One monolayer-periodic oscillations in the magnetization induced second harmonic generation signals during the growth of Co films on Cu(001)

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Magnetization induced optical second harmonic generation is used to investigate the surface magnetic properties during the layer-by-layer growth of Co films on Cu(001). The second harmonic (SH) signal changes as a function of Co thickness and an oscillatory behavior of the SH asymmetry with one monolayer period is observed for the transverse Kerr-geometry and *s*-polarized incident light, while the average SH signal has no such oscillations. The magnetization induced SH signal at half filled layers is enhanced with respect to that of filled layers, which agrees with the enhancement of magnetic moment of edge atoms and the higher step density at half filled layers. Its dependences on the growth rate and temperature are discussed.
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I. INTRODUCTION

The magneto-optical Kerr effect has been considered as a fruitful and powerful technique to study the magnetism of multilayers and ultrathin films. In the past few years after the first theoretical paper by Pan, Wei, and Shen[1] and experimental work by Reif *et al.*,[2] nonlinear optics has been introduced into the study of magnetic properties of ultrathin films,[3–11] which is called magnetization induced second harmonic generation (MSHG).

It is well known that in materials with inversion symmetry the second harmonic generation (SHG) is generated only at the surface and buried interfaces, where the inversion symmetry of the electronic density is locally broken. The presence of magnetization in materials does not break the inversion symmetry of the bulk, but lowers the symmetry at surfaces/interfaces resulting in more nonzero tensor elements of the tensor of second order susceptibility $\chi^{(2)}$. Therefore MSHG is surface/interface sensitive and has been used to study the magnetism of surface and interfaces. It has been shown that the thickness sensitivity is 1 to 2 monolayers (ML).[6,8]

The dimensional effect on the magnetism of ultrathin films has been widely investigated.[13–15] It is generally confirmed that the magnetic moment in ferromagnets like Fe, Co, and Ni is enhanced with reduced number of nearest neighbors due to the decrease of the bandwidth.[16,17] It was found that the magnetic moment of a free surface is enhanced with respect to that of the bulk, and that a rough surface may have a larger magnetic moment than a smooth surface. The early work of Fe(110) on W(110) by Albrecht *et al.*[17] showed that the magnetic moment of a real step atom was enhanced by roughly $0.5\mu_B$ with respect to the atomic moment in a smooth surface. The calculation for 2.5 ML Co on Cu(100) gave an enhancement of about $0.1\mu_B$ of magnetic moment for the atom at the step site with respect to the 3 ML thick film.[18]

The growth of Co on Cu(001) at room temperature follows a layer-by-layer mode. Co islands periodically nucleate, grow in size, and then coalesce, giving rise to oscillations of the average step density during the growth. Consequently a periodically oscillating magnetic moment of the topmost layer of the film is expected.

Here we report on 1 ML period oscillations of the magnetization induced second harmonic (SH) signals during the growth of Co on Cu(001). From the measured data, we derive that the magnetization induced surface component of $\chi^{(2)}$ is enhanced at half filled layers, which agrees with the discussion above.

II. EXPERIMENT

The Co films were grown on Cu(001) in a molecular beam epitaxy (MBE) apparatus with a base pressure of less than 4×10^{-11} mbar. A growth rate of 7 ML/h was used at room temperature for the study of the 1 ML periodic oscillations in MSHG, while other growth rates and substrate temperatures were chosen for comparison experiments.

For the MSHG measurement, the light pulses from a Ti:sapphire laser ($\lambda=840$ nm, 80 fs pulse width) was focused onto the sample in the MBE chamber. Only experimental results with *s*-polarized incident light and transversal geometry were discussed in this article. The optical plane was parallel to the $\langle\bar{1}10\rangle$ azimuthal direction of the Cu crystal, and the external magnetic field of 70 Oe was applied to magnetically saturate the film along the $\langle110\rangle$ direction, the easy axis of magnetization. During the growth of the Co film, the SH light generated in the sample was detected by a photomultiplier and recorded for the two opposite directions of magnetization.

