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Magnetization correlations and random magnetic anisotropy in nanocrystalline films $Fe_{78}Zr_{10}N_{12}$

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Abstract. The quantitative analysis of static ferromagnetic correlations in nanocrystalline films $Fe_{78}Zr_{10}N_{12}$ was performed by two methods: the correlation magnetometry technique and magnetic force microscopy. The data, obtained by both methods, prove to be in good agreement.

Introduction

The excellent soft magnetic properties of nanocrystalline (NC) ferromagnets are realized in a case, when the magnetic correlation length or stochastic domain size $2R_L$ are larger, than the average crystallite size $2R_c$, considered to be a ‘‘structural correlation length’’ [1-5]. Thus, it is important to quantify the volume-averaged magnetic correlation length or stochastic magnetic domain size in NC soft magnets. To provide the image of magnetic microstructure in the near vicinity of the specimen surface, the following microscopic methods are used: Lorentz microscopy [6], Kerr microscopy [4], and magnetic force microscopy [7]; some information about the magnetic microstructure of the bulk magnetic materials can be obtained by means of magnetic small-angle neutron scattering [1, 3, 7].

Unlike these methods, a new technique for the quantitative analysis of static ferromagnetic correlations in NC bulk ferromagnets has been developed [8-9]. This technique, based on the evaluation of the correlation function of magnetization parameters, is used in this paper for quantitative analysis of static ferromagnetic correlations in Fe-Zr-N films, exhibiting an unique combination of high saturation induction (up to 1,8 T) and low coercive force (0.3 - 0.4 Oe) [10]. The magnetic correlation length or stochastic domain size $2R_L$ was evaluated in this paper by magnetic force microscopy method as well. The soft magnetic properties of these films are formed due to interplay of random magnetic anisotropy and magnetization correlations [6, 11-12]. Thus, these films can be considered as the good candidates for fundamental investigations of magnetization correlations.

Theoretical background

The inhomogeneous orientation of the magnetic moment $\vec{M}(\vec{x})$ in nanomagnets is described by the correlation function $K_m(\vec{r})$ of the magnetization components, orthogonal to applied magnetic field (Eq. 1a). According to [8-9] the correlation function $K(\vec{r})$ of the orientation of local anisotropy axes (Eq. 1b) may be written as

$$\left\langle \vec{m}_\perp(\vec{x})\vec{m}_\perp(\vec{x}+\vec{r})\right\rangle = K_m(\vec{r}) \qquad (1a)$$

$$K_m(\vec{r}) = \frac{(K/A)^2}{8\pi k_H}\int K(\vec{r}')\cdot e^{-k_H|\vec{r}-\vec{r}'|}d\vec{r}' \qquad (1b)$$

here $\vec{m}(\vec{x}) = \vec{M}(\vec{x})/M$ is the magnetization unit vector, K is the effective anisotropy energy, A is the exchange parameter, $k_H = (M_s H/2A)^{1/2}$ is the wave number of exchange correlations, and H is an external magnetic field.

The function of magnetic correlation $K_m(r)$ decreases exponentially: $K_m(0)\cdot\exp(-r/R_m)$; here $K_m(0)$ is the magnetization dispersion, R_m is the correlation radius of magnetization, herein the quantity $K_m(0)$ and R_m depend upon the applied field. This dependence can be visualized by characteristic fields $H_L=2A/M_sR_L^2$ and $H_R=2A/M_sR_c^2$. In the "weak" fields ($H< H_L$) correlation radius $R_m = R_L$, in the "strong" fields ($H> H_R$) $R_m = R_c$. At $H_L<H<H_R$ the value of $R_m = R_H = (2A/M_sH)^{1/2}$, here $R_H = 1/k_H$ is known to be characteristic length of the magnetization ripple [6, 8-9]. Using analytic theory and numerical calculation and taking into account stochastic domain formation, one can present the field dependence of the magnetization dispersion $K_m(0) = d_m(H)$ in a form [9]:

$$d_m(H) = \left(\frac{aH_a}{H}\right)^2 \left(\frac{H_L^{1/2} + H^{1/2}}{H_R^{1/2} + H^{1/2}}\right)^d \equiv a^2 \left(\frac{R_c}{\delta}\right)^4 \frac{(R_H / R_c)^{4-d}}{(1 + R_c / R_H)^d} \cdot \left(1 + \frac{R_H}{R_L}\right)^d \qquad (2)$$

