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Transport Properties of the Europium Chalcogenides

Abstract: Transport properties of pure and doped Eu chalcogenides are reviewed to determine the mechanisms responsible for the anomalous behavior near T_c, the Curie temperature. It is found that, whereas the scattering theory of simple metals accounts for the behavior of materials containing impurities in excess of 2×10^{20} cm^{-3}, several models for transport have been proposed for smaller concentrations. The impurity hopping model appears to be consistent with the data for very dilute systems.

Introduction

The transport properties of europium chalcogenides are unique in their strong dependence on the magnetic state of the system. One result is the appearance, near the Curie temperature, of an anomalous peak in the resistance, whose size depends critically on the carrier concentration. Another is a large negative magnetoresistance.[1-4] The large field-dependent resistance changes have hampered the investigation of the Hall effect except in samples with high carrier concentrations,[5,6] where the anomalous transport behavior is weakest. Nonetheless, these studies, in conjunction with other experiments that yield information about the conduction process in perhaps a less direct manner, have permitted the testing of various theoretical ideas concerning the mechanism of electron transport.

The chalcogenides are semiconductors with a fundamental absorption edge greater than 1 eV in all cases. The absorption edge has been interpreted variously as being a transition from the 4f state to the conduction band or to an excitonic $4f^6 5d$ state.[7-9] From this it cannot be decided whether the conduction band minimum has s or d character. Since the electric-dipole transition from f states to s states is forbidden, the energy gap between the f states and the bottom of the conduction band may be somewhat smaller than the optically observed 4f-5d transition energy and may have s character. In either case conduction due to intrinsic processes is expected to be negligible and "pure" samples have resistivities up to 10^{10} Ω-cm at room temperature. It is possible, however, to dope these materials and cause them to behave as extrinsic semiconductors. Several such alloys are indicated in Table 1. This does not represent a complete list, but the table does demonstrate that dopants are not confined to the trivalent rare earths. The rather surprising fact is that, wherever a thorough investigation of the resistivity has been made, the gross features are relatively insensitive to the nature of the dopant but very dependent on dopant concentration.

Table 1 Conducting Eu chalcogenide alloys.[a]

Material	*Solubility limit*
$Eu_{1-x}Gd_xO$	$x < 0.10$
$Eu_{1-x}Gd_xS$	$x = 1$
$Eu_{1-x}Gd_xSe$	$x = 1$
$Eu_{1-x}Gd_xTe$	$x = 1$
$Eu_{1-x}La_xO$	$x < 0.05$
$Eu_{1-x}La_xS$	
$Eu_{1-x}Nd_xS$	$x = 1$
$Eu_{1-x}La_xSe$	$x = 1$
$Eu_{1-x}O$	$x < 0.001$
$Eu_{1+x}S$	$x < 0.1$
EuS_{1+y}	$y < 0.002$
$EuS_{1-y}Cl_y$	$y < 0.06$
$EuS_{1-y}Br_y$	$y < 0.07$
$EuSe_{1-y}Cl_y$	$y < 0.06$
$EuSe_{1-y}Br_y$	$y < 0.007$

[a] From S. Methfessel et al., *Proc. Rare Earth Conference*, Grenoble 1969, to be published in *Compt. Rend.*

I shall try to show that when the doping level is high enough to produce a degenerate semiconductor the well-established theories of conductivity for simple metals suffice to explain all present results. For low concentrations of impurities on the other hand, conduction is either nonmetallic (in the sense that σ, the conductivity, approaches zero with decreasing temperature) or band-like

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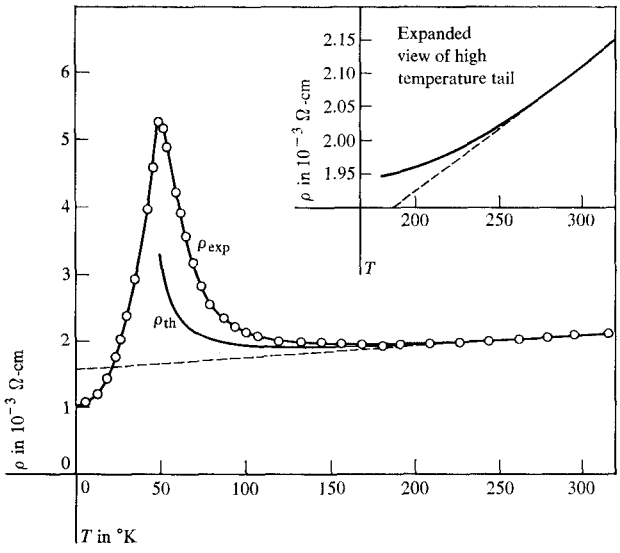


