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Influence of quadratic contributions in magnetization-induced second harmonic generation studies of magnetization reversal

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Magnetization-induced optical Second Harmonic Generation (MSHG) from an exchange-biased CoO/Fe multilayer produces an asymmetrical hysteresis loop that indicates different magnetization reversal behaviour between the interface and the bulk ferromagnet. A more careful analysis of the data demonstrates that this asymmetry is in fact due to a quadratic dependence on the magnetization of the MSHG intensity.

1 Introduction

Magnetization-induced second harmonic generation (MSHG) was demonstrated 14 years ago [1] as a sensitive technique for magnetic surface and interface studies and since then it has been successfully applied for the examination of surface and interface magnetism [2]. Of particular interest is the combination of MSHG with the magneto-optical Kerr effect (MOKE), since this provides an all optical, non-invasive, way to investigate and compare simultaneously surface/interface and bulk properties of magnetic materials. The basis for this comparison is provided by the shape of the magnetization reversal, i.e. the hysteresis loop.

An interesting example of a phenomenon in which the interface and the bulk magnetization are expected to behave in a different manner is the exchange bias effect. Exchange bias occurs when a ferromagnetic/antiferromagnetic (FM/AFM) system is heated to a temperature above the Néel temperature (T_N) of the AFM but below the Curie temperature (T_C) of the FM and then is cooled in the presence of an external magnetic field. As a result, a shift of the hysteresis loop (H_E) away from the zero field position is observed. Although it has been discovered almost 50 years ago [3], there is still no complete theoretical explanation for this effect. Because of its numerous applications in spin-valves and magnetic sensors, it has recently become the subject of a lot of renewed research interest [4, 5]. One of the main reasons for this lack of understanding is believed to be the fact that there are relatively few experimental techniques able to probe buried interfaces. Therefore this problem is particularly suitable for a simultaneous investigation with MSHG and MOKE.

Our results revealed a clear difference in the magnetization hysteresis loop, depending on whether it was measured with MSHG or MOKE. Furthermore, this difference was found to be in good qualitative agreement with the expected magnetic behaviour at the interface and in the bulk in the case of exchange

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bias effect. We prove, however, that the observed difference is due to a quadratic dependence on the magnetization in the MSHG intensity [6] and most likely is not of magnetic origin.

2 Experimental details

MSHG and MOKE measurements were done using a Ti-Sapphire laser at 800 nm with pulse width duration ~100 fs and a repetition rate of 82 MHz. The laser power was attenuated with a 1/20 chopper blade to about 8 nW and was focused to a spot with diameter of ~100 μm. The angle of incidence was 30° and the magnetic field was applied in the longitudinal configuration. The incoming light was S-polarized (0°), and in order to maximize the MSHG contrast, the analyser was positioned at +45°. After reflection, a dichroic mirror was used to separate the fundamental from the second harmonic signal and both were plotted simultaneously.

The composition of our thin-film samples was Si(111)/Fe/CoO/Au. Initially, 6 nm Fe was deposited by molecular beam epitaxy (MBE) on hydrogen-passivated Si(111). After the preparation of 2 nm CoO [7, 8], the sample was covered by a 6 nm Au cap layer to prevent contamination from the atmosphere. Exchange bias was induced by cooling the sample from a temperature of 300 K (for CoO T_N = 291 K), in the presence of an external magnetic field of 2.5 kOe.

3 MSHG and exchange bias

For intense electromagnetic fields, such as those generated by a pulsed laser beam $\boldsymbol{E}(\omega)$ incident on a thin multilayer film, the polarization at the harmonic frequency 2ω is given by

$$\boldsymbol{P}_i^{(l)}(2\omega) = \chi_{ijk}^{(l)} \boldsymbol{E}_j^l(\omega)\, \boldsymbol{E}_k^l(\omega) \tag{1}$$

where χ_{ijk} is a third order polar tensor describing the non-linear optical susceptibility at the symmetry breaking interface between the centrosymmetric films and l numbers the interfaces in our sample. We can distinguish two types of contributions to the susceptibility: the "odd" (χ^{odd}) and "even" (χ^{even}), depending on whether the tensor elements associated with them change sign upon reversal of the magnetic moment.

In the presence of a magnetic field, the second harmonic intensity is then given by [2]

$$I(2\omega, \boldsymbol{M}) \propto \left(|\chi^{\text{even}}|^2 + |\chi^{\text{odd}}(\boldsymbol{M})|^2 \pm 2|\chi^{\text{even}}|\,|\chi^{\text{odd}}(\boldsymbol{M})| \right) \cdot I^2(\omega)\,, \tag{2}$$

which is linearly dependent on the magnetization for $|\chi^{\text{odd}}(\boldsymbol{M})| / (2|\chi^{\text{even}}|) \ll 1$.

