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ON THE NEW TYPES OF COMPOSITE METAMATERIALS

This paper is the review of our study published in J Appl Phys **115**, 233509 (2014) and J Mater Sci **53**, 2034 (2018). In it, we examined the possibility of fabricating the metamaterial, which is both gyrotropic and of the simultaneously negative permittivity and permeability. Our idea was to use the three-component mixture of ingredients, where one of them is responsible for the negative effective permeability μ_{eff} of hypothetical metamaterial, while all three are responsible for the negative value of effective permittivity ε_{eff}. At first we considered the following composite: the first component was the "swarm" of single-domain ferromagnetic nanoparticles, immersed in a mixture of other two, silver and mercury cadmium telluride. Then, as fabrication of the $Hg_{1-x} Cd_x Te$ is related to using mercury which is very poisoning, we tried to exclude this material substituting it by $Pb_{1-x} Sn_x Te$. Additionally, taking into account that silver is relatively expensive material, we have also used Cu and Al particles as the cheaper substitute of it. We have shown by computer simulations that by the proper fitting of the parameters, e.g., the radius of nanoparticles, their magnetic moments, the relative concentration of ingredients, etc., it is possible to obtain the double-negative metamaterial, that is with negative refraction index in a relatively broad range of temperatures and magnetic fields. The last seems to be very promising in terms of practical applications of metamaterials.

Keywords: negative refraction index, three-component gyromagnetic metamaterials, single-domain ferro-magnetic nanoparticles, superparamagntism, Bruggeman approximation

1. INTRODUCTION

In recent years, we have been witnessed of the explosion of interest in a field of research, which is termed as metamaterials. This area of research is characterized by an exponential growth of a number of publications and it is a hopeless task to refer to even the most important of them in a short Introduction. However, to mention just a few, there are two monographs [1, 2] and the references therein. According to Ref. [1], the term "metamaterials" can be used in a more general, as well as in a more specific sense. In the more general sense, these are materials possessing "properties unlike any naturally occurring substance" or simply "not

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observed in nature." More specifically, these are the materials with a negative refractive index, whose existence and properties were discussed for the first time by Veselago [3]. Many of the researchers apply the term ''metamaterials'' to composites which contain inclusions of certain resonance properties and characteristic sizes of less than the wavelength, such as highly conducting needles, split rings, spirals. In 1967 V. Veselago [3] considered theoretically the medium, which has simultaneously negative real parts of permittivity and permeability, Re [ε] and Re [μ], respectively. Veselago himself called these materials 'left-handed'; left-handedness here refers to the fact that, when the refractive index is negative, the electric field vector E, the magnetic field vector H, and the wave-vector k of a plane wave make a left-handed triad. Since the term left-handedness sometimes is confused with chirality, it is not universally accepted among the researches. In the nice paper [4] Agranovich briefly discussed how people came to the understanding of the phenomenon and clearly pointed out that negative refraction occurs at the interfaces as a natural consequence of the negative group velocity of waves propagating in one of the media. It is worth mentioning that there is no unanimity as for the term negative group velocity materials also. Some authors prefer the term negative phase velocity materials. This is because in case of such materials the phase velocity and group velocity are directed against each other and which direction is positive and which is negative is the matter of convention. The results of Veselago on the other hand, confirmed that this type of medium has to have the negative refractive index, and thereby could exhibit a lot of extraordinary optical properties. The necessary requirement for the material to become negative refractive index material, as it was shown by Veselago and others is the negativity of both the real part of permittivity and real part of permeability, that is why we decided to use in this paper the term double-negative metamaterial (DNMM). Despite the theoretically envisaged possibility for this type of materials to exist, there was no experimental evidence of metamaterials occurring in nature. In the end of 1990s, Smith et al. [5] and Pendry with co-workers [6] published the seminal papers, in which they have shown that these types of materials can be produced in totally artificial way in laboratory.

It is interesting to note that already in his very first publication, Veselago anticipated that negative refraction index materials (or negative phase-velocity (NPV) materials) could be searched among gyrotropic materials. Optical activity, which is the ability of the medium to rotate the polarization plane of electromagnetic waves, has always been a phenomenon of great importance to many areas of research. Mackay and Lakhtakia, [7] Pendry [8], and Tretyakov [9] predicted that a strong optical activity may result in the negative refraction. Consequently, artificial gyrotropic media have started to attract a lot of attention as potential candidates for achieving negative refraction [10–12].

