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Indirect Exchange Interaction Fields in Magnetic Nanogranular Films

Yu.I. Dzhezherya, A.F. Kravets*, I.M. Kozak†, A.Ya. Vovk, A.M. Pogorily

Institute of magnetism, National Academy of Sciences of Ukraine, Vernadsky 36b, 03142 Kyiv, Ukraine

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The investigation results of the influence of non-magnetic matrix conducting properties on the magnetization process of magnetic nanogranular films are presented. Effective field in the films with conducting matrix influences the properties of the magnetic subsystem and facilitates the establishment of magnetic correlation in the ensemble of ferromagnetic granules. Polarization of the electron subsystem leads to the appearance of the exchange interaction between magnetic moments of the nearest magnetic granules. One can state about initiation of indirect exchange interaction in the case when the field of indirect exchange will exceed the value of local magnetostatic fields.

Keywords: Magnetic nanogranular films, Magnetization, Indirect exchange interaction, Indirect exchange interaction field, Structural percolation.

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1. INTRODUCTION

Magnetic nanogranular materials containing nanogranules of metallic ferromagnet in non-magnetic matrix (metal or dielectric) attract a lot of attention due to numerous striking physical effects appearing there in the applied magnetic field. Thus, in magnetic nanogranular materials one can observe the giant magnetoresistance [1-5], tunneling magnetoresistance [6-10], giant Hall effect [11], magneto-refractive effect [12-15], magnetoinduced nonlinear optical effect [16], etc. These effects are of great interest both from the fundamental point of view and for the practical application in magnetic field sensors [17], magnetic media for high-density information recording [18] and high-frequency devices [19].

If differences in the transport properties of magnetic nanogranular films with different types of matrices are evident [20, 21], then interpretation of distinctions in the behavior of such systems during magnetization faces certain difficulties.

Collective behavior of magnetic moments of separate granules depends on the interaction between them.

Indirect exchange interaction of the Ruderman-Kittel-Kasuya-Yosida (RKKY) type [22] can be one of the most known interactions between magnetic granules in conducting non-magnetic medium. However, as the analysis shows, the RKKY interaction in the ensemble of nanogranules is relatively small due to extraordinarily little impact on a separate magnetic granule from the side of neighboring magnetic granules, as well as because of fast spatial oscillations of the RKKY exchange interaction integral and its averaging to zero over volume.

Another mechanism of indirect exchange can be connected with the polarization of subsystem of conduction electrons due to the *s-d*-exchange in the system with the long-range magnetic order [23]. Indeed, if magnetize the ensemble of magnetic nanogranules staying initially in the superparamagnetic state by an external field, then magnetic polarization of *s* conduction electrons will be also realized in the whole material volume. Influence of the magnetopolarized medium filling the whole space is identical to the effective magnetic field which facilitates the establishment of the magnetic ordering in ensemble of nanogranules [24]. In contrast to metal nanogranular films, collective behavior of the ensemble of nanogranules in metal-dielectric films becomes substantial only if the structural percolation threshold is achieved [23].

Based on the analysis of the results of ferromagnetic resonance in magnetic nanogranular films with different types of non-magnetic matrixes it was discovered in the work [23] that additional correlation mechanism exists in materials with conducting matrix between magnetic moments of nanogranules. It can be connected with indirect exchange interaction through the conduction band electrons. Contrariwise, in nanogranular materials with dielectric matrix, indirect exchange is characterized by a small value, since at concentrations of the magnetic material lower than the structural percolation threshold it is conditioned by the effects of spin-dependent tunneling of electrons from a granule into a granule through a high energy barrier [6-10].

It was shown for CoFe-Cu granular alloys [25] that magnetic and transport properties can be explained by: (a) dipolar interactions between closely spaced magnetic particles in metal matrix, (b) indirect ferromagnetic exchange through the metal matrix due to CoFe doping which disappears during annealing, (c) perpendicular to the plane uniaxial magnetic anisotropy following from the strain of the crystal lattice which disappears after annealing. The model of superparamagnetic interaction which was successfully tested in different magnetic systems is used in the dipole-dipole interacting metal and metal-dielectric granular films [26].

