determines the Reynolds number. In principle, once these choices are made, the dissipation rate and through it the time constant follows. However, since the spectrum $E(k)$ is truncated, the dissipation rate in Eq. 5.24 is not the one that follows from the Kolmogorov length $\eta$ (or the Kolmogorov wavenumber $k_\eta$). The value of $\epsilon$ determines the time scale of the generated velocity field. Several choices are possible to determine a value for $\epsilon$. From the rms velocity $u$ and the relation for the 1D spectrum,

$$\frac{3u^2}{2} = \int_{k_c}^{k_\eta} E(k) \, dk,$$ (5.25)

1

87
5 Two-point correlation statistics

it follows with Eq. 5.24 that

$$\varepsilon = \left(3u^2/ \left[2C \int_{k_c}^{k_\eta} k^{-5/3} dk \right] \right)^{3/2}. \quad (5.26)$$

The integral length scale can, rather than from the velocity correlation function, also be calculated from the spectrum as [49]:

$$L = \frac{3\pi}{4} \int_{k_c}^{k_\eta} k^{-1} E(k) dk. \quad (5.27)$$

Together with the Kolmogorov scale from Eq. 2.12 this particular choice of \( \varepsilon \) sets the Kolmogorov time

$$\tau_\eta = (2\pi)^{2/3} \varepsilon^{-1/3} k_\eta^{-2/3}, \quad (5.28)$$

the effective dissipation constant,

$$C_\varepsilon = \frac{L \varepsilon}{u^3}, \quad (5.29)$$

and the effective Taylor microscale Reynolds number

$$Re_\lambda = \sqrt{15} C_\varepsilon^{-2/3} \left(\frac{L}{\eta}\right)^{2/3}. \quad (5.30)$$

The equations (5.26) through (5.30) represent one particular choice. Another choice could start from the experimental value of \( C_\varepsilon \) from which \( \varepsilon \) would follow. The possibility of choices reflects the ambiguity of kinematic simulations because these simulations do not deal explicitly with dynamics. In the simulation, the values of the discrete set of wavenumbers \( k_n \) are chosen by discretizing wavenumber space into a finite number of modes, geometrically distributed over the range of \( k_c \) to \( k_\eta \).

$$k_n = k_c \left(\frac{k_\eta}{k_c}\right)^{n-1/n_k} \quad n = 1 \ldots N_k. \quad (5.31)$$

The \( N_k \) vectors \( \vec{k}_n \) are chosen from a spherical shell of radius \( k_n \), with a direction chosen randomly on the sphere. Now \( \vec{a}_n \) and \( \vec{b}_n \) have to be determined in such a way that each mode represents a part of the energy spectrum. To this aim, their directions are picked randomly and their lengths are taken from the energy shells \( E(k_n) \) as

$$\frac{3}{2} a_n^2 = \frac{3}{2} b_n^2 = E(k_n) \Delta k_n \quad (5.32)$$

with

$$\Delta k_n = \frac{k_{n-1} - k_{n+1}}{2}. \quad (5.32)$$
5.4 Kinematic simulations

In the simulated velocity field $\vec{u}(\vec{x}, t)$ we track material particles by integrating

$$\frac{d\vec{x}}{dt} = \vec{u}(\vec{x}, t)$$

(5.33)

using a 4\textsuperscript{th} order Runge-Kutta scheme. Clearly, the time step $\tau$ in the numerical integration should be small enough to resolve the fastest changing velocity on the smallest length scales, the Kolmogorov velocity $v_\eta$ and the Kolmogorov length scale $\eta$. According to [64] a time step of $1/10$ of the Kolmogorov time $\tau_\eta$ is small enough to safely resolve all flow features.

5.4.3 Line simulation

The numerical experiment is set up with an inertial range that is bounded by the (normalised) large scale $L = 1$ and small scale $\eta = 10^{-3}$, a range of scales that approximately corresponds to our jet. This inertial range is set up by 100 independent Fourier modes and the rms velocity is set to unity. In each realisation a string consisting of 1000 particles is placed into the synthetic flow field and tracked for a set time. For each of the time delays 1000 realisations are generated, where each realisation has a different set of randomly picked Fourier components.

As we use just one camera in our experiments, we cannot obtain a complete representation of the line and we are for the moment limited to its 2D projection. This means that velocimetry is done from 2 dimensional projections of a line, and it is this situation that must be simulated here. As in the experiments, the line is created in the $x$-direction and the displacement in the $y$-direction is observed.

There are several problems with this kind of velocimetry, which all disappear if the time interval $\Delta t$ is made small enough.

For longer times ($> \tau_\eta$), elongated and deformed lines can curl in such a way that the 2D projections of the line will display self-intersections. Such apparent self-intersections will become more numerous for delay times that are much longer than the Kolmogorov time. Clearly, the assignment of the displaced point $\vec{x}$ will be ambiguous and we have to think of how to deal with these situations. A possible strategy is sketched in Fig. 5.12. First, the displaced line is resampled in the regularly distributed $x$-coordinates of the material points that made the original written line. Second, in the case of (apparent) self intersections, we take the vertical displacement to be the average $(y_1 + y_2 + y_3)/3$ of the points 1, 2 and 3. This form of averaging mimics the averaging done in the experiment, which is the consequence of fitting a Gaussian to an intensity profile. Here, an important remark must be made: the occurrence of self intersections on short time scales will only occur in simulations of ideal material lines. In reality the written lines in air diffuse as fast in real space as in momentum space (through viscosity), therefore intersections at small scales will never be seen. This is discussed in Chapter 4.

Since the smallest eddies turn over quickest, it is expected that the problems sketched above affect the large wavenumbers first. This is indeed the case, as is
Two-point correlation statistics

Figure 5.12: Schematic picture of a self-intersection. To remove self-intersections in the simulations, we take the vertical displacement to be the average \((y_1 + y_2 + y_3)/3\). This form of averaging mimics the averaging done in the experiment. The solid line shows the actual line, the dotted line shows the line after averaging points with equal \(x\) position.

shown in Fig. 5.13. In this figure we present spectra that are calculated from line displacement compared with a spectrum that is directly derived from the velocity field. The dotted line corresponds to the average spectrum calculated from 1000 realisations of the directly obtained velocity field \(u(y)\) (equivalent to the velocity calculated from \(\bar{x}_{Dt} - \bar{x}_0\) when \(\Delta t \to 0\)). The spectrum shows an inertial range from \(k^* = 2 \cdot 10^{-2}\) to \(k^* = 1\). Even though the spectrum is defined directly in the simulations (by choosing the correct values of \(\bar{v}(\vec{k}_n)\) and \(\bar{w}(\vec{k}_n)\) for \(\vec{k}_n\)) the retrieved scaling exponent in the inertial range of \(-1.75\) differs from \(-5/3\). This is due to the limited number of Fourier modes that has been calculated in order to “fill” the spectrum. The solid lines correspond to average spectra calculated from 1000 realisations of the displacement of the line during increasing intervals \(\Delta t/\eta\). The inertial range scaling exponent in the spectra displays a strong dependency on the tag-probe interval and it is clear that the inertial range is not reflected well for the larger wavenumbers. For \(k^* > 0.05\) the scaling exponent is underestimated. This effect is already visible for time delays that are a small fraction of the Kolmogorov time \(\Delta t = 0.1\tau_\eta\) and becomes more prominent for longer times.

With increasing delay time, the simulated spectra are cut off at decreasing values of \(k^*\). In order to quantify this effect we plot in Fig. 5.14 the compensated spectrum \(k^{1.75}E(k)\), and define two cut-off wavenumbers by setting two arbitrary thresholds as indicated in Fig. 5.14a. The corresponding cut-off wavenumbers \(k_{\text{co}}\) are plotted in 5.14b. These wave numbers define a length scale \(k_{\text{co}}^{-1}\), which is seen to increase nearly linearly with delay time, a property that is independent of the cutoff threshold. The erosion of the spectrum is due to relative motion of points along the line, which can even give rise to apparent self-intersections in the 2D projection of the tagged line. At small times, the relative distance travelled is linear in the elapsed time. This explains the linear dependence of \(k_{\text{co}}^{-1}\) on the delay time.

In order to better approximate the APART measurements the simulation was extended to allow for diffusion. This was done by including a random displacement

\[
\Delta x_{\text{brownian}} = \zeta \sqrt{2D\tau}
\]  

(5.34)
Figure 5.13: Spectra of a synthetic velocity field. The dotted spectrum is directly derived from the simulated turbulent velocity field. The solid line spectra are calculated from the displacement of the line during intervals $\Delta t/t_\eta = 0.1, 0.2, 0.3, 1.0$ and $1.5$. 
Figure 5.14: The upper figure shows the dependency of the measured inertial range on the tag-probe interval (ranging from 0.1 to 1.5) in compensated spectra of the velocity field. The wavenumber at which the spectral energy falls below the threshold is considered the cut-off wavenumber. The dashed lines denote the two cut-off thresholds of 0.35 and 0.40. The lower figure shows the cut-off wavenumbers for delays from 0.1 to 1.5. The data is fitted with $k^{-1}_{co} = a\Delta t^b + c$ where $b$ is found to be $0.84 \pm 0.05$ and $0.87 \pm 0.05$, respectively.
5.4 Kinematic simulations

after advecting the particle during a time step $\tau$. The random vectors $\zeta$ were drawn from a uniform distribution $\zeta \in [-1, 1]$ [65]. In the numerical simulations, physical parameters such as the kinematic viscosity are not explicitly given, but enter through the choice of smallest and largest wavenumbers. As explained above, this choice implies a value for $\varepsilon$. This value was used to determine the kinematic viscosity $\nu = (\varepsilon \eta^4)^{1/3}$ from which the mass diffusion coefficient follows from

$$D = \frac{\nu}{Sc},$$

(5.35)

where the Schmidt number $Sc$ was taken unity.

The initial line is constructed by sprinkling $1 \cdot 10^6$ particles, mimicking the tracer particle distribution in the experiments albeit, obviously, in much smaller concentrations. This distribution is approximated by a constant density along the line and a radial Gaussian density distribution with constant width perpendicular to this line. The motion of the particles in the synthetic flow is calculated for a specific time delay and an image is created of the 2D projection of the particle density. This is done for 1000 lines and the resulting line images are fitted in the same manner as is done with the experimental images. The spectra calculated from the processed data are shown Fig. 5.15. The upper curve shows a thin-line spectrum for delay $\Delta t = \tau_\eta$ and the lower curve displays the finite volume-line spectra for the same delay.

The simulated spectrum of the diffusing line bears a striking similarity to the actual measured spectrum, which was shown in Fig. 5.4. The spectrum at high wavenumbers is severely eroded, and a bump appears at the highest wavenumbers $k^* > 2$. However, this bump is not as pronounced as in the experiment. The explanation for the presence of the bump in these simulations is similar to that for the bump observed in measurements. Due to the limited amount of tracers ($10^6$) used to construct a thick line, the profile appears noisy, comparable to the photon noise in measurements (see Fig. 5.16). This compromises the line fitting, which leads to a background in the spectrum.

Finally, Fig. 5.17 shows the normalised second-order structure function of the kinematic simulation. Thin mathematical lines that are advected over $\tau_\eta$ now display a scaling behaviour $G_2(r) \sim r^{\zeta_2}$, with $\zeta_2 = 0.67$ which is indistinguishable from the predicted scaling behaviour $\zeta_2 = 2/3$. At small scales both curves display the scaling of a smoothed vector-field $G_2(r) \sim r^2$. However, this extends to much larger scales ($r/\eta \approx 400$) for the diffusing lines. It should be noted that, like in the experimentally determined structure functions, the equivalent of the high-wavenumber bump in Fig. 5.15 is absent in the structure function (compare with Fig. 5.4 and Fig. 5.6). Apparently, the nature of the fit errors introduced by photon(-like) noise is such that its influence is greater in wavenumber space than in real space.
Figure 5.15: Comparison of spectra for time $\Delta t/\tau_H = 1$, the upper spectrum is calculated from displacement of thin lines, the lower is calculated from lines with finite thickness with a Gaussian profile, subject to diffusion.

Figure 5.16: Thick line distorted by turbulence in a kinematic simulation. The inset shows a magnification of a part of the line. The profile appears noisy due to the limited amount of tracers used to construct the line.
Figure 5.17: The second order structure functions. The dotted line shows the structure function $G_2(r)$ as calculated from the velocities $u(ax)$ directly probed from the synthetic flow field. The upper solid curve shows $G_2(r)$ found from thin lines and the lower one shows $G_2(r)$ from diffusing lines.
5 Two-point correlation statistics

5.5 Conclusions

In this chapter it has been shown that APART molecular tagging velocimetry can be used to measure streamwise velocity increments transverse to the separation. The first order measurements of turbulence, such as the velocity probability density functions in Ch. 3, yield results as are generally expected. However, we do find a significantly different behaviour for second order statistics.

The MTV spectrum is significantly different from the synthetic spectrum (Fig. 5.4). At large wave numbers the measured spectrum displays a background, most probably caused by photon-noise, while at low wave numbers the effect of the finite extent of the images is felt. Compared to the synthetic spectrum, the integral scale is still an order of magnitude away. Additionally, diffusion is shown to have a very significant effect on the spectrum. These effects make that the experimental spectrum lacks a clear $k^{-5/3}$ behaviour.

The second order structure function (Fig. 5.6) looks much better, although it does not increase rapidly enough at the small scales. It was shown that the scaling exponent, $G_2(r) \sim r^{\zeta_2}$, $\zeta_2 \simeq 0.77$ is significantly larger than the self-similar one, $\zeta_2 = \frac{2}{3}$. At the largest separations the measured structure function is still far from the asymptotic value, $2u^2$.

We have considered how different types of image artifacts may result in erroneously resolved line centres and may indirectly influence the shape of the spectrum. In practice removal of these image artifacts will have some effect on the velocity statistics, but not significantly so when the differences between the measured spectra and the true spectra are considered. Although the “hump” at the smallest scales is slightly reduced, there is no real improvement in retrieving an inertial range exponent that better resembles the expected $k^{-5/3}$. We have determined that this is mainly attributable to diffusion.

By means of kinematic simulations we have investigated the deviation of two point correlations from the expected results. In Ch. 4 it was shown that when using molecular tracers, the smallest scales of turbulence will inherently remain unrecognizable since (at the Kolmogorov time) the thermal diffusion length scale will be of the same order as the smallest turbulent scale. Using numerical simulations the effect of diffusion on the spectrum was studied. The simulated spectrum (Fig. 5.15) of the diffusing line bears a striking similarity to the actually measured spectrum, which was shown in Fig. 5.4. The spectrum at high wavenumbers is severely eroded and a bump appears at the highest wavenumbers. The bump can be explained by photon-like noise in the simulation compromising the line fitting procedure. The deviating inertial range exponent found in the numerical simulations with diffusion corroborates the hypothesis that deformation of the spectrum is indeed mostly due to thermal and turbulence induced diffusion.

Additionally, the finite time between tracer creation and read-out results in an average over the small-scale velocity. This effect is significant since the tag-probe time
is of the same order as the Kolmogorov time $\tau_\eta$ (12 $\mu$s). The numerical simulations show that especially the small scales are affected and with increasing time, this will extend to larger scales in the spectrum. The largest scale that is affected depends on the tag-probe time as $1/k^* \simeq t^{0.8}$. This is reasonably close to what is expected: for short times, the separation between particles grows linearly with time.
5 Two-point correlation statistics
6 Intermittency

6.1 Introduction

In this chapter we will focus on higher order statistics. In particular, we will examine probability distribution functions for velocity increments over different separations and demonstrate a non-Gaussian distribution of the velocity increments for small separations. This anomalous behaviour is indicative for intermittency, a turbulence property we will explore in Section 6.3 by means of higher-order structure functions. We will investigate how higher-order statistics can be measured using the APART technique, and we will pay attention to how sources of distortions, such as smearing-out of structures due to diffusion and temporal averaging of the velocities during the write-read delay, affect the higher order statistics. Also, errors occurring during the fit procedure result in perceived high velocities. Such falsely found high velocities can lead to large velocity increments on the smallest scales, and will mostly affect the higher order structure functions. For the highest order statistics even very infrequent fit errors may influence the results.

