COMPUTATIONAL PROBLEMS OF ELECTRICAL ENGINEERING

Vol. 2, No. 2, 2012

MAGNETIC METAMATERIALS AS PERSPECTIVE MATERIALS OF RADIOELECTRONICS

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Abstract: Electromagnetic properties of 3D-nanocomposites based of the opal matrixes containing particles of one or two transitive metals are investigated. Phase analysis of the nanocomposites is carried out. Microwave measurements are executed in the frequency interval of 26-38 GHz. Field dependences of the factors of passage and reflexion are obtained. Spectra of a magnetic resonance and antiresonance are restored. Frequency dependences of an amplitude of the resonance and antiresonance are obtained. It is established, that in the nanocomposites containing particles of two transitive metals, the magnetic resonance amplitude is much larger than in the nanocomposites containing particles of one metal.

Key words: opal matrix, metamaterials, microwave properties.

1. Metamaterials – a new class of materials with unique electromagnetic properties

Opal matrixes are considered as one of the most perspective classes of materials for application in devices of optical and microwave ranges. Now, linear and nonlinear optical properties of the opal matrixes, the photo induced absorption in them, changes of factor of refraction, and also a variation of intensity, polarization and coherence occurring are intensively investigated when passing through matrixes of powerful coherent radiation [1]. The structure, and physical properties of opal matrixes filled with metal or ferromagnetic nanocorpuscles have been investigated in detail [2]. The specificity of optical properties of two- and three-dimensional objects on the basis of opal matrixes [3] has also been considered. The greatest interest is caused by the properties of ensembles of various microspheres and matrixes as photon crystals. Interaction of high-frequency electromagnetic waves and magnetophonon crystals considered as metamaterials is the most actual direction in the specified area. Applied aspects of obtaining a negative refrection indicator at the millimetric range frequencies, using the phenomenon of magnetic resonance, are considered to be promising. Earlier the formulation of a solution to the problem of interaction of electromagnetic waves and stratified environment [4], in particular, consisting of cores with a negative refraction indicator has been proposed. Propagation of the directed waves in the environment has been studied, and application of such an environment in linear and dipole antennas has been considered. The character of waveguide propagation of waves in planar waveguides prepared on the basis of a 3D layered photon crystal [5] has been investigated. It is shown, that it is possible to achieve conditions of full passage of a wave through such a waveguide structure.

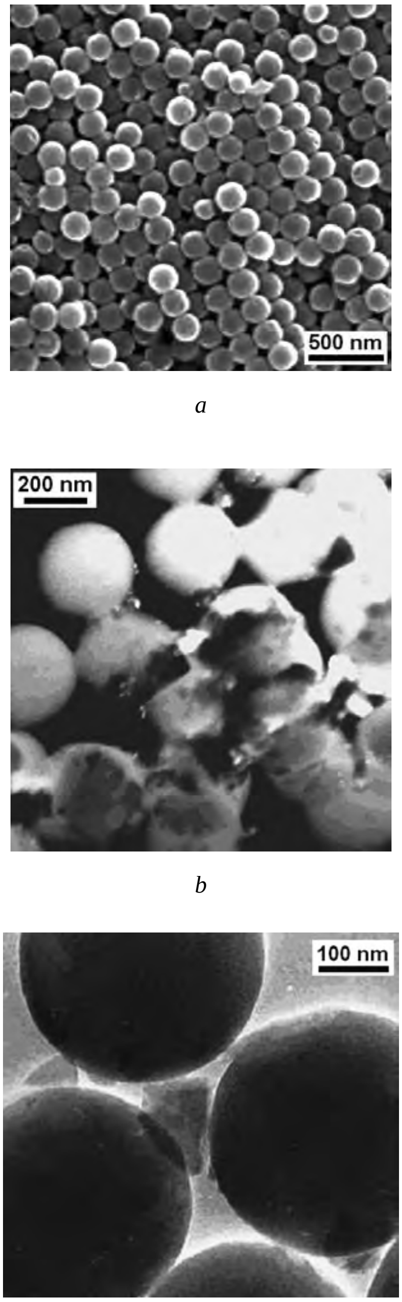
Although a standard definition of metamaterial has not been developed yet, we will use, therefore, the following one. A metamaterial is a two- or three-dimensional environment, discrete on the nanoscale sizes and consisting of a component, considerably differing in electromagnetic properties. Such components can be, for example, metal and dielectric, ferromagnetic and paramagnetic, etc. The most interesting line of a metamaterial considers its ability to get, under certain conditions, a negative sign on the valid part of a refraction indicator. A special class of metamaterials is made by environments from a dielectric constant close to zero or so-called ENZ (Epsilon-Near-Zero) – materials. The effect of "supercommunication" (supercoupling) represents one of vivid examples of abnormal propagation of waves in such environments [6, 7]. The specified phenomenon represents wave tunneling through narrow channels and the bends connecting two wave guides in conditions when a usual wave passage is impossible. In the area where dielectric permeability is close to zero, and, accordingly, the refraction indicator is close to zero because of the large phase speed and almost homogeneous wave front the reflexion factors are insignificant.

