Quantum Well States and Oscillatory Magnetic Anisotropy in Ultrathin Fe Films

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The magnetic anisotropy of Fe film grown on vicinal Ag(1,1,10) surfaces studied with the *in situ* magneto-optic Kerr effect shows oscillatory behavior. We found that both the in-plane step-induced uniaxial anisotropy and the perpendicular anisotropy measured at low temperature show strong oscillations as a function of Fe thickness with a period of $\sim$5.7 monolayers. Such novel oscillation of the anisotropy is attributed to the quantum well states (QWS) of d-band electrons at the Fermi level in the Fe film. These QWS have been observed by angle-resolved photoemission spectroscopy from a Fe wedge films grown on Ag(001) surface.

Index Terms—**Magnetic anisotropy, quantum well states, ultrathin magnetic film.**

I. Introduction

INVESTIGATIONS on layered magnetic nanostructures have attracted great interest in the past decades after the discoveries of interlayer coupling [1] and giant magnetoresistance [2]. The essential physics are expected in these layered magnetic nanostructures due to the formation of spin-polarized quantum well states (QWS) by electron confinement perpendicular to the films [3]–[5]. The formation of QWS can modify the density of states (DOS) around the Fermi level, resulting in many oscillatory physical properties as a function of film thickness, such as the oscillation of the interlayer exchange coupling [6]–[8], giant magnetic resistance [9], tunneling magnetic resistance [10], magneto-optic effect [11], and magnetic anisotropy [12], [13]. Exploring such a quantum size effect opens up an opportunity to manipulate various magnetic properties in magnetic nanostructures.

Magnetic anisotropy is one of the key properties for the applications of magnetic materials, in particular, for their applications in magnetoelectronics. 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 [12], [13]. While the QWS in the NM overlayer can modulate the electronic structure of ferromagnetic (FM) films only through the interfacial hybridization, the QWS inside the FM film itself can directly alternate its electronic structure and modulate the magnetic anisotropy more strongly. Theory predicts such an effect in Fe/Au(001) superlattices [14] and in Co/Cu(001) systems [15], [16].

In this paper, we review our systematic studies of the thickness dependent anisotropy for Fe films grown on a Ag(1,1,10) vicinal surface, in particular of the step-induced uniaxial anisotropy which oscillates with the Fe thickness with an oscillation period of $\sim$5.7 monolayers (ML) [17]. By studying the tilting angle off normal from the vicinal surface, we prove that the perpendicular anisotropy of Fe film also oscillates with the film thickness with the same period [18]. Moreover, we show that the amplitude of the anisotropy oscillation can be enlarged by increasing density of the substrate steps [19]. The oscillations of the magnetic anisotropy are attributed to the QWS of the Fe d-band with minority spins.

The QWS in thin films can be studied directly by measuring the electronic structure, but there are very few experimental studies on QWS in FM films. For example, QWS were observed in Co and Fe films by inverse photoemission, spin-polarized electron reflection [20]–[23] and two photon photoemission [24], [25] in an unoccupied electron band only. The spin dependent quantum well resonance in Fe film was predicted [26] and also observed [27] in fully epitaxial magnetic tunnel junctions with Fe/MgO/Fe/Cr(001) structure by differential conductance spectra measurements. 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 grown on W(110) substrate by angle-resolved photoemission spectroscopy (ARPES) [28]. Here, we also show our first observation of the QWS by the ARPES from a wedge Fe film grown on a Ag(001) surface.

II. Experimental Setup

The anisotropy studies were performed at the Max-Planck-Institut für Mikrostrukturphysik in Halle. The films were grown and analyzed in a multi-chamber ultrahigh vacuum system with a base pressure better than 5×10^{-11} mbar. The Ag(1,1,10) substrate with a vicinal angle of 8° was prepared with cycles of 1 keV Ar^+ ion sputtering and subsequent annealing at 600°C. The prepared surface showed nearly equidistant and regular monoatomic steps along the [110] direction measured by scanning tunneling microscopy, as shown in Fig. 1(a). The low-energy electron diffraction image [Fig. 1(b)] shows very sharp double splitting diffracting spots characteristic for a

Manuscript received January 08, 2011; accepted January 12, 2011. Date of current version May 25, 2011. Corresponding author: Y. Wu (e-mail: wuyizheng@fudan.edu.cn).

