

## Oscillatory magnetic anisotropy due to quantum well states in thin ferromagnetic films (invited)

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Magnetic anisotropy depends strongly on the density of states at the Fermi level. If significant contributions to magnetocrystalline anisotropy energy (*MAE*) are due to spin-polarized quantum well states (*QWS*), a significant increase of *MAE* can occur periodically as a function of film thickness. The oscillation period *L* is determined by the wavelength of the corresponding electron waves. The uniaxial magnetic anisotropy of fcc-Co is found to oscillate with a period of 2.3 ML. In contrast, in bcc-Fe, the uniaxial magnetic anisotropy oscillates with a period of 5.9 ML. We attribute such oscillations to *QWS* in a minority-spin *d*-band at the Fermi level. © 2012 American Institute of Physics. [doi:10.1063/1.3670498]

### I. INTRODUCTION

Magnetocrystalline anisotropy is one of the key properties of ferromagnetic (*FM*) thin films and is of particular importance for their application in magnetic recording and spintronics. Magnetocrystalline anisotropy is caused by spin-orbit coupling of electrons, which can be interpreted as a relativistic coupling between the spin of a moving electron and the electric field created by all nuclei and other electrons present in the system. There exist several concepts how to manipulate magnetic anisotropy in order to achieve the desired anisotropy configuration. The magnetocrystalline anisotropy energy (*MAE*) in *FM* films is determined by *d*-electrons. It depends on the separation energies between the various states from the electron *d*-band and, thus, it can be changed by varying the symmetry of the atomic arrangement, for instance, by tetragonal distortion.<sup>1,2</sup> More generally, any manipulation of the *d*-electron bands, which results in occupied and unoccupied states close to the Fermi level ( $E_F$ ), can lead to a significant increase of *MAE*. In particular, quantum well states (*QWS*) can lead to such an effect at film thicknesses at which the *QWS* are placed close to  $E_F$ .<sup>3</sup>

In this article, we show how *QWS* formed in *FM* thin films influence their magnetocrystalline anisotropy. We show how the magnetic anisotropy oscillates with film thickness, how the oscillation period depends on electronic structure, and how the oscillation amplitude changes with varying temperature.

### II. MAGNETIC ANISOTROPY AND QUANTUM WELL STATES

#### A. Magnetocrystalline anisotropy at stepped surfaces

In the absence of perpendicular magnetic anisotropy, the magnetocrystalline anisotropy of thin films grown on single-

crystalline substrates reflects the symmetry of the crystal surface. However, the symmetry can be decreased by growing the film on a vicinal/stepped surface.<sup>4</sup> Such a symmetry reduction is often described as an additional uniaxial magnetic anisotropy with the easy magnetization axis in the film plane, oriented along or perpendicular to the step direction. In case the steps are oriented along one of the easy axes of the four-fold anisotropy of a *FM*(001) film, one of them becomes the easy magnetization axis and the other the intermediate magnetization axis. In first approximation, the effective step-induced uniaxial anisotropy energy can be expressed as

$$K_{S,eff}(N) = K_{S,vol} + K_{S,surf}/N, \quad (1)$$

where  $K_{S,vol}$  and  $K_{S,surf}$  describe volume and surface contributions to the step-induced anisotropy, respectively. Since  $K_{S,vol}$  is independent of *N*, it is interpreted as the result of the structural distortion in the film volume above the step edges.  $K_{S,surf}/N$  contains information on how the uniaxial step-induced anisotropy depends on film thickness (it originates from the loss of translational symmetry at the step edges and, thus, depends on thickness as  $1/N$ ).<sup>5,6</sup>

In *FM* films grown on vicinal surfaces, there is also competition between the magnetocrystalline anisotropy, which prefers an orientation of the magnetization along the principal crystallographic directions (i.e., in the terraces plane) and the shape anisotropy, which prefers an orientation of the magnetization in the film plane (which, for vicinal surfaces, is not equivalent to the principal crystallographic planes).<sup>4,7</sup> As a result, if the magnetization is oriented perpendicular to the step edges, it can be tilted by a small angle away from the film plane toward the terrace plane.

