Dimensionality of a System of Exchange-Coupled Grains and Magnetic Properties of Nanocrystalline and Amorphous Ferromagnets

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Characteristics of random magnetic anisotropy in ferromagnetic films of amorphous $Co_{90}P_{10}$ and nanocrystalline Ni₇₅C₂₅, Fe₈₀B₄C₁₆, and Co₈₀C₂₀ alloys and also in multilayer films $[Co_{93}P_7(x)/Pd(14 \text{ Å})]_{20}$ and $[Co_{90}P_{10}(x)/Pd(14 \text{ Å})]_{20}$ obtained by various technological procedures were studied experimentally. It was found that the spatial dimensionality (*d*) of the system of ferromagnetically coupled grains $(2R_c)$ in the materials under study determined the exponent in the power dependence of the approach of magnetization to saturation

in the region of fields $H < 2A/MR_c^2$. The dependence $\Delta M \sim H^{-1/2}$ was observed for nanocrystalline and amorphous films with a three-dimensional grain arrangement. The approach to saturation in multilayer films with a two-dimensional grain arrangement in an individual magnetic layer follows the law $\Delta M \sim H^{-1}$. The main micromagnetic characteristics of random anisotropy, such as the ferromagnetic correlation radius R_f and the average anisotropy $\langle K \rangle$ of a ferromagnetic domain with a size of $2R_f$, were determined for multilayer Co/Pd films. Correlation was found between the coercive field and these characteristics of random anisotropy. © 2000 MAIK "Nauka/Interperiodica".

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Considerable success in understanding the magnetic structure and soft magnetic properties of amorphous and nanocrystalline ferromagnets has recently been achieved based on the notions of random magnetic anisotropy (RMA). Thus, it was shown experimentally in [1] that soft magnetic properties of materials with a grain size $2R_c$ smaller than the exchange correlation length $\delta = (A/K)^{1/2}$ depend on R_c as follows: the coercive force $H_c \sim R_c^6$, and the initial permeability $\mu \sim R_c^{-6}$. These relationships for soft magnetic properties are conditioned by chaos in the direction of local magnetic anisotropy *K* and by the possibility of describing the magnetic structure of such materials by a set of weakly coupled magnetic domains with the size $2R_f$ and the average domain anisotropy

$$\langle K \rangle = \frac{K}{\sqrt{N}} = K \left(\frac{R_c}{R_f} \right)^{d/2}.$$
 (1)

Simple theoretical estimates [1–3] give the following equations for the average anisotropy of a magnetic domain and the ferromagnetic correlation radius:

$$\langle K \rangle \sim K(R_c/\delta)^{2d/(4-d)},$$
 (2)

$$R_f \sim \delta(\delta/R_c)^{d/(4-d)}$$
(2')

depending on the spatial dimensionality d of the arrangement of the grain system $2R_c$. For a three-

dimensional arrangement of exchange-coupled grains (d = 3), according to Eqs. (2) and (2'), we obtain $\langle K \rangle \sim R_c^6$, $R_f \sim R_c^{-3}$.

The main micromagnetic properties of amorphous and nanocrystalline ferromagnets $\langle K \rangle$ and R_f , as well as the local anisotropy characteristics *K* and R_c , can be directly determined from magnetization curves in the region of approach to saturation. The ideas of these measurements are as follows. The approach of magnetization to saturation in sufficiently high magnetic fields ($H > 2A/MR_c^2$) is described by the Akulov law for a polycrystal [4]

$$\frac{\Delta M}{M_s} = \left(\frac{2D^{1/2}K}{HM_s}\right)^2,\tag{3}$$

independent of the dimensionality *d* of the grain arrangement. Here, *D* is a numerical symmetry factor (see [5]), and the condition on the field *H* is determined by the inequality $R_c > R_H$, where the magnetic correlation radius $R_H = (2A/M_H)^{1/2}$. In fields below the socalled exchange field $H_{ex} = 2A/MR_c^2$, the inequality $R_H > R_c$ is fulfilled. Substituting $\langle K \rangle$ obtained according to Eq. (1) into the Akulov law, with regard to effective averaging over grains within the region $2R_H$, one can obtain the following dependence of magnetic moment scattering on the field:

$$\frac{\Delta M}{M_S} = \left(\frac{2D^{1/2}\langle K\rangle}{HM_S}\right)^{(4-d)/2} \equiv \left(\frac{R_H}{R_f}\right)^{4-d}.$$
 (4)

Thus, an analysis of the magnetization curve M(H) throughout the entire range of magnetic fields allows the values of K, R_c , R_f , and $\langle K \rangle$ to be measured and the dimensionality of the system d to be determined.