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Digital Object Identifier 10.1109/TMAG.2011.2108273

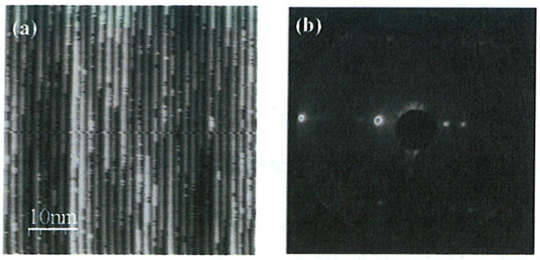

Fig. 1. (a) STM image and (b) LEED image of a clean Ag(1,1,10) surface. The electron beam energy in (b) is 43 eV.

vicinal surface with regular steps. The Fe films were epitaxially grown on a Ag(1,1,10) vicinal surface by molecular beam epitaxy at room temperature (RT) and then annealed at 150°C for 30 min in order to improve the surface morphology [29], [30]. The Fe films were grown as wedge samples with a slope of ~4 ML/mm and with a thick shoulder in order to determine the wedge position. Magnetic properties were probed by *in situ* longitudinal MOKE. Kerr ellipticity was measured with an s-polarized laser beam (wavelength 670 nm) of <0.2 mm diameter and an incident angle of 30° with respect to the sample normal.

The quantum well states in Fe films can be directly measured by angle-resolved photoemission spectroscopy (ARPES). ARPES data were measured at 22 K at beamline 7.0.1 of the Advanced Light Source (ALS) using a Scienta R4200 analyzer. The wedge Fe film was grown on a Ag(001) single crystalline substrate. Thickness dependent ARPES measurements were systematically performed on the wedged Fe film by taking advantage of the small photon beam size of ~ 50 μm diameter [31], [32]. The 110 eV photon energy was selected in order to optimize the QWS signal during the measurement.

III. Results

A. Oscillatory In-Plane Magnetic Anisotropy

It is well known that the atomic steps can induce an in-plane uniaxial anisotropy for Fe films grown on a Ag(001) vicinal surface [33], [34]. The magnetic hysteresis loops usually show rectangular shape when the external field is parallel to the step direction; double split loops can be observed if the magnetic field is perpendicular to the step direction, as shown in Fig. 2. The split loops can be characterized by a shift field (Hs) which is usually treated as proportional to the uniaxial anisotropy [33]–[35]. Thus, the in-plane uniaxial magnetic anisotropy can be obtained even quantitatively from this single-loop measurement and the thickness dependent magnetic anisotropy can be quantitatively studied across one wedge sample utilizing the small laser spot in the MOKE measurement.

Usually the step-induced magnetic anisotropy shows a monotonic change as a function of film thickness if measured at room temperature. However, Fig. 2 shows the magnetic hysteresis loops of Fe films with different thicknesses, which clearly prove the oscillation of the shift field Hs. These oscillations only appear at low temperature. Fig. 3 shows the thickness-dependent Hs measured at different temperatures. The oscillation with