In the theoretical part of the present work we have proposed the phenomenological approach to the indirect exchange interaction in magnetic nanogranular materials with conducting non-magnetic matrix. Effective field of indirect exchange for granular films is determined from the field and temperature dependences of the magnetization. Calculation results are compared with the experimental data of switching in granular films with different non-magnetic matrixes.

* anatolii@kth.se
† irinkm@ukr.net

2. DETERMINATION OF THE INDIRECT EXCHANGE FIELD VALUE IN MAGNETIC NANOGRANULAR FILMS

The value of the effective field of indirect exchange can be determined in a simple way from the analysis of magnetization curves of nanogranular films measured at different temperatures.

To make the calculations more general, we suppose that ferromagnetic granules possess anisotropy and write their energy in the following form:

$$U_i = -\frac{\beta_i}{2v_i}\left(\mathbf{m}_i\mathbf{n}_i\right)^2 - \mathbf{m}_i\left(\mathbf{H}_i^m + \mathbf{H} + \mathbf{h}\right), \qquad (1)$$

where index "*i*" indicates the granule number; $\mathbf{m}_i$ is the granule magnetic moment; v_i is the granule volume; β_i is the constant of easy-axis anisotropy (can be defined by the form-factor or type of the crystal structure of granule material); n_i is the unit vector along the direction of the granule anisotropy axis; $\mathbf{H}_i^m$ is the magnetostatic field in the region of granule disposition; h is the indirect exchange interaction field, the determination of which is the subject of investigation of the given section.

For the development of the algorithm for the determination of the h value, we will use the elements of the Langevin theory considering that system of granules is in the superparamagnetic state.

At the constant module of the granule magnetic moment, its average value is defined by the expression

$$\left\langle \mathbf{m}_i \right\rangle = \frac{\int d\Omega_i \mathbf{m}_i \exp\left(-U_i/kT\right)}{\int d\Omega_i \exp\left(-U_i/kT\right)}, \qquad (2)$$

where k is the Boltzmann constant; T is the system temperature; $d\Omega_i$ is the solid angle element; integration is performed over all possible directions of the magnetic moment.

Expression (1) for the potential energy contains the term which takes into account the magnetodipole contribution $\mathbf{H}_i^m$. Since this value depends on the mutual arrangement of granules and the spatial orientation of their magnetic moments, then precise accounting of the given contribution is difficult. Therefore, in the future when writing $\mathbf{H}_i^m$ we will use the mean field approximation assuming that magnetic granules are located in space without significant density fluctuations [27, 28]. Then, approximately we can write

$$\mathbf{H}_i^m \approx -4\pi \cdot M_z \mathbf{e}_z. \qquad (3)$$

The given value is defined by the shape of the studied system. In our case, the sample represents a thin film, whose plane is orthogonal to the *Oz*-axis. Since in the mean field approximation the value of $\mathbf{H}_i^m$ is the same for all granules, then henceforth we will omit the index, which determines the granule number.

When considering we will assume that magnetic energy of granules considerably less than the thermal one ($|U_i| << kT$). This condition can be always satisfied at sufficiently high temperatures.

We will perform the expansion in powers of U_i/kT to quadratic terms inclusive in the numerator and denominator of expression (2)

$$\left\langle \mathbf{m}_i \right\rangle \approx \left(1/4\pi\right)\int d\Omega_i \mathbf{m}_i \left(1 - U_i \Big/ kT + \frac{1}{2}\left(U_i/kT\right)^2\right) + \left(1/4\pi\right)^2 \left(\int d\Omega_i U_i/kT\right)\left(\int d\Omega_i \mathbf{m}_i \left(1 - U_i/kT\right)\right). \qquad (4)$$