III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the SH intensities $I_\uparrow(2\omega)$ and $I_\downarrow(2\omega)$ as a function of the Co film thickness for the two opposite magnetization directions. The SH signal from the pure Cu substrate is very small. After an initial decrease, the intensities increase very rapidly until at about 2 ML they reach a

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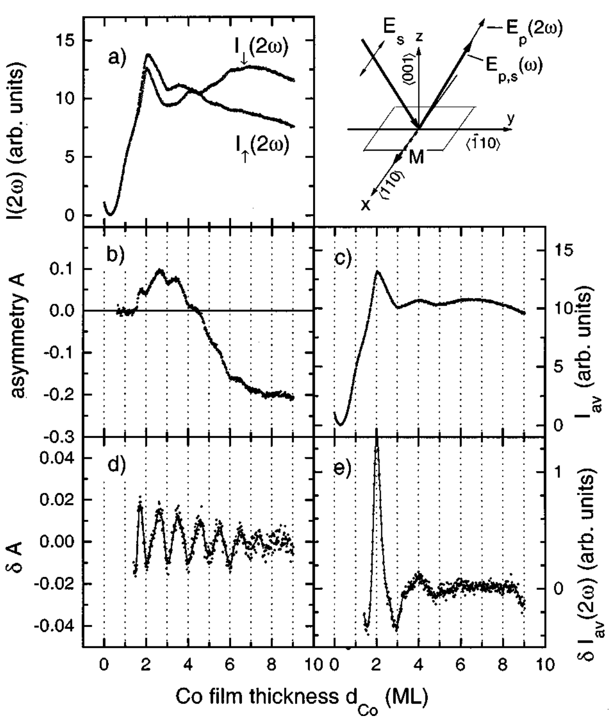

FIG. 1. (a) Measured total SH intensity as a function of the Co film thickness for *s*-polarized incident light in the transversal Kerr geometry for the magnetization in opposite directions. (b) Magnetic asymmetry and (c) average SH intensity calculated from the SH intensities in (a). In (d) the difference of the magnetic asymmetry and its running average over 50 points is plotted. In (e) the similar difference as in (d) is plotted for average SH intensity I_{av}.

maximum. The curves for $I_\uparrow(2\omega)$ and $I_\downarrow(2\omega)$ separate at about 1.5 ML where the Co film begins to order ferromagnetically.

In Fig. 1(b) the magnetization asymmetry $A=(I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow)$ calculated from the SH intensities in Fig. 1(a) is plotted, where oscillations with a period of 1 ML are clearly seen as a function of the Co film thickness, superimposed on top of a smooth curve. In the transverse geometry the magnetic asymmetry can be written as[5,12]

$$A=\frac{2x}{1+x^2}\cos\varphi, \tag{1}$$

where $x=\chi^{(2)}_{\mathrm{mag}}/\chi^{(2)}_{\mathrm{nonmag}}$, $\chi^{(2)}_{\mathrm{mag}}$ and $\chi^{(2)}_{\mathrm{nonmag}}$ are effective tensor elements of the magnetization induced and nonmagnetic nonlinear susceptibility, respectively, and φ is the phase difference between these two parts. During growth, the periodic nucleation, growth, and coalescence of Co islands result in oscillations of the average step density. This causes the periodic change of both the morphology and the magnetic properties of the Co surface. It is not guaranteed a priori that the morphology induced variation in $\chi^{(2)}_{\mathrm{nonmag}}$ does not affect the asymmetry.

However, the average SH intensity $I_{\mathrm{av}}=(I_\uparrow + I_\downarrow)/2$ in Fig. 1(c), which depends almost entirely on $\chi^{(2)}_{\mathrm{nonmag}}$, exhibits only some long periodic variations with the Co film thickness after a strong increase up to 2 ML. No 1 ML oscillations can be detected even in the enlarged view of Fig. 1(e), where the difference between the average intensity I_{av} and the smooth curve averaged over 50 points is plotted. By contrast, the 1 ML periodic oscillations for the asymmetry can be well observed in Figs. 1(b) and 1(d). Therefore morphology induced changes in $\chi^{(2)}_{\mathrm{nonmag}}$ are absent in the present experimental configuration. The 1 ML period is entirely caused by changes in $\chi^{(2)}_{\mathrm{mag}}$.

The observed asymmetry is not entirely due to the surface, but results from the combined contributions of the Co surface and the Co/Cu interface. From the analysis of a thick Co film,[10] we know that the surface and interface contributions have nearly opposite phases. An increase of surface part of $\chi^{(2)}_{\mathrm{mag}}$ increases the asymmetry while the interface part decreases it. The sign change of the asymmetry at about 4 ML is only caused by the gradual increase of the interface contribution relative to the surface contribution with increasing Co thickness, which results from the variation of the electronic structure of the Co film with thicknesses. At 4 ML a complete destructive interference of surface and interface magnetization induced SH light occurs. However, the 1 ML variation of the asymmetry always exhibits a maximum at half filled layers. Therefore, we can conclude that in the whole range of the Co thickness, the magnetic SHG signal at the surface is enhanced at half filled layers, with respect to filled layers.

