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Nonlinear Magneto-Optical Effect in Fe/Au Superlattices

K. Sato, S. Mitani[a], K. Takanashi[a], H. Fujimori[a], A. Kirilyuk[b], A. Petukhov[b] and Th. Rasing[b]
Tokyo University of Agriculture and Technology, Koganei, Tokyo 184-8588, Japan
[a]Institute for Materials Research, Tohoku University, Sendai 980-77, Japan
[b] Research Institute for Materials, University of Nijmegen, 6500 GL Nijmegen, The Netherlands

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Nonlinear magneto-optical effects were measured in $[\mathrm{Fe}(n\mathrm{ML})/\mathrm{Au}(n\mathrm{ML})]_N$ superlattices prepared by MBE on a MgO (100) substrate. Nonlinear magneto-optical effects were measured using Ar$^+$-laser-pumped Ti-sapphire laser (λ=760 nm) with suitable combination of filters, polarizer-analyzer and photon-counting apparatus. A well-defined four-fold symmetry pattern was observed in the polar plot. Reversal of magnetization results in a reflection of the azimuthal pattern with respect to a certain axis. Such a symmetrical property cannot be understood if only electric dipole terms are taken into account in the analysis. The azimuthal pattern is explained in terms of electric quadrupole terms for non-magnetic contribution and dipole terms for magnetic contribution.

Keywords: nonlinear magneto-optical effect, azimuthal pattern, Fe/Au superlattice, surface, interface, electric quadrupole

1. Introduction

Nonlinear magneto-optical effect is a magnetically induced second harmonic generation (MSHG) in magnetic materials.[1)] This effect is very sensitive to surfaces and interfaces, since SHG in the materials with an inversion symmetry becomes allowed only at surfaces and interfaces where such symmetry is broken. This is the reason why MSHG has been applied to studies of magnetic thin films and multilayers.[2), 3)] However, previous MSHG studies on multilayers were concentrated in polycrystalline films prepared by sputtering technique.

In this study we have applied for the first time the MSHG technique to Fe/Au superlattices with atomically controlled epitaxial layers. The superlattice with a modulation of mono-atomic layers of Fe and Au has been known to show an artificial order with an $L1_0$ structure that does not exist in nature. [4)] Such an artificial structure persists at interfaces between Fe and Au layers even for superlattices with longer modulation period. [5)] The linear magneto-optical spectra of the superlattices have been studied intensively, suggesting formation of the band structure peculiar to the artificial structure. [6)]

2. Experimental

The $[\mathrm{Fe}(n\ \mathrm{ML})/\mathrm{Au}(n\ \mathrm{ML})]_N$ superlattices (n=1, 2, 3, 4, 5, 6, 8, 10 and 15; ML stands for the monolayer) were prepared by UHV deposition on cleaved MgO (100) substrates with a Au(100) buffer layer. The base pressure of the deposition system was 3×10^{-10} Torr. A Fe seed layer of 1 nm followed by a Au buffer layer of 50 nm was deposited at 200°C and subsequently annealed for 30 min to 1 h at 500 °C. The orientation of the Au buffer layer was (100) for MgO substrates. The Fe seed was necessary to control the orientation of the Au layer. The details were described elsewhere. [4)] The samples we used for MSHG experiments were those with n=15, N= 7; n=6, N=17; n=1, N=100.

MSHG measurements were performed using an Ar$^+$-laser-pumped mode-locked Ti-sapphire laser (λ=760 nm) with suitable combination of filters, polarizer-analyzer and photon-counting apparatus. The incident angle of the laser beam was 45° to the normal. The spot size of the laser beam focused on the sample was approximately 80 μm and the beam intensity was 1 GW/cm^2. Magnetic field up to about 0.2 T was applied in the longitudinal Kerr geometry. Sample was rotated around the beam spot to get the azimuthal dependence of the MSHG signal.

