New Magneto-Optical Transition in Ultrathin Fe(100) Films

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Magneto-optical polar Kerr effects in ultrathin bcc Fe(100) films sandwiched by Au layers were investigated in the 1.6–5.3 eV photon energy range at room temperature. A new peak appeared in the off-diagonal conductivity spectrum at around 3.5–4.5 eV for the Fe films thinner than 10 Å. The peak energy is lower for thinner Fe films. The absolute value of ε_{xy} at 3.7 eV in the 3 Å thick Fe film is 2 times as large as that in the bulk Fe.

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Studies on ultrathin magnetic films and artificial superlattices are useful in testing the limits of band theory, exploring new phenomena, and establishing a technology to control the electronic properties of materials. For these purposes, surface sensitive measurement techniques, such as spin-resolved photoemission spectroscopy, have been used. Recently, it has been shown that the magneto-optical effect is a useful phenomenon when utilized for the highly sensitive measurement of the magnetization of ultrathin films. This measurement is referred to as the surface magneto-optical Kerr effect (SMOKE) method [1]. Magneto-optical effects of ultrathin films were measured *in situ* for Fe/Au [1], Fe/Ag [2,3], and other systems [4], and *ex situ* for Fe/Cu [5,6] and Co/Au systems [7].

These magneto-optical measurements were carried out at a fixed wavelength (employing a laser light source) and no wavelength-dependence information was reported except in a few cases [5]. The electronic contribution to magneto-optical properties has also not been discussed, though large enhancements of the effects were found [6,7]. A spectroscopic measurement of magneto-optical effects provides important information on magnetic band structures [8] and is also interesting from an application point of view because a high density rewritable magneto-optical memory requires a large magneto-optical effect in short-wavelength visible and ultraviolet regions.

From the wavelength dependence of the Kerr rotation in thin films and multilayered films, Katayama *et al.* showed that the magneto-optical Kerr rotation was enhanced at the plasma edge of noble metals which were multilayered with magnetic metals [5,9]. This phenomenon was well explained by the classical optics multilayer model for the films thicker than 20 Å [5,10] and is not related to the modification of electronic structures.

In this Letter, we present the photon energy dependence of magneto-optical conductivity in ultrathin epitaxial Fe(100) films sandwiched between Au(100) layers. A new peak observed in the spectra is discussed in relation to the results of band calculations [11–14] and photoemission spectroscopies [15,16].

The films were made by the molecular beam epitaxy (MBE) technique employing electron beam gun sources for Fe and Au and a Knudsen cell for Ag [17]. The MBE chamber was maintained in the 10^{-10} Torr range during deposition. (100) cleaved surfaces of MgO single crystals were used as substrates. After a thermal flash at 900 °C, a 2000 Å thick fcc Ag(100) buffer layer and a 2000 Å thick fcc Au(100) layer were deposited at room temperature. These films were annealed at 350 °C for several minutes. After cooling to room temperature, a bcc Fe(100) layer was deposited and capped by the deposition of a 20 Å thick Au(100) layer. The room-temperature growth of an Fe layer on the Au(100) surface prevents surface segregation and alloying [18] and provides a sharp diffraction pattern [1]. The thicknesses of the films were measured by a quartz thickness monitor and controlled by a shutter system. The film thickness and growth mode were monitored by reflective high-energy electron diffraction (RHEED) intensity oscillation. The error of the average thickness is less than 0.2 Å. Several samples with Fe layers of various thicknesses from 2 to 100 Å were prepared.

The room-temperature polar Kerr rotation of the samples was measured *ex situ* by a Kerr rotation spectrometer using a Faraday modulator (JASCO-250) in the photon energy range from 1.5 to 5.4 eV. The incident angle was 10° from the surface normal. Kerr ellipticity was calculated from the value of the Kerr rotation using Kramers-Kronig (KK) relations [Eq. (40) in Ref. [19]]. Integration was performed only for the measured range and no extrapolation was used. To check the validity of the KK transformation using a spectrum of limited energy range, the magneto-optical ellipticity of a sample with a 3 Å thick Fe layer was directly measured by the equipment using a piezoelastic modulator [20]. Deviation between the measured spectrum and the transformed spectrum is about 7% of the full scale of the spectrum. This deviation does not influence the following discussion.

