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Characteristics of the Magnetic Microstructure of Amorphous and Nanocrystalline Ferromagnets with a Random Anisotropy: Theoretical Estimates and Experiment

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An experimental determination (both direct and indirect) of the characteristics of the magnetic microstructure, namely, the ferromagnetic correlation radius R_f and the rms fluctuation of the mean anisotropy $D^{1/2}\langle H_a\rangle$, is performed for amorphous and nanocrystalline ferromagnets with a random anisotropy characterized by the quantities R_c and $D^{1/2}H_a$, respectively. The magnetization curves of amorphous and nanocrystalline ferromagnets are found to exhibit a dependence on H that is caused by the alignment of the magnetizations of individual magnetic blocks with the field. © *2000 MAIK "Nauka/Interperiodica".*

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1. Amorphous and nanocrystalline ferromagnetic alloys can be represented as an ensemble of clusters or grains of size $2R_c$, which are bound together by the exchange interaction and have randomly oriented easy axes. In the approximation of a continuous medium, such a system is described by the internal energy density

$$U = \frac{1}{2}\alpha(\nabla\mathbf{M})^2 - \frac{1}{2}\beta(\mathbf{Ml})^2 - \mathbf{HM}, \qquad (1)$$

where the magnetization M is characterized by a constant magnitude M_s, the exchange parameter $\alpha = 2A/M_s^2$ is determined by the exchange interaction constant A, the parameter $\beta = H_a/M_s = 2K/M_s^2$ is determined by the local anisotropy constant K, $\mathbf{l}$ is the unit vector of the easy axis of this anisotropy, and H is the external magnetic field.

It is known that, in a ferromagnet, an orientational irregularity of the magnetic anisotropy of any origin (crystallographic, elastic, or other) gives rise to the formation of an inhomogeneous state of the magnetic moment $\mathbf{M}(\mathbf{x})$ [1–10]. This state is called a stochastic magnetic structure (SMS). The parameters of the SMS are determined by the relations between three characteristic fields: the external field H, the exchange field $H_{ex} = 2A/M_sR_c^2$, and the rms fluctuation of the local anisotropy field $D^{1/2}H_a$, where D is the symmetry factor equal to 1/15 for a uniaxial anisotropy [4]. In the case of large grains satisfying the inequality

$$R_c > D^{-1/4}(A/K)^{1/2}, \quad H_{ex} \le D^{1/2}H_a, \qquad (2)$$

the correlation properties of the inhomogeneous state of the orientation of $\mathbf{M}$ always coincide with the correlation properties of the local anisotropy fluctuations. In this case, the approximation of crystallites without exchange interaction between them is valid. Beginning from the publications [11, 12], this approximation was used for calculating the law of magnetization approach to saturation:

$$\Delta M/M_s = (\sqrt{D}H_a/H)^2, \quad H > D^{1/2}H_a. \qquad (3)$$

For amorphous and nanocrystalline ferromagnets, the other inequality is valid:

$$R_c < D^{-1/4}(A/K)^{1/2}, \quad H_{ex} > D^{1/2}H_a, \qquad (4)$$

If this inequality is satisfied, the correlation properties of the SMS fundamentally differ (in the fields $H < H_{ex}$) from those of the local anisotropy: the deviations of the magnetization $\mathbf{M}(\mathbf{x})$ from the direction of the external field are correlated in space and form a static wave with the characteristic wavelength $R_H = (2A/MH)^{1/2}$. The changes occurring in the correlation properties of the SMS in the vicinity of the field H_{ex} lead to a change in

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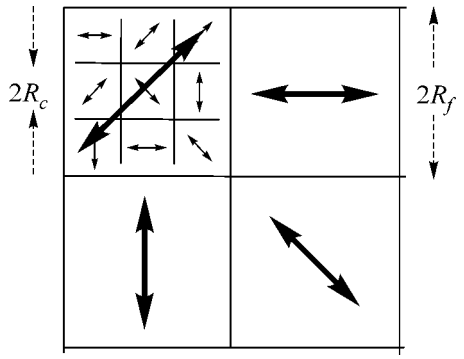

Fig. 1. Schematic representation of a ferromagnet with a random anisotropy. The small arrows indicate random orientation of the local magnetic anisotropy **l**(**x**), and the large arrows show random orientation of the mean anisotropy of a magnetic block **n**(**x**).

