LOW-DIMENSIONAL SYSTEMS AND SURFACE PHYSICS

Optical and Magneto-optical Properties of CoFeB/SiO₂ and CoFeZr/Al₂O₃ Granular Magnetic Nanostructures

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Received March 19, 2004

Abstract—The optical and magneto-optical properties of the new granular nanocomposites (CoFeB)/(SiO₂) and (CoFeZr)/(Al₂O₃), which are grains of amorphous ferromagnetic alloys embedded in dielectric matrices, have been studied. The dependence of the optical, magneto-optical, and magnetic properties of the nanocomposites on their qualitative and quantitative composition, as well as on the conditions of their preparation, was investigated. Spectra of the dielectric functions $\epsilon = \epsilon_1 - i\epsilon_2$ were obtained by the ellipsometric method in the range 0.6–5.4 eV. Above 4.2 eV, the absorption coefficient of the (CoFeB)/(SiO₂) composites was found to be close to zero for all magnetic-grain concentrations. The polar Kerr effect measured at a photon energy of 1.96 eV in dc magnetic fields of up to 15 kOe reaches values as high as 0.25°–0.3° for these nanocomposites and depends only weakly on the conditions of preparation. On the other hand, the (CoFeZr)/(Al₂O₃) nanostructures reveal a considerable difference in the concentration dependences of the Kerr effect between samples prepared in a dc magnetic field and in zero field. © 2004 MAIK “Nauka/Interperiodica”.

1. INTRODUCTION

In recent years, there has been an increasing interest in the area of the physics of condensed matter that deals with investigating various structures on the nanometer and even smaller scales. Structures that are made up of grains a few nanometers in size embedded in nonmagnetic metallic or dielectric matrices are materials that border between classical physics of the solid state and nanophysics.

Interest in granular nanostructures has experienced explosive growth due to a number of their physical properties that have scientific significance and also application potential. Among them the magnetoresistance effect [1], inverse or positive magnetoresistance [2], spin-dependent electron tunneling, high dielectric losses in the microwave range [3], the anomalous Hall effect [4], and the large magnetorefractive effect [5] should be mentioned. A number of papers aimed at revealing and investigating possible correlations between the magnetic properties, magnetotransport, and linear and nonlinear optical and magneto-optical properties of granular nanostructures have appeared [5–7]. Particular attention centers presently on the behavior of nanostructures near the percolation threshold, because in this region the most pronounced changes in their characteristics are observed.

By studying the optical properties of materials, one can gain valuable information on their electronic structure, an approach widely employed in optics of the

solid state. Magneto-optical phenomena (in particular, the Kerr and Faraday effects) can also yield information on the magnetic properties of materials. At the same time, certain features observed in the optical and magneto-optical properties of granular nanostructures are still not fully understood [8, 9].

Until recently, most studies of the properties of granular films dealt with crystalline grains of a metal or alloy. Nanocomposites with grains of amorphous materials have attracted less interest [10]. Compared to grains with crystal structure, amorphous grains have a lower anisotropy energy, whose magnitude noticeably affects the main magnetic properties of nanocomposites [11]; in some cases, this influence may become a favorable factor, for instance, for the manifestation of magnetoresistive properties [12]. This accounts for the increasing interest over the past several years in the magnetoresistive characteristics of such structures [2, 12], as well as in their properties in the microwave range [4].

As for the optical and magneto-optical properties of structures with amorphous grains, there are only a few reports on relevant studies. The steady-state and dynamic Kerr effects in (CoNbTa)/(SiO₂) nanocomposites are investigated in [13], where a correlation between the dynamic and steady-state magneto-optical properties of these structures was revealed. A study of the correlations between the magnetorefractive effect and magnetoresistance in (CoFeZr)/(SiO₂) is given in

