Russian Physics Journal, Vol. 49, No. 9, 2006

ANALYSIS OF THE MICROWAVE MAGNETIC PERMEABILITY SPECTRA OF FERRITES WITH HEXAGONAL STRUCTURE

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This paper presents the results of theoretical and experimental investigations of the microwave magnetic permeability spectra of polycrystalline ferrites with hexagonal crystal structure. It is shown that the observed nonmonotone frequency dependence of the permeability is due to the features of the natural ferromagnetic resonance in the presence of a domain structure. The calculations of the contribution of the rotation of the magnetization vector to the permeability performed in the approximation of independent grains adequately describe experimental data.

INTRODUCTION

To solve many practical problems it is necessary to create materials with prescribed electromagnetic characteristics. For the magnetic materials used in microwave devices, these characteristics are the spectra of their magnetic permeability $\mu(\omega) = \mu'(\omega) - i\mu''(\omega)$ and dielectric permittivity $\varepsilon(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega)$ (MP and DP, respectively). The dielectric permittivity of the majority of ferrites is related to the ionic mechanism of polarization and weakly varies in the microwave range. The frequency dependences of MP are more complicated. Several regions of anomalous dispersion are generally detected that are related to the volumetric resonance observed in large specimens; to the oscillations of domain boundaries (DB's) which may be both resonance and relaxation in character [1]; to the natural ferromagnetic resonance (NFMR) [2, 3] caused by the rotation of the magnetization vector in the effective internal field of magnetic anisotropy, and to the exchange resonance in the internal exchange field in the presence of two or more magnetic sublattices [4]. It is well-known that the NFMR region in magnetics with cubic structure can bifurcate due to the magnetic interaction of magnetization oscillations in neighboring domains which are differently oriented in relation to the variable magnetic field [5]. Similar observations have been made for ferrites with hexagonal structure [6]. Under certain conditions all mentioned regions of anomalous dispersion of MP can lie in any band of the microwave frequency range, creating additional maxima and steps in the frequency dependences $\mu'(\omega)$ and $\mu''(\omega)$ [7–9].

The main source of information about MP spectra is experiment since theoretical ideas which could be used to calculate the frequency dependences of MP by known physical parameters, for example, by static parameters such as saturation magnetization, fields of magnetic anisotropy, etc., have not yet been developed. The experimental error hampers the interpretation of the obtained data and selection of anomalous dispersion regions related to one or another physical mechanism. This leads to inaccuracy in the determination of resonance frequencies and complex values of MP and gives no way of precisely predicting the chemical composition of a material, its magnetocrystal structure and domain structure, hampering the production of materials with prescribed properties.

This work considers the method of separation of dispersion regions by the data of an actual experiment processed by the technique reported elsewhere [10] with the use of the Cramers–Kronig relations.

EXPERIMENTAL

The region of the complicated behavior of the MP spectra of hexaferrites has the features of a significant frequency range and a large dynamic range of measurands, leading to the problem of examination of materials with great losses

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($\tan\delta_m > 0.5$); therefore, it is insufficient to use only one measuring procedure. In this connection, for the low-frequency range (0.1–1 GHz) the coaxial waveguide method was used with a specimen of the test material shaped as a thin washer of outer diameter 16 mm, inner diameter 6.95 mm, and thickness 1 mm. For the high-frequency range (0.9–17 GHz) rectangular transmission resonators with type H_{10n} oscillations were used in which a specimen shaped as a thin cylinder of diameter 1.6 mm and length equal to that of the wide wall of the resonator was placed [11]. The wide bandwidth of the resonance measurements was attained with a set of resonators by using the frequency variation method with multimoding. For the intermediate range, where methods with distributed and lumped parameters are equally applicable (0.2–1 GHz), measurements were additionally performed on a nonregular microstrip resonator [12] made of high-dielectric-permittivity ceramics with a specimen shaped as a thin plate having the dimensions of the air gap between the strip lines, 10×10×1 mm. The static initial permeability was determined by the inductance of a toroid measured at a frequency of 1 kHz. The measurement error estimated for all methods used was not above 2–5% for the real component and 5–15% for the imaginary component of the MP.

