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MAGNETIC DICHROISM IN EuSe

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Magnetic dichroism is the magnetic-fieldinduced difference in optical absorption coefficients for light with different senses of polarization. Magnetic circular dichroism, which occurs for light propagating along the direction of magnetization, is well known and produces the ellipticity associated with the Faraday effect (magnetic circular birefringence). Similarly, magnetic linear dichroism, which occurs for light propagating at right angles to the magnetization, must coexist with the Cotton-Mouton effect (magnetic linear birefringence), although both of these effects are usually so small as to escape observation. This paper describes a remarkably large magnetic circular and linear dichroism exhibited by a single crystal of EuSe at 4.2°K. These effects reflect not only the wavelength dependence and magnetic shift of the broad-band absorption but also the various spin structures exhibited by EuSe.

EuSe is unusual in that the antiferromagnetic spin configuration of Eu⁺⁺ ions at temperatures below the ordering temperature ($\sim 6^{\circ}$ K) can be readily altered by a magnetic field. Combined measurements of magnetization¹ and powder neutron diffraction² suggest the following picture: In zero field the spins in each (111) plane are ferromagnetically aligned, but angles between spins of successive (111) planes are approximately 90°, 180° , -90° , 180° , 90° , ... At intermediate applied fields (1-2 kOe for powder specimens), the spins of two of the four sets of (111) planes flop into parallel alignment with the other two and yield a nonoscillatory ferromagnetic component. Above ~8 kOe pure ferromagnetic alignment remains. The other europium chalcogenides exhibit either ferromagnetic order (EuO and EuS) where the nearest-neighbor ferromagnetic exchange J_1 dominates, or pure antiferromagnetic order of the second kind (EuTe) where next-nearest-neighbor antiferromagnetic superexchange J_2 dominates. For EuSe the molecular-field analysis³ suggests $J_1 \approx -J_2$. Then, since a given Eu⁺⁺ ion is coupled to a neighboring (111) plane by three nearest-neighbor interactions (J_1) and three second-nearest-neighbor interactions (J_2), it is not surprising that the turn angles deviate from the usual antiferromagnetic values of 180°.

Our measurements of magnetic dichroism were made on a single crystal of EuSe immersed in liquid helium (the same crystal described in reference 4). Figure 1 shows the absorption coefficients (α_1 and α_r) for left and right circularly polarized light of wavelengths 680, 700, and 800 m μ versus magnetic field H applied parallel to the direction of light propagation and normal to the crystal plate. These absorption data are not corrected for reflection losses.⁵ The large changes in absorption (corresponding to factors as large as 30 in transmitted intensity) are readily visible to the eye. We note that EuSe exhibits three distinct regions of optical behavior which seem to correlate with the three types of spin configuration discussed above. For $H \ge 10$ kOe (the ferromagnetic region) α_r is enhanced significantly, while α_l is relatively unaffected by the field. At intermediate applied fields $(3 \approx H \approx 10 \text{ kOe})$, both

 α_l and α_r go through a broad maximum near 6.5 kOe. In the low-field region (below ~3 kOe), both absorption coefficients show small changes which are quite sensitive to sample misorien-tation.

The magnetic linear dichroism of EuSe is shown in Fig. 2 which gives the absorption coefficients for light of wavelengths 680, 700, and 800 m μ propagating normal to the plane of the specimen while the applied field (and the magnetization) lies in the plane of the specimen. The curves labeled α_{\perp} and α_{\parallel} refer to the plane of polarization of the incident light being perpendicular and parallel, respectively, to the field. The structure at low fields again appears to reflect the transitions between magnetic phases, but the critical applied fields are now much smaller than in Fig. 1 because of the smaller demagnetizing field. The regions of characteristic behavior appear at about the same internal fields as may be noted from the top scale⁶ in Figs. 1 and 2.

With increasing wavelength of the incident light the dichroism diminishes rapidly. Figure 3 gives the variation with wavelength of the absorption coefficients α_l , α_r , α_{\parallel} , and α_{\perp} measured in an applied field of 15 kOe. The absorption coefficient in zero field is also given for 4.2°K, 77°K, and room temperature. All curves are normalized to the room-temperature absorption for 800 m μ as measured using a Cary Model 14 spectrophotometer.



FIG. 1. Magnetic circular dichroism of EuSe single crystal of thickness 157μ at 4.2° K.



FIG. 2. Magnetic linear dichroism of EuSe single crystal at 4.2° K.



FIG. 3. Absorption edge in EuSe single crystal in zero-field paramagnetic state [α (0) at 77°K and room temperature], in zero-field antiferromagnetic state [α (0) at 4.2°K], and in high-field ferromagnetic state (15 kOe at 4.2°K).

In the paramagnetic temperature region, the absorption curve shifts towards shorter wavelengths with decreasing temperature as observed in other (nonmagnetic) materials. However, the curve of $\alpha(0)$ at 4.2°K shows that in the temperature region of spin ordering, a reverse shift (to longer wavelengths with decreasing temperature) occurs in agreement with observations on EuO and EuS by Busch, Junod, and Wachter.⁷

The anomalous maximum in the observed absorption in the intermediate field region (Figs. 1 and 2) may arise from scattering of the light out of the optical aperture due to the magnetic structure. The intermediate-field spin structure may in fact be an inhomogeneous mixture of a simple antiferromagnetic structure (of the type MnO) with the high-field ferromagnetic structure.⁸ The height of the absorption maximum varies in the same sense but more rapidly than λ^{-4} of a simple Rayleigh scattering law, but, since the absorption of the ferromagnetic phase has a strong dependence on wavelength, the change in scattered intensity with wavelength will be enhanced. Another mechanism capable of removing light from the main beam is diffraction of light by

magnetic domains as observed in CrBr_3 .⁹ Attempts to observe scattered light have not yet been successful.