It is worth mentioning that most of the proposed ever since designs of metamaterials were characterized by ever increasing sophistication of fabrication methods. Contrary to these, in our previous work [13], we proposed a relatively simple way to produce metamaterial using the mixture of three ingredients, where the one was responsible for the negativity of Re [$\mu(\omega)$] and the other two for the negativity of Re [$\varepsilon(\omega)$].

This paper comprises an excerpts of our papers published previously [13, 14], in which we addressed the issue of the possibility to fabricate three-component artificial composite gyrotropic metamaterial and demonstrate by numerical simulations, what are the domains (relative to the concentrations of individual components as well as to other important parameters) of its existence.

2. THE MODEL

Let us assume that we have a mixture of three materials, each having granular or powder form, such that the grain sizes are much smaller than the electromagnetic wavelength propagating in the medium. We are trying to match the properties of ingredients in such a way that ε, the dielectric permittivity of the composite would be determined by the three components, while μ, magnetic permeability mainly by one of them responsible for the magnetic properties of the mixture. Let us start from the third ingredient mentioned above, which by the assumption should determine the effective permeability of the hypothetical material and suppose it to be metal magnetic nano-particles (or grains; we shall use these two words interchangeably). We treat these metallic grains as immersed or dispersed in a weakly conducting matrix. If the metallic particles are supposed to be single-domain, then we can take into account only the orientational alignment of their intrinsic magnetic moments and do not need to take into account their induced magnetic moments, as it can be easily proved (see Ref. [15], Chap. 82).

The size of the single-domain particle depends on the material and contributions from different anisotropy energy terms. If we assume nano-particle shape to be spherical, then typical values for the critical radius *a* are about 15 nm for Fe and 35 nm for Co, for g – Fe_2O_3 it is about 30 nm, while for $SmCo_5$, it is as large as 750 nm [16]. Now we can treat the suspension of metallic grains as a kind of “frozen paramagnetic macromolecules,” where the metallic nano-particles play the role of “macromolecules.” The magnetic moments of these single-domain nano-particles at room temperature are distributed at random and we can describe their behavior in the framework of Langevin theory of paramagnetism. Note that the “swarm” of magnetic nanoparticles immersed into another medium was already considered in scientific literature and even the term for describing this situation was already coined, namely, superparamagnetism [17]. The point is that such system behaves like a paramagnet, with one notable exception that the independent moments are not that of a single atom, but rather of a single-domain ferromagnetic particle, which may contain more than 10^5 atoms.

Thus, in the external magnetic field $\vec{H}_0$, an averaged magnetic moment of the unit volume of such medium is equal

$$\vec{M}_0 = \chi_0 \vec{H}_0 \tag{1}$$

where $\chi_0 = (N\mu^2_0)/3k_BT$; N, k_B are the concentration of magnetic nano-particles and Boltzmann constant, respectively, and T is a temperature. Suppose now that we put this hypothetical material in an external magnetic field $\vec{H} = \vec{H}_0 + \vec{h}(t)$, where the second term is the time dependent magnetic field of the electromagnetic wave, propagating in the medium.

Then, the magnetization of the medium can be represented as $\vec{M} = \vec{M}_0 + \vec{m}(t)$, where the second term arises due to time dependent component of the magnetic field and the equation
of motion is of the form

$$\frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{H} \tag{2}$$

where γ is the gyromagnetic ratio. Supposing $h(t) << H_0$ and $m(t) << M_0$ one can make the linearization of Eq. (2) and after some algebra (see for details [1]) one arrives at

$$\vec{m} = \chi \vec{h}_t - \vec{G} \times \vec{h}_t \tag{3}$$

where

$$\chi = \chi_0 \frac{\omega_0^2}{2i\Gamma}\left(\frac{1}{\varpi_1 - \omega} - \frac{1}{\varpi_2 + \omega} \right),$$

$$G = \chi_0 \frac{\omega_0^2}{2i\Gamma}\left(\frac{1}{\varpi_1 - \omega} - \frac{1}{\varpi_2 + \omega} \right) H_0 \; .$$