In this chapter we also investigate how an increase in the write-read delay will influence the statistics. The sources of possible artifacts have already been discussed in some detail in Chapter 5: increased diffusion, time-integrated rather than instantaneous velocities and self intersections of the line. We will examine if and how these have an effect on the higher-order statistics that will be presented.

6.2 Probability distribution functions

6.2.1 Theory

The second hypothesis of Kolmogorov states that small-scale turbulence is isotropic and homogeneous for large Reynolds numbers, with statistical properties only depending on the viscous dissipation rate $\epsilon$ and the scale. If we consider the probability distribution function (PDF) $P_r(\Delta u')$ of fluctuations of non-dimensionalised velocity increments over small scale separations, it follows that these should be universal, that is, independent of large scale geometry, Reynolds number and separation $r$. The velocity is non-dimensionalised by $\epsilon$ and $r$ as $\Delta u' = \Delta u(r)/(r\epsilon)^{1/3}$. The evolution of the shape of the PDF as a function of $r$ results in a non-trivial behaviour of
the moments. These moments, or so-called structure functions, are defined as:

\[ G_p(r) = \int P_r(\Delta u)\Delta u^p d\Delta u. \]  

(6.1)

If \( P_r(\Delta u) \) has a form which is parametrized by a single \( r \)-dependent variable (its Gaussian width, for example) the moments of different order \( p \) are trivially related. Therefore, any breaking of scale invariance also implies that the structure functions of different orders are no longer related in a self-similar manner.

In our molecular tagging scheme we infer velocities from the displacement of lines. Because we measure velocities perpendicular to the line, we only have access to the PDF of transverse velocity increments, from which we can derive the transverse structure functions. Velocity increments

\[ \Delta u_r(x) = u_2(x + \vec{e}_i \cdot \vec{r}) - u_2(x) \]  

(6.2)

can be collected for all lines and all points on each line. Because of homogeneity and reflection symmetry, the PDF of \( \Delta u_r \) does not depend on \( x \) and is symmetric. In passing, we notice that the longitudinal PDF’s \( P_{rL}(\Delta u) \) where the separation vector \( \vec{r} \) points in the same direction as the velocity is asymmetric. This can be understood from a symmetry argument put forward in [68]: Consider the two points and their longitudinal velocities. Within the frame moving along with velocity \( u \), the possibility to convey information from \( x \) to point \( x + r \) depends on the sign of the velocity increment \( \Delta u \). Hence, there must be a difference between the positive and negative side of the distribution.

### 6.2.2 Results

Figure 6.1 shows probability density functions \( P_r(\Delta u) \) measured for two different separations, \( r = 1.2 \cdot 10^{-4} \text{ m} \) and \( r = 4.1 \cdot 10^{-3} \text{ m} \), respectively. With \( \eta = 1.4 \cdot 10^{-5} \text{ m} \), this corresponds to a separation inside the dissipative range \( (r/\eta = 8.6) \) and one inside the inertial range \( (r/\eta = 295) \), respectively.

The velocity increments inferred from line displacements were accumulated for \( 4 \cdot 10^3 \) lines and two delay times \( \Delta t = 10 \mu s \) and \( \Delta t = 30 \mu s \). Especially for the longer delays, \( \Delta t/\tau_\eta = 30 \mu s/13 \mu s \simeq 2.5 \), where lines are severely warped, this is a daunting procedure (see Sec. 6.4). Still, the PDF’s at the two delay times agree well with each other. We also observe that very large velocity differences are observed over very short distances: they have been found as large as \( 20 \text{ ms}^{-1} \) over a mere \( 120 \mu \text{m} \). This must be compared to the mean velocity \( U = 40 \text{ ms}^{-1} \) and rms velocity fluctuation of \( 13 \text{ ms}^{-1} \). In Section 6.4 we will present information about the nature of these large events.

Finally, we notice that the PDF’s are perfectly symmetric, which is demonstrated by overlaying \( P_r(\Delta u) \) and \( P_r(-\Delta u) \). At smaller separations \( r \) the measured line displacements are influenced by diffusive broadening, and no longer represent true
Figure 6.1: PDF of transverse velocity increments. Plot a) shows velocity data visualized with $\Delta t = 10\,\mu s$ write-read delay and plot b) with $\Delta t = 30\,\mu s$ delay time. In both graphs the widest PDF corresponds with separation $r = 4.1 \cdot 10^{-3}$ m and the narrowest with $r = 1.2 \cdot 10^{-4}$ m. Additionally, all PDF’s are mirrored, (gray lines) to check for symmetry.

hydrodynamic velocity variations. Because the largest separation is still smaller than the integral length scale, the corresponding PDF has not yet returned to Gaussian.

In order to emphasize the non-Gaussian shape of the PDF’s, we show in Fig.
6.2 the same PDF’s, but now with $\Delta u/\left\langle (\Delta u(r))^2 \right\rangle^{1/2}$ as horizontal axis, so that every Gaussian velocity distribution is represented by the same parabola (on semi-logarithmic axes). For large separations the plots show a near-perfect Gaussian distribution function.

One way to quantify the deviation of our PDF’s from a Gaussian distribution is by representing the tails of the PDF as stretched exponentials

$$P_r(\Delta u) = \beta e^{-\alpha |\Delta u|^n},$$

where for a Gaussian $n$ takes on the value 2. In Fig. 6.2 the fitted exponentials are shown as the bold lines. The figure illustrates that these give an accurate representation, with $n = 0.34$ at $r = 1.2 \cdot 10^{-4} \text{m}$, and $n = 1.0$ at $r = 4.1 \cdot 10^{-3} \text{m}$. At smaller separations, no satisfying fit with stretched exponentials could be found.

This indicates that at these scales the line-wrinkles follow a different statistical behaviour and thus are caused by a process that is different from Kolmogorov turbulence. If we compare the PDF’s at the two delays, we see that at $2.5\tau_\eta$ the plots are slightly more noisy than at $0.8\tau_\eta$. Furthermore, we see that the stretched exponentials of the tails at $2.5\tau_\eta$ are smaller than those for $0.8\tau_\eta$. This is related to fit ambiguities that are inherent to line-fitting for longer delays. This point is further explored in Sec. 6.4.

6.3 Structure functions

6.3.1 Theory

From the probability density functions of the transverse velocity increments, the structure functions follow as

$$G_{\perp}^p(r) = \int P(\Delta u) \Delta u^p d(\Delta u),$$

where in the transverse arrangement that is accessible in our molecular tagging scheme, the velocity components that make the increments $\Delta u$ are oriented perpendicular to the separation vector $\vec{r}$, which we indicate by the superscript $\perp$; if they are oriented along the separation vector $\vec{r}$, we indicate this by the superscript $\parallel$.

The second order structure functions are related to the correlation functions that were introduced in Section 5.2.1 by Eq. 5.12 and Eq. 5.5

$$u^2 f(r) = u^2 - \frac{1}{2} G_{2}(r), \quad u^2 g(r) = u^2 - \frac{1}{2} G_{2}^\perp(r),$$

where $u$ is the rms turbulence velocity, $G_2 = G_{11}$ and $G_2^\perp = G_{22}$.

There is little one can say about the structure functions without solving the turbulence problem; so far this has proven to be an elusive goal. However, for the
6.3 Structure functions

Figure 6.2: PDF’s of transverse velocity increments, normalized with the \( u_{rms} \) value for its respective separation \( r \): \( \langle (\Delta u (r))^2 \rangle^{1/2} \). To check for symmetry, the horizontal mirror images of the PDF’s are added (gray lines). In the upper figure the write-read delay is 10 \( \mu \)s, in the lower the delay is 30 \( \mu \)s. The bold lines show the fitted stretched exponentials.
3rd order longitudinal structure function, an exact result was found by von Kármán and Howarth [69]:

\[
G_3^\parallel(r) = -\frac{4}{5} \langle \varepsilon \rangle r + 6\nu \frac{\partial G_3^\parallel(r)}{\partial r}.
\]  
(6.6)

where \( \langle \varepsilon \rangle \) is the mean energy dissipation. For this result to hold, the following conditions must apply: turbulence must be homogeneous and isotropic, with a clear separation between the scales where energy is injected and the scales \( r \) where Eq. 6.6 holds.

When the scales that are considered are separated far from the small scales where dissipation reigns, we may set \( \nu = 0 \) in Eq. 6.6 and find:

\[
G_3^\parallel(r) = -\frac{4}{5} \langle \varepsilon \rangle r.
\]  
(6.7)

Inspired by this result, Kolmogorov postulated a similar behaviour for all other moments \( p \neq 3 \)

\[
G_p^\parallel(r) = C_p \langle \langle \varepsilon \rangle r \rangle^{p/3},
\]  
(6.8)

where the scaling exponent (called \( \zeta_p \)) is \( p/3 \). No exact result can be derived for orders \( p \neq 3 \), and no exact result exists for the transverse structure functions. However, in isotropic turbulence the second order functions \( G_2^\parallel \) and \( G_2^\perp \) are related in the way described in Sec. 5.2.1, which forces the scaling exponents to be the same.

In Kolmogorov’s refined similarity hypothesis [70], velocity distributions are assumed to be determined by the local energy dissipation of order \( p \): \( \varepsilon_r^p \), spatially averaged over a sphere with size \( r \). The scaling of the velocity increments over a distance \( r \) (that is, the velocity increments in vortices of size \( r \)) is related to the scaling of the energy dissipation inside a sphere of size \( r \) by

\[
\langle \varepsilon_r^p \rangle \sim r^{\tau_p}.
\]  
(6.9)

The factor \( \tau_p \) is related to the structure functions scaling exponent by

\[
\zeta_p = p/3 + \tau_p/3.
\]  
(6.10)

In fact, this observation can be used to devise a model for \( \zeta_p \). A recent model proposed by She and Leveque [71] suggests a description of the scaling of the turbulent field in terms of a sequence of moment ratios of the dissipation rate \( \varepsilon_r \) at scale \( r \). These moment ratios are related by

\[
\varepsilon_r^{(p)} = \frac{\langle \varepsilon_r^{p+1} \rangle}{\langle \varepsilon_r^p \rangle},
\]  
(6.11)

with a hierarchy determined by a specific function \( \varepsilon_r^{(p)} \) such that it leads to a scaling exponent

\[
\tau_p = -2p/3 + 2[1 - (2/3)^p],
\]  
(6.12)
and therefore
\[ \zeta_p = p/9 + 2[1 - (2/3)^{p/3}]. \] (6.13)

It was further shown by She and Leveque [72] that such a scaling model corresponds with a Log-Poisson statistical distribution of the energy dissipation.

### 6.3.2 Results

In Fig. 6.3, we have plotted transverse structure functions of orders 1 to 8 in a log-log plot, again for both long (30 µs) and relatively short (10 µs) delays.

The structure functions for both delays display a power law dependence in the inertial range for separations larger than 40 \( \eta \), where we are able to discern an exponential scaling range of at least one decade. Although this is a fairly small region for fitting the scaling behaviour, we are still able to find accurate scaling exponents. All plots have been reduced by plotting \( G_{1/p}^{1/p}(r) \) so that in the K41 prediction all orders would have the same scaling exponent of 1/3. In the scaling range, the scaling component is fitted on a log-log scale and thus retrieved by

\[ \frac{d \log \left( G_{1/p}^{1/p}(r) \right)}{d \log r} = \zeta_p/p \] (6.14)

We see from Fig. 6.3 that the reduced slopes \( \zeta_p/p \) decrease for increasing orders \( p \). For orders above 8 the statistical convergence is insufficient and the exponents rapidly deteriorate. We observe furthermore that in the dissipation range an additional hump is present, where we would have expected a cut-off. It is noted that the small scale artifacts, again in the shape of a hump, are considerably less prominent than those found in spectral measurements. The same conjectures concerning the source of this phenomenon that we have put forward in the discussion of the energy spectrum as discussed in Sect. 5.2, apply to this effect. Both diffusion and image noise will influence the smallest scales. One would expect that this artifact does not affect structure functions much at the inertial range.

It is interesting to observe the differences between interrogation delays of 10 µs and 30 µs. In the latter case, the small-\( r \) behaviour at the low-order structure functions shows a steeper decline. This is because longer time delays give a larger displacement signal so that small displacements are more significant with respect to discretization. However, the larger moments emphasize the rare events and will emphasize the increasing ambiguity of a velocity that is inferred from a displaced line. It is noteworthy that otherwise the two sets of structure functions look similar. This means that for a delay as long as 30 µs even the extreme velocities, which are rare, and more prone to fit errors, come out correctly.

In Fig. 6.4, we have plotted the exponents as a function of \( p \). The solid circles show the measurements done at 10 µs delay, the solid squares show the data at 30 µs. Since in our lab-setup turbulence is generated at moderate Reynolds numbers,
Figure 6.3: Log-log plot of reduced transverse structure functions of moment orders 1 to 8. In the upper figure the write-read delay is 30 $\mu$s, in the lower the delay is 10 $\mu$s. The structure functions display a power law dependence for separations larger than 40 $\eta$. 
6.4 Violent events

So far, we have only considered turbulent velocities perpendicular to the line, in the plane of view. However, in isotropic turbulence the velocity fluctuations lead to a redistribution of the concentration of tagged molecules. In fact, in strong straining regions, the concentration may become so low that the line appears to be cut. In most of these cases, also the deformation of the line in the $x-y$ plane is very large.

Figure 6.5 shows 6 “difficult” lines at a delay of 30 $\mu$s that were selected from $4 \cdot 10^3$
lines using the criterion that their minimum intensity should be below a set value close to the background noise level, with the average intensity above a threshold value. These lines do indeed show very strong deformations and can hardly be used for velocimetry. Indeed, they were excluded from the measurement of statistical quantities.

Although it is possible that the line has been displaced so much that it is not illuminated anymore by the “read” laser or has moved outside of the focal plane of the camera, we observe the same patches in 2D projections of strained lines generated in kinematic simulations (where laser intensity and defocussing do not play a role). An example of such a line is shown in Fig. 6.6. It should be noted that the time scale on which the experiment is done ($\Delta t = 2.5\tau_\eta$) is much shorter than in numerical simulations ($\Delta t = 10\tau_\eta$), although the difference in write-read delay can be smaller if an alternative definition is used for $\tau_\eta$ (see Section 5.4.2).

### 6.5 Conclusions

The probability distribution functions for velocity increments show that for decreasing separations, the distributions turn from Gaussian into distributions with stretched-exponential tails. This change of the shape of the distribution function is indicative of intermittency. The same intermittent behaviour is seen in the structure functions. We observe that just as in spectral measurements artifacts present in the line displacements also play a role in structure functions: in the dissipation range a “bump” is present, where we would have expected a cut-off at the dissipative scale. However, contrary to the spectral measurements, the inertial range shows a proper scaling behaviour. We have discussed a model for the scaling exponents based upon the refined similarity hypothesis of Kolmogorov.

The results show that the set delay between the writing and reading of a line indeed has an influence on the higher order statistics. Due to straining, the lines can seem to be cut off. Also, large deformations can be found, making fitting ambiguous. Other experiments [73, 68, 74] and simulations [75, 71] have shown that the high order longitudinal and transversal structure functions may have different scaling exponents, but (for short times) we have not observed a discernible difference with the (longitudinal) Log-Poisson model. The same was also found from the transverse velocity measurements by Noullez, Miles and Frisch [29].
Figure 6.5: Violent events in lines subject to strong high turbulence ($Re_\lambda = 500$). Measurements are done at long delays ($\Delta t = 30 \mu s$). Large deformations and low line intensity make accurate fitting difficult.
Figure 6.6: Violent events and patchy structure in kinematic simulations at very long delays ($\Delta t = 10\tau_\eta$).
7 Line stretching

7.1 Introduction

An important characteristic of turbulence is its ability to transport and mix fluid very effectively. This phenomenon can be expressed in terms of the evolution of the material surface or line separating two differently marked 3D or 2D regions of the flow, respectively.

Although the subject of surface deformation has often been investigated, it has mostly been done through numerical simulations [76, 77]. With the technique of APART we can experimentally study the increase of length of a line by photochemically creating a thin line of NO molecules in a flow, and subsequently measuring the deformed line after a delay time by LIF detection. In this chapter we will discuss these measurements and their experimental difficulties.

