MODELLING OF THERMAL ARGON PLASMAS

PROEFSCHRIFT

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Chapter 1

General Introduction

When matter in the gaseous state is supplied with sufficient heat it turns into a plasma, which consists of a mixture of atoms, ions and electrons. Since the basic properties of a plasma are mainly determined by the presence of electric charges the principal plasma parameters are the concentration and the mean energy of the charged particles. It is well known that plasmas used in laboratories cover a wide range of these parameters. Especially in the last four decades the technological and industrial applications in the field of plasma physics have been developed considerably and are still growing.

In a wide number of applications one deals with plasmas at temperatures lower than $30000 \, \text{K}$, such as magnetohydrodynamic generators, microwaves discharges arc plasmas and so on. Important from the fundamental point of view is that these laboratory plasmas in general are in a state of non-equilibrium. The main reason is that a laboratory plasma is not a closed system so that the plasma which in almost any case is sustained by an electromagnetic energy source, continuously loses energy by radiation, diffusive and convective transport to the environment. Therefore it is obvious that there is no thermodynamic equilibrium since the energy input is transferred to other energy forms.

A special class of plasmas is formed by the so-called thermal plasmas. They are characterized by relatively high particle densities and they have ionization degrees varying between $10^{-4}$ - $10^{-1}$ while the temperature has typical values in the range of $6000 - 25000 \, \text{K}$. The use of thermal argon plasmas can be found in various applications. The inductively coupled plasma (ICP) and the expanding cascaded arc are well-known in the field of spectrochemical analysis, plasma chemical synthesis, heating of materials, lightsource technology and deposition of carbon or silicon based films. In the theoretical study of these plasmas the processes which cause deviations from local thermal equilibrium (LTE) and especially their influence on the flow and temperature fields are investigated using macroscopic models. Most thermal plasma models are based on the LTE assumption [1, 2]. Accurate measurements of the electron density and electron temperature and excited state population densities in the cascaded arc [3, 4] and the ICP [5] revealed the fact that the LTE assumption in these plasmas is not a sound basis. Moreover,
from analysis of the instantaneous response of excited levels using the relaxation technique [6] in the ICP, BYDDER et al. [7] and FEY et al. [8] could determine the ratio of the electron and heavy particle temperature. Therefore a non-LTE model of the thermal plasma is required to get information of both the electron and heavy particle temperature and electron densities with respect to elementary balances and transport phenomena.

In the group Equilibrium and Transport in Plasmas at the Eindhoven University of Technology the fundamental processes in thermal plasmas are studied both experimentally and theoretically. In many cases models were constructed to describe specific phenomena in thermal plasmas. However, since there are many interfering processes in a plasma it is better to construct a grand model for the plasma as a whole. With such a global model we can investigate the fundamental interplay between the microscopic and macroscopic processes. Moreover models are useful to investigate different operating conditions so that excessive empiricism can be avoided. Modelling of the cascaded arc was a part of the work of BEULENS [9]. We will concentrate on the modelling of the ICP.

The ICP which is a special type of high-pressure gas discharge, is created and sustained by a high frequency (RF) magnetic field. This RF-field is generated by an induction coil surrounding the plasma vessel and induces an electric current in the plasma. The power is dissipated in the active zone which has an annular shape due to the electromagnetic skin effect. In Fig. 1.1 a typical spectrochemical ICP set-up is shown in which the plasma confinement vessel is a torch [10]. The outer flow serves as a cooling flow to protect the quartz torch wall, the intermediate flow can be used to lift the plasma, while in the central flow samples can be introduced for spectrochemical purposes. In these open configurations, which generally operate under atmospheric pressures, forced convective energy transport is an important energy loss mechanism, besides radiation and conductive energy losses. For obvious reasons a closed confinement vessel is used if
the ICP is used as a light source. A commercial low pressure ICP light source is already available and the high pressure light sources are under development at Philips Forschungslabor Aachen [11]. In all applications of the ICP an important advantage is that it is a clean source of relatively high energy content. The purity is due to the absence of eroding electrodes.

Outline of the thesis

It is convenient to divide the plasma kinetics into fast microscopic activity, which concerns collisions (elastic and inelastic) and radiative processes, and the relatively slow macroscopic transport processes such as conduction, convection and diffusion which tend to drive the system out of translational equilibrium. When there is a clear distinction between the fast and slow processes the plasma can be considered as a fluid and local thermodynamic quantities can be defined. The plasma is then described by hydrodynamical equations. Chapter 2 deals with the conditions for the validity of the hydrodynamical approach in modelling plasmas.

It is important to realize that the non-equilibrium state of microscopic activity manifests itself on a macroscopic scale. Ionization and recombination must be accompanied by the inward and outward transport of neutral particles, respectively. Another important fact is that most of the ionization/recombination and excitation/de-excitation processes are effectuated by electrons. Free electrons effectively interact with bound electrons in atoms and ineffectively transfer kinetic energy to the atoms as a whole due to the large mass difference. As a consequence there might be a significant difference between the electron and heavy particle temperature while the internal state distribution function of the atoms is ruled by the electrons. Van der MULLEN [12, 13] has made a classification of the electron governed microscopic processes in atomic plasmas in terms of proper and improper elementary balances. With this classification of excitation and ionization equilibra more insight was obtained in the interplay between the non-elastic electron heavy particle collisions as described by collisional radiative (CR) models. Moreover, the calculation of the effective ionization and recombination with numerical CR-models could be simplified by using the fact that the excitation flow through the upper part of the atomic system is analytically known. The details concerning this matter are given in chapter 3.

In chapter 4 the so-called hybrid collisional radiative model is applied to recombining plasmas and the numerical results are compared with the experimental data of van de SANDEN [14] and with analytical expressions for the effective recombination coefficients under these conditions.

Another important aspect of the state of non-equilibrium will be manifested and created by the radiation loss of the plasma. The model of WILBERS et al. [15] for the total radiation loss is extended in that it allows for equilibrium departures.
of the atomic state distribution function as well. The results are presented such that they can be easily implemented in a macroscopic model for non-equilibrium plasmas. This will be discussed in chapter 5.

With the various models for the ionization/recombination and radiation loss the problem of macroscopic modelling of thermal plasmas can be tackled. Semi-empirical formulae for the ionization/recombination coefficients, a model for the calculation of the electromagnetic field in the ICP and the transport coefficients will be given in chapter 6.

In chapter 7 the numerical model for the macroscopic description of the plasma will be outlined. A stationary two-dimensional model, inspired by the work of MILOJEVIC, has been developed in orthogonal curvilinear coordinates so that different geometrical configurations can be handled with the same model. A method to construct a curvilinear coordinate system will be discussed. Further an overview of the SIMPLE algorithm of PATANKAR to solve the system of transport equation will be given [16].

The macroscopic model in which the results of chapters 2-6 are incorporated, will be applied to the inductively coupled plasma as used by FEY [10] and the group of BOULOS [17]. The results of the model calculations are presented in chapter 8.

References

Chapter 2

Physical model

2.1 Introduction

In this thesis the macroscopic description of thermal plasmas is based on the hydrodynamical equations. The validity of this approach will be studied in this chapter. Since the hydrodynamical equations are derived from the Boltzmann transport equation (BTE) the study of the validity of the hydrodynamical approach can be traced back for each term in the BTE. The analysis can in cases be formulated in characteristic times and it is important to distinguish between the microscopic and hydrodynamic timescales of the system under study. The fastest significant timescale deals with the time \( \tau_c \) between successive collisions of particles, while the slowest timescale is the hydrodynamical relaxation time \( \tau_{\text{hyd}} \). The latter timescale is associated with macroscopic transport such as diffusion and convection. Only \( \tau_{\text{hyd}} \) is experimentally accessible in the experimental plasma set-ups of the ETP group. The discussion and examples will mainly refer to the inductively coupled plasma (ICP) and cascaded arc-generated plasmas.

2.2 Basic Balances and Equilibrium Departure

To use the classical kinetic theory of dilute gases for the description of the dynamical behaviour of the plasma, the statistical Debye potential concept for plasmas must be applicable. The condition for this requires that \( n_e \lambda_D^2 \gg 1 \) where \( n_e \) is the number density of the charged particles and \( \lambda_D \) is the Debye-radius. Then the average interaction energy is much smaller than the kinetic energy so that the plasma behaves as an ideal gas. Using the expression \( \lambda_D = \sqrt{\varepsilon_0 kT/n_e e^2} \) it is found for the ICP that the number of electrons within the Debye-sphere equals \( \pm 40 \) whereas for the cascade arc this number is found to be approximately \( \pm 20 \). So we may conclude that the classical kinetic theory gives a good description of the plasmas under study.

Of primary interest in the description of non-equilibrium properties in plasmas is the one particle distribution function \( f(\vec{r}, \vec{v}, t) \), which is defined in such a way
Chapter 2

that \( f(\vec{r}, \vec{v}, t) \, d\vec{r} \, d\vec{v} \) represents the number of particles within a volume element \( d\vec{r} \) at position \( \vec{r} \), and within the velocity space element \( d\vec{v} \) centred around \( \vec{v} \) at time \( t \). The distribution function for species \( \alpha \) can be determined by the Boltzmann equation and reads

\[
\frac{\partial f_\alpha}{\partial t} + \vec{v} \cdot \nabla_r f_\alpha + \frac{\vec{F}_\alpha}{m_\alpha} \cdot \nabla_v f_\alpha = \left( \frac{\partial f_\alpha}{\partial t} \right)_{el} + \left( \frac{\partial f_\alpha}{\partial t} \right)_{in} \tag{2.1}
\]

where the first term on the left-hand side of Eq. (2.1) describes the temporal behaviour, the second and the third describe the influences of spatial inhomogeneities and macroscopic forces \( \vec{F}_\alpha \), respectively. In a plasma the forces will be mainly electromagnetic in nature. The right-hand side of Eq. (2.1) describes the changes in the distribution function as a result of inelastic and elastic collisions. Radiative processes may be incorporated in the inelastic processes. Further we confine ourselves to monoatomic partially ionized plasmas which consist of electrons, ions and atoms of one element, i.e. \( \{\alpha = e, i, a\} \). The distribution function contains information of the state of equilibrium (departure). Therefore it is useful to consider Eq. (2.1) in order to understand in global terms the influences of the sources of equilibrium departure. A first glance at (2.1) shows that the macroscopic phenomena are grouped at the lhs (the second and third term) whereas the elementary processes are gathered at the rhs. An important type of elementary process is that of the self-collision. For those species for which the frequency of collisions between alike particles is high, maxwellization will be established. For our class of plasmas this certainly applies to the atoms for which the gradients and external forces may be neglected. Ions are different in the sense that they are sensitive to electromagnetic forces. However, the momentum and energy exchange with atoms occurs so effective that we may assume that the ions and atoms have the same translational distribution function. The maxwellization of electrons is less simple and will be discussed in the next section.

The above discussion applies to the translation aspect of atoms and ions. The internal state distribution is much more determined by electron collisions rather than heavy particle collisions. At the other hand these inelastic collisions might disturb the maxwellization of the electron gas. To study the electron energy distribution function (EEDF) one should solve the BTE together with the atomic state distribution function (ASDF) in a self-consistent manner [1, 2, 3]. However, this is beyond the scope of this study. To study the relationship between the ASDF and EEDF in global terms we will follow Van der MULLEN who has made a classification of equilibrium departures in electron excitation kinetic plasmas (EEK) [4], i.e. atomic plasmas dominated by electron kinetics. The classification is based on the so-called improper and proper balances. The latter describes the competition between production and reverse destruction processes.
At this point certain proper balances are of particular importance. A proper balance is in equilibrium when per unit volume and per unit time the number of forward transitions equals the number of backward transitions (cf. (2.2), (2.3) and (2.5)). The equilibrium distribution for the translational energy is effectuated by elastic collisions and is depicted by the following scheme of forward and backward processes,

$$X(E_1^i) + Y(E_2^i) \rightarrow M \rightarrow X(E_1^f) + Y(E_2^f)$$

(2.2)

where $E$ is the kinetic energy and $E_1^i + E_2^i = E_1^f + E_2^f$. The purely elastic collisions between like particles, for which the characteristic timescale is the momentum relaxation timescale $\tau_m$, will drive the translational energy distribution function to the well-known Maxwell distribution. These processes are included in the rhs of (2.1).

The classification of the distribution over the excited states in atoms and ions, i.e. the ASDF, is founded on two kinds of proper balances. The first balance deals with collisional excitation and de-excitation by electrons and is called the Boltzmann balance. It can be represented by

$$A(p) + e(E) \rightarrow B \rightarrow A(q) + e(E - E_{pq})$$

(2.3)

where $A(p)$ denotes an atom in state $p$ and $E_{pq} = E_p - E_q$ is the difference between the excitation energies of state $p$ and $q$. When the Boltzmann balance is in equilibrium the relation between the population density of the excited states is given by

$$\frac{n^B(q)}{g(q)} = \frac{n^B(p)}{g(p)} \exp\left(-\frac{E_{pq}}{kT_e}\right)$$

(2.4)

in which $g(p)$ is the statistical weight of state $p$. For the ground state the notation $n(1) = n^B(1)$ will be used. For our class of plasmas we may assume that $n_a = \sum_{q \geq 1} n(q) \approx n(1)$, i.e. that the density of atoms is in good approximation equal to that of the density of atoms in the ground state. In the expression for $n_a$, $n(q)$ represents the actual density of excited state $q$.

The second, the Saha balance, describes the competition between collisional ionization (forward) and three particle recombination (backward), i.e.

$$A(p) + e \rightarrow S \rightarrow A^+ + e + e$$

(2.5)

The equilibrium state of this balance determines the density

$$\frac{n^S(p)}{g(p)} = \frac{n_a n_+}{2 g_+} \left(\frac{\hbar^2}{2 \pi m_e kT_e}\right)^{3/2} \exp\left(\frac{E_p}{kT_e}\right)$$

(2.6)
The fact that (de)excitation and ionization/recombination are governed by electrons is reflected by the presence of the electron temperature in the denominator of the exponent in Eqs. (2.4) and (2.6). If the Boltzmann and or Saha balances are out of equilibrium the Maxwell distribution can be disturbed. With the knowledge of the translational energy distribution function and the ASDF the plasma transport and the effective ionization/recombination coefficients can be calculated. In the case of near equilibrium the macroscopic properties can also be determined from the distribution function [5].

2.2.1 Deviations of the EEDF from Maxwell distribution, relaxation times

The study of the presence of Maxwell equilibrium for the electron gas will be guided by a simple analysis of the electron Boltzmann transport equation. By comparing the various terms with the electron-electron (e-e) collision validity criteria will be discussed. The leading term in this analysis, i.e. \( (\partial f_e/\partial t)_{ee} \) which tends the electron gas towards a Maxwell distribution is incorporated in the rhs of the BTE (Eq. (2.1)). Including all the various elastic and inelastic processes this rhs contains

\[
\begin{align*}
(\frac{\partial f_e}{\partial t})_{el} &= (\frac{\partial f_e}{\partial t})_{ee} + (\frac{\partial f_e}{\partial t})_{ei} + (\frac{\partial f_e}{\partial t})_{ea}, \\
(\frac{\partial f_e}{\partial t})_{in} &= (\frac{\partial f_e}{\partial t})_{ei} + (\frac{\partial f_e}{\partial t})_{ea},
\end{align*}
\]

(2.7)

(2.8)

All other terms, together with the two transport terms at the lhs of (2.1) deal with equilibrium disturbing processes. Before discussing the validity criteria for the presence of Maxwell equilibrium we will give some comments on the general structure of the electron BTE of a thermal plasma for which we consider a phase space element \( \delta \Gamma \). The occupation of \( f_e(\delta \Gamma, t) \) of this element is the result of various production (P) and destruction (D) processes of which the contribution of the elastic e-e collisions may be written as \( (\partial f_e/\partial t)_{el} = P(\delta \Lambda) - \nu_{ee} f(\delta \Lambda) \) which is justified provided \( \delta \Gamma \) is sufficiently small. Now suppose that the e-e collisions are dominant then the BTE can be written as

\[
\frac{1}{\nu_{ee}} \frac{\partial f_e(\delta \Gamma)}{\partial t} = \frac{1}{\nu_{ee}} P(\delta \Gamma) - f(\delta \Gamma) + \text{other terms}
\]

(2.9)

which expresses that a local disturbance, for instance an overpopulation of \( \delta \Lambda \), will disappear in a time-duration of typically \( 1/\nu_{ee} \). The overpopulation will be redistributed over other elements of the electron phase space and all balances of type (2.2) will equilibrate eventually. This process of maxwellization in which e-e
collisions are the leading processes only describes the redistribution of electrons over the phase space. It does not give information on the electron density $n_e$, mean velocity or mean kinetic energy. The value of $T_e$ is determined by the acceleration of the electric field at the one hand and the collisions with heavy particles at the other hand i.e. the third term at the lhs and the second and third term at the rhs of the BTE (cf Eq. (2.7)). However, if $\nu_{ee}$ supersedes all other frequencies the external forces and the friction with heavy particles will not disturb the maxwellization process but determine the total energy content of the electron gas, i.e. an integral quantity of the electron gas as a whole. That is the reason why these influences can best be described by the moments of the BTE. The fact that these moments and the hydrodynamic quantities can be formulated is due to the fact that maxwellization is effective. Concluding we may state that if the e-e collisions are dominant the electron gas can be treated as a hydrodynamic fluid. The properties of this gas, i.e. the density and temperature are determined by the small terms in the BTE. The slow changes in hydrodynamic quantities are followed quasi instantaneously by the maxwellization.

The characteristic relaxation time to establish a Maxwellian distribution function is determined by the self-collision time. The self-collision time for charged particles as derived by SPITZER [6], is defined as the transverse diffusion time for particles with velocity $v = \sqrt{3kT_e/m_e}$. It is given by

$$
\tau_e = \frac{3^{3/2}}{8\pi \zeta} \left( \frac{e^2}{4\pi \varepsilon_0 m_e} \right)^{-2} \left( \frac{kT_e}{m_e} \right)^{3/2} \frac{1}{n_e n_\Lambda}
$$

$$
= 2.66 \times 10^5 \frac{T_e^{3/2}}{n_e n_\Lambda} \text{ [sec]},
$$

(2.10)

where the physical quantities are expressed in MKS-units, $\zeta$ is of the order unity, and $\ln \Lambda$ is the Coulomb logarithm, which for thermal plasma has typical values between 5 and 8. For 1eV thermal atmospheric plasmas $\tau_{ee}$ is of the order $10^{-10}$ sec. A characteristic relaxation time closely related to $\tau_e$ is the slowing down time. Up to a numerical factor of the order unity and $T_e$ replaced by $E/k$, Eq. (2.10) also gives the slowing down time $\tau_s$ of suprathermal electrons ($E \gg kT_e$).

The statement that the EEDF is nearly Maxwellian can be translated into the condition that the equilibrium disturbing terms in Eq. (2.7) and (2.8) are much smaller than the $(\partial f_e/\partial t)_{el}^{ee}$ term. An order of magnitude of each term will be given in terms of characteristic times for energy transfer.

1. **Elastic electron-ion collisions.** The relaxation time $\tau_{ei}^e$ for energy transfer from electron to ions is $\tau_{ei}^e = (m_i/2m_e)\tau_e \gg \tau_e$. Due to the charge neutrality of the plasma, i.e. $n_e = n_i$, $(\partial f_e/\partial t)_{el}^{ei} \ll (\partial f_e/\partial t)_{el}^{ee}$ so that the
influence of the elastic e-i collisions on the maxwellization of the electron gas can be neglected. In Fig. 2.1 the relaxation frequency for energy transfer $\nu_{ei}^{\alpha}/n_a$ versus $E$(eV) is shown for different ionization degrees ($10^{-2}$ and $10^{-4}$) and $ln\Lambda$ is taken to be 10.

2. Elastic electron-atom collisions. The electron-atom collision time for energy transfer is given by $\tau_{ea}^{\alpha}(v) = [2(m_e/m_a)n(1)Q_{ea}^{m}(v)\nu]^{-1}$, where $Q_{ea}^{m}(v)$ is the e-a cross-section for momentum transfer. Figure 2.1 also shows $\nu_{ea}^{\alpha}$ versus $E$(eV) where the minimum is due to the Ramsauer minimum in the momentum transfer cross-section. The condition that the bulk of the electron distribution function is Maxwellian implies that the demand $\tau_e \ll \tau_{ea}^{\alpha}$ leads to a condition

$$\left( \frac{n_e}{n_a} \right) \gg 2.0 \times 10^{17} \left( \frac{2m_e}{m_a} \right) \frac{\hat{E}^2 Q_{ea}^{m}}{ln\Lambda} \equiv \alpha_{ea}(\hat{E})$$ (2.11)

for the ionization degree $\alpha$ where $\hat{E}$ is expressed in eV and $\alpha_{ea}$ is a critical ionization degree for which $\tau_{ea}^{\alpha} = \tau_e$ holds. In principle condition (2.11) must hold for the whole energy range. A critical ionization degree ($\alpha_{ea}^{\text{crit}}$) is obtained by putting the boundary value of this energy range into (2.11). The boundary value is given by the onset of inelastic collisions at the first excitation energy $E_{12}$. Substituting $\hat{E} = \hat{E}_{12} = 10$eV in (2.11) it is found that $\alpha_{ea}^{\text{crit}}$ equals $7 \times 10^{-6}$. The ionization degree in thermal plasmas varies from $10^{-4}$ to $10^{-1}$ so that the elastic e-a collisions will only have a negligible effect on the EEDF.
3. Inelastic e-heavy particle collisions. In literature much attention has been paid to the solution of the electron BTE including inelastic collisions [7, 1, 2, 3]. Shaw et al. [1] showed that for a broad variety of plasmas the main inelastic contribution comes from inelastic collisions involving the ground level, i.e. $e + A_1 \rightarrow e + A_{j>1}$ which is in accordance with Eq. (3.2) [8]. Further van der Mullen showed that the excitation processes of ions do not perturb the EEDF significantly due to the charge neutrality [9] so that we only have to consider the ground level excitation process. If the electron thermal energy is much smaller than the first excitation energy, i.e. $3kT_e/2 < E_{12}$, the analysis of the electron BTE can be simplified considerably. For the noble gases and atomic hydrogen $E_{12}$ is of the order 10 eV. In this way it is convenient to divide the EEDF into 2 parts, which are separated by the first threshold energy $E_{12}$. The first part represents the large bulk of the EEDF and is dominated by elastic collisions of the electrons. The second part represents the small tail which might be affected by the (de)excitation and ionization (recombination) processes of the atom and ion (cf. Fig. 2.3). Knowledge of the tail of the distribution function is needed in view of the rate of effective ionization of the ground level. To estimate the influence of inelastic collisions on the EEDF and on the ionization degree we shall only consider ionizing plasmas. In ionizing plasmas the excitation processes will dominate over the reverse processes, i.e. de-excitation to the ground level, since the excited states are underpopulated with respect to the Boltzmann population (Eq. (2.4)). An electron with energy $E > E_{12}$ will excite an atom and will lose an amount of energy equal to the excitation energy. The mean frequency of this excitation process, i.e. the depopulation
frequency of the tail of the EEDF, is calculated with

$$\left| \frac{\partial n_e^T}{\partial t} \right|_{1-2} \simeq n_e^T (\nu_{12}) = n_e n_a (\nu Q_{12}), \quad (2.12)$$

where

$$n_e^T = n_e \int_{E_{12}}^{\infty} f(E) dE \quad (2.13)$$

is the number of electrons in the tail and \( (\nu Q_{12}) \) is the rate coefficient for the ground level excitation. For a Maxwellian distribution we have

$$n_e^T = n_e \left[ \sqrt{\frac{E_{12}}{kT_e}} \exp \left( -\frac{E_{12}}{kT_e} \right) + \frac{\sqrt{\pi}}{2} \text{erfc} \left( \frac{E_{12}}{kT_e} \right) \right] \quad (2.14)$$

$$\cong n_e \sqrt{E_{12}/kT_e} \exp \left( -E_{12}/kT_e \right), \quad (2.15)$$

where \( \text{erfc}(x) \) is the complementary error function. A Maxwellian EEDF will prevail if the population frequency \( \nu_{ee}^T \) of the tail by means of elastic e-e collisions dominates over \( (\nu_{12}) \). An estimation of \( \nu_{ee}^T \) can be obtained by considering the reverse process in equilibrium, i.e. the rate at which a high energy electron loses energy due to collisions with bulk electrons \((1/\tau_s)\) (cf. Eq. (2.10)). A critical ionization degree \( \alpha_{inel}^{crit} \) can be defined by equating \( (\nu_{12}) \) and \( \tau_s^{-1}(E_{12}) \),

$$\alpha_{inel}^{crit} = 3.1 \times 10^{11} \frac{(\nu Q_{12})}{ln \Lambda} \hat{E}_{12} \hat{T}_e^{0.5} \exp \left( \hat{E}_{12}/\hat{T}_e \right), \quad (2.16)$$

above which the tail of the EEDF will be almost Maxwellian. In Fig. (2.2) \( \alpha_{inel}^{crit} \) is shown versus \( kT_e \). It is obvious that in the energy range of interest the inelastic collisions have much more effect on the EEDF than the elastic
collisions and that two regimes can be distinguished. In the low and intermediate ionization degree regime, $\alpha_{el}^{\text{crit}} \ll \alpha \ll \alpha_{inel}^{\text{crit}}$, the bulk of the EEDF will be still Maxwellian. However, the tail is depleted and will affect the rate coefficients for excitation and ionization of the ground level. In the “high” ionization degree regime, $\alpha_{inel}^{\text{crit}} \ll \alpha$, the whole EEDF will be Maxwellian.

Comparison with the $\alpha_{inel}^{\text{crit}}$ determined with Eq. 2.16 and the model of VLČEK in which the BTE is calculated self-consistently with a collisional-radiative model [3]. The calculations with this model shows that for a 0.8eV thermal argon plasma with ionization degree $\alpha = 3 \times 10^{-4}$ the effective ionization coefficient $S_{CR}$ is a factor 2 lower than the $S_{CR}$ calculated with a Maxwellian distribution function. Then for $\alpha > 3 \times 10^{-4}$ the rate coefficients for excitation and ionization of the ground level are not affected considerably by deviations from Maxwell in the tail [3, 10]. Since the ionization degree in the active zone, i.e. the ionizing region of the inductively coupled plasma is typically $10^{-3}$ we may retain the Maxwellian values of the rate coefficients.

Since laboratory plasmas are open systems gradients in these thermodynamic properties will be present. In order to estimate the importance of the macroscopic transport on the bulk of the EEDF we shall discuss each term on the left hand side of Eq. (2.1). The demand that the maxwellization processes will not be disturbed substantially induces boundary conditions for macroscopic parameters like the strength and frequency of the electric field and the gradient lengths. Depending on how the electric power is coupled into the plasma the electric field will have a DC component or a high frequency (RF) component. In the case of RF plasmas both the amplitude and the frequency of the electric field may influence the EEDF. Therefore the first and third term on the lhs of Eq. (2.1) are considered together.

4. Characteristic of external electromagnetic forces is that they diminish the chaotic behaviour of charged particles. In order to assure an isotropic EEDF, the collision time must be much smaller than the time needed to accelerate an electron to thermal velocities, i.e. $\tau_e \ll \nu_{th}/eE_f$, where $E_f$ is the electric field strength. A condition for $E_f$ can be derived, viz.

$$\frac{eE_f\lambda_{ee}}{kT_e} \ll 7 \times 10^{-14} n_e T_e^{-1} \ln \Lambda \equiv E_{f,\text{crit}} \quad \text{[V/m].}$$

(2.17)

It should be noted that criterion (2.17) addresses mainly the bulk of the EEDF. Under thermal plasma conditions $E_{f,\text{crit}}$ can be of the order $10^5$V/m. In the ICP and cascaded arc set-ups the amplitude of the electric fields are
of the order 1000V/m so that condition (2.17) is easily fulfilled. For RF generated plasmas, the question arises what the influence of the frequency is. When the angular frequency $\omega$ of the RF-field is much smaller than the electron-neutral collision frequency for momentum transfer, the effective RF-field in the electron BTE is equal to that associated with a DC-field with amplitude $E_f = E_0^{RF}/\sqrt{2}$ [11]. The influence of the RF nature of the E-field on the EEDF is then negligible and the electron transport coefficients can then be calculated with DC expressions. On the other hand when $\omega \gg (2m_e/m_a)\nu_{ea}$, the energy transfer from the electrons to the heavy particles does not change much during one cycle of the field oscillation so that the EEDF will remain stationary. To summarize, we shall consider RF-fields of which the frequency satisfies the following conditions

$$\nu_{eh}^e \ll \omega \ll \nu_{eh}$$  \hspace{1cm} (2.18)

In an atmospheric ICP $\nu_{eh}$ and $\nu_{eh}^e$ are of the order $10^{10}$Hz and $10^{5}$Hz, respectively. Condition (2.18) then implies an angular frequency in the range $1$MHz $< \omega < 1$GHz. In the ICP used in the ETP group [12, 13] the angular frequency $\omega = 2\pi \times 100$ MHz so that a stationary treatment of the EEDF can be used.

5. The demand that spatial inhomogeneities in the plasma as described by the second term on the lhs of Eq. (2.1) have little effect on the equilibrium restoring e-e collisions implies that

$$\tau_e \ll L_e / v_{the},$$  \hspace{1cm} (2.19)

where $L_e$ is a characteristic length to be specified. When this condition is met a local Maxwellian distribution function with small correction is possible [14, 15] so that a hydrodynamical approach is justified. By substituting Eq. (2.10) in Eq. (2.19) one finds

$$L_e \gg 1.4 \times 10^{17} n_e^{-1} T_e^2 (eV).$$  \hspace{1cm} (2.20)

In a plasma with low Mach number $L_e$ is a characteristic plasma dimension as determined by the boundary conditions and Eq. (2.19) will in general be satisfied in thermal plasmas. Globally we may distinguish between two main transport phenomena, namely convection and diffusion. The latter include apart from diffusion of particles also the conduction of heat. With each phenomena a macroscopic timescale can be introduced. The diffusion and convection timescales are

$$\tau_{dif} = \frac{L_e^2}{D_e} = \frac{L_e}{\lambda_{ee} v_{th}}, \quad \tau_{conv} = \frac{L_e}{v_{pl}}$$  \hspace{1cm} (2.21)
where $D_e$ and $v_{pl}$ are the diffusion coefficient and the plasma bulk velocity, respectively. Typical values are $L_e \simeq 4 \times 10^{-4} \text{m}$ [16] and $D_e \simeq 10^{-3} \text{m}^2/\text{sec}$ and $v_{pl} \simeq 10 \text{m/sec}$ so that $\tau_{\text{diff}} = 1.6 \times 10^{-4} \text{sec}$ and $\tau_{\text{conv}} \simeq 10^{-4} \text{sec}$. This is an important timescale in the macroscopic description of the plasma. If the particle flux by diffusion or convection is the dominant transport process it must be compensated by ionization and recombination. The relaxation times for ionization and recombination will be treated later.

With the aforementioned conditions the EEDF is almost Maxwellian. If the assumption is justified that the ion and neutral particle translational energy distribution function are also Maxwellian, then we can conclude that the plasma is in a state in which the translational equilibrium departures are small. Sufficient information of the plasma behaviour can be obtained from the macroscopic properties which are defined as (velocity) moments of the distribution function (Eq. (2.1)). In the next section the macroscopic transport equations are presented.

