# Effect of quantum well states in Cu overlayer on magnetic anisotropy of Fe and Co films revisited

S. Manna,[1] P. L. Gastelois,[1,2] M. Dąbrowski,[1] P. Kuświk,[1] M. Cinal,[3] M. Przybylski,[1,4,*] and J. Kirschner[1,5]

[1]*Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle, Germany*

[2]*Serviço de Nanotecnologia, Centro de Desenvolvimento da Tecnologia Nuclear, 31270-901 Belo Horizonte, MG, Brazil*

[3]*Institute of Physical Chemistry, Polish Academy of Sciences, 01-224 Warsaw, Poland*

[4]*Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, 30-059 Kraków, Poland*

[5]*Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany*

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The effect of quantum well states (QWS) formed in a Cu overlayer on the magnetic anisotropy of Fe and of Co films underneath is investigated at low temperatures. An oscillatory magnetic anisotropy is observed below 50 K, with a period of $L_{Cu} = 5.8$ ML, and it is the same for Cu/Fe and Cu/Co. We attribute this effect to QWS from *sp* electrons in Cu which hybridize to *d* electrons at the Fermi level in Fe (and Co) and thus modify the magnetic anisotropy at low temperatures. This is why the effect was not so pronounced in the experiments reported 15 years ago showing the low-amplitude oscillations detected at 170 K [Ch. Würsch *et al.*, Nature (London) **389**, 937 (1997)].

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## I. INTRODUCTION

Electrons can be confined perpendicular to the film plane and form quantum well states (QWS).[1–3] The formation of QWS can alter electronic structure and result in oscillatory physical properties as a function of film thickness.[4,5] In particular, layered magnetic structures have attracted great interest after the discoveries of an interlayer exchange coupling through a nonferromagnetic spacer[6] and giant magnetoresistance (GMR).[7] Both phenomena are believed to be due to the formation of spin-polarized QWS by electron confinement in the nonferromagnetic spacer perpendicular to the structure plane.[1–3]

Magnetic anisotropy energy (MAE) in ferromagnetic films is determined by *d* electrons. 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, QWS can lead to such an effect (i.e., the magnetic anisotropy can be particularly large for specific thicknesses for which QWS formed from *d* electrons appear at $E_F$; Ref. 8).

There are two options for considering the effect of QWS on magnetic anisotropy of the ferromagnetic film (FM): (a) bilayers nonmagnet (QWS)/ferromagnet, i.e., the effect of QWS formed in a nonmagnetic overlayer/underlayer, and (b) ferromagnetic films, i.e., the effect of QWS formed in the ferromagnet itself.

Cinal and Edwards theoretically analyzed the oscillations of magnetic anisotropy in a Pd(*N*)/Co/Pd(*N*) system caused by QWS existing in the nonmagnetic Pd layers of a varying number of atomic layers *N*.[9,10] Experimentally, there is only one report on the effect of QWS in a nonmagnetic overlayer on magnetic anisotropy of ferromagnetic film.[11] The experiment was performed at 170 K for Cu films grown step by step on top of the Co/Cu(001) vicinal surface. The observed oscillations of the anisotropy field (i.e., the field necessary to align all the spins along the harder direction) consist of two electron wavelengths, 2.4 and 5.4 monolayers (ML),[11,12] which correspond to two extremal radii of the Cu Fermi surface, at the neck and the belly,[13] respectively, and had a small amplitude (only a few oersteds).

Almost identical periods, in particular, $L_{Cu} \approx 6$ ML, were obtained from an inverse-photoemission experiment on Cu films grown on single-crystalline Fe(001) and on Co(001).[13–17] The effect of QWS formed in Cu films on magnetic anisotropy of Cu(*N*)/Fe and Cu(*N*)/Co was not calculated up to now.

Instead, Szunyogh *et al.*[18] found oscillatory magnetic anisotropy in a purely ferromagnetic system like Co(*M*)/Cu(001) with a varying number *M* of Co atomic layers. Cinal[19] examined the role of QWS in the same system. His careful analysis with a parametrical tight-binding model revealed that the total magnetic anisotropy energy oscillations (with a period of 2.12 ML) result from QWS formed mainly from *d* electrons near the $\bar{\Gamma}$ point. Only recently were these predictions experimentally confirmed.[20] The effect of anisotropy oscillations, induced by QWS, was also experimentally observed for Fe films grown on vicinal surfaces of Ag(001),[21–24] where an oscillation period of 5.7 ML was found. Interestingly enough, in both cases of fcc Co and bcc Fe, the oscillatory magnetic anisotropy was clearly proven at temperatures below 170 K.