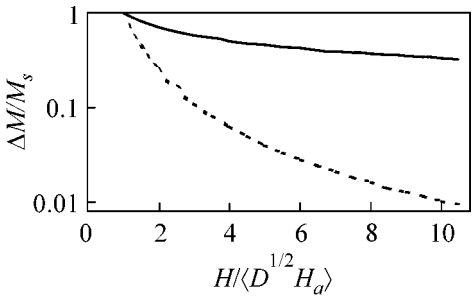

Fig. 2. Theoretical dependences of the variance of magnetic moment on the external magnetic field in the $D^{1/2}\langle H_a\rangle$ units: the solid curve corresponds to Eq. (11) and the dashed curve to Eq. (14).

the law of magnetization approach to saturation. For three-dimensional and isotropic inhomogeneities of anisotropy, the following relationship was obtained [4, 7–9]:

$$\begin{aligned}\Delta M/M_s &= (D^{1/2}H_a/H_{ex})^2(H_{ex}/H)^{1/2}\\ &= (D^{1/2}H_a/H_{ex})^2(R_H/R_c),\\ H &< H_{ex} \text{ or } R_H > R_c.\end{aligned} \tag{5}$$

For anisotropic and low-dimensional inhomogeneities, the dependence on H may be different [6, 7, 13–16].

As the magnetic field further decreases ($H \ll H_{ex}$), the situation changes. In low magnetic fields, the magnetic system of amorphous and nanocrystalline ferromagnets exhibits the well-known Imry–Ma effect [17]. This effect consists in the instability of the ferromagnetic state with respect to the randomly oriented local magnetic anisotropy. In this case, the ferromagnetic order is characterized by the correlation length $R_f = R_c(H_{ex}/D^{1/2}H_a)^2$ [5, 10], so that the magnetic structure of such a material can be described by an ensemble of weakly coupled magnetic blocks (Fig. 1). The block size is $2R_f$, the mean anisotropy in the block is $\langle K\rangle = K/N^{1/2} = K(R_c/R_f)^{3/2}$, and the unit vector **n** of this anisotropy is randomly oriented. In the approximation of a continuous medium, such a system can be described by the internal energy density represented in the form

$$\mathbf{U} = -\frac{1}{2}\beta^e(\mathbf{Mn}) - \mathbf{HM}, \tag{1'}$$

where the parameter $\beta^e = \langle H_a\rangle/M = 2\langle K\rangle/M^2$ is determined by the constant $\langle K\rangle$ characterizing the mean anisotropy in the magnetic block. In zero field, the magnetization of a magnetic block is oriented along the unit vector **n**. Therefore, in this case, the correlation properties of the irregular orientation of **M**(**x**) completely reproduce the correlation properties of the fluctuations of mean anisotropy $\langle K\rangle$. This means that the magnetization curve in low magnetic fields should be described by dependence (3) modified as follows:

$$\begin{aligned}\Delta M/M_s &= (D^{1/2}\langle H_a\rangle/H)^2 = (R_H/R_f)^4,\\ H &> D^{1/2}\langle H_a\rangle \text{ or } R_H < R_f.\end{aligned} \tag{3'}$$

Simultaneously, using the definition of the characteristics of the system of magnetic blocks (R_f and $D^{1/2}\langle H_a\rangle$) in terms of the characteristics of the grain system (R_c and $D^{1/2}H_a$), Eq. (5) can be represented in the form

$$\begin{aligned}\Delta M/M_s &= (D^{1/2}\langle H_a\rangle/H)^{1/2} = R_H/R_f,\\ R_c &< R_H \ll R_f.\end{aligned} \tag{5'}$$

The aim of our work is the experimental study of the aforementioned effects. Its significance is determined by the fact that the experimental measurement of dependence (3') is a direct, rather than indirect [see Eq. (5')], proof of the existence of magnetic blocks and allows one to directly measure the characteristics of the magnetic microstructure (the quantities $\langle H_a\rangle$, $\langle K\rangle$, and R_f) of amorphous and nanocrystalline ferromagnets.

