

## PHYSICS OF MAGNETIC PHENOMENA

### ANALYSIS AND CORRECTION OF THE MAGNETIC PERMEABILITY SPECTRA OF $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ HEXAFERRITE BY USING CRAMERS–KRONIG RELATIONS

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UDC 621.317.023, 538.62

*The frequency dependences of the magnetic permeability of  $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$  ferrite with hexagonal crystalline structure are given which have been obtained by waveguide and resonator methods in the frequency range 0.1–11 GHz. The data on the magnetic spectra of this material available in the literature have been analyzed to select the most reliable experimental results. The expert judgement on the spectra was made using the Cramers–Kronig dispersion relations. It has been shown that the Cramers–Kronig relations can be used to correct complex magnetic permeability measurements by reciprocal recalculation of the frequency dependences of the real and imaginary parts.*

#### INTRODUCTION

The current development of microelectronics is related to further mastering of the high-frequency and superhigh-frequency wavelength ranges. This is promoted by miniaturization of mobile communication devices, development of high-speed digital photo- and videocameras and wireless computer networks as well as by the increase in operation speed of personal computers.

Much attention is now given to the search for new radiomaterials and nonconventional technologies of their production and to the modification of the properties of already available radiomaterials for their efficient use in the microwave range of electromagnetic radiation. In this connection, ferrimagnetic materials have a special place. They are used in manufacturing multilayer chip inductors [1–5], to increase the efficiency and miniaturize antennas of communication facilities, including mobile phones [6, 7], as radioswallowing coatings and as materials ensuring electromagnetic compatibility between the gigahertz processors of personal computers and wireless computer communication facilities [8, 9] and also between details and units of mobile phones [6].

The frequency band of NiCuZn spinel-structured ferrites, which are conventionally used for these purposes, is restricted by 300 MHz frequencies [3, 5, 10]. In the low-frequency microwave range, ferrites with hexagonal crystalline structure (hexaferrites), synthesized on the basis of the  $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$  ( $\text{Co}_2\text{Z}$ ) compound [1–12], which possesses high magnetic permeability and rather high dispersion frequency, show promise. The frequency at which the magnetic losses of  $\text{Co}_2\text{Z}$  hexaferrite synthesized by conventional ceramic technology are maximum is close to 1.4 GHz at room temperature [10]. The use of this material in integral microelectronic devices is hindered by the high temperature of final roasting (1260°C) which makes them unsuitable for use in the planar technology of deposition of ferrite materials immediately on strip conductors. In this connection, various ways of reducing this temperature are proposed, such as the low-temperature sol-gel-synthesis [3, 4, 11, 12] or adding temperature-reducing glass-forming components to the stock at the stage of final roasting [1, 2, 5–7].

The information necessary for practical use of ferrimagnetics can be gained by studying their dynamic characteristics – the spectra of the complex values of magnetic permeability (MP)  $\mu^*(f) = \mu'(f) - i\mu''(f)$ . Since the theory that

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would describe the MP spectra of polycrystalline ferrites in the microwave range is yet to be developed, the sole way of obtaining informations about them is to determine experimentally the  $\mu^*(f)$  values in the desired frequency band. Reviewing the relevant literature [1–9, 11, 12], one can conclude that the  $\mu^*(f)$  spectra of  $\text{Co}_2\text{Z}$  hexaferrite obtained by different authors are substantially different. Many experts compare their results to the  $\mu^*(f)$  spectra given in the monograph by Smith and Wijn [10]. Our investigations have shown that the results reported in [10], as a rule, do not correlate with the measured spectra [1–9, 11, 12].

In this paper, the frequency dependences of the complex magnetic permeability of  $\text{Co}_2\text{Z}$  hexaferrite taken from various publications are analyzed. Their reliability is evaluated and the way of coordinating the spectra by a correction procedure based on the Cramers–Kronig relations is shown. The effect of this correction on the results of calculations for radioswallowing coatings is discussed. The corrected data taken from the literature are compared to the magnetic permeability spectra of  $\text{Co}_2\text{Z}$  hexaferrite measured by us.

