MAGNETOOPTICAL SPECTRA IN MULTILAYERS

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1. Introduction

There have been a rapid development in the field of magnetooptical(MO) recording in recent years[1]. MO media suited for short wavelength recording to get higher recording density of next generation are extensively studied. For this purpose, number of studies in the periodic structure of ultra-thin films, such as Pt/Co and Pt/Fe multilayers have been attempted[2-4], because not only they show a large MO effect in short wavelength region around 300 nm but also they have perpendicular anisotropy.

Only a few works have been done on the physical origin of MO effect in these multilayers[5-7]. In order to get further insight into the electronic structures which causes the short-wavelength MO effect, it is necessary to investigate MO spectra for wide range of photon energies.

This chapter is devoted to show the importance of MO spectra for studies of multilayers, as well as to introduce a measuring technique for spectra of magnetooptical rotation and ellipticity.

2. Electromagnetic Theory of Magnetooptical Effect[8]

In a phenomenological treatment, a medium through which electromagnetic waves traverse is treated as a continuum with a dielectric permeability tensor ϵ_{ij}. If the medium is isotropic, only the diagonal elements ϵ_{ii} survive with three diagonal elements being degenerate; i.e., $\epsilon_{xx}=\epsilon_{yy}=\epsilon_{zz}$. When a magnetic field is applied or a magnetization appears, say, in the z-direction, the dielectric tensor becomes to possess non-vanishing off-diagonal elements, $\epsilon_{xy}=-\epsilon_{yx}$ and the degeneracy of diagonal terms is lifted in the z-direction, thus leading to the expression,

$$\hat{\epsilon} = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ -\epsilon_{yx} & \epsilon_{xx} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix}. \qquad (1)$$

The permeability tensor elements ϵ_{ij} can be related to the conductivity tensor elements σ_{ij} by the following expression:

$$\epsilon_{ij} = \delta_{ij} + i\sigma_{ij}/\omega\epsilon_o, \qquad (2)$$

the conductivity tensor being suited for discussing metallic materials in which dielectric constant diverges towards low frequencies.

The propagation of electromagnetic waves in the medium with the dielectric permeability tensor expressed by eq.(1) can be treated by Maxwell equation.

$$\text{curl } E = -\frac{\partial B}{\partial t}$$
$$\text{curl } B = \mu_0 \frac{\partial D}{\partial t} = \epsilon\epsilon_0\mu_0 \frac{\partial E}{\partial t} = \frac{\epsilon}{c^2}\cdot\frac{\partial E}{\partial t} \tag{3}$$

We assume the following exponential time- and position-dependence as expression of the electric and magnetic field vector of the electromagnetic wave propagating along z-direction.

$$E = E_0 \exp(-i\omega t) \exp(i\omega \hat{N} z/c)$$
$$B = B_0 \exp(-i\omega t) \exp(i\omega \hat{N} z/c) \tag{4}$$

Here $\hat{N}$ is the complex refractive index given by $\hat{N} = n + ik$. Applying eq.(4) to eq.(3) and eliminating B, we get the following equation.

$$-\hat{N}^2(E\cdot k)\, k + (\hat{N}^2 I - \hat{\epsilon})\, E = 0, \tag{5}$$

where k stands for the unit vector for z-direction.

In order to obtain a non-trivial solution the following secular equation should hold.

$$\begin{vmatrix} \hat{N}^2 - \epsilon_{xx} & -\epsilon_{xy} & 0 \\ \epsilon_{xy} & \hat{N}^2 - \epsilon_{xx} & 0 \\ 0 & 0 & -\epsilon_{zz} \end{vmatrix} = 0 \tag{6}$$

From this equation we get eigenvalues of $\hat{N}^2$ as follows:

$$\hat{N}_\pm^2 = \epsilon_{xx} \pm i\epsilon_{xy}, \tag{7}$$

the plus and minus signs corresponding to eigenstates with right-circular polarization (RCP) and left-circular polarization (LCP), respectively. Equation (7) shows that the off-diagonal dielectric element is essential to optical activity, since RCP and LCP are degenerate when ϵ_{xy} vanishes.

