Interfaces contributions to the nonlinear magneto-optical response of quantum well states

A. Kirilyuk and Th. Rasing
Research Institute for Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands

R. Mégy and P. Beauvillain
Institut d'Electronique Foundamentale, Université Paris-Sud, 91405 Orsay Cédex, France

The nonlinear optical response of a noble metal overlayer on top of a magnetic film shows an oscillatory behavior dominating the total second harmonic output. The oscillation periods are twice as large as those observed with linear Kerr effect measurements. To explain this effect, we decompose the total nonlinear susceptibility in different interface contributions. © *1997 American Institute of Physics.* [S0021-8979(97)45608-6]

In ultrathin films, due to the electronic potential discontinuities at interfaces, the perpendicular component of the wave vector can become quantized. Those quantum well states (QWS) may act as the mediator for the exchange coupling in magnetic sandwiches leading to a characteristic oscillatory behavior of the coupling.[1] Photoemission experiments have shown direct evidence of such QWS in thin metal films.[2,3] In addition, these states can become spin polarized even in nonmagnetic metals.[4] As a result, magnetic properties of the ferromagnetic metal surface depend on the thickness of the (thin) nonmagnetic overlayer evaporated onto it. For example, magneto-optical Kerr effect (MOKE) measurements demonstrated small oscillatory changes of the Kerr angle as a function of the overlayer film thickness for the Au(111)/Co[5] as well as for the Cu(111)/Co systems.[6] However those oscillations, though measurable, have a very small amplitude. This fact complicates the measurements and explains the small number of the studied systems.

Magnetization induced second harmonic generation (MSHG) is a nonlinear version of the MOKE technique.[7,8] Recently, it demonstrated an enhanced sensitivity to the very narrow film interface regions[9,10] and a very large magneto-optical response.[11] As electronic structure calculations have shown that these QWS can possibly be localized at the film interfaces[12] (although every QWS wave function extends through the whole film, the local density of states may have maximum at the interfaces), the interface sensitivity of MSHG is obviously of a direct use. Also the enhanced magneto-optical response could probably increase the observability of QWS.

In this article, we report the unambiguous observation of QWS in Au(111) and Cu(111) overlayers on Co(0001) (see also Refs. 13 and 14). The oscillations are found in both the SHG intensity and in the nonlinear magneto-optical effects measured as a function of the gold or copper overlayer thickness. Very large effects in MSHG are in agreement with the interface localization of QWS. For magnetic measurements, we used both polar and transversal configurations, giving rise to polarization rotations and MSHG intensity changes, respectively. The periods of the observed oscillations are *twice* the periods measured with linear MOKE. This can be explained by a simple model taking into account the symmetry of the QWS wave functions.

Our samples were step-shaped wedges of Au(111) or Cu(111) grown on top of a thin [5–20 monolayers (ML)] Co(0001) film on a Au(111) substrate. The copper wedge was covered by 10 ML of gold for protection.

For the MSHG measurements, a pulsed laser beam from a Ti-sapphire laser (100 fs pulses with a repetition rate of 82 MHz) was focused onto the sample while the latter could be moved with the help of a stepping motor in a magnetic field that was either in plane or perpendicular to the sample. After proper filtering, the outcoming specular second harmonic (SH) light was detected with a photomultiplier. In the polar configuration, a Kerr rotation of the SH polarization was measured similar to what has been described in Ref. 11. In the transversal configuration, we checked that for both P and S incoming light polarizations the SH output was always strictly P polarized (i.e., in the plane of incidence). As a magnetic signal, we measured the normalized intensity difference for the magnetization up and down (see below).

The second harmonic polarization $\mathbf{P}(2\omega)$ of a magnetic medium is generally described by a third rank polar tensor χ^{cr}_{ijk} for the crystallographic contribution and a fourth rank axial tensor χ^{magn}_{ijkl} for the magnetization-induced part. In centrosymmetric materials, both tensors are nonzero only at surfaces and interfaces, where the inversion symmetry is broken. For a particular magneto-optical configuration, e.g., polar or transversal (and for a high-symmetry surface), one may consider only one simplified third rank tensor with different components, that are either even in the magnetization $\mathbf{M}$, describing the crystallographic part, or odd and thus relate to the magnetization-induced contribution.[7] $\mathbf{P}(2\omega)$ can then be written as