Figure 1 Temperature dependence of the resistivity ρ of nominal $Eu_{0.95}Gd_{0.05}S$. The true concentration is $Eu_{0.99}Gd_{0.01}S$. (From S. Methfessel et al., *Proc. Rare Earth Conf.*, Grenoble 1969, to be published in *Compt. Rend.*)

but in a band made up primarily of the wave functions of the impurities. Our understanding of this range is minimal. The situation is analogous to that encountered in ordinary semiconductors such as Si or Ge where, as the doping level is increased, the conduction process changes from hopping to band-like. For Ge and Si this transition occurs when the ratio of the mean distance between impurity sites, R_i ($R_i = N_i^{-\frac{1}{3}}$, where N_i is the number of impurities per cm^3), to the effective Bohr radius of the impurity electron, a_0, is about 5 (Ref. 10). One major difference is that, in the case of europium chalcogenides, a_0 is smaller than the lattice parameter and is difficult to estimate accurately. Nonetheless, assuming the applicability of the familiar formula

$$a_0 = \frac{\epsilon \hbar^2}{m^* e^2},$$

with the high frequency dielectric constant $\epsilon \approx 5$ (Ref. 11), and the effective mass m^* equal to the free electron mass, one obtains $a_0 \approx 2.5$ Å. When $R_i/a_0 \lesssim 5$, conduction is probably metallic. Thus, with $N_i \approx 2 \times 10^{20} y$ cm^{-3}, where y is the percent doping, the condition $R_i/a_0 \lesssim 5$ can be rewritten as $y \gtrsim 2.5$. Similarly, impurity concentrations of one percent or less probably result in non-metallic properties.

I wish to introduce one other concept at this point. It should be recognized that the impurities in a doped semiconductor form, in a sense, a random lattice. Anderson[12] and later Mott[13,14] pointed out that, given an impurity band of width J and a varying perturbing potential at each impurity site of average value W, impurity electrons are localized and no unassisted transport can occur for W/J greater than approximately 5. It may therefore be useful to think of the doped europium chalcogenides as ordinary impure semiconductors with additional perturbing potentials. These are of two types: (a) interactions with acoustic and optical phonons giving rise to polaron effects of the usual kind and (b) exchange interactions between the spins of impurity electrons and the localized spins of the Eu^{2+} ions ($S = 7/2$). As I shall try to demonstrate later, it is the exchange interaction that dominates the transport properties of lightly doped chalcogenides at temperatures greater than the Curie temperature T_c. For the temperature $T \ll T_c$, the material can be treated as an ordinary semiconductor and, depending on the ratio W/J, hopping or band-like conductivity may be expected.

Resistivity

The transport properties of EuS doped nominally with 5% Gd have recently been studied[5] and the temperature dependence of the resistivity is shown in Fig. 1.[15] Two characteristic features of the data are to be noted: (1) The high temperature results indicate a linear dependence of the resistivity on temperature and, (2) the resistivity has a sharp peak at 49°K. In addition, an investigation of the temperature dependence of the Hall effect facilitated the separation of the normal Hall constant R_0 from the anomalous portion R_1. The values obtained were $R_0 = -2.85 \times 10^{-10}$ Ω-cm/Oe and $R_1 \approx -30 \times 10^{-10}$ Ω-cm/Oe.

The interpretation of these results has been carried out in the spirit of the free-electron model and I shall summarize it here. From R_0 one derives an effective number of free carriers, $n = 2.2 \times 10^{20}$ cm^{-3}. This is, on the assumption that each Gd ion contributes a conduction electron, in good agreement with microprobe analysis, which yields a value[16] $N_i = (2.4 \pm 0.4) \times 10^{20}$ cm^{-3}. Furthermore, the relatively small value of the anomalous Hall constant R_1 is consistent with the assumption of s character at the bottom of the conduction band.

