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Resistivity and Hall Effect of EuS in Fields up to 140 kOe†

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The dc electrical resistivity ρ and Hall effect were measured in n-type EuS single crystals. Data were taken from 300 to 2 K in magnetic fields H up to 140 kOe. At room temperature all samples had resistivities of the order 10^{-2} Ω cm and contained $n \sim 10^{19}$ conduction electrons/cm^3. Negative magnetoresistance was observed at all temperatures. At 77 K the negative magnetoresistance was due mostly to an increase in the mobility μ, although a small increase in n with increasing H was also observed. The zero-field resistivity as a function of temperature T exhibited a very large peak near the Curie temperature $T_C \cong 19$ K. The data are consistent with the interpretation that this peak was due largely to a decrease in μ. In the presence of a magnetic field the resistivity peak decreased in magnitude, became broader, and shifted to a higher temperature. A very large negative magnetoresistance was observed near T_C. At moderate fields this negative magnetoresistance was due largely to an increase in μ, but at high fields the changes in μ and n were comparable. Well below T_C, the resistivity and Hall coefficient exhibited hysteresis as a function of H, and were constant at fields above magnetic saturation. In the field-increasing part of the hysteresis cycle ρ decreased monotonically with time. A resistivity "elbow," similar to the one observed earlier in Eu-rich EuO, was observed also in the Eu-rich EuS samples. With increasing H the elbow shifted to higher temperatures. Hall-effect measurements at 4.2 K indicated that at fields above magnetic saturation the anomalous Hall term was small compared to the normal term. The Hall data also showed that a large part of both the hysteresis in ρ and the resistivity elbow were due to a change in n. The various data are compared with theoretical models and earlier electrical-transport measurements on the Eu chalcogenides.

I. INTRODUCTION

Magnetic semiconductors are materials which exhibit spontaneous magnetic order at low temperatures on one hand, and semiconductor characteristics on the other. In such materials the electrical transport properties are often strongly affected by the state of magnetic order, and some of the magnetic properties are influenced by the presence of charge carriers. In the last several years there has been a great deal of interest in magnetic semiconductors. The present knowledge in this field was summarized in two reviews by Methfessel and Mattis, [1] and by Haas. [2]

The europium chalcogenides (EuO, EuS, EuSe, and EuTe) form an important group of magnetic semiconductors. These compounds have the rock-

salt structure. EuO and EuS are ferromagnetic, while EuTe is antiferromagnetic. In EuSe the ferromagnetic and antiferromagnetic interactions are comparable, and the material exhibits several magnetic phases with different magnetic order. Pure and stoichiometric EuS has a Curie temperature $T_C = 16.3$ K.[3] However, the Curie temperature is higher when charge carriers are present.[1,4,5] The magnetic properties of pure and stoichiometric EuS are due to well-localized spins on the Eu^{++} ions. The interactions between the localized spins have been attributed to a ferromagnetic exchange interaction of each Eu^{++} ion with its 12 nearest neighbors, and to a weaker antiferromagnetic interaction with its six next nearest neighbors.[1,2] When a sample of EuS contains charge carriers there is an additional indirect exchange interaction between the Eu^{++} spins.

In all the Eu chalcogenides there is a strong interaction between charge carriers and the Eu^{++} ions. This interaction manifests itself in two ways. First, the magnetic properties are sensitive to the presence of charge carriers.[5-8] Second, and of greater interest here, the electrical-transport properties are strongly affected by the state of magnetic order. In the next few paragraphs we summarize some earlier experimental and theoretical results which are relevant to the present work.

Measurements of the electrical resistivity and Hall coefficient in the Eu chalcogenides have been carried out by several groups. Heikes and Chen[5] measured the resistivity of several lanthanum-doped samples of EuS as a function of temperature and observed a very large peak in the resistivity near T_C. Similar results were obtained by Methfessel *et al.*[9] in $Eu_{0.95}La_{0.05}S$. These workers also found that the magnitude of the resistivity peak was reduced considerably when magnetic fields up to 15 kOe were applied. A peak in the resistivity was also observed by von Molnar and Kasuya in gadolinium-doped EuS.[10,11] By analyzing their Hall data, in fields up to 12 kOe, these authors concluded that the resistivity peak was due mainly to a decrease in the mobility. Large peaks in the resistivity as a function of temperature were also observed in gadolinium-doped samples of EuSe.[6,12] Hall-effect measurements on $Eu_{0.95}Gd_{0.05}Se$ showed that the resistivity peak was not accompanied by an appreciable change in the concentration of charge carriers.[12]

An interpretation of the electrical transport measurements in EuS and EuSe was given by Kasuya, von Molnar, and co-workers. They distinguished between the case of low impurity concentration, where electrical conduction in the paramagnetic phase is accomplished by a hopping process, and the case of high impurity concentration with band conduction.[10,13,14] The first situation was discussed in detail by Kasuya and Yanase.[14] According to these workers, the interaction of a charge carrier near an impurity site with the neighboring Eu^{++} ions causes an alignment of the spins of these ions. This is called the magnetic impurity state (MIS). The properties of the MIS, which are influenced by temperature and magnetic field, have a strong effect on the activation energy of hopping from one impurity site to another.

According to von Molnar and co-workers, the change in the conduction process from hopping to band conduction occurs at impurity concentrations of the order of 1 at.%.[10,11,13] In the case of band conduction, one expects that the charge carriers will be scattered by spin fluctuations.[15] Near T_C this scattering mechanism should result in a resistivity peak.[2,16-18] A second mechanism which may lead to a resistivity peak near T_C is the scattering of those electrons which participate in band conduction by the spin clusters of the MIS.[10] This assumes that a fraction of the electrons are localized and form the MIS. In addition, magnetic polaron effects may be important near T_C.[13]

Studies of the resistivity in EuO revealed two separate phenomena.[19-23] At temperatures slightly above $T_C = 69$ K the zero-field resistivity $\rho(0, T)$ exhibited a peak. This peak was due mostly to a decrease in the mobility μ and not to a change in the concentration n of charge carriers. A second phenomenon was observed in EuO samples grown from Eu-rich solutions. As these samples were cooled below T_C, $\rho(0, T)$ decreased very rapidly (sometimes by many orders of magnitude) near 50 K. Oliver *et al.* called this phenomena the resistivity "elbow." Further studies showed that the resistivity elbow was due primarily to a change in n and not to a change in μ. To explain the change in n, Oliver *et al.* proposed a model which involves a trap level and a conduction-band edge whose energy decreases with magnetic order. Near 50 K the energy of the conduction-band edge drops below that of the trap level, which leads to an increase in the number of conduction electrons. A similar model was proposed earlier by Lehmann to explain his results in n-type $CdCr_2Se_4$.[24] It has been recently proposed that the trap level in EuO is an oxygen vacancy which traps two electrons.[21,23]

The present paper describes measurements of the dc resistivity and Hall effect in n-type EuS single crystals. The measurements were carried out from 2 to 300 K and in magnetic fields from 0 to 140 kOe. The results can be divided into three categories: (i) results which are similar to those observed earlier in EuS, e.g., the resistivity peak near T_C; (ii) results which were not observed earlier in EuS, but which are similar to those observed in other Eu chalcogenides, e.g., a resistivity "elbow"; (iii) results which were not observed earlier in any of the Eu chalcogenides,

e.g., hysteresis and time dependence of the resistivity at liquid-helium temperatures. Preliminary results of the present work appeared in an abbreviated form earlier.[25]

II. EXPERIMENTAL TECHNIQUE

Measurements were made on several samples which were obtained from four single crystals grown using techniques which were described earlier.[26] Two of the single crystals (Nos. 1 and 4) were grown from a nominally stoichiometric melt of EuS, whereas the others (Nos. 2 and 3) were grown from Eu-rich solutions of Eu and S. The two types of crystals will be called "nominally stoichiometric" and "Eu rich," respectively. None of the crystals was intentionally doped with impurities, but the presence of small amounts of such impurities cannot be overruled. The origin of the charge carriers in the various samples (i.e., whether they are due to nonstoichiometry, impurities, or various crystal defects) and their degree of compensation are not known. However, it is likely that at least a fraction of the charge carriers in the Eu-rich samples were due to the excess Eu.

