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The effect of optical non-reciprocity in multilayer nanostructure systems based on Bi

© A.M. Shadrin, D.D. Noskova, F.A. Pudonin, I.A. Sherstnev¶, A.P. Boltaev

Lebedev Physical Institute, Russian Academy of Sciences,
Moscow, Russia

¶ E-mail: shersntevia@lebedev.ru

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In multilayer non-magnetic structures with island layers Bi ($[Bi-Al_2O_3]_N$ — at room temperature, an abnormally large optical non-reciprocity effect was detected. It is shown that this effect can be closely related to the behavior of dielectric permittivity in Bi nanostructured layers. It is found that in these multilayer island systems, the metallic nature of the optical response of the dielectric constant ($Re(\varepsilon) < 0$) is observed < 0) in the optical range when the thickness of the dielectric layer of Al_2O_3 is less than 1.6 nm. It is assumed that the island systems $[Bi-Al_2O_3]_N$ exhibit the properties of „left" metamaterials with negative μ and ε in the optical range.

Keywords: insular film, thin film, percolation, exchange interaction, permittivity.

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1. Introduction

Nowadays, various chiral systems are being extensively studied. Chirality is the absence of symmetry between right and left sides of the object. Chiral objects include, for example, DNA molecules, Dirac semimetals, some metamaterials, etc. [1]. Chiral objects also include some two-dimensional metallic (magnetic) nanostructures. One of the chirality manifestations is the effect of optical nonreciprocity, that is when, for example, the polarization plane of the reflected light rotates by different angles when the direction of the incident light on the object is reversed. This effect is due to breaking of the time-reversal symmetry [2]. Previously, the optical nonreciprocity effect has been found in magnetic metal nanoparticles with vortex type of magnetization, where the same vortex direction was set for all particles by an external magnetic field. In this case, the effect of nonreciprocity is associated with the unequal scattering of light when the magnetic vortex direction changes [3–5]. The magnitude of this effect is of the order of magnitude of the magnetooptical Kerr effect (about $1-2$ arc minutes). Recently, an anomalously large (up to 2 arc degrees) effect of optical nonreciprocity has been found in $[FeNi-Al_2O_3]_7$ multilayer systems of magnetic nanoislands at room temperature [6]. This effect has been assumed to be associated with the formation of a supervortex type of magnetization in multilayer magnetic nanoisland systems [7–9]. The effect disappeared when the FeNi films became continuous. It is important to note that the magnetic nanoislands in these structures are almost round in shape [6], therefore, the nonreciprocity effect was not related to the anisotropy of the geometric shape of islands. Also, the nonreciprocity effect in these nanoisland systems did not depend in any way on the applied external magnetic field of up to $\sim 1.5\,T$, in contrast to what was observed, for example, for specially shaped Co or CoFe nanoparticles [3]. Such independence on magnetic field can be associated with the presence of some additional, yet unknown, mechanism that is not associated with vortex or supervortex magnetization of the magnetic nanoisland system. This means that the optical nonreciprocity effect can also be observed in multilayer systems of nonmagnetic nanoislands. We believe that in this case the nonreciprocity effect can be associated with the permittivity features in nonmagnetic island systems [10]. Also, there are other systems, where nonreciprocity is independent from magnetic field, for example [11–13]. This work presents the results of studies of the optical nonreciprocity effect and the permittivity in multilayer structures of diamagnetic $Bi-[Bi-Al_2O_3]_N$ nanoislands.

2. Measurements and results

Bismuth is a diamagnetic material ($\mu < 0$) with a strong spin-orbit interaction. However, with a critical thickness of ~ 30 nm Bi can transit from the semimetallic state to the semiconductor state [14]. Also, surface metallic states can exist in ultrathin Bi(001) films, which has been confirmed by broadband terahertz spectroscopy studies [15] and spectral ellipsometry studies [16].

The percolation threshold is a critical film thickness, below which the film has island structure during its growth and above which it becomes continuous. To determine the percolation threshold for Bi, a series of Bi films of various thicknesses were deposited by the RF-sputtering method. Two series of multilayer structures were grown to study the optical nonreciprocity effect and permittivity features. In the first series of $[Bi-Al_2O_3]_7$ structures, the effective thickness of Bi layers varied in the range of $0.8-2.6$ nm with the unchanged thickness of Al_2O_3 layers of ~ 3 nm. In the second series, the thickness of Bi island layers was fixed at~ 1.4 nm and the thickness of Al_2O_3 layers varied from 0.7 to 5 nm. All structures were grown with a single process. Layer deposition rates were determined immediately before the deposition of structures and were 14.3 nm/min for Bi layers and 7 nm/min for dielectric layers of Al_2O_3. The effective layer thickness was determined by the deposition time with an accuracy of not worse than 5%. The process of such multilayer structures growing was similar to that described previously in [6,16]. Polished ceramic plates (sitall) with TiO_2 (rutile [17]) as their main component were used as substrates. Roughness of the substrates was about $1-3$ nm [18]. All the grown structures were covered with a protective layer of Al_2O_3 with a thickness of ~ 2 nm to protect them against the external atmosphere.