In Ref. 19 it is shown that the nonlinear magneto-optical Kerr susceptibility is proportional to the magnetization at the surface or interface, respectively. This suggests that the enhancement of $\chi^{(2)}_{\mathrm{mag}}$ of the surface at half filled layers reflects the enhancement of the surface magnetization which agrees with the argument that the magnetic moment of step atoms has an enhanced value relative to that of atoms within the flat surface.

It is known that the growth conditions, such as growth temperature and evaporation rate, influence the growth mode, the structure, and the magnetic properties of the film. Figures 2 and 3 compare experimental results for different evaporation rates and different growth temperatures, respectively, with otherwise identical conditions to the measurement in Fig. 1. In all cases the average signal I_{av} does not show any 1 ML periodic oscillations. For ease of comparison, the result of Fig. 1 is shown again in column (a) of Fig. 2 and column (b) of Fig. 3.

In the whole range of the evaporation rate investigated, we see the 1 ML oscillations in the asymmetry as the Co film thickness increases, indicating that the growth is layer by layer up to 40 ML/h of the evaporation rate. It was only possible to vary the evaporation rate by a factor of less than 6. Therefore, the island size is expected to decrease only by a factor of less than 2. No significant increase in the amplitude of δA is observed although the average surface properties are changed as can be seen most easily at the shift of the zero crossing point of the asymmetry to lower thickness with increasing evaporation rate indicating a reduced surface contribution. Further experiments are necessary to clarify this growth rate dependence.

By contrast to the deposition rate, the change of the substrate temperature has a much stronger influence. At higher temperature (350 K), the growth is still layer by layer, but the oscillations are less pronounced than those at room tem-

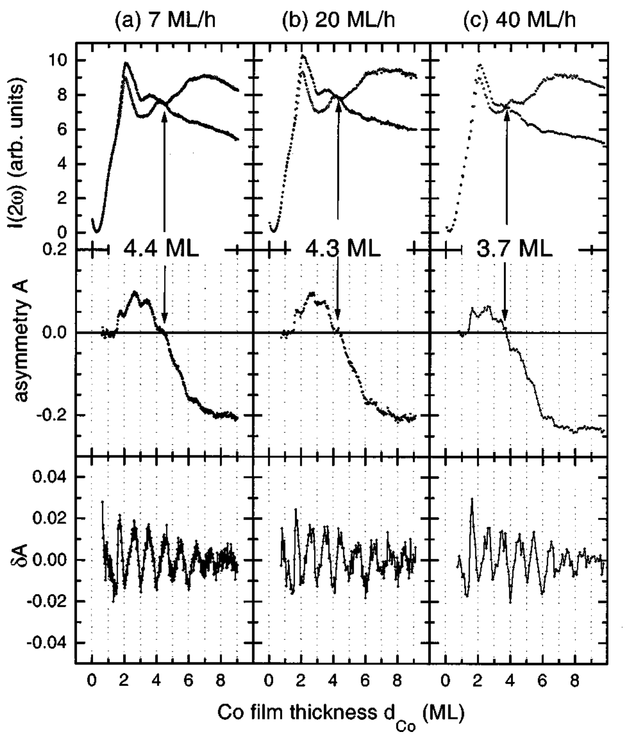

FIG. 2. Measured SH intensities $I(2\omega)$, magnetic asymmetries A, and the differences δA between the asymmetry and its running average over 50 points as a function of the Co film thickness for the magnetization in opposite directions. Figures in columns (a), (b), and (c) have been obtained with evaporation rates of 7, 20, and 40 ML/h. Otherwise, the measurement conditions are identical to those of Fig. 1.