2. Let us theoretically estimate the correlation properties of the irregular orientation of **M**(**x**). The main characteristic of these properties is the correlation function $K_m(\mathbf{r})$ or the spectral density $S_m(\mathbf{k})$ related to the correlation function through the Fourier transform:

$$\begin{aligned}\langle \mathbf{m}_\perp(\mathbf{x})\mathbf{m}_\perp(\mathbf{x}+\mathbf{r})\rangle &= K_m(\mathbf{r});\\ \langle \mathbf{m}_\perp(\mathbf{x})\mathbf{m}^*_\perp(\mathbf{x}+\mathbf{r})\rangle &= S_m(\mathbf{k})\delta(\mathbf{k}-\mathbf{k}');\\ K_m(\mathbf{r}) &= \int S_m(\mathbf{k})e^{i\mathbf{kr}}d\mathbf{k},\end{aligned} \tag{6}$$

where $\mathbf{m}_\perp(\mathbf{x})$ are the transverse components of the unit vector of magnetization $\mathbf{m}(\mathbf{x}) = \mathbf{M}(\mathbf{x})/\mathbf{M}$. The magneti-

zation curve is related to $K_m(\mathbf{r})$ and $S_m(\mathbf{k})$ by the standard relationships

$$\Delta M/M_s \equiv d_m(H) = K_m(\mathbf{r})|_{\mathbf{r}=0} = \int S_m(\mathbf{k})d\mathbf{k}. \quad (7)$$

The general expression for $S_m(\mathbf{k})$ through the arbitrary spectral density $S(\mathbf{k})$ of the fluctuations of local anisotropy axis has the form [3, 4, 17]

$$S_m(\mathbf{k}) = \left(\frac{K}{A}\right)^2 \frac{S(\mathbf{k})}{(k_H^2 + k^2)^2}, \quad (8)$$

where $k_H = 1/R_H$ is the wave number of exchange correlations. If we model the stochastic properties of the orientational irregularity of magnetic anisotropy by the simplest correlation function

$$K(r) = De^{-k_c r}, \quad S(k) = \frac{Dk_c}{\pi^2(k_c^2 + k^2)^2}, \quad (9)$$

where $k_c = 1/R_c$, we obtain a symmetric expression for $S_m(\mathbf{k})$:

$$S_m(k) = \frac{1}{\pi^2}\left(\frac{K}{A}\right)^2 \frac{Dk_c}{(k_H^2 + k^2)^2(k_c^2 + k^2)^2}. \quad (10)$$

In this case, the expression for the variance d_m has the form [4]

$$d_m(H) = \frac{(D^{1/2}H_a)^2}{H^{1/2}(H_{ex}^{1/2} + H^{1/2})^3}. \quad (11)$$

One can see that, for $H \gg H_{ex}$, Eq. (11) yields expression (3) for both inequalities (2) and (4) and, for $H \ll H_{ex}$, Eq. (11) yields expression (5).

We now consider a random function $\langle \mathbf{m}(\mathbf{x})\rangle_{R_f}$ that is obtained by averaging the random function $\mathbf{m}(\mathbf{x})$ over the space interval $(\mathbf{x} - |R_f|, \mathbf{x} + |R_f|)$:

$$\langle \mathbf{m}(\mathbf{x})\rangle_{R_f} = \frac{1}{(2R_f)^3} \int_{x-|R_f|}^{x+|R_f|} \mathbf{m}(\mathbf{x})d\mathbf{x}. \quad (12)$$

