THE MAGNETO-OPTICAL QUANTUM SIZE EFFECT IN bcc-Fe(001) AND (110) ULTRATHIN FILMS

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Abstract. Recently, the oscillation of the magneto-optical effect with respect to the ferromagnetic layer thickness was observed for bcc-Fe(001) ultrathin films. The oscillation periods have been explained by the theory concerning the formation of quantum well states in the ferromagnetic layer. In this paper, a physical picture how the electron confinement produces the quantum size effect of the magneto-optical responses is introduced, and experimental results on the magneto-optical effects of the bcc-Fe films of different orientations are presented. The (001) oriented films show clear oscillation. On the other hand, the (110) oriented films does not show any oscillation. The reason is discussed from the band structure and growth modes.

1. INTRODUCTION

By continuous efforts of many researchers in the field of magnetic ultrathin films and multilayers, now, it is available to control the thickness of magnetic layers within monatomic layer level, although it is really possible only for several special combinations of substrates and overlayers. This new technology allows us to investigate some basic physical properties like oscillating magnetic interlayer coupling, giant magneto-resistance and interface magnetism, and also gives us opportunities to create new application devices based on such physical properties of the magnetic ultrathin films and multilayers [1]. In those findings and inventions, the interference of the scattered electron in the ultrathin metallic multilayers have been playing a central role.

In a well defined ultrathin film, an interference of the electrons which are scattered at the interfaces composes a confined state (quantum well states and resonance states) in the film. The oscillation of the exchange coupling of two ferromagnetic layers through non-magnetic spacer-layer [2] can be explained by the occurrence of spin polarized quantum well states (QWS) at the Fermi energy in the nonmagnetic spacer layer [3]. The exchange coupling oscillates also as a function of ferromagnetic layer thickness [4] due to an occurrence of QWS at the Fermi energy in the ferromagnetic layers [5]. A lot of experimental data on the influence of the spacer layer on the coupling character can be found in literature. Such QWS's of occupied states and unoccupied states in metallic systems have been observed directly by the photoemission spectroscopy [6] and the inverse photoemission spectroscopy [7].

In 1992, it was also shown that the QWS's could have large contributions to the magneto-optical (MO) effect [8]. Oscillations of magneto-optical Kerr rotation and/or ellipticity as a function of ferromagnetic layer thickness in Au/Fe/Au(001) [9] and the nonmagnetic layer thickness in Fe/Cu/Fe(001) [10], Fe/Au(Ag)/Fe(001) [11], Au/Co/Au(111) [12], and Ru/Co(0001) [13] have been reported. These results suggest the existence of quantum size effects in the MO response of magnetic metals. Oscillation periods observed for MO effect are usually different from that of magnetic interlayer coupling and have a photon energy dependence [9, 11, 12], excepting some cases [10, 13]. The oscillation of the MO effect as a function of Fe (001) layer thickness shows a clear energy dependence and was explained by a concept of QWS formation along to the band dispersion of the bcc-Fe [9, 14]. Experiments on the MO properties
as a function of the Au thickness in Au/Co/Au(111) system evidence the existence of QWS's of completely confined in a Au over layer [12]. These results may be understood as an effect of the QWS formation to the interband transition by photon absorption.

On the other hand, recently, Carl et al. found a sharp quantum-well like resonances in the field dependent polar Kerr effect [13]. They explained this effect by a QWS formation at Fermi energy. The QWS at Fermi energy affects to interband transition through Pauli paramagnetic behavior in the Ru layer. This may have essential relation with the giant magneto-resistance effect [1] and will open one of very interesting issue of the thin film magnetism.

These results are interesting not only for the basic physics but also MO applications. These phenomena provide a conventional and powerful method to study the QWS's in the materials and offers a possibility to design new magneto-optical recording media with large magneto-optical Kerr rotation. One of a key point to improve recording densities of the MO recording media (photo rewritable magnetic recording disk which is written by the heat of laser spot and read out using MO effect) is to realize larger MO effect at shorter wavelength region.

In this paper, we will concentrate to describe the quantum size effect in MO responses which has been observed in the Fe ultrathin layers. A physical picture why and how QWS's contribute to the MO oscillations will be given demonstrating experimental results on the MO properties of Fe (001) and (110) ultrathin films.

2. THEORY

Magneto-optical Kerr effect is an effect that is observed in the reflection of the light on the surface of magnetic materials. For a linear polarized incident light, polarization of the reflected light becomes elliptic (Kerr ellipticity: ηK) and its principal axis rotates (Kerr rotation: ΦK) by the difference in absorption of the right circular polarized light from left circular polarized light [15].

