Journal of Magnetism and Magnetic Materials 148 (1995) 295–297

ELSEVIER

Journal of magnetism and magnetic materials

Direct comparison of nonlinear and linear Kerr-effect measurements on thin Co films on Cu(001)

R. Vollmer [a,*], A. Kirilyuk [a], H. Schwabe [a], J. Kirschner [a], H.A. Wierenga [b], W. de Jong [b], Th. Rasing [b]

[a] *Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle / Saale, Germany*
[b] *Research Institute for Materials, Toernooiveld, 6525 ED Nijmegen, The Netherlands*

Abstract

It is shown that magnetization-induced second harmonic generation (SHG) is a powerful tool for investigating surfaces and buried interfaces. The source of the SHG is bound to the interface within a region of less than 2 monolayers. Changes in the electronic structure of the Co and especially the Cu-covered Co films as a function of the Co/Cu film thickness are observed.

Second harmonic generation (SHG) is a nonlinear optical technique that derives its interface sensitivity from the breaking of symmetry at the boundaries between centrosymmetic media [1]. Because the optical penetration depth in metals is of the order of 100 nm, not only surfaces but also buried interfaces can be investigated by this method. Theoretically, it has been shown that magnetic effects should be detectable with SHG [2,3]. First experimental evidence for this nonlinear Kerr effect on surfaces was given by Reif et al. [4] and for buried interfaces by Spierings et al. [5]. In this experiment we compare directly linear magneto-optical Kerr-effect (MOKE) measurements with nonlinear Kerr measurements of the magnetization-induced second harmonic generation (MSHG) on Co films epitaxially grown on Cu(001). It is shown that MSHG is very surface sensitive. A two monolayer (ML) thick Co film gives essentially the same MSH signal as a 20 ML thick film. The MSH signal of Cu-covered Co films depends strongly and in a complicated way on the Cu coverage.

Co grows in the fcc structure on Cu(001) and the easy axis of magnetization is along the ⟨110⟩-directions in the surface plane. Most MSHG measurements were carried out in the transverse geometry, i.e. the plane of incidence is perpendicular to the magnetization of the film. The angle of incidence was ~ 35° measured to the surface normal. It was verified experimentally that the SH outgoing light is purely p-polarized. Therefore, in this geometry the total SH signal is recorded. For the longitudinal geometry, in which the optical plane contains the direction of the magnetization, a polarizer was used in the outgoing beam to separate the polarizations. No compensation for ellipticity was made. The linear Kerr measurements were taken in the longitudinal geometry with an angle of incidence of about 45°. For the MSHG measurements we used an Ar^+ ion laser pumped femtosecond Ti–sapphire laser at a wavelength of 800 nm and for linear Kerr measurements a HeNe laser.

The relative magnetic SH signal ρ and the linear MOKE signal as a function of Co film thickness is shown in Fig. 1. ρ is defined as $\rho = (I(+M) - I(-M))/(I(+M) + I(-M))$, with $I(\pm M)$ the SH intensity for the magnetization in opposite directions. While the linear MOKE signal increases nearly linearly with the film thickness the MSHG does not, directly proving the interface sensitivity of MSHG. ρ is nearly constant for p-polarized incoming light except for a small bump for thicknesses around 3–4 ML, while for s-polarized light ρ is initially positive and changes sign at around 5 ML. There is almost no change in the average signal $I_{av} = (I(+M) + I(-M))/2$ for s-polarized light and a small decrease towards thicker films for p-polarization. We speculate that the strong thickness dependence of the relative magnetic signal for s-polarization as well as the small bump at 3–4 ML for p-polarization is related to finite size effects of the electronic structure of the film, as will be discussed further below.

Actually the Co film has two interfaces: the vacuum/Co and the Co/Cu(001) interface. At both interfaces the SH is generated and the observed signal is the superposition of

* Corresponding author. Fax: +49 (0345) 5511-223; e-mail: vollme@secundus-mpi-msp-halle.mpg.de.

SSDI 0304-8853(95)00244-8

both sources [7]. By modifying one interface and observing the change in the SH signal one can partly separate the two contributions. By exposing the Co film to 2 Langmuir (L) CO the average SH signal is nearly unchanged while ρ changes. For a 10 ML Co film for p-polarization of the incident beam, ρ *increases* from 0.3 of the clean film to 0.7. For s-polarized light ρ changes sign at around 1 L. This implies that the magnetic part of $\chi^{(2)}$ (the part which changes its sign when reversing the magnetization of the film) of the two interfaces partially cancel each other for the clean film.

Another way of modifying the upper interface is to evaporate Cu on top of the Co layer. Just by symmetry considerations one would expect that the nonmagnetic part of $\chi^{(2)}$, the part which does not change its sign when reversing the magetization, cancels completely, as can be seen in Fig. 2. The average SH signal for incident p-polarization drops by more than an order of magnitude within 4 ML. There is only a minor decrease in the average SH signal for s-polarization. However, the initial SH intensity is already ten times lower than for p-polarization.

The relative magnetic SH signal ρ shown on the left side of Fig. 2 behaves completely differently. ρ oscillates between $+0.6$ and -0.6 in the range of 2–10 ML. Similar large changes are observed for s-polarized light. Also, the average signal for both s- as well as p-polarized incident light, shows one bump ranging from ~ 7 to ~ 18 ML Cu coverage. This system, Cu films on fcc Co(001), is well known for its appearance of quantum well states. Ortega and Himpsel [6] have found an oscillating behavior of the density of states at the Fermi surface as a function of the Cu film thickness and observed an oscillation period of 5.5 layers. Oscillations of period 2.3 can be attributed to quantum well states as well [8]. The experimental results seem to indicate that these finite size effects of the Cu film which modulate the electronic structure at the interface can be seen by SHG, in both the magnetic and nonmagnetic parts.

