OPTICAL PROPERTIES OF $\text{Fe}_x\text{O}_{1-x}$ OXIDE-METAL COMPOSITE FILMS

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Abstract—Optical and magneto-optical properties of $\text{Fe}_x\text{O}_{1-x}$ ($x>0.5$) oxide-metal composite films were measured in photon energy range from 1.5eV to 4eV by ellipsometry and polar Kerr effect. The observed complex reflectivity ratio $R_p/R_s$ were compared with those calculated from anisotropic effective dielectric constant by a model that fine Fe metallic ellipsoids are precipitated in dielectric matrix ($\text{FeO}$). Volume fractions of metallic phase thus obtained are in good agreement with those from magnetization measurement, but depolarization factor of metallic precipitates does not explain the perpendicular magnetic anisotropy of the films. Kerr rotation angle, $\theta_K$, increases linearly with average magnetization of the composite films up to 20 min for $\sigma=100$ emu/g at photon energy 2eV. The ratio $\theta_K/\sigma$ is twice as large as that for pure Fe.

KEYWORD: $\text{Fe}_x\text{O}_{1-x}$ COMPOSITE FILMS, PERPENDICULAR ANISOTROPY, ROTATION ELLIPSOIDS, EFFECTIVE DIELECTRIC CONSTANT

INTRODUCTION

Binary $\text{Fe}_x\text{O}_{1-x}$ thin films prepared by rf-sputtering become ferromagnetic for $x>0.5$ and show perpendicular magnetic anisotropy.$^{(1,2)}$ They are considered to be a metal-oxide composite which composed of fine ferromagnetic Fe metal precipitate distributed in non-magnetic FeO matrix. The perpendicular magnetic anisotropy may be interpreted by internal shape effect arising from elongated fine ferromagnetic precipitate with their long axes normal to the films plane in FeO matrix. Optical properties of such composite films in which metal fine particles are suspended in dielectric matrix may be treated as continuous media with an effective dielectric constant since the metal particles are much less than the optical wave length.$^{(3)}$ According to this approach, Maxwell Garnett explained the colors of glass containing metallic particles and very thin gold or silver films with island structure.$^{(4,5)}$ David improved the model to get anisotropic effective dielectric constant by treating metallic particles as ellipsoids of revolution with their axes perpendicular to the film plane$^{(6)}$, and Yamaguchi et al. took into account the interaction between the particles.$^{(7)}$ Ellipsometry gives us the ratio of complex reflectivity for p and s polarized light with oblique incidence condition. By comparing the experimental results with appropriate model calculation, we may get not only a volume fraction of metallic phase but also an information about the shape of the metallic precipitates. Kerr effect is originated from off-diagonal component of dielectric tensor. The experimental study of polar Kerr rotation in these composite system would open a new approach to the effective dielectric tensor with off-diagonal components in magnetic composite system.

Fig.1. X-ray diffraction pattern of samples using Fe-Kα radiation
**EXPERIMENTAL PROCEDURES**

Thin film samples were prepared by a conventional rf-sputter system (ANELVA SPF-210A) using a composite targets of Fe$_2$O$_3$ and metallic Fe under the Ar pressure of 2 Pa. The number of small Fe chips (about 25 mm$^2$ area) on a sintered Fe$_2$O$_3$ disk (80 mm$^2$) was adjusted to change average metal contents, $x$, in Fe$_x$O$_{1-x}$ films. Six film specimens were prepared which were named as A to F with increasing metal content, $x$. X-ray diffraction was carried out with Fe-K$_\alpha$ radiation. Magnetization of the film specimens was measured at room temperature by vibrating sample magnetometer in both parallel and perpendicular field to the film plane, from which both the saturation magnetization and uniaxial anisotropy energy were derived.

Optical constant, that is, the complex reflectance ratio for $s$ and $p$ polarized light was measured by a photometric scanning ellipsometry system with rotating analyser. It covers optical wave length from 300 nm to 750 nm. The accuracy of complex reflectance ratio $P$ was about 0.2% in visible region. Polar Kerr rotation angle was measured by polarization modulation technique in applied field up to 9kOe. The accuracy for rotation angle was about 0.002 degree in the visible region.

**RESULTS & DISCUSSION**

X-ray diffraction patterns of the six samples, A to F with increasing Fe content, $x$, are shown in Fig. 1. There appear (111) reflection of FeO phase in all samples and (200) peak in sample A and B. (110) reflection of bcc Fe metal phase becomes clear in sample D, E and F, but it is broad and weak in B and C suggesting that the metallic phase is very fine or might be amorphous state.

It is possible to deduce the volume fraction, $q$, of ferromagnetic Fe phase in non-magnetic FeO from the saturated magnetization as shown in Table 1, assuming the appropriate density, 7.87 and 5.97 for metallic Fe and FeO phase, respectively. If Fe precipitates have very thin needle like shape with their axes normal to the film plane, the perpendicular magnetic anisotropy energy due to internal shape effect may be simply calculated as

$$K = \pi q (1 - q) M_F^2$$

The observed anisotropy energies in our film specimens are plotted versus the magnetization in Fig. 2. They are all positive, but much less than the values expected from Eq. (1) which gives the maximum perpendicular anisotropy due to internal shape effect. This implies that Fe precipitates in our films do not satisfy the ideal condition for Eq. (1).

