Probing structure and magnetism of CoNi/Pt interfaces by nonlinear magneto-optics

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Magnetic CoNi/Pt interfaces are studied as a function of their preparation conditions by magnetization-induced second-harmonic generation (MSHG) measurements. A detailed method has been developed to decompose the total MSHG response into magnetic and crystallographic contributions for each interface. Although the bulk magnetism of the CoNi film (3 nm thick) shows only a subtle dependence on the sputtering Ar pressure, the interfaces appear to be dramatically affected. It can be shown that the crystallographic part probes the increase in the interface roughness while the magnetic one clearly reveals a maximum in the in-plane magnetization of the interface.
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The magneto-optical Kerr effect (MOKE) has been established as a powerful and simple technique to study ultrathin magnetic films and multilayers, with a magnetic sensitivity down to less than a monolayer. Recently, a complementary nonlinear magneto-optical tool has been developed to allow for the study of film interfaces: magnetization-induced second-harmonic generation (MSHG). It has been shown to combine extreme surface/interface sensitivity[1,2] with very large magneto-optical effects.[3]

To make devices out of magnetic multilayers, like giant magnetoresistance and spin-tunneling-based sensors as well as magnetic and magneto-optical recording media, a knowledge of the interface structure is essential. Many techniques used for these studies at present require either very complicated equipment (like synchrotons for x-ray circular magnetic dichroism) or at least should be carried out in UHV conditions. The study of buried interfaces is especially difficult, and here an optical technique can be very helpful due to the relatively large penetration depth.

In this letter, we show how the interface-specific magneto-optical MSHG technique can be used to correlate the sputtering-induced changes in the interface morphology with the changes in the interface magnetic properties of the magneto-optical recording material CoNi/Pt. We developed a method that allows the unambiguous determination of the nonlinear magneto-optical tensor $\chi^{(2)}$ for each interface of the multilayer structure. The relation between the interface roughness and intermixing and the crystallographic contribution to MSHG is demonstrated. We find that the interface magnetization has a dramatic dependence on the Ar sputtering pressure, while the bulk magnetism is relatively unperturbed.

In the electric-dipole approximation, SHG is expressed through the second-order polarization $\mathbf{P}(2\omega)$ induced in a medium by an incident electromagnetic wave $\mathbf{E}(\omega)$:

$$P_i(2\omega)=\chi^{(2)}_{ijk}E_j(\omega)E_k(\omega). \tag{1}$$

The third-rank polar tensor $\chi^{(2)}$ vanishes in any centrosymmetric medium. Hence, a symmetry breaking surface or interface is a source of SHG, giving rise to the extreme interface sensitivity of the technique. The presence of a magnetization does not influence the *bulk* inversion symmetry but does change the symmetry of the *interface*, making the magnetic probing also interface sensitive.[4] For an isotropic [or (001)] surface in the transversal magneto-optical configuration ($\mathbf{M}\|y$, xz is the plane of incidence) the nonlinear magneto-optical tensor $\chi^{(2)}$ can be written as

$$\chi^2=\begin{pmatrix} \mathbf{xxx} & \mathbf{xyy} & \mathbf{xzz} & 0 & xzx & 0 \\ 0 & 0 & 0 & yzy & 0 & \mathbf{yxy} \\ zxx & zyy & zzz & 0 & \mathbf{zzx} & 0 \end{pmatrix}. \tag{2}$$

The elements shown in bold are *odd* in the magnetization (roughly proportional to it[5]). Thus, the nonlinear magneto-optical properties of an isotropic interface are characterized by ten (complex) numbers.

A crucial challenge is how to derive $\chi^{(2)}$ from the experimental data. For this, multiple scattering calculations (based on a transfer matrix technique[6,7]) are used to determine the electric-field $\mathbf{E}(\omega)$ for every interface; the same calculation procedure is used afterwards to compute the outgoing SHG intensity. The tensor components $\chi_i^{(2)}$ for each interface i are the fitting parameters to describe the experimental data.