Here d is the dimension of exchange–coupled areas in the grain; $\delta=(A/K)^{1/2}$; $H_a=2K/M_s$ is local magnetic anisotropy field and a is the symmetry coefficient, equal to $1/15^{1/2}$ for uniaxial anisotropy. It can be seen, that the character of the field dependence $d_m(H)$ changes in the vicinity of characteristic fields H_R and H_L. The dispersion $d_m(H)$ determines the magnetization approach to saturation law: $M/M_s \approx 1 - d_m(H)$. The measurement of dispersion in the course of magnetization approach to saturation and the following it's interpretation it in the terms of Eq.2 allows one to determine the fields H_a, H_R, H_L and evaluate the lengths $2R_c$ and $2R_L$. This is the basic point of correlation magnetometry technique.

Experiment

For the investigation, we prepared the film of compositions $Fe_{78}Zr_{10}N_{12}$ ~ 0.8 μm thickness on a heat-resistant glass substrate.The film was prepared by dc reactive magnetron sputtering of Fe - 10 at.% Zr target. The following preparation conditions were used for the film: the preliminary vacuum in the chamber was $P_0 = 1\cdot10^{-6}$ dyn/cm^2, Ar+N_2 mixture pressure was $P_{(Ar+N2)} = 6.65$ dyn/cm^2 and the nitrogen pressure was $P_{N2} = 3.3\cdot10^{-2}$ dyn/cm^2; the nitrogen content in the gas mixture was 0.5 %. The cathode power equals $1.5\cdot10^9$ erg/s; the sputtering time was τ = 100 min. The sputtered film were annealed at 400° C during 1 h in vacuum of ~$2.7\text{x}10^{-2}$ dyn/cm^2. Magnetic measurements were carried out with the vibrating sample magnetometer The Quantum Design PPMS in the fields up to 90 kOe applied along the film plane and in the temperature range from 2 K to 300 K. The $M(T)$ measurement were carried out in the applied field 20 kOe that corresponds to practically uniform magnetization i.e. ferromagnetic saturation. Magnetic force microscopy (MFM) data were carried out on atomic force microscope NT-MDT Smena with Co-coated cantilever in two pass mode, the second pass at a height of 62.2 nm above the surface. It was shown by X-ray diffraction that the obtained films had a random mixed (nanocrystalline + amorphous) structure without any texture [10]. The ferromagnetic phase, which was a nitrogen supersaturated bcc alfa-FeN solid solution, is nanocrystalline with the grain size in the bcc phase, depending on the annealing conditions, changes from ~ 3 to ~25 nm.

Results and discussion

The approaches magnetization to saturation curves at temperatures 2 K and 300 K are close to each other. In the fields $H>$ 50 kOe the magnetization tends to the saturation value (M_s = (1060 ± 30) G at 2 K and M_s = (940 ± 30) G at 300 K) and after that it is constant (fig.1). These values allow us to calculate the magnetization dispersion as $d_m(H) \approx 1 - M/M_s$ to interpret the magnetization approach to saturation in the terms of Eq.2. (see the theoretical background). The log-log plots of $d_m(H)$ revealed the three different parts that corresponded to the three different

power dependences and separated by two field values $H_R = (22 \pm 1)$ kOe and $H_L = (1.3 \pm 0.1)$ kOe (fig.2). There are two linear areas in the log-log plots of $d_m(H)$ (fig.2) characterized by different power dependences and the third part corresponds well to eq.2: the first area is placed at applied field less then $H_L = (1.3 \pm 0.1)$ kOe, the second one is placed at applied field between H_L and H_R, and the third one is placed at applied field higher then $H_R = (22 \pm 1)$ kOe.