After substitution of U_i into (4) and integration over all possible directions of the magnetic moment we have

$$\left\langle \mathbf{m}_i \right\rangle = \frac{M_i^2}{3kT}\left(1 - \frac{\beta M_i^2}{6v_i kT}\right)\left(\mathbf{H} + \mathbf{H}^m + \mathbf{h}\right) + \mathbf{e}^\alpha \frac{\beta_i}{2v_i} n_i^\lambda n_i^\gamma \left(H^\nu + H^{m\nu} + h^\nu\right) \times \times \frac{1}{4\pi}\int d\Omega_i m_i^\alpha m_i^\lambda m_i^\nu m_i^\gamma. \qquad (5)$$

Indices denoted by the Greek letters $\alpha, \lambda, \gamma, \nu$ show the spatial components of vectors. Summation is carried out over double indices.

Then, we take into account the known relation

$$\frac{1}{4\pi}\int d\Omega_i m_i^\alpha m_i^\lambda m_i^\nu m_i^\gamma = \frac{m_i^4}{15}\left(\delta_{\alpha\lambda}\delta_{\nu\gamma} + \delta_{\alpha\gamma}\delta_{\nu\lambda} + \delta_{\alpha\nu}\delta_{\lambda\gamma}\right), \qquad (6)$$

where $\delta_{\alpha\lambda}$ is the Kronecker symbol.

Whereupon, expression for the average magnetic moment of a granule takes the form

$$\left\langle \mathbf{m}_i \right\rangle = \left(m_i^2/3kT\right)\left(\mathbf{H} + \mathbf{H}^m + \mathbf{h}\right) + \mathbf{e}^\alpha \frac{\beta_i}{v_i}\frac{m_i^4}{45\left(kT\right)^2}\left(H^\nu + H^{m\nu} + h^\nu\right)\left(3n_i^\alpha n_i^\nu - \delta_{\alpha\nu}\right). \qquad (7)$$

Expression (7) determines the average value of the magnetic moment of a separate granule with account for the spatial orientation of the anisotropy axis. However, only the integral characteristic of magnetization can be obtained in the measurements. Therefore, based on the expression (7) we present the average magnetization of the material $\mathbf{M} = \left(1/V\right)\sum_i \left\langle \mathbf{m}_i \right\rangle$ in the form

$$\mathbf{M} = \chi_0\left(T\right)\left(\mathbf{H} + \mathbf{H}^m + \mathbf{h}\right) + \mathbf{e}_\alpha \cdot \chi_{\alpha\nu}\left(T\right)\left(H^\nu + H_i^{m\nu} + h^\nu\right), \qquad (8)$$

where $\chi_0\left(T\right) = V^{-1}\sum_i m_i^2/3kT$ and

$$\chi_{\alpha\nu}\left(T\right) = V^{-1}\sum_i \left(\beta_i/5v_i\right)\left(m_i^2/3kT\right)^2\left(3n_i^\alpha n_i^\nu - \delta_{\alpha\nu}\right).$$

Noting further that values β_i/v_i, m_i and n_i^α for the ensemble of granules are statistically independent, we rewrite the values of coefficients χ_0 and $\chi_{\alpha\nu}$ as

$$\chi_0\left(T\right) = \rho\left\langle m^2\right\rangle \Big/ 3kT, \quad \chi_{\alpha\beta} = \frac{1}{45\left(kT\right)^2}\rho\left\langle m^4\right\rangle\left\langle \beta/v\right\rangle\left\langle 3n_\alpha n_\nu - \delta_{\alpha\nu}\right\rangle, \qquad (9)$$

where ρ is the number of granules in the volume unit.

Angular brackets in (9) denote averaging.

It is not too difficult to make sure that in the case of isotropic in directions distribution of anisotropy axes of

granules expression for $\chi_{\alpha\nu}$ becomes zero. If directional processes took place in the production of the samples, it would be affected the tensor components $\chi_{\alpha\nu}$. One should also pay attention to the connection between its diagonal components $\chi_{xx}+\chi_{yy}+\chi_{zz}=0$.

It follows from expression (9) that with increasing temperature tensor components $\chi_{\alpha\nu}$ decrease faster than the value of χ_0 that substantially simplifies consideration for high-temperature region.