The straining of material surfaces was first studied in 1952 by Bachelor [78], where he assumed that straining is persistent and lines orient towards the axis of maximum strain. As measurements on material surfaces using classical techniques had proven to be difficult, and were even considered impossible because of “insuperable experimental difficulties” [45], it was not until numerical simulation techniques and computers had sufficiently matured that material surfaces and lines could be followed and that these conjectures could be tested by numerical experiments [79, 45, 80, 77]. There it was shown, amongst others, that the line elements orient differently than expected. This point will be further discussed in Sec. 7.4.2. Additionally, local curvature of the line was introduced as a typical property of material surfaces. Although curvature is not as directly related to a relevant physical property in turbulence, such as mixing, as the stretching rate is, it has been shown to be related to line stretching properties [81].

In another branch of research on surfaces and lines [82, 83], it was shown that the structure of material lines could be explained with the concept of the fractal dimension [84]. This work was further expanded [85, 86, 87] and an expression was found to describe the increasing fractal dimension of a deforming surface in time. We will discuss this further in Sec. 7.4.1.

The theoretical prediction of the evolution of the stretching rate as a function of time predicted in [77] was confirmed in extensive and careful simulations by Kida and Goto [88, 89, 90]. There, it was also shown that there is a significant difference in the measured mean of flow properties along a true material line/surface, compared to the average over elements on randomly chosen positions. We will discuss the
7 Line stretching

argumentation in Section 7.4.2 and Appendix A.

7.1.1 Stretching rate

For points close together in a fully developed turbulent flow, the distance between them increases exponentially with time. One would then assume the line length will also increase exponentially in time. However, there is an interesting twist: The line length increase does not follow a simple mono-exponential trend, but the exponent will change as a function of time [91]. The origin of the transient behaviour of the stretching rate will be discussed in Section 7.4.2 where a simple model will be used to describe its characteristic features. Therefore, let us describe the line growth as

\[ L(t) = L_0 e^{\tilde{\Gamma}(t)/\eta}, \quad (7.1) \]

where \( \eta \) is the Kolmogorov time and \( \tilde{\Gamma}(t) = \Gamma(t)/\eta \) is the reduced exponent. The exponent is defined in this way to find a stretching rate which is (largely) independent of the Reynolds number. In order to visualize the evolution of the line length, we introduce a non-dimensional, derivative, stretching “rate” as:

\[ \gamma(t) = d\tilde{\Gamma}(t)/dt. \quad (7.2) \]

This derivative stretching rate will change in time until, ultimately, a constant value is reached. As the derivative of the growth is sensitive to noise, we will for practical purposes mostly use an alternative definition of the stretching rate, namely

\[ \tilde{\gamma}(t) = \tilde{\Gamma}(t)/t. \quad (7.3) \]

The advantage of definition 7.3 is that the asymptotic growth rate is better defined, but for short times the error is still very significant.

7.1.2 Fractal scaling

We have seen in previous chapters that the lines that are produced with the APART technique are subject to different forms of artifacts, and that these artifacts are mostly apparent on the smallest scale. This is something to consider when we determine the lengths of these lines. It is very well conceivable that, if we zoom in close enough on the line, much of the length that is contained in the smallest wrinkles of the line may be the result of noise. In order to resolve this error, we determine the total line length by covering it with straight line elements of a predetermined length (Fig. 7.1). These line elements, which we will refer to as rulers, are given a length that is greater than the length-scale dominated by noise.

The usage of rulers to measure line length is not merely in order to filter out noise, it also gives insight in a characteristic property of a material line, its fractal nature.
Figure 7.1: Estimating the line length using rulers. If the rulers (the grey line elements) are longer than the smallest features of the line, its length is underestimated.

In general, if the line has a fractal dimension $d_f$, the number of rulers of length $l_0$ to cover the line with length $L$ scales as

$$N(l_0) = \left( \frac{l_0}{L} \right)^{-d_f}. \quad (7.4)$$

Material lines in turbulence are fractals, and Eq. 7.4 can be used to infer the true length from the $l_0$ behaviour. The inferred length then follows from

$$\tilde{L} = l_0 N(l_0) \sim L \left( \frac{l_0}{L} \right)^{1-d_f}. \quad (7.5)$$

Typically the dependence $\tilde{L}(l_0)$ will look as sketched in Fig. 7.2. For lines without noise, the line is smooth on scales smaller than $l_0 \approx \eta$, and the inferred line length $\tilde{L}(l_0)$ with $l_0 < \eta$ becomes constant and equal to the true line length $L$ (see Fig. 7.2). For lines with (white) noise, the line length does not reach an asymptote when $l_0$ goes to zero. In this chapter we propose to use the algebraic behaviour of $\tilde{L}(l_0)$ to estimate the line length of $l_0 = \eta$ by extrapolation. We will investigate how exactly the line length scales with time and with ruler length.
7.1.3 The stretching rate in a 2D projection within a bounded region

The next point that we need to address is the following: in our single camera setup, we can only capture a two dimensional projection of the line. The consequence is that we cannot observe the line fluctuations in the direction perpendicular to the plane of view (the $z$-direction). We will assume that the fluctuations in the $z$-direction are statistically equivalent to those in the $y$-direction, i.e. the direction of the mean flow. This means that we can make a prediction of the length increase in the unobserved $z$-direction. In the next section we will discuss how this affects the found line length and stretching rate. Closely related to this is the problem of line growth in the parallel direction along the line and, thereby, out of the region imaged by the camera. Because these issues cannot be settled in the experiment we will address them in the context of kinematic simulations of a turbulent field.

7.1.4 Transient behaviour

With our experimental technique we can only investigate line deformation for relatively short times (a few $\tau_\eta$). For these times, it could be that we can still interpret the line displacement in an Eulerian frame, and thus it could be argued that the line length $L(t)$ is related to the already known Eulerian properties. In Section 7.2.5 we will see that, for short time approximations, we could indeed link line stretching to the Eulerian structure function by:
7.2 Experimental considerations

\[ \frac{L(t)}{L_0} = \left( 1 + \frac{2G_2(l_0)t^2}{l_0^2} \right)^{\frac{1}{2}}, \quad (7.6) \]

thereby making line stretching at these times a trivial representation of an already known Eulerian property. We will investigate if, for times that are not infinitesimal, but relatively short, the line length will still be trivially related to the second order structure function, or if the mechanics (the interplay between rotation and strain) that govern line stretching will show another behaviour. In Section 7.4.2 we will test this by comparing the stretching rate calculated using the directly measured line length with the stretching rate indirectly calculated from \( G_2 \) of the dataset.

7.2 Experimental considerations

7.2.1 Fitting

In the evaluation of the experiments we fit mathematical lines to the observed “sausages” of tagged molecules. The line center position is determined (with sub-pixel resolution) for each vertical strip \( i \), so that each point on the line has an \( x- \) and \( y\)-position \( x(i), y(i) \). The initial value for \( x(i) \) is chosen \( x(i) = i, i = 1 \ldots n \) where for our camera \( n = 1024 \).

It has already been discussed in Sect. 6.4, that the vertical fit technique may experience problems in fitting written lines that are far from horizontal. Since we aim to follow line growth to as long time scales as possible, we see that lines can deform to such an extent that simple vertical fitting will not suffice anymore. Thus, in a second pass, the results found for \( (x(i), y(i)) \) are used as initial values for a fit in the direction locally perpendicular to the line. Note that, in this second pass, the line center will be refitted along a diagonal slice, so that now \( x(i) \neq i \). The total line length for line \( m \) is now calculated using ruler increments that scale as \( r_0(k) = 2^{k-1} \), where \( k \) is an integer value.

\[ L_j(k) = \sum_{i=1}^{N_{r_0}} l_{r_0(k)}(i), \quad N_{r_0} = n/r_0(k) \quad (7.7) \]

with:

\[ l_{r_0}(i) = \left( \Delta x_i(r_0)^2 + 2\Delta y_i(r_0)^2 \right)^{1/2}, \quad (7.8) \]

\[ \Delta x_i(r_0) = x_{i-r_0} - x_{(i-1)-r_0}, \quad (7.9) \]

\[ \Delta y_i(r_0) = y_{i-r_0} - y_{(i-1)-r_0}. \]

The factor of two multiplying the \( y\)-component is to account for the missed fluctuations in the \( z\)-direction, as will be justified later. Note that where in the previous
definition of the ruler $l_0$ was of fixed length, now the horizontal separation is nearly constant (but not quite, due to the second stage of the perpendicular fit) and line element lengths will slightly vary for $l_{r_0}(i)$, $i = 1 \ldots n$ with constant increments $r_0(k)$. Because of the constant horizontal separation, the number of rulers $N_{r_0}$ is now fixed for a constant value of $r_0(k)$. In numerical simulations it was checked that this gives the same outcome.

Line properties are now calculated as the mean over $M$ measured lines, recorded under the same conditions (time delay, laser alignment, etc.). From the obtained values of $L_{j}^{(t)}(i)$, the stretching rate $\tilde{\gamma}_k(t)$ is derived as

$$
\tilde{\gamma}_k(t) = \frac{\tau_0}{t} \frac{1}{M} \sum_{i=1}^{M} \ln \left( \frac{L_{j}^{(t)}(i)}{L_{ref}^{(t=0)}(i)} \right),
$$

where $L_{j}^{(t)}$ are lines at time $t$ and $L_{ref}^{(t=0)}$ is the unperturbed reference line. Note that for a completely straight line, $L_{ref}^{(t=0)}$ is independent of $k$.

### 7.2.2 Fit problems

As mentioned above, in order to better fit deformed lines, we have used a second stage of fitting, where a perpendicular fit is applied. However, in severely contorted lines, problems will most likely occur in the first stage. The crux of our method lies in the fact that evenly spaced steps are taken along the $x$-axis. This will be untenable when the line folds back or self-intersects, and a parametric description is needed.

This will require a different type of line detection. Since careful fitting is done in the second stage using perpendicular fits, the first stage can be relatively crude. A scheme that simply traces pixels of maximum intensity would therefore be sufficient. A possible algorithm is illustrated in Fig. 7.3.

Around a previously found point on the line, a square grid is formed with a grid size comparable to the line width, and within this grid the cell with maximum intensity is sought. In this grid cell the actual point position can be further specified by finding the intensity median. From this location the procedure is repeated for a next point, until the complete line is traced. Such a scheme has not yet been implemented but is considered as a future improvement.

A promising alternative that is currently tested [92] is an active contour model in which an energy-minimizing spline follows image contours while being influenced by constraint forces [93, 94]. This technique of boundary detection and tracking is regularly used in medical applications of vessel tracking [95].

It remains to be investigated to what extent such a new technique will help us to follow lines until longer delay times, since there are other experimental difficulties as well as fundamental limitations, especially when analyzing severely deformed lines. As we have seen in Fig. 6.5 lines will start widening due to diffusion, but we also
see the creation of patches of higher and lower intensity. Some initial measurements with the active contour model indicate that the resulting line stays locked onto lines, even in patchy regions, when a certain tension term is prescribed to the spline.

For now, we use the perpendicular fitting routine followed by meticulous filtering of lines containing evident errors. First, it is determined whether the minimization routine employed during fitting of a strip terminates with an error. This usually indicates that no Gaussian peak could be found in the cross-section. This can be the case because the initial guess of the location of the peak position was incorrect but also because the image does not contain a line of sufficient intensity. If the former is the case, the problem can sometimes be remedied by using another algorithm for the initial guess. If this does not work, an error will be reported and the line is ignored.

Secondly, the intensity of the line as a whole is checked. If the intensity is not sufficiently high, the light from the oscillator and amplifier stages in the write laser probably did not coincide either spatially or temporally resulting in a low intensity, broadband laser pulse. Thus, when the average line intensity lies below a set threshold the line is discarded.

Before we can turn to line stretching experiments, we first have to discuss some potential limitations of our method.

### 7.2.3 2D projection

Using only one camera, the displacement in the $z$-direction cannot be observed, and we have to restrict ourselves to a 2D projection of the line. We can, however,
7 Line stretching

make a reasonable estimation of the extra contribution of the missing dimension. As expressed in Eq. 7.8, we can represent the missing information by allowing for a factor 2 in the stretching rate. We will now justify this assumption.

Let us consider a line in 3 dimensions and additionally assume that the rulers used to measure its length yield increments \( \Delta x^2_i, \Delta y^2_i \) and \( \Delta z^2_i \), where the latter is defined similar to Eq. 7.9. Then, one realized line has a length

\[
L_j = \sum_i \left( \Delta x_i^2 + \Delta y_i^2 + \Delta z_i^2 \right)^{1/2},
\]

(7.11)

while the original line (at \( t = 0 \)) has a length \( L_{ref} = \sum_i \Delta x_i \). Therefore

\[
\frac{L_j}{L_{ref}} = \frac{\sum_i \Delta x_i \left( 1 + \left( \frac{\Delta y_i}{\Delta x_i} \right)^2 + \left( \frac{\Delta z_i}{\Delta x_i} \right)^2 \right)^{1/2}}{\sum_i \Delta x_i}.
\]

(7.12)

\[
\simeq 1 + \frac{1}{2} \sum_i \left( \frac{\Delta y_i}{\Delta x_i} \right)^2 \Delta x_i + \frac{1}{2} \sum_i \left( \frac{\Delta z_i}{\Delta x_i} \right)^2 \Delta x_i.
\]

(7.13)

where the approximation is justified if the slopes of the line are small. The mean stretching rate over a set of lines is defined as

\[
\langle \ln \frac{L_j}{L_{ref}} \rangle \simeq \frac{1}{2} \left( \frac{\sum_i \left( \frac{\Delta y_i}{\Delta x_i} \right)^2 \Delta x_i}{L_{ref}} \right) + \frac{1}{2} \left( \frac{\sum_i \left( \frac{\Delta z_i}{\Delta x_i} \right)^2 \Delta x_i}{L_{ref}} \right).
\]

(7.14)

In isotropic turbulence the averaged terms are equal. Therefore, if the local slopes are small and the approximations are valid, the stretching rate from line lengths

\[
L_j = \sum_i \left( \Delta x_i^2 + 2 \Delta y_i^2 \right)^{1/2}
\]

(7.15)

is the same as that of Eq. 7.11, at least on average. Since \( \Delta x_i^2 \) is (very nearly) constant, the stretching rate derived from actual 3D line lengths would be two times larger than that derived from 2D projections. The key assumption in this argument is that everywhere along the line the slope is small, so that \( \frac{\Delta y}{\Delta x} \ll 1 \) and \( \frac{\Delta z}{\Delta x} \ll 1 \). Especially for longer time scales this need not be true. To judge how well this approximation holds in practice, kinematic simulations have been performed.

In these simulations initially straight lines were allowed to deform in a 3 dimensional flow field and were evaluated at fixed times. This was done for a set of a 1000 lines. At these times the line lengths were measured in three dimensions as well as the lengths of the 2-dimensional projection. Both were averaged over all lines. In the last case a procedure was followed to deal with apparent self-intersections.
7.2 Experimental considerations

Figure 7.4: Kinematic simulations of line stretching. The lines show the stretching rate $\gamma$ for the 3D line length and the line length of the 2D projection of the same lines. The inset shows the ratio between the two.

This procedure has been described in Sect. 5.4.3. In Fig. 7.4 the stretching calculated from these 3D and 2D line lengths is shown as a function of time. The inset shows the ratio between the two, which is almost perfectly 2, except for very short times. In later experiments we will see that the relation holds even for much longer times ($t = 10\tau_\eta$). This is remarkable, since for such long delays the line slopes are significant.

