EuO. II. Dependence of the Insulator-Metal Transition on Magnetic Order

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Three previously considered relations for the dependence of the activation energy Δ on magnetic order, in "activated" EuO samples, are tested. These relations are: (i) $\Delta = \Delta_0 - (1 + \alpha)\delta_R$, where $\Delta_0 \equiv \Delta(T \gg T_C, H = 0)$, δ_R is the red shift of the optical absorption edge due to magnetic order, and α is a constant; (ii) $\Delta = \Delta_0(1 - a\sigma)$, where σ is the reduced magnetization and a is a constant; (iii) $\Delta = \Delta_0(1 - b\eta)$, where η is the nearest-neighbor two-spin correlation function and b is a constant. The resistivity and Hall data in the preceding paper, and additional Hall data, are analyzed in detail and compared with these relations. New high-field magnetization and optical red-shift data are presented and used in this comparison. The analysis includes the H dependence of the Hall coefficient at $T \gg T_C$, which is sensitive to the dependence of Δ on magnetic order. It is shown that neither relation (ii) nor relation (iii) represents the data accurately. Penney, Shafer, and Torrance proposed relation (ii) on the basis of more limited data. Relation (i) was tested only at 297 K. At this temperature relation (i) agreed with the H dependence of the Hall coefficient. More extensive measurements of $\delta_R(H, T)$ and further analysis are necessary to test the validity of relation (i).

I. INTRODUCTION

The general features of the insulator-metal transition (IMT) in EuO and the basic model for this transition were discussed in the preceding paper.[1] In the present paper we analyze the results for the *activated* samples in detail. For these samples the carrier concentration n is determined primarily by the activation energy Δ, which is related to the energy separation ϵ between the trap level and the edge of the lowest conduction band. When $\Delta \gg kT$, n obeys the relation

$$n = N(T)e^{-\Delta/kT}, \tag{1}$$

where $N(T)$ is a slowly varying function of T. The temperature and magnetic field variation of n (and to a large extent also of the resistivity ρ) is governed therefore by the dependence of Δ on T and H.

All available experimental data show that Δ decreases with increasing magnetic order, but the detailed dependence of Δ on magnetic order is a matter of controversy. Oliver *et al.*[2] assumed that Δ varied linearly with the optical red shift δ_R. Later, Penney, Shafer and Torrance[3] (PST), and Holtzberg *et al.*[4] argued that Δ was not linear with δ_R, but, instead, was linear with the magnetization M. PST also concluded that Δ did not vary linearly with the nearest-neighbor two-spin correlation function $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$.

In the present paper we analyze our data for $\rho(H, T)$ and the ordinary Hall coefficient $R_0(H, T)$ and determine the dependence of Δ on H and T. We then examine whether Δ varies linearly with M, or with $\langle \vec{S}_1 \cdot \vec{S}_2 \rangle$, or with δ_R. The earlier analysis of PST is discussed in detail. We find that their conclusion concerning the linear dependence of Δ on M does not hold in general.

II. PROPOSED RELATIONS BETWEEN Δ AND MAGNETIC ORDER

According to Oliver *et al.*,[2] Δ should vary linearly with the optical red shift δ_R, i.e.,

$$\Delta = \Delta_0 - (1 + \alpha)\delta_R, \tag{2}$$

where $\Delta_0 = \Delta(T \gg T_C, H = 0)$, and α is a constant. The reasoning behind Eq. (2) is as follows. The optical transition is assumed to be from the $4f^7$ level to the lowest conduction band. The red shift δ_R in this picture represents the downward energy shift of the edge of the lowest conduction band. In addition, the energy of the trap level is assumed to vary linearly with δ_R. It then follows that the energy separation ϵ between the conduction-band edge and trap level is linear in δ_R. The activation energy Δ should be equal to ϵ or $\frac{1}{2}\epsilon$, depending on whether or not the sample contains a sufficient number of compensating acceptors. In either case one obtains an equation for Δ which has the form of Eq. (2). If $\Delta = \epsilon$ then $\alpha\delta_R$ is the energy increase of the trap level due to magnetic order. If $\Delta = \frac{1}{2}\epsilon$ then the shift of the trap level is $(1 + 2\alpha)\,\delta_R$. PST[3] have argued, in a different context not involving Eq. (2), that $\Delta = \epsilon$ for EuO samples with $\Delta_0 \cong 0.3$ eV.

The assumptions on which Eq. (2) are based are not universally accepted. According to Kasuya and Yanase[5,6] the lowest conduction band is the 6s band. Moreover, Kasuya,[6] and also Freiser *et al.*,[7] have argued that the final state of the optical transition involves an excitonlike 5*d* state. In this interpretation the red shift does not correspond to the shift of

the edge of the lowest conduction band.

The dependence of Δ on magnetic order was examined by PST,[3,4] who concluded that Δ is linear in the magnetization M (in a single domain), i.e.,

$$\Delta = \Delta_0(1 - a\sigma), \tag{3}$$

where $\sigma = M/M_0 = \langle S\rangle/S$ is the reduced magnetization, and a is a constant approximately equal to 1.3. Equation (3) was proposed on empirical grounds, namely, it appeared that this equation described accurately the T dependence of ρ near the IMT. The analysis which led PST to Eq. (3) is reviewed in Sec. IV A. It is difficult to see how Eq. (3) can be justified theoretically. Although first-order perturbation theory leads to a shift of the conduction-band edge which is linear in σ,[8] more realistic treatments lead to a more complicated dependence of the conduction-band edge on magnetic order.[9]

PST also imply that Eq. (2) does not give the correct dependence of Δ on magnetic order. Their argument is based, in part, on the assumption that δ_R is proportional to the nearest-neighbor two-spin correlation function $\langle \vec{S}_1 \cdot \vec{S}_2\rangle$, where $\vec{S}_1$ and $\vec{S}_2$ are neighboring spins in the Eu^{++} lattice. This assumption can be written

$$\delta_R = \delta_R(0)\eta\ , \tag{4}$$

where $\delta_R(0) \cong 0.25$ eV is the red shift at $T = 0$ (Refs. 7 and 10) and $\eta = \langle \vec{S}_1 \cdot \vec{S}_2\rangle/S^2$. Equations (2) and (4) give

$$\Delta = \Delta_0(1 - b\eta), \tag{5}$$

where b is a constant.

Equation (5) is similar in form to Eq. (3), except that the long-range order parameter σ is replaced by the short-range order parameter η. From the analysis of their data PST concluded that Eq. (3) was correct and that Eq. (5) was invalid. Since they regarded Eq. (5) as equivalent to Eq. (2), their conclusion implied that Eq. (2) was also invalid.

Relation (4) is based on the red-shift data of Freiser *et al.*,[7] and others,[10] at zero or low magnetic fields (2 kOe). These data show that at zero and low fields, δ_R is very nearly proportional to η. However, the validity of Eq. (4) for *all* values of H and T has not been established experimentally. In fact, some of the available red-shift data, discussed later in Sec. V B, disagree with Eq. (4). For this reason we regard Eqs. (2) and (5) as two different equations which are not necessarily equivalent to each other.

Each of the Eqs. (2), (3), and (5) represents a possible relation between Δ and the magnetic order. Of these only Eqs. (2) and (3) were proposed (by Oliver *et al.* and PST, respectively). In the following sections we shall examine whether any of the three equations describes the data accurately.

