Oscillatory Magnetic Anisotropy Originating from Quantum Well States in Fe Films

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The magnetic anisotropy of Fe film grown on vicinal Ag(1,1,10) surfaces was studied with the *in situ* magneto-optic Kerr effect. Below 200 K, strong oscillations of the uniaxial magnetic anisotropy as a function of Fe thickness with a period of 5.7 monolayers are found, which can even cause the easy magnetization axis to oscillate between perpendicular and parallel to the steps. Such novel oscillation of the anisotropy is attributed to the quantum well states of *d*-band electrons at the Fermi level in the Fe film. This is unlike the previously observed oscillatory behaviors of ferromagnetic films caused by the quantum well states in nonmagnetic interfacing layers.

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Modern magnetoeletcronic devices are based on metallic films with thicknesses on the nanometer scale. The electrons in such thin films could be confined perpendicular to the films to form quantum well states (QWS) [1–3]. The formation of QWS could result in oscillatory physical properties as a function of film thickness such as the oscillation of the interlayer exchange coupling [4–6], magneto-optic effect [7], tunneling magnetic resistance [8], and magnetic anisotropy [9,10]. Exploring such a quantum size effect opens an opportunity to manipulate various magnetic properties in magnetic nanostructures.

Magnetic anisotropy is one of the most important properties of magnetic materials. It can determine the orientation of the easy magnetization axis, and thus is crucial for magnetoelectronic technology and its applications. The magnetic anisotropy is caused by the spin-orbit coupling of the electrons and therefore is affected by an altered electronic band structure. In Cu/Co/Cu(001), the magnetic anisotropy was found to be modulated by the QWS in nonmagnetic (NM) Cu film [9,10]. While the QWS in the NM overlayer can modulate the electronic structure of ferromagnetic (FM) film only through the interfacial hybridization, the QWS inside the FM film itself can directly alternate its electronic structure and modulate the magnetic anisotropy much stronger. Theory predicts such effect in Fe/Au(001) superlattices [11] and in Co/Cu(001) system [12,13].

There are few experimental studies on QWS in FM films. For example, QWS were observed in Co and Fe films by inversed photoemission and spin-polarized electron reflection [14–17], however in unoccupied electron band only. The intrinsic physical properties of FM are related more to the electrons in the occupied band, but there is only one report on QWS below E_F observed in Fe films by angular resolved photoemission spectroscopy—ARPES) [18]. Up to now no experimental observation of anisotropy oscillations induced by QWS in FM film has been made. Thus, further experimental effort should be made to answer whether (and how) do the QWS in FM films affect the magnetic anisotropy.

In this Letter, we report on the oscillation of step-induced in-plane uniaxial anisotropy with the Fe thickness, d_{Fe}, for Fe films grown on a Ag(1,1,10) vicinal surface, with an oscillation period of 5.7 ± 0.2 monolayers (ML). Such an effect is attributed to the QWS of Fe *d*-band with minority spins, and it occurs only at low temperatures (LT), i.e., below 200 K. If measured at room temperature (RT), $1/d_{\mathrm{Fe}}$ dependence of in-plane uniaxial anisotropy is obtained.

The experiments were performed in a multichamber ultrahigh vacuum system with the pressure below 2×10^{-10} mbar during Fe deposition. A Ag(1,1,10) substrate was used with 8° off the (001) surface and the step edges along the $[\bar{1}10]$ direction. The Ag(1,1,10) substrate was prepared with cycles of 1 keV Ar ion sputtering and subsequent annealing at ~600 °C. The sharp double-splitting diffraction spots were formed in low-energy electron diffraction. Using scanning tunnelling microscopy, monoatomic steps along the $[\bar{1}10]$ direction were observed that were nearly equidistant and regular. The average terrace width is found to be ~2 nm. The Fe films were grown at RT by molecular beam epitaxy as a wedged shape with a slope of ~4 ML/mm along the $[\bar{1}10]$ direction of the Ag substrate, i.e., along the [100] direction of the Fe film. After growth, the films were annealed at 150 °C for 30 min in order to improve the surface morphology [19,20]. Magnetic properties were probed by the *in situ* longitudinal magneto-optical Kerr effect (MOKE) with a laser diode (wavelength 670 nm, beam diameter <0.2 mm).

