# Hydrostatic-pressure effect on electrical resistivity of EuO in the temperature range 6-300 K

J. P. Desfours\*

Centre d'Etudes d'Electronique des Solides, Université des Sciences et Techniques du Languedoc, Place E. Bataillon, 34060 Montpellier Cedex, France

C. Godart

Equipe de Recherche du Centre National de la Recherche Scientifique-Chemie métallurgie, et spectroscopie des Terres Rares, 92190 Meudon-Bellevue, France

# G. Weill

Laboratoire de Physique du Solide, Centre National de la Recherche Scientifique, 92190 Meudon-Bellevue, France

## M. Averous\*

Centre d'Etudes d'Electronique des Solides, Université des Sciences et Techniques du Languedoc, Place E. Bataillon, 34060 Montpellier Cedex, France

## C. Llinares

## Faculté des Sciences de Tunis, Tunis, Tunisia (Received 23 March 1978)

The critical behavior of the pressure coefficient of the resistivity has been studied in a large temperature range (6-300 K) under hydrostatic pressure up to 5 kbar. Near the critical temperature  $T_c$ , this pressure coefficient reverses its sign. This is explained by the pressure effect on the magnetic order which induces a shift of  $T_c$  towards high temperature. A positive value of the pressure coefficient of the resistivity has been found at low temperatures and at high temperatures. This behavior is observed for the first time in EuO and is explained by transfer of electrons from a higher-mobility band to a lower-mobility band. Below 60 K, the large decrease in the resistivity with increasing pressure has been explained by pressure effect on the self-trapping phenomenon of the conduction electrons.

## I. INTRODUCTION

Europium oxide has been extensively studied during recent years because it exhibits many interesting phenomena like a large red shift of the absorption edge with cooling below the Curie temperature  $T_c$ , a giant Faraday rotation, a giant negative magnetoresistance, and a large temperature dependence of the electrical resistivity. All these phenomena are related to the magnetic properties. In this paper, we give the first results on the variation of the resistivity under hydrostatic pressure in a large temperature range, namely, 6-300 K. The pressure coefficient of the resistivity  $\alpha$  $= d \ln \rho / dP$  shows a critical behavior near the ordering temperature. Three important points in these results are (i) two sign changes of the resistivity pressure coefficient; one near the resistivity peak, which can be related to the shift of  $T_c$ with pressure, and the other below 45 K, which corresponds to a positive value of  $\alpha$  as at room temperature; (ii) a large decrease of the resistivity under pressure between 60 and 50 K, which cannot be explained only by the shift of  $T_{C}$ , but involves also an effect of pressure on the self-trapping of electrons; (iii) a positive value of the pressure coefficient of the resistivity at high temperatures has been obtained, the same as at low temperatures. This sign is contrary to previous results<sup>1-3</sup> and is explained by a transfer, under pressure, of electrons from a higher-mobility band (presumably a 6s band) to a lower-mobility band. This transfer is confirmed by Hall-effect measurements in the paramagnetic-temperature phase. Uniaxial-piezoresistance measurements show that the lower-mobility band is a 5*d* band along the [100] axis of the Brillouin zone.

The critical behavior of the resisti ity under pressure is discussed throughout the temperature range (6-300 K) in terms of a band scheme.

#### **II. EXPERIMENTAL DETAILS**

The single crystal used in this work was grown by the Guerci and Shafer method.<sup>4</sup> It was cut by cleaving in the [100] direction for uniaxial-stress experiments. The crystallographic direction was x-ray controlled with an accuracy of less than one degree. The uniaxial-stress apparatus was similar to that used by Averous *et al.*<sup>5</sup> The reproducibility of the piezoresistance effect was repeatedly checked and found to be better than 1%.

For resistivity and Hall-effect measurements under pressure, the sample was placed in a Cu-

2750

© 1978 The American Physical Society

Be bomb with helium as the pressure-transmitting medium.<sup>6,7</sup> The bomb was placed in a liquid-helium cryostat and an external electromagnet provided a magnetic-field intensity up to 1 T.

