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Hysteresis, critical fields and superferromagnetism of the film with perpendicular anisotropy

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ABSTRACT

This paper is focused on the analysis of hysteresis and critical phenomena of magnetization reversal of superferromagnetic (SFM) state in nanogranular (NG) Co/Al_2O_3 film with perpendicular anisotropy. It was demonstrated that the transition from the multidomain SFM state to the homogeneous SFM state, during the magnetization process, occurs critically. The value of the field of critical transition to the homogeneous state depends on the demagnetization field, granular anisotropy and interparticle exchange anisotropy. It turned out that the temperature dependence of the coercive force of the film, despite its SFM state, accords with the Neel–Brown formula for anisotropic single-domain ferromagnetic particles, but has an anomalous angular dependence. It was concluded that domain wall motion affects these features of the coercive field. The domain wall movement may occur due to the overturn of magnetic moments of particles in the boundaries between the superdomains. At the same time, the main factors influencing the coercivity are the anisotropy of the particles, which blocks their magnetic moment reorientation, and demagnetizing factor of the film. Together they lead to the anomalous angular dependence of the coercive field.

1. Introduction

Films with growth-induced perpendicular anisotropy (where easy magnetization axes of granules are perpendicular to the film plane) are promising for application in microelectronics, spintronics, as well as RF devices [1–3]. They may be interesting as model systems with interparticle magnetic interactions realized on different spatial scales. Moreover, the choice of such films as the model systems is promoted by the identical anisotropy orientation for all particles in the ensemble.

Typically, there are two types of intergranular interactions in NG films: dipole-dipole and exchange interactions [4–6]. The dipole–dipole interaction provides long-range magnetostatic contribution, known as demagnetization field. Its value and orientation depends on the shape of the sample. Such contribution leads to collective easy-plane anisotropy for orientation of magnetic moment of the film as the whole. In addition, the dipole-dipole interaction between neighboring particles will create a microscopically inhomogeneous local field on each particle, which can affect the cross-correlation of the magnetic moments of the particles. The latter will be also fluctuating with the random arrangement of the particles and the lack of ordering of their magnetic moments directions.

When present, interparticle exchange-type interaction is proportional to scalar product of the magnetic moments of particles, promoting the correlation of the magnetic moments orientations. Interparticle ferro-type exchange interaction can contribute to the formation of superferromagnetic (SFM) state in NG film [7–14]. In such state, the average magnetic moment of the particle will be different from zero, and it will be directed along the easy magnetization axis of the ensemble at $T < T_{SF}$ [15], where T_{SF} is SFM ordering temperature. The SFM state, as ordered phase of superspins of granules, has common features with the conventional ferromagnetic phase of crystals. It should be mentioned, that the authors of [15] argued that magnetization curves are corresponding to Langevin magnetization even below the temperature of SFM ordering. But, such statement was not confirmed in subsequent reports [14,16]. The stability boundary of SFM phase in the system of the particles with oriented in plane anisotropy was determined both theoretically and experimentally in [17].

The mismatch in the directions of the easy magnetization axes of single-particle anisotropy and collective demagnetising anisotropy of particles ensemble as whole can cause their competition. That can be observed in films with perpendicular anisotropy. In such films, when its magnetic moment is oriented normal to the film plane, the positive energy of demagnetization prevents the formation of homogeneous SFM state with parallel orientation of the particles magnetic moments. Thus, the competition will result

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in an equilibrium multidomain SFM state [18] with codirectional magnetic moments of the particles within one superdomain. In such case, the magnetic moments of the granules inside superdomains are aligned parallel by intergranular ferromagnetic exchange-like interaction, and will be perpendicular to the film plane.

The magnetization reversal of the SFM state, with superdomains, requires additional research. The structure of the superdomain boundaries and their types were investigated only in few papers [19]. Magnetization reversal of such state should be essentially different from conventional magnetization process of multi-domain ferromagnet (FM). Unlike ordinary FM, SFM state is characterized by much lower interparticle exchange energy compared to the anisotropy energy. Due to this, the wall between superdomains can be very thin or contain no granules at all. Thus, the boundary of two superdomains can lie between two particles belonging to different superdomains.

There are two possible types of magnetization reversal. In the first one, all particles within the superdomain with magnetic moment directed against the applied magnetic field, can reverse their magnetization (turn their magnetic moments along the field) simultaneously. In the second one, the magnetization reversal occurs by the motion of the domain wall [20] and consistent magnetization reversal of single particles, similar to a conventional magnetization reversal of multi-domain FM. The coercive properties of the film should be determined by the nature of superdomain magnetization reversal. Moreover, the magnetic moments of the particles may be blocked by individual anisotropy. Such blocking is temperature-dependent and may affect the coercivity of the film [21,22].