All samples were rectangular parallelepipeds, prepared by cleaving larger single crystals. The faces of the samples were parallel to the {100} equivalent faces, and the dimensions of a typical sample were $1\times1\times4$ mm. Electrical contacts were made with indium solder using an ultrasonic soldering iron.

The procedures used in the electrical measurements, in the control and measurement of the temperature, and in the generation of magnetic fields, were very similar to those used in our earlier work on EuTe.[27] All measurements in the present work were carried out with the magnetic field $\vec{H}$ perpendicular to the direction of the electrical current (i.e., perpendicular to the long dimension of the sample). Corrections for the demagnetizing field were applied as in the case of EuTe. However, since EuS is ferromagnetic (unlike EuTe which is antiferromagnetic), the demagnetizing corrections were quite large in some cases. Much of the data at temperatures near T_C were taken with the samples immersed in liquid hydrogen.

III. RESULTS AND DISCUSSION

A. General Comments

Measurements of the dc resistivity $\rho(H, T)$ and Hall effect were carried out from 300 to 2 K in magnetic fields from 0 to 140 kOe. Many of the results indicate a strong correlation between the magnetic properties and the electrical transport properties. This correlation manifests itself most dramatically in the temperature variation of the zero-field resistivity $\rho(0, T)$, an example of which is shown in Fig. 1. The very large peak occurs either at or near T_C. A correlation between the magnetic properties and the electrical behavior is also found both at $T \gg T_C$ and $T < T_C$. The presentation below is divided into three parts corresponding to the three temperature ranges: $T \gg T_C$, $T \cong T_C$, and $T < T_C$. Each of these three ranges is associated with different phenomena.

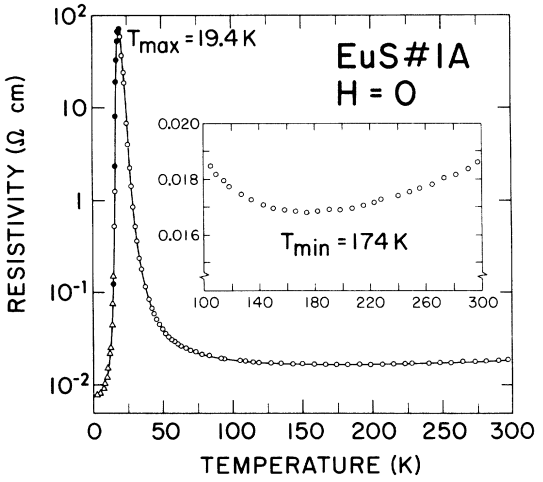

FIG. 1. Temperature variation of the zero-field resistivity $\rho(0, T)$ in sample 1A (semilog scale). The insert shows $\rho(0, T)$ vs T near the resistivity minimum (linear scale).

Several remarks should be made concerning the scope and limitations of the discussion of the experimental data. (a) The discussion consists essentially of a comparison of the data with available theoretical models and previous experimental results in the Eu chalcogenides. (b) While some of the experimental data agree with theoretical predictions, other data either remain totally unexplained or their interpretation is uncertain. (c) The art of preparing EuS single crystals has not reached the point where one has a good control over the concentrations of donors and acceptors. The lack of detailed knowledge of the types and concentrations of donors and acceptors, and the degree of compensation of each sample, makes the interpretation of the data still more difficult.

B. Results for $T \gg T_C$

1. Zero-Field Resistivity

The room-temperature resistivities of all samples were of order 10^{-2} Ω cm. Hall-effect measurements indicated that the samples were n-type, with $\sim 10^{19}$ conduction electrons/cm^3 at 300 K. The Hall mobilities at 300 K were about 30 cm^2/V sec, except for one sample with a mobility $\mu = 8$ cm^2/V sec.[28] The values for the mobilities at 300 K suggest that electrical conduction at this temperature was band conduction, rather than conduction by a hopping

TABLE I. Electrical properties of the various samples.[a]

Sample No.	1A	1B	2A	2B	2C	3	4A	4B
$\rho(0, 300\ K)$ Ω cm	1.9×10^{-2}	2.2×10^{-2}	1.6×10^{-1}	5.0×10^{-2}	2.9×10^{-2}	4.2×10^{-2}	1.7×10^{-2}	1.6×10^{-2}
R(100 kOe, 300 K) cm^3/C	-0.59	-0.66	-1.30	···	-1.11	-1.20	···	···
$n(300\ K)$ cm^{-3}	1.06×10^{19}	9.5×10^{18}	4.8×10^{18}	···	5.6×10^{18}	5.2×10^{18}	···	···
$\mu(300\ K)$ cm^2/V sec	31	30	8	···	38	29	···	···
$\rho(0, 77\ K)$ Ω cm	2.2×10^{-2}	2.3×10^{-2}	4.3×10^{-1}	1.1×10^{-1}	5.4×10^{-2}	4.9×10^{-1}	1.7×10^{-2}	1.5×10^{-2}
R_0(100 kOe, 77 K) cm^3/C	-0.58	-0.62	-1.53	···	-1.15	-1.43	···	-0.49
ρ(100 kOe, 4.2 K) Ω cm	3.7×10^{-3}	5.0×10^{-3}	1.7×10^{-2}	2.0×10^{-3}	1.4×10^{-3}	7×10^{-3}	4.6×10^{-3}	5.0×10^{-3}
R_0(100 kOe, 4.2 K) cm^3/C	-0.33	-0.39	-0.19	···	-0.15	···	···	-0.35
T_{max} K	19.4	···	···	18.9	18.8	19.4	···	···
ρ_{max} Ω cm	73	···	···	3.6×10^3	1.6×10^3	3.7×10^3	···	···

[a] $\rho(H, T)$ is the resistivity at the field H and temperature T. The accuracy of ρ varies between 25% for sample Nos. 1 and 4, and a factor of 2 for sample Nos. 2 and 3. The relative accuracy of ρ in a given sample, as a function of H and T, is much higher. $R(H, T)$ and $R_0(H, T)$ are the Hall coefficient and normal Hall coefficient, respectively. The accuracy of R and R_0 is about 5%. n is the carrier concentration at room temperature, derived from R. μ (300 K) is the room-temperature mobility. The accuracy of μ is limited by the uncertainties in ρ and R. T_{max} is the temperature where $\rho(0, T$ is maximum. ρ_{max} is $\rho(0, T_{max})$.

process. The room-temperature electrical characteristics of the samples are listed in Table I.

The temperature variation of the zero-field resistivity $\rho(0, T)$ was measured in samples 1A (nominally stoichiometric) and 2C (Eu rich) from 300 to 2 K. The results in both samples were qualitatively similar. The data for sample 1A are shown in Fig. 1. On cooling from room temperature, $\rho(0, T)$ first decreased, then went through a minimum, and finally increased at a rate which became progressively higher as T approached T_C. The resistivity minimum for sample 1A occurred at $T_{min} \cong 174$ K, as shown in the insert of Fig. 1. At the minimum, ρ was 10% lower than at 300 K. For sample 2C, $T_{min} \cong 198$ K, where ρ was 14% lower than at 300 K.

The positive value of $\partial\rho(0, T)/\partial T$ above T_{min} is explainable by the ordinary increase of the scattering of electrons by phonons with increasing T. However, as T decreases towards T_C, the influence of the magnetic ions on $\rho(0, T)$ becomes progressively stronger. Several mechanisms whereby the magnetic ions influence ρ will be described later. Below T_{min}, the interaction with the magnetic ions causes $\rho(0, T)$ to increase with decreasing T. The superposition of the effects of both the phonons and the magnetic ions on $\rho(0, T)$ leads to a resistivity minimum.

