Magneto-Optical Spectra in Fe(xML)/Au(xML) Artificially Ordered Superlattices

K. Sato, T. Kondo,[a] J. Abe, H. Ikekame, M. Sano,* S. Mitani,*
K. Takanashi,* and H. Fujimori*

Faculty of Technology, Tokyo University of Agriculture and Technology,
2-24-16 Nakacho, Koganei, Tokyo 184
*Institute for Materials Research, Tohoku University,
Katahira, Aoba-ku, Sendai, Miyagi 980-77

The spectra of the magneto-optical Kerr effect and optical constants were measured in artificially ordered superlattices consisting of 1 to 5 monoatomic layers of Fe and Au. The Kerr rotation spectra showed a prominent structure around 4 eV, which undergoes a shift with layer thickness, the spectral features being quite different from those calculated by the virtual optical constant method which assumes that the superlattice is composed of pure metallic layers. This result suggests the formation of a new ordered alloy with a period extending up to 5 mololayers, which do not exist in the binary phase diagram of thermal equilibrium. The magneto-optical structure around 4 eV is attributed to the Au 5d → Fe 3d transition, on the basis of results of the energy band calculation.

Key words: artificial superlattice, Fe(x mololayers)/ Au(x monolayers) ($x=1, \cdots, 5$) ordered structure, magneto-optical effect, energy band structure

1. Introduction

Recently magnetic and optical properties in ultrathin Fe films grown epitaxially on Au substrates are attracting much interest. Enhanced magnetic moment of Fe has been proposed in this system based on the results of band calculations.[1] Magneto-optical spectra in ultrathin Fe of films of a few monolayers (ML) thickness show anomalous structures, the energy positions of which are dependent on the film thickness.[2] The spectral features have been interpreted in terms of the quantum size effect, by which the electronic wavefunctions are confined to form spin-polarized quantum levels.[3]

The equilibrium binary phase diagram of Fe-Au system has been known to show a complete immiscibility between Fe and Au, the fact being in favor of the formation of ideal Fe/Au superlattices since the intermixing between neighboring layers could be minimized. For Fe/Au superlattices with the period thicker than 5 nm, optical spectra were successfully explained by the combination of two metallic layers using classical optical treatments.[4] However only a few studies have been conducted on Fe/Au superlattices with period of ML thickness due to difficulty in preparation of well-defined superlattice structures. Suzuki and Katayama studied Fe(3Å)/Au(4Å) and Fe(3Å)/Au(8Å) superlattices and found a new magneto-optical transition around 4 eV and explained in terms of the quantum confinement effect.[5]

Recently we have succeeded in preparaing good quality superlattices of [Fe(1ML)/Au(1ML)]$_n$ by the MBE technique, in which formation of ordered structure of L1$_0$ type has been confirmed by X-ray analyses.[6]

In order to clarify whether the electronic structure of the material can be expressed simply by the stacking of two different band structures of respective metallic layers or by that of the compound between Fe and Au, systematic optical and magneto-optical studies were conducted in [Fe(xML)/Au(xML)]$_n$ superlattices with different layer thickness x. The rest of the paper describes the results of these measurements.

2. Experimental

The samples used in the present study were prepared by the MBE technique on MgO substrates with thick Au buffers (500 nm), the details of which were described in ref. 6. The designed layer thickness and the number of period are listed in Table 1. Magneto-optical Kerr rotation and ellipticity were measured at room temperature between 1.2 and 6 eV using the specially designed Kerr spectrometer.[7] The maximum field applied was 1.7 T. Optical reflectivity was measured using a Hitachi U-3410 spectrophotometer, from which optical constants were deduced by the Kramers-Kronig analysis with the help of the data values measured by a Woollam type WVASE spectroscopic ellipsometer between 2 and 5 eV.

3. Results and Discussion

Figure 1 shows spectra of magneto-optical Kerr rota-

[a] Present address: Tokyo Institute of Technology, *Nagatsuta, Yokohama 227.*

tion θ_K (solid curves) and ellipticity η_K (dotted curves) in the [Fe(xML)/Au(xML)]$_n$ (x=1-5) superlattices. The peak value of rotation below 4 eV do not exhibit a drastic change with layer thickness. A prominent dispersion-type structrue centered at 3.6 eV is observed in the Kerr rotation spectrum for x=1, which undergoes a systematic "blue shift" as x is increased. The total energy shift between x=1 and 5 samples amounts to approximately 1 eV. An additional structure at low energy region seems to show a systematic "red shift" with thickness, the tendency being obvious in thicker layers.

