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Nonlinear magneto-optical Kerr effect study of quantum-well states in a Au overlayer on a Co(0001) thin film

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We have measured the polar nonlinear magneto-optical Kerr rotation and the total generated second harmonic intensity from a perpendicularly magnetized Co(0001)/Au(111) thin film (6 ML) versus the thickness of a Au overlayer. For both experiments we find a clear oscillation with a period of about 13.5 ML. This behavior can be interpreted as arising from quantum-well states (QWSs) in the Au overlayer, though interestingly, the observed period is twice the expected one. Especially for the reflected intensity this oscillation is very pronounced: the intensity changes by a factor of 10 when the Au overlayer thickness changes from 7 to 13 ML. These strong effects make this nonlinear technique very suitable for the study of these QW oscillations. © *1996 American Institute of Physics.* [S0021-8979(96)19808-5]

Since the discovery that magnetic films separated by a nonmagnetic spacer layer could be coupled antiferromagnetically,[1] and the subsequent discovery that this coupling could oscillate between ferromagnetic and antiferromagnetic,[2] there have been intense efforts to understand these phenomena. It was shown that in ultrathin films, due to the electronic potential discontinuities experienced by electronic states at interfaces, the perpendicular component of the wave vector can become quantized, giving rise to resonances in the density of states. Those quantum-well states (QWSs) may act as the mediator for this magnetic coupling.[3] Magneto-optical Kerr effect (MOKE) and direct- and inverse photo emission experiments have shown direct evidence of such QWSs in thin noble metal films on magnetic substrates (Fe and Co),[4–6] and in a bcc Fe(100) layer on Au(100).[7,8] More recently, oscillations of the Kerr angle with changes of overlayer film thickness have been observed for a Au/Co/Au system.[9] These results showed the existence of quantum size effects in the thin Au overlayer and also gave an indication of possible interface contributions.

Due to the fact that optical second harmonic generation (SHG) is known to be surface sensitive on an atomic scale, it seems that SHG studies might be particularly suitable for improving our knowledge of the electrodynamics of ultrathin metallic films. Though the absolute nonlinear signals are small, the nonlinear magneto-optical effects can be large: compared to the linear Kerr angle, enhancements up to a factor of 1000 have been observed.[10] Wierenga *et al.*[11] were the first to report about the possibilities of this nonlinear optical technique for detecting QW oscillations. They have found very strong oscillations in the magnetization-induced SHG signal for the Cu/Co/Cu system as a function of the thickness of the Cu overlayer, which are probably related to QWSs in these films.[11] These strong effects are directly related to the extreme interface localization of the nonlinear response.

In this paper we report the unambiguous observation of QWSs in a Au(111) overlayer on Co(0001). The oscillations are found in both the polar nonlinear MOKE as well as in the total generated second harmonic intensity, as measured as a function of the Au overlayer thickness. These are the first nonlinear MOKE results in the polar geometry, showing an enhancement of two orders of magnitude with respect to its linear equivalent.

For our experiments we used a Ti:sapphire (Tsunami) laser operating at a repetition rate of 82 MHz and a pulse width of about 100 fs. The incoming laser light was filtered and focused onto the sample. Polarization control of both incident fundamental and reflected SH light was achieved by means of polarizers. Appropriate optical filtering was used before the signal was detected by a photon counter. Our sample consists of a six monolayer (ML) Co(0001) film on a 30 nm thick Au buffer layer, covered with a stepped Au wedge, consisting of 13 terraces of $t_{\mathrm{Au}}=6-18$ ML. The terraces have a width of 1.5 mm, so the laser beam can easily be focused on each separate step. Details about the sample preparation can be found in Ref. 9. Note that this sample has a perpendicular easy magnetization axis.

