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Peculiarities of temperature behavior of magnetization in Co/Ge/Co films

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Abstract

In Letter the results of experimental and theoretical investigations of magnetic properties in trilayer Co/Ge/Co films are represented. The temperature dependences of films, being two phases in respect of magnetism, are studied. Experimental results are explained in the frameworks of impurity model, when the hexagonal cobalt grains are considered as quasi-Ising particles distributed chaotically and dissolved within an isotropic matrix of cubic cobalt. The fields of magnetization rise on the $T-H$ plane are determined.

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Interest to film materials in a *transition metal/semiconductor* system is caused, first of all, by possible practical application in spintronics devices [1]. But the investigation of physical properties presents no smaller interest in respect to problems of fundamental physics of solid state and magnetism. The possibility of control for properties of such structures by picking out of active layer and spacer materials and also creation of mesostructure required (granularity, modulation of active elements concentration and so on) permits not merely to construct new materials, but to reveal new physical phenomena.

Granular multilayer films are of interest in connection with existing in them large magneto resistive effect [2], as compared with films having homogeneous magnetic layers. General regularities of resistive properties of granular films are understudied on the whole, but the influence of magnetic structure on electron transport mechanisms in such systems is weakly studied. Also the question about mechanisms responsible for forming of magnetic state in such films is left in abeyance.

Earlier, it was found by us in trilayer Co/Ge/Co films that at magnetic layer thickness more 5 nm the hexagonal phase (hcp) is appeared together with the metastable cubic face-centered one (fcc). At the same time the ratio depends on both semiconducting layer thickness and magnetic layer one [3]. In the first case at the cobalt thicknesses $5 \leqslant t_{\mathrm{Co}} \leqslant 10$ nm the hexagonal phase portion rises with increasing of germanium thickness t_{Ge}. In the last case in all films investigated by us at every semiconducting spacer thickness the portion of hexagonal phase rises with increasing of total magnetic layer thickness. The semiconducting interlayer is peculiar catalyst under forming of cubic cobalt phase, inasmuch as, the direct correlation is observed between structure of the germanium interlayer and it of cobalt close-fitting. These results were earlier obtained by transmission electron microscopy (TEM) and confirmed by nuclear magnetic resonance measurements (NMR). It turned out, that the hexagonal cobalt is uniformly distributed on film volume as grain in the form of like spherical. Consequently, with respect to magnetic structure we have situation, when the strongly anisotropic magnetic particles (hexagonal cobalt) are distributed in practically isotropic medium (cubic cobalt).

Exactly, the present investigation is given up to study of peculiarities of magnetic behavior in such system.

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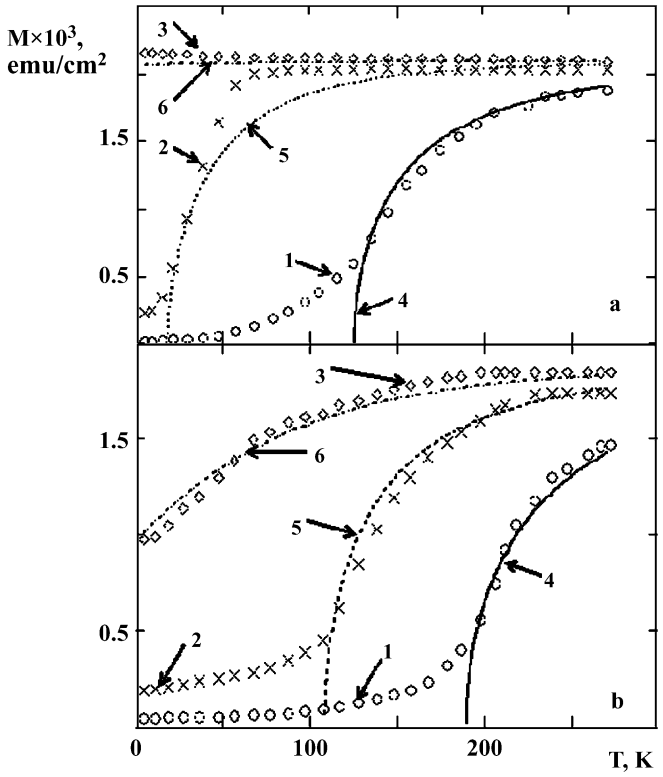