In Fig. 1 dipole transitions from flat and completely spin polarized d(x*iy)z bands to a cosine like pz band is shown to explain the MO effect. By the selection rule, the right (left) circular polarized light can excite an electron in the d(x*iy)z (d(x*iyr)) band to the pz band. If d(x*iyr) bands are separated, ΔSO, by the spin-orbit coupling, those absorption do not cancel each other and produce MO effect. For a small ΔSO, the MO effect is proportional to ΔSO and energy differentiation of the density of state (DOS) of the pz band [16]. In the case of ultrathin films of ferromagnetic materials, the pz band separates to 2D-subbands and the DOS will have many steps caused by band edges of the 2D-subbands as shown in Fig. 2. An energy differentiation the DOS of the film, thus MO spectrum, shows many peaks at edge of the 2D-subbands. The position of the peaks changes with film thickness and yield oscillation of the MO effect as a function of the film thickness [17].

![Fig. 1 Schematic picture of a band structure of ferromagnetic material giving magneto-optical effect. Fermi energy is in between spin split dxy, yz bands. The dxy, yz bands are split to dxy and dxy, yz bands by the spin-orbit coupling. By the selection rule, the right (left) circular polarized light can excite an electron in the dxy, yz band to the pz band.](image1)

![Fig. 2 Schematic picture of a density of states (DOS) of an ultrathin film. By a formation of 2D sub-bands, DOS has steps. Dotted line shows a case of infinite film thickness. Lower part of the figure shows an energy differentiation of the DOS corresponding to a MO spectrum.](image2)
Next we will consider how this oscillation period is determined. To do so, let us consider a ferromagnetic ultrathin film of thickness $d$ (see Fig. 3). An electron traveling rightward will be reflected toward the leftward by the right side surface. Superposition of the going wave (wave vector $k_z^+$) and the reflected wave (wave vector $k_z^-$) makes a standing wave confining in the film (Fig. 4). Phase change of the wave function of the electron during a travel from center of the film to the center of the film via both surfaces is:

$$
\Delta \theta = (k_z^+ - k_z^-) \cdot d + \text{Arg}(r^+) + \text{Arg}(r^-),
$$

where $r^+$ ($r^-$) is a complex reflectivity of the electron at the right (left) surface. Absolute value of the complex reflectivity gives a degree of the electron confinement [18] and its argument gives a phase change by a reflection. A standing wave can be constructed only when this phase change equals to $2n\pi$ ($n$: integer). As a result, if we change the film thickness, a standing wave (quantum well state / resonance state) appears periodically at given energy level and modulates magnitude of the DOS. The period is:

$$
\Lambda = \frac{2\pi}{q},
$$

where $q$ equals to a scattering vector ($q = k_z^+ - k_z^-$). For the magnetic coupling problem of the magnetic multilayers, we considered electrons at Fermi energy so $q$ was equal to $2k_F$. Now we are considering about an optical transition in the film. Therefore, $q$ should be determined by Fermi's golden rule and $k$-select rule ($A$ relaxation of the $k_z$ selection rule by limited film thickness will be discussed later).

$$
\epsilon_f(k_z^+,k_z^+) - \epsilon_i(k_z^+,k_z^+) = \hbar \omega,
$$

where $\epsilon_f(\mathbf{k})$ ($\epsilon_i(\mathbf{k})$) is the energy band dispersion of the host material of the ferromagnetic ultrathin film. Suffix $f$ and $i$ denote a final and initial state of the optical transition, respectively. $k//\text{ is an inplane wave vector}$. This equation define an 'iso-energy-difference surface' in the $k$-space. The initial point and final point of the scattering vector, $q$, are on the iso-energy-difference surface. From eq. (3), $q$ is a function of $k//\text{ and photon energy}$. It means that for a

![Fig. 3 Interference of the electron in an ultrathin film. $r^+$ represents complex reflectivity of the electron at surfaces.](image)

![Fig. 4 Sketch of the quantum well states (QWS's) and optical transition.](image)
given photon energy, each point in 2D-Brillouin zone has a contribution to the formation of a quantum well state with its own oscillation period. What we observe by the MO effect is a summation of these different oscillations over a 2D-Brillouin zone. As a result, only several \( q \)-vectors which have the stationary value with respect to a small change of \( k_\parallel \) have finite contribution to the resultant oscillation. The condition to be a stationary spanning \( q \)-vector is:

\[
\frac{\partial q}{\partial k_\parallel} = 0, \tag{4}
\]

This means that the tangential planes of the iso-energy-difference surface at initial and final point of the \( q \)-vector should be parallel for the stationary spanning \( q \)-vectors (see Fig. 5). As we can imagine easily, if a curvature of the iso-energy-difference surface at initial and final points of \( q \)-vector is small, the area in the 2D-Brillouin zone which contribute to the effect is wide and the effect will be large. These discussions are the same as that of the magnetic interlayer coupling \([18, 19]\) alternating the Fermi surface by an iso-energy-difference surface.