The law of approach to saturation for isotropic (three-dimensional) inhomogeneities was theoretically derived in [5] based on the canonical expansion of random functions. The approach to saturation in ferromagnets with two-dimensional anisotropy inhomogeneities was theoretically considered in [6, 7], and Eq. (4) for inhomogeneities with an arbitrary dimensionality was actually obtained in [8].

Equations (1), (2), and (4) indicate that the dimensionality of a system of exchange-coupled grains determines the character of the approach of magnetization to saturation in the region $R_H > R_c$. Hence, it also determines the level of soft magnetic properties of amorphous and nanocrystalline ferromagnets through $\langle K \rangle$. This work is devoted to an experimental study of the effects considered above.

Experiment. This work reports the results of studying magnetization curves for films of amorphous $Co_{90}P_{10}$ (thickness t = 2000 Å) and nanocrystalline $Co_{80}C_{20}$ (t = 1200 Å), $Fe_{80}B_4C_{16}$ (t = 500 Å), and $Ni_{75}C_{25}$ (t = 600 Å) alloys and also for multilayer films $[Co_{93}P_7(x)/Pd(14 Å)]_{20}$ (where x = 30 Å, 45 Å, 55 Å, and 80 Å) and $[Co_{90}P_{10}(x)/Pd(14 Å)]_{20}$ (where x = 20 Å, 45 Å, 60 Å, 65 Å, and 115 Å) obtained by various technological procedures [5, 9, 10]. Small additions of phosphorus in the cobalt layers of Co/Pd multilayer films were used for obtaining ferromagnetic layers differing in the short-range structure. In Co(P) layers prepared by the chemical deposition technique, an fcc structure is obtained at concentrations of 5–8 at. % P and an amorphous state of Co layers is obtained above 9 at. % P [5].

Magnetic measurements were carried out using a vibrating-coil magnetometer with a superconducting solenoid in fields up to 30 kOe and temperatures from 4.2 to 200 K. The film substrate was measured separately, and its contribution to the total signal (~1%) was subtracted.

Results and discussion. Figure 1 shows high-field regions of magnetization curves M(H) for films of nanocrystalline and amorphous alloys. The thicknesses of the films for which these curves were measured considerably exceeded the sizes of the constituent grains or clusters ($2R_c \sim 100$ Å). Thus, magnetic domains with a size of $2R_f$ were arranged in these films as grains with a size of $2R_c$ in a three-dimensional way. In this case, according to Eq. (4), the approach of magnetization to saturation in these materials must follow the power

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Fig. 1. High-field regions of magnetization curves M(H) for amorphous and nanocrystalline films with d = 3: (1) $\text{Co}_{80}\text{C}_{20}$ (t = 1200 Å), (2) $\text{Co}_{90}\text{P}_{10}$ (t = 2000 Å), (3) $\text{Fe}_{80}\text{B}_4\text{C}_{16}$ (t = 500 Å), and (4) $\text{Ni}_{75}\text{C}_{25}$ (t = 600 Å).

dependence $\Delta M/M_s \sim H^{-1/2}$. The rectilinear regions on the coordinates ($\Delta M/M_s$, $H^{-1/2}$) in Fig. 1 actually indicate that this power dependence of the approach of magnetization to saturation is actually fulfilled in these samples within the range of fields of 1–10 kOe. According to Eq. (4), this fact points to a three-dimensional arrangement of exchange-coupled grains.

Figure 2 displays magnetization curves for multilayer Co(P)/Pd films. The sizes of the grains or clusters that compose individual magnetic layers of these films are of the order of the thickness of such layers. The filling of an individual layer along the height of one grain leads to the formation of a two-dimensional system of ferromagnetically coupled grains in these films. An essential point here is the fact that the exchange coupling between grains within an individual layer considerably exceeds the coupling between neighboring individual layers (by at least an order of magnitude) studied in [10]. For the tasks of this work, the multilayer arrangement was only a way of accumulating the experimental signal for studying the small effect of magnetic moment scattering in the vicinity of saturation in ultrathin layers by the induction method. According to Eq. (4), the approach of magnetization to saturation in a two-dimensional arrangement of grains corresponds to the law $\Delta M/M_s \sim H^{-1}$. The rectilinear regions in the high-field magnetization curves of multilayer Co(P)/Pd films with both amorphous and nanocrystalline Co layers on the coordinates $(\Delta M/M_s H^{-2}, H)$ point to the fulfillment of the dependence $\Delta M/M_s \sim H^{-1}$ in the range of fields up to 20-25 kOe.