7.2.4 Window influence

When lines are deforming, they will not only grow in the direction perpendicular to the line, but also in the parallel direction along the line and, thereby, out of the region imaged by the camera. As is schematically drawn in Fig. 7.5, this may result in an addition to the total line length that is not captured in the measurements. The effect of the window on the stretching rate was estimated by using kinematic simulations in the same way as described in the previous section. However, in the present simulations a box is defined that bounds the unperturbed lines at $t_0$. When the line has deformed for the set time, both the full line length and the length of the line part bounded by the box were determined. The dashed line in Fig. 7.6 shows the stretching rate of complete lines. The solid line shows the stretching rate for the part
7. Line stretching

Figure 7.5: Lines are truncated when stretching occurs longitudinally resulting in underestimated line length and stretching rate. The longitudinal line elongations $\Delta l_1$ and $\Delta l_2$ are not captured by the camera and do not contribute to the line length.

of the lines that is contained within the bounding box (the reference line was also bounded). It is clear that the stretching rate of the full line is only marginally larger than that of the truncated line. Not only does this indicate that line truncation due to bounded images does not pose a serious problem in measurements, it is also an indication that the line length increases mostly by the smallest scales.

7.2.5 Relation second order structure function to line length

For the short time delays in our experiment (up till a few times $\tau_p$), an Eulerian interpretation of the tagged molecule displacement is still possible. Assuming that the time delays are small enough for the extraction of Eulerian quantities, the line length can be computed from Eq. 7.15 and setting $\Delta x_i = l_{r_0}$, $\Delta y_i = t \Delta u_i(r_0)$ and $\Delta u_i(r_0) = u_{i-r_0} - u_{(i-1)-r_0}$:

$$L_j(t) = \sum_{i=1}^{N_{r_0}} \left[ \frac{t^2}{l_{r_0}} + 2 t^2 (\Delta u_{r_0}(i))^2 \right]^{1/2} L_{ref} = N_{r_0} l_{r_0}, \quad (7.16)$$

with $l_{r_0}$ the ruler length and $N_{r_0}$ the number of rulers needed for computation of the length. The factor 2 accounts for the unobserved out of plane deformation of the line. The stretching rate involves an ensemble average of $\ln(L(t)/L(0))$. In order to relate this to the Eulerian second order structure function, we make the following two drastic assumptions:

$$\langle \Gamma \rangle = \left< \ln \left( \frac{L_j(t)}{L_0} \right) \right> \approx \ln \left( \frac{\langle L_j(t) \rangle}{L_0} \right) \quad (7.17)$$
7.3 Measurements

7.3.1 Data validation

For a set of write-read delays, ranging from 1 $\mu$s to 30 $\mu$s, lines have been recorded in a very strongly turbulent flow ($U = 65\,\text{ms}^{-1}$). For each delay time $t_i$, a set of 4000 (single shot) images are acquired. Since we will be pushing the possibilities of the technique to its maximum when measuring at very long delays, we have taken considerable care to write lines of as high quality as possible. The write laser was
optimally tuned, so that the shot-to-shot line intensity fluctuation was minimal and (nearly) every laser shot displayed a narrow-band spectrum. The readout laser was not focused, in order to capture every displaced molecular tracer line. The quality of the line set is demonstrated in Fig. 7.7 and Fig. 7.8. In order to check if our line data is suited for line stretching measurements, velocimetry must come out correctly as well. Therefore we plotted, respectively, the probability distribution functions (Fig. 7.7) and structure functions (Fig. 7.8) for 2 relatively large delay-times, $\Delta t = 20 \mu s (1.5 \tau_\eta)$ and $\Delta t = 30 \mu s (2.3 \tau_\eta)$.

Both graphs show that velocimetry still comes out reasonably well for these long times. We do see that the histograms in Fig. 7.7 are not fully symmetrical at these time scales, and that the asymmetry is stronger for $\Delta t = 2.3 \tau_\eta$. This can be attributed to the gradient in the camera sensitivity. For lower velocities, the lines are located in the lower part of the image, where the camera sensitivity is lowest. Located there, lines are least intense and are more likely to be removed based on the minimal line intensity criterion. For longer delays the separation between “slow” and “fast” lines is greater, hence the larger asymmetry in Fig. 7.7b compared to Fig. 7.7a.

In the structure functions in Fig. 7.8 we do see a somewhat different scaling behaviour at the small scales; the difference in slopes being the greatest for the largest time delay. However, this happens on the same scales where all of our measurements are influenced by diffusion, photon noise, etc. We have already attributed this to different sources of errors and ambiguity in Chapter 5. Most important is that for
7.3 Measurements

![Figure 7.8: Structure functions (Eq. 6.1) for long time delays: a) $\Delta t = 1.5 \tau_\eta$ and b) $\Delta t = 2.3 \tau_\eta$. The orders shown correspond, from bottom to top, to $p = 2 \ldots 8$.](image)

the largest scales it can be seen that the structure functions come out right, and that they are similar to those discussed in section 6.3.2. With this information we are confident that even for these delays, that are at the edge of what is possible to track, the data is still qualitatively correct. However, we have seen that care must be taken in interpreting line stretching at the smallest scales, or short ruler lengths.

### 7.3.2 Stretching rate dependence on time and ruler length

In Fig. 7.9 we have plotted the stretching rate of lines as a function of time. For each of the lines the full length is calculated as the sum of all line elements using Eq. 7.8 with a fixed ruler length $r_0(k) = 2^k$, $k = 2 \ldots 8$. Individual line lengths are now averaged for every write-read delay and from the average line length evolution in time the stretching rate $\gamma$ is calculated using Eq. 7.10. From Fig. 7.9 it is noticed that the stretching rate increases with time. The trend of the time evolution of $\gamma$ will be discussed in Sect. 7.4.2. We also see that the growth-rate decreases with increasing ruler size, but the trend in time remains similar, except for the very shortest ruler lengths. Since the stretching rate using ruler lengths of 1 and 2 pixels is dominated by noise, we are not considering them. For $r_0 = 2^2$ the evolution of the stretching rate is still by no means smooth. For these rulers the effect of errors in the line fits due to photon noise or otherwise, will be significant, especially when the time delay is short, resulting in a small flow-induced line displacement relative to noise-induced displacement. This is especially apparent in the sharp peak at $t = 0.6 \tau_\eta$ for $r_0 = 2^2$. It is striking, that for the longest times ($t \geq 2.5 \tau_\eta$) the stretching rate decreases.
Figure 7.9: Stretch-rate $\bar{\gamma}$ as a function of time for increasing ruler-length: $r_0 = 2^k$ pixels with $k = 2...8$ or $r_0/\eta = 2.0$, 3.9, 7.9, 15.7, 31.5, 63, 126, as indicated on the right.

linearly for exponentially increasing ruler length from $r_0 = 2^4$ and up.

This logarithmic dependency of the stretching rate on the ruler length is shown explicitly in Fig. 7.10. Turning back to Eq. 7.5 and realizing that the stretching rate involves the logarithm of the line length, we see that the approximate lin-log dependence in Fig. 7.10 reflects the fractal character of the stretched line, with the slope of the lines equal to $1 - d_f$. Apparently, the fractal dimension $d_f$ increases with increasing delay times, and thus the simple description in Eq. 7.5 does not fully describe the fractal scaling. In Section 7.4 we will compare the behaviour observed here with theoretical work that does include time dependency as well as numerical simulations. It is apparent that for short rulers, the stretch rate is severely influenced by noise.

### 7.3.3 Non-trivial behaviour of the stretching rate

Recall that we discussed in Section 7.2.5 that for short time delays in our experiment (up till a few Kolmogorov times $\tau_\eta$), an Eulerian interpretation of the tagged molecule displacement may still be possible. This interpretation may result in an accurate prediction of the line length as a function of time and rulersize, based on the second order structure function alone. We will now see if there is any new information to be found in line stretching.

Recall that the relation that was made in Eq. 7.17 is only justified when $L(t) \approx L(0)$, while approximation Eq. 7.18 is only justified when the local slopes of the lines
7.3 Measurements

Figure 7.10: Stretch-rate $\tilde{\gamma}$ of lines in turbulence as a function of ruler length for increasing delays: From bottom to top: $t/\tau_\eta = 0.8, 1.1, 1.5, 1.9, 2.3, 2.7$.

are small.

Effectively, these slopes will decrease if they are measured with longer rulers. Therefore, the interpretation of the stretching rate in terms of the second order structure function is expected to break down for long times and short rulers. In Fig. 7.11 we plotted the stretching rate derived from the line lengths calculated using Eq. 7.18. Since we extract velocities from displacements, it is important to also compute the structure functions at a given time delay from the observed displacement.

When comparing this graph with Fig. 7.9, the breakdown of the assumptions linking $G_2$ to the stretching rate can be seen: for most ruler sizes, line lengths obtained through the second order structure function are overestimated for $t/\tau_\eta < 2$. For large rulers ($k = 6 \ldots 8$) the directly and indirectly calculated stretching rates are nearly corresponding.

Another interesting point is that the Eulerian interpretation 7.19 predicts a quadratic time-dependence of the logarithm of the line length, while the experiments point to a saturation of $\tilde{\gamma}$ (i.e. a linear time-dependence). The conclusion is that the stretching rate offers new information, which just starts to show at the delay times accessible in this experiment.
7 Line stretching

![Graph showing stretch-rate as calculated from the second order structure functions for increasing ruler-lengths: $r_0 = 2^k$ pixels with $k = 2 \ldots 8$ as indicated on the right.]

7.4 Interpretation

7.4.1 Line stretching as a multiplicative process

Theory

An early attempt to measure the shape of material lines in turbulence is reported by Villermaux and Gagne [85]. Since their approach is different from the one that is followed here, we will restate their results in the context of our experiments. In their paper, Villermaux and Gagne looked at the deformation of the leading edge of smoke ribbons released in a very weakly turbulent wind-tunnel flow. The Reynolds numbers of the turbulence used were very small, $Re_\lambda = 12 \ldots 21$. In fact, they were much smaller than the ones where the flow can be considered as being fully developed [96].

The authors consider line stretching as a multiplicative inertial range process. It is assumed that length increase will only occur longitudinally so that two points that are initially separated by $r = r_0$ have a mutual distance after a time $\tau$ given by:

$$r(\tau) = r_0 + \delta u(r_0)\tau,$$

(7.20)

where $\delta u$ is the RMS velocity difference of the two points, along their axis of separation. If an initially straight line has a length $L_0$, the number of rulers $r_0$ to cover the line after one time-step $\tau$ is, on average,
7.4 Interpretation

\[ N_\tau = \frac{\langle L(\tau) \rangle}{r_0} = L_0 \left( \frac{r_0 + \delta u(r_0) \tau}{r_0} \right) \tau. \]  

(7.21)

After a time \( t \) this process has repeated \( t/\tau \) times. We set the time-step to the Kolmogorov time \( \tau = \tau_\eta \) and the resulting line length is,

\[ L_{r_0}(t) = N(t)r_0 = L_0 \left( 1 + \frac{\delta u(r_0)}{r_0} \right)^{t/\tau_\eta}, \]

(7.22)

In [85] the ensemble average is assumed to be given by

\[ \frac{\langle L_{r_0}(t) \rangle}{L_0} = \left( 1 + \frac{\delta u(r_0)}{r_0} \right)^{t/\tau_\eta}, \]

(7.23)

which gives for the stretching rate

\[ \tilde{\gamma} = \ln \left( 1 + \frac{\delta u(r_0)}{r_0} \right). \]

(7.24)

Since below the Kolmogorov length lines are smooth, all contributions to the stretching rate are found in the scales \( > \eta \). Thus, in Eq. 7.24 the maximum stretching rate is reached when \( r_0 = \eta \). We then approximate the velocity increments with the Kolmogorov velocity, where we interpret the average as root mean square:

\[ \langle \delta u^2(\eta) \rangle^{\frac{1}{2}} = \left( \frac{1}{15} \frac{\eta^2}{\nu} \right)^{\frac{3}{2}} = \left( \frac{1}{15} \frac{\eta^2}{\tau^2} \right)^{\frac{3}{2}}. \]

(7.25)

In line with Eq. 7.20 we have considered longitudinal increments only\(^1\), in which case we find for the stretching rate

\[ \tilde{\gamma} = \ln \left( 1 + \left( \frac{1}{15} \right)^{\frac{3}{2}} \right) \approx 0.23. \]

(7.26)

As we have seen in Sect. 7.3 and will be further explained in Sect. 7.4.2 the true stretching rate shows a transient behaviour, and reaches its asymptote only after several Kolmogorov times. The result from Eq. 7.26 gives us merely an estimation of the asymptote and we miss the initial growth of \( \tilde{\gamma} \). According to K41 [46] the longitudinal velocity scales with distance as \( \delta u(r_0) \propto (\epsilon r)^{1/3} \), so that in the right hand side of Eq. 7.22 the dependence on the ruler length \( r_0 \) is

\[ \left( \frac{\delta u(r_0)}{r_0} \right) \propto r_0^{-\frac{2}{3}}. \]

(7.27)

\(^1\)If we were to additionally consider both transverse separations for which equal velocity increments are assumed, this would increase the stretching rate to 0.30.
This means that the stretching rate \( \tilde{\gamma} \) decreases with increasing \( r_0 \). It also implies that the dependence of \( N_t (r_0) \) on \( r_0 \) in Eq. 7.21 does not exhibit the power law \( N_t (r_0) \sim r_0^{-d_f(t)} \) that is associated with fractal scaling. However, Villermaux and Gagne [85] state that, when including a term to describe the viscosity-dominated regime, the velocity difference can be described as

\[
\delta u(r_0) = f(r_0/\eta)(r_0)^{1/3},
\]

with \( f \) a crossover function, as first introduced by Benzi et al. [97, 98]. Villermaux and Gagne have introduced an approximation to this function \( f \) which has the property that \( N_t (r_0) \) now does show a “quasi power law dependence” [85] on \( r_0 \), and as a result the fractal dimension can be calculated as

\[
d_f = 1 + \Delta d_f \frac{t}{\tau_\eta},
\]

where \( \Delta d_f \approx 0.10 \) the increment of the fractal dimension in time. All the Reynolds number dependence is now contained in \( \tau_\eta \).

Let us finally recapitulate the consequences for the line length. The combination of Eqs. 7.5 and 7.29 yields

\[
L_{r_0}(t) = L_\eta(t) \left( \frac{r_0}{\eta} \right)^{-0.10 \frac{\Delta}{\tau_\eta}},
\]

For ruler length \( \eta \) all line wrinkles are resolved and thus the line length \( L_\eta(t) \) is the full, ruler size independent, line length. The stretching rate of this line was already described by Eq. 7.1. It then follows that

\[
L_{r_0}(t) = L_0 e^{\tilde{\gamma}_{r_0}(t) \frac{t}{\tau_\eta}}.
\]

with

\[
\tilde{\gamma}_{r_0}(t) = \tilde{\gamma}_\eta(t) - \Delta d_f \ln \left( \frac{r_0}{\eta} \right),
\]

where \( \tilde{\gamma}_\eta(t) \) is the “true” stretching rate and the second term constitutes the influence of usage of rulers that do not fully resolve all line wrinkles. As stated above Villermaux and Gage have found a value for the fractal dimension of \( \Delta d_f = 0.10 \). We will now see how this compares with our simulations and measurements.

**Comparison with simulations and measurements**

Since we feel that the step from non-algebraic length increase towards an algebraic one may be arguable, we want to ascertain if numerical simulations support the relation of Eq. 7.32 and show likeness to the APART data. We will use kinematic
7.4 Interpretation

Figure 7.12: Stretch-rate $\tilde{\gamma}$ as a function of time for increasing ruler-lengths in kinematic simulations. From top to bottom: $r_0 = 2^k$ with $k = 0 \ldots 8$ or $r_0/\eta = 0.02 \ldots 20.5$. Plot a) shows the stretch-rate in 3D, plot b) shows the fitted 2D projection.

Simulations to calculate the line formation. The lines were described as strings of particles, where we initially spaced the particles by distance $r_0$. For each of these “ruler lengths” $r_0$, 4000 instances of the line are followed during evolution, and at fixed times snapshots are made. This is done for different ruler lengths.

Fig. 7.12a shows the stretching rate of three-dimensional lines versus time calculated according to Eq. 7.2. The simulations show similar features as the measurements depicted in Fig. 7.9: the growth rate increases with time, and increases with decreasing ruler size $r_0$. Like in the APART data of Fig. 7.9, it is clear that for exponentially increasing ruler lengths the stretching rate decreases approximately linearly (when $r_0 > 2^3$ and $t/\tau_\eta > 6$). A striking feature in the simulations is that we can actually find negative stretching rates when points on the line are separated far enough. Fig 7.12b is based on the same lines, but now the line length of the 2D projections of the 3D lines are used. As seen before, the stretching rate in three dimensions is half of that of the full 3D lines (for $\gamma > 0$).