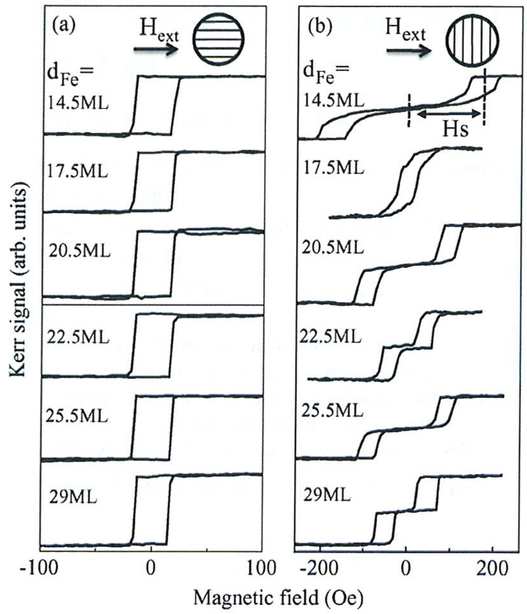

Fig. 2. Representative hysteresis loops for different Fe thicknesses measured at $T = 5$ K with the magnetic field applied (a) parallel and (b) perpendicular to the steps. The results indicate the oscillation of the in-plane uniaxial anisotropy.

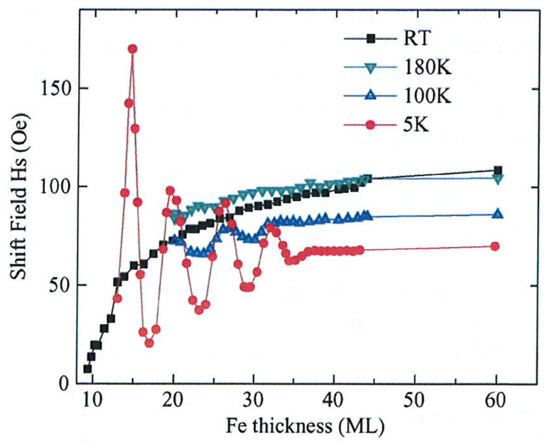

Fig. 3. Shift fields Hs as a function of Fe film thickness measured at different temperatures.

a period of ~5.7 ML only exists at a temperature lower than 180 K. At RT, only a monotonic change of Hs can be observed.

The oscillation of the step-induced in-plane magnetic anisotropy are expected to originate from the QWS at E_F in Fe film. The oscillation period of QWS at E_F is determined by the Fermi wave vector, k_F, as $\Lambda = \pi/k_F$ [3]–[5]. We consider the theoretical bcc Fe band structure along the $\Gamma - H$ direction, i.e., along the direction that the electrons are confined in the Fe films [27]. In this case, the k_F of the Fe minority-spin d-band with the symmetry Δ'_2 is estimated to be $0.2k_{BZ}$, 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 strong relation to the minority-spin d-band with Δ'_2 symmetry. Such d electrons with Δ_2' symmetry in Fe film can be confined to form the QWS, since there are no d electron states around E_F in the fcc Ag(001) substrate.

It should be pointed out that QWS in NM films can appear in many systems at RT, such as in Cu/Co(001) and Ag/Fe(001) [3]–[5]. However, Fig. 3 shows that the QWS affects the magnetic anisotropy in Fe films only at low temperature. So the low temperature measurement is crucial to observe the effect of QWS in Fe film, which may be the reason why such oscillatory behavior has not been observed before. The absence of the QWS effect above 180 K is unlikely to be attributed to thermal fluctuation. From the theoretical bcc Fe band structure [27], we estimate that for 15 ML thick Fe film, the energy separation of QWS around E_F in the Δ_2' electron band is ~160 meV. This energy is much higher than the thermal energy at RT, but no QWS effect was observed for 15 ML Fe films at RT.

The short electron mean free path (MFP) in Fe films could be a possible reason to explain the absence of the QWS effect at high temperature. In order to form QWS, the MFP should be larger than the film thickness. The MFP for Fe at the Fermi surface is only ~2 nm at RT [36], so it is hard to form QWS at RT for an Fe film with the thickness larger than 10 ML. However, the MFP can be increased by reducing the electron-phonon scattering at low temperature, then the QWS can appear under the condition where the MFP is large enough compared to the film thickness. For 40 ML Fe film, the MFP is still not large enough, even at the lowest temperature, so no QWS can be observed in 40 ML Fe film, as shown in Fig. 3.