III. LOW- AND HIGH-TEMPERATURE REGIONS

In the present work, the resistivity and Hall effect for activated samples were measured in two temperature regions: $4.2 \leq T \lesssim 120$ K, and $230 \lesssim T \lesssim 300$ K. Between ~120 and ~230 K the resistivity was so high, even at the highest magnetic fields, that electrical measurements could not be made with our apparatus. In terms of the trap model (Ref. 1, Sec. V A), the high resistivity between 120 and 230 K is a consequence of the high values of Δ/kT in this temperature region.

There are several qualitative differences between the results for the low-temperature region below 120 K and those for the high-temperature region above 230 K:

(i) Below 120 K, kT was sufficiently small so that measurements could be made only when $\Delta(H, T)$ was below $\sim 0.5\Delta_0$. Above 230 K, where measurements could be made at all fields, the H dependence of R_0 reflected the decrease of Δ from Δ_0 at $H = 0$ to $\sim 0.8\Delta_0$ at the highest field. Thus the data in the low-T and high-T regions gave information for low and high values of Δ, respectively.

(ii) Since δ_R and η have not been measured as a function of H in the low-T region, only the relationship between $\Delta(H, T)$ and $\sigma(H, T)$ was investigated in this region. For $T > 230$ K, the H dependence of η could be calculated. In addition, the H dependence of δ_R was measured at 297 K. Thus in the high-T region the electrical transport data could be used to test all three relations between Δ and magnetic order.

(iii) It will be shown below that relations (2), (3), and (5) lead to very different predictions for the H dependence of the Hall coefficient at $T \gtrsim 230$ K. Therefcre, the high-T data are useful for testing the various models.

(iv) Since the magnetic susceptibility is much smaller at $T \gtrsim 230$ K than at $T \lesssim 120$ K, the corrections due to the demagnetizing field are much less important in the high-T region than in the low-T region. For example, at 297 K the difference between B and H_{ext} is only ~1%.

IV. ANALYSIS OF RESISTIVITY AND HALL DATA FOR $T \lesssim 120$ K

In this section the data for $\rho(H_{ext}, T)$ and $R_0(H_{ext}, T)$ in the range $4.2 \leq T \lesssim 120$ K and $0 \leq H_{ext} \leq 140$ kOe are used to test Eq. (3). The relevant resistivity and Hall data are shown in Figs. 13–15 of Ref. 1. The method of analyzing the resistivity data is similar to the one used by PST.[3,4] However, contrary to PST we find that Δ is not uniquely determined by σ and, therefore, Eq. (3) (with parameters Δ_0 and a which are independent of H and T) does not give an accurate description of the dependence of Δ on magnetic order. Our conclusion

is based for the most part on additional data at much higher magnetic fields than those available earlier (our experimental data at zero and low magnetic fields are similar to those of PST). In addition, we show that the analysis of PST contains some weaknesses which make the exact agreement with Eq. (3) questionable even at zero and low magnetic fields.

Because the procedure used by PST is the starting point of our own analysis, we review their procedure in detail.

A. PST Method of Analysis

PST[3,4] measured the temperature dependence of ρ at external magnetic fields $H_{ext} = 0$ and 20 kOe. To determine the $\Delta(H_{ext}, T)$ from the data for $\rho(H_{ext}, T)$, they used the equation

$$\rho(H_{ext}, T) = \rho_0 e^{\Delta(H_{ext}, T)/kT} \quad (6a)$$

or

$$\Delta(H_{ext}, T) = kT \ln[\rho(H_{ext}, T)/\rho_0] , \quad (6b)$$

where ρ_0 was taken to be the resistivity at 4.2 K. Values of $\sigma(T)$ at $H_{ext} = 0$ were obtained from Mössbauer data. A plot of $\Delta(T)$ vs $\sigma(T)$ at $H_{ext} = 0$ gave a straight line, but a plot of $\Delta(T)$ vs $\eta(T)$ did not. On this basis it was concluded that Δ obeyed Eq. (3) but not Eq. (5). A value $a \cong 1.3$ for the parameter in Eq. (3) was obtained.

The sample used by PST was nonellipsoidal, so that in the presence of an external magnetic field, $\vec{H}_{ext}$, the magnetization M inside the sample was nonuniform. PST argued that in the presence of H_{ext} the resistance of their sample was determined by the regions of maximum ρ, which corresponded to the regions of lowest M. Values of Δ obtained from Eq. (6) were regarded as characteristic of these regions. The demagnetizing factor N for these lowest-M regions was set equal to the maximum possible value 4π, i.e., the reduced magnetization which appears in Eq. (3) was set equal to that of a thin disk perpendicular to $\vec{H}_{ext}$. With this choice of N, the results at $H_{ext} = 20$ kOe satisfied Eq. (3) with the same values of Δ_0 and a as for $H_{ext} = 0$.

The conclusion of PST that Δ obeys Eq. (3) but not (5) is based primarily on Fig. 2 of Ref. 3. However, as this figure shows, the two equations do not predict widely different behaviors in this case. It is therefore important to examine whether the uncertainties involved in the method of data analysis are significant. One source of uncertainty is considered below. In addition, we consider the demagnetizing factor used to construct Fig. 3 of Ref. 3.

a. Changes in the mobility μ. When $\Delta \gg kT$, the carrier concentration n is given by Eq. (1). Since $\rho = (ne\mu)^{-1}$, the parameter ρ_0 in Eq. (6) is given by

$$\rho_0(T, H) = [N(T) e \mu(T, H)]^{-1} . \quad (7)$$

PST set ρ_0 equal to $\rho(4.2\ \mathrm{K})$. Implicit in this choice are two assumptions. First, the T and H dependences of μ are ignored, i.e., $\rho(T)/\rho(0)$ is set equal to $n(0)/n(T)$ [see Eq. (1) of Ref. 3]. Second, $N(T)$ is equated to $n(4.2\ \mathrm{K})$.

As shown in the preceding paper,[1] for any fixed H_{ext}, μ decreases with increasing T in the region of the IMT. Although the T dependence of μ at $H_{ext} = 0$ was not determined in the present work, the data for $H_{ext} = 25$ kOe (Fig. 8 of Ref. 1) suggest that μ at $H_{ext} = 0$ decreases by an order of magnitude or more between 50 and 69 K (T_C). Had the T dependence of μ been taken into account by PST, the agreement of the zero-field results with Eq. (5) would have improved, whereas the excellent agreement with Eq. (3) would not have been maintained.

