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VUV-reflectivity, magneto-optical spectra and band structure in single crystals of Cr_3Te_4

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The diagonal and off-diagonal elements of the optical conductivity tensor in Cr_3Te_4 were obtained from the reflectivity spectra measured with synchrotron radiation and the magneto-optical Kerr spectra. These values were compared with the joint density of states evaluated from the energy band structure calculated by Dijkstra. Fairly good agreement was obtained between experiment and calculation.

Chromium tellurides have crystal structures derived from the nickel arsenide structure showing several types of superstructures with the ordering of the cation vacancies and are known to show diverse magnetic and electrical properties [1]. Among these compounds Cr_3Te_4 is ferromagnetic with a Curie temperature of 325 K. It also shows an order-to-order magnetic transition around 80 K, below which it shows canted antiferromagnetic properties. The electrical properties in this compound are metallic.

Single crystals of Cr_3Te_4 were prepared by means of the chemical vapor transport technique, as described in our previous paper [2]. The reflectivity spectrum of the crystal was measured for photon energies between 0.3 and 23 eV. The reflectivity below 4 eV was measured using a conventional spectroscopic system with a specially designed reflectivity attachment, as used for the studies in pyrite type compounds [3]. For energies higher than 4 eV we employed VUV light from the storage ring at the Synchrotron Radiation Laboratory of ISSP at the University of Tokyo. Both spectra were connected and calibrated using the optical constants evaluated by ellipsometry. The experimental details are described elsewhere [4].

Fig. 1 illustrates the reflectivity spectra between 0.3 and 23 eV in the Cr_3Te_4 crystal at room temperature. The real and imaginary parts of the conductivity spectra were evaluated by Kramers–Kronig analysis. For this purpose, the reflectivity for below 0.3 eV was extrapolated using the Drude formula with the parameters determined so that the real part of the conductivity at the zero-frequency limit is consistent with the dc conductivity. For energies above 23 eV, the reflectivity spectrum was extrapolated using the E^{-4}-formula. The real and the imaginary part of the diagonal element of the optical conductivity thus obtained are plotted in fig. 2 by solid and dotted lines, respectively. The optical constants n and k were also determined for photon energies between 0.3 and 23 eV.

The polar magneto-optical Kerr rotation and Kerr ellipticity spectra in the Cr_3Te_4 crystal were measured by means of the polarization modulation technique employing a piezobirefringent modulator [5]. From these spectra the real and imaginary part of the off-diagonal element of the conductivity tensor were evaluated using the optical constants obtained above. In fig. 3 are illustrated the real and imaginary part of the off-diagonal conductivity in Cr_3Te_4.

The absorptive part of the diagonal conductivity, σ'_{xx}, below 5 eV is given in fig. 4a together with the JDOS (joint density-of-states)/photon energy spectra evaluated from the DOS curve calculated by Dijkstra [6]. A striking similarity is observed in the overall features of the two spectra. The structures a, b and c marked in the experimental spectrum seem to correspond to a′, b′ and c′ in the calculated spectrum. The structures a′ and b′ correspond to transitions from the occupied high-DOS states just below the Fermi level derived from the chromium 3d orbitals to the empty states above the Fermi level derived from tellurium 5p orbitals in the majority spin band, while the structure c′ is due to the transition from the occupied 5p states of tellurium to the empty 3d states above the Fermi

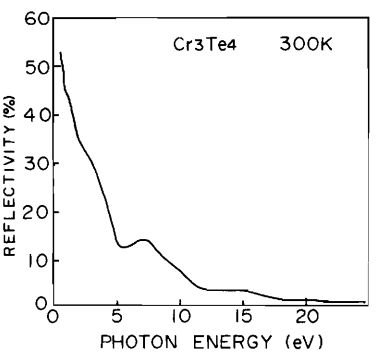

Fig. 1. Normal incidence reflectivity of a single crystal of Cr_3Te_4 measured at room temperature.

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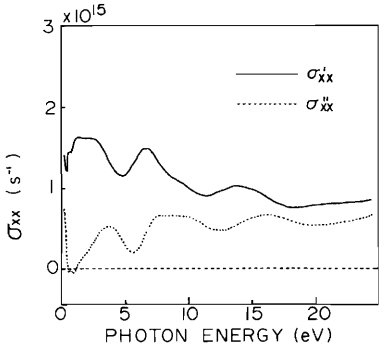

Fig. 2. Real and imaginary part of diagonal element of the conductivity tensor in Cr_3Te_4, deduced from the reflectivity.

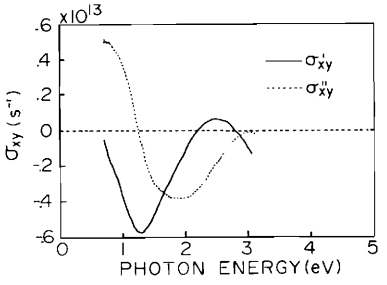

Fig. 3. Spectra of the real and imaginary part of the off-diagonal conductivity in Cr_3Te_4.

level in the minority spin band, which are the exchange-split counterpart of the majority-spin 3d states.

The absorptive part of the off-diagonal element of conductivity, σ''_{xy} is given in fig. 4b together with the energy derivative of the JDOS curve [7], which is considered to correspond to the difference in the absorption spectra for two opposite circular polarizations. A similarity in the overall lineshape between experiment and calculation is found, although some difference in the energy position of the structures is seen.

In conclusion, the diagonal and the off-diagonal elements of the conductivity tensor in Cr_3Te_4 deduced from our optical experiments were found to be qualitatively explained by the band structure. This means that the d-electrons of the chromium atoms in this material are not localized and that the magnetic behavior in this compound should be treated in terms of itinerant electron magnetism.

The authors are indebted to Dr. M. Fujisawa of the Synchrotron Laboratory of the ISSP, the University of Tokyo for the VUV reflectivity measurements. They

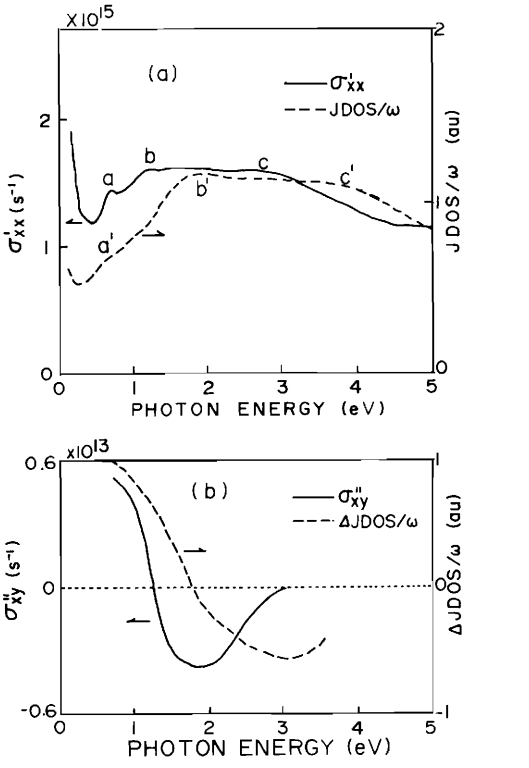

Fig. 4. (a) Absorptive part of the diagonal conductivity σ'_{xx} (solid curve) and the calculated JDOS divided by the photon energy (dashed curve). (b) Absorptive part of the off-diagonal conductivity σ''_{xy} (solid curve) and the calculated energy-derivative of the JDOS divided by the photon energy (dashed curve).

are also grateful to Mr. T. Fukazawa of JASCO Inc. for spectroscopic ellipsometry.

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