Magneto-optical properties of Co-based nanocomposites

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Abstract

Magneto-optical properties of DC magnetron sputtered Co-based granular films have been studied in the transversal Kerr effect (TKE) geometry in the photon energy range 0.5 – 4.5 eV at room temperature. The TKE vs. concentration plots demonstrate a strong non-linear dependence on metal fraction with a maximum at the percolation threshold for Co\textsubscript{x}(TiO\textsubscript{2})\textsubscript{1-x} and Co\textsubscript{x}(Sm\textsubscript{2}O\textsubscript{3})\textsubscript{1-x} films. A correlation between the TKE concentration dependences and the giant magneto-resistance (GMR) data has been found. The results show that the magnitude and shape of the TKE concentration dependences (as well as GMR) depend not only on the metal volume fraction but also on the dielectric matrix composition (TiO\textsubscript{2} or Sm\textsubscript{2}O\textsubscript{3}).

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1. Introduction

Ferromagnetic metal-metal and metal-insulator granular alloys have been subject of great interest in the last ten years, since they exhibit unique physical properties. The features inherent to nanocomposites reveal themselves not only in magneto-transport properties, giant magneto-resistance (GMR), tunneling magneto-resistance and giant extraordinary Hall effect [1,2], but also in the linear and nonlinear optical and magneto-optical (MO) effects [3,4].

The magneto-optical studies are sensitive to the changes of magnetic and electronic structures, the mechanisms of dispersion, the characteristic sizes, the form and topology of nanoparticles [5,6]. The essential growth of magneto-optical response near the percolation threshold, as well as enhanced magneto-resistance, was observed in a number of nanocomposite systems [7-10]. This behavior of MO effect was rather successfully described within the framework of the modified effective medium theory. However, there is no even qualitative understanding of MO spectra for the case of ferromagnetic MnAs clusters embedded in the GaAs semiconductor matrix and for discontinuous multilayers.[3,4] The main problem now is the search for nanocomposite
systems possessing an optimum combination of magneto-transport and magneto-optical properties what demands careful studies of composites with various dielectric and semi-conductor matrixes. For example, one of the most promising materials is Co-doped TiO$_2$ anatase in which the room-temperature ferromagnetism has been recently discovered [11]. It should be relevant to investigate its properties for a wide interval of metal component concentrations. Though the problem of interaction of Co particles with different matrixes was studied in many works [12,13], till now there is no full understanding how the surroundings influences the dispersion of the metal in the dielectric or semiconductor medium.

Therefore it was of interest to study the dependence of MO and MR properties of nanocomposites on matrix composition and their changes resulting from annealing. In this work we present the results of investigation of magneto-optical, magnetic and magneto-transport properties of granular ferromagnetic-insulator composites, in which the ferromagnetic nanometer-sized polycrystalline Co granules are embedded into either TiO$_2$ or Sm$_2$O$_3$ insulating matrix, both in as-deposited state and after annealing.

2. Experiment

Co$_x$(TiO$_2$)$_{1-x}$ and Co$_x$(Sm$_2$O$_3$)$_{1-x}$ granular films (where $X$ is the concentration of Co ranging from 25 to 86 at.%) approximately 400 nm thick were prepared by a tandem deposition method using rf-magnetron sputtering from Co and TiO$_2$ (or Sm$_2$O$_3$) targets. The films were sputtered onto glass substrates (Corning No. 1737). The base pressure in the chamber was $5 \times 10^{-5}$ Torr and a flow of high purity argon with pressure of $2 \times 10^{-4}$ Torr was used during sputtering. The samples were annealed at 200°C for 30 minutes. The MO properties were investigated in the transverse Kerr effect (TKE) geometry. The TKE spectra were measured by a dynamic method in the 0.5 – 4.5 eV energy range in the applied magnetic field up to 3.0 kOe. Magnetic properties of the films were measured by vibrating magnetometer in magnetic field up to 20 kOe. The magneto-resistance (MR) was investigated using standard four-point probe geometry.