The subject of investigation was $Co_{1.2}Zn_{0.8}W$ hexagonal ferrite synthesized by standard ceramic technology, whose ferromagnetic resonance range falls in the chosen frequency band. At room temperature this material is characterized by magnetic anisotropy of "easy-plane" type with the following principal magnetic parameters: anisotropy field $H_\Theta = -4$ kOe and saturation magnetization $4\pi M_0 = 3.6$ kG. The solid-material hydrostatic density ρ is 3.77 g·cm^{-3} and porosity makes 29%. The single-phase nature of the material is confirmed by x-ray structure analysis, which has shown that a test specimen contained 95% of the master W phase [13, 14].

SEPARATION OF DISPERSION RANGES

At the borderline between the methods used some disagreement between the results was observed which could be caused by both the different approximations used for different measuring and the systematic errors inherent in these procedures. To coordinate the results, the Cramers–Kronig relations were used in the technique proposed by Polivanov [15], such that the frequency range is divided into bands for which piecewise linear approximation of one of the permeability components is possible, and the integrals for the second component are calculated analytically. It is well known that the wider the frequency range of the measurements, the more adequate information is provided by the Cramers–Kronig relations. The error builds up as the edges of the measurement range are approached. If we do not apply some artificial methods, we have either to resign ourselves with the losses of part of experimental data, or to complete the frequency dependences of the imaginary and real components outside the frequency range of the experimental research based on some physical suppositions. We used a similar procedure [10] to analyze the MP spectrum of Co_2Z hexaferrite with one dispersion range. The study performed has also shown that for the case where the MP spectra of the imaginary and real components are measured independently it is possibile to use the Cramers–Kronig relations to refine experimental data with the purpose to reduce the actual experimental error.

We use the dispersion relations to analyze and process the measurements of the MP spectrum of $Co_{2-x}Zn_xW$ ($x = 0.8$) polycrystalline hexaferrite, initially supposing the presence of two dispersion ranges in the spectrum: the low-frequency range associated with the processes of displacement of domain boundaries ($\mu_d(\omega)$) and the high-frequency range associated with the rotation of the magnetization vector in the effective field of magnetic anisotropy ($\mu_r(\omega)$). This supposition is most often used in analyzing experimental data [7–9]. For $Co_{1.2}Zn_{0.8}W$, these ranges are so close to each other that their overlapping is observed. In this case, a problem arises in determining exactly the NFMR frequency and the maximum value of μ''. To eliminate this problem, we propose a procedure of separation of the two dispersion ranges after preliminary processing of experimental data with the use of the Cramers–Kronig relations.

This procedure is illustrated by Fig. 1. In Fig. 1*a*, the dark symbols designate the measurements of the real part of the MP and the lines with open circles present a piecewise linear approximation of the spectrum. Assuming the absence of ranges of anomalous dispersion in the low-frequency part of the spectrum outside the measurement range ($f < 0.1$ GHz), we considered that $\mu'(\omega)$ retains its value, equal to the static permeability $\mu(0) = 5.55$, up to the test frequency range and tends to unity at frequencies above the test range. The value of $\mu'(\omega)$ at frequencies above 50 GHz and up to 500 GHz was taken equal to unity. This extension is necessary to prevent an undesirable effect of the jump in $\mu'(\omega)$ from unity to zero at the upper limit of the approximation range on the $\mu''(\omega)$ spectrum to be reconstructed. The results of the reconstruction of $\mu''(\omega)$