### 2.3 Basic equations

In a collision dominated multicomponent plasma each constituent is in partial translational equilibrium and must be described by its particle density, average velocity and temperature. The calculation of these thermodynamic quantities is based on the averaging of the thermal motion of the particles. It should be kept in mind that the thermal motion of a particle of species $a$ can be defined relatively to the mean velocity of species $a$, $\bar{V}_a$, or relative to the plasma bulk velocity $\bar{V}$ (cf. Eq. (2.31)). Therefore, two velocity representations can be used (cf Fig. 2.4),

$$\bar{v}_a^{(1)} \equiv \bar{V}_a + \bar{u}_a^{(2)} \equiv \bar{V} + \bar{c}_a.$$  \hspace{1cm} (2.22)

In the first representation the velocity is decomposed in a random component $\bar{u}_a$ relative to the mean velocity $\bar{V}_a$ for particle species $a$. By taking the average of Eq. (2.22) over the species $a$ the following relation between the species velocity $\bar{V}_a$ and the plasma velocity $\bar{V}$ is found

$$\langle \bar{v}_a \rangle \equiv \bar{V}_a = \bar{V} + \langle \bar{c}_a \rangle.$$  \hspace{1cm} (2.23)
In this subsection we consider the continuity, momentum and intrinsic energy equations for particle species $\alpha$ worked out for the $\alpha$ system. They are obtained by taking the zeroth, first and second moment of the Boltzmann transport equation Eq. (2.1) and using the first representation in Eq. (2.22). It is found that \[ \frac{\partial n_\alpha}{\partial t} + \nabla \cdot (n_\alpha \bar{V}_\alpha) = \langle \left( \frac{\partial f_\alpha}{\partial t} \right)_{CR} \rangle, \]  
(2.24) 

\[ \frac{\partial}{\partial t} (m_\alpha n_\alpha \bar{V}_\alpha) + \nabla \cdot (m_\alpha n_\alpha \bar{V}_\alpha \bar{V}_\alpha) + \nabla \cdot \bar{\pi}_\alpha^* + \nabla \cdot P_\alpha^* = n_\alpha \varepsilon_\alpha (\bar{E} + \bar{V}_\alpha \times \bar{B}) + \bar{\mathbf{R}}_\alpha, \]  
(2.25) 

\[ \frac{\partial}{\partial t} (n_\alpha \varepsilon_\alpha^*) + \nabla \cdot [n_\alpha \varepsilon_\alpha^* \bar{V}_\alpha] + P_\alpha^* \nabla \cdot \bar{V}_\alpha + \bar{\pi}_\alpha^* : \bar{V}_\alpha \bar{V}_\alpha + \bar{V}_\alpha \overline{q}_\alpha^* = Q_\alpha. \]  
(2.26) 

The electric $\bar{E}$ and magnetic field $\bar{B}$ must satisfy the Maxwell equations [18]. The right hand side of the continuity equation Eq. (2.24) represents the particle production and destruction rate due to collisional-radiative processes. The quantities $\overline{\pi}_\alpha^*$, $\varepsilon_\alpha^*$ and $\overline{q}_\alpha^*$ in the momentum Eq. (2.25) and the energy equation Eq. (2.26) are the viscosity tensor, the intrinsic energy per particle and heat flux. They are defined as 

\[ \langle \overline{\pi}_\alpha \rangle_{ij} = -m_\alpha \int \left( (\bar{u}_\alpha)_i (\bar{u}_\alpha)_j - \frac{u_\alpha^2}{3} \delta_{ij} \right) f_\alpha d\bar{u}_\alpha, \]  
(2.27) 

\[ \frac{3}{2} P_\alpha^* = n_\alpha \varepsilon_\alpha^* = \frac{1}{2} m_\alpha \int u_\alpha^2 f_\alpha d\bar{u}_\alpha, \]  
(2.28) 

\[ \overline{q}_\alpha^* = \frac{1}{2} m_\alpha \int u_\alpha^2 \bar{u}_\alpha f_\alpha d\bar{u}_\alpha. \]  
(2.29) 

The pressure $P_\alpha^*$ is related to $\varepsilon_\alpha^*$ by means of the equation of state for ideal gases (2.28). Note that in the intrinsic energy equation 2.26 the systematic kinetic energy related to $\bar{V}_\alpha$ is eliminated. The dynamical change of systematic kinetic energy can be obtained from the momentum equation. In the right hand sides of Eqs. (2.25) and (2.26) $\bar{\mathbf{R}}_\alpha$ and $Q_\alpha$ represent the exchange of momentum and heat between the different species by means of collisions, respectively (cf. chapter 6). The asterisk "*" means that these quantities defined in the $\alpha$ system are not partial quantities. By this it is meant that the unit surfaces across which a flux quantity is transferred, have different orientations for the different species so that for example $\sum_\alpha P_\alpha^*$ can not be considered as the exact total pressure. The reason is that in the "*" (or $\alpha$) system the thermal motion is defined relatively to the species averaged velocity $\bar{V}_\alpha$ which might be different for different species.
2.4 Equations for a conducting fluid

In the second, i.e. the plasma representation of Eq. (2.22) we first have to define the total mass density $\rho$ and the plasma bulk velocity $\vec{V}$. They are defined as

$$\rho = \sum_\alpha n_\alpha m_\alpha ,$$

(2.30)

$$\rho \vec{V} = \sum_\alpha n_\alpha m_\alpha \vec{V}_\alpha .$$

(2.31)

From Eqs. (2.22) and (2.31) we can derive

$$\sum_\alpha n_\alpha m_\alpha \langle \vec{c}_\alpha \rangle = 0 .$$

(2.32)

The electric field seen by charged particles in the plasma frame is

$$\vec{E}' = \vec{E} + \vec{V} \times \vec{B} .$$

(2.33)

In the plasma velocity representation all peculiar velocities $\vec{c}_\alpha$ are defined relative to the same bulk velocity $\vec{V}$. Then, for example, the total pressure $P$ is given by the sum of the partial pressures $P_\alpha$. The same holds for the viscosity tensor, intrinsic energy and heat flux. The definitions of these quantities in both velocity representations are interrelated by

$$\left( \frac{\rho a}{a} \right)_{ij} - \left( \frac{\rho a}{a} \right)_{ij} = n_\alpha m_\alpha \left( \langle (\vec{c}_\alpha)_i \rangle \langle (\vec{c}_\alpha)_j \rangle - \frac{1}{3} \langle c_\alpha \rangle^2 \delta_{ij} \right) ,$$

(2.34)

$$n_\alpha \epsilon_\alpha - n_\alpha \epsilon_* = \frac{1}{2} m_\alpha n_\alpha \langle c_\alpha \rangle^2 ,$$

(2.35)

$$\vec{q}_\alpha - \vec{q}^* = \frac{5}{2} \langle \vec{c}_\alpha \rangle P_\alpha + \frac{\vec{\pi}_\alpha}{c_\alpha} \cdot \langle \vec{c}_\alpha \rangle + \frac{1}{2} m_\alpha n_\alpha \langle c_\alpha \rangle^2 \langle \vec{c}_\alpha \rangle .$$

(2.36)

In a collision-dominated plasma these differences are unimportant. The reason is that the diffusion velocities $\langle \vec{c}_\alpha \rangle$ are proportional to the gradients of thermal properties which are assumed to be small (cf. §2.2.1 item 5). It is then justified to neglect the quadratic correction terms in Eqs. (2.34)-(2.36). The transport equations in the plasma bulk velocity frame are obtained by substituting Eq. (2.23) in Eqs. (2.24)-(2.26) and using the definitions in Eqs. (2.34)-(2.36), viz.

$$\frac{\partial n_\alpha}{\partial t} + \vec{V} \cdot (n_\alpha \vec{V}) + \vec{V} \cdot (n_\alpha \langle \vec{c}_\alpha \rangle) = \left( \frac{\partial f_\alpha}{\partial t} \right)_{CR} ,$$

(2.37)

$$\frac{\partial}{\partial t} \left( m_\alpha n_\alpha \langle \vec{c}_\alpha \rangle \right) + \vec{V} \cdot (m_\alpha n_\alpha \vec{V}(c_\alpha)) + \vec{V} \cdot \vec{\pi}_\alpha + \vec{V} \cdot P_\alpha +$$

$$m_\alpha n_\alpha \left[ \frac{\partial \vec{V}}{\partial t} + \left( \langle \vec{V} + \langle \vec{c}_\alpha \rangle \rangle \cdot \vec{V} \right) \right] = n_\alpha \epsilon_\alpha [\vec{E}' + \langle \vec{c}_\alpha \rangle \times \vec{B}] + \vec{R}_\alpha .$$

(2.38)
\[
\frac{\partial n_\alpha e_\alpha}{\partial t} + \vec{\nabla} \cdot (n_\alpha e_\alpha \vec{V}) + \vec{\nabla} \cdot \vec{q}_\alpha + m_\alpha n_\alpha \langle \vec{e}_\alpha \rangle \left[ \frac{\partial \vec{V}}{\partial t} + (\vec{V} \cdot \vec{\nabla})\vec{V} \right] \\
+ P_\alpha \vec{\nabla} \cdot \vec{V} + \vec{P}_\alpha : \vec{\nabla} \vec{V} = n_\alpha e_\alpha \vec{E} \cdot \langle \vec{e}_\alpha \rangle + Q_\alpha.
\]

(2.39)

We now have two systems of transport equations (2.24-2.26), and (2.37-2.39) for particles, momentum and intrinsic energy. In view of the computational model and the accompanying numerical solution method it is convenient to choose for the plasma system of transport equations. Hence our plasma model is based on Eqs. (2.37-2.39).

### 2.4.1 Continuity and momentum equations

Solving Eqs. (2.37-2.39) is still a complicated task. A more convenient way to handle Eqs. (2.37-2.39) is to take linear combinations of the various species components of Eqs. (2.37) and (2.38). Additional assumptions lead to simpler equations. The equations for the determination of the plasma bulk flow are obtained by summing Eqs. (2.37) and (2.38) over all species \(\alpha\), and are known as the Navier-Stokes equations and are given in Eqs. (2.40-2.41). The equations for the difference between the electron and ion component of Eq. (2.38)/summing will lead to the so-called generalized Ohm's law. The third equation generated by (2.38) describes the "sum" of the electron and ion components and is related to diffusion.

The global continuity equation, the equation for the bulk velocity and the electric current are [15, 19]

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

(2.40)

\[
\frac{\partial}{\partial t} (\rho \vec{V}) + \vec{\nabla} \cdot (\rho \vec{V} \vec{V}) = -\vec{\nabla} P - \vec{\nabla} \cdot \vec{\nabla} + \vec{J} \times \vec{B},
\]

(2.41)

\[
\vec{J} = \sum_\alpha n_\alpha e_\alpha \langle \vec{e}_\alpha \rangle,
\]

(2.42)

respectively. As stated before the Navier-Stokes equations (2.40-2.41) are obtained by summing (2.37) and (2.38) over all species \(\{\alpha = e, i, a\}\). Equation (2.42) merely is a definition equation. It should be noted that the definitions of the electric current density in both velocity representations are equivalent provided the plasma is neutral.

Equation (2.24) or (2.37) represents three equations for the electrons, ions and neutrals. In addition to the total continuity equation (2.40) two equations for the electrons and ions must be considered. Before discussing this point it is better to
consider first the momentum equations from which the diffusion velocities \( \langle \vec{c}_e \rangle \) and \( \langle \vec{c}_+ \rangle \) can be found. Only two instead of three equations generated by (2.25) or (2.38) must be used since the solution procedure of the transport equations is based on the total momentum equation (2.41) [20]. In order to calculate the Ohmic dissipation using the second velocity representation an equation for \( \vec{J} \) is needed so that the difference between the \( \langle \vec{c}_+ \rangle \) and \( \langle \vec{c}_e \rangle \) is known. This can be achieved by multiplying Eq. (2.38) with \( e_\alpha/m_\alpha \), summing over \( \alpha \), neglecting the viscous force terms and taking into account that \( m_e < m_+ \), which gives

\[
\frac{m_e}{n_e e^2} \frac{\partial \vec{J}}{\partial t} - \vec{E}' + \frac{1}{n_e e} \vec{J} \times \vec{B} - \frac{1}{n_e e} \vec{v} P_e + \frac{1}{n_e e} \vec{R}_e = 0. \tag{2.43}
\]

Equation (2.43) is known as the generalized Ohm's law and is applicable for DC as well as for RF plasmas. For negligible influence of the \( \vec{B} \)-field and close coupling between the ion- and neutral drift velocities the friction force \( \vec{R}_e \) between the electron and heavy particles can be written in the form [14, 17]

\[
\vec{R}_e = e n_e \frac{\vec{J}}{\sigma_e} - \beta n_e \vec{v} kT_e, \tag{2.44}
\]

where \( \sigma_e \) is the electron electrical conductivity and \( \beta \) is a numerical factor of order unity. Since the thermal friction is much smaller in the direction of the current it can be neglected.

For the ICP configuration in which the external electric field only has an RF-component the generalized Ohm's law can be simplified. It is assumed that the DC-component of \( \vec{J} \) can be neglected. Let \( \vec{F} \) denote the RF component of \( \vec{F} \). The friction force \( \vec{R}_e \) only has a contribution due to \( \vec{J} \). Substituting (2.44) in (2.43) it is found that \( \omega m_e \sigma_e / n_e e^2 \approx \omega / \nu_{eh} \ll 1 \) so that the first term on the lhs of Eq. (2.43) can be neglected. Typical values are \( \vec{J} \approx 10^6 \text{ A/m}^2 \), \( \sigma \approx 750/\Omega \text{m} \), \( \vec{B} \approx 10^{-3} \text{ Tesla} \), \( n_e \approx 10^{21} \text{ m}^{-3} \) so that \( | \vec{J} \times \vec{B} / e n_e | \ll | \vec{J} / \sigma | \). Collecting these results and taking into account that the plasma velocity \( \vec{V} \) and \( \vec{B} \) have more or less the same direction, Eq. (2.43) reduces to

\[
\vec{J} = \sigma \vec{E}. \tag{2.45}
\]

With Eqs. (2.41) and (2.43) still one equation is needed to specify \( \langle \vec{c}_e \rangle \) or \( \langle \vec{c}_+ \rangle \) which then can be used in Eq. (2.37). The ion component of (2.38) provides an equation for \( \langle \vec{c}_+ \rangle \) in principle [21] so that with the knowledge of \( \vec{J} \), \( \langle \vec{c}_e \rangle \) can be determined. At this point we use another approach by using the fact that in a collision-dominated plasma \( \langle \vec{c}_e \rangle \) and \( \langle \vec{c}_+ \rangle \) will be almost equal. This approach is used for ICP's and cascaded arc plasmas [22, 23]. The small difference between
\( \langle \bar{c}_e \rangle \) and \( \langle \bar{c}_+ \rangle \) which is proportional to the electric current density may not be neglected in the description of DC plasmas since the electric current is necessary to sustain the plasma. In order to maintain quasi-neutrality in the plasma, with the exception of a thin wall boundary layer whose thickness is of the order of the Debye length, the fluxes \( n_e \langle \bar{c}_e \rangle \) and \( n_+ \langle \bar{c}_+ \rangle \) must be equal. As a result we only have to consider one equation for the electrons or ions on the level of particle transport. In this regime of ambipolar diffusion the electron and ion particle diffusion are coupled by the onset of the ambipolar electric field. The particle diffusion velocities are then given by \([17, 19]\)

\[
\langle \bar{c}_e \rangle = \langle \bar{c}_i \rangle = D_A \frac{\bar{\nabla} (P_e + P_i)}{2P_i} = \frac{D_A}{2} \left( 1 + \frac{T_e}{T_i} \right) \frac{\bar{\nabla} n_e}{n_e} + \frac{D_A T_e}{2} \frac{\bar{\nabla} T_e}{T_e} + \frac{D_A}{2} \frac{\bar{\nabla} T_i}{T_i},
\]

(2.46)

where \( D_A \) is the ambipolar diffusion coefficient. In most cases the thermal diffusion in Eq. (2.46) can be neglected since the gradient length associated with \( n_e \) is much smaller than that associated with \( T_e \). Therefore, for thermal plasmas where the electron and ion temperatures are not too different we get

\[
n_e \langle \bar{c}_e \rangle = D_A \bar{\nabla} n_e.
\]

(2.47)

### 2.4.2 Energy equations

The intrinsic energy for a species \( \alpha \) depends on its internal degrees of freedom. As mentioned in section 2.2 electrons only have translational energy while the ASDF reflects the additional internal degrees of freedom. Since we deal with monatomic particles the electronic excitation is the remaining degree of freedom. In the multicomponent description of electron-dominated plasmas the intrinsic energy equation for the heavy particles can be formulated in such a way that the heavy particles only have translational energy. The intrinsic energy of excitation (de-excitation) and ionization (recombination) is then considered as an energy loss (gain) in the electron energy equation \([24]\). We have the following relation between the intrinsic energy and the temperature

\[
\epsilon_\alpha = \frac{3}{2} kT_\alpha.
\]

(2.48)

The finite extent of laboratory plasmas implies that energy conversion takes place, which results in non-equilibrium effects. The following scenario makes this more clear. In EM-sustained plasmas the electrons will pick up this EM energy. By means of collisions electrons transfer their energy to the heavy particles. An
important non-equilibrium feature is the difference between the electron and heavy particles temperatures in the regions of plasma creation and destruction. The very ineffective energy exchange due to the large mass ratio between the electrons and heavy particles is responsible for this temperature difference. The heavy particles transfer their energy to the environment by means of conduction and convection.

It is further assumed that due to the effective energy transfer between the ions and atoms, they have almost the same temperature. The intrinsic energy equation for the heavy particles is obtained by summing Eq. (2.39) over the heavy particles. The heavy particles will be denoted by the subscript \( h \). Since the energy of the particles is mainly thermal the fourth term on the lhs of Eq. (2.39) can be neglected. For the electrons the viscous dissipation term can also be neglected. The intrinsic energy equations for the heavy particles and electrons are

\[
\frac{\partial}{\partial t} \left[ \frac{3}{2} (n_e + n_a) kT_h \right] + \nabla \cdot \left[ \frac{3}{2} (n_e + n_a) kT_h \mathbf{v} \right] + \nabla \cdot \mathbf{q}_h + P_h \nabla \cdot \mathbf{v} + \overline{\mathbb{F}}_h : \nabla \mathbf{v} = Q_h, \tag{2.49}
\]

\[
\frac{\partial}{\partial t} \left[ \frac{3}{2} n_e kT_e \right] + \nabla \cdot \left( \frac{3}{2} n_e kT_e \mathbf{v} \right) + \nabla \cdot \mathbf{q}_e + P_e \nabla \cdot \mathbf{v} = \mathbf{J}_e \cdot \mathbf{E} + Q_e, \tag{2.50}
\]

respectively. To close the system of equations for mass, momentum and energy transport the heat flux \( \overline{\mathbb{q}}_\alpha \) and viscosity tensor \( \overline{\mathbb{F}} \) must be expressed in terms of \( n_\alpha \), \( \mathbf{v} \) and \( T_\alpha \). Since \( \overline{\mathbb{F}} \) and \( \overline{\mathbb{q}}_\alpha \) deal with momentum and energy transport as a result of spatial inhomogeneities they are expressed in gradients of \( \mathbf{v} \) and \( T_\alpha \) [25], i.e.

\[
\overline{\mathbb{F}}_{ij} = -\mu \left[ \left( \frac{\partial V_i}{\partial x_j} + \frac{\partial V_j}{\partial x_i} \right) - \frac{2}{3} \nabla \cdot \nabla \delta_{ij} \right], \tag{2.51}
\]

\[
\overline{\mathbb{q}}_\alpha = -\kappa_\alpha \nabla T_\alpha. \tag{2.52}
\]

Here \( \mu \) and \( \kappa_\alpha \) represent the molecular viscosity and thermal conductivity coefficients, respectively.

In chapter 6 explicit expressions for the particle and energy source terms and for the electron transport coefficients will be given as functions of \( n_\alpha \) and \( T_\alpha \). The physical model as presented in this chapter will be applied to ICP.

### 2.5 Final form

We conclude this chapter with an overview of the transport equations to be solved. Supplied with appropriate boundary conditions the system of equations (2.37, 2.40, 2.41, 2.49, 2.50) can be solved. With the expressions for the viscosity tensor (2.51) and the heat flux (2.52) these equations can be written down in
conservative form, i.e. they have the following structure

\[
\frac{\partial \rho \Phi}{\partial t} + \nabla \cdot (\rho \nabla \Phi) - \nabla \cdot (\mu \phi \nabla \Phi) = S^\phi
\]

\[
\frac{\partial n_\alpha \Phi_\alpha}{\partial t} + \nabla \cdot (n_\alpha \nabla \Phi_\alpha) - \nabla \cdot (\mu_\alpha \phi \nabla \Phi_\alpha) = S_{\alpha}^\phi.
\]  

(2.53)

The field variables $\Phi, \Phi_\alpha$ and the corresponding transport coefficients $\mu^\phi, \mu_\alpha^\phi$ are summarized in the following Table 2.1. The source terms $S^\phi, S_{\alpha}^\phi$ and the transport coefficients $\mu^\phi$ and $\mu_\alpha^\phi$ are function of all quantities as represented by $\Phi$, which make the coupled system of equations (2.53) non-linear. In conjunction with the

<table>
<thead>
<tr>
<th>$\Phi, \Phi_\alpha$</th>
<th>transport coefficient $\mu^\phi, \mu_\alpha^\phi$</th>
<th>Balance</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$D_A$ = diffusion coefficient</td>
<td>total continuity</td>
</tr>
<tr>
<td>$n_e$</td>
<td>$\mu$ = viscosity coefficient</td>
<td>electron balance</td>
</tr>
<tr>
<td>$\vec{V}$</td>
<td>$\kappa_e$ = thermal conductivity</td>
<td>total momentum balance</td>
</tr>
<tr>
<td>$T_e$</td>
<td>$\kappa_h$ = thermal conductivity</td>
<td>electron energy balance</td>
</tr>
<tr>
<td>$T_h$</td>
<td></td>
<td>heavy p. energy balance</td>
</tr>
</tbody>
</table>

Table 2.1. Field variables and transport coefficients in the transport equations (2.53).

boundary conditions the system of equations (2.53) must be solved numerically. Because of the conservative form of the equations the control volume method will be used. Chapter 7 is devoted to the numerical solution method.

References

Physical model


Chapter 3

A novel collisional radiative model with a numerical bottom and an analytical top

Abstract—A cut-off procedure for numerical collisional radiative (CR) models is developed and is based on knowledge of the excitation flow in the collision-dominated part of an atomic system as revealed by analytical models. This new cut-off technique makes it possible to reduce the number of levels in a numerical CR-model drastically in comparison with the older, numerical cut-off technique used by Bates et al., in which the truncation level was demanded to be in Saha equilibrium. The model is applied to an ionizing and recombining argon plasma as an example of a more general technique which can also be applied to other atomic systems. The results are in good agreement with the analytical top model developed previously in our group and also with the experiments and numerical calculations of Vlček and Pelikán.

3.1 Introduction

The atomic state distribution function (ASDF), which describes how atoms and ions are distributed over the various excited states, is an important feature of plasmas. The ASDF is related to basic plasma parameters such as the electron density $n_e$, electron temperature $T_e$ and groundstate density $n_1$ and contains information on the transport of radiation and particles.

Experimentally determinable by using well-established spectroscopic techniques, modelling of the ASDF is of renewed interest in the fields of spectrochemical analysis, recombination lasers, investigations of impurity compositions of thermonuclear plasmas, astrophysics, etc. This interest is based on the fact that comparisons of experimental data with analytical and numerical calculations may reveal important plasma properties. In the 1930’s, astrophysicists such as BAKER and MENZEL [1] studied analytically the spectra of recombining hydrogen systems in various astrophysical objects. Although the advent of the computer opened the

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new possibility of a numerical approach initiated by the paper of Bates et al. [2], there still remained interest in analytical treatments of the problem in various groups, e.g. Fujimoto [3], Biberman et al. [4] and van der Mullen et al. [5].

The advantage of the analytical approach is that it clarifies the influence of various features of plasmas, without the necessity of using tables and graphs. The disadvantage is that it cannot provide a general prediction of population densities of excited states under all possible conditions. In numerical approaches, we are not faced with this problem. However, in numerical models such as that proposed by Bates et al. [2], the number of excited states has to be very large since the upper state should be in partial local Saha equilibrium (pLSE) to ensure that the truncation does not affect the population of the lower levels [6].

In this paper, we combine analytical and numerical techniques in such a way that the lower excited states can be described numerically, whereas and analytical expressions are used for a careful truncation at the top levels. The procedure is based on the technique described in Ref. [6]. This more rigorous hybrid technique can be applied to ionizing as well as to recombining systems. The aim of this study is to reduce drastically the number of levels that must be taken into account so that the technique can also be used to calculate the particle source terms in plasma-transport models. In this study, the hybrid technique is applied to an argon plasma. Applications to other atomic systems will be essentially the same.

### 3.2 Theory

#### 3.2.1 Fundamental Equations

The ASDF is mainly determined by elementary collisional and radiative (CR) processes. The manner in which these processes manifest themselves in the ASDF is described by CR-models such as those given in Refs. [9, 8, 2, 4, 7]. The solution method, which is based on the fact that transport phenomena of atoms in excited levels can be neglected in a large variety of plasmas, is called the quasi-steady-state solution (QSSS). The atom and ion ground-level population densities, for which transport can not be neglected, are input parameters for the CR-models. We use the following notation: $n_1$ and $n_+$ are the densities of the ground level of a general $(Z-1)$-fold ionized atomic system and the subsequent ionic $(Z)$ system, respectively; $p$ is used to label the excited levels; if $p$ appears as a variable in a function, it represents the effective principal quantum number. The temperature and the densities $n_1$ and $n_+$ are the main input parameters for the CR-models. The densities $n_1$ and $n_+$ are determined by the following zeroth
moment of the Boltzmann transport equation:

\[
\frac{\partial n_1}{\partial t} + \nabla \cdot (n_1 \mathbf{w}_1) = \left[ \frac{\partial n_1}{\partial t} \right]_{\text{CR}} \tag{3.1}
\]

\[
\frac{\partial n_+}{\partial t} + \nabla \cdot (n_+ \mathbf{w}_+) = \left[ \frac{\partial n_+}{\partial t} \right]_{\text{CR}} \tag{3.2}
\]

where \( \mathbf{w}_1 \) and \( \mathbf{w}_+ \), are the mean velocities of the atom and ion ground levels, respectively. The rhs contains the changes due to CR processes.

Under a wide range of conditions, the relaxation time of the excited levels is much shorter than the relaxation time of the atom and ion ground levels. In addition, the mean thermal energy is assumed to be much less than the first excitation energy [4] so that, for the excited levels, the relation

\[
\sum_{p>1} n(p) \ll n_1, n_+ \tag{3.3}
\]

holds. Under these circumstances, the distribution of the densities of the excited levels is determined by the QSSS. This statement means that the excited levels follow the relatively slow changes (\(10^{-3} \sim 10^{-4} \text{ sec}\)) of the ground-level densities of the atom and the ion with relatively short time constants (\(10^{-7} \sim 10^{-8} \text{ sec}\)). Therefore, it is justified to neglect temporal and spatial relaxation for excited levels so that the QSSS of the ASDF is given by

\[
\left. \frac{\partial n(p)}{\partial t} \right|_{\text{CR}} = 0, \quad p > 1, \tag{3.4}
\]

for all excited levels, while \( n_1, n_+, n_e \) and \( T_e \) are input parameters. We will confine ourselves to plasmas to which Eq. (3.4) is applicable.

### 3.2.2 Basic assumptions

We will now describe an example showing how the analytical and numerical methods for an argon plasma are combined when the following processes are important. Applications to other systems and processes are straightforward. In our example, the following CR processes are taken into account:

1. Excitation and de-excitation by electronic collisions given by

\[
A(p) + e \leftrightarrow A(q) + e. \tag{3.5}
\]

The rate coefficients are, respectively, \( K(p, q) \) for the transitions from left to right and \( K(q, p) \) for those from right to left.
2. Ionization and three body recombination,

\[ A(p) + e \leftrightarrow A^+ + e + e. \]  

(3.6)

The rate coefficients are \( S(p) \) and \( K_+(p) \), respectively.

3. Bound-bound absorption and spontaneous emission,

\[ A(p) + h\nu_{pq} \leftrightarrow A(q). \]  

(3.7)

The rates are \( (1 - \Lambda_{qp})A(q,p)n(q) \) and \( A(q,p)n(q) \), respectively, where \( A(q,p) \) is the Einstein coefficient for spontaneous emission and \( \Lambda_{qp} \) the optical escape factor. This notation convention has the effect that the net result of emission and absorption transitions between \( q \) and \( p \) may be written as a modified decay rate \( n(q)\Lambda_{qp}A(q,p) \).

4. Photo-ionization and radiative recombination,

\[ A(p) + h\nu \leftrightarrow A^+ + e. \]  

(3.8)

The rate coefficients are \( A(p,+) \) and \( A(+,p) \), respectively. In this study, we will take into account only radiative recombination.

The density of a level will be denoted by \( n(p) \). In general, a level \( n(p) \) consists of \( g(p) \) states. The state density is denoted by \( \eta(p) = n(p)/g(p) \). We assume that all states in the same level are equally distributed [2]. Stimulated emission and transitions induced by ion-ion, ion-atom and atom-atom collisions will be neglected. We assume that this procedure is justified when the ionization degree \( \alpha \approx 10^{-3} \) [4]. In this example of using the cut-off procedure, we will use a Maxwellian electron energy distribution function (EEDF). However, this constraint is not really needed as long as the bulk of the EEDF is Maxwellian. The calculation of rate coefficients for transitions between excited levels is straightforward and the principle of detailed balance is valid for the forward and corresponding backward processes. Vlček and Pelikán [10] showed that for a broad class of plasmas, only the high-energy tail of the EEDF is affected so that our cut-off procedure has a wide range of applicability.

### 3.2.3 Solution of the problem

The population density \( n(p) \) of an excited level \( p \) is given by Eq. (3.4). Explicitly,

\[
\begin{align*}
    n_e \sum_{q \neq p} n(q)K(q,p) + \sum_{q > p} n(q)A(q,p)\Lambda_{qp} + n_e n_+ [n_e K_+(p) + A(+,p)] \\
- n(p) \sum_{q < p} A(p,q)\Lambda_{pq} - n_e n(p) \sum_{q \neq p} K(p,q) - n_e n(p)S(p) &= 0.
\end{align*}
\]  

(3.9)
In principle, Eq. (3.9) constitutes an infinite set of linear equations, whereas the summation runs over an infinite number of levels. In order to obtain the solution of Eq. (3.9) including the bottom levels, we write each component of Eq. (3.9) in the following form:

$$\sum_{q>1} C_{qp} n(q) = n_+ C_{+p} + n_1 C_{1p},$$

(3.10)

where the coefficients $C_{qp}$ are functions of $K(p, q)$, $A(p, q)$, $\Lambda_{pq}$, $T_e$, and $n_e$. It should be noted that $q = p$ is included in Eq. (3.10) and that $n_1$ and $n_+$ are considered to be independent variables. Therefore, the population density of a level $p$ is determined only by atom and ion ground-level contributions, i.e.

$$n(p) = n^+(p) + n^1(p),$$

(3.11)

where $n^+(p)$ is the solution of Eq. (3.10) if there are no ground-level atoms ($n_1 = 0$) and $n^1(p)$ is the solution of Eq. (3.10) when there are no ions ($n_+ = 0$). The values of the ground levels $n_+$ and $n_1$, which are not predicted by the CR-model, can be calculated by using Eqs. (3.1) and (3.2) or they may be determined experimentally. It is convenient to relate $n(p)$ to equilibrium quantities by relating $n^+(p)$ to the Saha population density and $n^1(p)$ to the Boltzmann population density given by

$$\eta^+(p) = r^+(p) \eta^S(p), \quad \eta^1(p) = r^1(p) \eta^B(p),$$

(3.12)

where

$$\eta^S(p) = \eta_e \eta_+ \left( \frac{\hbar^2}{2\pi m_e k T_e} \right)^{3/2} \exp(E_{pi}/k T_e)$$

(3.13)

and

$$\eta^B(p) = \eta_1 \exp(-E_p/k T_e).$$

(3.14)

For the electrons, we have $\eta_e = n_e/2$. The quantity $E_{pi}$ is the ionization energy of level $p$. The majority of the excited levels is oriented with respect to the ground level of the ion rather than with respect to the ground level of the atom because of the large energy gap between the ground level of the atom and the first excited level. An instructive notation can be found by dividing Eq. (3.11) by $n^S(p)$, viz.

$$b(p) = n(p)/(n^S(p)) = \eta(p)/\eta^S(p),$$

(3.15)

$$= r^+(p) + r^1(p) b(1),$$

(3.16)

where $b(p)$ is the overpopulation ($b(p) > 1$) or underpopulation ($b(p) < 1$) factor with respect to Saha. The population coefficients $r^+(p)$ and $r^1(p)$ reflect a purely
recombining and a purely ionizing plasma, respectively. In this framework, knowledge of the \( r^1(p) \) and \( r^+(p) \) population coefficients and of the CR rate coefficients enables us to determine the effective CR recombination (\( \alpha_{CR} \)) and ionization (\( S_{CR} \)) coefficients. These two-body rate coefficients are obtained by substituting Eq. (3.16) and the left-hand side (lhs) of Eq. (3.9) applied to \( p = 1 \) into Eq. (3.1). The result can be written as

\[
\frac{\partial n_1}{\partial t} + \nabla \cdot (n_1 \vec{w}_1) = n_e n_+ \alpha_{CR} - n_e n_1 S_{CR},
\]

(3.17)

in which

\[
\alpha_{CR} = n_e K_+(1) + A(+, 1) + \sum_{q>1} \frac{n^S(q) r^+(q)}{n_e} \{n_e K(q, 1) + A(q, 1) \Lambda_q 1\},
\]

(3.18)

\[
S_{CR} = \sum_{q>1} K(1, q) + S(1) - \sum_{q>1} \frac{n^S(q) r^1(q)}{n_e n^S(1)} \{n_e K(q, 1) + A(q, 1) \Lambda_q 1\}.
\]

(3.19)

Similarly, if one substitutes Eq. (3.16) and the lhs of Eq. (3.9), with \( p \) now replaced by the ion level \( n_+ \), into Eq. (3.2), one obtains

\[
\frac{\partial n_+}{\partial t} + \nabla \cdot (n_+ \vec{w}_+) = -n_e n_+ \alpha_{CR} + n_e n_1 S_{CR},
\]

(3.20)

where

\[
\alpha_{CR} = \sum_{q>1} \{n_e K_+(q) + A(+, q)\} - \sum_{q>1} \left\{ \frac{n^S(q) r^+(q)}{n_+} S(q) \right\},
\]

(3.21)

\[
S_{CR} = \sum_{q>1} \left\{ \frac{n^S(q) r^1(q)}{n^S(1)} S(q) \right\} + S(1).
\]

(3.22)

It is not difficult to show that Eqs. (3.18) and (3.19) are equivalent to Eqs. (3.21) and (3.22), respectively. Equation (3.18) expresses the total recombination coefficient as a sum of direct recombinaton to the atom ground level and de-excitation of that part of the population of excited levels which originates from the ion ground level. Equation (3.21) states that not every recombination to an excited level leads to effective recombination to the ground level. A part of the population of an excited level originating from the ion ground level is ionized again. In Eq. (3.19), not every excitation of the atomic ground level leads to effective ionization. A part of the population of the excited levels...
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originating from the ground levels is de-excited back to the ground level, i.e. 
\{n^S(q)r^1(q)/[n_e n^S(1)] \times [n_e K(q, 1) + A(q, 1)\Lambda_q]\}. The second expression 
for the ionization coefficient in Eq. (3.22) expresses this same result but in a 
complementary manner. In plasma-transport models, \(\alpha_{CR}\) and \(S_{CR}\) are important 
parameters.

3.2.4 The Boundary Conditions

Condition for Saha-equilibrium

In view of the main goal of this paper, i.e., a drastic reduction of the work required 
for calculation of the ASDF, it is important to know under what conditions the 
ASDF will approach its equilibrium value and how this approach is affected by 
the various processes. In other words, the question is how the various departures 
from equilibrium affect the ASDF.

Two primary disturbing phenomena may be distinguished which cause equi­
librium departures of the ASDF, namely, transport of radiation and transport of 
particles. The Saha density is the best candidate for the norm of the ASDF under 
equilibrium conditions. It can be proved that if the balance 

\[ A(p) + e \leftrightarrow A^+ + e + e \]  

(3.23)

is in equilibrium, then the population density of the excited level \(p\) obeys the 
Saha relation of Eq. (3.13). To state this differently, the density approaches the 
Saha value in the limit in which the ratio of the rates of the equilibrium-disturbing 
to the equilibrium-restoring processes tends to zero. Since the ionization and 
recombination rate coefficients increase for increasing \(p\) values, the Saha balances 
will approach the equilibrium state more and more closely for increasing \(p\). The 
upper levels for which Eq. (3.23) holds at equilibrium are said to be in pLSE (in 
the usual nomenclature the abbreviation pLTE is used). If a disturbing process 
has a radiative character, the demand for the presence of Saha equilibrium and 
thus the validity of the Saha formula (3.13) may be implemented by writing 

\[ \tilde{N}_e(p) \equiv \frac{n_e K(p)}{A(p)} \gg 1 \]  

(3.24)

where \(K(p)\) and \(A(p)\) refer, respectively, to the total collisional and radiative 
depopulation processes of level \(p\). The dimensionless quantity \(\tilde{N}_e\) represents 
the number of electron-induced transitions per radiative lifetime [6]. This well 
known criterion sets an \(n_e\)-dependent boundary \(p_{CR}\) in the atomic system for 
which \(\tilde{N}_e(p_{CR}) = 1\). It should be noted that the \(K(p)\) in the criterion of 
Eq. (3.24) includes the rate coefficients of all (de)excitation processes, of which
ionization is only a minor part. This result means that \( p_{CR} \) is not a boundary between radiative levels and pLSE but rather between radiatively- and collisionally-determined levels. It is the boundary between the corona balance (CB) and the excitation-saturation balance (ESB) in ionizing plasmas and between the capture radiative-cascade (CRC) balance and the de-excitation saturation-balance (DSB) in recombining plasmas [3, 9, 5].

The (de)excitation balances ESB and DSB differ only in the direction of the (de)excitation flow. This flow, which is important in the cut-off procedure, will be discussed presently. When we refer to the common properties of the (de)excitation flow, we use the abbreviation D/ESB.