$$\mathbf{P}(2\omega,\pm\mathbf{M})=(\chi_{even}(\pm\mathbf{M})\pm\chi_{odd}(\pm\mathbf{M}))E^2(\omega), \qquad (1)$$

where χ_{even} and χ_{odd} are linear combinations of, respectively, even and odd tensor elements and $\mathbf{E}(\omega)$ is the incoming light field. In the polar geometry, χ_{even} and χ_{odd} are orthogonal to each other so the vector sum within the brackets results in a SH polarization plane rotation of $\alpha\approx|\chi_{odd}|/\chi_{even}|$. In the transversal geometry, in contrast, both χ_{even} and χ_{odd} act along the same direction, and Eq. (2) shows then a change in the absolute value of $\mathbf{P}(2\omega)$, i.e., a change in the SH intensity $I_{SH}\propto|\mathbf{P}(2\omega)|.^2$ Then, the magnetization contrast can be defined as

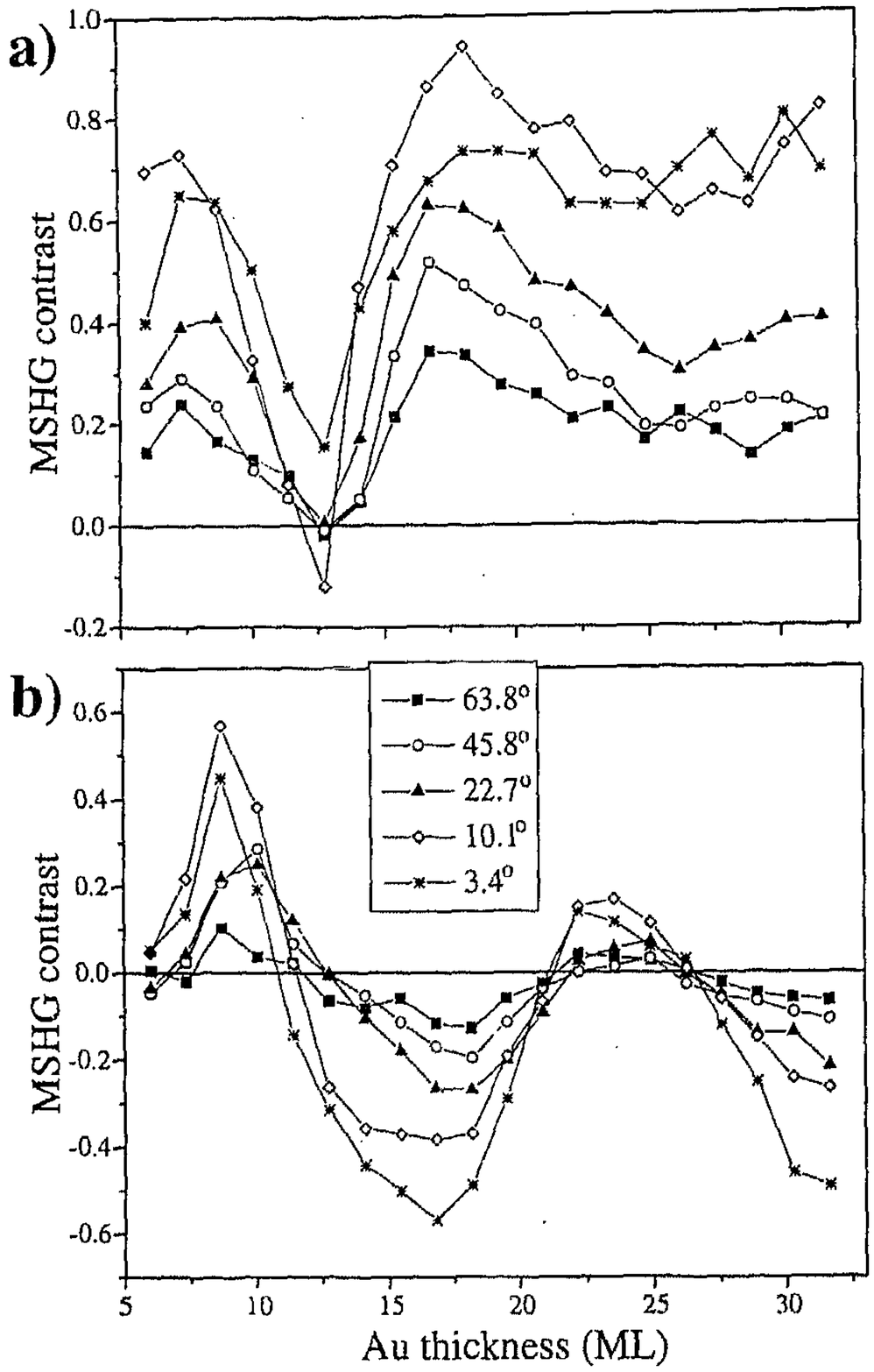

FIG. 1. MSHG relative signal as a function of the gold overlayer thickness for (a) $S_{in}P_{out}$ polarization and (b) $Q_{in}S_{out}$ polarization, for different angles of incidence (indicated in the figure).

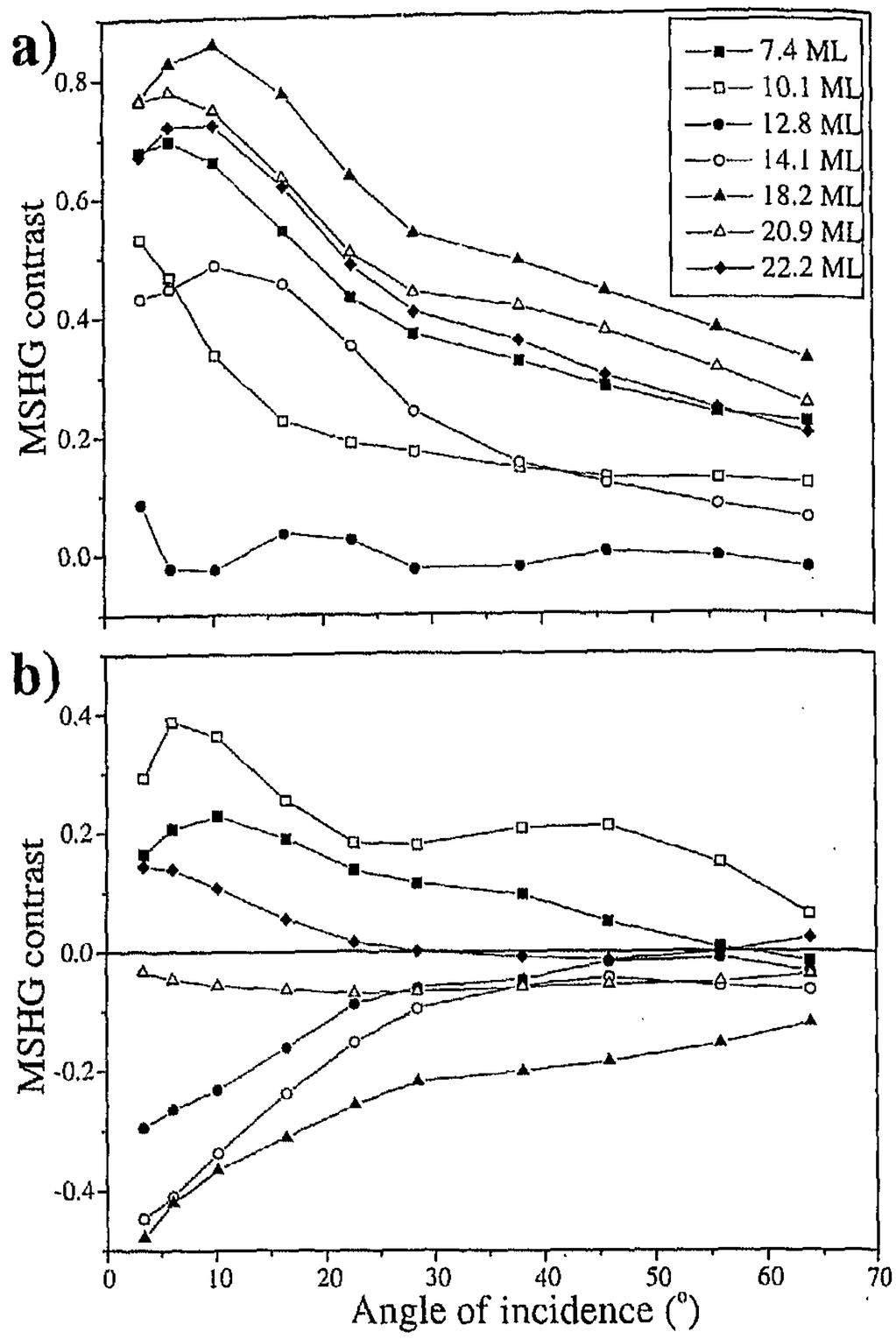

FIG. 2. MSHG relative signal as a function of the angle of incidence for (a) $S_{in}P_{out}$ polarization and (b) $Q_{in}S_{out}$ polarization, for different gold overlayer thicknesses (indicated in the figure).