The choice $N(T) = n(4.2\ \mathrm{K})$ is, in principle, not valid in general, since $N(T)$ depends on the number of traps, on the number of compensating acceptors, and on the density of states of the conduction band $N_c(T)$.[11] However, in practice, the error introduced by setting $N(T) = n(4.2\ \mathrm{K})$ was not very important for our samples, and this was probably also the case for the samples used by PST.

b. Demagnetizing factor N. The choice $N = 4\pi$ is questionable. It is based on the assumption that the measured resistance was determined by the highest-resistivity regions. However, it is doubtful that a sample can be represented by an equivalent circuit in which the low-resistivity regions are *in series* with the high-resistivity regions. It should also be noted that the geometry of the sample used by PST leads to an average demagnetizing factor $N_{av} < 2\pi$. In the region of the IMT, the calculated demagnetizing field NM can be as large as 15 kOe if one lets $N = 4\pi$. Therefore, the demagnetizing corrections are important for the field

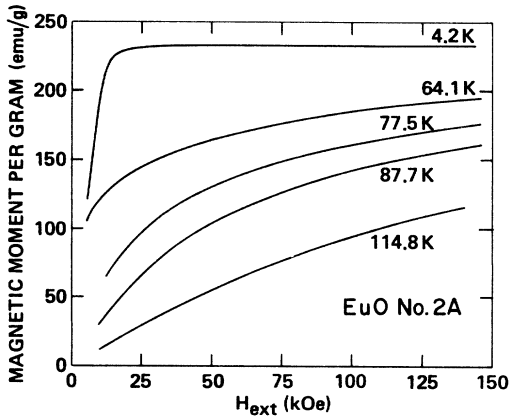

FIG. 1. Magnetization of sample 2A as a function of external (applied) magnetic field H_{ext} at several temperatures. The average demagnetization factor in this case is approximately equal to 6.

H_{ext} = 20 kOe used by PST. (These corrections are less important at the highest fields used in the present work.)

B. Analysis of Our Resistivity Data by the Method of PST

The resistivity data in Figs. 13 and 14 of Ref. 1 were analyzed following a procedure similar to the one used by PST, i.e., values of $\Delta(H_{ext}, T)$ were obtained using Eq. (6) with ρ_0 set equal to $\rho(4.2\ \mathrm{K})$. Values for $\sigma(H_{ext}, T)$ were obtained from magnetization measurements, except at $H_{ext} = 0$ where, following PST, we used the expression

$$\sigma(T) = 1.13\,(1 - T/T_C)^{0.36} \tag{8}$$

and the values of T_C for our samples (Table I of Ref. 1). The magnetization measurements were carried out in liquid helium, liquid nitrogen, liquid argon, and liquid natural gas. Some of the magnetization data are shown in Fig. 1. Values for σ at temperatures intermediate between those of the various cryogenic liquids were obtained by interpolation.

The above procedure of obtaining Δ and σ differs from that of PST in one respect. Our values for σ at $H_{ext} \neq 0$ correspond to the measured magnetization, with the sample oriented relative to $\vec{H}_{ext}$ as in the resistivity measurements. This corresponds to a demagnetizing factor N equal to the average demagnetizing factor N_{av} for the sample (see Ref. 12).

The results for Δ as a function of σ at various values of H_{ext} are shown in Fig. 2. Note that the results for the two samples (obtained from two different single crystals) are very similar. At $H_{ext} = 0$ the results for sample 2A follow Eq. (3) with $a = 1.39$ and $\Delta_0 = 0.274$ eV, except at low values of Δ. The corresponding results for sample 3 give $a = 1.38$ and $\Delta_0 = 0.289$ eV for $H_{ext} = 0$. These values for a and Δ_0 are close to the values obtained by PST.[3,4] However, Fig. 2 shows that for a given value of σ, Δ increases with increasing H_{ext}. This means that Δ is not uniquely determined by σ and therefore cannot be described by Eq. (3) with parameters Δ_0 and a which are independent of H_{ext}. For example, the data at 140 kOe extrapolate to $\Delta_0 = 0.38$ eV for sample 2A and to $\Delta_0 = 0.41$ eV for sample 3, whereas the corresponding values obtained from the results at $H_{ext} = 0$ are ~40% lower. The T dependence of the zero-field resistivity near room temperature gives $\Delta_0 = 0.32$ eV for both samples (see Table I of Ref. 1).

Since the results of Fig. 2 do not follow Eq. (3) with fixed Δ_0 and a, it follows either that Eq. (3) does not give an accurate description of the dependence of Δ on magnetic order, or that the procedure of evaluating Δ and/or σ from the data is inaccurate, or both. We now consider sources of error in the procedure of obtaining Δ and σ.

Consider first the demagnetizing factor N, which affects the values of σ. For samples 2A and 3, oriented with their long edge perpendicular to $\vec{H}_{ext}$, the average demagnetizing factor was approximately 2π. The values for σ in Fig. 2 correspond to this average N. Since N must always be between zero and 4π, the maximum possible error in N was $\sim 2\pi$. For $H_{ext} = 140$ kOe, a change of N by $\pm 2\pi$ would shift the values of σ in Fig. 2 only by ∓ 0.01 to ∓ 0.02. These shifts in σ are too small to make the line for Δ vs σ at 140 kOe coincide with that for $H_{ext} = 0$ [the values for $\sigma(T)$ at $H_{ext} = 0$ are not affected by the choice of N].

The other known sources of error in the analysis are related to the choice $\rho_0 = \rho(4.2\ \mathrm{K}) = \mathrm{const.}$ in Eq. (6). This choice neglects the changes in μ from its value at 4.2 K, and it also assumes that the parameter $N(T)$ in Eq. (1) is equal to $n(4.2\ \mathrm{K})$. The errors introduced by letting $\mu = \mu(4.2\ \mathrm{K})$ and $N(T) = n(4.2\ \mathrm{K})$ will be discussed separately.

For a fixed value of H_{ext}, μ decreases with increasing T. When the decrease of μ from its value at 4.2 K is taken into account, the resulting values of $\Delta(H_{ext}, T)$ are lower than those plotted in Fig. 2. This correction for Δ becomes progressively larger as T (or Δ) increases. Analysis of this correction shows that the difference between $\Delta(H_{ext} = 140\ \mathrm{kOe})$

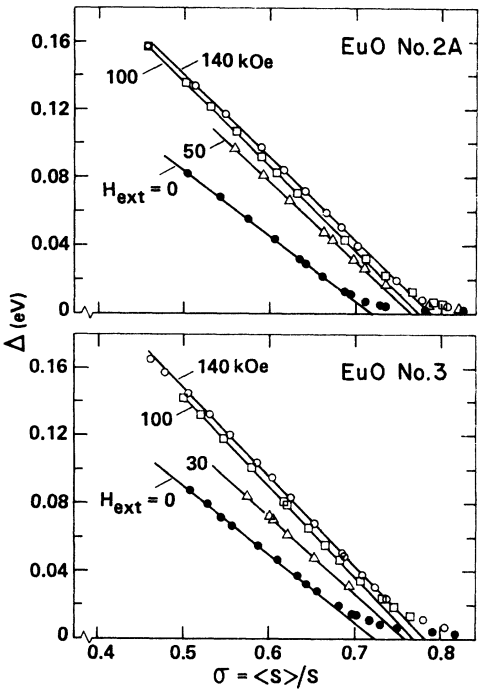

FIG. 2. Activation energy Δ as a function of reduced magnetization σ for various fixed values of H_{ext}. Values for Δ were obtained from data for ρ using Eq. (6) and setting $\rho_0 = \rho(4.2\ \mathrm{K})$. Values for σ were obtained from magnetization measurements, except at $H_{ext} = 0$, where Eq. (8) was used.

and $\Delta(H_{ext}=0)$, for a given value of σ, cannot be explained by the variation of μ.