In metamaterials with a dielectric constant close to zero there is one more unusual phenomenon – strengthening when the waves are passing through a small aperture [8]. The propagation of electromagnetic waves in the disperse environment with almost zero indicator of refraction is theoretically considered in [9], and both a mode of a continuous wave and transients are discussed. In this particular work, physical properties of a class of metamaterials, i.e. nanocomposite environments based of opal matrixes with inclusions magnetic or metal nanocorpuscles, will be considered. Microwave properties of such environments and effect of a magnetic field on them are considered in more detail. Synthesis of opal matrix samples with the diameter of nanospheres SiO2 ranging from 200 to 400 nm is executed with the use of the following technological operations [10]. Nanocorpuscles of the amorphous SiO_2 are produced by the accelerated technology based on the reaction of tetraether hydrolysis of optosilicon acid Si (OC2H5)4 with solution of ethanol C2H5OH at the presence of of ammonium hydroxide NH4OH which served as a catalyst. In the beginning of the hydrolysis reaction, there were formed small branched nanocorpuscles, and then, in the course of polycondensation, they transformed into particles of amorphous spherical-shaped silicon dioxide. After suspension and removal upholding hydrolysate, the ordered deposit is a hydrogel contaning the amount of liquid up to 50-60% of the total weight, chalky, breakable, and, therefore, it is necessary to consistently conduct various stages of thermal treatment for the purpose of hardening of the obtained opal matrixes. The control of correctness of packing nanospheres was carried out according to the form and width of Bragg reflexion bands.

One of the simplest and widely applied ways of introduction of various chemical elements (and connections) into opal matrixes is the impregnation method. The method is based on impregnation of an opal matrix by a substance – precursor with a certain chemical compound with the subsequent thermal treatment in the process of which, in interspherical emptiness of an opal matrix, the necessary chemical compound is formed. Substances – precursors should possess good solubility in water (or in other solvents) and turn into oxides (or in other compounds) at moderate temperatures of heat treatment. As such precursors it is possible to use soluble salts of metals (in the given work nitrates Fe, Ni were applied, Mn and Zn).

In the course of impregnation, salts' water solutions spontaneously, at the expense of capillary effect, fill up the pores of an opal matrix. Subsequently, thermal treatment is carried out where partial thermal decomposition of nitrate groups and complete free water extraction take place. In this case the thermal treatment was conducted within several hours in the air at the temperatures of 770-870 K. It was a repeated procedure (up to 20 impregnations) with gradual filling of interspherical space of the opal matrix with oxides and the subsequent high-temperature thermal treatment for obtaining a desired structure. To receive metal particles from the besieged oxides of metals we apply annealing in hydrogen atmosphere.