For the MSHG measurements, a pulsed laser beam from a Ti–sapphire laser (82 MHz×100 fs pulses) with a wavelength of 840 nm was focused onto the sample. After proper filtering, the outcoming specular second-harmonic light was detected with a photomultiplier. The asymmetry of the MSHG signal $A=(I^+-I^-)/(I^++I^-)$ (where $I^\pm$ is the

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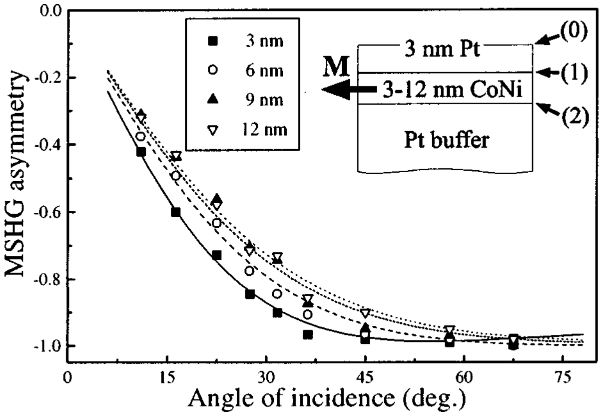

FIG. 1. Angle of incidence dependencies of the MSHG asymmetry A for samples with different CoNi layer thicknesses (indicated in the figure). Lines are the theoretical fits to the experimental points using one single set of tensor elements.

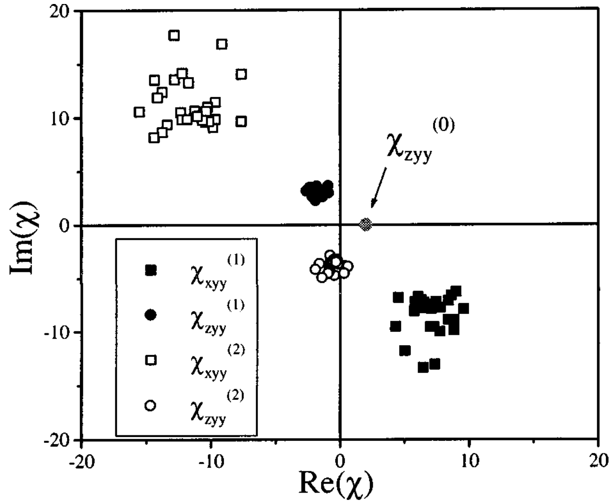

FIG. 2. The nonlinear magneto-optical tensor components determined from the fits of the experimental data of Fig. 1.

MSHG intensity for the magnetization up or down, respectively) was measured in the specular direction as a function of the angle of incidence.

The samples were prepared in a computer-controlled sputtering system, base pressure of 5×10^{-8} mbar, with argon as a sputtering gas. The deposition rates were kept low (1.7–2.0 Å/s for Pt and 0.4–0.6 Å/s for CoNi), to assure a smooth layer growth and a good control of the layer thickness. A 40 nm thick Pt buffer layer was deposited on a Si(001) substrate followed by a magnetic CoNi layer (thickness varied between 3 and 12 nm) and covered by a 3 nm thick Pt cap layer. Such samples were prepared at different Ar pressures (between 4 and 36 μbar). It was found that the magnetic interface properties considerably depend on the growth conditions, in particular, on the Ar pressure used for sputtering.

In order to determine the $\chi^{(2)}$ tensor for one given interface quality, a set of samples was used with different magnetic layer thicknesses, prepared under exactly the same conditions ($p_{\mathrm{Ar}}=12\ \mu$bar). We may, therefore, assume that the $\chi^{(2)}$'s are the same for the different samples, and the only thing that is changed are the local optical fields at the interfaces, due to absorption and multiple scattering.