3. Results and discussion

Figures 1 and 2 show the spectral dependences of the TKE in Co$_x$(TiO$_2$)$_{1-x}$ and Co$_x$(Sm$_2$O$_3$)$_{1-x}$ granular films. For these films, as well as for other granular alloys, it was found that the TKE spectra of nanocomposites significantly differ from the spectrum of homogeneous polycrystalline cobalt. Moreover, the amplitude of the magneto-optical response of the nanocomposites in some instances surpassed that of the bulk Co. Contrary to the amorphous metal-dielectric nanocomposites [10], the studied nanocomposites exhibit changes of the spectra shape and a substantial growth of the TKE amplitude not only in the infrared, but also in the ultra-violet range. With increase of the metal concentration the shape of the TKE spectra gradually changes and a new negative feature grows in the
near-infrared region as well as a new wide peak appears in the UV range. From comparison of the TKE spectra of the two systems follows that substitution of the matrix material does not lead to cardinal changes in the amplitude and shape of the TKE curves.

Figure 3 shows the TKE versus metal fraction dependence for our Co-based granular films. For the both systems there was observed a non-monotone dependence of TKE on the magnetic component concentration with a maximum near the percolation threshold. This maximum is most pronounced in the near infrared range.

The TKE versus metal fraction dependences exhibiting a maximum at the concentrations corresponding to the percolation threshold $X_{pc}$ were already observed in granular alloys such as $(\text{FePt})_{1-x} (\text{SiO}_2)_x$ [9], $(\text{CoFeB})_x (\text{SiO}_2)_1-x$ and $(\text{CoFeZr})_x (\text{SiO}_2)_1-x$ [10], Co(Al$_2$O$_3$)[14]. As it was shown in [10] this maximum is not connected with the growth of the MO activity but is caused by variation of the optical and MO parameters due to changes of topology and microstructure of nanocomposites.

Generalization of these results allows a conclusion that in nanocomposites the non-monotone behavior of the TKE versus concentration curves does not depend on the composition of metal or dielectric phases.

The comparison of the curves on fig.3 demonstrates that for nanocomposites the behavior of the concentration dependence TKE($x$) as well as MR($x$) depends on matrix material. It should be pointed out that the shapes of the peaks on the magneto-optical response versus concentration plots follow the shape of the magneto-resistance maxima both for Co$_x$(TiO$_2$)$_{1-x}$ and Co$_x$(Sm$_2$O$_3$)$_{1-x}$ thin films.

For the Co$_x$(TiO$_2$)$_{1-x}$ composites a wide negative maximum of TKE is observed in the metal component concentration range of $39-49\%$. However for the Co$_x$(Sm$_2$O$_3$)$_{1-x}$ granular alloy the MO response attains large magnitudes near the percolation threshold, with a maximum observed only at $X\sim 49\%$.

It is known that microstructure of the granulated alloys varies with the metal component concentration, what in turn causes changes in the magnetic, optical and magneto-optical properties. Below the percolation threshold composites represent an array of isolated magnetic granules introduced into a nonmagnetic matrix [15,16], and remain in the superparamagnetic state. The analysis of magnetic characteristics measured for Co$_x$(Sm$_2$O$_3$)$_{1-x}$ testifies that at $x = 33\%$ it exhibits hard magnetic properties (the magnetization curve comes to saturation in the fields $H_S \sim 14\, \text{kOe}$) and has a low saturation magnetization. Apparently, by this are determined small magnitudes of MO response observed in such nanocomposites, taking into consideration that the Kerr effect depends linearly on magnetization and the maximum amplitude of the magnetic field used in our Kerr effect measurements was $3\, \text{kOe}$. Beyond the percolation threshold composites consist of continuous metal regions divided by small nonmagnetic layers and they are in the ferromagnetic state [15]. Magnetization curves for the Co$_x$(Sm$_2$O$_3$)$_{1-x}$ film with $x=75\%$ indicate that it is ferromagnetic with $H_S \sim 1\, \text{kOe}$.