Let us see how the stretching rate changes with ruler size in these simulations. The stretching rate $\tilde{\gamma}$ is plotted as a function of $\ln r_0$ in Fig. 7.13a for three dimensions and in Fig. 7.13b for the two-dimensional projection. For larger separations the relation between the stretching rate and ruler size is, indeed, again exponential.

It is found from fitting the slopes in this figure that for increasing delay times (where a multiplicative process is expected to work), the fractal dimension increment asymptotes to $\Delta d_f = 0.020$ in 3D and $\Delta d_f = 0.012$ in 2D. Furthermore, the stretching rate saturates for ruler lengths smaller than $\eta$, as is expected for fractal
7 Line stretching

Figure 7.13: Stretch-rate $\tilde{\gamma}$ of lines in turbulence as a function of ruler-lengths in kinematic simulations. Plot a) shows the stretch-rate in 3D, b) shows the fitted 2D projection. The different lines show the decreasing slopes with increasing times ($t/\tau_\eta = 2 \ldots 10$). Note the logarithmic abscissa.

scaling with a cutoff length. This is easily understood, since no smaller scales of turbulence exist that can wrinkle the line. We found values of $\tilde{\gamma}(<\eta) = 0.11$ and $\tilde{\gamma}(<\eta) = 0.06$ for 3D and 2D, respectively.

This effect is not seen in the measurements, because there photon noise will continue to contribute to the growth rate even below the smallest scales, where no turbulence is present.

Although our numerical simulations are relatively simple, they do expose the fractal nature of deforming lines in turbulence, and corroborate the relation in Eq. 7.32.

Now, having seen that the time dependency of $d_f$ can be described by Eq. 7.32 (albeit with a very different value of $d_f$ as used by [85]), let us return to the APART data. By fitting the slopes shown in Fig. 7.10 we can find values for the maximal stretching rate (the stretching rate at the Kolmogorov scale $\tilde{\gamma}(\eta) \equiv \tilde{\gamma}_0$) and the fractal dimension increment $\Delta d_f$. As we discussed, the stretch rate could not be directly read from Fig. 7.9, since for small separations, the results are severely influenced by artifacts. However, by using our knowledge of the fractal scaling behaviour, we can extrapolate $\gamma$ from the larger scales. The result of the exercise is shown in Fig. 7.14a and Fig. 7.14b. We had already seen that the rate of stretching changes logarithmically with the ruler size and also saw that there is an additional time dependency present.
7.4 Interpretation

Figure 7.14: The maximum stretching rate $\gamma_0$ and fractal dimension $\Delta d_f$ as a function of time, as fitted from experimental data shown in Fig. 7.10. The horizontal lines display the maximum values $\tilde{\gamma}(\eta) = 0.085$ and $\Delta d_f = 0.017$, respectively.

In the next section we will discuss whether the maximum stretch rate $\tilde{\gamma}(\eta) = 0.085$ found in Fig. 7.14a already constitutes the asymptotic value, or that longer times should be explored. The fractal dimension was not expected to change with time delay, and the dependency found is believed to be related to the decreasing role of noise induced line length increase. The plot of $d_f$ seems to asymptote in Fig. 7.14b at a value of the fractal dimension increment of $\Delta d_f = 0.017$. This is very comparable to the value that we have found in numerical simulations. However, both values are more than a factor of 5 less than those found in experiments by [85]. It should be noted that there is a significant difference between our measurements and those by Villermaux and Gagne: the smoke line measurements were done at very low Reynolds numbers. It can be that this makes it easier to experimentally resolve all wrinkles. Also, the growth rate was a priori fixed at $\tilde{\gamma}(\eta) = 0.31$. It can be that setting the stretching rate to a fixed value resulted in finding a higher fractal dimension.

The stretch rate derived from our APART measurements is close to that found using kinematic simulations. However, as stated before the simulations should be considered with some caution. They are good for predicting trends, but due to the ambiguity in the determination of $\epsilon$ and thereby the Kolmogorov time, the absolute value of the stretching rate can not be confidently determined.
7.4.2 Stretching mechanism

Theory

Both in measurements and simulations we already noticed some transient behaviour in the stretching rate that we will now investigate further. In order to better observe any time dependency, we use the derivative stretching rate as introduced in Eq. 7.2. In Fig. 7.15 we show the typical behaviour of the stretching rate, where the derivative stretching rate $\dot{\gamma}$ (solid line) and the mean stretching rate $\bar{\gamma}$ (dotted line) are both plotted. This data was obtained by extensive direct numerical simulations on deforming lines performed by Goto and Kida [88, 89]. The trend in the rate of growth is easily discerned. Starting at 0, the stretching rate quickly increases within the first Kolmogorov time. Similar to a slightly underdamped oscillator, the stretching rate shows a single overshoot between $t = 1 \tau_\eta$ and $t = 5 \tau_\eta$ and reaches an asymptotic value of $\gamma = 0.17$ after approximately $t = 10 \tau_\eta$.

We can understand the transient by considering the interaction of individual line elements within the flow. Initially, all line elements that make up the line, are randomly oriented with respect to the eigenvector with largest eigenvalue of the local strain tensor. In time, line-elements will start to orient along the axis of maximum strain. The local vorticity, on the other hand, will tend to rotate line-elements away from the maximum strain, thereby decreasing the average stretching rate and, under specific circumstances, resulting in an overshoot.

This qualitative argument for the time dependence of the stretching rate can
be translated into a simple analytical model, which we have taken from [77]. Let us consider a material line element $\vec{e}$ that evolves according to the local velocity gradient:

$$\frac{de_i}{dt} = \sum_{j=1}^{3} \frac{\partial u_i}{\partial x_j} e_j.$$  \hfill (7.33)

The velocity gradient can be decomposed into strain and rotation as

$$\frac{\partial u_i}{\partial x_j} = S_{ij} + r_{ij},$$  \hfill (7.34)

with $\mathbf{S}$ the strain rate tensor $S_{ij} = (\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i})$ and $\mathbf{R}$ the rotation tensor $r_{ij} = \frac{1}{2} \epsilon_{ijk} \omega_k$ with $\vec{\omega}$ the vorticity and $\epsilon$ the third order isotropic tensor. Let $a_1$, $a_2$ and $a_3$ be the principal values of the symmetrical tensor $S_{ij}$, ordered such that $a_1 \geq a_2 \geq a_3$. Due to incompressibility we have $a_3 \leq 0$, but henceforth we will use $a_3 \equiv |a_3|$. We will now consider a simplified, two-dimensional model that describes the evolution of a line element perpendicular to the intermediate axis of strain, that is, the intermediate eigenvector $s_2$ (with eigenvalue $a_2$) of $S_{ij}$. This model is illustrated in Fig. 7.16a.

The Cartesian components of line element $\vec{e}$ then evolve according to

$$\frac{dy}{dt} = a_1 y - r_{13} x,$$

$$\frac{dx}{dt} = -a_3 x + r_{13} y,$$

with the substitutions

$$x(t) = r(t) \sin \alpha(t),$$

$$y(t) = \frac{r(t)}{\sin \alpha(t)}.$$

Figure 7.16: Orientation of line elements due to strain and rotation. The left image shows line elements randomly oriented with respect to the axis of maximum strain $s_1$ and the axis of minimum strain $s_3$. The middle image shows that if only strain is present, all elements will orientate toward the local axis of maximum strain. In the right image we see that in the presence of rotation, the lines are drawn away from the maximum strain axis.
7 Line stretching

\[ y(t) = r(t) \cos \alpha(t). \]  \hspace{1cm} (7.37)

Eq. 7.35 separates in an equation for \( r(t) \) and \( \alpha(t) \), from which the stretching rate follows as (Appendix B)

\[ \gamma(t) = \frac{d \ln r}{dt} = a_1 - (a_1 + a_3) \cos^2 \alpha(t), \]  \hspace{1cm} (7.38)

with \( \alpha(t) \) given by

\[ \tan \alpha(t) = \left( \frac{C e^{-2\delta t} + 1}{C e^{-2\delta t} - 1} \right) \delta - \rho, \]  \hspace{1cm} (7.39)

where \( \delta = (\rho^2 - 1)^{1/2} \), \( \rho = \frac{a_1 + a_3}{2r_{13}} \) and \( C \) a constant such that \( \alpha(0) \) is the initial angle of the line element. For \( \rho \leq 1 \) the angle of the line elements will display a periodic behaviour and they will not contribute to the transient peak in the stretching rate. Only when \( \rho > 1 \), that is, when the rates of strain are larger than the rotational effect: \( a_1 + a_3 > 2r_{13} \), the line element will contribute to the transient growth peak and finally reach a steady state. It can be shown that the asymptotic angle \( \alpha(\infty) \) is obtained by solving \( \sin 2\alpha = \frac{1}{\rho} \) (see appendix B). Plugging in the resulting expression for \( \cos^2 \alpha(\infty) \) in Eq. 7.38 results in

\[ \gamma(t) = a_1 - (a_1 + a_3) \frac{1 - \sqrt{1 - 1/\rho^2}}{2}. \]  \hspace{1cm} (7.40)

The type of transient behaviour depends on the combination (\( \rho \)) of our model parameters, but the precise stretching rate \( \gamma(t) \) depends on all three of them. Since these parameters take on random values in turbulence, one wonders how this model survives averaging. In Pope & Girimaji [77] the average over the initial orientation (where \( \alpha(0) \) has a spherically symmetric distribution) is discussed, under which the transients survive. However, no further averaging (such as the lengths of the different vectors) has been considered.

Therefore, there is not a simple way to parametrize measured stretching rates from data at small time scales, that is, predicting the asymptote \( \gamma(\infty) \), the position of the bump and the shape of the curve.

Comparison with DNS

Detailed investigations of the stretching of lines have been performed using direct numerical simulations by Goto and Kida [76]. In this article the point is made that a significant difference exists between calculating the mean stretching rate of uncorrelated line elements in a flow-field and calculating the evolution of the line-elements of a true line within the flow.

The argument for this is that, contrary to the Bachelor assumption, the statistics of material lines/surfaces are not the same as those of infinitesimally thin ones. The
line average of a quantity $g$ along a material line defined by a line integral can be approximated by

$$\langle g \rangle_{\text{line}} = \frac{\sum_{i=1}^{I} g^{(i)}(t) l^{(i)}(t)}{\sum_{i=1}^{I} l^{(i)}(t)}, \quad (7.41)$$

where $I$ is the number of line elements. Bachelor and Townsend [78] assumed that, in stationary homogeneous turbulence, all line elements are statistically equivalent and $\langle g \rangle_{\text{line}}$ can be approximated by the line-element average,

$$\langle g \rangle_{\text{line-element}} = \frac{1}{I} \sum_{i=1}^{I} g^{(i)}(t). \quad (7.42)$$

If this were true, the line stretching rate would trivially be related to the growth of small line-elements averaged over the flow, and it would no longer be necessary to follow real material lines. That this is not the case, and that Bachelor's argument is essentially flawed, can be appreciated by rewriting Eq. 7.41 as

$$\langle g \rangle_{\text{line}} = \frac{\langle g l \rangle_{\text{line-element}}}{\langle l \rangle_{\text{line-element}}}. \quad (7.43)$$

Comparison of Eq. 7.43 with Eq. 7.42 shows that the line average and line-element average are only identical if $g$ and $l$ are statistically independent of each other.

This is clearly not true for the stretching rate, as the local shape and the stretching rate are directly related. This argument is further explored in Appendix A. The stretching rate as computed by Kida and Goto [88] was already shown in Fig. 7.15, in addition we have calculated the corresponding values of $\tilde{\gamma}$, as given in Fig. 7.15 as well.

It seems that the data is in agreement with the model: the stretching rate exhibits transient behaviour and the competition between rotation and strain indeed results in a clear overshoot at approximately $2 \tau_{\eta}$. We can now ask ourselves how this corresponds with our kinematic simulations. The results are shown in Fig. 7.17 where we have plotted, aside from the integral stretching rate $\tilde{\gamma}$, also the derivative stretching rate $\gamma$. This yields a surprising result: the derivative stretching rate also exhibits an overshoot, albeit not so pronounced as in the DNS data. This was not foreseen, since the simulations do not explicitly define coherent structures. Apparently, the random interaction between the wave modes leads to an average of $\gamma(t)$ over many $r_{13}$, $a_1$ and $a_3$ values, with their respective orientations so that $\rho > 1$.

Because the overshoot is less well-defined, it is hard to compare a maximum in stretching rate with the one found in the DNS data. We do see that the model and kinematic simulations both predict an overshoot and a (suspected) asymptotic stretching rate. In the DNS data this asymptote is found beyond approximately $9 \tau_{\eta}$, but in the kinematic simulation the stretching rate is still decreasing at $t = 10 \tau_{\eta}$. This difference may be (partly) accounted to the less than well-defined $\tau_{\eta}$ in kinematic simulations.
This also means that it is likely that we have not yet found the asymptote of the growth-rate in our measurements. It will prove difficult to increase the read-write delays. Aside from any experimental limitations, the influence of diffusion in smearing out small-scale line wrinkles will also increase. The found value of \( \bar{\gamma}(r = \eta, t = 2.5\tau_\eta) = 0.10 \) should be considered a lower limit.

### 7.5 Conclusions

In this chapter measurements of the growth rate of material lines are presented. It was shown that from the measured two-dimensional lines within a bounded region, the true stretching rate of three-dimensional, unbounded, lines can be inferred. The measurements show that there is a definite time dependency in the stretching rate, and that the growth rate is sensitive to the chosen length of the rulers used to determine the line length.

This ruler dependency can be satisfactorily described by assigning a fractal dimension of \( \Delta d_f = 0.017 \) to the line. This value is comparable to our kinematic simulations (\( \Delta d_f = 0.020 \)), but considerably lower than the value (indirectly) derived in [85] of \( \Delta d_f = 0.10 \). However, it should be taken into consideration that there is a significant difference between our APART measurements and the smoke line measurements by Villermaux and Gagne [85]. The latter were done at very low Reynolds numbers and the growth rate was a priori fixed at \( \bar{\gamma}(\eta) = 0.31 \). This value
is very much different from what is found from our measurements: \( \bar{\gamma}(\eta) = 0.085 \). Although our experimentally found stretching rate is almost comparable with our kinematic simulations \( \bar{\gamma}(\eta) = 0.11 \), it does not agree well with the very careful DNS measurements by Goto and Kida, who found a stretching rate of \( \bar{\gamma}(\eta) = 0.17 \). We believe that we need to be able to resolve longer delays in order to find the stretching rate asymptote.

The time dependency of the growth-rate can be explained by the competition between strain and rotation as proposed by Girimaji and Pope [77]. The stretching transients described in the model are seen in full direct numerical simulations as well as our kinematic simulations. Unfortunately, the APART data is not sufficiently accurate for finding the derivative stretching rate, so recovery of the overshoot is not possible. We do see a pronounced peak at \( t = 0.2 \tau_\eta \) in Fig. 7.9, but this is likely to be attributable to measurement artifacts rather than a real turbulence feature.

Finally, we have shown that line stretching results are not trivial, in the sense that they can be derived from the second order structure function, even for relatively short times \( < \tau_\eta \).
7 Line stretching
8 Outlook

8.1 Introduction

In this thesis a new scheme of molecular tagging velocimetry (MTV) based on the creation of nitric oxide molecules, was introduced as a tool for the study of turbulence. It has been a goal to develop this scheme into a measuring technique and to see how far the technique could be extended in terms of accuracy. A second goal was to investigate how far the unique features of this technique could be used to measure flow properties that have been hard or impossible to measure using conventional techniques.

Although multiple other MTV techniques have been developed within the last decades, only few have really been used to extensively investigate fluid flows. In fact, only one other scheme, RELIEF, based on the creation of excited oxygen as tracer molecules, was used to study turbulent flows.

It is our belief that in this thesis work advances have been made (both practical and theoretical) that will help to turn APART into a usable technique in flow measurement, and that this work has resulted in more insight into specific advantages and disadvantages.

