Multilayer Co/Pd Films with Nanocrystalline and Amorphous Co Layers: Coercive Force, Random Anisotropy, and Exchange Coupling of Grains

R. S. Iskhakov, S. V. Komogortsev, A. D. Balaev, and L. A. Chekanova

Kirensky Institute of Physics, Siberian Division, Russian Academy of Sciences, Krasnoyarsk, Russia e-mail: rauf@iph.krasn.ru

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Abstract—The values of saturation magnetization M_s , exchange coupling constant A, local magnetic anisotropy field H_a , random anisotropy correlation radius R_c , and coercive force H_c were independently measured for multilayer Co/Pd films with nanocrystalline and amorphous Co layers. It is shown that variation of the coercive force $H_c(t_{Co})$ as a function of the cobalt layer thickness t_{Co} is related to changes in characteristics of the magnetic microstructure. The main factor determining changes in the ferromagnetic correlation radius R_f and the average anisotropy $\langle K \rangle$ of a magnetic block in multilayer Co/Pd films is variation of exchange coupling constant $A(t_{Co})$. © 2002 MAIK "Nauka/Interperiodica".

The magnetic structure of nanocrystalline and amorphous ferromagnets is determined by their random magnetic anisotropy (RMA) and results from a competition of the ordering action of exchange interactions and the disordering effect of random local anisotropy K breaking the long-range ferromagnetic order [1]. In such materials, the ferromagnetic order is established at a characteristic length $R_{\rm f} = R_{\rm c} (H_{\rm ex}/D^{1/2}H_{\rm a})^2$, where $H_{\rm a} =$ $2K/M_s$ is the local magnetic anisotropy field, R_c is the random anisotropy correlation radius, H_{ex} is the exchange field, D is a numerical coefficient of the symmetry (equal to 1/15 in the case of a uniaxial anisotropy), and M_s is the saturation magnetization. The resulting magnetic structure can be described in terms of an ensemble of weakly coupled magnetic blocks [1, 2] with an average dimension of $2R_{\rm f}$, an average block anisotropy of $\langle K \rangle = K/N^{1/2} = K(R_c/R_f)^{3/2}$, and randomly oriented unit anisotropy vectors **n**.

To the present, researchers engaged in the magnetic properties of such materials believed that this type of magnetic structure is just what accounts for the observed values of characteristics of the magnetically soft amorphous and nanocrystalline alloys [2–4]. It should be noted that an analysis of the behavior of the coercive force H_c within the framework of the RMA model used in [2–4] is directed mostly to the study of H_c as a function of the nanograin size, $H_c(R_c)$, thereby assuming that the values of anisotropy $K(R_c)$, exchange coupling $A(R_c)$, and saturation magnetization $M_s(R_c)$ are constant (or their changes can be ignored). However, now there are prerequisites for undertaking complex measurements of all the parameters entering into theoretical expressions for H_c (R_c , H_a , A, and M_s) and

revealing the corresponding dependences. In particular, the RMA and magnetic block structure can be studied using small-angle neutron diffraction [5–7] and measurements of magnetization curves in the region of magnetizations close to saturation [8–10]. The exchange coupling constants are widely determined using methods based on the spin-wave resonance (SWR) [11] and Bloch's $T^{3/2}$ law [12].

The results of our preliminary investigation of the magnetization behavior M(H) in multilayer Co/Pd film structures [9] showed that these films are characterized by a two-dimensional inhomogeneity of the magnetic anisotropy. In particular, it was found that the experimental curve of M(H) measured in the range of field strengths from 5 to 25 kOe can be described by the expression

$$\frac{\Delta M}{M_{\rm s}} = \frac{D^{1/2} \langle H_{\rm a} \rangle}{H},\tag{1}$$

which corresponds to a ferromagnet with an anisotropy inhomogeneity dimension d = 2 and an experimental condition of $H < H_{ex}$. Applying expression (1) to the analysis of the experimental M(H) curve, we determined the values of $\langle H_a \rangle$ and $R_f = (A/D^{1/2}\langle K \rangle)^{1/2}$ (where $\langle K \rangle = M_s \langle H_a \rangle/2$ and the coefficient *D* was selected equal to 1/15 [8]). It was established that the value of H_c correlates to $\langle H_a \rangle$ (Fig. 1), which implies that $\langle H_a \rangle$ plays the role of effective anisotropy in the Co/Pd films studied. Therefore, an analysis of the effective anisotropy and the related coercive force in these systems can be performed using expressions obtained within the framework of the RMA model.