The condition \( p > p_{CR} \), i.e. that levels are collisionally instead of radiatively dominated, is in most cases needed but not at all sufficient for the presence of pLSE [6]. A second demand for the presence of Saha equilibrium deals with the transport of particles. In an ionizing system in which the ground level \( n_1 \) is overpopulated with respect to the Saha-equilibrium value, i.e. \( b(1) \gg 1 \), there will be a net excitation flow \( J(p) \) over the excited levels in the atomic system to support the outward transport \( \vec{V} \cdot (n_+ \vec{w}_+) \) of charged particles. This flow can be described analytically for levels \( p \) for which stepwise excitation and de-excitation processes are dominant, i.e. if

\[
p > p_{hc} = Z \sqrt{Ry/kT_e \delta_e},
\]

where \( Ry \) designates the Rydberg energy and \( \delta_e \) represents a parameter depending on the atomic system [6]. For hydrogenic systems, the value of \( \delta \) is model-dependent (\( \delta_e = 1.5 \) in BIBERMAN et al. [4] and \( \delta_e = 3.83 \) in MANSBACH and KECK [11]). For non-hydrogenic systems, the level spacing may be smaller and \( \delta_e \) may be somewhat greater. In fact, \( p_{hc} \) defines a hot-cold boundary level for which the excitation and de-excitation processes are in balance [12]. This dependence of the (de)excitation kinetics on the temperature was discussed by BYRON et al. [12].

Concerning \( J(p) \cong \vec{V} \cdot (n_+ \vec{w}_+) \cong n_+ w_+ / \lambda_+ \) as a disturbance of the Saha balance, the second criterion for the presence of pLSE is

\[
\frac{n_e S(p) \lambda_+}{n_+ w_+} \gg 1,
\]

in which \( \lambda_+ \) is a characteristic geometrical length. It is noteworthy that this criterion is \( p \)-dependent. If the criterion in Eq.(3.26) is fulfilled, then the collision processes responsible for restoring Saha equilibrium for the level \( p \) dominate over stepflow [5] at level \( p \), which is generated by the particle transport. In contrast to the first or GRIEM criterion [13] in Eq. (3.24), the second criterion in Eq. (3.26)
is seldom considered. It should be stressed that especially for laboratory plasmas for which $\lambda_+$ may be rather small, Eq. (3.26) may be the most stringent criterion; the disturbing influence of particle transport on the ASDF is important [5, 9, 14]. This result means that the level $p_{es}$ for which $n_e S(p_{es}) = n_+ w_+ / \lambda_+$ may be very high. The cut-off technique of BATES et al. [2] is based on the presence of pLSE and is such that many more levels have to be taken into account than the maximum of $p_{CR}$ and $p_{es}$. In our cut-off technique, we take advantage of the fact that collision-dominated levels can be treated analytically if $p > \max(p_{hc}, p_{CR})$ by using the analytical top model (ATM) [5] so that we can lower the cut-off level to the first collisional hot level.

The Analytical Top Model (ATM)

The fact that the cut-off procedure can be used for ionizing as well as for recombining plasmas is based on what is called the complementary principle [6], as will be illustrated presently. In Eq. (3.16), we introduced the population coefficients $r_1(p)$ and $r_+(p)$. We first consider the situation of large $n_e$-values, for which the $r_1(p)$ and $r_+(p)$ coefficients are determined by electron collisions only. The $r_1(p)$ value is obtained by setting $n_+ = 0$. As a result, there can be no Saha equilibrium since the recombination process in Eq. (3.23) is missing. The population density of an excited level $p$ will be mainly determined by a so-called improper balance between excitation from level $p-1$ to $p$ and excitation from level $p$ to $p+1$ (Fig. 3.1a). By analogy, the $r_+(p)$ coefficient describes the ASDF for a purely recombining plasma. For sufficiently large $n_e$-values, all levels are collisionally dominated and a downward recombination flow is created. This flow is characterized by the reverse improper balance (Fig. 3.1b) such that the two flows cancel each other for the case that $b(1) = 1$ so that the relation $b(p) = r_1(p) + r_+(p) = 1$ should hold (see also Fig. 3.2) [7]. This complementary principle allows us to treat $r_1(p)$ and $r_+(p)$ in a complementary manner provided that $n_e$ is sufficiently
Figure 3.2. For the levels below $p_{CR}$, radiative decay is important. The densities of these levels must be calculated numerically. This procedure defines the boundary for the top levels. Above the boundary level $p_{CR}$, the excitation flow (a) or the de-excitation flow (b) are analytically determined. For increasing electron densities, $p_{CR}$ will decrease.

large so that, if $r^1(p)$ is known we immediately have $r^+(p) = 1 - r^1(p)$.

In the case of a purely ionizing, collisionally-dominated, hot system, it can be shown by using the ATM [5] that

$$r^1(p) = f(p) = b_0 p^{-x}. \quad (3.27)$$

The function $f(p)$ is the solution of a differential equation derived from the continuity equation for the excitation flow $J(p)$ in the excitation space, i.e.

$$\nabla \cdot \vec{J}(p) = R - I, \quad (3.28)$$

where $R$ and $I$ represent, respectively, the recombination and ionization sinks. The excitation flow $J(p)$ is described by stepwise collision processes and their rate coefficients. The exponent $x$ in Eq. (3.27) is determined by the competition between ionization and excitation. Its minimum value is 5 when there is no ionization and, in general, it takes a value of about 6 for ionizing [7] as well as for recombining plasmas [5]. It is interesting to note that a differential equation, comparable to Eq. (3.28), has been set up by SEATON in the treatment of planetary nebulae [15]. The overpopulation expressed in Eq. (3.27) decreases rapidly for increasing $p$ values. For a purely recombining, collision-dominated plasma, the ASDF is given by

$$r^+(p) = 1 - f(p) = 1 - b_0 p^{-x}, \quad (3.29)$$
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in view of the complementary principle. It can be shown that this solution satisfies Eq. (3.28) as well [6].

For decreasing $n_e$-values, the influence of radiative transitions at the bottom of the system will become increasingly important. However, Eqs. (3.27) and (3.28) can be retained for the top levels. The $b_0(n_e)$ value is then $n_e$-dependent. The boundary value $b_0$ in ionizing systems is determined by the boundary with CB and by the boundary with CRC for recombining systems. This power law for the collisional part of the ASDF has indeed been observed in various experiments (Fig. 3.3) [16, 7].

Summarizing, we may state that collisionally-dominated levels in both ionizing and recombining systems are described by the same differential equation (3.28). Only the boundary conditions are different. This information and especially knowledge of the step flow will be used to construct the cut-off procedure.

3.2.5 A Hybrid Cut-off Technique

For the collisionally-dominated part of the atomic system, an analytical solution may be derived for the ASDF for levels $p > \max(p_{hc}, p_{CR})$ in the ESB and pLSE. However, peculiarities in the lower part of the atomic system such as the presence of metastable and resonant states cannot be accounted for analytically.

In the numerical approach, we are not faced with these problems but with a complementary problem. In the ATM, a boundary condition for the ASDF is needed in the lower part of the atomic system, whereas for numerical models we need a boundary condition in the upper part to reduce the number of excited levels. The truncation of the atomic system at a certain cut-off level $N$ stops the (de)excitation flow and leads to erroneous pLSE-like results because, for the level $N$, there is only the Saha balance [17]. To overcome this problem, we combine the ATM with the numerical model. Knowledge of the (de)excitation flow $J(p)$ revealed by the ATM gives us a prescription for the upper boundary condition for the cut-off in numerical CR-models while, at the same time, the numerical model will give a lower boundary condition for the ATM. This cut-off procedure, in which we use the boundary value of an excitation flow rather than that of a level density, can be implemented in the well-known procedure given in the publication of Bates et al. [2]. The authors in Ref. [2] use as the boundary condition the requirement that there exists a cut-off level $N$ such that collisional ionization and recombination processes are dominant and in equilibrium. Stated in another way: in their cut-off procedure, it is assumed that the pLSE balance is valid for the cut-off level $N$. This level and all higher levels obey the Saha relation of Eq. (3.13), which is equivalent to the condition $b(u \leq N) = 1$ or $r^1(p) = 0$. In view of this, we group the terms involving the upper levels with the terms...
Figure 3.3. Relative overpopulation $\delta b(p)$ as a function of the effective principal quantum number for argon (van der MULLEN et al. [16]; $n_e = 1.7 \times 10^{19} \text{m}^{-3}$; o, $n_e = 6.7 \times 10^{19} \text{m}^{-3}$; , $n_e = 1.5 \times 10^{20} \text{m}^{-3}$; $T_e = 3$ to 5 eV.

Involving the continuum (see Ref. [6]). Then Eq. (3.9) becomes

\[
n_e n(p) \sum_{q \neq p}^{N} K(p, q) + n_e n(p) \left[ \sum_{u > N} K(p, u) + S(p) \right]
+ n(p) \sum_{q < p} A(p, q) \Lambda_{pq} - \sum_{1 < q < p} n_e n(q) K(q, p) - \sum_{q > p} n(q) A(q, p) \Lambda_{qp}
= n_e n_{+} [n_e K_{+}(p) + A(+, p)] + \sum_{u > N} n^S(u) A(u, p) \Lambda_{up}
\]
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\[ + \sum_{u>N} n_e n^S(u)K(u, p) + n_e n_1 K(1, p). \]  

(3.30)

With the principle of detailed balance, we can write this last equation in the form of an equation for \( b(p) \), viz.

\[
\begin{align*}
&n_e b(p) \left[ \sum_{q \neq p} K(p, q) + \sum_{u>N} K(p, q) + S(p) \right] \\
&+ b(p) \sum_{q<p} A(p, q) \Lambda_{pq} - \sum_{1<q<p} n_e b(q) K(q, p) - \sum_{q>p} \frac{b(q)n^S(q)}{n^S(p)} A(q, p) \Lambda_{qp} \\
&= \sum_{u>N} n_e K(p, u) + n_e [S(p) + n_+ A(+, p)/n^S(p)] \\
&+ \sum_{u>N} \frac{n^S(u)}{n^S(p)} A(u, p) \Lambda_{up} + n_e b(1) K(p, q). \\
\end{align*}
\]  

(3.31)

A curve drawn through the solution \( b(p) \) as a function of \( p \) must be tangential to \( b(p) = 1 \). If this is not the case, the excitation flow over the excited levels is mutilated by the cut-off. To avoid this problem, \( N \) should be increased. Depending on the conditions of the plasma, \( N \) can be very large. Many authors use this cut-off procedure [8, 18]. In this paper, we employ a different approach for the cut-off procedure. In our approach, the flow predicted by the ATM is used as the upper boundary condition.

The excitation flow across the cut-off level is mainly determined by step processes and may be written as

\[ J(N, N + 1) = n_e n(N) K(N, N + 1) - n_e n(N + 1) K(N + 1, N). \]  

(3.32)

Using the principle of detailed balance and the factor \( b(N) \), we find

\[ J(N, N + 1) = n_e n^S(N) K(N, N + 1)[b(N) - b(N + 1)]. \]  

(3.33)

The step flow across the cut-off level for the case when \( b(N + 1) \) is set equal to unity would be given by

\[ J^{\text{cut}}(N, N + 1) = n_e n^S(N) K(N, N + 1)[b(N) - 1] \]  

(3.34)

so that we may write

\[ J(N, N + 1) = J^{\text{cut}}(N, N + 1) \left[ \frac{b(N) - b(N + 1)}{b(N) - 1} \right] \]  

(3.35)
From analytical and experimental results, it is known that [6]

\[
\frac{b(N) - b(N + 1)}{b(N) - 1} = 1 - \frac{N^6}{(N + 1)^6} \equiv \gamma(N).
\] (3.36)

We note that the correction factor \( \gamma(N) \) tends to zero if \( N \to \infty \). If we rewrite the boundary flow as

\[
J(N, N + 1) = n_e K(N, N + 1) \gamma(N) [n(N) - n_s^N(N)],
\] (3.37)

it is clear that the factor \( \gamma(N) \) can be used to correct the rate coefficients involved at the cut-off level. When the first term on the rhs of Eq. (3.37) is added to the lhs of Eq. (3.31) and the second term to the rhs of Eq. (3.31), we find for the cut-off level \( N \) the following relation:

\[
n_e b(N) \sum_{q}^{N-1} K(N, q) + n_e b(N) \left[ \sum_{u \geq 2N} K(N, u) + S(N) + \gamma(N) K(N, N + 1) \right] + b(N) \sum_{q < N} A(N, q) \Lambda_{Nq} - \sum_{q > 1} \sum_{N} n_e b(q) K(N, q)
\]

\[= [n_e K(N, N + 1) \gamma(N) + n_e S(N)] + \frac{n_e n_s}{n_s^N(N)} A(+, N) + \frac{1}{n_s^N(N)} \sum_{u > N} n_s^N(u) A(u, N) + n_e b(1) K(N, 1).\] (3.38)

In this cut-off procedure, a simple correction of the rate coefficient for the cut-off level \( K(N, N + 1) \) enables us to lower the upper boundary level to almost the first collisionally-dominated level. This procedure drastically reduces the number of levels that must be considered.

### 3.3 The argon system and the rate coefficients

The numerical CR-model will be applied to an argon plasma composed of atomic Ar and singly-ionized Ar\(^+\) in the ground level. In the CR-models of Katsonis [19] and of Vlček and Pelikan [10], the Ar atom is divided into two subsystems according to the two core quantum states \( ^2P_{1/2} \) (primed system) and \( ^2P_{3/2} \) (unprimed system). In our Ar model, we also separate the two resonant and the two metastable levels of the 4s and 4s' groups in accord with the model of Vlček [18]. However, in view of our main goal to observe the effect of the cut-off procedure, it is not necessary to take into account the separate subsystems and
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an extensive electron-coupling scheme between the various groups in the neutral argon system. For levels higher than 4s, the two subsystems corresponding to \( nl \) and \( nl' \) are lumped together into one effective level as follows:

\[
\sum_n g_n E_n / \sum_n g_n .
\]  

(3.39)

The energy difference between \( nl \) and \( nl' \) in the two subsystems is almost constant and amounts to 0.18 eV. For plasmas with temperatures of 3eV, this difference will be negligible. The energy of the lumped effective levels, which are confined to the \( s, p, d, \) and \( f \) groups, is \( 1/3 \times 0.18 \) eV above the subsystem \( nl(2P_{3/2}) \), which agrees with the statistical weights of both subsystems. It is not difficult to extend our Ar model and to incorporate the special features of the two subsystems. In our computations, the Ar atom has at most 46 effective levels (see Table 3.1) which, because of the cut-off procedure, can be reduced to 14 effective levels.

<table>
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<th>Level number ( n )</th>
<th>Effective ( pqn )</th>
<th>Excitation energy eV</th>
<th>Statistical weights ( g_n )</th>
<th>Group</th>
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Since there is a lack of experimental data for electronic transitions between excited levels, semi-empirical (SE) expressions are required for the inelastic electron-atom collisions. DRAWIN [20] proposed SE formulae for the cross-sections based on extensions to the Bethe formulae [21]. These formulae contain parameters for optically-allowed, spin-allowed and parity-forbidden transitions between the excited levels and have been calculated for Ar by KIMURA et al. [22]. The computations of Ref. [10] are based on these data. In our study, we employ the SE excitation cross-section formulae proposed by VRIENS and SMEETS [23] for neutral hydrogen and alkali atoms. However, for electronic transitions between the ground level and the first 7 excited levels \( \{4s[3/2]_2, 4s[3/2]_1, 4s'[1/2]_0, \\
 \}

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Table 3.1. Data for the Ar-I system used in the model

3.3.1 Excitation cross-sections

\[
\begin{align*}
23 & \quad 6.994 & 15.481 & 84 & 480 & 7f \\
24 & \quad 7.317 & 15.505 & 36 & & 9p \\
25 & \quad 7.706 & 15.530 & 60 & & 8d \\
26 & \quad 7.845 & 15.538 & 12 & & 10s \\
27 & \quad 7.991 & 15.546 & 84 & 660 & 8f \\
28 & \quad 8.330 & 15.563 & 36 & & 10p \\
29 & \quad 8.668 & 15.578 & 60 & & 9d \\
30 & \quad 8.841 & 15.585 & 12 & & 11s \\
31 & \quad 8.997 & 15.591 & 84 & 864 & 9f \\
32 & \quad 9.337 & 15.603 & 36 & & 11p \\
33 & \quad 9.652 & 15.613 & 60 & & 10d \\
34 & \quad 9.856 & 15.619 & 12 & & 12s \\
35 & \quad 10.000 & 15.623 & 84 & 1098 & 10f \\
36 & \quad 10.348 & 15.632 & 36 & & 12p \\
37 & \quad 10.690 & 15.640 & 60 & & 11d \\
38 & \quad 10.875 & 15.644 & 12 & & 13s \\
39 & \quad 11.069 & 15.648 & 84 & 1344 & 11f \\
40 & \quad 11.327 & 15.653 & 36 & & 13p \\
41 & \quad 11.662 & 15.659 & 60 & & 12d \\
42 & \quad 11.841 & 15.662 & 12 & & 14s \\
43 & \quad 11.965 & 15.664 & 84 & 1620 & 12f \\
44 & \quad 12.362 & 15.670 & 36 & & 14p \\
45 & \quad 12.649 & 15.674 & 60 & & 13d \\
46 & \quad 12.958 & 15.678 & 84 & 1920 & 13f 
\end{align*}
\]
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4s'[1/2], 4p, 3d, and 5s}, experimental data sets of CHUTJIAN and CARTWRIGHT [24], TACHIBANA [25], and PETERTSON and ALLEN [26] are employed. These were also incorporated in the CR-model of VLČEK [18].

The radiative transition probabilities are taken from WIESE et al. [27]. For the ionization cross-sections, the SE formulae of Ref. [23] are employed as well. The SE formulae of Ref. [19] were used for radiative recombinations. The plasma will be transparent except for the resonance lines and free-bound radiation. For these levels, the escape factor in a plasma with cylindrical configuration with radius R is given by (HOLSTEIN [28], KLEIN [29])

\[
\Lambda_{1n} = g_0 T(R)
\]  

where \(g_0\) is of the order unity and the transmission factor [18, 29]

\[
T(R) = T_D \exp \left( -\frac{\pi T_{LD}^2}{4T_L} \right) + T_L \text{erf} \left( \frac{\sqrt{\pi T_{LD}}}{2T_L} \right),
\]

where \(T_D\) and \(T_L\) are, respectively, the transmission coefficients for Doppler-broadened and Lorentz-broadened spectral lines. The quantity \(T_{LD}\) is the transmission coefficient for a Voigt profile, i.e.

\[
T_D = \frac{1}{k_0 R \sqrt{\pi \ln(k_0 R)}}, \quad T_L = \frac{a}{\pi k_0 R},
\]

\[
T_{LD} = \frac{2a}{\pi \sqrt{\ln(k_0 R)}},
\]

where \(a\) is the ratio of Lorentz to Doppler broadening and \(k_0 R\) is the optical depth.

3.4 Results

3.4.1 An ionizing plasma

The first computations have been carried out for an ionizing Ar plasma under the following conditions:

\[
n_e = 6.7 \times 10^{19} \text{m}^{-3}, \quad n_1 = 10^{19} \text{m}^{-3}, \quad T_e = 3 \text{eV},
\]

for which the collisional-radiative and the hot-cold boundary are, respectively, given by

\[
p_{CR} \simeq 3.5, \quad p_{hc} \simeq 1.
\]
Chapter 3

Figure 3.4. The $r^1(p)$ coefficient as a function of the effective principal quantum number obtained with a double truncation. The plasma conditions are $n_e = n_i = 6.7 \times 10^{19} \text{m}^{-3}$, $T_e = 3\text{eV}$, $n_1 = 10^{19} \text{m}^{-3}$, $T_a = 1\text{eV}$; Δ, 46 effective levels; +, 34 effective levels; V, 26 effective levels; ◊, 14 effective levels.

This approach means that the principal quantum number of the cut-off level must be at least 3.5.

For a correct cut-off procedure, the ASDF must not depend on the number of excited levels. We will demonstrate the consequences of an inadequate cut-off and compare this procedure with the novel hybrid cut-off procedure. Figure 3.4 shows four calculations of the ground level contribution $r^1(p)$ of the ASDF as a function of the effective pqn with 14, 26, 34, and 46 effective levels, respectively, using an incorrect cut-off. Using the Ar-atom model of Table (3.1) with normal statistical weights for $s$, $p$, $d$, and $f$, namely, 12, 36, 60, and 84, means that a double truncation is executed: one truncation at the effective pqn side and one at the orbital quantum number side because $g$, $h$, . . . effective groups with $l \geq 4$ are not taken into account. As can be seen in Fig. 3.4, the population of the levels in the neighbourhood of the cut-off level are increased artificially. The reason is that the excitation flow $J(p)$ is truncated totally and thus incorrectly; it
Figure 3.5. The $r_1(p)$ and $\delta b(p)$ coefficients as functions of the effective $pqn$ obtained with the hybrid cut-off technique and the same conditions as in Fig. 3.4. The $r_1(p)$ calculated with 46, 34, 26, and 14 effective levels are represented as follows: (Δ) 46 effective levels; (+), 34 effective levels; (Ο) 26 effective levels; (◇) 14 effective levels. The $\delta b$ are represented as follows: (+), 46 effective levels; (Ο) 34 effective levels; (▽) 26 effective levels; (□) 14 effective levels, the experimental values (van der MULLEN et al. [16] are shown by (■). Curve: calculations of VLČEK and PELIKÁN [10].

becomes zero at the truncation level since for the cut-off level in this calculation, there is only the Saha balance left. The slope of the $r_1(p)$ coefficient varies from $-4.0 \pm 0.1 \ (N = 46)$ to $-2.5 \pm 0.2 \ (N = 14)$, whereas a constant slope of -6 is to be expected according to analytical and experimental results. The level 14 shows $r_1$-values of $2.0 \times 10^{-5} \ (N = 14)$ and $3.7 \times 10^{-6} \ (N = 46)$, which differ by a factor of 6. These calculations show that if $J(N)$ is suppressed, the overpopulation of the ASDF in ionizing systems decreases too slowly. As a
consequence, the ASDF depends also on the number of levels $N$.

To avoid the truncation at the orbital quantum number side, we include the $g$, $h$, ... by increasing the statistical weight of the $f$-group artificially, i.e. we use $12p^2 - 108$ as statistical weight of the $f$-group. The $12p^2$ stands for the statistical weight of the whole Ar energy shell with effective $pqn$, while $108 = 12 + 36 + 60$ represents the sum of the statistical weights of the $s$, $p$, and $d$ groups, as previously retained in the model. When the truncation of the orbital quantum number is removed, the slope of the $r^1(p)$ coefficient varies from $-4.7 \pm 0.1$ ($N = 46$) to $-2.8 \pm 0.2$ ($N = 14$) and is still too low.

In Fig. 3.5, the $r^1(p)$ and $\delta b(p) = b(p) - 1$ coefficients are shown as functions of the effective $pqn$ when the hybrid cut-off technique is applied. The slope shown in Fig. 3.5 is calculated with all 46 levels included. However, the first excited levels are not collisionally dominated but are influenced by radiative processes. The slope of the $r^1(p)$ for $p > p_{CR}$ varies from $-6.1 \pm 0.2 (N = 46)$, $-6.2 \pm 0.3 (N = 34)$ to $-5.7 \pm 0.5 (N = 26)$. For $N = 14$, it makes no sense to carry out a linear regression analysis for the collision-dominated part. But the ASDF as a whole, calculated with $N = 14$, coincides within 2% with the ASDF calculated with $N \geq 26$. Furthermore, the slope of $r^1(p)$ agrees well with the results predicted by the ATM and the experimental [16] data. It is clear that the slope of the $\delta b(p)$ coefficient is the same as the slope of the $r^1(p)$ as predicted by the theory [6] and found in experiments [16] and is independent of the number of levels. As can be seen, the level densities of the $d$ and $f$ groups are systematically a little too high. This result is probably due to the fact that we were using the VRIENS and SMEETS [23] cross-section formulae. These give large cross-sections for electron excitation between the $p - d$ and $d - f$ groups. Also shown are some experimental values measured in a hollow cathode-arc discharge [16] for the following plasma parameters: $n_e = 6.7 \times 10^{19} m^{-3}$, $n_1 = 10^{19} m^{-3}$, $T_e = 3$ to $5 eV$, and a radius of 1 cm. Our numerical calculations agree very well with the calculations of Ref. [10] (curve). In our calculations, it may be sufficient to take only 14 effective levels to calculate the constant $b_0$ in the analytical formula (3.27).

The two-body rate coefficients $\alpha_{CR}$ and $S_{CR}$ [see Eqs. (3.18) and (3.19)] are the same for all calculations with $N = 14, 26, 34, and 46$, namely,

$$S_{CR} = 2.11 \times 10^{-16} m^3 sec^{-1}, \quad \alpha_{CR} = 1.05 \times 10^{-19} m^3 sec^{-1}.$$

### 3.4.2 Recombining plasma

We have applied the hybrid cut-off procedure for a recombining plasma under conditions corresponding to an inductively-coupled plasma (see Nowak et al.
Figure 3.6. The $r^1(p)$ and $1 - r^+(p)$ coefficients as functions of the effective principal quantum number obtained with the hybrid cut-off technique. The conditions are $n_e = n_1 = 4 \times 10^{20} \text{m}^{-3}$, $T_e = T_a = 0.6 \text{eV}$ and $n_1 = 5 \times 10^{23} \text{m}^{-3}$. The $1 - r^+$ are represented as follows: (Δ) 46 effective levels; (+), 34 effective levels; (▲) 26 effective levels; (○) 14 effective levels. The $r^1$ are represented as follows: (+), 46 effective levels; (o), 34 effective levels; (△) 26 effective levels; (□) 14 effective levels.

For these conditions, the highest boundary value for $p$ is $\max(p_{hc}, p_{CR}) = p_{hc} \approx 3$. In Fig. 3.6, the $r^1(p)$ and $1 - r^+(p)$ coefficients are plotted as functions of the effective pqn. The slope of the $r^1(p)$ coefficient is $-5.9 \pm 0.2$ and is in good agreement with the expected value $-6$. The $1 - r^+(p)$ has a slope of $-4.8 \pm 0.1$ and seems to be too small in absolute value since the $r^+(p)$ and $r^1(p)$ should have
Figure 3.7. The $r^1(4p)$ as functions of the electron density ($T_e = 3\text{eV}$ and $T_\alpha = 1\text{eV}$). Results: --------, $n_1 = 10^{17}\text{m}^{-3}$; ---, $10^{18}\text{m}^{-3}$; ----, $10^{19}\text{m}^{-3}$; -----, $10^{20}\text{m}^{-3}$; -------, $10^{21}\text{m}^{-3}$; the curve A shows the calculations of VLČEK and PELIKÁN; [10] •, experimental results from van der SliDE et al.[31]

the same slope. In this numerical calculation, we did not take photo-ionization into account so that radiative recombination is not a balanced process. The slope of the $r^+(p)$ calculated without radiative recombination and photo-ionization amounts to $-5.4\pm0.2$ and this is in better agreement with the expected value of $x = -6$.

In this case, 14 effective levels are found to be sufficient to calculate $b_0$ and the remaining top part of the ASDF can be expressed analytically. The two-body rate coefficients $S_{CR}$ and $\alpha_{CR}$ are found to be

$$S_{CR} = 9.34 \times 10^{-24}\text{m}^3/\text{sec}, \quad \alpha_{CR} = 2.04 \times 10^{-19}\text{m}^3/\text{sec}.$$ 

3.4.3 Comparison with another numerical model and with experimental data

In Fig. 3.7, the population coefficient $r^1(4p)$ calculated with $N = 26$ is shown as a function of $n_e$ for $T_e = 3\text{eV}$ with different ground-level densities ($n_1 = 10^{17}$ to $10^{21}\text{m}^{-3}$). The indicated range corresponds to an opacity range from optically-
thin to optically-thick plasmas for resonant radiation. The plasma column has a radius of 0.01 m. For low and intermediate $n_e$-values, the $r^1(4p)$ is proportional to $n_e$ in the CB, as can be proved by analytical calculations [6, 7]. For high $n_e$-values, the $r^1(4p)$ saturates, i.e. it is completely dominated by electronic collisions in the ESB since both production from lower and destruction to a higher level are proportional to $n_e$. The numerical results of the extensive 65-level model of Ref. [18] for the single $4p'[1/2]_1$ are also shown. It should also be noted that the calculations have been carried out with 4p representing a whole group. If we take into account the energy shift of the effective 4p and $4p'[1/2]_1$ levels, the result lies systematically 35% above the result of Ref. [10]. This difference may at least partly be due to different cross-section data (see Ref. [20] for the work of Ref. [10] and Ref. [23] for ours). Also shown are the experimental values of $r^1(4p'[1/2]_1)$ of the 4p group in Ar I obtained in a plasma column generated by a hollow cathode-arc discharge measured by van der SIIDE et al. [31]. Our computations agree well with these experimental data. For a comparison with other numerical CR-models [19, 17], we refer to Ref. [10].

3.5 Conclusions

The use of an inadequate cut-off in numerical CR-models results in an incorrect ASDF. In this work, a numerical CR-model with a hybrid cut-off procedure [6] has been applied to an ionizing plasma (hollow cathode-arc discharge) and a recombining plasma. The main conclusions which can be drawn from our computational results are the following:

1. The hybrid cut-off technique, based on knowledge of the analytical excitation flow [6, 5], may be used successfully in reducing the number of excited levels below the number of levels needed in the cut-off technique proposed in Ref. [2]. With this new technique, the computational effort required to calculate the two-body ionization and recombination rate-coefficients and the ASDF is relatively small.

2. In calculations of the ASDF, it is sufficient to deal with only 14 effective levels for an ionizing as well as a recombining Ar plasma.

3. The calculated ASDF agrees well with the experimental values of Ref. [31] for an ionizing plasma.

4. The $r^1(p)$ for the whole 4p group calculated with 26 effective levels is typically a factor of 1.35 greater than the $r^1(4p)$ for the $4p'[1/2]_1$ level calculated with an extensive 65-level Ar-model [10]. This difference may be due to the fact that the calculations of Ref. [10] are based on excitation cross-sections of Refs. [20] and [22] and not on Ref. [23] as are ours.
Acknowledgement — We would like to thank the Institute for Continuing Education (IVO) for financial support of D. Benoy. These investigations in the program of the Foundation for Fundamental Research on Matter (FOM) have been supported by the Netherlands Technology Foundation (STW). The calculations were performed on an IBM RT 6150 RISC WS, as a part of a joint study agreement with IBM NV Netherlands.

References

Chapter 4

Application of a Hybrid Collisional Radiative Model to Recombining Argon Plasmas

Abstract - A collisional radiative model, in which a hybrid cut-off technique is used, is applied to recombining plasmas to study the atomic state distribution (ASDF) and the recombination coefficient. Computations of the ASDF using semi-empirical rate coefficients of Vriens and Smeets (V-S) and Drawin (D) are compared with experimental values measured at various positions in a free expanding argon arc jet. Apart from the shock position, where the calculated results are too low, the model calculations are higher than the experimental results. The volumetric recombination coefficient has a $T_e^{-4.2}$ and a $T_e^{-4.8}$ dependence when semi-empirical rate coefficients of, respectively, $V-S$ and $D$ are used. The differences between the models based on the rate coefficients of $V-S$ and $D$ indicate that the recombination flow is sensitive to the low temperature behaviour of the rate coefficients.

4.1 Introduction

Knowledge of the atomic state distribution function (ASDF) is of fundamental importance in the field of plasma spectroscopy, recombination lasers, plasma-transport models, astrophysics, and the study of impurities in thermonuclear plasmas. The ASDF describes how excited states in atoms and ions are populated in relation to a given electron temperature $T_e$, neutral ground-state density $n_1$ and electron density $n_e$. With this function it is possible to calculate the coefficients of total recombination and ionization [1], which are required for the particle- and energy source terms in plasma-transport equations [2]. At present, there is no general analytical solution for the ASDF under various conditions. On the other hand, several calculations exist for specific density and temperature ranges where various sets of semi-empirical expressions for the cross sections related to electron induced transitions are used.

In a recent paper of Benoy et al. [3], a new technique was developed in

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which the analytical solution for the upper part of atomic systems can be used to simplify the numerical part of the ASDF of argon drastically. This so-called hybrid technique based on a combination of analytical and numerical techniques, as described by Van der MULLEN [4], makes it possible to reduce the number of atomic levels needed in a numerical model drastically [5]. As a result the computational effort is minimized so that the hybrid technique is very suitable for the determination of the source terms in transport models. Moreover, it is a practical method for a comparative study of ASDF's generated by different sets of semi-empirical rate coefficients.

In Ref. [3] the hybrid technique has been applied to conditions corresponding to ionizing plasma regions [6, 8, 7]. In the present paper, the characteristics of a recombining argon plasma are studied using two different hybrid CR-models which differ from each other with respect to the set of semi-empirical rates. One of the models is based on formulae published by VRIENS and SMEETS [9] and the other uses the rates as given by DRAWIN [10]. In both cases the ASDF as well as the recombination coefficients are calculated. The result of both hybrid models will be compared with experimental results obtained from a cascaded, arc-created expanding plasma. The comparative study will reveal substantial differences between the two different models with respect to the ASDF as well as the coefficient of total recombination. This is essentially based on the fact that the kinetics of a cold recombining plasma are determined by the unknown threshold behaviour of electron collisions. The experimental results are not suitable for selecting one of the sets of semi-empirical rate coefficients.

We will start with an outline of the hybrid technique.

4.2 The hybrid cut-off technique

4.2.1 The general set-up of the Collisional-Radiative models

The calculation of the two different sets of ASDFs is done using numerical collisional-radiative (CR) models, as initiated by BATES et al. [11]. The number of levels used in the CR models is reduced drastically by providing the cut-off level with a stepwise ionization/recombination flow as prescribed in Ref. [4] and worked out in Ref. [3] for an ionizing argon system. The construction of a model in which the lower part of the atomic system is calculated numerically while the upper part obeys an analytical relation, is denoted by the hybrid model. This will be described in Sec. 4.2.2. This section is devoted to the general set-up of CR models.