SHG arises from the nonlinear polarization $\mathbf{P}(2\omega)$ induced by an incident laser field $\mathbf{E}(\omega)$. In the electric dipole approximation, this polarization can be written as

$$P_i(2\omega)=[\chi^{+}_{ijk}(M)+\chi^{-}_{ijk}(M)]E_j(\omega)E_k(\omega), \tag{1}$$

where $\chi^{+}_{ijk}(M)$ and $\chi^{-}_{ijk}(M)$ are the even and odd elements of the nonlinear susceptibility tensor that fulfill $\chi^{\pm}_{ijk}(-M)=\pm\chi^{\pm}_{ijk}(M)$, as suggested by Pan *et al.*[12] Since this susceptibility tensor has to reflect the symmetry of the crystal,[13] the nonzero elements are easily derived from the invariance of $\chi(M)$ under symmetry operations. Table I

TABLE I. The nonzero elements of the SH susceptibility tensor for an isotropic surface in the polar (**M**‖z) configuration and for a nonmagnetized interface. The two columns list the elements that are even and odd in the magnetization, respectively.

Nonmagnetized	
$xxz=xzx=yyz=yzy$	
$zxx=zyy$	
zzz	
Polar (**M**‖z)	
Even in **M**	Odd in **M**
$xxz=xzx=yyz=yzy$	$xyz=xzy=-yxz=-yzx$
$zxx=zyy$	$zxy=zyx$
zzz	

shows the relevant tensor elements for our experiments. For s-polarized incident light (i.e., parallel to the y axis, see the inset to Fig. 1), only the even element can be excited. Therefore, no Kerr rotation can be expected for this configuration. For p-polarized incident light the Co/Au interfaces have two odd components, both giving rise to s-polarized SHG, and three even components, which produce p-polarized SHG. From this it follows that the SHG polarization ellipses for $\pm\mathbf{M}$ are each other mirror images in the plane of incidence, and we can define a nonlinear Kerr angle $\Phi_K^{(2)}$, analogous to the linear Kerr rotation. Experimentally, the Kerr angle is found by fitting a $\cos^2\alpha$ dependence to the measured intensity as a function of the analyzer angle α, for opposite directions of the magnetic field. This leads to a precision in the nonlinear Kerr angle of ±0.12°.

The experimental variation of $\Phi_K^{(2)}$ vs t_{Au} is given in Fig. 1, showing a clear oscillatory behavior. These measurements have been done with p-polarized incident light at a wavelength of 740 nm and at an angle of incidence of 45°. The observed amplitude of the oscillation is about two orders of magnitude larger than its linear counterpart, as measured on the same sample.[9] The solid line in the figure is a least-squares fit to the expression