Fig. 1. Micromagnetic structure parameters (1) $\langle H_a \rangle$ and (3) R_f determined from the experimental M(H) curve approximated by Eq. (1) [9] (open symbols) and calculated by formulas (2) and (3) (small black symbols) and (2) coercive force H_c of multilayer Co/Pd films: (a) $[Co_{90}P_{10}(t_{C0})/Pd (14 \text{ Å})]_{20}$; (b) $[Co_{93}P_7(t_{C0})/Pd (14 \text{ Å})]_{20}$.

However, the aforementioned method of determining $\langle H_a \rangle$ and R_f cannot disclose the relationship between the changes of these parameters and variations of the main magnetic quantities (H_a, A, M_s) and the main parameter of the magnetic nanostructure (R_c) . Indeed, the theoretical expressions for $\langle H_a \rangle$ and R_f of an amorphous or nanocrystalline ferromagnet with a twodimensional inhomogeneity of the magnetic anisotropy are as follows [9]:

$$R_{\rm f} = \frac{2A}{\sqrt{D}H_{\rm a}M_{\rm s}R_{\rm c}},\tag{2}$$

$$\langle H_{\rm a} \rangle = \frac{\sqrt{D} H_{\rm a}^2 M_{\rm s} R_{\rm c}^2}{2A}.$$
 (3)

According to formula (3), in a material with equiaxial grains ($2R_c = t_{Co}$, where t_{Co} is the cobalt layer thickness) and constant values of H_a , A, and M_s , the values of $\langle H_a \rangle$

and H_c must increase in proportion to t_{Co}^2 . This is at variance with the experimental curves of $\langle H_a \rangle$ and H_c , which are described by decreasing functions of t_{Co} (Figs. 1a and 1b).

In this study, the saturation magnetization M_s , exchange coupling constant A, local magnetic anisotropy field H_a , random anisotropy correlation radius R_c , and coercive force H_c were independently measured for multilayer Co/Pd films with nanocrystalline and amorphous Co layers. The aim of these measurements was to determine the contribution of each of these parameters to the change of magnetic microstructure characteristics R_f and $\langle H_a \rangle$ and, hence, to the change of coercive force H_c .

The thermomagnetic curves and magnetization curves of these ferromagnetic films were measured in a wide range of temperatures (0-200 K) and magnetic fields (0-30 kOe) using an automated vibrating-sample magnetometer with a superconducting coil.

Multilayer Co/Pd films were prepared by sequential chemical deposition of components onto glass substrates. We studied two series of samples: (i) $[Co_{93}P_7(t_{Co})/Pd (14 \text{ Å})]_{20}$, where $t_{Co} = 6$, 30, 45, 55, or 80 Å and (ii) $[Co_{90}P_{10}(t_{Co})/Pd (14 \text{ Å})]_{20}$, where $t_{Co} = 20$, 45, 60, 65, or 115 Å. Small phosphorus additives in the cobalt layers of Co/Pd ensured the obtaining of ferromagnetic layers composed of metastable Co(P) solid solutions featuring various short-range order structures [13]. Previously [13], it was established that Co(P) layers containing 5–8 at.% P possess an fcc structure, while the layers containing above 9 at.% P occur in the amorphous state.

The experimental temperature dependence M(T) of multilayer Co/Pd films measured in the temperature range from 50 to 200 K can be described by the equation

$$M(T) = M_{s0}(1 - BT^{3/2}).$$
(4)

The values of magnetization M_{s0} and constant A, calculated from the Bloch constant B by the formula

$$A = \frac{k_{\rm B}}{8\pi} \left(\frac{g\mu_{\rm B}}{M_{\rm s0}} \right)^{1/3} \left(\frac{2.612}{B} \right)^{2/3},\tag{5}$$

are listed in the table.

In order to determine the parameters H_a and R_c characterizing the orientational inhomogeneity of the magnetic anisotropy, it is necessary to measure the magnetization curves both in the fields smaller than the exchange field H_{ex} and in the fields significantly exceeding H_{ex} , that is, to find two experimental asymptotes M(H). Indeed, in the fields $H > H_{ex}$, the magnetization approaches the saturation level according to the law

$$\frac{\Delta M}{M_{\rm s}} = \left(\frac{D^{1/2}H_{\rm a}}{H}\right)^2.$$
 (6)

Measuring this asymptotic region of the M(H) curve, we can determine the value of $D^{1/2}H_a$. The field strength at which the behavior according to Eq. (1)

changes to the behavior described by Eq. (6), $H_{\rm ex} = 2A/MR_{\rm c}^2$, is used to calculate the correlation radius $R_{\rm c}$.