Kerr rotation and ellipticity were simulated by means of the virtual optical constant method[8] using the reported values of optical constants of Fe and Au, as well as the values of magneto-optical constants of Fe described in the literature. The calculation in the Fe(1ML)/Au(1ML) superlattice was performed using the a'_{Fe} and a'_{Au} thicknesses of Fe and Au layers, respectively, calculated from the lattice spacing $2d_{200}$ obtained by XRD, as follows:

$$a'_{Fe(1ML)} = 2d_{200} \times \frac{a_{Fe}}{a_{Fe}+a_{Au}}$$

$$a'_{Au(1ML)} = 2d_{200} \times \frac{a_{Au}}{a_{Fe}+a_{Au}}. \tag{1}$$

where a_{Fe} and a_{Au} represent lattice constants of Fe and Au crystals, respectively.

For $x \geq 2$, it was assumed that atoms at the interface and those in the layers sandwitched by interfaces take different thickness values; i.e., the same value as in x= 1 was used as the thickness of interfacial Fe and Au, while atomic spacings of bulk Fe and Au were used for atom-layers sandwitched between two interface layers. Layer thicknesses used in the calculation are also listed in Table 1, in which the third and fourth columns provide values of the assumed layer thickness of Fe and Au, respectively, and the fifth column gives the total thickness for one period. The spectra of magneto-optical Kerr rotation and ellipticity simulated in this way are illustrated in Fig. 2, for x=1 and 5. Both rotation and ellipticity for x=5 show remarkable agreement with those for x=1. A sharp peak around 2 eV and two small peaks around 4-5 eV are observed in θ_K,

Table 1 Structure parameters of the Fe(xML)/Au(xML) superlattices

Structure Fe(xML)/ Au(xML)	Number of Period (Å)	Thickness of Fe (Å)	Thickness of Au (Å)	Total thickness (Å)
x=1	100	1.568	2.232	3.8
x=2	50	3.001	4.271	7.272
x=3	33	4.434	6.310	10.744
x=4	25	5.867	8.349	14.216
x=5	20	7.300	10.388	17.688

regardless of the unmber of monolayers x. The peak at 2 eV may be ascribed to the plasma enhancement of Kerr effect. It is quite obvious that the experimental spectra shown in Fig. 1 cannot be reproduced by the simple optical calculations.

In order to clarify whether or not the magneto-optical structures observed around 4 eV in Fig. 1 are related to the plasma frequecies shifted from the original Au position, real and imaginary parts of dielectric permeability of the superlattices were evaluated from the reflectivity spectra. The result for x=1 is shown in Fig. 3 (No drastic change was observed for samples with $x \geq 2$). This spectrum clearly denies the presence of plasma resonance around 4-5 eV, since the real part of the dielectric functions was not observed to cross the abscissa. From experimentally obtained data of θ_K, η_K,
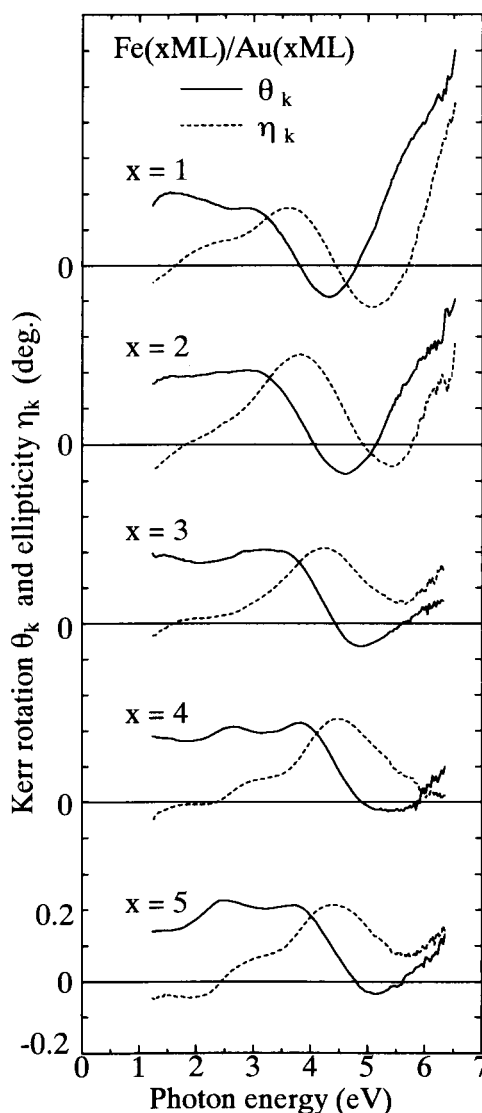