In this paper we report on the experimental and theoretical evidence of the quantum origin of anisotropy oscillations observed at low temperatures in Fe and Co films due to QWS formed in a Cu overlayer. Several requirements were fulfilled to observe such an effect. First, our ferromagnetic films were grown on vicinal surfaces, introducing an additional uniaxial in-plane anisotropy due to the symmetry lowered by the steps.[11,12,25–27] As a result, so-called split loops were measured when the magnetic field was applied along the harder axis.[20,21,23,25,28] The loops were characterized by a shift/anisotropy field $H_s$, a measure of uniaxial anisotropy introduced to the film by the steps (see Fig. 1). Positive and negative $H_s$ indicate that the easy magnetization axis is oriented along or perpendicular to the steps, respectively. In addition, Cu films are of well-defined (i.e., sharp) surfaces and interfaces with Fe (and Co) film underneath. Finally, since the

 

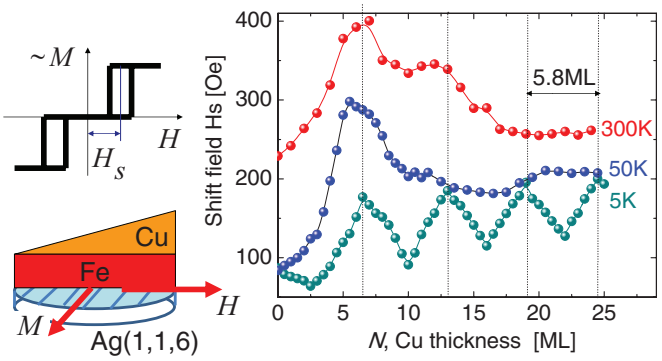


FIG. 1. (Color online) Shift field $H_S$ (defined as shown in the left panel) measured at different temperatures (from 300 to 5 K), plotted vs the thickness of the Cu overlayer for $M = 10$ ML of Fe on Ag(1,1,6) (right panel). Positive $H_s$ corresponds to the easy magnetization axis oriented along the steps.

effect of QWS on magnetic anisotropy is a low-temperature phenomenon,[8,20,21] the magnetic properties were probed at temperatures down to 5 K.

## II. EXPERIMENTAL DETAILS

The experiments were performed in a multichamber ultrahigh vacuum system with pressure below $2 \times 10^{-10}$ mbar during deposition. Ag(1,1,6) and Cu(1,1,13) substrates were prepared with cycles of 1 KeV Ar ion sputtering and subsequent annealing at ~775 and at ~900 K, respectively. The Fe and Co films were grown at room temperature (RT) and at 220 K, respectively, by molecular beam epitaxy. After growth, the films were warmed up to 450 and 300 K, respectively, in order to improve the surface morphology. In both cases, the easy axes of the fourfold anisotropy were oriented along and perpendicular to the step edges (Fe is rotated by 45° with respect to the [100] direction of Ag; the easy magnetization axis of Co on Cu(001) is oriented along [110]). Cu on Fe/Ag(1,1,6) was grown at RT, whereas on Co/Cu(1,1,13) it was grown at 170 K as a wedge shape with an ~3 ML/mm slope along the $[\bar{1}10]$ direction of the Ag(1,1,6) and Cu(1,1,13) substrates. Growth of Cu on Fe(001) is complex since fcc Cu is grown on bcc Fe. Some structural relaxation is expected near a thickness of 10 ML, above which the fully relaxed fcc Cu continues to exist.[29,30] Growth of Cu on Co(001) seems to be straightforward (a well-known and well-investigated system). However a long-time-scale interdiffusion can play an important role.[31]

Magnetic properties were probed by the *in situ* longitudinal magneto-optical Kerr effect (MOKE) with a laser diode (wavelength of 670 nm, incidence angle of 21° and beam diameter < 0.2 mm). The thickness resolution was limited by the size of the laser beam and the averaging over the thickness range resulting from the wedge slope. The MOKE measurements were performed in a wide range of temperatures from RT down to 5 K.

