Optical and magneto-optical spectra of insulating granular system Co-Al-O

E. Gan'shina, R. Kumaritova, A. Bogoroditsky, M. Kuzmichov, S. Ohnuma*.

Physics Department, Moscow Lomonosov University, 119899, Russia.

*The Research Institute of Electric and Magnetic Materials, Sendai982, Japan.

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Optical and magneto-optical spectra of the insulating granular system Co-Al-O prepared by reactive sputtering are measured in the energy range 0.5–3.8 eV. The transverse Kerr effect magnitude in the $\text{Co}_{52}\text{Al}_{20}\text{O}_{28}$ film attain large values. The data have been interpreted in the framework of the effective medium approximations: Bruggeman (BA) and symmetrised Maxwell-Garnett approximation (SMG). The best fit between experimental data and theoretical prediction was obtained in SMG. It is shown that magneto-optical spectra of ferromagnetic composites are highly sensitive to composite microstructure.

Keywords: magneto-optical spectra, metal-insulator composites, effective medium approximation.

1. Introduction

Recently giant magnetoresistance (GMR) has been reported in the insulating granular Co-Al-O thin films¹⁻³. The value of the GMR of Co-Al-O films depends on oxygen concentration and takes maximum at a critical oxygen concentration, just above percolation threshold³. This is possibly connected with complex modification in microstructure of these composite material in the vicinity of the percolation transition. Further detailed investigation is needed to clarify this mechanism.

The optical methods are widely used for investigation of the microstructure of these insulating granular alloys^{4, 5}. For magnetic granular films one can obtain the magneto-optical (MO) spectra, as well as the optical one. Recently we have shown that MO investigation can give useful information about the concentration and shapes of the magnetic granulars⁶⁻⁸.

In this paper we present the results of the optical and magneto-optical spectra measurements of superparamagnetic insulating granular Co-Al-O thin films and the analysis of these spectra in the framework of the effective medium approximations: Bruggeman (BA)^{4, 8} and symmetrised Maxwell-Garnett approximations (SMG)⁹.

2. Experimental

The Co-Al-O granular thin films were prepared on glass substrates by a reactive sputtering technique. The structural, magnetic and electrical transport properties of these samples were reported previously^{1, 2}. Microstructural observation revealed that these films were composed of Co-based metallic granules and Al_2O_3 -based insulating network-like boundaries. The diameter of Co-particles is

about 20 Å or slightly larger and the width of the boundaries is 5–10 Å.

We investigated three samples of composition near to $\text{Co}_{52}\text{Al}_{20}\text{O}_{28}$, which gives the largest magnetoresistance (MR). The values of the specific electrical resistance (ρ) and magnetoresistance for investigated samples are given in table 1. The ratio of Co/Al of all samples is kept constant (75/25) and concentration of oxygen becomes higher in the order of the samples 1, 2, 3.

Table.1

Sample	$\rho, \mu\Omega\text{cm}$	MR%(10kOe)
1	$6.8 \cdot 10^4$	-7.2%
2	$1.2 \cdot 10^5$	-9.2%
3	$5 \cdot 10^5$	-7.2%

The optical constants n and k were measured using the Beattie-polarization method. The MO properties were measured in the geometry of transverse Kerr effect (TKE) in the energy range 0.5–3.8 eV, the light incidence angles being 60° and 73°. A dynamic method of the recording of the magneto-optical effect was used. The relative change in the intensity of the reflected light, which is equal to $\delta = I(H)/I(0) - 1$, was directly measured in the experiment, where $I(H)$ and $I(0)$ are the intensities of the reflected light in the presence and in the absence of the magnetic field, respectively. The maximum amplitude of the ac magnetic field was equal to 3 kOe. For the granular films the parameter δ at this field amplitude is about 80–90% of their maximum value.

3. Results and discussion

Fig. 1 shows the spectral dependence of TKE for the Co-Al-O and Co⁷ films. As well as for other granular alloys⁶⁻⁸ one can see the drastic change of MO spectra in the

Fig. 1 The experimental TKE spectra of the samples 1, 2, 3 ($\phi=60^\circ, 73^\circ$) and pure Co ($\phi=73^\circ$).