When an external magnetic field is applied to the FM/AFM system at a temperature T such that $T_N < T < T_C$, the FM part aligns with the field, while the AFM one remains disordered. Then, as the system is cooled below T_N, the AFM orders. However, because of the strong magnetic field in the immediate neighbourhood of the FM, some of the AFM spins become pinned in the direction of the external applied field. When reversing the external field, the "bulk" FM spins will follow the field while those at the FM/AFM interface will remain in their previous orientation due to their interaction with the pinned AFM spins. In other words, in order to reverse completely, the system will have to overcome this interaction and therefore a larger external field needs to be applied. When completing the hysteresis loop, the FM/AFM interaction at the interface will now play an opposite role, i.e. facilitating the reversal process.

4 Results and discussion

Figure 1a shows the magnetization reversal observed by MOKE and MSHG. We can see a clear difference between the two curves. First, the exchange bias value is different – it is larger for the MSHG. Second, the shape of the hysteresis curves differs – while the magnetic reversal observed with MOKE is symmetrical, the MSHG curve presents a sharp first reversal and then a more gradual second reversal.

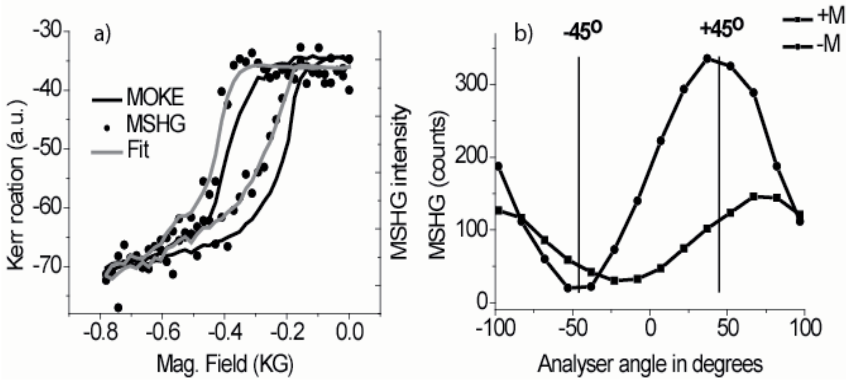

Fig. 1 In (a), simultaneous MOKE (black line) and MSHG (dots) measurements of the exchange biased samples at temperature 50 K. The grey line is a numerical fit for the MSHG intensity data. In (b), the MSHG intensity as function of analyser rotation for +***M*** and –***M***.

Since the MOKE signal is related to the "bulk" magnetization, while the MSHG is more sensitive to interface effects, the difference between the two curves can be related to their different origin. Indeed, as the exchange bias results from the AFM pinning at the interface, it is reasonable to say that its value is higher at the interface and hence the larger value of loop shift observed with MSHG.

Furthermore, we know that the AFM pins the FM at the interface [9]. When the "bulk" of the FM starts to reverse, the interface spins remain pinned in the direction of the bias. At some point though, the external field overcomes the exchange interaction and then the interfacial spins reverse sharply. This gives rise to the square behaviour during the first reversal with MSHG. While completing the hysteresis, the interfacial spins start their reversal sooner, since the exchange interaction and the external field act together, and this produces a more gradual second reversal as seen again with MSHG.

These results were reproducible on several samples with different composition and seemed to provide evidence that our observation was of general character, i.e. intrinsically related to the exchange bias effect.

The explanation given above assumes that the MSHG signal is a direct (linear) measure of the magnetization at the interface. However, it could very well be that the bulk and the interface behave in the same manner but that there is a quadratic component in the MSHG signal that distorts the hysteresis loop, making it asymmetrical. Indeed, it follows directly from Eq. (2), that if the condition $|\chi^{\mathrm{odd}}(\boldsymbol{M})|/(2|\chi^{\mathrm{even}}|) \ll 1$ is not fulfilled, the quadratic terms in the intensity dependence become important.

Figure 1a shows a numerical fit for the MSHG intensity data based on Eq. (2) assuming a quadratic dependence on ***M***. The magnetization behaviour was given by the results observed with MOKE and we chose $\chi^{\mathrm{even}} = 12$. Although it is clear that the fit accounts very well for the MSHG results, this does not prove anything; it simply supports the possibility of an alternative explanation for our results. In order to demonstrate which explanation is the real one, we performed two more measurements.

In the first measurement, we modified the optical part of the experiment, while keeping the magnetic part unchanged. This was done by rotating the analyser from the +45° to the –45° position.

Figure 2a gives a schematic representation of our hysteresis loop emphasizing the asymmetry. In Fig. 1b, the MSHG intensity dependence on the analyser rotation is plotted and we can see that between

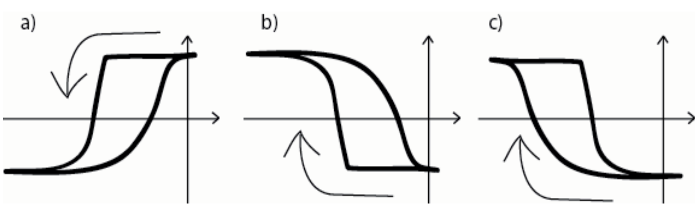

Fig. 2 Schematic representation of the MSHG hysteresis loop asymmetry for analyser at +45° (a) and at –45°: for a magnetic origin of the asymmetry (b) and for an optical origin (c).

