

Experimental confirmation of quantum oscillations of magnetic anisotropy in Co/Cu(001)U. Bauer,^{1,*} M. Dąbrowski,¹ M. Przybylski,^{1,2,†} and J. Kirschner¹¹Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany²Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, al. Mickiewicza 30, PL-30-059 Kraków, Poland

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The effect of quantum well states on the magnetic anisotropy of Co films grown on vicinal Cu(001) substrates was studied by *in situ* magneto-optic Kerr effect in a temperature range of 5 K to 365 K. The uniaxial magnetic anisotropy is found to oscillate as a function of Co thickness with a period of 2.3 atomic layers, modulated exactly as theoretically predicted by L. Szunyogh *et al.* [*Phys. Rev. B* **56**, 14036 (1997)] and independently by M. Cinal [*J. Phys. Condens. Matter* **15**, 29 (2003)] and M. Cinal and A. Umerski [*Phys. Rev. B* **73**, 184423 (2006)]. Sub-monolayer Au coverage was used to fine-tune magnetic anisotropy and to provide convenient experimental access to anisotropy oscillations. The anisotropy oscillations, which are present up to room temperature, are attributed to quantum well states in the minority *d* band at the Fermi level of Co.

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I. INTRODUCTION

In dimensionally reduced magnetic systems, such as ultrathin films, electrons can be confined perpendicular to the film plane and form quantum well states (QWSs).⁴⁻⁶ The formation of QWSs can directly alternate the electronic structure at the Fermi level and therefore result in oscillatory physical properties such as magnetic anisotropy.^{2,3,7-11}

For (001) face-centered-cubic (fcc) Co films, theory predicts oscillations of the magnetocrystalline anisotropy energy (MAE) as a function of film thickness.^{1,2} Szunyogh *et al.*¹ performed layer-resolved *ab initio*-like calculations for Co slabs consisting of *N* atomic layers (monolayers, ML). Their results showed oscillations of the layer-resolved MAE for fixed film thickness as well as oscillations of the MAE averaged over the Co film thickness. In the latter case the calculations yielded oscillations with a clear period of 2 ML.

Independently, Cinal² examined the role QWSs play in purely ferromagnetic (FM) systems such as *N* ML of Co on Cu(001). A careful analysis with a parametrical tight-binding (TB) model revealed that the total MAE oscillations in Co(*N*)/Cu(001) are a superposition of two oscillatory contributions: the dominating one coming from the neighborhood of the Γ point with a period of 2.12 ML and the other originating in the region around the *M* point with a larger period of 5.15 ML, but of significantly smaller amplitude. Both oscillatory contributions are clearly attributed to QWSs formed in the FM Co layer. Later, the calculations were extended to Co films on vicinal Cu(001), where the uniaxial magnetic anisotropy was found to oscillate with a period close to 2 ML, as well.³

These predictions, made nearly 15 years ago, have so far not been confirmed experimentally. This is not surprising since oscillations with a period of 2 ML can be easily diminished if the vertical amplitude of surface roughness is large. Moreover, the QWSs responsible for the anisotropy oscillations correspond to small in-plane wave vectors *k*, and thus require flat surfaces over large areas to propagate well.² Up to now, anisotropy oscillations induced by QWSs in a FM film were experimentally observed only for Fe films grown on vicinal surfaces of Ag(001) at temperatures below 200 K.⁹⁻¹¹ An oscillation period of 5.9 ML was found but could not be verified by theory which is not available for this system.

The Co(*N*)/Cu(001) system is the only system for which oscillatory magnetic anisotropy due to QWSs in FM layers has been investigated by theory and therefore allows direct comparison to experiments. Furthermore, anisotropy oscillations are predicted to be present even at room temperature (RT),² which makes this system particularly interesting from a technological point of view.

In this article we report on the experimental confirmation of magnetic anisotropy oscillations due to QWSs in fcc Co films. In order to be sensitive to small changes of the anisotropy, the fourfold anisotropy of Co films grown on Cu(001)^{12,13} was modified by growing the films on Cu(1,1,13) vicinal substrates.^{7,8,14} Changes of both uniaxial in-plane and perpendicular anisotropy were followed for uncovered and Au-covered films between 5 K and 365 K. In particular, the oscillation periods were derived and compared to theoretical predictions.