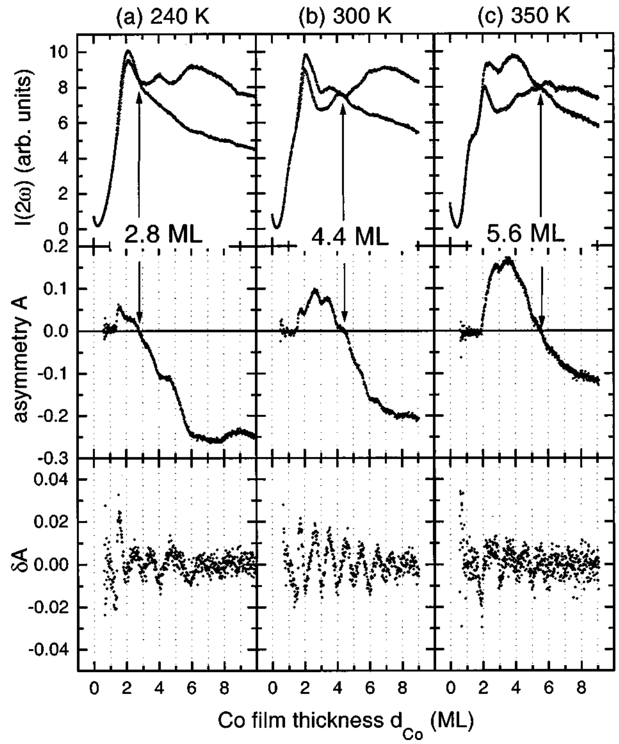

FIG. 3. Measured SH intensities $I(2\omega)$, magnetic asymmetries A, and the differences δA between the asymmetry and its running average over 50 points as a function of the Co film thickness for the magnetization in opposite directions. Figures in columns (a), (b), and (c) have been obtained at growth temperatures of 240, 300, and 350 K.

perature in agreement with the (expected) increased Co island size. When the film grows at low temperature (240 K) the 1 ML oscillations in magnetic asymmetry are kept up to 4 ML. The further course of the asymmetries seem to indicate that the sixth layer starts to grow before completion of the fifth layer.

In conclusion, oscillations with a period of 1 ML are found in the asymmetry of the MSHG signal for *s*-polarized incident light in transversal geometry, which we attribute to the periodic change of the magnetic surface susceptibility due to the periodic variation of the number of step edge atoms during the growth of Co. Additional experiments show that the evaporation rate does not significantly change the 1 ML periodic signal in the range investigated. The reduction of the growth temperature might indicate a change to bilayer or three-dimensional growth at 240 K. Different growth conditions move the zero crossing points in the magnetic asymmetry, indicating a change of the average properties of the thin Co film.

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[1] R.-P. Pan, H. D. Wei, and Y. R. Shen, Phys. Rev. Lett. **39**, 1229 (1989).
[2] J. Reif, J. C. Zink, C. M. Schneider, and J. Kirschner, Phys. Rev. Lett. **67**, 2878 (1991).
[3] H. A. Wierenga, M. W. J. Prins, D. L. Abraham, and Th. Rasing, Phys. Rev. B **50**, 1282 (1994).
[4] H. A. Wierenga, W. de Jong, M. W. J. Prins, Th. Rasing, R. Vollmer, A. Kirilyuk, H. Schwabe, and J. Kirschner, Phys. Rev. Lett. **74**, 1462 (1995).
[5] R. Vollmer, M. Straub, and J. Kirschner, Surf. Sci. **352–354**, 937 (1996).
[6] R. Vollmer, A. Kirilyuk, H. Schwabe, J. Kirschner, H. A. Wierenga, W. de Jong, M. W. J. Prins, and Th. Rasing, J. Magn. Magn. Mater. **148**, 295 (1995).
[7] B. Koopmans, M. G. Koerkamp, Th. Rasing, and H. van den Berg, Phys. Rev. Lett. **74**, 3692 (1995).
[8] M. Straub, R. Vollmer, and J. Kirschner, Phys. Rev. Lett. **77**, 743 (1996).
[9] A. Kirilyuk, Th. Rasing, R. Mégy, and P. Beauvillain, Phys. Rev. Lett. **77**, 4608 (1996).
[10] Q. Y. Jin, H. Regensburger, R. Vollmer, and J. Kirschner, Phys. Rev. Lett. **80**, 4056 (1998).
[11] T. Luce, W. Hübner, A. Kirilyuk, Th. Rasing, and K. H. Bennemann, Phys. Rev. B **57**, 7377 (1998).
[12] R. Vollmer, in *Nonlinear Optics in Metals*, edited by K.-H. Bennemann (Oxford University Press, London, in press).
[13] S. Ohnishi, A. J. Freeman, and W. Weiner, Phys. Rev. B **28**, 6741 (1983).
[14] E. Tamura, R. Feder, G. Waller, and U. Gradmann, Phys. Status Solidi B **157**, 627 (1990).
[15] O. Hjortstam, J. Trygg, J. M. Wills, B. Hohansson, and O. Eriksson, Phys. Rev. B **53**, 9204 (1996).
[16] U. Gradmann, in *Handbook of Magnetic Materials*, edited by K. H. J. Buschow (Elsevier, Amsterdam, 1993), Vol. 7, Chap. 1, p. 1.
[17] M. Albrecht, U. Gradmann, T. Furubayashi, and W. A. Harrison, Europhys. Lett. **20**, 65 (1992).
[18] A. V. Smirnov and A. M. Bratkovsky, Phys. Rev. B **54**, R17371 (1996).
[19] U. Pustogowa, W. Hübner, and K. H. Bennemann, Surf. Sci. **307–309**, 1129 (1994).