## **III. EXPERIMENTAL RESULTS**

Resistivity measurements were made under hydrostatic pressure up to 5 kbar in the temperature range (6-300 K). The chosen single EuO crystal contained an excess of Eu. The resistivity-versus-temperature curve at zero pressure (Fig. 1, curve 1) exhibits a sharp peak near 75 K and an insulator-metal transition (IMT) of about three orders of magnitude around 50 K. An elbow in the curve can be noticed at this temperature. Curve 2 in Fig. 1 shows the resistivity versus temperature for an applied pressure of 5 kbar. From these curves, three points can be made: (i) At high temperatures, the resistivity increases with pressure. This behavior is observed for the first time and is different from that obtained earlier with other types of samples.<sup>1-3</sup> At very low temperatures, the resistivity again increases with pressure. (ii) The resistivity peak is shifted by pressure towards higher temperatures at a rate of 0.48 K/kbar (Fig. 2). Near 76 K, the two  $\rho$ -vs-T curves (Fig. 1) cross over and then for tempera-



FIG. 1. Curve 1: resistivity vs temperature at P=0; Curve 2: resistivity vs temperature at P=5000 bars; Dashed line:  $\rho$ -vs-T curve at zero pressure, shifted at a rate of  $dT_C/dP=0.48$  K/kbar.



FIG. 2. Resistivity peak temperature vs pressure up to 5000 bars.

tures below the crossing temperature, the resistivity decreases with pressure, which corresponds to a negative pressure coefficient. (iii) The increase in the temperature of the insulator-metal transition with pressure is about 6 K for an applied pressure of 5 kbar.

For more accuracy in the determination of the resistivity pressure coefficient we have measured the resistivity variation with pressure for different values of regulated temperatures. The results are given in Fig. 3, where it is noted that  $\ln \rho$  varies linearly with pressure. The derived variations of the pressure coefficient of the resistivity,  $\alpha$ , with temperature are shown in Fig. 4,  $\alpha$  is positive in the paramagnetic and ferromagnetic temperature ranges. It exhibits a critical behavior near  $T_c$  with an inversion of sign and a large decrease below 60 K.

In order to test the model proposed in the following discussion, we have measured the Hall ef-



FIG. 3. Resistivity vs pressure at different regulated temperatures.



FIG. 4. Pressure coefficient of resistivity vs temperature.

fect under pressure in the paramagnetic region and the resistivity under uniaxial stress applied along the [100] crystallographic direction. For the Hall effect (80 K), the results are:

$$\begin{split} P &= 200 \text{ bars, } R_{H} = 0.48 \text{ cm}^{3}/\text{C}, \quad \sigma = 0.49 \ (\Omega \text{ cm})^{-1}, \\ P &= 2525 \text{ bars, } R_{H} = 0.52 \text{ cm}^{3}/\text{C}, \quad \sigma = 0.41 \ (\Omega \text{ cm})^{-1}, \\ P &= 5010 \text{ bars, } R_{H} = 0.52 \text{ cm}^{3}/\text{C}, \quad \sigma = 0.33 \ (\Omega \text{ cm})^{-1}. \end{split}$$

For the piezoresistance (300 K), the modulated uniaxial stress X was about  $2 \times 10^7$  dyn/cm<sup>2</sup> along the [100] axis, with a static stress of about 1.4  $\times 10^8$  dyn/cm<sup>2</sup>. When the current is parallel to the stress direction and parallel to the [100] direction, the measured piezoresistance coefficient is equal to

$$\pi_{11} = \frac{1}{X} \frac{d\rho}{\rho} = 1.4 \times 10^{-12} \text{ cm}^2/\text{dyn}$$

in the Smith notation.<sup>8</sup> The coefficient  $\pi_{12}$  can be determined by means of hydrostatic-pressure experiments using the relationship

$$d \frac{\ln \rho}{dP} = \pi_{11} + 2\pi_{12} = \alpha .$$

For our sample  $\pi_{12} = 29 \times 10^{-12} \text{ cm}^2/\text{dyn}$  at 300 K.

