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MAGNETO-OPTICAL SPECTRUM AND ELECTRONIC STRUCTURES OF CoS_2

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Magneto-optical and infrared reflectivity spectra of a pyrite-type ferromagnetic compound CoS_2 have been measured. A strong magneto-optical effect was observed at 0.8 eV below T_c (~ 120 K). From theoretical analyses of these spectra it turns out that both $d\epsilon$ and $d\gamma$ bands are narrow although CoS_2 shows a metallic conductivity.

The pyrite-type compound CoS_2 has been known as a ferromagnet ($T_c \sim 120$ K) with a metallic conductivity [1]. It has been considered that in this compound $3d\epsilon$ states are filled with six electrons and $3d\gamma$ states are partly filled contributing to the metallic conduction as well as to the ferromagnetism. In order to get detailed information of the 3d bands we have performed measurements and analyses of magneto-optical spectra and infrared (IR) reflectivity spectra of this compound.

Single crystals of CoS_2 have been obtained by the chemical transport technique with chlorine as a transporting agent. Reflectance magneto-circular dichroism (RMCD) were measured at temperatures below T_c under applied magnetic field of 4 kOe, by using the polarization modulation technique [2]. Near-normal-incidence IR reflectivity was measured at the University of Tsukuba for photon energies between 0.1 and 1 eV at room temperature.

In fig. 1 is illustrated a spectrum of the measured RMCD ($\Delta R/R$) by a solid curve. In the same figure the Kerr rotation ϕ_K calculated from $\Delta R/R$ by using a dispersion relation derived by D.Y. Smith [3] is also plotted by a dotted curve. Three main structures have been observed, of which the most prominent is the one at 0.8 eV, where the Kerr rotation reached a value of

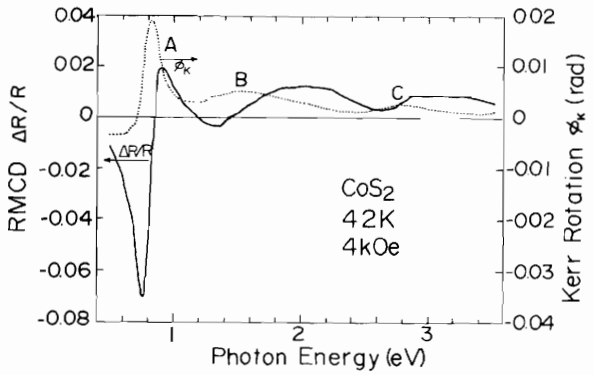

Fig. 1. Solid curve: RMCD ($\Delta R/R$) spectrum of single crystal of CoS_2 measured at 4.2 K under the magnetic field of 4 kOe; dotted curve: Kerr rotation ϕ_K spectrum calculated from the RMCD spectrum.

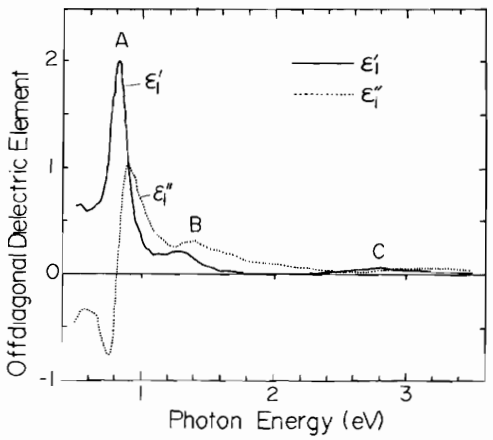

Fig. 2. Spectra of ϵ_1' and ϵ_1'' calculated from the experimental data of $\Delta R/R$ and ϕ_K. Solid curve: ϵ_1'; dotted curve: ϵ_1''.

0.019 rad (~ 1.1°). Fig. 2 shows real and imaginary parts of the off-diagonal element of the dielectric tensor ϵ calculated from $\Delta R/R$ and ϕ_K, where ϵ is defined as

$$\epsilon = \begin{bmatrix} \epsilon_0 & -i\epsilon_1 & 0 \\ i\epsilon_1 & \epsilon_0 & 0 \\ 0 & 0 & \epsilon_z \end{bmatrix}.$$

A prominent diamagnetic line shape is observed at 0.8 eV, as well as weaker structures around 1.5 and 2.8 eV.

