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Room-temperature perpendicular magnetic anisotropy of MgO/Fe/MgO ultrathin films

A. Koziół-Rachwał,1 W. Słkowroński,2 T. Ślezak,1 D. Wilgocka-Ślezak,3 J. Przewoźnik,1 T. Stobiecki,2 Q. H. Qin,4 S. van Dijken,4 and J. Korecki1,3
1Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, al. Mickiewicza 30, 30-059 Kraków, Poland
2Department of Electronics, AGH University of Science and Technology, al. Mickiewicza 30, 30-059 Kraków, Poland
3Jerzy Haber Institute of Catalysis and Surface Chemistry, Polish Academy of Sciences, ul. Niezapominajek 8, 30-239 Kraków, Poland
4NanoSpin, Department of Applied Physics, Aalto University School of Science, P.O. Box 15100, FI-00076 Aalto, Finland

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We used the anomalous Hall effect to study the magnetic properties of MgO/Fe(i)/MgO(001) structures in which the Fe thickness \( t \) ranged from 4 Å to 14 Å. For the iron deposited at 140 K, we obtained perpendicular magnetization at room temperature below the critical thickness of \( t_c = (9 \pm 1) \text{ Å} \). In the vicinity of \( t_c \), the easy magnetization axis switched from an out-of-plane orientation to an in-plane orientation, and the observed spin-reorientation transition was considered in terms of the competition among different anisotropies. The perpendicular magnetization direction was attributed to magnetoelastic anisotropy. Finally, the temperature-dependent spin-reorientation transition was analyzed for Fe thicknesses close to \( t_c \).

I. INTRODUCTION

Perpendicular magnetic anisotropy (PMA) is a desirable property of ferromagnetic nanostructures; it is interesting for fundamental research, and it has essential applications in high-density magnetic random access memory.1,2 Following the early theoretical prediction of PMA in ultrathin films,3 many experimental studies have demonstrated that perpendicular spin alignment exists in various types of ferromagnetic films, especially 3d elemental and alloy films, transition-metal/rare-earth alloys, multilayers, and superlattices, among others.4 PMA has recently been observed for CoFeB/MgO5,6 and Co/oxide7 interfaces, which indicates that these metal oxide structures are promising candidates for ferromagnetic electrodes in next-generation high-density non-volatile memory. Although perpendicular magnetic order was postulated using first-principles calculations for a Fe/MgO interface in the ultrathin Fe-thickness range,8 most experimental studies have reported results concerning the in-plane easy magnetization axis down to the thinnest films of Fe.9–11 Nevertheless, we recently observed PMA in a MgO/Fe/MgO structure at low temperatures for an Fe thickness of \( (4 – 10) \text{ Å} \).12,13 However, as the temperature increased, the magnetic properties of the system became obscured by superparamagnetism. The superparamagnetic behavior of Fe/MgO is attributed to the 3-dimensional cluster growth of Fe on MgO(001), which is thermodynamically preferred because MgO has a lower surface energy (1.16 J/m²) than Fe (2.9 J/m²).14 On the other hand, Fahsold et al.15 have shown that the cluster formation in Fe/MgO(001) can be suppressed at low temperatures because of the reduced mass transport. They reported that the MgO(001) substrate was almost completely covered by a monolayer (ML) of Fe for a deposition temperature as low as 140 K.

The present paper focuses on the magnetic properties of ultrathin Fe films that were deposited on MgO(001) at low temperatures. We used the anomalous Hall effect (AHE), which is sensitive to the normal component of magnetization,16,17 to study the magnetic anisotropy in MgO/Fe(i)/MgO(001) as a function of the Fe thickness in the range of 4 Å < \( t < 14 \) Å. We confirmed the existence of perpendicular magnetization at room temperature (RT) below a critical thickness of \( t_c = (9 \pm 1) \text{ Å} \), and we monitored the spin-reorientation transition (SRT) from the out-of-plane direction to an in-plane direction with increasing Fe thickness. For a Fe thickness just above the critical thickness, we showed that the magnetization switched from the in-plane orientation to the perpendicular orientation when the temperature was decreased.

