QUANTUM WELL STATES IN NOBLE METALS: A SECOND HARMONIC GENERATION STUDY

A. Kirilyuk, M. Groot Koerkamp 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 Fondamentale, Universite Paris-Sud, 91405 Orsay Cedex, France

Abstract - The nonlinear magneto-optical response of a magnetic metal as a function of the thickness of a noble metal overlayer shows that an oscillatory behavior dominates the total generated second harmonic output. The oscillation periods are approximately twice as large than those observed with linear Kerr effect measurements or estimated from the Fermi surface. The spectral dependence of the signal indicates that the thickness independent background is due to interband transitions while the oscillatory part persists at lower photon energies.

KEYWORDS: NONLINEAR MAGNETO-OPTICS, QUANTUM WELL STATES, INTERBAND TRANSITIONS

INTRODUCTION

In ultrathin films, due to the electronic potential discontinuities 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 (QWS) may act as the mediator for the exchange coupling in magnetic sandwiches leading to a characteristic oscillatory behaviour 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/Co/Au(111) system [5]. 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 [6,7]. Recently it demonstrated an enhanced sensitivity to the very narrow film interface regions [8,9] and a very large magneto-optical response [10]. As electronic structure calculations have shown that these QWS can possibly be localized at the film 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 paper we report the unambiguous observation of QWS in Au(111) and Cu(111) overlayers on Co(0001). 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. For magnetic measurements, we used both polar and transversal configurations, giving rise to polarization rotations and MSHG intensity changes, respectively. The spectral dependencies of the observed signals show that interband transitions in the overlayer metal are responsible for the thickness independent part of the second harmonic intensity which is therefore reduced at longer wavelengths. In contrast, the oscillatory part does not considerably depend on the photon energy in the low-energy part of the spectrum.

SAMPLES

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, because all measurements were done ex-situ. Cobalt films have also been grown as steps, with few different thicknesses. Because of a strong interface-induced perpendicular magnetic anisotropy in this system [11], we had a possibility to use different (polar or transversal) magneto-optical configurations depending on the cobalt thickness.

EXPERIMENTAL SETUP

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 outgoing 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. [10]. 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”.

RESULTS AND DISCUSSION

MSHG is generally described by a third rank nonlinear susceptibility tensor \( \chi^{(2)} \):

\[
P(2\omega) = \chi^{(2)}_{ijk} E_i(\omega) E_j(\omega),
\]

where \( P(2\omega) \) is a second harmonic polarization of the medium created by the incident light wave \( E(\omega) \). In magnetic media, \( \chi^{(2)} \) becomes magnetization dependent: \( \chi^{(2)}(M) \) [6]. From symmetry considerations, tensor elements can be separated in even and odd with respect to the magnetization. Eq. (1) is then written as

\[
P(2\omega, M) = (\chi_{\text{even}}(M) + \chi_{\text{odd}}(M))E^2(\omega),
\]

where \( \chi_{\text{even}} \) and \( \chi_{\text{odd}} \) are linear combinations of respectively even and odd tensor elements.

In experiments, the SH intensity \( I_{\text{SH}} \sim |P(2\omega)|^2 \) and the polarization of the outgoing light is measured. Similar to MOKE, in the polar geometry we deal with a polarization plane rotation as a magneto-optical response. For a P-polarized fundamental beam, the angle of rotation is proportional to the ratio between odd and even tensor elements. In the transversal geometry, in contrast, only the SH intensity depends on the sample magnetization. Then, the magnetization contrast can be defined as:

\[
\eta = \frac{[I(+M)-I(-M)]}{[I(+M)+I(-M)]}
\]

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

The measured SH intensity displays a strong oscillatory behaviour as a function of the overlayer thickness. Figs. 1a,b show the SH intensity measured at different wavelengths for the Au(111) and Cu(111) overlayers, respectively. Evidently, these oscillations are a dominating feature of the curves, especially for longer wavelengths. Similar oscillatory behaviour has been found for the magnetization induced contributions to SHG, both for the polarization rotation in the polar configuration [12] and for the intensity changes in the transversal setup (eq. (3)); see Fig. 2 for the Cu(111) overlayer.