However, we also believe this work has left potential improvements as well as applications still untouched. In this outlook we will briefly discuss how we think that APART can be further developed into a mature and easy-to-use measuring technique, and what kind of experiments may be interesting to conduct in the future.

8.2 Experimental setup

Currently, the measuring setup is large and complex. Two lasers need to be used, and both are expensive and complicated to handle and maintain. If an alternative MTV scheme could be developed for which only mainstream lasers (like the Nd:YAG laser) would be required to tag and visualize molecules, the technique would become much more accessible to “normal” fluid mechanics laboratories, like PIV and LDA have. Research into such alternatives is, in fact, already ongoing in our laboratory [58]. One approach involves the creation of a tracer molecule that can both be written and read by Nd:YAG lasers. Another approach focuses on phosphorescent tracer molecules, that require only a single laser for the whole process.
8.3 Diffusion

One of the main topics of this thesis is the interaction between diffusion and turbulence. It is interesting to observe how these two processes interact: one occurs at the molecular level and the other on a macroscopic level, but the effects on a molecular tracer distribution are nevertheless almost equal in magnitude. We have discussed the effect on energy spectra and structure functions, and it may be interesting to investigate how this interaction will also affect other statistical properties.

On the other hand, for some applications it will be more interesting to explore how the effect of diffusion may be decreased, as diffusion makes measuring detailed flow properties much more difficult. Unfortunately, as the argument posed in Section 2.4 indicates, increasing the spatial scale of the experiment will not help to reduce the relative effect of diffusion if the Schmidt number remains near unity. A worthwhile path to pursue is increasing the weight of tracer molecules. We have shown that heavier molecules will reduce the effect of diffusion without losing the ability to follow local accelerations in the flow. This does, however, mean that the flow will have to be seeded, but gas phase molecular seeding does not perturb the flow, and when a molecular species is used that can be efficiently tagged and visualised, the amount of seeding particles will not be a serious impediment.

A main challenge will be to find suitable molecules that on the one hand are heavy enough to limit the importance of diffusion, but still can efficiently be brought into the gas phase. One of the possible candidates on which research has already started in our group [58] involves lanthanide chelates. These complexes, with molecular weight of typically over 200, are about ten times heavier than NO, and thus have diffusion rates that are about an order of magnitude lower. The specific case of \( \text{Tb}^{3+}-\text{dipicolinic acid} \) has already been demonstrated as a tracer molecule in liquids [99], and it was noted that this phosphorescent molecule has a relatively long lifetime of \( \approx 1 \text{ ms} \) and a high phosphorescence efficiency.

8.4 Increased time delays

An example of an experiment that will benefit from a lowered diffusion rate is tracking the stretching and deformation of material lines in a flow. Line stretching is a turbulence property that has proven to be very difficult to investigate experimentally at high Reynolds numbers, and we believe that MTV may be particularly suited for this task. Reduced diffusion, and the resulting diminished line blurring, will significantly increase the time over which line evolution can be tracked. Together with more advanced line tracing techniques this will make it possible to track line deformation over more than approximately ten Kolmogorov times and thereby enabling us to verify theory and simulations concerning surface deformation [77, 89].

Work is carried out by Verkuijlen [92] and others to implement an active contour plot algorithm to trace even the most strongly deformed lines. The algorithm
8.5 Multiple lines

One of the promises that APART holds, is the possibility to write multiple lines. The combination of horizontal and vertical lines will allow us to measure both velocity components in a crossing, as well as velocity derivatives. This gives us access to other flow properties such as vorticity. We could also perceive the line crossings as points written in the flow. If we are able to follow these points for sufficiently long times we can look at flow properties in a Lagrangian framework. This becomes even more interesting if we are able to make more than one crossing “point”: If we track the created points we can, for example, study particle pair separation [100]. In the analysis of written lines it has proven to be difficult to find and distinguish multiple lines in an image. Regardless of which line fitting algorithm is used, some a priori knowledge about the line orientation and position is needed. This knowledge could be provided through the Hough transform [101]. For successful line discrimination, it may be necessary for the written line to have sharply defined edges. To this end an edge filter, such as the canny filter [102], could be applied first.

Work on this subject is currently being performed. While it has proven to be difficult to write multiple lines and at the same time retaining sufficient energy per line to create NO, Pashtrapanska et al. [103] have recently been able to show (preliminary) results of images with two line crossings.

It is clear that this outlook does not give an exhaustive view on all the developments and possibilities of “Measuring Lines in Turbulence using APART”, but we hope it does give some indication of its potential.
A Thin and finite volume material lines

A convenient manner to derive statistics of material lines is just to observe randomly positioned line elements instead of laboriously following the line elements within a true line. On first sight, this should not pose a problem, since the flow is statistically equivalent at all the locations of the line elements. However, it has been argued that this will introduce a form of bias, and in the case of the growth rate, result in a significant underestimation. In order to understand this, we will have a look at the difference between the line average of a physical property $g$ and the thin limit of the volume average. The volume average of the property $g(x, t)$ over a line is expressed as:

$$\langle g \rangle_{\text{line}} = \lim_{\text{dia}(V_t) \to 0} \frac{1}{V_t} \int_{V_t} F(x, t) \, dV$$  \hspace{1cm} (A.1)

whereas the volume average in the thin limit is found by integration over a blob that, at a time $t$, is shaped as $V_t$:

$$\langle g \rangle_{\text{tube}} = \lim_{\text{dia}(V_0) \to 0} \frac{1}{V_t} \int_{V_t} F(x, t) \, dV,$$  \hspace{1cm} (A.2)

where integration is carried out over line $L_t$. Although both descriptions seem very similar, the resulting value can differ significantly for $t > t_0$:

$$\langle g \rangle_{\text{line}} \neq \langle g \rangle_{\text{tube}}.$$  \hspace{1cm} (A.3)

The difference lies in the property that the local beam waist remains constant (and therefore uniform) for the line average and will change for the thin tube average. This difference is exemplified in Fig. A.1. Here a uniform line is defined at an initial time $t_0$. This line will stretch and deform in time. Note that, although the flow is taken to be incompressible in 3 dimensions, the 1-dimensional line is not incompressible. The uniform tube is thus advected by an incompressible flow into a deformed tube $V_t$ with non-uniform cross-section. This is reflected in the particle distribution that makes up the line: the particles that where initially placed at equal distance now have been redistributed non-uniformly along the line. This means that the values of property $g$ in the stretched part of the line will be represented less in the thin tube average. In Fig. A.1 particles are distributed uniformly along the line, equivalent to the representation of a line average. Given a sufficiently large number of particles [76] the contribution of the nonuniform particle distribution can be calculated by
A Thin and finite volume material lines

Figure A.1: Nonuniform line stretching. The upper figure shows the initial line, undeformed and with uniform cross-section. The center figure shows a deformed line at a certain time \( t \). This line has a nonuniform volume and corresponds to a line average. The lower figure shows a new uniform tube around the same centerline as the middle one, corresponding to a volume average.

weighing the line property \( g \) with a factor proportional to the distance between neighbouring particles.

Let us rephrase this argument in a way that corresponds better with our form of measurements: the line average of a quantity \( g \) along a material line defined by a line integral can be approximated by

\[
\langle g \rangle_{\text{line}} = \sum_{i=1}^{I} g^{(i)}(t) l^{(i)}(t) / \sum_{i=1}^{I} l^{(i)}(t),
\]

where \( I \) is the number of line elements. Batchelor [78] now assumed that, in stationary homogeneous turbulence, all line elements are statistically equivalent and \( \langle g \rangle_{\text{line}} \) could be approximated by the line-element average

\[
\langle g \rangle_{\text{line-element}} = \frac{1}{I} \sum_{i=1}^{I} g^{(i)}(t).
\]

This assumption makes the study of material lines in turbulence drastically more simple, since it becomes unnecessary to follow the evolution of real material lines. Eq. A.4 can also be written as:

\[
\langle g \rangle_{\text{line}} = \frac{\langle g l \rangle_{\text{line-element}}}{\langle l \rangle_{\text{line-element}}},
\]

144
Comparison of Eq. 7.43 with Eq. 7.42 shows that the line average and line-element average are only identical if \( g \) and \( l \) are statistically independent of each other.

It is obvious that the line average and line-element average are only identical if \( g \) and \( l \) are independent of each other and it follows that the observed variable may differ between tracking these uncorrelated line elements rather than tracking the evolution of true lines. In this light, let us again look at line property \( g \): the stretching rate \( \gamma \). The total length of the line is expressed as Eq. B.1. The stretching rate can be calculated for every line element as \( \gamma_t^{(j)} = \frac{d \ln l_t^{(j)}}{dt} \tau_\eta \), so that the stretching rate of the complete line becomes

\[
\zeta_L = \sum_{i=1}^{I} \gamma_t^{(i)} l_t^{(i)}.
\]  

(A.7)

We thus see that the weight factor is the segment length \( l_t^{(j)} \). Numerical simulations by Goto et al. [76] have shown that the difference between the line average and the thin volume average is significant (\( \gamma = 0.17 \) for lines against \( \gamma = 0.14 \) for uncorrelated line elements).
A Thin and finite volume material lines
The stretching mechanism

A line will stretch and deform in time under influence of turbulent advection. The total length of the line is expressed as

\[ L_t = \sum_{j=1}^{l} l_t^{(j)}, \]  

where \( l_t^{(j)} \) is the length of line element \( j \) at time \( t \). The stretch-rate can then be calculated as

\[ L(t) = L_0 e^{\gamma t/\tau}, \]

where \( L_0 \) is the initial length. In order to get a qualitative understanding of the evolution of lines, let us consider a material line element \( \vec{e} \) that evolves according to the local velocity gradient:

\[ \frac{d\vec{e}_i}{dt} = \sum_{j=1}^{3} \frac{\partial u_i}{\partial x_j} \vec{e}_j. \]

It is observed in numerical simulations that the mean length over line elements \( e_j, \sum |e_j| \) does not increase uniformly in time. This transient growth is due to the competition between strain and rotation: strain tries to orient \( \vec{e} \) in the direction of fastest growth; rotation tries to direct \( \vec{e} \) away from this, resulting in a lower stretch-rate than when rotation would be absent. In order to understand this, we will analyse a simple model. We will follow Girimaji & Pope [77], but will indicate necessary extensions.

The velocity gradient can be decomposed into strain and rotation as

\[ \frac{\partial u_i}{\partial x_j} \vec{e}_j = S_{ij} + r_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) + \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} - \frac{\partial u_j}{\partial x_i} \right), \]

with \( S \) the strain rate tensor \( S_{ij} = \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \) and \( R \) the rotation tensor \( r_{ij} = \frac{1}{2} \epsilon_{ijk} \omega_k \) with \( \omega \) the vorticity and \( \epsilon \) the third order isotropic tensor. Let \( a_1, a_2 \) and \( a_3 \) be the principal values of the symmetrical tensor \( S_{ij} \), ordered such that \( a_1 \geq a_2 \geq a_3 \). Due to incompressibility we have \( a_3 \leq 0 \), but henceforth we will use \( a_3 \equiv |a_3| \). We will now consider a simplified, two-dimensional model that describes the evolution of a line element perpendicular to the intermediate axis of strain, that
Figure B.1: 2D Stretch rate model with rotation. The material line element $\vec{e}$ evolves according to the local strainrate and rotation $r_{13}$.

is, the intermediate eigenvector $s_2$ (with eigenvalue $a_2$) of $S_{ij}$. This is shown in Fig. B.1. The Cartesian components of $\vec{e}$ then evolve according to

$$\frac{dy}{dt} = a_1 y - r_{13} x,$$  \hspace{1cm} (B.5)  

$$\frac{dx}{dt} = -a_3 x + r_{13} y.$$  \hspace{1cm} (B.6)  

We solve Eq. B.5 separately for the angle $\alpha$ and the length $r$ of the line elements. With $\tan\alpha(t) = y(t)/x(t)$, we have

$$\frac{d}{dt} (\tan\alpha(t)) = \frac{1}{x} \frac{dy}{dt} - \frac{y}{x^2} \frac{dx}{dt},$$  \hspace{1cm} (B.7)  

$$= \frac{y}{x} (a_1 + a_3) - r_{13} \left(1 + \frac{y^2}{x^2}\right),$$  \hspace{1cm} (B.8)  

resulting in$^1$

$$\frac{d\alpha}{dt} = \frac{1}{2} (a_1 + a_3) \sin 2\alpha(t) - r_{13}$$  \hspace{1cm} (B.9)  

We will see later on how to reach an analytic solution for it. For the length $r^2 = x^2 + y^2$ of the line we use Eq. B.5:

$$y \frac{dy}{dt} = a_1 y^2 - r_{13} xy,$$  \hspace{1cm} (B.10)  

$$x \frac{dx}{dt} = -a_3 x^2 + r_{13} yx.$$  \hspace{1cm} (B.11)  

Adding these two equations then leads to

$$\frac{1}{2} \frac{dr^2}{dt} = r^2 \left(a_1 - (a_1 + a_3) \cos^2 \alpha(t)\right),$$  \hspace{1cm} (B.12)  

$^1$The parameter $\alpha$ in Eq. B.9 is equal to $\alpha = \frac{\pi}{2} - \Gamma$ as defined by Girimaji & Pope [77].
or
\[
\frac{1}{2} \frac{d \ln r}{dt} = \zeta(t) = a_1 - (a_1 + a_3) \cos^2 \alpha(t).
\] (B.13)

In the absence of rotation, $\alpha \to \pi/2$ and $r$ grows according to the largest eigenvalue $a_1$. This scenario is shown in Fig. B.1. However, if $r_{13} \neq 0$ and Eq. B.9 reads a stationary solution, the asymptotic growth rate will be smaller than $a_1$. If $d(t)$ is then (transiently) close to $\pi/2$, a transient peak in the growth rate is observed. Whether this is the case, depends on the type of solutions of Eq. B.9. Therefore we will briefly discuss these solutions. First notice that $r_{13}$ can be used to scale the time so that a single parameter $\rho$ results in
\[
\frac{d\alpha}{dt} = \rho \sin 2\alpha - 1,
\] (B.14)

with $\rho = \frac{a_1 + a_3}{2r_{13}}$. It is now useful to go back to the variable $y = \tan \alpha$, $\frac{dy}{dt} = (1 + \tan^2 \alpha)$, so that
\[
\frac{d}{dt} (\tan \alpha) = \rho (2 \sin \alpha \cos \alpha) \left(1 + \frac{\sin^2 \alpha}{\cos^2 \alpha}\right) + \left(1 + \frac{\sin^2 \alpha}{\cos^2 \alpha}\right),
\] (B.15)

and
\[
\frac{dy}{dt} = 2\rho y + (1 + y^2).
\] (B.16)

The right-hand side of Eq. B.16 can be written as
\[
(y + \rho - (\rho^2 - 1)^{1/2})(y + \rho + (\rho^2 - 1)^{1/2}).
\] (B.17)

Introducing $y' = y + \rho$, we now have
\[
\frac{dy}{dt} = (y - \delta)(y + \delta),
\] (B.18)

with $\delta = (\rho^2 - 1)^{1/2}$. This equation can readily be integrated to
\[
y(t) = \left(\frac{Ce^{-2\delta t} + 1}{Ce^{-2\delta t} - 1}\right) \delta.
\] (B.19)

If $\rho < 1$ we find no transient dwelling near $\alpha = \pi/2$, so only the case $\rho > 1$ will give a transient growth peak. For this case, Eq. B.14 gives the asymptotic orientation $\sin 2\alpha = \frac{1}{\rho}$. For the asymptotic growth rate, we need to solve for $\cos^2 \alpha$
\[
\cos^2 \alpha = \frac{1 \pm \sqrt{1 - 1/\rho^2}}{2},
\] (B.20)
where we select the $-$ sign as $\alpha = \pi/2$ for $\rho \to \infty$ ($r_{13} \to 0$) for the asymptotic growth rate it then follows from Eq. B.12.

\[
\gamma(\infty) = a_1 = (a_1 + a_3)\left(1 - \sqrt{1 - 1/\rho^2}\right).
\]  

(B.21)

Clearly, the shape of the growth function $\gamma(t)$ (or the actual line length $e^{\gamma(t)t}$) depends on all three parameters of our simple model. However, the form of the angle dependence $\alpha(t)$ depends on a single parameter $\rho$ (apart from the scaling of the time axis). Using the simple model we have identified three parameters that determine the shape of the average line length. It should also be realized that the growth rate from Eq. B.12 indicates the growth-rate for a single line element, but in order to arrive at a prediction for the full line growth rate we must average over all line elements. There is not a simple way to parametrize measured stretching rates from data at small timescales, that is, predicting the asymptotic $\gamma(\infty)$, the position of the bump and the shape of the curve.
Bibliography


M. Pashtrapanska. private communication, 2005.
Summary

From the 16th century, turbulence has remained a topic of continuous study, stemming both from academic and industrial interest. In this thesis a new scheme of Molecular Tagging Velocimetry (MTV) called APART (Air Photolysis And Recombination Tracking) for measuring this turbulence is introduced. In MTV a pulsed laser is used to ‘write’ a pattern of molecules in the flow field. After a set time delay the pattern, that has been altered by the flow field, is read back. The velocity field now follows from the displacement and the deformation of the pattern. In order to resolve the displacement of the pattern it is necessary to do careful image analysis. By employing a two-stage fitting technique the line center of all but the most deformed lines can be found.

