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Azimuth Angle-Dependence of MSHG in Magnetic Superlattices and its Symmetry Analysis

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Abstract- Magnetic second harmonic generation (MSHG) has been measured in Fe/Au and Co/Ru superlattices. Well-defined 4-fold symmetry was observed in azimuth angle-dependence of MSHG and nonlinear Kerr effect of the Fe/Au superlattices, while isotropic azimuthal dependence was found in the Co/Ru superlattices. Azimuthal patterns are analyzed in terms of nonlinear susceptibility tensors taking into account the symmetry of superlattices.

I. INTRODUCTION

Magnetic second harmonic generation (MSHG) has been studied extensively because of its sensitivity to surfaces and interfaces of magnetic materials.[1] In our previous MSHG studies on Fe[xML]/Au[xML] superlattices with integer x, we have elucidated that polar plots of the azimuth angle-dependence of second harmonic (SH) intensity shows a well-defined 4fold symmetry pattern, which undergoes a mirror reflection with respect to a certain axis on the reversal of magnetic field.[2] Similar azimuthal patterns were also found in Fe[xML]/Au[xML] superlattices with noninteger values of x.[3] Recently we have measured azimuth angle-dependence of nonlinear Kerr rotation in the same superlattice system. It is found that the nonlinear Kerr effect also shows 4-fold symmetry pattern in the azimuth angle-dependence.[4] Analysis using nonlinear susceptibility tensor lead to the conclusion that electric quadrupole contribution should be included in addition to the dipolar contribution to account for the anisotropic behavior of MSHG.

Usually only isotropic MSHG has been studied and very few examples of anisotropic MSHG studies have been reported in literature.[5, 6] It is important to note that earlier MSHG anisotropy can be fully described within the dipole formalism of χ tensor. On the contrary, our results cannot be explained solely by the dipolar term but require incorporation of electrical quadrupole contribution to the χ tensor.

In this paper, experimental results of azimuth angle-dependence of MSHG in Fe/Au and Co/Ru superlattices are presented and discussed in terms of symmetry analysis of dipole and quadrupole χ tensors.

II. EXPERIMENTAL

The Fe/Au and Co/Ru superlattices were prepared epitaxially using ultra high vacuum (UHV) deposition technique on MgO(100) and Al_2O_3(1120) substrates, respectively. The base pressure of the deposition system was 3×10^{-10} Torr. As a buffer layer, Au (100) layer was employed in the Fe/Au superlattice, while Ru (0001) layer was used in the Co/Ru superlattice. Details of preparation conditions have been published elsewhere.[7]

MSHG measurements were performed using a mode-locked Ti-sapphire laser (λ=810 nm). A 5W diode-pumped YVO_4-SHG laser (Coherent VERDI) with the wavelength of 530 nm was employed as an excitation source for the Ti-sapphire laser (Coherent MIRA). The pulse width of the Ti-sapphire laser was 150 fs and the repetition rate was 80 MHz. To avoid sample damage by the laser irradiation, the averaged power of the light beam was reduced to 1/10-1/20 of the original intensity using a rotating light chopper. The spot size of the laser beam focused on the sample was 40 μm in diameter and the peak power density was estimated to be 0.5 GW/cm^2. The incident angle of the laser beam was fixed at 45° to the sample normal. Magnetic field up to about 0.3 T was applied in the longitudinal Kerr geometry. Details of experimental setup have been published elsewhere.[3]

The sample was mounted on a computer-controlled rotating stage to obtain the azimuthal angle-dependence of the MSHG signal, which was measured for all the four combinations of input-output polarization; i.e., P_{in}-P_{out}, S_{in}-P_{out}, P_{in}-S_{out}, S_{in}-S_{out}, where the notations P and S denote the polarizations parallel and perpendicular to the incident plane of reflection, respectively. For measurements of the nonlinear Kerr rotation a computer-controlled rotating analyzer was employed. The MSHG signal shows a sinusoidal dependence on the analyzer angle, but is subjected to a phase difference between two opposite directions of magnetization ($\pm M$). The angle of the nonlinear Kerr rotation $\theta_K^{(2)}$ was obtained as a half of the difference of the phases $\psi_\pm$ for $\pm M$ determined using the Fourier analysis. The dependence of the nonlinear rotation $\theta_K^{(2)}$ on the sample azimuth φ was also measured.