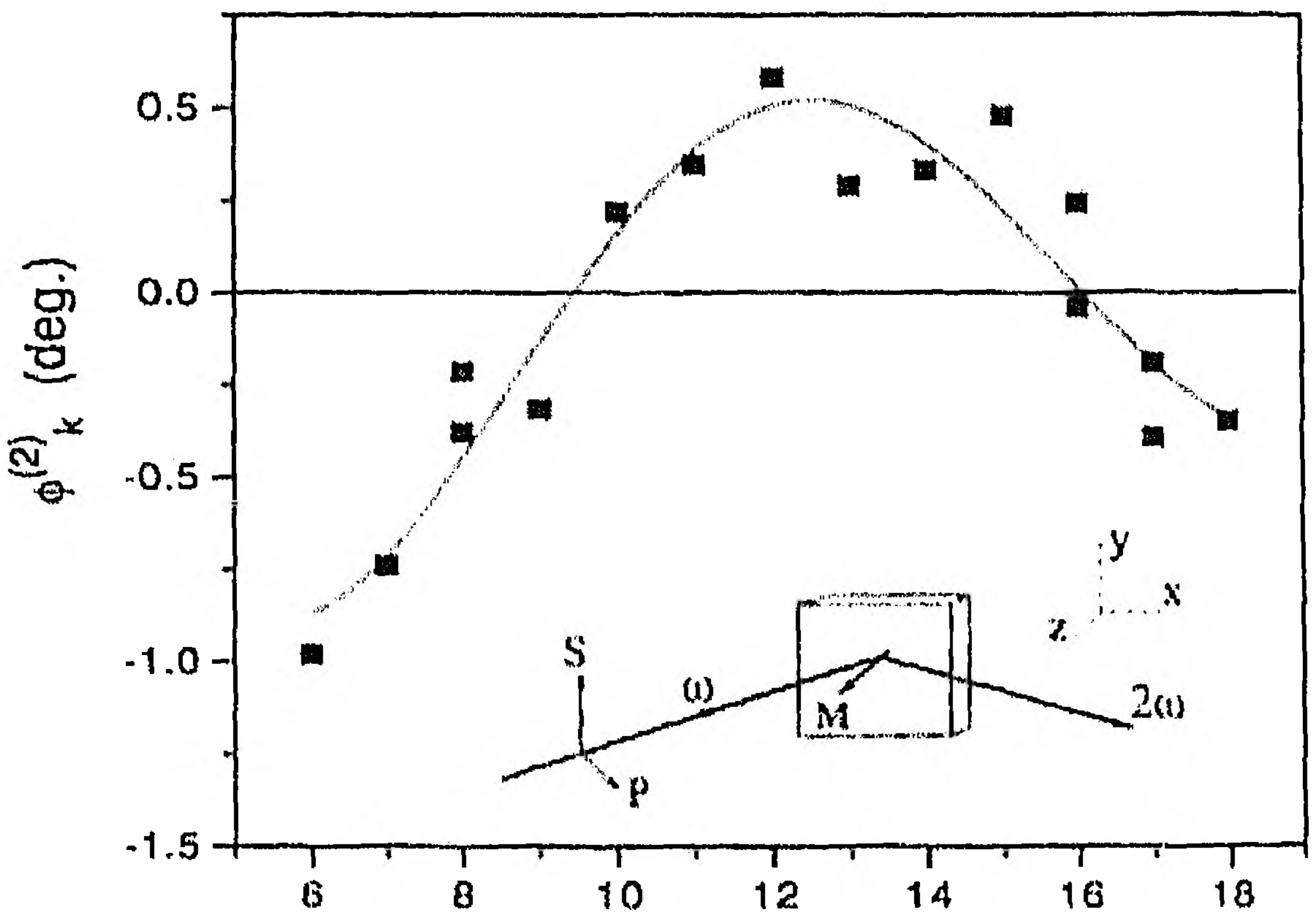

FIG. 1. Nonlinear polar Kerr rotation as a function of the Au overlayer thickness. The solid line is a fit to Eq. (2). The inset shows the experimental configuration.