The structure of an opal matrix before magnetic nanocorpuscles are embeded into interspherical emptiness is an ordered nanosphere ensemble forming a densely packed periodic structure. After the introduction procedure, the most part of the brought substance is concentrated in the space between nanospheres. That is proved by the results of X-ray analysis and electronic microscopy executed on the scanning electron microscope (SEM) CARL ZEISS LEO 1430 VP and transmission electronic microscope (TEM) JEM-200CX specify. The X-ray analysis has shown that in the brought substance there are some ferriferous phases. In particular, in a nickel-zinc ferrite nanocomposite, most of reflexes refer to the phases of ZnFe2O4 and (NiXZn1-X)Fe2O4 type, having the crystal structure of a spinel. Particles of the entered phases have a polycrystalline structure and are characterized by the wrong form sized from 5 to 70 nm. In Fig. 1, the structure of opal matrix and nanocomposites with inclusions of nickel-zinc-ferrite and metal cobalt particles is shown. Volume concentration of the brought phases does not exceed 5-15 %.



a

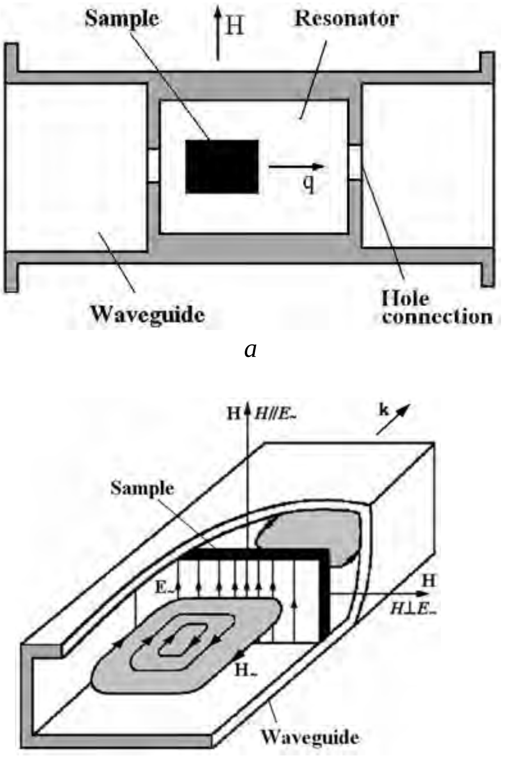
b

c

Fig. 1. The SEM (a) and TEM (b, c) image of structure opal matrix (a) and mechanical disruption of nanocomposites with nickel-zinc-ferrite (b) and metal cobalt (c) particles.

2. Microwave properties of nanocomposites. Magnetic resonances

Microwave measurements have been executed placing the sample both in the resonator and in the wave guide. In the latter case the measurements are made according to the scheme shown in Fig. 2. The effect of an external constant magnetic field on the transfer factor has appeared various at different orientation of this field relative to a microwave magnetic field. There were measured a relative change in an external magnetic field of the module of transfer factor $d_m = \left[|D(H)| - |D(0)| \right] / |D(0)|$ and change of the module of reflexion factor $r_m = \left[|R(H)| - |R(0)| \right] / |R(0)|$, where $D(H)$ and $R(H)$ are the factors of passage into and reflexion from the sample, measured in the field H.



a

b

Fig. 2. The scheme of an arrangement of the sample in resonator (a) and wave guide (b).

Comparison of the results of measurement of field dependence of passage and reflexion factors for a nanocomposite, containing nickel-zinc ferrite, at $H \perp H_{\sim}$ is presented in Fig. 3. The obtained dependences have appeared to have a similar changes magnitude, and dependence form. With the frequency increasing, the position of resonant feature is necessary in stronger fields; with the frequency growing, the amplitude of a resonance increases. The spectra of a magnetic resonance have been achived by measuring the resonant fields at different frequencies.