II. EXPERIMENTAL DETAILS

The experiments were performed in a multichamber ultra-high vacuum system with a pressure below 2×10^{-10} mbar during Co deposition. A Cu(1,1,13) substrate [6.2° off the (001) surface] with the step edges along the $[\bar{1}10]$ direction was used. The vicinal Cu(001) substrate was prepared with cycles of 1 keV Ar ion sputtering and subsequent annealing at ~ 600 °C. Sharp split diffraction spots were detected in low-energy electron diffraction (LEED). Using scanning tunneling microscopy (STM), monoatomic regular steps along the $[\bar{1}10]$ direction with an average terrace width of ~ 2 nm were observed. Co films were grown at 190 K by molecular beam epitaxy in wedge shape with a small slope of ~ 1.6 ML/mm along the $[110]$ direction. After growth, the films were warmed up to RT in order to improve surface morphology, and only then covered with Au. Magnetic properties were probed by the *in situ* longitudinal magneto-optical Kerr effect (MOKE) with a laser diode (wavelength 670 nm, incidence angle 21°, and beam diameter < 0.2 mm).

Epitaxial growth of Co films on vicinal Cu(001) substrates results in a step-induced uniaxial anisotropy with the easy magnetization axis oriented along or perpendicular to the step

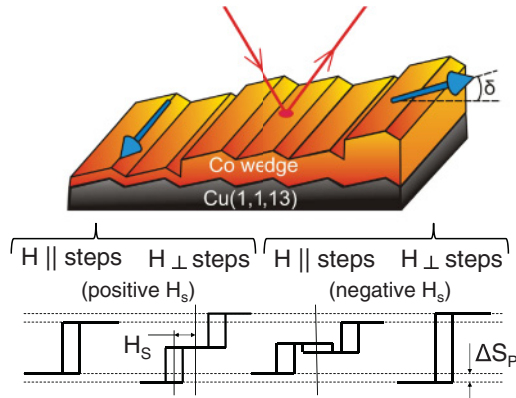


FIG. 1. (Color online) Schematics of Co wedge on Cu(1,1,13) with easy magnetization axis [thick (blue) arrows] oriented parallel or perpendicular to the steps. Thin (red) arrows represent incoming and outgoing laser beam, and H is the magnetic field. Representative hysteresis loops for longitudinal MOKE measurements for the easy magnetization axis oriented either parallel (left panel) or perpendicular (right panel) to the steps are also shown. ΔS_p denotes additional polar MOKE signal due to tilting of the easy axis relative to the sample plane by angle δ .

edges. If the steps are oriented parallel to one of the easy axes of the fourfold anisotropy, the two easy axes become nonequivalent. In this case so-called split hysteresis loops are measured when the magnetic field is applied along the harder of those two axes.^{9,15,16} These hysteresis loops are characterized by a shift field H_s which is a measure of the uniaxial anisotropy introduced by the steps. Positive or negative H_s refers to the situation where the easy magnetization axis is oriented along or perpendicular to the steps, respectively (see Fig. 1). Perpendicular to the steps, the easy axis can be tilted relative to the film plane by an angle δ which gives rise to the additional polar MOKE signal ΔS_p . If the easy magnetization axis is oriented perpendicular to the steps and hysteresis loops are measured along the steps, this perpendicular magnetization component can result in additional features at low fields (Fig. 1).¹⁶

III. RESULTS

For uncovered Co films on vicinal Cu(001) it is known that at RT, the easy magnetization axis is initially oriented parallel to the steps and, after an initial small rise, the shift field H_s decreases with increasing Co thickness (N) (compare Fig. 2). In particular, an abrupt decrease of H_s occurs around a thickness of strain relaxation at 15 ML,¹³ which is accompanied by a switch of the easy magnetization axis to perpendicular to the steps and a saturation of H_s above 30 ML of Co.¹³

Covering Co films with a nonmagnetic material such as Au changes the local atomic configuration at the Co surface and modifies the interface contribution to the step-induced anisotropy.^{9,10,17,18} In the case of Au, the interface contribution becomes negative (i.e., forces the magnetization to be oriented perpendicular to the steps) but, as expected, the volume contribution (due to structural distortion of the film volume above the steps) remains unchanged.¹⁰ Figure 2 illustrates