$$\rho = \frac{I(+\mathbf{M}) - I(-\mathbf{M})}{I(+\mathbf{M}) + I(-\mathbf{M})}, \tag{2}$$

where $I(\pm\mathbf{M})$ are the SH intensities measured for opposite directions of the sample magnetization.

The measured SH intensity displays a strong oscillatory behavior as a function of the overlayer thickness. Figures 1(a) and 1(b) show the MSHG contrast ρ measured at different angles of incidence for the Au(111) overlayer. Similar oscillations have been found for the absolute SH intensity. To get a more quantitative idea about these curves, we fitted all the dependencies with a formula

$$I_{SH} = A \exp\left(-\frac{d_{Au,Cu}}{\delta}\right) \sin\left(2\pi \frac{d_{Au,Cu}}{\Lambda} + \gamma\right) + B. \tag{3}$$

A puzzling point here is the value of the periods which appeared to be rather large. For both the Au(111) and Cu(111) overlayers, the observed period (12–16 ML) is approximately twice the value obtained with linear MOKE.

To explain the period doubling, we will use a model based on the mirror symmetry of the opposite film interfaces.[14] Indeed, for a thin film, the corresponding tensor elements on the opposite film interfaces are related to each other by a mirror symmetry relation and therefore they differ only by a phase factor of 180°. Hence the resulting total SHG signal arises from the competition between the signals from the two film interfaces that mostly cancel each other and does only depend on the difference in their local fields. Within this approach, the influence of QWS on MSHG would be largely cancelled also because every QWS contributes symmetrically (via its local density of states) to the χ tensor elements of both interfaces. This is contrary to the experimental observations of a total domination of QWS on the SHG response. To understand the observed double period, one has to take the symmetry of the QWS wavefunctions into account, and the fact that the two interface contributions are *interdependent*.

Therefore, we tried to decompose the total MSHG signal into the contributions from different interfaces. For this purpose, first the angle-of-incidence dependencies of the MSHG signals were measured for each gold overlayer thickness value (see Fig. 2). Next, a fit of these data was performed, using the χ tensor elements as fitting parameters. For the fit to be unique, one has to use either $S_{in}P_{out}$ or $Q_{in}S_{out}$ (Q is the polarization in between P and S) polarization combinations. In both cases, there is only one even (and one odd) χ component per (magnetic) interface.

The major part of the signals can very well be described with a model taking into account only the interface MSHG. Oscillatory behavior of the signals naturally originates from the corresponding behavior of the tensor components. Figure 3(a) shows the individual χ components of the two gold