Here, we consider about an effect of a relaxation of the \( k_\parallel \)-selection rule. In Fig. 6 several quantum well states are indicated by small circles. From eq. (1) separation of the quantum well states in the \( k \)-space is \( \pi/D \). Heisenberg's uncertainty theorem, however, requires the uncertainty of the \( k_\parallel \)-vector is the same order as the separation of the quantum well states and seems to be able to merge the contributions from several quantum well states to the optical responses and kill the quantum size effect. Actually, for a transition from a flat band to a parabolic band, for example, relaxed \( k_\parallel \)-selection rule can merge transitions from several quantum well states on the flat band (right side of Fig. 6). On the other hand, we can excite only one quantum well state on the parabolic final band by a fixed photon energy because the transitions to the other quantum well states needs different excitation energy. As a result, we can only observe separate contribution from a quantum well state made from the bands that have larger \( z \)-direction group velocity of the electron.

Finally, the oscillation of the polar MO effect should be expressed as follows approximately:

\[
\phi_{k} + im_{k} = \sum_{\gamma} A_{\gamma} e^{i(k_{z} - \delta k_{z})}, \tag{5}
\]

![Fig. 5. Iso-energy-difference surface and its stationary spanning vector.](image)

![Fig. 6. Effect of the relaxation of the \( k_{z} \)-selection rule caused by limited film thickness. QWS's are indicated by open circles. QWS's on the flat band cannot be separated by the optical transition. But QWS's on the steep band will be separately observed by the optical transition because of energy selectivity of the measurement.](image)
where \( v \) represents a point in 2D-Brillouin zone in which \( q \) vector has its stationary value. \( A_v \) is an amplitude, \( \kappa_v \) is a constant which gives exponential decay of the oscillation. The exponential decay of the oscillation is related to a limited life time of the optical excitation. Critical thickness, \( d_c \), where oscillation amplitude reaches to \( 1/e \) is express by using the life time of the excitation, \( \tau \):

\[
d_c = \frac{\kappa_v}{\tau \times v_z}.
\]

Note that \( 2d_c \) equals to a distance that the electron travels during the life time.

From above discussions, it is expected that; (1) polar MO signal oscillates with respect to a change of the ferromagnetic layer thickness of ultrathin layered system, (2) the oscillation periods are given by inverse of stationary spanning vector on the iso-energy -difference surface, (3) The \( q \)-vector accompanying small curvature of the iso-energy-difference surface at its initial and final points has large contribution to the MO oscillation, (4) the quantum well states and resonance states made from the band which has large \( z \)-direction group velocity will contribute to the oscillation, and (5) oscillation amplitude decreases exponentially caused by a limited life time of the optical excitation.

3. EXPERIMENTAL METHOD

Bcc-Fe (001) and (110) ultrathin films and wedges sandwiched by Au layers were grown using the molecular beam epitaxy (MBE) technique. The vacuum of the growth chamber during growth was \( 10^{-10} \) Torr range. Polished MgO (001), (111) and cleaved MgO (001) single crystal were used as substrates. The cleaved MgO substrate was introduced to the MBE chamber just after cleaving in air, while the polished MgO substrate was cleaned by the following procedure before loading in the system: 15 minutes ultrasonic cleaning in acetone, 30-45 seconds etching in 5 vol% HNO₃ ethanol solution, 15 minutes ultrasonic cleaning in acetone, drying with hexane. All substrates were attached to the Mo sample holder by using In solder.

A buffer layer of Ag (~2000Å) and a seedlayer of Au (~2000Å) were deposited and annealed at 450°C preceding to the Fe layer growth. These layers grow epitaxially on the substrate with the same orientation as the MgO single crystal substrate. The Fe wedges were made by moving the sample under a shutter during deposition. The substrate temperature was room temperature during deposition. The deposition rate was measured and controlled to be constant by a crystal thickness monitor. Slopes of the wedges were typically 1 or 0.5 Å/min. All the Fe films were covered with a Au cap layer of 20 Å in order to prevent oxidation.