## III. EXPERIMENTAL RESULTS

In a simplified model of magnetization reversal considering the single-domain switching, the shift field $H_s$ is proportional to the in-plane uniaxial magnetic anisotropy $K_u$.[32] This relation is proven to be a reliable approximation, particularly for small variations of $K_u$.[11,12,21,33] A detailed description of the magnetization reversal, however, is more complex. It demands taking into account the domain wall energy and precluding the separation of the in-plane fourfold and in-plane twofold anisotropies.[34,35] For the magnetization oriented in a direction close to the sample plane, the oscillatory behavior of $H_s$ measured experimentally[20] reflects oscillatory changes of the magnetic anisotropy calculated theoretically[18,19,33] (of exactly the same period). For the magnetization oriented perpendicular to the steps, the competition between magnetocrystalline and shape anisotropies can tilt magnetization out of the film plane, and thus the magnetization reversal (and the corresponding split hysteresis loops) can be more complex.[28]

The surface of an ultrathin film (e.g., of Fe) grown on stepped surfaces [e.g., of Ag(001)] reproduces these steps at the substrate. This results in different local atomic configurations at the step edges (in comparison to the Fe atoms at the flat surface), both at the ferromagnet/substrate and nonmagnet-ferromagnet interfaces. Thus, there is a surface/interface contribution to the step-induced anisotropy (i.e., there is a $H_{s,surf}$ contribution to $H_s$), which decreases with increasing thickness $M$ of a ferromagnetic film as $1/M$.[21,23] The volume contribution to $H_s$, $H_{s,vol}$, resulting from the structural distortion in the film volume above the step edges is independent of $M$. Obviously, covering ferromagnetic film with Cu changes the $H_{s,surf}$ contribution to $H_s$, mainly due to the difference between the surface (of Fe or Co) and the interface (Cu/Fe or Cu/Co) contributions to the uniaxial anisotropy. Rather surprisingly, it is found that not only is the covering material important, but also the thickness of the covering layer (of Cu in this case) plays a role in determining the magnetic anisotropy of the system. For example, for a constant thickness of Ni film on Cu(001), coercivity increases gradually with an increasing thickness of Cu.[36] The variation in coercivity is described in terms of the contribution to the magnetic anisotropy induced by the Cu overlayer via strain.[36]

We performed a detailed analysis for Cu films in the thickness range between $N = 0$ and $N = 25$ ML grown on $M = 10$ ML thick Fe films on Ag(1,1,6) and on $M = 10$ ML thick Co films on Cu(1,1,13). A minimum thickness of 10 ML Fe was chosen because below this thickness, there is a strong polar Kerr contribution to the loops due to spin reorientation transition observed at a thickness of 6 ML.[37] There is no such limitation for Co films on Cu(001) surfaces. The hysteresis loops were measured perpendicular to the easy magnetization axis, i.e., perpendicular to the steps. In all cases the loops are split, and $H_s$ can be easily derived. $H_s$ is plotted vs the thickness of the Cu film in Figs. 1 and 2 for Cu($N$)/Fe/Ag(1,1,6) and for Cu($N$)/Co/Cu(1,1,13), respectively.

For Cu($N$)/Fe/Ag(1,1,6), at RT, $H_s$ shows some dependence on the thickness of Cu($N$), which is, however, difficult to interpret as "oscillatory" (Fig. 1). Initially, i.e., up to $N \sim 6$ ML, $H_s$ increases due to a change from the Fe surface to the Cu/Fe interface contribution to $H_s$. Then $H_s$ decreases and saturates above a thickness of $N \sim 17$ ML. Interestingly, at RT, the saturated $H_s$ value is similar to $H_s$ for an uncovered Fe film of the same thickness, which means that a sufficiently

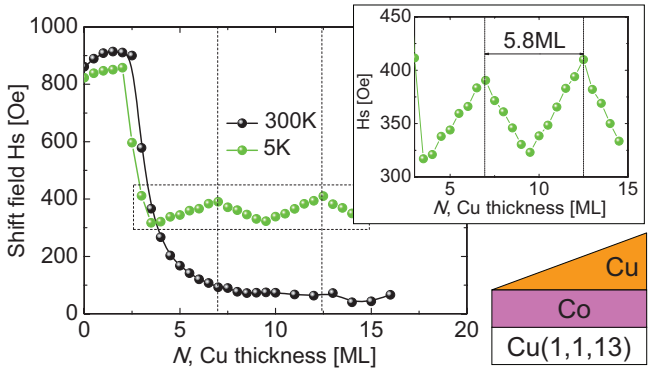


FIG. 2. (Color online) Shift field $H_S$ measured at 300 and 5 K, plotted vs Cu overlayer thickness for $M = 10$ ML of Co on Cu(1,1,13). The oscillatory behavior of $H_s$ at 5 K is more visible in the inset. The oscillation period is exactly the same as for a Cu overlayer on Fe/Ag(1,1,6), as shown in Fig. 1.

thick Cu overlayer is inert for the magnetic anisotropy of Fe on Ag(1,1,6).