The analysis leading to the calculated curve in Fig. 1 assumes that scattering processes are independent of one another and that their contributions to the resistivity are additive. Thus the total resistivity is written as

$$\rho_{tot}(T) = \rho_i + \rho_L(T) + \rho_m(T) \quad (1)$$

where ρ_i is a temperature independent term due to neutral impurity scattering, ρ_L is due to acoustical phonon scattering and is linear in temperature, and ρ_m is a scattering term whose origin is the effective exchange interaction I_{sf} between the conduction electron and the localized 4f spins associated with each Eu^{2+} ion (or for that matter a Gd^{3+} ion). An experimental curve for ρ_m is then obtained simply by subtracting ρ_i, the low temperature limit of the resistivity, and $\rho_L(T)$, which is determined from the high temperature slope, from the measured total resis-

tivity. The remaining magnetic contribution is constant at high temperature, peaks near T_c, and approaches zero with decreasing temperature. This temperature dependence was first predicted by de Gennes and Friedel,[17] the anomaly near T_c resulting from critical scattering of the electrons by correlated spins. At high temperatures the expression for the resistivity is of the exact form[18]

$$\rho_m(\infty) = 4.3 \times 10^{-4}(10^{23}/N)\mu^2 S(S+1)(I_{sf}^2/E_F), \quad (2)$$

where N is the number of magnetic ions per cm^3, μ is the ratio of the effective mass m^* to the free electron mass and E_F is the Fermi energy. This formula is useful because an estimate of I_{sf} can be made if $\rho_m(\infty)$ is known. Substitution of $S = 7/2$, $\mu = 1$, $E_F = (\hbar^2/2m)(3\pi^2 n)^{\frac{2}{3}}$ and $\rho_m(\infty) = 0.56 \times 10^{-3}$ Ω-cm yields $I_{sf} = 0.043$ eV. As T_c is approached the expression for $\rho_m(T)$ involves correlation effects and is of the form

$$\rho_m(T) = \rho_m(\infty) \int_0^{\pi} \tfrac{1}{2} \sin\theta(1 - \cos\theta) \times \sum_i \gamma(\mathbf{R}_i) \exp(i\mathbf{K}\cdot\mathbf{R}_i), \quad (3)$$

where $\mathbf{K}$ is the momentum transfer, $\mathbf{R}_i$ the vector between the spins in question and $\gamma(\mathbf{R}_i)$ is the spin-spin correlation function. Mathematical evaluation of γ is difficult and, as can be seen from Fig. 1, the curve calculated with the simplest form of γ, that derived for a nearest-neighbor ferromagnet, predicts a much smaller contribution than is observed experimentally. To account for this difference it was suggested that an additional contribution to scattering of the quasifree carriers might be expected from localized impurity states at the bottom of the band. These localized impurities form spin clusters and therefore enhance the effective scattering cross section. A systematic investigation of γ that includes correlation due to exchange interactions between localized impurity electrons and the Eu^{2+} spins has been undertaken by Kasuya and Yanase,[19] and agreement between theory and experiment has improved considerably.

Caution must be exercised, however, in making such an analysis. Perhaps the most fundamental question is whether or not expressions (2) and (3), results of the first Born approximation, are valid in our case. The criteria may be written[20] as follows:

If $ka < 1$,

$$\frac{m^* Va^2}{\hbar^2} < 1 \text{ must be satisfied, and} \quad (4a)$$

if $ka > 1$,

$$\frac{m^* Va}{\hbar^2 k} < 1 \text{ must be satisfied,} \quad (4b)$$

where k is the magnitude of the Fermi wave vector, a the radial extent of the spherical scattering potential, m^* the effective mass, and V the strength of the potential.

A rough calculation for the case of the doped EuS and spin-disorder scattering [Eq. (2)] can be made if a is known. On the assumption that a represents the average radial probability distribution of the 4f electron we have $a \approx 0.5$ Å and $ka = (3\pi^2 n)^{\frac{1}{3}} a \approx 0.1$. Thus condition (4a) applies,

$$\frac{m^* Va^2}{\hbar^2} = \frac{m^* I_{sf} Sa^2}{\hbar^2 N} \times \frac{3}{4\pi a^3} \approx \frac{I_{sf}(\text{eV})}{a(\text{Å})} \times 6.3 = 0.54,$$

and the approximation is valid.

Although the numerical values cannot be regarded as being particularly accurate, this calculation does demonstrate that the use of the Born approximation is questionable for s-d exchange interactions, which are probably encountered in the chromium spinels and which are much larger than I_{sf}.