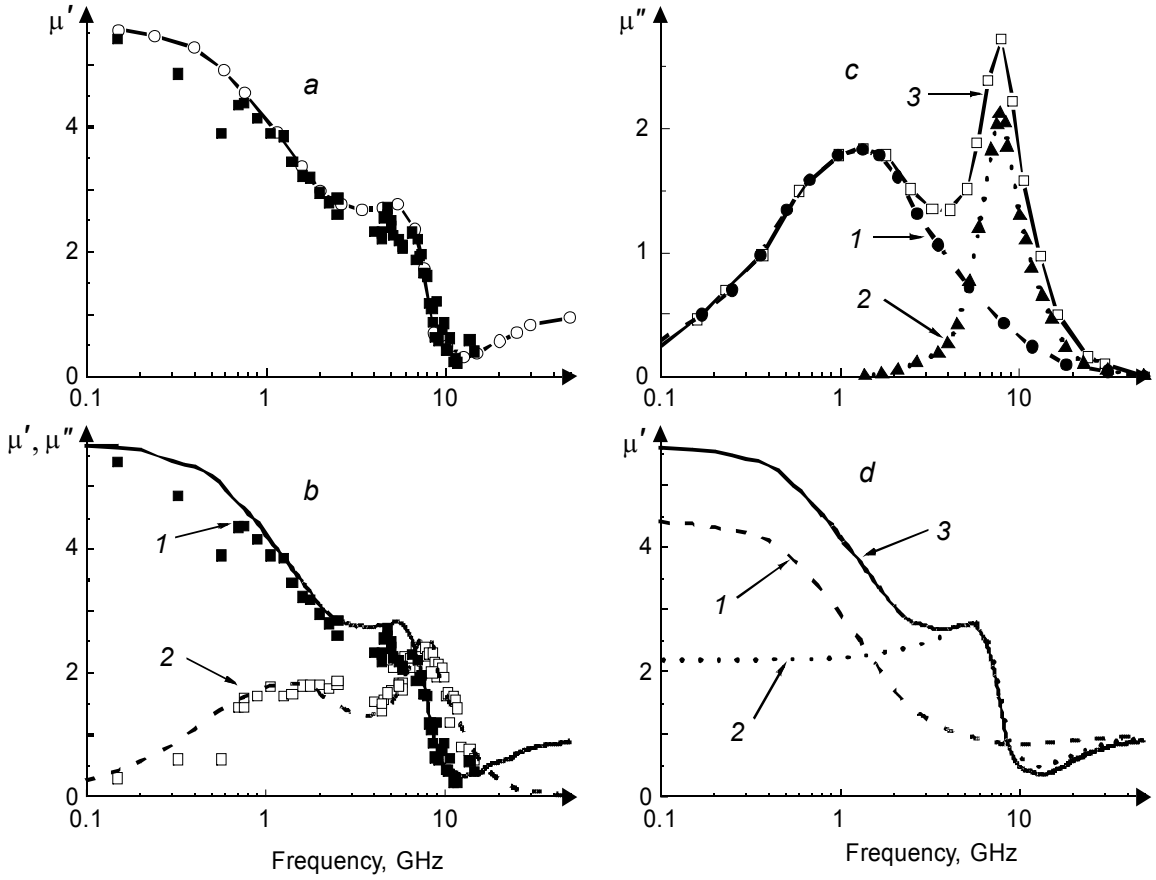

Fig. 1. Processing of the spectrum of $Co_{1.2}Zn_{0.8}W$ hexaferrite by the Cramers–Kronig relations: (*a*) measurements of μ′ and their piecewise linear approximation; (*b*) the real (curve *1*) and the imaginary part (curve *2*) of the permeability (dots: experiment; lines: calculation); (*c*) separation of dispersion ranges: $\mu_d''(\omega)$ (curve *1*), $\mu_r''(\omega)$ (curve *2*); the initial smoothed spectrum of the imaginary part of the permeability (curve *3*), and (*d*) calculation of the real parts of the permeabilities from their imaginary parts: $\mu_d'(\omega)$ (curve *1*), $\mu_r'(\omega)$ (curve *2*), and the total permeability (curve *3*).

by the real part of the permeability are given in Fig. 1. Note that the coordination of the measured spectra was performed both by the calculation of $\mu''(\omega)$ from $\mu'(\omega)$ and by the inverse re-calculation. The obtained smoothed frequency dependences of the complex MP are presented by lines in Fig. 1.

