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Nonlinear magneto-optical effect in Fe/Au superlattices modulated by noninteger atomic layers

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Magnetic second harmonic generation (MSHG) signals from the Fe/Au superlattices grown on MgO (100) substrate have been investigated. The measurements were carried out in the longitudinal Kerr geometry with a 45° incidence. The magnetic field of ± 0.2 T was applied to provide a magnetic saturation. The polar plot of azimuth dependence of the MSHG intensity showed a four-fold symmetry pattern similar to that observed in Fe/Au superlattices modulated by integer atomic layers. Azimuthal patterns show 45° rotation by reversing the magnetic field. Nonlinear Kerr rotation angle of 23.2° was observed in the $x=3.75$ superlattice. [S0021-8979(00)49108-5]

Studies of nonlinear magneto-optical effects, in particular, of magnetic second harmonic generation (MSHG), are attracting interest of researchers of magnetism, since the phenomenon is quite sensitive to surfaces and interfaces where inversion symmetry is broken. This is the reason why MSHG has been applied to studies of magnetic thin films and multilayers.[1] However, previous MSHG studies on multilayers were concentrated in polycrystalline films prepared by sputtering technique. We have applied the MSHG technique to Fe/Au superlattices with atomically controlled epitaxial layers.

Takanashi *et al.* succeeded in preparation of Fe/Au superlattice composed of mono-atomic layers of Fe and Au and showed that an artificial order exists with a $L1_0$ structure.[2] Mitani *et al.* prepared Fe/Au superlattices with longer modulation period and found from x ray and magnetic studies that the $L1_0$ structure remains at interfaces between Fe and Au layers.[3] Recently, $[\mathrm{Fe}(x\mathrm{ML})/\mathrm{Au}(x\mathrm{ML})]_N$ superlattices modulated by noninteger number ($x=1.25,1.5,1.75,\cdots$) of atomic layers were also prepared. X-ray diffraction (XRD) measurements indicate that coherent layered structures are formed also in the noninteger case.[4] The linear magneto-optical spectra of the Fe/Au superlattices modulated by integer and noninteger number of atomic layers have been studied intensively.[5,6] It is elucidated from these studies that a novel energy band structure is formed, which is completely different from that of the constituent materials. An *ab initio* band calculation by Yamaguchi *et al.* provides theoretical support for the experimental findings.[7]

In our previous work, we measured the MSHG of Fe/Au superlattices modulated by integer atomic layers and observed strong MSHG signals, the azimuthal pattern of which shows a prominent four-fold symmetry reflecting that of the substrate.[8] The azimuthal pattern was qualitatively explained in terms of combination of electric quadrupole for the nonmagnetic term and electric dipole for magnetic term.

The specimens used in the present study were prepared on MgO (100) substrates by the deposition technique in an UHV chamber. 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. Multilayers with N periods, each period consisting of x ML Fe and x ML Au, were deposited in the UHV system at 70 °C on the Au buffer. The layer thickness was controlled using a quartz thickness monitor. Physical parameters (x, N, one period thickness l, and lattice spacing d) of the superlattices employed in this study are summarized in Table I.

The XRD pattern was explained by a theoretical calculation assuming fractional occupation of Fe and Au in each layer, suggesting a formation of a coherent layered structure even for noninteger values of x. Details of the x-ray analysis were described in Ref. 5.

MSHG measurements were performed using a mode-locked Ti-sapphire laser ($\lambda=810$ nm). A 5 W 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 (MIRA). The pulse width of the Ti-sapphire laser was 150 fs and the repetition rate was 80 MHz. The averaged power output of the laser was approximately 600 mW. To avoid possible sample damages by the laser irradiation, the averaged power of the light beam was reduced to 1/20 of the original intensity by a 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 0.2 T was applied in the longitudinal Kerr geometry.

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TABLE I. Physical parameters of the Fe/Au superlattices modulated by noninteger number of atomic layers. For reference, the superlattice with $x=1$ is also prepared.

x (ML)	N (periods)	Thickness l for one period (nm)	Observed d_{002} value (nm)
1	30	0.3840	0.1915
15	7	5.2408	0.1698
1.25	30	0.4324	0.1775
1.50	30	0.4845	0.1745
1.75	57	0.5865	0.1795
2.25	30	0.8544	0.179
2.50	30	0.8963	0.176
2.75	36	0.9605	0.178
3.25	30	1.1428	0.1771
3.50	30	1.2726	0.1761
3.75	30	1.3007	0.1772

Azimuthal angle dependence of the MSHG signal was measured for P_{in}–P_{out} configuration. Measurements of the nonlinear Kerr rotation angle were performed employing a computer-controlled rotating analyzer for P polarization of incident beam.

The second harmonic (SH) light was effectively filtered using two blue filters (Hoya-Schott BG39) and guided to a photomultiplier (Hamamatsu type R464), the output of which was guided to a preamplifier (Hamamatsu type C5594) with the gain of 36 dB and the bandwidth of 1.5 GHz and a photon counting apparatus (Stanford Research, SR-400).