II. EXPERIMENTAL

The sample was prepared in an ultra-high vacuum (UHV) chamber, which was equipped with molecular beam epitaxy evaporators with metal vapor sources, an electron-beam evaporator for MgO deposition, and a quartz thickness monitor. A one-side polished MgO(001) single-crystal was used as a substrate. After annealing the MgO substrate crystal for 1 h at 770 K, a 100 Å thick MgO buffer layer was deposited at 720 K. The sharp low-energy electron diffraction (LEED) pattern that was collected at this stage confirmed that the MgO(001) surface structure was well ordered. The MgO buffer layer ensured a contamination-free (mainly carbon) surface. Using a movable shutter, a wedge-shaped Fe layer with a thickness of 4 Å to 14 Å was deposited at a rate of 1.5 Å/min (approximately 1 ML/min) on the substrate.
which was maintained at 115 K. The resulting slope of the Fe wedge was 1 ML/mm. Figure 1 shows the LEED pattern that was collected along the Fe wedge. The diffraction patterns for a Fe thickness of 4 Å resemble those of the MgO substrate (Fig. 1(a)). This resemblance can be explained by the formation of small Fe clusters during the initial growth on the MgO. The Fe(001) diffraction pattern appeared for an Fe coverage of approximately 6 Å (Fig. 1(b)) and became more intense and sharper with increasing Fe thickness (Figs. 1(c) and 1(d)). The Fe wedge was finally capped with a 100 Å thick MgO layer. On this sample, standard Hall bars that were 40 μm wide (along the Fe gradient) and 700 μm long (along the constant Fe thickness) were fabricated using photolithography and ion-beam etching. The 100 μm × 100 μm contact pads for the Hall bars were manufactured from 50 nm thick Al in a separate lithography step.

III. RESULTS AND DISCUSSION

The RT AHE hysteresis loops were measured as a function of the Fe thickness, where the external magnetic field was perpendicular to the surface of the Fe stripes. Representative AHE loops are shown in Fig. 2(a). For Fe \( t < 6 \) Å, the loops present no hysteresis and exhibit an s-shape, which is the characteristic of superparamagnetism, as shown by an exemplary loop for \( t = 5.9 \) Å. For higher thicknesses, hysteretic behavior appears. For \( t = 6.5 \) Å, we observed a 40% remanence. Above this thickness, the loops indicated an out-of-plane easy magnetization axis with a domain structure that evolved with the Fe thickness. In particular, for \( t = 8 \) Å, the hysteresis loop exhibits a full remanence, a small out-of-plane coercivity of 15 Oe, and a characteristic “hourglass” shape. This hysteretic loop is typical for a material in which a metastable single-domain state exists at remanence. A rapid decrease of PMA for 8.6 Å manifests in a characteristic loop with almost zero remanence, which is typical for fine-structured perpendicular domains. Further increase of the iron thickness resulted in a transition to the non-hysteretic hard loops that are typical of ultrathin Fe/Mg(001) films, which indicated an SRT to an in-plane easy axis (Fig. 2(b)).

By simulating the hard-axis loops, we quantitatively determined the anisotropy constants of the investigated system as a function of the Fe-film thickness. In the simulations, the single-domain model was assumed, in which the layer was described by the saturation magnetization \( M_s \) and the effective anisotropy constants \( K_1^{eff} \) and \( K_2^{eff} \), which included all contributions to the anisotropy energy density from various sources. The use of a second-order anisotropy constant was necessary to reproduce the shape of the loops. The equilibrium magnetization direction, which is defined in terms of

![FIG. 1. LEED pattern collected after the deposition of 4 Å (a), 6.0 Å (b), 10 Å (c), and 13 Å (d) of Fe on MgO(001) at an electron energy of 120 eV.](image1)

![FIG. 2. (a) and (b) Normalized AHE hysteresis loops measured at RT for different Fe thicknesses. The hard loops in (b) are shown alongside the simulation results (see text).](image2)
the angle $\theta$ measured from the surface normal, was determined by minimizing the free energy density, which is given by the following formula:

$$E(\theta) = -M_s H \cos(\theta - \phi) - K_1^{\text{eff}} \cos^2 \theta - K_2^{\text{eff}} \cos^4 \theta,$$

where $\phi$ is the polar angle that indicates the direction of the external magnetic field ($\phi = 0$ for the given experimental geometry). The first term in $E(\theta)$ denotes the interaction of the Fe-layer magnetization with the external magnetic field $H$, and the other two terms represent the first- and second-order contributions to the magnetic anisotropy energy.

