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Spin-glass-like behavior of low field magnetisation in multilayer $(Gd/Si/Co/Si)_n$ films

G.S. Patrin^{a,b,*}, V.O. Vas'kovskii^c, D.A. Velikanov^a, A.V. Svalov^c, M.A. Panova^b

^a L.V. Kirenskii Institute of Physics, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk, 660036, Russia
^b Krasnoyarsk State University, prospect Svobodnyi, 79, Krasnoyarsk, 660041, Russia
^c Ural State University, prospect Lenina, 51, Ekaterinburg, 620083, Russia

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Abstract

The results of experimental investigations of magnetic properties of multilayer $(Gd/Si/Co/Si)_n$ films in low magnetic fields are represented. The spin-glass-like behavior of magnetization is found. The role of biquadratic exchange coupling in a forming of magnetic state of system is discussed.

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The use in multilayer magnetic films active layers of different chemical elements and manners of stacking of the components allows directionally to control the magnetic properties of constructed materials and very widens the diversity of observed effects [1]. In multilayer films, where the layers of 3d- and 4felements are combined, the materials are used with very different magnetic properties. Apart from a display of individual properties of layer material, the behavior of the whole system depends on the interaction at the interfaces. Modification of interfaces, by means of selection of technological regimes or introducing another interlaying material (for instance, semiconductor), influences on the exchange interactions and (or) magnetic anisotropy. In this plane the $(Gd/Si/Co/Si)_n$ magnetic films are interesting that the compensation point of magnetization is found with

Corresponding author. *E-mail address:* patrin@iph.krasn.ru (G.S. Patrin). the temperature changing and it is typical for the bulk ferrimagnet alloys. This effect is observed both in the films without silicon interlayer [2,3] and with it [4]. Only the compensation temperature (T_{comp}) of magnetization depends on the thickness of silicon interlayer. Also in these films the peculiarity in the temperature dependence of magnetization is experimentally found that is explained by magnetic field influence on the interlaying interaction [5]. The effects of magnetic field on the interlaying interaction and the temperature dependence of the exchange parameters are connected with existence of biquadratic contribution into exchange coupling [6,7]. In this number one can note the paper [8], where it is corresponded about inducing of the biquadratic exchange by magnetic field in the (Fe/SmCo) bilayer films. In such systems it is possible the display of properties not inherent to systems described by simple interactions because of the competitive contributions of opposite signs into interlayer interaction.

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Fig. 1. Temperature dependences of the magnetization of the film with $t_{Si} = 10$ Å received with heating. FC: cooling regime $H \neq 0$, ZFC: cooling regime H = 0. The field of cooling and measuring is H = 20 Oe.

In the present Letter the results of investigations of magnetic properties in the $(Gd/Si/Co/Si)_{20}$ multilayer films in low magnetic fields in the temperature range T = 4.2-300 K are represented. In the films investigated the thickness of each of cobalt and gadolinium layers were $t_{Co} = 30$ Å and $t_{Gd} = 75$ Å, respectively, and the thickness of the silicon interlayer was varied in the range $t_{Si} = 0-10$ Å. The technology of the films preparation, its certification, and measurement methodic are described in [5]. The magnetic field was in the film plane. Note, that at given thickness the magneto-active layers were in the amorphous state [9].

By performing temperature measurements of magnetization in the film with silicon interlayer $t_{Si} =$ 10 Å it was found that in low magnetic fields (H <100 Oe) the curves have different shapes in dependence on thermo-magnetic prehistory (the cooling of sample was in magnetic field (FC) or without it (ZFC)) (Fig. 1). In these measurements the magnetic field was H = 20 Oe at cooling and heating. In the case FC at T < 100 K the curve is copy both at heating of film and cooling it. Such behavior is similar to that is observed in spin glasses [10]. In Fig. 2 the temperature dependences of magnetization are represented in films $t_{Si} = 2, 5, 10$ Å (parts (a), (b), (c), respectively) received in the zero field cooling and measured in the fields H = 10, 20, 50, and 100 Oe (curves 1, 2, 3,