Thus, at high temperatures the processes of system magnetization can be described by equation

$$\mathbf{M}=\chi_0(T)\cdot(\mathbf{H}-4\pi\mathbf{e}_z M_z+\mathbf{h}). \qquad (10)$$

If external field is oriented in the film plane, then expression for the indirect exchange field **h** can be represented in the following form:

$$\mathbf{h}=\frac{\mathbf{M}}{\chi_0(T)}-\mathbf{H}. \qquad (11)$$

In order to determine h on the ground of relation (11), we consider two magnetization curves obtained at rather high temperatures T_1 and T_2 (Fig. 1). Absence of hysteresis implies that system of nanogranules is in the superparamagnetic state. Plot on the graph a straight line corresponding to a certain value of magnetization M. This straight line will cross magnetization curves in the points with abscissas H_1 and H_2. Thus, for one and the same sample, the states defined by the parameters (H_1, T_1) and (H_2, T_2) are characterized by the same magnetization value and, therefore, by the same value of indirect exchange field.

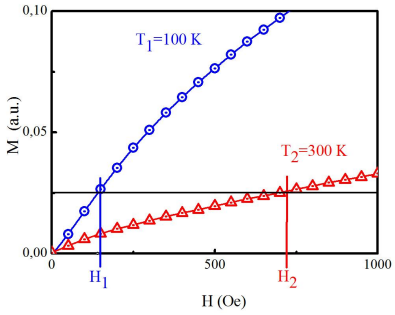

Fig. 1 – Scheme for the calculation of the effective value of indirect exchange field h_{eff} in magnetic nanogranular films

Here, for the determination of h value, one can obtain the formula based on the experimental data

$$\frac{h+H_1}{h+H_2}=\frac{T_1}{T_2}. \qquad (12)$$

Finally we have

$$h=\frac{H_2T_1-H_1T_2}{T_2-T_1}. \qquad (13)$$

We should especially emphasize that one can state about the manifestation of indirect exchange interaction only if h in magnitude will exceed the value of local magnetostatic fields. Therefore, quantitative criterion of the usability condition of the developed theory can be formulated as follows:

$$h >> 4\pi\langle M\rangle. \qquad (14)$$

Otherwise, the result (in magnitude) will be of the same order of magnitude as the errors introduced in the derivation of formula (13).

The proposed model was tested on two series of nanogranular films with metal $(Co_{50}Fe_{50})_xAg_{1-x}$ and dielectric $(Co_{50}Fe_{50})_x(Al_2O_3)_{1-x}$ matrixes (x is the volume fraction of magnetic material). These films were deposited under the same technological conditions in an oil-free vacuum of ~ 10^{-4} Pa using the method of electron-beam co-evaporation of magnetic and non-magnetic components from two sources. Details of the deposition process, structural and magnetic properties of nanogranular films are presented in [10, 15]. Magnetic hysteresis loops were measured at 10, 100, and 300 K using the SQUID magnetometer in magnetic fields to 50 kE. Magnetic measurements were carried out in the temperature range of 5-300 K in magnetic field of 50 E. It was shown that films of the composition $x < 0.2$ should be considered as the ensemble of superparamagnetic nanoparticles at the temperature higher than 75 K. The average size of magnetic nanoparticles determined from the measurements of the magnetic susceptibility is the same for both Ag and Al_2O_3 matrixes. In particular, magnetic nanoparticles of the size of ~ 1.5 nm are typical for films of the composition $x = 0.13$ [29]. Thus, topology and spatial arrangement of magnetic material for metal and metal-dielectric granular films of the given composition are the same. Low-field parts of the magnetization curves for metal $(Co_{50}Fe_{50})_{0.13}Ag_{0.87}$ and metal-dielectric $(Co_{50}Fe_{50})_{0.13}(Al_2O_3)_{0.87}$ films are represented in Fig. 2a and 2b, respectively. Calculated values of the indirect exchange field as the function of medium magnetization are shown in Fig. 3. As seen, the exchange interaction in the films with conducting metal matrix considerably higher in comparison with the dielectric matrix. One can observe a clear linear dependence of h on the magnetization M. This allows to introduce the indirect exchange interaction constant $k = h / M$ (see Fig. 4). This constant for metal nanogranular films is almost one order of magnitude larger in comparison with metal-dielectric films.

