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Nonlinear and linear Kerr studies of Co/Cu multilayers

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Abstract

Magnetization induced optical second harmonic generation (MSHG) in combination with the magneto-optical Kerr effect (MOKE) was used to study thin Co films and Cu/Co multilayers of thicknesses between 1 and 20 monolayers (ML) grown on a Cu(001) substrate. From the different dependence of MSHG and MOKE on Co film thickness we get direct information about the interface sensitivity of MSHG. Modifying the vacuum/Co interface by gas adsorption yields the relative contributions of the vacuum/Co and the Co/Cu(001)-substrate interfaces. Both MSHG and MOKE data can be accurately described by a multiple reflection model.

Keywords: Cobalt; Copper; Magnetic interfaces; Metal-metal magnetic thin film structures; Second harmonic generation

1. Introduction

The magnetic properties of thin films and multilayer systems containing ferromagnetic material provide a fascinating field of research and are a subject of great current interest [1]. Besides their technological significance, a number of extraordinary phenomena are observed such as the change of the magnetization from normal to in plane for thin Fe films [2], a spin behaviour at clean surfaces that is different from the bulk [1,3], and oscillatory exchange coupling through non-magnetic spacers [4-6]. Because the interfaces appear to play an important role in the magnetic properties of such multilayers, a magnetization and interface sensitive probe would be very interesting. Recently, it has been shown that the interface sensitive nonlinear optical technique of second harmonic generation, is also sensitive to the magnetization [7-11].

SHG arises from the nonlinear polarization $P(2\omega)$ induced by an incident laser field $E(\omega)$.

$$P_j(2\omega) = \chi^{(2)}_{jkl} E_k(\omega) E_l(\omega) + \text{higher order terms.} \tag{1}$$

The lowest order term describes an electric dipole source. Symmetry considerations show that $\chi^{(2)}$ is zero in a centrosymmetric medium, thus limiting electric dipole radiation to the interfaces where inversion symmetry is broken. The magnetic properties of the material are included by introducing a magnetization dependent nonlinear susceptibility tensor: $\chi^{(2)}_{jkl}(M)$, as was suggested by Ru-Pin Pan et al. [8]. A symmetry analysis shows that we can distinguish tensor elements that are respectively even and odd in the magnetization [8]. Thus we may write:

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$$E(2\omega) = (\chi_+^{(2)}(M) + \chi_-^{(2)}(M)) : E(\omega)E(\omega), \quad (2)$$

where $\chi_+^{(2)}(M)$ and $\chi_-^{(2)}(M)$ are linear combinations of the tensor elements, with $\chi_\pm^{(2)}(-M) = \pm\chi_\pm^{(2)}(M)$.

We now define the relative magnetic effect for MSHG as:

$$\rho = \frac{I(2\omega, M^+) - I(2\omega, M^-)}{I(2\omega, M^+) + I(2\omega, M^-)}, \quad (3)$$

where $I(2\omega, M^+)$ and $I(2\omega, M^-)$ are the SH-intensities for opposite directions of the remanent magnetization.

In this paper we use magnetization induced second harmonic generation (MSHG) in combination with MOKE to study thin Co films of thicknesses between 1 and 20 monolayers (ML) and Cu/Co multilayers grown on a Cu(001) substrate. We find that the relative magnetic effect ρ, as determined from MSHG reaches a constant value at 3 ML, in contrast to the MOKE signal that increases linearly with thickness. From their different behavior with Co film thickness we get direct information about the surface/interface sensitivity of MSHG. Modifying the vacuum/Co interface by carbonmonoxide adsorption yields the relative contributions of the vacuum/Co and the Co/Cu(001)-substrate interfaces. These results can be directly compared with the results obtained from the Cu/Co/Cu(001) system. Both the MOKE and the MSHG data can be described accurately by a multiple reflection model.

2. Sample preparation and experiment

We have studied two sets of samples:

(i) Co/Cu(001) with varying cobalt film thickness.

(ii) 10 ML Cu/Co/Cu(001) with varying cobalt film thickness.

The samples were prepared in a UHV system with a base pressure of 5×10^{-11} Torr. Cleaning the Cu(001) substrate consisted of several cycles of Ar^+ sputtering followed by annealing at 600°C. The Co films were grown at a rate of approximately 1 ML/min, while the Cu(001) substrate was kept at a temperature of approximately 100°C. The Cu overlayers were grown at the same rate, while the substrate was kept at about 70°C. It has been shown that both the growth of Co on Cu(001) and the growth of Cu on Co(001) is pseudomorphic [4]. Epitaxial growth was verified for every film by monitoring the (0,0) medium energy electron diffraction (MEED) spot intensity while depositing. After preparation the film quality was checked by Auger electron spectroscopy (AES); all contaminations were below 1 at%, except carbon, which was typically 2–3%.