Although several different schemes of creating and visualising molecular tracers have been developed, APART is one of the most promising. In APART tagging is done by photosynthesis of NO molecules out of $N_2$ and $O_2$ molecules in air. No additional seeding of air is needed to create these molecular tracers. Furthermore, NO is a stable molecule and is thus long-lived.

Although one may naively expect that, since APART employs extremely small tracers particles, that is molecules, this allows the resolution of even the smallest turbulence scales. However, we show that, since molecules reside at such different scales, they are governed by different mechanics, resulting in diffusion. For NO lines in air it is shown that, no matter how thin initially, these will broaden by thermal diffusion to the size of the Kolmogorov length within one Kolmogorov time, and will thus smear out turbulence on the smallest scale.

In order to compare APART with known flow properties, fully characterised turbulence is used. It was created by means of a free turbulent jet where, at approximately 45 nozzle diameters, $Re_\lambda \approx 460$. All macro scale parameters of the flow were determined and linked to the micro scale properties by the relation between the rms velocity $u_{rms}$ and the dissipation rate $\epsilon$. Histograms of the velocity indicate that the velocity distribution is almost perfectly Gaussian for all positions along the written line, even though other line properties such as intensity and width are position dependent.

Though APART is an unseeded optical technique, it can not be considered fully non-intrusive. That is, energy absorption along the path of the laser beam, mainly due to oxygen leads to local heating. Through different techniques (energy absorption, LIF spectroscopy and line width analysis) it is shown that the rise in temper-
ature within the first microseconds is approximately 400 K. Not only does the laser based heating increase the thermal diffusion coefficient, it also results in convection. These effects have been calculated and are shown to produce super-diffusive line widening. Indeed, this effect is observed in measurements. Another effect that was predicted and has been measured is that the diffusion constant $D$ does not go to zero for infinite pressure ($1/p \to 0$).

Since APART allows us to write lines and we can observe the displacement perpendicular to the line, we can measure true transverse velocities, and thereby, amongst others, energy spectra of transverse velocity increments. Whereas the measured velocity PDF’s are in full compliance with theory, the measured energy spectra look dissimilar. Several effects that contribute to this effect have been uncovered. The photon noise in the images adds a significant background to the spectrum over the full spectral range. Diffusion of the line mainly affects the inertial subrange, so that no $k^{-5/3}$ scaling can be found.

If we consider higher order statistics in the form of structure functions, we obtain much better quality statistics. Although the smallest scales remain affected by artifacts and diffusion, it does not significantly influence the larger scales. We find that for a write-read delay of 10 $\mu$s the scaling exponents of our transverse structure function actually correspond very nicely with the Log-Poisson model.

An important characteristic of turbulence is its ability to transport and mix fluid effectively and this phenomenon can be expressed in terms of the evolution of the line separating two different marked regions of the flow. The tracking of such lines is nearly impossible by means of conventional techniques, but APART is well suited for this application. It is expected from theory that the line length increases exponentially with an exponent $\tilde{\gamma}/\tau_\eta$, where $\tilde{\gamma}$ is $\text{Re}$ independent, but time dependent, as can understood by the model of Girimaji and Pope [77]. It has also been observed that the wrinkled line has a fractal dimension $d_f > 1$ and thus the stretching rate $\tilde{\gamma}$ found depends on the fractal dimension of the line and the length of the line elements (“rulers”) that are used to determine the total length. The fractal dimension that is found is $d_f = 1.017$, a value that is comparable to our kinematic simulations ($d_f = 1.020$) but considerably lower than the value (indirectly) derived by Villermaux et al. [85] of $d_f = 1.10$. The experimentally found stretching rate, extrapolated to ruler size $\eta$, is found to have a maximum $\tilde{\gamma}_0 = 0.085$, lower than those found in DNS simulations by Goto and Kida [89] ($\tilde{\gamma}_0 = 0.17$).
Vereenvoudigde samenvatting

Stromingen en turbulentie

In dit promotieonderzoek is een nieuwe techniek voor het meten van stroming van gassen en, in het bijzonder, het meten van turbulentie geïntroduceerd. Wanneer men denkt aan turbulentie is de eerste associatie vaak de vliegreis door een turbulent atmosfeer. Hoewel dit klopt, is het maar een klein onderdeel van de rol die turbulentie speelt in ons dagelijks leven. Turbulentie is bijvoorbeeld essentieel voor het effectief mengen van twee gassen of vloeistoffen. Dit kun je zien wanneer je in je kopje koffie roert (turbulentie maakt), maar hetzelfde principe geldt ook voor hoogovens waar gesmolten metalen worden samengevoegd om een legering te maken.

Hiermee wil ik aangeven dat de invloed van turbulentie overal om ons heen is, vaak ongemerkt. Bovendien is dit een voorbeeld van een toepassing, maar ook vanuit een theoretisch oogpunt is turbulentie erg interessant. Je kunt je afvragen, “waarom zijn turbulente stromingen nog niet helemaal begrepen?”. Dit is geen gekke vraag. Immers, de eerste observaties werden al zo'n 500 jaar geleden gemaakt door Leonardo da Vinci. Een van zijn schetsen is te zien in afbeelding 2.2.

Nu is het inderdaad zo dat er een wiskundige omschrijving is gevonden die klopt voor elke stroming: dit is de, bij natuurkundigen, bekende Navier-Stokes vergelijking. Zoals hieronder te zien is, betreft het een vrij complexe formule, maar als je deze uiteindelijk hebt opgelost weet je precies hoe de gas of vloeistof stroomt.

$$\rho \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + \mu \nabla^2 \mathbf{v} + \mathbf{F}$$

We kunnen op deze manier de bewegingen van ‘rustige’ stromingen voorspellen (zoals bijvoorbeeld water dat langzaam door een breed kanaal stroomt). Er is echter een probleem: als de stroming sneller wordt, zal deze voorspelling op een bepaald moment “instabiel” worden. Denk hierbij weer aan stromend water, maar deze keer een snel stromende rivier: het water kolkt en wervelt en geen moment is de stroming hetzelfde als het voorgaande moment. Wanneer dit gebeurt noemen we de stroming turbulent. We kunnen deze mate van (in)stabiliteit van een stroming samenvatten in een enkel getal, het zogenaamde Reynolds getal. Een hogere waarde hiervan geeft een meer turbulent stroming aan.
Leonardo schreef hierbij: "Zie hoe de beweging van het wateroppervlak, dat gelijkenissen vertoont met haar, twee bewegingen toont. De een is veroorzaakt door het gewicht van het haar, het ander is de richting van de krullen. Zodoende heeft het water een krullende beweging, deels veroorzaakt door de hoofdstroom, deels door de willekeurige en tegengestelde stroming."

Deze instabiliteit vormt het probleem als we proberen de stroming te berekenen met de Navier-Stokes vergelijkingen. We kunnen nog wel een oplossing voor onze formule vinden, maar een minuscule wijziging in de beginomstandigheden, resulteert in een volledig ander stromingsveld. Dit wordt ook wel eens uitgelegd aan de hand van het beroemde vlindereffect: De vleugelslag van een vlinder aan de andere kant van de wereld kan hier het verschil uitmaken tussen een windvrije dag en een langsstormende orkaan.

We hebben nu geen kant en klare oplossing meer voor het stromingsveld. Het enige wat we kunnen doen is het stromingsveld niet slechts een enkele keer meten, maar de meting vele malen te herhalen en in al deze metingen proberen patronen te herkennen. In de statistiek van turbulente stromingen is te zien dat het stromingsveld zich niet volledig willekeurig gedraagt. Belangrijk werk op dit gebied is gedaan door de Engelsman Richardson en de Rus Kolmogorov. Zij hebben zo'n halve eeuw geleden de basis gelegd voor modellen die het gedrag van de stromingen in veel gevallen nauwkeurig omschrijven.

Het onderliggende idee komt op het volgende neer: Neem de kop koffie weer in gedachten. De energie die de turbulentie veroorzaakt (hier de energie die het ons kost om het lepeltje te bewegen) wordt omgezet in grote wervels in de stroming (ongeveer de grootte van het kopje). Deze grote wervels breken op in kleinere wervels.
en deze kleinere wervels vallen op hun beurt ook weer uit elkaar in nog kleinere. Dit gaat net zo lang door totdat de wervels zo klein zijn geworden dat de viscositeit (de stroperigheid) van de vloeistof of gas ze in haar greep krijgt, en ze door wrijving verdwijnen. De kleinste schaal waarop de wervels nog bestaan noemen we Kolmogorov schaal. We moeten ook bedenken dat terwijl enerzijds wervels aan het opbreken zijn, er anderzijds tegelijkertijd ook weer nieuwe, grote, wervels gecreëerd worden. Dit betekent dat op elk moment het stromingsveld bestaat uit wervels van verschillende grootte die naast, maar ook over elkaar heen bestaan. Dit beeld is geschetst in figuur 2.3.

Dit simpele model is in de loop der tijd verder uitgebreid. Echter, geen van de tot nu toe ontwikkelde modellen geeft een complete beschrijving van de stroming. Dit betekent dat er op het gebied van (turbulente) stromingsleer nog steeds genoeg te doen is.

Een apocrief verhaal onder natuurkundigen die zich met stromingsleer bezighouden is dan ook dat van Albert Einstein die op zijn sterfbed ligt. Hij is van plan God twee vragen te stellen: “Waarom is er relativiteit en waarom is er turbulentie?”. “Ik geloof echt,” zei de beroemde fysicus, “dat hij het antwoord heeft op de eerste vraag”.

Een volgende vraag die we ons kunnen stellen is: waarom gebruiken we geen computers om turbulentie te berekenen? Immers, we kunnen aan de hand van de Navier-Stokes vergelijkingen het hele turbulente stromingsveld calculeren. Computers worden inderdaad veel ingezet om stromingsvelden door te rekenen, en vaak met veel succes. Dit gebeurt echter vaak met behulp van aannames (modellen) waar-
door een aanzienlijk deel van het rekenwerk kan worden overgeslagen. Dit is omdat de benodigde berekeningen van het stromingsveld erg onvoordelig schalen met de turbulentie graad. In de praktijk betekent dit dat zelfs de grootste supercomputers (nog) niet de vele realisaties (afbeeldingen van het stromingsveld) die nodig zijn voor correcte statistieken van een sterk turbulent stromingsveld kunnen doorrekenen.

Nu we vast hebben gesteld dat, wetenschappelijk gezien, de experimentele bepaling van de turbulentie graad nog steeds interessant is, kunnen we gaan kijken wat voor werktuigen ons daarvoor ter beschikking staan. Dit zijn er te veel om in dit bestek te behandelen, maar een moeten we er toch noemen.

De hittedraad snelheidsmeting is één van de bekendste en meest gangbare technieken voor het meten van turbulentie. Hierbij wordt een uiterst dunne draad in de stroming geplaatst en elektrisch verwarmd. De afkoeling van de draad is afhankelijk van de snelheid waarmee de lucht langs de draad stroomt. Hoewel dit een uiterst nauwkeurig manier van snelheden meten is, heeft het ook enige nadelen: de draad kan geen ruimtelijke verschillen in snelheden waarnemen, en kan slechts wachten op wat langsgeblazen wordt. Bovendien kan de draad slechts relatief trage fluctuaties in de snelheid waarnemen. Niettemin levert deze techniek bijzonder nauwkeurige resultaten op en we zullen onze metingen ook gaan ijkken aan deze techniek.

Lijnen schrijven met APART

Hiermee zijn we aanbeland bij de nieuwe techniek die de basis vormt voor dit proefschrift, genaamd APART. Dit staat voor "Air Photolysis And Recombination Tracking". We kunnen de techniek iets prozaïsch betitelen als "schrijven in lucht", waarbij we de laser als pen kunnen zien. We schrijven lijnen in lucht door gebruik te maken van een zogenaamde Argon-Fluoride excimer laser. Door precies de juiste kleur licht te kiezen (wat een maat is voor de energie) is het mogelijk om zuurstof te splitsen en uit stikstof een electron te verwijderen. Door een chemisch proces vormen deze vervolgens het molecuul stikstof oxide (NO). Het is gebleken uit metingen, maar ook uit chemische berekeningen dat de hoeveelheid NO na enkele microseconden op haar maximum is en daarna langzaam verminderd. Omdat de laser bundel 50 µm breed is (dit is slechts vijf-honderdste millimeter!) ontstaat er een zeer dunne lijn van deeltjes langs het pad van de laser. Doordat de hoeveelheid stikstof oxide moleculen die zich normaal gesproken in lucht bevinden verwaarloosbaar is, vormt de NO concentratie langs de lijn een groot contrast met NO concentratie van de omgeving.

Aangezien de deeltjes zijn aangemaakt in een turbulente stroming, zal deze de deeltjes meevoeren. De snelheid in de stroming zal overal anders zijn, en dus zullen sommige moleculen sneller verplaatsen dan anderen. Dit betekent dat wanneer we op een vastgestelde tijd na het schrijven de lijn gaan bekijken, we zien dat de oorspronkelijk rechte lijn (zoals getekend in figuur 2.4 als de zwarte lijn) nu niet alleen verplaatst, maar ook vervormd is (aangegeven als de donkergrijze lijn in figuur 2.4).

We gaan er van uit dat alle moleculen, met de hoofdstroom mee, omhoog bewegen en de beweging zijwaarts verwaarloosbaar is. Dan kunnen we uit de verplaatsing (zie de pijlen in figuur 2.4), in combinatie met de verstreken tijd, de locale snelheid bepalen.

Echter, om dit te kunnen bepalen moeten we eerst een manier hebben om de NO deeltjes zichtbaar te maken. Indien we met het blote oog naar de lijn kijken is deze, net als lucht, transparant. Om de moleculen toch zichtbaar te maken gebruiken we een tweede laser, die precies de juiste kleur heeft om de deeltjes te laten oplichten. Deze zogenaamde dye laser produceert een “uitlees” laserbundel die dusdanig breed is dat hij de gehele geschreven lijn omvat. Door weer de juiste kleur licht te kiezen zullen de NO moleculen fluoresceren. Deze fluorescentie lijkt op wat te vinden is bij de wijzers van sommige horloges, die in het donker oplichten. Het verschil met een horloge is echter dat het fluorescerende licht van NO ultraviolet is. Dat wil zeggen, het ligt buiten het bereik waarbinnen onze ogen licht kunnen waarnemen.

Dit fluorescentie signaal wordt vervolgens opgenomen met behulp van een zeer lichtgevoelige camera. De camera is in nog een opzicht speciaal: hij is in staat om ultraviolet licht op te nemen.

De gebruikte opstelling is geschetst in afbeelding 2.5. hier is te zien dat er naast de al beschreven elementen ook een filter aanwezig is. Deze houdt het directe laserlicht tegen, en zorgt ervoor dat alleen het fluorescentie signaal van de lijn door wordt gelaten.