The effect of replacing $N(T)$ by $n(4.2\ \mathrm{K})$ was estimated in the following way. The T dependence of ρ $(H_{ext}=0)$ for $T \gg T_C$ gives $\Delta_0 = 0.32$ eV. Substituting this value into Eq. (6) and using the measured value of ρ $(H_{ext}=0)$ at 297 K we calculated ρ_0 (297 K). For samples 2 and 3 this value was one order of magnitude larger than $\rho(4.2\ \mathrm{K})$. To evaluate the T dependence of $\rho_0(T)=[N(T)e\mu]^{-1}$, it was assumed, following PST, that the samples contained compensating acceptors which pinned the Fermi level at the trap level. In this case $N(T)$ is proportional to $T^{3/2}$.[11] Thus aside from the variation of μ, $\rho_0(T) \propto T^{-3/2}$. To calculate $\rho_0(T)$ in the region of the IMT we approximated μ by its average value near the IMT at high magnetic fields, which was roughly equal to $3\mu(297\ \mathrm{K})$. With these approximation

$$\rho_0(T) = \tfrac{1}{3}(297/T)^{3/2}\rho_0(297\ \mathrm{K}). \tag{9}$$

In the region of the IMT, values for $\rho_0(T)$ calculated from Eq. (9) are one or two orders of magnitude larger than $\rho(4.2\ \mathrm{K})$. When these values are substituted in Eq. (6), the calculated values of $\Delta(H_{ext}, \sigma)$ are lower than those shown in Fig. 2. However, the difference $\Delta(140\ \mathrm{kOe}) - \Delta(0)$, for a fixed σ, is only ~25% smaller than that in Fig. 2. Thus the new choice for $\rho_0(T)$ does not improve the agreement with Eq. (3) significantly.

For completeness, we have also considered the possibility that there are no compensating acceptors. Then $\rho_0(T)$ is proportional to $T^{-3/4}$.[11] This choice of $\rho_0(T)$ does not lead to substantially different results from those obtained from Eq. (9), i.e., the disagreement with Eq. (3) remains.

In conclusion, it does not appear that the disagreement between the results in Fig. 2 and the predictions of Eq. (3) (with fixed Δ_0 and a) can be accounted for by the known sources of error in the procedure used to obtain Δ and σ.

C. Analysis of Hall Data

In principle, Hall data are more useful than resistivity data in determining the dependence of Δ on magnetic order, because R_0 is directly related to n and the question of the variation of μ does not arise. However, in practice, Hall data are more difficult to obtain than resistivity data. In the present work, Hall data near the IMT of activated samples were obtained only at $H_{ext} \gtrsim 100$ kOe. These Hall data provide only a limited test of Eq. (3).

The temperature variation of R_0 in sample 3, for $H_{ext}=100$ and 140 kOe, is shown in Fig. 15 of Ref. 1. These data were analyzed by means of Eq. (1) in order to obtain $\Delta(H_{ext}, T)$. The analysis was complicated by the fact that the parameter $N(T)$ in Eq. (1) was not known exactly. Three different methods of obtaining $N(T)$ were used. In the first method $N(T)$ was set equal to $n(4.2\ \mathrm{K}) = 1/eR_0(4.2\ \mathrm{K})$. This choice is analogous to setting $\rho_0 = \rho(4.2\ \mathrm{K})$. The results obtained with this choice of $N(T)$ are shown in Fig. 3.

The other two choices for $N(T)$ are based on the zero-field value of $R_0(297\ \mathrm{K})$ and the value $\Delta_0 = 0.32$ eV at $T \gg T_C$. When these values are substituted into Eq. (1) they give $N(T)$ at $T = 297$ K. The variation of $N(T)$ with T depends on whether or not the sample contains compensating acceptors.[11] If compensating acceptors are present then

$$N(T)/N(297\ \mathrm{K}) = (T/297)^{3/2}, \tag{10}$$

whereas in their absence,

$$N(T)/N(297\ \mathrm{K}) = (T/297)^{3/4}. \tag{11}$$

The Δ-vs-σ plots obtained for these two choices of $N(T)$ are shown in Fig. 4(a) and 4(b). In both cases negative values of Δ are obtained for the highest values of σ. This indicates either that these choices are inaccurate or, more likely, that Eq. (1) does not apply to the low-temperature "tail" of the IMT [Eq. (1) is only valid when $\Delta \gg kT$].

The results in Figs. 3, 4(a), and 4(b) are similar, and show that for each value of H_{ext} (i.e., 100 and 140 kOe), Δ obeys Eq. (3) with parameters Δ_0 and a, which are not very sensitive to the choice $N(T)$. Moreover, the parameters Δ_0 and a for the 140 kOe data are only slightly different from those for the 100 kOe data. However, the important question, whether Eq. (3) with the parameters Δ_0 and a obtained at $100 \le H_{ext} \le 140$ kOe also describes the Hall data at $0 \le H_{ext} \le 100$ kOe, cannot be answered on the basis of our limited Hall data. [As shown earlier, Eq. (3), with fixed Δ_0 and a, does not describe the resistivity data at all values of H_{ext}.]

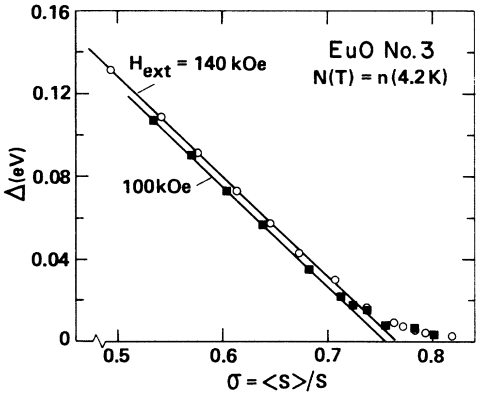

FIG. 3. Activation energy Δ for sample 3 as a function of σ at $H_{ext}=100$ and 140 kOe. Δ was calculated from results for $R_0 \sim n^{-1}$ using Eq. (1) and setting $N(T)=n(4.2\ \mathrm{K})$.

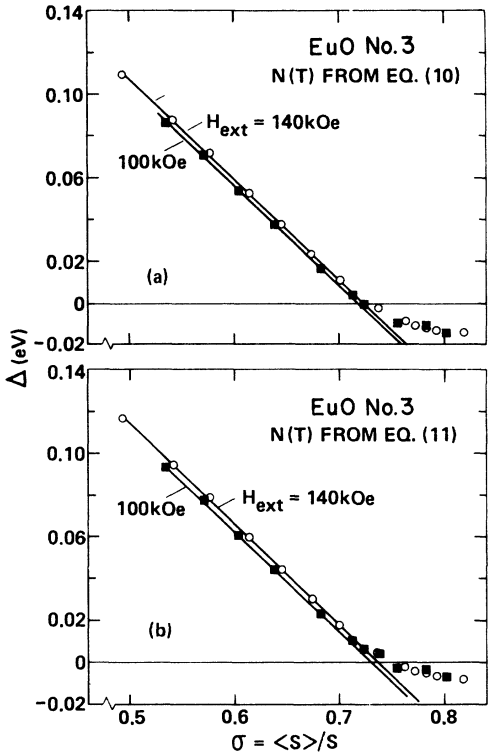

FIG. 4. Δ vs σ for sample 3 at H_{ext} = 100 and 140 kOe. Δ was calculated from results for $R_0 \sim n^{-1}$ using: (a) Eqs. (1) and (10), and (b) Eqs. (1) and (11).