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## B. Magnetocrystalline anisotropy in view of second order perturbation theory

The *MAE* results from the anisotropy of the spin-orbit interaction, i.e., it is the difference,

$$E_{\text{MA}} = E(\theta_2, \varphi_2) - E(\theta_1, \varphi_1), \quad (2)$$

of the total energies obtained from the Hamiltonian, including the spin-orbit coupling term, with the magnetization pointing in two different directions defined with the corresponding polar ( $\theta$ ) and azimuthal ( $\varphi$ ) angles. Since spin-orbit coupling  $H_{\text{SO}}(\theta, \varphi)$  is small in transition metals, it can be treated as a perturbation and perturbation theory can be applied to calculate the energy  $E(\theta, \varphi)$  and, subsequently, *MAE*. For systems with reduced symmetry, like thin films (perfectly flat or deposited on stepped surfaces), the dependence of the system energy  $E = E^{(0)} + E^{(2)} + E^{(4)} + \dots$  on spin orientation is already present in the second-order energy correction,<sup>8,9</sup>

$$E^{(2)}(\theta, \varphi) = \frac{1}{2} \sum_{\mathbf{k}_{\parallel}} \sum_{n\sigma} \sum_{n'\sigma' \neq n\sigma} \frac{f(\varepsilon_{n\sigma}(\mathbf{k}_{\parallel})) - f(\varepsilon_{n'\sigma'}(\mathbf{k}_{\parallel}))}{\varepsilon_{n\sigma}(\mathbf{k}_{\parallel}) - \varepsilon_{n'\sigma'}(\mathbf{k}_{\parallel})} \times \left| \langle n\sigma \mathbf{k}_{\parallel} | H_{\text{SO}}(\theta, \varphi) | n'\sigma' \mathbf{k}_{\parallel} \rangle \right|^2, \quad (3)$$

where  $f(\varepsilon)$  is the Fermi occupation factor. Thus, *MAE* is expressed by matrix elements of the spin-orbit interaction between occupied and unoccupied states ( $|n\sigma \mathbf{k}_{\parallel}\rangle$ ,  $|n'\sigma' \mathbf{k}_{\parallel}\rangle$ ) and their energies ( $\varepsilon_{n\sigma}(\mathbf{k}_{\parallel})$ ,  $\varepsilon_{n'\sigma'}(\mathbf{k}_{\parallel})$ ), with the same or opposite spins  $\sigma$ ,  $\sigma'$  and lateral wavevectors  $\mathbf{k}_{\parallel} = (k_x, k_y)$  (for flat films) from the whole two-dimensional Brillouin zone (*BZ*). Since the individual terms in *MAE* are inversely proportional to the energy difference between occupied and unoccupied states, their contribution to *MAE* is particularly large if energies of such pairs of states are close to each other. Thus, a significantly enhanced *MAE* can be expected for an electron configuration where one of the states is placed just below and one just above the Fermi energy  $E_F$ . Such states can be responsible for strong changes of *MAE* due to the factors that shift their energies with respect to  $E_F$ . Therefore, magnetocrystalline anisotropy can depend strongly on film thickness,<sup>9</sup> tetragonal distortion<sup>1,2</sup> or local structure relaxation, and crystal field splittings (*d*-orbital energies) near step edges.<sup>10</sup> The *MAE* contributions from pairs of states with energies placed less than a few  $k_B T$  around  $E_F$  are also strongly affected by finite temperature  $T$ .

## C. Quantum well states and magnetic anisotropy oscillations in thin ferromagnetic films

In a thin film, electron motion is confined by the upper and lower surface of the film. In this case, electrons may form standing waves if their wavelength fits into the thickness of the film. Such standing waves represent states called quantum well states (*QWS*).<sup>11–13</sup> In other words, *QWS* in thin films come from the Bloch states in the bulk, which are reflected at the boundaries of the film. As a consequence, the  $z$  component,  $k_z$ , of the three-dimensional wavevector  $\mathbf{k} = (k_x, k_y, k_z) = (\mathbf{k}_{\parallel}, k_z)$  is quantized and the *QWS* energies are placed near  $E_F$  periodically, i.e., at specific thicknesses,

$$N_m = N_0 + m \cdot L, m = 1, 2, \dots \quad (4)$$

As it was mentioned before, any change of the electronic structure of the *3d* states close to  $E_F$  can result in a change of magnetocrystalline anisotropy. An exciting manifestation of this direct correlation between the density of states at  $E_F$  and magnetocrystalline anisotropy would be the oscillation of magnetic anisotropy with film thickness with a period  $L$  (see Fig. 1). Such an effect indeed takes place, since, once a *QWS*  $|n\sigma \mathbf{k}_{\parallel}\rangle$  crosses  $E_F$ , its occupancy changes from occupied to unoccupied or vice versa. In the resulting electron configuration, this *QWS* couples (see Eq. (3)) to a different set of states  $|n'\sigma' \mathbf{k}_{\parallel}\rangle$  (e.g., occupied instead of unoccupied), so that its contribution to *MAE* becomes different. This effect is particularly enhanced if the moving *QWS* couples to (i.e., forms a pair with) another state  $|n'\sigma' \mathbf{k}_{\parallel}\rangle$  placed close to  $E_F$ . In such a case, a pair of occupied and unoccupied states exists for which the energies are very close to each other (Fig. 1). Thus, a modification of magnetocrystalline anisotropy can occur due to such *QWS*, but only at the specific thicknesses  $N_m$  [see Eq. (4)].