3. Results

The MSHG signal was quite sensitive to the surface quality of samples. If surfaces or interfaces were deteriorated or damaged the MSHG signal lost intensity and reproducibility was lacking, especially in the azimuthal dependence of the signal on rotating the sample.

As shown in the polar plot of the azimuthal pattern of Fig. 1 the MSHG signal of the sample with n=1, i.e., $[\mathrm{Fe}(1\mathrm{ML})/\mathrm{Au}(1\mathrm{ML})]_{100}$ was very week (only 1000 counts per 10 s) and deviates from point to point. The change of the MSHG signal on reversal of the magnetic field was not systematic. Such a poor pattern seems to be caused by deterioration of the Fe layers, since thickness of the uppermost Au layer was so thin (1 ML) that the Au layer could not act as a sufficient protection layer.

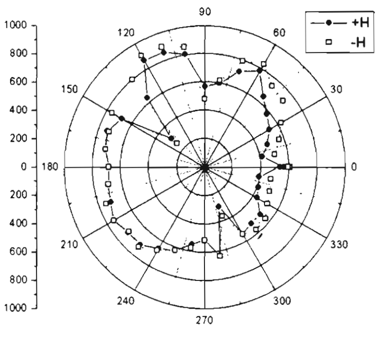
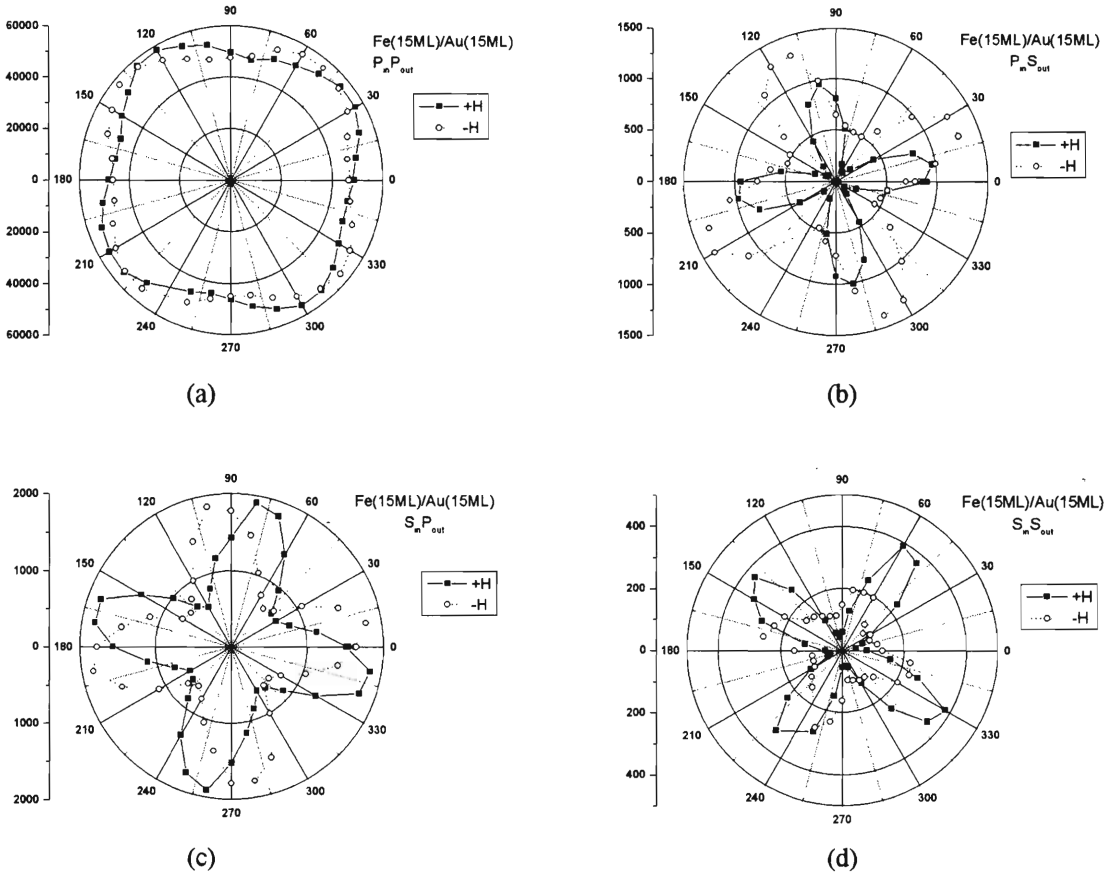