The stochastic properties of the orientational irregularities of the mean anisotropy $\langle K\rangle$ of magnetic blocks are determined by the rules described in [18]. In the case $R_f \gg R_c$, these properties are described by the correlation function and the spectral density of the following types:

$$\tilde{K}(r) = D\left(\frac{k_f}{k_C}\right)^3 e^{-k_f r}, \quad \tilde{S}(k) = \frac{Dk_f}{\pi^2(k_f^2 + k^2)^2}\left(\frac{k_f}{k_c}\right)^3, \quad (13)$$

where $k_f = 1/R_f$. Substituting Eqs. (13) into Eq. (8) and then into Eq. (7), we obtain the following expression

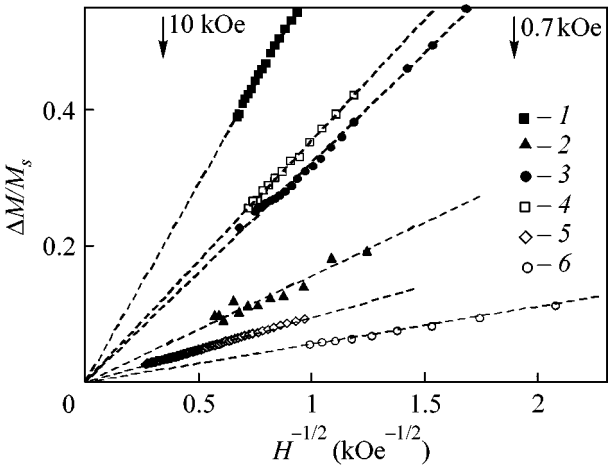

Fig. 3. High-field portions of the magnetization curves $M(H)$: (*1*) $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ and (*2*) $Co_{80}Zr_{10}$ amorphous tapes; (*3*) a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ nanocrystalline tape; $Co_{90}P_{10}$ amorphous films with t = (*4*) 500 and (*5*) 2000 Å; and (*6*) a $Co_{90}P_{10}$ amorphous coating with t = 30 μm.

for the variance of the random deviations of $\langle \mathbf{m}(\mathbf{x})\rangle_{R_f}$:

$$d'_m(H) = \frac{(D^{1/2}H_a)^2}{H^{1/2}(H_f^{1/2} + H^{1/2})^3}\left(\frac{R_c}{R_f}\right)^3 = \frac{(D^{1/2}\langle H_a\rangle)^2}{H^{1/2}(H_f^{1/2} + H^{1/2})^3}. \quad (14)$$

Here, the rms fluctuation of anisotropy in a magnetic block is $D^{1/2}\langle H_a\rangle = D^{1/2}H_a/(R_c/R_f)^{3/2}$ and the field H_f is determined as $H_f = 2A/MR_f^2$. A direct substitution of R_f yields $H_f \equiv D^{1/2}\langle H_a\rangle$. Hence, Eq. (14) is valid only for the fields $H > H_f = D^{1/2}\langle H_a\rangle$. In this field range, Eq. (14) is reduced to Eq. (3'). Figure 2 shows dependences (11) and (14) as functions of magnetic field in $D^{1/2}\langle H_a\rangle$ units. One can see that, up to $H \approx 10D^{1/2}\langle H_a\rangle$, the variations of d_m are insignificant, whereas d'_m drops to zero (within the experimental error). This means that the magnetization of amorphous and nanocrystalline ferromagnets occurs through the alignment of the mean magnetizations of the blocks with the field [according to Eqs. (3') and (14)] and only after that the decrease in the amplitude of $\mathbf{m}_\perp(\mathbf{x})$ [described by Eqs. (5), (5'), and (11)] takes place.

3. Figure 3 presents the high-field portions of the magnetization curves $M(H)$ for films and foils of amorphous and nanocrystalline alloys produced by different techniques (fast quenching of the melt or chemical deposition). The magnetization curves were obtained using vibrating-coil magnetometers with an electromagnet for fields of up to 15 kOe and with a superconducting