Fig. 1 Spectra of Kerr rotation θ_K (solid curve) and Kerr ellipticity η_K (dotted curve) in the Fe(xML)/ Au(xML) superlattices for $1 \leq x \leq 5$.

and ε_{xx} we evaluated ε_{xy} using the following formula,[9]

$$\theta_K + i\eta_K = \frac{\varepsilon_{xy}}{(1-\varepsilon_{xx})\sqrt{\varepsilon_{xx}}} . \qquad (2)$$

From ε_{xy} we further calculated real and imaginary parts of $\omega\sigma_{xy}$, which is known to be useful for discussion of magneto-optical effect in terms of the electronic structures.[10] The obtained spectra are given in Fig. 4.

It is found that $\omega\sigma_{xy}''$ takes an approximately constant value ($\sim -2\times10^{29}$ s^{-2}) between 1 and 4 eV, on which a peak around 4–5 eV is superposed. Erskine and Stern argued that constant value of $\omega\sigma_{xy}''$ can be correlated to the spin polarization of conductions electrons.[11] This means electronic states of the present material at the Fermi surface are quite different from those of simple Fe or Au.

We believe that the 4 eV transition is originated from band-to-band transition in the new energy band scheme of the superlattice. To confirm this point we made a rough estimate of $\omega\sigma_{xy}''$ using the density of states (DOS) curve obtained by the electronic band structure calculations in the Fe[1ML]/Au[1ML] super-lattice.[12] The spectrum of $\omega\sigma_{xy}''$ was evaluated by taking an energy-derivative of the joint density of states derived from a convolution between the filled and the empty states in the DOS curves, the same procedure having been described elsewhere[13] The estimated spectrum of $\omega\sigma_{xy}''$ is shown in Fig. 5. A distinct peak is found around 4 eV in the spectrum, which arises mainly from
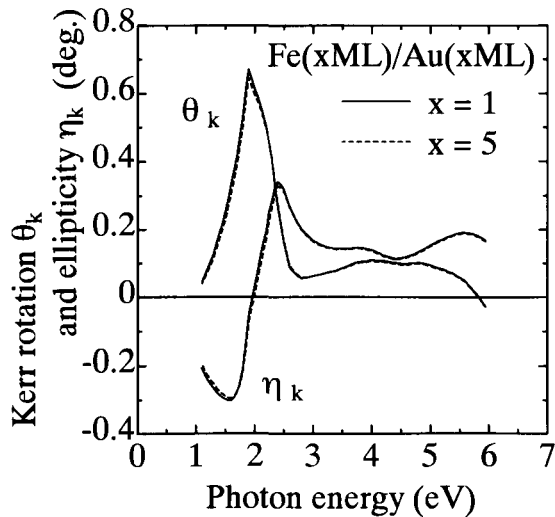

Fig. 2 Simulated spectra of Fe(xML)/Au(xML) super-lattices for $x=1$ (solid curve) and $x=5$ (dotted curve).
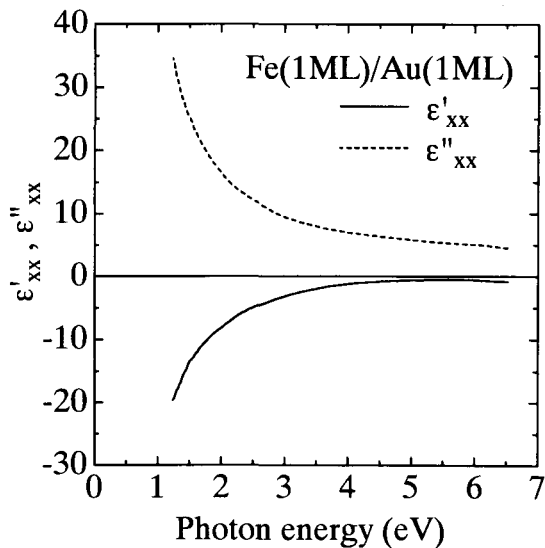
Fig. 3 Spectra of real (solid) and imaginary (dotted) parts of the diagonal dielectric tensor element.
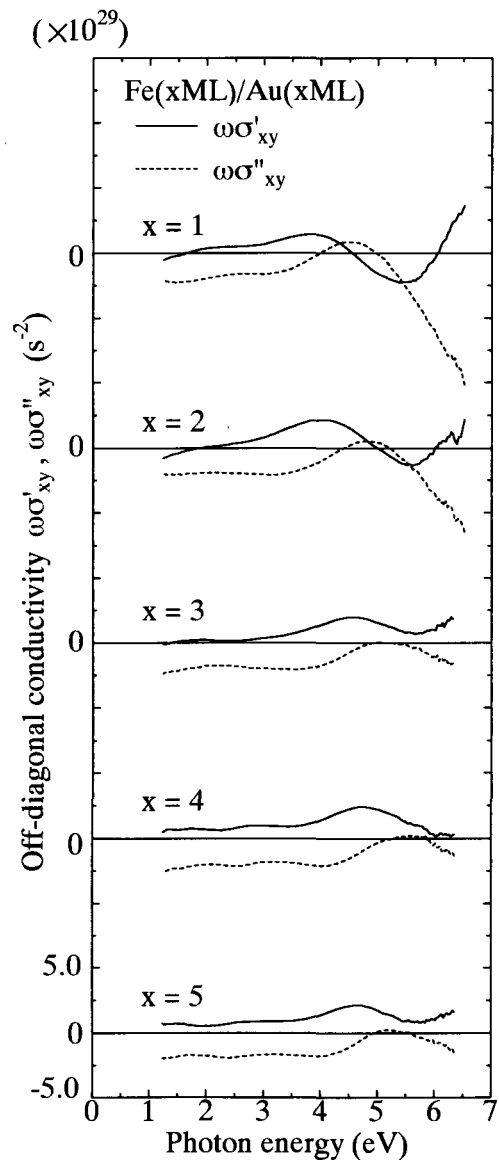