B. Oscillatory Perpendicular Magnetic Anisotropy

The effect of QWS on the perpendicular anisotropy (perpendicular to the surface) in a FM film has been theoretically predicted [14]–[16]. In this section, we show that the perpendicular magnetic anisotropy also oscillates with the thickness for Fe film grow on a Ag(1,1,10) vicinal surface [18].

In order to detect the perpendicular anisotropy, usually the large shape anisotropy should be overcome. Such a large magnetic field is usually not easy to make compatible with the UHV system. However, for magnetic film grown on a vicinal surface, the variation of the perpendicular anisotropy can be estimated. In general, magnetic anisotropy consists of the crystalline anisotropy and the shape anisotropy. Usually the shape anisotropy forces the magnetization to lie in the surface plane, and the easy axis of the crystalline anisotropy is related to the principal crystallographic axis which is not in the surface plane for the vicinal surface. So if the magnetic field is perpendicular to the step direction, the magnetization may tilt away from the surface with a small angle δ resulting from the competition between the crystalline anisotropy and the shape anisotropy [37], [38]. Such a small tilting angle δ can be detected by the MOKE measurement due to very strong polar magneto-optical Kerr effect.

Fig. 4(a)–(c) shows the three different experimental geometries of the MOKE measurement. The in-plane magnetic field H is perpendicular to the steps in α^+ geometry [Fig. 1(a)] and α^- geometry [Fig. 4(c)]. The Ag[001] direction is 8° away from the normal direction of the surface toward the left side ($\alpha > 0$) in α^+ geometry [Fig. 1(a)], but the Ag[001] direction tilts toward

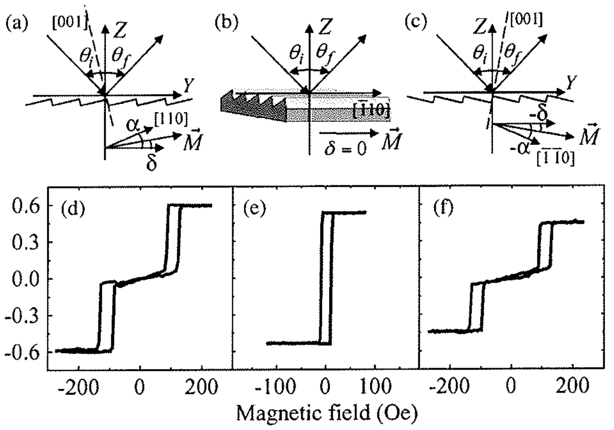

Fig. 4. Schematic of the MOKE experiment at three geometries: (a) α^+, (b) α'', and (c) α^-. (d)–(f) corresponding hysteresis loops of 45 ML of Fe on Ag(1,1,10) representative for the experimental geometries (a)–(c), respectively [18].

the right side in α^- geometry [Fig. 1(c)], i.e., with $\alpha < 0$. Obviously, in these two geometries, the magnetizations tilt away from the surface plane with opposite tilting angle δ, thus generating opposite polar Kerr signals. At α'' geometry with the magnetic field applied along the steps [Fig. 4(b)], the magnetization is both in the terrace plane and in the film plane; thus only the longitudinal Kerr signal can be measured.

Fig. 4(d)–(f) shows the representative hysteresis loops measured for 45 ML of Fe grown on a Ag(1,1,10) surface in three MOKE geometries, respectively. The hysteresis loops at α^+ geometry and α^- geometry show typical double splitting loops with very different saturation Kerr signals. Such difference of the Kerr signal is due to the opposite polar contributions in these two measurement geometries. Thus, the longitudinal signal ϕ_l and polar Kerr signal ϕ_p can be obtained from

$$\phi_l = (\phi_{\alpha+} + \phi_{\alpha-})/2 \tag{1}$$
$$\phi_p = (\phi_{\alpha+} - \phi_{\alpha-})/2. \tag{2}$$