Parameters of the nanocomposites under study

Series	Grain alloy	Matrix	Grain concentration, at. %	Preparation conditions
I-1	Co ₄₁ Fe ₃₉ B ₂₀	SiO ₂	From 34 to 68	Ar, P _{Ar} = 8.0 × 10 ⁻⁴ Torr
I-2	Co ₄₁ Fe ₃₉ B ₂₀	SiO ₂	From 34 to 68	Ar + N ₂ , P _{N₂} = 1.8 × 10 ⁻⁴ Torr
I-3	Co ₄₁ Fe ₃₉ B ₂₀	SiO ₂	From 34 to 68	Ar + O ₂ , P _{O₂} = 2.8 × 10 ⁻⁵ Torr
II-1	Co ₄₅ Fe ₄₅ Zr ₁₀	Al ₂ O ₃	From 21 to 54	Ar + O ₂ , P _{O₂} = 2.8 × 10 ⁻⁵ Torr
II-2	Co ₄₅ Fe ₄₅ Zr ₁₀	Al ₂ O ₃	From 21 to 54	Ar + O ₂ , P _{O₂} = 2.8 × 10 ⁻⁵ Torr; in-plane magnetic field, forced substrate cooling

[14]. A comparative investigation of the equatorial Kerr effect and magnetoresistance in SiO₂ matrices with embedded FeCoB, CoFeZr, and CoNbTa nanoparticles is discussed in [15].

We report here on a study of granular nanocomposites based on CoFeB and CoFeZr amorphous ferromagnetic alloys. Their optical and magneto-optical properties were investigated over a broad range of concentrations, both below and above the percolation threshold. We also investigated the effect of various conditions of nanocomposite preparation on their properties, namely, the presence of oxygen or nitrogen in the preparation process, the substrate temperature, and the magnetic field. We are not aware of any publications on either the spectra of dielectric functions of such granular structures or the polar Kerr effect.

2. EXPERIMENT

All samples were prepared by ion beam sputtering [10]. Two groups of nanostructures were investigated. Group I included structures based on grains of a Co₄₁Fe₃₉B₂₀ amorphous ferromagnetic alloy embedded in amorphous dielectric SiO₂ films. Nanocomposites of

group II were films of amorphous dielectric Al₂O₃ with embedded nanograins of a Co₄₅Fe₄₅Zr₁₀ amorphous ferromagnetic alloy. The main characteristics of these groups are listed in the table. Samples of the first group differed in that part of them were prepared in an argon environment (subgroup I-1), while the other part was grown in an atmosphere with nitrogen (I-2) or oxygen (I-3). The magnetic-phase concentration varied from 34 to 68 at. %. Samples of the second group were fabricated in an argon or oxygen environment. Some of them were prepared in a dc magnetic field under forced cooling of the substrate (subgroup II-2), and the remainder, in zero magnetic field without forced cooling (I-1). The concentration of ferromagnetic grains in samples of this group varied from 21 to 54 at. %. The nanocomposite film thicknesses ranged from less than 1 μm for the (CoFeB)/(SiO₂) samples to 5–10 μm for (CoFeZr)/(Al₂O₃). The (CoFeB)/(SiO₂) films were deposited on glass substrates, and the (CoFeZr)/(Al₂O₃) films, on ceramic substrates. The ferromagnetic particles grew in size from 2 to 7 nm with increasing content of the magnetic phase. Such grain sizes are less than the typical critical diameters separating the superparamagnetic regime from the regime with blocked particles in granulated structures for specific temperatures and measurement times [11, 16]. Primary characterization of the samples was accomplished by measuring their electrical resistivity. Figure 1 shows the electrical resistivity of (CoFeB)/(SiO₂) structures as a function of grain concentration. This shape of the concentration dependences of electrical resistivity is typical of percolation systems [10].

The optical properties of granular nanocomposites were studied by reflection ellipsometry. The choice of reflection geometry was motivated by the strong absorption in nanocomposites with high grain concentrations. The essence of the method, which is discussed in detail, e.g., in [17], consists in measuring the ellipsometric angles ψ and Δ . The physical meaning of these angles can be readily grasped from the main relation of reflection ellipsometry

$$\tan \psi \exp(i\Delta) = r_{pp}/r_{ss}, \quad (1)$$

Fig. 1. Electrical resistivity of (CoFeB)/(SiO₂) nanocomposites plotted vs. grain concentration.

where $r_{pp}(r_{ss})$ is the amplitude reflectance for light polarized parallel (perpendicular) to the plane of incidence. Assuming the simplest model of a semi-infinite isotropic sample in a medium with a real refractive index of unity (air), the real and imaginary parts of the dielectric function $\epsilon = \epsilon_1 - i\epsilon_2$, as well as the refractive index n and absorption index k $[(n - ik)^2 = \epsilon]$, can be determined from measured ellipsometric angles using the relation

$$\epsilon = \sin^2 \varphi \left[1 + \tan^2 \varphi \left(\frac{1 - r_{pp}/r_{ss}}{1 + r_{pp}/r_{ss}} \right)^2 \right], \quad (2)$$

where φ is the angle of incidence.