Under quasi-steady state (QSS) conditions, i.e. when the CR-relaxation times for excited levels ($10^{-7} \sim 10^{-8}$ sec) are much shorter than the hydrodynamical
relaxation times \((10^{-3} \sim 10^{-4} \text{ sec})\), the equation for the population density \(n(p)\) for excited levels reads

\[
\frac{\partial n(p)}{\partial t} \bigg|_{CR} = 0, \quad p > 1. \tag{4.1}
\]

The symbol \(p\) is used to number the levels in an atomic system or it represents the effective principal quantum number \((\text{pqn})\) [4]. The densities \(n_1, n_+\), the electron density \(n_e\) and the electron temperature \(T_e\), which are the main input parameters for CR-models, follow from experiments [6, 8, 7] or from plasma-transport models. In the CR-model the following CR processes are considered: electron-heavy particle inelastic collisions, line radiation and radiative recombination. The absorption of resonant radiation and the photo-ionization are described with an escape factor. The effects of stimulated emission and inelastic collisions between heavy particles will be neglected. To calculate the rate coefficients for electron-induced transitions, a Maxwellian electron energy distribution function (EEDF) is used. The argon system of levels being used is described in Ref. [3]. As stated before, we will construct two different CR models which only differ with respect to the rate of excitation processes of excited levels. The model denoted by \(V\) uses the rates of VRIENS and SMEETS [9] whereas model \(D\) is based on cross-sections of DRAWIN [10]. In these semi-empirical expressions, known values for the optical transition probabilities of WIESE et al. [12] or hydrogen-like values are implemented [4]. The remaining part of the models is the same. Experimental data of TACHIBANA are used for the ground-state excitation [13], whereas the escape factor for radiative transitions to the ground-state employs a model of KLEIN [14]. For all other radiative transitions, the plasma is taken to be optically thin. For radiative recombination we use data of KATSONIS [15].

The population density of an excited level \(p(2, \ldots, N)\) can be described by the following equation:

\[
\sum_{q > 1}^{N} C_{qp} n(q) = n_+ C_+ + n_1 C_{1p}. \tag{4.2}
\]

The coefficients \(C_{qp}\) are known functions of the rate coefficients, \(T_e, n_e,\) and escape factors. The solution of Eq. (4.2) can be written in standard form, i.e. in terms of a ground-state \(n^1(p)\) and an ion contribution \(n^+(p)\), since Eq. (4.2) is a linear set of equations, viz.,

\[
n(p) = n^+(p) + n^1(p). \tag{4.3}
\]

It is convenient to relate the ground-state contribution to the corresponding Boltzmann population \(n^B(p)\), \(n^1(p) = r^1(p)n^B(p)\) and the ion contribution to the
corresponding Saha population \( n^S(p) \), \( n^+(p) = r^+(p)n^S(p) \), where

\[
\frac{n^B(p)}{g_p} = \frac{n_1}{g_1} \exp(-E_{1p}/kT_e),
\]  

(4.4)

\[
\frac{n^S(p)}{g_p} = \frac{n_e n_+}{2g_+} \left( \frac{\hbar^2}{2\pi m_e kT_e} \right)^{3/2} \exp(E_{p+}/kT_e).
\]  

(4.5)

In these formulae, \( E_{1p} \) is the excitation energy, \( E_{p+} \) the ionization energy of level \( p \) and \( g_p \) the statistical weight of level \( p \). The coefficients \( r^+(p) \) and \( r^1(p) \) are the population coefficients for a purely recombing and ionizing plasma, respectively. Another instructive expression for the excited state population density is obtained when Eq. (4.3) is divided by \( n^S(p) \), i.e.

\[
b(p) = r^+(p) + r^1(p)b(1),
\]  

(4.6)

where \( b(p) = n(p)/n^S(p) \). A level \( p \) is said to be over or under-populated with respect to Saha when \( b(p) > 1 \) or \( b(p) < 1 \), respectively.

4.2.2 The Cut-off Procedure

An important method to simplify a CR-model is the reduction of the number of levels. In the model of Bates et al. [11] a cut-off of the atomic system is used at a level which is supposed to be in Saha equilibrium. Because of the increasing collisional rate coefficients for ionization and recombination for increasing \( p \) values, the higher levels in an atomic or ionic system reach Saha equilibrium more easily than the lower levels. A level \( p \) and all higher-lying levels are said to be in partial local Saha equilibrium (pLSE) if they are populated according to Eq. (4.5) which is equivalent to \( b(p) = 1 \). However, such a level can be very high in the atomic system so that the number of levels to be treated in the CR-model must be large. Moreover, small deviations from the Saha density as given by Eq. (4.5) may cause large excitation flows in the upper part of the system which will affect the ionization or recombination coefficients and thus the density of lower levels. This is due to the fact that the rate coefficient for stepwise excitation scales with \( p^4 \).

The discussion of finding the lowest level in pLSE is often guided by the GRIEM criterion [16] which states that for a level \( p \) in pLSE the total collisional depopulation should be larger than the total radiative depopulation. This condition defines a critical level \( p_{cr} \) dependent on \( n_e \), which is a boundary level between the radiative and collisional dominated part of the atomic system. Explicit expressions for \( p_{cr} \) have been derived for hydrogen-like systems in Refs. [17, 18]. However,
although GRIEM's condition is nearly always needed, it is hardly ever sufficient for the presence of pLSE. There are two situations in which the incompleteness of this condition can be demonstrated. First, as can be seen from Eq. (4.5), $b(p)$ depends on the magnitude of $b(1)$, so that when $b(1) \gg 1$ in an ionizing plasma, the excited level populations are governed by the ground-state population. Apart from an $n_e$ criterion, $n_1$ must also be specified. The second situation deals with cold recombining systems where, although $b(1) \ll 1$, even large $n_e$-values can not prevent that the ASDF differs substantially from Saha equilibrium. This result is caused by the fact that for low $T_e$-values, the de-excitation of lower levels supercedes excitation due to the fact that electrons are missing sufficient translational energy for excitation [16]. The result implies that apart from $n_e$ and $b(1)$, we must also put a demand on the $T_e$-value in order to mark out the validity regime of pLSE. This $T_e$-related boundary condition can be obtained by the requirement that for the boundary level the excitation process $p \rightarrow p + 1$ has the same probability as the de-excitation process $p \rightarrow p - 1$, which leads to the so-called hot-cold boundary level [19]

$$p_{hc} = Z\sqrt{Ry/kT_e\delta},$$

(4.7)

where $Ry$ is the Rydberg energy and $\delta$ represents a parameter which depends on the atomic system and the adopted theory. For hydrogen-like systems, BIBERMAN et al. prescribe $\delta = 1.52$ [20]; in the model of MANSBACH and KECK, $\delta = 3.83$ [21]; in the work of FUJIMOTO, $\delta = 3$ [1, 17].

When GRIEM's condition and $p > p_{hc}$ are fulfilled, the system is ruled by the so-called (de)excitation saturation balance (DSB/ESB) and is dominated by stepwise processes. This stepwise (de)excitation flow over the collision-dominated excited levels can be described analytically both for ionizing and recombining systems. The change of the excitation flow over the system of levels can be described using a continuity equation in excitation space [4]. The difference between the recombining and ionizing case is then reduced to a difference in boundary condition. In both cases it can be shown that the Saha decrement $\delta b(p) = b(p) - 1$ scales with $|\delta b(p)| \sim p^{-6}$ which means for the ionizing case that [4, 17]

$$\delta b(p) \simeq r^1(p)\delta b(1) \sim p^{-6}. \quad (4.8)$$

On the other hand, for recombining systems, the relation

$$- \delta b(p) \simeq 1 - r^+(p) \sim p^{-6} \quad (4.9)$$

holds. This is closely related to the fact that for sufficiently large $n_e$-values, the superposition of ionization and recombination flows of the same magnitude gives
the relation $r^1(p) + r^+(p) = 1$, which is the so-called complementary property of $r^1(p)$ and $r^+(p)$ [4, 17]. In the hybrid cut-off technique, the analytical expressions (4.8) and (4.9) are used for the cut-off level to construct the (de)-excitation flow. These provide the coupling of the highest level with the continuum. In ionizing systems, this is the stepwise ionization while in recombining systems it is the stepwise recombination. It should be noted that in the hybrid cut-off technique an excitation flow is used as boundary condition rather than a population density as in Ref. [11]. The principal quantum number at cut-off $N$ in Eq. (4.2) should be at least $N \geq \max(p_{hc}, p_{cr})$. This number is much smaller than that prescribed by the method of BATES et al. [11]. In the CR-models for Ar of KATSONIS [15] and VLČEK and PELIKÁN [22], cut-off principal quantum numbers of 50 and 19 are used, respectively, while in our CR-model $N = 13$. Depending on the conditions the number of levels can even be reduced. In Sec. 4.4, the hybrid cut-off technique is compared with the technique proposed in Ref. [11]. We shall call the technique in which the cut-off level $N$ has only an ionization recombination channel a stagnation cut-off. The reason is that when $b(N) \neq 1$ the excitation current is obstructed. For obvious reasons, we will denote the new technique as the conductive cut-off.

### 4.3 Cascaded arc-created expanding plasma

In Ref. [3] the hybrid cut-off technique has been applied to specific (ionizing) plasma conditions according to the work of van der MULLEN [4, 8, 7]. This study is focussed on recombining systems which can be found in non-current carrying plasmas or plasma regions such as afterglows, outer regions of flames [6], recombination lasers and so on. Our main attention goes to the recombining regions of a freely-expanding plasma generated by an cascaded arc. This plasma source can be applied in the field of light source technology [23], deposition devices [24] and particle sources which are important in thermonuclear research [25]. For a successful application of the cascaded arc and from a fundamental point of view, the physical state of the plasma has to be understood. It has been the subject of a large number of investigations [22, 26, 27]. In the study of TIMMERMANS et al. [26] and the CR-model calculations of VLČEK and PELIKÁN and references therein [22], the nonequilibrium processes in the cascaded arc are investigated. Typical plasma parameters in the high pressure cascaded arc are $n_e = 10^{21} \sim 2 \times 10^{23} \text{ m}^{-3}$, $n_1 = (0.2 \sim 1) \times 10^{24} \text{ m}^{-3}$ and $T_e \approx 10000 \text{ K}$ [28].

Studies of KIMURA et al. [28] and LIMBAUGH [29] were devoted to the elucidation of nonequilibrium and non-ideal effects in the freely expanding plasma jet. Recent spectroscopic measurements of van de SANDEN [30] supplied us with
Application of a hybrid collisional radiative model ...

reliable data of the local plasma parameters at various positions in the expanding plasmajet. Values for the plasma parameters at axial positions $z/D > 4$ from the exit of the cascaded arc, where $z$ is the distance to and $D$ is the diameter of the arc orifice, are $n_e < 10^{20}\text{m}^{-3}$, $n_1 \leq 10^{21}\text{m}^{-3}$ and $T_e \leq 3500\text{K}$. The measurements were performed with an arc current of 45 A and an argon flow of 58 scc/sec. The plasma flows through an orifice of $D = 4\text{mm}$ and the low pressure background is about 40 Pa.

4.4 Results

4.4.1 The atomic state distribution function

In this subsection the experimental results of Ref. [30] will be used to study the influence of the rate coefficients, radiative decay and radiation trapping on the ASDF of an expanding plasma jet. The expanding plasmajet is a low temperature argon plasma with relatively high $n_e$ values (cf Table 4.1). Only for the 4s and 4p

<table>
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<tr>
<th>Z (mm)</th>
<th>$T_e(K)$</th>
<th>$n_e(\text{m}^{-3})$</th>
<th>$n_1(\text{m}^{-3})$</th>
<th>$T_e(K)$</th>
<th>$n_e(\text{m}^{-3})$</th>
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<td>$6.6^{20}$</td>
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Table 4.1. Values of electron temperatures (K), electron and neutral particle densities at various axial (Z) and radial (R) positions in the expanding plasma arcjet. The notation $4.4^{19}$ means $4.4 \times 10^{19}$.

levels will the radiative decay compete with the electron-induced transitions. For higher states the electron-induced de-excitation process will be dominant. These levels are in the so-called de-excitation saturation balance [4]: the level population $n(p)$ is a result of balance between de-excitation of higher levels giving rise to production of $n(p)$ and de-excitation from $p$ to lower levels. A level $p$, for which the condition $p > \text{max}(p_{cr}, p_{hc})$ holds, will be populated according to Eq. (4.9). The lower lying levels have to be calculated with the hybrid cut-off technique, taking apart from the electron-induced transitions the effect of radiation and radiation trapping into account. In the calculations, 46
effective levels are included which corresponds to a cut-off at principal quantum number \( N = 13 \). In the CR-model all 4p and 4p' levels have been lumped into one effective 4p level.

Table 4.1 gives an overview of the local values of the plasma parameters in the expanding plasmajet for which the CR-model is applied. The axial positions \( z = 20 \) mm and \( 40 \) mm correspond to positions before and in the shock front, while the position at \( z = 70 \) mm is located behind the shock front. Line-emission measurements are used to determine the population densities \( n(p) \) of the excited states listed in Table 4.2. A tomographic technique is used to Abel-invert the

<table>
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<th>spectroscopic notation</th>
<th>( \lambda ) (nm)</th>
<th>( E_{p^+} ) (eV)</th>
<th>( g_p )</th>
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<tr>
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<td>518.8</td>
<td>0.455</td>
<td>5</td>
</tr>
</tbody>
</table>

Table 4.2. Spectral lines of Ar I used in the measurements, \( E_{p^+} (eV) \) is the ionization energy, \( \lambda \) the wavelength of the transition and \( g_p \) the statistical weight of the upper level.

lateral profiles. The accuracy of the \( n(p) \)-values thus obtained is better than 25% for the 4p-levels, while for the higher lying levels it is due to the uncertainty in the A-value rather poor, say a factor 2. The Saha-values \( n^S(p) \) are obtained using \( n_e \) and \( T_e \) values which are determined by a Thomson-Rayleigh scattering set-up [30]. With this technique where the scattered photons are dispersed over more than hundred pixels of an intensified optical multichannel analyzer an accuracy in \( n_e \) an \( T_e \) is reached within 7%. The Rayleigh scattering is used to get the ground-state density. This value of \( n_1 \) is needed to calculate the escape factor for the radiative decay of the 4s levels to the ground-state. The uncertainty in \( b(p) = n(p)/n^S(p) \) for higher levels is mainly determined by the inaccuracy
Application of a hybrid collisional radiative model...

Figure 4.1. The $b(p)$ factor vs $p$ taking radiation trapping into account (full line) and optically-thin plasma (broken line); • experimental values of van de Sanden [30]. On the right axis, the $p^{-6}$ dependence of $1 - b(p)$ obtained by the model is investigated for highly excited levels. The curves denoted by $V$ are obtained with rate coefficients of VRIENS and SMEETS [9] and those denoted with $D$ are obtained with rate coefficients of DRAWIN [10] a) $Z = 20$ mm and $R = 0$ mm, b) $Z = 20$ mm and $R = 9$ mm, c) $Z = 40$ mm and $R = 0$ mm, d) $Z = 40$ mm and $R = 9$ mm, e) $Z = 70$ mm and $R = 0$ mm.

in $n(p)$ while for lower levels the error in $T_e$ will affect $n^S(p)$ via the factor $\exp(E_{p+}/kT)$. This means that an error in $b(p)$ of a factor 2 is typical for the whole $p$-range.

In Figs. 4.1a – e, where $b(p)$ versus $p$ is shown, the results of the hybrid model are compared with the experimental results obtained at the locations given in Table 4.1. To investigate the influence of the rate coefficients two different sets of rate coefficients were used. The results of the set of VRIENS and SMEETS are denoted by $V$ those using DRAWIN are denoted by $D$ (full lines).

It turns out that at all positions the $b(p)$-factors for the lower lying levels are much smaller than 1, which indicates that the expanding plasmajet is in a non-equilibrium recombining state. It is also observed that in both cases $V$ and $D$ the value $b = 1$ is approached for increasing $p$ which supports the statement...
that higher lying levels are in pLSE. Moreover, as indicated by the quantity $\delta b$ depicted at the right hand side of the Figs 4.1a – b, it is shown that the approach of pLSE obeys the relation $\delta b \propto p^{-6}$ for both cases $V$ and $D$. This justifies the cut-off technique.

In general, the ASDF using Drawin rates turns out to have higher $b$-values. This is related to the fact that especially for low $T_e$-values the excitation and de-excitation rates for highly excited states of $D$ may exceed those of $V$ by more than a factor 60, while de de-excitation rates of the lower levels are in both variants dictated by the values of experimental values of TACHIBANA [13]. Comparison shows that the density of the lowest 4p level tends to be lower than the lowest model result (model $V$), with an important exception at the center of the shock. This might suggest that the depopulation rate of 4p is larger than predicted by the models. Probably there is an extra depopulation process not included in our model which is not operative in the shock. To investigate the influence of radiation trapping we used both variants $V$ and $D$ to calculate the ASDF for an optically thin plasma putting $\Lambda = 1$ (broken curves). It turns out that the effect of radiation trapping on the calculated Saha-decrement is only substantial for the
lower lying levels. The lowering of the 4p group is due to the radiative decay of the higher lying levels 3d and 5s. The occupation of the 4s level appears to have no influence on that of the 4p level. It can be seen that there is a reasonable agreement between the measurements and the optically thin model V. Again, this is not true for the shock position for which the optically thick model D gives a good fit of the experimental results.

4.4.2 Recombination coefficient

The total recombination rate coefficient $\alpha_{cr}$ defined such that $n_en_+\alpha_{cr}$ represents the number of recombination processes per unit of volume and time is an important parameter in modelling flowing plasmas. The explicit expressions for $\alpha_{cr}$ in terms of the $r^+$ coefficients as given in Refs. [1, 3, 4],

$$\alpha_{cr} = \sum_{q \geq 1} \{n_eK_+(q) + A(+, q)\} - \sum_{q > 1} n^S(q)r^+(q)S(q)/n_+,$$

(4.10)
makes it possible to use the numerically calculated ASDF to compute $\alpha_{cr}$. In the above equation $K_+(q)$ and $A(+, q)$ are the rate coefficients for three-body and radiative recombination, respectively, whereas $S(q)$ is the rate coefficient for ionization. Each summation contains only levels for which the effective $pqn$ is smaller than that of the cut-off level. The recombination to the cut-off level can be increased with the stepwise recombination flow in the way as described in Ref. [4]. This is the basis of the conductive cut-off technique.

In the high $n_e$-limit, radiative processes will be unimportant so that $\alpha_{cr}$ scales with $n_e$ [cf. Eq. (4.10)]. Therefore it is useful to introduce the parameter $\kappa_+ = \alpha_{cr}/n_e$, the total three-body recombination rate coefficient, to study collisional-dominated recombination. In various studies it is argued that for low $T_e$ the electron excitation kinetics can be expressed in terms of $E/kT_e$ solely. The simplest approach is that given by Thomson which for a hydrogenic system predicts [4, 20]

$$\kappa_+ = 2.6 \times 10^{-39}T_e^{-4.5},$$

(4.11)
where $\dot{T}_e$ is expressed in eV. Note that $\kappa_+$ does not depend on $n_e$. It refers to a situation where the whole atomic system is dominated by the cold DSB. Calculations in Refs. [4, 20, 21] showed the same temperature dependence of the three-body recombination coefficient and are confirmed experimentally by Hin­nov and Hirschberg in recombining helium plasmas [31]. For a comparison with experimental $\kappa_+$ values for various metals we refer to Refs. [4, 20]. There it is found that Eq. (4.11) reproduces experimental results within a factor two [4]. A general observation in these works is that the experimental values are higher. The simple formula Eq. (4.11) for $\kappa_+$ will serve as a guideline in the discussion of the numerically calculated $\alpha_{cr}$ for which we should keep in mind that its validity region is limited to low $T_e$ and high $n_e$-values. Two series of $\kappa_+$ values will be discussed. The first one (cf. Fig. 4.2), denoted by case $V$ is obtained employing Eq. (4.10) to the $r^+$ values of a CR model based on rates of VRIENS and SMEETS. The second series is obtained using the rate coefficients of DRAWIN (case $D$ cf. Fig. 4.3). In both cases, we consider $\kappa_+$ vs $T_e$ for an optically thin plasma with
Figure 4.2. Recombination coefficient $\kappa_{CR}$ vs $T_e$ for various $n_e$ and an optically-thin plasma (case V). Full line: conductive cut-off technique; broken line: stagnation cut-off technique. A) $n_e = 10^{17} \text{ m}^{-3}$, B) $n_e = 10^{18} \text{ m}^{-3}$, C) $n_e = 10^{19} \text{ m}^{-3}$, D) $n_e = 10^{20} \text{ m}^{-3}$, E) $n_e = 10^{21} \text{ m}^{-3}$. Dashed-dotted line: Eq. (4.11). Dotted line: optically-thick calculation with $n_e = 10^{21} \text{ m}^{-3}$.

First we will investigate the influence of the cut-off technique on the recombination coefficient. Figure 4.2 shows case V based on 26 effective levels which corresponds to a cut-off level with pqn $N = 8$. The full line represents calculations with the conducting cut-off. The broken lines refer to a stagnation cut-off, i.e. the coupling between the highest level with the continuum is affected by direct ionization/recombination solely. The full curve lies above the broken one especially for $T_e < 0.3$ eV. This is based on the fact that the de-excitation flow and thus the effective stepwise recombination over the cut-off boundary is rather high. If the calculations are performed using 46 effective levels it is found that the broken line moves upwards while the full curve is unaltered. The same observation was found for case D for both cut-off methods.

To conclude, we may state that the conductive cut-off method is successful in reducing the number in the atomic system especially in the low temperature range. It is a robust technique and independent of the set of rate coefficients.

$n_1 = 10^{20} \text{ m}^{-3}$ and $n_e = 10^{17}, 10^{18}, 10^{19}, 10^{20}, 10^{21} \text{ m}^{-3}$.
Application of a hybrid collisional radiative model...

Figure 4.3. Recombination coefficient $\kappa_{CR}$ vs $T_e$ for various $n_e$ and an optically-thin plasma (case D). Full line: conductive cut-off technique; broken line: stagnation cut-off technique. A) $n_e = 10^{17}$ m$^{-3}$, B) $n_e = 10^{18}$ m$^{-3}$, C) $n_e = 10^{19}$ m$^{-3}$, D) $n_e = 10^{20}$ m$^{-3}$, E) $n_e = 10^{21}$ m$^{-3}$. Dashed-dotted line: Eq. (4.11). Dotted line: optically-thick calculation with $n_e = 10^{21}$ m$^{-3}$.

The dashed-dotted lines in Figs. 4.2-3 depict Eq. (4.11). As stated before, this value of $\kappa_+$ is obtained for a completely collisional dominated system where recombination is supported by the fact that for free electrons with low temperature de-excitation will dominate over excitation. It is found in both cases $V$ and $D$ that the $\kappa_+$ value approaches a limit for $n_e \to \infty$ which is reached within 1% for $n_e = 10^{21}$ m$^{-3}$ and $T_e = 1$ eV. For lower $T_e$-values, this limit is approached for lower $n_e$-values. However, comparison shows that the limit values for case $V$ and $D$ do not coincide with each other nor with the dashed-dotted line (Eq. (4.11)). For low $T_e$ values the $\kappa_+$-value of case $V$ is a factor 2 too low whereas that of case $D$ is factor 1.5 too high. The recombination coefficient scales with $T_e^{-4.2}$ in case $V$ while it scales with $T_e^{-4.8}$ for case $D$. This difference between $V$ and $D$ is related to the different low-$T_e$ behaviour of the corresponding rate coefficients. Both semi-empirical cross-section formulae are derived from the BORN-BETHE expression, $\sigma_{pq}(E) = [A_{pq} \ln(E/E^*) + B_{pq}] / E$, where the term with $A_{pq}$ represents the optically allowed and the term with $B_{pq}$ the optically forbidden transitions. In
the formulae of DRAWIN \( E^* = E_{pq} \) so that the logarithm is always positive [10] while in the formulae of VRIENS and SMEETS \( E^* = 2 \times Ry \) so that this term can be negative at low electron energies and is corrected by the positive \( B_{pq} \) term. This results in a reduced cross-section [9].

Concerning the influence of radiative processes on the recombination, Figs. 4.2 and 4.3 reveal that for low \( T_e \)-values radiative processes are relatively unimportant. The basic reason is that there is not much competition between radiative decay and the de-excitation processes due to electron collisions, both are downwards. For relatively high \( T_e \)-values, we see that radiative processes are important. It is e.g. found for \( T_e = 1 \text{eV} \) and \( n_e < 10^{19} \text{m}^{-3} \) that \( \kappa_+ \) scales with \( n_e^{-1} \), i.e. \( \alpha_{CR} \) is constant, which means that recombination is essentially determined by the capture-radiative cascade balance, of which the capture process \( e + A^+ \rightarrow A(p) \) is the driving population source [4].

To study the influence of trapping of radiation we added the \( \kappa_+ \) results based on CR model results in which all the escape factors were set to \( \Lambda = 0 \) for all resonance transitions and \( n_e = 10^{21} \text{m}^{-3} \). This refers to a situation in which the recombination is almost completely confined by the collisional depopulation of the lower excited states. As indicated by the dotted line in Figs. 4.2 and 4.3 it is shown that for \( T_e > 0.4 \text{ eV} \) the \( \kappa_+ \) will stay below the value predicted by Eq. (4.11). This is closely related to the fact that the excitation cross section of Ar is much smaller than that of H. Therefore the same applies to the de-excitation rate. However, for decreasing \( T_e \) the influence of the \( n(4s) \) and \( n(3d) \) on the rest of the system will become unimportant and the recombination flow will be controlled to the upper part of the system which is essentially hydrogen-like. So the discrepancies between case \( V \), case \( D \) and Eq. (4.11) are not due to the fact that we are dealing with Ar instead of hydrogen but due to the fact that the hydrogenic rate coefficients are questionable for low \( T_e \) values. This is closely related to the the unknown threshold behaviour of excitation cross sections.

### 4.5 Conclusions

The hybrid cut-off technique developed in Ref. [3], where it was successfully applied to ionizing systems, is applied to recombining systems. We can draw the following conclusions:

1. The conductive cut-off method is also valid for recombining systems. This is justified by the fact that the quantity \( \delta b \) scales with \( p^{-6} \) so that the number of levels in the CR-model can be reduced in a recombining plasma as well.
2. The experimental ASDFs of a cold recombining plasma in an expanding arc jet as measured by van de Sanden [30] lie between the ASDF calculated
with case $V$ and $D$ in most cases, but are not able to differentiate between sets of rate coefficients as given by VRIENS and SMEETS [9] and DRAWIN [10]. Only the calculated 4p level overestimates the lowest experimental 4p levels, except at the shock position. This suggests that there might be an extra depopulation process of 4p which is not included in our model.

3. Since the effective recombination is determined by the upper part of the level system, an accurate description of the excitation flow in the upper part is required which is accomplished by using the conducting cut-off technique.

4. In the limit of high $n_e$, the recombination coefficient scales with $T_e^{-4.2}$ and $T_e^{-4.8}$ for case $V$ and $D$, respectively, while $T_e^{-4.5}$ is expected. This difference is caused by different threshold behaviour of the cross-sections according to DRAWIN and to VRIENS and SMEETS.

References

Chapter 5

Radiative Energy Loss in a Non-Equilibrium Argon Plasma

Abstract - The total radiative loss in atmospheric argon plasmas is calculated admitting for deviations from LTE. We have taken into account non-equilibrium excited state populations using numerical and analytical collisional-radiative models. Simple expressions for the different radiation loss mechanisms are given in terms of the electron density, electron temperature and ionization degree. These quantities together with the heavy particle temperature also define the deviation from equilibrium. In the recombining zones the presence of non-equilibrium will have a significant influence on the total radiative loss due to line radiation. The influence results from the fact that in a recombining plasma the excited states are much smaller populated than the value predicted by Saha. The results of this study can also be used for non-atmospheric argon plasma provided that \( n_e > 5 \times 10^{19} \text{ m}^{-3} \) and \( n(1)d > 10^{20} \text{ m}^{-2} \) in which \( d \) is the plasma dimension.

5.1 Introduction

Thermal argon plasmas can be found in various applications. Cascaded arcs are well-known in the field of deposition of carbon- or silicon-based films and radiation source technology. Inductively coupled plasmas (ICP) are used for spectrochemical analysis, plasma spraying, material synthesis and as light sources. For a proper understanding of these types of plasmas it is needed to compare experimental results with theoretical models. By means of this comparative study it might be possible to obtain scaling laws and to optimize the various plasma applications.

Plasma systems have in general an ionizing part in which the plasma is created and a recombining part, i.e. the afterglow, where deposition (cascaded arcs) or spectroscopic analysis (ICP) takes place. In both plasma parts significant deviations from local Saha equilibrium (LSE) are known to exist, i.e. excited states populations deviate from Saha equilibrium (in the usual nomenclature the

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1This article has been submitted for publication in J. Phys. D: Applied Physics.
Plasma radiation, which is an important transport coefficient in modelling [1], is sensitive to deviations from LSE. However, in the typical temperature range of 4000 \( \sim \) 12000K found in thermal argon plasmas reliable radiative loss data are lacking and the data available are based on the LSE assumption. Recently, WILBERS et al. [2] calculated the total radiative loss for isobaric argon plasmas allowing some departures from LSE which were incorporated using a two-temperature model and a non-equilibrium value of the neutral ground state population \( n(1) \). The so-called \( b(p) \) factors, defined by

\[
b(p) = \frac{n(p)}{n^S(p)} \quad (5.1)
\]

where

\[
n^S(p) = n_e n_+ \frac{g_p}{2 g_+} \left( \frac{\hbar^2}{2 \pi m_e k T_e} \right)^{3/2} \exp \left( \frac{E_{p+}}{k T_e} \right) \quad (5.2)
\]

describe the deviation of the population of the excited levels from the Saha population. The symbols used in the formulas are explained in the Nomenclature. Using Eq. (5.1) we can qualify the plasma studied in Ref. [2] by stating that \( b(1) = 1 \) whereas \( b(p) \) and \( T_e/T_h \) are allowed to differ from unity. This condition is denoted by partial local Saha equilibrium (pLSE). It was found that over a wide range of \( T_e \) and \( T_h \) values \( \varepsilon_{rad}/n_e^2 \), where \( \varepsilon_{rad} \) is the total emission coefficient, was a function of electron temperature \( T_e \) only and that the radiation loss term is mainly determined by line radiation of which especially the lines in the \( 4p - 4s \) transitions are the most important. Since pLSE is assumed this means that \( n(4p) = n^S(4p) \) which has the typical \( n_e^2 \) dependence in singly ionized plasma \( (n_e = n_+) \). For \( T_e > 10000K \) the results were in agreement with the experimental results of EVANS and TANKIN [3] fitted by MILLER and AYEN [4].

OWANO et al. [5] reported experimental results for radiative losses in argon down to 6000 K. Comparison with the models of Refs. [2, 3, 4] show a good agreement. However, in the interpretation of the results, they relied on equilibrium relations of Boltzmann and Saha in determining the temperature and electron density. Again the conclusion was that the radiation is predominantly generated by line transitions.

The dominancy of line radiation is important since it can be used for a proper understanding of non-pLSE situation of radiation loss. To construct a model which allows further deviations from LSE we must admit for \( n(p) \) departures from Saha equilibrium. This can be done using a combination of experimental and theoretical results.

Measurements of excited state populations in helium afterglows [6] and in a freely expanding cascaded argon arc plasma [7] revealed the fact that these excited
states were underpopulated with respect to the Saha population as described in Ref. [8]. This will result in a decreased radiative loss in comparison with the pLSE situation and it might affect the dominancy of the line radiation. On the other hand overpopulation of the excited levels in an ionizing low pressure argon plasma were reported in Refs. [9] and [10]. In this situation the radiative loss is increased compared to the LSE situation. Therefore, to be as general as possible, we should also consider these types of pLSE deviations in thermal plasmas.

The aim of this paper is to include non-equilibrium effects in the excited state population and to construct a radiative loss term for a relatively large range of plasma conditions as found in thermal argon plasmas. The radiative loss can be used as an energy loss term in plasma modelling. In order to cover a wide range of conditions we will construct four expressions to calculate the different radiation loss contributions, namely atom and ion free-free, recombination and line radiation.

5.2 Radiative Loss Term

5.2.1 pLSE

Before studying non-LSE conditions it is useful to study the equilibrium results as obtained in previous studies. Therefore we reproduce in Fig. 5.1 the results as obtained by Ref. [2] where the radiative loss due to line, recombination and free-free radiation are compared to each other. The left vertical axis shows that the 3 contributions are normalized to $n_e^2$. This presentation is possible for the free-free and recombination emission provided the plasma is singly ionized. The same applies for line radiation if pLSE is present whereas the extra demand for the $n_e^2$ normalization of the neutral free-free radiation is that LSE is present ($b(1) = 1$) or that the $b(1)$ is constant or known as a function of temperature over the whole temperature range.

We first consider the contribution of the line radiation which is given by

$$\varepsilon_{\text{line}} = \sum_{p,q} n_p A_{pq} \frac{\hbar \nu_{pq}}{4\pi} = \sum_{p,q} b(p)n_s^s(p)A_{pq} \frac{\hbar \nu_{pq}}{4\pi}. \quad (5.3)$$

For the calculation the plasma is assumed to be optically thin except for the resonance radiation for which we assume that the plasma is completely optically thick. As can be deduced from Fig. 5.1 in the temperature range $5000K \lesssim T_e \lesssim 12000K$ line radiation is the main contribution to the total radiative loss under pLSE conditions. The background of the dominancy of line radiation is that for pLSE conditions $n(p) = n_s^s(p) \propto n_e^2 \exp(E_{p+}/kT_e)$ (cf. Eq. (5.2)) which increases with decreasing $kT_e$. Equation (5.3) indeed shows that due to the $n_e^2$
Figure 5.1. Contributions to the total radiative loss normalized to $n_e^2$ under atmospheric pLSE condition, as a function of temperature. The ff contribution is due to electron-neutral interaction. ($- -$) absolute and (-----) relative contributions of the lines in Table 5.1 to the total line emissivity.

In $n^S(p)$ the line radiation can also be normalized to $n_e^2$ provided $b(p) = 1$ or a function of $T_e$ only. Also of interest is the fact that line radiation is mainly determined by $4p - 4s$ transitions. The broken line represents the radiative loss calculated with the 8 strongest lines (cf. Table 5.1). As can be seen these lines represent already 60 - 70% of the line radiation as shown on the right axis.

As stated before, it is a characteristic feature of the Saha formula (Eq. (5.2)) that the line emissivity increases for decreasing electron temperatures if the $n_e$ value is kept constant. This can be illustrated in the power interruption experiment as initiated by GUREVICH et al. [11]. The switch-off of the power generator causes a cooling of the electrons which results in an increase of the line emission. This rapid increase with a time constant $\tau_{T_e}$ is followed by a much slower decay associated with recombination of the plasma. The results of the power interruption experiment in an ICP of FEY et al. [12] demonstrate that the excited levels of argon are governed by the Saha balance at an electron temperature ($T_e \approx 0.8$eV) higher.
Table 5.1. The 8 strongest lines in the Ar-I system

<table>
<thead>
<tr>
<th>level</th>
<th>Statistical weight</th>
<th>Energy (eV)</th>
<th>A-value ($\times 10^8$)</th>
<th>Wavelength (Å)</th>
</tr>
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<td>13.072</td>
<td>0.366</td>
<td>8115.3</td>
</tr>
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<td>13.168</td>
<td>0.274</td>
<td>7635.1</td>
</tr>
<tr>
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<td>13.091</td>
<td>0.233</td>
<td>8424.7</td>
</tr>
<tr>
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<td>12.904</td>
<td>0.212</td>
<td>9123.0</td>
</tr>
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<td>13.298</td>
<td>0.244</td>
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</tr>
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<td>13.149</td>
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<td>8103.7</td>
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</tr>
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<td>3</td>
<td>13.279</td>
<td>0.196</td>
<td>7948.2</td>
</tr>
</tbody>
</table>

than the heavy particle temperature ($T_h \approx 0.6\text{eV}$). For lower temperatures the deviations from Saha equilibrium increase and the excited state populations are expected to be underpopulated with respect to the Saha population [8]. Therefore it is possible that under recombining non-pLSE conditions the dominancy of line emission might be affected since only line radiation is affected by this departure from Saha.