Figure 1 shows RHEED intensity oscillations during

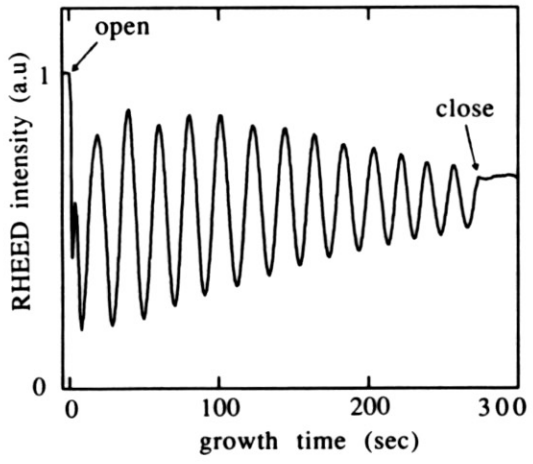

FIG. 1. RHEED intensity oscillations during the growth of a 20 Å (13.95 ML) thick Fe(100) layer on a Au(100) surface at room temperature. The shutter system was controlled by a quartz oscillator thickness monitor. The deposition rate was 0.74 Å/sec. The specular spot intensity was observed under the following conditions: $E=20$ keV, $\theta=0.3°$, electron beam∥[100] azimuth of Au.

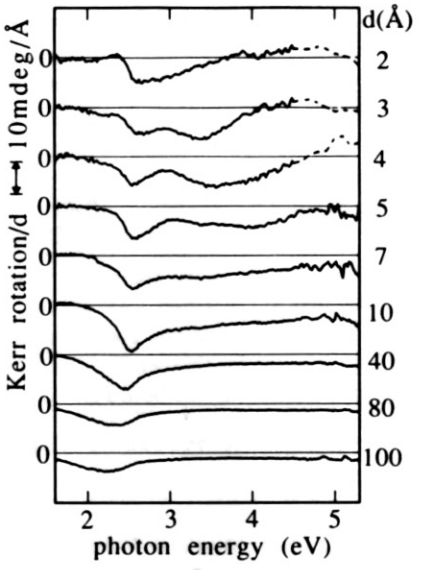

FIG. 2. Magneto-optical polar Kerr rotation spectra for the Au(20 Å)/Fe(d Å)/thick-Au(100) sandwiched films at room temperature.

the growth of a 20 Å thick Fe film on a Au(100) surface. The first, unexpected peak is considered to be related to the disappearance of the reconstruction pattern of the Au(100) surface at 0.3 ML (monolayer). The long lasting intensity oscillation indicates the layer-by-layer growth of the Fe film.

Figure 2 shows the Kerr rotation spectra normalized by the Fe film thickness at room temperature for several Fe film thicknesses. Only one large peak at 2.5 eV was observed for the film with an Fe thickness of 100 Å. The peak coincides with the coupled plasma edge of Au. The spectrum is similar to the spectra observed for the Fe/Au multilayered films prepared by the sputtering method [9], and is attributed to an interference effect [5,10]. No other peak was observed for the films with Fe thicker than 40 Å. For the films with thinner Fe layers, however, a new peak appeared at around 4 eV. The peak energy depends on the thickness of the Fe layer. It shifts to lower energy in thinner films. The 2 Å thick Fe film is not ferromagnetic at room temperature. Rotation is the value at 7.6 kOe here.