Fig. 1. Temperature dependences of magnetization of films 1 and 2 of composition Co/Ge/Co measured in different magnetic fields. Points are experiment; curves are calculations in formula (5). 1, 2—$H = 50$ Oe; 2, 5—$H = 200$ Oe; 3, 6—$H = 800$ Oe.

Temperature dependences of magnetization in low magnetic fields of such films are found unusual. Procedure of experimental data getting is expounded in paper [3]. Here note only, that before receiving of every experimental curve, from the beginning the sample was demagnetized and in zero magnetic fields was cooled to liquid helium temperature. Then magnetic field was switched on and, at this field fixed, the sample was heated. In Fig. 1 the magnetization dependences $M(T)$ are represented for films Co–Ge system with different thicknesses of magnetic and nonmagnetic layers (film 1—$t_{\mathrm{Co}} = 12$ nm, $t_{\mathrm{Ge}} = 2.4$ nm; film 2—$t_{\mathrm{Co}} = 13.2$ nm, $t_{\mathrm{Ge}} = 3.74$ nm) and, correspondingly, with different volume ratios of hexagonal and cubic phases $(q/(1-q))$. It is seen that in low magnetic field the magnetization is practically equal to zero up to some temperature (T_c), above that the sharp rise of magnetization and approaching to saturation are began. Such behavior is typical for all films investigated of this system. The difference is only in the fact that the rise of magnetization is occurred at different temperatures and magnetization values are different.

To analyze temperature properties of magnetization the model is proposed, when the strong anisotropic magnetic particles are solved in isotropic matrix and exchange coupled with it (expansion of Stoner–Wolhfarth model [4]). In Fig. 2 the model structure of film magnetic layer are represented. Here the direction of external magnetic field is chosen as Z-axis. In the general case for such situation the magnetic energy can be

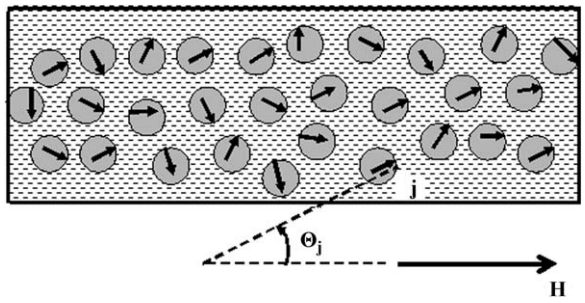

Fig. 2. Sketch of mesostructure of magnetic layer with strong anisotropic grains (spheres with arrows inside) in external magnetic field and orientation of grain anisotropy axes.

wrote in form

$$E = -tM_0H\cos\varphi - H\sum_j \mu_j \cos\alpha_j - \lambda M_0 \sum_j \mu_j \cos(\varphi - \alpha_j) - \sum_j D_j \cos^2(\Theta_j - \alpha_j), \quad (1)$$

where $M_0 = M_0(H)$ is magnetization of matrix in given magnetic field, φ is angle, determinative the magnetization direction, α_j is angle between direction of magnetic moment of jth grain and external magnetic field, $\mu_j = \mu_j(T, H)$ is granular magnetic moment, depending on temperature an magnetic field, λ is constant of exchange coupling between matrix and particle, $D_j > 0$ is grain constant of anisotropy, t is effective thickness of magnetic layer. Summation is made on all grains of system.