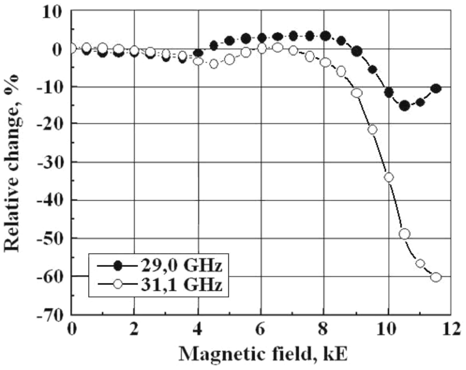
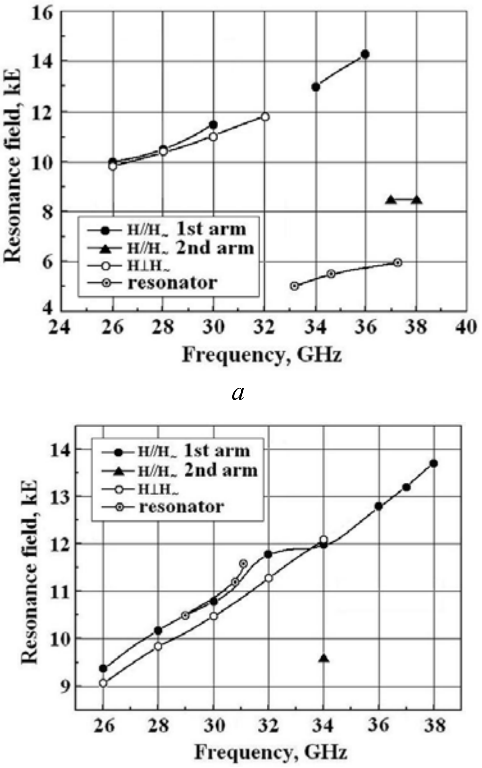


Fig. 3. The magnetic resonance measured by means of refleting and passing the microwaves through the nanocomposite sample, containing nickel-zinc ferrite, $H \perp H_{\sim}$. Measurements done in the resonator.

The measurements are executed both in a wave guide, and in resonators of various widths. Results for the nanocomposite, containing nanocorpuscles of ferrite manganese-zinc and nickel-zinc, are shown in Fig. 4, a, b. While being measured, the sample of a nanocomposite was placed in the resonator centre. The resonance fields created due to the waveguide technique, and referring to an acoustic branch of a resonant spectrum, are shown in Fig. 4,b by round symbols. Asterisks note the resonances concerning other branches of the spectrum.



a

b

Fig. 4. A spectrum of magnetic resonance for the nanocomposite sample, containing manganese-zinc-ferrite (a) and nickel-zinc-ferrite (b).

Given the measurements of passage and reflexion factors without a magnetic field, the attenuation factor in several nanocomposites has been defined. The attenuation factor is an important parameter of a material defining real losses of a microwave device. Frequency dependences of the attenuation factor in a millimetric range of wave lengths are shown in Fig. 5.

Changes in the value of the factors in a magnetic field can be great. For example, for a nanocomposite with particles of cobalt-zinc of ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$, the maximum change in the value of the passage factor reaches 19 % (see Fig. 6).

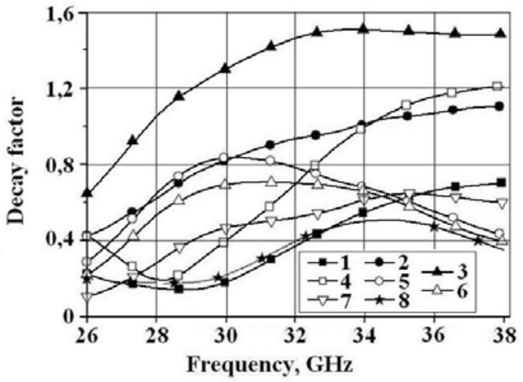


Fig. 5. Frequency dependence of attenuation factors in 3D-nanocomposites:
1 – $Co_{0,35}Zn_{0,65}Fe_2O_4$, 2 – $Co_{0,5}Zn_{0,5}Fe_2O_4$, 3 – $Ni_{0,5}Zn_{0,5}Fe_2O_4$, 4 – $Mn_{0,5}Zn_{0,5}Fe_2O_4$, 5 – $Mn_{0,3}Co_{0,3}Zn_{0,4}Fe_2O_4$, 6 – $La_{0,3}Co_{0,3}Zn_{0,4}Fe_2O_4$, 7 – $Nd_{0,3}Co_{0,3}Zn_{0,4}Fe_2O_4$, 8 – an opal matrix without ferrite.