## ANALYSIS OF THE SPECTRA BY MEANS OF THE CRAMERS–KRONIG RELATIONS

In our opinion, it is reasonable to perform the fitting of the measurements of the real and imaginary parts of the permeability and also the interpolation and extrapolation of experimental data with the help of the Cramers–Kronig dispersion relations, which are valid for the response functions of arbitrary linear systems. For the components of complex magnetic permeability they have the form [10]

$$\mu'(f) - 1 = \frac{2}{\pi} \int_0^{\infty} \frac{f_1 \mu''(f_1)}{f_1^2 - f^2} df_1, \quad \mu''(f) = -\frac{2f}{\pi} \int_0^{\infty} \frac{\mu'(f_1)}{f_1^2 - f^2} df_1. \quad (1)$$

Note that the integrals in (1) are evaluated in the sense of the Cauchy principal value.

In finding the spectrum  $\mu'(f)$  by a known relation for  $\mu''(f)$  (or vice versa), to simplify the calculations, it is possible to apply a piecewise linear approximation of the imaginary (real) component in the frequency band under consideration, which was proposed by Polivanov [13]. The entire frequency range of  $\mu''(f)$  ( $\mu'(f)$ ) is subdivided into intervals with coordinates  $f_i$  ( $i = 1, 2, \dots, N$ ) in which the corresponding function can be approximated by linear expressions:

$$\mu_i'' = k_i f_i + d_i \quad \text{or} \quad \mu_i' = k_i f_i + d_i. \quad (2)$$

Substituting (2) in (1) and evaluating the integrals, we obtain formulas (3) for recalculation of the permeabilities. To avoid ambiguities, the calculation of spectra by these formulas is performed for the frequencies  $f$  that do not coincide with the boundary points  $f_i$  of the approximation intervals. The fitting of measurements was carried out by reciprocal recalculation of  $\mu'(f)$  by  $\mu''(f)$  and vice versa:

$$\begin{aligned} \mu'(f) &= 1 + \frac{1}{\pi} \sum_{i=1}^N \left\{ 2(\mu_{i+1}'' - \mu_i'') + k_i f \ln \left| \frac{(f_{i+1} - f)(f_i + f)}{(f_{i+1} + f)(f_i - f)} \right| + d_i \ln \left| \frac{f_{i+1}^2 - f^2}{f_i^2 - f^2} \right| \right\}, \\ \mu''(f) &= -\frac{1}{\pi} \sum_{i=1}^N \left\{ k_i f \ln \left| \frac{f_{i+1}^2 - f^2}{f_i^2 - f^2} \right| + d_i \ln \left| \frac{(f_{i+1} - f)(f_i + f)}{(f_{i+1} + f)(f_i - f)} \right| \right\}. \end{aligned} \quad (3)$$

The Cramers–Kronig relations (1) are strictly fulfilled in an infinite interval of frequencies, which is unattainable in actual physical experiment. Therefore, their replacement by formulas (3) involves an error related to the boundedness of the frequency interval in which approximation is performed. The results of calculations by the Cramers–Kronig relations are rather sensitive to jumps in frequency dependences of permeability. The boundedness of the frequency range of measurements implies that such jumps occur at boundary points, which is physically not justified. This error can be minimized by adding on the missing intervals with smooth curves, assuming that these missing intervals are ranges of

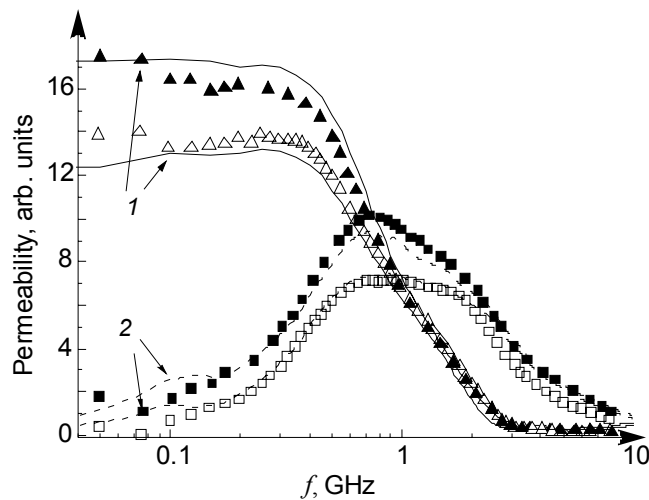


Fig. 1. Magnetic spectra of  $\text{Co}_2\text{Z}$  hexaferrite samples [8]: real (1) and imaginary parts of permeability (2). Points: experiment, lines: calculation.

