LOW FREQUENCY MAGNETIC LOSSES IN IRON-RICH SINGLE-CRYSTAL FERRITES

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Abstract—The initial magnetic permeability spectra of the single-crystal manganese-zinc ferrite and two titanomagnetites were measured at audio frequencies from room temperature down to 10 K. The observed maxima in the magnetic loss factor can be ascribed to the passing of the anisotropy constant through zero and to the relaxation processes connected with the presence of charge carriers in the octahedral sites of the spinel lattice.

INTRODUCTION

Manganese-zinc ferrites range among the important materials used in video tape heads, high permeability ferrites, etc. The understanding of the processes occurring during magnetization is very important for tailoring the material for desired purposes. In the present paper low frequency (20 Hz to 20 kHz) and temperature (10 to 300 K) dependences of the initial permeability of three single-crystal cubic ferrites are studied in order to clarify the processes encountered in these materials.

SAMPLES AND MEASUREMENT

The first sample investigated was Bridgman type single-crystal manganese-zinc ferrite $\text{Mn}_{0.51}\text{Zn}_{0.36}\text{Fe}_{2.13}\text{O}_4$ (henceafter denoted as MnZn), widely used for video tape heads; the crystal was made by the Victor company of Japan. The other two samples were single-crystal titanomagnetites of the compositions: $\text{Ti}_{0.1}\text{Fe}_{2.9}\text{O}_4$ (Ti1) and $\text{Ti}_{0.2}\text{Fe}_{2.8}\text{O}_4$ (Ti2). Both titanomagnetites were prepared by the arc-image floating-zone technique from presintered rods as described earlier [1]. From the crystals small hollow cylinders (5 mm outer and 2 mm inner diameter, 3 mm in height) were prepared by grinding and etching and provided with the primary and the secondary toroidal coils to enable measurements of real (μ') and imaginary (μ'') parts of the initial permeability $\mu = \mu' - j\mu''$. More detailed description of the method of measurement is given elsewhere [2].

RESULTS

Manganese-zinc ferrite

Frequency and temperature dependences of both components of the initial permeability of the manganese-zinc ferrite are plotted in Fig. 1.

Fig. 1 Temperature and frequency dependences of the real (a) and the imaginary (b) parts of the initial permeability MnZn ferrite measured with magnetizing field amplitude of 0.8 A/m (≈ 10 mOe).

It was found earlier that the measured values of the permeability depend on the amplitude of the alternating magnetizing field. Such an effect can be seen in Fig. 2 where the permeability is measured at higher magnetizing field. It is seen that not only the magnitude but also the shape of the temperature dependences of μ are significantly influenced by the amplitude of the magnetizing field.

Fig. 2 The same as in Fig. 1 at magnetizing field amplitude of 4 A/m (≈ 50 mOe).

Corresponding magnetic loss factors, $\tan \delta = \mu''/\mu'$, are plotted in Fig. 3. Although the overall temperature dependences possess some common features great differences could be seen.

First it is found that in all the curves a frequency independent maximum occurs at about 210 K. As it was pointed out earlier [3, 4], such "secondary" maxima occur often in MnZn ferrites and are connected with the vanishing of the magnetocrystalline anisotropy. It is interesting to note that on increasing the amplitude of magnetizing field from 0.8 to 4 A/m this frequency independent maximum almost vanishes at low frequencies.

The other maxima seen in Fig. 3 depend on frequency which is characteristic for thermally activated relaxation processes. The relaxation seen at about 100 K in Fig. 3a seems to merge with the maximum at 210 K in Fig. 3b while an indication of a low-temperature relaxation seen in Fig. 3a at high frequencies and low temperatures becomes well developed near 20 K in Fig. 3b.

Titanomagnetites

In Fig. 4 the initial permeability dependences for Ti1 sample are represented. Here sharp maxima in μ' and μ'' in the region

Fig. 4 Temperature and frequency dependences of the real (a) and imaginary (b) parts of the initial permeability of $\text{Ti}_{0.1}\text{Fe}_{2.9}\text{O}_4$ sample measured at magnetizing field of 4 A/m.

50–100 K, both dependent on frequency, could be seen. They resemble relaxation maxima in Mn- or Zn-ferrites with compositions close to magnetite [5]. Measurements of μ for the Ti2 sample are represented in Fig. 5. Values of μ are two orders of magnitude smaller than those for Ti1. It is also seen that the relaxation is composed of several components. Let us note that no relaxations were found in titanomagnetites with higher Ti contents.

Fig. 5 The same as in Fig. 4 for $\text{Ti}_{0.2}\text{Fe}_{2.8}\text{O}_4$ sample.

Fig. 3 Temperature and frequency dependences of magnetic loss factor $\tan \delta$ of MnZn ferrite at magnetizing fields of 0.8 (a) and 4 A/m (b).

Fig. 6 Temperature and frequency dependences of magnetic loss factor $\tan \delta$ for $\text{Ti}_{0.1}\text{Fe}_{2.9}\text{O}_4$ (a) and $\text{Ti}_{0.2}\text{Fe}_{2.8}\text{O}_4$ (b).

Magnetic loss factors, $\tan\delta$, for titanomagnetite samples are shown in Fig. 6. At least two relaxation processes can be recognized. The results for both titanomagnetites were not significantly influenced by the amplitude of the magnetizing field and, consequently, only dependences for magnetizing field of 4 A/m are shown.

DISCUSSION

The origin of various relaxation mechanisms in iron-rich ferrites is ascribed to the hopping of electrons among Fe ions in octahedral positions ($\text{Fe}^{2+} \longleftrightarrow \text{Fe}^{3+}$). It is the same mechanism which is responsible for the electrical conductivity in these materials. As a proof of the same origin of the processes the proximity of the activation energies of both effects [5] is considered. The activation energy of the relaxation process can be determined from the Arrhenius relation expressing the temperature dependence of the maxima in the $\tan\delta$ vs. T curves for particular frequencies $f=1/2\pi\tau$

$$\tau = \tau_0 \exp(\epsilon/kT) \quad (1)$$

where k is the Boltzmann constant, ϵ is the activation energy and τ represents the time constant for $T \rightarrow \infty$. The activation energies for all observed processes are summarized in Table 1.

TABLE 1
Activation energies of the relaxation processes observed in MnZn, Ti1 and Ti2 samples

Sample	$\epsilon_1(\text{eV})$	$\tau_{01}(\text{sec})$	$\epsilon_2(\text{eV})$	$\tau_{02}(\text{sec})$
MnZn	0.083	1.39×10^{-9}		
Ti1	0.084	5.41×10^{-13}	0.146	1.52×10^{-14}
Ti2	0.098	1.37×10^{-12}	0.108	7.58×10^{-11}

Contrary to MnZn ferrite where only one relaxation is observed in the 50–100 K region (see Fig. 3a) the magnetic losses of titanomagnetites display besides the main peak an additional smaller relaxation (see Fig. 6). Using (1) the parameters of the main and the subsidiary relaxations were determined and are listed in Table 1.

Based on a similar behaviour of the frequency dependent maxima of the MnZn ferrite sample and of both Ti-samples and on proximity of the activation energies to those determined from the electrical conductivity [1] it is concluded that the hopping of electrons among Fe^{2+} and Fe^{3+} in octahedral positions is responsible for the relaxation mechanism with the activation energy ϵ_1 . The two processes in titanomagnetites with close activation energies can be understood in view of the different nearest neighbours caused by the presence of Ti^{4+} ions [5].

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