For Fe films grown on Ag(1,1,10) vicinal surface, the competition between the surface anisotropy and the shape

anisotropy results in a spin reorientation transition (SRT) at a film thickness of ~7 ML Fe [21]. Above the SRT thickness, the Fe film shows in-plane uniaxial anisotropy with the easy axis parallel to the step direction. The longitudinal hysteresis loops show rectangular shape when the external field is parallel to the step direction; the double split loops can be observed when the magnetic field is perpendicular to the step direction, as shown in the inset of Fig. 1(a). The split loops are characterized by a shift field (H_s) which is proportional to the uniaxial anisotropy [9,10,21,22], so the in-plane uniaxial magnetic anisotropy can be obtained quantitatively from this single-loop measurement across the Fe wedge.

Figure 1(a) shows that the shift field H_s increases with the Fe thickness and saturates above 60 ML of Fe when measured at RT. The saturation value of the uniaxial anisotropy refers to the volume contribution, H_s^{volume}, due to the strain induced by the atomic steps. The strain originates from the large step-height difference between film and substrate due to the bcc or fcc character of the system [22]. The thickness-dependence of H_s can be well fitted by a $1/d_{\text{Fe}}$ dependence formula $H_s(d_{\text{Fe}}) = H_s^{\text{volume}} + H_s^{\text{surf}}/d_{\text{Fe}}$, with the parameter $H_s^{\text{volume}} = 126$ Oe and $H_s^{\text{surf}} = -1090$ Oe · ML. Although the measured magnetic easy axis of the Fe film is along the step direction, the negative sign of H_s^{surf} indicates that the total contribution from the Fe/Ag interface and vacuum-Fe surface prefers to align the magnetic moment perpendicular to the step direction.

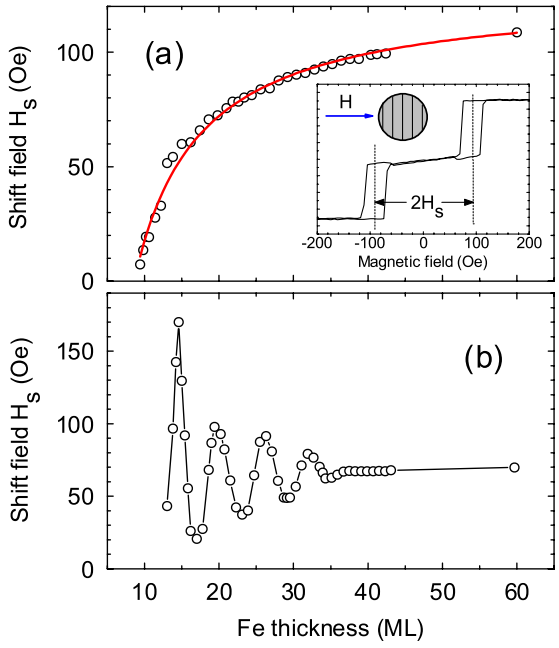

FIG. 1 (color online). Shift field as a function of Fe thickness measured for Fe/Ag(1, 1, 10) at (a) RT and (b) 5 K. Red line in (a) is a result of fitting as described in the text. Inset shows geometry of the MOKE experiment and a representative hysteresis loop.

At $T = 5$ K we found that the SRT thickness shifts to ~10 ML. The thickness dependence of the shift field H_s does not show any resemblance to the $1/d_{\text{Fe}}$ relation observed at RT, but exhibits a large amplitude oscillation with the Fe thickness with a period of 5.7 ± 0.2 ML. The oscillation amplitude around 15 ML of Fe is about 150 Oe, 1 order of magnitude larger than the anisotropy oscillation in Cu/Co/Cu(001) system [9,10]. Above 35 ML no oscillation behavior can be observed. The thickness-dependent oscillation of a physical property usually originates from either surface morphology [23] or the QWS in the films [4–10]. However, the oscillation of the surface morphology should have exactly 1 ML period, which is different from our experimental results. Therefore we attribute the observed thickness-dependent oscillation of the uniaxial anisotropy to the QWS in the Fe thin film. The QWS are expected to relate to the electron wave confined perpendicularly to the film surface. A possible effect on the magnetic anisotropy of the lateral electron confinement perpendicular to the step edges [24] is assumed to be negligible since it would originate at the film surface. Such QWS can modulate periodically the density of states at the Fermi level and thus modulate the magnetic anisotropy.