Here $\vec{h}_t$ is the component of $\vec{h}(t)$ perpendicular to $\vec{H}_0$, vectors $\vec{H}_0$ and $\vec{G}$ in Cartesian coordinates are of the form $\vec{H}_0$ =(0, 0, H_0), $\vec{G}$ = (0, 0, G), $\Gamma = \tau^{-1}$ (τ is relaxation time), $\varpi_1 = -i\Gamma + \sqrt{\omega_0^2 - 2\Gamma^2}$, $\varpi_2 = i\Gamma - \sqrt{\omega_0^2 - 2\Gamma^2}$, ω - is the frequency of the electromagnetic wave incident of the medium and $\omega_0 = \gamma H_0$. Using Eq. (3), one can write down the next formulae for m_x and m_y, components of $\vec{m}$:

$$m_x = \chi h_x + iGh_y \; , \; m_y = \chi h_y - iGh_x$$

Introducing the tensor

$$\chi_{\alpha\beta} = \begin{pmatrix} \chi & iG & 0 \\ -iG & \chi & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

one can rewrite the above expression (3) as

$$m_\alpha = \chi_{\alpha\beta} h_\beta \ , \ \alpha, \beta = \{x, y\}$$

The conditions $\chi_{\alpha\beta} = \chi^*_{\alpha\beta}$ and $\chi_{\alpha\beta}(\vec{H}_0) = \chi_{\beta\alpha}(-\vec{H}_0)$ mean that the medium is gyrotropic [15]. Following the line of reasoning of Ref. [13], since our material is gyrotropic one, we can write down the formulae for the magnetic susceptibilities as follows:

$$\chi_\pm = \chi \pm G \ ,$$

$$\chi = \frac{\chi_0 \omega_0^2}{\omega_0^2 - 2i\omega\Gamma - \Gamma^2 - \omega^2}, \ G = \frac{\chi_0 \omega\omega_0}{\omega_0^2 - 2i\omega\Gamma - \Gamma^2 - \omega^2}$$

These two susceptibilities correspond to two possible circular polarizations of electromagnetic waves which are the eigenmodes of the gyrotropic medium.

In general, the propagation of light in gyrotropic medium is rather complicated and for the medium characterized by the magnetic susceptibility tensor mentioned above it will be considered elsewhere. But the situation becomes more lucid and simple when the wave vector of light is aligned along external magnetic field. Then it turns out, that in such medium two circularly polarized waves with $\vec{k} \parallel \vec{H}_0$ propagate which are characterized by two refraction indices. Thus, for this alignment we can consider the medium as isotropic except the two waves propagate in it.

Then, the expressions for real and imaginary parts of these magnetic susceptibilities, (χ_+ and χ_-) are of the form:

$$\mathrm{Re}\,[\chi_+] = \frac{\chi_0\omega_0\,(\omega_0^2 - \Gamma^2 - \omega^2)(\omega_0 + \omega)}{(\omega_0^2 - \Gamma^2 - \omega^2)^2 + 4\omega^2\Gamma^2},$$

$$\mathrm{Im}\,[\chi_+] = \frac{2\chi_0\omega_0\omega\Gamma(\omega_0 + \omega)}{(\omega_0^2 - \Gamma^2 - \omega^2)^2 + 4\omega^2\Gamma^2},$$

$$\mathrm{Re}\,[\chi_-] = \frac{\chi_0\omega_0\,(\omega_0^2 - \Gamma^2 - \omega^2)(\omega_0 - \omega)}{(\omega_0^2 - \Gamma^2 - \omega^2)^2 + 4\omega^2\Gamma^2},$$

$$\mathrm{Im}\,[\chi_-] = \frac{2\chi_0\omega_0\omega\Gamma(\omega_0 - \omega)}{(\omega_0^2 - \Gamma^2 - \omega^2)^2 + 4\omega^2\Gamma^2}.$$