Another feature which must be reckoned with in magnetic semiconductors is the band splitting below T_c. This exchange splitting, $2I_{sf}S \approx 0.3$ eV in Eu chalcogenides, produces a redistribution of the carriers and affects the calculation of any scattering process. An estimate of E_F in the ferromagnetic region of our samples yields $E_F = (\hbar^2/2m^*)(6\pi^2 n)^{\frac{2}{3}} \approx 0.2$ eV and leads to the interesting conclusion that all conduction electrons are polarized in one spin direction below T_c. This notion may prove to be useful in tunneling studies. Thompson et al.[21] have fabricated Schottky barriers between degenerate EuS and In metal. It is unlikely that spin-flip scattering will destroy much of the polarization during the tunneling process and the junction may therefore act as a source of spin-polarized electrons at temperatures much lower than T_c.

Scattering theory of carriers in a simple parabolic band apparently explains the available transport data on Eu chalcogenides with carrier concentrations in excess of about 2×10^{20} cm^{-3} for $T > T_c$. Although minor quantitative features need to be explored further, the predicted magnitude and temperature dependence of the resistivity anomaly are consistent with experiment. It should also be pointed out that 2×10^{20} carriers per cm^3 is approximately a one-percent impurity concentration and means that our previous estimate of the "metal-nonmetal" transition was somewhat conservative.

Any interpretation of transport properties in europium chalcogenides containing a small number of carriers (produced by dopants such as trivalent lanthanum and gadolinium), vacancy states (chalcogen vacancies act as donors), excess metal, or photoexcited electrons must deal consistently with the following set of experimental facts observed in EuS, EuSe (see Figs. 2 and 3) and EuTe:[7]

1) The resistivity increases exponentially with decreasing temperature in the paramagnetic region.

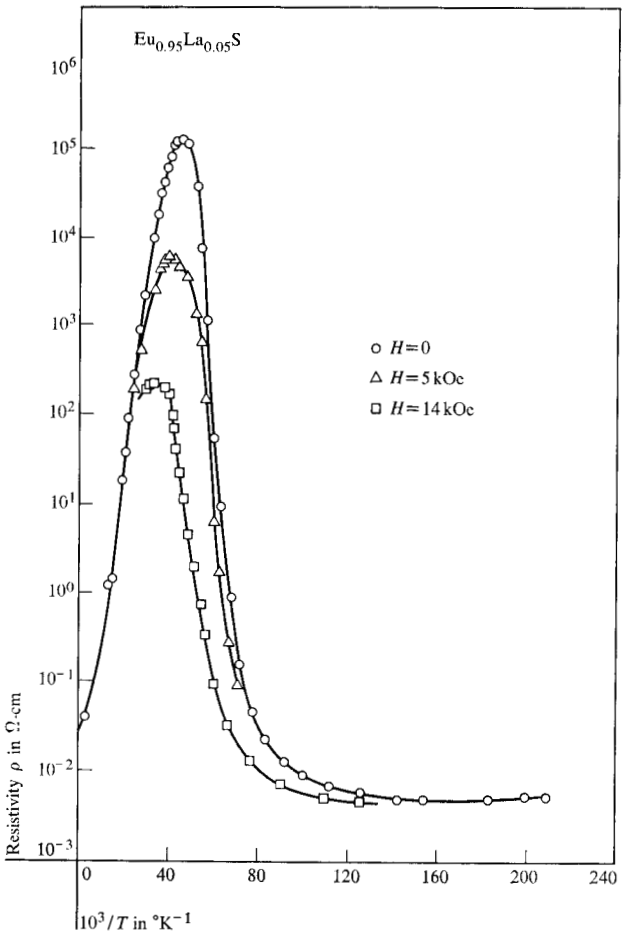


Figure 2 Temperature dependence of the resistivity ρ of nominal $Eu_{0.95}La_{0.05}S$ for several applied magnetic fields H. (From S. Methfessel et al., *Proc. Rare Earth Conf.*, Grenoble 1969, to be published in *Compt. Rend.*)

