The Symmetrised Maxwell-Garnett Approximation for Magneto-Optical Spectra of Ferromagnetic Composites

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We develop the symmetrised Maxwell-Garnett (SMG) approximation for magneto-optical spectra calculations by introducing an average over two following random units: inclusion of A coated by shell of B and inclusion of B coated by shell of A. Thus the symmetrised Maxwell-Garnett approach is the modified version of both Maxwell-Garnett and Bruggeman theories for arbitrary values of ferromagnetic volume fraction. The calculated spectra of metal-metal granular alloys in Bruggeman and SMG approaches are closed to each other, but this is not the case for metal-insulator composites, especially when close to percolation threshold. It is shown that magneto-optical response of the ferromagnetic composites is extremely sensitive to the value of the percolation threshold and hence to the microstructures of the composites.

Keywords: effective medium theory, percolation threshold, magneto-optical spectra, metal-insulator composites.

1. Introduction

Ferromagnetic metal-metal and metal-insulator granular alloys have been a subject of great interest in the last few years, since they exhibit unique physical properties: giant magnetoresistance (GMR)¹⁾, tunneling magnetoresistance²⁾ and giant extraordinary Hall effect³⁾. Recently it was shown that magneto-optical spectra (MOS) at visible wavelength of both granular $\text{Co}_x\text{Ag}_{1-x}$, $\text{Co}_x\text{Cu}_{1-x}$, $(\text{CoFe})_x\text{Ag}_{1-x}$ alloys with GMR and metal-insulator composites $\text{Co}_x(\text{CuO})_{1-x}$, $\text{Co}_x(\text{Al}_2\text{O}_3)_{1-x}$ are quite different from those of homogeneous alloys^{4) 5)}. Notwithstanding the fact that optical spectra of heterogeneous alloys, metal-insulator composites in particular, were extensively discussed in literature in framework of effective medium theories^{6)–9)}, there is no at present even qualitative understanding of MOS for granular ferromagnets, especially for ferromagnetic composites.

Optical spectra of solids are determined by diagonal part of the dielectric function $\epsilon_{xx}(\nu)$, where ν is the frequency of light. MOS can be measured in Faraday or Kerr effect geometry and they depend on both diagonal $\epsilon_{xx}(\nu)$ and nondiagonal $\epsilon_{xy}(\nu)$ terms. Therefore MOS are more complex and more sensitive to the electronic and ionic structure and can give additional and useful informations about granular alloy microstructure.

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The two basic Maxwell-Garnett (MG) and Bruggeman (BA) approaches were used for interpretation of MOS of granular ferromagnets^{4), 10) 11)}. The first approach is valid only for the small volume fraction of one or another component. In the framework of MG approximation there is no percolation threshold for electrical conductivity in metal-insulator composites⁸⁾. The Bruggeman approach (BA), being equivalent to the effective medium approximation or to the coherent potential approximation (CPA) theory of random alloys applies to a greater extent to the aggregate structure than to the granular alloys^{7) 8)}.

In this paper we develop the symmetrised Maxwell-Garnett (SMG) approximation for MOS calculations and present the results showing the advantages of this approach over MG and Bruggeman theories.

2. Theory

In linear approximation on magnetic field and magnetization the dielectric function of granular ferromagnet magnetized along the z direction ($\hat{\epsilon}$), its ferromagnetic ($\hat{\epsilon}_m$) and nonmagnetic ($\hat{\epsilon}_0$) constituents can be written as

$$\hat{\epsilon} = \begin{pmatrix} \epsilon & i\gamma & 0 \\ -i\gamma & \epsilon & 0 \\ 0 & 0 & \epsilon \end{pmatrix}; \quad \hat{\epsilon}_m = \begin{pmatrix} \epsilon_1 & i\gamma_1 & 0 \\ -i\gamma_1 & \epsilon_1 & 0 \\ 0 & 0 & \epsilon_1 \end{pmatrix};$$

$$\hat{\epsilon}_0 = \begin{pmatrix} \epsilon_0 & 0 & 0 \\ 0 & \epsilon_0 & 0 \\ 0 & 0 & \epsilon_0 \end{pmatrix}. \quad (1)$$

All the parameters in Eq.(1) contain real and imaginary parts and are the functions of frequency ν , for example $\epsilon_1(\nu) = \epsilon'_1(\nu) - i\epsilon''_1(\nu)$, $\gamma_1(\nu) = \gamma'_1(\nu) - i\gamma''_1(\nu)$. Knowing the parameters ϵ and γ for composite, MOS is calculated by well known relations. For example transverse Kerr effect (TKE) parameter δ is