Since the thickness dependence of magnetic anisotropy in an Fe/Ag(1, 1, 10) system is very different at RT and LT, we systematically studied how changes in temperature influence the QWS effect on the magnetic anisotropy. To avoid the influence of possible contamination during the measurement with long periods of time, a new capped sample Au(4 ML)/Fe/Ag(1, 1, 10) was prepared showing only in-plane magnetization for $d_{\text{Fe}} > 5$ ML. Figure 2 shows the representative loops for different Fe thicknesses measured at 5 K in the magnetic field, H, applied either perpendicular to the steps or parallel to the step edges. It is seen that the easy axis direction of the uniaxial magnetic anisotropy oscillates with the Fe thickness. When $d_{\text{Fe}} =$ 11.5, 17.5, and 23.5 ML, the loops with H perpendicular to the steps show square shape, and if we measure the loops with H parallel to the steps, the double-splitting loops can be obtained. When $d_{\text{Fe}} = 15$, 20.5 and 27.5 ML, the loops show double-splitting loops for H perpendicular to the steps, and the square loop can be obtained for H parallel to the steps. A small portion of out-of-plane magnetization is observed for the hard-axis loops in Fig. 2(b). This is due to the competition between the shape anisotropy and the magneto-crystalline anisotropy for the magnetization perpendicular to the steps [25,26].

The thickness-dependent H_s at different temperatures is plotted in Fig. 3(a). The shift field H_s is defined as negative when the double-splitting loops occurs for H parallel to the steps (Fig. 2). The QWS effect on the Fe magnetic anisotropy remains after Au capping. It is clear from Fig. 3(a) that not only the quantitative value, but also the sign of the shift field H_s oscillates with the Fe thickness for $T = 5$ K. The period of H_s oscillations is the same as for the uncovered sample, i.e. ~5.7 ML. When the temperature is increased, the amplitude of the oscillation decreases and

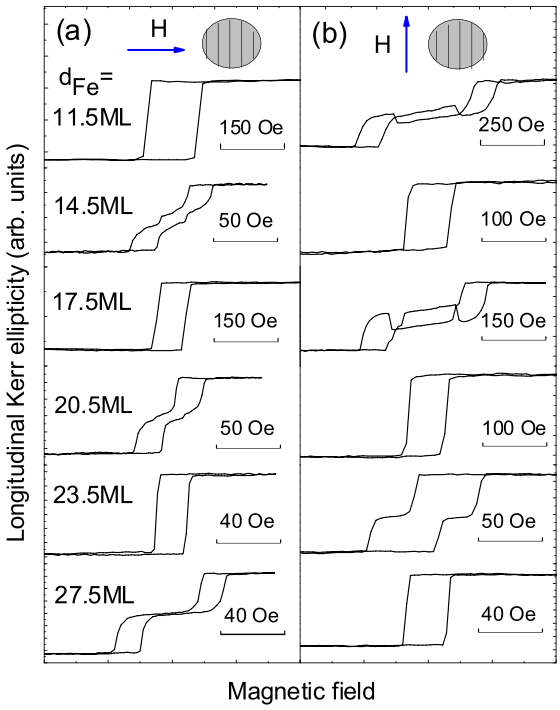

FIG. 2 (color online). Representative hysteresis loops for different Fe thicknesses measured at $T = 5$ K in a magnetic field applied (a) perpendicular and (b) parallel to the steps. The results indicate easy magnetization axis of the film oscillating with Fe thickness.