Fig. 2. Temperature dependences of the magnetization of the films received with heating. Cooling conditions—(ZFC). (a) $t_{Si} = 2 \text{ Å}$, (b) $t_{Si} = 5 \text{ Å}$, (c) $t_{Si} = 10 \text{ Å}$. Curves 1, 2, 3, 4 are obtained in fields H = 10, 20, 50, 100 Oe, respectively.

and 4, respectively). It is seen that spin-glass behavior becomes more obvious with the thickness silicon interlayer increasing. So, in the film with $t_{Si} = 2$ Å the cusp is only observed in the magnetic fields $H \le 15$ Oe. In the films with $t_{Si} = 5$ Å and $t_{Si} = 10$ Å the measurement fields, when the cusp is not revealed, equal $H \approx 100$ Oe and $H \approx 150$ Oe, respectively. In all cases at T = 4.2 K the FC curves reach on the values approximately equal to values which are at reverse motion on a curve of magnetization in the given measurement field (Fig. 3). In the (Gd/Co)₂₀ film a nothing of the kind is observed in the same experimental conditions. Also, at different thickness of silicon interlayer the field dependences of magnetization have



Fig. 3. Field dependences of the magnetization of the films (a) $t_{Si} = 2$ Å, (b) $t_{Si} = 5$ Å, (c) $t_{Si} = 10$ Å. Cooling conditions—ZFC. T = 4.2 K. Arrows indicate directions of the temperature variation.

different shape (Fig. 3). It is seen that the t_{Si} increasing causes the rising of magnetic field saturation and broadening of hysteresis region. At first, such shape of field dependences in the films with different thickness t_{Si} gives reason to affirm, that with the temperature lowering there is no "lockout" of a semiconductor interlayer, and the interaction between layers is maintained. Second, in the given range of silicon thicknesses the role of a semiconductor interlayer is boosted with increasing of its thickness.

As it is known [10], the spin glass state arises in systems with a many-minimum energy distribution, among their number, a series of the spin glass properties can be explained in the model of two-minimum potential [11]. With reference to our case the possibility of such situation is caused by existence of the biquadratic exchange interaction between magnetic lay-



Fig. 4. Surfaces of the magnetic energy (*E*) depending on the angles of magnetizations in layers φ_1 and φ_2 . Column 1: $\eta = 0.7$. Column 2: $\eta = 1.7$. (a) H = 5 Oe, (b) 50 Oe, (c) 300 Oe, (d) 600 Oe.

ers. If to assume, that the film consists of a lot of layers, so that it is possible not to take into account the effects connected with influence of boundary conditions, the magnetic energy of system with consideration of the Zeeman interaction has form:

$$E = E_l \cos(\varphi_1 - \varphi_2) + E_q \cos^2(\varphi_1 - \varphi_2) - H [M_1 \cos(\varphi_H - \varphi_1) + M_2 \cos(\varphi_H - \varphi_2)],$$

where E_l and E_q are the surface densities of energy of bilinear and biquadratic exchange, respectively, two last members are the Zeeman energy (E_{Zi}) of magnetic moments in different layers. Here *H* is external magnetic field, M_i is surface density of magnetic moment in *i*th kind of layers, φ_i are angles of corresponding magnetizations, φ_H determines the direction of external magnetic field, i = 1, 2 and signifies Gd

