Experimental observation of quantum oscillations of perpendicular anisotropy in Fe films on Ag(1,1,10)

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An oscillatory behavior of the tilting angle of magnetization as a function of film thickness at low temperatures is reported for the Fe films grown on Ag(1,1,10). A competition between magnetocrystalline anisotropy and magnetic shape anisotropy tilts the magnetization from the surface plane toward the terrace plane when the magnetization is oriented perpendicular to the step edges. The tilting angle changes with the variation in the magnetocrystalline anisotropy. The oscillation period is exactly the same as for the oscillations of the in-plane anisotropy.

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I. INTRODUCTION

Magnetic anisotropy is one of the key properties for applications of magnetic materials, in particular, for their applications in magnetoelectronics. Great efforts have been undertaken in order to understand magnetic anisotropy and to learn how it can be manipulated. The magnetic anisotropy in thin films can be modulated by film thickness,[1,2] film temperature,[3] and the growth temperature.[4] The anisotropy can also be determined by a chosen combination of film/substrate, in particular, due to interfacial hybridization[5] and strains.[6] Moreover, magnetic anisotropy can be modified by an electric field,[7,8] which opens the possibility to reverse the magnetization with an electric field applied to magnetic thin films. For magnetic thin films with a thickness on the nanometer scale, quantum effects may play a particularly important role for their magnetic properties.[9,10] In a Cu/Co/Cu(001) system, the in-plane magnetic anisotropy was found to be modulated by quantum well states (QWSs) in the nonmagnetic Cu layer through interfacial hybridization.[11,12] QWS formed inside the ferromagnetic (FM) layer are supposed to modulate the magnetic anisotropy directly and thus is expected to magnify the effect of QWS on the magnetic anisotropy. Indeed, for Fe films grown on Ag(1,1,10) and Ag(1,1,6) vicinal surfaces,[13,14] the step-induced in-plane magnetic anisotropy of the Fe film was discovered to be strongly oscillating with Fe film thickness at an oscillation period of 5.7 ML. Such an effect is attributed to the QWS of an Fe *d* band with minority spins. The effect of QWS on the perpendicular anisotropy (perpendicular to the surface) in a FM film has been theoretically predicted[15–17] but so far there is still a lack of experimental evidence.

In this paper, we report on the magnetic anisotropy perpendicular to the surface which oscillates in Fe thin films grown on Ag(1,1,10). The competition between the magnetocrystalline anisotropy and the magnetic shape anisotropy tilts the magnetization from the surface plane toward the terrace plane when the magnetization is oriented perpendicular to the step edges.[18,19] This allows to follow changes in the perpendicular component of magnetization. Since the vicinal angle is small (8°), the tilting angle and more so the changes in the tilting angle are expected to be very small. Fortunately, the magneto-optical Kerr effect (MOKE) can detect such tiny tilting angle by measuring the polar Kerr effect which is roughly one order of magnitude stronger than the longitudinal Kerr effect.

II. EXPERIMENT

The experiments were performed in a multichamber ultrahigh vacuum system with a base pressure better than 5×10^{-11} mbar. The Ag(1,1,10) substrate with the vicinal angle of 8° was prepared with cycles of 1 keV Ar ion sputtering and subsequent annealing at 600 °C. Sharp double-splitting diffracting spots were formed in low-energy electron diffraction, and nearly equidistant and regular monoatomic steps along the [110] direction were observed by scanning tunneling microscopy. 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.[1,20] 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 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.

III. RESULTS

We performed MOKE measurements at three different experimental geometries as shown in Figs. 1(a)–1(c). In Figs. 1(a) and 1(c), the in-plane magnetic field H is perpendicular to the steps (α^+ and α^- geometries). However, in α^+ geometry [Fig. 1(a)] the Ag [001] direction is 8° away from the normal direction of the surface toward the left side ($\alpha > 0$), whereas the Ag [001] direction tilts toward the right side in

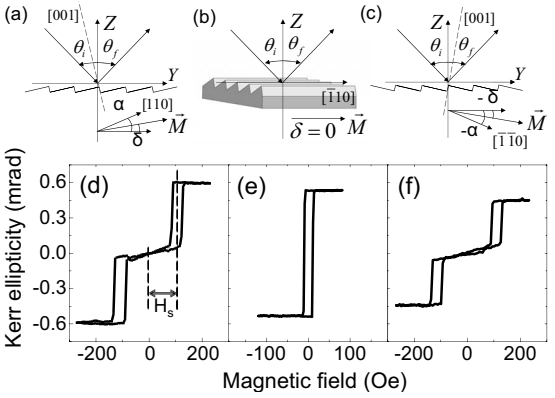

FIG. 1. Schematic of the MOKE experiment at three geometries: (a) α^+, (b) $\alpha^{\parallel}$, and (c) α^-. [(d)–(f)] corresponding hysteresis loops of 45 ML of Fe on Ag(1,1,10) representative for the experimental geometries (a)–(c), respectively.