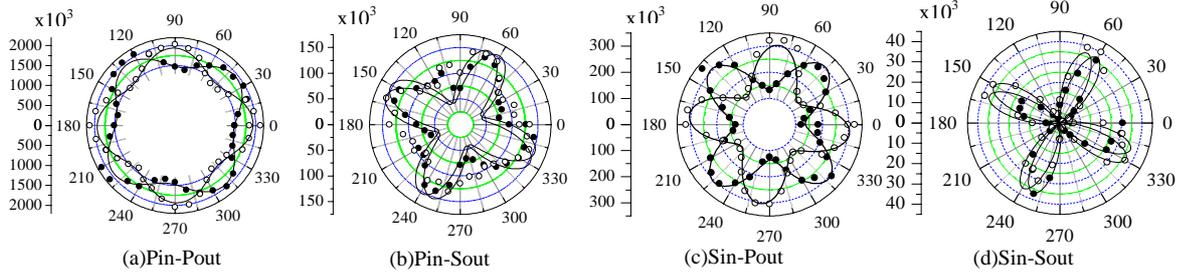| (a)Pin-Pout | (b)Pin-Sout | (c)Sin-Pout | (d)Sin-Sout |

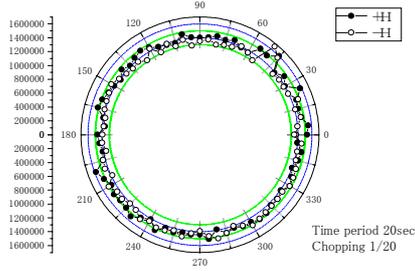Fig. 1 Azimuth angle dependence of MSHG intensity in Fe(3.5ML)/Au(3.5ML) superlattice

Fig. 2 Azimuthal angle dependence of MSHG intensity in Co(5ML)/Ru(5ML) superlattice

III. RESULTS AND DISCUSSION

Figure 1 shows typical examples of azimuth angle-dependence of MSHG intensity from the [Fe(3.5ML)/Au(3.5ML)]$_{30}$/Au(001)/MgO(001) super-lattice for four different combinations of input and output polarizations; i.e., P$_{in}$-P$_{out}$, P$_{in}$-S$_{out}$, S$_{in}$-P$_{out}$ and S$_{in}$-S$_{out}$. Open and closed symbols are experimental data points for two opposite directions of the applied field and the lines are results of simulation, which will be described later. All the azimuthal patterns are anisotropic with 4-fold symmetry. On the other hands, azimuthal MSHG pattern of [Co(5ML)/Ru(5ML)]$_{20}$/Ru(0001)/Al$_2$O$_3$ superlattice plotted in Fig. 2 for P$_{in}$-P$_{out}$ configuration exhibits only an isotropic azimuthal pattern. No detectable MSHG signal was observed in S$_{in}$-S$_{out}$ and P$_{in}$-S$_{out}$ configurations.

X-ray diffraction and electron diffraction analyses have elucidated that the Fe/Au superlattices follow the symmetry of MgO(001) substrate and have the 4/mmm symmetry for bulk and 4mm for surface. On the other hand, Co/Ru superelattices have the 6/mmm symmetry for bulk and 6mm for surface following the hexagonal symmetry of the Ru(0001) buffer.