The results of the measurements together with the fitting curves are shown in Fig. 1 for the $S_{\mathrm{in}}P_{\mathrm{out}}$ polarization combination. The number of fitting parameters is determined by the polarization used. Thus, for $S_{\mathrm{in}}P_{\mathrm{out}}$ we fix $\mathbf{zyy}_0$ (neither the absolute intensity nor the optical phase of MSHG is taken into account), hence, $\mathbf{zyy}_{1,2}$ and $xyy_{1,2}$ are the only components left to be determined. This gives eight parameters (2 interfaces×2 complex components) to fully describe these data. The linear (magneto-) optical parameters of similar samples were measured previously.[8] The uniqueness of the fits was checked for both $S_{\mathrm{in}}P_{\mathrm{out}}$ and $P_{\mathrm{in}}P_{\mathrm{out}}$ polarization combinations by randomizing the initial choice of the fit parameters. Figure 2 shows the $\chi^{(2)}$ tensor components obtained from the fits of Fig. 1. The convergence of the parameters is evident. An interesting point is that the tensor components show different signs for the subsequent magnetic layer interfaces (1) and (2). This is an independent experimental confirmation of a strict requirement from symmetry and provides a strong support for the model used in the calculations. Also, the crystallographic and magnetic contributions to $\chi^{(2)}$ appear to be of the same order of magnitude, in strong contrast to the linear case.

To determine the dependence of $\chi^{(2)}$ on the interface quality, the sample with a 3 nm thick CoNi layer was measured for different Ar sputtering pressures. The assumption was then made that all tensor components changed in a similar way, i.e., the scaling parameters $\mathscr{M}$ and $\mathscr{C}$ were defined as

$$\chi^{(2)}_{\mathrm{magn}}(p_{\mathrm{Ar}})=\mathscr{M}(p_{\mathrm{Ar}})\chi^{2}_{\mathrm{magn}}(p_0), \tag{3}$$

$$\chi^{(2)}_{\mathrm{cr}}(p_{\mathrm{Ar}})=\mathscr{C}(p_{\mathrm{Ar}})\chi^{(2)}_{\mathrm{cr}}(p_0), \tag{4}$$

with $p_0=12\ \mu$bar. To fit the data for any new sample, only the two complex parameters $\mathscr{M}$ and $\mathscr{C}$ are used (actually, this only gives three parameters in total because one phase can still be fixed). The possibility to fit the data for any Ar pressure (see Fig. 3) supports our assumption that all $\chi^{(2)}$'s are changed in a similar way.

The parameters $\mathscr{M}(p_{\mathrm{Ar}})$ and $\mathscr{C}(p_{\mathrm{Ar}})$ represent the dependence of the nonlinear magneto-optical interface properties on the interface structure (controlled via the sample preparation conditions). The value of $\mathscr{C}$ is proportional to the crystallographic contribution to the MSHG, expressed via

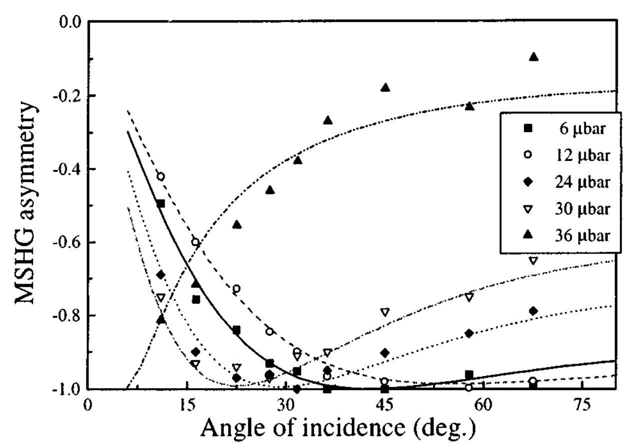

FIG. 3. MSHG asymmetry A as a function of angle of incidence for samples prepared at different Ar pressures (indicated in the figure).