Values for $\rho(0, 77\ K)$ in the various samples are listed in Table I. In the nominally stoichiometric samples, $\rho(0, 77\ K)$ was approximately equal to $\rho(0, 300\ K)$, while in the Eu-rich samples $\rho(0, 77\ K)$ was significantly higher than $\rho(0, 300\ K)$. The Hall-effect data at 300 and 77 K are described below. These data, which were taken only at fields above several kOe, do not give directly the concentration n of charge carriers at $H=0$. However, the value of $n(H=0)$ can be estimated by extrapolating the data at finite H to $H=0$. On this basis we conclude that in the nominally stoichiometric samples $n(H=0)$ did not change appreciably between 300 and 77 K, whereas in the Eu-rich samples $n(H=0)$ was lower at 77 than at 300 K. However, this decrease in n accounts only for a part of the difference between $\rho(0, 77\ K)$ and $\rho(0, 300\ K)$. A comparable or a larger part of this difference was due to a change in the mobility.

2. *Magnetoresistance and Hall Effect*

A negative magnetoresistance was observed at room temperature in all samples which were mea-

sured. At 100 kOe the fractional change in the resistivity $\Delta\rho/\rho_0$ ranged from −10% in sample No. 3 to −3% in sample 4A. Larger magnetoresistance was observed at 77 K where at 100 kOe $\Delta\rho/\rho_0$ varied from −56% for sample No. 3 to −24% for samples 4A and 4B. The Eu-rich samples exhibited a larger negative magnetoresistance at high fields than the nominally stoichiometric samples, which actually had a small positive magnetoresistance at low fields. Figure 2 shows the results in sample 2C (Eu-rich) and in samples 1A and 4A (nominally stoichiometric).

Hall-effect measurements were carried out at room temperature and at 77 K. Some of the results at 100 kOe are summarized in Table I. At room temperature the Hall coefficient $R(H)$ was approximately independent of H and the experimental uncertainty in $R(H)$ was several percent, which is comparable to $\Delta\rho/\rho_0$ at the highest fields. For this reason it was not possible to separate the contribution to the magnetoresistance due to a change in μ from the contribution due to a change in n.

At 77 K the Hall coefficient showed a slight variation with the intensity of the applied field. The biggest variation was observed in the Eu-rich samples 2A, 2C, and 3 for which R decreased with increasing H. Some of the results for $R(H)$ at 77 K are shown in Fig. 3. Comparison of these data with the corresponding data for $\rho(H)$ in Fig. 2 indicates that ρ decreased considerably more rapidly with increasing H than R. This means that while n changed with H, the principal cause for the negative magnetoresistance was the increase in the mobility μ. This conclusion applies to all samples which were studied at 77 K.

The decrease of R with increasing H in the Eu-rich samples means (in a one-band model) that n increased with increasing alignment of the Eu^{++}

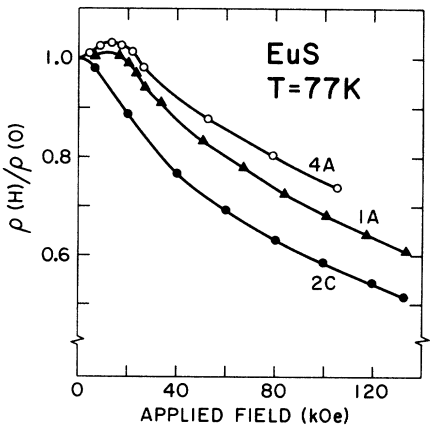

FIG. 2. Magnetic field variation of the resistivity of samples 1A, 2C, and 4A at 77 K.

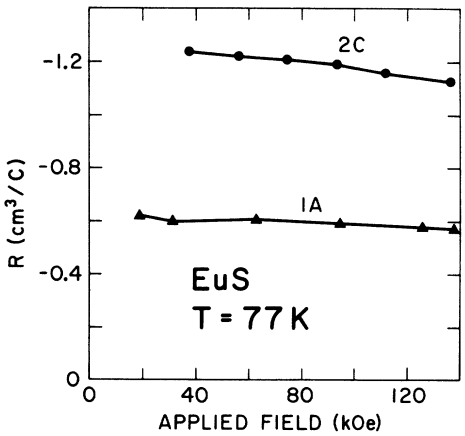

FIG. 3. H dependence of the Hall coefficient R of samples 1A and 2C at 77 K.

spins. This trend was more apparent at lower temperatures where a given magnetic field produced a larger alignment of the Eu^{++} spins. The exact cause of the H dependence of n is unknown. However, in general terms, the change in n apparently resulted from the redistribution of electrons between states which were in the conduction band (or, possibly, in an impurity band) and states which were localized near impurity sites. As is well known, both the spins of the conduction-band electrons and those of electrons localized near impurity sites are coupled to the spins of the Eu^{++} ions. An alignment of the Eu^{++} spins may therefore cause shifts in the energy levels of both the conduction and the localized electrons. In particular, a parallel alignment of the Eu^{++} spins causes a Zeeman splitting of the conduction band, which results in a lowering of the conduction-band edge. The variation of the energy of an impurity state with spin alignment depends on the nature of the impurity. The energy difference between a given localized electron and an electron in the conduction band should change with the alignment of the Eu^{++} spins, which leads to a redistribution of the electrons.

The principal contribution for the negative magnetoresistance at 77 K was the increase of μ with increasing H. The increase of μ with H was larger at lower temperatures. The exact mechanism of the increase in μ is not known. Some of the possible mechanisms are (i) a decrease in scattering due to spin fluctuations as the magnetic order increases with H and (ii) a decrease in the scattering by spin clusters associated with the MIS. As H increases the number of such clusters may decrease because some electrons move from localized states into the conduction (or impurity) band. Also, as H increases the difference between the spin alignment inside and outside the cluster decreases, which may lead to a weaker magnetic scattering.

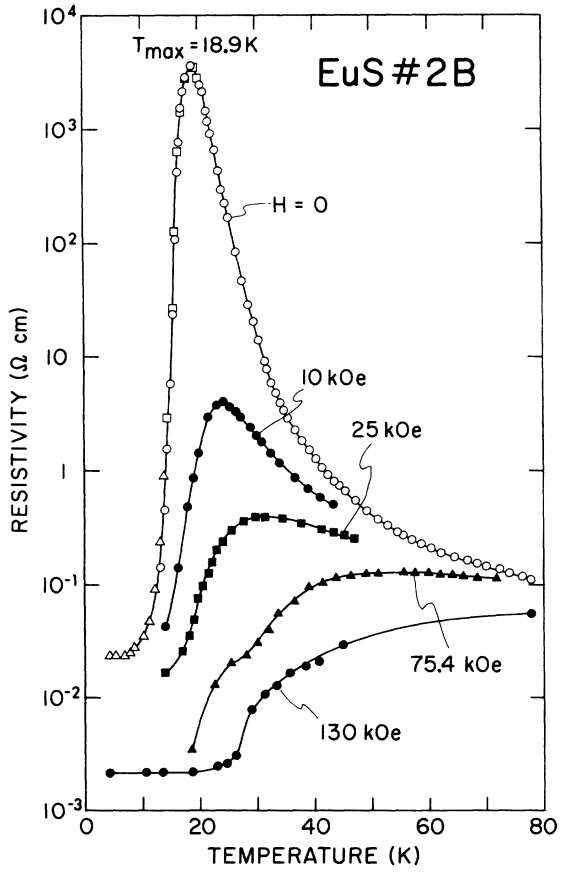

FIG. 4. Temperature variation of the resistivity of sample 2B at several fixed values of the applied magnetic field (semilog scale).