The morphology of composites near the percolation threshold is of such kind that the dielectric layer dividing the ferromagnetic granules from each other is minimal [15,17]. In spite of the fact that magnetic particles in such alloy still do not contact with each other directly, there were investigations confirming that magnetic moments of the granules already start to cooperate. Apparently, enhancement of the MO response near the percolation threshold is caused by a unique combination of magnetic and optical properties.

Evolution of the magneto-transport and MO properties of Co$_x$(TiO$_2$)$_{1-x}$ and Co$_x$(Sm$_2$O$_3$)$_{1-x}$ films after annealing 30 min at $200\, \text{C}$ has been examined in order to study the process of structural relaxation. The study of the MR concentration dependence in the composites has shown that annealing differently influences the GMR magnitude depending on the matrix composition. For the both systems the MR magnitude decreased for the composites with concentrations beyond the percolation threshold.

![Fig. 3. The comparison of concentration dependencies of TKE (E~1eV) and magneto–resistance: a – for Co$_x$(TiO$_2$)$_{1-x}$ and b – for Co$_x$(Sm$_2$O$_3$)$_{1-x}$ nanocomposite.](image-url)
That is caused by integration of cobalt granules and formation of metal chains. Consequently, electroconductivity in the granulated alloys results not only from tunneling between granules, but is also determined by the transport of carriers along the metal channels formed of the integrated ferromagnetic particles. In the composites with ferromagnetic component concentration below the percolation threshold the MR magnitude practically was not changed. The most significant changes of the MR and magneto-optical properties resulting from the annealing were observed for the nanocomposites near the percolation threshold that is in case when the distances between granules are minimal. The MR magnitude for the Co<sub>x</sub>(TiO<sub>2</sub>)<sub>_1-x</sub> system with x within the percolation range increased after annealing by 1 %, while for Co<sub>x</sub>(Sm<sub>2</sub>O<sub>3</sub>)<sub>_1-x</sub> it decreased by 0.4 %.

Annealing influenced much more strongly the magneto-optical properties than the magneto-transport ones, and the TKE spectra evolution of the composites was found to depend on the matrix material. The TKE spectra of Co<sub>x</sub>(Sm<sub>2</sub>O<sub>3</sub>)<sub>_1-x</sub> show little changes after annealing, nevertheless a reduction of the MO response amplitude by approximately 2 times was observed in the IR range. For the Co<sub>x</sub>(TiO<sub>2</sub>)<sub>_1-x</sub> films after annealing the TKE amplitude has decreased by more than 6 times (energy of incident light E=1 eV). Moreover, the sign of the MO effect has changed. (Fig.4)

According to the publication available interactions of Co with titanium oxide and samarium oxide can be different. Moreover a strong metal–surrounding interaction has been found to affect the metal dispersion [13,18]. As a result, the nanocomposition microstructures forming during magnetron sputtering depends on the matrix composition. Magneto-optical properties are highly sensitive to deformations of the alloy microstructure. That is why the shapes of the TKE spectra are so much different for the annealed Co<sub>x</sub>(TiO<sub>2</sub>)<sub>_1-x</sub> and Co<sub>x</sub>(Sm<sub>2</sub>O<sub>3</sub>)<sub>_1-x</sub> granular systems.

The changes caused by structural relaxation begin at the surface and are most strongly manifested in the near-surface layer, what brings about greater changes in the MO properties in comparison with the magneto-transport ones.

4. Conclusion

The magnetic, magneto-transport and magneto-optical properties of Co<sub>x</sub>(TiO<sub>2</sub>)<sub>_1-x</sub> and Co<sub>x</sub>(Sm<sub>2</sub>O<sub>3</sub>)<sub>_1-x</sub> nanocomposites have been investigated in a wide concentration region in as-deposited state and after annealing at 200°C. For the both systems in the vicinity of the percolation threshold an enhancement of the MO response connected with the changes of morphology of the composites and transition from superparamagnetic to ferromagnetic state has been observed. A correlation between the TKE concentration dependences and the GMR data has been established. It has been found that annealing influences the magneto-optical properties much more strongly than the magneto-transport ones and the evolution of TKE spectra of the composites depends on the matrix material.
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References