D. Additional Comparisons with Eq. (3)

Other comparisons of the experimental data with Eq. (3) have also been made. In one comparison, Eq. (3) was used to predict the shift of the IMT to higher temperatures caused by H_{ext} = 100 kOe. For this purpose the zero-field resistivity data were analyzed by the method of PST and the results for Δ vs σ were fitted to Eq. (3). Values for Δ_0 and a were obtained from this fit. The variation of ρ with T at 100 kOe was then calculated from Eqs. (3) and (6), using data for σ(100 kOe) vs T and setting $\rho_0 = \rho(4.2\ \mathrm{K})$. The predicted and observed curves for ρ(100 kOe) vs T are shown in Fig. 5. It is clear that although Eq. (3) gives a rough estimate of the field-induced shift of the IMT to higher temperatures, it overestimates this shift.

Further comparisons with Eq. (3) were made by considering the field dependence of ρ and R_0 at fixed temperatures between 65 and 88 K. From Eqs. (1) and (3), at a fixed temperature

$$R_0(H_2)/R_0(H_1) = e^{(a\Delta_0/kT)(\sigma_1-\sigma_2)}\ , \qquad (12)$$

where σ_1, σ_2 are the reduced magnetizations at H_1 and H_2. If the field dependence of μ is neglected, then the right-hand side of Eq. (12) is also equal to $\rho(H_2)/\rho(H_1)$. In applying Eq. (12) we used the value

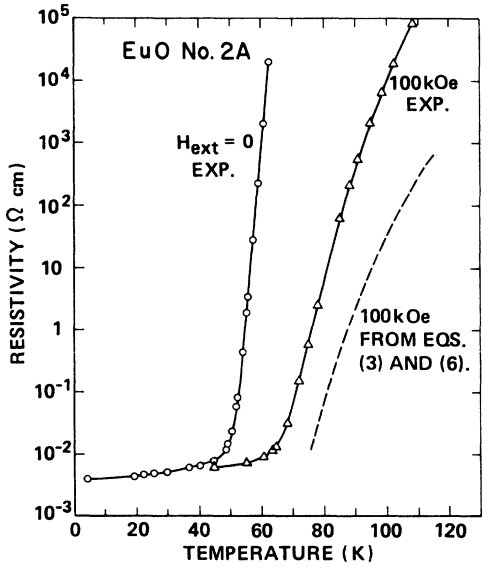

FIG. 5. Effect of an external field equal to 100 kOe on the IMT. The solid lines represent the measured resistivity ρ at H_{ext} = 0 and 100 kOe. The dashed line is calculated from Eq. (6), with $\rho_0 = \rho(4.2\ \mathrm{K})$, assuming Eq. (3). Δ_0 and a in Eq. (3) are obtained from the zero-field results in Fig. 2.

for $(a\Delta_0)$ which was obtained from analysis of the zero-field resistivity. The data for $R_0(H_2)/R_0(H_1)$ and $\rho(H_2)/\rho(H_1)$ showed that Eq. (12) gave a reasonably good estimate of these ratios. One case in which the agreement was particularly good is shown in Fig. 6.

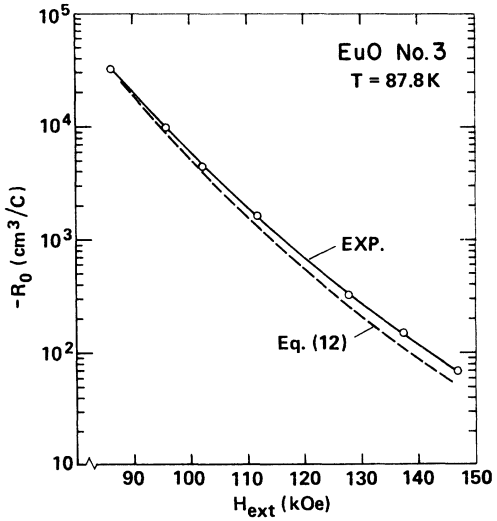

FIG. 6. Ordinary Hall coefficient R_0 vs H_{ext} in sample 3 at 87.8 K. The dashed curve is calculated from Eq. (12), with values for Δ_0 and a obtained from the zero-field results in Fig. 2, and σ vs H_{ext} from Fig. 1. The calculated curve was forced to pass through the experimental point at the lowest field.

Although Eq. (12) is derived from Eq. (3), the success of Eq. (12) does *not* imply that Eq. (3) (with the zero-field values for Δ_0 and a) holds at constant T. The good agreement of data for $R_0(H_2, T)/R_0(H_1, T)$ with Eq. (12) only implies that the isotherm $\Delta(H_{ext})$ vs $\sigma(H_{ext})$ in the Δ-σ plane is parallel to the straight line $\Delta(T)$ vs $\sigma(T)$ for zero field. Curves for Δ vs σ at constant values of H_{ext}, or at constant temperatures, are shown in Fig. 7. This sketch is based on analysis of our resistivity data by the method of PST. Note that an isotherm such as T_2 can be roughly parallel to the line for $H_{ext} = 0$.

E. Conclusions

Analysis of the resistivity data shows that Eq. (3) with fixed Δ_0 and a does not give an accurate description of the T dependence of Δ for *all* fields. However, for a fixed value of H_{ext}, Δ is approximately linear in σ over the range of our measurements (see Fig. 2). Thus the results for each value of H_{ext} can be approximated by Eq. (3) with coefficients Δ_0 and a which depend on H_{ext}. Figure 2 shows that the variations of Δ_0 and a with H_{ext} are most rapid at low fields and are fairly slow between 50 and 140 kOe.

V. ANALYSIS OF HALL DATA AT $T \sim 4T_C$

Hall measurements at $230 \lesssim T \lesssim 300$ K provide a sensitive means of testing Eqs. (2), (3), and (5), because these equations lead to very different variations of n with H at these temperatures. In addition, demagnetization corrections are not important at these temperatures.

A. Predictions for R vs H on the Basis of Eqs. (2), (3), and (5)

In considering the H dependence of R at a fixed temperature near $4T_C$, we first assume that the conduction band is a single band and that the density-of-states effective mass is independent of H. In this case,

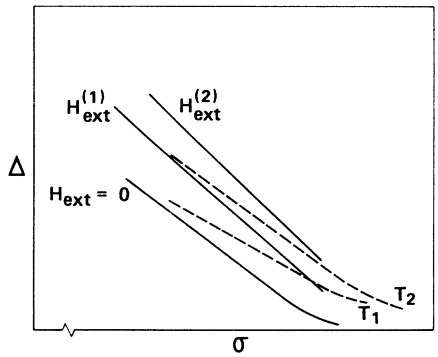

FIG. 7. Schematic showing $\Delta(H_{ext}, T)$ vs $\sigma(H_{ext}, T)$ at fixed values of H_{ext} (solid lines) and at fixed values of T (dashed lines). Note that an isotherm, such as T_2, can be nearly parallel to the curve for Δ vs σ at $H_{ext} = 0$.

$$R(H)/R(0) = n(0)/n(H) = e^{[\Delta(H) - \Delta(0)]/kT}. \quad (13)$$

The single-band assumption will be examined and modified later.

Using Eq. (13), the various relations in Sec. II lead to the following equations for R vs H at fixed T:

(i) If Δ varies linearly with the optical red shift [Eq. (2)] then

$$R(H) = R(0)\, e^{-(1+\alpha)\delta_R(H)/kT}, \quad (14)$$

where $\delta_R(H)$ is the red shift caused by the field H.