C. Resistivity Peak and Hall Effect near T_C

1. Resistivity Peak

One of the most prominent features of the resistivity data is the very large peak in $\rho(0, T)$ vs T at or near T_C. Figures 1 and 4 show this peak in a nominally stoichiometric sample and in a Eu-rich sample, respectively. A similar behavior was observed earlier by several workers.[5,9-11]

The resistivity peak in sample 1A occurred at $T_{max} = 19.4$ K. Ultrasonic-attenuation measurements on the same sample indicated that the Curie temperature T_C was at or close to T_{max}.[29] While insulating samples of EuS have a Curie temperature $T_C = 16.3$ K, higher values for T_C were reported in samples which contained charge carriers (Ref. 1, p. 514). Table I gives the values for T_{max} and the corresponding values of $\rho_{max} \equiv \rho(0, T_{max})$ in several samples. Note that ρ_{max} was orders of magnitude larger than the room-temperature resistivity.

A very large negative magnetoresistance was observed at temperatures near T_{max}. The magnetoresistance was studied by measuring ρ (a) as a function of T in several fixed fields, and (b) as a function of H at fixed temperatures. The results in sample 2B for ρ vs T (at several fixed fields) are shown in Fig. 4. Qualitatively, the magnetic field reduced the magnitude of the resistivity peak, broadened the peak, and shifted the peak to a higher temperature. Earlier measurements on $Eu_{0.95}La_{0.05}S$ in fields up to 14 kOe also showed a reduction of the resistivity peak with H, and a shift of the peak to higher temperatures.[9] Similar effects were also observed in EuSe doped with Gd,[12] and in EuO.[21] Note that in EuS the resistivity at 130 kOe increases rapidly near 28 K. This effect will be discussed later.

Measurements of ρ as a function of H at fixed temperatures near T_{max} were performed on various samples. The results for samples 1A and 2C are plotted on a semilog scale in Figs. 5 and 6, respectively. The large negative magnetoresistance shown in these figures is consistent with Fig. 4. The rate of decrease of ρ with increasing H, $|d\rho/dH|$, was larger at low fields than at high fields. Also, at the highest fields ρ appeared to approach saturation, although complete saturation was not achieved at 130 kOe.

Near T_{max} the resistivity in sample 2C decreased fairly abruptly with H at ~72 kOe (Fig. 6), whereas a similar effect was not observed in sample 1A (Fig. 5). The abrupt change of ρ at ~72 kOe was also observed in two other Eu-rich samples. This effect will be discussed further in Sec. III D.

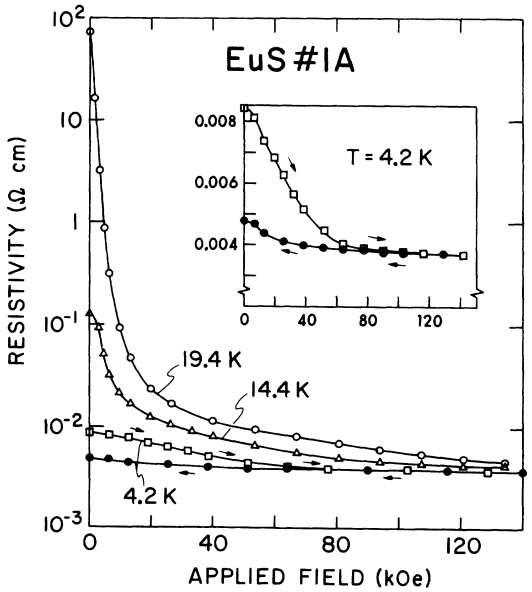

FIG. 5. Magnetic field variation of the resistivity of sample 1A at 19.4, 14.4 and 4.2 K (semilog scale). The insert shows the hysteresis at 4.2 K more clearly (linear scale).

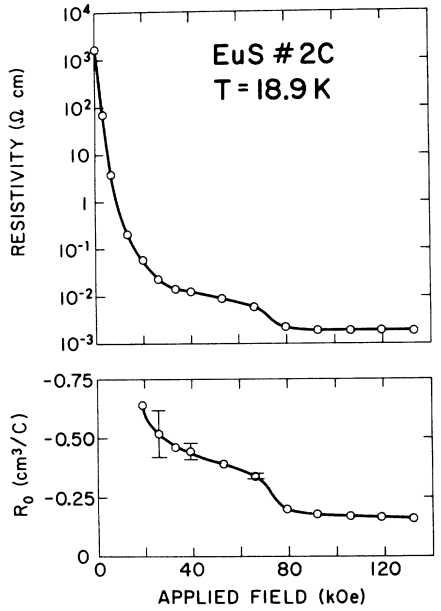

FIG. 6. H dependence of the resistivity and normal Hall coefficient of sample 2C at 18.9 K. Some typical uncertainties in R_0 are indicated by the error bars. These uncertainties become progressively smaller as the field increases.

2. *Hall Effect*

In interpreting the Hall data, we shall assume that electrical conduction is due to carriers in a single band. As is well known, the resistivity in a one-band model is related to the concentration n of conduction carriers and to the mobility μ by the relation

$$\rho = (ne\mu)^{-1}, \tag{1}$$

where e is the magnitude of the charge of the electron. Often, one tries to separate the contributions of n and μ to ρ by determining n from the Hall effect. In nonmagnetic materials the Hall voltage V_H is given by

$$V_H = RH_{ext}(I/s), \tag{2}$$

where R is the Hall coefficient, H_{ext} is the external (applied) magnetic field, I is the electric current through the sample, and s is the thickness of the sample along $\vec{H}$. In a one-band model $n = 1/|Re|$.

In magnetic materials the Hall voltage is often expressed as[30]

$$V_H = (R_0B + R_1M)(I/s), \tag{3}$$

where B is the magnetic induction inside the sample, and M is the magnetization. The coefficients R_0 and R_1 are the normal and anomalous Hall coefficients, respectively. In a one-band model $n = 1/|R_0e|$, so that Eq. (1) gives

$$\rho = R_0\mu^{-1}. \tag{4}$$

The coefficient R_0 cannot be determined uniquely from the measured Hall voltage V_H because the relative contributions of R_0B and R_1M to the right-hand side of Eq. (3) are unknown. The usual procedure in the case of a ferromagnet below T_C is to assume that R_0 and R_1 are field independent. At fields above magnetic saturation R_1M is then a constant, and R_0 is proportional to the slope of V_H vs applied field. This procedure was used by von Molnar and Kasuya who concluded that the resistivity peak in EuS was due mostly to a decrease in μ and not to a decrease in n.[10] However, the assumption that R_0 and R_1 are H independent is questionable in the case of EuS, in view of the large dependence of the transport properties on H. The data shown below are consistent with a marked dependence of R_0 on H. Thus it is unlikely that a value of R_0 which is obtained from data at fields above magnetic saturation (where ρ is much lower than at $H=0$) gives a reliable measure of n at $H=0$. In addition, magnetization measurements on the present samples (see, for example, Fig. 7) showed that even at $T \ll T_C$ the magnetization was saturated only at fields in excess of 75 kOe, which are much higher than the maximum field of 12 kOe used by von Molnar and Kasuya.

An alternative procedure for obtaining R_0 was used in the present study and in the earlier work on EuTe.[27] This procedure is based on experimental results at 4.2 K which show that at fields above magnetic saturation the term R_1M in Eq. (3) is small compared to R_0B. The Hall-effect data at 4.2 K will be presented below. These data also suggest (but do not prove) that at 4.2 K and at fields below magnetic saturation $R_1M \ll R_0B$. On this basis

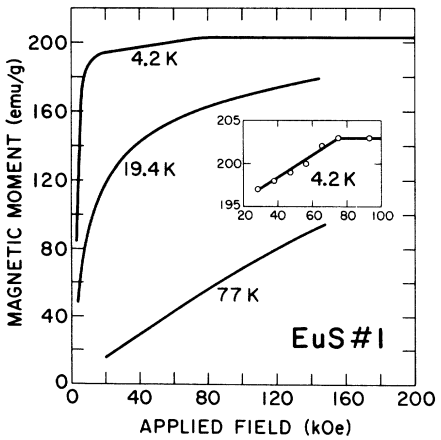

FIG. 7. Magnetization curves for sample No. 1 at 4.2, 19.4, and 77 K. The insert shows more clearly the approach to magnetic saturation at 4.2 K (after Foner and McNiff).

we assumed that $R_1 M \ll R_0 B$ at all temperatures and fields so that Eq. (3) is well approximated by

$$V_H = R_0 B(I/s). \quad (5)$$

All Hall-effect data were analyzed using Eq. (5). To obtain B from the external (applied) field we used the magnetization data of Foner and McNiff[31] (examples of which are shown in Fig. 7), and the appropriate demagnetizing factors for the various samples. At $T \gg T_C$, $M \ll H_{ext}$ so that the coefficient R_0 derived from Eq. (5) was nearly equal to the coefficient R obtained from Eq. (2). However, at $T \lesssim T_C$, and especially at $T \ll T_C$ and at low fields, the difference between R_0 and R was substantial.

