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Optical Reflectance Study of Magnetic Ordering Effects in EuO, EuS, EuSe and EuTe

Abstract: The reflectivity of the ferromagnetic semiconductors EuO, EuS and EuSe has been measured as a function of temperature and light polarization in an orienting magnetic field. In the energy range from just above the absorption edges to about 5.0 eV, there are two prominent features, E_1 and E_2 , which change with the magnetic order, indicating exchange splittings of the 5d states. Antiferromagnetic EuTe shows a different structure in the E_1 and E_2 peaks. However, with a large magnetic field applied (>40 kOe) the spectrum becomes characteristic of the other ferromagnetic europium chalcogenides suggesting a phase transition to parallel spin alignment at $H \approx 80$ kOe. The effect provides direct experimental confirmation of the band theory prediction that there are superlattice splittings in the band structure of an antiferromagnetic crystal.

We have been investigating the electronic structure of the magnetic semiconductors EuO,¹ EuS,^{2,3} and EuTe,⁴ by means of optical reflectance studies. The experiments were performed on single crystals grown from the melt.¹ Near normal incidence reflectivity was measured from cleaved (100) faces, oriented with the magnetic field normal to the surface. Reproducible results were obtained from a large number of different crystals, indicating that surface strain effects were not important. As Fig. 1 shows, the reflectance spectrum is rich in structure and, in particular, we have measured the influence of magnetic ordering on the first two peaks, E_1 and E_2 . In a domain-orienting magnetic field, the ferromagnetic materials EuO, EuS and EuSe exhibit polarization-dependent splittings in E_1 and E_2 at temperatures near and below the Curie point, T_c . Near T_c , the E_1 peak splits first into right and left circularly polarized components (σ_R and σ_L) and then, at temperatures well below T_c , each component shifts and splits further with the peaks forming an overlapping quartet. The behavior of the E_2 peak is more complex in that it is already split in zero field at temperatures well above T_c (e.g., c.f. Fig. 1). In addition, however, E_2 also shows a polarization dependence in a magnetic field at temperatures near and below T_c .

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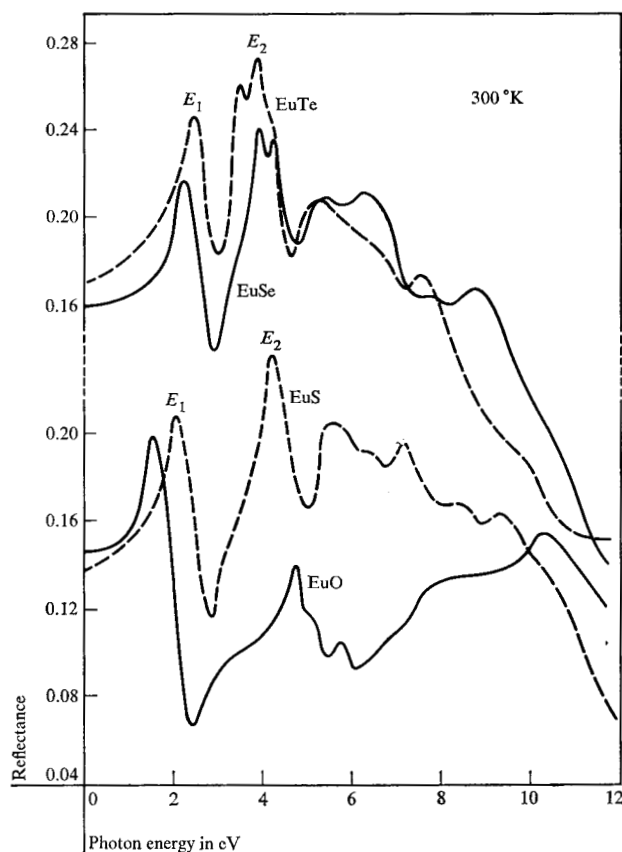


Figure 1 Reflectance spectra of EuO, EuS, EuSe and EuTe at 300°K in zero magnetic field.

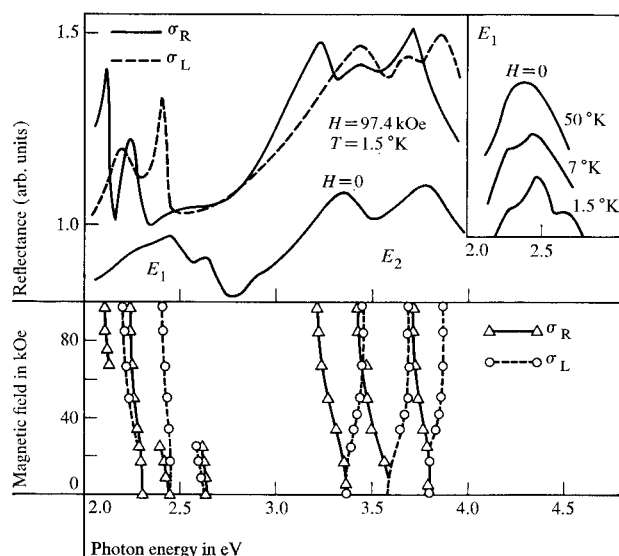


Figure 2 Polarized E_1 and E_2 reflectance peaks of EuTe for $H = 97.4$ kOe and $H = 0$ at 1.5°K . The $H = 0$ curve has been shifted for clarity. Also shown are the positions of the peaks in the E_1 and E_2 multiplets as a function of magnetic field. The inset shows the temperature dependence of the E_1 peak at zero magnetic field.