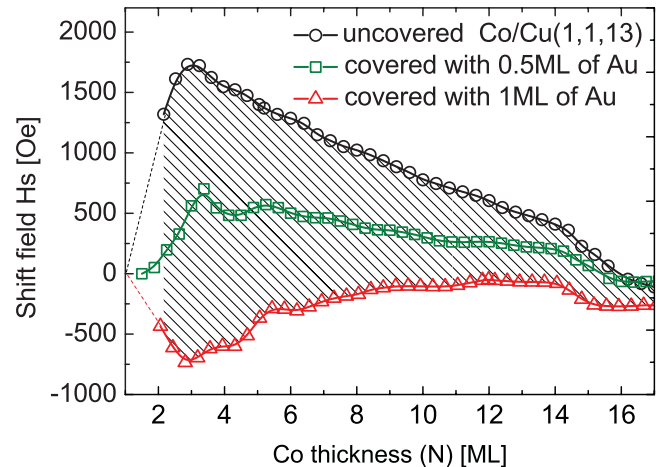


FIG. 2. (Color online) Shift field H_s measured at RT vs Co thickness for Co/Cu(1,1,13) covered by different amounts of Au. The dashed area illustrates the values of H_s which are accessible by sub-monolayer Au coverage.

how the dependence of H_s on N evolves with sub-monolayer Au coverage for Co films grown on Cu(1,1,13). The shaded area represents the values of H_s which are accessible by Au coverage between 0 and 1 ML. By carefully depositing minute amounts of Au on uncovered Co films, the magnitude of H_s can be significantly reduced and brought close to $H_s = 0$ over a wide thickness range (e.g., for 1 ML Au coverage between 8 and 14 ML Co). This offers two advantages: It results in a much higher sensitivity to small variations in H_s in the MOKE experiment and allows changing the easy magnetization axes by small variations in H_s .

Independent of the Au thickness, the system always shows in-plane magnetization in agreement with theoretical predictions for Au/Co/Cu(001).¹⁹ The H_s dependence on N (Fig. 2) always starts from $H_s = 0$ at $N = 0$ and reaches a maximum absolute value at $N = 3$ ML (independent of the orientation of the easy magnetization axis). This reflects the formation of continuous Co films up to $N = 3$ ML and well-developed bottom and upper interfaces only above this thickness. Note that above the maximum at 3 ML of Co the height of the dashed region (i.e., the range of accessible H_s) in Fig. 2 scales as $1/N$ which confirms that covering with Au introduces an additional term to the $H_s(N)$ dependence, which is of interface origin.

Since the anisotropy oscillations are expected to be strongest at low temperature,^{2,9,10} we first investigate the dependence of H_s on N for Co/Cu(1,1,13) covered by 0.5 ML Au at $T = 5$ K. Here, $H_s(N)$ still shows resemblance to the dependence at RT with the same maximum at 3 ML; however, it oscillates with increasing Co thickness with a period of 2.3 ± 0.3 ML (Fig. 3). The oscillation amplitude below 15 ML of Co is about 300 Oe, i.e., almost two orders of magnitude larger than the anisotropy oscillations caused by QWSs in the Cu overlayer in the Cu/Co/Cu(001) system.⁸ This clearly confirms the volume character of the observed anisotropy oscillations which are due to QWSs propagating in the Co film. Note that the same oscillatory behavior is observed for uncovered Co/Cu(1,1,13) but is less prominent due to the overall much higher magnitude of H_s (compare Fig. 2). Since

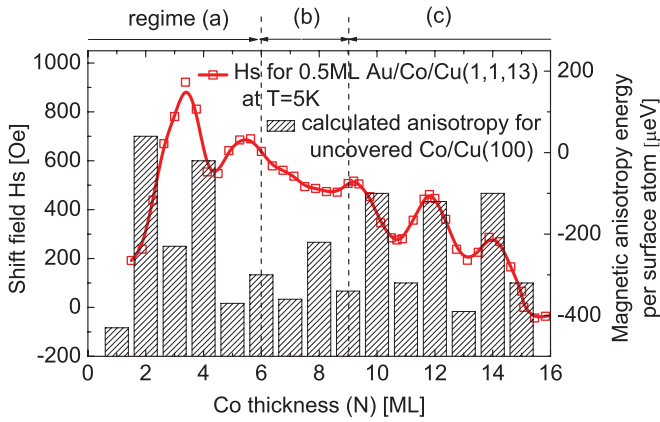


FIG. 3. (Color online) Shift field H_s measured at 5 K vs Co thickness for Co/Cu(1,1,13) covered with 0.5 ML of Au. Experimental data are compared to the band energy contribution to the magnetic anisotropy energy calculated per surface atom by Szunyogh *et al.*¹

covering with Au does not change the volume contribution to the uniaxial anisotropy, it does not meaningfully impact the oscillation amplitude.^{3,10} In Fig. 3, the experimental data are compared to the band energy contribution to the MAE calculated per surface atom by Szunyogh *et al.*¹