Subetituting the expressions

$$\hat{N}_+^2 = n_+ + ik_+$$
$$\hat{N}_-^2 = n_- + ik_- \tag{8}$$

into eq.(7), and then putting $\Delta n = n_+ - n_-$ and $\Delta k = k_+ - k_-$, we get the following relations between $\epsilon_{xy} = \epsilon_{xy}' + i\epsilon_{xy}''$ and Δn or Δk.

$$\epsilon_{xy}' = n\Delta k + k\Delta n$$
$$\epsilon_{xy}'' = k\Delta k - n\Delta n \tag{9}$$

Faraday rotation θ_F and Faraday ellipticity η_F can be related to Δn and Δk by next equations:

$$\theta_F = -\Delta n l/2c$$
$$\eta_F = -\Delta k l/2c. \qquad (10)$$

Therefore, we can associate real and imaginary part of off-diagonal element of dielectric permeability tensor with Faraday rotation and ellipticity as follows:

$$\epsilon_{xy}' = -(2c/\omega l)(n\eta_F + k\theta_F)$$
$$\epsilon_{xy}'' = -(2c/\omega l)(k\eta_F - n\theta_F). \qquad (11)$$

Next, we deal with the MO effect of reflection (MO-Kerr effect). Complex amplitude reflectivity $\hat{r}_\pm$ for RCP(+) and LCP(−) can be given by the next formula:

$$\hat{r}_\pm = (1 - N_\pm)/(1 + N_\pm). \qquad (12)$$

Through a simple derivation, we obtain an expression for the complex Kerr rotation $\Phi_K = \theta_K + i\eta_K$ as given by

$$\Phi_K = -(1/2)\ln(\hat{r}_-/\hat{r}_+). \qquad (13)$$

Substituting the $\hat{r}_\pm$ in eq.(13) by eq.(12), we obtain to the first order of ϵ_{xy},

$$\Phi_K = \epsilon_{xy}/(1 - \epsilon_{xx})\sqrt{\epsilon_{xx}}. \qquad (14)$$

This expression tells that MO Kerr effect can become large with the same value of the off-diagonal element, when the denominator in eq.(14) takes a small value. This occurs at the plasma frequency where the real part of ϵ_{xx} vanishes.

From eq.(14) real and imaginary part of ϵ_{xy} are formulated in terms of linear combination of Kerr rotation and ellipticity using n and k as in the following relations:

$$\epsilon_{xy}' = A\,\theta_K - B\,\eta_K$$
$$\epsilon_{xy}'' = B\,\theta_K + A\,\eta_K, \qquad (15)$$

where A and B can be expressed by

$$A = n(1 - n^2 + 3k^2)$$
$$B = k(1 - 3n^2 + k^2). \qquad (16)$$

Thus from the experimentally obtained Kerr rotation and ellipticity values one can calculate the real and imaginary part of off-diagonal element of permeability tensor, provided that optical constants n and k are known.

For the sake of discussing MO materials, it is important to obtain off-diagonal element of dielectric tensor, since ϵ_{xy}, therefore σ_{xy} as well, can be directly correlated with the electronic structure as will be described in the next section.

3. Quantum Theory of Magnetooptical Effect

Off-diagonal element of the optical conductivity σ_{xy} evaluated by quantum mechanics is given by

$$\sigma_{xy} = (e^2/2\hbar m^2) \sum_{\substack{kn \\ (o)}} \sum_{\substack{km \\ (u)}} \{|(\pi_-)_{mn}|^2 - |(\pi_+)_{mn}|^2\}/\{\omega_{mn}^2 - (\omega+i\Gamma)^2\} \qquad (17)$$

where $\pi_i = P_i + i s_i \times \nabla V(r_i)$ is a momentum operator including spin-orbit interaction[10]. In this formula, (o) and (u) stands for the summation on occupied and unoccupied states.

For the purpose of discussing the magnetooptical spectra evaluation of imaginary part σ_{xy}'' (absorptive part) is sufficient, since σ_{xy}' can be calculated by Kramaers-Kronig relation. By taking a limit of $\Gamma \to 0$ in eq.(17), we obtain

$$\sigma_{xy}'' = (\pi e^2/4m^2\hbar) \sum_{\substack{kn \\ (o)}} \sum_{\substack{km \\ (u)}} \{|(\pi_-)_{mn}|^2 - |(\pi_+)_{mn}|^2\} \delta(\omega - \omega_{mn}). \qquad (18)$$

he spectrum of σ_{xy} can be obtained by evaluating the transition matrix element $(\pi_+)_{mn}$ for entire Brillouin zone k-points in the spin-polarized energy band calculation taking into account the spin-orbit interaction. Such calculation performed for Fe[11] and Ni[12] by Callaway's group showed fairly good agreement with experimental spectra as shown in Figs. 1(a) and 1(b). Recent relativistic calculation by Ebert's group provided σ_{xy}'' spectra for PtFe and PdFe alloys[13].