The idea of the subsequent calculations (see for details Ref. [13, 14]) is the following. First, the 'swarm' of magnetic nanoparticles can be treated as paramagnetic in the frame of Langevin theory of paramagnetism, because in the absence of an external magnetic field their magnetic moments are distributed at random. Being placed in magnetic field, magnetic moments of individual grains, treated in terms of classical physics start to precess, that is why the frequency domain where Re [$\mu(\omega)$] < 0 appears in the vicinity of resonance ($\omega_0 \approx \omega$). Second, being in external magnetic field the whole ensemble behaves as gyrotropic material. Third, the magnetic moments of single-domain ferromagnetic nanoparticles are huge in comparison with atomic magnetic moments, that is why the 'swarm' of ferromagnetic nanoparticles forms 'super-paramagnetic'. As a result, the frequency ω_0 can be made greater in order to look for such frequency domains, where the dielectric permittivity and magnetic permeability are simultaneously negative.

As it was mentioned in the Introduction, in Ref. [13] for one of the mixture components was chosen $Hg_{1-x}Cd_xTe$ semiconductor compound. The reasoning behind this choice was the following. The electrical properties of this material are crucially dependent on cadmium concentration x. If $x = 0$, that is in case of HgTe, the material is semimetal with energy gap $E_g < 0$, while in case of $x = 1$ (CdTe) material becomes semiconductor with wide energy gap of about 1.5 eV at 300 K. Thus, changing the concentration of cadmium, one can change the energy gap, and hence the concentration of electrons. In terms of our model, it means that one can pass smoothly and continuously from Lorentz model for dielectric permittivity, where the electrons are almost tightly bounded to Drude model, where they are almost free to move. As a result, cadmium concentration becomes an important parameter of the model; by means of it–among others–one can control the frequency range where the real part of dielectric permittivity can be made negative and forced it to be overlapped with the frequency domain, where magnetic permeability is negative.

3. CALCULATION OF EFFECTIVE PERMITTIVITY AND PERMEABILITY

In our calculations of the effective permittivity and subsequent computer simulation of the hypothetical metamaterial, we consider a mixture consisting of three components: silver, mercury cadmium telluride $Hg_{1-x}Cd_xTe$ (here, x denotes a cadmium fraction), and the third one, which is the aforementioned “swarm” of magnetic nano-particles. We denote the permittivity of each component as $\varepsilon_1(\omega)$, $\varepsilon_2(\omega)$ and $\varepsilon_3(\omega)$, respectively, which depends on the angular frequency ω. It is known that there are several alternative approaches to the description of effective macroscopic characteristics such as the conductivity, permittivity, etc., of the composite media. Among them are Maxwell-Garnett theory also known as Clausius-Mosotti approximation [18-20] and the Bruggeman approximation often called as the effective medium theory [21] (see also Ref. [1]). We choose for our purposes the last one, because the Bruggeman theory is most widely known and used to calculate an effective permittivity of a composite medium. Its main asset is that all ingredients of a mixture by assumption are treated on the same footing in a symmetric way and none of them plays a privileged role. In this approximation, the effective permittivity can be calculated as the root of the following third-order algebraic equation (see Ref. [1] for details):

$$f_1 \frac{\varepsilon_1 - \varepsilon_{eff}}{\varepsilon_1 + 2\varepsilon_{eff}} + f_2 \frac{\varepsilon_2 - \varepsilon_{eff}}{\varepsilon_2 + 2\varepsilon_{eff}} + f_3 \frac{\varepsilon_3 - \varepsilon_{eff}}{\varepsilon 3 + 2\varepsilon_{eff}} = 0.$$

As we consider a dissipative medium, we always choose the root of Eq. (13), which has a positive imaginary part. Using the permittivity values of particular ingredients (see [13]), we were able to calculate the effective permittivity of the mixture as well as its dependence on the frequency. As for the calculation of the permeability of mixture, we use the following formula:

$$\mu_{eff} = f_1\mu_1 + f_2\mu_2 + f_3(\chi+1) \approx f_{12} + (1 - f_{12})(\chi+1)\,,$$

where $f_{12} = f_1 + f_2$. It can be easily shown that if the permeabilities μ_1, μ_2 of two other non-magnetic components are ≈ 1, which is the case, then this formula follows directly from the formulae obtained in the framework of the Bruggeman theory [22].