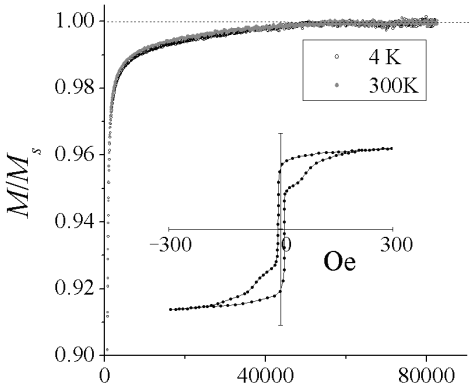

Fig.1. Approach magnetization to saturation in $Fe_{78}Zr_{10}N_{12}$ film.

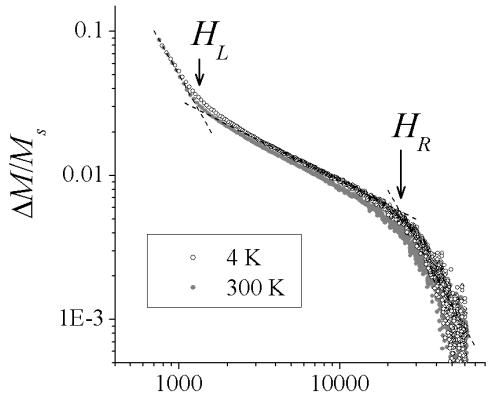

Fig.2. Log-log $\Delta M(H)/M_s$ plots of $Fe_{78}Zr_{10}N_{12}$ film.

The part of the log-log plot for the fields above H_R (fig.2) is consistent with Eq.2 and it could be processed by Akulov's law for magnetization approach to saturation $d_m(H) = (aH_a/H)^2$. The determination of the anisotropy field values by fitting of this part by C/H^2 expression and taking in account the value $a = 1/15^{1/2}$ allow to determine the anisotropy energy $K = H_aM_s/2$: $K = (3.8 \pm 0.1)\cdot 10^6$ erg/cm^3 at 2 K and $K = (3.3 \pm 0.1)\cdot 10^6$ erg/cm^3 at 300 K. The linear part of log-log plot for the fields below H_L is characterized by tangent equal to -2 (fig.2). This part also is consistent with Eq.2 and corresponds to the regime of stochastic domains formation. The tangent of linear part of the log-log plot for the fields between H_L and H_R is about 0.5 which is consistent with Eq.2 with $d = 3$. The value of R_c is estimated by equation $R_c = (2A/M_sH_R)^{1/2}$ using H_R, M_s and the value of $A = (6.1 \pm 0.4)\cdot 10^{-7}$ erg/cm measured for this film [12], it's equal to $R_c = (2.2 \pm 0.1)$ nm. The magnetization correlation radius of the magnetic microstructure of the films was estimated, in accordance with [8-9], to be $R_L = 4A^2/K^2R_c^3 = (0.130 \pm 0.04)$ μm for the 300 K.

