

"classical" critical indices  $\eta=0$  and  $\nu=\frac{1}{2}$ .

<sup>7</sup>Equation (4) is rigorous for  $d=2$ , and confirmed numerically, but unproven, for  $d=3$ . See Fisher and Burford, Ref. 3; and M. E. Fisher, in *Critical Phenomena*, edited by M. S. Green and J. V. Sengers (National Bureau of Standards, Washington, D. C., 1966).

<sup>8</sup>M. F. Sykes, J. L. Martin, and D. L. Hunter, Proc. Phys. Soc. (London) **91**, 671 (1967).

<sup>9</sup>D. R. Hartree, *Numerical Analysis* (Oxford University Press, London, England, 1952).

<sup>10</sup>For more distant sites the number of nonzero  $q_n(\vec{r})$ ,  $n \leq 12$ , is insufficient for worthwhile extrapolation.

<sup>11</sup>Scaling depends only on the variation with temperature of  $\kappa(T) = \kappa_0 \epsilon^\nu$ . In drawing Fig. 1 we have used  $\kappa_0 a = 2.30 \pm 0.03$ , which consistently fits the exponential part of the large- $r$  behavior, i.e., correctly gives the slope of  $\ln[(r/a)\epsilon^{-\nu}] \Gamma(\vec{r}, T)$  vs  $-\epsilon^\nu r/a$  [see Eq. (2)].

<sup>12</sup>Fisher, Ref. 7.

<sup>13</sup>V. G. Vaks, A. I. Larkin, and Yu. N. Ovchinnikov, Zh. Eksperim. i Teor. Fiz. **49**, 1180 (1965) [translation: Soviet Phys.-JETP **22**, 820 (1966)].

<sup>14</sup>Static scattering experiments measure  $\chi(\vec{k}, T)$ , the Fourier transform of  $\Gamma(\vec{r}, T)$ . Strong scaling predicts  $\chi(\vec{k}, T) = F(k/\kappa) k^{2-\eta}$  for small  $k$  and  $\kappa$  regardless of the ratio  $k/\kappa$ . Reference 3 gives scattering approximations of this type. Our prediction is that there are small nonscaling departures from this behavior, notably at fixed  $k$  as  $\epsilon \rightarrow 0$ . It seems likely that recent scattering experiments on lattice systems do not have sufficient resolution to detect such effects. See L. Guttman and H. C. Schnyders, Phys. Rev. Letters **22**, 520 (1969); also, J. Als-Nielsen and O. W. Dietrich, Phys. Rev. **153**, 706, 711, 717 (1967).

<sup>15</sup>M. E. Fisher, Rept. Progr. Phys. **30**, 615 (1967).

## SPIN-POLARIZED SPLITTINGS IN THE TEMPERATURE-DEPENDENT REFLECTANCE OF EuO†

J. Feinleib, W. J. Scouler, J. O. Dimmock, J. Hanus, and T. B. Reed

Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts 02173

and

C. R. Pidgeon

Francis Bitter National Magnet Laboratory, \* Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 5 May 1969)

The reflectance of EuO from above the Curie point at 69°K down to 1.5°K shows splittings in the two main peaks near 1.5 and 4.7 eV. The first peak is a polarization-dependent triplet at 1.5°K and a doublet at 70°K. The data suggest that the 1.5-eV peak is associated with the absorption edge and arises from the transition  $4f^7(^8S_{7/2}) \rightarrow 4f^6(^6F_J)5d$  ( $t_{2g}$ ). The narrow  $t_{2g}$  sub-band is exchange split by 0.25 eV at low temperatures.

Previous optical studies of the magnetic semiconductors, EuO and other europium chalcogenides, have shown unusually large magneto-optical effects<sup>1</sup> and an anomalous red shift of the absorption edge upon cooling through the Curie point.<sup>2</sup> These effects have been ascribed to transitions from the localized europium  $4f$  electron states to  $5d$  energy states.<sup>3</sup> We have further examined the effects of magnetic ordering on the band structure by optical reflection studies of EuO beyond the absorption edge in two sets of experiments: (1) measurement of the reflectance from 0.6 to 5.2 eV at several temperatures through the Curie point,  $T_C = 69^\circ\text{K}$ , down to  $1.5^\circ\text{K}$  and (2) examination of the first peak in the reflectance with circularly polarized light and in a magnetic field at 70 and  $1.5^\circ\text{K}$ . We believe the splittings of the reflectivity peak observed in these measurements indicate that the transitions are to conduction-band states which spin split at low temperatures. To our knowledge, this is the first time such structure has been seen by direct

reflectance measurements through the Curie point in any ferromagnetic material.