Figure 8 shows R_0 vs H for sample 1A (nominally stoichiometric) at $T_{max} = 19.4$ K. The error bars show some typical experimental uncertainties at the lowest fields. The uncertainties at these fields were largest because the Hall voltage was small and the superimposed resistive voltage (due to the unintentional offset in the positions of the Hall leads) was large. Figure 9(a) shows the field dependence of ρ, R_0, and μ^{-1}, all normalized to their values at 130 kOe. The values of μ^{-1} were calculated using Eq. (4). Comparison of $R_0(H)$ and $\mu^{-1}(H)$ indicates that at $16 < H \lesssim 80$ kOe the negative magnetoresistance was due largely to an increase of μ with increasing H, while above ~80 kOe the fractional change in $n(H)$ was comparable to the fractional change in $\mu(H)$. Although reliable Hall data could not be obtained below ~15 kOe, extrapolation of the data at higher fields to the low-field region suggests that the large negative magnetoresistance at low fields was due largely to a variation in μ.

The H dependence of R_0 for sample 2C (Eu rich) near T_C is shown in the bottom part of Fig. 6. The

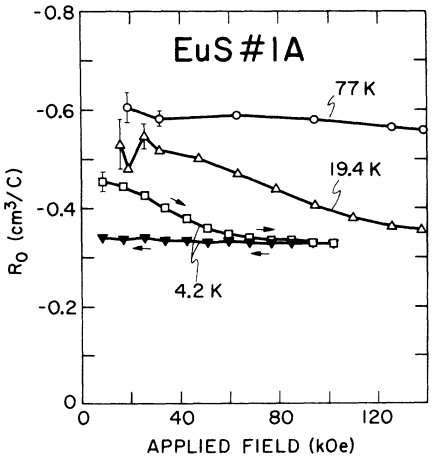

FIG. 8. H dependence of R_0 in sample 1A at 4.2, 19.4, and 77 K. The uncertainties at the lowest fields are indicated by error bars. The uncertainties at higher fields are much smaller. Note the hysteresis at 4.2 K.

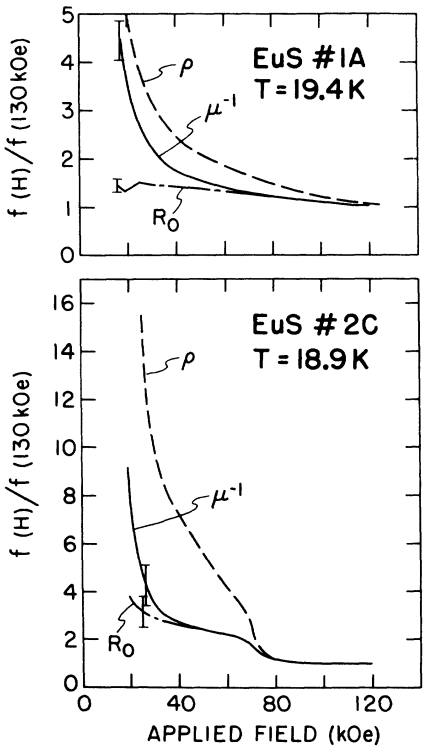

FIG. 9. H dependence of ρ, R_0, and μ^{-1} in samples 1A and 2C at a temperature near T_{max}. The various functions $f = \rho$, R_0, μ^{-1} were normalized to their values at 130 kOe. The uncertainties at the lowest fields are indicated by bars. The uncertainties at higher fields are much smaller.

data indicate a monotonic increase of n with increasing H. Near 72 kOe, where ρ decreases rapidly, R_0 also decreases rapidly. The contributions of n (or R_0) and μ to the magnetoresistance are shown in Fig. 9(b). Below 40 kOe the mobility contribution to the negative magnetoresistance appears to dominate, although the large uncertainty in R_0 below ~30 kOe makes this conclusion somewhat uncertain. Above ~40 kOe, $\mu^{-1}(H)$ and $R_0(H)$ change at approximately the same rate. Hall-effect measurements at 18.8 K on another Eu-rich sample (No. 3) showed a similar dependence of R_0 on H. The results also indicate that in the Eu-rich samples 2C and 3, the carrier concentration varied more strongly with H than in the nominally stoichiometric sample 1A.

3. *Discussion*

A peak in the zero-field resistivity was observed earlier in all the Eu chalcogenides which are ferromagnetic, namely, EuO,[19–21] EuS,[5,9–11] and Gd-doped EuSe.[12] The first question which arises in connection with this peak is whether it is due primarily to a sharp decrease in the mobility μ or to a sharp decrease in the concentration of charge carriers. In earlier studies it was concluded on the basis of Hall-

effect measurements[10,12] and optical measurements[21] that the peak in $\rho(0, T)$ was due primarily to a decrease in μ. In the present work the measurements of R_0 at temperatures near T_{max} did not extend to fields below ~ 15 kOe. Therefore, we cannot separate the contributions of μ and n to the *zero-field* resistivity near T_{max}. However, the experimental evidence discussed below is consistent with the conclusion that the peak in $\rho(0, T)$ is due mostly to a decrease in μ.

Results for the variation of $\mu(H, T)$ with T are limited to fields where Hall-effect data are available. The lowest field H_L for which reliable Hall-effect data were obtained near T_{max} was ~ 20 kOe for sample 2C, and ~ 15 kOe for sample 1A (see Figs. 6 and 8). An analysis of the results for $\rho(H_L, T)$ and $R_0(H_L, T)$ as a function of T indicates that the large decrease in $\rho(H_L, T)$ between T_{max} and 4.2 K was primarily due to an increase in μ and only secondarily to an increase in n. This suggests that the same was true for the change in $\rho(0, T)$ between T_{max} and 4.2 K.

Further evidence for the low zero-field mobility at temperatures near T_{max} is given by the H dependence of ρ and R_0. Although Hall-effect data at very low fields are not available, the curves in Fig. 9 suggest that the total rise in ρ as H changed from 130 kOe to zero was largely due to a decrease in μ.

Several mechanisms which may contribute to a resistivity peak near T_C have been discussed in the literature. (i) Critical scattering of electrons by spin fluctuations was considered by de Gennes and Friedel[16] and by others.[2,17,18] This scattering decreases the mobility of the electrons which participate in band conduction. (ii) In the case where some electrons participate in band conduction while others participate in the formation of the MIS, the scattering of the band electrons by the spin clusters of the MIS should increase near T_C.[10,11] In addition, the probability of forming a MIS may also increase near T_C, so that some band electrons may become localized near T_C. This will have the effect of both decreasing the number of electrons which participate in band conduction and increasing the scattering of those electrons which remain the band. (iii) Electrical conduction by electrons associated with the MIS was considered by Kasuya and Yanese.[14] Conduction in this case is carried out by a hopping process with an activation energy which is temperature dependent and which decreases rapidly below T_C. (iv) The possibility of forming magnetic polarons near T_C was discussed by von Molnar and Kasuya.[13] The relative contributions of these various mechanisms to the resistivity peak observed in the present study are uncertain. It is also not known whether in addition to these four mechanisms there are others which contribute to the resistivity peak.