Corresponding author Fax: +7-095-932-8820;
e-mail: eagan@magn.phys.msu.su

Fig. 2 Lines show the calculated optical spectra for different values of Co volume fraction f in the framework of Bruggeman approach ($L=0.33$). Circles and squares show experimental ones of sample 2.

comparison to that of homogeneous Co. In these superparamagnetic films the TKE magnitude attained values larger than it was for bulk Co and previously investigated granular alloys $\text{Co}_x\text{CuO}_{1-x}$ ⁸⁾. Note that the curves for all samples are quite similar in UV and visible range, but there is a significant difference among them in the near infrared range and at incident angle $\varphi=73^\circ$.

The behavior of the measured optical spectra was similar for all samples and was in agreement with the data for granular films of $\text{Co-Al}_2\text{O}_3$ ⁴⁾ and $\text{Ni-Al}_2\text{O}_3$ ⁵⁾. Fig.2 shows real and imaginary parts $\epsilon' = n^2 - k^2$ and $\epsilon'' = 2nk$ of dielectric tensor for the sample 2. The values ϵ' and ϵ'' slowly increase with wavelength. It is related to the percolation behavior. Obtained values of ϵ' for our samples are less than the one for evaporated $\text{Co-Al}_2\text{O}_3$ composite⁴⁾.

To understand these data we calculated the MO spectra of these composites in the effective medium approximation (BA)⁴⁾. According to this approach the MO spectra $\delta(\hbar\omega)$ can be calculated as a function of the consistent optical and MO parameters, volume fraction f of magnetic particles and their shape L . The scheme of the TKE calculation is following^{6,7)}.

In linear approximation at magnetic field and magnetization the effective dielectric tensor ($\hat{\epsilon}$) of granular ferromagnet magnetized along z-direction can be written as:

$$\hat{\epsilon} = \begin{pmatrix} \epsilon & i\gamma & 0 \\ -i\gamma & \epsilon & 0 \\ 0 & 0 & \epsilon \end{pmatrix}. \quad (1)$$

The two self-consistent equations for the diagonal and non-diagonal elements of effective tensor in the BA are as follows^{4,6)}

$$f(\epsilon_1 - \epsilon^{BA})(\epsilon^{BA} + (\epsilon_1 - \epsilon^{BA})L_j) + (1-f)(\epsilon_0 - \epsilon^{BA})(\epsilon^{BA} + (\epsilon_0 - \epsilon^{BA})L_j) = 0, \quad (2)$$

$$f(\gamma^{BA} - \gamma)[\epsilon^{BA} + (\epsilon_1 - \epsilon^{BA})L_j]^2 + (1-f)\gamma^{BA}[\epsilon^{BA} + (\epsilon_0 - \epsilon^{BA})L_j]^2 = 0, \quad (3)$$

where $\epsilon_1 = \epsilon'_1 - i\epsilon''_1$ and $\gamma_1 = \gamma'_1 - i\gamma''_1$ are diagonal and non-diagonal dielectric tensor elements of the magnetic component, respectively and $\epsilon_0 = \epsilon'_0 - i\epsilon''_0$ is the dielectric constant of the non-magnetic component. These equations should be solved and then it is easy to calculate TKE by well known relations

$$\delta = (a\gamma' + b\gamma'') \frac{2 \sin 2\varphi}{a^2 + b^2}, \quad (4)$$

where φ is the light incident angle,

$$a = \epsilon''(2\epsilon' \cos^2 \varphi - 1); \quad b = \cos^2 \varphi(\epsilon''^2 - \epsilon'^2 + 1) + \epsilon' - 1.$$

The optical and MO parameters used in the calculations were taken from⁷⁾ for Co and from¹⁰⁾ for Al_2O_3 . We used the experimental value for the volume fraction $f_{\text{exp}}=0.58$ and spherical particles ($L=0.33$) as a first step to fit the experimental data shown in Fig. 1. The calculation of the volume fraction f_{exp} was done under the assumption that Al_2O_3 was completely formed in the film $\text{Co}_{52}\text{Al}_{20}\text{O}_{28}$ ²⁾. Comparing the calculated and the experimental results, we put into account that the ratio $\delta/\delta_{\text{max}}$ is about 0.8 at the magnetic field 3 kOe.

