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Nonlinear magneto-optical Kerr measurements on Fe(110)

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Abstract

For the clean Fe(110) surface Kerr angles close to 90° are observed in the frequency doubled light and for the oxygen covered surface asymmetries up to 100%. In the wavelength range from 740 to 840 nm a sizable amount of the second harmonic light is possibly generated by the bulk quadrupole like nonlinear polarization especially for p-polarized incident light. For s-polarized incident light a strong surface contribution is found.

Keywords: Iron; Low index single crystal surfaces; Second harmonic generation

1. Introduction

It is well known that for cubic crystals there is no second harmonic generation (SHG) in the bulk as far as electric dipole contributions are considered. At the surface the inversion symmetry is broken and therefore SHG is possible [1–3]. Recently this method was applied to ferromagnetic samples and it was demonstrated that the SHG yield depends on the magnetization of the sample [4–6]. Together with the surface sensitivity of SHG itself it may be used as a surface and interface sensitive probe of magnetism at surfaces and in thin films.

To the lowest order the total surface nonlinear polarization at 2ω induced at the surface is given by:

$$P_i(2\omega) = \sum_{jk} \chi_{ijk} E_j(\omega) E_k(\omega), \qquad (1)$$

where $E(\omega)$ is the electric field amplitude of the incident light, χ_{ijk} the third rank tensor of the nonlinear susceptibility. The number of independent tensor elements of χ is reduced by the symmetry of the surface. A magnetization M reduces the symmetry and consequently more tensor elements become nonzero. These additional tensor elements are odd with respect to reversal of the magnetization direction while the former do not change their sign. The physical origin of the new tensor element contributions can be understood in similar terms as for the linear Kerr effect: The spin–orbit interaction causes a rotation of the direction of the (nonlinear) polarization induced in the medium by the incident light with respect to the polarization in the nonmagnetic state. Depending on the orientation of the magnetization with respect to the optical plane two cases can be distinguished: A magnetization parallel to the optical plane leads to a rotation of the outgoing (frequency doubled) light while a magnetization perpendicular to the optical plane reduces or enhances the SH intensity due to the reduced or enhanced projection of the nonlinear polarization onto the direction of the E-vector of the outgoing light. In analogy to the linear Kerr effect the former is called longitudinal

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and the latter is called transversal nonlinear Kerr effect. Note, however, there is an important difference between the linear and the nonlinear Kerr effect: Even in the nonmagnetic state the polarization of the outgoing (frequency doubled) light is not necessarily in the same direction as that of the incident light.

For a fixed geometry the outgoing SH intensity can be expressed as a function of an effective nonmagnetic, χ_{nm}, and magnetic, χ_m, matrixelement:

$$I^{\pm}(2\omega) \propto |\chi_{nm} \pm \chi_m|^2, \tag{2}$$

with the + and − for the magnetization in opposite directions. Defining an asymmetry $A := (I^+(2\omega) - I^-(2\omega))/(I^+(2\omega) + I^-(2\omega))$ one gets for the transversal geometry:

$$A_t = \frac{2|\chi_m/\chi_{nm}|}{1 + |\chi_m/\chi_{nm}|^2} \cos\varphi, \tag{3}$$

with φ the phase difference between the even and odd part of the nonlinear susceptibility. For the longitudinal geometry it can be shown that for the approximation given in Eq. (1) the nonmagnetic part of χ results in an entirely p-polarized SH light amplitude while the magnetic part is entirely s-polarized independently of the input polarization. By placing an analyser in the outgoing beam path in this case both, the relative amplitude $|\chi_m/\chi_{nm}|$ and the phase φ can be determined by measuring the asymmetry A as defined above as a function of the analyser angle. In this case the asymmetry is given by:

$$A_l(\alpha) = \frac{2|\chi_m/\chi_{nm}|\tan\alpha}{1 + |(\chi_m/\chi_{nm})\tan\alpha|^2} \cos\varphi. \tag{4}$$

A polarizer angle $\alpha = 0$ corresponds to p-polarization of the SH light.