In the case in which the Co film is covered by 1 ML of Au, the magnitude of H_s is reduced even further and stays close to $H_s = 0$ between 8 and 14 ML of Co (compare Fig. 2). Figure 4 shows representative hysteresis loops for different Co thicknesses, measured at 5 K with the magnetic field H applied either parallel or perpendicular to the steps. It can be seen that due to the reduced magnitude of H_s , the oscillation amplitude

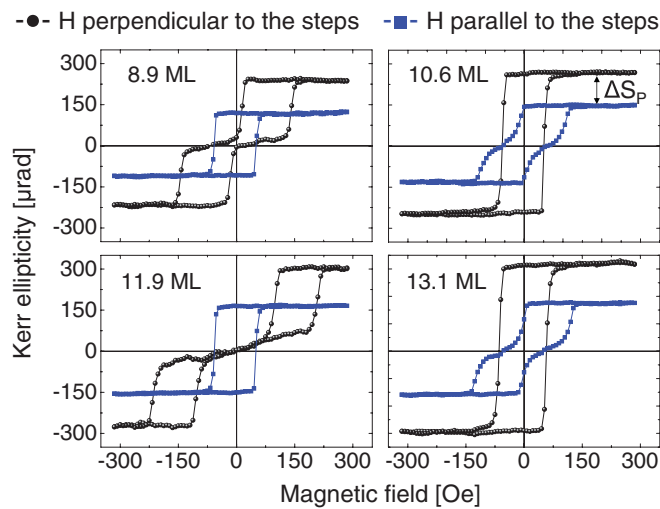


FIG. 4. (Color online) Representative hysteresis loops at different Co thicknesses N for Co/Cu(1,1,13) covered by 1 ML Au measured at 5 K. Measurements were performed under a magnetic field H applied perpendicular [(black) circles] and parallel [(blue) squares] to the steps. The hysteresis loops show that the easy magnetization axis oscillates as a function of Co film thickness. The difference in saturation Kerr signal of Co for perpendicular and parallel geometries is due to the additional polar MOKE signal ΔS_p which contributes only if the magnetic field is applied perpendicular to the steps (compare Fig. 1).

of H_s is now sufficiently large to change the orientation of the easy magnetization axis several times with increasing Co film thickness N . For $N = 8.9$ and 11.9 ML the hysteresis loops with H applied parallel to the steps show square shape whereas the loops obtained with H applied perpendicular to the steps are split. For $N = 10.6$ and 13.1 ML the loops measured with H parallel to the steps are split whereas the loops measured with H perpendicular to the steps show square shape. The difference in longitudinal Kerr signal in saturation of the hysteresis loops measured with H applied perpendicular or parallel to the steps is due to the additional polar MOKE signal ΔS_p which contributes only if the magnetic field is applied perpendicular to the steps (compare Fig. 1).^{10,11,18,20} This is due to the competition between shape, in-plane, and perpendicular magnetic anisotropy which tilts the magnetization off the sample plane by an angle δ . The value of ΔS_p increases more-or-less linearly with increasing Co thickness and, in agreement with previous experimental results,²⁰ corresponds to a small δ of $\sim 2^\circ$.

After a careful inspection, we find that independently of the Au coverage, there is a low-amplitude oscillation with a period of 2.3 ± 0.3 ML superimposed on the linear dependence of the saturation signal on N if measured perpendicular to the steps at 5 K. Thus, ΔS_p oscillates with N (Fig. 5) and shows a regular amplitude corresponding to a variation of δ of $\sim 1^\circ$. Interestingly, the maxima of ΔS_p are not in phase with the maxima of H_s (Fig. 3). There is a phase difference of about $+\pi/2$ in regime (a) and of about $-\pi/2$ in regime (c). Additionally, ΔS_p shows pronounced maxima and minima even in the thickness range where H_s does not oscillate [i.e., regime (b)]. This implies that the observed oscillation of ΔS_p is not a simple consequence of the oscillatory step-induced in-plane anisotropy, but originates from the perpendicular magnetic anisotropy.