The energy position of the most prominent structure has been related to the energy at which the transition from $d\epsilon$ band to $d\gamma$ band starts [4]. We calculated the magneto-optical line shape for this transition assuming that it is expressed in terms of the localized transition between multiplets; $^2E(d\epsilon^6 d\gamma) \rightarrow {}^2T_1(d\epsilon^5 d\gamma^2)$. Taking the spin–orbit interaction into account we get an energy level diagram as shown in fig. 3a. Calculation of the off-diagonal element of the dielectric tensor was performed for this energy scheme. The details of the calculation have been published elsewhere [5]. The obtained spectra are illustrated in figs. 3b and 3c together with the experimental ones. It is found that the best fit is obtained with a reasonable set of parameters: the

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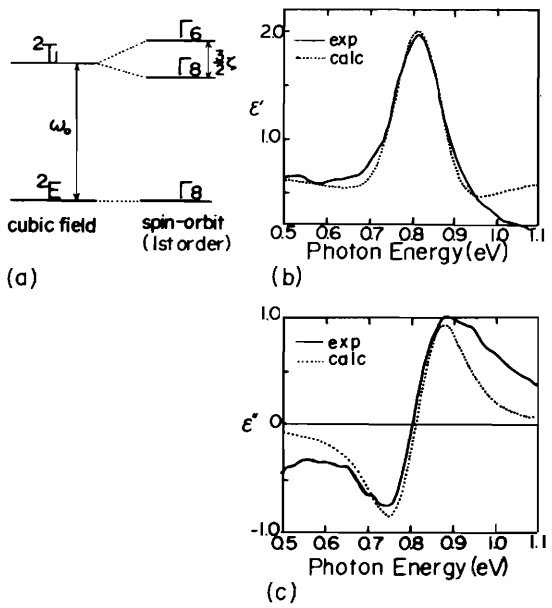

Fig. 3. (a) Energy level diagram of related transition. (b) Calculated magneto-optical line shape of ϵ_1' compared with experimental one. (c) Calculated magneto-optical line shape of ϵ_1'' compared with experimental one.

spin–orbit parameter $\zeta = 533\ \mathrm{cm}^{-1}$ (free Co^{2+} ion value) and the line width $\Gamma = 1000\ \mathrm{cm}^{-1}$. The fact that the magneto-optical line shape is successfully described by a transition between multiplets shows that the exciton effect is essential in the related transition, which reflects the localized nature of the $d\epsilon$ holes.

In the next place, the solid curve of fig. 4 shows a reflectivity spectrum of CoS_2 for 0.1–2.5 eV. In this spectrum we can clearly see a rapid decrease of the reflectivity with the increase of photon energy in the low energy region, which is characteristic of the plasma resonance of the conduction electrons. The spectrum was analyzed taking both the Drude term and the interband terms into consideration. The interband terms were approximated by Lorentzian oscillators having center energies at 1.16, 1.77 and 3.14 eV. As shown by the dotted curve in fig. 4 the best fit was obtained with an effective mass $m^* = 2.66\ m_0$, and a lifetime $\tau = 1.7 \times 10^{-15}$ s, assuming that one $d\gamma$ electron is supplied

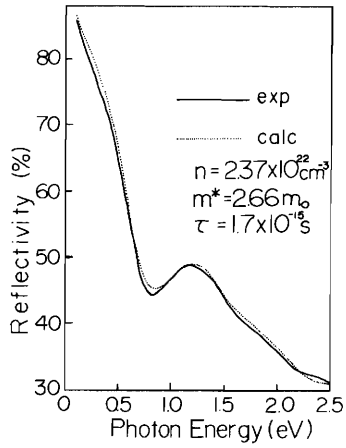

Fig. 4. Experimental reflectivity spectrum and calculated spectrum assuming Drude term and interband term.

from every one Co atom. This means that the $d\gamma$ conduction electrons are so heavy that they can hardly be treated as 'free' electrons.

Approximation of interband terms by Lorentzian oscillators may be justified by the localized nature of related transitions.

It is concluded from the above analyses that both the $d\epsilon$ band and $d\gamma$ band are not so broad but rather localized, even though the crystal shows a metallic conductivity. This may be related to the strong correlation of the d electrons in the pyrite type compound.

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