α^- geometry, i.e., with $\alpha<0$ [Fig. 1(c)]. In $\alpha^{\parallel}$ geometry [Fig. 1(b)], the magnetic field H is applied along the steps in the sample plane.

Figures 1(d)–1(f) shows representative hysteresis loops measured for 45 ML of Fe grown on a Ag(1,1,10) surface at α^+, $\alpha^{\parallel}$, and α^- geometries, respectively. It is well known that in the Fe/Ag(1,1,10) system the atomic steps can modify the anisotropy, which manifests as an additional in-plane uniaxial anisotropy with the easy axis parallel to the steps above a certain thickness of Fe.[13,14,21] So the hysteresis loops show a rectangular shape when the magnetic field (H) is applied parallel to the steps [Fig. 1(e)] and double-split loops can be measured when a magnetic field is applied perpendicular to the steps [Figs. 1(d) and 1(f)]. The shift field H_s characterizing the loops is related to the uniaxial anisotropy induced by the steps.[22] However, the saturation Kerr signals measured at the α^+, $\alpha^{\parallel}$, and α^- geometries from the same sample are obviously different. This is due to the competition between the magnetocrystalline anisotropy (which favors the magnetization in the terrace plane) and the shape anisotropy (which favors the magnetization in the film plane). When the magnetization is perpendicular to the steps [as shown in Fig. 1(a), α^+ geometry and in Fig. 1(c), α^- geometry], the film plane is obviously different from the terrace plane. So the competition between both anisotropies tilts the magnetization from the film plane toward the terrace plane by a tilting angle δ. Therefore, the Kerr signals measured at α^+ and α^- geometries consist of both longitudinal and polar signals. At $\alpha^{\parallel}$ geometry with the magnetic field applied along the steps [Fig. 1(b)], the magnetization is both in the terrace plane and in the film plane; thus, there is no perpendicular component of the magnetization. The perpendicular component of the magnetization has different signs for α^+ geometry and α^- geometry, thus generating opposite polar Kerr signals. Since the magnitude of the polar Kerr signal is almost one order larger than that of the longitudinal signal, MOKE is more sensitive to the perpendicular component than to the in-plane component of magnetization. So the polar and longitudinal Kerr contributions may be similar, even if δ is small. Thus, the differences in Kerr signals at apparent magnetic saturation seen in Figs. 1(d)–1(f) can be understood as originating

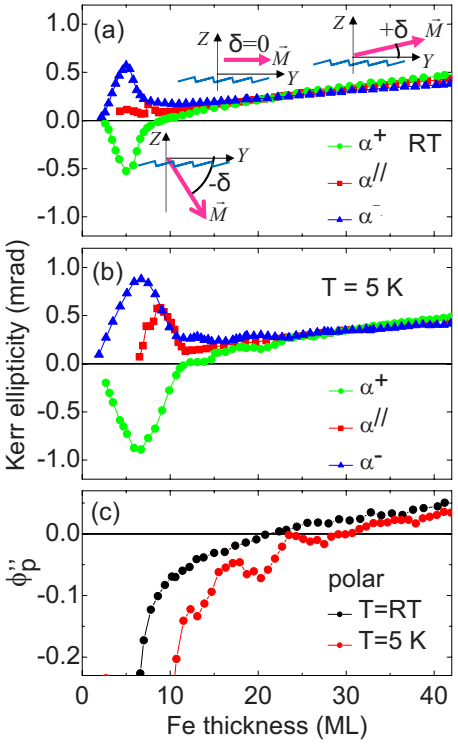

FIG. 2. (Color online) Kerr ellipticity vs thickness measured at α^+, $\alpha^{\parallel}$, and α^- geometries for Fe films on Ag(1,1,10) at: (a) RT and (b) 5 K. (c) Polar Kerr signal at RT and 5 K, all at $H=300$ Oe. Schematic in (a) show magnetization orientation at different thicknesses of Fe when measured in α^+ geometry.