The $H_s(N)$ is different if Cu is grown on top of Co/Cu(1,1,13) (Fig. 2). At a Co thickness of $M = 10$ ML, the easy axis is oriented along the steps, and $H_s$ is large (∼860 Oe).[20] With a covering of Cu, local atomic configurations at the Co surface change, which in turn influences the interface contribution to the step-induced anisotropy. As a result, Co films exhibit an easy magnetization axis still oriented along the step edges, but $H_s$ is much smaller (at RT it decreases smoothly and reaches a value of $H_s \sim 80$ Oe at ∼21 ML). This value is close to the $H_{s,vol}$ contribution to $H_s$ of Co/Cu(1,1,13). This confirms that the effect of a Cu covering is purely an interface effect resulting only from the $H_{s,surf}$ contribution to $H_s$, which is different for a Co surface and a Cu/Co interface.[11] This allows us to conclude that a large $H_s$ reported for thin Co on Cu(1,1,13)[20] is due more to the surface of Co than the Co/Cu(1,1,13) interface. At RT the transition spreads out to nearly 20 ML, which is a Cu thickness range similar to that for the Cu($N$) overlayer on Fe/Ag(1,1,6). However, the effect is not the same since a Cu covering of Co/Cu(1,1,13) results in a distinct change of $H_s$.

The $H_s(N)$ dependence for Cu on both Fe/Ag(1,1,6) and Co/Cu(1,1,13) changes remarkably with decreasing temperature (Figs. 1 and 2). More importantly, high-amplitude oscillations of $H_s$ take place, particularly at 5 K. The oscillations disappear at 50 K, which is a much faster decay of oscillations with increasing temperature than we observed previously for oscillatory magnetic anisotropy of ferromagnetic films [of Fe (Refs. 8, 21, and 23) and Co (Refs. 8 and 20); in both cases, the oscillations of $H_s$ were detectable even at 150 K] due to QWS formed in FM. It should be mentioned that the use of the wedge samples made it difficult to reproduce small-amplitude oscillations found 15 years ago for step-by-step-deposited Cu on a Co film grown on a vicinal surface of Cu(001) by MOKE measurements carried out at 170 K.[11,12] However our goal was to examine how magnetic anisotropy of ferromagnetic films behaves below 170 K (i.e., in the temperature range in which the effect of QWS formed in FM was clearly observed).[8] In order to measure $H_s$ at different temperatures wedge samples were essential, which results, however, in averaging the Kerr signal over the thickness range corresponding to the size of the laser spot. We probed the anisotropy not only of $M = 10$ ML thick Fe and Co films (Figs. 1 and 2) but also of the $M = 20$ ML thick Co film. At this thickness of Co, there are no more oscillations with increasing $N$ even at 5 K. This is because the electronic structure of $M = 20$ ML of Co on Cu(1,1,13) can be different from the electronic structure of $M = 10$ ML of Co due to the structural relaxation occurring near the thickness of 15 ML.[38]

The oscillatory dependence of $H_s$ on $N$, for both Cu($N$)/Fe/Ag(1,1,6) and Cu($N$)/Co/Cu(1,1,13), shows distinct maxima/minima (see Figs. 1 and 2). For simplicity, for Cu($N$)/Fe/Ag(1,1,6), in Fig. 1 we show only $H_s$ values measured at 5, 50, and 300 K. A similar behavior occurs for Cu($N$)/Co/Cu(1,1,13). The oscillation amplitude is, however, a bit smaller (Fig. 2). In both cases [Cu($N$)/Fe/Ag(1,1,6) and Cu($N$)/Co/Cu(1,1,13)] there are no oscillations even at 50 K, which suggests that even a small temperature spread of the Fermi-Dirac occupation function is sufficient to suppress the oscillatory character of the contribution of QWS to magnetic anisotropy.[8]

It can be observed, in particular in Fig. 1, that $H_s$ for $N = 0$ significantly decreases at 5 K with respect to its value at RT. This effect corresponds to QWS in FM, which contribute to magnetic anisotropy and modify $H_s$ at low temperatures. The change of $H_s$ with decreasing temperature at $N = 0$ is less prominent in the case of Co (Fig. 2) since the QWS contribution to the magnetocrystalline anisotropy in Co is smaller. One should keep in mind that $d$-electron QWS can be formed in Co and Fe films and can affect the electronic structure near $E_F$ for specific thicknesses.[8,21,23] Therefore, the question is whether the period of magnetic anisotropy oscillations vs the Cu thickness should be dependent on the thickness of a Co and/or Fe film or not. This question can be answered experimentally. In the case of Cu($N$)/Co($M$)/Cu(1,1,13), it is difficult to probe the oscillation period for different thicknesses of Co. This is due to the oscillation period for a Co film with no Cu($N$) overlayer on top, which is $L_{Co} = 2.3$ ML only,[20] while in reality, even for a perfect layer-by-layer growth, our thin Co films always consist of the areas which are $M$ and $(M + 1)$ ML thick. Another difficulty is a long-time-scale interdiffusion, which plays an important role for Cu/Co/Cu(1,1,13), resulting in the absence of $H_s$ oscillations a few hours after the sample is grown.[31]