The effect of a magnetic field on the resistivity peak is shown in Fig. 4. These data are qualitatively similar to results obtained by Oliver *et al.* in EuO.[21] In the presence of a low or an intermediate field the resistivity peak is reduced, becomes broader, and moves to a higher temperature. The rate at which the magnitude of the peak is reduced with H is largest at low fields. Thus a field of 10 kOe reduces the peak in sample 2B by three orders of magnitude, while a further increase of H to 25 kOe causes an additional reduction of only one order of magnitude. The large decrease in ρ caused by the application of a low or a moderate magnetic field near T_C appears to be due largely to a field-induced increase in the mobility, although a decrease in R_0 is also observed (see Fig. 9).

Near T_{max} both ρ and R_0 appear to approach saturation at the highest magnetic fields. This approach to saturation is apparently related to the fact that the magnetization at T_{max} is approximately 90% saturated at 140 kOe (see Fig. 7). The saturation of ρ and R_0 at high fields is more clearly observed at $T \ll T_C$. However, already at temperatures near T_{max} the high-field value of R_0 is significantly lower than (i) the low-field value of R_0 near T_{max}, and (ii) the values of R_0 at 300 or 77 K (compare the values for R_0 in Figs. 6 and 8 and the values at 77 and 300 K in Table I). This means that a substantial fraction of the electrons which participate in band conduction when the magnetization is saturated (or nearly saturated) do not participate in band conduction when the magnetization is far below saturation. This aspect is discussed further in Sec. III D in connection with the resistivity elbow.

D. Hysteresis, Time-Dependent Resistivity, and Resistivity Elbow at $T < T_C$

Measurements at temperatures well below T_C revealed three experimentally interrelated phenomena: hysteresis in ρ and R_0 as a function of H, time dependence of ρ, and a resistivity "elbow."

1. Hysteresis in ρ and R_0

Hysteresis in ρ as a function of H was first observed in sample 1A at 4.2 K. After cooling the sample to 4.2 K at $H = 0$, the magnetic field was increased from 0 to 140 kOe. The increase in H caused a monotonic decrease in ρ. Subsequently, when the field was decreased gradually from 140 kOe to zero, the dependence of ρ on H was markedly different from the dependence observed when H was first increased from zero. These results are shown in the insert of Fig. 5. The hysteresis was observed only below ~ 75 kOe. At these fields ρ was higher when H first increased from zero than when H decreased from high fields. In the following discussion the hysteresis cycle will be divided into two parts. The part where H increases from zero to high field will be referred to as the $H\uparrow$ part of

the hysteresis cycle. The part where H decreases from high fields towards zero will be called the $H\downarrow$ part. Note that the symbols $H\uparrow$ and $H\downarrow$ do not indicate opposite directions of the applied field.

Hysteresis in ρ was observed only in the first field up-down cycle after the sample was cooled to liquid-helium temperatures at $H=0$. Once one field cycle was completed, the behavior of $\rho(H)$ in subsequent cycles (for both increasing and decreasing fields) was as in the $H\downarrow$ part of the first cycle. Also, the results in subsequent cycles did not depend on whether the field was oriented along the same direction as in the first cycle or along the opposite direction. Following the first hysteresis cycle, a second equivalent cycle in $\rho(H)$ could be observed only by first warming the sample above ~10 K and then cooling it back to liquid-helium temperatures at $H=0$. The hysteresis in $\rho(H)$ was larger in the Eu-rich samples than in the nominally stoichiometric samples. Results for one of the Eu-rich samples are shown in Fig. 10.

As mentioned above, hysteresis in the resistivity of sample 1A at 4.2 K was observed only at fields below ~75 kOe. Measurements of the magnetization $M(H)$ of the same sample at 4.2 K were carried out by Foner and McNiff.[31] Their results, which are shown in Fig. 7, indicate that $M(H)$ became saturated only at ~75 kOe, which is an unusually high saturation field for a ferromagnet with low anisotropy at $T \ll T_C$. No hystersis in $M(H)$ was observed in this sample.

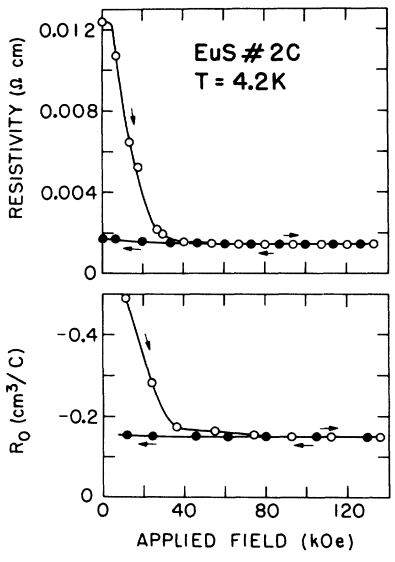

FIG. 10. H dependence of ρ and R_0 in sample 2C at 4.2 K. These results were obtained after the sample was cooled to 4.2 K at $H=0$. The data in the $H\uparrow$ part of the hysteresis cycle were taken 5 min after the magnetic field reached a given fixed value.

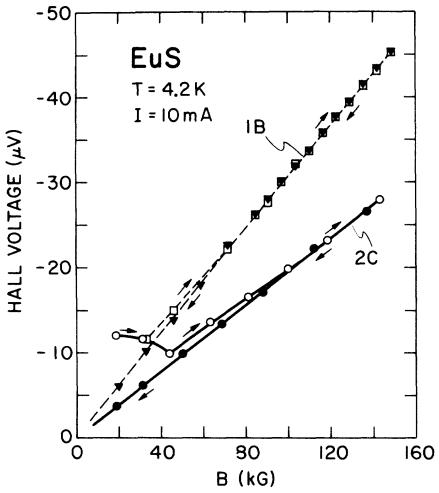

FIG. 11. Hall voltage in samples 1B and 2C as a function of the magnetic induction B at 4.2 K. These data show the hysteresis cycle which is observed after cooling the sample to 4.2 K at $H=0$.

Hall-effect data at 4.2 K were taken on samples 1A, 1B, 2A, 2C, and 4B. The Hall voltage of samples 1B and 2C as a function of the magnetic induction B inside the sample is shown in Fig. 11. Three features should be noted:

(a) The Hall voltage as a function of B exhibited hysteresis. This hysteresis was particularly pronounced in sample 2C at $B \lesssim 40$ kG. The resistivity of this sample also had a large hysteresis in the same range of magnetic field (Fig. 10).

(b) The Hall voltage at fields above magnetic saturation ($B \gtrsim 85$ kG) was approximately proportional to B. The data for V_H vs B at these high fields were fitted to Eq. (3) using the least-squares method and assuming that R_0 and R_1 were field independent *at fields above magnetic saturation*. The fits showed that in both samples 1B and 2C, R_1M/R_0B was less than 5% in fields $B \gtrsim 85$ kG. Similar fits for samples 1A and 2A at fields above magnetic saturation gave R_1M/R_0B less than 7% and less than 3%, respectively. The Hall-effect data in sample 4B were not extensive enough to warrant a least-squares fit, but they showed that the hysteresis in V_H was less than 3%, and that V_H was nearly proportional to B at all fields. The fact that in all five samples R_1M was very small compared to R_0B at fields above magnetic saturation is the basis on which we assumed that Eq. (3) can be approximated by Eq. (5).

(c) In all five samples which were studied, the Hall voltage in the $H\downarrow$ part of the hysteresis cycle was proportional to B at all fields, including fields below magnetic saturation. The simplest interpretation of this experimental result is that

(i) V_H is well described by Eq. (5) (i.e., $R_1M \ll R_0B$ at all fields), and (ii) R_0 is independent of B, so that n is field independent in the $H\downarrow$ part of the cycle. The alternative interpretation is that (i) Eq. (5) is invalid at fields below magnetic saturation (i.e., R_1M is not very small compared to R_0B), and (ii) both R_0 and R_1 vary with B, but in such a way that V_H remains proportional to B at all fields. We regard this alternative interpretation as implausible.