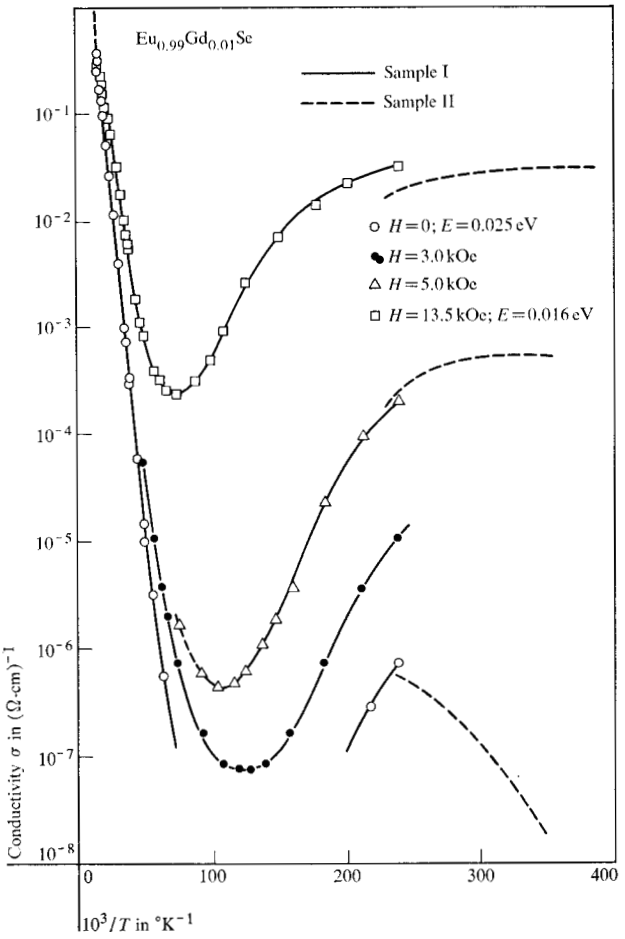


Figure 3 Temperature dependence of the conductivity σ of nominal $Eu_{0.99}Gd_{0.01}Se$ for several applied magnetic fields H. [Adapted from S. von Molnar et al., *J. Appl. Phys.* **38,** 959 (1967).]

2) A resistivity peak occurs near the magnetic ordering temperature T_c.
3) The peak position in temperature, the peak height and the apparent activation energy for transport in the paramagnetic region, are very sensitive to externally applied magnetic fields.
4) The influence of magnetic fields on resistivity is observable up to $2T_c$ or higher temperatures.

Hall effect and photoconductivity

The question of whether or not the exponential behavior in the paramagnetic region arises from changes in carrier concentration or mobility led to an investigation of the Hall effect in Gd-doped EuSe.[4] Although measurements were limited to samples with apparent carrier concentrations in excess of 10^{19} cm^{-3} for experimental reasons, the Hall results indicated that variations in the mobility and not in carrier concentration account for the observed temperature dependence. This suggests thinking of transport in terms of hopping in samples with small carrier concentrations, such as the nominally 1% Gd-doped EuSe depicted in Fig. 3, but the Hall effect has not been measured for such high impedance samples. The best evidence, so far, that exponential mobility variations dominate transport in EuSe and EuS comes from photoconductivity experiments.[22,23] Figure 4 depicts the results of photoconductivity measurements by Penney[23] in undoped EuSe. A comparison with Fig. 3 illustrates the remarkable agreement between the two measurements in the paramagnetic region. Not only the activation energies E derived from the slope of the curves, but also the changes in E with applied magnetic field are comparable for both figures. Whereas in doped materials the quantity measured is $\sigma = ne\mu$, photoconductivity experiments measure the mobility-lifetime product for a given quantum efficiency. It is therefore reasonable to conclude that μ, the common factor, dominates the temperature dependence in both measurements. The alternative explanation requires that

n, the mobile carrier density of the doped material, and τ_l, the lifetime of the photoexcited electron, have the same temperature and magnetic field dependence. This is an extremely unlikely possibility.

Theoretical models

• *Impurity hopping*

Three models have been suggested to explain transport properties in lightly doped chalcogenides. The most detailed is the impurity model described by Kasuya and Yanase.[24] They interpret the resistivity as an impurity hopping phenomenon with an activation energy whose major contribution comes from the difference in local spin order between the neighborhood of the empty state and that of the occupied impurity state. This difference arises because the impurity wave function is expected to have sufficient overlap with the nearest-neighbor Eu^{2+} ions to enhance the exchange interaction and produce a cluster of ordered spins. The model calculation[24] actually represents the low concentration scheme, with $J \lesssim W \ll E$, where E, the magnetic activation energy, is, roughly speaking, the difference in energy between occupied and unoccupied impurity states. Difficulty in obtaining a quantitative fit for all temperatures arises because of the temperature dependence of E. Kasuya obtained a good fit to the data (Fig. 3) in the paramagnetic region where E is relatively constant and large. In the neighborhood of T_c and below, however, E is expected to decrease, and vanishes well below the Curie temperature because the spin lattice is ordered regardless of the presence or absence of an electron at the impurity site. If $J \gtrsim W$, impurity banding occurs. This appears to be responsible for the abrupt decrease in and flattening out of the resistivity below the Curie temperature (see Fig. 2). (I should point out that the conductivity decrease observed in Fig. 3 at temperatures below 4.2°K and for $H = 0$ does not represent the condition $J \ll W$, but reflects the remanence of the complex spin structure of pure EuSe.[25])