Strong and reproducible SH signals were observed in all the Fe(xML)/Au(xML) superlattices studied. The polar patterns of the azimuthal angle dependence of MSHG intensity for $x=1$ ($L1_0$ artificial alloy), 15 are plotted in Fig. 1, while those for $x=1.25$, 1.5, 1.75, 2.25, 2.5, 2.75, 3.25, 3.5, and 3.75 are shown in Fig. 2.

The azimuthal pattern of SH signal from the $x=1$ superlattice shows a well-defined four-fold symmetry. The similar pattern is also observed in $x=15$ superlattice. The reversal of the magnetic field direction resulted in a slight change of the azimuthal pattern in the $x=1$ superlattice, in contrast to a big change observed in the $x=15$ superlattice. The observed four-fold symmetry clearly reflects the symmetry of the MgO (100) substrate. Theoretical analysis has revealed that surface nonmagnetic second-harmonic generation (SHG) signal of the electric dipole origin provides only isotropic contribution but does not give rise to the four-fold anisotropic signal assuming 4/mmm symmetry for the surface. The experimentally observed symmetry can be interpreted in terms of the interference between surface magnetic and bulk nonmagnetic contributions.[8]

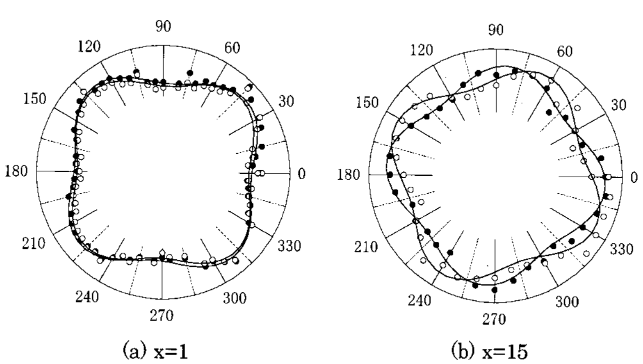

FIG. 1. Polar plots of the azimuthal angle dependence of MSHG intensity in Fe(xML)/Au(xML) superlattices with (a) $x=1$ and (b) $x=15$.

In the superlattices with noninteger values of x, the similar anisotropic behaviors were observed. However, both the anisotropy and the magnetic effect seem less prominent for thin layers ($1<x\leqslant 2.5$) than for thick layers ($x\geqslant 2.75$). In the former, the ratios of the fractionally occupied layers to the fully occupied layers are larger than in the latter, which may be the reason why the anisotropy becomes obscure in the former.

The theoretical analysis resulted in a conclusion that the P_{in}–P_{out} MSHG intensity I of the longitudinal Kerr geometry could be expressed by the following equation:[8]

$$I=|A+B\cos 4\phi \pm C\sin 4\phi|^2. \tag{1}$$

The surface nonmagnetic SHG response of the electric dipole origin gives rise to isotropic signal and contributes to the parameter A. The bulk nonmagnetic SHG response of the electric quadrupole origin causes an anisotropic contribution (the parameter B) for uniaxial 4/mmm symmetry (the present case). The surface magnetic response comes from the electric dipole term expanded by magnetization and contributes to the parameter C. Details of the analysis will appear in later publications.

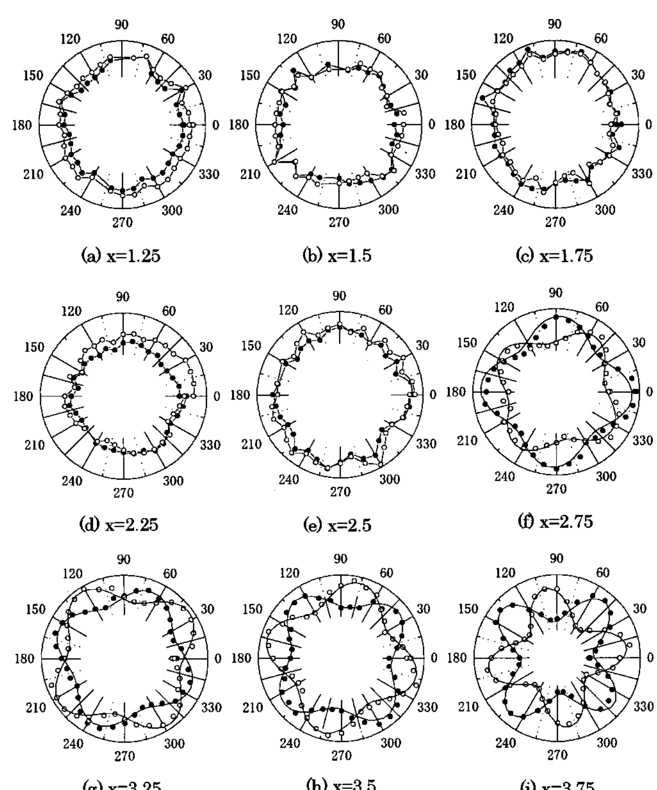

FIG. 2. Polar plots of azimuthal patterns of MSHG signal in superlattices with (a) $x=1.25$, (b) $x=1.5$, (c) $x=1.75$, (d) $x=2.25$, (e) $x=2.5$, (f) $x=2.75$, (g) $x=3.25$, (h) $x=3.5$, and (i) $x=3.75$.