The matrix magnetization is determined as $M = M_s \cos\varphi_0$, where M_s and φ_0 are magnetization of saturation and equilibrium angle, respectively. One can wait that the contribution from granular subsystem will be small. Estimation of these will be made later. Equilibrium angles are found from conditions of energy minimum:

$$\frac{\partial E}{\partial \varphi} = tM_0H\sin\varphi + \lambda M_0 \sum_j \mu_j \sin(\varphi - \alpha_j) = 0, \quad (2a)$$

$$\frac{\partial E}{\partial \alpha_j} = H\mu_j \sin\alpha_j - \lambda M_0 \mu_j \sin(\varphi - \alpha_j) - D_j \sin\bigl[2(\Theta_j - \alpha_j)\bigr] = 0, \quad (2b)$$

under the stipulation that $(\partial^2 E/\partial\psi^2)_0 > 0$, $\psi = \{\varphi, \alpha\}$.

It is obvious that this system is not analytically solvable without simplifying assumptions. Now let us remember, that the energy of magnetic crystallographic anisotropy of the hexagonal cobalt is well above then it of the cubic cobalt. In low magnetic fields range the inequality $D_j \gg (H\mu_j, \lambda m\mu_j)$ is right. Under such conditions every particle, numbered by j index, behaves similar to quasi-Ising particle with local axis of anisotropy determined by Θ_j angle. As a zero approximation, neglecting by small items, from (2b) it follows, that $\alpha_j \cong \Theta_j$. To determine the φ angle we receive expression

$$\sin\varphi = \frac{1}{tM_0}\sum_j \mu_j \sin\Theta_j. \quad (3)$$

Next step consists in averaging-out of expression (3) on all directions of granular anisotropy axes. Also, for simplicity we

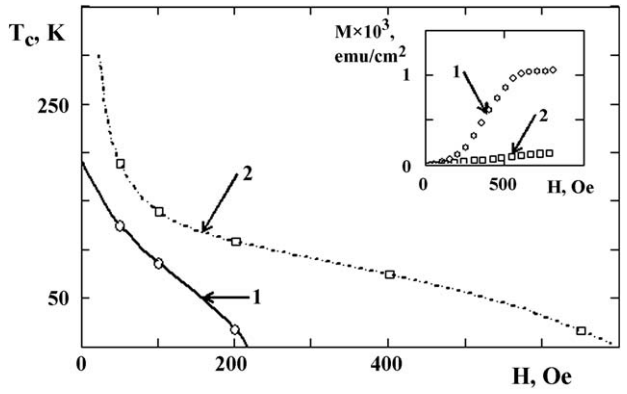

Fig. 3. Temperature of rise of film magnetization versus external magnetic field value. Points are extrapolation of experimental dependences up to intersection with abscise axis, curves are calculations (see text). In insert the field dependences of magnetization are given in these films at $T = 4.2$ K.

assume that all particles are identical, i.e., $\mu_j = \mu_0$, then their number on unit area of film equals $N = qt/v_0$ (here v_0 is grain volume). Since particles are represented as quasi-Ising, every particle has only two states. This implies that the average value of magnetic moment on the direction of external magnetic field is given by following expression [5]:

$$\mu(T, H, \Theta) = \mu_0(T) \tanh\left[2\frac{(H + \lambda m(H))\mu_0(T)\cos\Theta}{K_B T}\right]. \quad (4)$$

Finally we receive expression for magnetization:

$$M = 2tM_s\sqrt{1 - \left(\frac{q}{(1-q)}\frac{m(T)\langle\sin\varphi(T, H)\rangle}{M_0(H)}\right)^2}, \quad (5)$$

where $\langle\sin\varphi(T, H)\rangle = \frac{1}{\mu_0(T)}\int_0^{\pi/2}\sin^2\Theta\mu(T, H, \Theta)\,d\Theta$ and integration is carried out on upper semisphere [6], $\mu_0(T) = m(T)\frac{4\pi}{3}r^3$, $m(T)$ is magnetization of grain material, r is grain radius.