The saturation magnetization value was an essential simulation parameter, and its uncertainty was the main source of experimental errors. We used two approaches to determine the range of plausible variation in $M_s$ as a function of the film thickness. First, the magnetization was determined as a function of temperature from Vibrating Sample Magnetometer (VSM) measurements that were performed using a Quantum Design Physical Property Measurement System (PPMS). During the measurements, a magnetic field of up to 20 kOe was applied in the Fe-film plane. The VSM measurements of two reference samples yielded $M_s = (1710 \pm 100) \text{ emu/cm}^3$ for 13 Å and $M_s = (1515 \pm 100) \text{ emu/cm}^3$ at RT for 9 Å of Fe. The values of $M_s$ for the intermediate Fe thicknesses were determined by assuming a linear $M_s(t)$ relation. The main contribution to the given $M_s$ errors arises from the determination of the sample volume. Additionally, the magnetization data must be corrected by considering the strong diamagnetic signal from the MgO substrate, which might overlap on a slowly saturating superparamagnetic component and contribute an additional systematic error, especially for the smaller thickness. On the other hand, our measurements of the nuclear resonance scattering of synchrotron radiation indicated that the average hyperfine magnetic fields, which are a measure of the local magnetization, remained bulk-like and independent of thickness down to thicknesses as small as 7 Å. Therefore, these two experimental observations were used as the limiting cases for the variation in $M_s$.

The simulation results are presented in Fig. 3 using the anisotropy-space diagram that is spanned by the anisotropy $K_s^{\text{eff}}$ and $K_2^{\text{eff}}$. The structure of the anisotropy space corresponds to three magnetic phases with different easy axes:\textsuperscript{19} perpendicular ($\theta = 0^\circ$), in-plane ($\theta = 90^\circ$), and canted ($0^\circ < \theta < 90^\circ$). For positive $K_1^{\text{eff}}$, the magnetization is perpendicular when $K_1^{\text{eff}} > -0.5 K_2^{\text{eff}}$ (phase I) and canted when $K_2^{\text{eff}} < -0.5 K_1^{\text{eff}}$ (phase II). For negative $K_2^{\text{eff}}$, two magnetization alignments are possible: an in-plane phase (III) for $K_2^{\text{eff}} < -0.5 \times K_1^{\text{eff}}$ and a “coexistence” phase (C) for $K_2^{\text{eff}} > -0.5 \times K_1^{\text{eff}}$, where the in-plane and the out-of-plane phases coexist as a result of the presence of two local minima in the anisotropy energy. The experimental effective anisotropy constants for successive Fe thicknesses were accessible for $K_1^{\text{eff}} < 0$ and $K_2^{\text{eff}} < -0.5 \times K_1^{\text{eff}}$, i.e., for the in-plane magnetization. Within the limits set by the $M_s$ uncertainty, the experimental points lay on a straight line that defines the SRT trajectory in anisotropy space and provides insight into the SRT mechanism. In general, three SRT scenarios can be distinguished:\textsuperscript{20} via the origin of the anisotropy space, via a canted phase, and via the coexistence of in- and out-of-plane phases. For the present MgO/Fe/MgO structure, within the experimental uncertainty, we observed an SRT from an in-plane direction to an out-of-plane direction through the narrow region of the coexistence phase state; however, the existence of an SRT path through the origin of the anisotropy space cannot be excluded.