In this way, *QWS* lead to an oscillatory dependence of *MAE* on the film thickness. The oscillation period  $L$  is then determined by the wave vector  $k_{z0}$  of the electron waves (i.e.,  $k_z$  of the bulk *d*-band, corresponding to the *QWS* that cross  $E_F$ ) and can be used to identify the electronic states which contribute to the *MAE*. In particular, the oscillatory term in *MAE* can occur due to the *QWS*, which are derived from a pair of bulk states with  $\Delta_5$  symmetry. These *QWS* form intrinsic pairs which have energies very close to each other for  $\mathbf{k}_{\parallel}$  around the  $\bar{\Gamma}$  point. Thus, they contribute

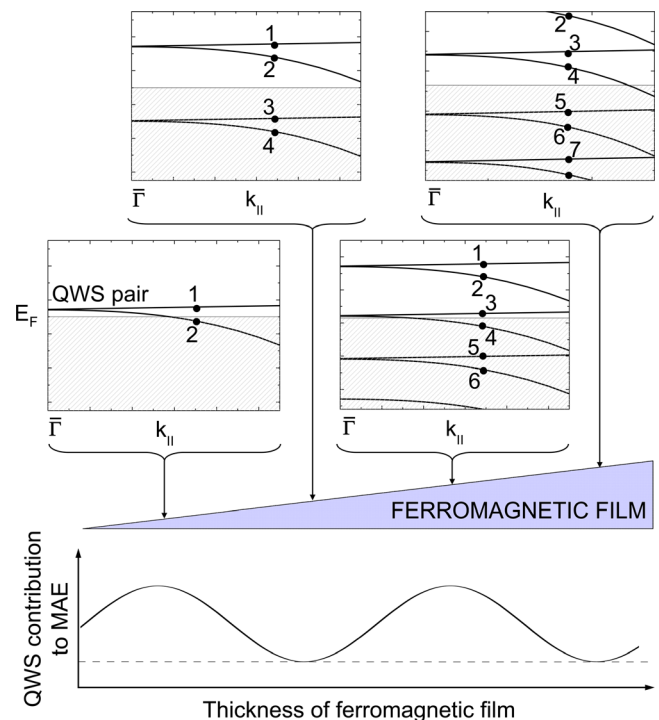


FIG. 1. (Color online) Schematic representation of how *QWS* are placed with respect to  $E_F$  and how their contribution to *MAE* changes with increasing film thickness. The *QWS* contribution to *MAE* (solid line) is plotted with respect to the *MAE* without *QWS* (dashed line).

strongly to  $MAE$  only if  $E_F$  lies in between the energies of the pair states. This particular mechanism has been theoretically identified to be responsible for the oscillations of magnetic anisotropy in (001) fcc-Co and Co/Pd films<sup>14–16</sup> (see Sec. IV A). The amplitude of  $MAE$  oscillations coming from such pairs of  $QWS$  decreases strongly with temperature, since the energy separation between the pair states is of the order of a few  $k_B T$  over a significant region of the  $BZ$  around the  $\bar{\Gamma}$  point.

$QWS$  can be formed not only in the  $FM$  film, but also in the nonmagnetic (NM) overlayer. Thus,  $QWS$  can affect both the volume and the interface contributions to the magnetic anisotropy of the  $NM/FM$  system. The effect of  $QWS$  in the  $NM$  overlayer on the underlying  $FM$  film is reported both from a theoretical and experimental point of view.<sup>16–18,29</sup> While this effect is relatively weak for  $Cu(N)/(001)fcc-Co$  films, much more pronounced  $MAE$  oscillations (due to large spin-orbit coupling of Pd atoms) are predicted for  $Pd(N)/(001)fcc-Co$  films, with an amplitude comparable to that observed due to  $QWS$  formed directly in the  $FM$  film.<sup>16</sup>

The oscillations of  $MAE$  can originate only from  $QWS$  states with wavevectors lying in the vicinity of the high-symmetry point of the  $BZ$ , since the latter are the stationary points of the energy bands<sup>3</sup> (the same condition is required for oscillations of interlayer exchange coupling<sup>19</sup>). Then, the wavevectors  $k_{z0}$ , which define the  $MAE$  oscillation periods, are extremal radii of the three-dimensional Fermi surface in the  $z$  direction, and, thus, several periods corresponding to different  $d$ -bands and different high-symmetry points can exist.

