

Is the organic metal $(\text{ET})_4\text{Hg}_3\text{Br}_8$ a quasi-2D superconductor?

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Two classes of organic superconductors have now been identified: the quasi-1D salts tetramethyltetraselenafulvalene [TMTSF (Bechgaard's salts) D. Jerome and H. I. Schultz, *Adv. Phys.* **31**, 299 (1982)] and the quasi-2D salts *bis* (ethylenedithiolo) tetrathiofulvalene (ET) {S. S. Parkin *et al.*, *Phys. Rev. Lett.* **50**, 270 (1983); I. F. Schegolev *et al.*, *Mol. Cryst. Liq. Cryst.* **126**, 365 (1985); I. M. Williams *et al.*, *Inorg. Chem.* **23**, 3839 (1984); R. N. Lyubovskaya *et al.*, *Pis'ma Zh. Eksp. Teor. Fiz.* **42**, 380 (1985) [JETP Lett **42**, 468 (1985)]}.

The significant anisotropy in the properties of the salts of the first class allows an interpretation of their low-temperature behavior in terms of strong superconducting fluctuations at temperatures significantly higher than the transition temperatures T_c which are actually observable.¹ However, we do not yet have any convincing data for a 1D nature of the superconductivity in the salts of this class, since all of their superconducting properties can be explained by the ordinary BCS theory.⁶ The salts of the second class, with essentially quasi-2D structures and properties, are also 3D superconductors,⁶ despite their pronounced anisotropy. In this letter we report the properties of the first ET salt with an exchange anion $[\text{Hg}_3\text{Br}_8]^{-2}$ which goes superconducting at atmospheric pressure and which is apparently a quasi-2D superconductor.

We have found that at least three phases exist in the system of ET with the brommercurate anion. One of these phases is an insulating phase even at room temperature; another becomes an insulator below ~ 180 K; and the third, with the composition¹⁾ $(\text{ET})_4\text{Hg}_3\text{Br}_8$, remains a metal as $T \rightarrow 0$. The crystals of the latter phase are generally rhombi with typical dimensions of 0.5–2 mm along their diagonals and typical thicknesses of 0.1–0.5 mm.

The resistivity of the crystals in the transverse direction, measured directly for four of the samples by pasting two electrodes to each plane, lies in the range $(1.5-3) \times 10^2 \Omega \cdot \text{cm}$ at $T = 300$ K. The temperature dependence of the resistivity is shown by curve 1 in Fig. 1. The behavior $\rho_{\perp}(T)$ is nonmetallic over the temperature range 18–300 K.

The longitudinal resistivity ρ_{\parallel} is measured by placing electrodes on one plane of the crystal; its values for ten crystals for which measurements were carried out lie between 0.2 and $2\Omega \cdot \text{cm}$ at $T = 300$ K. The anisotropy in the plane of the crystal does not exceed a factor of two or three. Curve 2 in Fig. 1 shows the temperature dependence of ρ_{\parallel} . We see that this dependence is metallic over the entire temperature range.

The qualitative difference in the behavior of $\rho_{\parallel}(T)$ and $\rho_{\perp}(T)$ indicates that the

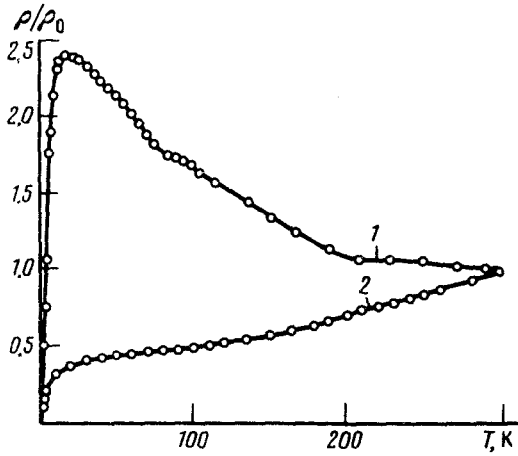


FIG. 1. Resistivities ρ_{\parallel} and ρ_{\perp} versus the temperature.

anisotropy of the conductivity of the $(\text{ET})_4\text{Hg}_3\text{Br}_8$ crystals increases substantially as the temperature is lowered. The large value of ρ_{\parallel} (300 K) and its pronounced scatter among the various crystals may be caused in part by the circumstance that in calculating the conductivity from the measured resistivity we ignored the particular features of the current spreading in highly anisotropic materials. Consequently, the values given here for ρ_{\parallel} (300 K) should probably be regarded as estimates. The actual value of the longitudinal-transverse anisotropy $\rho_{\perp}/\rho_{\parallel}$ is apparently considerably larger than its apparent value $\sim 10^3$.