Here, the $\phi_{\alpha+}$ and $\phi_{\alpha-}$ are the Kerr signals measured at α^+ and α^- geometries, respectively. Thus, the small tilting angle δ of the Fe magnetization can be estimated from the Kerr signals measured in three different geometries. According to (1) and (2), the tilting angle δ can be estimated by

$$\tan\delta = \frac{M_z}{M_y} = \frac{\phi_p}{\phi_l} \bullet \frac{\phi_l^{\text{sat.}}}{\phi_p^{\text{sat.}}} \tag{3}$$

where $\phi_l^{\text{sat.}}$ and $\phi_p^{\text{sat.}}$ are the saturation Kerr signals in longitudinal and polar geometries. $\phi_p^{\text{sat.}}$ is difficult to measure due to the limited applied magnetic field. However, the theory of the magneto-optical Kerr effect in ultrathin FM films has been well developed [39], so the tilting angle δ can be calculated by the ratio $\phi_l^{\text{sat.}}/\phi_p^{\text{sat.}}$ calculated theoretically. The tilting angle δ for 45 ML of Fe on Ag(1,1,10) was calculated as equal to 0.7° [18].

Fig. 5 shows the representative hysteresis loops for different Fe thicknesses measured at RT in α^+ and α^- geometries. Due to the strong perpendicular surface anisotropy, the Fe magnetization is oriented perpendicular to the film plane up to a thickness near 6 ML [29], [33]. For 5 ML Fe films [Fig. 5(a) and (b)], the

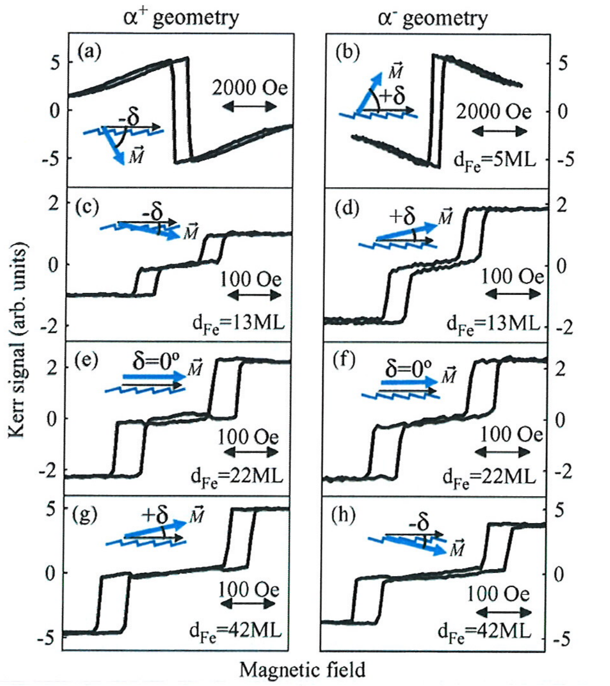

Fig. 5. Representative hysteresis loops for different Fe thicknesses measured at RT in α^+ geometry (left column) and α^- geometry (right column). The schematic drawing in each figure shows the orientation of the Fe magnetization.

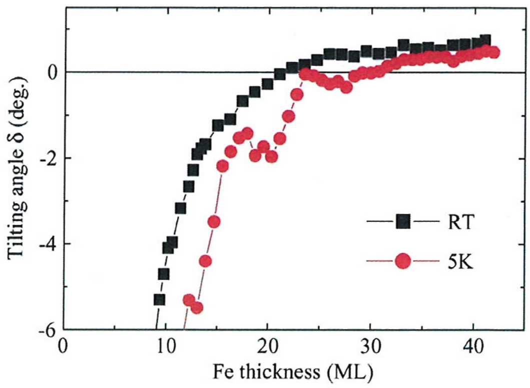

Fig. 6. The calculated tilting angles δ of the Fe magnetization as a function of Fe thickness at RT and 5 K for Fe films on Ag(1,1,10) [18].