5.2.2 Non-pLSE

To investigate the influence of non-equilibrium values of the $b(p)$ factors, (i.e. $b(p) \neq 1$) on the line emissivity we can use collisional-radiative (CR) models. An instructive expression for the excited state population is

$$b(p) = r^1(p)b(1) + r^+(p),$$

[8] where $r^+(p)$ and $r^1(p)$ are the relative population coefficients for a purely recombining and ionizing plasma, respectively. It is clear that we have to distinguish between two cases, i.e. ionizing and recombining plasma parts. In the ionizing region where the plasma is created under relatively high temperature conditions the $b(1)$ is larger than 1. Typical values for $b(1)$ are $b(1) \sim 10^{-3}$ in the active zone of the ICP and cascaded arcs. In low pressure ionizing plasmas the $b(1)$ can be as high as $10^6$. However, CR-models show that the population coefficient $r^1(4p)$ is of the order of a few $10^{-4}$ [13] so that the ground state contribution
to the excited state population can remain small for high pressure ionizing systems and we may expect that the pLSE condition is approached closely under high temperature, i.e. ionizing conditions. However to be as general as possible we should be prepared for strong ionizing conditions for which a large $b(1) \gtrsim 10^4$ value propagates to excited levels. From CR-models we know that $r^1(p)$ scales with $p^{-6}$ where $p = \sqrt{Ry/E_{p+}}$ is the effective principal quantum number, if the system is collisionally dominated, [8, 13]

$$r^1(p) = b_0 p^{-6}. \quad (5.5)$$

In general the coefficient $b_0$ is a function of $T_e$ and $n_e$ so for a given $n_e$, $T_e$ and $n_1(1)$ value the $b(1)$ can be computed using Eqs. (5.1, 5.2). For sufficiently large $n_e$ values, i.e. $n_e \gtrsim 10^{20} \text{m}^{-3}$, $b_0$ is a function of $T_e$ solely (cf. Eq. (5.13)). Equation (5.5) provides information on the enhanced population density, i.e. enhanced line radiation.

On the other hand, in the recombining region such as the afterglow we meet with the low temperature situation, where due to exponential increases with temperature (cf. Eq. (5.1)) the radiative loss term normalized with $n_e^2$ becomes relatively important. Van de Sanden [7] has measured excited state populations of argon in a free expanding plasma jet. In Fig. 5.2 the $b(p)$ factors are shown versus $\epsilon_p = E_{p+}/kT_e$ for different positions in the expanding plasma jet corresponding to different temperatures (a) [7] and for an helium afterglow (b) [6]. The $b(p)$ factors are smaller than 1 indicating that the excited states are underpopulated with respect to Saha population. Values up to $10^{-4}$ are attained so that the emissivity of the line in question is reduced by a factor of $10^{-4}$! Symbols indicated with an arrow represent the same argon line transition ($4p'(1/2, 7503 \text{ A})$) for different $T_e$-values. Also shown in Fig. 5.2 are theoretical atomic state distribution functions (ASDF) according to calculations of Mansbach and Keck [15] and Biberman et al. [14] (modified diffusion approximation). The theoretical ASDF's are based on the so-called cold de-excitation saturation balance in which collisional de-excitation prevails over radiation [8]. The ASDF calculated by Biberman et al. is derived for hydrogen like systems and reads

$$b_{Bib}(\epsilon_p) = b(1)\chi(\epsilon_p) + [1 - \chi(\epsilon_p)] \quad (5.6)$$

where

$$\chi(x) = \frac{4}{3\sqrt{\pi}} \int_0^x e^{-t^2/2} dt$$

Note that Eq. (5.6) is of the same form as Eq. (5.4). Recombining systems are characterised by $b(1) \to 0$. For $\epsilon_p > 1$ the function $1 - \chi(\epsilon_p)$ can be approximated
Figure 5.2. The $b(p)$ factor as function of $E_{p+}/kT_e$. (a) Experimental values for $b(p)$ in argon for different temperatures in a free expanding cascaded arc jet [7]. \( \bullet: T_e = 0.144\text{eV}, \quad \circ: T_e = 0.213\text{eV}, \quad \Delta: T_e = 0.260\text{eV}, \quad \Box: T_e = 0.204\text{eV}. \) (b) Experimental $b(p)$ values in a helium afterglow [6]. \( \mp: T_e = 0.13\text{eV}, \quad \Delta: T_e = 0.19\text{eV}, \quad \circ: T_e = 0.27\text{eV}. \) Also shown are the ASDF of Ref. [14] (full line) and of Ref. [15] (broken line).

by

$$
\frac{b_{Bib}(\epsilon_p)}{4} \approx \frac{3}{3\sqrt{\pi}} \epsilon_p^{3/2} e^{-\epsilon_p} \left( 1 + \frac{3}{2\epsilon_p} + \ldots \right)
$$

(5.7)
The ASDF of Ref. [15] can be approximated by

\[ b_{MK}(\epsilon_p) \simeq \left( \frac{\epsilon_p^3}{3!} + \frac{\epsilon_p^2}{2!} + \epsilon_p + 1 \right) e^{-\epsilon_p} \]  

Both Eqs. (5.7, 5.8) state that the underpopulation depends solely on \( \epsilon_p \), which means that a level with large \( E_{p+} \) value in a high temperature plasma should behave in the same way as a level with low \( E_{p+} \) value at low temperature. This is confirmed by the fact that the various experimentally obtained values \([6, 7, 9]\) are found to be function of \( \epsilon_p \) solely. The difference between the two theoretical ASDF’s is due to the different set of cross-section used in these models. For high \( \epsilon_p \) values it is not clear which theoretical ASDF is the most appropriate. For low \( \epsilon_p \) values the experimental results \([7]\) favor the ASDF of Ref. \([15]\). The same conclusion can be drawn from the measurements of HinnoV and Hirschberg in a helium afterglow (cf. Fig. 5.2b) \([6]\). For \( \epsilon_p \lesssim 6 \) the results of Ref. \([15]\) are in excellent agreement with Eq. (5.8) \([8]\). For the 4p levels of argon which are dominant in the line radiation loss this corresponds with a temperature range of \( T_e > 5000K \). Concluding we may state that for recombining plasmas \( b(p) \) decreases with decreasing temperature so the line emissivity normalized to \( n_e^2 \) will increase much more slowly than predicted by the pLSE assumption. Therefore the dominancy of the line emissivity becomes questionable and we have to consider the different radiation loss mechanisms separately. In the next section we will use Eqs. (5.5, 5.7 and 5.8) to calculate the line contribution to the radiative losses.

### 5.3 Results

In this section the results of free-free and line radiation will be presented as separate equations. The reason is that in a broad range of non- LSE conditions \( n(1), n_e \) and \( T_e \) are decoupled. We start with the emissivity contributions due to electron-ion free-free and recombination transitions which do not depend on the state of equilibrium departure. These contributions can be found in Ref. \([2]\) and are fitted by (cf. Table 5.2)

\[ \log_{10} \left( \frac{\epsilon_{fi}^e + \epsilon_{fi}^a}{n_e^2} \right) = \sum_{i=0}^{3} f_i T_e^i. \]  

The quantities are expressed in MKS units.

For a proper treatment of the line radiation the line emissivity must be divided into a part originating from the ground state which is related to \( r^i(p) \) and from the continuum related to \( r^+(p) \) (cf. Eq. (5.4)). Figure 5.3 compares the line radiation
Radiative energy loss ...

Figure 5.3. Radiation losses due to line transitions, neutral free-free and recombination radiation. (---): recombination radiation. Line transitions are shown by thick lines: (---): pLSE, (----): Ref. [15], (--.--): Ref. [14]. Neutral free-free radiation is calculated with $\alpha = \max(\alpha_{LSE}, \alpha_{min})$. A: $\alpha_{min} = 10^{-4}$; B: $\alpha_{min} = 10^{-5}$; C: $\alpha_{min} = 10^{-6}$; D: $\alpha = \alpha_{LSE}$.

loss for the ASDF according to BIBERMANN et al. (dashed line), MANSBACH and KECK (dashed dotted line) and to Saha (full line). The latter, added for the sake of completeness, makes a comparison of this study and those of Refs. [2]-[4] possible. It should be noted that at $4000 \, K$ the differences between BIBERMANN and MANSBACH and KECK are more than one order of magnitude ($b_{MK}(4p[5/2]) \sim 0.5$ and $b_{Bib}(4p[5/2]) \sim 0.01$) whereas the differences between Saha and BIBERMANN value accord for more than 4 orders of magnitude. The data of the radiative losses originating from the continuum contribution ($r^+(p)$), calculated with the ASDF of BIBERMANN et al. can be fitted by

$$\varepsilon_{Bib}^+(T_e, n_e) = n_e^2 10^{-27.52-1.18\ln(T_e)} \, \text{Wm}^{-3} \text{sr}^{-1}$$

while the radiative losses calculated with the ASDF of MANSBACH and KECK leads to

$$\varepsilon_{MK}^+(T_e, n_e) = n_e^2 10^{-22.78-1.66\ln(T_e)} \, \text{Wm}^{-3} \text{sr}^{-1}.$$
Note that in all cases collisional de-excitation must be dominant which means that \( n_e \gtrsim 5 \times 10^{19} \text{m}^{-3} \). In the transition region between ionizing and recombining systems we meet the situation that \( \epsilon_p \lesssim 6 \). This value corresponds to a \( 4p \) level at 5000 K. For \( T_e \gtrsim 5000K \) we recommend the use of Eq. (5.11) to calculate the line radiation due to the part of the excited level densities which originates from the ions (cf \( r^+(p) \) in Eq. (5.4)).

The line radiation originating from the ground state contribution of the excited levels can be calculated by substituting \( b(1)b_0p^{-6} \) into Eq. (5.3), which yields

\[
\epsilon^1 = \frac{n(1)b_0}{g_1Ry^3} \sum_{p,q} \exp(-E_p/kT_e)E_p^3 E_{p+g_pA_{pq}} \frac{h\nu_{pq}}{4\pi} \tag{5.12}
\]

It is obvious that the sum in Eq. (5.12) is a function of \( T_e \) only. For \( n_e \gtrsim 5 \times 10^{19} \text{m}^{-3} \) the coefficient \( b_0 \) is almost independent of \( n_e \) [8, 16]. The coefficient \( b_0 \) is calculated using the CR-model of Ref. [13] and can be fitted by (cf. Table 5.2)

\[
\log_{10}b_0(T_e) = \sum_{i=0}^{3} a_i T_e^i \tag{5.13}
\]

where \( T_e \) is expressed in K. Equation (5.12) is then only a function of \( n(1) \) and \( T_e \)

\[
\epsilon^1(n(1), T_e) = 6.29 \times 10^{-30} n(1)b_0(T_e)F(T_e)
\]

\[
F(T_e) = \sum_{i=0}^{6} d_i T_e^i. \tag{5.14}
\]

For low temperatures Eq. (5.14) can be used even for \( n_e \) values lower than \( 10^{20} \text{m}^{-3} \) since the ionizing contribution will then be negligible. When ionizing equilibrium departures are small Eq. (5.14) is insignificant compared with other radiation loss contributions. Only strong ionizing conditions for which \( b(1) \gtrsim 10^4 \) may cause Eq. (5.14) to contribute substantially.

The neutral free-free emissivity depends on the product \( n_e n(1) \) in contrast to all the other emissivity contributions which depend on \( n_e^2 \). However, to make comparison possible we write

\[
\frac{\epsilon_{ff}^{ee}}{n_e^2} \sim \frac{n_e n(1)}{n_e^2} = \frac{n(1)}{n_e} = \frac{1}{\alpha}, \tag{5.15}
\]

which shows that the ionization degree \( \alpha \) is a suitable parameter. For thermal plasmas with higher temperatures it is plausible that the LSE value of \( \alpha \) gives a reasonable description for \( \epsilon_{ff}^{ee} \). However, for recombining conditions it is
expected that there will be an underpopulation of \( n(1) \) with respect to its Saha value. This again can be related to the power interruption experiment of Gurevich and Podmoshenskii where it is found that for argon the temperature relaxation time is much smaller than the plasma recombination time so that \( n_e \) and \( T_e \) are decoupled in contrast with the LSE state [12]. The spectral emissivity \( \varepsilon_{ff}^{ea} \) can be integrated over the frequency domain yielding

\[
\frac{\varepsilon_{ff}^{ea}(T_e, n_e/n(1))}{n_e^2} = \left( \frac{6C_2k}{hc} \right) T_e^{5/2} \left( \frac{n(1)}{n_e} \right) Q(T_e),
\]

(5.16)

which is an increasing function of \( T_e \) at constant \( n_e/n(1) \). Also shown in Fig. 5.3 are the \( \varepsilon_{ff}^{ea} \) contributions (thin lines indicated with C, B and A) for various minimum ionization degrees \( \alpha_{min} \) (\( 10^{-6}, 10^{-5} \) and \( 10^{-4} \)). In representing \( \varepsilon_{ff}^{ea}/n_e^2 \) we assumed an atmospheric LSE plasma for the higher temperature domain. When \( \alpha_{LSE} = (n_e/n(1))_{LSE} < \alpha_{min} \), Eq. (5.16) is used to calculate \( \varepsilon_{ff}^{ea} \) with

\[ \alpha = \alpha_{min}. \]

If \( \alpha_{LSE} > \alpha_{min} \) we choose for \( \alpha_{LSE} \).

The radiation losses as predicted by formulae (5.9), (5.10), (5.11) and (5.16) are compared to each other in Fig. 5.3. From Fig. 5.3 we may conclude the following: 1) line radiation using non-LSE formulae for a recombining ASDF is lower than those predicted by Saha. However, it is still dominant provided \( \alpha_{min} > 10^{-5} \). 2) With respect to the continuum radiation we may state that the neutral free-free is larger than the recombination radiation for an \( \alpha \) value larger than \( 10^{-4} \).

We now have 4 simple expressions (5.9, 5.11, 5.14 and 5.16) to calculate the total radiative loss which can be used in plasma modelling. The input parameters are \( T_e, n_e \) and \( n(1) \). It would be convenient to compare the total radiative loss with results from various authors. However, a proper comparison is not possible since \( T_e, n_e \) and \( n(1) \) are decoupled in our case. In Fig. 5.4 the total radiative loss for an atmospheric argon plasma versus temperature is shown. The ionization degrees correspond with those of Fig. 5.3. The curve marked with \( \alpha_{LSE} \) is obtained when \( n_e, n(1) \) and \( T_e \) would be related by Saha's relation. In the temperature range \( T_e \lesssim 12000K \) line radiation is the main loss contribution. This is partly based on the fact that for lower temperatures the population density of excited levels will increase. However, one should keep in mind that this increase is much smaller than the value predicted by Saha. Therefore the radiation due to Eq. (5.11) will be lower than predicted by Ref. [2]. The main reason of the dominancy of line radiation in a recombining plasma is based on the fact that the \( n_e \)-value in such a plasma will be much larger than the value predicted by Saha. The results of this study can also be used for non-atmospheric conditions provided that \( n_e > 5 \times 10^{19} \text{ m}^{-3} \) and \( n(1)d > 10^{20} \text{ m}^{-2} \), where \( d \) is the plasma dimension. The reason for
Figure 5.4. The total radiative loss. For ionization degrees see Fig. 5.3.

Table 5.2. Fit coefficients

<table>
<thead>
<tr>
<th>Eq. (5.9)</th>
<th>Eq. (5.13)</th>
<th>Eq. (5.14)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_0$</td>
<td>$a_0$</td>
<td>$d_0$</td>
</tr>
<tr>
<td>$f_1$</td>
<td>$a_1$</td>
<td>$d_1$</td>
</tr>
<tr>
<td>$f_2$</td>
<td>$a_2$</td>
<td>$d_2$</td>
</tr>
<tr>
<td>$f_3$</td>
<td>$a_3$</td>
<td>$d_3$</td>
</tr>
</tbody>
</table>

the first demand is that the ASDF must be collision dominated and the second demand deals with the fact resonant radiation should be trapped.
5.4 Conclusions

The model of Ref. [2] for the total radiative loss has been extended by admitting larger deviations from LSE. Deviations from LSE are manifested by two important effects. First we take into account the effect of non-equilibrium ASDF’s (cf. Eq. (5.4)). The ground state contribution of the non-equilibrium excited state population is calculated with the numerical CR-model of Ref. [13], while for the continuum contribution the analytical ASDF of Ref. [15] has been used. Secondly $n_e$ and $T_e$ are decoupled especially in the recombining zone. A suitable parameter to account for this deviation from LSE is the ionization degree and plays an important role in the neutral free-free radiation. In the recombining zone the influence of non-LSE on the total radiative loss may be substantial and is due to the fact that line radiation depending on the ASDF does not obey the Saha equation. However, the line radiation will remain dominant. The background is that for low temperatures the actual electron density is higher than the Saha value for the electron density which is obtained using the ground state density and the temperature. An additional LTE deviation is the difference between the heavy particle and electron temperature which can be accounted for by the pressure. The total radiative loss is then a function of the electron density, electron temperature and ionization degree. Analytical expressions and numerical fits for the different radiative loss mechanisms are given (5.9, 5.11, 5.14, and 5.16) which can then be used in plasma modelling.

References

Table 5.3. Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{pq}$</td>
<td>transitions probability</td>
</tr>
<tr>
<td>$b(p)$</td>
<td>nonequilibrium parameter</td>
</tr>
<tr>
<td>$c$</td>
<td>velocity of light</td>
</tr>
<tr>
<td>$C_2$</td>
<td>Electron-neutral continuum constant ((1.03 \times 10^{-34} \text{Wm}^2\text{K}^{-3/2}\text{sr}^{-1}))</td>
</tr>
<tr>
<td>$d$</td>
<td>plasma dimension</td>
</tr>
<tr>
<td>$E_p$</td>
<td>excitation energy of level p</td>
</tr>
<tr>
<td>$E_{p^+}$</td>
<td>ionization energy of level p</td>
</tr>
<tr>
<td>$g_p, g^+_{p}$</td>
<td>statistical weight</td>
</tr>
<tr>
<td>$h$</td>
<td>Planck's constant</td>
</tr>
<tr>
<td>$k$</td>
<td>Boltzmann constant</td>
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<tr>
<td>$n(1)$</td>
<td>neutral particle density</td>
</tr>
<tr>
<td>$n_e$</td>
<td>electron density</td>
</tr>
<tr>
<td>$n_h$</td>
<td>heavy particle density</td>
</tr>
<tr>
<td>$n(p)$</td>
<td>excited state density</td>
</tr>
<tr>
<td>$n^+_p$</td>
<td>ion density</td>
</tr>
<tr>
<td>$n^S(p)$</td>
<td>Saha population density</td>
</tr>
<tr>
<td>$p$</td>
<td>principal quantum number</td>
</tr>
<tr>
<td>$Q$</td>
<td>electron-atom cross-section</td>
</tr>
<tr>
<td>$R_y$</td>
<td>Rydberg energy</td>
</tr>
<tr>
<td>$r^1(p), r^+(p)$</td>
<td>relative population coefficients</td>
</tr>
<tr>
<td>$T_e$</td>
<td>electron temperature</td>
</tr>
<tr>
<td>$T_h$</td>
<td>heavy particle temperature</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>ionization degree</td>
</tr>
<tr>
<td>$\epsilon_p$</td>
<td>$E_{p^+}/kT_e$</td>
</tr>
<tr>
<td>$\epsilon$</td>
<td>emissivity</td>
</tr>
<tr>
<td>$\nu$</td>
<td>frequency</td>
</tr>
</tbody>
</table>
Chapter 6

Source terms, transport coefficients

In this chapter the source terms of the particle (section 6.1), momentum and energy (section 6.2) transport equations will be expressed in such a way that they can be used in the numerical model. The model as presented in chapter 2 will be applied to an inductively coupled plasma (ICP). For this purpose the electromagnetic (EM) field must be specified in order to calculate the Ohmic dissipation ($\mathbf{J} \cdot \mathbf{E}$) and the Lorentz force ($\mathbf{J} \times \mathbf{B}$). In section 6.3 two models are presented for the calculation of the EM-field in the ICP. It will be shown that for relatively high frequencies a two-dimensional formulation is needed for the EM-field. To complete the macroscopic model the transport coefficients have to be specified. The electron transport coefficients for partially ionized plasmas are discussed in section 6.4. In this discussion we are guided by the ionization degree.

6.1 Particle source terms

The collisional-radiative production term $\langle (\partial f_e/\partial t)_{CR} \rangle$ of the electron gas is the result of excitation/de-excitation, ionization/recombination and radiation processes and reads

$$
\langle \left( \frac{\partial f_e}{\partial t} \right)_{CR} \rangle = n_e n_a S(1) - n_e n_+ \left[ n_e K_+(1) + A_+(1) \right] + n_e \sum_{q>1} n(q) S(q) \\
- n_e n_+ \sum_{q>1} \left( n_e K_+(q) + A_+(q) \right),
$$

(6.1)

where $S(q)$, $K_+(q)$ and $A_+(q)$ are the rate-coefficients for ionization, three particle and two particle (radiative) recombination, respectively. Under the assumption that the relaxation time for the atomic excited states is much shorter than the diffusion-convection time the population densities of the excited states are only determined by population/depopulation balances with the ground and ion ground levels (cf. QSS as discussed in chapter 3) [1]. It can be shown (cf. Eqs. (3.17)
and (3.20)) that under QSS conditions Eq. (6.1) can be written as

\[
\left(\frac{\partial f_e}{\partial t}\right)_{CR} = n_e n_a S_{CR} - n_e n_+ \alpha_{CR}.
\] (6.2)

In chapter 3 a combined analytical numerical collisional-radiative model is discussed to compute the ASDF, the effective ionization \( S_{CR} \) and recombination coefficients \( \alpha_{CR} \) for an argon plasma. The coefficients \( S_{CR} \) and \( \alpha_{CR} \) coefficients are functions of \( n_e, n_a \) and \( T_e, T_a \). The fact that \( S_{CR} \) and \( \alpha_{CR} \) also depend on \( n_a \) and \( T_a \) is connected with absorption of resonance radiation. The difference of \( T_e \) and \( T_a \) is not of significance for radiation absorption and \( n_a \) can be determined from

\[
p = n_a k T_a = 1 \text{ atm. For } n_e > 10^{19} \text{ m}^{-3} \text{ the system is in the excitation saturation balance (ESB) and the population coefficient } r^1(p) \text{ is given by Eq. (3.27), i.e. } r^1(p) = b_0 p^{-6} [2]. To incorporate the hybrid CR-model in the plasma transport model the } b_0 \text{ coefficient has been calculated for a large number of plasma conditions. Under ESB conditions } b_0 \text{ is almost independent of the electron density so that } b_0 \text{ is only a function of electron temperature [1, 2]. The results for } S_{CR} \text{ are based on a fit for } b_0 \text{ and a polynomial fit for } \log S_{CR} \text{ by using Eq. (3.22) is obtained as function of the electron temperature, in which the fit coefficients are given in Table 6.1.}

\[
\log_{10} S_{CR}(T_e) = \sum_{i=0}^{6} m_i T_e^i,
\] (6.3)

This equation can be used in the temperature domain \( 2000 \leq T_e \leq 19000 \text{ K} \). In

| \( m_0 \) | \(-85.560\) | \( m_4 \) | \(-8.560 \times 10^{-14}\) | \( z_0 \) | \(-18.352\) |
|\( m_1 \) | \(3.195 \times 10^{-2}\) | \( m_5 \) | \(3.570 \times 10^{-18}\) | \( z_1 \) | \(-9.840 \times 10^{-5}\) |
|\( m_2 \) | \(-7.789 \times 10^{-6}\) | \( m_6 \) | \(-6.095 \times 10^{-23}\) | \( z_2 \) | \(4.374 \times 10^{-9}\) |
|\( m_3 \) | \(1.083 \times 10^{-9}\) | & | & | \( z_3 \) | \(-7.862 \times 10^{-14}\) |

Table 6.1. Fit coefficients \( m_i \) in Eq. (6.3) and \( z_i \) in Eq. (6.6).

Fig. (6.1) the effective ionization coefficient \( S_{CR} \) versus temperature is shown. For comparison the rate-coefficients for the excitation the effective 4s-level \( (K_{1-4s}) \), and the direct ionization \( (K_{1+}) \) are also shown in Fig. (6.1).

The recombination is due to three particle \( \alpha_{3,CR} \) and radiative recombination,

\[
\alpha_{CR} = \alpha_{3,CR} + \sum_{q \geq 1} A_+ (q).
\] (6.4)
Figure 6.1. Rate coefficients for the argon ground level excitation versus temperature. Full line: effective ionization coefficient $S_{CR}$, ($-$ -): $K_{1,4s}$, ($-$ -): $K_{1,+}$.

For the same plasma conditions as for $S_{CR}$, the effective recombination coefficient has been calculated according to Eq (3.21). The results for $\alpha_{3,CR}$ are fitted by a semi-empirical expression which reads,

$$
\alpha_{3,CR} = n_e 10^{a_0(n_e)} (T_e/11608)^{a_1(n_e)} \quad [\text{m}^3/\text{sec}]
$$

$$
a_0(n_e) = -13.982 - 1.275 \log_{10}(\min(10^{20}, n_e))
$$

$$
a_1(n_e) = -27.624 - 1.675 \log_{10}(\min(10^{20}, n_e)). \quad (6.5)
$$

In Fig. 6.2 $\alpha_{3,CR}$ versus temperature is shown for different electron densities. For comparison the result of THOMSON for $\alpha_{3,CR}$ [1] is also reproduced in this figure. The rate coefficient for radiative recombination $\sum_q A_+(q) = A_+(T_e)$ can be obtained by dividing the spectral free-bound emissivity $\epsilon_{FB}$ by the photon energy and integrating over all wavelengths, i.e.

$$
A_+(T_e) = \frac{4\pi}{n_en_+} \int \frac{\epsilon_{FB}(\lambda, T_e) \lambda}{hc} d\lambda \quad [\text{m}^3/\text{s}]
$$

$$
\log_{10}[A_+(T_e)] = \sum_{i=0}^{3} z_i T^i_e, \quad (6.6)
$$

where $\epsilon_{FB}$ can be found in Ref. [3] and the fit coefficients $z_i$ are given in Table 6.1. The integration is performed over a wavelength interval 100nm - 100\mu m
which implies that resonance radiation is totally trapped. In Fig. 6.3 the radiative recombination versus temperature according to Eq.(6.6) and Ref. [4] is shown. The $\epsilon_{FB}$ scales with $n_e n_+$ so that the radiative recombination coefficient does not depend on the electron density.

In the field of thermal plasma modelling [5] the experimental results of recombination measurements by DESAI and CORCORAN have been used for the effective recombination coefficient [6]. The recombination coefficient was measured in an atmospheric argon plasma jet and their results could be fitted by the following
expression

\[ \alpha_{CR}(T_e, n_e) = 8.86 \times 10^2 T_e^{-1.8} 10^{-3410/T_e} n_e^{-0.64} \text{ m}^3/\text{sec}. \]  

(6.7)

The negative power of \( n_e \) is explained by a dominant dissociative recombination channel. The reaction mechanism of dissociative recombination is thought to be

\[
A^+ + A(1) + A(1) \leftrightarrow A_2^+ + A(1) \\
A_2^+ + e \leftrightarrow A_2^* \rightarrow A(1) + A^*
\]

where \( A_2^* \) is an excited argon molecule. The differences between (6.4) and (6.7) for the recombination coefficient might be three orders of magnitude at \( T_e \sim 3000K \) for the same electron density.

### 6.2 Energy source

The energy transfer \( Q_h \) as a result of elastic collisions between heavy particles and electrons is proportional to the difference of the electron and heavy particle temperature. The expression for \( Q_h \) is [7]

\[
Q_h = n_e \sum_{\alpha = i, a} \left( \frac{2m_e}{m_\alpha} \right)^{3/2} k(T_e - T_\alpha) \bar{\nu}_{ea},
\]

(6.8)

where \( \bar{\nu}_{ei} \) and \( \bar{\nu}_{ea} \) are the average collision frequencies for momentum transfer between electron-ion and electron-atom collisions, respectively. The \( \bar{\nu}_{ei} \) is given by [7]

\[
\bar{\nu}_{ei} = \frac{4\sqrt{2}\pi}{3} \left( \frac{e^2}{4\pi \varepsilon_0 m_e} \right)^2 \left( \frac{m_e}{kT_e} \right)^{3/2} n_+ ln\Lambda.
\]

(6.9)

Fitting the experimental data of MILLOY [8] the following semi-empirical expression for the average electron-neutral collision frequency for momentum transfer in argon is obtained

\[
\bar{\nu}_{ea} = \sqrt{\frac{8kT_e}{\pi m_e}} n_a \left[ \frac{5 \times 10^{-20}}{(1 + 1.7 \times 10^{-3} T_e)^2} - 3 \times 10^{-21} \\
+ 2.8 \times 10^{-24} T_e - 4.1 \times 10^{-34} T_e^3 \right].
\]

(6.10)

The electron source term \( Q_e \) accounts apart from the elastic energy transfer to the heavy particles \( Q_h \), for radiation losses \( Q_{rad} \) and for collisional excitation/de-excitation and ionization/recombination \( Q_{IR} \) [9]. In chapter 5 the total radiative
losses are expressed in the main plasma parameters. Under QSS conditions \( Q_{IR} \) can be simplified by using the effective ionization and recombination coefficients. Then \( Q_{IR} \) is given by

\[
Q_{IR} = (n_en_aS_{CR} - n_en_n\alpha_{CR})E_+^*.
\]  

(6.11)

The quantity \( E_+^* = E_+ - \Delta E \) is the ionization potential \( E_+ \) corrected with the ionization potential lowering \( \Delta E \) [10]. Under ICP conditions it is found that \( \Delta E = 0.014\text{eV} \) typically while for argon \( E_+ = 15.759\text{eV} \).

### 6.3 Electromagnetic field

#### 6.3.1 A one-dimensional EM-field formulation

An essential feature of the present model is the calculation of the EM-field generated by the coil. This is necessary to obtain an explicit form of the EM source terms in Eqs. (2.41) and (2.50). We start with the Maxwell equations given in the left hand column which can be simplified to the equation at the right hand column.

\[
\nabla \cdot \bar{E} = \frac{\rho_e}{\varepsilon_0} \tag{6.12}
\]

\[
\nabla \times \bar{E} = -\frac{\partial \bar{B}}{\partial t} \tag{6.13}
\]

\[
\nabla \times \bar{H} = \frac{\mu_0}{\varepsilon_0} \left( \bar{J} + \varepsilon_0 \frac{\partial \bar{E}}{\partial t} \right) \tag{6.14}
\]

\[
\nabla \times \bar{H} = \bar{J} \tag{6.15}
\]

The simplification in Eq. (6.14) is justified since electromagnetic effects are neglected. The reason is that \( \omega_p^2 \gg \nu_{eh}\omega \), where the plasma frequency \( \omega_p \) and the frequency of the EM-field are of the order \( 10^{12} \) and \( 2\pi \times 10^8 \), respectively, and the wavelength of the EM-field is much larger than the characteristic dimensions of the plasma. Further, magnetization effects will be neglected so that we may take \( \mu_r = 1 \).

In Refs. [11, 12] the calculation of the EM-field is further simplified by assuming that the magnetic field only has an axial component \( H_z \) and the electric field only an azimuthal component \( E_\phi \). We will discuss this model and show that for our case there are too many drawbacks so that a proper treatment needs a different approach, which will be dealt with in section (6.3.2). In order to facilitate the discussion a cylindrical coordinate system will be used. In the one-dimensional
model the Maxwell equations read

\[
\frac{1}{r} \frac{\partial}{\partial r}(r E_\phi) = -\mu_0 \frac{\partial H_z}{\partial t} \tag{6.16}
\]

\[
\frac{\partial H_z}{\partial r} = -\sigma E_\phi, \tag{6.17}
\]

where Ohm’s law (2.45) is used. Substituting \(E_\phi = E_\phi^A \exp[i(\omega t - \phi_E)]\) and \(H_z = H_z^A \exp[i(\omega t - \phi_H)]\) into Eqs. (6.16) and (6.17) equations for \(E_\phi^A\) and \(H_z^A\) and the phase difference \(\Delta = \phi_H - \phi_E\) are obtained,

\[
\frac{\partial r E_\phi^A}{\partial r} = -r \mu_0 \omega H_z^A \sin \Delta, \tag{6.18}
\]

\[
\frac{\partial H_z^A}{\partial r} = -\sigma E_\phi^A \cos \Delta, \tag{6.19}
\]

\[
\frac{\partial \Delta}{\partial r} = \left( \frac{\sigma E_\phi^A}{H_z^A} \right) \sin \Delta - \left( \frac{\mu_0 \omega H_z^A}{E_\phi^A} \right) \cos \Delta. \tag{6.20}
\]

The boundary conditions along the torch axis \(r = 0\) are given by

\[
E_\phi^A(0, z) = 0, \tag{6.21}
\]

\[
H_z^A(0, z) = \frac{NI}{2(\ell_2 - \ell_1)} \left[ \frac{\ell_2 - z}{\sqrt{R_c^2 + (\ell_2 - z)^2}} - \frac{\ell_1 - z}{\sqrt{R_c^2 + (\ell_1 - z)^2}} \right], \tag{6.22}
\]

\[
\Delta(0, z) = \pi/2, \tag{6.23}
\]

where \(I\) is the coil current, \(N\) is the number of windings, \(\ell_1\) and \(\ell_2\) are the distances of the top and bottom of the coil and \(R_c\) is the coil radius. (cf Fig. 6.4).