### III. EXPERIMENTAL DETAILS

The experiments discussed in this article were performed in a standard multi-chamber ultrahigh vacuum ( $UHV$ ) system with a base pressure below  $2 \times 10^{-10}$  mbar. Single crystalline substrates (of Ag and Cu) were prepared by several cycles of 1 keV Ar ion sputtering and subsequent annealing, usually at 500–600 °C. In case of vicinal surfaces, using scanning tunneling microscopy ( $STM$ ), nearly equidistant and regular monoatomic steps along the [110] direction were observed on the surface. Fe and Co films were grown at room temperature ( $RT$ ) and at 190 K, respectively, by molecular beam epitaxy. Some samples were capped with a Au layer (0.5–4 ML).

An ideal method for studying magnetic anisotropy of thin films is provided by the magneto-optical Kerr effect ( $MOKE$ ). This method can be easily applied *in situ* under  $UHV$  conditions and therefore allows the study of uncovered films. Qualitatively,  $MOKE$  gives direct information on the magnetic state of the sample, i.e., it is relatively easy to determine whether the magnetization is probed along an easy or hard direction. Quantitatively, one can measure Kerr rotation or ellipticity by  $MOKE$ , which is proportional, but not equal to the magnetization,  $M$ . Thus, in order to obtain quantitatively the  $MAE$  ( $=\mu_0 \cdot H_A \cdot M_S$ ), a value for  $M_S$  must be assumed. Another quantitative limitation of  $MOKE$  is related to the anisotropy field,  $H_A$ , i.e., the field at which magnetization saturates if the magnetic field is applied along the hard axis. Since  $H_A$  is usually large and not sharply

defined, it is often difficult to determine its value precisely and to follow small variations of  $H_A$  by  $MOKE$ .

For magnetic thin films grown on stepped surfaces, the superposition of four-fold and uniaxial anisotropy has the consequence that so-called split hysteresis loops can be measured when the magnetic field is applied along the intermediate magnetization axis (Fig. 2).<sup>4–6,20</sup> Split hysteresis loops are characterized by a shift field ( $H_S$ ), which is defined as half of the distance between two constituent loops (Fig. 2). The more the magnetization prefers an orientation along the easy axis, the larger the anisotropy and the larger  $H_S$ . Therefore,  $H_S$  can be taken as a measure of the anisotropy modification introduced by the substrate steps. Positive or negative  $H_S$  refers to the situation where the easy magnetization axis is oriented along or perpendicular to the steps, respectively. In contrast to the anisotropy field  $H_A$ , the shift field  $H_S$  can be easily determined by  $MOKE$  and can be used to estimate the uniaxial anisotropy induced by the steps on the substrate surface.

As mentioned before, perpendicular to the steps, competition between the magnetocrystalline and shape anisotropy can tilt the magnetization out of the film plane. Accordingly, if the magnetization is oriented perpendicular to the steps, it will have a small component normal to the film plane, whereas the magnetization will be completely in the film plane if it is oriented parallel to the steps.<sup>20,21</sup> Since the polar Kerr effect is much stronger than the longitudinal Kerr effect ( $S_L$  in Fig. 2), even a small normal component of the magnetization can give a significant polar contribution ( $\Delta S_P$  in Fig. 2) to the total Kerr signal.

In the experiments reported in this article, magnetic hysteresis loops were probed by *in situ* longitudinal (fixed incidence angle 21°)  $MOKE$  with a laser diode of wavelength 670 nm and beam diameter  $< 0.2$  mm. In the experimental  $MOKE$  setup, the sample can be rotated in the film plane