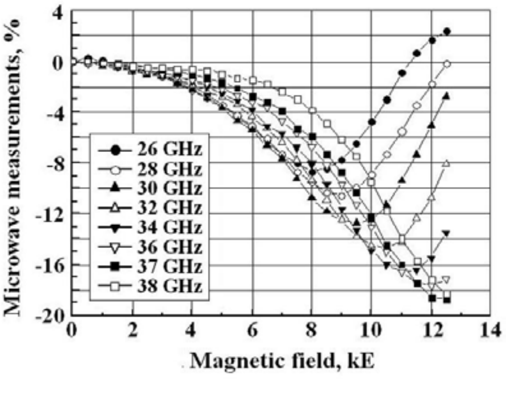
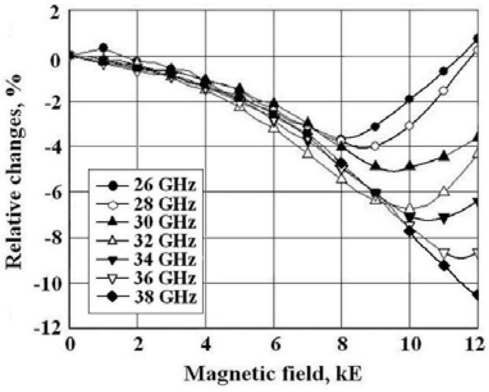


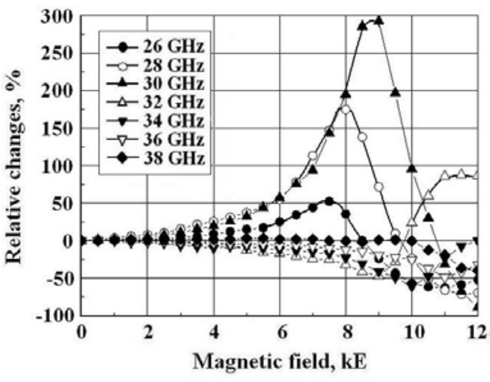
Fig. 6. Dependence of factor of passing through a nanocomposite with particles of cobalt-zinc-ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$ on the intensity of an external magnetic field.

Quite a big change of the passage factor for a nanocomposite with particles of ferrite neodymium-zinc-cobalt is showen in Fig. 7. A physical reason for such a change of the passage factor illustrated in Fig. 6 and 7,a is the magnetic resonance.

With the magnetic field changing, changes of the signal reflected from nanocomposites are especially great. Field dependences of changes of the reflected signal for the sample containing nanocorpuscles of nickel-zinc of ferrite, at $H \perp H_{\sim}$ are shown in Fig. 7,b. Much attention should be paid to very big up to 4 times changes of the reflected signal which can be defined as huge. Signal reduction in a resonance visually (Fig. 7, b) seems to be less considerable, but actually it reaches - 90%. In other words, in the field of a resonance, the reflected signal 10 times decreases.



a



b

Fig. 7. Field dependence of reflexion factor for a nanocomposite, containing particles of $Nd_{0,3}Co_{0,3}Zn_{0.4}Fe_2O_4$ (a) and $Ni_{0,5}Zn_{0,5}Fe_2O_4$ (b).

The further part of discussion is devoted to the development of a method for calculation of factor of passing a wave through a nanocomposite and its dependences on the intensity of an external constant magnetic field. An ultimate goal of such calculation is acquiring the data on field dependences of magnetic permeability and refraction factor. The relationship between the parts of complex wave number for the homogeneous wave travelling along a wave guide axis in the material under investigation, containing a ferromagnetic phase, can be written down as follows:

$$\beta_2 = \sqrt{q^2 - \chi^2} \qquad (1)$$

where β_2 are the wave number in a wave guide, χ stands for the cross-section wave number, $q = q^{'} - iq^{"}$ represents the complex wave number of the electromagnetic wave extending in the unlimited environment. The wave number q is defined by the formula:

$$q = \frac{\omega}{c}\sqrt{\varepsilon_{ef}\,\mu_{ef}}\ , \tag{2}$$

where the effective dielectric permeability ε_{ef} and effective magnetic permeability μ_{ef} are complex values:

$$\varepsilon_{ef} = \varepsilon_1 - i\varepsilon_2 \text{ and } \mu_{ef} = \mu_1 - i\mu_2 .$$