Detailed microscopic, atomic force and X-ray studies of similar multilayer systems with Bi island layers have been previously presented in [16]. Fig. 1 shows SEM-image of the $[Bi(1.4\,nm)-Al_2O_3(1.6\,nm)]_7$ island multilayer structure.

Previously [8,16,17,19] it has been shown by X-ray reflection methods that multilayer structures composed of alternating island metal layers are indeed periodic systems, and the cross-section of such structures [19] demonstrates no noticeable mutual diffusion of structure layers.

The percolation threshold for Bi was determined as a thickness of d^*, that films with a thickness of $d < d^*$ have metal type of their temperature dependence of conductance, and films with a thickness of $d > d^*$ have island type of their temperature dependence of conductance, when the conductance is of dielectric nature. Previously, we have suggested a method to determine d^* of metal films [20] also based on the fact that island metal films are characterized by $\mathrm{Re}(\varepsilon) > 0$, i.e. the optical response is of dielectric nature, however, films with metal type of conductivity have $\mathrm{Re}(\varepsilon) < 0$. $\mathrm{Re}(\varepsilon)$ was determined by ellipsometry method in the wavelength range of $\lambda = 0.4-1.1\,\mu$m [21] (angle of light incidence on the sample was 70°). Fig. 2 shows dependence of $\mathrm{Re}(\varepsilon)$ on the Bi film thickness at a wavelength of $\lambda = 0.6\,\mu$m. It can be seen that $d^* \approx 3.9$ nm for Bi films.

Usually, to study the nonreciprocity effect, coefficients of polarized light reflection from the structure are compared for opposite directions of incidence. If these coefficients are not equal to each other, the optical nonreciprocity effect takes place. Precision measurement requires, for example, that the incident light intensity remains unchanged throughout the entire process of measurement, which is difficult to implement in many cases. Therefore, in this study, as well as in [6], the change in the rotation angle of the polarization plane $\Delta\omega$ was measured for the reflected p-polarized light, while the sample rotated about its axis by 180° from the initial position. This method is less sensitive to variations of the incident light intensity on the structure. To observe the optical nonreciprocity effect,

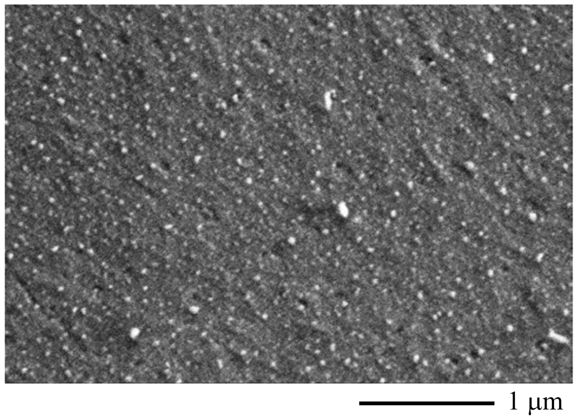

Figure 1. Scanning electron microscopy of a typical multilayer structure of $[Bi-Al_2O_3]_7$ on sitall substrate.

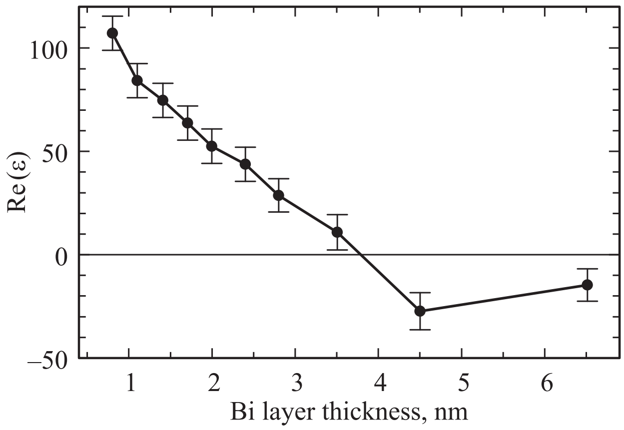

Figure 2. Dependence of $\mathrm{Re}(\varepsilon)$ on the thickness of layer for a single-layer Bi film.

