Superconducting properties of β-(BEDT-TTF)$_2$I$_3$ crystals

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The temperature dependence and anisotropy of the critical magnetic field in an organic superconductor β-(BEDT-TTF)$_2$I$_3$ are measured. The value of H_{c2} when $\overline{H}\,\|\overline{a}$ is approximately equal to the value of H_{c2} when $\overline{H}\,\|\overline{b}$ and exceeds H_{c2} by almost an order of magnitude when $\overline{H}\,\|\overline{c}$. The state density is estimated to be $N(0)\cong 0.25\times 10^{34}\ \mathrm{erg}^{-1}\cdot\mathrm{cm}^{-3}$. The critical temperature of the superconducting transition increases from 1.3 K to 4.5 K as a result of thermal cycling from liquid-helium temperature to room temperature. The sample goes superconducting at $T\cong 7$ K.

The observation of a superconducting transition in a new organic metal (BEDT-TFF)$_2$I$_3$, the structure of this organic material, and its critical magnetic fields were

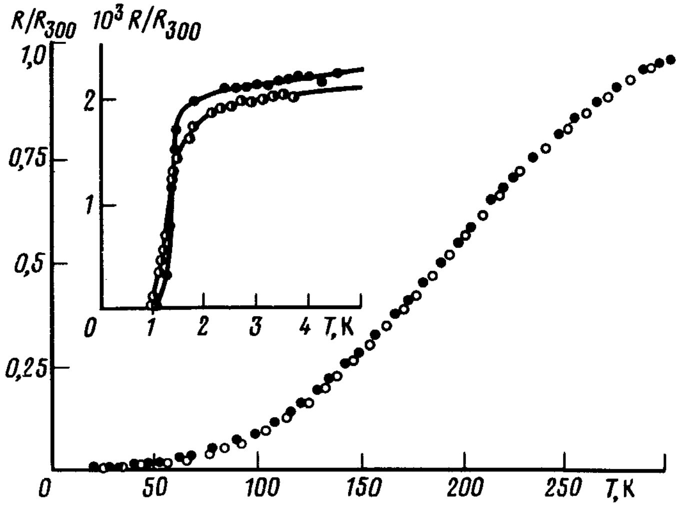

FIG. 1. Temperature dependence of the resistance of two β-(BEDT-TTF)$_2$I$_3$ samples. The inset shows the superconducting transitions of two samples recorded during the first cooling cycle.

reported in previous experimental studies.[1–3] As pointed out by the authors of these studies, various modifications, which differ in structure and in superconducting-transition temperature, can be produced by changing the conditions under which single crystals are synthesized and grown. For example, samples of a triclinic modification β-(BEDT-TTF)$_2$I$_3$ have a superconducting-transition temperature $T_c = 1.5$ K and samples of orthorhombic modification γ-(BEDT-TTF)$_2$I$_3$ have $T_c = 2.5$ K. In both cases, the superconducting transition occurs at standard atmospheric pressure.

We have studied the critical magnetic fields of β-(BEDT-TTF)$_2$I$_3$ crystals, which were grown in the shape of needles with typical dimensions $0.01\times0.05\times2$ mm. The magnetic field at which the resistance of the sample is equal to $0.5R_{\text{tip}}$ was assumed to be the critical field at a given temperature. The electrical resistance was measured by a four-contact method with a direct current ($I = 100\ \mu$A). The resistance along the direction of the $\bar{a}$ axis was measured in all experiments. The average conductivity of the crystals was $\sigma_{300} \simeq 30$ mho/cm at room temperature.

Figure 1 shows the resistance of two samples plotted as a function of temperature. The measurements were taken in successive cycles of cooling the samples from room temperature to liquid-helium temperature. The ratio of the resistance at room temperature to liquid-helium temperature is $R_{300}/R_{4.2} = 400$. The temperature dependences of $R(T)/R_{300}$ for various samples which were measured in several experiments are reproduced well in the temperature interval 300–8 K. In the temperature interval 8–80 K, we have $R(T) \propto T^2$ and above 200 K we have $R(T) \propto T$. The curves for the superconducting transitions in both samples obtained during the first cooling cycle are shown in the inset in Fig. 1. The superconducting transitions occur rather abruptly in both samples at the critical temperature of 1.3 K.