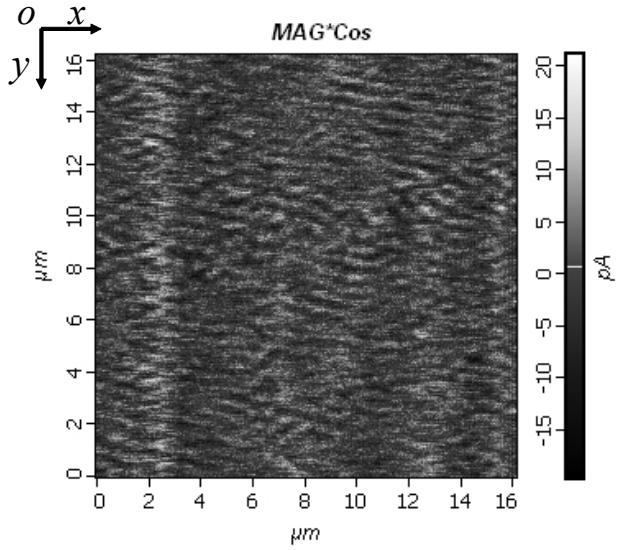

Fig.3. MFM image of $Fe_{78}Zr_{10}N_{12}$ film.

The MFM film surface image at zero applied fields reveals the contrast, visualizing the film magnetization heterogeneity (fig.3). We assume that each pattern in fig.3 represents a ferromagnetic correlation volume with fluctuating magnetization direction due to the average random magnetocrystalline anisotropy of the grains as it discussed in theoretical background of the paper.

There is no strict conformity MFM image contrast and magnetization heterogeneity and yet the main scales characterize uniform areas and statistical properties of both types of heterogeneities would be the same. Contrast correlation functions are calculated in the following way:

$$C(r) = \left\langle \left(z(r') - \langle z \rangle\right)\left(z(r'+r) - \langle z \rangle\right) \right\rangle, \quad (3)$$

here $z(r')$ is the grayscale intensity of the pixel with coordinates r'. Because there is anisotropy of MFM image contrast we choose two perpendicular axes (*ox* and *oy*) corresponded long and short contrast waves and then calculate $C(r)$ averaging by eq.3 along each axes (fig.4, 5).

The exponential decrease of correlations ~ exp(-r/R_L) along *ox* axis and damped oscillations like ~ $(1+r/R_L)^{-3/2}\cos(r/R_L)$ along *oy* are revealed. The magnetic correlation radii R_L^{ox}=(0.170 ± 0.009) μm and R_L^{oy} = (0.090 ± 0.005) μm are in a good quantitative agreement with that one, estimated by correlation magnetometry.

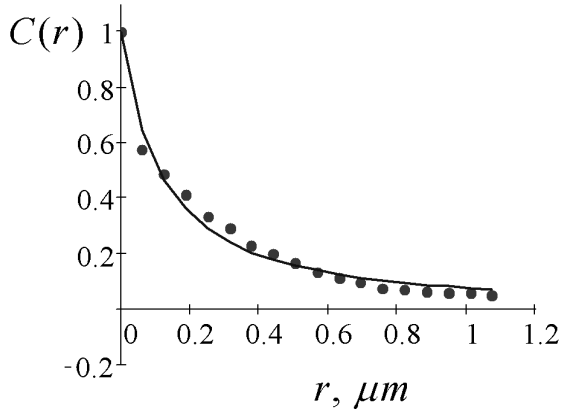

Fig.4. MFM image contrast correlations along *ox* axis. Solid line is fitting by $C(r)=\exp(-r/R_L)$.

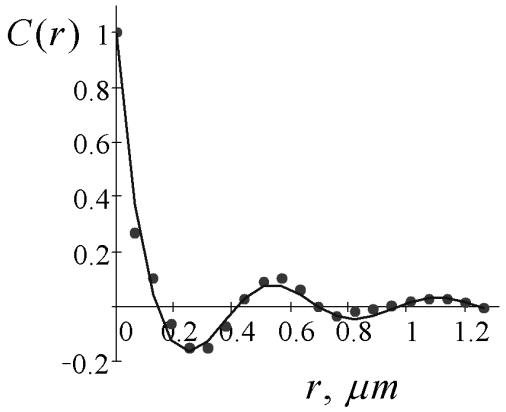

Fig.5. MFM image contrast correlations along *oy* axis. Solid line is fitting by $C(r)=(1+r/R_L)^{-3/2}\cos(r/R_L)$

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