The H dependence of R_0 at 4.2 K for samples 1A and 2C is shown in Figs. 8 and 10, respectively. The values for R_0 were obtained using Eq. (5). The results indicate that in the $H\uparrow$ part of the hysteresis cycle n increases monotonically with H, and that n assumes a constant value at fields above magnetic saturation. In the $H\downarrow$ part of the cycle n is field independent and is higher than in the $H\uparrow$ part of the cycle. The data for $\rho(H)$ and $R_0(H)$ show that in sample 1A (nominally stoichiometric) the hysteresis in ρ is due to comparable changes in both n and μ. However, in sample 2C (Eu rich) the hysteresis in ρ is largely due to a hysteresis in n, with a smaller contribution due to a hysteresis in μ.

2. *Time Dependence of ρ*

Another phenomenon associated with the hysteresis cycle is the time dependence of ρ. This phenomenon was observed only in the $H\uparrow$ part of the first field cycle, after cooling at $H = 0$ to liquid-helium temperatures. In this part of the first cycle, the resistivity at a fixed value of H decreased monotonically with time t. In some cases, the time dependence of ρ was studied for more than 1 h. The rate at which ρ decreased with t became progressively slower as t increased. The function $\rho(t)$, for a fixed H, could not be described by a simple exponential function involving one time constant. A time dependence of ρ was observed only at low and intermediate fields where the resistivity in the $H\uparrow$ part of the cycle was higher than in the $H\downarrow$ part.

Figure 12(a) shows some results for $\rho(t)$ in the $H\uparrow$ part of the hysteresis cycle. These data were obtained on sample 2C at two fields and at two temperatures. Note that at 17.4 kOe the resistivity was still changing appreciably after 1 h. Note also that for small t, $|d\ln\rho/dt|$ was larger at the higher field and at the higher temperature. Figure 12(b) shows in greater detail the variation of ρ in the first 15 min at 4.2 K. This figure also shows the results, at the same fields, in the $H\downarrow$ part of the hysteresis cycle where ρ was independent of t. At 29.6 kOe, the resistivity in the $H\uparrow$ part of the cycle seemed to approach the resistivity in the $H\downarrow$ part asymptotically with time, although a difference of several percent still remained after 70 min.

The results in Fig. 12 need some clarification. The data for a given fixed field H_0 were taken immediately after H was increased from 0 to H_0. Typically this increase in field took about 30 sec, but this time varied by as much as a factor of 2 from run to run. The time t was always measured from the instant at which H reached the value H_0. The data for $\rho(t)$ obtained in this manner depended on the speed S at which the field was increased to H_0. This dependence of $\rho(t)$ on S was most pronounced at times near $t = 0$, where $d\rho/dt$ was largest. However, for $t \gg 1$ min, $\rho(t)$ was not sensitive to a small change in S, because in all cases $\rho(t)$ did not change appreciably in ~30 sec at times $t \gg 1$ min.

Due to the time dependence, ρ was not uniquely determined by H in the $H\uparrow$ part of the hysteresis

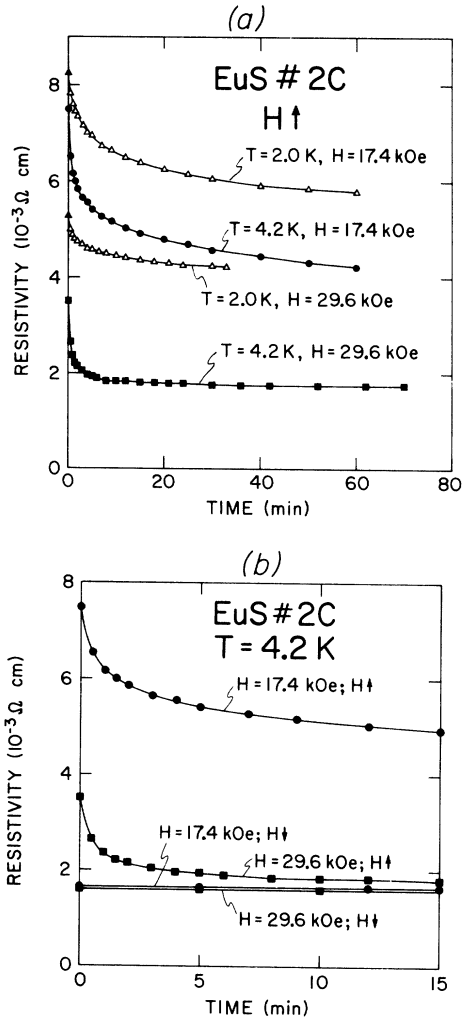

FIG. 12. Time dependence of ρ in sample 2C. (a) Results for the first hour in the $H\uparrow$ part of the hysteresis cycle. These data were taken at both 4.2 and 2.0 K in fixed fields of 17.4 and 29.6 kOe. (b) Results at 4.2 for the first 15 min in both the $H\uparrow$ and $H\downarrow$ parts of the hysteresis cycle.

cycle. To obtain consistent resistivity data which could be compared with Hall-effect data, we chose to take both types of data at $t = 5$ min. This procedure was used in obtaining results such as those shown in Fig. 10.

The time dependence of ρ in the $H\uparrow$ part of the hysteresis cycle suggests that in this part of the cycle the sample is in a metastable state. This higher-resistvity metastable state drifts with time towards a lower-resistivity state. At fields above magnetic saturation the sample reaches a stable state with a lower ρ and a higher n. The stable state remains as H is decreased isothermally back to zero. To obtain the metastable state again, it is necessary to warm the sample above ~ 10 K and then cool it back to liquid-helium temperatures at zero or low magnetic field.

The origin of the metastable state is unknown. It is possible that it is related to the presence of domain walls. However, the persistence of the metastable state to fields as high as 75 kOe makes this explanation unlikely, although it does not rule it out completely. Ultrasonic measurements, with shear waves, did not reveal any strong attenuation due to domains, except at fields below ~ 10 kOe.[29] Thus the question of the origin of the metastable state remains open.

3. *Resistivity Elbow*

A third phenomenon observed at $T \lesssim T_C$ is the resistivity "elbow." The term "elbow" was used by Oliver *et al.* to describe the large sharp increase in ρ with increasing T in EuO.[19–21] The same phenomenon has also been referred to as a metal-insulator transition.[23] In EuO the resistivity elbow occurs at ~ 50 K, which is well below T_C, and the phenomena associated with the elbow are easily separated from the phenomena associated with the resistivity peak near T_C. It has been suggested by Petrich *et al.* that a resistivity elbow should not occur in EuS, in agreement with their resistivity data.[23]

In the present work, the first measurements of the resistivity at temperatures below T_C were carried out in the absence of a magnetic field at any phase of the measurements. These early data for $\rho(0, T)$ vs T did not reveal a resistivity elbow. However, after observing the hysteresis in $\rho(H)$ at 4.2 K, new measurements of $\rho(0, T)$ were performed. In these measurements a high magnetic field was first applied at 4.2 K and was then reduced back to zero. The zero-field resistivity was subsequently measured as a function of increasing temperature. A clear elbow in $\rho(0, T)$ was observed at ~ 9 K. The results for sample 2C are shown in Fig. 13. The dashed curve in this figure represents $\rho(0, T)$ as measured by cooling from high temperatures at $H = 0$, while the solid curve marked by $H = 0$ was

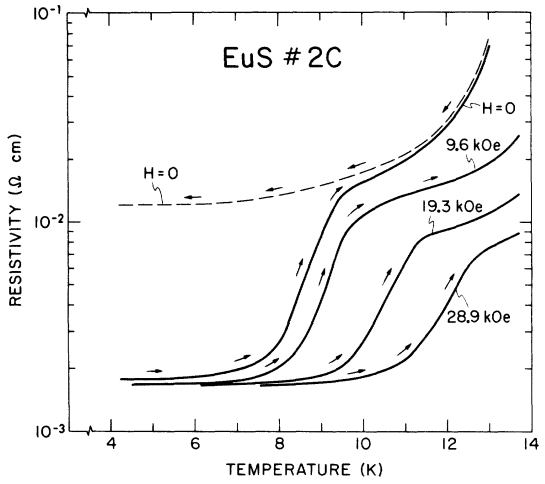

FIG. 13. Resistivity of sample 2C as a function of T at various fields. The dashed curve shows data obtained by cooling at $H = 0$. The solid curves show data obtained by warming at a fixed H after an 83-kOe field was first applied at 4.2 K and then reduced to the given fixed value.

obtained by warming the sample from 4.2 K after an 83-kOe field was applied at 4.2 K and then removed. This solid curve shows an elbow at ~ 9 K. The magnitude of the resistivity elbow at $H = 0$ was approximately equal to the difference between the zero-field resistivities at 4.2 K before and after a high field was applied. Since the hysteresis in $\rho(H)$ at 4.2 K was much larger in the Eu-rich samples, all studies on the resistivity elbow were carried out on these samples.