Fig. 1 A polar plot of the azimuthal dependence of the MSHG signal in Fe(1ML)/Au(1ML) superlattice.



(a) (b)

(c) (d)

Fig. 2 Polar plot of azimuthal pattern of MSHG signal.
(a) Pin-Pout, (b) Pin-Sout, (c) Sin-Pout, (d) Sin-Sout.

On the contrary, for thicker layers like *n*= 6 and 15 the MSHG signal was intense and the polar pattern was quite clear. The dependence of MSHG signal on azimuthal angle was measured for 4 combinations of input-output polarization; i.e., P_{in}-P_{out}, S_{in}-P_{out}, P_{in}-S_{out}, S_{in}-S_{out}. The results for *n*=15 and 6 are quite similar. Here we show only the result for *n*=15 superlattice. Azimuthal patterns in the Fe(15ML)/Au(15ML) superlattice for the 4 polarization geometry are shown in Figs. 2(a) to 2(d). Here zero of the azimuthal angle is taken arbitrarily.

Analyzer angle-dependence of MSHG signal shows a sinusoidal curve with a shift for two oposite directions of magnetic field, from which the nonlinear Kerr rotation as large as 3° was obtained.

4. Discussion

The four-fold pattern clearly reflects the symmetry of the MgO (100) substrate. This suggests that the Fe/Au superlattice is perfectly epitactic to the substrate. No such 4-fold symmetry in the nonmagnetic SHG signal of crystallographic origin is expected in the electric dipole scheme even for the surface, since $\chi^{(2)}_{ijk}$ vanishes as far as two mirror planes exist. Electric quadrupole term is necessary to explain the 4-fold symmetry.

Three contributions were separately discussed; (1) nonmagnetic contribution from the bulk (cubic), (2) nonmagnetic contribution from the surface, and (3) magnetic contribution mainly from the surface.

(1) Nonmagnetic bulk contribution

It is found that bulk contribution to SHG is described in terms of two independent elements of the nonlinear susceptibility of rank 4 for both cubic and uniaxial (4/mmm) symmetry; one is an isotropic term $\chi_2 \equiv \chi^Q_{ijij}$ and the other is an anisotropic term $\zeta \equiv \chi^Q_{iiii} - \chi^Q_{ijij} - 2\chi^Q_{iijj}$.

The azimuthal angular dependence of the SHG intensity was found to be described by a quadratic expression of an even function, $|A+B\cos 4\varphi|^2$ for S_{in}-P_{out} and P_{in}-P_{out} configurations and by a quadratic expression of an odd function, $|C\sin 4\varphi|^2$ for S_{in}-S_{out} and P_{in}-S_{out} configurations. The isotropic term contributes only to the isotropic parameter A, while the anisotropic term contributes to the isotropic parameter A, as well as to the anisotropic parameters B and C.

(2) Surface contribution to SHG

The symmetry of the surface of the cubic centrosymmetric materials is reduced to (4mm) due to the rupture of mirror symmetry at the surface. In this case rank 3 tensor due to electric dipole term survives. Symmetry consideration reveals that nonmagnetic surface contribution is zero for S_{in}-S_{out} and P_{in}-S_{out}

geometry, while it is described by a parameter $\chi^{S}_{zyy} \equiv \chi^{S}_{zxx}$ for S_{in}-P_{out} and all the three parameters $\chi_{xxz}, \chi_{zxx}, \chi_{zzz}$ for S_{in}-S_{out} geometry. Only isotropic contribution survives and no azimuthal angular dependence appears.

