

Influence of Magnetic Ordering on the Faraday Effect in CdCr_2Se_4

Abstract: Measurements of the magneto-optical rotation in CdCr_2Se_4 at wavelengths between 0.9 and 1.3 μm are reported. In the paramagnetic state, the rotation passes a minimum with decreasing wavelength and changes sign at the absorption edge. The Verdet constant is proportional to the paramagnetic susceptibility ($V = -18 \text{ deg/kOe-cm}$ at 300°K and $\lambda = 1.05 \mu\text{m}$). In the ferromagnetic state the Faraday effect is large and the sign reversal very pronounced; e.g., the rotation changes by $6.5 \times 10^4 \text{ deg/cm}$ in a wavelength interval of 400 Å in saturated material at 93°K. The photon energy at which the sign reversal occurs shows a strong temperature-dependent red shift; the temperature dependence is similar to that of the absorption edge. At low temperatures an additional rotational minimum appears that is subject to the same red shift.

It is the purpose of this paper to report on measurements of the Faraday rotation in the semiconducting spinel CdCr_2Se_4 at wavelengths near the optical absorption edge and at temperatures above and below the ferromagnetic ordering temperature ($T_c \approx 130^\circ\text{K}$). From earlier magneto-optical work¹⁻⁴ large, strongly temperature- and wavelength-dependent rotations were expected.

Our crystals were grown by slow cooling of a solution of CdCr_2Se_4 in a PbCl_2 - CdCl_2 flux. They were characterized by chemical and x-ray analysis and by thermoelectric power and magnetization measurements as follows: The Pb content was less than 0.005 weight-percent. The lattice constant was $(10.746 \pm 0.001) \text{ Å}$.^{2,5,6} The conduction was p type at about 200°C. The Curie temperature was 130°K. Platelets (0.025 ± 0.005) mm thick were ground parallel to the (111) growth faces of crystals with an edge length of 3 mm. They were glued with Canada balsam on glass substrates. The rotational spectra were recorded with the samples in a magnetic field up to 9.3 kOe, parallel to the light beam, and at temperatures between 41°K and 300°K. The absolute temperature values are accurate to only about ± 5 degrees; however, the temperature could be kept constant within ± 0.1 degree. Figure 1 gives the Faraday rotation as a function of wavelength for various temperatures. All these curves were measured with the same magnetic field, 4.6 kOe, which was sufficiently strong to saturate the samples at the lowest temperature. At high temperatures, in the paramagnetic

state, the rotation passes through a broad minimum with decreasing wavelength, then increases rapidly and changes sign close to the absorption edge. With decreasing temperature the rotations increase in magnitude due to the rising magnetization, with a particularly rapid increase below T_c . At liquid-nitrogen temperature the Faraday effect amounts to several times 10^4 deg/cm . Near the wavelength at which the rotation reverses its sense, the value changes by about $6.5 \times 10^4 \text{ deg/cm}$ in a wavelength interval of 400 Å. Those parts of the curves where the rotation increases abruptly are subject to a large temperature-dependent red shift, with the temperature dependence similar to that of the absorption edge.^{1,2} An additional shift can be induced at temperatures near T_c by increasing the magnetic field.² At 130°K this shift amounts to 15 deg/kOe-cm. Below about 140°K a second, more pronounced rotational minimum appears that is shifted together with the steep edge of the curve and increases rapidly in magnitude with decreasing temperature. In the paramagnetic range at wavelengths where the Faraday effect shows little dispersion, the rotation was found to be proportional to the applied field. At high temperatures, between 180°K and 300°K, where the susceptibility is sufficiently field-independent, the Verdet constant measured at the wavelength 1.05 μm is proportional to the molar susceptibility χ as measured by Baltzer et al.⁵:

$$V = \chi \times 540 \text{ deg/kOe-cm.}$$

At 300°K the Verdet constant is -18 deg/kOe-cm . From this proportionality we conclude that up to room tem-