It is seen from (5) that to realize concrete calculations it is necessary to know: portion of hexagonal phase q, grain radius r, constant of exchange coupling λ between grain and matrix, dependences of magnetization of grain $m(T)$ and matrix $M_0(H)$ materials. Further we have taken q values from NMR experiments [3] and they are equal $q_1 = 0.12$ (for film 1) and $q_2 = 0.35$ (for film 2). As is the convention in other papers (for instance, the bases are explained in [7]), we shall be to consider that the grain magnetization is the same as of bulk hexagonal cobalt. The magnetization of bulk hexagonal cobalt at temperatures $T < 300$ K [8] is well approximated by function $m(T) = m_0(1 - 0.479(T/T_c)^{1.19})$, where $m_0 = 1442$ emu/cm^3 and T_c is Curie temperature. Matrix magnetization will be taken from experiment (insert in Fig. 3 for $M_0(H)$ and Fig. 2 for M_s) believing that it does not depend on temperature for both films (it is right for cubic cobalt in this temperature range). Then the r and λ are fitting parameters.

In Fig. 1 the temperature dependences, calculated on formula (5) for film 1 (part (a)) and for film 2 (part (b)) in different magnetic fields, are represented (curves 4, 5, 6). It turned out, that for film 1 value $\lambda_1 = 0.1$ and for film 2 $\lambda_2 = 62$ and as it was to be expected $\lambda > 0$. The gain radius is about the same $r \cong 1.91 \pm 0.001$ nm for both cases. It is seen that with fitting values of r and λ the experimental dependences $M(T)$ and calculations are rather well satisfied. The availability of nonzero magnetization at temperatures below T_c one can explain by a residual magnetization (owing to nonuniform distribution of grains in film volume) and dimension yield of grains. The last circumstance has essential influence on the $M(T, H)$ dependence. Other things being equal, the smaller grain dimensions, the lower temperature T_c. The rise of exchange coupling with increasing of hexagonal cobalt portion, evidently, is due to increasing of total area of surface of granular subsystem, yet the interaction between gains and matrix occurs on the phase interface. Decrease of the saturation magnetization is explained by lessening of total cubic cobalt portion, which gives main contribution to magnetization. The fact that good result is with using of magnetization values from experimental field dependences (insert in Fig. 3), it is pointed out that the grains feel the mean field by matrix caused. Proper contribution of granular subsystem to magnetization is determined in the following way $M_g = \langle\cos\alpha\rangle\mu_0(T, H)N$. The substitution of numerical data in this expression at $T = 5$ K and $H = 50$ Oe gives additional value of order $< 10^{-5}$, but no experimental 10^{-3}.

In Fig. 3 the theoretical temperature dependences of beginnings of nonzero magnetization on value of measurement field and experimental points, received by approximation of experimental curves, are represented. The T_c values are solution of equation which results from condition of equality to zero of radicand in (5). This result can consider as plotting of phase diagram in T–H coordinates. Here the $T_c(H)$ line is boundary separating state where the magnetization equals to zero (under the $T(H)$ curve) and it differs from zero (above the $T(H)$ curve). One can estimate minimal portion of impurity phase when the influence of granular subsystem begins to become apparent. With the set of parameters for film 1 at $T = 4.2$ K and $H = 1$ Oe we obtain the value $q_c = 0.0026$. One can assume, that this is concentration by amounting to which in the absence of external influences the system becomes spin-glass-like due to the competition of exchange coupling between matrix and granular subsystem and effect of external magnetic field on matrix magnetization. Physical meaning of results received is simple and consists in the fact that with temperature increasing the effect of granular subsystem on matrix diminishes due to decreasing of its magnetic energy. On reaching T_c the magnetic energy of matrix begins to prevail and the system acquires nonzero mean magnetic moment.

We become aware of roughness of model used. But, on initial stage the knot of the matter, determinative unusual behavior of magnetization in two phase system in presence of strong anisotropic granular subsystem, is understood. Further advancement on the path to insight of magnetic properties of such systems requires the overrunning of main field approximation taking into consideration finite quantity of magnetic anisotropy and its dependence on temperature. Also, it is necessary to continue experimental investigations with the purpose of control of parameters of granular subsystem (change grain and matrix material, making of structure modulated and so on).

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