The anisotropy oscillations disappear with increasing N and are not observed above 22 ML. This can be due to structural quality (rougher surface with increasing film thickness) and/or electronic origin (the energies of the QWS pairs move closer together with increasing thickness). In order to investigate the influence of structural quality on the anisotropy oscillations, two identical samples were grown on Cu(1,1,13). One was

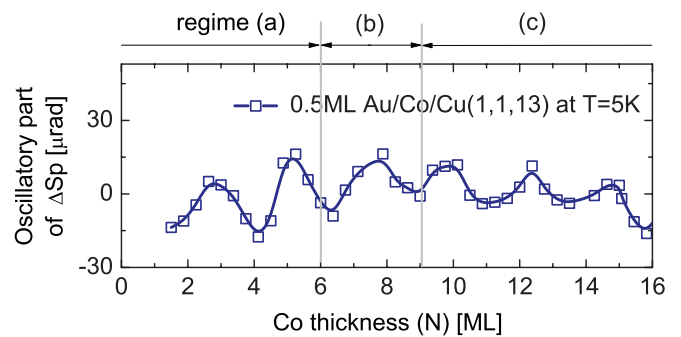


FIG. 5. (Color online) Oscillatory part of ΔS_p obtained from the saturation signal measured vs. Co thickness for Co/Cu(1,1,13) covered by 0.5 ML Au. Measurements were performed at 5 K with the magnetic field applied perpendicular to the steps. ΔS_p is the additional polar MOKE signal which results from the tilting of the easy magnetization axis with respect to the film plane.

grown at 190 K, the other at RT, and both were covered with Au at RT. It can be expected that a higher growth temperature results in more interdiffusion at the Co/Cu interface and therefore more interfacial roughness. At RT, the $H_s(N)$ characteristics of both samples are very similar (not shown here), but not exactly the same. This is most likely due to a slightly different mode of growth, and finally not exactly the same uniaxial magnetic anisotropy. Moreover, conditions to create QWSs are not equally fulfilled in both cases. Nevertheless, at $T = 5$ K, anisotropy oscillations are observed for both samples. The difference in growth temperature does not change the position of the maxima of H_s ; however, it influences the oscillation amplitude strongly. The sample grown at RT exhibits an oscillation amplitude reduced by a factor of ~ 3 . This observation is in agreement with interlayer exchange coupling experiments in Co/Cu(N)/Co(001), which showed that increased interfacial roughness does not significantly change the phase of the QWSs (i.e., does not modify the coupling peak positions) but that it can significantly decrease the oscillation amplitude.⁵

IV. DISCUSSION

Comparison to theory of Szunyogh *et al.*¹ shows an agreement with the data for all the investigated systems, in particular for N ML of Co on Cu(001) uncovered (shown in Fig. 3) and covered with Cu. The calculated oscillation period is 2 ML. There are three different regimes of oscillations visible in both experiment and theory: (a) for $N < 6$, with a pronounced maximum at 3 ML and 5.5 ML; (b) $6 < N < 9$, with no clear maxima; and (c) $N > 9$, with three distinct maxima at about 9, 12, and 14 ML. The agreement is almost perfect in this case, except for a large anisotropy energy calculated for 2 ML, but not observed experimentally. This is most likely due to the growth of Co on Cu(001), which is initially not layer-by-layer as is assumed for theoretical calculations. The different oscillation amplitude in regimes (a), (b), and (c) can be understood as a consequence of adding the band energy contributions to the anisotropy energy from all the atomic layers forming the film.¹ The lack of clear oscillations in regime (b) is attributed to a successive dying out of interface-interface interactions.¹ In view of QWSs, this is due to the fact that the energies of QWS pairs are not necessarily close to E_F in this thickness range and therefore do not contribute significantly to the MAE.²

It is expected that anisotropy oscillations vanish with temperature when the spread of the Fermi function ($\sim 4 k_B T$) becomes comparable to the energy difference between the two states of each QWS pair contributing to the MAE.² This contribution is large only for those QWSs which are close to E_F at $\bar{\Gamma}$ [i.e., at $k = (k_x, k_y) = 0$]. Thus, a strong dependence on temperature should be expected only for those N for which the QWSs contribute strongly to the anisotropy. Calculations by Cinal² and Cinal and Umerski³ predict this effect for $N = 9, 11, 13$, and 15 ML, but not for $N = 10, 12$, or 14 ML. To investigate the temperature dependence of $H_s(N)$, a careful MOKE analysis within the Co thickness range of 8 to 18 ML was performed (see Fig. 6). Note that for this experiment the sample was covered with 1 ML of Au to further reduce the magnitude of H_s (compare Fig. 2). In excellent