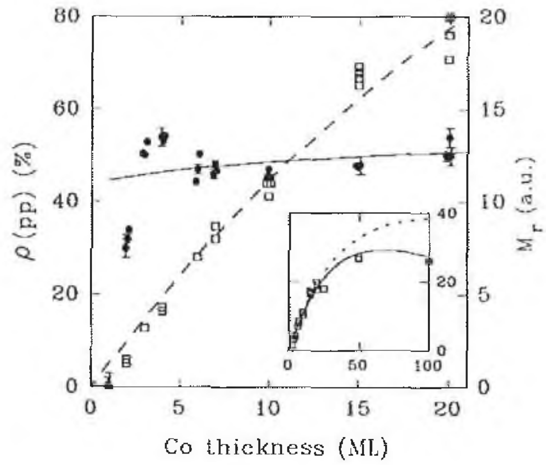

Fig. 1. Co film thickness dependence of the relative nonlinear magnetic effect ρ(pp) and the MOKE amplitude M_r in Co/Cu(001); dots/solid line - experimental ρ(pp) data and theoretical fit; open symbols/dashed line - MOKE data and fit. The inset shows the MOKE data and fits for thicker Co films; dashed line: Lambert Beer's law, solid line: multiple reflection model.

For the SHG experiments we used the output of a Ti:Sapphire laser at 800 nm. The pulse intensity of the incoming beam was about 16 μJ cm^{-2}. At an angle of incidence of 35°, we have studied the $p_{in}p_{out}$ polarization combination (i.e. both fundamental and second-harmonic beams are polarized in the plane of incidence). No analyzer was needed, because the s-polarized SH-output was negligible, in accordance with theory [8,11,12]. The magnetization was parallel to the (110) direction of the Co film, the easy axis of the film, and perpendicular to the optical plane. The MOKE measurements were done in the longitudinal configuration.

3. Results and discussion

Fig. 1 shows the relative magnetic effect ρ and the MOKE amplitude M_r as a function of the Co thickness for Co on Cu(001). The difference between the

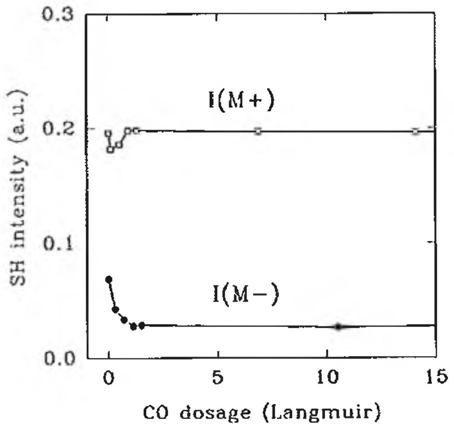

Fig. 2. The pp SH-intensities of a 7 ML Co-film on Cu(001) for positive and negative magnetic saturation as a function of CO-dosage. The signals at 15 L CO remain constant until at least 40 L.

two sets of data is striking: M_r increases almost linearly with Co thickness, wheras ρ hardly changes after 3 ML of Co. The origin of these different behaviours lies in the probing depth of the two techniques. MOKE is a bulk probe, and the total Kerr rotation is proportional to the amount of material, taking absorption losses into account. On the other hand MSHG is an interface sensitive probe, that is independent on the bulk film thickness. The deviation of the linear thickness dependence of M_r is a result of the absorption in the thin films. Usually this is taken into account by a Lambert–Beer's type of analysis. A more accurate approach is to use a multiple reflection model, similar to Moog et al. [13] and Lissberger et al. [14]. For thicknesses below 20 ML, both approaches give an equally good fit. Above 20 ML, the effects of multiple reflections start to play a noticable role, as can be seen in the inset of Fig. 1. For the calculation, we used the bulk indices of refraction as listed in Ref. [15]. From the close agreement between experiment and calculation, we can conclude that the MOKE results can accurately be described by bulk refractive indices for Co thicknesses above 3 ML.

To analyze the MSHG results one should realize that there are two interfaces that contribute to the SH signal: the Co/Cu and the vacuum/Co interface. To determine the relative strength of the SH-signal from these two interfaces we measured the SH-signal from a Co film on Cu(001) as a function of carbonmonoxide exposure as gas adsorption is known to strongly reduce the SHG from metal surfaces [16]. Fig. 2 shows the pp SH-intensities of a 7 ML Co film on Cu(001) for positive and negative magnetization as a function of CO dosage. We observe that the signals change until a dosage of 1 langmuir (1 L = 10^{-6} Torr s), whereafter they become constant until at least 40 L. The original value of $\rho \sim 45\%$ has increased to $\rho \sim 70\%$. Comparable effects have been observed on adsorbing O_2 and for different Co film thicknesses. We have observed that a dosage of a few langmuirs of O_2 to magnetized Ni(110) and Fe(110) crystals reduces the pp SH-signal, generated from a 532 nm Nd-YAG beam, by up to a factor of 20 depending on initial cleanliness. We therefore come to the reasonable assumption that the CO eliminates all SH-contributions by the Co/vacuum-interface. The total response can then be calculated using a multiple reflection model that includes the relevant nonlinear tensor elements and the boundary conditions for nonlinear sources at the interfaces [12]. The line in Fig. 1 shows the result of such a fit, that is seen to give a good description of the experimental results above 5 ML. For the analysis we used one odd and one even term for each interface.

