Simulation of magneto-optical properties of nanocomposites 
\((\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}\)

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Abstract. The magnetooptical spectra of the transverse Kerr effect of \((\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}\) nanocomposites are calculated within the framework of the symmetrized Maxwell-Garnett (SMG) approximation. The quasi-classical size effect and the size distribution of the granules were taken into account. The calculation results for the spectral range 0.5-3.5 eV and different concentrations of the magnetic component are in semi-quantitative agreement with the experimental data. A possible reason for the existing discrepancy is the difference between the optical and magneto-optical parameters of ferromagnetic granules from those corresponding for bulk samples.

1. Introduction and background

Magnetic nanocomposites “ferromagnetic metal-dielectric” have various magnetic, magnetotransport, optical and magneto-optical properties depending on their chemical composition, component concentration, crystal structure, size and shape of the granules and so on [1, 2]. At the same time, in such structures for compositions near the percolation threshold, it is possible to enhance such promising effects as magnetoresistance, extraordinary Hall effect (EHE), high-frequency magnetic permeability, magneto-optical transverse Kerr effect (TKE), nonlinear magneto-optical effects, etc. [3-6]. Magneto-optical spectroscopy is an effective method for studying the magnetic microstructure of ferromagnetic materials, including nanocomposites. However, in the case of nanocomposites, the interpretation of magneto-optical spectra is associated with significant difficulties and the effective medium method is practically the only one to be implemented [7]. Among the different variants of the effective medium theory, the symmetrized Maxwell-Garnett (SMG) approximation [8] allows to take into account the nanocomposite microstructure most fully at an arbitrary concentration of the components.

In this paper, the SMG is used for calculations the TKE spectra of \((\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}\) nanocomposites for different values of the volume concentration of the magnetic component. In addition to the well-developed approach we took into account the influence of the quasi-classical size effect and the granular size distribution on the TKE spectra.
2. Calculation method

The SMG method is based on calculation of the probability to find the dielectric granule surrounded by metal (particle A) and the metal granule surrounded by dielectric (particle B) for the nanocomposite with the metal volume fraction \( X \) (figure 1) [8]. The SMG expressions for calculations of the diagonal (optical) component \( \varepsilon_{\text{EMA}}^{\text{MG}} \) and the nondiagonal (magneto-optical) component \( \gamma_{\text{EMA}}^{\text{MG}} \) of the dielectric permittivity tensor for nanocomposites are as follows [9]:

\[
\begin{align*}
P_A \frac{\varepsilon_{\text{EMA}}^{\text{MG}} - \varepsilon_{\text{EMA}}}{2(1 - L_A)(\varepsilon_{\text{EMA}}^{\text{MG}} - \varepsilon_{\text{EMA}})} + P_B \frac{\varepsilon_{\text{EMA}}^{\text{MG}} - \varepsilon_{\text{EMA}}}{2(1 - L_B)(\varepsilon_{\text{EMA}}^{\text{MG}} - \varepsilon_{\text{EMA}})} &= 0 \\
\frac{\gamma_{\text{EMA}}^{\text{MG}} - \gamma_{\text{EMA}}}{2(1 - L_A)(\varepsilon_{\text{EMA}}^{\text{MG}} - \varepsilon_{\text{EMA}})} + \frac{\gamma_{\text{EMA}}^{\text{MG}} - \gamma_{\text{EMA}}}{2(1 - L_B)(\varepsilon_{\text{EMA}}^{\text{MG}} - \varepsilon_{\text{EMA}})} &= 0
\end{align*}
\]

(1)

where \( P_A \) (\( P_B \)) is the probability of the presence of the ellipsoidal particle A (B) with the form-factor \( L_A \) (\( L_B \)), \( \varepsilon_{\text{EMA}}^{\text{MG}} \) is the effective permittivity obtained using the Maxwell – Garnett approximation [4], indexes 1 and 2 correspond to the metal and dielectric components.

\[
P_A = \frac{u_1}{u_1 + u_2}, \quad P_B = \frac{u_2}{u_1 + u_2},
\]