To get a more quantitative idea about these curves, we fitted all the dependencies with a formula

\[
I_{\text{SH}} = A \exp\left(-d_{\text{Au,Cu}}/\delta\right) \sin(2\pi d_{\text{Au,Cu}}/\Lambda + \gamma) + B
\]

It can be seen already from Fig. 1 that the phase of the oscillations \( \gamma \) changes while the period \( \Lambda \) remains approximately constant (at least, for the gold overlayer). Fig. 3a shows the period \( \Lambda \) for P and S incoming polarizations as a function of the second harmonic photon energy.
Fig. 2. Relative magnetic effect in transversal geometry as defined by eq. (3), for the 15 ML thick Co film with a Cu(111) overlayer. Dashed lines serve as guides for the eye.

A puzzling point here is the value of these periods which is rather large. For the Au(111) overlayer the observed period (12-16 ML) is approximately twice the value obtained with linear MOKE. For the sample with a Cu(111) overlayer, a MOKE study failed to find any oscillations of the Kerr angle. Also interesting is the observation that different light polarizations clearly indicate different periods. This might be due to the fact that light with different incoming polarizations excite different $\chi$ tensor elements which, in turn, depend on different wave functions. The general origin of the observed large periods will be discussed in detail elsewhere [13].

Fig. 3b shows the dependence of the oscillation amplitude $A$ relative to the background $B$ as a function of the SH photon energy. Also plotted is the optical absorption curve for gold taken from Ref. [14]. Thus, while the $A/B$ value for the S polarized incoming light remains approximately the same, 80-90%, the dependence for P polarization definitely shows an increase toward lower photon energies. This means that the non-oscillatory part of the signal is considerably reduced at lower energies. This increase actually coincides with the fundamental absorption edge in gold which strongly suggests optical interband transitions as the origin for the thickness independent part of the SH intensity.

For the Cu(111) overlayer, the periods and amplitudes behave in a way very similar to the case of gold. There is one exception, however: the fundamental absorption edge for copper is shifted to lower energies (2.1 eV instead of 2.5 eV for gold). In other aspects, the band structures of Cu and Au are very similar. Therefore, the observed change in relative amplitude is just shifted to lower energies (Fig. 3b). There is also a considerable change in the oscillation period occurring around 2.7 eV which can possibly be ascribed to the beginning of some type of optical interband transition at this energy.

Thus we can suppose that the strong oscillatory behaviour of SHG at lower photon energies is not related to the direct interband transitions. Therefore it is necessary to consider other possibilities, namely (i) the (spin-polarized) band structure of cobalt serves as a source for either initial or final states, or (ii) intraband transitions in gold are responsible for the effect. It has
been shown, however, that in the case of transitions between spin-polarized states, the SH intensity should be considerably different for the opposite directions of the sample magnetization [15]. This does not appear to be the case for the considered samples (the relative magnetic effect as defined by eq. (3), almost always oscillates around zero or very close to it, see Fig. 2), hence the direct influence of the cobalt band structure can be ruled out.

As for the intraband transitions, they might be of much more importance in very thin films in comparison to the bulk metals. Indeed, for ultra thin films the component of the wave vector perpendicular to the film plane becomes quantized. Therefore the requirement of its conservation is lifted, and the transitions between the subsequent QWS become allowed. Such transitions at low photon energy obviously take only place in the vicinity of the Fermi surface. This fact can naturally explain why the oscillation period does not considerably depend on the photon energy. The only strong change which is observed for the period of the Cu(lll) overlayer can therefore be explained by a crossover to interband transitions at higher photon energies.

CONCLUSION

Nonlinear magneto-optics has been demonstrated as a very powerful tool for the study of (spin-polarized) Quantum Well states. In the case of (111) oriented noble metal overlayers, oscillations of the MSHG signals as a function of the overlayer thickness persist even at low photon energies, where direct interband transitions are not allowed even for two-photon processes.

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REFERENCES