4. RESULTS OF COMPUTER SIMULATIONS

Our task now is to find the set of parameters: T – the temperature, $\vec{B}$ – external magnetic field, r – he radius of nano-particles, m – absolute value of the magnetic moment of nano-particles, x – the fraction of Cd in $Hg_{1-x}Cd_xTe$-compound semiconductor, f_1, f_2, and f_3 fractions of the compounds in the mixture, and a range of frequencies for which the following inequalities are simultaneously fulfilled:

$$\mathrm{Re}\,[\mu_{eff}(\omega)] < 0,\ \mathrm{Re}\,[\varepsilon_{eff}(\omega)] < 0,\ \mathrm{Im}\,[\mu_{eff}(\omega)] > 0,\ \mathrm{Im}\,[\varepsilon_{eff}(\omega)] > 0.$$

As one can see, we have seven independent parameters to be controlled at the simulations and hence, a vast enough searching space, regardless the fact that all of them are bounded from above as well as from below. Our strategy can be briefly outlined as follows: first, we search for the range of frequencies where Re [μ(ω)] is negative and then we check if for the same frequency interval Re[ε(ω)] is negative The process of selecting the values of the model parameters are carried out in several sequential steps (for details, see [13]).

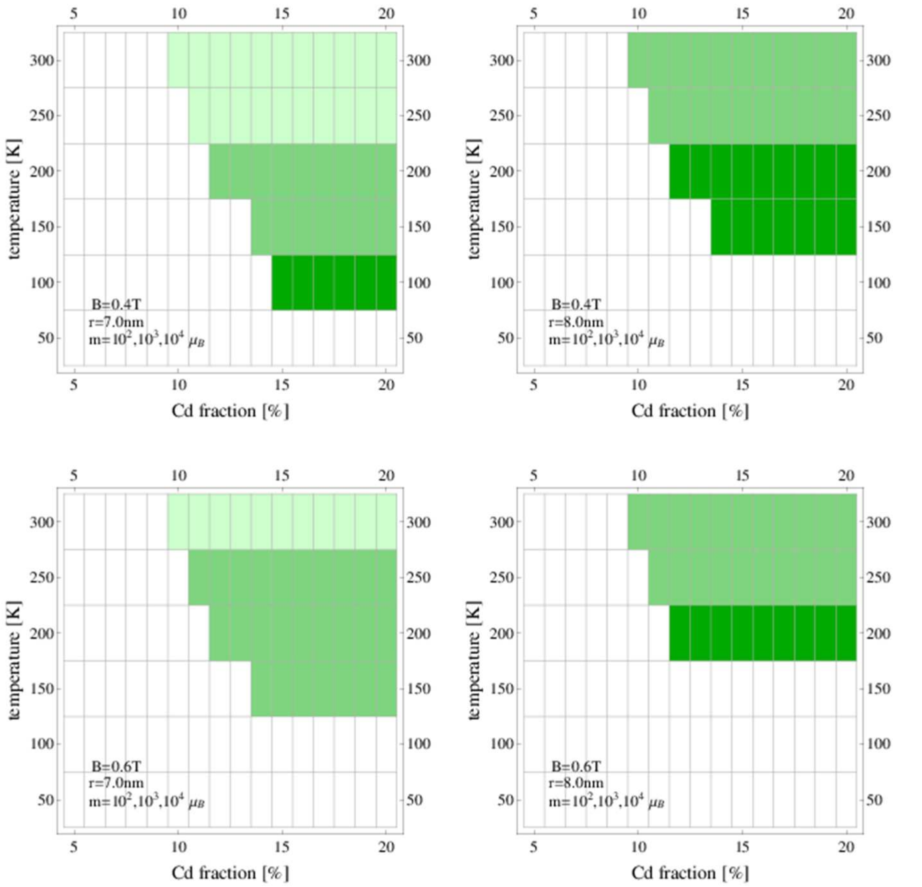

Fig. 1. The regions colored in different shade of green in *T–x* plane and for the different values of model parameters. *B*, *r*, and *m* (see in the main text) correspond to the domains in which at least one set of the filling factors f_1, f_2, and f_3 exists, such that the resulting mixture possesses Re [n] < 0