Fig. 4 Spectra of the real (solid) and imaginary (dotted) parts of the off-diagonal conductivity element multiplied by angular frequencry, $\omega\sigma_{xy}''$ in Fe(xML)/Au(xML) superlattices.
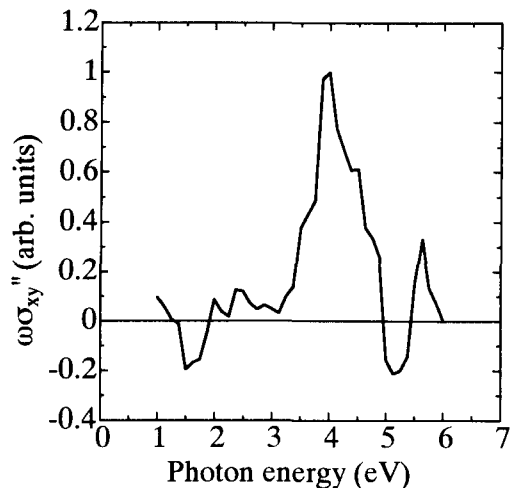
Fig. 5 A spectrum of $\omega\sigma''_{xy}$ estimated from the band calculation of the Fe(1ML)/Au(1ML) superlattice (see the text).

the filled 5 d states (down spin) of Au to the empty 3 d states (down spin) of Fe. This result supports the idea that the Fe[1ML]/Au[1ML] superlattice is not a simple stack of Fe and Au layers but a new ordered alloy between them.

Concerning the systematic shift of the magneto-optical structures with layer thickness, we simulated spectra for $x \geq 2$ by virtual optical constant method assuming that the superlattice consists of interfacial "$x=1$" alloy and "bulk" layers of Fe and Au. However, we failed to explain the shift, as the calculated peak remained at the same position up to $x=5$. This fact suggests the formation of ordered alloy with period extending to as far as 5 MLs. The formation of minizone due to the folding of the Brillouin zone may also be suspected as the origin of the systematic shift.

The peak-shift of $\omega\sigma''_{xy}$ around 4–5 eV may also be explained by quantum confinement effect as proposed in ref. 5. This interpretation is based on the assumption that the electronic structures of bulk Au and Fe are preserved still in the thin layers of the monoatomic thickness. However, it is obvious from the present experiments that the electronic structures of Au in this

material is quite different from bulk, since no plasma enhancement was observed and conduction electrons at the Fermi surface were found spin-polarized. Nevertheless, we cannot completely rule out the possibility of the quantum confinement effect for thicker layers. Further studies are necessary to clarify this point.

4. Conclusions

From the optical and magneto-optical spectra it has been deduced that the electronic structure of the Fe/Au superlattice is completely different from that estimated by the simple stack of Fe and Au layers. Spin polarization of conduction electrons is suggested from the off-diagonal conductivity. Magneto-optical structure around 4 eV was attributed to an optical transition between Au 5 d and Fe 3 d states.

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