In the first set of experiments no polarized light or magnetic field was used. The results of these measurements appear in Fig. 1.  $E_1$  centered at 1.44 eV at  $80^\circ\text{K}$  is the lowest energy structure near the absorption edge at 1.2 eV.<sup>2,4-6</sup>  $E_2$  is centered at 4.65 eV at  $80^\circ\text{K}$ .

As the temperature is lowered through  $T_C$  several striking features appear in the spectrum.

(1)  $E_1$  appears to narrow slightly with decreasing temperature above  $T_C$  but for  $T < T_C$ ,  $E_1$  broadens and a second peak  $E_1'$  splits off to lower energy. As noted later in our magneto-optical measurements, there is a third peak  $E_1''$  splitting off to higher energies, not resolved in Fig. 1 (see Fig. 2), which also contributes to the broadening.

(2)  $E_1$  has a red shift upon cooling similar to that of the absorption edge.<sup>6</sup> However, the peak shift is not as great as that of the edge, the peak shifting about 0.2 eV and the edge about 0.3 eV

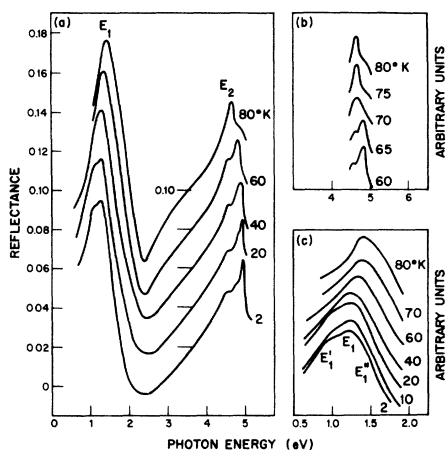


FIG. 1. (a) Reflectance versus photon energy at selected temperatures above and below the Curie point  $T_C = 69^\circ\text{K}$ . Each curve is arbitrarily displaced 0.02 from the one above. (b) Detail of  $E_2$  near the Curie point. (c) Detail of  $E_1$ .

between 80 and  $10^\circ\text{K}$ . It thus appears that if the low-energy tail of  $E_1$  is giving rise to the absorption edge as we believe, then the additional red shift of the absorption edge is caused by the splitting off of the transition  $E_1'$ .

(3)  $E_2$  does not appear to have a red shift as does  $E_1$ . However, there are distinct changes in it on cooling below  $T_C$ , as shown in Figs. 1(a) and 1(b). The peak at 4.65 eV and the high-energy shoulder resolve into a remarkably sharp peak at 4.95 eV with at least two lower energy shoulders.

In our second set of experiments the reflectance around  $E_1$  was examined using right and left circularly polarized light in a magnetic field. For measurements at  $1.5^\circ\text{K}$ , the sample was immersed in pumped liquid He with the magnetic field applied perpendicular to the reflecting face of the sample. In this configuration, a minimum field of 25 kOe is required to remove the domain structure. In our experiments, 42 kOe were used. The results are shown in Fig. 2. The structure of  $E_1$  in the ferromagnetic state is clearly resolved into three components. The central  $E_1$  peak is seen for both polarizations,  $E_1'$  is seen only in  $\sigma_R$  polarization and  $E_1''$  only in  $\sigma_L$  polarization. The convention used is that  $\sigma_R$  corresponds to a magnetic quantum number change  $\Delta m = -1$  while  $\sigma_L$  corresponds to  $\Delta m = +1$ . The energy splittings of  $E_1'$  and  $E_1''$  from the central  $E_1$  peak are equal and have the value  $\Delta E_1 = 0.25$  eV.