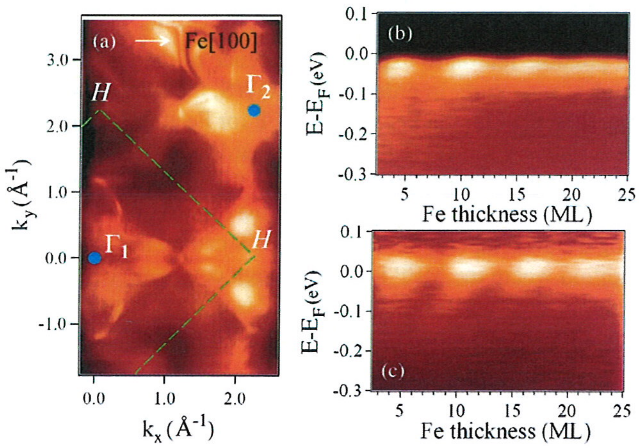

Fig. 7. (a) ARPES intensity at the Fermi level with $h\nu = 110$ eV. (b) Photoemission spectra and (c) the spectra as a function of Fe thickness at the Γ_2 point normalized with a Fermi function.

strong Kerr signal can be observed due to the large polar contribution, and the polar Kerr signals show the opposite hysteresis loops in α^+ and α^- geometries because of the opposite projection components of the in-plane fields in these two geometries. The Kerr signal will be reduced if the Fe magnetization is tilted towards the sample surface by the large in-plane magnetic field.

The competition between the perpendicular surface anisotropy and the volume anisotropy results in a spin reorientation transition (SRT), i.e., the magnetization continuously rotates toward the sample plane above the SRT thickness [40]. Fig. 5(c)–(h) show the loops of Fe films with different thicknesses of 13 ML, 22 ML, and 42 ML. The hysteresis loops with the same Fe thickness have the same shift fields in both α^+ and α^- geometries. However, for 13 ML Fe film, the Kerr signal in α^+ geometry is smaller than in α^- geometry, but for 42 ML Fe film, the Kerr signal in α^- geometry is relatively larger. Our results indicate that the 13 ML Fe has a negative tilting angle δ in α^+ geometry, and the 42 ML Fe has a positive δ in α^+ geometry. The tilting angle crosses zero at $d_{Fe} \sim 22$ ML, so the Kerr signals in both α^+ and α^- geometries show the same value for 22 ML Fe [Fig. 5(e), (f)].

The tilting angle δ can be calculated as a function of the Fe thickness [18], as shown in Fig. 6. Clearly, the calculated value of δ at RT increases monotonically with the Fe thickness and crosses zero at $d_{Fe} \sim 22$ ML. However, if measured at 5 K, the tilting angle has a very clear oscillation added to an increasing background, and the oscillation period is also ~5.7 ML. From the MOKE measurements in α'' geometry, the Kerr signal changes linearly with the thickness, indicating that the magnetization and the optical constant do not oscillate with the Fe thickness, so the shape anisotropy will not oscillate with Fe thickness [18]. Since the tilting angle of the magnetization originates from the competition between the magnetocrystalline anisotropy and the shape magnetic anisotropy, our results show that the perpendicular magnetocrystalline anisotropy oscillates with the Fe thickness, in particular, at low temperature. The same period of the in-plane and perpendicular anisotropy oscillations indicates the same origin, which is related to the QWS in a minority-spin d-band with Δ_2' symmetry at the Fermi surface, and not from the QWS in d-band with Δ_5 symmetry as predicted by the theoretical calculations for an Fe/Au multilayer [14].