Figure 2 shows low-temperature parts of the resistivity curves. Comparison of the

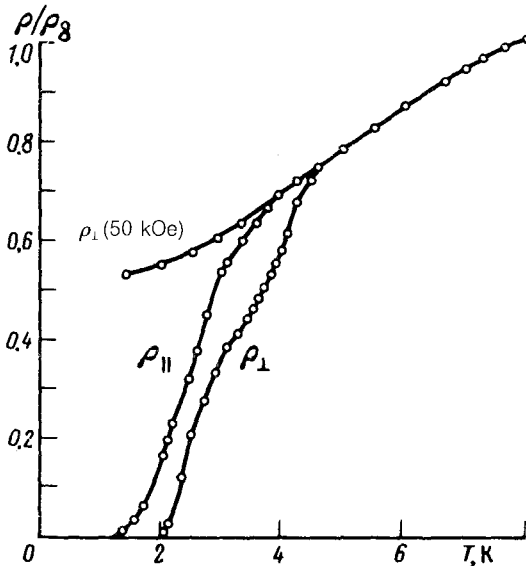


FIG. 2. Temperature dependence of ρ_{\parallel} and ρ_{\perp} near the superconducting transition; effect of a magnetic field (50 kOe) on the resistivity ρ_{\perp} (curve 1).

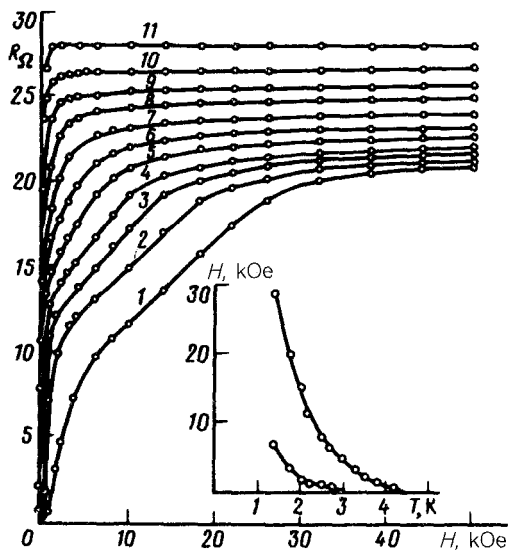


FIG. 3. The resistivity R_1 versus the magnetic field H_1 over the temperature interval 1.4–4.22 K. 1—1.4 K; 2—1.77 K; 3—2.04 K; 4—2.19 K; 5—2.51 K; 6—2.74 K; 7—3.01 K; 8—3.3 K; 9—3.53 K; 10—3.81 K; 11—4.22 K. The inset is an H - T diagram for the two superconducting transitions.

behavior of the resistivity ρ_1 in a zero field and in a field $H = 50$ kOe, directed perpendicular to the plane of the crystal, shows that the superconducting transition begins at $T \approx 4.5$ K. For ρ_1 , the transition is completed at $T = 2$ K. For ρ_1 the transition begins at a slightly lower temperature ($T \approx 4$ K) and is completely ended at $T = 1.4$ K. The fact that the transitions occur over a significant temperature interval and the slight change in slope on the $R(T)$ curves near 3 K are evidence of an inhomogeneity of the sample, which is a consequence of the existence of either two superconducting phases or regions with significant internal stresses which reduce the value of T_c .

The possible existence of two phases is also suggested by an analysis of the curves illustrating the disruption of the superconductivity by a magnetic field, i.e., the curves of $R_1(H_1)$ (H_1 is the field perpendicular to the plane of the crystal), shown in Fig. 3. Over the temperature interval from 4.2 to 3 K, there is a single change in slope on the curves, which probably corresponds to a high-temperature superconducting transition. At lower temperatures, at weak fields, yet another change in slope occurs; this one corresponds to a second superconducting transition. Under the assumption that the position of the change in slope on these curves characterizes the critical field H_{c2} at the given temperature, we can construct curves of $H_{c2}(T)$ for both transitions. The results are shown in the inset in Fig. 3.

The $(\text{ET})_4\text{Hg}_3\text{Br}_8$ lattice is isostructural with the lattice of the compound $(\text{ET})_4\text{Hg}_3\text{Cl}_8$, studied previously,⁵ and the plane of the crystal is therefore the plane of the conducting layers of ET molecules. In this case, the field H_1 will be an easy-direction field. Interestingly, the magnitude of H_{c2} in the case of the high-temperature transition is significantly greater than the corresponding values of H_{c2}^* in other ET salts.⁷ The temperature dependence $H_{c2}(T)$ in our case is also somewhat unusual: It has a positive curvature, and it has no significant linear region at $T \lesssim T_c$.

These results could hardly be a consequence of the particular method used to

determine the values of $H_{c2}(T)$, since the same method leads to the same results⁸ in the case of β - ET_2I_3 , for which the results were subsequently confirmed by an ordinary measurement of $H_{c2}(T)$ in homogeneous samples.⁹ The probable explanation is the pronounced anisotropy of the $(\text{ET})_4\text{Hg}_3\text{Br}_8$ crystals, which is also manifested in the qualitative difference in the behavior of ρ_{\perp} and ρ_{\parallel} as functions of the temperature, mentioned above. We are thus apparently dealing with a quasi-2D metal, whose transverse conductivity should vanish in the limit $T \rightarrow 0$. The observed decrease in ρ_{\perp} as the temperature is reduced below 18 K may be a consequence of transverse "short circuits" in this case.

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¹The composition was determined from x-ray structural data by T. G. Takhirov, O. A. D'yachenko, and L. A. Atovmyan.

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