As was noted above, the values of H_{ex} for the multilayer Co/Pd films studied varied within 20-25 kOe, which is very close to the maximum field strengths used in our experiments. Therefore, direct determination of the $H_{\rm a}$ and $R_{\rm c}$ values from the asymptotic regimes using Eqs. (1) and (6) was impossible. However, measurements in the fields of 25-30 kOe revealed certain deviations of the experimental M(H) curves from the power dependence $M(H) \sim H^{-1}$, which were related to the onset of the transition of the magnetic system to the regime described by expression (6). Based on these deviations, we determined the parameters H_a and R_c using approximation of the portions of M(H) curves in the fields from 5 to 30 kOe by a theoretical expression obtained previously [14] for a two-dimensional inhomogeneity of the magnetic anisotropy. This analytical expression (the law of magnetization variation close to saturation) for d = 2 is as follows:

$$\frac{\Delta M(H)}{M_{\rm s}} = \frac{DH_{\rm a}^2}{H\left(\frac{2A}{MR_{\rm s}^2} + H\right)}.$$
(7)

Figure 2 shows a typical experimental magnetization curve approximated by expression (7); the inset presents a plot of the square deviation s^2 of the experimental plot M(H) from the theoretical curve (7) in relative units versus the fitting parameter R_c . The data refer to the M(H) measurements for a $[Co_{90}P_{10}(t_{Co})/Pd$ $(14 \text{ Å})]_{20}$ sample with $t_{Co} = 115 \text{ Å}$. The minimum of s^2 (in absolute units) agrees well with the experimental mean-square error of M_s . The values of H_a and R_c corresponding to the s^2 minimum for the Co/Pd films studied are presented in the table.



Fig. 2. Magnetization curve of the $[Co_{90}P_{10}(t_{Co})/Pd (14 \text{ Å})]_{20}$ sample with $t_{Co} = 115$ Å measured in the region of approaching saturation (points) and approximation using expression (7) (solid curve). The inset shows a plot of the square deviation s^2 of the given experimental plot M(H) from the theoretical curve in relative units versus the fitting parameter R_c .

Once the numerical values of H_a , A, M_s , and R_c are determined, we can calculate $\langle H_a \rangle$ and R_f using formulas (2) and (3). The results of these calculations are presented by small black symbols in Fig. 1. As can be seen, the $\langle H_a \rangle$ and R_f vales determined directly via Eq. (1) and those calculated by formulas (2) and (3) show a good coincidence. An analysis of the dependences $H_a(t_{Co})$,

Type of samples	$t_{\rm Co}, {\rm \AA}$	A, 10^{-6} erg/cm^3	$M_{\rm s}, {\rm G}$	H _c , Oe	$R_{\rm c}$, Å	$D^{1/2}H_{\rm a}$, kOe
Nanocrystalline Co, $[Co_{93}P_7(t_{Co})/Pd (14 \text{ Å})]_{20}$	30	0.37	800	49	21	2
	45	0.63	870	34	26	1.4
	55	0.92	885	28	30	2.1
	80	0.93	914	11	26	1.5
Amorphous Co, [Co ₉₀ P ₁₀ (<i>t</i> _{Co})/Pd (14 Å)] ₂₀	20	0.17	483	300	18	3.3
	45	0.34	720	85	18	2.4
	60	0.51	916	76	24	2.2
	65	0.62	913	76	25	2.2
	115	0.63	888	33	26	1.6

The main magnetic constants and parameters of the random anisotropy, magnetic microstructure, and coercive force of Co/Pd films with amorphous and nanocrystalline Co layers

 $R_{\rm c}(t_{\rm Co})$, $A(t_{\rm Co})$, and $M_{\rm s}(t_{\rm Co})$ presented numerically in the table shows that a maximum gradient is characteristic of the exchange coupling constant *A*, which varies by a factor of almost 4. This is evidence that the main factor determining changes in the characteristics $\langle H_{\rm a} \rangle$ and $R_{\rm f}$ of the magnetic microstructure of multilayer Co/Pd films and, hence, of their coercive force $H_{\rm c}$, is variation of the exchange coupling constant *A* (one of the main magnetic parameters) related to a decrease in the thickness of Co layers, rather than a change in the anisotropy correlation radius $R_{\rm c}$ or the grain size. We believe that a similar situation takes place in nanocrystalline ferromagnetic alloys of the finemet type, extensively developed and studied in recent years.

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