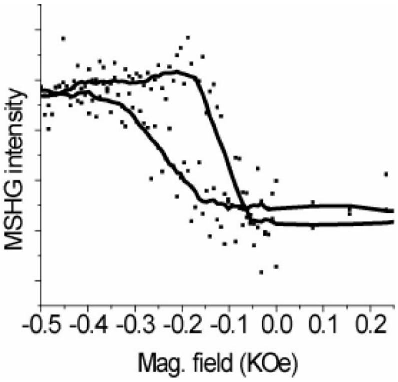

Fig. 3 MSHG intensity as function of magnetic field for temperature 50 K with analyser at –45°.

+45° and –45° the magnetic contrast is reversed. Therefore, if the asymmetry in the hysteresis loop has a magnetic origin, we can expect the first reversal to be sharp, because of the action of the pinned AFM spins (Fig. 2b). On the other hand, if the origin is of optical nature, the reversal will reproduce the magnetic behaviour of the curve in Fig. 2a, with only a symmetry change that accounts for reversing the MSHG intensity levels, as in Fig. 2c.

The obtained experimental result is plotted in Fig. 3. There is a clear similarity between this curve and the one in Fig. 2c.

This is in agreement with the assumption of a quadratic contribution distorting the signal.

In our second measurement, we modified the magnetic part of the experiment while keeping the optical one unchanged. For this purpose, we field-cooled the sample in an opposite field. The first consequence of this manipulation will be a positive loop shift, as represented on Figs. 4b and c. Figure 4a gives again a schematic representation of our initial hysteresis loop. And again, if the reason for the asymmetry is magnetic, we expect the first reversal to be sharp, as in Fig. 4b, while an optical cause of the asymmetry will reproduce the initial curve, as shown in Fig. 4c.

The obtained results are shown in Fig. 5, where it is clearly visible that the shape of the two hysteresis curves is similar, in the same manner as Fig. 4a resembles Fig. 4c.

5 Conclusions

We have demonstrated that the asymmetry of the MSHG hysteresis curve from a CoO/Fe system is due to a quadratic dependence of the MSHG intensity on ***M*** and is not directly (linearly) representative of the magnetic behaviour at the exchange-biased FM/AFM interface. Thus, although our data seemed initially in excellent agreement with the intuitively appealing model of the magnetic phenomenon that we were trying to investigate, they turned out to be produced by a magneto-optical nonlinearity.

In conclusion, we can say that although the technique of magnetization-induced second harmonic generation shows great potential for the simultaneous studies of surface/interface and bulk effects, it should be employed carefully.

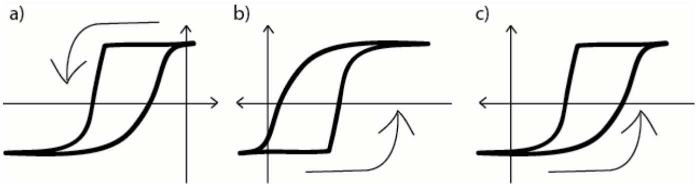

Fig. 4 Schematic of the hysteresis loop shape after positive field-cooling (a). After negative field-cooling, in the case of real magnetic effect (b), and in case of an optical origin (c).

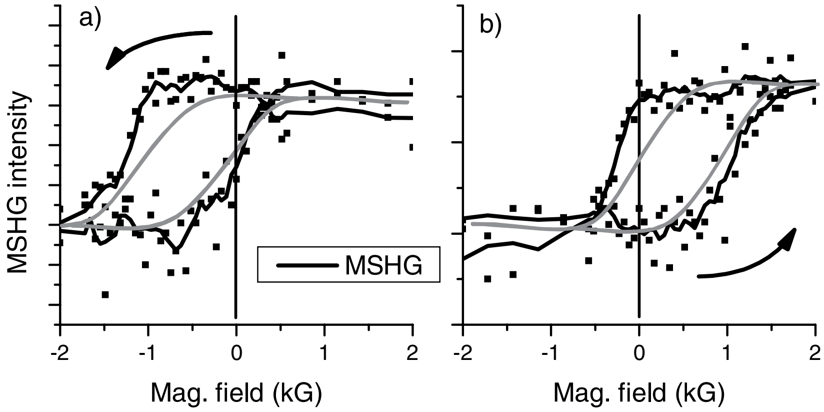

Fig. 5 MSHG intensity as function of applied magnetic field, at a temperature of 10 K for positive (a) and negative (b) field-cooling. The hysteresis grey lines are guides to the eye representing a symmetrical reversal.

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