The separation of the contributions of the DB and NFMR displacements to the MP spectra was carried out as follows. According to the data reported in [1], the contribution of the processes of displacement to $\mu''(\omega)$ on the frequency logarithmic scale is described by a near-Gaussian symmetric curve. Therefore, the high-frequency flank in $\mu_d''(\omega)$ was completed symmetrically relative to the low-frequency maximum in the spectrum of the smoothed imaginary part of the MP. Then the spectrum $\mu_d''(\omega)$ was point-by-point calculated by the spectrum of full losses $\mu''(\omega)$ and thus the contribution of the processes of rotation of the magnetization vector $\mu_r''(\omega)$ was found. The results of this procedure are presented in Fig. 1*c*. The real parts of the permeabilities $\mu_d'(\omega)$ and $\mu_r'(\omega)$ were calculated with the use of the Cramers–Kronig relations by the known $\mu_d''(\omega)$ and $\mu_r''(\omega)$. The spectra obtained are given in Fig. 1*d*. Curve *3* represents the total spectrum $\mu'(\omega) = (\mu_d'(\omega) - 1) + (\mu_r'(\omega) - 1) + 1$. It practically coincides with the spectrum of the MP real part obtained as a result of smoothing that given in Fig. 1.

Thus, as a result of the analysis performed, smoothed experimental frequency dependences of the MP of $Co_{1.2}Zn_{0.8}W$ hexaferrite have been obtained which can be used for practical calculations of various microwave devices, in particular, of radioabsorbing materials and coatings, for a given frequency band with sufficient accuracy. Besides, the

application of dispersion relations has made it possible to separate the contributions of the processes of DB and NFMR displacement to the spectra, and this opens prospects for target control of the properties of hexaferrites in various bands of the microwave range. This can be attained either by modifying their microstructure, which primarily affects $\mu_d(\omega)$, or by varying the fields of magnetocrystalline anisotropy, which mainly determines the $\mu_r(\omega)$ spectrum [1].

ALLOWANCE FOR THE DOMAIN STRUCTURE ON MAGNETIC PERMEABILITY SPECTRA

It should be noted that careful comparison of the experimental MP spectra of the hexaferrite under investigation in the frequency range above 1 GHz with their description by two simple smooth curves reveals appreciable discrepancies. In the experimental data some peculiarities – additional local maxima and steps in the frequency dependences $\mu'(\omega)$ and $\mu''(\omega)$ – are seen which fall outside the limits of the experimental error. It is natural to suppose, that these peculiarities are due to the effect of the domain structure on the natural ferromagnetic resonance. The problem of the effect of the DS on the MP spectra of hexaferrites in the NFMR range was already considered theoretically [16].

The magnetic anisotropy fields of the hexaferrites are, as a rule, greater in magnitude than the saturation magnetization, and to analyze the behavior of these materials on the microwave scale, the model of independent grains is applicable. In this case, the starting point of calculations is deriving an expression for the MP of an individual monocrystalline grain. Let a grain have the shape of an ellipsoid of revolution with the domain structure and anisotropy of the easy magnetization plane (EMP) type. The axis of revolution of the ellipsoid coincides with the *c*-axis of the hexagonal crystal, and the DS is a system of plane-parallel domains normal to the hexagonal axis. The magnetic permeability of such a specimen, after averaging over the domain structure in the directions of the easy axes in the base plane can be written in the form of a diagonal tensor with components