The question could be more reliably answered for Cu($N$)/Fe($M$)/Ag(1,1,6). Thus, a sample of two thicknesses of Fe was grown ($M = 18$ and 22 ML). The results of the MOKE experiment are shown in Fig. 3. The oscillations are of exactly the same period, and the maxima correspond to the same thicknesses of Cu for both Fe thicknesses. However, as expected for different thicknesses of Fe, $H_s$ oscillates with respect to different "base" values. This is because $H_s$ for Fe itself is modified by the QWS formed in Fe. As seen from Fig. 3, there are specific thicknesses of Cu at which the QWS from Cu do not contribute to the magnetic anisotropy of Fe ($N = 6$ and 12 ML). However, there are also thicknesses of Cu at which QWS from Cu contribute, causing a lowering of $H_s$. These Cu thicknesses should correspond to strong hybridization of QWS in Cu with QWS in Fe while in the

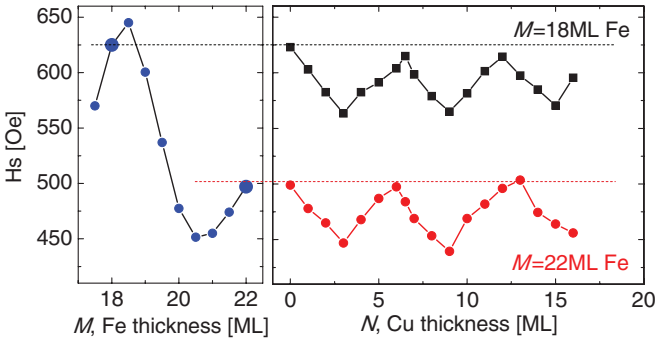


FIG. 3. (Color online) Shift field $H_S$ measured at 5 K, plotted vs Cu overlayer thickness for $M = 18$ and 22 ML of Fe on Ag(1,1,6) (right panel). Dashed horizontal lines represent $H_s$ values measured at 5 K for uncovered Fe films of the same thicknesses (left panel). This means that QWS formed in Cu modify the magnetic anisotropy of Fe films already modified by QWS formed in the Fe films.

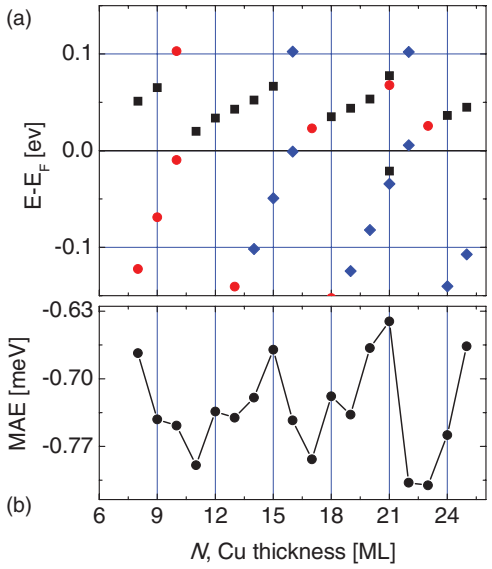


FIG. 4. (Color online) (a) Energies of minority-spin states at the $\mathbf{k}_{||} = (0.13,0)\pi/a$ point, which are localized in Co with the probability $x > 0.65$ and in Cu with the probability $0.03 < y < 0.1$ (black squares), $0.1 < y < 0.2$ (red circles), or $0.2 < y < 0.35$ (blue diamonds; note that $x + y = 1$); (b) perpendicular magnetic anisotropy energies at $T = 30$ K, both obtained for the free-standing (001) fcc Cu($N$ ML)/Co(11 ML) bilayer within the tight-binding model.

former case of the 6- and 12-ML-thick Cu overlayers such hybridization is expected to be much weaker. The oscillation amplitude seems to be almost independent of the thickness of Fe, at least in the investigated thickness range (Figs. 1 and 3).