It is noteworthy that Fig. 3 shows a very large shift of all 4.2°K absorption curves towards longer wavelengths when the fields become large enough to produce the ferromagnetic phase. For right circularly polarized light, the shift in the absorption curve at 4.2°K produced by an applied field of 15 kOe (an internal field of about 5 kOe) is close to 1000 cm⁻¹. Shifts of this magnitude are consistent with the observations of Busch, Junod, and Wachter⁷ who deduced from their diffuse reflection measurements on powdered EuO a shift in the absorption edge between 90 and 20°K of about 1900 cm^{-1} . (The Curie temperature of EuO is 69°K so that at 20°K the magnetization is near its saturation value for 0° K.) These results imply state splittings of the order of one or two thousand cm^{-1} . Since the exchange splitting inferred from T_c of the $4f^7$ (⁸S) ground state of Eu⁺⁺ is for EuSe only of the order of 10 cm^{-1} , and for EuO of the order of 100 cm^{-1} , splitting of the excited states must be responsible for the absorption shift.

EuSe is particularly interesting since at 4.2° K in zero field the spins have the antiferromagnetic order described previously. Therefore the excited states should exhibit exchange splittings even in the absence of an applied field. The fact that with the transformation to a ferromagnetic phase a shift to <u>lower</u> energies is nonetheless observed implies that the exchange interaction of the excited ions with the neighboring ions is ferromagnetic.

We propose that the transition involved here is a charge-transfer transition in which an felectron from one Eu ion is excited into a state made up of 5d orbitals on the nearest-neighbor Eu ions. The excited Eu ion then has the configuration $4f^{6}$, while the twelve nearest neighbors have a mixture of $4f^7$ and $4f^75d$. This transition has already been described by Methfessel.¹⁰ The $4f^75d$ state has been invoked by Goodenough¹¹ to account for the ferromagnetic interaction between nearest-neighbor Eu ions in the chalcogenides. Suppose, then, that the magnetic energy of the excited d electron is given by $H_m = -2(J/12)\overline{\sigma}_d \cdot \sum \overline{S}_n$, where $\overline{\sigma}_d$ is the spin of the *d* electron, \overline{S}_n is the spin of the 4f shell of a neighbor, and J is the intra-atomic f-d exchange integral. For the paramagnetic state H_m has the expectation value zero (neglecting short-range order). For the ferromagnetic ground state with $S = \frac{7}{2}$, $\langle H_m \rangle = -3.5J$. The choice of $J = 540 \text{ cm}^{-1}$ then yields the shift of 1900 cm⁻¹ observed⁷ in EuO. For the antiferromagnetic phase of EuSe, one obtains $\langle H_m \rangle = \frac{1}{4}(-3.5J)$. The expected shift at 0°K upon transformation to the ferromagnetic phase is then $\frac{3}{4}(-3.5J)$, that is, 1420 cm^{-1} . At 4.2° K and 15 kOe, however, the magnetization of EuSe is about 0.8 of its saturation value at 0°K.¹ Since $\langle H_m \rangle$ is approximately proportional to M^2 , we would therefore expect a shift in the band edge at 4.2°K of about 900 cm⁻⁻¹, in reasonable agreement with the 1000 cm^{-1} reported above. The value of $J = 540 \text{ cm}^{-1}$ for the f-d exchange in Eu⁺ is plausible in view of the results of Cal $lahan^{12}$ in the isoelectronic Gd^{++} which yield a value of J = 1013 cm⁻¹. The contraction of the orbitals in the more highly charged Gd^{++} ion would account for this larger exchange.

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MAGNETIC COUPLING BETWEEN TWO ADJACENT TYPE-II SUPERCONDUCTORS

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It is possible for a type-II superconductor to be in a resistive, yet superconductive, state. According to Anderson's¹ flux-creep model, the resistive behavior is associated with the motion of quantized flux vortices (fluxons) present in Abrikosov's² theory. This Letter deals with an experiment which shows that it is indeed permissible to interpret literally the voltage drop along a type-II superconductor as arising from the motion of flux.

The experimental arrangement is shown in Fig. 1. First, an Sn film a few thousand angstroms thick is evaporated onto a microscope glass slide. I shall refer to this film as the primary. The center section of the primary is reduced in width by outlining it with a razor blade. Next, a thin film of SiO_2 approximately 100-200 A thick is evaporated over the primary film. Finally, a second Sn film is evaporated over the SiO_2 layer. I shall refer to this film as the secondary. The secondary film is made as thin as possible, i.e., of the order of 500-1000 Å, and it must be narrower than the primary film. The two metal films are electrically separated

by the SiO_2 film, i.e., the measured resistance between them approaches infinity.

The experiment consists of passing a dc current I_{D} along the primary film and measuring the dc voltage developed both in the primary film V_p and in the secondary film V_s . Because of the narrow section in the center of the primary, the voltage drop in the primary is limited to a short length that is completely paralleled by the secondary. The current and voltage connections are shown in Fig. 1. When both the films are normal, or when the secondary film is in the normal state and the primary film is in the superconducting state, no voltage can be detected in the secondary loop as shown in Fig. 2. However, as soon as both films are superconducting, a dc voltage is seen in the secondary loop as well as in the primary circuit. This behavior is displayed for three different temperatures in Fig. 2. If the primary current is increased such that the film enters the normal state, the primary voltage jumps to a high value (of the order of 0.5 V) while the secondary voltage abruptly drops to zero.