Nonlinear polarization is expressed by

$$P_i(2\omega) = \chi_{ijk}^D E_j(\omega)E_j(\omega) + \chi_{ijkL}^D E_j(\omega)E_j(\omega)M_L$$
$$+ \chi_{ijkl}^Q E_j(\omega)\nabla_k E_l(\omega) \qquad ,(1)$$

where the index L (=X,Y) refers to the magnetic moment direction which is the axial vector. The first two terms are of electric dipole origin, while the last term is of electric quadrupole origin. The 3rd rank tensor vanishes for centrosymmetric crystal structures. Therefore, the first term in the above expression

survives only at the surfaces and interfaces where inversion symmetry is broken. The second term is of the surface magnetic origin. On the other hand, the last term results from the nonlocal (quadrupole-allowed) contribution and survives even in a centrosymmetric bulk. Significance of this term has been known in cubic nonmagnetic metals such as Cu.[8] Azimuthal dependence of MSHG for experimental configurations can be calculated using transformation matrix $A_{i'i}$, which connects the laboratory coordinate (i',j',k') with the sample coordinate (i,j,k). The 3rd rank tensor transforms as $\chi_{i'j'k'}^D = \sum_{ijk} A_{i'i}A_{j'j}A_{k'k}\chi_{ijk}^D$ and the 4th rank tensor as $\chi_{i'j'k'l'}^Q = \sum_{ijkl} A_{i'i}A_{j'j}A_{k'k}A_{l'l}\chi_{ijkl}^Q$, where

$$A_{i'i} = \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix}. \qquad (2)$$

The first term in eq. (1) gives an isotropic response for Pin-Pout and Sin-Pout configurations while it vanishes for Pin-Sout and Sin-Sout configurations in either 4mm or 6mm surface symmetry, because a 3rd-rank tensor is unable to yield the 4-fold and 6-fold symmetric rotational anisotropy. The second term (magnetic contribution) and the last term (quadrupole contribution) involve 4th rank tensor, which leads to 4-fold anisotropic azimuthal patterns for the surface 4mm symmetry and the bulk 4/mmm symmetry, respectively. On the other hand, the 4th rank tensor never gives rise to a 6-fold anisotropic azimuthal pattern since the rank is less than 6. This explains why azimuthal pattern of Co/Ru superlattice shown in Fig. 2 is not anisotropic.

The symmetry analysis results in the azimuth angle-dependence of MSHG in Fe/Au superlattice for four combinations of polarizations as follows: [9]

$$I^{ss}(\varphi, \pm M) = \left| \pm A^{ss} + B^{ss}\sin 4\varphi \pm C^{ss}\cos 4\varphi \right|^2$$

$$I^{sp}(\varphi, \pm M) = \left| A^{sp} - B^{sp}\cos 4\varphi \pm C^{sp}\sin 4\varphi \right|^2$$

$$I^{ss}(\varphi, \pm M) = \left| \pm A^{ps} - B^{ps}\sin 4\varphi \mp C^{ps}\cos 4\varphi \right|^2 \qquad (3)$$

$$I^{pp}(\varphi, \pm M) = \left| A^{pp} + B^{pp}\cos 4\varphi \mp C^{pp}\sin 4\varphi \right|^2$$

where the double sign corresponds to two opposite magnetization directions. The terms with B parameters

are originated from the nonlocal (qudrupole-allowed) contribution, whereas those with $\pm C$ are from surface magnetic contribution. For relatively small angle of incidence, the following approximate relation holds between parameters:

$$B^{SS} \approx B^{SP} \approx B^{PS} \approx B^{PP} \approx B$$

and

$$C^{SS} \approx C^{SP} \approx C^{PS} \approx C^{PP} \approx C \ .$$

We made a fitting procedure on the experimental azimuthal patterns using eq. (3). Since anisotropic behavior is the most obviously observed in the S_{in}-P_{out} pattern, we employed this pattern (Fig. 1(c)) to determine anisotropic parameters B and C by fitting. Parameters thus determined are A^{sp}=460, B=26 and C=-88. The lines in Fig. 1(c) show the best fit to the experiment. Using the B and C thus determined, we simulated other three patterns and obtained the best fit only by adjusting the isotropic A parameters. The observed anisotropic shape, as well as the relative magnitude of intensity, were satisfactorily reproduced for (a) P_{in}-P_{out}, (b) P_{in}-S_{out} and (d) S_{in}-S_{out} with the parameters as A^{PP}=1310, A^{PS}=-300 and A^{SS}=100, respectively.