from the different polar contributions to the total Kerr signal in different MOKE geometries. Similar behavior has been reported for Co films grown on vicinal surfaces of Cu(001).[18,19]

The analysis in α^+, $\alpha^{\parallel}$, and α^- geometries has been extended for Fe films thinner than 45 ML. The resulting thickness dependencies of the Kerr signals at $H=300$ Oe studied at RT and 5 K are shown in Fig. 2. A large signal at low thicknesses corresponds to the polar contribution which is due to a strong perpendicular surface anisotropy forcing the easy-magnetization axis to be oriented perpendicular to the film plane up to a thickness near 6 ML.[1] This critical thickness for Fe films grown on vicinal surfaces of Ag(001) is slightly larger than that of Fe films grown on a flat surface of Ag(001).[1,14,21] The competition between the perpendicular anisotropy and the shape anisotropy results in a spin-reorientation transition (SRT), i.e., above the critical thickness of the SRT the magnetization continuously rotates toward the sample plane.[23] The polar Kerr contribution decreases and crosses zero at $d_{\mathrm{Fe}}\sim 22$ ML, indicating sign reversal of δ at this thickness. In the α^- geometry, δ has the opposite sign than in the α^+ geometry. In the $\alpha^{\parallel}$ geometry, and for a magnetic field applied along the steps, the magnetization lies in the film plane and the longitudinal Kerr signal increases linearly with Fe thickness [except at thicknesses a little above the thickness of the SRT where the switching of the perpendicular magnetization component results in a polar contribution to the measured Kerr signal—Fig. 2(b)].

To explore whether the perpendicular anisotropy can be influenced by the QWS formed in Fe films, we performed a

MOKE measurement at 5 K, as shown in Fig. 2(b). In comparison to the experimental results at RT, the maximum Kerr signal at the SRT is shifted to thicker films, which simply indicates a stronger perpendicular anisotropy at low temperature. However, above the SRT thickness, the Kerr saturation signals measured in α^+ and α^- geometries are discovered to oscillate with the Fe thickness, whereas the saturation signal measured in the $\alpha^{\parallel}$ geometry does not oscillate. This indicates the oscillatory polar Kerr contribution.

Quantitatively, if the magnetization is probed in α^+ and α^- geometries, the polar Kerr effect is additive and subtractive, respectively (see Fig. 1).[12] 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 ϕ_{α^+}'' and ϕ_{α^-}'' are the Kerr ellipticities measured at α^+ and α^- geometries, respectively. In general, the longitudinal signal measured perpendicular to the steps, i.e., calculated as ϕ_l'' from Eq. (1) (which corresponds to the in-plane component of the Fe magnetization) can be different from the saturation longitudinal signal measured in the $\alpha^{\parallel}$ geometry (which corresponds to the total Fe magnetization). However, since the vicinal angle is small, the in-plane component averaged from the measurements in α^+ and α^- geometries should be very similar to the saturation magnetization measured in the $\alpha^{\parallel}$ geometry. This is the case as one can see in Figs. 2 and 3 and it can be used as evidence that the anisotropic in-plane Kerr effect is very small, if it exists. The polar signal ϕ_p'' is plotted vs. Fe film thickness in Fig. 2(c). The oscillatory polar Kerr contribution just indicates that the perpendicular component of the Fe magnetization oscillates with the Fe thickness.

To exclude any influence of a possible contamination of the Fe surface during the MOKE experiment, we also measured the sample covered with a 4 ML Au protection layer (as shown in Fig. 3). The Au capping layer reduces the perpendicular anisotropy, so the Fe magnetization is aligned in the film plane, and no strong polar Kerr component can be observed for low Fe coverage at RT anymore. However, the Kerr signals in the α^+ and α^- geometries are still different from that measured in the $\alpha^{\parallel}$ geometry. This means that the Fe magnetization in this case also tilts away from the film surface when the magnetic field is applied perpendicular to the steps in the film plane. If measured at $T=5$ K, a small polar Kerr component below a thickness of 10 ML can be observed so the perpendicular anisotropy becomes stronger at low temperature. Above the SRT thickness, the Kerr signals measured in α^+ and α^- geometries show oscillatory behavior. Meanwhile, the Kerr signal in $\alpha^{\parallel}$ geometry only increases linearly with the Fe thickness.