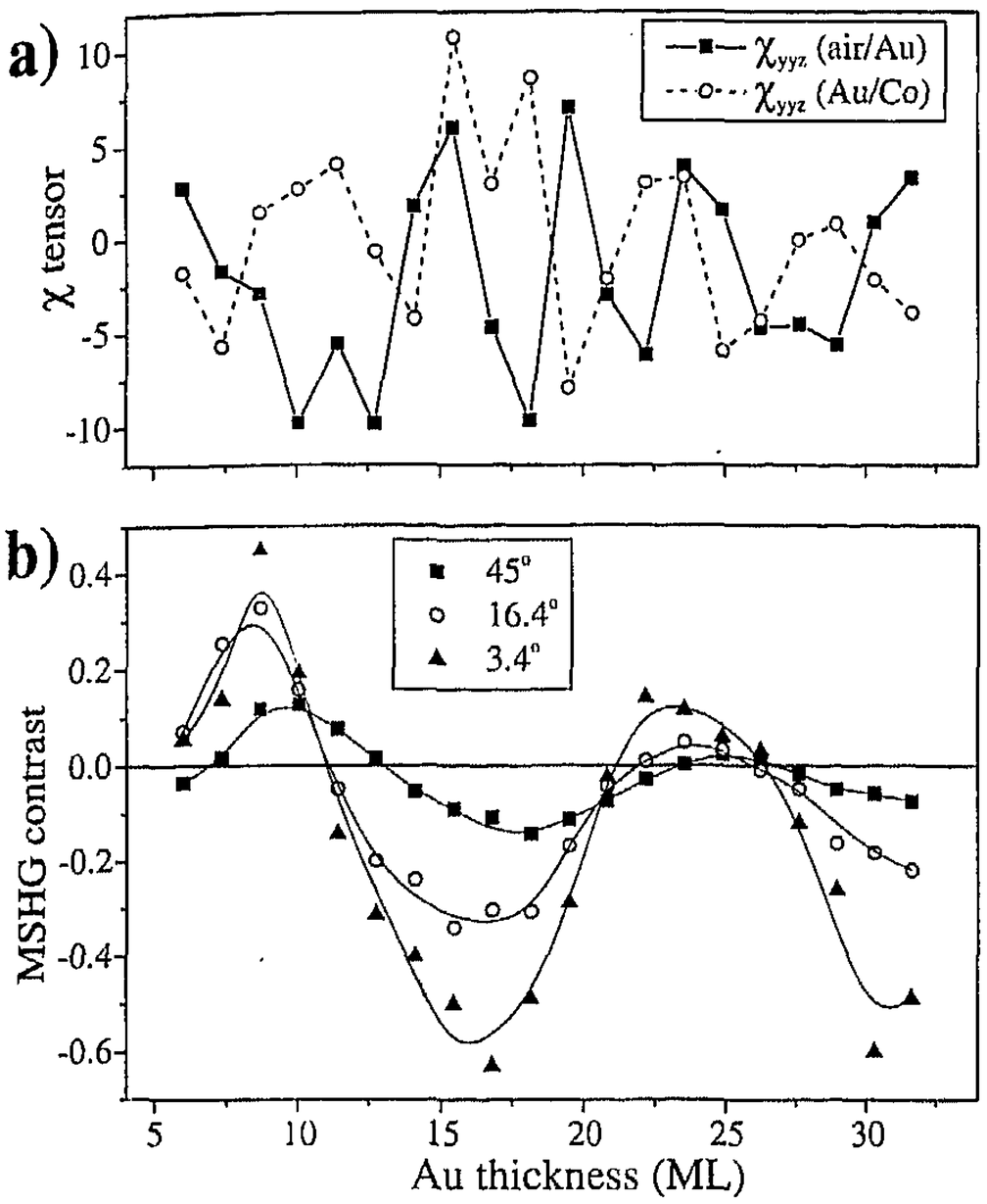

FIG. 3. (a) Tensor components of different Au(111) film interfaces as a function of the film thickness. (b) Theoretical fit of the MSHG contrast using the parameters shown in (a).

layer interfaces as a function of the film thickness, displaying an oscillatory behavior with a mean period of around 6–8 ML, i.e., the same period as observed by MOKE. However the resulting MSHG signal perfectly fit the experimentally observed slowly oscillating behavior with the double period [see Fig. 3(b)]. This means that while the local density of states and hence the χ tensor at each interface show the standard QWS period, the resulting total response includes also the phase between the corresponding elements and therefore allows a slower variation. This is possible, of course, only because the interfaces are not independent as soon as every QWS wave function is located at both interfaces simultaneously. Although it is not exactly correct to talk about "contributions of different interfaces" in such conditions, it is still possible to formally use this model.

As for the linear MOKE technique, although it has "bulk" sensitivity, the oscillatory part of the signal is provided by the narrow region along the Au/Co interface, where spin polarization of electrons is affected the most. Therefore it is related to the local density of states at this interface only which oscillates with a single QWS period.

The unusually large influence of QWS on SHG is perfectly explained by the interface sensitivity of the technique[8,10] plus the calculated interface localization of QWS.[12] Noble metals seem to be the most convenient materials for such studies because of their simple Fermi surfaces and high interband transition thresholds. It appears that the mirror symmetry of a thin film can lead to the doubling of the oscillation period. To further develop this subject, theoretical calculations are necessary, taking into account realistic eigenfunctions of a thin metal film forming a quantum well.

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