## **IV. DISCUSSION**

### A. Paramagnetic-temperature range

From the piezoresistance experiments described in Sec. III, the difference  $\pi_{11} - \pi_{12}$  is not zero. Thus, we can say from the work of Keyes,<sup>9</sup> that conduction occurs principally in a band centered along the [100] direction of the Brillouin zone. From the band calculation of  $Cho^{10}$  we may assume that it is a 5*d* band.

From Hall measurements in the paramagnetic range it appears that when hydrostatic pressure is applied the number of carriers remains constant, while the Hall-mobility product  $R_{\mu\sigma}$  decreases.

For the degenerate sample studied, some electrons may be in the upper conduction band which is, according to Cho,<sup>10</sup> a 6s band centered at the  $\Gamma$  point. When a pressure up to 5 kbar is applied, the relative position of the subbands is changed without any deformation of their shapes.

According to these explanations, the observed increase of resistivity under pressure may be explained by a transfer of electrons from a highermobility band to a lower-mobility band. Because these electrons transfer at the same energy in a degenerate sample, the ratio of mobilities in the two bands is

$$\mu_{6s}/\mu_{5d} = m_{5d}^*/m_{6s}^*$$

The effective masses for the 6s and 5d bands are  $m_{6s}^* = 0.2m_0$ ,<sup>11</sup> and  $m_{5d}^* = 0.4m_0$ .<sup>10</sup> Thus the mobility in the 5d band is smaller than that in the 6s band, which is in agreement with the above explanation of a positive value of the pressure coefficient of the resistivity. This explanation is consistent with a 6s-band pressure coefficient smaller in absolute value, or positive, compared to that of the 5d band.

The behavior explained above is not in disagreement with the negative values of the pressure coefficient of the resistivity obtained on other samples.<sup>1-3</sup> In fact for stoichiometric EuO, only the 5d band is occupied, and near room temperature these samples exhibit an activation energy in their resistivity. When a hydrostatic pressure is applied, the 5d band is lowered at a rate of -4.4meV/kbar,<sup>12</sup> the activation energy decreases at the same rate,<sup>1-3,13</sup> and thus the number of carriers activated into the conduction band is increased, which leads to a negative pressure coefficient of the resistivity (samples M2, M4, F5, Ref. 3).

When the Eu concentration is increased the activation energy of the resistivity is decreased<sup>3,14</sup> while the 6s band becomes populated. When a pressure is applied the resistivity variation is a result of two effects: an increase in carrier density and a decrease of mobility by transfer of electrons from the 6s to the 5*d* band. Thus the pressure coefficient of the resistivity can be slightly negative (sample F4, Ref. 3), positive (present work), or zero (sample M3, Ref. 3) depending on the mobility difference of the two bands at the Fermi level.

A		Pressure coefficient of $T_C$ derived from susceptibility measurements (K/kbar)	Pressure coefficient of $T_C$ derived from Grüneisen- constant measurement (K/kbar)
	Sokolova <i>et al</i> .	$0.4 \pm 0.1$	
	(Ref. 15)		0.24
	(Ref. 16)		0.04
	Stevenson et al.	0.46	
	(Ref. 17)		
	Mc Whan et al.	0.37	
	(Ref. 18)		

TABLE I. Different values of  $\triangle$  from susceptibility and Grüneisen-constant measurements.