IV. DISCUSSION

The small tilting angle δ of the Fe magnetization can be estimated from the Kerr signals measured in three different geometries. According to Eqs. (1) and (2), the tilting angle δ can be estimated by

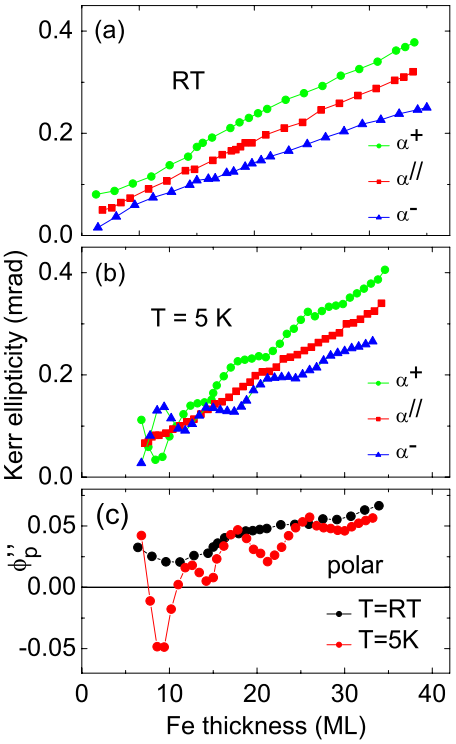

FIG. 3. (Color online) Kerr ellipticity measured at 300 Oe for three geometries at: (a) RT and (b) 5 K as a function of Fe film thickness for the sample covered with 4 ML of Au. (c) Polar Kerr signal at RT and 5 K.

$$\tan\delta = \frac{M_z}{M_y} = \frac{\phi_p''}{\phi_l''}\frac{\phi_l^{s''}}{\phi_p^{s''}}, \tag{3}$$

where $\phi_l^{s''}$ and $\phi_p^{s''}$ are the saturation Kerr signals in longitudinal and polar geometries, respectively. The saturation longitudinal Kerr signal $\phi_l^{s''}$ can be obtained from the Kerr signal measured in $\alpha^{\parallel}$ geometry, but usually we cannot measure the saturation polar Kerr signal due to the limitation of the magnetic field which can be applied. Fortunately, since the theory of magneto-optical Kerr effect in ultrathin FM films has been well developed,[24] δ can be calculated using the ratio between the longitudinal and polar saturation signals calculated theoretically. By utilizing the value of the indices of refraction $n_{\mathrm{Fe}}=2.87+3.36i$ and $n_{\mathrm{Ag}}=0.27+4.18i$ (Ref. 25) and also the Voigt constant of $Q_{\mathrm{Fe}}=0.376+0.0066i$,[26] the ratio was calculated to be 0.11 (it changes by no more than 0.1% within the investigated thickness range). Then δ for 45 ML of Fe on Ag(1,1,10) can be calculated as equal to 0.7°. Such a small tilting angle is difficult to be detected by any other experimental method.

The calculated δ as a function of Fe thickness is shown in Fig. 4. At RT, it is clear that the tilting angle increases with the Fe thickness and reverses its sign at $d_{\mathrm{Fe}}\sim 22$ ML. At $T=5$ K the tilting angle shows an oscillatory behavior similar to that of the polar contribution shown in Fig. 2(b). The oscillation amplitude is not larger than 3°.

As it is shown in Fig. 4, the tilting angle as a function of Fe thickness can also be obtained from Eq. (3) for the Au-

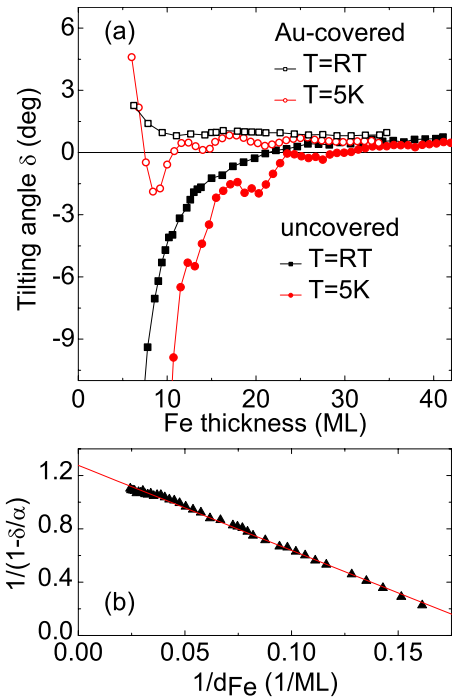

FIG. 4. (Color online) (a) Calculated tilting angle as a function of Fe thickness at RT for uncovered and Au-covered samples (square and open square, respectively) and at 5 K for uncovered and Au-covered samples (circle and open circle, respectively). (b) $1/(1-\delta/\alpha)$ as a function of $1/d_{\mathrm{Fe}}$ at RT, solid line is a result of linear fitting.