anomalous dispersion in the low-frequency and high-frequency parts of the spectra outside the measurement range. In doing this, it is necessary to take advantage of the analytic properties of the function  $\mu^*(f)$ : (a)  $\mu''(f)$  tends to zero as  $f \rightarrow 0$  and  $f \rightarrow \infty$  and the area bounded by the curve  $\mu''(f)$  (it is proportional to the saturation magnetization) is finite and (b)  $\mu'(f)$  is bounded: as  $f \rightarrow 0$ ,  $\mu'(f)$  tends to  $\mu_0$ , the static initial permeability, which is determined by independent experiments (it is also proportional to the saturation magnetization), and  $\mu'(f) \rightarrow 1$  as  $f \rightarrow \infty$ .

Numerical experiments with the spectra obtained by solving the Landau–Lifshits equation of motion for a magnetic moment [10] and with those measured in experiments have shown the following. The added segments of spectra, if the jumps are not abrupt, weakly affect the magnetic permeability calculated for the basic range. To provide reasonable accuracy for the restoration of the corresponding characteristics for spectra with one region of abnormal dispersion, it well suffices to have 25–30 nonregular partitions of the frequency interval in which piecewise linear approximation is performed.

Owing to finiteness of the area bounded by the curve  $\mu''(f)$ , the added frequency interval required in restoring the real part by the imaginary one is considerably smaller than that required in the inverse restoration. This interval is chosen by the condition that the restored and measured values of the static initial permeability or the low-frequency ranges of the restored and measured spectra  $\mu'(f)$  are close to each other.

In restoring the imaginary part of the permeability by the real one it is necessary to extend the high-frequency boundary of the added-on spectral range to frequencies of  $\sim 500$  GHz. For this range, not coming into conflict with the up-to-date physical notions,  $\mu'(f)$  can be taken equal to unity. The necessity of such extension is related to that the drop in  $\mu'(f_N)$  from unity to zero near the upper bound of the analyzed spectrum leads to appreciable distortions in the restored relation  $\mu''(f)$  at these frequencies.

## PROCESSING OF THE EXPERIMENTAL MP SPECTRA OF $\text{Co}_2\text{Z}$ HEXAFERRITE BY MEANS OF THE CRAMERS–KRONIG RELATIONS

To illustrate the application of formulas (3) to processing experimental data, we shall consider the spectra of  $\text{Co}_2\text{Z}$  hexaferrite given in [8]. Synthesis of materials for the experiment described in [8] was performed by the ceramic technology in oxygen. The MP spectra obtained were noticeably different from those given in [10]. Figure 1 presents the  $\mu^*(f)$  spectra measured on samples synthesized at an oxygen pressure  $P_{\text{O}_2} = 21.3$  (open circles) and 101.3 kPa (solid circles). The ferrite synthesized at the greater  $\text{O}_2$  pressure shows greater values of the static initial permeability and MP imaginary part at the