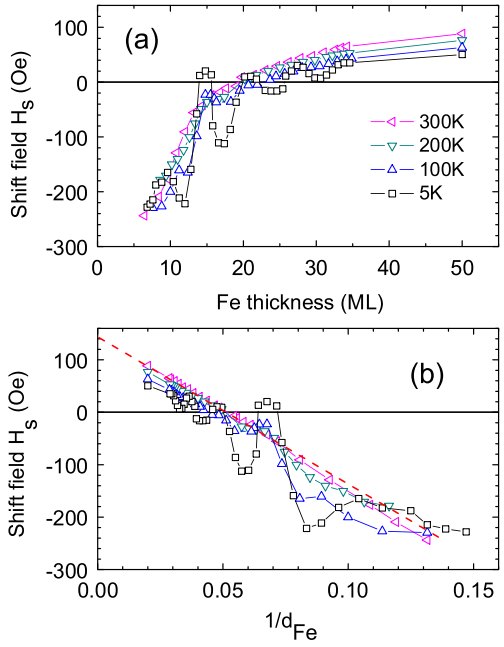

FIG. 3 (color online). Shift field as a function of (a) Fe film thickness and (b) of $1/d_{Fe}$ measured at different temperatures for Au/Fe/Ag(1, 1, 10). Straight dashed line in (b) is a guide to the eyes.

only a monotonic change of the magnetic anisotropy can be observed for $T > 200$ K .

At RT, we found that the easy axis runs along the step direction for thick film, similar to the uncovered sample. However, for $d_{Fe} < 19$ ML there is an in-plane spin reorientation transition and the easy axis switches to a direction perpendicular to the steps. To directly compare the experimental data with the $1/d_{Fe}$ dependence, the shift fields as a function of $1/d_{Fe}$ were also plotted in Fig. 3(b), which shows a good linear relation at RT for Fe films thicker than 10 ML. The fitted H_s^{volume} contribution (~150 Oe) is close to that of the uncovered sample, whereas the H_s^{surf} contribution (-3004 Oe · ML) is strongly altered by the Au covering. Such different H_s^{surf} values for uncovered and Au-covered samples are responsible for the different background of the H_s variation with the Fe thickness at LT as shown in Fig. 1(b) and 3.

Based on the result shown in Fig. 3, the QWS effect appears only at LT and increases with decreasing temperature. To make the temperature-dependent effect more clear, the sample Au(4 ML)/Fe/Ag(1, 1, 10) with uniform Fe thicknesses was investigated (Fig. 4). For a thick film ($d_{Fe} = 40$ ML), the shift field gradually decreases with decreasing temperature. However, for $d_{Fe} = 27$ ML, one sudden increase of H_s can be observed at 60 K, and for $d_{Fe} = 15$ ML, the transition temperature is 110 K (Fig. 4). Such a transition should be related to the quantum size effect according to the results of Fig. 3. Obviously, the effect of QWS occurs at lower temperatures for thicker Fe film.

It should be pointed out that QWS in NM films can appear in many systems at RT, like in Cu/Co(001) and Ag/Fe(001) [1–3]. However, QWS affects the magnetic anisotropy in Fe films only at LT. One possible explanation could be the short electron mean free path (MFP) in Fe films. In order to form QWS, the MFP should be larger than the film thickness. At RT, the MFP for Fe spin-up and spin-down electrons at the Fermi surface are 1.5 and 2.1 nm respectively [27], and may be different for s and d electrons. Such small MFP can cancel the QWS at RT for thick