The components of the wave number β_2 are set by the following ratios:

$$\beta_2^{'} = \sqrt{\frac{1}{2}\left(\sqrt{(\Re^4 + \Im^4)} + \Re^2\right)},$$

$$\beta_2^{''} = \sqrt{\frac{1}{2}\left(\sqrt{(\Re^4 + \Im^4)} - \Re^2\right)} \tag{3}$$

where

$$\Re = \sqrt{(\frac{\omega}{c})^2(\varepsilon_1\mu_1 - \varepsilon_2\mu_2) - \chi^2}\ ,$$

$$\Im = \frac{\omega}{c}\sqrt{(\varepsilon_2\mu_1 + \varepsilon_1\mu_2)} \tag{4}$$

Equivalent resistance for the wave of TE10 type is defined by the ratio below:

$$Z_{10} = \frac{\omega\mu_{ef}\,\mu_0}{\beta_{10}} \tag{5}$$

where the wave number $\beta_{10} = \beta_{10}^{'} - i\beta_{10}^{''}$ is determined from the formulas (3), (4), considering that cross-section wave number for a mode of TE10 type is equal to

$$\chi_{10} = \frac{\pi}{a}$$

Let's write down the equation of magnetization movement in the form of Hilbert:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M}\times\mathbf{H} + \frac{\alpha}{M}\mathbf{M}\times\frac{\partial \mathbf{M}}{\partial t} \tag{6}$$

where $\mathbf{M}$ is the magnetization vector, whose module is equal to M, $\mathbf{H}$ is the vector of intensity of a magnetic field, α is the dissipation parameter, $\gamma = \frac{g|e|}{2mc}$ is the gyromagnetic relation, g stands for the factor of spectroscopic splitting. In the linearized equation (6) tensor of high-frequency magnetic permeability looks like [11]

$$\vec{\mu} = \begin{pmatrix} \mu & i\mu_a & 0 \\ -\mu_a & \mu & 0 \\ 0 & 0 & \mu_{\parallel} \end{pmatrix} \tag{7}$$

where $\mu = 1 + 4\pi\chi$, $\mu_a = 4\pi\chi_a$ and $\mu_{\parallel} = 1 + 4\pi\chi_{\parallel}$.

Here:

$$\chi = \chi' - i\chi'' =$$

$$= \gamma M m_F \frac{\omega_H\left[\omega_H^2 - \left(1-\alpha^2\right)\omega^2\right] - i\alpha\omega\left[\omega_H^2 + \left(1+\alpha^2\right)\omega^2\right]}{D} \tag{8a}$$

$$\chi_a = \chi_a' - i\chi_a'' =$$

$$= \gamma M m_F \omega \frac{\left[\omega_H^2 - \left(1+\alpha^2\right)\omega^2\right] - i2\alpha\omega\omega_H}{D}\ , \tag{8б}$$

$$\chi_{\parallel} = -\frac{i\alpha\gamma m_F M}{\omega - i\alpha\omega_H} \tag{8в}$$

where $D = \left[\omega_H^2 - (1+\alpha^2)\omega^2\right]^2 + 4\alpha^2\omega^2\omega_H^2$, $\omega_H = \gamma H_0$, m_F represents the mass fraction of a ferromagnetic phase in the substance volume.

The intensity of magnetic field H_0 corresponds to a constant magnetic bias field which is parallel to the axis 0z. Effective magnetic permeability for an electromagnetic wave, provided $\mathbf{q} \perp \mathbf{H}_0$, is defined by the following relation:

$$\mu_{ef} = \mu - \frac{\mu_a^2}{\mu} \tag{9}$$

Let's calculate the module of factor of the wave passage through a plate of the nanocomposite, accepting that the wave numbers are defined by the formulas (3) – (4), and effective magnetic permeability looks like (9). In further numerical calculations it is accepted, that magnetization of particles of cobalt M = 1194 Gs. For massive samples of cobalt M = 1424 Gs. Conductivity and dielectric permeability of the environment: $\sigma = 0.36$ Sm/m, $\varepsilon_1 = 3.29$ have been defined from the frequency dependences of the passage factor without an external magnetic field. Thickness of the sample $d = 1$ mm, and the volume fraction of a ferromagnetic phase in it is accepted equal to $m_F = 0.07$. For the frequency *f*=26 GHz a value of dissipative parameter is chosen equal to $\alpha \approx 0.166$, so that the line widths of the settlement and experimental dependences near the resonance coincide.