## B. Resistivity-peak temperature range

From Fig. 1 one can see that the resistivity peak is shifted by pressure towards higher temperatures at a rate of 0.48 K/kbar. The shift is of the same order of magnitude as the shift of  $T_{c}$  under pressure determined by different authors<sup>15-18</sup> and listed in Table I. Assuming that the resistivityversus-temperature curve is shifted in the same way as  $T_C$  by pressure, one obtains the dashed curve in Fig. 1. For every temperature, the calculated values of  $\alpha = d \ln \rho / dP$  are represented on Fig. 4 by the dashed line. The shifted curve of Fig. 1 and the experimental curve merge into one another between 60 and 90 K, thus the values of  $\alpha = d \ln \rho / dP$  calculated from the  $\rho$ -vs-T curve at zero pressure and the shifted one are in good agreement with experimental values in this temperature range.

From this analysis it can be said that the behavior of resistivity and the shift of  $T_c$  under pressure are basically due to the same physical phenomenon, i.e., the effect of pressure on magnetic interactions. The simple shift of the  $\rho$ -vs-T curve is in fact sufficient to explain the inversion of the sign of the pressure coefficient of the resistivity,  $\alpha = d \ln \rho / dP$ .

## C. Insulator-metal transition

Near 50 K, the shift of the  $\rho$ -vs-T curve is not enough to explain the large decrease in resistivity under pressure as in Figs. 1 and 4, which show a large minimum of the pressure coefficient of the resistivity. This can be explained in the following way: for temperatures below 50 K, the Eu<sup>2+</sup> 4f spins are all parallel (ordered phase) and the electrons are free. When the temperature increases the Eu<sup>2+</sup> 4f spins become disordered. In order to keep the crystal ordered, the free electron couples its spin with the Eu<sup>2+</sup> spins, then loses some en-

ergy and becomes "self-trapped" on magnetic polarons.<sup>19-24</sup> The number of free carriers therefore decreases. The self-trapping of electrons has been pointed out previously by Shapira et al.25 For their sample 4A, which is similar to that studied in the present paper, the self-trapping of electrons occurs at 45 K for an applied magnetic field of 25 kOe. When a greater field is applied the trapping occurs at higher temperatures (60 K). This behavior is explained by the fact that the magnetic field reorders these Eu<sup>2+</sup> spins, which frees the electrons. Pressure has the same effect on carrier density because it increases the magnetic interactions (see the shift of  $T_{c}$ ). When pressure is applied, the self-trapping occurs at higher temperatures. The effect of pressure on the mobility and carrier density is illustrated in Fig. 5.



FIG. 5. Schematic diagram of the pressure effect on the mobility and carrier density in the self-trapping temperature range.

For a given temperature the conductivity at zero pressure is  $\sigma_0 = (en_0\mu_0)^{-1}$  and at pressure p it is  $\sigma_p = (en_p \mu_p)^{-1}$ . Because  $n_p > n_0$  and  $\mu_p > \mu_0$ , it follows that  $\sigma_p \gg \sigma_0$  or  $\rho_p \ll \rho_0$ . This scheme shows that pressure acts not only on the mobility, as shown in the resistivity-peak region, but also on the carrier density near the temperature of the insulator-metal transition. This explains the important decrease of the resistivity with pressure for temperatures below 60 K.

### D. Low-temperature range

At very low temperature, all the Eu<sup>2+</sup> 4f spins are ordered, and the electrons are free. The effect of pressure on the mobility of each band is negligible ( $T \ll T_C$ ). The only effect of pressure on the resistivity is an effect on carriers which are transferred from the 6s band to the 5d band, giving an apparent change in total mobility. This explanation is confirmed by the fact that the pressure coefficient of the resistivity has the same value as at high temperatures.

#### V. CONCLUSION

The pressure coefficient of the resistivity has been measured in a large temperature range (6– 300 K). A critical behavior of the pressure coefficient of the resistivity,  $\alpha$ , has been observed around  $T_c$  because  $T_c$  is shifted by pressure towards high temperatures. In addition, around the insulator-metal transition temperature, the pressure effect on the self-trapping of electrons on magnetic polarons must be taken into account to explain the large absolute value of  $\alpha$ .