The ellipsometric angles ψ and Δ were measured on a spectroscopic ellipsometer at photon energies ranging from 0.6 to 5.4 eV. The spectral responses of the dielectric functions and of the indices of refraction and absorption were calculated from Eqs. (1) and (2).

A magneto-optical investigation of the nanocomposites was performed using the linear magneto-optical Kerr effect in polar geometry and the Faraday effect. These phenomena are of the same nature and consist in rotation of the plane of polarization and the appearance of ellipticity of the light reflected (in the case of the Kerr effect) from a medium magnetized perpendicular to the surface or of the light transmitted through such a medium (in the case of the Faraday effect). The angle of rotation of the plane of polarization is determined by both diagonal and off-diagonal elements of the permittivity tensor of the medium (in the isotropic case) [18]:

$$\hat{\epsilon} = \begin{pmatrix} \epsilon & -i\epsilon_{xy}(M) & 0 \\ i\epsilon_{xy}(M) & \epsilon & 0 \\ 0 & 0 & \epsilon \end{pmatrix}. \quad (3)$$

In particular, the rotation of the plane of polarization in the polar Kerr effect, θ_K , can be found from the relation

$$\theta_K = \frac{-B\epsilon'_{xy} + A\epsilon''_{xy}}{A^2 + B^2}, \quad (4)$$

where $\epsilon_{xy} = \epsilon'_{xy} + i\epsilon''_{xy}$, $A = n^3 - 3nk^2 - n$, and $B = k^3 - 3kn^2 + k$.

In the case of transmitted light, the rotation of the plane of polarization is given by

$$\theta_F = -\frac{\pi l}{\lambda(n^2 + k^2)}(n\epsilon'_{xy} - k\epsilon''_{xy}), \quad (5)$$

where l is the layer thickness and λ is the wavelength of light.

Measurements were performed at photon energies of 1.96 and 2.54 eV (helium–neon and argon lasers) in a dc magnetic field of ± 15 kOe applied perpendicular to the sample surface. The angle of rotation of the polarization plane θ of the light reflected from a magnetized sample was measured with a polarimeter setup (Fig. 2)

Fig. 2. Experimental setup for measurement of the magneto-optical Kerr effect. PC stands for computer.

using a modulation technique. All the studies were performed at room temperature.

3. RESULTS AND DISCUSSION

3.1. Optical Properties

Figure 3 plots spectral responses of the real and imaginary parts of the dielectric function of (CoFeB)/(SiO₂) nanocomposites with magnetic-phase concentrations below (35 at. %) and above (62 at. %) the percolation threshold. The absence of absorption at energies above 4.2 eV is a remarkable feature observed at all concentrations of the magnetic phase. The inset to Fig. 3 displays the dependences of the indices of refraction n and absorption k on the concentration of CoFeB nanograins at 3 eV. The refractive index for these nanostructures is $n \approx 1.5$ and does not depend on the magnetic phase concentration or photon energy. The absorption index k and the imaginary part of the dielectric function ϵ_2 undergo the most pronounced changes with increasing concentration in the energy range below 4.2 eV. The nonmonotonic pattern of the spectral response of the optical parameters observed at low energies should apparently be assigned to the interference occurring in nanocomposite films with a thickness less than 1 μm . This assumption is borne out by the fact that this effect is most clearly pronounced in samples with a low concentration of the magnetic phase (Fig. 3a) and, hence, with low absorption.