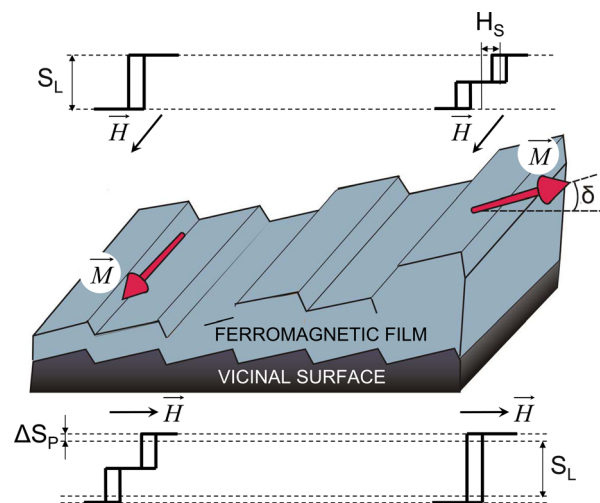


FIG. 2. (Color online) Schematics of  $FM$  film grown in wedge geometry on a vicinal surface with easy magnetization axis [thick (red) arrows] oriented parallel or perpendicular to the steps. Representative hysteresis loops for longitudinal  $MOKE$  measurements for the easy magnetization axis oriented either parallel or perpendicular to the steps are also shown.  $S_L$  denotes longitudinal  $MOKE$  signal and  $\Delta S_P$  denotes additional polar  $MOKE$  signal due to tilting of the easy axis relative to the sample plane by angle  $\delta$ .

with respect to the magnetic field and the plane of the incoming and outgoing laser beam.

#### IV. ANISOTROPY OSCILLATIONS DUE TO QWS

Since it is not trivial to observe anisotropy oscillations due to *QWS*, a stepped surface can be intentionally used to lower the symmetry of the substrate. In particular, step-induced anisotropy is well known for the Co/Cu(001)<sup>14,17,22</sup> and Fe/Ag(001)<sup>4,23</sup> systems. As already mentioned, the shift field,  $H_S$ , is a measure of the step-induced uniaxial anisotropy and is therefore thickness-dependent. According to Eq. (1), the thickness dependence of  $H_S$  at *RT* can be described as a sum of two contributions:  $H_{S,RT}(N) = H_{S,vol} + H_{S,surf}/N$ . In this approach,  $H_{S,vol}$  is the value which  $H_{S,RT}$  approaches in the limit of thick films, whereas  $H_{S,surf}$  determines  $H_{S,RT}$  in the limit of thin films (Fig. 3). Both,  $H_{S,vol}$  and  $H_{S,surf}$  depend strongly on the density of the surface steps. The uniaxial anisotropy becomes larger when the density of steps increases.<sup>4,6</sup> For instance, we found that the value of  $H_{S,vol}$  for Fe films grown on Ag(1,1,6) is by a factor of 4 larger than the value observed for Fe films grown on Ag(1,1,10) (Fig. 3).<sup>6</sup> Usually, the surface/interface contribution,  $H_{S,surf}$ , is much larger than  $H_{S,vol}$ . For instance, covering with Au introduces a strong interface contribution to the step-induced uniaxial magnetic anisotropy (Fig. 3). Similar to covering with Cu,<sup>24</sup> a minute amount of Au can remarkably reduce  $H_S$  and even change the easy magnetization axis from parallel to perpendicular to the step edges. Covering the film modifies  $H_{S,surf}$ , whereas  $H_{S,vol}$  remains unchanged.<sup>6</sup> The above description, in particular, how  $H_{S,RT}$  depends on  $N$ , is valid at *RT*, i.e., if there are no or only weak anisotropy oscillations present. A smooth thickness dependence of  $H_{S,RT}$  is not surprising. It is expected that anisotropy oscillations vanish with temperature when the spread of the Fermi function ( $\sim 4 \cdot k_B T$ ) becomes comparable to the energy difference between the two states of each *QWS* pair contributing to the *MAE*.<sup>3</sup> This contribution is large only for those *QWS* states which are close to  $E_F$  at  $\bar{\Gamma}$  (i.e., at  $\mathbf{k}_{||} = (k_x, k_y) = (0, 0)$ ). As a consequence: (1) strong oscillations of magnetocrystalline anisotropy are expected only at

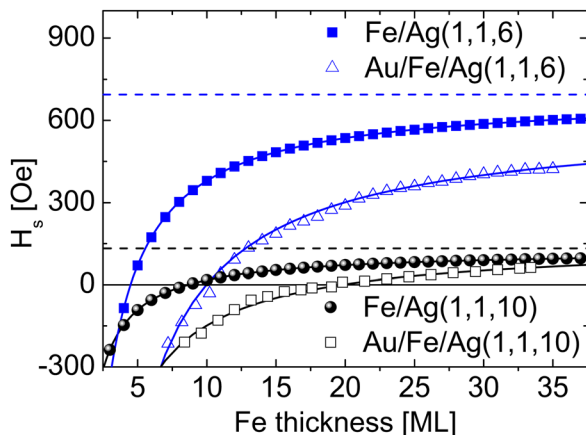


FIG. 3. (Color online) Shift-field  $H_S$  measured at *RT* and plotted vs Fe thickness for Fe/Ag(1,1,10) (Ref. 5) and Fe/Ag(1,1,6) (Ref. 6) uncovered and covered with 4 ML of Au. Dashed lines represent the values of  $H_{S,vol}$  for Fe films grown on Ag(1,1,6) (upper) and Ag(1,1,10) (lower).