It has been shown in the past by several authors that for metals in general it is not justified to exclude contributions from the bulk [3]. Expressions for the SHG from surfaces of cubic crystals including these contributions to the lowest order have been derived by Sipe et al. [8] in the nonmagnetic case. For the magnetic case Koopmans et al. [9] performed a symmetry analysis for an isotropic medium which is equivalent to the symmetry of a cubic crystal if the magnetization is along a [100] direction. Without giving a complete analysis we state that Eq. (4) still describes the asymmetry in the longitudinal geometry but now with different values for χ_m and χ_{nm} including bulk and surface terms. For the transversal geometry Eq. (3) remains true in the same sense.

In this paper we present results on the nonlinear Kerr effect for different geometries and polarization combinations of the incident fundamental and outgoing SH light. We will show that surface sensitivity is not generally guaranteed and significant contributions of SHG due to higher-order terms than the electric dipole term are important for certain geometries and polarization combinations of the incident and SH light.

2. Experiment

An elliptically shaped Fe(110) sample of 2 mm thickness was clamped onto a soft iron yoke so that the magnetic field is along the [100] direction, the easy axis of bcc iron. A current of a few 100 mA running through a coil wound around the iron yoke was sufficient for magnetization saturation. The crystal together with the iron yoke could be rotated about the surface normal. The crystal was cleaned in ultra-high vacuum (UHV), base pressure 4×10^{-11} mbar) by repeated 500 eV Ar^+ bombardment at nearly grazing incidence followed by annealing to 850 K. The carbon build up during sputtering was removed by oxygen titration at 750 K as described in Ref. [7]. After this cleaning procedure all contaminations were below 1 at% as checked by Auger electron spectroscopy. The optical set-up was similar to that described in Ref. [5]. The light of a femtosecond Ti-sapphire laser was focused down to ~ 50 μm diameter onto the sample through a fused silica window of the UHV chamber. Using a Babinet–Soleil compensator as $\lambda/2$ wave plate the polarization axis of the linearly polarized light of the laser could be rotated. The frequency doubled light generated at the surface of the Fe crystal left the UHV chamber through a second window and was detected by a photomultiplier. The fundamental light reflected from the Fe surface was blocked by Schott BG36 colored glass filters. Possibly generated SH light in the incident beam was absorbed by a OG570 colored glass filter. The polarization of the generated SH

light could be measured by placing a polarizer in the outgoing beam path.

3. Results

Fig. 1 shows the SH intensity (bottom panels) as a function of the O_2 exposure for the longitudinal (left panels) as well as for the transversal geometry (right panels) together with the resulting asymmetries (top panels) as defined above. The open and solid symbols in the bottom panels are for the magnetization of the sample in opposite directions while the dashed line is the arithmetic average of the two curves. The incident angle of the light with respect to the surface normal was about 38° as for all measurements described in this paper. For the longitudinal geometry the analyser was set to $\alpha = 70°$ and for the transversal geometry the total SH intensity was recorded. Using Auger ratios of the O(KLL) (510 eV) and the Fe($L_{2,3}$VV) (651 eV) peaks and comparing with the results of Ref. [10] we find an oxygen coverage of