The results of our calculation for the various sets of parameters are shown in Figs. 2-3. Comparison of the experimental data and calculated curves in Figs. 1-3 shows that calculated TKE is very small for experimental volume fraction $f=0.58$ and it is in qualitative agreement with experimental data for volume fraction about 0.42. The magnitude of TKE increases as volume fraction f decreases. The calculated optical spectra are in better agreement with the experimental data for smaller Co-volume fraction too. Such difference between f_{exp} and f was observed for many other investigated granular alloys and connected with the fact that parts of the ferromagnetic metal particles are in an oxidized state and that some of Co may remain dispersed in atomic form.

The simple theory BA can give a qualitative description of the observed behavior of MO spectra but does not explain the spectral profile of the experimental TKE curves in the near IR range. Recently it was shown⁹⁾ that the MO spectra for ferromagnetic metal-insulator composites can be more correctly described in SMG approach¹¹⁾. Its approximation takes into account the presence of separate

Fig. 3 The calculated TKE spectra for different Co volume fraction f in the framework of Bruggeman approach ($L=0.33$)

particles in the medium like in Maxwell-Garnet (MG) approximation ⁴⁾. However, in contrast to MG the SMG approach is applicable for arbitrary values of ferromagnetic volume fraction.

In this approach effective tensor is calculated as an average over two following random units: unit A - an inclusion of A coated by a shell of B, unit B - an inclusion of B coated by a shell of A (Fig.4)¹¹⁾.

Fig.4 Random units A and B in the effective medium (SMG approximation)¹¹⁾.

The probabilities to find these units in composites are $P_A = u_1 / (u_1 + u_2)$ and $P_B = u_2 / (u_1 + u_2)$,

where $u_1 = (1 - f^{1/3})^3$ and $u_2 = [1 - (1 - f)^{1/3}]^3$.

Then it is easy to calculate the dielectric functions for both types of units in MG approximation and then SMG approximation can be obtained introducing these values in the Bruggeman eqs. (2-3). (You can see the calculation in detail in ref. 9).

The simulated MO spectra in SMG approach shows that the amplitude profile and even the sign of TKE are extremely sensitive to a small changing of parameters and rather in narrow range of values f , L_A and L_B correlate to the obtained experimental data.

The final results for comparison to calculated TKE spectra with the experimental data are presented in Fig. 5. One can see that SMG approach gives a correct description of the MO spectra, if $f=0.46$ and an effective medium is built from spherical magnetic granules ($L_B=0.33$) and needle-like non-magnetic components ($0.1 < L_A < 0.2$). Percolation threshold f_c in this case is approximately equal to 0.47.

Fig.5. Lines show the calculated TKE spectra for different unit A shape factor (L_A) in the framework of SMG ($f=0.46$, $L_B=0.33$). Symbols show the experimental TKE spectra of samples 1,2,3.

Fig. 6 Lines show the calculated optical spectra for different unit A shape factor (L_A) in the framework of SMG ($f=0.46$, $L_B=0.33$). Symbols show the experimental optical spectra of sample 2.

The results of the calculations suggest that the differences in the TKE spectra of the samples in near IR range are related to changing of their microstructure - the thickness of boundaries and its shape varies with changing of the oxygen concentration. Note that calculated optical spectra (Fig.6) depend only weakly on the shape of the nonmagnetic component L_A .

Consequently we can conclude that the magneto-optical spectra are more sensitive to microstructure of inhomogeneous media by reference to optical spectra and SMG approach gives correct description of the MO spectra by reasonable values of concentration and shape of magnetic and nonmagnetic particles.

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