A thickness-induced polar SRT is the result of competition among two or more anisotropies (e.g., volume, surface, magnetoelastic, etc.) that favor different easy magnetization directions, as has been shown for various systems.\textsuperscript{20} For the MgO/Fe/MgO structure, the perpendicular magnetization, which was observed experimentally at low temperatures, was attributed to either the epitaxial strains\textsuperscript{12} or to the interfacial anisotropy, which competes with the volume anisotropy.\textsuperscript{21} In the latter case, the surface character of the PMA is verified if the effective anisotropy constants can be phenomenologically separated into surface/interface and volume contributions that obey the following relations, which were postulated by Gradmann and Müller:\textsuperscript{21}

$$K_1^{\text{eff}} = K_{s1} + 2K_{s1}/t,$$

$$K_2^{\text{eff}} = K_{s2} + 2K_{s2}/t,$$

where $K_{s1}$ and $K_{s2}$ are the volume contributions, and $K_{s1}$ and $K_{s2}$ are the surface contributions (the prefactor of 2 is related to the assumption of two identical interfaces). The volume contribution $K_{s1}$ is usually dominated by the shape anisotropy ($-2\pi M_s^2$) and favors the in-plane magnetization alignment. If the surface term $K_{s1}$, which results from broken symmetry, is positive, it dominates at low thicknesses and perpendicular magnetization is observed. In Fig. 4, $K_1^{\text{eff}}/t$ and $K_2^{\text{eff}}/t$ are plotted as functions of the Fe thickness $t$. The critical thickness $t_c$, at which the effective anisotropy constant is zero, was determined to be $(9 \pm 1)$ Å. Within the experimental error, this critical thickness value is universal for $K_1^{\text{eff}}$ and $K_2^{\text{eff}}$ because the Fe-thickness range of the phase
coexistence is quite narrow. The observed value of $t_c$ is close to the value of 8 Å that has recently been found for MgO/Cr/Fe/MgO.\textsuperscript{22} The second-order anisotropy constant $K_2^{\text{eff}}$, which is responsible for the coexistence state during the SRT, has the opposite sign to $K_v^{\text{eff}}$, and its magnitude is approximately ten times smaller than the magnitude of $K_v^{\text{eff}}$.

The shape anisotropy contribution $K_{sh}$ can be eliminated from the effective anisotropy $K_1^{\text{eff}}$, and Eq. (2) can be rewritten as

$$K_1^{\text{eff}} - K_{sh} = K_{v1}' + 2K_{s1}/t,$$

where $K_{v1}'$ includes all volume anisotropy contributions except the shape anisotropy ($K_{v1}' = K_{v1} - K_{sh}$). Both $K_{v1}'$ and $K_{s1}$ can be obtained from the plot of $(K_1^{\text{eff}} - K_{sh})$ vs. $t$ as a function of $t$, where the slope indicates $K_{v1}'$, and the Y-intercept indicates the surface anisotropy $K_{s1}$. In the present study, the quantitative analysis of the contributions from $K_{v1}'$ and $K_{s1}$ is ambiguous because of the large uncertainty of the shape anisotropy term. Assuming, based on the VSM measurements of $M_s$, that the shape anisotropy is dependent on the Fe thickness, one obtains the plot $(K_1^{\text{eff}} - K_{sh})$ vs. $t$ versus $t$. This plot is represented by the blue circles in Fig. 5, and it yields $K_{v1}' = +1.92 \times 10^7$ erg/cm$^3$ and $2K_{s1} = -0.5$ erg/cm$^2$. However, if a thickness-independent $M_s$ is considered, as suggested by the hyperfine magnetic field data,\textsuperscript{13} the resulting anisotropy constants that are derived from the red triangles in Fig. 5 are $K_{v1}' = +9.7 \times 10^6$ erg/cm$^3$ and $2K_{s1} = +0.76$ erg/cm$^2$. Obviously, the magnitude of the shape anisotropy contribution and its thickness dependence are crucial to the quantitative analysis of the surface and volume anisotropy constants. However, in both cases that were considered, the slope of $(K_1^{\text{eff}} - K_{sh})$ vs. $t$ is positive, which proves that $K_{v1}'$ contains a large positive term on the order of $10^7$ erg/cm$^3$, which counterbalances a small negative magnetocrystalline contribution (equal to $-4.8 \times 10^5$ erg/cm$^3$ in bulk Fe) and effectively favors the out-of-plane magnetization direction. Such a positive contribution can be attributed to magnetoelastic anisotropy, which has been postulated for Fe/MgO(001) in previous experimental studies.\textsuperscript{12} Although we cannot reach a definite conclusion regarding $K_{s1}$ in this study, according to our previous results\textsuperscript{13} and theoretical predictions,\textsuperscript{23} we expect a positive value of the surface anisotropy.