The applicability of the impurity model depends on the condition that the impurity levels lie deeper than kT, where k is Boltzmann's constant, below the bottom of the conduction band. Kasuya and others have assumed that the impurities or other defects have a binding energy of the order of 0.5 eV. Although high temperature measurements in doped EuSe did not show the onset of conductivity by thermal activation of carriers from such states into the conduction band, more recent work on EuO doped with excess Eu confirms the existence of such states. Examples of the data are shown in Fig. 5. The Hall effect in Eu-doped EuO changes in proportion with the resistivity ρ, and the activation energies, calculated by assuming $\rho = \rho_0 \exp \Delta E/kT$, vary between about 0.2 eV and 0.4 eV. This spread in energy is to be expected since the position

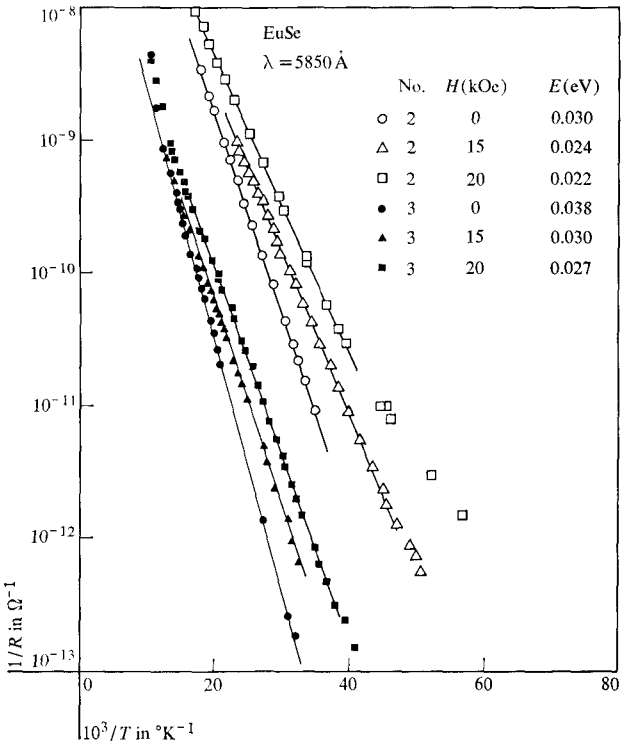
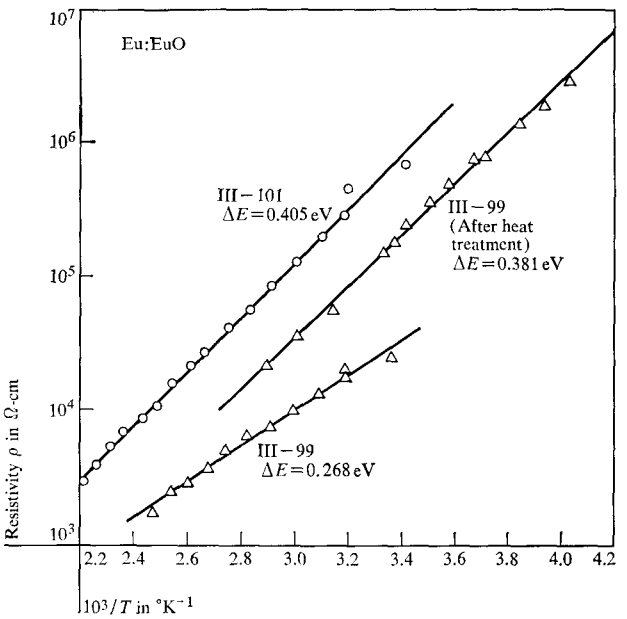


Figure 4 Photoconductance $1/R$ of two EuSe samples at various magnetic fields H. (From T. Penney, *Proc. III Int. Conf. Photoconductivity*, Stanford 1969).