low temperatures (*LT*), (2) magnetocrystalline anisotropy for the thicknesses  $N_m$  [Eq. (4)] should exhibit a strong dependence on temperature, whereas, for those thicknesses  $N$  for which no pairs of *QWS* contribute to, the anisotropy should exhibit only very weak temperature dependence. In order to investigate those predictions, a complete temperature-dependent *MOKE* experiment was performed for two systems: fcc-Co films grown on vicinal surfaces of Cu(001) and bcc-Fe films grown on vicinal surfaces of Ag(001).

#### A. QWS and oscillatory anisotropy in Co/Cu(001)

The Co/Cu(001) system is the only system for which oscillatory magnetic anisotropy due to *QWS* in *FM* layers has been investigated by theory<sup>3,10,25</sup> and, therefore, allows direct comparison to experiments.<sup>26</sup> Cinal<sup>3</sup> explicitly postulated and examined the role that *QWS* play in purely *FM* systems, like  $N$  monolayers of fcc-Co on Cu(001). A careful analysis with a parametrical tight-binding (*TB*) model<sup>3,10</sup> revealed that the oscillations of the total *MAE* in Co/Cu(001) are a superposition of two oscillatory contributions. The dominating contribution comes from the neighborhood of the  $\bar{\Gamma}$ -point, more precisely from the *QWS* originating from the doubly degenerate  $\Delta_5$  band, which crosses  $E_F$  at  $k_{z0} = 0.528 \cdot 2\pi/a$  and, thus, corresponds to a period of 2.12 ML. The second contribution is of significantly smaller amplitude and originates from the region around the *M*-point, more precisely from the  $Z_3$ -band, which crosses  $E_F$  at  $k_{z0} = 0.194 \cdot 2\pi/a$  and  $k_{z0} = 0.806 \cdot 2\pi/a$ , and, thus, corresponds to a larger period of 5.15 ML. Later, the calculations were extended to Co films on vicinal surfaces of Cu(001), where the uniaxial magnetocrystalline anisotropy was found to oscillate with a period close to 2 ML, as well.<sup>10</sup>

Only recently,<sup>26</sup> we were able to confirm experimentally these predictions, which were made nearly 10 years ago. We performed the experiment for uncovered and Au-covered Co films grown on Cu(1,1,13) (i.e., a vicinal surface of Cu(001)). Covering with Au does neither influence the period nor the amplitude; however, it changes the “base value” ( $H_{S,RT}$ ) at which the anisotropy oscillations occur at *LT*. Depending on how much the Co film is covered with Au, oscillations of the same amplitude do or do not result in an oscillatory easy magnetization axis, i.e., an easy axis which changes its orientation from parallel to perpendicular to the steps. In particular, we investigated the thickness dependence of  $H_S$  for Co/Cu(1,1,13) covered with 0.5 ML of Au at  $T = 5$  K.<sup>26</sup> Here,  $H_{S,LT}$  oscillates with increasing Co thickness with a period  $L = 2.3 \pm 0.3$  ML (Fig. 4), in excellent agreement with theory.<sup>3,10,27</sup> The maximum oscillation amplitude (in the thickness range below 15 ML of Co, i.e., below the thickness of strain relaxation<sup>28</sup>) is about 300 Oe, i.e., almost two orders of magnitude larger than the anisotropy oscillations caused by *QWS* in the Cu overlayer in the Cu/Co/Cu(001) system.<sup>24</sup> This confirms the volume character of the observed anisotropy oscillations, which are clearly due to *QWS* formed in the Co film.