a modified laser ellipsometer (LEF-2, $\lambda = 632.8$ nm) was used. The sample was placed on a goniometer table and could be rotated in its plane. The P-polarized light irradiated the sample at an angle of 70°, and the reflected light from the structure passed through the second polarizer (the analyzer) and then it was received by a photomultiplier. With the sample rotation about its axis (φ), the analyzer angle (ω) was determined at a minimum signal on the photomultiplier. This angle was measured from the s-position. Then the $\Delta\omega = \omega(\varphi) - \omega(\varphi + 180^\circ)$ was calculated, i.e. the difference between polarization angles for opposite directions of light incidence (rotation of the sample by 180°). If $\Delta\omega = 0$, then there is no nonreciprocity effect, if $\Delta\omega = 0$, then the optical nonreciprocity effect takes place in the system. Fig. 3 shows $\Delta\omega(\varphi)$ dependence for different Bi layer thicknesses. It can be seen that for the structures with island type of Bi layers and a dielectric layer thickness of ~ 3 nm the optical nonreciprocity effect takes place with a magnitude not more than 2, which is above the $\Delta\omega$ for magnet island systems [6]. The performed studies of magnetooptical Kerr effect have shown that these structures

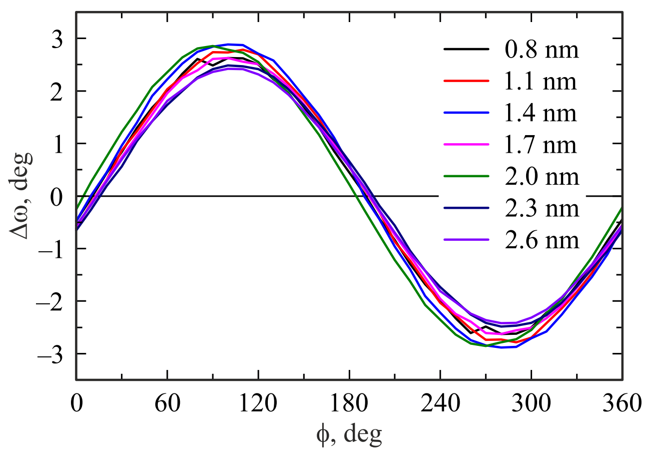

Figure 3. The magnitude of nonreciprocity $\Delta\omega$ for $[Bi{-}Al_2O_3]_7$ as a function of sample rotation φ for different thicknesses of the Bi layer.

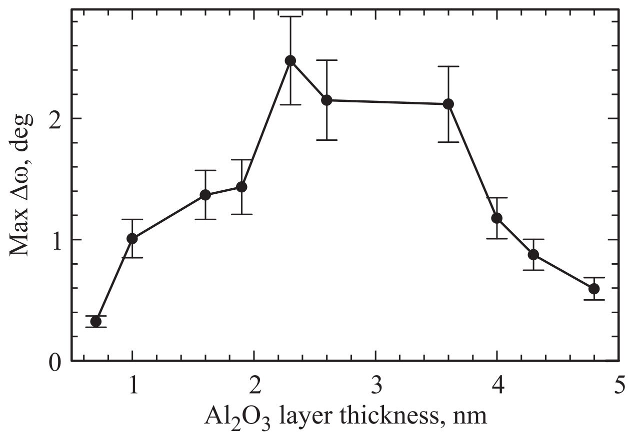

Figure 4. Maximum nonreciprocity $\Delta\omega$ for $[Bi{-}Al_2O_3]_7$ depending on the thickness of Al_2O_3 layer.

are nonmagnetic. Thus, an optical nonreciprocity effect with anomalously large magnitude is found in structures with island-type Bi layers, when there are neither vortex or supervortex magnetizations nor island shape anisotropy. It is worth to note that magnitude of the optical nonreciprocity effect was not dependent on external electric or magnetic fields. Physical causes for the emergence of this effect in such structures are not fully understood so far, however, some assumptions can be made.