The effective dielectric constant of a system in which a metal particles of revolutional ellipsoids

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**Table 1** Saturation magnetization $\sigma_s$, composition $x$, and volume fraction $q$ of samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\sigma_s$ (emu/g)</th>
<th>$x$</th>
<th>$q_{(mag)}$ (mag.)</th>
<th>$q_{(opt)}$ (opt.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>10.0</td>
<td>.51</td>
<td>.03</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>25.5</td>
<td>.54</td>
<td>.07</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>40.1</td>
<td>.55</td>
<td>.12</td>
<td>.13</td>
</tr>
<tr>
<td>D</td>
<td>74.3</td>
<td>.60</td>
<td>.23</td>
<td>.22</td>
</tr>
<tr>
<td>E</td>
<td>90.2</td>
<td>.63</td>
<td>.29</td>
<td>.28</td>
</tr>
<tr>
<td>F</td>
<td>99.1</td>
<td>.65</td>
<td>.33</td>
<td>.33</td>
</tr>
</tbody>
</table>

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**Fig-2.** Observed and calculated perpendicular magnetic anisotropy $K$ vs $\sigma_s$ (emu/g).

**Fig-3.** Wave length dependence of complex reflectance ratio ($\rho = \rho_r + i\rho_i$) of samples.
are suspended in dielectric matrix with their axes parallel to z direction normal to the film plane as proposed by Jarrett and Ward as follows.\(^9\)

\[
\begin{align*}
ex &= e_y = e_z = e_0 + q(e_r - e_0) / [1 + f_z(1 - q)(e_r - e_0) / 2e_0] \\
e_z &= e_0 + q(e_r - e_0) / [1 + f_z(1 - q)(e_r - e_0) / e_0]
\end{align*}
\] (2, 3)

where \(f_z\) is the depolarization factor for z direction of metal particles, and \(e_r\) and \(e_0\) represent the dielectric constant of the Fe metal and FeO, respectively. The system is anisotropic except the case where \(f_z = 1/3\), that is, suspended metal particles are spherical.

Fresnel's reflection coefficients with incident angle \(\theta\) for s and p polarized light for optically anisotropic media are expressed as follows.\(^{10}\)

\[
\begin{align*}
Rs &= (Y_0 - Y_1) / (Y_0 + Y_1) \\
Rp &= (Z_0 - Z_1) / (Z_0 + Z_1)
\end{align*}
\] (4, 5)

where,

\[
\begin{align*}
Y_0 &= \cos \theta \\
Y_1 &= (e_x - S^2) / e_x
\end{align*}
\] (6, 7)

The complex reflectance ratio \(R_p/R_s\) can be derived from the experimental results of ellipsometry. The real and imaginary part of it are plotted as a function of the wave length of light in Fig.3. The observed complex reflectance ratio should be compared with those estimated from Eq. (4) and Eq. (5) by inserting Eq. (3) and Eq. (4) with appropriate values for \(q\) and \(f_z\), or we may get \(q\) and \(f_z\) as fitting parameters. Examples are shown in Fig.4, where both calculated and observed \(R_p/R_s\) are plotted in the complex plane. There are good agreement between the volume fraction of metallic phase, \(q\), estimated from magnetization and that from ellipsometry. The depolarizing factor, \(f_z\), which is related to the shape of the metal particles affects the calculated ratio \(R_p/R_s\) as shown in Fig.4. The observed ratios seem to close to those

\[Z_e = \cos \theta\]
\[Z_i = (e_x - S^2 e_x / e_z)^{1/2} / e_x\]
\[S^2 = (\sin \theta / e)^2\] (8, 9, 10)

**Fig-4. Examples of observed and calculated complex reflectance ratio \((\rho = \rho_r + i\rho_i)\) for \(f_z = 0, 1/3, 1.\)**

**Fig-5. Polar Kerr rotation \(\theta_k\) vs photon energy (eV) with applied field \(9\) kOe.**

**Fig-6. Absolute value of polar Kerr rotation \(\theta_k\) vs \(\sigma_g\) (emu/g) at photon energy 2 and 3 eV.**
$f_z=1$ (flat disk shape) rather than those with $f_z=0$ (thin needle like shape with its long axis normal to the film plane). The model adopted here might be too simplified to be successful to explain the perpendicular magnetic anisotropy. The shape of metal precipitates must be complex and their long axes have distribution, which would not necessarily give the same effects to the magnetic anisotropy and to the optical anisotropy. Polar Kerr rotation, $\Theta_k$, of the Fe–FeO composite films are shown in Fig. 5 with in photon energy range from 1.5 to 3.0 eV. The absolute values of the rotation angles of all samples increase with decreasing photon energy same as pure metallic Fe. Oscillation seen in sample A may probably be due to an interference effect with the reflection from the substrate. In Fig. 6 the Kerr rotation angles at photon energy 2 eV and 3 eV are plotted versus the magnetization of the samples. The absolute values of $\Theta_k$ increase linearly with the average magnetization of composite films up to 20 min for $\sigma_a=100$ emu/g at photon energy 2 eV. It should be noted, however, that the ratio, $|\Theta_k|/\sigma_a$ is about twice as large as that for pure Fe.

REFERENCES