(3)

\[
u_1 = \left(1 - X^{\frac{1}{3}}\right)^3, \quad u_2 = \left(1 - \left(1 - X^{\frac{1}{3}}\right)\right)^3
\]

(4)

In these expressions, all optical and magneto-optical parameters of the metal and dielectric components are determined from the literature data for the bulk samples. It is obvious that these parameters for the granules may differ from the bulk ones. One, but not the only mechanism leading to such a difference, is the quasi-classical size effect associated with scattering on the surface of the granules. The method of accounting for this effect is developed in [8-11]. The role of the quasi-classical size effect in magneto-optics is important in the infrared region of the spectrum. Its contribution to the magneto-optical parameter is determined by the ratio \( R_s/R_{\text{bulk}} \), that is, the ratio of the anomalous Hall effect coefficient of the granule surface to the same coefficient for the bulk sample. Modeling of magneto-optical spectra and comparison with experiment allows to find this parameter. Since the quasi-classical size effect depends on the size of the granules, the simulation takes into account the size distribution of the granules through the lognormal distribution.

SMG expressions are determined by the volume concentration of the metal \( X \) (see (4)), whereas experimental data are usually given for atomic concentration \( X_A \). To compare the results of the calculation with the experiment, it is necessary to convert the atomic to the volume concentration \( X \)
according to the formula:

\[ X = \frac{V_1 \cdot X_d}{V_1 \cdot X_d + V_2 \cdot (1 - X_d)} \]  

(5)

where \( V_1 \) (\( V_2 \)) is the molar volume of the metal (dielectric) component.

The calculation results were compared with the experimental data for the spectral dependences of the transverse Kerr effect (TKE) of \((\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}\) nanocomposites given in figure 2. Details of samples fabrication, their structural and magnetic properties are given in [12]. All measurements were carried out at room temperature in a field of about 2 kOe. In fact, the atomic concentration of metal \( X_d \) for nanocomposite thin film can differ from the nominal atomic concentration \( x \) because of nonstoichiometric matrix and dispersed magnetic ions in it, but we did not take this difference into account.

3. Calculation results and discussion

The obtained spectral dependences for different values of the atomic concentration of the magnetic component are shown in figures 3-5. We varied the form-factors of particles \( A \) and \( B \) in such a way as to correctly reproduce the value of the percolation threshold (that is very critical for the region of average concentrations), as well as the \( R/R_{\text{bulk}} \) parameter, and the average size of the granules. Note that the best agreement with the experiment was obtained with the average size of the granules \( r_0=2.35 \) nm, which is in good agreement with the structural data [12]. \( R/R_{\text{bulk}} \) cannot be determined experimentally and was modeled based on the best correspondence with the measured TKE spectra. Thus, the work carried out an assessment of the parameter \( R/R_{\text{bulk}} \), that is important for the theory of the anomalous Hall effect.

**Figure 2.** Experimental spectral dependences of the transverse Kerr effect for \((\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}\) nanocomposites with different atomic concentrations of metal: 18.5\% - triangles, 28.7\% - circles and 62.1\% - squares.

**Figure 3.** Experimental TKE spectrum for \((\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}\) nanocomposites (triangles) in comparison with model spectrum (solid line) \((X=0.063, R/R_{\text{bulk}} = -10, L_A=0.49, L_B=0.33, r_0=2.35 \) nm).
Figure 4. Experimental TKE spectrum for $(\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}$ nanocomposites (triangles) in comparison with model spectrum (solid line) ($\chi=0.106$, $R/R_{\text{bulk}} = -6$, $L_A=0.44$, $L_B=0.33$, $r_0=2.35$ nm).

Figure 5. Experimental TKE spectrum for $(\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}$ nanocomposites (triangles) in comparison with model spectrum (solid line) ($\chi=0.325$, $R/R_{\text{bulk}} = -10$, $L_A=0.49$, $L_B=0.33$, $r_0=2.35$ nm).

