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The magnetization model of multilayered composite thin films: Beyond the effective-medium theories

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ABSTRACT

Multilayered composites consisting of many thin ferromagnetic films with in-plane magnetic anisotropy separated by non-magnetic dielectric layers of different sizes are experimentally and theoretically investigated. Thin samples as well samples with transverse sizes comparable with longitudinal ones are used. The measured static magnetic properties of the bulk sample are found to be different from the properties of constituent thin films. This is an evidence for strong interactions between the magnetic layers in the sample, which interact at distances exceeding greatly the distance between adjacent magnetic layers. A theoretic model is developed taking into account magneto-dipole interactions between iron films in a multi-layer system. The model explains the anomalously high demagnetization field of the sample observed in the measurements.

1. Introduction

Ferromagnetic multilayered films attract much attention because of their applications in spintronics [1], high-density magnetic recording media, and microwave devices [2]. For example, bulk magnetic materials with high microwave permeability needed for many technical applications are of interest recently [2–5]. A promising approach to develop such materials employs composites based on thin ferromagnetic films [5,6]. However, microwave magnetic properties degrade with the growth of the film thickness due to decrease in microwave permeability and higher magnetic losses at low frequencies. Conventionally, this is attributed to the effect of eddy currents [7]. To avoid this degradation, multi-layer thin films composed of alternating magnetic and dielectric layers are used [8]. Besides of high microwave permeability, such a composite materials consisting of alternating magnetic and non-magnetic dielectric or metallic thin films demonstrate extraordinary magnetic and transport properties, such as tunneling magnetoresistance, spin injection, etc. Both magnetostatic and high-frequency properties of these materials are determined in many respects by the interaction between ferromagnetic layers.

In this paper we study magnetostatic properties of composite films consisting of up to several hundreds thin Fe layers, separated by non-magnetic dielectric layers of different widths. The detailed description of samples under study is given in the next section. Since the system is quite inhomogeneous in transverse direction, effective-medium theories cannot explain quantitatively experimental results. We proposed a quite simple model taking into account a magnetostatic interaction between ferromagnetic layers, assuming that exchange interaction between ferromagnetic layers is negligible in comparison to magnetostatic one. We show that magnetostatic interaction of two layers logarithmically decreases with the distance d between them, if d is much smaller than longitudinal sizes L of layers. Due to this long-range character of magneto-dipole interaction we should take into account an interaction between all ferromagnetic layers in the system.

2. Experiment

2.1. Description of samples

Films under study are deposited by the magnetron sputtering onto a $\Delta_0 = 10\,\mu$m thick flexible mylar substrate [9,10]. The preparation technique of the samples is described in detail in Ref. [11]. We measure two types of multi-layered film samples. The sample of the first type, 20-layered film sample, is produced by deposition of alternating iron layers of thickness $\Delta_m = 70$ nm and SiO_2 interlayers of the same thickness $\Delta = 70$ nm. The films consisting of 10 such iron layers are deposited onto *both* sides of the mylar substrate of thickness $\Delta_0 = 10\,\mu$m. The total thickness

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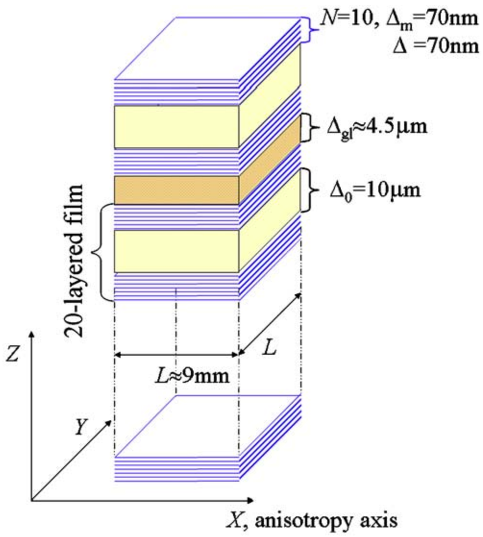

Fig. 1. (Color online) Schematic illustration of the 'bulk' sample consisting of N_0 2×10-layered composite films glued together. The thickness of the glue layers is $\varDelta_{gl} \approx 4.5\,\mu m$. Each 2×10-layered film consists of iron layers of thickness $\varDelta_m = 70\,nm$ alternating with SiO_2 layers of the same thickness $\varDelta = 70\,nm$ deposited onto a flexible mylar substrate of the thickness $\varDelta_0 = 10\,\mu m$. The in-plane easy axes of ferromagnetic layers are parallel to each other.

of 10-layered composite film is $D_{10} = 10\varDelta_m + 9\varDelta = 1.33\,\mu m$. As a result we have two 10-layered ferromagnetic composites films separated by non-magnetic layer of the thickness $\varDelta_0$ much (about 7.5 times) grater than the total thickness of the ferromagnetic films (20-layered film). The longitudinal sides of the films are $L \times L \approx 0.9 \times 0.9\,cm^2$. Films have in-plane magnetic anisotropy with easy axis parallel to one of their longitudinal side. The samples of the second type, 'bulk' samples, consist of N_0 number of such 20-layered films stacked together with the easy axes *parallel* to each other and glued under pressure. The thickness of the glue layers is $\varDelta_{gl} \approx 4.5\,\mu m$. We consider the sample of the second type, containing $N_0 = 36$ such 20-layered films. Thus, we have 72 stacks of 10-layered ferromagnetic composites films separated by alternating non-magnetic layers of thicknesses $\varDelta_0$ and $\varDelta_{gl}$ several times grater than the thickness D_{10} of 10-layered films. The schematic illustration of samples is shown in Fig. 1.