A consequence of this formulation of the electric and magnetic field is that there is no axial component of the Lorentz force. The radial component of the Lorentz force is \(\sigma \mu_0 E_\phi^A H_z^A \cos \Delta\). The radial dependence of the phase difference is due
to the presence of the induced electric current in the plasma. Equation (6.22) for \( H_z^A \) on the axis corresponds to the axial magnetic field along the axis of a finite solenoid in free space. The validity of this expression will be discussed using the dimensionless parameter \( \Psi = R/\delta \) where \( R \) is the plasma radius and \( \delta = \sqrt{2/\mu_0 \sigma \omega} \) the skin depth. Since \( \Psi \) scales with \( \sqrt{\omega} \), there will be almost no shielding of the magnetic field by the plasma for low frequencies, i.e. when \( \Psi \leq 1 \). This results in a more or less uniform energy dissipation distribution. In this case the one-dimensional model is a reasonable approach in the sense that the axial magnetic field can be approximated by Eq. (6.22). In Fig. (6.5) the axial magnetic field in an infinite homogeneous solenoid is shown versus \( \Psi \). It is clear that for \( \Psi < 1 \) the magnetic field is almost not shielded. Since a typical value for the electrical conductivity in induction plasmas operating with argon is \( 700/\mu \text{m} \) we find for a plasma with a characteristic radius of 0.01m, that \( \Psi = 1 \) corresponds to \( \omega \approx 2 \times 10^7 \). However, apart from the limitations to low field frequencies

![Figure 6.5. Axis value of the magnetic field of an infinite solenoid versus \( \Psi = R \sqrt{\mu_0 \sigma \omega}/2 \).](image)

the one-dimensional EM-field formulation has other important drawbacks. The neglect of the radial component of the magnetic field is not always justified if the ratio of the length and diameter of the coil is not large and if the coil consists of a few turns only. Furthermore, a pure axial magnetic field as given by Eq. (6.22) does not satisfy Eq. (6.3.1). But the most important point is the fact that for higher frequencies, i.e. \( \Psi > 1 \), the axial magnetic field distribution along the axis will deviate appreciably from the magnetic field of a finite coil in free space [13, 14]. This implies that we need a different approach for the computation of the 100MHz ICP.
6.3.2 A two-dimensional EM-field formulation

The drawbacks of the one-dimensional EM-field model are circumvented if a two-dimensional EM-field formulation is used as proposed in Refs. [13, 15]. In this model the EM-field is formulated in terms of the vector potential $\vec{A}$. The magnetic field is then given by the relation $\vec{B} = \nabla \times \vec{A}$. Substituting this expression in Eqs. (6.14) and (6.15) and if the Lorentz gauge, which is equivalent to the Coulomb gauge when the electrostatic field can be neglected is employed, the following equations are obtained for the vector potential and the electric field,

$$
\nabla^2 \vec{A} = -\mu_0 \vec{J} \quad (6.24)
$$

$$
\vec{E} = -\delta \vec{A}/\delta t. \quad (6.25)
$$

The Ohmic dissipation and Lorentz force averaged over a period of a RF cycle, are

$$
\vec{J} \cdot \vec{E} = \frac{1}{2} \sigma \omega^2 |A_\varphi|^2, \quad (6.26)
$$

$$
\vec{F}_{\text{Lorentz}} = \frac{1}{2} \sigma \text{Re}(\vec{E} \times \vec{B}^*), \quad (6.27)
$$

respectively. The notation $\text{Re}$ stands for the real part and $*$ for the complex conjugate. The general solution of Eq. (6.24) is [16]

$$
\vec{A}(\vec{r}, t) = \frac{\mu_0}{4\pi} \int_V \frac{\vec{J}(\vec{r}', t)}{|\vec{r} - \vec{r}'|} d\vec{r}' \quad (6.28)
$$

and the current density $\vec{J}(\vec{r}, t)$ consists of a contribution originating from the plasma and one from the coils. For an axisymmetrical excitation coil the vector potential, the electric field and the current density only have an azimuthal component. In the following we will use $A_\varphi$ to refer to the $\varphi$ component of the vector potential. Using Eq. (2.45) the equation for the vector potential is

$$
\nabla^2 A_\varphi = i \mu_0 \sigma \omega A_\varphi. \quad (6.29)
$$

In order to solve this elliptic equation we have to specify the boundary conditions. At the axis of symmetry $A_\varphi(0, z) = 0$. For a boundary position, represented by $\vec{R}_w$, the vector potential is given by

$$
\vec{A}(\vec{R}_w) = \frac{\mu_0}{4\pi} \int_\text{plasma} \frac{\vec{J}(\vec{r}', t)}{|\vec{R}_w - \vec{r}'|} d\vec{r}' + \frac{\mu_0}{4\pi} \sum_n I_n \int \frac{d\vec{l}'_n}{|\vec{R}_w - \vec{r}'|}. \quad (6.30)
$$
where the coil current has been written as \( \mathbf{j} \, d\mathbf{r} = \sum_n I_n \, d\mathbf{l}_n \) and \( d\mathbf{l}_n \) is an elementary arclength along a coil turn. The summation is to be taken over the number of windings. In the present model it is assumed that the coil current is not affected by the plasma so that the integral over the coil can be calculated analytically. The first term on the rhs of Eq. (6.30) describes the induced EM-field in the plasma. The problem is how to calculate the plasma contribution to the vector potential at the boundary, because the plasma current is not known. However, in view of a numerical solution an iteration process can be used to compute \( A_\varphi \). Starting from a guessed \( A_\varphi \) distribution the plasma current is computed and successively \( \mathbf{A}(\mathbf{R}_w) \) is calculated and Eq. (6.29) is solved. This process is repeated until a converged solution is obtained.

### 6.4 Transport coefficients

With the specification of the source terms of the hydrodynamical equations in the previous sections, only the transport coefficients are needed to specify the problem. Since the two-temperature nature of the plasma the LTE values of the transport coefficients cannot be used, i.e. the transport coefficients are functions of \( T_e, T_a, n_e \) and \( n_a \). A general approach and method to calculate transport coefficients is given by HIRSCHFELD et al. [17]. DEVOTO has derived simplified expressions for partially ionized argon plasmas. The background of the simplification is that the electron-heavy particle collisions are neglected in the calculation of the ion and atom transport properties [18]. In the present plasma model the heavy particle transport properties as given by DEVOTO are implemented and will not be discussed any further. The electron transport properties will be discussed in the next subsection.

#### 6.4.1 The electron transport coefficients

For the calculation of the electron transport coefficients three regimes as characterized by the ionization degree \( \alpha \) can be distinguished. At low ionization degrees (\( \alpha \lesssim 10^{-6} \)) the Coulomb interactions between electrons and ions and the mutual electron interaction can be neglected. In the so-called Lorentz limit in which the electrons only collide with (neutral) massive particles, the electron transport coefficients have the form [7, 19]

\[
\beta_n^e = G_n \int_0^\infty e^n \left( - \frac{\partial f_e^m(e)}{\partial e} \right) \, de, \tag{6.31}
\]
where $f_e^m$ is the Maxwellian distribution. For $n = 3$, $\beta^3$ represents the electrical conductivity and with $n = 5$, $\beta^5$ the thermal conductivity $\lambda_e$ can be calculated

$$\lambda_e = \lambda_e' - T_e \phi_e^2 / \sigma_e,$$  

(6.32)

where

$$\lambda_e' = \frac{4\pi n_e k}{3} \int_0^\infty \frac{c^4 f_M}{\nu_{ea}} \left[ \frac{m_e c^2}{2kT_e} - \frac{5}{2} \right]^2 dc$$  

(6.33)

$$\phi_e = \frac{4\pi n_e e}{3T_e} \int_0^\infty \frac{c^4 f_M}{\nu_{ea}} \left[ \frac{m_e c^2}{2kT_e} - \frac{5}{2} \right] dc.$$  

(6.34)

The electron-neutral particle collision frequency $\nu_{ea}$ is given by $n_a c Q_{ea}(c)$ and $Q_{ea}(c)$ is the cross-section for momentum transfer.

On the other hand, in the limit of fully ionization (FI, $\alpha > 10^{-1}$) the transport coefficients are worked out by SPITZER [20]. In addition to the electron-ion collisions also the effect of the electron-electron collisions, is taken into account. The expression for the electrical conductivity is then

$$\sigma_{FI} = \frac{32e^2}{elnA} \sqrt{\frac{\pi}{eme}} \left( \frac{2kT_e}{e} \right)^{3/2} \gamma_E = \frac{2.632 \times 10^{-2}}{elnA} T_e^{3/2},$$  

(6.35)

where $\gamma_E$ is a factor expressing the fact that $\sigma$ is determined by of electron-electron collisions and is equal to 0.582 [20]. In the nomenclature of BRAGINSKI, where the electron transport coefficients are expressed in terms of a modified electron-ion collision frequency, we have $\sigma_{FI} = n_e e^2 / 0.506 m_e (\nu_{ei})$ [21]. Also the factor $\beta$ in the electron thermal friction term (2.44) has been calculated by BRAGINSKI and equals 0.71 [21]. As in the case of very low ionization degrees the electron transport coefficients can be written in terms of pure collision frequencies.

The situation is much more complicated for any degree of ionization, i.e. $10^{-6} \leq \alpha \leq 10^{-1}$ because of the electron-electron, electron-ion and electron-atom interactions. In this regime of the ionization degree the electron transport coefficients cannot be expressed in terms of a collision frequency. A rigorous treatment of the electron transport coefficients has been given by SPITZER and HARM [22] for non-magnetized DC plasmas. BRAGINSKI generalized their results to magnetized RF plasmas [23, 24]. FROST [25] has proposed a Lorentz-like expression for the transport coefficients, i.e. in terms of pure collision frequencies (cf. Eq. (6.31)), valid in the whole range of ionization degrees by fitting the numerical results of [23]. The expressions of FROST make use of a modified electron-ion collision frequency. The FROST mixture rules for the electron
properties can then be summarized by

\[
\Gamma_n[e] = A_n \int_0^\infty \frac{e^{-w} w^{5/2}(w - 2.5)^n}{\frac{2kT_e}{m_e} w \nu_e^1(w) + K_n(n_e, T_e)} \, dw, \tag{6.36}
\]

where

\[
K_n(n_e, T_e) = B_n(8\pi n_i) \left( \frac{m_e}{2kT_e} \right)^{1/2} \left( \frac{e^2}{4\pi \varepsilon_0 m_e} \right)^2 \ln \Lambda \tag{6.37}
\]

and \( K_n \) contains the modified electron-ion collision frequency due to the electron-electron collisions. The electron-neutral momentum transfer collision frequency \( \nu_e^1(w) \) is expressed by \( \nu_e^1(w) = n_a \left( \frac{2kT_e}{m_e} w \right)^{1/2} Q_{ea}(kT_e w) \). Note that for pure electron-ion collisions \( \nu_{ei} \) scales with \( v^{-3} \). The dimension of the integral is \( m^{-2}s^3 \).

In Table (6.2) an overview is given of the various constants. The constants \( B_n \) are such that the FROST expression for the electron properties in the limit of fully ionization are the same as the SPITZER expressions.

<table>
<thead>
<tr>
<th>n</th>
<th>( \Gamma_n[e] )</th>
<th>( A_n = \frac{8e}{3\sqrt{\pi}} \left( \frac{e}{kT_e} \right)^{1-n} n_e T_e^{2-n}! )</th>
<th>( B_n )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>( \sigma_e \left[ \frac{1}{\Omega m} \right] )</td>
<td>( \frac{8e^2}{3\sqrt{\pi}} \frac{k}{m_e^2} n_e T_e )</td>
<td>( 6.425\times10^{-1} n_e T_e \frac{1}{\Omega m} \frac{m^2}{s^3} )</td>
</tr>
<tr>
<td>1</td>
<td>( \Phi_e \left[ \frac{A}{mK} \right] )</td>
<td>( \frac{8e}{3\sqrt{\pi}} \left[ \frac{k}{m_e^2} \right]^2 n_e T_e )</td>
<td>( 5.39\times10^{-5} n_e T_e \frac{A}{mK} \frac{m^2}{s^3} )</td>
</tr>
<tr>
<td>2</td>
<td>( \lambda_e \left[ \frac{W}{mK} \right] )</td>
<td>( \frac{8}{3\sqrt{\pi}} \left[ \frac{k^3}{m_e^2} \right] n_e T_e^2 )</td>
<td>( 4.775\times10^{-9} n_e T_e^2 \frac{W}{mK} \frac{m^2}{s^3} )</td>
</tr>
</tbody>
</table>
Source terms, transport coefficients ... 

References

Chapter 7

Solution procedure

In chapter 2 the physical transport equations for the macroscopic description of thermal plasma were formulated. The characteristic non-equilibrium features of the ASDF as described by the source terms in the particle and energy transport equations are given in chapter 6. Further it is assumed that the physical system under study is

1) stationary,
2) axisymmetric so that we have in fact a two-dimensional problem.

An important feature of the present computer model is related to the geometry of the physical system. The transport equations are expressed in a generalized orthogonal curvilinear coordinate system. A method to determine a boundary fitted coordinate system will be given in section 7.1. In section 7.2 the general form of the discretized equations obtained with the finite volume discretization method, will be presented. A co-located formulation [1] of the SIMPLE algorithm of PATANKAR [2] is used and will be treated in section 7.3. This chapter is concluded with a note about the development of the program (section 7.4).

7.1 Geometry

7.1.1 Need for appropriate grid

In order to solve the Eqs. (2.37, 2.41, 2.49, 2.50) they have to be written in a specific coordinate system. The equations of the physical model have a relatively simple form if they are worked out in cartesian coordinates. However, to deal with axial-symmetric problems it is more convenient to apply the cylindrical coordinate system as can be found for example in Ref. [3]. To be prepared on various difficulties as to be expected under several plasma conditions we prefer to be as general as possible with respect to geometry. Therefore the physical model is expressed in a generalized coordinate system. There are two reasons: 1) it is better to describe the boundary of the plasma as a coordinate surface, i.e. the collection of points of which one of the coordinates, say $\xi$, is constant. Since this boundary might have a capricious form, it is not sufficient to deal with cartesian,
cylindrical and spherical coordinates only. 2) In the inner space, i.e. the plasma volume we need a fine grid there where steep gradients are expected.

An example of a system with a curved boundary is the plasma generated by the cascaded arc in the expansion zone [4]. An example of steep gradients can be found in the ICP where a "small" boundary layer is confined between the active zone in which the energy is dissipated and the torch wall [5] (cf. chapter 8).

The main problem in grid generation is the determination of the mapping between the gridpoints \((x, y)\) in the physical domain and gridpoints \((\xi_1, \xi_2)\) in the computational domain (cf. Fig. 7.1) [6]. The main constraints are that the transformation is a one-to-one mapping and that the grid lines must be smooth in order to provide continuous transformation derivatives. These are needed to calculate the change in metric during the transformation. With the determination of the coordinate system and the metric tensor, the geometrical scale functions can be calculated, which are needed to express the macroscopic transport equations in generalized coordinates. Since the governing transport equations have a simpler form we will confine ourselves to orthogonal coordinate systems.

Different techniques are developed to determine the mapping between \((x, y)\) and \((\xi_1, \xi_2)\) coordinate system. These techniques are [6]:

1) complex variable method,
2) algebraic method,
3) elliptic partial differential equations.

The complex variable method corresponds with the conformal mapping method which is too restrictive in the sense that the transformation scale factors in the coordinates are equal (grid aspect ratio \(AR = 1\)). Therefore we will focus our

![Figure 7.1](image-url)
attention to the one-dimensional algebraic and two-dimensional elliptic partial differential equation methods. POPE [7] used method 3 but it is still rather restrictive in the sense that the AR is constant throughout the calculation domain. MOBLEY and STEWART [8] extended the method of POPE by introducing an extra algebraic transformation of the computational coordinates \((\xi_1, \xi_2)\) to \((\chi, \zeta)\) resulting in a grid AR which does not need to be constant. For this reason the method of MOBLEY and STEWART is used in our computer model.

### 7.1.2 Ortho-curvilinear computational coordinate system

Let the coordinates \((\xi_1, \xi_2, \xi_3)\) be functions of \((x, y, z)\), which are assumed to be single-valued and to have continuous derivatives. The invariant elementary arc length in an ortho-curvilinear coordinate system is defined by

\[
d s^2 = g_{ij} d\xi^i d\xi^j \quad \text{orthogonal to } \ell_1 (l_1 d\xi_1)^2 + (l_2 d\xi_2)^2 + (l_3 d\xi_3)^2, \tag{7.1}
\]

where \(g_{ij}\) is the metric tensor, and \(l_i\) represents the geometrical scale factors. The Jacobian \(J\) of the transformation is given by \(\sqrt{g}\) the square root of the determinant of \(g_{ij}\). For an orthogonal coordinate system the metric tensor elements are given by \(g_{ij} = \delta_{ij} \ell_i^2\). Under the assumption of axisymmetry the transport equations will not depend on \(\xi_3 = \varphi\) and the notation \(\xi_1 = \xi\) and \(\xi_2 = \eta\) will be adopted.

### 7.1.3 Two methods for orthogonal grid generation

This subsection deals with two methods of grid generation. 1) The one-dimensional (1-D) algebraic transformation and 2) the 2-D elliptical partial differential equation method. The first method can be used to extend the applicability domain of analytical (e.g. cartesian, cylindrical and spherical) coordinate systems without affecting the orthogonality. By means of an algebraic transformation it is possible to generate along an existing coordinate curve a dense grid in those regions where

![Figure 7.2. The coordinate curves of the computational coordinate system \(\xi_1, \xi_2\) drawn in the coordinate system \((x, y)\). Along the \(\xi_1\) curve the \(\xi_2\) and \(\varphi\) are constant. The geometrical scale factor \(\ell_3 = y\) corresponding to the \(\varphi\) coordinate has the meaning that a small rotation \(\delta\varphi\) creates in \(P\) an arclength \(\ell_3\delta\varphi\) perpendicular on the \(\xi_1, \xi_2\) plane.](image)
steep gradients are to be expected. This method illustrated in Fig. 7.3 will be used for the ICP configuration. Results of computations with transport equations formulated in an algebraic generated grid will be discussed in section 8.5.
and configuration of the coordinate system [9]. There is much freedom in the choice of these functions and it should be noted that in general Eq. (7.2) does not necessarily produce conformal or orthogonal grids. Since the macroscopic transport equations must be expressed in general coordinates, in which $\xi$ and $\eta$ are the independent variables, we have to transform Eq. (7.2) to the computational domain by interchanging the role of $(\xi, \eta)$ and $(x, y)$. After a considerable amount of algebra we get

$$
\alpha \frac{\partial^2 x}{\partial \xi^2} - 2\beta \frac{\partial^2 x}{\partial \xi \partial \eta} + \gamma \frac{\partial^2 x}{\partial \eta^2} = -J \times \left( \frac{\partial x}{\partial \xi} + \frac{\partial x}{\partial \eta} \right)
$$

$$
\alpha \frac{\partial^2 y}{\partial \xi^2} - 2\beta \frac{\partial^2 y}{\partial \xi \partial \eta} + \gamma \frac{\partial^2 y}{\partial \eta^2} = -J \times \left( \frac{\partial y}{\partial \xi} + \frac{\partial y}{\partial \eta} \right)
$$

(7.3)

with

$$
\alpha = x_\xi^2 + y_\xi^2 = g_{22}, \quad \beta = x_\xi x_\eta + y_\xi y_\eta = g_{12}, \quad \gamma = x_\xi^2 + y_\eta^2 = g_{11},
$$

$$
J = x_\xi y_\eta - x_\eta y_\xi = \sqrt{g},
$$

(7.4)

where $g$ equals the determinant of $g_{ij}$ and the subindices $\xi$ and $\eta$ denote partial derivatives. The elements $\alpha$, $\beta$ and $\gamma$ are the metric tensor elements and $J$ is the Jacobian of the transformation. Equation (7.3) simplifies by the orthogonality constraint, which means that $\beta = 0$. In that case, to which we confine ourselves, the geometrical scale factors $\ell_1$, $\ell_2$ and $\ell_3$ are given by

$$
\ell_1^2 = \gamma, \quad \ell_2^2 = \alpha, \quad \ell_3 = y.
$$

(7.5)

The latter relation is demonstrated in Fig. (7.2). The quantities $\ell_1$ and $\ell_2$ are depicted in Fig. 7.4 where the vector $(x_\xi, y_\xi)$ along which $\ell_1 \Delta \xi$ is directed is perpendicular to the carrier of $(x_\eta, y_\eta)$ and $\ell_2 \Delta \eta$, making the inner product $\beta = (x_\xi, y_\xi) \cdot (x_\eta, y_\eta) = 0$. Moreover the orthogonality implies that $\cos \theta = \ell_2 \Delta \eta / \ell_1 \Delta \xi$ and $\sin \theta = y_\eta \Delta \eta / \ell_2 \Delta \eta$. Figure 7.4. Geometrical meaning of $\ell_1$, $\ell_2$.

$$
x_\xi / \ell_1 = y_\eta / \ell_2 \text{ i.e. that the aspect ratio}
$$

$$
AR = \ell_2 / \ell_1 = y_\eta / x_\xi = -x_\eta / y_\xi
$$

(7.6)

The last equality is found via $\sin \theta$.

The freedom in the choice of $P(\xi, \eta)$ and $Q(\xi, \eta)$ in Eq. (7.2) will be exploited
in the following [9]. Suppose that \((\xi, \eta)\) represents a grid with AR = 1 satisfying Eq. (7.2). Then we may effectuate an algebraic transformation along the \(\xi\)-curve by introducing \(\chi\) and along the \(\eta\)-curve by introducing \(\zeta\) where

\[
\xi = f(\chi) \quad \text{and} \quad \eta = g(\zeta)
\]  

(7.7)

are monotonically increasing functions (see Fig. 7.3) Now if we choose \(P\) and \(Q\) such that

\[
P = \frac{\partial}{\partial \chi} \left( \frac{1}{f'(\chi)} \right), \quad Q = \frac{f'}{g'} \frac{\partial}{\partial \zeta} \left( \frac{1}{g'} \right)
\]

where the prime denotes a first derivative, then an equation for the transformation between \((x, y)\) and the new computational domain \((\chi, \zeta)\) is obtained. By introducing the following operator which is obtained by substituting the previous expressions for \(P\) and \(Q\) into Eq. (7.3) [8],

\[
\Gamma = \frac{\partial^2}{\partial \chi^2} + \left[ \frac{f'(\chi)}{g'(\zeta)} \right]^2 \frac{\partial^2}{\partial \zeta^2} - \frac{f''(\chi)}{f'(\chi)} \frac{\partial}{\partial \chi} - \left[ \frac{f'(\chi)}{g'(\zeta)} \right]^2 \frac{g''(\zeta)}{g'(\zeta)} \frac{\partial}{\partial \zeta}.
\]

(7.8)

Eq. (7.3) becomes

\[
[\chi] = \Gamma[y] = 0.
\]

(7.9)

As can be seen \(x\) and \(y\) are apparently independent. However, there are constraints on this independency since \(x\) and \(y\) are coupled through the application of the boundary conditions and the orthogonality constraint. From the orthogonality constraint (cf. Eq. (7.6)) and the fact that the AR in the \((\xi, \eta)\) system equals unity, we can derive that

\[
y_{\zeta} = \frac{g'}{f'} x_{\chi}, \quad x_{\zeta} = \frac{g'}{f'} y_{\chi}.
\]

(7.10)

In fact the function \(f\) and \(g\) determine via the ratio of their derivatives the AR of the \((\chi, \zeta)\) coordinate system. This method has the big advantage that the search for the orientation of a curvilinear system (a 2-D problem) is separated from the refinement along the coordinate lines (a 1-D aspect) The relations (7.10) are used to formulate the boundary conditions for Eq. (7.9) (cf. Fig. 7.5). With the specification of the boundary function \(y^+(x), y^-(x), x^+(y)\) and \(x^-(y)\) and the algebraic transformation functions \(f(\chi)\) and \(g(\zeta)\), Eq. (7.9) can be solved.

In Fig. 7.6 an example of grid generation using Eq. (7.9) is shown. The geometry of this grid is relevant to that of a thermal plasma expanding in free space. In this case the objective was searching a grid in the region bound by the curves \(y^-(x) = 0, y^+(x) = 0.5 + 1.5x^2 - x^3, x^-(y) = 0\) and \(x^+(y) = x_{max}\) with AR = 1; i.e. the algebraic transformations were taken to be unit transformations \((\xi = f(\chi) = \chi\) and \(\eta = g(\zeta) = \zeta\)). The boundary conditions related to the
Solution procedure

Figure 7.5. Boundary conditions for $x$ and $y$ in Eq. (7.9). The meaning of the boundary conditions in the physical system are shown in Fig. 7.6.

Figure 7.6. Application of grid-generation using Eq. (7.9) in which the normalized boundary function is given by $y^+(x) = 0.5 + 1.5x^2 - x^2$. The line $\eta = 0$ corresponds with the symmetry axis.

solution of Eq. (7.9) for $x(\xi, \eta)$ and $y(\xi, \eta)$ are depicted in Fig. 7.5. The grid generated inside the boundary contour is a result of the computer program. The fact that the AR in the figure is not unity is caused by the AR of the print program.

The scale factors $\ell_i$ can be computed with Eq. (7.5). If a 1-D algebraic transformation is sufficient to determine the grid, the scale factors $\ell_i$ transform according to

$$
\ell_x = \ell_1(\xi, \eta) \frac{\partial f}{\partial \chi}, \quad \ell_\xi = \ell_2(\xi, \eta) \frac{\partial g}{\partial \xi}, \quad \ell_\eta = \ell_3(\xi, \eta).
$$

(7.11)

In the following $\ell_1$ stands for $\xi$ or $\chi$ and $\ell_2$ for $\eta$ or $\zeta$. 
7.2 Numerical solution procedure

With the specification of the coordinate system the metric tensor elements $\ell_1^2$, $\ell_2^2$ and $\ell_3^2$ (cf. Eq. 7.5) are known and the system of transport equations (2.53) can be expressed in an orthogonal curvilinear coordinate system. From differential geometry it is known that the use of generalized coordinates leads to the so-called contravariant and covariant components of tensors. In an orthogonal coordinate system the covariant basis vectors $\hat{\ell}_i$, which are tangent to the coordinate lines and the contravariant basis vectors $\hat{\ell}^i$ which are perpendicular to coordinate surfaces, have the same direction. However, there is still a difference between the covariant and contravariant basis vectors in the sense that they do not need to be unitvectors, but $|\hat{\ell}_i| = |\hat{\ell}^i|^{-1}$. For this reason it is convenient to use the physical basis vectors $\ell_i$ which are unit vectors, and parallel to $\hat{\ell}_i$ and $\hat{\ell}^i$. There is a simple relation between the different vector component representations

$$ V_j = \ell_j \hat{V}^i = \hat{V}_j / \ell_j , \quad (7.12) $$

where $\hat{V}^i$ and $\hat{V}_j$ are the contravariant and covariant components of a vector, respectively. The relation between the physical-component derivative and the covariant derivative of a tensor can be found in Ref. [7].

Writing the transport equation (cf section 2.5)

$$ \vec{\nabla} \cdot \vec{J}(\Phi) = S(\Phi) \quad \text{with} \quad \vec{J} = \rho \vec{v} \Phi - \mu \Phi \vec{V} \Phi \quad (7.13) $$

in physical components we obtain [7]

$$ \frac{\partial}{\partial \xi_1} \left\{ \ell_2 \ell_3 \left[ \rho v_1 \Phi - \frac{\mu_\Phi}{\ell_1} \frac{\partial \Phi}{\partial \xi_1} \right] \right\} + \frac{\partial}{\partial \xi_2} \left\{ \ell_1 \ell_3 \left[ \rho v_2 \Phi - \frac{\mu_\Phi}{\ell_2} \frac{\partial \Phi}{\partial \xi_2} \right] \right\} = S_\Phi \ell_1 \ell_2 \ell_3 . \quad (7.14) $$

The possible quantities $\Phi$ are given in Table 2.1. Since Eq. (7.13) is written in a conservative form the finite-volume method is particular useful to discretize the Eq. (7.14). Integrating over a finite volume and using Gauss’ theorem we get for a typical control volume (cf. Fig. 7.8)

$$ I_e - I_w + I_n - I_s = S_\Phi \ell_1 \ell_2 \ell_3 \Delta \xi_1 \Delta \xi_2 \quad (7.15) $$

where the lower indices of the fluxes refer to the boundary points “east”, “west”, “north” and “south” as depicted in Fig. 7.8. The west-east direction corresponds with $\xi_1$ coordinate lines and the south-north direction with the $\xi_2$ coordinate lines. The fluxes $I(\Phi)_{e,w,n,s}$ are given in Table 7.1. Due to the assumption of axial symmetry the fluxes along the $\ell_3 = \varphi$ curve are not incorporated in Eqs. (7.14, 7.15). However, the $\ell_3$ is still present in Eqs. (7.14, 7.15). The reason is that the
elementary flux areas scale with the radial distance, i.e. $l_3$. In Fig. 7.7 this is illustrated for the radial flux area.

Note that the continuity equation is obtained by putting $\Phi = 1$ in Eq. (7.15),

$$F_e - F_w + F_n - F_s = 0.$$  \hfill (7.16)

There is an essential difference between the boundary points of a control volume denoted by small letters and the grid points denoted by capitals. The former $e, w, n, s$, serve as intermediates to get via (7.15) an interrelation between the quantities $\Phi$ at $P$ and the neighbouring grid points $E, W, N, S$. This relationship

$$a_P \Phi_P = a_E \Phi_E + a_S \Phi_S + a_W \Phi_W + a_N \Phi_N + S_\Phi \ell_1 \ell_2 \ell_3 \Delta \xi_1 \Delta \xi_2$$  \hfill (7.17)

in the computational domain will be found by using an appropriate discretization procedure to get $\Phi_e$ from $\Phi_E$ and $\Phi_P$ etc., and is described by the discretization coefficients $a_{E,S,W,N}$.

A guideline in the construction of a suitable discretization scheme is the numerical stability in discretized convection-diffusion equations [2]. PATANKAR prescribes a numerical stable discretization scheme for the control boundary values of $\Phi$ which is obtained by multiplying Eq. (7.16) with $\Phi_P$ and subtracting it from Eq. (7.15) [2],

$$(I_e - F_e \Phi_P) - (I_w - F_w \Phi_P) + (I_n - F_n \Phi_P) - (I_s - F_s \Phi_P) = S_\Phi \ell_1 \ell_2 \ell_3 \Delta \xi_1 \Delta \xi_2$$  \hfill (7.18)
The hybrid scheme is used, in which the convective term \( F \) is determined by a first-order upwind scheme, i.e. \( F_e \Phi_e = (\Phi_P, 0) - \Phi_E \max(-F_e, 0) \), when the absolute value of the grid Peclet number \( (P = F/D) \) is larger than 2 or by a second-order central difference otherwise [10]. Substituting this scheme in the expressions of \( I \) in Table 7.1 and in (7.18), it is found that

\[
\begin{align*}
I_e - F_e \Phi_P &= [D_e \max(0, 1 - 0.5|P_e|) + \max(-F_e, 0)](\Phi_P - \Phi_E) \\
I_w - F_w \Phi_P &= [D_w \max(0, 1 - 0.5|P_w|) + \max(F_w, 0)](\Phi_W - \Phi_P) \\
I_n - F_n \Phi_P &= [D_n \max(0, 1 - 0.5|P_n|) + \max(-F_n, 0)](\Phi_P - \Phi_N) \\
I_s - F_s \Phi_P &= [D_s \max(0, 1 - 0.5|P_s|) + \max(F_s, 0)](\Phi_S - \Phi_P). 
\end{align*}
\]

The discretization coefficients in Eq. (7.17) are then given by

\[
\begin{align*}
a_N &= \max \left[ \frac{|F_n|}{2}, D_n \right] - \frac{F_n}{2} \geq 0 \\
a_E &= \max \left[ \frac{|F_e|}{2}, D_e \right] - \frac{F_e}{2} \geq 0 \\
a_S &= \max \left[ \frac{|F_s|}{2}, D_s \right] + \frac{F_s}{2} \geq 0 \\
a_W &= \max \left[ \frac{|F_w|}{2}, D_w \right] + \frac{F_w}{2} \geq 0 \\
ap &= a_E + a_S + a_W + a_N > 0
\end{align*}
\]

Due to the non-linearity of the transport equations (7.13), the discretization coefficients \( a_E, S, W, N \) are functions of \( \Phi \), so that an iterative solution method will be used. The source term \( S_\Phi \) can be treated explicitly, which means that in a new iteration cycle the current value of \( \Phi \) is used to compute \( S_\Phi \). The convergence can be increased by linearizing the source term \( S_\Phi \ell_1 \ell_2 \ell_3 \Delta \xi_1 \Delta \xi_2 = \hat{S}_P \Phi_P + \hat{S}_C \)
so that the source term is treated more implicitly. Equation (7.17) is then written as [2]

\[
(a_P(\Phi^n) - \hat{S}_P(\Phi^n))\Phi_P^{n+1} = \alpha \sum_{NB} a_{NB}(\Phi^n)\Phi_{NB}^{n+1} + \alpha \hat{S}_C + (1 - \alpha)a_P(\Phi^n)\Phi_P^n
\]

(7.21)

in which an under-relaxation factor \(0 < \alpha < 1\) is introduced and \(NB\) stands for neighbour grid points. The superscript \(n\) means that the quantity \(\Phi^n\) is calculated in the \(n\)-th iteration cycle. Equation (7.21) represents a system of algebraic discretization equations for each variable \(\Phi\) and is solved with the modified strongly implicit procedure [11, 12].

### 7.3 The pressure correction equation

An iterative scheme is used to solve the momentum equations which is based on the so-called pressure-correction procedure. In this method the pressure and the velocity are decoupled in a way as described by Patankar [2]. In this pressure based method (PLE pressure linked equations) the iteration process is started with an estimated pressure field \(p^n\). Inserting \(p^n\) in a linearized momentum equation a velocity field \(\tilde{v}^{n+1}\) is generated due to the \(\nabla p\) term. This calculated \(\tilde{v}^{n+1}\) will generally not satisfy the continuity equation so that the pressure and velocity fields must be corrected. This is done using the continuity equation.