We calculated the spectral response of the dielectric functions for the CoFeB alloy from the spectral response of these functions obtained for the nanocomposite and available optical parameters of the matrix. The calculation was based on the effective-medium approximation [19] and showed that the increased transparency observed above 4.2 eV is accounted for by the properties of the alloy rather than of the nanocomposite (Fig. 4). The fact that the calculations performed

Fig. 3. Spectral response of the real (ϵ_1) and imaginary (ϵ_2) parts of the dielectric function for (CoFeB)/(SiO₂) nanocomposites (samples I-1) with a magnetic phase concentration of (a) 35 and (b) 62 at. %. Inset: concentration dependences of the refractive index n and absorption index k for (CoFeB)/(SiO₂) nanocomposites at a photon energy of 3 eV.

Fig. 4. Spectral response of the real, ϵ_1 (filled symbols), and imaginary, ϵ_2 (open symbols), parts of the dielectric function for a Co₄₁Fe₃₉B₂₀ alloy. Plotted for comparison are spectral responses for pure iron and cobalt [20].

for different magnetic phase concentrations yielded the same results corroborates the validity of the spectra displayed in Fig. 4.

The spectral and concentration relations of the (CoFeZr)/(Al₂O₃) nanocomposites followed a different pattern. Figure 5 presents graphs of the real and imagi-

nary parts of the dielectric function obtained for nanocomposites of series II-1 and II-2 with a grain concentration of 51 at. %. It should be stressed that the absorption index of these nanocomposites does not vanish at any concentration of the magnetic phase, which sets this situation apart from the one observed in the (CoFeB)/(SiO₂) nanocomposites. This fact probably indicates that the matrix plays an essential role in forming the optical properties of a nanocomposite. Note that the spectral response of (CoFeZr)/(Al₂O₃) is more monotonic than that of (CoFeB)/(SiO₂) samples. The principal feature of the dielectric function spectral responses is a clearly pronounced structurization of spectra in the region of 3.5 eV at magnetic phase concentrations higher than 45 at. %. On the whole, the spectral responses of the dielectric functions of the nanocomposites prepared in a dc magnetic field under substrate cooling (samples II-2) and in zero field without cooling (II-1) are similar. In our opinion, the main difference lies in that the above-mentioned structurization of dielectric function spectra is more distinct in samples fabricated in zero field without cooling, which is demonstrated convincingly by Fig. 5. It should be pointed out that, in contrast to (CoFeB)/(SiO₂), the refractive index of the (CoFeZr)/(Al₂O₃) nanocomposites turns out to be dependent on photon energy, decreasing slightly with an increase in this energy, while remaining practically independent of concentration for the samples obtained in a magnetic field. Note that, at low energies, the spectra of nanocomposites II-1 are nonmonotonic, but this feature disappears as the particle concentration increases. We explain this feature in the same way as in the case of the (CoFeB)/(SiO₂) samples; namely, we assign it to interference. The smaller interference period in the samples of (CoFeZr)/(Al₂O₃) compared to that in the (CoFeB)/(SiO₂) nanocomposite correlates well with the larger thickness of the (CoFeZr)/(Al₂O₃) films. There is no interference, however, in samples II-2.

3.2. Magneto-optical Properties

Figure 6 displays concentration dependences of the Kerr effect obtained in a field of 15 kOe with a photon energy of 1.96 eV (solid lines) for the three types of (CoFeB)/(SiO₂) nanocomposites. Obviously, the presence of oxygen or nitrogen impurities does not have a noticeable effect on the maximum magnitude of the Kerr rotation angle, which is 0.25° for nanocomposites I-1 and I-2 and 0.3° for I-3. The concentration dependences also retain their general pattern for all samples. Note the presence of a sharp maximum at concentrations of 37, 40, and 37.5 at. % for samples I-1, I-2, and I-3, respectively. Because the percolation threshold for these nanocomposites was found to be about 46 at. % [12], the observed maximum is not related to percolation processes. This maximum lies in the range of magnetic-grain concentrations where the spectral responses of optical parameters behave nonmonotonically; there-

Fig. 5. Spectral response of the real, ϵ_1 , and imaginary, ϵ_2 , parts of the dielectric function for $(\text{CoFeZr})/(\text{Al}_2\text{O}_3)$ nanocomposites obtained (a) without a magnetic field and cooling (II-1) and (b) in a magnetic field under cooling (II-2).

fore, this behavior may originate from an interference phenomenon. The maximum is not associated with a change in an off-diagonal element of the permittivity tensor in Eq. (3), because the Faraday effect measured by us in this concentration range has the expected monotonic dependence on concentration (dashed line in Fig. 6c). Faraday rotation measurements could be performed only at magnetic-grain concentrations less than 42 at. % because of strong absorption of light setting in at higher concentrations.