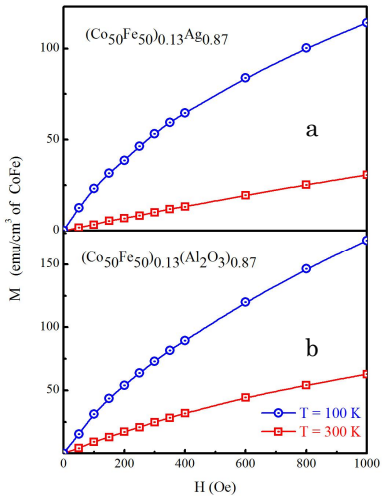

Fig. 2 – Low-field parts of the magnetization curves for metal $(Co_{50}Fe_{50})_{0.13}Ag_{0.87}$ (a) and metal-dielectric $(Co_{50}Fe_{50})_{0.13}(Al_2O_3)_{0.87}$ (b) nanogranular films measured at 100 K and 300 K

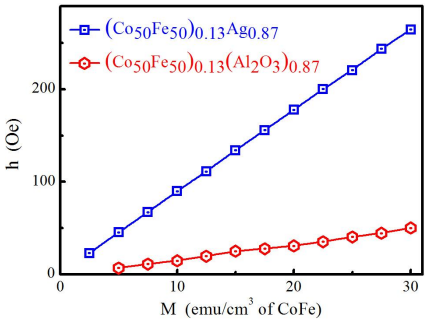

Fig. 3 – Dependences of the effective values of indirect exchange interaction fields h_{eff} on the magnetization M for metal $(Co_{50}Fe_{50})_{0.13}Ag_{0.87}$ and metal-dielectric $(Co_{50}Fe_{50})_{0.13}(Al_2O_3)_{0.87}$ films

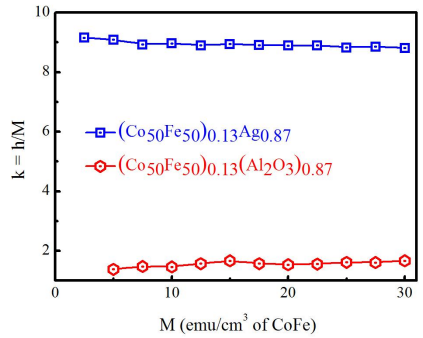

Fig. 4 – Dependences of the indirect exchange interaction constant k on the magnetization M for metal $(Co_{50}Fe_{50})_{0.13}Ag_{0.87}$ and metal-dielectric $(Co_{50}Fe_{50})_{0.13}(Al_2O_3)_{0.87}$ films

As could be expected, the value of effective interaction field of magnetic granules in the medium with dielectric matrix is insignificant and comparable with the error value of the theory developed in the work. Magnetic interaction in such system becomes appreciable only near the structural percolation threshold. At the same time, effective field in the films with conducting matrix substantially influences the properties of magnetic subsystem and facilitates the establishment of magnetic correlation in the ensemble of ferromagnetic granules.

3. CONCLUSIONS

The phenomenological model of the indirect intergranular exchange interaction in magnetic nanogranular materials is proposed in the work. In the framework of the given model it is shown that at small concentrations of ferromagnetic material collective behavior of the ensemble of nanogranules depends largely on the indirect exchange interaction with polarized subsystem of free electrons. The given interaction is exhibited in the systems with conducting matrix. Otherwise, in nanogranular materials with dielectric matrix indirect exchange has a small value, since below the structural percolation threshold it is conditioned by the effects of spin-dependent tunneling of electrons from a granule into a granule through a high energy barrier. Conclusions of the theoretical calculations obtained within the model of indirect exchange are confirmed experimentally.

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