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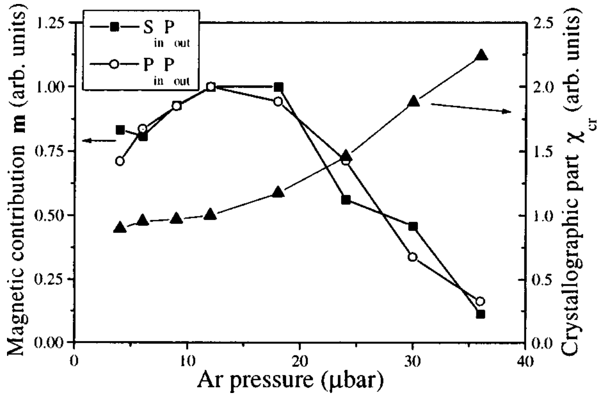

FIG. 4. The ratio $\mathbf{m}=\mathscr{M}/\mathscr{C}$ as a function of the sputtering Ar pressure independently derived for the two different polarization combinations (indicated in the figure).

the local symmetry breaking induced by the interface. It is incorrect to say, however, that $\mathscr{M}$ represents the purely magnetic part of MSHG. Indeed, all the ''magnetic'' elements of $\chi^{(2)}$ are only nonzero in the presence of the crystallographic symmetry breaking, i.e., the same factor influences both χ_{cr} and χ_{magn}. Hence, one may write $\mathscr{M}\propto\mathscr{C}\cdot\mathbf{M}$.

To extract information on the interface magnetic properties from our results, we take the ratio $\mathbf{m}=\mathscr{M}/\mathscr{C}$. In Fig. 4, **m** is plotted as a function of the sputtering Ar pressure for the $S_{in}P_{out}$ and $P_{in}P_{out}$ polarizations. The precise coincidence of the **m** dependency for both polarization combinations once again supports the model used for the derivations.

Figure 4 shows that the crystallographic contribution $\chi_{cr}\propto\mathscr{C}$ increases rapidly above 15 μbar, while staying almost constant below this pressure. The increase of χ_{cr} indicates an increasing interfacial roughness for higher sputtering pressures. Though the crystallite size is known to stay constant in the whole pressure range, the crystallites may become slightly misoriented.[9] This increase of χ_{cr} due to the increasing interface roughness can schematically be understood as being due to the increase of the effective surface area of the interface. For stronger roughnesses, other mechanisms may play a role.[10]

In contrast to the crystallographic one, the magnetic contribution **m** shows a clear maximum at pressures of 15–20 μbar. At very low Ar pressures the interface layers become slightly intermixed due to the high energies of sputtered atoms. This intermixing hardly affects the crystallographic part of MSHG but clearly suppresses the magnetic one. Note that the maximum in the interface magnetization does not have to coincide with the sharpest interface. Evidently, the drop of **m** for large p_{Ar} is related to a decreasing in-plane magnetic moment of the rough interface. A possible explanation here is that the increasing roughness changes the local coordination of the Co atoms, which may even lead to an out-of-plane lifting of the *local* interface magnetic moments. This explanation is supported by our observation of a specular *S*-polarized MSHG output at higher Ar pressures. Such a MSHG yield can only be nonzero in the presence of a perpendicular (out-of-plane) magnetization component. In addition, polar MOKE hysteresis loops also show a small remanence ($\leqslant$10% of M_s) for the sample sputtered at P_{Ar}=36 μbar, confirming our MSHG results.

To summarize, we have shown that nonlinear magneto-optics is clearly able to follow the (subtle) changes in the interface structure, both crystallographic and magnetic. For the case of Pt/CoNi/Pt, we found the optimum sputtering pressure that yielded a maximum in-plane interface magnetization with only a small change in interface morphology. With further increase of the sputtering pressure, the interface roughness clearly increases while the in-plane interface magnetic moment decreases. The latter appears to be accompanied by the appearance of an out-of-plane magnetization component at the interface. It should be underlined that for all studied samples, no difference in magnetization was observed with either MOKE or vibrating-sample magnetometer. Microscopic theory is required for the further development of the MSHG method in order to make an unambiguous correlation of the observed dependence of the magneto-optical response with the magnetization (exchange constant, spin-orbit coupling, etc.) at the interface.

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