The possibility that the shape of the resistivity elbow at $H = 0$ was influenced by a time dependence of ρ was studied. It was found that below 7 K and above 10 K the resistivity did not change by any measurable amount during 5 min. However, between 8 and 9 K (i.e., at the elbow) ρ increased slowly with time. Thus the exact shape of the resistivity elbow at $H = 0$ depended on the speed at which data were taken as the sample was warming.

The remaining solid curves in Fig. 13 show data obtained by first applying an 83 kOe field at 4.2 K, then reducing it isothermally to a fixed lower value (9.6, 19.3, and 28.9 kOe), and finally warming the sample at the fixed field. Note that the resistivity elbow shifted to higher temperatures as H increased. For samples 2B and 2C the elbow at an external field of 130 kOe occurred near 28 K (see the curve for 130 kOe in Fig. 4).

The data for $\rho(0, T)$ depended very strongly on whether these data were obtained by cooling at $H = 0$ or by warming from liquid-helium temperatures after a high magnetic field was first applied and then removed. However, as the intensity of

the magnetic field increased, the data for $\rho(H, T)$ as measured by these two procedures (cooling at a fixed H, or warming at the same H after a high field was first applied) became progressively closer to each other. As a consequence, at moderate and high fields the resistivity elbow could also be observed when the sample was cooled at a fixed field. Also, the shape of the resistivity elbow at moderate or high fields did not depend on the speed at which the temperature was changing near the elbow.

The resistivity elbow was also observed by measuring ρ as a function of H at a fixed temperature. These measurements were performed in liquid hydrogen. A recorder tracing of the elbow is shown in Fig. 14 (see also Fig. 6). Figure 6 indicates that the normal Hall coefficient $R_0(H)$ also had an "elbow."

The H dependence of the temperature T_e of the resistivity elbow was studied in fields up to 130 kOe. $T_e(H)$ was defined as the midpoint in the elbow of $\log\rho(H, T)$ vs T, for a fixed H. Similarly, the field $H_e(T)$ at the elbow was defined as the midpoint in the elbow of $\log\rho(H, T)$ vs H, for a fixed T. The dependence of T_e on H (or H_e on T) in sample 2C is shown in Fig. 15. Measurements of H_e as a function of T were also carried out in sample No. 3 at liquid-hydrogen temperatures. The results for $H_e(T)$ were close to those in sample 2C. $H_e(T)$ were close to those in sample 2C.

The resistivity elbow in EuO was discussed in detail by Oliver *et al.*[19–21] and by Petrich *et al.*[23] Both groups employed a similar model which involves a trap level and a conduction-band edge whose energy decreases with magnetic order. According to this model, the mechanism for the resistivity elbow at $H = 0$ is as follows. As T decreases below T_C, the ferromagnetic alignment of the Eu^{++} spins increases gradually. At a certain temperature (~ 50 K in EuO) the energy of the conduction-band edge becomes lower than that of the trap level. When this happens, electrons which were previously trapped move into the conduction band. The increase in the number of electrons in the conduction band causes ρ to decrease. Thus in this model the resistivity elbow is due primarily to a change in n, in agreement with the available

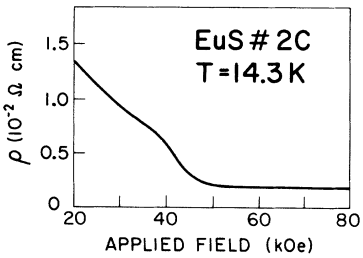

FIG. 14. Recorder tracing of the resistivity of sample 2C as a function of applied magnetic field at 14.3 K. Note the "elbow" at ~ 43 kOe.

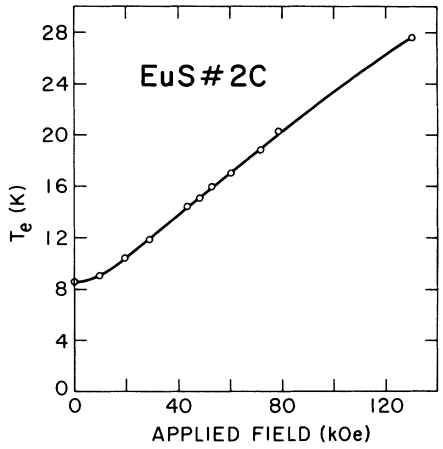

FIG. 15. Dependence of the temperature T_e at the resistivity elbow on applied magnetic field in sample 2C.

experimental data in EuO. The model also predicts that the resistivity elbow will move to higher temperatures as H increases.[21]

The model used by Oliver *et al.* indicates that the behavior of ρ at temperatures well above the elbow depends strongly on whether the number of traps is larger or smaller than the number of electrons. If the number of traps is larger, conduction at high temperatures is due to electrons which are thermally excited from the trap level to the conduction band. If the number of traps is smaller than the number of electrons, conduction at high temperatures is due largely to the excess electrons which always remain in the conduction band. It appears that in the present samples the number of traps was smaller than the number of electrons.

The present experimental data for the resistivity elbow in EuS are similar to those in EuO, with a few notable exceptions. In EuO the resistivity elbow can be observed by cooling the sample at $H = 0$, whereas in EuS the elbow cannot be observed by this procedure (except, possibly, if the cooling process is very slow). On cooling EuS at $H = 0$ to liquid-helium temperatures one apparently obtains a metastable state. This metastable state gives rise to hysteresis and time dependence in ρ, which are phenomena not observed in EuO. However, aside from those features which are associated with the metastable state, the data on the elbow in EuS bear a close resemblance to those in EuO.

The Hall-effect data in EuS indicate that the resistivity elbow is accompanied by an appreciable change in n. Thus the constant value of R_0 in the $H\downarrow$ part of the hysteresis cycle at 4.2 K is considerably lower than (i) the values of R_0 at 77 and 300 K, and (ii) the low-field value of R_0 near T_{max} (see Figs. 6 and 10 and Table I). The elbow in

$R_0(H)$ vs H at liquid-hydrogen temperatures also shows that the resistivity elbow is associated with a change in n (see Fig. 6). These observations are consistent with the model used to explain the resistivity elbow in EuO.[19-21,23] At present this model provides the only available explanation of the resistivity elbow.

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[28]The lower value for μ in sample 2A may be due to a systematic error in the measurement of ρ. Such a systematic error could result, for example, from an internal cleavage inside the sample.

[29]The ultrasonic data in EuS were qualitatively similar to the data in EuO [See, Y. Shapira and T. B. Reed, J. Appl. Phys. 40, 1197 (1969)]. In EuO, the ultrasonic data gave the correct value for T_c.

[30]In the more usual expression for V_H in magnetic materials, the magnetic induction B which appears in Eq. (3) is replaced by the internal field H_{int}. However, the form of Eq. (3) used here is more convenient because the data at fields above magnetic saturation shows that $R_1 M$ is very small compared to $R_0 B$ (see also Ref. 27). Moreover, from the theory one expects that the effective field which acts on the electron is $\vec{B}$ [see, C. Kittel, Phys. Rev. Letters 10, 339 (1963)].

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