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Study of Magnetic Correlations in Nanostructured Ferromagnets by Correlation Magnetometry

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We propose a theoretically justified experimental magnetometric technique for determining the size of stochastic domains spontaneously formed in the spin system of nanostructured ferromagnets and for evaluating the effective anisotropy in these magnetically correlated regions. The method is based on monitoring the $\Delta M \sim H^{-2}$ relationship in the low-field part of the integral magnetization curve. © *2003 MAIK "Nauka/Interperiodica".*

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1. INTRODUCTION

Nanocrystalline and amorphous ferromagnets belong to the class of nanostructured materials extensively studied both in basic aspects and in view of possible applications. A currently important problem is the establishing of relationships between macroscopic and microscopic parameters of these materials. For nanostructured ferromagnets, such a relationship has been described within the framework of the so-called random anisotropy model [1–4]. According to this approach, the spin system of a ferromagnet is described in terms of the following microscopic parameters: exchange interaction A, magnetization M_s, local anisotropy K, and the length $2R_c$ of its homogeneous orientation. It is assumed that the easy axes of local anisotropy in the grains (clusters) of a ferromagnet are randomly oriented. Some cases of spatially inhomogeneous anisotropy, exchange, and magnetization have been also considered (see, e.g., [2]).

A characteristic feature of the random anisotropy model in systems of dimension $d < 4$ is instability of the ferromagnetic state with respect to the appearance of an arbitrarily small random anisotropy even at a zero temperature, which is completely analogous to breakage of the ferromagnetic order in the random field model [5]. In both cases, the ferromagnetic order is established over a characteristic distance—the magnetic orientation coherence length $2R_L$ ($R_L \gg R_c$). Both models predict an increase in the length of magnetic correlations with decreasing correlation length of the random perturbations.

Thus, a magnetic structure of nanostructured ferromagnets can be represented by an ensemble of stochastic domains (with a size of $2R_L$) obeying an approximation analogous to the model of exchange-independent grains in polycrystalline solids. This well-known approximation has been widely used for calculating macroscopic parameters such as coercive force, susceptibility, and residual magnetization in the region of irreversibility of the magnetization curve (beginning with the original paper of Stoner and Wohlfarth [6]). The same approximation was used to describe the law of the magnetization approach to saturation employed (beginning with the works of Akulov [7, 8]) for determining the microscopic anisotropy K from the reversible magnetization curve. Herzer [9] showed that this method provides an adequate description of nanostructured ferromagnets: determination of the coercive force as a function of the grain size, $H_c \sim R_c^6$, was equivalent to estimating the macroscopic anisotropy in a stochastic domain as $K \sim (R_c/R_L)^{3/2}$ and the domain size as $R_L \sim A^2/K^2 R_c^3$.

It should be emphasized that R_L is a very important parameter. Unfortunately, the possibilities of experimental determination of the magnetic correlation length are rather restricted. It is commonly accepted that the main experimental method is offered by small-angle neutron scattering (SANS). Using this method, Löffler *et al.* [10] recently measured the value of $L_m = 2R_L$ and studied the dependence of $L_m$on the grain size $D = 2R_c$ for nanostructured Fe. Ryne [11] used SANS to measure L_m and studied the dependence of L_m on the applied magnetic field H in a $TbFe_2$ amorphous alloy.

This paper demonstrates the possibility to measure the magnetic correlation length R_L in nanostructured ferromagnets and to evaluate the macroscopic anisotropy in these magnetically correlated regions using a magnetometer—a commonly available instrument.

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2. THEORETICAL JUSTIFICATION OF THE PROPOSED METHOD

For an inhomogeneous ferromagnet characterized by a local magnetic anisotropy of arbitrary origin and symmetry, the law of the magnetization approach to saturation can be written as

$$\langle M_z\rangle/M_s \approx 1 - d_m(H), \tag{1}$$

where d_m is the normalized mathematical dispersion (variance) of the transverse magnetization components [2]:

$$d_m = \frac{\langle M_\perp^2\rangle}{M_s^2} = (aH_a)^2\int\frac{S(k)d^3k}{((2A/M_s)k^2 + H)^2}. \tag{2}$$

Here, $H_a = 2K/M_s$ is the local anisotropy field, a is the symmetry factor (for uniaxial anisotropy, $a = 1/15^{1/2}$) and $S(k)$ is the normalized spectral density of the correlation function $K(r)$ of orientation of the local anisotropy axes.