C. *Quantum Well States Measured Directly by ARPES*

The most direct method to measure the Quantum well states is ARPES, and we performed the ARPES measurement on a wedged Fe film grown on Ag(001) in ALS. Due to the dipole transition selection rule [41], the photon excitation from Δ_2' band along $\Gamma - \mathrm{H}$ direction is forbidden, so we did the ARPES measurement on a second Γ point with a high electron emission angle. The 110 eV photon energy was selected to optimize the photoemission at the second Γ point [42]. Fig. 7(a) shows the Fermi surface of bcc Fe. We then measured the thickness dependent photoelectron spectra at different $K_{//}$ with different elec-

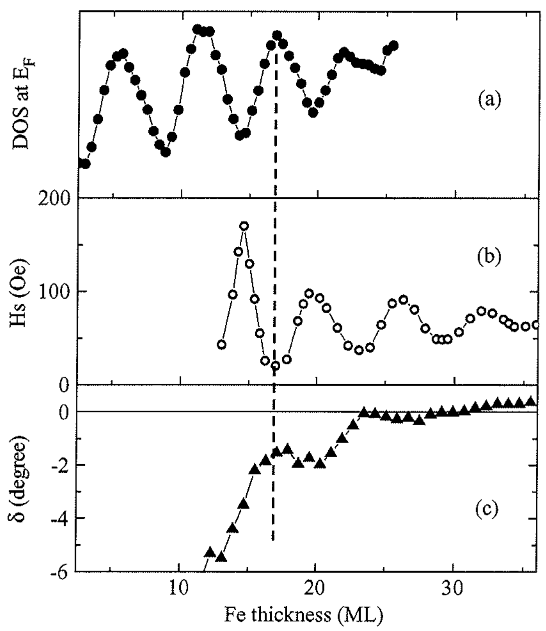

Fig. 8. The thickness dependencies of (a) density of states at E_F, (b) the shift field and (c) the tilting angle δ. The vertical dashed line is a guide of eyes.

tron emission angles, and tried to determine whether the DOS can oscillate with the film thickness. Indeed, we found a QWS at the second Γ point, not at the first Γ point. As shown in Fig. 7(b), the DOS at Γ_2 around E_F clearly oscillates with the Fe film thickness, and the oscillation period is about 5.6 ML, similar to the oscillation period of magnetic anisotropy. To highlight the QWS at the Fermi surface, we normalized the spectra of Fe thin films by a Fermi function and show the result in Fig. 7(c). It is interesting to note that the QWS only exist around E_F, and almost disappear for electron energy lower than −0.1 eV. The QWS in the Fe/W(110) system also shows similar behavior [28], and this behavior is very different with the QWS observed in the NM films, such as Cu/Co(001) and Ag/Fe(001) systems [3]–[5], in which the QWS can extend to several eV below the Fermi surface. This may be attributed to the strong correlation effect in 3d transition ferromagnetic metal [43], such that the electron lifetime away from E_F is strongly suppressed by the many-body effect [44], [45], resulting in a much small electron mean free path. So the QWS cannot form for those electrons at lower energy, if the electron MFP is smaller than the film thickness.

In Fig. 8, we plot the thickness dependent DOS at the Fermi surface, as well as the thickness dependence of the shift field Hs and the tilting angle δ. It is very clear that all the oscillations have the same period. DOS and Hs have the anti-phase oscillation. When the DOS reach maximum, the shift field has the lowest value. Here, it should be remembered that the positive shift field means the easy axis of the in-plane uniaxial anisotropy is along the step direction, so that we can then conclude that the QWS at the Fermi surface induce the in-plane anisotropy with easy axis perpendicular to the steps. On the other hand, when the QWS at E_F appears, the tilting angle reaches a maximum, indicating the QWS at E_F try to force the Fe magnetization to lie in the terrace plane.