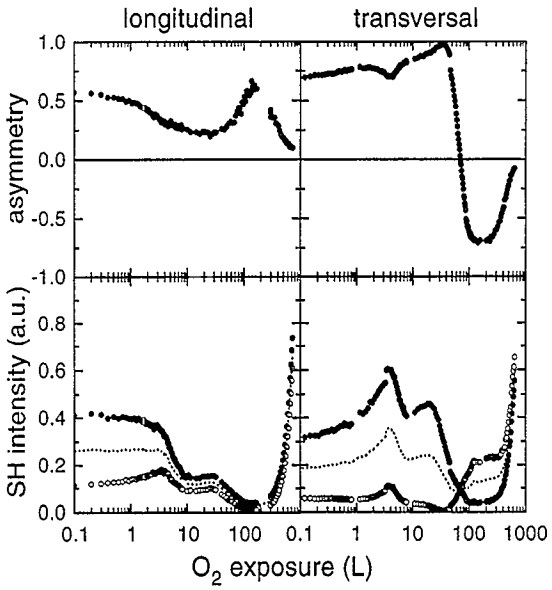

Fig. 1. SH intensity (two bottom panels) and the measured asymmetry (two upper panels) as a function of the O_2 coverage for the longitudinal (left panels) and the transversal (right panels) geometry. For the measurements in longitudinal geometry the analyser polarizer was set to an angle $\alpha = 70°$ with respect to p-polarization. For the transverse geometry the total SH output is shown. The solid and open circles in the two lower panels denote the SH intensity for the magnetization in two opposite directions while the dashed line represents the average SH intensity.

0.25 for 3 Langmuir (L) and a full monolayer (ML) for an exposure of 24 L. At higher coverage oxide formation sets in. Surprisingly the average SH intensity does not drop with the O_2 exposure for low coverage but stays constant up to an oxygen exposure of 3–4 ML or even increases for the transverse geometry. There are two known oxygen superstructures commonly denoted as $c(2 \times 2)$ and $c(3 \times 1)$ at a coverage of 0.25 and 0.33 ML. These superstructures are maximally developed at 3 and ~ 5 L [10] and are not resolved as individual peaks in Fig. 1. A second maximum at around 25 L seen in both geometries coincides nearly with the completion of the full monolayer coverage. On further oxygen exposure the chemical structure of the surface becomes quite complicated because of oxide formation [10] and we will not further discuss this range. Looking at the asymmetries of the SH intensity one finds that for the longitudinal geometry the asymmetry decreases with increasing oxygen coverage while for the transversal geometry it increases up to 100% at full monolayer coverage. There is a small dip around 3 L mainly caused by an increase of the average SH intensity due to the oxygen superstructure. At higher oxygen exposure oxide formation quite drastically changes the asymmetry.

For the longitudinal geometry the ratio $|\chi_m/\chi_{nm}|$ can be derived from measured asymmetries $A_l(\alpha)$ using Eq. (4). The result is shown in Fig. 2 for p-polarized incident light as in Fig. 1 (solid symbols) and for s-polarized incident light (open symbols) for a wavelength of 790 nm of the incident light. Clearly, one observes only little influence of the oxygen on $|\chi_m/\chi_{nm}|$ for p-polarized incident light while there is a strong decrease of this ratio for the s-polarized incident light. We note that for p-polarized incident light the p-polarized SH signal shows a maximum around 3 L as pronounced as in the transversal geometry while the s-polarized part drops monotonously. Obviously the magnetization induced part of the SH light is not affected significantly by the oxygen superstructures.

The exact behavior at very low coverage is complicated by the preparation of the clean Fe surface. Very low coverage of oxygen changes the ratio $|\chi_m/\chi_{nm}|$ quite dramatically. For surfaces with a small carbon contamination the SH light for s-input polarization was almost entirely s polarized which

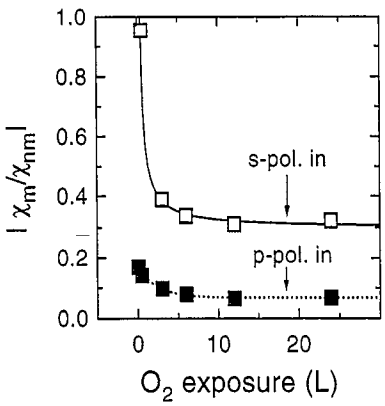

Fig. 2. Ratio of the magnetic and nonmagnetic part of the nonlinear susceptibility χ_m / χ_{nm} derived according to Eq. (4). The wavelength of the incident light was 790 nm. Solid symbols represent data taken with p-polarized incident light and open symbols with s-polarization. The solid and dashed curves are guidelines to the eye.

corresponds to a Kerr angle close to 90°. Very small amounts of oxygen hardly detectable in an Auger spectrum result in a strong increase of the p-polarized component of the SH light and a strong reduction of the Kerr angle.