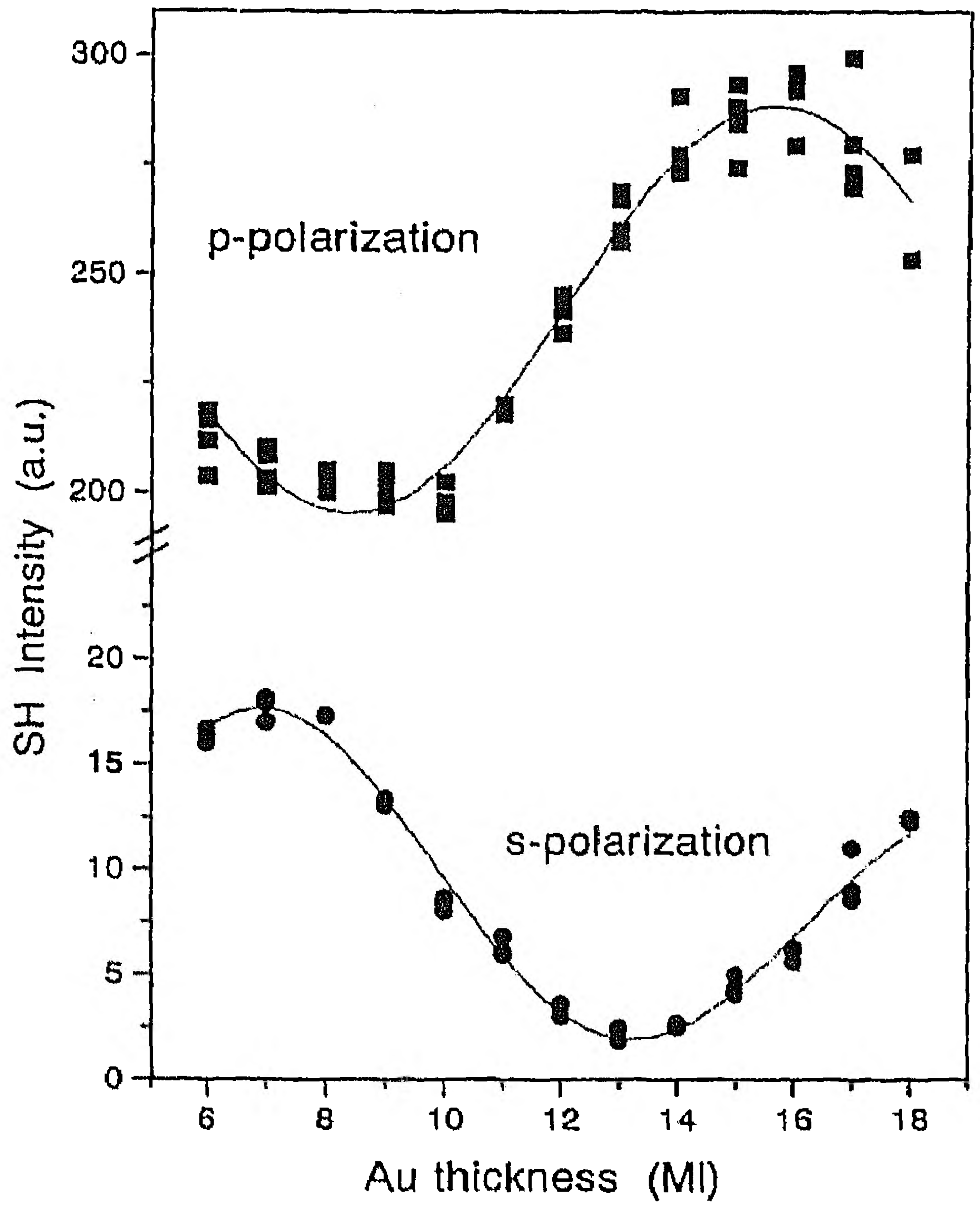

FIG. 2. Total generated SH intensity as a function of the Au overlayer thickness. Dots: s-polarized incoming light, squares: p-polarized incoming light. The solid lines are fits to Eq. (2).

$$\Phi_K^{(2)}=A\times\exp\frac{-t_{\mathrm{Au}}}{\delta}\times\cos\left(2\pi\frac{t_{\mathrm{Au}}}{\Lambda}+\gamma\right)+\beta. \tag{2}$$

From this fit the period Λ of the oscillation is found to be 14±3 ML with an attenuation length δ of 18±4 ML. The maximum measured Kerr angle is about 1.0°. Although this value is a factor of 10 larger than its linear counterpart, the rotation is not as large as one would expect from earlier nonlinear Kerr experiments.[10] One reason for this is the 180° phase difference between the tensor components of the symmetric interfaces Au/Co and Co/Au. Because of the small thickness of the Co film (6 ML), the SH signals from these interfaces partly cancel each other and reduce the nonlinear magneto-optical effects.

The observed oscillatory character of the Kerr angle suggests that the quantum size effects have an influence on the tensor components. Since these effects will not only appear in the magnetic (odd), but particularly in the nonmagnetic tensor elements, the QWSs should also be observable in the total SH intensity. In Fig. 2 it can be seen that for both s- and p-input polarizations we found a clear oscillation in the total reflected SH signal. In the case of s-polarized incident light, we only have one nonzero tensor element χ_{zyy} present at both the Au/air and Au/Co interfaces, which explains the small signals here. The oscillation, however, comes out much more pronounced than in the Kerr measurements. By varying the Au overlayer thickness from 7 to 13 ML, the generated SH intensity drops by a factor of 10. The solid line in the figure is a least squares fit of the measured data according to Eq. (2). From this fit the period is found to be 12.6 ML, with an attenuation length of 18.3 ML. For p-polarized incident light, a clear oscillation can be seen on top of a large offset.