As shown above, the polar SRT in MgO/Fe/MgO is induced by competing anisotropies that prefer different magnetization axes. In such a situation, if the involved anisotropies follow different temperature behaviors, SRT can also occur as a function of temperature.\textsuperscript{24} Figure 6 shows the AHE hysteresis loops that were measured as a function of temperature for selected Fe thicknesses. For $t = 8.2$ Å, i.e., below the critical thickness at RT, as the temperature decreased, the loops became rectangular with full remanence, and the coercive field monotonically increased to 200 Oe at 15 K (Fig. 6(a)). This behavior signifies an enhancement of the perpendicular magnetic anisotropy, which was directly observed for the Fe layers that were above the critical SRT thickness at RT (Figs. 6(b) and 6(c)). For $t = 10$ Å, the loop changed in character from hard to easy at 100 K, and at 15 K, the remanence reached almost 90%, which highlights the temperature-induced reorientation of the easy magnetization axis from an in-plane direction to an out-of-plane direction. For $t = 12.2$ Å, the magnetization remained in the plane, but the saturation field decreased considerably with decreasing temperature. Assuming that the lattice structure remains unchanged during cooling (heating), the temperature-driven SRT should be attributed to the corresponding temperature-induced changes in the material constants: the anisotropy constants and/or the magnetization. Consequently, there is a certain ambiguity in the simulations of the measured hard loops because they could be
reproduced using different sets of $M_s$ and $K_{\text{eff}}$. Assuming the $M_s(T)$ dependence that was obtained from the VSM measurements (we observed a 6% linear increase in $M_s$ for $t = 13$ Å and a 20% increase for $t = 9$ Å by decreasing the temperature from 300 K to 10 K), we simulated hysteresis loops for three Fe thicknesses: 10 Å, 12 Å, and 13.4 Å. The extracted $K_1^{\text{eff}}$ values are shown in Fig. 7. The gradual decrease in the $K_1^{\text{eff}}$ magnitude with decreasing temperature implies the increase of a positive contribution to $K_1^{\text{eff}}$. Although we cannot explicitly extract $K_{2s}(T)$ and $K_{v1}(T)$, we believe that the volume (magnetoelastic) anisotropy contribution determines the $K_1^{\text{eff}}(T)$ dependence. For the surface anisotropy, because magnetic properties depend on temperature more strongly on the surface than in the bulk, one would expect the temperature effects to be enhanced for thinner films; however, Fig. 7 suggests the opposite behavior. Simultaneously, the strong $K_1^{\text{eff}}(T)$ dependence can be explained in terms of magnetoelastic effects by considering that the epitaxial strains that were established during the low-temperature deposition were at least partially released at elevated temperatures because of the different thermal expansion coefficients of Fe and MgO. Remarkably, for all considered Fe thicknesses, $K_2^{\text{eff}}$ was temperature-independent, which is consistent with general observations concerning higher-order anisotropy constants. In the anisotropy space, the temperature-driven SRT occurs via the coexistence of out-of-plane and in-plane phases, similar to the thickness-driven transition.

IV. CONCLUSIONS

In summary, we showed that by applying low-temperature deposition, we suppressed the superparamagnetism in MgO/Fe/MgO. Under these circumstances, we confirmed the existence of perpendicular magnetic anisotropy at room temperature up to a critical thickness of 9 Å, for which we observed the spin-reorientation transition from an out-of-plane direction to an in-plane direction. We attributed the perpendicular magnetization to the magnetoelastic anisotropy that dominates at small thicknesses and possibly also to a smaller surface anisotropy term. The dominating origin of the PMA is different from that of surface anisotropy in systems in which the epitaxial strain is substantially reduced by a vanadium or chromium buffer layer between the Fe and the MgO substrate. Our analysis of the SRT in anisotropy space demonstrated that the transition occurred in a narrow thickness range via the state of coexisting phases. Finally, the temperature-driven SRT was discussed for Fe thicknesses close to the SRT critical thickness.

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