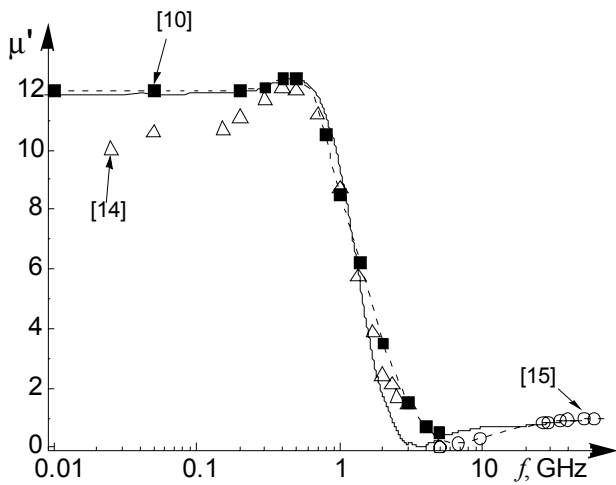


Fig. 2

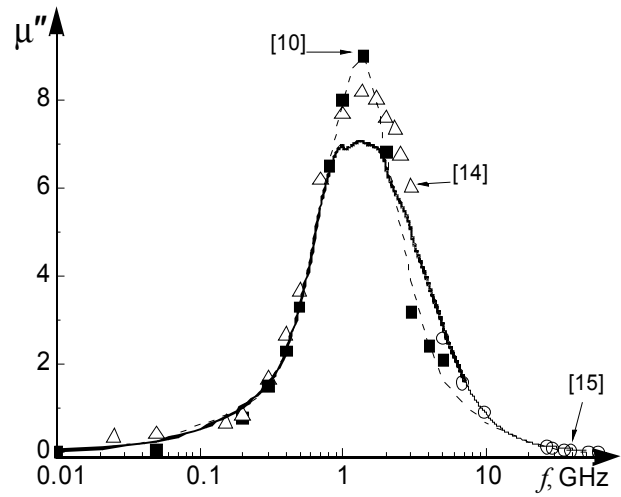


Fig. 3

Fig. 2. Frequency dependence of the real part of the permeability of  $\text{Co}_2\text{Z}$  hexaferrite. Dashed line: piecewise linear approximation; solid line: restored spectrum.

Fig. 3. Frequency dependence of the imaginary part of the permeability of  $\text{Co}_2\text{Z}$  hexaferrite. Dashed line: piecewise linear approximation; solid line: recovered spectrum.

greatest losses. The frequency dependences of the imaginary part of the permeability have two maxima. The low-frequency maximum is treated by the authors of [8] to be caused by the processes of displacement of domain boundaries, and the step in the high-frequency dip is considered to be related to natural ferrimagnetic resonance. The piecewise linear interpolation of the corresponding characteristics in the measurement range, necessary for calculations by formulas (3), was performed immediately over the experimental points. The solid lines in Fig. 1 represent the real parts of the permeability restored by the experimental spectra  $\mu''(f)$  and the dashed lines represent the imaginary parts calculated by the relations  $\mu'(f)$ .

Let us consider the spectrum of the hexaferrite produced at  $P_{\text{O}_2} = 21.3$  kPa. It can be seen that the restoration of the real part of MP gives an underestimated value of the permeability at low frequencies compared to its measured value. In the region of the greatest losses, the fall in  $\mu'(f)$  occurs at lower frequencies than this takes place for the measured spectrum. Calculations of  $\mu''(f)$  by  $\mu'(f)$  show that the low-frequency portion of the spectrum is in rather good agreement with experiment, except for the position of a small additional maxima at a frequency of  $\sim 0.1$  GHz, which is due to the presence of a step in the curve  $\mu'(f)$ . For the high-frequency dip, the calculated spectrum is observed to appreciably shift relative to the measured one toward the high frequencies. Similar comparison of the spectra of the hexaferrite synthesized at  $P_{\text{O}_2} = 101.3$  kPa shows that the calculation of  $\mu'(f)$  by  $\mu''(f)$  gives an overestimated value of  $\mu'$  at frequencies lower than 1 GHz. By the inverse calculation of  $\mu''(f)$  by  $\mu'(f)$ , underestimated values of  $\mu''$  are naturally obtained in the region of the greatest losses. Thus, the measured MP spectra of both materials require corrections.

Since we measured spectra for samples synthesized by the conventional ceramic technology in the air at a final roasting temperature of  $1260^\circ\text{C}$ , we could not invoke present-day publications [1–9, 11, 12] for their comparison with the data available in the literature. In the experiments described in [1–9, 11, 12], either microcrystalline materials with grain sizes of about several micrometers were obtained by different synthesis technologies, or the conditions of synthesis of  $\text{Co}_2\text{Z}$  hexaferrite were substantially different from ours. In this connection, for the analysis and comparison purposes we have taken the spectra measured for nontextured polycrystalline samples of  $\text{Co}_2\text{Z}$  hexaferrite at frequencies of up to 5 and 3 GHz [10, 14], respectively, and from 5 to 60 GHz [15]. The frequency dependences of the real and imaginary parts of MP taken from these publications are represented in Figs. 2 and 3, respectively. According to Fig. 2, the experimental data on  $\mu'$