The agreement between theory and experiment is almost perfect, in particular, in the thickness range between 9 and 15 ML (with three distinct maxima at about 10, 12, and 14 ML, Fig. 4). According to our expectations, a strong temperature dependence of magnetocrystalline anisotropy should

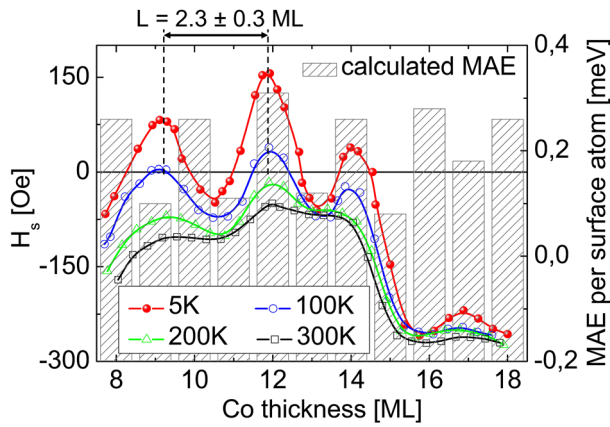


FIG. 4. (Color online) Shift-field  $H_S$  measured at varying temperature and plotted as a function of Co thickness for Co/Cu(1,1,13) covered with 1 ML of Au. Experimental data (Ref. 26) are compared to the band energy contribution to the magnetic anisotropy energy calculated per surface atom (Ref. 3).

occur only for those thicknesses for which the  $QWS$  contribute strongly to the electronic structure at  $E_F$ .<sup>3,10</sup> Experimentally, it is found (see Fig. 4) that the anisotropy at  $N = N_m = 9.3, 11.9, 14.0,$  and  $16.6$  ML [Eq. (4)] depends strongly on temperature, whereas, for  $N = 10.6, 13,$  and  $15.6$  ML, the anisotropy changes only little with temperature,<sup>26</sup> which is in excellent agreement with theory. The theoretically calculated oscillation amplitude of the  $MAE$  of  $\Delta H_S = 140 \mu\text{eV}$  was obtained per step atom for the Co/vicinal-Cu(001) system at  $RT$ .<sup>3,10</sup> The change of the anisotropy energy corresponding to the experimentally observed (maximal)  $\Delta H_S$  of 300 Oe is estimated to be  $230 \mu\text{eV}$ . Considering that the experiments were performed at 5 K, this is in very good agreement with theory. The oscillation amplitude decreases with increasing temperature and vanishes completely a little above  $RT$ .

## B. QWS and oscillatory anisotropy in Fe/Ag(001)

To prove whether the period of the anisotropy oscillations is related to the electronic structure, we carried out another *MOKE* experiment for bcc-Fe films grown on vicinal surfaces of Ag(001). It is expected that the amplitude of the oscillations depends on how strongly the anisotropy is modified by the steps. In order to prove this concept, we used two different vicinal surfaces of Ag(001), one with a larger (Ag(1,1,10)<sup>5</sup>) and one with a smaller (Ag(1,1,6)<sup>6</sup>) terrace width. For the Au/Fe/Ag(1,1,6) sample, at 5 K,  $H_{S,LT}$  clearly oscillates with Fe thickness (Fig. 5) with a period  $L = 5.9 \pm 0.4$  ML, which is exactly the same as observed for Fe films grown on Ag(1,1,10). This is natural, since there is the same ultrathin film of Fe grown on the same Ag(001) substrate. In both cases, the width of each succeeding maximum of  $H_{S,LT}$  increases with increasing film thickness. This is in part due to the surface quality, which degrades with increasing film thickness, and in part due to the increasing number of  $QWS$  pairs contributing to the anisotropy. The oscillation amplitude  $\Delta H_S$ , i.e., the difference between the first maximum and the first minimum of  $H_{S,LT}$  measured for Au/Fe/Ag(1,1,6) is about 550 Oe, i.e., nearly 4 times larger than for Au/Fe/Ag(1,1,10)

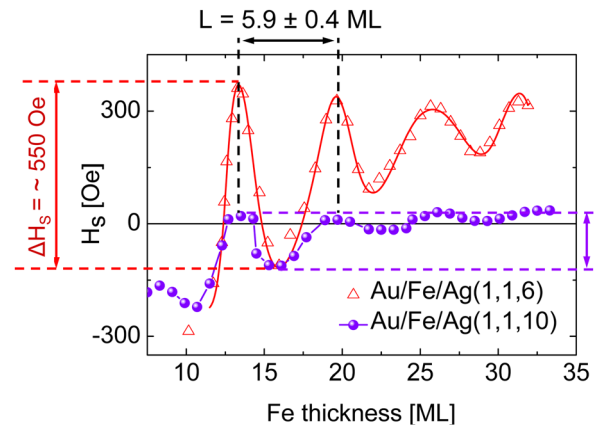


FIG. 5. (Color online) Shift-field  $H_S$  measured at 5 K and plotted vs Fe thickness for Fe/Ag(1,1,10) (Ref. 5) and Fe/Ag(1,1,6) (Ref. 6) covered with 4 ML of Au.