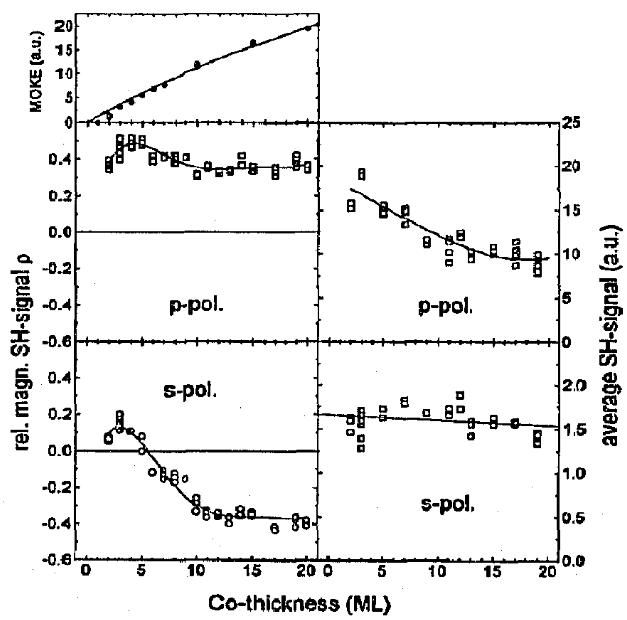

Fig. 1. Dependence of linear MOKE (left topmost panel) and nonlinear MSH signals (bottom four panels) from Co films on Cu(001) as a function of the layer thickness. The linear measurements were taken in the longitudinal geometry, while for the MSHG measurements the transverse geometry was chosen.

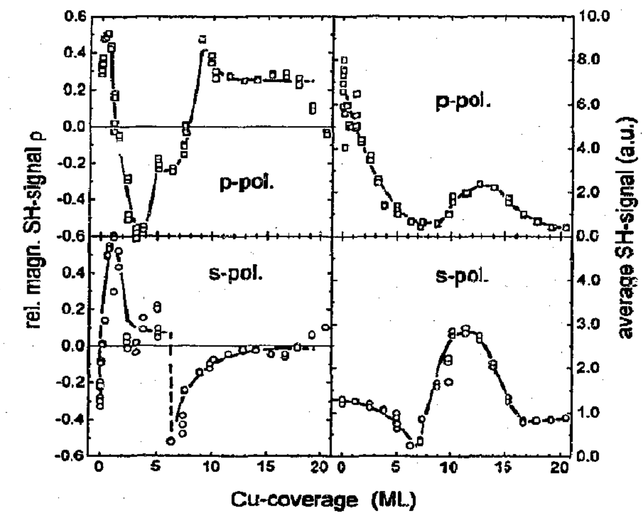

Fig. 2. Relative magnetic SH signal ρ (left side) and the average SH signal (right side) as a function of the Cu coverage for a 10 ML Co film in transverse geometry for both p-polarized (top) and s-polarized (bottom) incoming light.

First measurements for the longitudinal geometry are carried out. In this geometry the nonmagnetic components of $\chi^{(2)}$ lead to a p-polarized SH outgoing beam, both, for s- and for p-polarized incident light, while the magnetic component of $\chi^{(2)}$ leads to entirely s-polarized SH outgoing light. Therefore, by measuring the relative magnetic signal as a function of the polarizer angle, which is placed in the outgoing beam, from the relative magnetic signal ρ alone one can derive the relative amplitude of the magnetic and nonmagnetic parts of $\chi^{(2)}$ and the relative phase factor ϕ betweem them. For example, a 15 ML thick Co film on Cu(001) exposed to 5 L CO and s-polarized incoming light, gives $|\chi^{(2)}_{\rm mag}|/|\chi^{(2)}_0| \approx 0.43$ and $\phi \approx 50°$. These values correspond to a Kerr rotation of 18°. For p-polarized incident light the Kerr angles are found to be smaller.

In conclusion, we have shown that magnetization-induced second harmonic generation is a powerful tool for investigating surfaces and buried interfaces. It is very surface sensitive, because the source of the SHG is bound to the interface within a region of less than 2 monolayers. Detailed information about the specific tensor elements of $\chi^{(2)}$ can be obtained from the measurements at different geometries. Changes in the electronic structure of the Co and especially the Cu-covered Co films as a function of the Co/Cu film thickness are observed.

References

[1] Y.R. Shen, The Principles of Nonlinear Optics (Wiley, New York, 1984).
[2] Ru-Pin Pan, H.D. Wei and Y.R. Shen, Phys. Rev. B 39 (1989) 1229.
[3] W. Hübner and K.H. Bennemann, Phys. Rev. B 40 (1989) 5973.
[4] J. Reif, J.C. Zink, C.M. Schneider and J. Kirschner, Phys. Rev. Lett. 67 (1991) 2878.
[5] G. Spierings, V. Koutsos, H.A. Wierenga, M.W.J. Prins, D. Abraham and Th. Rasing, Surf. Sci. 287 (1993) 747; J. Magn. Magn. Mater. 121 (1993) 109.
[6] J.E. Ortega, F.J. Himpsel, G.J. Mankey and R.F. Willis, Phys. Rev. B 47 (1993) 1540; Phys. Rev. Lett. 69 (1992) 844.
[7] H.A. Wierenga, M.W.J. Prins and Th. Rasing, Physica B 204 (1995) 281.
[8] M.T. Johnson, S.T. Purchell, N.W.E. McGee, R. Coehoorn, J. aan de Stegge and W. Hoving, Phys. Rev. Lett. 68 (1992) 2688.