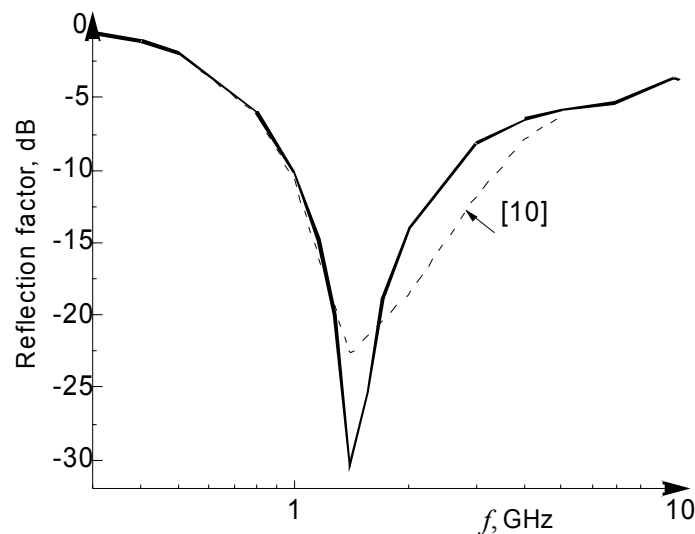


Fig. 4. Frequency dependence of the reflectivity for a  $\text{Co}_2\text{Z}$  hexaferrite plate placed on a metal. Explanations are given in the text.

correlate among themselves except for the discrepancy between the measurements at low frequencies given in [10] and in [14]. The experimental data on spectra  $\mu''(f)$  taken from different publications (see Fig. 3) are close to each other. For analysis we used in the main the spectra presented in [10].

The dashed lines in Figs. 2 and 3 present the results of the piecewise linear approximation and the solid lines refer to the calculations by formulas (3). For the calculation of  $\mu''(f)$  by  $\mu'(f)$  (see Fig. 3) at frequencies of up to 1 GHz, the calculated curve practically coincides with experiment. However, the maximum of  $\mu''$  at resonance is noticeably smaller and the breadth of the line is greater than for the experimental spectrum. The relation  $\mu'(f)$  restored by  $\mu''(f)$  at frequencies below 1 GHz also coincides with experiment, however falling more abruptly in the region of resonance. The reverse recalculation by the restored  $\mu''(f)$  spectrum gives a curve of  $\mu'(f)$  which almost coincides with the approximating curve. These spectra were taken as the corrected spectra when we compared our results with the data available in the literature. The effect of the correction of spectra is illustrated by Fig. 4 in which the calculated frequency dependences of the coefficient of reflection from  $\text{Co}_2\text{Z}$  hexaferrite layers of thickness 3.2 mm applied on a metal are presented. The permittivity was taken equal to  $\epsilon^* = 15 - i1$ . The dashed curve is constructed by the experimental data of [10]; the solid line represents the corrected spectrum. A considerable difference in reflectivities in the region of resonance can be seen.

The region of anomalous dispersion for  $\text{Co}_2\text{Z}$  hexaferrite, which is characterized by a significantly decreased real component of the MP and by increased magnetic losses, occupies a wide frequency band; therefore, measurements were carried out by several methods. For the low-frequency range (0.1–1 GHz), the coaxial waveguide method was used with the sample of the material under investigation shaped as a thin washer (outside diameter 16 mm, inside diameter 4.6 mm, thickness 1–2 mm). Measurements were performed by a routine procedure [16] on a measuring line. For the high-frequency range (0.9–11 GHz), transmission-type rectangular resonators with type  $H_{10n}$  oscillations were used in which samples shaped as thin cylinders (diameter 1–2 mm, length 23–75 mm) [17] were placed. For the range where methods with distributed and lumped parameters are applicable (0.2–1 GHz), measurements were performed on a nonregular microstrip resonator [18] made of high-permeability ceramics with the sample shaped as a thin plate of width 5 mm, length 10 mm, and thickness 1 mm. The use of three various measuring methods with superimposed frequency ranges on variously shaped samples made the experimental results obtained more reliable.