(ii) If Δ varies linearly with the magnetization [Eq. (3)], then

$$R(H) = R(0)\, e^{-a\Delta_0\sigma(H)/kT}, \quad (15)$$

where we have used $\sigma(T > T_C, H = 0) = 0$. At $T \sim 4T_C$, $\sigma(H)$ is proportional to H at fields below 150 kOe, so that Eq. (15) leads to a linear variation of ln R with H.

(iii) If Δ varies linearly with the nearest-neighbor two-spin correlation function [Eq. (5)] then

$$R(H) = R(0)\, e^{b\Delta_0[\eta(0) - \eta(H)]/kT}. \quad (16)$$

The H dependence of η is discussed in detail in the following paper.[13] For $T \gg T_C$ one expects a ferromagnet to behave approximately like a paramagnet consisting of noninteracting spins. In this case $\eta \equiv \langle \vec{S}_1 \cdot \vec{S}_2 \rangle / S^2 = \langle \vec{S}_1 \rangle \cdot \langle \vec{S}_2 \rangle / S^2 = \sigma^2$. Since for a paramagnet $\eta = \sigma = 0$ at $H = 0$, $[\eta(H) - \eta(0)] = \sigma^2$. The effect of the interaction between the spins on the relation between η and σ is calculated in the following paper on the assumption that EuO can be approximated by a Heisenberg ferromagnet with only nearest-neighbor interactions.[14] Above T_C and at low fields, where σ is linear in H, we find that $[\eta(H) - \eta(0)] = A(T)\sigma^2$, where the parameter A depends only on T/T_C and is of order one. In the limit $T \gg T_C$, $A = 1$. At $T \sim 4T_C$, where our Hall data were taken, A is only a few percent below unity. Therefore, we shall let $[\eta(H) - \eta(0)] = \sigma^2$. The use of this approximation in Eq. (16) is satisfactory as long as $(b\Delta_0\sigma^2/kT) \lesssim 1$, which is the case for our experiments at $T \sim 4T_C$. Equation (16) therefore reduces to

$$R(H) = R(0)\, e^{-b\Delta_0\sigma^2(H)/kT}. \quad (17)$$

Equations (15) and (17) lead to very different predictions for the behavior of $R(H)$. First, since σ is proportional to H, Eq. (17) predicts a quadratic variation of ln R with H, instead of the linear variation predicted by Eq. (15). There is also a large difference between the magnitudes of the H-induced change in R predicted by Eqs. (15) and (17). At $T = 4T_C$, $\sigma \cong 0.15$ at 150 kOe, so that $\sigma^2 \ll \sigma$ for fields below 150 kOe. It is shown below that the coefficients a and b in Eqs. (15) and (17) are comparable. Therefore, Eq. (17) leads to a much

smaller H-induced change in R than Eq. (15).

The discussion thus far was based on the assumption of a single conduction band. This assumption is probably not valid at $T \sim 4T_C$ because at $T > T_C$ a magnetic field is expected to split the conduction band into spin-up and spin-down subbands separated by $\delta(H, T)$. At low fields $\delta \lesssim kT$, so that both subbands should be considered, i.e., $n = n^{(+)} + n^{(-)}$ where $n^{(+)}$ and $n^{(-)}$ are the electron concentrations in the upper and lower subbands, respectively.

In the experiments of Oliver *et al.* and of PST, electrical conduction near the IMT was due to electrons in the lower subband (because $\delta \gg kT$). These authors interpreted Δ as a measure of the energy separation ϵ between the trap level and the edge of the lowest conduction subband. Therefore, we shall assume that in Eqs. (2), (3), and (5), Δ refers to the activation energy for the lower subband, and that the activation energy for the upper subband is $\Delta + \delta$. We shall also assume that the density-of-states effective mass for each subband is independent of H.

In the limit of high magnetic fields, where $\delta \gg kT$, only the lower subband contributes to the Hall effect. The density of states $N_c^{(-)}(E)$ for this subband is equal to one-half of the density of states $N_c(E)$ for the unsplit conduction band at $H = 0$, where in each case the energy E is measured from the bottom of the band. Therefore, in the limit $\delta \gg kT$ the right-hand sides of Eqs. (13)–(17) should be multiplied by a factor of 2, to take into account the effect of spin splitting.

It was shown in Fig. 18 of Ref. 1 that at $T \cong 4T_C$ a field of 150 kOe increases the average Hall mobility by only ~ 25%. This suggests that the mobilities $\mu^{(+)}$ and $\mu^{(-)}$ of the two spin subbands are not widely different, and that setting $\mu^{(+)} = \mu^{(-)}$ will not result in a serious error in the calculation of $R(H)$.[15] With this additional assumption

$$\frac{R(H)}{R(0)} = \frac{n(0)}{n^{(+)}(H) + n^{(-)}(H)} = \frac{e^{-\Delta_0/kT}}{\frac{1}{2}e^{-\Delta/kT} + \frac{1}{2}e^{-(\Delta+\delta)/kT}} \tag{18}$$

or

$$\frac{R(H)}{R(0)} = \frac{2e^{(\Delta-\Delta_0)/kT}}{1 + e^{-\delta/kT}}, \tag{19}$$

where Δ and δ depend on H. The factor $e^{(\Delta-\Delta_0)/kT}$ is equal to $R(H)/R(0)$ for an unsplit band [see Eq. (13)]. It follows from Eq. (19) that the effect of spin splitting of the conduction band is to multiply the right-hand side of Eq. (13), [and those derived from it, i.e., Eqs. (14)–(17)] by a factor of $2(1 + e^{-\delta/kT})^{-1}$, which varies between 1 (when $\delta = 0$) and 2 (when $\delta \gg kT$).

More detailed predictions for the effect of the spin splitting on $R(H)$ can be made only if one knows the H dependence of δ. Physically, one expects that the "center of gravity" of the two subbands will not increase in energy as H increases. Assuming that compensating acceptors are present, this means that $0 \leq \delta \leq 2(\Delta_0 - \Delta)$. If the center of gravity of the two subbands is independent of H, $\delta = 2(\Delta_0 - \Delta)$ and Eq. (19) gives

$$\frac{R(H)}{R(0)} = \frac{1}{\cosh[(\Delta - \Delta_0)/kT]}. \tag{20}$$

If one assumes, for example, that Eq. (3) describes the variation of Δ then Eq. (20) gives

$$R(H) = R(0)/\cosh(a\Delta_0\sigma/kT) \tag{21}$$

for $\delta = 2(\Delta_0 - \Delta)$, whereas the case $\delta = 0$ is given by Eq. (15). In the limit of high magnetic fields, where $(a\Delta_0\sigma) \gg kT$, Eq. (21) gives a value for $R(H)$ which is twice that given by Eq. (15), as expected.

Again, it should be emphasized that if one assumes $\mu^{(+)} = \mu^{(-)}$ then, for any δ, $R(H)$ cannot exceed twice the value given by the formula for an unsplit conduction band.

B. Optical Red Shift

To compare the data for R vs H near room temperature with Eqs. (2) or (14), it is necessary to know the H dependence of δ_R. However, reliable results for δ_R vs H are difficult to obtain for reasons which are discussed below.