We also measured the SH intensity for s-polarized light in the transversal geometry. Here, an exposure of 10 L completely cancels the asymmetry but the average SH intensity remains nearly constant.

4. Discussion and conclusions

The behavior of the SH intensity especially for p-polarized incident light in the transversal geometry is quite opposite to that expected from a simple interpretation taking only surface sources of SHG like in Eq. 1 into account. Generally, it is believed that reactive gases like oxygen reduce SHG [2,11]. Also it is known that oxygen strongly reduces the magnetization at the surface [7]. Moreover, in the present case there is only one contributing interface to the surface SHG and interference effects like in multilayers can be excluded [6]. In the present experiments we observe a persistence of the magnetization induced SHG up to very high oxygen coverage for all geometries and input polarizations. Therefore, it seems very probable that the SH light from the Fe(110) crystal is generated to a sizable amount due to quadrupole like bulk contributions. This is further corroborated by the observation that for the longitudinal geometry the Kerr angle initially drops rapidly but it stays nonzero up to the highest oxygen coverage.

There are a few examples of SH enhancement due to resonances of the fundamental or SH light with electronic transitions of the adsorbates [12,13]. In the present case little difference in the SHG is found in the wavelength range between 740 and 840 nm implying that these photon energies are at least not very close to an electronic resonance. But smaller off resonance contributions from the oxygen cannot be ruled out and are very likely to contribute as it will be discussed in the following. The maxima observed at the $c(2\times2)/c(1\times3)$ superstructures and at the completion of the monolayer might be caused by such SH sources from the oxygen itself. Mainly the nonmagnetic part of the SH response is affected as it is expected from the consideration that oxygen usually carries no or only a small induced magnetic moment.

Quite interesting is the behavior of the asymmetry and the Kerr angle for the transversal and longitudinal geometry, respectively. Their rapid change-especially strong for s-polarized incident light-at low oxygen coverage below ~ 5 L are probably dominated by the reduction of the surface magnetization finally resulting in a complete cancellation of the magnetic surface contribution of SHG from the iron. The expected reduction of the nonmagnetic part due to the decreased polarizability of the iron surface may be compensated by the oxygen contribution. From this picture only a small change in the average SH intensity and a decrease of the asymmetry reaching a constant value at the same oxygen coverage is expected for all geometries and polarizations of the incident light. And this is what we observe in almost all cases with the exception of the transversal geometry with p-polarized incident light. However, it is possible to explain the latter case consistently with the above ideas: It differs from the other in the extremely large magnetic contribution $|\chi_m|$. With the observation of asymmetries up to nearly 100% it seems not too unlikely to assume a ratio $|\chi_m/\chi_{nm}|$ even larger than 1 which according to Eq. (3) results in an increasing asymmetry with decreasing magnetization. Disregarding the oxygen induced peak at about 3 L, the only weakly increasing average SH

intensity suggests that not the larger, magnetic component is changed with oxygen exposure but the nonmagnetic part is increased as it is observed for the longitudinal geometry. The above explanation is by far not unambiguous and would imply a reduction of the nonmagnetic SH component in the region of the occurrence of the superstructures. Alternative explanations with $|\chi_m/\chi_{nm}| < 1$ are also possible but would imply a change of the phase between nonmagnetic and magnetic SH component, too.

In summary we have shown that the Fe(110) surface shows large Kerr angles and asymmetries. However in the present case the magnetization induced SH light is generated to a sizable amount by bulk SH sources especially pronounced for p-polarized incident light.

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