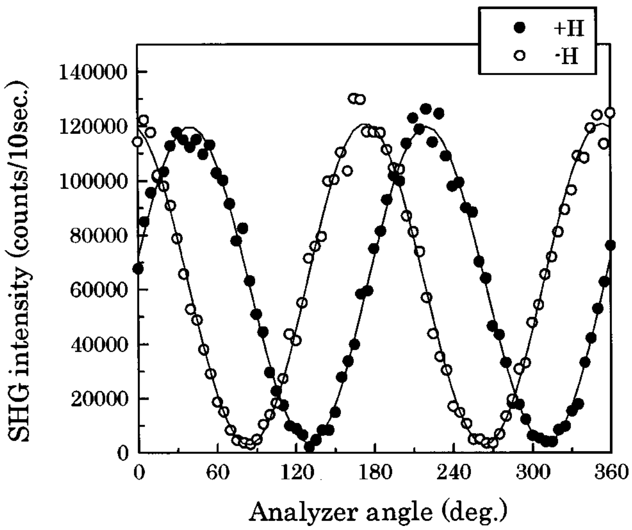

FIG. 3. Azimuthal patterns calculated using Eq. (1) for four sets of parameters.

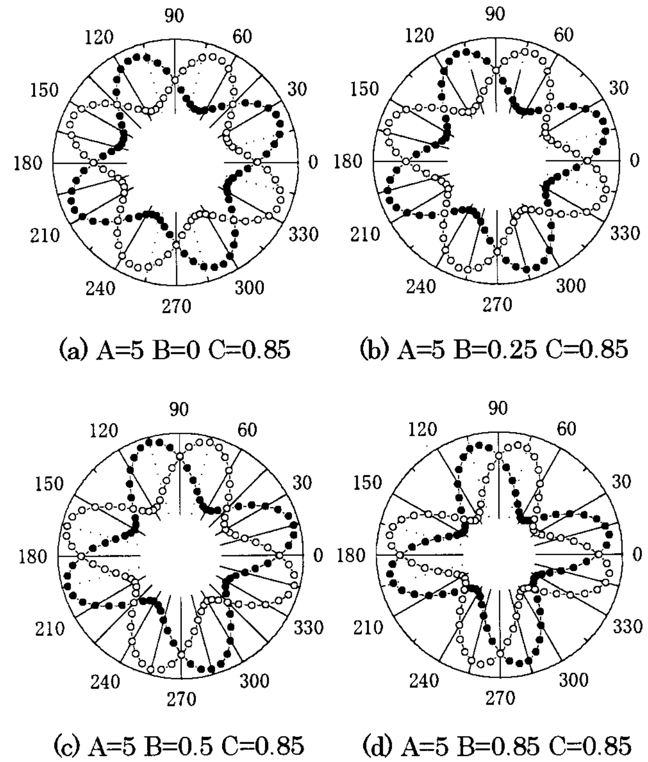

FIG. 4. Analyzer angle dependence of SH signal in Fe(3.75 ML)/Au(3.75 ML) superlattice for two opposite directions of the magnetic field.

The simulated results using Eq. (1) for $A=5$ and $C=0.85$ are plotted in Fig. 3 for $B=0$, 0.25, 0.5, and 0.85. It is found that the change of azimuthal pattern due to the field reversal depends strongly on the relative contribution of the parameters B and C. For B much smaller than C, the polar patterns show 45° rotation for the magnetization reversal, while for B comparable to C, the patterns undergo a smaller rotation.

The solid lines in Figs. 1 and 2 are the simulated curves that give the best fit to the experimental points using Eq. (1). Keeping the parameter $A=5$, the parameters B and C were determined as follows: $B=0.28$ and $C=0.02$ for $x=1$; $B=0.21$, $C=0.18$ for $x=15$; $B=0.14$ and $C=0.85$ for $x=3.75$; $B=0.18$ and $C=0.36$ for $x=2.75$.

The analyzer angle dependence of SH signal from the $x=3.75$ superlattice is plotted in Fig. 4. From this figure the nonlinear magneto-optical Kerr rotation was calculated. The rotation angle was determined as 23.2°, while the values for $x=1$ and $x=15$ superlattices were 2.43° and 3.2°, respectively. The nonlinear Kerr rotation is fairly large compared with linear Kerr rotation, which is no greater than 0.3°. Theoretical studies are now underway to understand why the fractional superlattice shows a large nonlinear Kerr rotation.

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