solenoid for fields of up to 30 kOe. One can see that, in the $(\Delta M/M_s, H^{-1/2})$ coordinates, the experimental curves can be described by linear dependences (of the type $y = ax$) determined by Eqs. (5) and (5'). This means that inequality (4) is valid for the amorphous and nanocrystalline alloys under study. The experimental curves also suggest that the numerical value of the coefficient of linear dependence is determined by the characteristics of the random anisotropy, R_c and $D^{1/2}H_a$. To calculate the latter, it is necessary to measure the magnetization curve in the fields $H > H_{ex}$. By recording the part of the curve described by dependence (3), one can determine $D^{1/2}H_a$. The revealed crossover of $M(H)$ (the transition from $\Delta M \sim H^{-1/2}$ to $\Delta M \sim H^{-2}$) provides the possibility to measure H_{ex}. Substituting the exchange interaction constant A, which is calculated from the low-temperature thermomagnetic curves $\Delta M \sim (T/A)^{3/2}$, into H_{ex}, one can determine R_c. However, in many cases this program cannot be implemented. Many amorphous and nanocrystalline ferromagnetic alloys are characterized by the values of H_{ex} that exceed the maximal fields used in the experimental measurements of $M(H)$ (see, e.g., [16]). Then, the grain size (or the cluster size) $2R_c$ can be determined by direct methods such as the X-ray structural analysis or transmission electron microscopy and the values of H_{ex} and $D^{1/2}H_a$ can be calculated (the latter is obtained from the linear dependence $\Delta M \sim H^{-1/2}$ similar to that shown in Fig. 3).

For the characteristics of the magnetic microstructure (R_f and $D^{1/2}\langle H_a\rangle$) the situation is entirely different. For their indirect determination, it is sufficient to record the linear dependences shown in Fig. 3. The measurement of the coefficient of linear dependence allows one to calculate the rms fluctuation of the anisotropy field in a magnetic block, $D^{1/2}\langle H_a\rangle \equiv H_f$. The substitution of A into H_f provides the value of R_f. For the amorphous and nanocrystalline alloys presented in Fig. 3, the following values of $D^{1/2}\langle H_a\rangle$ and R_f were obtained: 340 Oe and 130 Å, respectively, for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous tape 20 μm thick; 25 Oe and 640 Å for a $Co_{80}Zr_{10}V_{10}$ amorphous tape 30 μm thick; 100 Oe and 240 Å for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ nanocrystalline tape 20 μm thick; 120 Oe and 380 Å for a $Co_{90}P_{10}$ amorphous film 500 Å thick; 9 Oe and 1430 Å for a $Co_{90}P_{10}$ amorphous film 2000 Å thick; and 3 Oe and 2460 Å for a $Co_{90}P_{10}$ amorphous coating 30 μm thick. To directly determine the values of R_f and $D^{1/2}\langle H_a\rangle$, it is necessary to record the part of the dependence $M(H)$ that is described by Eqs. (3') and (14) rather than by Eqs. (5), (5'), and (11). With a vibrating-coil magnetometer, such a measurement is apparently impossible, because real amorphous and nanocrystalline alloys contain microcracks, pores, and inclusions of a second phase, giving rise to magnetostatic mechanisms of scattering for the magnetic moment [these mechanisms are not included in Eqs. (1) and (1')]. As a result the true values of H inside the sample do not coincide with the external magnetic field strength at $H \sim H_f$.

This difficulty can be overcome with the use of an experimental technique that allows one to exclude the effect of magnetostatic mechanisms. Such a technique is realized by a magneto-optic micromagnetometer [19] allowing the measurements of the local magnetization curves by using the equatorial Kerr effect $\delta(H) \sim M(H)$ with the light spot diameter of 1 to 30 μm in magnetic fields of up to 200 Oe. In this technique, the external magnetic field is applied along the sample surface normally to the plane of light incidence. Before measuring the local magnetization curves $M(H)$, the system is tuned so as to depart from the significant magnetostatic sources. For this purpose, it is necessary to construct the distribution curves $\delta(L)/\delta_s \sim M(L)/M_s$ at a constant external magnetic field H (much lower than the saturation field H_s). The curves are obtained by scanning the light spot over the sample surface along an arbitrarily chosen direction. (The scan length L is chosen so as to exceed the spot diameter by two to three orders of magnitude). Typical distribution curves can be found in [20]. They exhibit irregular deviations of M from the mean magnetization $\langle M\rangle$. As the field is increased and the scanning along L is repeated, the value of $\langle M\rangle$ increases and the deviations decrease, but the spatial scale of deviations is retained. The field H_s is determined as the one at which the amplitude of the deviations is of the order of experimental error. For our samples, the following deviations were observed: 200–300 μm for a 500 Å-thick $Co_{90}P_{10}$ amorphous film; 120–150 μm for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous tape; and 50–70 μm for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ nanocrystalline tape. The local magnetization curves $M(H)$ were obtained from the light spot, 20–30 μm in diameter, set at the center of a soft magnetic region. It was believed that the deviations of $\mathbf{M}(\mathbf{x})$ in this region are caused by the scattering due to the chaotic orientation of the axis $\mathbf{n}(\mathbf{x})$ of a magnetic block.