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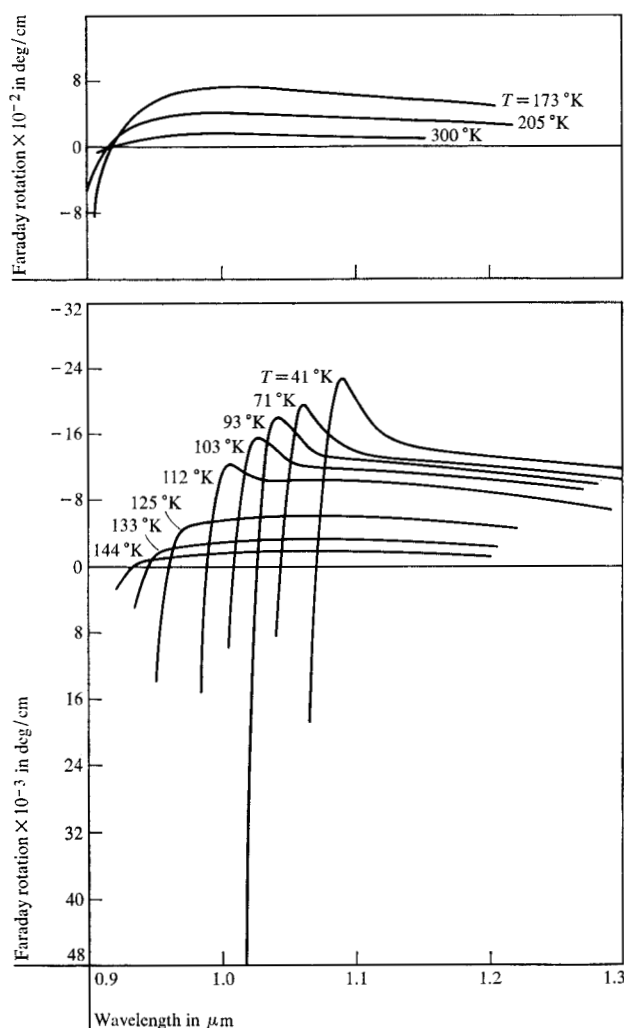
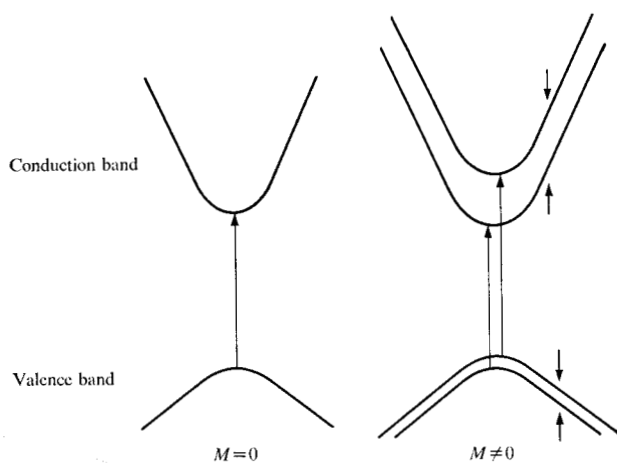


Figure 1 Faraday rotation in CdCr_2Se_4 as a function of wavelength for various temperatures with an applied magnetic field of 4.6 kOe. Note that the scale factors in the upper and lower parts of the figure are different.

Figure 2 Spin splitting of energy bands and spin-allowed transitions.



perature the observed Faraday effect is related to the magnetic moments of the Cr^{3+} ions and depends on the magnetic field only through its influence on these ionic moments. The direct interaction between electrons and magnetic field, which in nonmagnetic semiconductors is the origin of magneto-optical effects, does not contribute measurably in CdCr_2Se_4 in the temperature range studied.

From the facts that the sharp sign reversal and the extra peak in the rotational spectra occur close to the absorption edge and show roughly the same temperature-dependent red shift as the absorption edge transitions, it is evident that the observed rotational structure is due to these transitions. The absorption edge has previously been considered⁷ to be formed either by charge transfer from Se to Cr or by transitions between valence and conduction bands that are split into pairs of spin-polarized sub-bands by exchange interactions with the 3d (Cr^{3+}) electrons,^{4,8} whereas the lowest transitions from the Cr^{3+} ground state are expected at higher energies.^{4,7} In view of its complexity the charge-transfer model is not discussed here. However, we can show that the simple split-band model cannot account for the observed shape of the rotational spectra without additional assumptions. We consider two spin-split pairs of parabolic bands^{4,8} as indicated in Fig. 2. In addition, we assume a weak spin-orbit coupling.⁹ The rotational dispersion due to the two spin-allowed transitions (Fig. 2), assumed to be of equal intensity, is calculated⁹ as if the effective masses, the transition probability, the band splittings (which are proportional to the magnetization) and the damping were adjustable parameters.

In order to explain our spectra qualitatively it was necessary to superimpose two dispersion curves of the kind discussed above, both centered at roughly the same frequency at high temperatures. The damping and splitting parameters for the two curves are very different. This means that more electronic states than just those sketched in Fig. 2 are involved in the band-edge transitions.

The damping parameter is of particular interest because, through its temperature dependence, it will indicate whether the excited states are localized or band states. Since in the latter case the damping is due mainly to spin-disorder scattering, it should be drastically reduced below T_c . From the order of magnitude of the relaxation rates calculated by Haas⁸ one might speculate that the appearance of the sharp rotational peak below T_c is related to the temperature dependence of spin-disorder scattering. However, in order to establish a clear correlation between the optical and magnetic effects, magnetization measurements are necessary on samples from the same batch and under the same conditions as in the optical experiments. These measurements are now in progress.

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