Semi-quantitative agreement between theory and experiment is obtained. At low volume concentration of ferromagnetic granules (figure 3) good agreement takes place in the entire investigated frequency range. At the same time a certain value of the $R/R_{\text{bulk}}$ parameter corresponds well to the concept of the spin-orbital interaction enhancement at the surface of nanoparticles. At a low concentration of ferromagnetic granules, that is, far away from the percolation threshold, the values of form-factors do not strongly affect the spectral dependence, however, the values found show that the shape of the granules is close to spherical, which corresponds to the structural data. With increasing concentration of granules (figures 4 and 5) for the same set of variable parameters it is not possible to describe in detail the spectra over the entire frequency range. Thus, at $\chi=0.106$ deviations are visible in the middle region of the spectrum (figure 4), and near the percolation threshold ($\chi=0.325$) in the infrared region of the spectrum (figure 5). There may be several reasons for this behavior. The optical properties of small particles can differ significantly from those of bulk samples both due to quantum size effects and due to surface plasma oscillations and changes in the electronic structure. It is impossible to exclude the formation of an oxide layer on the surface of metal granules. Therefore, the values of the optical parameters of the nanocomposite from the direct optical experiment by ellipsometry should be taken for modeling the magneto-optical spectra. In addition, the size distribution of the granules can affect not only the quasi-classical size effect, but also the magneto-optical parameters. Finally, the formation of aggregates of granules in combination with superparamagnetic and superferromagnetic entities is not taken into account in the framework of the effective medium theory.

4. Conclusions

The TKE spectra of $(\text{CoFeZr})_x(\text{Al}_2\text{O}_3)_{1-x}$ nanocomposites were calculated in the framework of the SMG and taking into account the quasi-clasical size effect and the granular size distribution. The
values of the form-factor ($L$), mean particle size ($r_0$) and the ratio of the anomalous Hall effect coefficient of the granule surface to the similar coefficient in the bulk sample ($R_s/R_{bulk}$) were varied in the calculations of the spectra. The first two parameters ($L$ and $r_0$) correspond to the available structural data, and the third ($R_s/R_{bulk}$) was modeled based on the best correspondence with the measured TKE spectra. It is important to note that only the account of the quasi-classical size effect allowed to describe well the experimental spectra of TKE at the qualitative level. Therefore, the work carried out an assessment of the parameter $R_s/R_{bulk}$. Thus, the SMG can be successfully used to describe the magneto-optical properties of nanocomposites, as well as their other properties. Modeling by the SMG method taking into account the quasi-classical size effect and distribution of granules in size allows not only to describe the experimental data, but also to determine important microscopic parameters.

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References
[1] Granovsky A B, Sukhorukov Yu P, Gan’shina E A, Telegin A V 2013 Series in Materials Science vol 178, p 107 ed. M Inoue, M Levy, A V Baryshev (Berlin: Springer)
[2] Vyzulin S A, Gorobinsky A V, Kalinin E Yu, Lebedeva E V, Sitnikov A V, Sariev N E, Trofimenko I T, Chekriguina Y I, Shipkov I G 2013 Izvestiya RAN, Physical Series 74 10
[3] Gan’shina E, Granovsky A, Dieny B et al 2001 Physika B 299 260
[4] Kravets V G, Petford-Long A K, Kravets A F 2000 Journal of Applied Physics 87 1762
[5] Murzina T V, Gan’shina E A, Guschin Vet al 1998 Applied Physics Letters 73 3769
[6] Gan’shina EA, Vashuk M V, Vinogradov A N et al 2004 JETP 98 1027
[7] Niklasson G A, Granqvist C G 1984 J. Appl. Phys 55 3382
[8] Sheng P 1980 Phys. Rev. Lett. 45 60
[9] Granovsky A, Kuzmichev M, Clerc J P 1999 J. Magn. Soc. Japan 23 382
[10] Granovsky A B, Kuz’michev M V, Yurasov A N 2000 Bulletin of Moscow State University, Series Physics. Astronomy 6 67
[11] Yurasov A N 2016 Russian technological journal 1 10
[12] Aleshnikov A A, Kalinin Yu E, Sitnikov A V, Fedosov A G 2012 Promising materials. “Interkontakt Nauka” 5 68
[13] Yurasov A N, Yashin M M 2018 Russian technological journal 6 2
[14] Yurasov A, Gan’shina E, Sokolov A, Granovsky N, Zazymkina D 2018 EPJ Web of Conferences 185 02009