covered sample. At RT, the tilting angle decreases with the Fe thickness, indicating that the Au/Fe/Ag(1,1,10) system has a totally positive perpendicular anisotropy which favors the in-plane magnetization alignment. It is worth mentioning that for thick Fe films δ approaches the value of $\sim 0.7°$ independently whether the Fe film is covered with Au or not. At $T=5$ K, the tilting angle clearly oscillates with the Fe thickness. For $d_{\mathrm{Fe}} \sim 9$ ML, the tilting angle can even oscillate with a change of the sign.

Now we would like to discuss the tilting angle δ, and, in particular, its dependence on Fe thickness, which provides more information on the perpendicular magnetic anisotropy. To describe how the perpendicular anisotropy develops with Fe thickness when it is probed perpendicular to the steps (the in-plane uniaxial anisotropy can be neglected in this case), we introduce only the perpendicular (with respect to the terrace plane) magnetocrystalline anisotropy, i.e., we assume that the in-plane fourfold anisotropy is much weaker. Such perpendicular anisotropy can be expressed as $K_u = K_u^v + K_u^s/d_{\mathrm{Fe}}$, where K_u^v and K_u^s are volume and surface contributions to the total anisotropy energy, respectively. Note, that such volume contribution (probed perpendicular to the steps) can vary with thickness due to varying the in-plane uniaxial anisotropy forcing the magnetization to be oriented perpendicular or parallel to the steps. Due to the competition between the magnetocrystalline anisotropy and the magnetic shape anisotropy, for magnetization perpendicular to the steps the relation between tilting angle δ and vicinal angle α can be derived as following:[18]

$$\delta = \left(1 - \frac{1}{1 + \frac{K_u^v}{2\pi M^2} + \frac{K_u^s}{2\pi M^2} \times \frac{1}{d_{\mathrm{Fe}}}}\right) \times \alpha \qquad (4)$$

or

$$\frac{1}{1-\delta/\alpha} = 1 + \frac{K_u^v}{2\pi M^2} + \frac{K_u^s}{2\pi M^2} \times \frac{1}{d_{\mathrm{Fe}}}. \qquad (5)$$

Thus $\frac{1}{1-\delta/\alpha}$ should show a $1/d_{\mathrm{Fe}}$ thickness dependence which can be proven experimentally as shown in Fig. 4(b). Taking the value of $M_{\mathrm{Fe}} = 1.71 \times 10^3$ Gauss,[1] the linear fitting results in $K_u^v = 4.9 \times 10^6$ erg/cm^3 and $K_u^s = -1.6$ erg/cm^2. Here the negative sign of K_u^s means that the surface contribution to the perpendicular anisotropy has the easy axis normal to the surface. The thickness of SRT, d_c, can be calculated as $d_c = \frac{-K_u^s}{K_u^v - 2\pi M^2} \sim 5$ ML, which is consistent with the Fe thickness where the polar Kerr signal starts to decrease [see Fig. 2(a)].

Our results indicate that the volume contribution to the perpendicular anisotropy existing in this system (and forcing the magnetization to be oriented in the terrace plane) is strong. Most likely the dominant contribution to this anisotropy originates from the strain in Fe film. The lattice mismatch between fcc Ag and bcc Fe structure is about $\sim 0.8\%$ so the Fe film grown epitaxially on Ag substrate expands into the film plane and is compressed vertically.

Finally, the question is why δ oscillates? Principally speaking, the tilting angle of the magnetization originates from the competition between the magnetocrystalline and the shape magnetic anisotropy. From the MOKE measurements in $\alpha^{\parallel}$ geometry, the Kerr signal changes linearly with the thickness indicating that the magnetization and the optical constant do not oscillate with the Fe thickness. Thus, the shape anisotropy will not oscillate with Fe thickness and our results show that the perpendicular magnetocrystalline anisotropy oscillates with the Fe thickness, in particular, at low temperature.