(Fig. 5). The anisotropy is stronger the more the film is structurally perturbed by the steps. Therefore, increasing the step density results in an amplification of the film anisotropy,  $H_S \approx H_{S,RT} + \Delta H_S$ . This is why  $H_{S,RT}$  (Fig. 3) and the oscillation amplitude  $\Delta H_S$  (Fig. 5) scale in the same way with increasing step density.<sup>6</sup>

There are no theoretical calculations available to which we can compare our experimental results on the oscillatory magnetic anisotropy in bcc-Fe films. Nevertheless, the oscillation period of  $QWS$  is determined by  $k_{z0}$  and thus can be estimated from the electronic structure of Fe, in particular along the  $\Gamma$ -H direction, which is the direction along which the electrons are confined in the Fe films. In this case,  $k_{z0}$  of the Fe minority-spin  $d$ -band with  $\Delta_2'$  symmetry is estimated to be  $0.2 \cdot 2\pi/a$ .<sup>29</sup> Thus,  $QWS$  at  $E_F$  formed in this electronic band should have an oscillation period of 5 ML, which is very close to our experimentally observed value. A very similar value was obtained for the period of a strong oscillation of magnetic coupling as a function of the thickness of one of the Fe electrodes for the Fe/Cr/Fe system.<sup>30,31</sup> This confirms that  $QWS$  formed in the same electron band are responsible for the oscillatory behavior observed for magnetic anisotropy and for interlayer coupling. However, only  $\Delta_5$  bands are doubly degenerate along  $\Gamma$ -H, whereas the  $QWS$  from the  $\Delta_2'$  band have to couple (i.e., form pairs) with other states. Thus, such pairs, where one of the states is a  $QWS$  from the  $\Delta_2'$  band, are not degenerate at  $\bar{\Gamma}$  and thus do not necessarily contribute to  $MAE$  strongly enough to give a large oscillatory term in the  $MAE$  dependence on Fe film thickness.

Finally, as described in Sec. III, there can be a polar contribution to the hysteresis loops measured by *MOKE* in longitudinal geometry. Since the polar Kerr effect is larger than the longitudinal one, the hysteresis loops are sensitive, even to a small perpendicular component of the magnetization. Thus, longitudinal hysteresis loops allow detection of tiny changes in the orientation of the magnetization, and even a small tilting angle can be reasonably well determined. It was found for both Co and Fe films grown on vicinal surfaces of Cu(001) and Ag(001), respectively, that at 5 K the tilting angle shows very clear oscillations with periods similar to the ones observed for the oscillations of  $H_S$ .<sup>7,26</sup> From *MOKE*

measurements along the steps, it is known that the Kerr signal increases linearly with increasing film thickness. This indicates that the magnetization (and thus the shape anisotropy) as well as the optical constants do not oscillate with film thickness. Since the tilting angle of the magnetization originates from the competition between magnetocrystalline anisotropy and shape anisotropy, these results show that also the perpendicular magnetocrystalline anisotropy oscillates at  $LT$ .<sup>7</sup>

## V. CONCLUSIONS

It is known that magnetic anisotropy depends strongly on the density of states at the Fermi level.  $QWS$  formed in Fe and Co thin films can result in occupied and unoccupied electronic states close to  $E_F$  and thus contribute to magnetic anisotropy. The uniaxial magnetic anisotropy of fcc-Co is found to oscillate as a function of Co thickness with a period of  $2.3 \pm 0.3$  ML. In contrast, in bcc-Fe, the uniaxial magnetic anisotropy oscillates with a period of  $5.9 \pm 0.4$  ML. We attribute such oscillations to the  $QWS$  in a minority-spin  $d$ -band at the Fermi level. The oscillation periods are different for Co and Fe, due to their different electronic structure and thus different  $k_{z0}$  for the  $d$ -band electrons of  $\Delta_5$  and  $\Delta_2'$  symmetry, respectively. These observations directly confirm the correlation between  $QWS$  and oscillatory magnetic anisotropy.

The oscillation amplitude of  $H_S$  can be engineered by the density of surface steps: the larger the density of steps, the larger the amplitude of the oscillation. Since  $H_S$  depends on the covering material, a proper choice of the amount of the covering material can result in oscillations of  $H_S$  at  $LT$ , which do not only modify its value, but also its sign. This means that the easy magnetization axis can change its orientation from parallel to perpendicular to the steps periodically with increasing film thickness.

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