The discretized momentum equation in a control volume boundary point \(e\) in a comparable way as Eq. (7.21) [2]

\[
a_e(\tilde{v}^n, p^{n,+})v_e^{n+1,+} = \sum_{nb} a_{nb}(\tilde{v}^n, p^{n,+})v_{nb}^{n+1,+} + b + A_e(p^{n,+} - p_E^{n,+}), \quad (7.22)
\]

where the source term is split up in the \(\nabla p\) term and \(b = S_{ve}(\ell_1\ell_2\ell_3)_e(\Delta \xi_1 \Delta \xi_2)_e\) including the source terms other than the pressure force. The + sign in the superscript means that \(\tilde{v}^{n+1,+}\) does not necessarily satisfies the continuity equation. The guessed velocities \(v^{n,+}\) and pressures \(p^{n,+}\) must be corrected to satisfy the continuity equation. Thus,

\[
p^{n+1} = p^{n,+} + p', \quad v^{n+1} = v^{n+1,+} + v'. \quad (7.23)
\]

We have to derive a decoupled equation for the pressure and the velocity correction. The continuity equation is turned into an equation for the pressure correction. In order to do this we have to make a link between \(v'\) and \(p'\). Substituting Eqs. (7.23) in Eq. (7.22) it is found that

\[
a_e v'_e = \sum_{nb} a_{nb} v'_{nb} + A_e(p_P' - p_E'). \quad (7.24)
\]
A similar equation holds for the other velocity components. By substituting Eqs. (7.23) and (7.24) into the continuity equation one obtains an equation for the pressure correction \( p' \). To solve the pressure correction equation obtained in this way would cost a lot of expensive computer time compared with the solution of the momentum equation Eq. (7.22). The reason is the presence of the term \( \sum_{nb} a_{nb} v'_{nb} \) which relates the pressure correction, say \( p'_P \), with all other grid points, so that the pressure-correction equation would have a much more complicated form than Eq. (7.17). In literature much attention has been paid to overcome this numerical bottleneck [2, 13, 14]. PATANKAR proposed a method in which the link between \( p' \) and \( v' \) is made as explicit as possible, by dropping the term \( \sum_{nb} a_{nb} v'_{nb} \) in Eq. (7.24). To acknowledge the omission of this term this step is denoted by Semi-Implicit (SIM) because it does not include the implicit influence of the pressure correction at the neighbour points. Therefore the algorithm is called SIMPLE. Using Eq. (7.23) we get the following expression for \( v_e \)

\[
v_e = v_e^+ + h_e (p'_P - p'_E), \tag{7.25}
\]

where \( h_e = A_e/a_e \). With the expression Eq. (7.25) for \( v_e \) and similar expressions for \( v_w, v_n, \) and \( v_s \) we can transform the continuity equation (7.16) into a Poisson equation for the pressure correction. The discretized form of the pressure correction equation is (cf. Eq. (7.17))

\[
ap_p \rho'_P = a_E \rho'_E + a_W \rho'_W + a_N \rho'_N + a_S \rho'_S + S_{\text{press}} \tag{7.26}
\]

where

\[
\begin{align*}
a_E &= (\rho Ah)_e \\
a_W &= (\rho Ah)_w \\
a_N &= (\rho Ah)_n \\
a_S &= (\rho Ah)_s \\
ap &= a_E + a_W + a_N + a_S \\
S_{\text{press}} &= (\rho v^+_1 A)_w - (\rho v^+_1 A)_e + (\rho v^+_2 A)_s - (\rho v^+_2 A)_n
\end{align*}
\]

(7.27)

It should be noted that \( S_{\text{press}} \) is zero when the velocity field satisfies the continuity equation. By solving the pressure correction equation Eq. (7.26) we can update the pressure field using Eq. (7.23) and by using Eq. (7.25) we can update the velocity \( v^{n+1} \). The updated \( v^{n+1} \) does not fulfill completely the linearized momentum equations. In SIMPLE a new iteration cycle is started and this process is repeated until a converged solution is found. Other algorithms are developed in which more complicated pressure and velocity corrections are used.

**Staggered vs. co-located grid** In using the control-volume method it is natural to define the velocity components at other locations than the scalar quantities (such
Solution procedure

as pressure, density and temperature). This is made clear in Eq. (7.15) in which the convection terms \((F_e, F_w, F_n \text{ and } F_s)\) are not defined on the main grid points but at the boundaries of the control volumes. The same applies for the diffusive terms. Moreover, the staggered grid configuration suppresses non-physical oscillations of the pressure and the velocity fields [2]. However, RHIE et al. [1] developed successfully a method in which all physical quantities are defined on the main grid points (co-located grid method). Co-located grids are more appropriate when the physical grid is curved so that there is no need to evaluate curvature terms. Since we use orthogonal curvilinear coordinates the co-located grid method is used. Then special steps have to be taken to suppress the numerical oscillations of the pressure and the velocity fields. As can be seen from e.g. Eq. (7.26) we have to know the guessed velocities at the boundaries of the control volumes. The control volume surface values of \(v^+\) must be interpolated from the nodal values. With respect to the stability of the algorithm it is necessary to introduce an under-relaxation factor in Eq. (7.22), i.e.

\[
    a_P(\bar{v}^n, p^n)v_P^{n+1} = \alpha \sum_{NB} a_{NB}(\bar{v}^n, p^n)v_{NB}^{n+1} + (1 - \alpha)a_P(\bar{v}^n, p^n)v_P^n + \alpha b + \alpha \xi_2 A_P(p_{w}^n - p_e^n). \tag{7.28}
\]

Crucial for the success of the co-located grid method is how the interpolation scheme is defined. A linear interpolation scheme can lead to oscillatory pressure and velocity fields so that a special interpolation scheme must be used. MAJUMDAR [15] calls this the momentum interpolation. It was observed that the converged result was dependent of the under-relaxation factor. To avoid this undesirable feature of the method we write Eq. (7.28) as

\[
    v_e^{n+1} = \left[ \alpha_v \sum_{NB} a_{NB} p_{e}^{n+1} + b \right] \left[ \frac{1 - \alpha_v}{a_P} \right]_e + (v_e^n - v_e^{n+1})(1 - \alpha_v) - \alpha_v \frac{\delta \xi_2}{a_P} (p_{w}^n - p_e^n) \right] A_e, \tag{7.29}
\]

where the overline denotes linear interpolation, for example \(\bar{\Phi}_e = \frac{1}{2}(\Phi_P - \Phi_E)\). An important feature of the present interpolation scheme is the replacement of the pressure term by

\[
    \frac{\delta \xi_2}{a_P} (p_{e}^w - p_{e}^w) \right] A_e = \frac{\delta \xi_2}{a_P} (p_{e}^w - p_{e}^w) A_e. \tag{7.30}
\]

For the point \(e', \bar{a_P}\) is obtained from linear interpolation of the \(a_P\)'s calculated in the points P and E shown in Fig. 7.8. It is important to note that the control-volume velocity depends on the pressure at 2 neighbouring points so that indeed
oscillatory fields will be suppressed.

For compressible flows a pressure change leads to a density change according to the equation of state

\[(\rho v)_e = (\rho^+_e + \rho^e_0)(v^+_e + v^e_0) \cong (\rho^+_v + \rho^+ v' + \rho' v^+_e), \quad (7.31)\]

where \(\rho^+\) and \(v^+\) are guessed values and the velocity corrections are given by Eq. (7.25). The density corrections are related to the pressure corrections by

\[\rho' = (RT)^{-1}p' \equiv Kp'. \quad (7.32)\]

A first-order upwind scheme is used for the density correction [16], i.e.

\[v^+_e K_e p'_e = K_P p'_P \max(v^+_e, 0) - K_{EPE} \max(-v^+_e, 0) \quad (7.33)\]
\[v^+_w K_w p'_w = K_W p'_W \max(v^+_w, 0) - K_{PPW} \max(-v^+_w, 0). \quad (7.34)\]

Substituting similar expressions for \(v^+_n K_n p'_n\) and \(v^+_s K_s p'_s\) into the continuity equation we obtain a similar equation as (7.26), but the discretization coefficients are

\[a_P = [\rho^+_e h_e + K_P \max(v^+_e, 0)]A_e + [\rho^+_w h_w + K_P \max(-v^+_w, 0)]A_w + [\rho^+_n h_n + K_P \max(v^+_n, 0)]A_n + [\rho^+_s h_s + K_P \max(-v^+_s, 0)]A_s\]
\[a_E = [\rho^+_e h_e + K_E \max(v^+_e, 0)]A_e\]
\[a_W = [\rho^+_w h_w + K_W \max(-v^+_w, 0)]A_w\]
\[a_N = [\rho^+_n h_n + K_N \max(v^+_n, 0)]A_n\]
\[a_S = [\rho^+_s h_s + K_S \max(-v^+_s, 0)]A_s\]
\[S_{press} = -\rho^+_e v^+_e + \rho^+_w v^+_w - \rho^+_n v^+_n + \rho^+_s v^+_s. \quad (7.35)\]

With the solution for the pressure correction the new pressure and velocities can be calculated according to Eqs. (7.23). The new pressure and velocity fields are used in the next iteration cycle until consecutive changes are sufficiently small.

### 7.3.1 SIMPLE Algorithm and convergence criterion

The SIMPLE procedure can be summarized by the following scheme:

1. Guess the initial pressure field \(p^+\).
2. Solve the momentum equations to get \(v^+_1\) and \(v^+_2\).
3. Solve the pressure correction equation (7.26) with discretization coefficients (7.35).
4 Correct the pressure field and the velocity field using the pressure and velocity correction formules (7.23).

5 Solve the discretization equations for other variables described by equations of the type (7.14).

6 Regarding the corrected pressure field $p$ as a new guess $p^+$, return to step 2 and repeat the procedure until convergence.

Since an iterative solution method is used a convergence criterion must be defined. The following stop-criterion is used in the computer program. For each field quantity $\Phi$ a convergency parameter is calculated. For this purpose a slightly different form of Eq. (7.21) is used to calculate

$$Res(\Phi^n) = \sum_P \left\{ [a_P(\Phi^n) - S_P(\Phi^n)]\Phi^n_P - \left[ \alpha \sum_{NB} a_{NB}(\Phi^n)\Phi^n_{NB} + \alpha S_C + (1 - \alpha)a_P(\Phi^n)\Phi^n_P \right] \right\}.$$  

(7.36)

The sum over $P$ runs over all internal gridpoints and the convergency parameter is defined by

$$\epsilon(\Phi^n) = \frac{Res(\Phi^n)}{Res(\Phi^1)}.$$  

(7.37)

The demand for convergence is then

$$\max(\epsilon(\Phi^n_1), \ldots, \epsilon(\Phi^n_N)) \leq \epsilon,$$  

(7.38)

where $N$ is the number of physical field variables. A typical value for $\epsilon$ is $10^{-5}$. The total number of iterations required for convergence depends on the number of equations which have to be calculated. If for example $\Phi = n_e, T_e, T_h, p, \vec{v}, A_\Phi$ then the number of iterations may be as high as 20000.

7.4 Program

In a first attempt to perform modelling calculations FEY used the existing SEPRAN package based on finite elements [17]. It was concluded that this code was not appropriate to model compressible plasma flows and that a control volume based method as used in the work of MILOJEVIC was more suitable. Inspiration was drawn by the Fortran code of MILOJEVIC based on the work of PERIĆ et al. [18]. However, it was not easy to extend in this model the mass, momentum and energy equations. Therefore it was decided to construct a modular program such that for each physical quantity in Table 2.1 a separate module was developed. In this way
the program is very flexible in the sense that extensions of the program are not
difficult to implement for future applications.

For reasons of effective code generation and the portability of the program,
the C-programming language has been used. All calculations were performed on
an IBM RS-6000 RISC system.

References

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Chapter 8

Results

This chapter is divided into three parts. In the first part our attention is devoted to the code validation using analytical problems to test the computer program (section 8.1). Before discussing the two main other parts a short review of the ICP set-up and the global model is given in section 8.2 and 8.3. The second part deals (section 8.4) with the application of the numerical model to a relatively low frequency inductively coupled plasma. In the third part (section 8.5) a high-frequency inductively coupled plasma is considered. In section 8.6 general conclusions are given.

8.1 Code validation

Since the numerical solution procedure of the system of equations (7.14) is based on the SIMPLE-algorithm the basis of the program consists in the solution of the Navier-Stokes (NS) equations, i.e. the momentum and continuity equation. In the SIMPLE algorithm the pressure is treated as a primary variable. The continuity equation is then converted into a pressure correction equation (7.26). Using the finite volume method to discretize the transport equations, PATANKAR [1] recommend staggered grids to avoid unphysical oscillatory solutions. However, co-located grids can be used if a special interpolation scheme is incorporated to avoid possible numerical oscillations [2]. The success of the co-located method in the SIMPLE algorithm is shown by PERIĆ et al. [3].

Test of pressure correction module

In our work the analytical problem of Poiseuille flow is used to test the implementation of the pressure correction module in combination with the velocity components \( v_1 \) and \( v_2 \). Poiseuille flow is the laminar flow of an incompressible medium with viscosity \( \mu \) in a long pipe with length \( L \). The radial velocity \( v_2 \) is
zero and the axial velocity $v_1$ is given by

$$v_1(r) = \frac{P_{in} - P_{out}}{4\mu L} (R^2 - r^2),$$

(8.1)

where $R$ is the radius of the pipe and $P_{in}$, $P_{out}$ the inlet and outlet pressures, respectively. In the test case the following parameters were used: $L = 1$m, $\Delta p = p_{in} - p_{out} = 100$Pa, $\mu = 2 \times 10^{-5}$kg/m/sec and $R = 0.02$m so that $v_1(0) = 500$m/sec. The test of the code is demonstrated in Fig. 8.1 in which $v_1(0)$ versus number of radial grid points is shown. In these calculations the number of axial grid points (30) was kept constant. It must be stressed that in using a two-dimensional computer model to solve the one-dimensional problem numerical source terms originating from finite discretization are introduced in the one-dimensional equation. Therefore $v_1(0)$ approaches for an increasing number of radial grid points to a value which differs only slightly from the analytical value. Increasing the number of axial grid points the effect of the numerical source terms is reduced. A significant number to trace the convergence in the SIMPLE algorithm is the convergency parameter $\epsilon(p')$ for the pressure correction $p'$ (cf. Eq. (7.37). This parameter represents the relative mass defect (residue) of the continuity equation,

$$\epsilon(p') = \left| \frac{\sum_{gp} S^p_{\text{press}}(gp)}{\sum_{gp} S^1_{\text{press}}(gp)} \right|$$

(8.2)

where $S^p_{\text{press}}(gp)$ is the pressure correction source term in the grid point $gp$ at the $n$-th iteration. The rate of convergence of the pressure correction equation is shown in Fig. 8.2 for different number of radial grid points.
Results

Test of the grid module

In order to investigate the validation and the implementation of the grid generation module the same analytical problem of Poiseuille flow is used. Only the results of the test calculations including algebraic transformations for the combination of the $v_1$ and the pressure correction will be presented. The radial transformation function being implemented is

$$ r/R = g(\xi_2) = \ln(1 + N\xi_2)/\ln(1 + N) \quad (8.3) $$

which produces a grid similar to that shown in Fig. 8.18. The number of grid points is taken to be $N_Z = 30$ and $N_R = 40$. In Table 8.1 the comparison between the analytical and numerical results based on the transformation Eq. (8.3) with $N = 5$, for the Poiseuille flow is shown. The agreement between the numerical and analytical results is excellent. With a “normal” grid the difference between the analytical and numerical value for $v_1(0)$ was 1%, whereas with a radial transformation the difference is found to be 0.2%. This comparison shows that the computation is improved by using a transformed grid.

Other modules

The modules for $n_e$, $T_e$ and $T_h$ have been successfully tested separately using cylindrical and spherical coordinate systems including algebraic transformation.
Table 8.1. Comparison between analytical and numerical results including a radial transformation.

<table>
<thead>
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<th>$\xi_2$</th>
<th>$r$ (m)</th>
<th>$v_{\text{an}}$ (m/sec)</th>
<th>$v_{\text{num}}$ (m/sec)</th>
</tr>
</thead>
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<tr>
<td>0.000</td>
<td>0.0000</td>
<td>500.00</td>
<td>501.02</td>
</tr>
<tr>
<td>0.232</td>
<td>0.0086</td>
<td>407.55</td>
<td>408.25</td>
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<td>0.411</td>
<td>0.0125</td>
<td>304.69</td>
<td>306.29</td>
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<td>0.625</td>
<td>0.0158</td>
<td>187.95</td>
<td>187.42</td>
</tr>
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<td>0.0200</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

for both coordinate systems. The vector potential module is tested using the analytical problem of the magnetic field determination in an infinite solenoid with uniform load. This module is of particular importance in the inductively coupled plasma.

It is not possible to compare the results of the global interplay of all modules with analytical results. Therefore existing model calculations are used as a reference to compare the results obtained with our program [4]. The discussion concerning this matter will be a part of section 8.4.

8.2 ICP modelling

The model for the description of thermal plasmas outlined in chapters 2-7 is applied to atmospheric inductively coupled plasmas (ICP's). The ICP is created and sustained by induction from a RF magnetic field generated by an induction coil. Figure 8.3 shows a schematic view of an open ICP configuration suitable for spectroscopic applications. For a more detailed description of the ICP configuration we refer to chapter 1 and Ref. [5]. An important parameter determining the operation of the ICP is the oscillator frequency. For this reason two ICP configurations as shown in Fig. 8.3 will be considered in this study. First the set-up as used by Boulos [6, 7] is discussed. In this set-up the ICP is driven by a 3MHz RF-field. The frequency is such that the electromagnetic skin depth, as discussed in section 6.3.1 is relatively large, i.e. the skin depth $\sim$ torch radius. An important consequence is that the energy dissipation takes place in a relatively large region so that large gradients are not expected. The other configuration as used by Fey [5], is driven by a much higher frequency, namely 100MHz. In this configuration the skin depth is smaller than the plasma torch radius so that a very local energy dissipation will lead to relatively large gradients.
For the theoretical investigation of the ICP, models have been developed in order to study the various physical processes and their interrelationships. An obvious advantage of models is that the effect of different operating conditions can be investigated, which is important with respect to the optimization of the various applications. In the first attempts to describe the ICP one-dimensional models were developed [8]. Of principal interest were the radial temperature distributions at the coil position. These simplified models were based on the balance between energy dissipation and conductive energy transport and radiative losses, while convective transport and axial conduction were omitted. Important drawbacks of one-dimensional models are the fact that the temperature outside the active zone (cf. Fig. 8.3) and the flow field cannot not be determined. Therefore two-dimensional models were developed [6, 9] which are further extended in this work.

In the next section the transport equations are formulated in a cylindrically symmetric oriented coordinate system.

8.3 Model

In chapter 2 the conditions for a hydrodynamical description of the plasma were formulated and it was demonstrated that these conditions are fulfilled so that the plasma can be treated as a fluid. To calculate the flow, particle density and temperature distributions a laminar model is used in which the plasma is considered to be stationary, two-dimensional axisymmetric and quasi-neutral. The assumption of a laminar flow is validated if the Reynolds ($Re$) number, defined as $Re = L \rho v_0 / \mu$, is smaller than 2000 [10]. In the ICP set-ups the characteristic length $L$ and velocity $v_0$ are 0.01m and 10m/sec [5], respectively,
while $\rho \sim 0.2\text{kg}/\text{m}^3$ and the molecular viscosity $\mu \sim 2 \times 10^{-4}\text{kg}/\text{msec}$ [11] so that $Re \sim 10^2 \ll 2000$.

To account for non-equilibrium effects in thermal plasmas a two-temperature approach is adopted (Ref. [7, 12]) in which the plasma is considered as being composed of electrons and heavy particles (neutrals and ions). The RF power is coupled in the electron gas and is dissipated during electron-heavy particle collisions. Radiation and energy transfer to the heavy particles by means of elastic and inelastic collisions are the main energy loss mechanisms for the dissipated power $\sigma E^2 = \sigma \omega^2 A_r^2$. In their turn the heavy particles transfer energy to the environment by conductive and convective energy transport.

The continuity, momentum and energy equations have the following general form:

$$\frac{\partial}{\partial \xi_1} \left\{ \ell_2 \ell_3 \left[ \rho v_1 \Phi - \frac{\mu_\phi}{\ell_1} \frac{\partial \Phi}{\partial \xi_1} \right] \right\} + \frac{\partial}{\partial \xi_2} \left\{ \ell_1 \ell_3 \left[ \rho v_2 \Phi - \frac{\mu_\phi}{\ell_2} \frac{\partial \Phi}{\partial \xi_2} \right] \right\} = S_\phi \ell_1 \ell_2 \ell_3, \quad (8.4)$$

and a cylindrical oriented coordinate system will be used. The coordinates $\xi_i$ and scale factors $\ell_i$ for a pure cylindrical coordinate system are given in Table 8.2. It is not difficult to include a one-dimensional algebraic transformation which can be performed by transforming the $\ell_i$ according to Eq. (7.11). Then the coordinate lines hold their orientation and the scale factors $\ell_i$ are functions of one coordinate variable only, i.e. $\ell_1 = \ell_1(\xi_1)$, $\ell_2 = \ell_2(\xi_2)$ and $\ell_3 = \ell_3(\xi_2)$.

The terms with $v$ in Eq. (8.4) account for convective transport while those containing $\mu_\phi$ represent conductive/diffusive transport. The field variables $\Phi$, transport coefficients $\mu_\phi$ and the source terms are given in Table 8.3. In view of future application of the model to the 100MHz ICP the computation of the electromagnetic field is based on the self-consistent two-dimensional vector potential model (section 6.3.2).

The system of equations (8.4) is solved using the SIMPLE algorithm based on the finite volume discretization method (cf. chapter 7) [1]. In order to solve Eqs (8.4) appropriate boundary conditions must be specified. The boundary conditions for a plasma configuration shown in Fig. 8.4 are summarized in Table 8.4.
<table>
<thead>
<tr>
<th>$\Phi$</th>
<th>$\mu_\Phi$</th>
<th>Source term $S_\Phi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1$</td>
<td>$0$</td>
<td>$n_e n_\alpha S_{CR} - n_e n_+ \alpha_{CR}$</td>
</tr>
<tr>
<td>$\frac{n_e}{\rho}$</td>
<td>$DA$</td>
<td>$\frac{-1}{\ell_1} \partial p \frac{\partial p}{\partial \xi_1} + \frac{\partial}{\partial \xi_1} \left[ \frac{\mu}{\ell_1} \partial v_1 \right] + \frac{1}{\ell_1 \ell_2 \ell_3} \frac{\partial}{\partial \xi_2} \left[ \mu_3 \partial v_2 \right] + (\bar{J} \times \bar{B})_1$</td>
</tr>
<tr>
<td>$v_1$</td>
<td>$\mu$</td>
<td>$\frac{-1}{\ell_2} \partial p \frac{\partial p}{\partial \xi_2} + \frac{\partial}{\partial \xi_2} \left[ \frac{\mu}{\ell_2} \partial v_1 \right] + \frac{1}{\ell_1 \ell_2 \ell_3} \frac{\partial}{\partial \xi_2} \left[ \mu_3 \partial v_2 \right] - 2\mu v_2 \left[ \frac{\partial}{\partial \xi_2} \left( \frac{1}{\ell_2 \ell_3} \partial \ell_3 \right) \right] + \rho v_3 v_3 \frac{\partial}{\partial \xi_2} \left( \frac{\partial}{\partial \xi_2} \right)$</td>
</tr>
<tr>
<td>$v_2$</td>
<td>$\mu$</td>
<td>$\frac{-v_3 \mu}{\ell_2 \ell_3} \left[ \partial \ell_3 \left( \frac{1}{\ell_2 \ell_3} \partial \ell_3 \right) \right] + \frac{1}{\ell_2 \ell_3} \left[ \frac{\partial}{\partial \xi_2} \left( \frac{\partial}{\partial \xi_2} \right) \right] - \frac{v_3 v_2 \rho}{\ell_2 \ell_3} \left[ \frac{\partial}{\partial \xi_2} \right]$</td>
</tr>
<tr>
<td>$\frac{5 n_e k T_e}{2 \rho}$</td>
<td>$\kappa_e$</td>
<td>$\frac{v_1}{\ell_1} \frac{\partial}{\partial \xi_1} + \frac{v_2}{\ell_2} \frac{\partial}{\partial \xi_2} + \frac{1}{2} \alpha^2</td>
</tr>
<tr>
<td>$\frac{5 n_h k T_h}{2 \rho}$</td>
<td>$\kappa_h$</td>
<td>$\frac{v_1}{\ell_1} \frac{\partial}{\partial \xi_1} + \frac{v_2}{\ell_2} \frac{\partial}{\partial \xi_2} + Q_{eh}$</td>
</tr>
</tbody>
</table>

Table 8.3. Field variables, transport coefficients and source terms in the transport equations 8.4 and $n_h = n_\alpha + n_e$. The source terms are given in a cylindrical geometry oriented coordinate system. This means that a one-dimensional algebraic coordinate transformation can be included so that $\ell_1 = \ell_1(\xi_1)$, $\ell_2 = \ell_2(\xi_2)$ and $\ell_3 = \ell_3(\xi_3)$. For a purely cylindrical coordinate system the scale factors $\ell_i$ are given in Table 8.2.

### 8.4 3 MHz ICP

In this section the model calculations applied to an ICP generated by a 3 MHz RF-field are presented. This relatively low frequency condition is often used in modelling as found in literature. In Table 8.5 an overview is given of various oscillator frequencies as used by different groups.

By comparing the results found in literature with our results obtained under the same conditions we can test the implementation of the program. In the next subsection the reliability of our program is investigated using the results of Ref. [4].
Figure 8.4. The ICP torch geometry. The computational domain is enclosed by thick lines.

<table>
<thead>
<tr>
<th>$\Phi$</th>
<th>inlet</th>
<th>axis</th>
<th>torch</th>
<th>outlet</th>
<th>Eq. num.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_e$</td>
<td>$\frac{\partial n_e}{\partial z} = 0$</td>
<td>$\frac{\partial n_e}{\partial r} = 0$</td>
<td>$\frac{\partial n_e}{\partial r} = 0$</td>
<td>$\frac{\partial n_e}{\partial z} = 0$</td>
<td>8.5</td>
</tr>
<tr>
<td>$T_e$</td>
<td>$\frac{\partial T_e}{\partial z} = 0$</td>
<td>$\frac{\partial T_e}{\partial r} = 0$</td>
<td>$\frac{\partial T_e}{\partial r} = 0$</td>
<td>$\frac{\partial T_e}{\partial z} = 0$</td>
<td>8.6</td>
</tr>
<tr>
<td>$T_h$</td>
<td>$T_0$</td>
<td>$\frac{\partial T_h}{\partial r} = 0$</td>
<td>$\frac{\partial T_h}{\partial r} = \frac{\kappa_w}{\kappa_h \delta_w} (T_h - T_0)$</td>
<td>$\frac{\partial T_h}{\partial z} = 0$</td>
<td>8.7</td>
</tr>
<tr>
<td>$v_1$</td>
<td>$v_1 = \frac{Q_i}{\pi (R_i^2 - R_{i-1}^2)}$, $R_{i-1} &lt; r &lt; R_i$</td>
<td>$\frac{\partial v_1}{\partial r} = 0$</td>
<td>$v_1 = 0$</td>
<td>$\frac{\partial \rho v_1}{\partial z} = 0$</td>
<td>8.8</td>
</tr>
<tr>
<td>$v_2$</td>
<td>$v_2 = 0$</td>
<td>$v_2 = 0$</td>
<td>$v_2 = 0$</td>
<td>$\frac{\partial v_2}{\partial z} = 0$</td>
<td>8.9</td>
</tr>
<tr>
<td>$v_3$</td>
<td>$v_3 = (v_3)<em>i$, $R</em>{i-1} &lt; r &lt; R_i$</td>
<td>$v_3 = 0$</td>
<td>$v_3 = 0$</td>
<td>$\frac{\partial \rho v_3}{\partial z} = 0$</td>
<td>8.10</td>
</tr>
</tbody>
</table>

Table 8.4. The boundary conditions. $\kappa_w$, and $\delta_w$ are the thermal conductivity and the thickness of the torch wall, respectively. $T_0$ is the temperature outside the torch which is taken to be 350 K. $R_0 = 0$ and $(v_3)_i$ has to be specified.

8.4.1 First results

In order to obtain a global converged solution a suitable set of underrelaxation factors for each $\Phi$ in Table 8.3 must be chosen. In Ref. [1] specific values for the underrelaxation factors for the velocity components (0.7) and the pressure
**Results**

<table>
<thead>
<tr>
<th>group</th>
<th>Total Power input (W)</th>
<th>frequency</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>MOSTAGHIMI et al.</td>
<td>5k</td>
<td>0.3 MHz</td>
<td>[7]</td>
</tr>
<tr>
<td>MOSTAGHIMI and BOULOS</td>
<td>5k</td>
<td>3 MHz</td>
<td>[4]</td>
</tr>
<tr>
<td>CHEN</td>
<td>3 – 7k</td>
<td>3 MHz</td>
<td>[13]</td>
</tr>
<tr>
<td>MILLER and AYEN</td>
<td>2 – 6k</td>
<td>4 MHz</td>
<td>[9]</td>
</tr>
<tr>
<td>FEY (experimental)</td>
<td>600 – 800</td>
<td>100 MHz</td>
<td>[5]</td>
</tr>
</tbody>
</table>

Table 8.5. Various oscillator frequencies in the ICP: modelling and experimental corrections (0.3) were recommended.

In the first calculations both the global density $\rho$ and the temperature were underrelaxed. At that time we were faced with problems of convergence of the relative mass defect as defined by Eq. 8.2. The number $\epsilon(p')$ remained relatively constant and had a typical value between $1$ and $10^{-1}$. A considerable improvement of the convergence of $\epsilon(p')$ was obtained by noticing that in the SIMPLE algorithm $\rho$ is given by the equation of state rather than a differential equation of the type (8.4). Therefore it is recommended not to relax the global density $\rho$.

The specification of the torch dimension and the operating conditions are given in Table 8.6. The equations were solved using a $35 \times 24$ grid in the axial

<table>
<thead>
<tr>
<th>$R_1$</th>
<th>$R_2$</th>
<th>$R_3$</th>
<th>$R_{coil}$</th>
<th>$L_T$</th>
<th>$L_1$</th>
<th>$L_c$</th>
<th>$f$</th>
<th>$P_0$</th>
<th>$Q_1$</th>
<th>$Q_2$</th>
<th>$Q_3$</th>
<th>$p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0017 m</td>
<td>0.0188 m</td>
<td>0.0250 m</td>
<td>0.0330 m</td>
<td>0.170 m</td>
<td>0.010 m</td>
<td>0.070 m</td>
<td>f</td>
<td>3MHz</td>
<td>5kW</td>
<td>1 liter/min</td>
<td>3 liter/min</td>
<td>21 liter/min</td>
</tr>
</tbody>
</table>

Table 8.6. Torch specification (cf. Fig. 8.4) of the 3MHz ICP. The number of equidistant windings is 3. $L_1$ and $L_c$ denote the position of the first and last windings, respectively.

and radial direction, respectively. Results of our computations are compared with those published in Ref. [4]. It is important to note that the computations carried out in Ref. [4] were based on a LTE-model whereas ours are based on a two-temperature (2-T) model. The differences between the LTE and the 2-T model for
the axial temperature (heavy particle temperature in the 2-T model) are smaller than 4% [7, 12]. For this reason a comparison can be made between the axial heavy particle temperature obtained with the 2-T model and the axial temperature found in the LTE-model of Ref. [4]. In Fig. 8.5 the comparisons for the axial
temperature and $v_1$ distributions are shown. The agreement between the axial temperature calculated by us and Ref. [4] is excellent. Differences between the extreme values of $v_1$ on the axis of the order of 10% are observed. A comparison is also made for the radial distribution of the energy dissipation $\sigma \omega^2 |A_\varphi|^2$ at $z=0.052\text{m}$. In the next subsection it will be shown that according to the model for $r < 0.015\text{m}$ in the active zone the electron and heavy particle temperature are almost equal. For this inner region the 2-T and LTE predictions for $\sigma \omega^2 |A_\varphi|^2$ may not be too different. From Fig. 8.6 it is indeed observed that for $r < 0.015\text{m}$ the agreement is excellent. The differences for $r > 0.015\text{m}$ are due to the fact that our calculations are based on the 2-T model. In the 2-T calculations of Ref. [7] and ours presented in the next subsection it is demonstrated that near the torch wall both $T_e$ and $n_e$ are larger than the values obtained with a LTE-model so that the electrical conductivity $\sigma_{2-T}$ is also larger than $\sigma_{LTE}$. As a result the Ohmic dissipation $[\sigma \omega^2 |A_\varphi|^2]_{2T}$ will be larger than $[\sigma \omega^2 |A_\varphi|^2]_{LTE}$ which again has a positive feedback on $n_e$ and $T_e$.

Also shown in Fig. 8.6 is a typical course of the relative residues of $n_e$, $v_2$, $A_\varphi$ and the pressure correction versus number of iterations. After a transient “period” determined by the set of underrelaxation factors the global rate of the convergence is logarithmic. Concluding we may state that taking the existing literature as a reference the physical model outlined in chapters 2 – 6 is correctly implemented.

### 8.4.2 Discussion

In the majority of works concerning ICP modelling, the input parameters determining the configuration were varied [13, 14], such as the total power, input inlet gas flow rate, configuration of the coil, etc. In this study the influences of the particle source term (CR-models) needed in 2-T models and the radiative losses as developed in chapters 3 – 5 on the particle and temperature field will be investigated.

Experimental data for both the effective recombination coefficient and the total radiative loss measured by DESAI et al. [15] and EVANS et al. [16], respectively, were used by MOSTAGHIMI et al. [7]. The effective ionization coefficient $S_{CR}$ is then obtained via the principle of detailed balancing $S_{CR} = n_+ \alpha_{CR} / n^S(1)$.

With the operating conditions specified in Table 8.6 in which $L_1 = 0.030\text{m}$ and $L_e = 0.090\text{m}$, the following model calculations were carried out:

**Run:**

1. $S_{CR}$ and $\alpha_{CR}$: DESAI and CORCORAN [15].
   - Radiation loss: EVANS and TANKIN [16]. See Figures 8.7. (DC-ET)

2. $S_{CR}$ and $\alpha_{CR}$: DESAI and CORCORAN.
   - Radiation loss: WILBERS et al. [17]. See Figures 8.8. (DC-W)
A characteristic feature of relatively low frequency driven ICP's is that the energy dissipation takes place in a relatively large plasma volume, since the ratio \( \Psi \) of the torch radius and the skin depth \( \delta = \sqrt{2/\mu_0 \sigma \omega} \) is of the order 1 (cf. Fig. 6.3). This means that the Ohmic dissipation itself imposes in the inner plasma region relatively homogeneous profiles. The electric field increases towards the wall so that for the outer region \( \sigma E^2 \) will increase as well. However, in the outer plasma region near the torch wall conductive energy losses cool the plasma. The decrease in temperature reduces the electrical conductivity so that the Ohmic dissipation will show an off-axis maximum (cf. Fig. 8.6). Figures 8.7-8.10 show the electron and heavy particle temperatures and the electron density in an atmospheric argon plasma with the aforementioned source terms. The electron density has a clear off-axis maximum in all cases. Both \( T_e \) and \( T_h \) have an off-axis maximum as well but these are not always visible in the representation given because the contour steps are in some cases too large. However the profiles of \( T_e \) and \( T_h \) located inside the steep gradients close to the wall are flatter than those of \( n_e \).