Equation (2) is based on a picture in which the optical transition is to the lowest conduction band. δ_R is then identified as the energy shift of this band due to magnetic order. In the presence of a magnetic field the conduction band splits into two subbands and the interpretation of the measured red shift δ_M as a function of H is not straightforward because $\delta_M(H)$ is some weighted average of the H-induced energy shifts of the two subbands. On the other hand, the red shift δ_R, which appears in Eq. (2), corresponds to the shift of the lower subband so that, in general, $\delta_M(H) \leq \delta_R(H)$. This problem does not arise in the interpretation of optical data at $H = 0$ (except just below T_C) for the following reason. At $T > T_C$ the conduction band is not split, and at $T \ll T_C$ the splitting is sufficiently large that the upper subband has little effect on the optical absorption edge.

The experimental determination of δ_M vs H at $T > T_C$ is also complicated. It has been observed[7] that at $T > T_C$, $\delta_M(H)$ depends on the polarization of the light beam and its direction of propagation relative to the magnetization.

In the present work the optical-absorption edge was measured at ~ 297 K in fields up to 105 kOe. The experimental setup has been described previously.[16] An insulating EuO single crystal 0.4 mm thick was used. Unpolarized[17] light was transmitted through the sample in a direction perpen-

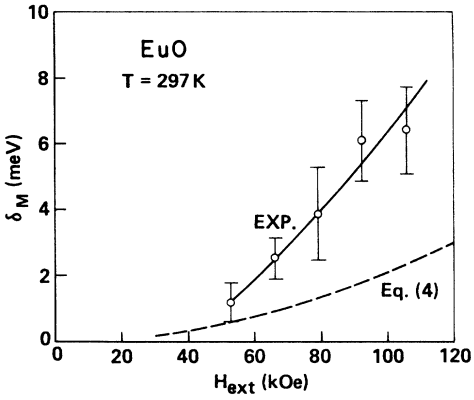

FIG. 8. Measured optical red shift δ_M vs H_{ext} at room temperature. The dashed line is calculated from Eq. (4) with $\delta_R(0) = 0.25$ eV and $[\eta(H_{ext}) - \eta(0)] = \sigma^2(H_{ext})$.

dicular to $\vec{H}_{ext}$ (and hence to $\vec{M}$). The optical transmission was measured from 0.9 to 1.0 eV, where the absorption coefficient β increased from ~1 to ~100 cm^{-1}. These values for β are similar to those observed by Freiser *et al.*[7] The measured red shift $\delta_M(H_{ext})$ was taken to be the energy shift due to H_{ext} at a fixed value of β. $\delta_M(H_{ext})$ was approximately independent of β for all values of β in the range 10–100 cm^{-1}. The results for $\delta_M(H_{ext})$ are shown in Fig. 8.

Figure 8 also shows the variation of δ_R calculated from Eq. (4) [which in the presence of a field reads $\delta_R(H, T) = \delta_R(T = 0)\eta(H, T)$], setting $[\eta(H) - \eta(0)] = \sigma^2$ and $\delta_R(T = 0) = 0.25$ eV. It is clear that the measured values of $\delta_M(H_{ext})$ are considerably higher than the calculated values of $\delta_R(H_{ext})$. The discrepancy may be even larger if the effect of spin splitting is included, because the measured shift $\delta_M(H_{ext})$ is expected to be smaller than $\delta_R(H_{ext})$.

It is noteworthy that earlier published data for the H-induced red shift at $T \gtrsim T_C$ also disagree with Eq. (4). According to Wachter, at $T = 74$ K, $[\delta_R(16$ kOe$) - \delta_R(0)] = 50$ meV (see Fig. 23 of Ref. 10). The corresponding value for $[\eta(H) - \eta(0)]$ can be deduced from the forced-isotropic-magnetostriction data of Argyle and Miyata[18] using the proportionality between the isotropic magnetostriction and η.[19] At 74 K these data give $[\eta(16$ kOe$) - \eta(0)] = 0.09$. Using this value one obtains from Eq. (4), $[\delta_R(16$ kOe$) - \delta_R(0)] \cong 23$ meV, which is a factor of 2 smaller than that observed. Similarly, according to Fig. 3 of Freiser *et al.*,[7] the red shift at 77 K, measured in the "linear parallel" configuration which these authors regard as best for measuring δ_R, is $[\delta_R(18$ kOe$) - \delta_R(0)] = 43$ meV. The corresponding value obtained from Ref. 18 and Eq. (4) is 21 meV, which is, again, a factor of 2 smaller than the measured value. The discussion above indicates that Eq. (4) is not valid at all temperatures and fields and, therefore, Eqs. (2) and (5) are not equivalent in general.

C. Comparison with Hall Data

It was shown in Sec. VI of Ref. 1 that near room temperature the Hall coefficient R in activated samples decreases in magnitude with increasing H. More extensive data for $R(H)$ at 297, 272, and 256.6 K are shown in Figs. 9–11. Note that the change in ln R induced by a given field H increases as T decreases. This trend is caused by two factors. First, as T decreases, a given field H produces a larger magnetic order (long range as well as short range), so that the factor $[\Delta(0) - \Delta(H)]$ which appears in Eq. (13) increases with decreasing T. In addition, the exponent in Eq. (13) contains the factor $1/kT$, which also increases with decreasing T.

We now compare the results in Figs. 9–11 with the predictions of the various relations for Δ. Consider first Eq. (3) which in the absence of spin-splitting leads to Eq. (15). To apply Eq. (15) it is necessary to know the product $(a\Delta_0)$. In the following analysis we let $a\Delta_0 = 0.445$ eV. This number is obtained from $\Delta_0 = 0.32$ eV, deduced from the T dependence of ρ at $230 < T < 300$ K, and $a = 1.39$, evaluated from the zero-field data in Fig. 2. This value for $a\Delta_0$ is also close to the average value deduced from the various straight lines in Fig. 2. (This figure gives $a\Delta_0 = 0.39$–0.48 eV for sample 2A, and 0.40–0.52 eV for sample 3.) The uncertainty of ±15% in $a\Delta_0$ does not affect the qualitative conclusions drawn from the analysis of $R(H)$. To use Eq. (15) it is also necessary to know $\sigma(H)$. This was obtained from magnetic susceptibility measurements.

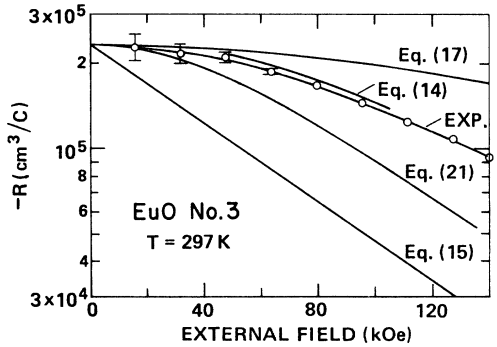

FIG. 9. Hall coefficient R vs H_{ext} at 297 K for sample 3. The curves representing Eqs. (15) and (21) were calculated with $a\Delta_0 = 0.445$ eV. The curve for Eq. (17) was calculated with $\Delta_0 = 0.32$ eV and $b = 1.5$. The curve for Eq. (14) was obtained from the data in Fig. 8 setting $(1 + \alpha) = 1.9$. Some of the experimental uncertainties for R are indicated. These uncertainties decrease with increasing H_{ext}.