(3) Magnetization-induced SHG

We use linear-in-M approximation and expand the tensors up to a linear term; $\chi(\mathbf{M}) = \chi(0) + \mathrm{X} \cdot \mathbf{M}$, where χ (0) is the nonmagnetic rank 3 tensor and X the rank 4 tensor which is the coefficient for magnetic term. The time reversal symmetry is lifted by magnetization. Mirror symmetry operations should be supplemented with an additional reversion of M. The azimuthal dependence is $|\pm A \pm B\cos 4\phi|^2$ for S_{in}-S_{out} and P_{in}-S_{out} geometry, while it is $|\pm C\sin 4\phi|^2$ for S_{in}-P_{out} and P_{in}-P_{out} geometry, where ± corresponds to direction of magnetization.

Results from the theoretical procedure are summarized in Table 1. It finally comes out that azimuthal dependence of MSHG for all of the four sets of polarization geometry can be described in the form of $|A + B\cos 4\phi \pm C\sin 4\phi|^2$ although the values of A, B and C are different for each configuration. The expression is visualized in Fig. 3 for two sets of parameters; (a) A=5, B=0.5 and C=0.25, (b) A=0.5, B=0.5 and C=0.25. Figs. 3(a) and 3(b) are quite similar to the experimental plots for P_{in}-P_{out} and S_{in}-P_{out}.

Table 1. Calculated azimuthal angle dependence of SHG and MSHG signals

α, β	surface, nonmagnetic	bulk, nonmagnetic	surface, magnetization-induced	Sum
s, s	0	$\lvert C\sin 4\phi\rvert^2$	$\lvert\pm A \pm B\cos 4\phi\rvert^2$	$\lvert\pm A \pm B\cos 4\phi \pm C\sin 4\phi\rvert^2$
s, p	$\lvert A\rvert^2$	$\lvert A + B\cos 4\phi\rvert^2$	$\lvert\pm C\sin 4\phi\rvert^2$	$\lvert A + B\cos 4\phi \pm C\sin 4\phi\rvert^2$
p, s	0	$\lvert C\sin 4\phi\rvert^2$	$\lvert\pm A \pm B\cos 4\phi\rvert^2$	$\lvert\pm A \pm B\cos 4\phi \pm C\sin 4\phi\rvert^2$
p, p	$\lvert A\rvert^2$	$\lvert A + B\cos 4\phi\rvert^2$	$\lvert\pm C\sin 4\phi\rvert^2$	$\lvert A + B\cos 4\phi \pm C\sin 4\phi\rvert^2$

5. Conclusion

Nonlinear magneto-optical effect (MSHG) was measured in Fe/Au superlattices. Strong MSHG signal was observed for four sets of polarization geometry. Prominent 4-fold symmetry reflecting that of the substrate was observed. The azimuthal pattern was qualitatively interpreted in terms of combination of electric quadrupole for the nonmagnetic term and electric dipole for magnetic term.

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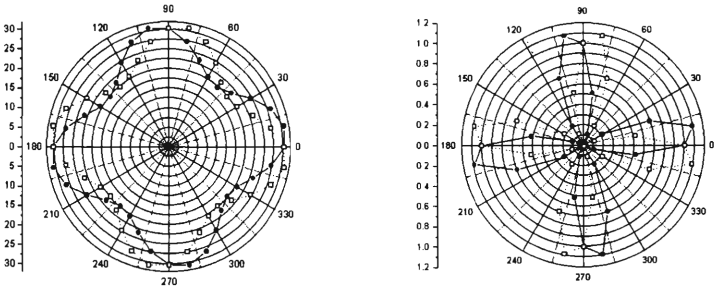

(a) A=5, B=0.5, C=0.25 (b) A=0.5, B=0.5, C=0.25

Fig. 3 Calculated azimuthal patterns for two sets of parameters.