IV. Discussion

In magnetic ultrathin films, the magnetic anisotropy consists of the volume anisotropy and the surface anisotropy (including the interface contribution), so that the thickness-dependence of Hs at RT usually can be well fitted by a $1/d_{FM}$ dependence formula. It would then be interesting to know whether the oscillation of Hs existing at low temperature originates from the volume anisotropy or from the surface anisotropy. Generally speaking, the QWS should extend into the whole Fe film, so the volume anisotropy should be influenced by QWS. In this case, the volume anisotropy should have little dependence on the capping layer, and indeed the Hs oscillation at low temperature should have a similar amplitude for both the Au-covered Fe film and the uncovered Fe film [17], [18]. Large amplitude quantum oscillations of magnetic anisotropy have been observed for Fe films grown on Ag(1,1,6) with high step densities [19], and this oscillation amplitude has a linear dependence on the volume anisotropy, indicating the volume character of the QWS effect on the magnetic anisotropy. However, the surface effect may not be completely ruled out in this measurement, since theoretically both volume contribution and surface contribution of the step induced anisotropy have a quadratic function of the step density [33], so both volume anisotropy and surface anisotropy can be modified by the step density on different vicinal surfaces. In a Cu/Co(001) system [12], [13], the magnetic anisotropy was found to be modulated by the QWS in nonmagnetic Cu films through the interfacial hybridization, thus this anisotropy oscillation should have interface character and will decay with the Co thickness.

The novel oscillation of magnetic anisotropy in Fe film is attributed to the QWS in a minority-spin d-band with the Δ_2' symmetry at E_F. A very similar value for the period of a strong oscillation of magnetic coupling as a function of the thickness of one of the Fe electrodes was obtained for the Fe/Cr/Fe(001) system [46]. This confirms that QWS formed in the same electron band are responsible for the oscillatory behavior observed for magnetic anisotropy and for interlayer coupling.

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 [26] and confirmed experimentally [27]. 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 ~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. On the other hand, there is a s electron band crossing the Fermi surface in fcc Ag(001), so the s electron in Fe film can penetrate into the Ag substrate, and cannot form the QWS in Fe film.

In fact, we also observed several other d-band QWS at different $K_{//}$ points with very different oscillation periods, especially the QWS in d-band with Δ_5 symmetry at E_F has been observed with a ~9.2 ML oscillation period [14]. Although such QWS in an Fe/Au(001) multilayer were predicted to influence the perpendicular anisotropy [14], our experimental observation did not support this prediction. Our results indicate that the electron state in the d-band with Δ_2' symmetry plays a more important role in the magnetic anisotropy in the Fe film than other electron bands. However, it still requires further theoretical studies on the relation between the magnetic anisotropy and the QWS in different d-bands, not only in Fe film, but also in other FM materials such as Co, Ni, etc. Nevertheless, our study shows that the QWS in a ferromagnetic ultrathin layer can be a useful method for engineering the magnetic anisotropy and other magnetic properties. Nowadays, the thickness of ferromagnetic layers in spintronics devices are usually at the nanometer scale, so our results indicate that the quantum effect may need to be considered in order to design the spintronics devices.

V. Conclusion

We studied the magnetic anisotropy of Fe films grown on a Ag(1,1,10) surface. We found that both the in-plane uniaxial anisotropy and the perpendicular anisotropy strongly oscillate with the Fe thickness with a period of ~5.7 ML, and this 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. We relate the observed oscillation of the magnetic anisotropy to the QWS in Fe film observed directly by ARPES measurements. We conclude that QWS in a minority-spin d-band with the Δ_2' symmetry at E_F induce an in-plane anisotropy with easy axis perpendicular to the steps. Our study provides a useful method for engineering the magnetic anisotropy, which is interesting for basic knowledge and possible applications in spintronics devices.

Acknowledgment

J. Li and Y. Wu thank Prof. J. Kirschner for offering the possibility to work in Max-Planck-Institut für Mikrostrukturphysik in Halle. Technical support from H. Menge (MPI Halle), G. Kroder (MPI Halle), Y.S. Kim (ALS Berkeley), and A. Bostwick (ALS Berkeley) is acknowledged. Y. Z. Wu also acknowledges the support of NSFC and MOST (Grants Nos. 2011CB921801, 2009CB931203 and 2010DFA52220) of China, by the Shanghai Education Development Foundation, by the Shanghai Science and Technology Committee, and by the Fok Ying Tong education foundation.

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