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

where φ is the light incident angle,

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

In the framework of effective medium theories the dielectric permeability $\hat{\epsilon}$ can be calculated as a function of the constituent dielectric permeabilities $\hat{\epsilon}_m$ and $\hat{\epsilon}_0$, vol-

ume fraction of ferromagnetic component f_i and granular shape. The corresponding sets of equations in MG (Eqs.(3,4)) and Bruggeman (Eqs.(5,6)) approximations are as follows (for details see ref. 4, 10, 11, 12)

$$\frac{\varepsilon^{MG} - \varepsilon_0}{\varepsilon_0 + (\varepsilon^{MG} - \varepsilon_0)L_j} - f_1 \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_0 + (\varepsilon_1 - \varepsilon_0)L_j} = 0 \quad (3)$$

$$\gamma^{MG} / [\varepsilon_0 + (\varepsilon^{MG} - \varepsilon_0)L_j]^2 - f_1 \gamma_1 / [\varepsilon_0 + (\varepsilon_1 - \varepsilon_0)L_j]^2 = 0 \quad (4)$$

$$f_1 (\varepsilon_1 - \varepsilon^{BA}) / (\varepsilon^{BA} + (\varepsilon_1 - \varepsilon^{BA})L_j) + (1 - f_1)(\varepsilon_0 - \varepsilon^{BA}) / (\varepsilon^{BA} + (\varepsilon_0 - \varepsilon^{BA})L_j) = 0 \quad (5)$$

$$f_1 (\gamma^{BA} - \gamma) / (\varepsilon^{BA} + (\varepsilon_1 - \varepsilon^{BA})L_j)^2 + (1 - f_1) \gamma^{BA} / (\varepsilon^{BA} + (\varepsilon_0 - \varepsilon^{BA})L_j)^2 = 0, \quad (6)$$

where L_j denotes the granular demagnetization factor for the direction along the magnetic field.

To develop the SMG approximation for MOS calculations we will follow the ideas of ref. 7-9. The SMG approach for diagonal dielectric function has proposed ⁷⁻⁹ by introducing an average over two following random units:

Fig.1 Random units A and B in the effective medium (SMG approximation).

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.1). The probabilities to find these units in composites (for confoal spheroidal units) are

$$P_A = u_1 / (u_1 + u_2); \quad P_B = u_2 / (u_1 + u_2) \quad (7)$$

where $u_1 = (1 - f_1^{1/3})^3$; $u_2 = [1 - (1 - f_1)^{1/3}]^3$.

Then it is easy to calculate the dielectric functions for both types of units in MG approximation by the following equations

$$\frac{\varepsilon_A^{MG} - \varepsilon_0}{\varepsilon_0 + (\varepsilon_A^{MG} - \varepsilon_0)L_{jA}} - f_1 \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_0 + (\varepsilon_1 - \varepsilon_0)L_{jA}} = 0 \quad (8)$$

$$\frac{\gamma_A^{MG}}{[\varepsilon_0 + (\varepsilon_A^{MG} - \varepsilon_0)L_{jA}]^2} - f_1 \frac{\gamma}{[\varepsilon_0 + (\varepsilon_1 - \varepsilon_0)L_{jA}]^2} = 0 \quad (9)$$

$$\frac{\varepsilon_B^{MG} - \varepsilon_0}{\varepsilon_0 + (\varepsilon_B^{MG} - \varepsilon_0)L_{jB}} - f_1 \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_0 + (\varepsilon_1 - \varepsilon_0)L_{jB}} = 0 \quad (10)$$

$$\frac{\gamma_B^{MG}}{[\varepsilon_0 + (\varepsilon_B^{MG} - \varepsilon_0)L_{jB}]^2} - f_1 \frac{\gamma}{[\varepsilon_0 + (\varepsilon_1 - \varepsilon_0)L_{jB}]^2} = 0 \quad (11)$$

(L_{jA} and L_{jB} denotes the demagnetization factors of units A and B, correspondingly).

Then SMG approximation can be obtained by introducing these values in the Bruggeman equations (5,6) putting $\varepsilon_1 = \varepsilon_A^{MG}$, $\varepsilon_0 = \varepsilon_B^{MG}$; $\gamma_1 = \gamma_A^{MG}$ and $f_i = P_A$:

$$P_A (\varepsilon_A^{MG} - \varepsilon) / (\varepsilon + 1/2 (1 - L_A)(\varepsilon_A^{MG} - \varepsilon)) + P_B (\varepsilon_B^{MG} - \varepsilon) / (\varepsilon + 1/2 (1 - L_B)(\varepsilon_B^{MG} - \varepsilon)) = 0 \quad (12)$$

$$P_A (\gamma - \gamma_A^{MG}) / [\varepsilon + 1/2 (1 - L_A)(\varepsilon_A^{MG} - \varepsilon)]^2 + P_B (\gamma - \gamma_B^{MG}) / [\varepsilon + 1/2 (1 - L_B)(\varepsilon_B^{MG} - \varepsilon)]^2 = 0 \quad (13)$$

where we have used the relations $L_{xx} = L_{yy} = 1/2(1 - L_z)$ and $L_{zz} = L^{(11)}$. The set of Equations (12, 13) is the final result of SMG approach for calculations of both optical and magneto-optical properties of granular solids.