During all the deposition steps the reflection high energy electron diffraction (RHEED) patterns were observed to be sure about crystal growth orientation. The magneto-optical polar Kerr

![Fig. 7 RHEED patterns of (a) 4.5 Å Fe (001) film grown on the Au (001) surface and (b) 23 Å Fe (110) film grown on the Au (111) surface. The electron beam is parallel to (a) Au [110] and (b) Au [110]. Primary energy is 20keV.](image-url)
Fig. 8 Epitaxial orientation relation of bcc-Fe grown on fcc-Au (001) and (111) surfaces. Large and small circles indicate Au and Fe atoms, respectively. There is no meaning on the relative position of Fe and Au atoms. The picture only indicates relative size of the lattice and orientations.

rotation and ellipticity were measured ex-situ by using Jasco 250 and 2500 spectrometers. These spectrometers are based on the modulation technique using a Faraday cell and piezoelectric modulator [20], respectively. The maximum applied field of our system is 1.9 Tesla. MO spectra, hysteresis curves and so called position scans were measured.

4. RESULTS

Fig. 7 shows RHEED patterns of the Fe layer grown on (a) Au (001) and (b) Au (111) surfaces. The patterns and X-ray diffraction done for thicker films indicate that bcc-Fe (001) atomic planes and (110) atomic plane grow on the fcc-Au (001) and (111) surface, respectively. Epitaxial orientations are (a) bcc-Fe (001) // fcc-Au (001) and bcc-Fe [110] // fcc-Au [110] for the Fe (001) films, and (b) bcc-Fe (110) // Au (111) and bcc-Fe [011] // fcc-Au [110] for Fe (110) films (Nishiyama-Wassermann orientation [21]) (see Fig. 8). The streaky RHEED patterns

![Diagram of epitaxial orientation relation](image)

(a) bcc-Fe(001)/fcc-Au(001)  (b) bcc-Fe(110)/fcc-Au(111)

Fig. 9 Polar ellipticity hysteresis curves of (a) Fe (001) and (b) Fe (110) wedges at several film thickness.

![Hysteresis curves](image)

(a) bcc-Fe (001)  (b) bcc-Fe (110)
indicate the surfaces are rather atomically flat. The pattern of the Fe grown on the Au(111) surface, however, broader, more spottier and shows only low contrast compared with that of Fe (001). This means that Fe (110) films contain a lot of in-plane grain boundaries and certain roughness of the surface. This result is quite normal because Fe (110) / Au (111) interface has large lattice mismatch ($\Delta a=20.8\%$ for Fe [110] direction, and $\Delta a=0.6\%$ for Fe [001] direction) and Fe makes domains on the Au surface corresponding to 3 possible orientations, while Fe (001) / Au (001) interface has very small lattice mismatch ($\Delta a=0.6\%$) and Fe grows as a single crystal.

In Fig. 9, the hysteresis curves observed using polar Kerr ellipticity measurements are listed for each film thickness and orientations. Always Fe (110) films show smaller saturation field, $H_s$. This smaller $H_s$ of Fe (110) will be attributed to the larger surface magnetic anisotropy [22]. In all the cases, there is almost no hysteresis and no remanence.

Fig. 10 shows normalized complex Kerr rotation spectra ($\sqrt{\phi_k^2 + \eta_k^2} / d$) for several film thicknesses, $d$, of the (a) Fe (001) and (b) Fe (110) ultrathin films. A large peak of the spectrum at around 2.5 eV is observed in all the samples and is due to coupled plasma edge of Au. At this energy, optical constants of the Au changes largely and enhances Kerr effect optically [23]. An important point in these spectra is that a new peak appears in 3.5-4.5 eV region for the Fe (001) films thinner than 10 Å and shifts toward lower energy's side with decreasing Fe layer thickness. The appearance of this new magneto-optical transition was well explained by a formation of the QWS's on the Fe $\Delta_1 (p_z)$ band [8]. While in the spectra of the Fe (110) films, there is no such large new peak. Although there appears small peak at 3.9 eV for the Fe (110) thinner than 6 Å, the energy of this peak does not change with change of the thickness.