Our results suggest that the E_1 and E_2 peaks arise from transitions corresponding to the transfer of localized europium 4f electrons into $5d(t_{2g})$ and $5d(e_g)$ states respectively (c.f. the accompanying paper by J. Dimmock).^{1,3,5} As the sample becomes magnetized the lowest, $M_s = -7/2$, $4f^7(^8S_{7/2})$ state becomes preferentially populated. A calculation of the transition probabilities to the manifold of spin-orbit split 7F_J states shows a resolution into right and left circularly polarized components under these circumstances.^{3,5} Below the magnetic ordering temperature the E_1 doublet structure is further separated to a triplet by the exchange splitting of the 5d states. The fact that this exchange splitting is different above and below T_c suggests that the 5d states are not localized on a single ion, since, if they were, the splitting would be independent of sample magnetization. The behavior of the E_2 peak is obviously more complex than that of E_1 . This should not be surprising since, in addition to the europium f to d transition, other transitions may also be involved (for example p to d or p to s band transitions across the fundamental energy gap). Also, the existence of a doublet at all temperatures suggests that there may be transitions associated with different critical points in the Brillouin zone.

Finally, we have observed the effects of spin alignment induced by a large external magnetic field on the reflectance spectra of antiferromagnetic EuTe at 1.5°K .⁴ Representative spectra for the E_1 and E_2 peaks, showing the zero-field and high-field spectra, are illustrated in Fig. 2.

A striking change in the E_1 peak is observed as the field is increased. A polarization independent broad triplet is observed at very low fields, but changes to a sharp polarization dependent triplet structure when the field exceeds about 60 kOe, with the peaks becoming more prominent, and the strengths saturating at about 80 kOe. Since this polarized triplet structure has been associated with ferromagnetic ordering, the saturation effect shows that field-induced spin alignment has been obtained. The E_2 structure also shows an anomalous change with field, with each peak splitting into σ_R and σ_L components. This effect again saturates at $H \approx 80$ kOe.

A further experiment has been carried out on the temperature dependence of the E_1 peak, shown inset in Fig. 2. The triplet at 1.5°K transforms to a doublet near the Néel temperature T_N , then gradually broadens, completely disappearing at room temperature. The temperature dependence above T_N is similar to that of the other chalcogenides, and, in particular, since the doublet vanishes at high temperatures, it appears that this structure is caused by antiferromagnetic ordering.

The lack of polarization dependence of the E_1 and E_2 structures at very low fields is to be expected for the antiferromagnetic state. At temperatures $\approx T_N$ there is a preferential population of the lowest spin-down state for one sublattice, and of the opposite spin state for the other sublattice. Thus, by contrast with the ferromagnetic case, we do not expect to see a polarization dependence of the spectra. For the antiferromagnetic state of the crystal there is a superlattice splitting of the bands at the new zone boundary, but zero splitting at $k = 0$ for the simple case.⁴ We associate the separation of the doublet into a triplet well below T_N with the superlattice splitting at the zone boundary. Since each d band has both up and down spin parts there will again be no polarization effect introduced by this. However, in addition to the basic antiferromagnetic superlattice-band splitting, in an applied magnetic field there is superimposed a net "ferromagnetic" interaction. This comes about above the spin flop field (≈ 4 kOe for EuTe^{6,7}) when the sublattice magnetizations are perpendicular to the field direction, and then slowly cant up toward the field direction as the field is increased. A simple molecular-field calculation using the exchange parameters given in Ref. 6 leads to an estimate $H_s \approx 70$ kOe for complete spin alignment, in good agreement with the experiment, and with earlier magnetization measurements.⁸

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References

1. J. Feinleib, W. J. Scouler, J. O. Dimmock, J. Hanus, T. B. Reed and C. R. Pidgeon, *Phys. Rev. Letters* **22**, 1385 (1969). A representative bibliography is given here.

2. C. R. Pidgeon, J. Feinleib, W. J. Scouler, J. Hanus, J. O. Dimmock and T. B. Reed, *Solid State Commun.* **7**, 1323 (1969).
3. W. J. Scouler, J. Feinleib, J. O. Dimmock and C. R. Pidgeon, *Solid State Commun.* (to be published).
4. J. Feinleib and C. R. Pidgeon, *Phys. Rev. Letters* **23**, 1391 (1969).
5. J. Hanus, J. O. Dimmock and J. Feinleib, 15 Ann. Conf. on Magn. and Mag. Materials (to be published); J. O. Dimmock, *IBM J. Res. Develop.* **14**, 301 (1970, this issue).
6. T. R. McGuire and M. W. Shafer, *J. Appl. Phys.* **35**, 984 (1964).
7. G. Busch, *J. Appl. Phys.* **38**, 1386 (1967)
8. G. Busch, P. Junod, P. Schwob, O. Vogt and F. Hüliger, *Phys. Letters* **9**, 7 (1964); I. S. Jacobs and S. D. Silverstein, *Phys. Rev. Letters* **13**, 272 (1964).

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