Similar measurements were made near  $T_C$  at  $70^\circ\text{K}$ . With 42 kOe applied,  $E_1$  is split into a dou-

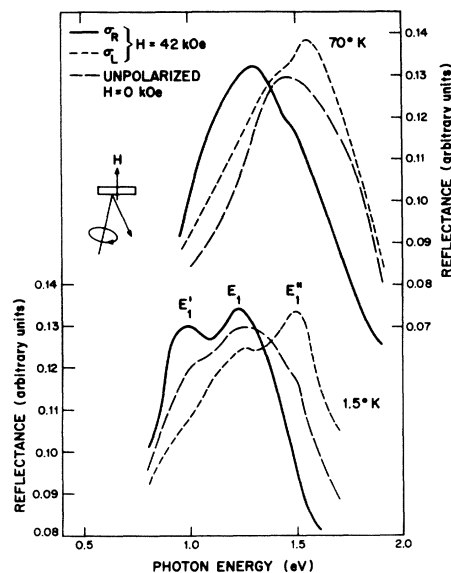


FIG. 2. Reflectance versus photon energy for right and left circularly polarized light at 70 and  $1.5^\circ\text{K}$ . The unpolarized reflectance is also shown.

plet with a total separation of 0.25 eV compared with the total separation of 0.5 eV at  $1.5^\circ\text{K}$ . We confirmed that the unpolarized peak is considerably broader at  $1.5^\circ\text{K}$  than at  $70^\circ\text{K}$  because of the increased splittings due to magnetic exchange.

In a recent review paper,<sup>7</sup> Methfessel and Mattis have documented the various extensive experimental results and conclude that the absorption edge in the Eu-chalcogenide semiconductors arises from a  $4f$ - $5d$  transition. The basic energy-band scheme is one in which the fundamental anion  $p$  band and a europium  $5d$ - $6s$  conduction band. The  $4f$  levels are highly localized and the ground state may be placed in this gap.

Using this model of localized  $4f$  states and  $5d$  bands for the electrons, as shown in Fig. 3, we have calculated, for right and left circularly polarized radiation, the transition probabilities for transitions from an all-spin-up ground state consisting of a  $4f^7(^8S_{7/2})$  configuration to excited states corresponding to the spin-up components of the  $4f^6(^7F_J)5d(t_{2g})$  states for the  $J=0, 1, 2, 3, 4, 5,$  and  $6$  configurations using essentially the same procedure as Freiser, Methfessel, and Holtzberg.<sup>8</sup> Our calculation differs from theirs in that they considered an  $e_g$  final state corresponding to the lowest  $5d$  state in  $\text{EuF}_2$  with a fluorite structure, while we have made the calculation for a  $t_{2g}$  final state corresponding to the lowest  $5d$  state in ferromagnetic  $\text{EuO}$  with rock-salt structure.<sup>6</sup> Also, instead of averaging over all

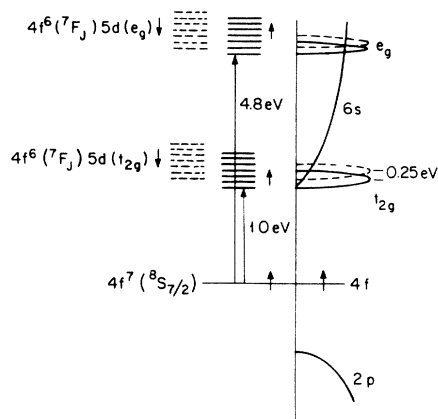


FIG. 3. Schematic band structure of ferromagnetic EuO. The right side shows the fundamental energy gap and the narrow  $5d$  bands. The left side shows the  $E_1$  and  $E_2$  reflectance structure as transitions from the localized  $4f^7$  ground state to a final state consisting of the ladder structure of a  $4f^6$  excited state and an electron in a spin-split component of the  $d$  bands.

$M_S$  quantum numbers as they did, we have considered only  $M_S = -3$  corresponding to the fully spin-polarized bands. When this is done, we find that the transitions to the higher  $J$ -value states are dominantly left circularly polarized while the transitions to the lower and intermediate  $J$ -value states are dominantly right circularly polarized. Consequently, simply due to the energy separations of these final-state configurations, we obtain a resolution of the transition into right and left circularly polarized components. The amount of this resolution can be estimated from the data on  $\text{EuF}_2$  to be approximately 0.3 eV. We believe that this value corresponds to the doublet splitting of 0.25 eV we observe near  $T_C$ . It should be recognized that the resolution into these polarized components depends on our considering only the  $M_S = -3$  state and that it disappears if one averages over all  $M_S$  values. In the model, then, the polarization effects near  $T_C$  are due to a preferential population of the spin-up  $^8S_{7/2}$  ground state in the presence of the magnetic field. The exchange splitting of the  $5d$  conduction-band state is assumed to be small when the temperature is near  $T_C$ . When full magnetic saturation is reached at low temperature, the resolved doublet at 70°K is further split, the various components corresponding to transitions to each of the two spin-split states of the  $5d$  band. Four transitions are expected, but because the exchange splitting is also  $\sim 0.25$  eV, the two central components of the quartet overlap giving a triplet. Thus, we obtain pure polarized components for

the extrema and a mixed central component, as observed experimentally. Although the model is obviously a simplified representation, it does give an adequate interpretation of the data.