3. Results and discussion

From Eqs.(12-13) and the definition of random units in Fig.1 it follows that SMG approach is equivalent to MG approximation at $f_1 \ll 1$ and $(1 - f_1) \ll 1$, and is modified version of both MG and Bruggeman methods for the arbitrary value of ferromagnetic volume fraction f_1 . Since the MOS calculations in SMG approach are rather tedious (as they include at the first stage the solution of Eqs. (8-11)) it is relevant to clarify the difference in results obtained by various effective medium theories. To answer this question we calculated the MOS in MG, Bruggeman and SMG approaches for metal-metal granular alloys $\text{Co}_f\text{Ag}_{1-f}$, $\text{Co}_f\text{Cu}_{1-f}$ and metal-insulator composites $\text{Co}_f(\text{CuO})_{1-f}$, $\text{Co}_f(\text{Al}_2\text{O}_3)_{1-f}$.

Fig. 2 Spectral dependencies of transverse Kerr effect (TKE) parameter δ for $\text{Co}_f\text{Cu}_{1-f}$ granular alloys at different ferromagnetic volume fraction f calculated in framework of Maxwell-Garnett (MG), Bruggeman (BA) and symmetrised Maxwell-Garnett (SMG) approximations ($\varphi = 70^\circ$, $L_A = L_B = 0.33$). The experimental data are from ref.4.

For all investigated systems the MOS obtained by these three schemes results at $f_1 \ll 0.1$ or at $(1-f_1) \ll 0.1$ do not differ noticeably. Moreover for metal-metal granular alloys the calculated spectra in Bruggeman and SMG approaches are close to each other for arbitrary values of ferromagnetic component (Fig.2). Thus for two-phase inhomogeneous systems when optical parameters of components do not differ significantly, SMG approach gives the same result for MOS as more simple effective medium theory does. Thus the conclusions made in ref. 4 from comparison of the experimental data with calculated spectra in Bruggeman approach are quite reasonable. For ferromagnetic metal-insulator composites the situation is more complex (Figs.3-5).

Fig. 3 Spectral dependencies of TKE parameter δ for composite $Co_f(CuO)_{1-f}$ at $f=0.866$ in SMG and Bruggeman approximations ($L_A=L_B=1/3$). The experimental data are from ref.5.

Fig. 4 Spectral dependencies of TKE parameter δ for composite $Co_f(CuO)_{1-f}$ at $f=0.34$ in SMG and Bruggeman approximations ($f_c^{BA} \leq f_c^{SMG}$). $L_A=L_B=1/3$, $f_c^{BA}=0.333$, $f_c^{SMG}=0.455$. The experimental data are from ref. 5.

The SMG and Bruggeman approaches give the different values of percolation threshold $f_c^{(8)}$ and MOS dramatically depend on this phase transition. Both Bruggeman and SMG theories give qualitatively (but not quantitatively) the same results at large enough metallic content when $f > f_c^{BA}$ and $f > f_c^{SMG}$ (Fig.3), but quite different in amplitude when $f_c^{BA} < f < f_c^{SMG}$ (Fig.4). Thus magneto-optical response of ferromagnetic composites is extremely sensitive to the value of the percolation threshold f_c and hence to

Fig. 5 Spectral dependencies of TKE parameter δ for composite $Co_f(CuO)_{1-f}$ at $f=0.4$ in SMG approach for different values of demagnetization factors L_A and L_B ($1-L_A=0.61$, $L_B=0.58$, $f_c=0.419$; $2-L_A=0.54$, $L_B=0.58$, $f_c=0.416$; $3-L_A=0.47$, $L_B=0.58$, $f_c=0.413$). The experimental data are from ref. 5 for $f=0.418$.

microstructures of the composites. The reliable effective medium theory of MOS must first of all to provide the correct value of f_c . Since SMG approach can give a wide range of values for f_c by putting reasonable values of demagnetizing factor (Fig.5) and more correctly describes the dependence of percolation threshold on composite microstructure, this theory is more general than Bruggeman approximation. The experimental study of MOS for composite ferromagnets near f_c can give a new insight in percolation problem and can shed light on the fundamental relations between microstructure and electromagnetic response of inhomogeneous media.

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