Nu missen we nog een belangrijk element, en dat is onze bron van luchtstroming of, exacter, onze bron van turbulentie. Hiervoor gebruiken we een turbulente jet. Dit is een luchtstroom die met een zeer hoge (ongeveer 1000 km/uur) maar gelijkmatige snelheid uit een tuit wordt geblazen. Doordat de omliggende lucht stilstaat
Figuur 2.5: Schematische afbeelding van de meet opstelling. De excimer (schrijf) laser schiet een bundel die gefocuseerd wordt door een lens. De dye (lees) laser verlicht de lijn, en het weerkaatste licht valt door een filter op de camera. Het filter houdt het directe licht van de lasers tegen.

Ontstaan er op het raakvlak van de stilstaande en de bewegende lucht hevige wervels. Op 40 centimeter boven de opening is de snelheid teruggezet naar ongeveer 140 kilometer/uur en is de luchtstroom ontwikkeld tot volledige turbulentie. Op deze hoogte gaan we dan ook lijnen schrijven.

In figuur 2.6 laten we enkele van deze lijnen zien. Zoals te verwachten valt, verplaatsen de lijnen zich steeds verder omhoog indien de tijd tussen schrijven en lezen langer wordt gemaakt. Wat ook opvalt is dat de vervormingen steeds groter worden. Deze kronkels in de lijn geven aan dat we inderdaad wervels in de turbulente stroming kunnen waarnemen. Ook is te zien dat er fluctuaties over zowel grote afstand (grote wervels) als over kleinere afstand (kleine wervels) aanwezig zijn in de lijnen.

Hoewel de afbeeldingen in figuur 2.6 goed laten zien dat we stromingen kunnen visualiseren, hebben we nog een stap nodig om hier kwantitatieve gegevens uit te halen. Om de waarden voor de locale snelheden te bepalen moeten we nauwkeurig het lijncentrum bepalen. Aangezien we later vele duizenden plaatjes gaan analyseren, zullen we deze stap moeten automatiseren. Dit doen we door elk beeld op te delen in verticale plakken (zie figuur 2.7 links) en voor elk van deze plakken het profiel te vergelijken met een van tevoren vastgesteld profiel (zie figuur 2.7 rechts). Hoewel de vorm van het profiel van tevoren vast ligt, kunnen we verschillende parameters wijzigen om het profiel optimaal te laten passen op de meting. De belangrijkste parameters zijn hoogte, breedte en de centrum positie. Als we al de centrumpositie van de net geschreven lijn (in deze betreffende plak) kenden en nu ook de positie van de verplaatste lijn, dan kunnen we vervolgens uit de verplaatsing en de vertraging de snelheid bepalen.
Figuur 2.6: Voorbeelden van geschreven lijnen. Bij elke lijn is er een andere wachttijd tussen het schrijven en lezen gekozen. Hoe langer de wachttijd is, des te vervormder de lijn wordt.
Figuur 2.7: Lijncentrum bepaling voor elke positie langs de lijn. Op elke locatie langs de lijn wordt een doorsnede gemaakt (linker afbeelding). In deze doorsnede wordt het gemeten intensiteitsprofiel vergeleken met een ideaal profiel (rechter afbeelding).

Chemie en fysica van APART

De creatie van NO atomen ligt ten grondslag aan de APART techniek. Maar wat gebeurt er als gevolg van de interactie van de laserpuls met de lucht, die er voor zorgt deze moleculen gemaakt worden? Het antwoord hierop is niet zo makkelijk te geven, maar het is belangrijk hier antwoord op te krijgen. Immers, dit proces bepaalt allerlei eigenschappen van de lijnen zoals de tijd tot het ontstaan, de tijd totdat ze weer verdwijnen en de snelheid waarmee ze verbreden.

Wat we ook kunnen zien is dat diffusie een belangrijke rol speelt in onze lijnen. Laten we even terug denken aan de kop koffie. Ook als hier niet in geroerd wordt, zal de druppel melk zich verspreiden. Hier ligt aan ten grondslag dat alle moleculen continu heen en weer bewegen en met elkaar botsen. Vergelijk het met de ballen op een biljart tafel, waar op afgestoten wordt, zoals in figuur 2.8. In molecuul-biljart vinden dit soort botsingen miljarden malen per seconde plaats en zullen de ballen zich al snel over het hele veld verdeeld hebben.

Wat we ook zien is dat een deel van de laserenergie wordt omgezet in warmte. Dit betekent dat in het lijncentrum de temperatuur stijgt, met zo’n 300 tot 600 graden Celsius. Dit is een aanzienlijke temperatuurverhoging, en dit heeft dan ook effect op de stroming. We zien dat het ervoor zorgt dat lijnen sneller verbreden dan bij kamer temperatuur. Dit is te begrijpen omdat een hogere temperatuur betekent dat moleculen sneller bewegen, en zich effectiever verspreiden. We kunnen daardoor ook niet zeggen dat onze meetmethode geen enkele invloed heeft op de te meten eigenschappen. Aan de andere kant, de verwarming vindt wel op plaats zeer kleine
Figuur 2.8: Een voorbeeld van diffusie. De biljart ballen liggen eerst op een kluitje, maar als een van de ballen de anderen raakt, komen ze steeds verder uit elkaar te liggen. Wanneer dit soort botsingen miljarden malen per seconde plaatsvinden zullen de ballen, of in ons geval moleculen, zich al snel gelijkmatig over het hele veld verdeeld hebben.

schaal (de geschreven lijnen zijn echter maar 50 µm breed) en de warmte is daarom na korte tijd alweer verdwenen.

Snelheidstatistieken

Zoals we al eerder besproken hebben, is het nodig om vele instanties van het stromingsveld te bekijken. In het geval van APART betekent dit het vele malen schrijven van lijnen (tien-duizenden) en alle te onderscheiden snelheden langs de lijn (zo’n 1000) eruit halen. Deze tientallen miljoenen (!) snelheden kunnen nu op verschillende manieren gebruikt worden. Als we kijken naar simpele statistiek zoals de gemiddelde snelheid en gemiddelde fluctuatie van de snelheid, vinden we precies terug wat we ook meten met de hittedraad techniek.

Een andere eigenschap waar we naar kijken is het snelheidsverschil tussen de snelheden op twee locaties langs de lijn ($u_1$ en $u_2$ in figuur 2.9). Het is goed voor te stellen dat wanneer we een afstand $\Delta x$ nemen tussen de twee locaties, het snelheidsverschil ons iets vertelt over wervels van grootte $\Delta x$ en groter (zoals wervel 1), maar niet over wervels kleiner dan $\Delta x$ (zoals wervel 2). Dit doen we voor alle lijnen en voor alle locaties langs de lijn. Op deze manier kunnen we de verschillende lengteschalen aftasten.

In deze metingen vinden we afwijkingen vergeleken met de waarden gemeten met behulp hittedraadmetingen (en zoals bekend uit de literatuur). Op de lengteschalen waar de kleinste wervels nog net aanwezig zijn (de zogenaamde Kolmogorov schaal), zien we namelijk dat in onze metingen diffusie een belangrijke rol speelt. De wervels op deze schaal worden uitgesmeerd door de diffusie.

Dit effect hebben we beter leren begrijpen door computers te gebruiken om onze meettechniek na te bootsen met behulp van een simpel model. Hier zien we in-
Figuur 2.9: Twee snelheidscomponenten op lijn met een onderlinge afstand $\Delta x$. Het snelheidsverschil tussen $u_1$ en $u_2$ vertelt ons iets over wervels van grootte $\Delta x$ (wervel 1). Wervels die kleiner zijn, kunnen niet tegelijkertijd invloed hebben op de plaats van $u_1$ en $u_2$.

derdaad dat wanneer de lijnen niet onderhevig waren aan diffusie, de uitkomsten overeen komen met hetgeen verwacht mocht worden uit literatuur, en dat dit beeld aanzienlijk verandert wanneer diffusie wel een rol speelt. Niettemin kunnen we met behulp van slim gebruik van net wat grotere schalen toch veel leren. Zo krijgen we bijvoorbeeld goede waarden uit complexe karakteristieken van de stroming, zoals zogenaamde intermittentie. Een belangrijk deel van dit proefschrift behandelt deze stromingseigenschap, maar deze materie voert te ver om hier te bespreken.

**Uitrekken van lijnen**

Denk nogmaals aan de inmiddels bekende druppel melk in de kop koffie. Door koffie te roeren zal de oorspronkelijk ronde druppel naar alle kanten uitgetrokken en vervormd raken, totdat de uitloper alle kanten van het oppervlak hebben bereikt. Een manier om de efficiëntie van dit process te beoordelen is door te kijken naar de snelheid waarmee de lengte van de scheidslijn tussen de melk en de koffie toeneemt, zie figuur 2.10.

Dit mechanisme kunnen we het bestuderen door een enkele lijn te schrijven en de vervorming ervan in de tijd vast te leggen. Dit is iets dat bijzonder moeilijk is te doen met conventionele methoden, maar waar APART uitermate geschikt voor is.

Een logische aannemer is dat de lijn in een zekere tijdstijd $t$ een zekere factor $\gamma$ langer wordt. Deze verlengde lijn zal in de volgende tijdstijd $t$ weer een factor $\gamma$ langer worden. Dit is goed vergelijkbaar met rente krijgen per jaar op een spaarrekening. Net zoals bij het effect van rente-op-rente-op-rente het banksaldo steeds sneller zal groeien, zal ook het langer worden van de lijn steeds sneller gaan verlopen. We noemen dit exponentiële groei. Echter, de literatuur en onze eigen computersimulaties leren ons, dat de lijngroei in het begin niet helemaal exponentieel verloopt.
Figuur 2.10: Voorbeeld van chaotische menging. In de eerste afbeelding wordt er een druppel melk in de koffie geschonken. In de tweede afbeelding is getoond hoe de druppel zich verspreidt door diffusie. In de derde afbeelding is getoond hoe de druppel zich verspreidt door te roeren: Door turbulentie zal de druppel melk gaan vervormen en zich steeds verder verspreiden.

Onze metingen hebben laten zien dat ook in de praktijk dit effect te zien is, hoewel de gevonden groei kleiner is dan verwacht.

Conclusie

Moleculaire schrijf methoden en met name APART zijn waardevolle technieken waarmee op nauwkeurige wijze stromingseigenschappen te meten zijn. Een met name waardevolle eigenschap van de methode is dat het mogelijk om is gewenste patronen te schrijven op precies de plaats en precies de tijd die men wil. Dit levert nieuwe mogelijkheden op ten opzichte van klassieke methoden (zoals de genoemde lijnrek metingen). Wel hebben we gezien dat de geschreven patronen onderhevig zijn aan diffusie. Dit kan in sommige omstandigheden ervaren worden als een nadeel, maar in een ander licht kunnen we dit ook als een interessant vraagstuk zien. Namelijk: wat is de interactie tussen turbulentie en diffusie als deze van dezelfde orde van grootte zijn?
Nawoord

Aan het einde van het schrijven van een proefschrift is het moeilijk om nog puf te vinden voor die laatste kleine stukjes werk. Dat geldt echter niet voor dit dankwoord. Het enige is probleem is waar ik moet beginnen. Laat mij dank eerst maar uit gaan naar Nico, een dagelijkse bron van inspiratie en de Graalburght goeroe op het gebied van optica en sarcastische opmerkingen. Ik ben bang dat van dat laatste zeker net zoveel is blijven hangen als van het eerste. Verder wil ik Hans bedanken, die het grote geheel in het oog hield, waar ik nog wel eens wilde verdwalen in de details van de meting van die dag. Echter, toen ik aan het einde van het schrijven blinde vlekken ontwikkelde voor de laatste foutjes en detail, zorgde jij juist voor de puntjes op de i. Het uiteindelijke “akkoord” met mijn manuscript had dan ook grote betekenis.

Zonder Willem was dit boekje er waarschijnlijk nooit geweest, en als het er wel was gekomen, was het nooit geworden wat het nu is. Hoewel ik niet altijd je briljante ingevingen direct begreep, leerde ik ze op waarde schatten en hebben ze het verhaal de vorm gegeven die het nu heeft. Daarnaast heb ik veel van je geleerd over uiteenlopende onderwerpen als statistische technieken, ideale schaatshouding en het programmeren in arcaïsche talen als Fortran. Het plezier van het maken van mijn eigen ponskaarten heb ik helaas nooit mogen proeven.

Het is eerder in dankwoorden gezegd, maar het is waar: een promotie doe je niet alleen. Als promovendus mag je dan ook in je handen knijpen als je kunt rekenen op mensen die veel handiger zijn dan jij. Ik ben erg dankbaar voor alle technische hulp van Arjan, Ad, Freek, Gerald, Jan en Gert en administratieve hulp (in mijn geval geen onaanzienlijke taak) van Ine, Marjan, Brigitte, Anita, Erna en Magda. My special thanks go out to David Osbourne, Susumu Goto and Christos Vassilicos, for generously sharing their kinematic simulation code and line stretching data.

Daarnaast heb ik het geluk gehad dat ik heb kunnen werken in twee bijzonder leuke groepen, turbulentie en vortex dynamica in Eindhoven en Molecuul- en laser fysica in Nijmegen, waarvan ik nu veel collega’s ook als vrienden beschouw. Ik ben bang als ik jullie nu allemaal zou gaan opnoemen ik één van jullie ga vergeten, maar schaats- en skatevrienden, mede-koffiegebruikers, filmavondbezoekers, volleyballers, computergekken en kroegtijgers, jullie weten zelf wel wie jullie zijn.

Aan alle vrienden die inhoudelijk niet met dit boekwerk te maken hebben, maar wel altijd bereid waren om de luisteren naar veel te lange verhandelingen over grote lasers en kleine wervels en zo om de week tijd vroegen “En, heb je al een datum?”, en in het bijzonder, Anke, Jolanda, Marjolein Simon, en Tamara: bedankt. Esther,
de rondjes naar Oortjeshekke waren altijd een welkome en leuke afwisseling van de weekenden proefschrift typen, nu nog die elfstedentocht. Maartje, beste buuf, bedankt voor de avonden analyseren wie er zou winnen bij Peking Express.

En dan natuurlijk de donderdagavond club. Ralph, Harm, Karel en Maikel, ik hoop dat we nog veel discussies gaan hebben over het leven, F14’s, persoonlijke hygiëne in het oude China, Smeg en de rest. Maarten, ik was van plan hier een leuke anekdote over je te schrijven maar ik betwijfel of, als ik daar aan begin, “één pagina dankwoord voldoende is”. Ook wil ik Gerard, Marja en Annemieke, mijn tweede familie, bedanken voor alle steun, vooral in mijn eerste studiejaren.

Irene, jij had voorzien dat het schrijven van dit boekje een flinke beproeving zou worden. Nu het af is kan ik zeggen, je had helemaal gelijk, maar het was de moeite waard!

Mijn kleine broertje heeft een speciale en noodzakelijke rol gehad in het ontstaansproces van dit werk. Guido, steeds als de stress zijn akelige hoofd opstak, zorgde jij voor de broodnodige ontspanning, van X-box nachten en tuinfeestjes bij “Rooie Arie” tot het verzinnen van raadseltjes (“het heeft een rode puntmuts en...”) tijdens zeiltochten. Bedankt dat je deze taak op je hebt genomen en ik hoop dat je dit zal blijven doen tot ver na deze promotie.

Uiteindelijk gaat mijn diepe dank uit naar mijn ouders die vanaf het moment dat ik besloot dat ik natuurkunde wilde studeren, maar nog niet overtuigd was dat ik het ook kon, vertrouwen in me hebben gehad. Dit boekje was er zonder jullie nooit gekomen.
Curriculum Vitae

5 January 1975  Born in Amsterdam, The Netherlands

1987-1993  SG Werenfridus VWO, Hoorn, NH

1993 - 2000  Student Applied Physics, Eindhoven University of Technology.

  • Internal traineeship in the Physics of Nanostructures Group
  • External traineeship in the Applied Molecular Physics Group at the Radboud University Nijmegen
  • Masters thesis project at the Maxima Medisch centrum.

2000-2005  Ph.D. student in a collaboration between the Turbulence & Vortex Dynamics Group at the Eindhoven University of Technology and the Applied Molecular Physics Group at the Radboud University Nijmegen