Figure 3(a) provides one quadrant of azimuth angle-dependence of nonlinear Kerr rotation $\theta_K^{2,S}$ for S_{in} configuration. The 4-fold symmetry is obviously observed. To evaluate $\theta_K^{2,S}$ theoretically we first describe the analyzer angle-dependence of SH intensity. Denoting the sp- and ss-electric field as E^{SP} and E^{SS}, respectively, the SH intensity of S polarization incidence for analyzer angle θ can be written as:

$$I^{Sin}(\theta)=|E^{SP}\cos\theta + E^{SS}\sin\theta|^2 \ . \quad (4)$$

Using expressions for E^{SS} and E^{SP} in terms of parameters obtained above, eq. (4) can be rewritten as

$$I^{Sin}(\theta)= (1/2)\{(A^{SP} \mp C\sin 4\varphi - B\cos 4\varphi)^2$$
$$+(\pm A^{SS} + B\sin 4\varphi \pm C\cos 4\varphi)^2\}\{1+\cos(2\theta - 2\psi_\pm^S)\} \quad (5)$$

where the phase $\psi_\pm^S$ is given by

$$\tan 2\psi_\pm^S$$
$$= \frac{2(A^{sp} \mp C\sin 4\varphi - B\cos 4\varphi)(\pm A^{ss} + B\sin 4\varphi \pm C\cos 4\varphi)}{(A^{sp} \mp C\sin 4\varphi - B\cos 4\varphi)^2 - (\pm A^{ss} + B\sin 4\varphi \pm C\cos 4\varphi)^2} \quad (6)$$

Then, the nonlinear magneto-optical Kerr rotation $\theta_K^{(2)S}$ is given by

$$\theta_K^{(2)S} = (\psi_+^P - \psi_-^P)/2 \quad (7)$$

The nonlinear magneto-optical Kerr rotation calculated by eq. (7) is plotted in Fig. 3(b) for S-polarization incidence. The calculated patterns show 4-fold symmetry in agreement with the experiment.

IV. CONCLUSION

Azimuth angle-dependence of MSHG was measured in Fe/Au and Co/Ru superlattices. Well-defined 4-fold anisotropic pattern was observed in the Fe/Au superlattice for all combination of the input-output polarizations, whereas only isotropic pattern
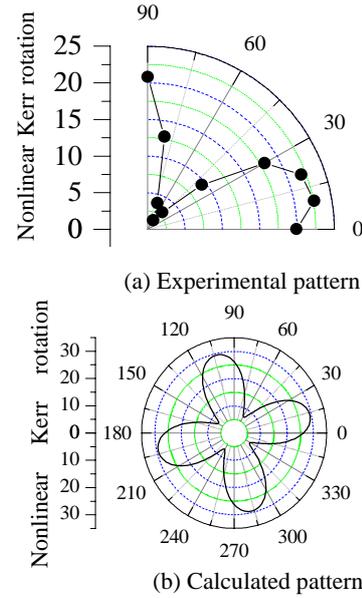

(a) Experimental pattern

(b) Calculated pattern

Fig. 3 Azimuthal dependence of nonlinear Kerr rotation for S_{in} configuration.

was observed in the Co/Ru superlattice. The absence of 6-fold symmetry in azimuthal pattern of Co/Ru system is reasonably explained taking into account the fact that 3rd and 4th rank χ-tensors are involved in the nonlinear polarization. The observed anisotropy in the azimuthal pattern of MSHG in Fe/Au for P_{in}-P_{out}, P_{in}-S_{out}, S_{in}-P_{out} and S_{in}-S_{out} configurations was successfully described by the tensor analysis.

Nonlinear Kerr rotation was also found to show the 4-fold symmetry and was explained theoretically by using the same parameters used to explain the anisotropy of the MSHG intensity.

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