For an exponential model correlation function of the type e^{-r/R_c}, we obtain

$$d_m = \frac{(aH_a)^2}{H^{1/2}(H_R^{1/2} + H^{1/2})^3}, \tag{3}$$

where $H_R = 2A/M_sR_c^2$ is a correlation field (for the reasons presented below, this parameter will be referred to as the "upper" correlation field) and R_c is the correlation radius of the anisotropy inhomogeneities. As can be seen, the character of the behavior of magnetization as a function of the field changes in the vicinity of H_R, so that

$$d_m = (aH_a)^2\begin{cases}H^{-2}, & H \gg H_R\\ H^{-1/2}H_R^{-3/2}, & H \ll H_R.\end{cases} \tag{4}$$

In the coordinates of $\log d_m$ versus $\log H$, this change is manifested by a characteristic bending of the experimental $M(H)$ curve in the region of $H \sim H_R$. Note that, if any other monotonically decreasing function (rather than exponent) is selected as the model correlation function, the analytical expression for $d_m(H)$ will change, but the asymptotic behavior (4) and the correlation field H_R remain the same.

Let us consider the physical reasons for the appearance of a characteristic point H_R in the $d_m(H)$ function. Deviations of the magnetization vector $\mathbf{M}(\mathbf{x})$ from the magnetic field direction in nanostructured ferromagnets are correlated within a region of size $R_H = (2A/M_sH)^{1/2}$, this value depends on the magnetic field and exchange. For $R_H \ll R_c$ (i.e., for $H \gg H_R$), the fluctuations of magnetization orientation are uncorrelated. On the contrary, for $R_H \gg R_c$, the magnetization vector orientation is strongly correlated. Therefore, $H \sim H_R$ corresponds to a change in the character of the magnetization curve. For this reason, the value of H_R and, hence R_c, can be experimentally measured. A method for processing the magnetization curve so as to extract data on the correlation radius and the effective magnetic anisotropy is called the correlation magnetometry [12].

The low-field effects related to the formation of stochastic domains are not manifested in formulas (3) and (4). This is explained by the fact that these expressions were obtained in the linear approximation of the perturbation theory that fails to be valid in the range of small fields. However, the correlation properties of the inhomogeneous orientation of $\mathbf{M}(\mathbf{x})$ in this range can be determined by numerical methods. In particular, the correlation function $K_m(r)$ and the corresponding correlation radius $R_m(H)$ were calculated [13] for a chain of exchange-coupled grains with random anisotropy. The main result of such numerical calculations consists in the appearance of another characteristic field $H_L = 2A/M_sR_L^2$, called the "lower" correlation field, where $2R_L$ is the size of a stochastic domain. In the range of $H > H_L$, the behavior of $R_m(H)$ is described by the linear theory, while for $H < H_L$, the $R_m(H)$ function tends to the constant value R_L (instead of infinitely increasing as predicted by the linear theory).

In order to take into account the formation of stochastic domains, we suggest modifying expression (3) as follows:

$$d_m = \left(\frac{aH_a}{H}\right)^2\left(\frac{H_L^{1/2} + H^{1/2}}{H_R^{1/2} + H^{1/2}}\right)^d, \tag{5}$$

where d is the dimension of the system of exchange-coupled ferromagnetic nanoparticles. This expression, in contrast to the exact formula (3), represents an interpolation and has to be verified in experiment. Indeed, in the range of $H > H_L$, expression (5) describes the experimental $M(H)$ curves for $d = 3$ [14], $d = 2$ [15], and $d = 1$ [16]. Below we will demonstrate that this expression also adequately describes the experimental data for $H < H_L$. To this end, we will need expressions for the asymptotic behavior of $d_m(H)$:

$$d_m = \begin{cases}\dfrac{(aH_a)^2}{H^2}\dfrac{H_L^{d/2}}{H_R^{d/2}} = \dfrac{(a\langle H_a\rangle_L)^2}{H^2},\\ a\langle H_a\rangle_L < H < H_L\\ \dfrac{(aH_a)^2}{H_R^{d/2}}\dfrac{1}{H^{(4-d)/2}} = \dfrac{(a\langle H_a\rangle_L)^2}{H_L^{d/2}}\dfrac{1}{H^{(4-d)/2}},\\ H_L < H < H_R\\ \dfrac{(aH_a)^2}{H^2}, \quad H_R < H\end{cases} \tag{6}$$

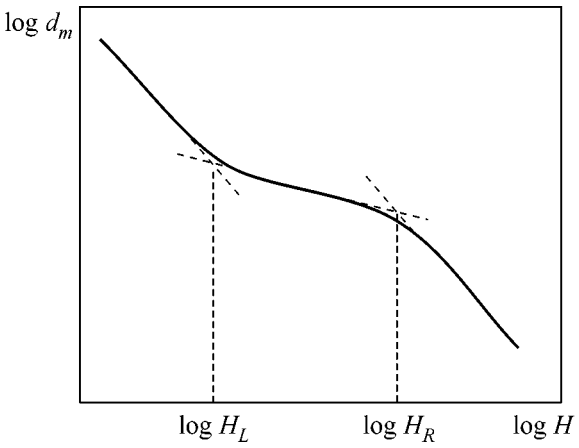

Fig. 1. A schematic curve of the magnetization dispersion $d_m(H)$ of a nanostructured ferromagnet.

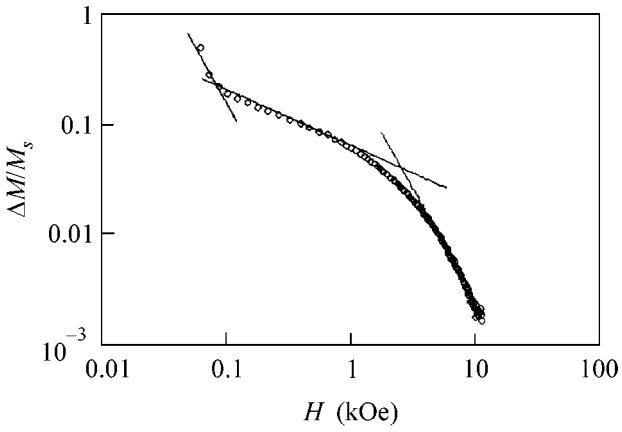

Fig. 2. A plot of the magnetization dispersion $\Delta M/M_s$ versus magnetic field strength for a 2000-Å-thick $Co_{90}P_{10}$ amorphous alloy film at $T = 4$ K ($H_L = 80$ Oe, $H_R = 3$ kOe).

Here, the macroscopic anisotropy field in a stochastic domain is given by the formula

$$\langle H_a \rangle_L = H_a(H_L/H_R)^{d/4} = H_a(R_c/R_L)^{d/2} = H_a/\sqrt{N}, \quad (7)$$

where N is the number of nanoparticles in this domain. Figure 1 schematically presents the function (5) and shows the asymptotes (6) on the double logarithmic scale. As can be seen, the experimental task reduces to monitoring the low-field crossover (bending) in d_m, the presence of which proves the formation of stochastic domains in the system.

3. RESULTS AND DISCUSSION

Figure 2 presents a double logarithmic plot of a reversible part of the magnetization curve $M(H)$ for a 2000-Å-thick $Co_{90}P_{10}$ amorphous alloy film. The films of amorphous and nanocrystalline alloys were obtained by chemical deposition onto cover glasses. The magnetization curves were obtained (with the substrate background subtraction) using a vibrating-sample magnetometer with a superconducting coil operating in a range of fields up to 30 kOe.