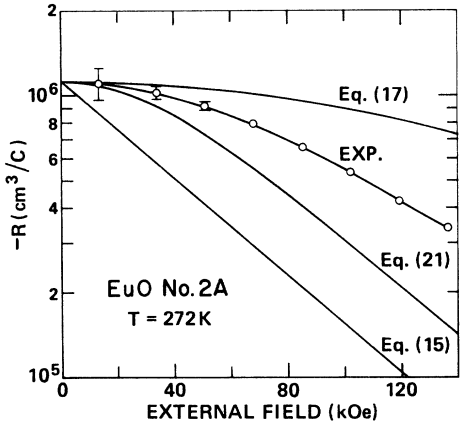

FIG. 10. R vs H_{ext} at 272 K for sample 2A. The theoretical curves represent Eqs. (15), (17), and (21).

The H dependence of R calculated from Eq. (15) is shown in Figs. 9–11. It is clear that this equation grossly overestimates the H dependence of R. This discrepancy cannot be resolved by considering the spin splitting of the conduction band because this splitting will increase the predicted values of $R(H)$ by a factor not exceeding 2. The qualitative effect of the spin splitting of the conduction band on the predictions for $R(H)$ is represented in Figs. 9–11 by the curves calculated from Eq. (21). At the highest fields, Eq. (21) gives values for $R(H)$ which are nearly twice as large as those obtained from Eq. (15), i.e., the maximum possible correction to Eq. (15) due to spin splitting. We therefore conclude that Eq. (3) does *not* represent the dependence of Δ on H near room temperature.

Consider next Eq. (17), which is based on Eq. (5). The parameter b in Eq. (17) can be estimated from the value of η at the onset of the IMT at zero field, i.e., near 50 K. Using the data of Argyle and Miyata[18] we obtain $b \cong 1.5$ (the uncertainty in b has no effect on the qualitative conclusions drawn below). The variation of $R(H)$ calculated from Eq. (17), with $b = 1.5$, is shown in Figs. 9–11. It is clear that this equation grossly underestimates the H dependence of R. The inclusion of spin splitting will make the agreement worse because the predicted values of $R(H)$ will be higher.

The analysis thus far shows that *the dependence of Δ on H is intermediate between those given by Eqs.* (3) *and* (5), i.e., intermediate between a linear dependence on the long-range order parameter σ and a linear dependence on the short-range order parameter η.

Finally, consider Eq. (14) which is based on Eq. (2). To evaluate the parameter $(1+\alpha)$ we use the data of Wachter,[10] which give $\delta_R \cong 0.17$ eV at $T \cong 50$ K (where Δ tends to zero). Substituting this value for δ_R into Eq. (2), setting $\Delta_0 = 0.32$ eV and $\Delta = 0$, we obtain $(1+\alpha) \cong 1.9$. Figure 9 shows the H dependence of R at 297 K calculated from Eq. (14) using the data in Fig. 8 and $(1+\alpha) = 1.9$. Here we assumed that $\delta_R(H_{ext}) = \delta_M(H_{ext})$. The agreement of the calculated curve with the experimental results is quite good. This indicates that at least in this case Eq. (2) gives a better description of the dependence of Δ on H than Eqs. (3) or (5). However, the good agreement of Eq. (14) with the Hall results at 297 K should not be regarded as conclusive evidence that Eq. (2) is correct, for several reasons. First, the comparison of $R(H)$ with Eq. (14) was made only at 297 K. Second, we assumed that $\delta_M(H_{ext}) = \delta_R(H_{ext})$, which is only true for an unsplit band. If spin splitting is present at 297 K then Eq. (14) must be modified. It is not clear that this modification can be made by simply replacing $\delta_R(H_{ext})$ by $\delta_M(H_{ext})$ in Eq. (14). Finally, there is some uncertainty in the value of $(1+\alpha)$. Oliver *et al.*[2] give $(1+\alpha) \cong 0.45$ eV$/\delta_R(T=0)$, which leads to $(1+\alpha) \sim 1.5$ to ~ 1.8, depending on the choice for $\delta_R(T=0)$. A still lower value, $(1+\alpha) \cong 1.2$, can be

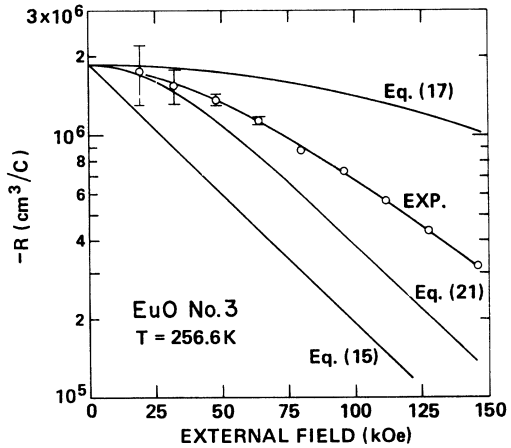

FIG. 11. R vs H_{ext} at 256.6 K for sample 3. The theoretical curves represent Eqs. (15), (17,) and (21).

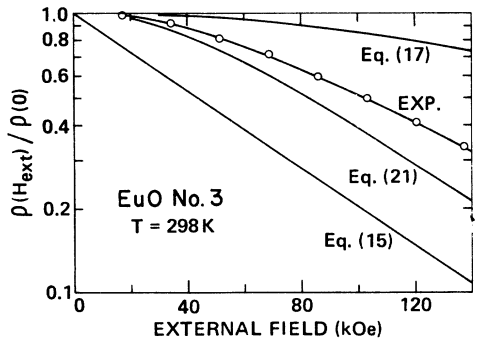

FIG. 12. $\rho(H_{ext})/\rho(0)$ vs H_{ext} at 298 K for sample 3. The theoretical curves represent Eqs. (15), (17), and (21), with $R(H)/R(0)$ replaced by $\rho(H)/\rho(0)$.

estimated from Fig. 20 of Ref. 2, on the assumption that Eq. (2) also describes the pressure dependence of Δ. Clearly, more extensive red-shift data as a function of H and T, and further analysis of the effects of spin splitting, are needed in order to check the validity of Eq. (2).

So far the analysis of the data near room temperature was confined to the H dependence of R, because R is more directly related to Δ than the resistivity ρ which depends not only on n but also on μ. The advantage of using R instead of ρ is also demonstrated by the behavior of $\rho(H)$ in *nonactivated* samples for which R is H independent near room temperature, whereas ρ decreases with H (see Ref. 1, Sec. VI).

For completeness we have also analyzed the H dependence of ρ in the activated samples. This is illustrated by Fig. 12 which shows $\rho(H_{ext})/\rho(0)$ at 298 K together with calculations based on equations similar to Eqs. (15), (17), and (21) [with $R(H)/R(0)$ replaced by $\rho(H)/\rho(0)$]. As can be seen, the agreement of the data in Fig. 12 with Eqs. (15) and (21) is better than that of the corresponding Hall data in Fig. 9, although a discrepancy still remains. Similar results were obtained at 272 K.

VI. CONCLUSIONS

Three relations for the dependence of Δ on magnetic order have been considered. It was shown that Eq. (3), with fixed values of Δ_0 and a, does not accurately describe the experimental results either at $T \lesssim 120$ K or at $T \sim 4T_C$, i.e., Δ is not linear in the long-range order parameter σ. The analysis also showed that Eq. (5) grossly underestimates the H dependence of R at $T \sim 4T_C$, i.e., Δ is not linear in the short-range order parameter η. Equation (2) gives a reasonably accurate description of the H dependence of R at 297 K, but it is uncertain whether Eq. (2) accurately represents the variations of Δ at other temperatures and fields.

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