Now we compare the thickness-dependent oscillation of the in-plane uniaxial anisotropy, described by H_s, and the tilting angle δ (Fig. 5). These two oscillations have the same period of ~ 5.7 ML and are *in phase*, i.e., the maximum of the absolute magnitude of δ coincides with the maximum of H_s. As can be seen in Fig. 5, at thicknesses at which the magnetization is more forced to be oriented along the step edges (i.e., at a maximum of H_s), the absolute magnitude of the tilting angle δ is larger than the absolute magnitude of the tilting angle at those thicknesses at which the magnetization is less forced to be oriented along the step edges, i.e., at a minimum of H_s. By contrast, one would expect *antiphase* oscillation in the sense that the larger the in-plane uniaxial anisotropy (and H_s), the smaller the magnitude of the tilting angle δ. In such case the magnetization should be less forced to be oriented in the terrace plane perpendicular to the steps, which is equivalent to a reduction of K_u^v and thus should result in a smaller δ—see Eq. (4). However, this expectation is not met according to Fig. 5, which means that the oscillating tilting angle cannot be explained as a simple consequence of the oscillatory uniaxial contribution to the in-plane

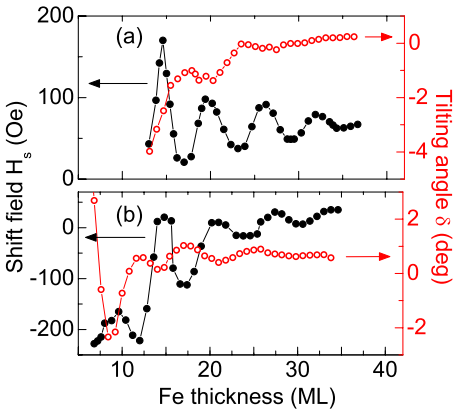

FIG. 5. (Color online) Shift field H_s (solid dots) and tilting angle δ (open dots) at 5 K as a function of Fe thickness for Fe film grown on Ag(1,1,10): (a) without Au capping layer and (b) with a 4-ML-thick Au capping layer.

anisotropy of the system. Instead this means that also the perpendicular anisotropy experiences oscillatory behavior of the same period. However, the QWS may provide a different effect on the atoms at the steps and the atoms at the terraces. The step atoms can influence the in-plane anisotropy[27] whereas the perpendicular anisotropy can be influenced differently by the atoms at both the terraces and the steps, which might be can result in the *in-phase* oscillation observed experimentally. However, the mechanism relating the oscillatory δ (i.e., the oscillatory perpendicular anisotropy) to the oscillatory H_s (i.e., to the oscillatory in-plane anisotropy) is not fully understood yet.

The in-plane magnetic anisotropy in the Fe film has been proven to be modulated by the QWS of *d*-band electrons at the Fermi level.[13] The same period of the in-plane and perpendicular anisotropy oscillations clearly proves that the oscillations have the same origin, which is related to the QWS in a minority-spin *d* band with $\Delta_{2'}$ symmetry at the Fermi surface,[13] and not from the QWS in *d* band with Δ_5 symmetry as predicted by the theoretical calculations for Fe/Au multilayer.[15] This is because the value of the Fermi wave vector, k_F, of the Fe minority spin *d* band with $\Delta_{2'}$ symmetry is estimated to be $0.2k_{BZ}$, where k_{BZ} is the Brillouin zone wave vector, providing an oscillation period of 5 ML. This is close to our experimental value. A similar oscillatory behavior of the spin-dependent transport in Fe/MgO/Fe tunnel junctions was also observed and attributed to the QWS in Fe(001) of minority electrons with Δ_1 symmetry (of *sp* character).[28] Since many QWS in different bands may exist in the Fe ultrathin film, our results indicate that only the electron state in the *d* band with $\Delta_{2'}$ symmetry plays an important role for the magnetic anisotropy in the Fe film. On other hand, our study provides a useful method to engineer the magnetic anisotropy, which is interesting for basic knowledge and possible applications in spintronics devices.

V. SUMMARY

In summary, our results show that the tilting angle of the Fe magnetization oscillates with the Fe thickness at 5 K. This oscillatory behavior has the same origin as the in-plane anisotropy oscillation, which is attributed to the QWS of *d*-band electrons at the Fermi level in Fe thin films.

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