## IV. THEORY

A thorough examination of the perpendicular magnetic anisotropy in Cu/Co bilayers using a realistic tight-binding model[19,39] reveals that the magnetic anisotropy energy oscillations with an increasing Cu thickness are mediated by the QWS formed from $d$ electrons in the Co film. This particular mechanism can be explained as follows: in a free-standing Cu slab, there exist $sp$ QWS of either spin that originate from the bulk Cu band with the $\Delta_1$ symmetry close to the $X$ point in the three-dimensional Brillouin zone.[15] However, no such minority-spin QWS, localized mainly in the Cu film, with $\boldsymbol{k}_{||} = (k_x,k_y)$ points close to $\bar{\Gamma} = (0,0)$ and energies close to or above $E_F$ (up to the top of the Co $d$ band), are present in the Cu/Co bilayer. This happens because the $sp$ states in Cu strongly hybridize with the minority-spin $d$ states (usually QWS) present in Co.[40–42] The resultant states spanning over the whole Cu/Co bilayer are localized mainly in Co but also have significant components in Cu. Within the Cu film, the probability amplitude of these states, or rather the corresponding layer- and orbital-projected probabilities (found in the tight-binding calculations), depends on the distance $z$ from the Cu surface in an oscillatory way that is characteristic for the $sp$ QWS in the Cu free-standing slabs.[41]

The phase of such QWS-like $sp$ waves localized in the Cu part of the bilayer is different at the Cu/Co interface for different Cu thicknesses while being almost fixed (thickness independent) at the Cu surface. As a result, the amplitude of these $sp$ waves at the Cu/Co interface oscillates as the Cu thickness increases. This modifies the $sp$-$d$ hybridization across this interface (i.e., between the minor $sp$-state component, localized in Cu, and the major $d$ one in Co) and thus leads to small oscillatory changes of the energies of the hybridized Co $d$ states with increasing Cu thickness; see Fig. 4(a).

The energy of the magnetic anisotropy is calculated as the difference 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 is small, it can be treated as a perturbation, and perturbation theory can be applied to calculate the energy $E(\theta,\varphi)$. For systems with reduced symmetry, like thin films (e.g., deposited on stepped surfaces), the dependence of the system energy $E = E^{(0)} + E^{(2)} + E^{(4)} + \cdots$ on spin orientation is already present in the second-order energy correction:[43,44]

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

where $f(\varepsilon)$ is the Fermi-Dirac occupation factor (dependent on temperature). Thus magnetic anisotropy energy is expressed by matrix elements of the spin-orbit interaction between occupied and unoccupied states ($|n\sigma\mathbf{k}_{||}\rangle$, $|n'\sigma'\mathbf{k}_{||}\rangle$) and their energies [$\varepsilon_{n\sigma}(\mathbf{k}_{||})$, $\varepsilon_{n'\sigma}(\mathbf{k}_{||})$], with the same or opposite spins $\sigma$, $\sigma'$ and corresponding (for flat films) to lateral wave vectors $\mathbf{k}_{||} = (k_x,k_y)$ from the whole two-dimensional Brillouin zone.

Since the energy $E^{(2)}(\theta,\varphi)$ depends on the magnetization orientation only through the matrix elements of spin-orbit coupling, different types of magnetic anisotropy energy, defined with the two corresponding magnetization directions, depend in a similar manner on the state energies through the same ratio $[f(\varepsilon_{n\sigma}) - f(\varepsilon_{n'\sigma'})]/(\varepsilon_{n\sigma} - \varepsilon_{n'\sigma'})$. Thus, it can be expected that various magnetic anisotropy energies (i.e., various anisotropy constants) vary in a similar way if the state energies change

(e.g., due to increasing film thickness). In particular, if the energy of the perpendicular anisotropy oscillates vs the film thickness due to quantum well states, similar oscillations with the same period (and the same or opposite phase) should also be expected for the energy of the step-induced anisotropy. This argument can be extended to predict the presence of similar anisotropy energy oscillations in flat films and films grown on stepped substrates (we assume similar quantum well states existing in both systems, although these QWS are somehow distorted in the film regions above the steps). This claim is supported by the theoretical predictions of oscillations with the same period of ∼2 ML for both perpendicular and step anisotropy energy in, respectively, flat and stepped fcc Co films.[19,33] Thus, it is concluded that the oscillations of the shift field $H_s$ (related to the step-induced uniaxial anisotropy energy) observed experimentally for Cu/Co and Cu/Fe on vicinal substrates can be explained by the same mechanism as the oscillations of the perpendicular anisotropy energy calculated for flat Co/Cu films [shown in Fig. 4(b)].