As was indicated above, the region of fields used for determining the system dimensionality d from the



Fig. 2. High-field regions of magnetization curves M(H) for multilayer Co/Pd films: (a) $[Co_{93}P_7(x)/Pd(14 \text{ Å})]_{20}$, x = (I) 30, (2) 45, (3) 55, and (4) 80 Å; (b) $[Co_{90}P_{10}(x)/Pd(14 \text{ Å})]_{20}$, x = (I) 20, (2) 45, (3) 60, (4) 65, and (5) 115 Å.



Fig. 3. Micromagnetic parameters $\langle H_a \rangle$ and R_f and coercive force H_c for multilayer Co/Pd films: (a) $[Co_{93}P_7(x)/Pd(14 \text{ Å})]_{20}$ and (b) $[Co_{90}P_{10}(x)/Pd(14 \text{ Å})]_{20}$.

observed dependence of the moment on the external field has an upper limit equal to the exchange field $H_{ex} = 2A/MR_c^2$. In order to estimate the value of H_{ex} , it is necessary to find the exchange interaction constant A. This constant, for the films studied in this work, was calculated from the measured low-temperature thermomagnetic curves (the Bloch law $T^{3/2}$ [9]). For the amorphous and nanocrystalline films studied in this work, the values of H_{ex} were found to be ~10–20 kOe. Therefore, the dependence of ΔM on H for these films also exhibited a crossover (transition from $\Delta M \sim H^{-1/2}$ to $\Delta M \sim H^{-2}$). This allowed us to measure H_{ex} directly, to calculate R_c , and to measure the value of K. For multilayer films, we estimated $H_{ex} \sim 20-30$ kOe, which is close to the value of the maximal fields used in our experiment.

Note that the law of approach to saturation $\Delta M/M_s \sim H^{-1/2}$ predicted in [5] is well known to magnetologists and is widely used for interpreting data on the approach of magnetization to saturation in amorphous and nanocrystalline alloys [11–15]. Here, a change of the exponent in the power dependence of the approach to saturation in nanocrystalline and amorphous materials with the dimensionality d = 2 was found experimentally. The results presented in Figs. 1 and 2 are described by Eq. (4). They demonstrate that the exponent in the power dependence of curves describing the approach of magnetization to saturation is not associated with the short-range order in the ferromagnetic material. It is determined only by such a substructure parameter as the dimensionality d of the arrangement of exchange-coupled grains. The fundamental possibility of experimentally determining the spatial dimensionality of the arrangement of a system of ferromagnetically coupled grains (in the general case, this dimensionality can be a noninteger number) is of great importance for the physics and technology of magnetic materials.

The slopes of the rectilinear regions in Figs. 1 and 2 characterize the mean-square fluctuations of the field of anisotropy $\langle K \rangle$ averaged over the region within which the magnetic moments of the grains are exchange-correlated: $D^{1/2}\langle H_a \rangle = 2D^{1/2}\langle K \rangle/M = D^{1/2}H_a/N^{1/2}$, where N is the number of structural units $2R_c$ incorporated into a magnetic domain. The magnetic correlation radius R_f can be estimated from Eq. (2) or from the following equation: $R_f = (AD^{1/2}\langle K \rangle)^{1/2}$. The values of $\langle H_a \rangle$ and R_f calculated in such a way for multilayer Co/Pd films differing in thickness of the ferromagnetic layer are given in Fig. 3 (the coefficient D was set equal to 1/15 [5]). The coercive forces H_c of these films are also given

here. A detailed analysis of the dependence of the micromagnetic parameters $\langle H_a \rangle$ and R_f in these films on the magnetic layer thickness is a subject of a separate comprehensive investigation [the observed variations of these quantities within a given thickness range are due to variations of both the magnetic constants (A, K)and structural parameters (R_c, d) , which nonlinearly enter into Eqs. (1) and (2)]. It is important to emphasize here the correlation of the coercive force H_c and the average anisotropy of the magnetic domain $\langle H_a \rangle$. Because there is no strong interaction between magnetic domains, the latter value represents the effective anisotropy of these materials. The correlation observed between the effective anisotropy and the coercivity of nanocrystalline and amorphous ferromagnets with a two-dimensional arrangement of magnetically coupled grains indicates that magnetic anisotropy fluctuations play the main role in the formation of soft magnetic properties in such systems.

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