Settlement dependences of the refraction factor of a nanocomposite with metal cobalt particles on the intensity of a magnetic field for several frequencies of a millimetric range are shown in Fig. 8. The magnetic resonance corresponds to a minimum of the passage factor. As Fig. 8 shows, the position of minima for settlement and experimental dependences is close and can be found on the area $H \approx 8.2 \div 8.4$ kE. The experimental dependence when the fields are smaller than the field of a resonance, shows the maximum of passage factor which corresponds to the minimum of wave absorption, namely, to antiresonance. At the frequency of 26 GHz a maximum position is in the field $H \approx 3.9$ kE. The settlement dependence is free of antiresonance, as in the simple variant of the theory used for calculation the latter cannot be obtained.

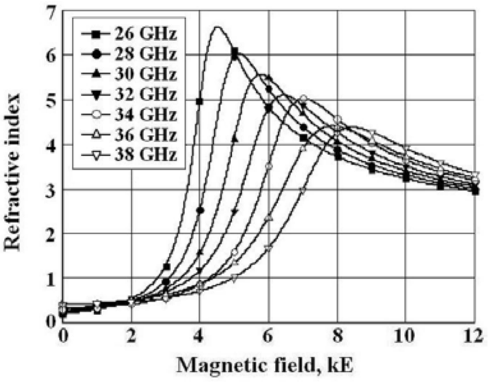


Fig. 8. Dependence of an indicator of nanocomposite refraction with particles of metal cobalt on the intensity of a magnetic field, calculated from experimental data concerning passing of electromagnetic waves.

3. Conclusions

In the given work electromagnetic properties of 3D-nanocomposites on the basis of an opal matrix with introduced in interspherical emptiness nanocorpuscles of ferrite-spinel are studied [12, 13]. Effective interaction of electromagnetic waves of a millimetric range with 3D-nanocomposites, consisting of opal matrixes, and containing both nanocorpuscles nickel-zinc and ferrite manganese-zinc is experimentally shown. Dependence of the microwaves transfer factor on the intensity of a magnetic field is defined by a magnetic resonance in magnetic nanocomposites. It is established, that spectra of a magnetic resonance contain an acoustic branch, as well as separate resonances out of this branch are fixed. Interaction of electromagnetic waves of a millimetric range in rectangular resonators and in a wave guide operating in the mode TE10, with the specified opal matrixes is studied in detail. Frequency dependence of the passage and reflexion factors on nanocomposites in the absence of an external magnetic field is measured. It has been established, that in the range of frequencies from 26 to 38 GHz, the reflexion factor as a whole decreases, and the passage factor as a whole increases when increasing the wave frequency. The absorbed share in the nanocomposite sample without an external magnetic field makes capacities from 5 to 20 %. The theoretical analysis of changes of passage and reflexion factors in a magnetic field is carried out. The obtained results create preconditions for working out the high-frequency devices operated by a magnetic field whose work will be based on the use of a microwave magnetic resonance in magnetic nanocomposites on the basis of opal matrixes. Such devices are structurally enough simple and can be quite effective in operation. In the work, it is established that to achive the greatest changes of a microwave signal it is necessary to carry out orientation of the fields $H \perp H_{\sim}$. The considered materials can find their application in the creation of operated attenuators, phase shifters and other devices of a millimetric range.

The work is carried out under the support of grant 12-07-12030-ofi_m and 13-02-90633–arm_a.

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МАГНІТНІ МЕТАМАТЕРІАЛИ ЯК ПЕРСПЕКТИВНІ МАТЕРІАЛИ РАДІОЕЛЕКТРОНІКИ

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Досліджено електромагнітні властивості тривимірних нанокомпозитних матеріалів на основі опалових матриць, які містять частинки одного або двох перехідних металів. Проведений фазовий аналіз нанокомпозитів. Мікрохвильові вимірювання здійснені в частотному діапазоні 26–38 ГГц. Отримано польові залежності коефіцієнтів проходження й відбивання. Відновлено спектри магнітного резонансу й антирезонансу, а також отримано частотні залежності їхніх амплітуд. Встановлено, що в нанокомпозитах, що містять частинки двох перехідних металів, амплітуда магнітного резонансу набагато більша, ніж у нанокомпозитах, що містять частинки одного металу.

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