When an Fe magnetic layer and a Au cap layer are grown on a Au substrate successively and the thicknesses of those layers are thin enough, the complex polar Kerr rotation for normal light incidence is approximated by the following equation [21]:

$$\phi+i\eta=i\frac{4\pi d}{\lambda}\frac{\varepsilon_{xy}}{1-\varepsilon_{xx}^{s}}, \tag{1}$$

where ϕ and η are the Kerr rotation and Kerr ellipticity of the reflected light, respectively. λ is the wavelength of light, d is the thickness of the magnetic layer, ε_{xy} is the off-diagonal part of the dielectric tensor of the magnetic layer, and ε_{xx}^{s} is the diagonal part of the dielectric tensor of the substrate. In the formula, there is no contribution from the diagonal parts of the dielectric tensor of a magnetic layer. Therefore, from normalized Kerr rotation, $(\phi+i\eta)/d$, we can clearly find a change in ε_{xy}.

Using (1) and KK relations, an off-diagonal element of the dielectric tensor of Fe, ε_{xy}, was calculated. In the calculation, the value of ε_{xx}^{s} reported by Johnson and Christy [22] was used. Figure 3 shows the real part of ε_{xy}, which corresponds to the imaginary part of σ_{xy}, in Fe layers for various film thicknesses, excluding the 80 and 100 Å thick Fe films. In those thick Fe films, the linear approximation of (1) will yield a large error because of absorption in the Fe layer. In the figure, $-\varepsilon_{xy}'$ is multiplied by the square of the photon energy to make the contribution from the intraband transition constant. The ε_{xy}' spectrum of bulk Fe is also shown for comparison [23]. The value of ε_{xy}' for the 40 Å thick Fe sample is in good agreement with those of bulk Fe. The peak of the Kerr rotation at around 2.5 eV is completely removed in the ε_{xy} spectra due to multiplication by $1-\varepsilon_{xx}^{s}$. This shows that the origin of the enhancement of the Kerr rotation at 2.5 eV is the plasma edge effect of Au layers.

For a 10 Å thick Fe layer, a new broad dispersion-type structure appears at 4.5 eV. A dispersion is also observed at the same energy for a 7 Å thick Fe layer. It shifts to lower energy for Fe films thinner than 5 Å. The center of the dispersion for a 2 Å thick Fe layer is about 3.5 eV. The amplitude of the dispersion is largest for the 4 Å thick Fe layer. The absolute values of ε_{xy} for the 3 Å thick film is about 2 times as large as that in the bulk Fe at 3.7 eV.

Ag/Fe/Au and Au/Fe/Ag sandwiched structures were also investigated in order to elucidate the effects of im-

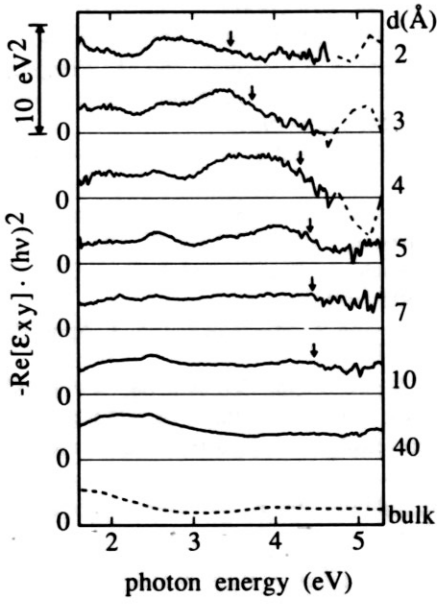

FIG. 3. Off-diagonal elements, $-\varepsilon'_{xy}$, of the dielectric tensor of Fe layers. $-\varepsilon'_{xy}$ is multiplied by the square of the photon energy. The broken line shows the data of bulk Fe after Krinchik and Artem'ev [23].

perfections in the layer structures of Fe. The 3 Å thick Fe layer which was grown on the Au(100) surface and covered by a Ag(100) layer, Ag/Fe/Au, shows a similar enhancement of ε_{xy} at around 4 eV. On the other hand, the sample with an Fe layer of the same thickness but which was grown on the Ag(100) surface and covered by a Au(100) layer, Au/Ge/Ag, does not show any enhancement of the ε_{xy} at around 4 eV.