The concentration dependence of the Kerr effect measured at a photon energy of 2.54 eV (dashed curve in Fig. 6a) is of the same character as the one observed for 1.96 eV but is shifted relative to the latter; in other words, the observed maximum is frequency-dependent and, thus, is not related to variation of the magnetic properties of the nanocomposites in the concentration range considered.

We calculated the concentration dependence of the rotation of the plane of polarization in reflection from a thin nanocomposite film (Fig. 7). The calculations were made in terms of the effective-medium approximation [21] using Eqs. (4) and (5) and included not only the Kerr effect for the ray reflected from the upper boundary of the nanocomposite film but also the Faraday effect for the ray transmitted through the film and reflected from the film/substrate interface. The calculated concentration dependence of the rotation of the

Fig. 6. Rotation of the plane of polarization plotted vs. magnetic grain concentration for $(\text{CoFeB})/(\text{SiO}_2)$ nanocomposites in a magnetic field of 15 kOe. (a) Kerr rotation for series I-1 measured at a photon energy of 1.96 eV (solid line, filled symbols) and 2.54 eV (dashed line, open symbols); (b) Kerr rotation for series I-2 measured at a photon energy of 1.96 eV; and (c) Kerr rotation (solid line, dark symbols) and Faraday effect (dashed line, open symbols) for series I-3 measured at a photon energy of 1.96 eV.

plane of polarization revealed that interference effects do indeed give rise to a strong enhancement of the magneto-optical effect at low magnetic-grain concentrations. The above appears to provide a convincing argument for the observed nonmonotonic behavior of the

Fig. 7. Calculated concentration dependences of the Kerr rotation angle θ_K (dash-dotted line), Faraday rotation angle θ_F (dashed line), and total rotation angle θ_Σ (solid line).

Fig. 8. Concentration dependences of the Kerr rotation for (CoFeZr)/(Al₂O₃) nanocomposites prepared in a dc magnetic field (samples II-2; solid line, open symbols) and without a magnetic field (samples II-1; dashed line, filled symbols).

Fig. 9. Field dependences of the Kerr rotation obtained for nanocomposites with magnetic phase concentrations below and above the percolation threshold. (a) (CoFeB)/(SiO₂) and (b) (CoFeZr)/(Al₂O₃). Inset: hysteresis for a (CoFeB)₆₈/(SiO₂)₃₂ sample.

concentration dependences below the percolation threshold being closely related to the interference effect.

Another remarkable feature was revealed in the concentration dependences of the Kerr effect for the

(CoFeZr)/(Al₂O₃) nanocomposites (Fig. 8). We believe the concentration dependence of the Kerr rotation for nanocomposites II-2, which has only one maximum near the percolation threshold, to be typical of nanocomposites. At the same time, samples II-1 reveal a reversal of sign of the effect at magnetic phase concentrations above 52 at. %. No such behavior is observed in nanostructures of the same composition but fabricated in a dc magnetic field under substrate cooling (samples II-2). As already mentioned, the optical spectra of these nanocomposites do not exhibit any radical differences; therefore, the observed difference in the magneto-optical properties may be assigned to a difference between the off-diagonal elements of the permittivity tensor in Eq. (3) for nanocomposites prepared in a magnetic field and without one.

It should be pointed out that the concentration dependences for samples II-1 reveal oscillations at low concentrations, which are not observed for samples II-2. This behavior reminds one of the case already discussed in connection with the optical properties of nanocomposites and is also related to interference effects.

