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Giant nonlinear Kerr effects

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Abstract

The nonlinear second harmonic reflectivity of magnetic surfaces is found to display longitudinal nonlinear Kerr rotations that are three orders of magnitude larger than their linear equivalent. In the transversal geometry, similar enhancements are observed for the intensity changes. The results are in good agreement with recent theoretical predictions. For ultra-thin Au overlayers on Fe, the nonlinear Kerr effect is extremely sensitive to the interface roughness.

The nonlinear optical technique of second harmonic generation (SHG) has been developed into a successful surface and interface probe, deriving its sensitivity from the symmetry breaking at interfaces between centrosymmetric media [1]. The presence of a magnetization $\boldsymbol{M}$ will alter the symmetry properties of the nonlinear optical response tensor, without affecting the inversion symmetry (as $\boldsymbol{M}$ is an axial vector) [2,3]. Recently it has been shown that SHG from magnetic surfaces or interfaces indeed leads to strong magnetic effects [4-6], with a magnetic dichroism that can be over 50%. Theoretically, it was also predicted that SHG from magnetic surfaces should lead to a nonlinear magneto-optical Kerr effect (NOMOKE) that is about an order of magnitude larger than its linear counterpart (MOKE) [7]. In this paper we will show that this enhancement can even be as large as three orders of magnitude, due to the difference in the symmetry properties of the nonlinear and linear optical response. We will also indicate how this may lead to a very sensitive probe for interface roughness.

For our experiment we used the 835 nm output of a Ti:sapphire 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 which was mounted in between the poles of an electromagnet. The magnetization could be switched in the y and in the x direction (see inset Fig. 1). Polarization control of both incident fundamental and reflected SH light was achieved by means of polarizors. Appropriate filtering was used before the signal was detected by a photomultiplier in combination with a lock-in amplifier. We did our experiments in air on the 100 surfaces of single-crystal Fe whiskers which were capped with an MBE grown thin Au or Cr film.

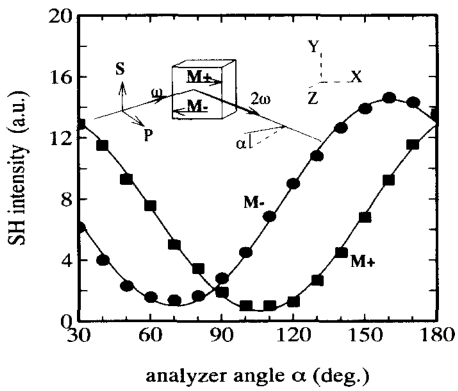

Fig. 1. Output polarization dependence of SHG reflection of the Fe/Cr sample on the s-input longitudinal configuration. The inset shows the experimental configuration.

Fig. 1 shows the polarization dependence of the SH signal from the Fe/Cr sample for an s-polarized input at an angle of incidence of 37° and for $\boldsymbol{M}$ along x and $-x$, respectively. The difference between the two minima of the curves corresponds to $2\Phi_K^{(2)} = 36°$, i.e. a nonlinear Kerr rotation $\Phi_K^{(2)}$ of 18°.

The SH fields are related to the incident laser fields through the nonlinear susceptibility $\chi_{ijk}^{(2)}$ [1]. For s-polarized incident light (i.e. parallel to y-axes), only the elements χ_{iyy} will be of importance (with $i = x, y, z$). In the longitudinal configuration with s-input we then only have two tensor elements: χ_{yyy}^{-}, that is odd in the magnetization and gives rise to s-polarized SHG and χ_{zyy}^{+}, that is even in the magnetization and gives rise to p-polarized SHG. From this it follows that the SHG polarization ellipses for $\pm\boldsymbol{M}$ are each other's mirror image in the plane of incidence,

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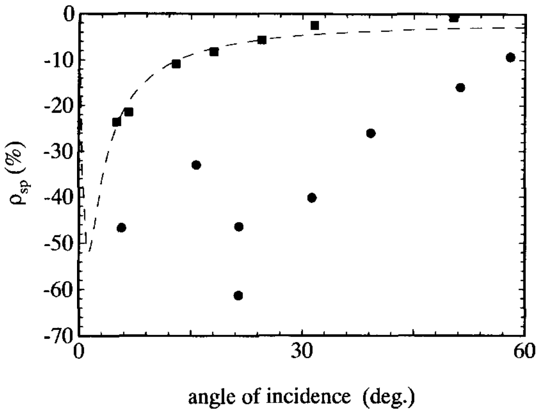

Fig. 2. Magnetic contrast ρ for the two different Fe/Au samples of the s-polarized input in the transversal configuration as a function of the angle of incidence. The dashed line is a theoretical fit for a smooth surface.

which defines a nonlinear Kerr angle $\Phi_K^{(2)}$, analogous to the linear Kerr rotation [8]. For the Fe/Au sample we find a maximum Kerr angle of 80° at an angle of incidence of 6°. This corresponds with an enhancement of more than a factor 10^3, compared with the value for the linear Kerr rotation of 0.03° as obtained in the same experimental set-up, using the fundamental incident beam.

To compare our results with the theoretical predictions we also measured the nonlinear Kerr rotation for a p-polarized input. At an angle of incidence of 45° we found $\Phi_K^{(2)} = 1.2°$ in excellent agreement with the theoretical prediction of 1.4° from Ref. [7].

In the transversal geometry, only magnetization induced intensity changes are expected (magnetic dichroism), as symmetry does not allow a Kerr rotation [8]. We used the transversal approach to study the intrinsic interface sensitivity of our NOMOKE technique. Fig. 2 displays the results for two different Fe/Au samples with 4 ML of Au, grown under different MBE conditions. The interface quality was checked with in situ STM studies [9]. The squares are for a sample grown at 493 K, showing a smooth Fresnel-like dependence of ρ on the angle of incidence. The dots are for the sample grown at 468 K, displaying a very irregular pattern. STM studies indicated a smooth surface for the first sample and the presence of holes in the caplayer of the second, showing the sensitivity of our nonlinear optical technique for this kind of surface roughness. A more quantitative study of this effect is in progress.

In conclusion, we have observed giant magnetic effects in the nonlinear optical response of magnetic interfaces. For the s-input longitudinal configuration we find an enhancement up to three orders of magnitude for the nonlinear relative to the linear Kerr rotation. By varying the angle of incidence this angle may be tuned at will between 0° and 90°. This nonlinear Kerr effect is an extremely sensitive interface probe, as demonstrated with results from differently grown Fe/Au samples.

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