This result shows that the new dispersion is very sensitive to the layer structure of Fe. Fe grows layer by layer on a Au(100) surface and also on a Ag(100) surface [24]. The surface free energy of Ag, however, is lower than that of Au, whereas the surface free energy of Fe is the largest in these three metals. Therefore, a perfect layer-by-layer growth is expected for the Fe/Au(100) system rather than for the Fe/Ag(100) system. Actually, the growth of Fe/Ag(100) shows considerable irregularity for the layer growth periods and broadening of diffraction spots [3,24], while the RHEED intensity oscillation of the Fe/Au(100) system has constant periods (see Fig. 1). These facts clearly manifest that the new dispersion at around 4 eV appears only in the samples having a perfect layer structure.

We believe that the new ε_{xy} structure is not caused by the optical interference effect since such an effect can be totally explained using (1), but is caused by a change in electronic structures due to the ultrathin layer arrangement.

Reim *et al.* [25] ascribed the magneto-optical structure observed at 1.9 eV in the FePd alloys to the effect of spin polarization in the Pd 5*d* band. Such an effect should also be considered as a cause of our new dispersion at 4 eV. However, we believe this is not the case in Au/Fe/Au for the following reasons. First, the magnetic moment induced at a Au atom adjacent to an Fe layer was estimated theoretically to be $(0.03–0.08)\mu_B$ [12], which is considerably smaller than the value of $0.35\mu_B$ [25] induced at a Pd atom in the FePd alloys. Second, there may be an objection that even though the induced moment at the Au atom is small, the large spin-orbit interaction in the Au atom will more than compensate and will introduce a magneto-optical effect comparable to the Pd in the FePd. We renounce this argument since we observed the distinct difference between Au/Fe/Ag and Ag/Fe/Au structures, which should bring about an identical effect if Au atoms adjacent to the Fe layer contribute to the magneto-optical dispersion.

Theoretical works on the electronic structures of ultrathin Fe layers [12,13] and surfaces [14] have predicted a narrowing of the bandwidth, enhancement of the magnetic moments of the Fe layer, induced moments in the nonmagnetic layer adjacent to the Fe layer, and an appearance of Fe *d*-surface bands between bonding and antibonding *d* bands. Experiments on photoemission spectroscopy have shown the appearance of a *d*-surface band [5] and narrowing of the *d* band [16]. These characteristic electronic structures should be reflected in the magneto-optical effect.

We tentatively attribute the new magneto-optical dispersion to the increased state density in the surface 3*d* band of the ultrathin Fe layer. Moreover, the energy shift of the new dispersion in the ε'_{xy} spectra seems to be related to the band narrowing. To clarify these points, a semirelativistic or a fully relativistic band calculation which can include spin-orbit interactions will be necessary.

Additionally, enhancements of the magneto-optical effects in the Fe/Cu [6] and the Co/Au [7] multilayers have been observed at 633 nm. In our experiments, ε_{xy} is small at 633 nm (2.0 eV) for the ultrathin Fe films. The mechanism of the enhancement at 2.0 eV should be different from that of the new dispersion around 3.5–4.5 eV.

In conclusion, we found a new dispersion in the ε'_{xy} spectra of the ultrathin bcc Fe(100) layers sandwiched by Au layers. This new dispersion appears at 4.5 eV for a 10 Å thick Fe film and shifts towards lower energy with decreasing thickness. The absolute value of ε_{xy} for a 3 Å thick Fe film is twice as large as that in bulk Fe at 3.7 eV. This phenomenon shows the possibility of controlling electronic and magnetic properties of metallic systems by forming ultrathin films and superlattices.

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