As can be seen from Fig. 2, the experimental field dependence $M(H)$ is well described by expressions (5) and (6) with $d = 3$. As the field H decreases, the magnetization sequentially obeys the high-field Akulov law ($\Delta M \sim H^{-2}$), the cooperative dependence ($\Delta M \sim H^{-1/2}$), and the low-field dependence ($\Delta M \sim H^{-2}$). The theoretical asymtotes described by expressions (6) are represented by straight lines. The points of intersection of these lines determine the characteristic fields $H_L \approx 80$ Oe and $H_R \approx 3$ kOe corresponding to the low- and high-field crossovers (bending points) in the magnetization curve $M(H)$. Upon substituting the values of exchange A and magnetization M_s into the expressions for H_L and H_R, we obtain the values of $R_L \approx 500$ Å and $R_c \approx 80$ Å. Using the portions described by the dependence $\Delta M \sim H^{-2}$, we determine the macroscopic and microscopic anisotropy fields: $\langle H_a \rangle_L \approx 150$ Oe and $H_a \approx 2$ kOe. Substitution of the measured and calculated values into expression (7) proves the validity of this relation.

Figure 3 shows a reversible part of the magnetization curve for a 100-Å-thick $Co_{93}P_7$ nanocrystalline alloy film. This curve reveals the low-field dependence $\Delta M \sim H^{-2}$, followed by a clearly identified dependence of the type $\Delta M \sim H^{-3/4}$ (for H from 0.2 to 2 kOe), a statistically reliable dependence of the type $\Delta M \sim H^{-1}$ (for H from 2 to 6 kOe), and a noisy signal in the fields above 10 kOe (reflecting the fact that the dispersion of magnetization becomes comparable with the accuracy of magnetization measurements).

The grain size in the $Co_{93}P_7$ nanocrystalline alloy is on the order of the film thickness. Therefore, this film features a two-dimensional system of ferromagnetically coupled grains ($d = 2$). According to expressions (5) and (6), this system has to exhibit a low-field crossover in $M(H)$ (reflecting the transition from $\Delta M \sim H^{-2}$ to $\Delta M \sim H^{-1}$). However, our experimental curve initially exhibits an intermediate transition from $\Delta M \sim H^{-2}$ to $\Delta M \sim H^{-3/4}$ and only then changes to $\Delta M \sim H^{-1}$. This behavior is not an artifact. Indeed, the rms deviation $d_m^{1/2}$ of magnetization has to be determined from the condition of minimum for the total energy including a magnetic dipole interaction (which is significant in the case of thin films). The most exhaustive theoretical analysis of this situation has been performed in [17–19]. In our notations, the final expression for d_m is as follows [19]:

$$d_m = \frac{1}{2}\frac{(aH_a)^2}{M_s^{1/2}H_R^{3/4}}\frac{1}{H^{3/4}} \equiv \frac{1}{2}\frac{(a\langle H_a \rangle_L)^2}{M_s^{1/2}H_L H_R^{-1/4}}\frac{1}{H^{3/4}}. \quad (8)$$

To our great surprise, an analysis of the available literature showed that the region of $\Delta M \sim H^{-3/4}$ in the

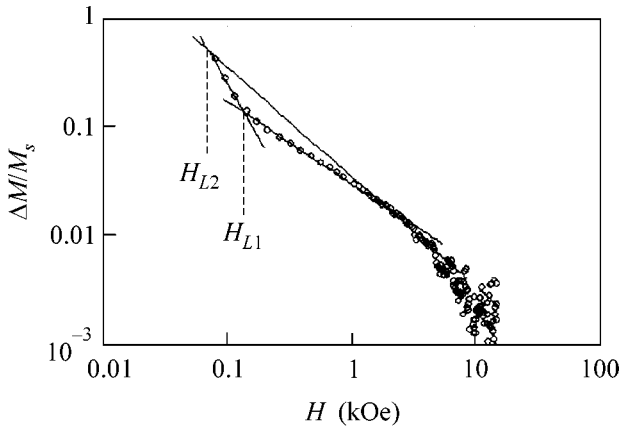

Fig. 3. A plot of the magnetization dispersion $\Delta M/M_s$ versus magnetic field strength for a 100-Å-thick $Co_{93}P_7$ film (T = 90 K).