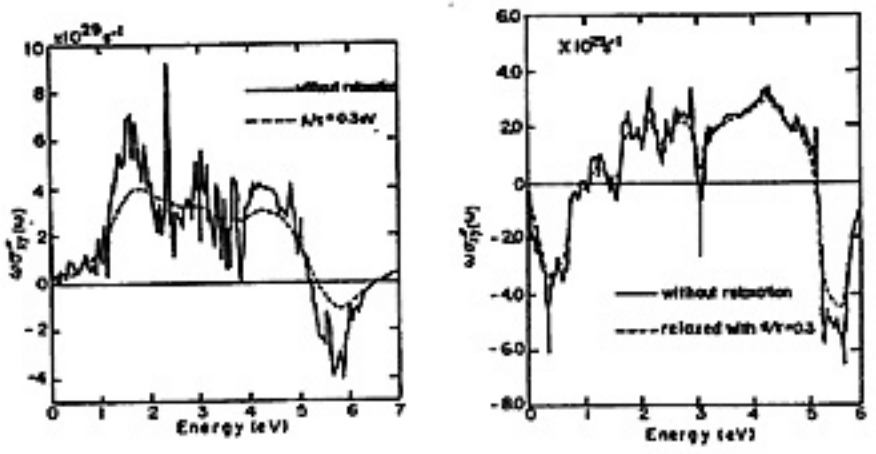
However, in most of the band calculations only the density of state (DOS) curve or the joint density of states at best are available. They lack such calculation of transition matrix in spin-orbit bands. As a convenient way of evaluation of σ_{xy},

$$\sigma_{xy}'' = (\pi e^2/4m^2\hbar)\{F_{nm}^{-} J_{nm}^{-} - F_{nm}^{+} J_{nm}^{+}\} \qquad (19)$$

where $F_{nm}^{\pm}$ is averaged transition matrix for RCP(+) and LCP(-) and $J_{nm}^{\pm}$ joint density of states as given by

$$F_{nm}^{\pm} J_{nm}^{\pm} = (1/8\pi^3) \int \{|\langle m|\pi_\pm|n\rangle|^2 + |\langle m|\pi_\pm|n\rangle|^2\} \delta(\omega - \omega_{nm}) d^3k. \qquad (20)$$

Subtraction between contributions of RCP and LCP in eq. (19) can be approximated by energy-derivatives of joint density of states, which provided a satisfactory explanation of MO spectra in Cr_3Te_4[14] and PtMnSb[15].



(a) (b)

Fig. 1 σ_{xy}'' of Fe (a) and Ni (b) evaluated by band calculation

4. Magnetooptical Measurements

Several kinds of measuring techniques have been known for evaluation of magnetooptical spectra: They are (1) Vibrating-polarizer technique, (2) Rotating-polarizer technique, (3) Faraday-rotator technique and (4) Retardation modulation technique. Since techniques (1)-(3) utilize modulation of the linear polarization, they are not suited for evaluation of ellipticity spectra. On the other hand, the technique (4) uses a modulation of the retardation, which enables spectroscopic measurement of both rotation and ellipticity. In the rest of this section, the fourth technique (often referred to as Sato technique) will be described.

4.1. Principles of the Retardation Modulation Technique

In this subsection, we briefly summarize the principles of technique described by the author in ref. 16.

A schematical illustration of the apparatus is shown in Fig. 2. The light from a monochromator is chopped by a rotating sector, polarized linearly by a polarizer with the transmission angle making 45° to the vertical axis and modulated by a photoelastic modulator (Hinds Inc. PEM-CF3). The modulator causes a retardation expressed by the following formula,

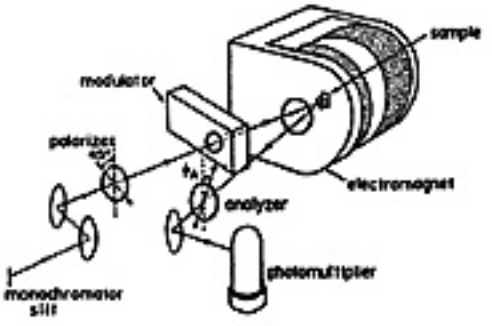


Fig. 2 A schematical illustration of retardation-modulation technique

$$\delta=\delta_0 \sin 2\pi pt, \qquad (21)$$

between vertical and horizontal polarizations, where δ_0 denotes the amplitude of the retardation and p the modulation frequency. The modulated light is reflected by a magnetized sample placed at the focal point of the mirror system and is passed through an analyzer with the transmission axis making an angle of ϕ_A to the horizontal plane and is detected by a photomultiplier.

As derived in ref. 16, the intensity I of the light detected by the photomultiplier is represented by