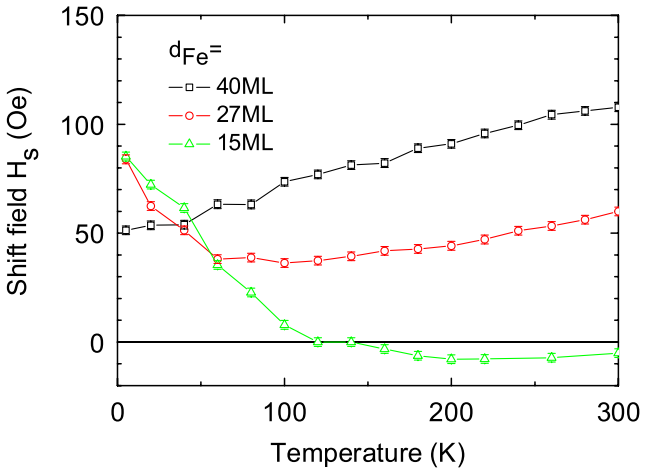

FIG. 4 (color online). Shift field H_s as a function of temperature for Au/Fe/Ag(1, 1, 10) with different Fe film thickness.

films. Usually the MFP increases at LT by reducing the electron-phonon scattering. Thus, the QWS can form at LT in thinner Fe films at the point the MFP could become larger than the film thickness. The effect of QWS on magnetic anisotropy should occur at higher temperatures for thinner films, which is consistent with the results of Fig. 4. Indeed, the first ARPES evidence of the QWS in Fe(110) films was also found at $T = 25$ K [18].

The oscillatory character of another important property, i.e., of the spin-dependent transport in Fe/MgO/Fe tunneling junctions was also predicted by theory [28] and confirmed experimentally [29]. The oscillation is attributed to the QWS of minority electrons with a Δ_1 symmetry (s character) of Fe(001). However the oscillation periodicity is $\sim$2 ML, which is different from our result. Since the magnetic anisotropy originates from the spin-orbit coupling, the more localized d-band electrons should contribute more to the magnetic anisotropy than the isotropic s character electrons. Therefore, it is very reasonable that the oscillation of the magnetic anisotropy is different to that of the spin-dependent tunneling of s electrons with Δ_1 symmetry in an Fe/MgO/Fe system.

The perpendicular magnetic anisotropy in Fe/Au multilayers is predicted theoretically to be modulated by the QWS in a d band of minority electrons with a Δ_5 symmetry [11]. However, the predicted oscillation period is $\sim$10 ML. The oscillation of the step-induced in-plane magnetic anisotropy may originate from the QWS in different d-electron band. The oscillation period of QWS at E_F is determined by the Fermi wave vector, k_F, as $\Lambda = \pi/k_F$ [1–3]. We consider the theoretical bcc Fe band structure along the ΓH direction, i.e., along the direction the electrons are confined in our Fe films [29]. In this case, the value of k_F of the Fe minority-spin d-band with $\Delta_{2'}$ symmetry is estimated to be $0.2k_{\mathrm{BZ}}$, where k_{BZ} is the Brillouin zone wave vector. Thus, QWS at E_F in this band should have an oscillation period of 5 ML, which is close to our experimental value. All other electron bands result in very different oscillation periods, so our result indicates that the magnetic anisotropy in Fe film should have a stronger relation to the minority-spin d-band with a $\Delta_{2'}$ symmetry. Although no experimental results on the QWS in this Fe d-band has been reported, it is very likely that the d electrons in Fe film can be confined to form the QWS, since there is no d electron states around E_F for Ag(001) substrate [1].

The absence of the QWS effect above 200 K is unlikely attributed to thermal fluctuation. From the theoretical bcc Fe band structure [29], we estimate that for 15 ML thick Fe film the energy separation of QWS around E_F in a d-electron band is $\sim$160 meV. This is much higher than the thermal energy at RT, but no QWS effect is observed at RT for Fe films of this thickness.

In summary, we studied the step-induced in-plane magnetic anisotropy of Fe film grown on a Ag(1,1,10) surface. A strong quantum oscillation of the uniaxial anisotropy is observed at LT with a period of 5.7 ± 0.2 ML. For the Au-covered Fe films, the easy magnetization axis also oscillates between perpendicular and parallel to the steps with the same period. Such a quantum oscillation is attributed to the QWS in a minority-spin d band at E_F. The lack of oscillation at RT may result from the thermal reduction of the MFP. At RT, the $1/d_{\mathrm{Fe}}$ behavior of the in-plane uniaxial anisotropy was observed.

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