## V. DISCUSSION

Assuming that the effect of oscillatory magnetic anisotropy in an Fe (and Co) film is due to QWS formed in a Cu overlayer, it should occur irrespective of the material from which the ferromagnetic film is made. The period of oscillatory magnetic anisotropy $L_{Cu}$ for Cu($N$)/Co(10 ML)/Cu(1,1,13) is exactly the same as that observed for Cu($N$)/Fe(10 ML)/Ag(1,1,6) and is equal to $L_{Cu} = 5.8 \pm 0.2$ ML. The changes in magnetic anisotropy with an increasing thickness of Cu fit perfectly to the results of the inverse-photoemission experiment carried out for Cu films grown on both Fe(001) and Co(001) single-crystalline substrates.[14–17] In both cases the maxima of the density of *sp* states at the Fermi level were detected with the same periodicity (i.e., every $L_{Cu} = 5.8 \pm 0.2$ ML) and also for the same thicknesses of Cu.[14–17] This clearly suggests that the QWS formed in Cu are responsible for the oscillatory magnetic anisotropy of the ferromagnetic (Fe or Co) film underneath. However, the mechanism relating *sp* QWS in Cu and the magnetic anisotropy of Fe (or Co) is complex, as explained in the previous section and discussed in more detail below.

In view of the second-order perturbation theory, each pair of the occupied and unoccupied $d$ states of the energies close to the Fermi level can contribute to the total magnetic anisotropy [Eq. (1)]. The occupied (unoccupied) quantum well state shown in Fig. 4(a) can contribute strongly to the anisotropy energy if coupled to any other unoccupied (occupied) electronic state of the energy close to $E_F$. The energy dispersion with increasing Cu film thickness is usually very weak for the latter states since they are mainly $d$ states localized almost entirely in the Co film. Thus the energy distance between both coupled states decreases when the energy of QWS approaches $E_F$. After crossing $E_F$, the QWS becomes unoccupied but can still couple, now to the occupied states. With increasing film thickness, the QWS shifts to higher energies. Thus the energy distance between both coupled states increases. It results in a maximum contribution to MAE for the thicknesses at which QWS cross $E_F$ (i.e., at which the energy distance between the coupled states is minimum). This is exactly what is shown in Fig. 4(b): for the energies of the quantum well state close to the Fermi level the MAE values are the largest, but negative. They are negative because MAE is calculated as the difference between the energy of the system for two different magnetization orientations (a negative MAE means that the magnetic energy of the system for the second orientation is larger than for the first one). The oscillatory contribution is expected only if the $d$ QWS in Co is distant from $E_F$ by less than the magnitude of its energy changes due to the varying Cu thickness. Since such an energy range is found to be minor, the magnetic anisotropy oscillations arise in small regions in the two-dimensional Brillouin zone near the $\boldsymbol{k}_{||}$ points at which QWS in Co cross $E_F$. This explains the small amplitude of the total magnetic anisotropy energy oscillations found in experiments for Cu/Co films on Cu vicinal substrates as well as in the tight-binding calculations for flat Cu/Co bilayers [Fig. 4(b)]. Thus the oscillations can disappear even with a small increase in temperature (i.e., if thermal fluctuations become comparable to the energy changes due to QWS in Cu). A similar mechanism for the magnetic anisotropy oscillations with increasing Cu thickness should also be valid for Cu/Fe bilayers. A possible extension of that is when *sp* QWS in Cu can hybridize with the QWS of either spin in Fe.

As mentioned in Sec. IV, it is not contradictory that the perpendicular anisotropy is calculated, whereas $H_s$ (which is a measure of the uniaxial step-induced anisotropy) is measured. All the terms contributing to the total magnetocrystalline anisotropy energy are related to the same electronic structure of the system (thin film, double layer, etc.), and they are calculated following the same approach [Eq. (1)]. Thus, all contributions to the total anisotropy energy which oscillate with thickness oscillate equally (i.e., with the same period, except there is no contribution from QWS due to symmetry reasons). In particular, it concerns the uniaxial in-plane anisotropy contribution which is directly related to $H_s$ measured in our experiment. For the same reasons, the oscillation amplitude should decay equally with increasing temperature for both the perpendicular and the uniaxial anisotropies. The temperature dependence is governed by the Fermi-Dirac occupation factors [Eq. (1)] of the same quantum well states contributing to both the perpendicular and the uniaxial anisotropies. Thus, one can say that the oscillations of $H_s$ decaying with increasing temperature reflect the behavior of perpendicular anisotropy, in agreement with theoretical predictions. A strong temperature dependence of the QWS effect on magnetic anisotropy, due to the QWS formed in ferromagnetic films, was previously reported for Fe and Co films on Ag(1,1,6) and Cu(1,1,13), respectively.[8,21,23] More generally, other oscillatory phenomena due to QWS, such as interlayer exchange coupling across a nonferromagnetic layer separating two ferromagnetic films, are strongly temperature dependent.[45] However, the effect is ascribed more to the thermal excitations of spin waves in ferromagnetic films/slabs, particularly at their interfaces, reducing the interlayer exchange.[46]