In the paramagnetic and ferromagnetic phases, the pressure coefficient of the resistivity is positive for the type of sample studied, because the electrons are transferred from the 6s band to a lower-mobility 5d band.

## ACKNOWLEDGMENT

The authors are grateful to Dr. J. P. Nadai for enlightening discussions.

\*Associé au CNRS.

- <sup>1</sup>M. R. Oliver, Ph.D. thesis (MIT, 1970) (unpublished).
- <sup>2</sup>M. R. Oliver, J. A. Kafalas, J. O. Dimmock, and T. B. Reed, Phys. Rev. Lett. <u>24</u>, 1064 (1970).
- <sup>3</sup>J. P. Desfours, J. P. Lascaray, C. Llinares, and M. Averous, Solid State Commun. 21, 441 (1977).
- <sup>4</sup>C. F. Guerci and M. W. Shafer, J. Appl. Phys. <u>37</u>, 1406 (1966).
- <sup>5</sup>M. Averous, J. Calas, C. Fau, and J. Bonnafe, J. Phys. (Paris) 37, 1347 (1976).
- <sup>6</sup>G. Weill, thesis (Université d'Orsay, 1977) (unpublished).
- <sup>7</sup>J. Calas, thesis (Université des Sciences et Techniques du Languedoc, 1974) (unpublished).
- <sup>8</sup>C. Smith, Phys. Rev. <u>94</u>, 42 (1954).
- <sup>9</sup>R. W. Keyes, Solid State Phys. <u>11</u>, 149 (1960).
- <sup>10</sup>S. J. Cho, Phys. Rev. B <u>1</u>, 4589 (1970).
- <sup>11</sup>E. Kaldis, J. Schoenes, P. Wachter, Proceedings of the International Conference on Magnetism, Chicago, 1971 (unpublished).
- <sup>12</sup>P. Wachter, Solid State Commun. 7, 693 (1969).
- <sup>13</sup>G. K. Sokolova, A. A. Samokhvalov, K. P. Rodionov, Sov. Phys. Solid State <u>13</u>, 1042 (1971).

- <sup>14</sup>C. Llinares, J. P. Desfours, J. P. Nadai, C. Godart, A. Percheron, and J. C. Achard, Phys. Status Solidi A <u>25</u>, 185 (1974).
- <sup>15</sup>G. K. Sokolova, K. M. Demchuk, K. P. Rodionov, and A. A. Samokhvalov, Sov. Phys. JETP <u>22</u>, 317 (1966).
- <sup>16</sup>B. E. Argyle, N. Miyata, T. D. Schultz, Phys. Rev. 160, 413 (1967).
- <sup>17</sup>R. Stevenson and M. C. Robinson, Can. J. Phys. <u>43</u>, 1744 (1965).
- <sup>18</sup>D. B. McWhan, D. B. Soners, G. Jura, Phys. Rev. <u>143</u>, 385 (1966).
- <sup>19</sup>T. Kasuya, A. Yanase, Rev. Mod. Phys. <u>40</u>, 684 (1968).
- <sup>20</sup>T. Kasuya, Solid State Commun. <u>18</u>, 51 (1976).
- <sup>21</sup>M. R. Oliver, J. O. Dimmock, A. L. McWhorter, and T. B. Reed, Phys. Rev. B 5, 1078 (1972).
- <sup>22</sup>M. W. Shafer, J. B. Torrance, and T. Penney, J. Phys.
- Solids <u>33</u>, 2251 (1971). <sup>23</sup>J. B. Torrance, M. W. Shafer, and T. R. McGuire,
- Phys. Rev. Lett. 29, 1186 (1972).
- <sup>24</sup>P. Leroux Hugon, Phys. Rev. Lett. <u>29</u>, 939 (1972).
- <sup>25</sup>Y. Shapira, S. Foner, and T. B. Reed, Phys. Rev. B <u>8</u>, 2299 (1973).