These results, along with optical rotation measurements near the absorption edge<sup>1</sup> and photoconductivity measurements,<sup>9</sup> strongly suggest that we are indeed observing transitions to the spin-polarized conduction-band structure. In ferromagnetic metals, reflectance structure associated with the spin-split bands is not readily observed because transitions to the Fermi surface do not usually give resolvable structure which can be followed through  $T_C$ . However, in the ferromagnetic semiconductor EuO, these transitions are observed because they occur at critical points in the joint density of states at the conduction-band minima and also because the  $5d$  conduction band is apparently quite narrow.

Cho's<sup>10</sup> latest band calculation for the Eu chalcogenides shows essentially the same features as this model except that there is a broad  $5d$  conduction band. However, our data as well as other results are more consistent with narrower, well separated,  $t_{2g}$  and  $e_g$  sub-bands. This is seen by comparing the two prominent features in the  $\text{EuF}_2$  absorption spectra.<sup>6</sup>

In  $\text{EuF}_2$ , sharp ladder structure in the lower energy peak is associated with transitions to the lower  $e_g$  component of the  $d$  band, while the well separated but less structured high-energy peak is associated with transitions to the  $t_{2g}$  state.<sup>8</sup> In EuO, this ladder structure appears in  $E_2$ , since  $t_{2g}$  and  $e_g$  are expected to be reversed.<sup>6</sup> Structure in the  $t_{2g}$  transitions is not resolved because of spin-orbit broadening which is absent in  $e_g$  transitions.<sup>8</sup> Thus, we believe  $E_2$  is due to transitions  $4f^7(^8S_{7/2}) - 4f^6(^7F_J)5d(e_g)$  although broader structure from other interband transitions ( $p - 5d$  and  $6s$ ) may be superimposed. The sharp structure of  $E_2$ , and the isolation of  $E_1$  which shows up more markedly in a plot of the imaginary part of the dielectric constant,<sup>5</sup> therefore lead us to believe that the transitions are to narrow, well separated  $t_{2g}$  and  $e_g$  states. We have not yet studied  $E_2$  in polarized light to determine if exchange splittings are also involved here.

Although spin-orbit effects were considered only for the  $4f$  states in our model, the spin-orbit splitting of the narrow  $t_{2g}$  band by  $\sim 0.3$  eV<sup>11</sup> should also contribute to the polarized doublet structure at  $T \sim T_C$  in much the same way that this effect causes the magnetic rotation in

$\text{CrBr}_3$ .<sup>12</sup> The origin of the triplet structure for  $T < T_C$  including this spin-orbit effect is still the splitting of the final  $5d$  conduction-band states by the exchange interaction as we described.

One must also consider the possibility that in  $\text{EuO}$  the  $5d$  levels do not form itinerant bands, but are localized since optical rotation effects in ferromagnetic insulators, such as  $\text{CrBr}_3$ ,<sup>12</sup> are due to transitions between localized states. In addition, the red shift of the absorption edge above the Curie point is most easily explained by localized short-range spin-split interactions.<sup>13</sup> However, we feel that if our model for the splitting of the  $E_1$  transition is valid, the change we observe from a doublet structure near  $T_C$  to a triplet structure at low temperatures requires the  $5d$  state to have delocalized character. Very simply, in a strongly coupled model, the energy state of the  $d$  electron will be lowest when aligned with the  $4f$  spin on the same ion. Therefore a spin splitting of a localized  $5d$  state should exist both above and below  $T_C$ . In the delocalized model, the  $d$  electron sees an average spin which is zero for  $T > T_C$  but is polarized for  $T < T_C$ . Therefore an exchange splitting will be seen only in the magnetic state, as we observe. Transport measurements<sup>14</sup> and photoconductivity<sup>9</sup> associated with the absorption edge also suggest a conducting  $d$  band. However, we have not ruled out the possibility that a broad  $6s$  band, degenerate with the  $5d$  band at the bottom, may be important for the transport measurements. Extension of this work to other Eu chalcogenides can help elucidate this problem.