$$\mu_{xx} = \mu_1, \quad \mu_{yy} = \mu_1, \quad \mu_{zz} = \mu_z , \tag{1}$$

where $\mu_{x(z)} = 1+\omega_M(\omega_1+i\alpha\omega)/[(\omega_1+i\alpha\omega)(\omega_2+i\alpha\omega)-\omega^2]$; $\mu_1 = (1+\mu_x)/2$; $\omega_M = \gamma 4\pi M_0$, ω is the circular frequency of the variable magnetic field, α is the damping constant in the equation of motion, γ is the gyromagnetic ratio, and M_0 is the saturation magnetization. Expressions for the frequencies ω_1 and ω_2 of the components μ_x and μ_z are given in Table 1, where the following designations are used: $\omega_\Theta = \gamma H_\Theta$, $\omega_\Phi = \gamma H_\Phi$. Here, $H_\Theta = 2(k_1+2k_2+3k_3-3k_4)/M_0$ and $H_\Phi = 36k_4/M_0$ are the fields of magnetic crystallographic anisotropy relative to the hexagonal axis and in the base plane, respectively; k_i is the anisotropy constant of the *i*th order in the expression for the energy of magnetic crystallographic anisotropy of the hexagonal crystal; N_t and N_l are the transverse and the longitudinal demagnetizing factor of a grain, respectively, and $2N_t + N_l = 1$. The resonance frequencies of the components $\mu_{x(z)}$ will be written in the form

$$\omega^2_{\text{res } x(z)} = \omega_1\omega_2 . \tag{2}$$

From formula (2) and Table 1 it can be seen that the resonance frequencies of the components μ_x and μ_z are different and they coincide only for a specimen shaped as a thin disk.

Similar calculations can be performed for a material whose anisotropy is of the easy magnetization axis (EMA) type. For a specimen shaped as an ellipsoid of revolution with the revolution axis coinciding with the hexagonal axis of the crystal and with a plane-parallel DS normal to the *y*-axis, the tensor averaged over the domain structure has a diagonal form with components

$$\mu_{xx} = \mu_x, \quad \mu_{yy} = \mu_y, \quad \mu_{zz} = 1. \tag{3}$$

The components $\mu_{x(y)}$ are determined by the same formula as $\mu_{x(z)}$, and the frequencies ω_1 and ω_2 are given in Table 2: $\omega_a = \gamma H_a$, where $H_a = 2k_1/M_0$. From Table 1 and Table 2 it can be seen that both the resonance frequencies and the permeability depend substantially on the demagnetization factors of the ellipsoidal grain. In actual ferrite materials there always exists a distribution of grains both in size and in shape, which is determined by the production technology. This

TABLE 1

	ω_1	ω_2
μ_x	$\omega_\Theta + \omega_M$	$\omega_\Phi + N_t\omega_M$
μ_z	ω_Φ	$\omega_\Theta + N_l\,\omega_M$

TABLE 2

	ω_1	ω_2
μ_x	$\omega_a + \omega_M$	$\omega_a + N_t\omega_M$
μ_y	ω_a	$\omega_a + N_t\omega_M$

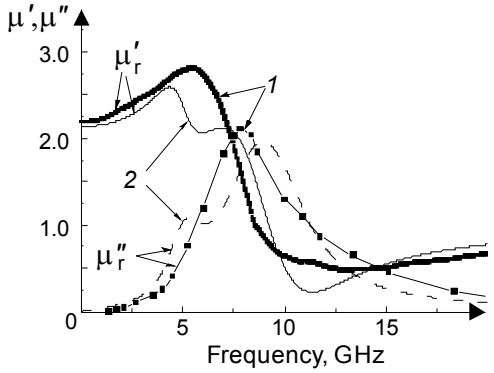

Fig. 2. Comparison of experimental smoothed spectra (curve *1*) with calculated ones (curve *2*).

results in a difference in resonance frequencies in different crystal grains, which must be taken into account when calculating the permeability of a polycrystal. In this connection, based on micrographs of metallographic speciments of the materials under investigation, the form of the distribution function $f(N_l)$ of demagnetization factors has been found for the grains, which were approximated by ellipsoids of revolution, and the permeability for the polycrystal was averaged by the formula

$$\mu_{av} = (1/3)\int f(N_l)(\mu_{xx} + \mu_{yy} + \mu_{zz})\, dN_l\,. \qquad (4)$$

The integration was carried out within the limits from $N_l = 1/3$ (grain shaped as a sphere) to $N_l = 1$ (thin disk).