Figures 1–5 illustrate the results of our simulation. Figure 1 presents in *T–x* plane, for different parameter values of *B*, *r*, and *m*, the domains where at least one set of the filling factors f_1, f_2, and f_3 exists, such that the resulting mixture has the real part of the refractive index less than zero. The different colors have the following meaning: denote by D_1 the region colored in dark green, the domain colored in the intermediate shade of green as D_2, and the region colored in light green as D_3. Then, D_1 corresponds to $m = 10^2$ μ_B, $D_1 \cup D_2$ corresponds to m = 10^3 μ_B and $D_1 \cup D_2 \cup D_3$ corresponds to $m = 10^4$ μ_B, where μ_B – Bohr magneton. We observe that for the greater values of nano-particle magnetic moment, the existence domain of metamaterial becomes larger.

The similar results are presented in Fig. 2 and Fig. 3, where different parameter values of *B*, *r*, and *m*, correspond to the domains where at least one set of the filling factors f_1, f_2, and f_3 exists, such that the resulting mixture has the real part of the refractive index less than zero. Now however, $Hg_{1-x}Cd_xTe$ is substituted in a mixture by $Pb_{1-x}Sn_xTe$ and silver is substituted by Al. The different colors have the following meaning: denote by D_1 the region colored in yellow, the domain colored in orange as D_2, and the region colored in red as D_3. Then, D_1 corresponds to $m = 10^2$ μ_B, $D_1 \cup D_2$ corresponds to $m = 10^3$ μ_B and $D_1 \cup D_2 \cup D_3$ corresponds to $m = 10^4$ μ_B.

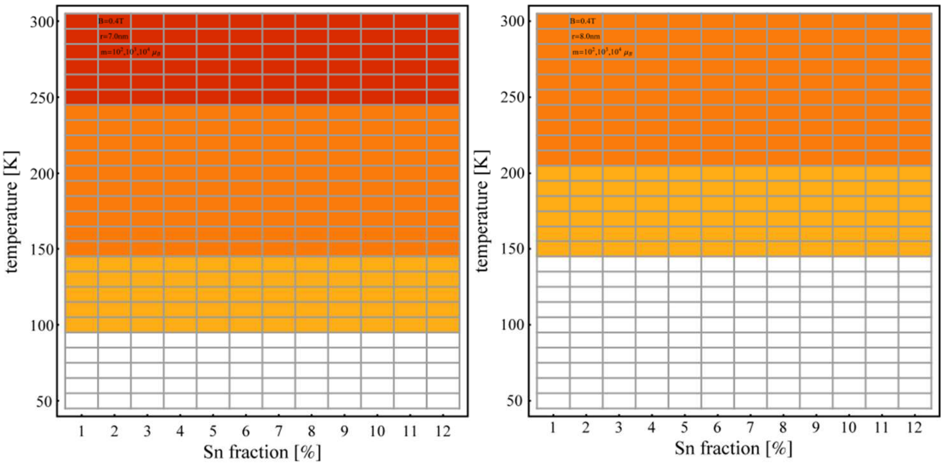

Fig. 2. The regions colored in red, orange and yellow (see text) in *T* - *x* plane and for the different values of model parameters for the mixture of $Pb_{1-x}Sn_xTe$, ferromagnetic nanoparticles and Al particles. *B*, *r* and *m* correspond to the domains in which at least one set of the filling factors f_1; f_2; f_3 exists, such that the resulting mixture possesses Re [n] < 0