We are indebted to S. J. Cho, S. von Molnar, D. Eastman, and P. M. Grant for discussion of their unpublished results. We thank B. Feldman for his invaluable technical assistance in conducting the experiments and R. E. Fahey for his as-

sistance in growing crystals.

†Work sponsored by the U. S. Air Force.

\*Work supported by the U. S. Air Force Office of Scientific Research.

<sup>1</sup>J. C. Suits and B. E. Argyle, *J. Appl. Phys.* **36**, 1251 (1965), and *Phys. Rev. Letters* **14**, 687 (1965); B. E. Argyle, J. C. Suits, and M. J. Freiser, *Phys. Rev. Letters* **15**, 822 (1965); J. H. Greiner and G. J. Fan, *Appl. Phys. Letters* **9**, 27 (1966); K. Y. Ahn and J. C. Suits, *IEEE Trans. Magnetics* **2**, 453 (1967); J. O. Dimmock, C. E. Hurwitz, and T. B. Reed, *Appl. Phys. Letters* **14**, 49 (1969).

<sup>2</sup>G. Busch and P. Wachter, *Physik Kondensierten Materie* **5**, 232 (1966).

<sup>3</sup>S. Methfessel, *Z. Angew. Phys.* **18**, 414 (1965); J. C. Suits, B. E. Argyle, and M. J. Freiser, *J. Appl. Phys.* **37**, 1391 (1966); S. Methfessel, F. Holtzberg, and T. R. McGuire, *IEEE Trans. Magnetics* **2**, 305 (1966).

<sup>4</sup>P. M. Grant and J. C. Suits, to be published.

<sup>5</sup>W. J. Scouler, J. Feinleib, J. O. Dimmock, and T. B. Reed, *Bull. Am. Phys. Soc.* **14**, 309 (1969).

<sup>6</sup>M. J. Freiser, F. Holtzberg, S. Methfessel, G. D. Pettit, M. W. Shafer, and J. C. Suits, *Helv. Phys. Acta* **41**, 832 (1968).

<sup>7</sup>S. Methfessel and D. C. Mattis, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, Germany, 1968), Vol. XVIII, Pt. 1, p. 389.

<sup>8</sup>M. J. Freiser, S. Methfessel, and F. Holtzberg, *J. Appl. Phys.* **39**, 900 (1968).

<sup>9</sup>R. Bachmann and P. Wachter, *Solid State Commun.* **6**, 771 (1968).

<sup>10</sup>S. J. Cho, private communication.

<sup>11</sup>F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1963).

<sup>12</sup>J. F. Dillon, H. Kamimura, and J. P. Remeika, *J. Phys. Chem. Solids* **27**, 1531 (1966).

<sup>13</sup>S. Methfessel, M. J. Freiser, G. D. Pettit, and J. C. Suits, *J. Appl. Phys.* **38**, 1500 (1967).

<sup>14</sup>T. Kasuya and A. Yanase, *Rev. Mod. Phys.* **40**, 684 (1968); C. E. Hurwitz, M. R. Oliver, J. O. Dimmock, and T. B. Reed, *Bull. Am. Phys. Soc.* **14**, 309 (1969).

## PRODUCTION OF $^{11}\text{B}$ AND $^{10}\text{B}$ BY PROTON SPALLATION OF $^{12}\text{C}$ †

Cary N. Davids, Helmut Laumer, and Sam M. Austin

Physics Department, Michigan State University, East Lansing, Michigan 48823

(Received 20 December 1968; revised manuscript received 9 May 1969)

Measurements of the  $^{10}\text{B}$  and  $^{11}\text{B}$  spallation production in  $^{12}\text{C}$  are presented. These cross sections are related to theories on the origin of the elements lithium, beryllium, and boron.

The nucleosynthesis of the light elements lithium, beryllium, and boron ( $\text{LiBeB}$ ) is a problem of great astrophysical interest.<sup>1-3</sup> Since these elements have very short lifetimes for destruction by stellar thermal protons, they will not

survive if produced in the inner regions of stars. It is generally believed that they are produced in a cool stellar environment by the proton-induced spallation of abundant heavier nuclei. The fast protons, with energies of several MeV and high-