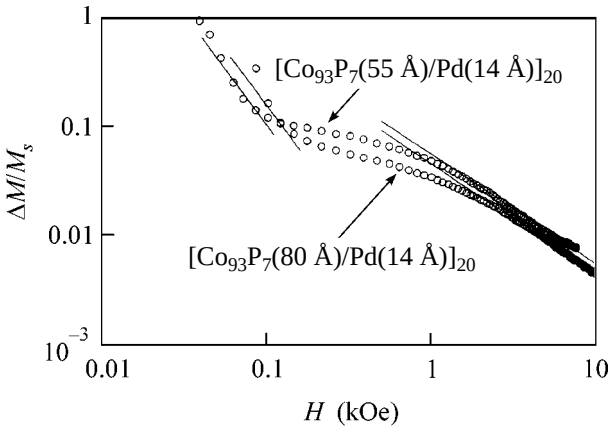

Fig. 4. A plot of the magnetization dispersion $\Delta M/M_s$ versus magnetic field strength for two multilayer $[Co_{93}P_7(x)/Pd(14\ Å)]_{20}$ structures with individual nanocrystalline Co layer thicknesses x = 80 and 55 Å (T = 4 K).

experimental curve of Fig. 3 is the first observation of the well-known theoretical dependence (8). The intersections of asymptotes $H^{-3/4}$ and H^{-1} with the line H^{-2} give two values of the characteristic field: $H_{L1} \approx 140$ Oe and $H_{L2} \approx 70$ Oe. This is indirect evidence of a modification of the shape of a stochastic domain in the thin film under consideration related to the magnetostatic effects, whereby a disk with a circular base existing at $H < H_{L2}$ transforms into a disk with an ellipsoidal base in higher fields.

Figure 4 presents the reversible parts of the magnetization curves $M(H)$ for two multilayer $[Co_{93}P_7(x)/Pd(14\ Å)]_{20}$ structures with individual nanocrystalline Co layer thicknesses x = 80 and 55 Å. Note that the total ferromagnetic layer thickness was 1600 or 1100 Å, respectively; hence, the useful signal magnitude was more than ten times greater as compared to that from a single ultrathin film. In the range of magnetic fields from 1–1.5 to 20 kOe, these samples obeyed the law $\Delta M \sim H^{-1}$ that confirmed the two-dimensional character of the system of ferromagnetically coupled grains. In the region of low fields, the magnetization follows the dependence $\Delta M \sim H^{-2}$, but the portion of the $M(H)$ curve in greater fields cannot be described by a power function. In our opinion, this is explained by more complicated magnetostatic phenomena (related to the magnetic dipole interactions between the magnetizations of individual ferromagnetic layers) as compared to those in a thin single-layer film.

Assuming that the characteristic fields H_L correspond to the H values at which the experimental data deviate from the low-field dependence $\Delta M \sim H^{-2}$, we conclude that a decrease in the ferromagnetic layer thickness (and, hence, in the R_c value) leads to a decrease in H_L (and to a corresponding increase in R_L), in agreement with the main stipulations of models [1–5]. Another physically reasonable observation is a decrease in the macroscopic anisotropy field $\langle H_a \rangle_L$ with increasing size of stochastic domains. Therefore, stochastic domains are also formed in individual ferromagnetic layers of multilayer structures, but the shape of these domains (like that in an ultrathin nanocrystalline ferromagnetic film) is significantly modified by magnetostatic interactions.

Thus, the proposed interpolation formula (5) and asymptotic expressions (6) qualitatively explain the effects observed on the magnetization curves measured in the entire range of magnetic fields. In the high-field range, $H \gg H_L$, formula (5) coincides with the exact theoretical expression (3), and in the low-field region, this formula describes the behavior of $M(H)$ related to the formation of stochastic domains in nanostructured ferromagnets. This approach allows effective quantitative characteristics (H_L, $\langle H_a \rangle_L$, R_L) to be introduced for description of the low-field magnetic correlations.

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