2.2. Experimental results

Magnetostatic properties were investigated using the vibrating sample magnetometer in magnetic field up to 9 kOe at room temperature [12]. The in-plane hysteresis loops in two orthogonal directions (along easy magnetization axis and along hard axis) were obtained. The results of measurements for single-layered, 20-layered Fe films, and for the 'bulk' sample are shown in Fig. 2a–c, respectively.

Coercive forces H_c are determined from these *in-plane* hysteresis loops. The saturation magnetizations for all samples, I_s, are evaluated in *out-of-plane* hysteresis loops of the films taking into account the demagnetization factor of thin film 4π, that is, $I_s = H_s/4\pi$, where H_s is the out-of-plane saturation field. We obtain $I_s = 1500 \pm 150\,Gs$ for all samples. The loop for single-layered sample (Fig. 2a) demonstrates the behavior typical for perfect magnetic films with in-plane magnetic anisotropy. The loop measured along the easy axis is almost square that indicates the absence of *in-plane* demagnetization field H_{dem} inside the film. The *in-plane* demagnetization factor of 20-layer film sample (Fig. 2b) can be estimated roughly as $N_x = 4\pi\delta/L \approx 2 \times 10^{-3}$, where $\delta = 20\varDelta_m = 1.4\,\mu m$ is a total magnetic thickness [13]. The corresponding demagnetization field $H_{dem} = N_x I_s \sim 3\,Oe$ is small enough. In a further theoretical study we will neglect it, changing the hysteresis loop measured for 20-layered film along the easy axis by step-like function. The 20-layered film sample has strongly pronounced in-plane magnetic anisotropy, as it can be seen from Fig. 2b. A small disorder in the directions of easy axes of different magnetic layers is affected in residual magnetization. However this effect is insufficient, and the hysteresis loops measured for 20-layered film look like loops of a single-layered ferromagnetic thin film.

In contrast to the previous cases the magnetization curve measured for the 'bulk' sample in the direction along in-plane easy axis is not square (see Fig. 2c). This is an indication of large demagnetizing field in the sample; its magnitude can be estimated as about $H_{dem} \sim 100\,Oe$ from the data in Fig. 2c. This demagnetizing field in 'bulk' sample arises from the interaction between ferromagnetic layers in different 10-layered films in it, although these films (of thickness $\varDelta_m = 70\,nm$) are separated from each other by distances up to several millimeters. Since the system is quite inhomogeneous in transverse direction, effective-medium theories cannot explain the experimental results quantitatively.

3. Theory of magnetization reversal

Here we propose a quite simple model taking into account a magnetostatic interaction of all ferromagnetic layers in the system. We suppose that easy axes of all layers are parallel to each other and directed along one of longitudinal sides of the sample (X axis). In addition we assume that each magnetic layer has uniform magnetization, I_s, and magnetic domains do not arise during magnetization reversal process. Even in framework of such simplified theory, one can explain quantitatively the hysteresis loops. We restrict ourself by the case of magnetization reversal *along* easy axis.

3.1. Magnetostatic interaction and magnetization reversal of two ferromagnetic layers

Let us consider first the system of two parallel uniformly magnetized square shaped ferromagnetic films of the thickness $\varDelta_m$ and the length L. The film's thickness $\varDelta_m$ and distance between centers of the films d are assumed to be much smaller than L. The energy of magneto-dipole interaction between ferromagnetic layers can be written as

$$U_{12} = I_s^2 \int_{V_1} d^3\mathbf{r}_1 \int_{V_2} d^3\mathbf{r}_2 \times \left[\frac{(\mathbf{e}_1\mathbf{e}_1)}{|\mathbf{r}_1 - \mathbf{r}_2|^3} - \frac{3(\mathbf{e}_1(\mathbf{r}_1 - \mathbf{r}_2))(\mathbf{e}_2(\mathbf{r}_1 - \mathbf{r}_2))}{|\mathbf{r}_1 - \mathbf{r}_2|^5}\right], \quad (1)$$

where unit vectors $\mathbf{e}_{1,2}$ describe the directions of magnetization vectors $\mathbf{I}_{1,2}$ of the films. The integration over $\mathbf{r}_1$ and $\mathbf{r}_2$ is performed over the films volumes $V_{1,2}$. We choose the coordinate system in such a way that Z axis is perpendicular to the plane of the films, and X and Y axes are directed along in-plane films sides. In the limit of large length L of the films in comparison to the their thickness $\varDelta_m$ and the distance d between them, one can derive an asymptotic formula for the interaction energy, which can be written as

$$U_{12} \cong 4I_s^2\varDelta_m^2 L \ln\left(\frac{0.461L}{d}\right) J(\mathbf{e}_1, \mathbf{e}_2), \quad (2)$$