For the Co/Cu interface we find a ratio of

$$\chi^{(2)}_{-,\mathrm{Co/Cu}}/\chi^{(2)}_{+,\mathrm{Co/Cu}} = 0.89.$$

This means that the magnetic contributions are of the same order of magnitude as the nonmagnetic ones. This accounts for the large relative effects ($\rho > 40\%$). Comparing the even and odd elements at the vacuum/Co and Co/Cu interfaces gives:

$$\chi^{(2)}_{-,\mathrm{vac/Co}}/\chi^{(2)}_{-,\mathrm{Co/Cu}} = -1.4$$

and

$$\chi^{(2)}_{+,\mathrm{vac/Co}}/\chi^{(2)}_{+,\mathrm{Co/Cu}} = -1.9.$$

This indicates that at the vacuum/Co and Co/Cu interfaces the amplitudes of both even and odd tensor elements are comparable. These ratios are found for any combination of one even and one odd tensor element [17].

These results show that above 5 ML both MOKE and MSHG results can accurately be described by a multiple reflection model, using bulk indices of refraction. The strong thickness dependence of the MSHG results below 5 ML can of course not be described by

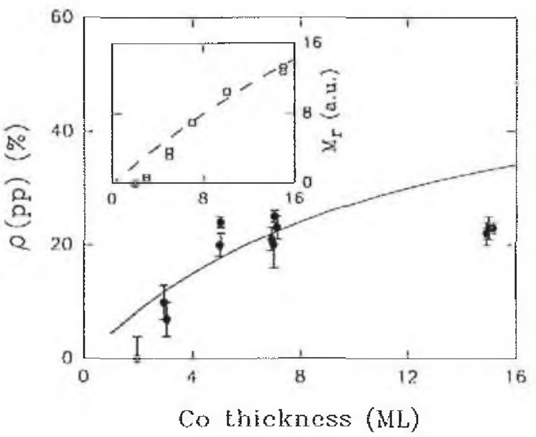

Fig. 3. Calculated and experimental Co film thickness dependence of ρ(pp) in Cu/Co/Cu(001); dots – experimental data; solid line – calculation based on fitting parameters of Fig. 1. The inset shows the experimental and calculated MOKE amplitude.

such an approach, as it takes at least 2 ML to define an interface.

The solutions for the Co/Cu interface can be used directly in the Cu/Co/Cu(001) trilayers to calculate ρ(pp) of these systems. Fig. 3 shows the experimental Co thickness dependence of ρ for Cu/Co/Cu(001) plus the theoretical prediction based on the results of Fig. 1. The only fitting parameter was the nonmagnetic nonlinear contribution of the Cu/vacuum interface, that was not present before. The agreement between experimental and predicted dependence is quite good. More importantly, Fig. 3 shows that such a multiple reflection model is *required* in order to understand the MSHG response of such a multilayer at all. Critical is that the Cu/Co/Cu trilayer is very symmetric and because of the mirror symmetry $\chi^{(2)}_{Cu/Co} = -\chi^{(2)}_{Co/Cu}$. Thus, without taking the multiple reflections into account, that yield different total fields at the two interfaces, their total response and thus ρ would have been zero. (Here we ignore possible differences that may exist between the absolute values of the two interface contributions, due to differences in the local structure.) Fig. 3 also shows the MOKE data for this system, with a theoretical fit including multiple reflections directly calculated from the Co/Cu results, using no adjustable parameters.

4. Conclusion

Epitaxially grown fcc Co/Cu(001) multilayers were studied by the magneto-optical Kerr effect (MOKE) and magnetization induced second harmonic generation (MSHG).

The results of the longitudinal MOKE experiments could be described by a simple model, that uses the well-established thin-film additivity law for the Kerr effect, while accounting for absorption and multiple reflections of the light in the multilayer. The model uses bulk refractive indices for the materials, and indicates that these indices accurately describe the optical properties of films above about 3 ML. In contrast to MOKE the relative magnetic effect in MSHG is nearly constant for Co films thicker than 3 ML. Similar behavior is observed in experiments on Cu/Co/Cu trilayers. The results clearly proof the interface sensitivity of MSHG. The multiple reflection theory of Ref. [12] gives a satisfying description of the Co film thickness dependence in the MSHG experiments on Co/Cu(001) and Cu/Co/Cu(001), and proves the importance of the local electromagnetic fields. These results show that the combination of linear and nonlinear optical experiments yields a powerful tool to study thin magnetic multilayers. Future plans include the application of spectroscopic MSHG, to study the (magnetic) interface states.

Acknowledgements

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[17] Recently, we have repeated these experiments with a new system (R. Vollmer, A. Kirilyuk, H. Schwabe, J. Kirschner, H.A. Wierenga, W. de Jong and Th. Rasing, to appear in J. Magn. Magn. Mater.). The results are in perfect agreement with those reported here, except for the fact that the background CO contamination was somewhat lower. This resulted in a larger Co/vacuum contribution and thus a smaller total magnetic effect, in accordance with the analysis given here.