Figure 4 shows the local magnetization curves $M(H)$ for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous foil and a $Co_{90}P_{10}$ amorphous film; the curves were measured for different parts of the samples. One can see that, in the $(\Delta M/M_s, H^{-2})$ coordinates, the experimental magnetization curves are described by linear dependences of the type of Eq. (3'). The slopes of these dependences characterize the values of the rms fluctuation of the mean anisotropy field in a magnetic block, $D^{1/2}\langle H_a\rangle$. For the curves presented in Fig. 4, we obtained: $D^{1/2}\langle H_a\rangle \approx 3$–$6$ Oe and $R_f \approx 970$–1300 Å for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ foil and $D^{1/2}\langle H_a\rangle \approx 60$–$70$ Oe and $R_f \approx 500$–570 Å for a $Co_{90}P_{10}$ film (500 Å thick). One can see that the difference between the values of $D^{1/2}\langle H_a\rangle$ and R_f determined for the $Co_{90}P_{10}$ amorphous film by the direct [Eq. (3')] and indirect [Eq. (5')] methods does not exceed 50%. The greater difference between the corresponding values obtained for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ tape is no surprise.

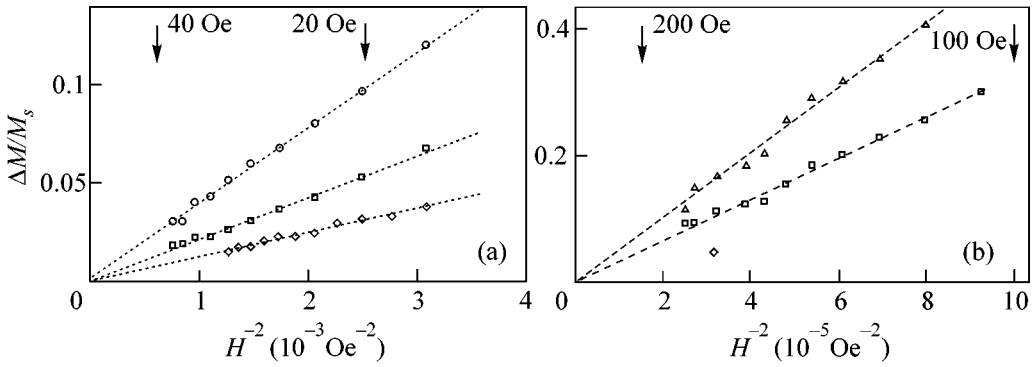

Fig. 4. Local magnetization curves $M(H)$ measured for different parts of (a) a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous tape (t = 20 μm) and (b) a $Co_{90}P_{10}$ amorphous film (t = 500 Å).

The point is that the local signal $\delta(H) \sim M(H)$ is obtained from a thin surface layer ~200 Å thick. Therefore, a coincidence between the "surface" signal $M(H)$ and the integral value of $M(H)$ should be expected only for films whose thickness is comparable to the penetration depth in the magneto-optic technique. The latter condition is fulfilled for the 500 Å-thick $Co_{90}P_{10}$ amorphous film and does not hold for the foils showing noticeable differences between the values of $D^{1/2}\langle H_a\rangle$ and R_f obtained for the bulk of the samples and for the surface layer of the material.

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