The oscillation period $L_{Cu} = 2\pi/(k_{z0}a)$ (expressed in ML), where $k_{z0} = k_{z0}(\boldsymbol{k}_{||})$ is found theoretically from the condition $\epsilon_b(\boldsymbol{k}_{||},k_z) = E_F$ with the energy $\epsilon_b$ of the *sp* states in bulk Cu (the dependence of $k_{z0}$ on $\boldsymbol{k}_{||}$ is discussed in Ref. 47). It is identical to the period with which QWS (with such $\boldsymbol{k}_{||}$) appear regularly at $E_F$ in a free-standing Cu slab as the Cu thickness

increases. Since the period $L_{Cu}$ is calculated for the $\boldsymbol{k}_{||}$ points at which the $d$ QWS in FM cross $E_F$ for the Cu/Co bilayer, it may be dependent on the ferromagnetic film thickness. Such dependence is found in the tight-binding calculations for the Cu/Co bilayer where the perpendicular magnetic anisotropy energy is predicted to oscillate with periods considerably smaller than 5.8 ML for Co film thicknesses corresponding to $d$ QWS crossing $E_F$ at $\boldsymbol{k}_{||}$ located relatively far from $\bar{\Gamma}$ (e.g., at 1/3 of the $\bar{\Gamma}$-$\bar{M}$ distance for $M = 10$ ML, which leads to a period of ∼4 ML). However, such dependence was difficult to confirm experimentally (see Sec. III). This can be explained by the above-mentioned fact that in reality, our Co($M$) films can consist of areas which are $M$ and $(M + 1)$ ML thick. In such a case, the dominating period close to 5.8 ML can arise from the film regions with thicknesses of $M + 1$ for which QWS cross $E_F$ at $\mathbf{k}_{||}$ close to $\bar{\Gamma}$, while the magnetic anisotropy oscillations with a shorter period coming from the regions with a thickness of $M$ are attenuated. This is because the lifetime for QWS of finite $\boldsymbol{k}_{||}$ is shorter than for $\boldsymbol{k}_{||} = 0$; their energy spread can be larger than the energy change due to the varying film thickness.

For Cu/Fe bilayers, the oscillation period is expected to be nearly independent of the Fe thickness and close to 5.8 ML since there exist QWS in Fe which cross $E_F$ at the $\boldsymbol{k}_{||}$ points close to $\bar{\Gamma}$. This is related to the weak $k_z$ dispersion (near $E_F$) of the majority-spin $d$ bulk band of $\Delta_5$ symmetry which gives such QWS in Fe. The independence of the oscillation period from the thickness of Fe is confirmed experimentally (see Sec. III, Fig. 3).

Finally, it is not surprising that the oscillations of magnetic anisotropy detected experimentally at 5 K are dominated only by one electron wavelength originating from $\boldsymbol{k}_{||} = (0,0)$ (and resulting in $L_{Cu} \sim 6$ ML). QWS with a shorter periodicity ($L_{Cu} \sim 2.3$ ML), originating from $\boldsymbol{k}_{||} = (0.805,0.805)\pi/a$, corresponding to the neck of the Fermi surface of Cu,[48] can be more effectively reflected at the Cu/Co (or Cu/Fe) interfaces. Thus there is no strong hybridization between these QWS and the $d$ states of Co (Fe).[49]

## VI. SUMMARY

In summary, we studied the effect of QWS in a Cu overlayer on the step-induced in-plane magnetic anisotropy of Fe and Co films [grown on Ag(1,1,6) and Cu(1,1,13), respectively]. An oscillation of the uniaxial anisotropy is observed at low temperatures (5–30 K), with a period of $L_{Cu} = 5.8 \pm 0.2$ ML. Such a quantum oscillation is attributed to the QWS in a $sp$ band of $\boldsymbol{k}_{||}$=(0,0) at $E_F$ in Cu strongly hybridized with the QWS formed by the $d$ electrons in Fe and Co films. Magnetic anisotropy oscillations with the shorter 2.3 ML period corresponding to QWS originating from the neck of the copper Fermi surface are not found, either in low-temperature experiment or theory calculations. The oscillation period of $L_{Cu} = 5.8 \pm 0.2$ ML fits perfectly to theory.

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*mprzybyl@mpi-halle.de

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