In Table 8.7 the values and the positions maxima of the \( T_e \) and \( n_e \) profiles are shown for the different runs. Radial distributions of \( n_e \) and \( T_e \) for run 1 at \( z=0.03 \),

<table>
<thead>
<tr>
<th>Run</th>
<th>( T_e(\text{max}) ) (K)</th>
<th>( n_e(\text{max}) ) ( \times 10^{-3} \text{m} )</th>
<th>( z_{\text{max}} ) ( \times 10^{-3} \text{m} )</th>
<th>( \int Q_{\text{rad}}dV ) (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1: DC-ET</td>
<td>9720</td>
<td>15.3</td>
<td>87.7</td>
<td>650</td>
</tr>
<tr>
<td>2: DC-W</td>
<td>9140</td>
<td>16.6</td>
<td>87.7</td>
<td>950</td>
</tr>
<tr>
<td>3: DC-B</td>
<td>9860</td>
<td>13.1</td>
<td>93.0</td>
<td>610</td>
</tr>
<tr>
<td>4: B-B</td>
<td>10650</td>
<td>13.1</td>
<td>87.7</td>
<td>715</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Run</th>
<th>( n_e(\text{max}) ) (m(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1: DC-ET</td>
<td>( 1.13 \times 10^{22} )</td>
</tr>
<tr>
<td>2: DC-W</td>
<td>( 5.90 \times 10^{21} )</td>
</tr>
<tr>
<td>3: DC-B</td>
<td>( 1.30 \times 10^{22} )</td>
</tr>
<tr>
<td>4: B-B</td>
<td>( 1.71 \times 10^{22} )</td>
</tr>
</tbody>
</table>

Table 8.7. Maximum values for the \( T_e \) and \( n_e \) profiles and the total radiative loss for the different runs.

0.09 and 0.15m are shown in Fig. 8.11. The first two axial positions correspond with the first and last winding of the coil. At the first winding position both \( n_e \)
Figure 8.7. Contour plots of electron density (a), temperature (b) and heavy particle temperature (c) with run DC-ET. The electron density contours are given in $10^{20}$ m$^{-3}$. The contour steps are: $\Delta n_e = 10^{21}$ m$^{-3}$, $\Delta T_e = 250$K and $\Delta T_h = 500$K.
Figure 8.8. Contour plots of electron density (a), temperature (b) and heavy particle temperature (c) with run DC-W. The electron density contours are given in $10^{20}$ m$^{-3}$. The contour steps are: $\Delta n_e = 10^{21}$ m$^{-3}$, $\Delta T_e = 250$ K and $\Delta T_h = 500$ K.
Results

Figure 8.9. Contour plots of electron density (a), temperature (b) and heavy particle temperature (c) with run DC-B. The electron density contours are given in $10^{20} \text{m}^{-3}$. The contour steps are: $\Delta n_e = 10^{21} \text{m}^{-3}$, $\Delta T_e = 250 \text{K}$ and $\Delta T_h = 500 \text{K}$. 
Figure 8.10. Contour plots of electron density (a), temperature (b) and heavy particle temperature (c) with run B-B. The electron density contours are given in $10^{20} \text{m}^{-3}$. The contour steps are: $\Delta n_e = 2 \times 10^{21} \text{m}^{-3}$, $\Delta T_e = 500 \text{K}$ and $\Delta T_h = 500 \text{K}$. 
Results

and $T_e$ do not show an off-axis maximum. Diffusion for electrons and conduction for the temperature and recirculation near the torch inlet are responsible for the fact that the profile has its maximum on the axis. In Ref. [4] and from Fig. 8.5 it can be seen that this recirculation is accompanied with a back flow near the axis of about 5m/sec. Using this value we can compare the axial convection component $|\vec{V} \cdot (n_e \vec{v})| \sim |5 \times 10^{21} \times (-5)/10^{-2}|$ with the radial diffusion component $|\vec{V} \cdot (D_A \vec{V} n_e)| \sim |5 \times 10^{21}(2 \times 10^{-3})/(5 \times 10^{-3})^2|$. It turns out that recirculation is the main transport mechanism for heating the central region near the torch inlet. Radial and axial distributions of $T_e$ and radial distributions of $T_h$ and $n_e$ are shown in Figs. 8.12 and 8.13, respectively. The radial distributions are shown for $z=0.087$m corresponding with the last winding of the coil and for which the maximum values are found. In the following the nomenclature "inner region" is used to denote the region $r < r_{max}$ where $r_{max}$ is the radial position of the profile maximum. Comparison of the radial distribution of $T_e$ and $T_h$ of the same runs reveals that $T_e \approx T_h$ in the inner region. This is due to the high collision frequency between electrons and heavy particles. For the outer regions the differences between $T_e$ and $T_h$ increase towards the torch wall. Due to these colder regions the energy transfer from electron to heavy particles decreases.

Influence of radiative losses. By comparing the results of runs 1 - 3 the influence of the radiative loss on the calculated profiles is investigated. A comparative study of the influence of the radiative loss is also performed by PROULX et al. [19] in which radiative loss data of CRAM were compared with data of [8, 16]. In Table 8.7 the volumetric radiative losses are also shown. The integration is performed over the whole computational domain. The computational data of WILBERS et al. [17] based on a non-LTE model allowing $T_e$ and $n_e$ to be decoupled but in which the excited states are populated according to Saha (cf. Eq. (2.6)), give the highest total radiative loss. The non-LTE model of BENYOY et al. [20] for the radiative loss in which also the atomic state distribution function is allowed to deviate from equilibrium and the resonant radiation is trapped, leads to a total radiative loss differing 6% with the calculations based on the experimental data of EVANS and TANKIN [16]. This apparent agreement between the run DC-ET and DC-B merely is a coincidence. The radiative loss data of Ref. [16] fitted by MILLER and AYEN [9] have a temperature threshold at 9500 K (cf. Fig. 5.4) and below this threshold the radiative loss is zero. Above this value the radiative loss increases more than the radiation loss predicted by BENYOY et al. [20]. In Fig. 5.4 the radiative loss ($Q_{rad}$) of Ref. [20] are also depicted from which it is clear that $Q_{rad}$ does not show a temperature threshold. The difference in radiation
loss above 9500 K between the two sets of data is compensated by the radiative loss in the lower temperature region in the data set of Ref. [20]. It should be stressed that the radiative loss data of Refs. [17, 20] are based on the assumption that the free-bound to the ground state may be neglected. This is a reasonable assumption since the free path of a resonant recombination photon \( \lambda_f = \frac{1}{n_\alpha \sigma_{ph}} \), where the ground state photo-ionization cross-section \( \sigma_{ph} \approx 3.5 \times 10^{-21} \text{m}^2 \) [21].
and $n_a \sim 10^{24} \text{m}^{-3}$, is about 0.003m which is smaller than the plasma dimension.

A larger radiation loss results in an overall lower temperature (electron and heavy particle) and $n_e$ field (cf. Figs. 8.7-8.9). The differences between the maximum $T_e$ are 600 $\sim$ 700K while the maximum values for $n_e$ differ with a factor of 2. On the axis the differences can be as high as 1000K (Fig. 8.12) while the differences in $n_e$ increase up to a factor of 4 (Fig. 8.13). The $n_e$ is much more sensitive for radiation losses than $T_e$. With accurate measurements of $n_e$ it is possible to distinguish between the different sets for the radiative loss data. We can conclude that $n_e$ is a more suitable parameter in tracing non-equilibrium processes than $T_e$ [22, 23].

Influence of ionization/recombination. In this part we consider the consequences of the differences between the data for ionization/recombination of DESAI and CORCORAN [15] and BENOY et al. [18] (cf. also chapter 6). Comparing Figs. 8.9-8.10 we see that in the hottest part where $T_e \approx T_h$ the temperature and $n_e$ differ 1000 K and a factor 1.4, respectively. The reason for this difference is found in the expression of the effective ionization coefficient $S_{DC}^{DC}$ using the data of DESAI and CORCORAN. From the expression for the recombination coefficient given in Eq. (6.7), we find

$$S_{CR}^{DC} = 2.56 \times 10^{25 - (3410/T_e)} T_e^{0.3} n_e^{-1.64} \exp \left( \frac{-E_+}{kT_e} \right), \text{m}^3/\text{sec}$$  (8.11)
A remarkable feature of this expression is the negative power of \( n_e \). The background is that \( S_{DCR} \) is based on dissociative recombination. Dissociative recombination is effective if the resulting excited atom makes a radiative transition. In order that radiative decay takes place \( n_e \) may not be too high otherwise excitation or ionization of this level would occur. These arguments make it plausible that dissociative recombination scales with a negative power of \( n_e \).

Ionization leads to an increase of \( n_e \) so that according to Eq. (8.11) \( S_{DCR} \) will decrease by the increase of \( n_e \) if \( T_e \) would not change. In contrast, the \( S_{CR} \) based on the collisional-radiative model of Ref. [18] does not show this \( n_e \) dependency. Hence, by using \( S_{DCR}^{CR} \) less electrons are created then when \( S_{CR} \) of Ref. [18] is used.

### 8.4.3 Conclusion

The results presented in this section demonstrate the usefulness of our model in tracing the effects of non-equilibrium aspects of ionization/recombination and radiation losses on the temperature and electron density profiles. In all cases the electron density is a suitable parameter to distinguish between the different models for the aforementioned processes. Accurate measurements of the electron density in this type of plasmas can give a decisive answer to the question of which model is the best in describing ionization/recombination and radiative loss.

### 8.5 100 MHz ICP

In this section we apply the model to a 100MHz ICP, which is mainly used as a spectroscopic analysis device [24, 23]. The torch dimension and the operating conditions of the ICP as used by FEY [5] are shown in Table 8.8. Typical values of the plasma parameters in the active zone are \( n_e \sim 1 - 3 \times 10^{21} \text{m}^{-3} \) and \( T_e \) has values between 5000 \( \sim \) 14000K [5, 25]. NOWAK et al. measured radial profiles of the electron density and temperature [25]. In Fig. 8.14 such profiles are shown for an axial position between the windings of the coil. It can be seen that the profile maxima are located at \( r \sim 0.005 \text{m} \). Using the power interruption method FEY determined the ratio between \( T_e \) and \( T_h \) in the active zone and found values of \( T_e/T_h \sim 1.35 \) [27]. With this experimental data at our disposal we can compare the results of our model calculations.

It is important to mention that the outer flow \( Q_3 \), which forms a cold sheath gas preventing the plasma reaching the torch wall, has a tangential component (cf. Fig. 8.3). It is empirically known that this so-called swirl has a stabilizing effect on the operation of the plasma [28]. Moreover, for the 100MHz ICP the swirl is
Results

Figure 8.14. Measured electron density (left) and temperature (right) as function of the radial position between the windings of the coil. The dashed curve gives the results obtained with $H_\beta$ broadening measurements [26] and the full lines measurements using the absolute line intensities. The upper curve is determined by assuming an overpopulation of the ground state density by a factor of 10 with respect to the Saha density, the middle one by assuming no overpopulation and the lowest one by assuming an underpopulation of a factor of 10 [25]. The temperature is obtained from $n_e$ and the extrapolation of the measured excited level population to the continuum [22].

necessary for the existence of the plasma. Therefore calculations were carried out with and without swirl.

The main difference with the ICP considered in the previous section is

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_1$</td>
<td>$7.5 \times 10^{-4}$ m</td>
</tr>
<tr>
<td>$R_2$</td>
<td>0.008 m</td>
</tr>
<tr>
<td>$R_3$</td>
<td>0.009 m</td>
</tr>
<tr>
<td>$R_{coil}$</td>
<td>0.0150 m</td>
</tr>
<tr>
<td>$L_T$</td>
<td>0.150 m</td>
</tr>
<tr>
<td>$L_1$</td>
<td>0.012 m</td>
</tr>
<tr>
<td>$L_c$</td>
<td>0.037 m</td>
</tr>
<tr>
<td>$f$</td>
<td>100MHz</td>
</tr>
<tr>
<td>$P_0$</td>
<td>800W</td>
</tr>
<tr>
<td>$Q_1$</td>
<td>0.2 liter/min</td>
</tr>
<tr>
<td>$Q_2$</td>
<td>0.6 liter/min</td>
</tr>
<tr>
<td>$Q_3$</td>
<td>12 liter/min</td>
</tr>
<tr>
<td>$p$</td>
<td>1 atm.</td>
</tr>
</tbody>
</table>

Table 8.8. Torch specification (cf. Fig. 8.4) of the 100MHz ICP. The number of equidistant windings is 2 [5].

the oscillator frequency. Since the skin depth $\delta \sim \omega^{-1/2}$ is of the order $\sqrt{2/(\mu_0 \times 700 \times 2\pi \times 10^8)} = 0.002$m we get for the ratio $\Psi = R_w/\delta = 4.5$. From Fig. 6.3 it can be seen that the magnetic field is much more shielded in comparison with the 3MHz ICP. As a result of increasing frequency it is to be
expected the Ohmic dissipation will take place in a decreasing plasma volume moving to the outer plasma regions near the torch wall [14].

Apart from the difference in frequency the mean power density in the volume determined by the height of the coil and the torch radius, \( P_0/V = P_0/[\pi R^2(L_c - l_1)] \) is also different. Using the torch specifications given in Tables 8.6 and 8.8 we find for the 3 MHz ICP \( P_0/V = 1.3 \times 10^8 \text{ W/m}^3 \), while for the 100 MHz ICP \( P_0/V = 4.0 \times 10^8 \text{ W/m}^3 \). Due to the relatively small as depicted in Fig. 8.3 and the higher power density larger gradients in the temperature and electron density are to be expected than in the 3 MHz ICP.

### 8.5.1 Discussion

In order to obtain a converged solution it was needed to adjust the set of under-relaxation factors in Eq. (7.38). Especially the under-relaxation factors dealing with \( n_e, T_e, T_h \) and the vector potential were given a lower value compared with the set of relaxation factors for the 3 MHz ICP. Having found a suitable set of relaxation factors it was observed that the rate of convergence was slower than for the other 3 MHz case. In Fig. 8.15 a typical rate of convergence for the 100 MHz case is shown in which the ionization/recombination data of DESAI and CORCORAN were used [15]. After 25000 iterations the relative residue for the pressure correction quantity is \( \sim 10^{-3} \) whereas \( \sim 10^{-8} \) was found for the 3 MHz case (Fig. 8.6). This is an indication that transport in the 100 MHz ICP is of much
more importance than in the 3MHz case.

Calculations without swirl

No grid transformation: Run 5. Calculations were done with the source terms specified by run DC-B in the previous section. The motivation for the use of the use of the data of DESAI and CORCORAN [15] for the ionization/recombination coefficients is that their expression is more suitable from the numerical point of view since the recombination coefficient has a weak functional dependence on $T_e$ at constant $n_e$ (cf. Fig. 6.2). In the previous section it was shown that the creation of electrons described by $S_{DC}^{CR}$ is stabilized by $n_e$, whereas $SCR$ of Ref. [18] results in an avalanche of electrons in the active.

In Fig. 8.16 the electron temperature and electron density profiles are shown. Both $T_e$ and $n_e$ show the characteristic feature of off-axis maxima in the ICP. In Table 8.9 the maximum values for $T_e$ and $n_e$ in the active zone and on the axis are shown (run 5). In order to compare the computational results with experimental data radial profiles at $z=0.038m$ of $T_e$, $T_h$ and $n_e$ are shown in Fig. 8.17. The following observations concerning $T_e$, $T_h$ and $n_e$ can be made:

- Reasonable agreement for the maximum value of the electron density in the active zone with the experimental data is found.
- Radial positions of the maxima are too close to the wall. The calculation shows maxima at $r_{\text{max}} \sim 0.0075m$ while according to experimental data of NOWAK et al. $r_{\text{max}} \sim 0.005 - 0.006m$.
- The global electron temperature profile is too low.
- The difference between $T_e$ and $T_h$ in the active zone is more than a factor 2 whereas the measurements of FEY et al. show a difference of a factor 1.35 [27].
- Unrealistic low values are found for the maximum axis values $T_{e(\text{max})}$ and $n_{e(\text{max})}$. The differences between $n_{e(\text{max})}$ and $n_e(\text{max})$ are two orders of magnitude while measurements reveal differences of a factor 2 only. The $T_{e(\text{max})}$ and $T_e(\text{max})$ differ more than a factor 2 while the difference in the experimental data is smaller than a factor 1.3.

We first investigate the location of the radial profile maxima. In Ref. [25] the radial gradient length for the electron density is estimated to be $4 \times 10^{-4}m$. The radial discretization length $\Delta r$ is $9 \times 10^{-3}/40 = 2.25 \times 10^{-4}m$ so that only 2 gridpoints are available in the active zone to describe the radial ambipolar diffusion. For this reason an algebraic coordinate transformation is carried out to increase the number of grid points near the torch wall and in the coil region. The transformation functions being used are Eq. (8.3) with $N = 6$ in the radial
Figure 8.16. Contour plots of electron temperature (a) and density (b) for run 5. The electron density contours are given in $10^{20} \text{m}^{-3}$. The contour steps are: $\Delta T_e = 500 \text{K}$ and $\Delta n_e = 2 \times 10^{20} \text{m}^{-3}$.

<table>
<thead>
<tr>
<th>run</th>
<th>$T_e(\text{max}) \text{K}$</th>
<th>$T_e^{\alpha_x}(\text{max}) \text{K}$</th>
<th>$n_e(\text{max}) \text{m}^{-3}$</th>
<th>$n_e^{\alpha_x}(\text{max}) \text{m}^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>7720</td>
<td>3330</td>
<td>$1.0 \times 10^{21}$</td>
<td>$1.4 \times 10^{19}$</td>
</tr>
<tr>
<td>6</td>
<td>7880</td>
<td>2290</td>
<td>$1.3 \times 10^{21}$</td>
<td>$7.2 \times 10^{18}$</td>
</tr>
<tr>
<td>7</td>
<td>8240</td>
<td>6080</td>
<td>$1.6 \times 10^{21}$</td>
<td>$7.1 \times 10^{19}$</td>
</tr>
</tbody>
</table>

Table 8.9. Maximum values in the active zone and on the axis for $T_e$ and $n_e$. Run 5: no algebraic transformation, run 6: with algebraic transformation in axial and radial direction, run 7: with swirl and algebraic transformation.

direction and

$$\frac{z}{L_T} = \frac{b}{a} + \frac{1}{a} \sinh \left[ \xi_1 (\sinh^{-1} (a - b) + \sinh^{-1} (b)) - \sinh^{-1} (b) \right]$$  \hspace{1cm} (8.12)

in the axial direction and $\xi_1$ is a normalized coordinate comparable with $\chi$ in Fig. 7.3.
Results

Figure 8.17. Radial distribution of $T_e$, $T_h$ (left) and $n_e$ (right) for $z=0.038m$. Full line: run 5, (---..-..): run 6

**With grid transformation: Run 6.** In Fig. 8.18 the resulting computational grid is shown in which $a = 6$ and $b = 1$. With the suggested transformation $\Delta r = 2 \times 10^{-4}m$ at $r = 0.006m$ and $\Delta r = 1.3 \times 10^{-4}m$ at $r = 7.5 \times 10^{-3}m$. Using the same source terms as in run 5 the results for $T_e$ and $n_e$ are shown in

Figure 8.18. Transformed grid

Fig. 8.19. The radial profiles of $T_e$, $T_h$ and $n_e$ are also include in Fig. 8.17 and the profile and axis-value maxima of $T_e$ and $n_e$ are shown in Table 8.9 as well. Comparing Fig. 8.16(a) and 8.19(a) it can be observed that the region in which the temperature is in excess of 3000K for run 5 (without transformation) is larger than for run 6 (with transformation). The effect of the axial transformation is that the contour lines between the windings of the coil are smoother, indicating that the discretization length $\Delta z$ before transformation is in fact too large in the
active zone. The effect of the radial transformation on the $T_e$, $T_h$ and $n_e$ profiles is clearly demonstrated in Fig. 8.17. The positions of the maxima of the radial $T_e$ and $n_e$ profiles at $z=0.038$ m are shifted towards the center and are $\sim 0.006$ m, which is in good agreement with the experimental observations [25].

Still the problems of too low axial values for the temperatures and electron density and the too large difference between $T_e$ and $T_h$ remain. In searching an explanation for this difference in $T_e$ and $T_h$ we consider the energy equations for the electrons and heavy particles. Using a global analysis of the electron energy equation FEY [5] showed that for $n_e > 10^{21} \text{m}^{-3}$ the elastic collisions between the electron and the heavy particles form the dominant process to transfer the electromagnetic energy via the electron to the heavy particles when $7000 < T_e < 10500$. Due to the relatively high $n_e$ in the active zone the collisional energy transfer between electrons and heavy particles will attempt to equilibrate $T_e$ and $T_h$. Since the energy dissipation takes places in a region near the torch wall and the heavy particle thermal conductivity is an increasing function of $T_h$ the radial
Results

Conductive energy transport will be considerable. To be more specific $\kappa_h$ scales with $\mu c_v$ [29] where $\mu$ is the viscosity which is proportional to $T_h^{1/2}$ for rigid sphere particles, and $c_v$ is the specific heat. Experimental values for the argon thermal conductivity reveal a higher temperature exponent due to the interaction between the heavy particles [11, 30]. If there is no sufficient cooling near the torch wall $T_h$ will be too high. As a result $\kappa_h$ will increase so that heat in the active zone can leak to the torch wall effectively rather than to the inner plasma regions (cf. Fig. 8.20(a)). The consequences will be that

1. $T_h$ is too low in the inner region, →
2. electrons are cooled by collisions with heavy particles $\rightarrow T_e$ decreases, →
3. the ionization will decrease, →
4. less electrons will be created, →
5. the coupling between the electrons and heavy particles reduces, so that differences between $T_e$ and $T_h$ will occur in the inner region.

From the experimental set-up we know that a swirl is needed for a stable operation. The swirl acts as a sheath layer between the torch wall and the active zone. Its purpose is to cool the plasma in the outer parts of the active zone thus protecting the torch wall from an excessive amount of heat (cf. Fig. 8.20(b)). Also the electrons are cooled and since the electric conductivity is mainly an increasing function of temperature the Ohmic dissipation $\sigma \omega^2 |A_\phi|^2$ in the outer part of the active zone will decrease. As a result the active zone will be displaced inwards.

Figure 8.20. Most heat generated in the active zone will be conducted to the torch wall in the absence of cooling swirl (a), while in the presence of such a swirl a larger part of the heat will be conducted to inner region of the plasma (b). The direction of the swirl is directed perpendicular to the plane of the paper.
The foregoing discussion demonstrates that for an adequate description of the 100 MHz ICP the swirl component of the flow field must be included in the model.

**Calculation With swirl: Run 7**

The same computational grid as in run 6 was used to perform a calculation with a swirl component. For this purpose the module for $v_3$ component was included. The boundary conditions for $v_3$ are given in Table 8.4. The values for $(v_3)_i$ are given in Table 8.10. For reason of convergency the values for $Q_3$ and $(v_3)_3$ are taken to be lower than the experimental values [27]. Still the convergency was not as good as for run 5 and 6. In Fig. 8.21 radial profiles for $T_e$ and $n_e$ are shown at $z=0.038$m for different number of iterations. From Fig. 8.21 it can be seen that first the energy dissipation takes place at $r/R_w \sim 0.9$ and that during the iteration process, inward diffusion and conduction increases $n_e$ and $T_e$ in the inner region, respectively. The differences for the $T_e$ profiles at 16000 and 20000 iterations are relatively small for all radial positions. For $n_e$ differences up to 13\% (at $r/R_w = 0.8$) are noticed in the active zone for 16000 and 20000 iterations. The differences are due to the fact that the total power input is underrelaxed as well. Since the total calculated power incoupling was found to be 795 while 800 W was the initial power input, it is expected that no significant changes will occur for higher number of iterations.

<table>
<thead>
<tr>
<th>$Q_i$</th>
<th>l/min</th>
<th>$(v_3)_i$</th>
<th>m/sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q_1$</td>
<td>0.2</td>
<td>$(v_3)_1$</td>
<td>0</td>
</tr>
<tr>
<td>$Q_2$</td>
<td>0.6</td>
<td>$(v_3)_2$</td>
<td>0</td>
</tr>
<tr>
<td>$Q_3$</td>
<td>6.0</td>
<td>$(v_3)_3$</td>
<td>5</td>
</tr>
</tbody>
</table>

Table 8.10. Specification of $Q_i$ and $(v_3)_i$ at the inlet.

Maximum values for $T_e$ and $n_e$ in the active zone and on the axis are shown in Table 8.9. From this and from a comparison between Figs. 8.17 and 8.21 it can be deduced that the presence of the swirl has a drastic influence on the $T_e$ and $n_e$ profiles. The value for $T_e^{ax}(max)$ is much higher than for run 5 (a factor 1.8) and run 6 (a factor 2.7) and the differences for $n_e^{ax}(max)$ are a factor 5.1 and 9.9 for run 5 and 6, respectively. However, the radial profiles for $T_e$ and $n_e$ are still too low compared with the experimental values of NOWAK et al. [25] (cf. Fig. 8.14). Since a calculation with $S_{CR}$ of Ref. [18] has resulted in a higher $T_e$ and $n_e$ distribution it is to be expected that this $S_{CR}$ will lead to increased $T_e$ and $n_e$ values for the 100 MHz ICP. At the present time it is not yet possible to obtain
Figure 8.21. Radial distribution of $T_e$ (a) and $n_e$ (b) for $z=0.038$ m at different number of iterations: (- - -) 4000, (---) 8000, (- - -) 12000, (- -) 16000, (---) 20000, for a computation with swirl.

a converged solution using the $S_{CR}$ of Ref. [18].
8.6 General conclusions

1. The fact that the energy incoupling region is much smaller and the power density consequently much higher makes the modelling of an ICP with high frequency (100 MHz) much more difficult than in the case of a low frequency (3 MHz). Transport processes and coefficients have to be known with high precision.

2. The presence of large gradients implies that the computational grid of a high-frequency ICP must be sufficiently fine. To avoid long computer time grid generation is needed.

3. The swirl, which is needed to avoid plasma wall interaction of a high frequency ICP, makes the convergence very difficult.

4. The ionization rate coefficient obtained by taking the detailed balance of the empirical found recombination coefficient of DESAI and CORCORAN is highly questionable from the physical point of view. A program using this rate coefficient generates lower $n_e$ and $T_e$ values. However, the negative power of $n_e$ (cf. Eq. 8.11) provides a computational stability. Electrons created by high $T_e$ values will temper further creation due to this negative power.

5. Including an ionization rate coefficient based on a collisional-radiative model [18] gives higher $n_e$ and $T_e$ values which are more in agreement with experimental data [31, 32, 33]. However, the avalanche nature of ionization, i.e. the fact that created electrons will improve energy incoupling resulting in additional creation of electrons, implies a computational stability problem. This can only be solved by an appropriate treatment of the transport processes.

6. The question might raise in how far the plasma is stable in reality.

7. The radiative loss term of EVANS and TANKIN [16] can not be realistic since it avoids the radiative loss below 9500 K. The radiative loss term of WILBERS et al. [17] creates too much losses for low temperatures.

8. Modelling of the 3 MHz ICP with own ionization and recombination rate coefficients gives $n_e$ and $T_e$ which significantly differ from those obtained with the rate coefficients of DESAI and CORCORAN [15]. Especially the measurements of $n_e$-values might provide a good method to determine the validity of the different rate coefficients.

9. This study has shown that for low frequency ICP’s in which transport processes are less important the influence of source and loss terms is significant and that the LTE approach is not justified. For high-frequency ICP’s the correct treatment of these source terms is much more essential. However, the treatment of transport processes is very critical.
References

Summary

This thesis describes the construction of a two dimensional time independent plasma simulation model. The model, describing the plasma and its boundaries as a whole by solving the conservation laws of mass, momentum and energy iteratively, can be used for the description of thermal plasmas. These are plasmas characterized by relative high energy and particle densities and temperatures in the range of 6000 — 25000 K.

The physics at the basis of the construction of the model ranges from the level of the microscopic activity of atomic collisional and radiative processes to the level of macroscopic fluid dynamics. The departure from equilibrium of elementary collision and radiative processes creates sources and sinks on the level of the fluid processes. In this way e.g. the study of ionization and recombination processes is of importance since these determine the electron particle and energy balance, whereas the escape of radiation implies an important energy loss term.

The validation of the model is effectuated by computations on well known inductively coupled plasmas (ICP), i.e. plasmas created in and sustained by the alternating magnetic field of a coil. Two configurations are dealt with:

1) a relatively large ICP created at low frequency (3 MHz) and
2) a relatively small ICP created at high frequency (100 MHz).

The description of ICP 1) turns out to be relatively simple. The model gives results which are comparable to those obtained by other groups. However, it is found that the plasma strongly depends on the choice of the source terms, i.e. the rate coefficients of ionization and recombination, and the radiative loss term. If the results of our collisional radiative model are implemented significantly larger values of the electron densities and temperatures are obtained.

The description of the small ICP (case 2) is much more complicated. Due to the high energy densities, steep gradients and consequently strong transport processes the exact knowledge of the transport processes is of crucial importance whereas the solution is numerically much less stable.

To describe the steep gradients in a proper way we constructed a grid generation module by which a fine grid can be created on those locations where the gradients are expected to be large. Calculations with this module give significantly other results than those without this module.
Samenvatting

Dit proefschrift beschrijft de constructie van een tweedimensionaal tijdsonafhankelijk plasma simulatie model. Het model, dat het plasma als geheel beschrijft door de behoudswetten van massa, impuls en energie in een iteratief samenspel op te lossen, kan worden gebruikt voor de beschrijving van thermische plasma's. Deze plasma's worden gekarakteriseerd door relatief hoge energiedichtheid en deeltjesconcentraties en temperaturen in het gebied van 6000 – 25000 K.

De fysica die ten grondslag ligt aan de constructie van dit model strekt zich uit van het niveau van de microscopische activiteit van atomaire processen tot dat van macroscopische van stromingsprocessen. De onderling niet in evenwicht zijnde elementaire botsings- en stralingsprocessen creëren daarmee bronnen en putten op het niveau van de stromingsprocessen. Zo is met name de studie van de ionisatie- en recombinatieprocessen van belang omdat zij in hoge mate de electronen deeltjes- en energiebalans bepalen, terwijl het ontsnappen van straling een belangrijke energieverliesterm impliceert.

Het model wordt aan de realiteit getoetst door berekeningen uit te voeren aan bekende inductief gekoppelde plasma's (ICP), plasma's gecreëerd in en in stand door het wisselend magnetisch veld in een spoel. Daarbij is gekozen voor twee configuraties:

1) een relatief groot ICP gecreëerd bij een relatief lage frequentie (3MHz) en
2) een relatief klein ICP gecreëerd bij een relatief hoge frequentie (100 MHz).

De beschrijving van ICP 1) blijkt relatief eenvoudig. Het model geeft vergelijkbare resultaten als die verkregen door andere groepen. Wel blijkt dat het plasma sterk afhankelijk is van de keuze van de ionisatie- en recombinatie-processen en de stralingsverliestermen. Indien voor deze bron- en verliestermen waarden worden geïmplementeerd die verkregen zijn door onze studie naar elementaire processen dan blijkt dit te leiden tot aanzienlijk hogere waarden voor de electronendichtheid en temperatuur.

De beschrijving van een hoogfrequent ICP is veel minder eenvoudig. Door de hoge energiedichtheden, steile gradiënten en daarmee gepaard gaande sterke transportprocessen is een grondige kennis van transportcoëfficiënten van cruciaal belang terwijl de oplossing numeriek weinig stabil is. Om de steile gradiënten goed te verdisconteren is een roostergeneratier module gecreëerd die een fijnmazig netwerk kan genereren op die plaatsen waar men steile gradiënten verwacht. Berekeningen met deze module geven significant andere resultaten dan berekeningen zonder de module.
Dankwoord

Vooraleer een punt te zetten achter vier jaar en vier maanden onderzoek, wil ik iedereen bedanken die heeft bijgedragen tot de instandkoming van dit proefschrift. Het zou onterecht zijn een aantal van hen niet met naam te noemen.

Mijn eerste promotor Daan Schram wil ik bedanken voor de mogelijkheid die hij me heeft gegeven om in een overwegend experimentele groep, theoretisch en voornamelijk modelleringswerk te verrichten. Zijn inzichten en discussies met hem hebben een zeer inspirerende invloed gehad op mij.

Veel dank gaat uit naar Joost van der Mull en die zowat vier jaar en half geleden een onbekende Belg binnenhaalde. De samenwerking met Joost en Frank Fey, mijn kamergenoot, was dan ook het meest intens, waardoor hun bijdrage tot dit werk niet weg te cijferen is!

Gert-Jan van Heijst, die optrad als tweede promotor en leescommissielid Frits de Hoog, die in korte tijd dit proefschrift hebben gelezen.

Van leescommissielid Jaroslav Vlček heb ik iets geleerd over het oplossen van de Boltzmann transportvergelijking zelfconsistent met botsings-stralingsmodellen. Hij gaf me tevens de mogelijkheid om gedurende een maand kennis te maken met het leven achter het gesneuvelde IJzeren Gordijn.

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Wetenschappelijk medewerker in werkgroep
Evenwicht en Transport in Plasma's
STELLINGEN

behorend bij het proefschrift

MODELLING OF THERMAL ARGON PLASMAS

door

D.A. Benoy

16 april 1993
Het gebruik van de vergelijking voor de deeltjesbalans in de macroscopische beschrijving van plasma's impliceert de quasi-stationaire oplossingsmethode voor botsings-stralingsmodellen.

*Dit proefschrift, hoofdstuk 2*

De atomaire verdelingsfunctie gegeven door partieel lokaal Saha-evenwicht en de veronderstelling van een quasi-stationaire oplossing voor botsingsstralingsmodellen, leiden niet tot een eenduidige recombinatiecoëfficiënt.

*Dit proefschrift, hoofdstuk 3*

Met de meting van de elektronendichtheid in atomaire plasma's kan onderzocht worden of de ionisatie- en recombinatiecoëfficiënt elektron-kinetisch bepaald zijn.

*Dit proefschrift, hoofdstuk 8*

De methode van de normalisering van de verdelingsfunctie voor de elektronenenergie (VFEE) voor de berekening van het elektrisch veld kan niet gebruikt worden als de VFEE naar de Maxwell-verdeling tendeert.

*R.M.M. Smits and M. Prins, Physica, 96C, 243, (1979).*


De uitdrukkingen voor de botsingsdoorsneden die VRIENS en SMEETS hebben opgesteld voor de door elektronen geïnduceerde overgangen tussen waterstof-niveaus zijn in principe ook te gebruiken voor overgangen tussen Rydbergachtige toestanden. Men kan dan echter negatieve waarden voor de overgangswaarschijnlijkheid vinden.

Vanwege de hoge deeltjes- en energiedichtheden is het open, hoogfrequente, inductief gekoppelde plasma uitermate moeilijk te modelleren maar is het zeer geschikt voor spectroscopische doeleinden.

Het feit dat meer dan 99% van de zichtbare materie in het heelal uit plasma bestaat, vindt op geen enkele wijze zijn weerslag in het aantal Nobelprijzen voor natuurkunde dat toegekend is voor plasmafysisch onderzoek.

Het achtereenvolgens gebruiken van meerdere computers voor het schrijven van een publikatie verhoogt de kans op schrijffouten in de uiteindelijke versie.

Ideale mobiliteit van kapitaal tussen landen leidt tot enorme kapitaalstromen bij het geringste verschil in rentevoet.

Vanuit het standpunt van de vooruitgang van de Europese ruimtevaart zou het beter zijn de militaire en civiele ruimtevaarttechnologie van de voormalige Sovjet Unie over te nemen in plaats van deze zelf te ontwikkelen.