Thus, previously it has been assumed that $\Delta\omega$ may be affected by features of the permittivity behavior in $[FeNi{-}Al_2O_3]_N$ magnetic island structures [10]. Therefore, an assumption has been made that in the case of structures with island-type Bi layers the emergence of the optical nonreciprocity effect may be related to the behavior of ε. In turn, the ε may be dependent on thickness of Al_2O_3 layers, which determines the degree of interaction between island-type Bi layers in a multilayer structure. Therefore, for $[Bi{-}Al_2O_3]_N$ structures with a fixed thickness of island-type Bi layers (~ 1.4 nm) the dependencies of $\Delta\omega$ on the thickness of Al_2O_3 layers were investigated. Fig. 4 shows graph of$\Delta\omega(d)$, where a strong dependence of the nonreciprocity effect $\Delta\omega$ on the thickness of Al_2O_3 layers can be seen. The nonreciprocity effect nearly disappears at a weak interaction between Bi layers (thickness of $Al_2O_3 > 4$ nm layers) and a strong interaction between Bi layers (thickness of $Al_2O_3 < 2$ nm layers).

We believe that this behavior of $\Delta\omega(d)$ may be related to the features of permittivity behavior depending on the thickness of Al_2O_3 layer. Therefore, the behavior of permittivity was studied with rotation of samples about their axes for the same structures with different thicknesses of the Al_2O_3 layer. For this purpose, the method of spectral ellipsometry was used. Here we used two calculation models. In the model of effective medium (the single-layer model), all the multilayer structure was considered as an effective single-layer medium and its effective value ε

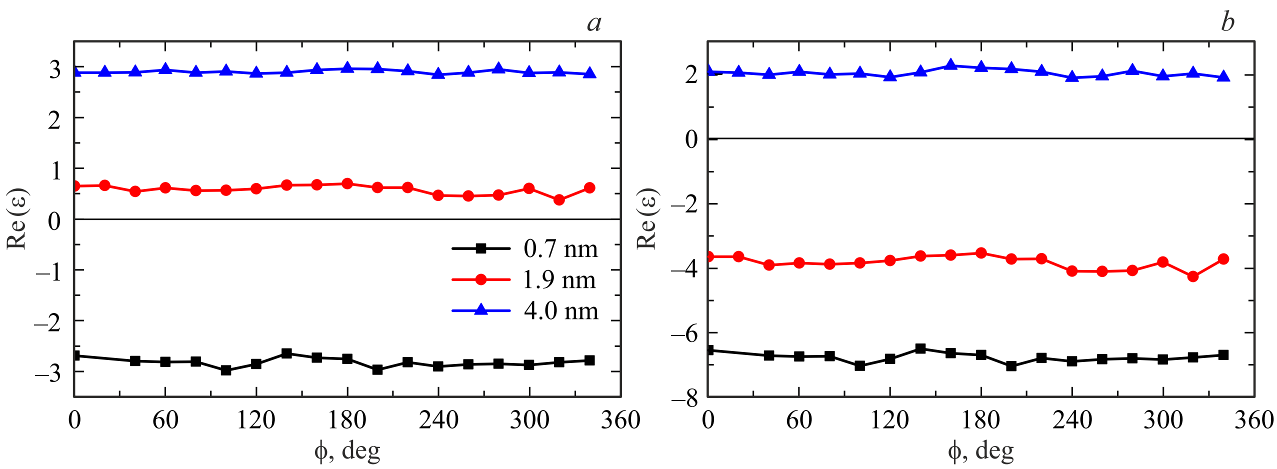

Figure 5. $Re(\varepsilon)$ for $[Bi{-}Al_2O_3]_7$ depending on the rotation of sample ϕ for different thicknesses of the Al_2O_3 layer. Two calculation models: (*a*) single-layer model, (*b*) multilayer model.

was calculated for the optical response of the structure as a whole. This model has already included automatically all the interactions between Bi layers. In the multilayer model, when ε is calculated for each Bi layer, no interlayer interactions are taken into account. Single-layer and multilayer calculations are described in detail in [22].