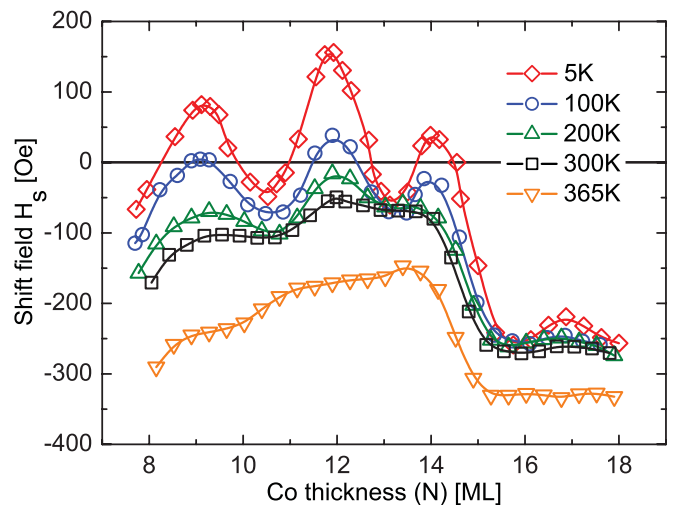


FIG. 6. (Color online) H_s as a function of Co thickness in regime (c), measured at different temperatures, for Co/Cu(1,1,13) covered by 1 ML of Au. The amplitude decreases with increasing temperature, but oscillations are still visible even at RT.

agreement with theory, the anisotropy at $N = 9.3, 11.9, 14.0$, and 16.6 ML depends strongly on temperature, whereas for $N = 10.6, 13$, and 15.6 ML the anisotropy changes only little. The oscillations do not disappear with temperature as strongly as was observed for Fe films on Ag(001).^{9,10} In agreement with theory,^{2,3} the anisotropy oscillations are still visible at RT and, e.g., at $N = 11.9$ ML show an amplitude of ~ 60 Oe (reduced from ~ 210 Oe at 5 K). Nevertheless, the amplitude decreases quickly above RT and the oscillations vanish at 365 K. This means that for $N = 9.3, 11.9, 14.0$, and 16.6 ML, at $T = 365$ K, the spread of the Fermi function becomes comparable to the energy difference between the two states of each QWS pair contributing to the MAE.

Szunyogh *et al.* calculated a period of 2 ML¹ and Cinal predicted periods of 2.12 ML and 5.15 ML² for anisotropy oscillations. The origin of the oscillation period of 2.12 ML is the intersection of the minority spin d band and the Fermi level at $k = 0.528 \frac{2\pi}{a}$ in the parametric TB model.² This is in good agreement with recent relativistic band structure calculations which yield $k = 0.505 \frac{2\pi}{a}$ and a corresponding oscillation period of 2.02 ML.²¹ The distance between the experimentally observed maxima is not constant and varies from 2.1 to 2.6 ML. This variation might be due to the non-perfect layer-by-layer growth and proximity to the structural transition at $N = 15$ ML, which was not considered in the theoretical calculations. Note that the oscillation amplitude at $N = 11.9$ ML is larger than at $N = 9.3$ and 14.0 ML. This can be due to beating since the oscillation period is not exactly equal to 2 ML. The lack of a clear oscillation with a period length of 5.15 ML is attributed to the significantly smaller amplitude of this contribution to the MAE.²

The calculated oscillation amplitude of the MAE is $\sim 250 \mu\text{eV}$ per surface atom at 0 K¹ and $\sim 150 \mu\text{eV}$ per surface atom at RT.² Also, a very similar result of $\sim 140 \mu\text{eV}$ was obtained per step atom for the Co(N)/vicinal-Cu(001) system at RT.³ In our experiment we follow the step-induced uniaxial anisotropy, which is only locally (i.e., at the steps) introduced

to the Co film,¹⁰ and thus our results should be considered per step atom. Accordingly, the change of the anisotropy energy corresponding to the experimentally observed H_s oscillation amplitude of maximally ~ 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.

V. SUMMARY

In summary, we studied the uniaxial in-plane and perpendicular magnetic anisotropy in Co films grown on Cu(1,1,13). Both oscillate as a function of Co thickness with a period of 2.3 ± 0.3 ML, even at RT. The oscillation period and

amplitude modulation are in excellent agreement with theoretical predictions¹⁻³ and are attributed to QWSs in the minority d band at E_F of Co. We find that sub-monolayer Au coverage is an effective tool to separate the anisotropy oscillations from the background of the step-induced uniaxial anisotropy. Film growth at low temperature significantly enhances the oscillation amplitude.

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