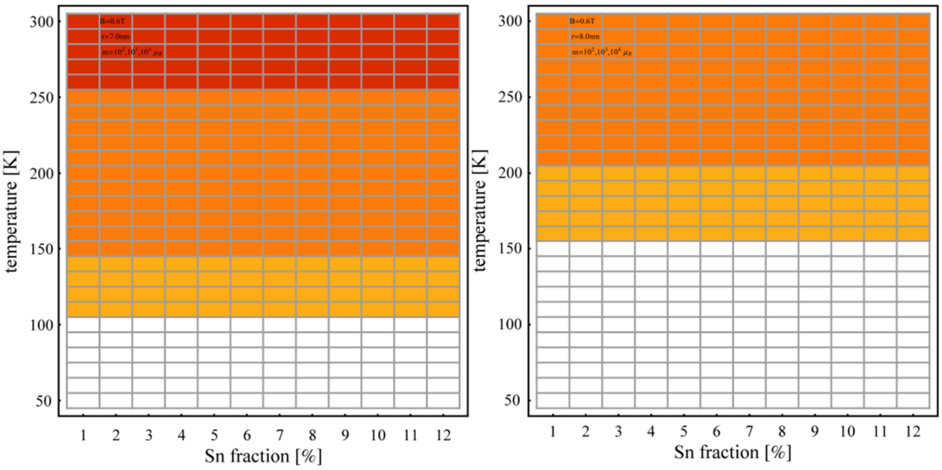

Fig. 3. The same as in Fig. 2, but for different values of *B*

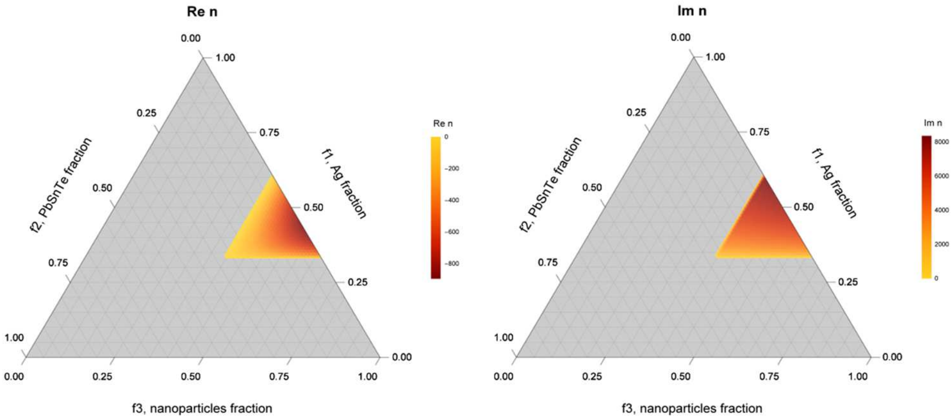

Fig. 4. Real and imaginary parts of the refractive index of the composite for selected cases presented on the ternary diagram. On each axis of the triangle, the relative contribution of individual components of the composite are indicated, which are expressed as the values of the filling factors f_1; f_2; f_3. The model parameters are : B = 0.2 T, r = 8 nm, $m = 10^4$ μ_B, T = 300 K, x = 0.08

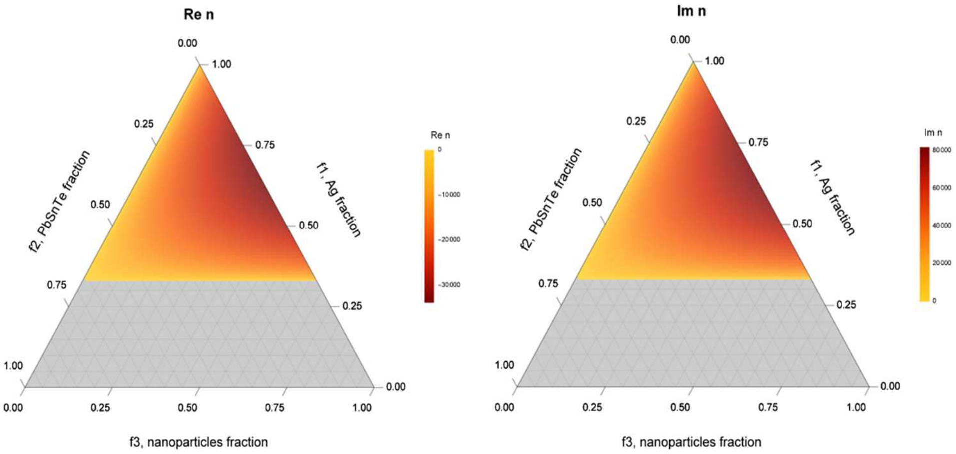

Fig. 5. The same as in Fig. 4, but for the model parameters $B = 0.4$ T, $r = 8$ nm, $m = 10^4\ \mu_B$, $T = 300\ K$, $x = 0.08$