The resonance measurements could be performed for a wide frequency range due to the use of a set of resonators (length variation method) and multimode conditions (frequency variation method) where oscillations in a resonator arise at a varied number of half-waves fitting in its length. A combination of these methods for increasing the density of the frequency network (set of resonators and multimode conditions) allows one to perform measurements for about a hundred

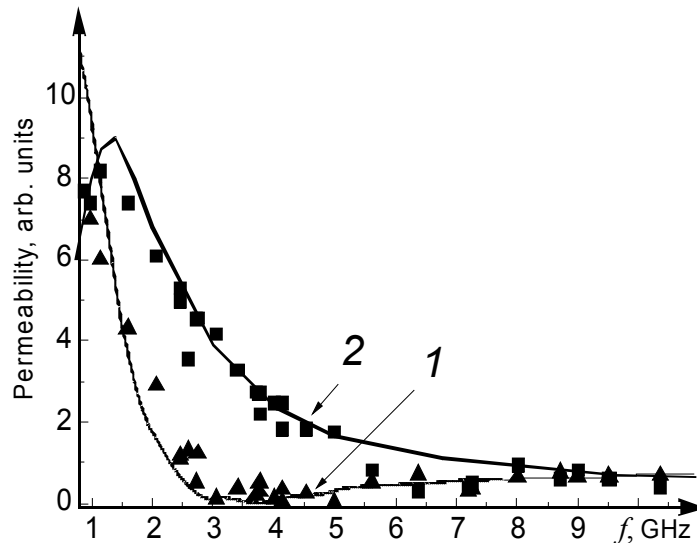


Fig. 5. Measurements of the MP spectra of  $\text{Co}_2\text{Z}$  hexaferrite: curve 1: real part, curve 2: imaginary part.

of discrete frequency points in the given range. The microwave duct was made as a set of elements of conventional meters of the voltage standing wave ratio. The data processing rate and the quality of the experiment were increased due to the use of a personal computer because the measurements were performed in real time [18].

The phase composition of the samples was examined by x-ray diffraction analysis; the basic Z-phase made 95%. The density was determined by hydrostatic weighing; it was equal to  $4.99 \text{ g/cm}^3$ . The measured complex permittivity was  $\epsilon^* = 15 - i1$  and weakly depended on frequency in the measurement range.

Because of the great values of permeability and permittivity, dimensional (volumetric) resonance might occur which could substantially distort the results. Our estimates obtained by the procedure proposed in [13] have shown that for the chosen sample dimensions (diameter of the cylinder or thickness of the washer) this effect makes less than 1% at frequencies of up to 11 GHz in the most unfavorable case.

The estimated measurement error made 3–5% for  $\mu'(f)$  and 15–18% for  $\mu''(f)$ . The greatest contribution to the measurement error was introduced by the instability of the frequency of the sweep generator which shows up when the sweep band, which is used for a detailed description of the resonance characteristic, is narrow.

The MP spectra of  $\text{Co}_2\text{Z}$  hexaferrite measured by us are given in Fig. 5. The lines in the figure represent the corrected spectra taken from [10, 14, 15]. The maximum and minimum present in the frequency dependence of the real component in the region where  $\mu'(f)$  varies abruptly shows that the spectrum is resonance rather than relaxation in character, as this could be supposed based on the data of [10].

## CONCLUSION

Our investigations have shown that the spectra of the magnetic permeability of  $\text{Co}_2\text{Z}$  hexaferrite available in the literature show discrepancies. Analysis of the spectra by using the Cramers–Kronig relations enables one to select the most reliable experimental results.

It has been shown that the Cramers–Kronig relations can be used not only to interpolate and extrapolate experimental data for the spectra, but also to check the correctness of MP spectra measurements and to fit the experimental data by the real and imaginary parts of the permeability. This correction noticeably changes the frequency dependences of the characteristics of various electrodynamic devices calculated by measured spectra.

The magnetic permeability spectra measured by us are in good agreement with the corrected data taken from the literature.

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