3.3. Magnetic Properties

Figure 9 presents magnetic field dependences of the polar magneto-optic Kerr effect obtained on (CoFeB)/(SiO₂) and (CoFeZr)/(Al₂O₃) nanostructures for magnetic phase concentrations below and above the percolation threshold. The pattern of the field dependences measured at low concentrations (in particular, the absence of hysteresis and of saturation in a field of 15 kOe) is typical of an ensemble of superparamagnetic particles. Magnetization curves of a superparamagnet are described, similar to the case of a paramagnetic material, by the Langevin function [11] with the magnetic moment of a paramagnetic atom replaced by the magnetic moment of a grain given as a single-domain ferromagnetic particle:

$$\theta_K = \theta_K^s \left(\coth\left(\frac{\mu H}{k_B T}\right) - \left(\frac{\mu H}{k_B T}\right)^{-1} \right), \quad (6)$$

where μ is the magnetic moment of the grain, H is the magnetic field strength, and T is the temperature.

Using function (6), we described the field dependences of the Kerr effect for all types of nanocomposites under study for concentrations below the percolation threshold. Thus, within this concentration region, these nanocomposites can be considered as an ensemble of noninteracting ferromagnetic single-domain particles with a negligible size dispersion for each concentration. At higher nanoparticle concentrations, the distances between the particles decrease, thus increasing the role of interactions between them and favoring the formation of clusters and chains, as a result of which the nanocomposites are no longer superparamagnets. The appearance of a weak hysteresis and saturation,

which become particularly pronounced in nanocomposites I-1, supports the existence of ferromagnetism in the nanocomposites above the percolation threshold.

We used Eq. (6) to analyze the field dependences of the Kerr rotation for samples with grain concentrations below the percolation threshold. The analysis showed that an increase in the magnetic phase concentration brings about an increase in the magnetic moment of the grains, which is caused by their growth in size. The presence of oxygen or nitrogen impurities in the (CoFeB)/(SiO₂) nanocomposites affects the grain magnetic moment substantially by reducing it. Because hysteresis and saturation at high concentrations are more pronounced in samples that are free of impurities, one may conclude that the presence of impurities also weakens intergrain interaction.

4. CONCLUSIONS

The main results of our study of the optical, magnetooptical, and magnetic properties of (CoFeB)/(SiO₂) and (CoFeZr)/(Al₂O₃) nanocomposites can be summed up as follows.

(i) It has been established that, at photon energies above 4.2 eV, (CoFeB)/(SiO₂) nanocomposites are transparent, irrespective of the magnetic-grain concentration. The CoFeB alloy was also shown to be transparent in this energy region. The spectral responses of the dielectric functions of the (CoFeZr)/(Al₂O₃) composites are more smooth, with a distinct structure appearing in them only above the percolation threshold.

(ii) A feature has been revealed in the concentration dependences of the Kerr effect in (CoFeB)/(SiO₂) nanocomposites, both containing oxygen and nitrogen impurities and without them; this feature can be reliably related to interference effects.

(iii) The diagonal elements of the permittivity tensor of the medium for the (CoFeZr)/(Al₂O₃) composite samples prepared in a magnetic field and without one have similar spectra. Therefore, the observed difference in behavior of the concentration dependences of the Kerr rotation between the two types of (CoFeZr)/(Al₂O₃) nanocomposites suggests a substantial change in the off-diagonal elements of the permittivity tensor. More comprehensive information can be obtained from a study of the spectral response of the Kerr effect for these nanocomposites.

(iv) The maximum magnitude of the Kerr effect in a field of 15 kOe is 0.2°–0.3°. This should be compared with the saturated Kerr effect in pure Co and Fe, which is about 0.32° and 0.43°, respectively [22]. In all nanocomposites, the Kerr rotation reaches a maximum (if we neglect local interference maxima in the region of low concentrations) in the concentration region corresponding to the percolation threshold.

(v) Below the percolation threshold, the nanocomposites under study behave like superparamagnets with no appreciable dispersion in grain size. Above the per-

colation threshold, the interaction between grains plays a substantial part, with the presence of oxygen and nitrogen impurities reducing the strength of this interaction. Impurities also reduce the magnetic moment of the grains.

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research (project no. 03-02-17575a), the Presidium of the Russian Academy of Sciences (program “Low-Dimensional Quantum Structures”), and the European program “Dynamics.”

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Translated by G. Skrebtsov