5. CONCLUSIONS

Up to now many metamaterial designs leading to a negative refraction index were proposed by the researchers. In 2000, it was shown experimentally that a metamaterial composed of periodically positioned scattering elements, all conductors, could be interpreted as showing simultaneously negative ε and μ in some frequency domain. An experimental observation of the negative refraction was reported for a metamaterial composed of wires and split-ring resonators deposited lithographically on the circuit board material [23]. All of the proposed ever since designs have their advantages as well as disadvantages, but it would be perhaps not an overstatement to say, that they are characterized by ever-increasing sophistication of fabrication methods. On the contrary, in our work [13], we proposed a comparatively simple way to produce a material which is at once gyrotropic and of negative-phase velocity. The idea is to make a mixture of three ingredients, where one of them would be responsible for the negativity of μ, while all three would be responsible for the negativity of ε. The first component of the mixture is the “swarm” of single-domain ferromagnetic nano-particles, immersed in a mixture of other two, silver and mercury cadmium telluride. The choice of silver is determined by the fact, that as it was shown, the permittivity of a mixture of silver and a dielectric material can be negative in some frequency domain. The other argument in favor of using silver is that it is a diamagnetic material. It means that considering the “swarm” of single domain ferromagnetic nano-particles suspended in a mixture containing silver, we can neglect the interaction between their magnetic moments and treat the whole mixture as superparamagnetic. The choice of mercury cadmium telluride is determined by the remarkable dependence of its energy gap on the fraction of cadmium in the compound. In its turn, it leads to the strong dependence of the electron concentration on this fraction as well as on the temperature. It enables to adjust to each other two frequency domains where ε < 0 and μ < 0 and make them simultaneously negative. In [13] , we carried out computer simulations in the frame of the proposed model in order to establish the domains of existence of such material, searching through a vast parameter space. We have seven parameters to be controlled in course of simulations, these are the temperature, external magnetic field, radius of nano-particles, their magnetic moments, fraction of cadmium in $Hg_{1-x}Cd_xTe$-compound, and the relative concentrations f_1, f_2, and f_3 of the components in a mixture. In total, there are eight parameters, but due to the relation $f_1 + f_2 + f_3 = 1$, only seven of them are independent. Despite as it seems, a relative simplicity of fabrication, if produced, such a metamaterial will show its disadvantages or limitations in use, too. First of all, a negative refraction can be achieved only if the material is in an external, although moderate magnetic field. In our calculations, B was restricted to ~ 0.8 T. On the other hand, in some circumstances it could be an advantage, since switching magnetic field on and off, one can trigger NPV-state on/off.

Another disadvantage is the use of $Hg_{1-x}Cd_xTe$, since its fabrication is related to using mercury, which is very poisoning. So, in the work [14] we substituted $Hg_{1-x}Cd_xTe$ by $Pb_{1-x}Sn_xTe$ and silver by Al and Cu, since they are also diamagnetic. On the other hand, silver is relatively expensive, while aluminum and copper are cheaper and have very similar conducting properties as silver. By computer simulations, we establish the domains where the material becomes negative refraction index material relatively to all parameters characterizing the mixture.

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NOWY TYP KOMPOZYTOWYCH METAMATERIAŁÓW

Praca stanowi przegląd prac autorów, opublikowanych wcześniej (*J Appl Phys* **115**, 233509 (2014) oraz *J Mater Sci* **53**, 2034 (2018)). W w/w pracach autorzy zbadali możliwość utworzenia metamateriału, który byłby jednocześnie giroskopowy oraz posiadał ujemne części rzeczywiste przenikalności magnetycznej i dielektrycznej. Ideą jest wykorzystanie w tym celu mieszanki trzech składników, z których pierwszy - to "rój" ferromagnetycznych nano- cząstek

Słowa kluczowe: ujemny współczynnik załamania, trójskładnikowy metamateriał giromagnetyczny, jednodomenowe ferromagnetyczne nano- cząstki, superparamagnetyzm, przybliżenie Bruggemana.

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