Fig. 5 shows dependencies of $\mathrm{Re}(\varepsilon)$ on the sample rotation angle ϕ for some thicknesses of Al_2O_3 layers in the approximation of effective medium (Fig. 5, *a*) and multilayer model (Fig. 5, *b*). Calculations are performed for the incident light wavelength of $\lambda = 630$ nm. It has been found that in island-type structures with a thickness of $Al_2O_3 < 1.6$ nm layer $\mathrm{Re}(\varepsilon)$ became negative as it took place in structures with metallic type of conductance. However, the island-type structure itself remains dielectric (thickness below the percolation threshold). With increase in thickness of Al_2O_3 layers the $\mathrm{Re}(\varepsilon)$ crosses the zero level and becomes positive, as it must be for the case of structures with dielectric type of conductance. It should be noted that previously this type of $\mathrm{Re}(\varepsilon)$ behavior has been also found in $[\mathrm{FeNi-Al_2O_3}]_N$ magnetic island structures with a thickness of $Al_2O_3 < 2$ nm [10]. This change in $\mathrm{Re}(\varepsilon)$ sign is observed in the whole spectral range. This behavior is observed for both models. Numerical values are expectedly different between the models, because the result of the multilayer model does not include the dielectric layer. Without the interlayer interaction, the numerical difference between the models can only be proportional. However, a difference in thickness can be seen where $\mathrm{Re}(\varepsilon)$ changes its sign. Hence the interlayer interaction is significant and should be taken into account. Also, it can be seen that there is no significant anisotropy of $\mathrm{Re}(\varepsilon)$

3. Results and discussion

The change in sign of the $\mathrm{Re}(\varepsilon)$ permittivity may be attributed to the size quantization of permittivity in island-type Bi layers in multilayer structures. Thus, previously Keldysh [23] has shown that Coulomb interaction in 2D semimetal layers grows with decrease in layer thickness and depends on the permittivity of media surrounding the semimetal layer. $E \propto 1/(\varepsilon_1 + \varepsilon_2)^2$, where ε_1 and ε_2 are permittivities of the medium above and below the semimetal layer. That is, as thickness of the semimetal film decreases, the effect of permittivity size quantization takes place, which manifests in the decrease in ε of the semimetal layer. In the case of $[\mathrm{Bi-Al_2O_3}]_N$ multilayer structures, effective permittivities of the media above and below each Bi layer, which are composed of several Bi layers and Al_2O_3 layers, will vary depending on the thickness of Al_2O_3 layers. It means, that ε of each Bi layer will change as well depending on the thickness of Al_2O_3 layers. Calculation of the change in permittivity in a multilayer structure is a complicated task and is beyond the scope of this experimental work. More so that further complications may arise from the fact that, for example, calculations in [23] are correct for $4\varepsilon \gg (\varepsilon_1 + \varepsilon_2)^2$, where ε is permittivity of the Bi layer, which may not be true in the structures under study.

As it has been found that magnitude of the nonreciprocity effect is dependent on thickness of Al_2O_3 layers (Fig. 4), an assumption can be made that the change in permittivity of Bi layers depending on the thickness of Al_2O_3 layer can be one of the causes of this effect. Here it is necessary to note that in the case of magnetic multilayer island systems of $[\mathrm{FeNi-Al_2O_3}]_N$ type there is a significant difference between permittivities calculated with single-layer and multilayer models of the effective medium. This means that the exchange interaction between FeNi layers in these systems plays a significant role in the emergence of the nonreciprocity effect.

It is necessary to make another important and interesting note. As Bi is a diamagnetic semimetal, it has diamagnetic permeability $\mu < 0$. At the same time, the permittivity of $[\mathrm{Bi-Al_2O_3}]_N$ structures with a thickness of Al_2O_3 layers less than 1.6 nm becomes negative in the frequency range of visible light. That is, these structures are „left-handed“ (with negative group velocity) metamaterials with simultaneous $\varepsilon < 0$ and $\mu < 0$ [24]. In these materials electric field, magnetic field and wave vector form a left-hand vector system. This feature allows the electromagnetic field to propagate against the phase velocity, that opens opportunities for a number of interesting practical applications [25,26]. It is worth to note here, that $\mu < 0$ is true for massive Bi, however, for island-type layers of Bi it is an assumption that needs further verification [27].

4. Conclusion

Thus, in multilayer structures of nonmagnetic island-type Bi layers an anomalously large optical nonreciprocity effect is found. The magnitude of this effect is significantly affected by thickness d of Al_2O_3 layers: $\Delta\omega$ has its maximum value at a thickness of $d \approx 3$ nm and drops abruptly at thicknesses of $d > 4$ nm and $d < 2$ nm. An assumption is made that the nonreciprocity effect is connected to the features of ε dependence on the thickness d of Al_2O_3 layers (size effect). We also assume that the metal-like optical response ($\mathrm{Re}(\varepsilon) < 0$) at $d < 1.6$ nm is a consequence of this size effect. If the island-type layers of Bi are assumed to remain diamagnetic, then $[\mathrm{Bi-Al_2O_3}]_N$ multilayer structures are „left-handed“ metamaterials.

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Conflict of interest

The authors declare that they have no conflict of interest.

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