and Co, respectively. It is possible to estimate a degree of influence of the biquadratic exchange on change of the magnetic energy of system. In Fig. 4 the values of the normalized surface density of energy E are represented in dependence on the angles φ_1 and φ_2 at miscellaneous relations of energies of biquadratic and bilinear exchanges $\eta = (E_q/E_l)$. (In calculations the following values of parameters were used: $E_l = 1 \text{ erg/cm}^2$ [7], $M_{\text{Gd}} = 12 \times 10^{-3} \text{ emu/cm}^2$, $M_{\text{Co}} = 4 \times 10^{-3} \text{ emu/cm}^2$, and the two last are taken from our measurements.) In the plane of film the angles φ_1 and φ_2 are counted off from a direction of external magnetic field, i.e., $\varphi_H = 0$. As it is seen in this figure (parts (a) and (b)), if an external field H is absent or very low, for small values η there are two energetic minima of type a "trough" on dependence $E(\varphi_1, \varphi_2)$, moreover, $\varphi_2 - \varphi_1 = \pi$ (Fig. 4(1a)). With the η increasing these minima are splitted each on two (Fig. 4(2a)). By switching on the magnetic field and its further increasing the energy surface assumes an intricate form with many minimums (Fig. 4(1b)-(1d) and (2b)-(2d)). And in a high magnetic field $(E_{Zi} > E_l, E_q)$ the absolute energetic minimum is at angles $\varphi_1 \cong \varphi_2 \cong 0$.

Now the behavior of magnetization is qualitatively understandable depending on cooling conditions. When the magnetic field is absent, all energy minima have identical depth, and distribution of directions of the magnetization in different layers is equally probability. With cooling of film the states, attributed these directions, have equal degree of population and, as a result, average magnetization of all system is equal to zero. The magnetic field switching on changes the depth of minima and selects preferable direction for magnetization in layers. With heating, as the temperature is raised, there is an "overflow" in this isolated minimum and nonzero average magnetization of system is arose. With cooling in rather high magnetic field the system reaches immediately the absolute energetic minimum and acquires nonzero average magnetization, which is subsequently observed.

From the analysis of experimental data it follows that the rising of silicon interlayer thickness increases the relative role of the biquadratic exchange. At first, with t_{Si} rising the spin glass properties are powerfully displayed, and namely, the field of "interlock" (H_b) is higher (see Fig. 2). (H_b is field when in the ZFC regime with magnetic field switching on the magnetization takes a value equal to one in the FC regime.) And it is connected with a height of barrier separating energetic minima. Secondly, at magnetization of films the saturation field is increased too with the t_{Si} rising (see Fig. 3). This fact is explained if to take into consideration that in the saturated state the system is in absolute energetic minimum and, to attain this minimum in the equilibrium conditions, it is necessary that in the magnetic field this minimum was lower all other minima. But a barrier height between individual minima is determined by the contribution of the biquadratic exchange.

By investigating the $(Co/Si)_{12}$ films with the silicon thickness $t_{Si} > 30$ Å the effect of thermo-magnetic memory was found in the temperature dependence of magnetization [12], that authors of paper quoted ascribe to transition of system into superparamagnetic state owing to "switching off" of coupling between magnetic layers with the temperature lowering. In our multilayer films at given silicon interlayer thickness we do not connect the temperature dependences of magnetization observed with superparamagnetic state. It is known [13], the field dependences of magnetization, measured at different temperatures, graphing in the dependence on (H/T) in consideration of the temperature dependencies of saturation magnetization, coincide and it is confirmed in numerous experiments. As it is easy to be convinced (see Fig. 3(a) and (b)) in our case this is not carried out. If the superparamagnetic state takes place, then the question rises concerning the nature of energetic barriers separating different magnetic states of the layers. In classical superparamagnets nonequivalent states arise because of presence of a magnetic crystallographic anisotropy in a microparticle. In limit of an experimental error we have not found any magnetic anisotropy in the plane of the film. Moreover, the previous measurements [5,6] in magnetic fields $H \gg H_b$ display, that the interaction between layers has antiferromagnetic character, and all system behaves as a ferrimagnet. Although the technological conditions for preparation of all multilayer films were the same, in the films without the silicon interlayer there is observed nothing of the kind.

In summary, once again we note, that as it follows from our experiments, the reason of existence of the spin-glass state of multilayer films $(Gd/Si/Co/Si)_{20}$ is the availability of the biquadratic exchange interaction. The investigation of mechanisms responsible for formation of the magnetic state and more detail theoretical description will be made elsewhere.

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