![Normalized complex polar Kerr spectra of Fe films](image)

Fig. 10 Normalized complex polar Kerr spectra of (a) Fe (001) and (b) Fe (110) ultrathin films of several film thickness. Arrows show new peaks appeared in these films.
The difference of these two orientations are also clear from the position scan of the samples. Fig. 11 shows thickness dependence of the normalized magneto-optical Kerr ellipticity for these two orientations at 4 eV. The Fe (001) wedge sample shows clear oscillation of the ηK as a function of the layer thickness. The oscillation period demonstrated here is about 3.4 Å (2.3ML). But it is not possible to find any clear oscillation for the Fe (110) wedge sample at any photon energy we observed (1.6-5.4eV). The Fe (001) wedge shows MO oscillations for all photon energy ranges (1.6-5.4eV) and the period changes with the photon energy [9].

5. DISCUSSIONS

In a very simple case where a film has only uniaxial anisotropy caused by a surface anisotropy, \( H_s \) times film thickness \( d \) for perpendicular magnetization process is expressed as follows [24]:

\[
\frac{1}{2} M_s H_s = \frac{1}{2} M_s^2 - \frac{2 K_{1}}{d}. \\
\therefore (H_s \cdot d) = M_s \cdot d - \frac{4K_s}{M_s}. \quad (7)
\]

Fig. 12 shows a plot of \((H_s \cdot d)\) versus film thickness for Fe (001) and (110) films. Straight line shows linear fit taking cuts of the y-axis as parameters, whereas slopes are taken as the \( 4\pi M_s \) of the bcc-Fe at 20°C (21.58 kG). Such linear fitting using bulk magnetization works well in the all thickness range for the Fe (001) films but only thick region \((d>15 \text{ Å})\) for the Fe (110) films. The surface anisotropies evaluated from cuts on the y-axis are about 0.5 erg/cm² for the Fe (001) sample and about 0.8 erg/cm² for the Fe (110) sample. Deviation from a linear dependence of the Fe (110) films seems to be caused by the unperfect layer structure of the (110) films.

We performed a band calculation of bcc-Fe using LMTO method and shows cross-sections of iso-energy-difference surfaces at 4 eV in Fig. 13. The graph was made taking some of the combinations of the bands which give most probable transitions between \( d \) and \( sp \) bands. The stationary spanning vectors corresponding to allowed transitions are also shown in the figure.
The vertical axis corresponds to the [001] direction of the bcc-Fe. The oscillation of the MO effect observed experimentally for the Fe (001) wedges ($\Lambda=3.4$ Å) is well explained by the contributions from $q_1$ ($\Lambda=4.1$ Å) and $q_2$ ($\Lambda=3.7$ Å) vectors. The $q_1$ vector corresponds to the optical transition from majority spin $\Delta S$ band ($d_{z^2}$) to $\Delta 1$ band ($p_2$).

Experimentally, no MO oscillation of the Fe (110) wedges has been observed. While the theory proposes some possible oscillation periods caused by $q_4$, $q_5$, and $q_6$ vectors in Fig. 13. The theory predicts, at the same time, that those oscillation amplitudes could be smaller than that of $q_1$ and $q_2$, because the curvature of the iso-energy-difference surface at the initial and final points of the $q_4$, $q_5$, $q_6$ vectors are larger than that of $q_1$ and $q_2$. To clarify a problem of the MO oscillation of different crystal orientations, it is necessary to find a good substrate-overlayer combination and produce perfect layer structure of several orientations.

6. CONCLUSION

A theory of the oscillation of the polar magneto-optical (MO) effect as a function of a ferromagnetic layer thickness was explained in detail. Experiments on the Fe (001) wedges show clear oscillation of the MO response with respect to the Fe film thickness. The oscillation period agrees with the theoretical prediction. While no oscillation was observed for the Fe (110) wedges. Corresponding to these results, the MO spectra of Fe (001) samples show a large new peak which shifts lower energy side for thinner Fe layers. However, in the MO spectra of the Fe (110) films, only a small peak is appear at fixed photon energy. This different behavior was discussed from the film structure and difference of the band structure. As a result, both structural and electronic reasons suggest weaker oscillation for the (110) films. Further experiments using better layer structure is necessary to clarify an influence of the crystal orientation to the quantum size effect of the MO responses.

The theory and experiments showed a possibility to design and control the electronic structure and properties of magnetic materials by making ultrathin films not only for the electronic states near to the Fermi energy but also for the occupied and unoccupied states in wide energy region.

Fig. 13. Iso-energy-difference surface of bcc-Fe for the energy difference of 4 eV. Definition of the "Iso-energy-difference surface" is given by eq. (3) in the text. Here, only some combination of the initial and final band in which MO transitions are allowed are shown.
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