Figure 5 Temperature dependence of the resistivity ρ of several samples of EuO doped with Excess Eu ($T \gg T_c$).



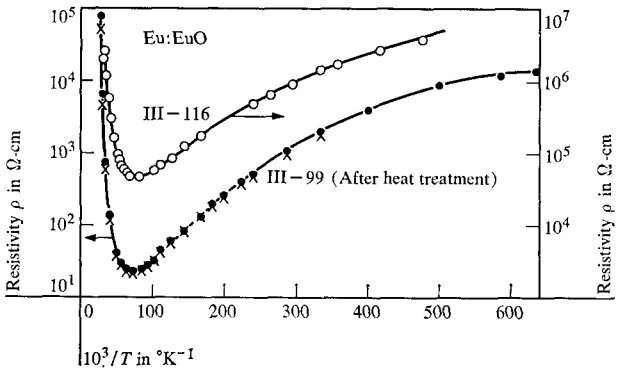


Figure 6 Temperature dependence of the resistivity ρ of several samples of EuO doped with excess Eu ($T \ll T_c$).

of the Fermi energy depends sensitively on the degree of compensation, as well as on the position of the various impurities and defects. Although the exact location in energy of the impurity states requires a more detailed investigation, we can assume that the states lie at energies lower than 0.2 eV below the bottom of the conduction band. Further evidence for deep states is the observation by Penney[23] of an exponentially decreasing dark conductivity in EuSe with an activation energy of about 0.5 eV. Low-temperature resistivity measurements of lightly doped EuO are shown in Fig. 6. The Hall effect was not measurable in these EuO samples at low temperature with available equipment and, if the effect exists, it puts an upper limit of about 2 cm^2/V-sec on the carrier mobility. After an initial drop the resistivity rises again with decreasing temperature. These data are also consistent with Kasuya's model in which the low temperature resistivity increase can be interpreted as due to impurity hopping; e.g., the condition $J \ll W$ is satisfied. As Mott[14] has pointed out, an apparent activation energy decrease with decreasing temperature is to be expected in this regime.

• *Magnetic polaron*

An even more qualitative argument which, in principle, gives an alternate description of many of the observed electrical effects involves the magnetic polaron.[4] The physical result, that of band electrons trapped in local spin clusters, is very similar to Kasuya's magnetic impurity state. But, whereas the mechanism for localization of the electron in Kasuya's model is the Coulomb attraction to the charged impurity, further stabilized by the exchange interaction with nearest neighbors, in the polaron model it is the exchange interaction alone. The high temperature data of Fig. 5 do not support the magnetic polaron model, at least for very low impurity concentrations. The application of the model also suffers in that a quantitative theoretical description is, so far, lacking. However, when $W \lesssim J \lesssim E$, e.g., for intermediate concentrations, but before the metallic description applies, the magnetic polaron may be an important effect. The photoconductivity data do not necessarily favor the magnetic polaron explanation since chalcogen vacancies provide impurity states in the undoped material.[23]

• *Localized impurity*

Finally, recent studies[26] of infrared absorption in doped EuO have been interpreted by the investigators as demonstrating that conduction occurs in a band at least 0.4 eV wide. They also suggest that the temperature dependence of the absorption and the sharp drop in resistivity below the Curie temperature in EuO are related to a carrier concentration change.[27] They consequently propose a model[28] of localized impurity levels below the conduction band edge in the paramagnetic region. As the temperature is lowered below the Curie temperature, the band splits and moves to lower energy, thereby sweeping the impurities into the band.

This model of band conduction is not consistent with the interpretation of the data of Fig. 6 in terms of hopping. The contradiction might be resolved, however, on the basis of impurity concentration. The samples used in the infrared work were generally of lower resistance than the samples of Figs. 5 and 6.

Summary

Although the scattering theory of simple metals accounts for the transport properties of chalcogenides containing impurities in excess of about 2×10^{20} cm^{-3}, the situation is as yet not nearly so clear for lower concentrations. When $J \lesssim W \ll E$, Kasuya's impurity hopping model applies and all the available data are consistent with the predictions. A final decision about the applicability of a particular model, however, must await an independent measurement of the temperature dependence, specifically below T_c, of the carrier concentration, the effective mass (polaron effects), or the drift mobility.

Acknowledgment

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