The solid line is again a fit by Eq. (2) with a period of 13.9 ML. In contrast with the s-input configuration, now no damping is found in the oscillation. This can possibly be explained by the different ways of penetration of the s- and p-polarized light into the material. For comparison we also measured the linear reflection as a function of the Au thickness. Within the experimental uncertainty of $\pm 2\%$, we found a constant value. Our observed period of about 13.5 ML is quite remarkable, as earlier linear Kerr measurements (at 632 nm) on the same sample gave a period of 7.8 ML.[9] Experiments on the oscillatory interlayer coupling between Co films across Au(111) gave a period of 6.5 ML.[14] The difference between the oscillation period in the linear and nonlinear Kerr rotation could possibly be due to the fact that these measurements were not done at the same wavelength, as Suzuki *et al.*[15] have shown that the period of the oscillation in the magneto-optical effect is energy dependent. To find out if there is any strong wavelength dependence, we repeated our Kerr and SH intensity measurements at different wavelengths between 740 and 1000 nm (the tuning range of our Ti:sapphire laser). In this range, however, no wavelength dependence of the period could be measured. A more likely explanation could be the difference between the selection rules involved in the linear and nonlinear optical response. However, this point needs more theoretical investigations. A final point of discussion is the interface roughness, as SHG is known to be extremely sensitive for this. The high quality of the thickness dependence fits suggests that the Co/Au interfaces are very smooth. The latter was also confirmed by other measurements.

In conclusion, we have observed large oscillations in both the nonlinear polar Kerr rotation and in the total generated SH intensity from a perpendicularly magnetized Au/Co/Au(111) sandwich versus the Au overlayer thickness. The period of the oscillation is for both experiments about 13.5 ML. The oscillations are believed to originate from QWSs in the Au overlayer. Specially for the total generated SH intensity, the quantum size effects can be very large: for the s-input polarization, the SH intensity drops by a factor of 10 when the Au overlayer thickness changes from 7 to 13 ML. These large effects make this nonlinear technique very suitable for detecting QW oscillations.

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[1] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986).

[2] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).

[3] M. D. Stiles, Phys. Rev. B **48**, 7238 (1993).

[4] J. E. Ortega, F. J. Himpsel, G. J. Mankey, and R. F. Willes, Phys. Rev. B **47**, 1540 (1993).

[5] D. Hartmann, W. Weber, A. Rampe, S. Popovic, and G. Güntherodt, Phys. Rev. B **48**, 16 837 (1993).

[6] T. Katayama, Y. Suzuki, M. Hayashi, and A. Thiaville, J. Magn. Magn. Mater. **126**, 527 (1993).

[7] F. J. Himpsel, Phys. Rev. B **44**, 5966 (1991).

[8] W. Geerts, Y. Suzuki, T. Katayama, K. Tanaka, K. Ando, and S. Yoshida, Phys. Rev. B **50**, 12 581 (1994).

[9] R. Mégy, A. Bounouh, Y. Suzuki, P. Beauvillain, P. Bruno, C. Chappert, B. Lecuyer, and P. Veillet, Phys. Rev. B **51**, 5586 (1995).

[10] B. Koopmans, M. Groot Koerkamp, Th. Rasing, and H. v.d. Berg, Phys. Rev. Lett. **74**, 3692 (1995).

[11] H. A. Wierenga, W. de Jong, M. W. J. Prins, Th. Rasing R. Volmer, A. Kirilyuk, H. Schwabe, and J. Kirschner, Phys. Rev. Lett. **74**, 1462 (1995).

[12] R.-P. Pan, H. D. Wei, and Y. R. Shen, Phys. Rev. B **39**, 1229 (1989).

[13] J. F. Nye, *Physical Properties of Crystals* (Oxford University Press, Oxford, 1957).

[14] V. Grolier, D. Renard, B. Bartenlian, P. Beauvillain, C. Chappert, C. Dupas, J. Ferré, M. Galtier, E. Kolb, M. Mulloy, J. P. Renard, and P. Veillet, Phys. Rev. Lett. **71**, 3023 (1993).

[15] Y. Suzuki and P. Bruno, J. Magn. Magn. Mater. **140–144**, 651 (1995).