The competition of perpendicular anisotropy vs anisotropy of demagnetization field leads to the inhomogeneous state of the film in certain range of fields, even without the SFM state and exchange interaction between the granules. The magnetic moments of one part of granules are oriented along the field direction (or close to it) while the other part has opposite direction. The transition from inhomogeneous magnetized state of the ensemble of single-domain granules to homogeneous magnetized state occurs critically under the influence of magnetic field. The transition from multidomain (nonuniformly magnetized) SFM state to single-domain (uniformly magnetized) state in films with perpendicular anisotropy should also occur critically. In addition, interparticle exchange interaction and temperature can modify the critical fields values comparing with those considered in [23] for film with perpendicular anisotropy of granules without SFM ordering.

Current paper is focused on experimental and theoretical analysis of the coercive and critical properties of magnetization reversal process of the films with perpendicular anisotropy in SFM-ordered state. The presence of SFM state in tested films was confirmed by magnetic force microscopy measurements.

In the paper there will be presented the results of experimental studies of magnetic properties of NG Co/Al_2O_3 film (ferromagnetic granules in a nonmagnetic matrix of aluminum oxide), with a relative content of cobalt 57.2 at%, slightly below the percolation threshold. The exchange interparticle interaction between the cobalt granules becomes notable along with the dipole–dipole interparticle interaction in the composite film with such high content of granular material. Previously, the occurrence of SFM state in films from this growth series with similar component content was shown in [24–26]. Other papers [27,28] demonstrated that such films possess growth-induced perpendicular magnetic anisotropy.

The previous research of Co/Al_2O_3 films with high content of granules were mainly conducted at high temperatures [29,30]. In the current research, the magnetic properties of the mentioned film will be analyzed in the wide temperature range, starting from high, when the reorientation of the magnetic moment of granules is not blocked, and ending with low, when thermally activated processes are blocked with anisotropy (are slowed relative to the measurement time). The effect of temperature on the critical transition fields from inhomogeneous SFM state to uniformly magnetized state of the film in tilted magnetic fields will be studied. It will be shown both experimentally and theoretically, that in the case, when the value of intergranular exchange interaction is sufficient for the formation of the SFM state and inhomogeneous magnetization of the film is realized through the formation of superdomains, such transition occurs critically. In addition, the detected abnormal angular dependence of the coercive force and its temperature dependence will also be discussed. For the explanation of the latter, an idea about the anisotropic intergranular exchange-like pair interaction will be involved.

2. The samples and the measurements methods

The samples of Co/Al_2O_3 NG composite films, about 5 μm thick, were prepared by ion-beam sputtering of composite targets on pyroceramics substrate (thickness about 1 mm) in the Voronezh Technical University (Russian Federation). As described in [27], the sputtering was performed in Ar atmosphere at the pressure of 3.2×10^{-5} Torr. For the obtaining of the set of films with different relative volume of granules, f_v, the components were sputtered on the long (≈ 25 cm) substrate. Co and Al_2O_3 targets were located near its opposite ends. The sputtering with such configuration provides wide range of component content in composite on one long film. The relative content of Co and Al_2O_3 varies in substrates length depending on the distance between the considered point and the targets. After the film deposition, the substrate was cut across its length into samples, the narrow strips (2×10 mm) with the different content of Co. The content of Co in the samples was determined by X-ray EPMA. The granules sizes were determined using transmission electron microscopy (TEM) of the film cross section. Typical TEM images for the number of samples of the series are given in [24,29,31]. The patterns of electron diffraction of granules were also shown there. They imply randomly oriented predominant Co cubic crystal structure in the granules. The crystallographic anisotropy of cubic Co is small [32]. The TEM images show the elongation of granules predominantly in the direction perpendicularly to the substrate. The average size of the long axis of the granules, estimated from TEM images of films with similar concentrations, is about 6 nm, and the long to short axis ratio is about 1.5 [24]. As it was argued in [31], such elongation leads to the presence of the magnetic anisotropy in the granules that is directed perpendicularly to the plane of the substrate.

One of the key parameters of NG composite films is relative volume (f_v), occupied by ferromagnetic granules in the film [33]. The threshold of contact percolation (when electrical contact between metal granules in the film occurs), f_{vp}, was determined [27], from the dependence of electrical conductivity of the film on the value of f_v. According to that, percolation threshold in Co/Al_2O_3 NG films is about 61 at% Co. Such high percolation threshold corresponds to the approximate relative volume of granules f_v in the film close to 0.66. This may serve as an argument in favor of the assumption that a dielectric shell, a surface layer of Co oxide in this case, coats each of the granules.

Despite the small spacing between the granules and such high relative volume of the granules in the film, $f_v \to 64$ at%, the cluster of electrically contacting granules did not form. In terms of electrical (contact) percolation, the investigated Co/Al_2O_3 film with 57.2 at% Co (and $f_v = 0.64$) was not percolated.

The estimation of the field of shape anisotropy H_A^{gr} for separate