To construct theoretical MP spectra of the hexaferrites by formula (4), it is necessary to know the static parameters, such as the fields of anisotropy H_Θ and H_Φ of the materials with EMP, H_a of the hexaferrites with EMA, and the saturation magnetization M_0. These quantities, alongside with the demagnetization factors of grains and their distribution, determine the values of the initial static permeability $\mu_r(0) = \mu_r'(0)$ and resonance frequencies of the spectra of polycrystalline hexaferrites, and an increase in saturation magnetization increases, while an increase in anisotropy fields decreases $\mu_r(0)$. The imaginary part of the MP under resonance is directly proportional to the saturation magnetization and inversely proportional to the damping constant α. The rate of fall of the real part of the permeability in the resonance range is also inversely proportional to the damping constant. The values of the anisotropy fields H_Θ and H_a and magnetization M_0 for the type $Co_{2-x}Zn_xW$ hexaferrites are taken by us from [13, 14]. The value of the saturation magnetization of the polycrystal that was substituted in the calculation formulas was obtained taking into account the correction for the porosity p of the material: $\langle M_0\rangle = M_0(1 - p)$. Figure 2 presents a comparison of the smoothed experimental spectra $\mu_r'(\omega)$ and $\mu_r''(\omega)$ of $Co_{1,2}Zn_{0,8}W$ hexaferrite (curves *1*) with the spectra calculated by formula (4) (curves *2*). In constructing the calculation curves, it was assumed that $H_\Theta = -4$ kOe, $4\pi\langle M_0\rangle = 3.6$ kG. The parameters that were varied in constructing theoretical curves were the damping constant α and the anisotropy field in the base plane, H_Φ. The spectra given in Fig. 2 have been obtained for $\alpha = 0.15$ and $H_\Phi = 0.8$ kOe. From this figure it can be seen that the presence of different resonance frequencies for the components of the MP tensor of a multidomain grain leads to a complicated structure of the spectrum in the NFMR range, which shows up in two maxima in the spectrum of the MP imaginary part and in a step in the dependence $\mu_r'(\omega)$.

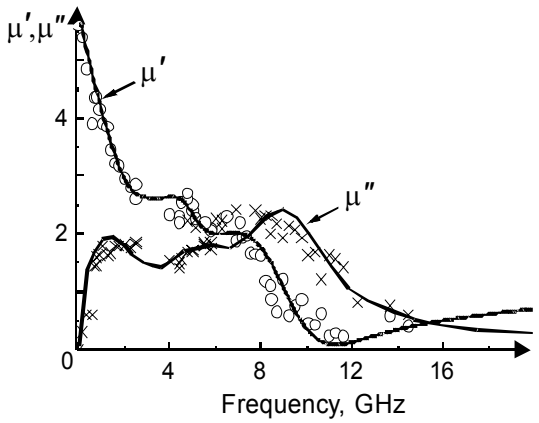

Fig. 3. The calculated (lines) and the experimental spectrum (points) of the MP of $Co_{1.2}Zn_{0.8}W$ hexaferrite.

Figure 3 presents the measured MP spectra of $Co_{1.2}Zn_{0.8}W$ hexaferrite and the calculation curves that sum up the contributions to the spectrum from the processes of displacement of DB's, $\mu_d'(\omega)$ and $\mu_d''(\omega)$ (see Fig. 1), and from the processes of rotation (curves *2* from Fig. 2).

The observed good agreement between the calculated spectra and experimental data suggests that the simple model of independent grains adequately describes the nonmonotone character of the MP spectra observed in experiment in the microwave range with a minimum number of adjustable parameters. Similar results have been obtained for other compositions of hexaferrites of the given type, which, at room temperature, show anisotropy of both EMP and EMA types. For the hexaferrites with EMP, the agreement between theory and experiment is better than that for the materials with EMA both in the position of additional maxima and in their magnitudes. This can be related to the stronger effect of magnetic interaction between grains, not considered in the theory, in hexaferrites with EMA than in materials with EMP. To refine the proposed approach to a description of MP spectra, it is necessary to invoke the results of the theory of composition mixtures.

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