The curves for superconducting transitions in the same sample which were obtained in five experiments are shown in Fig. 2. The sample was warmed to room

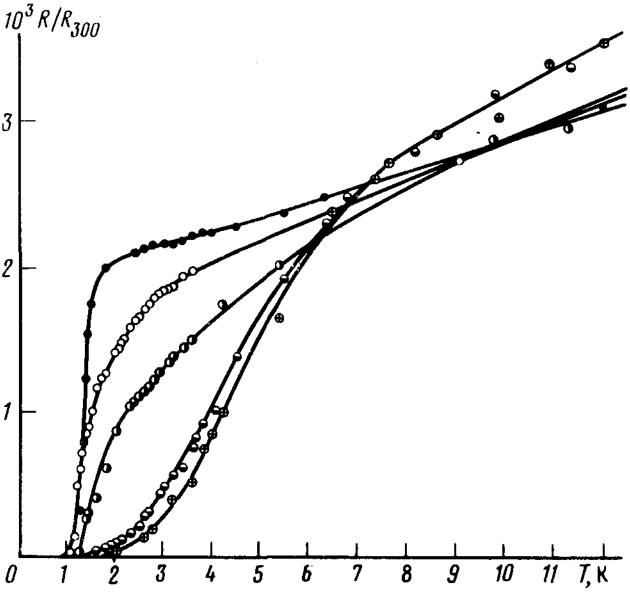

FIG. 2. Curves for the superconducting transitions of one sample, recorded during five cooling cycles. The curves are indexed in the order in which they were recorded.

temperature between two successive experiments. We see that the critical temperature T_c rises from 1.3 K in the first cycle to 4.5 K in the fifth cycle after each cooling cycle. The sample goes superconducting at $T \simeq 7$ K.

The anisotropy of the critical magnetic fields was studied by making use of a sample rotator on which a sample was mounted. The use of this arrangement allowed us to measure in a single experiment the dependence of the critical field on the angle of rotation only in one plane. To obtain a similar dependence in another plane, we had to warm the insert to room temperature and change the position of the module with the samples on the sample rotator. The superconducting-transition temperature, as we have mentioned, shifts slightly in this case.

Figure 3 shows the electrical resistance plotted as a function of the angle of rotation of the sample in a magnetic field. Because of the critical dependence of the resistance of the sample on the magnetic-field direction, the sample can be oriented in such a way that the preferred axis will be parallel to the magnetic field. Shown also in this figure is a plot of the critical magnetic field as a function of the angle of rotation in the *ac* plane; the $\bar{b}$ axis in this case remains perpendicular to the magnetic-field direction. At 0.5 K, the critical magnetic fields measured along the $\bar{a}$ and $\bar{c}$ axes are 14 and 2 kOe, respectively. Figure 4 shows the critical magnetic fields plotted as a function of temperature for a sample whose superconducting-transition temperature is 1.3 K and the magnetic field is directed along the $\bar{a}$ and $\bar{c}$ axes in the first experiment and for the same sample after its superconducting-transition temperature increases to 1.5 K and the magnetic field is directed along the $\bar{b}$ axis in the second experiment. The absolute values of the critical magnetic fields directed along the $\bar{a}$ and $\bar{b}$ axes unfortunately cannot be compared, since they were measured in two different experiments, and since

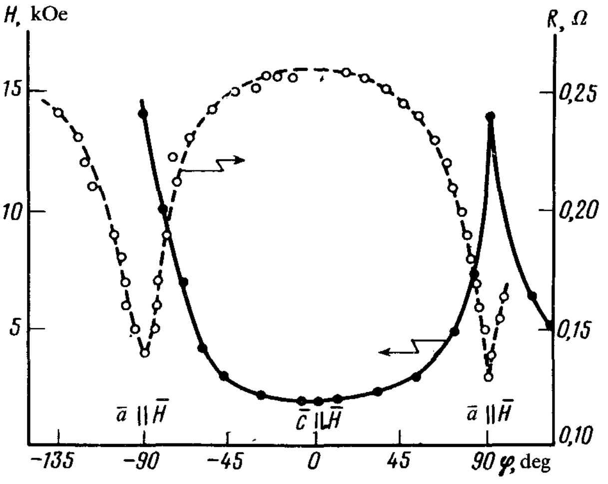

FIG. 3. Dependence of the resistance of the sample on the angle of rotation around the $\bar{b}$ axis in the ac plane, in a constant field $H = 14$ kOe at $T = 0.5$ K (dashed curve). Dependence of the critical magnetic field on the angle of rotation in the ac plane at $T = 0.5$ K (solid curve).

the superconducting-transition temperature was higher in the second experiment. The derivatives $\left.\frac{dH_{c2}}{dT}\right|_{T_c}$ of the critical magnetic fields directed along $\bar{a}$ and $\bar{b}$ axes are, however, in approximate agreement with each other, by analogy with the data of Ref. 3 for γ-(BEDT-TTF)$_2$I$_3$. The data in Fig. 4 can be used to estimate the values of $\left.\frac{dH_{c_2}}{dT}\right|_{T_c}$, which correspond to the directions $\bar{a}$, $\bar{b}$, $\bar{c}$. These values are ~37 kOe/K, ~37 kOe/K, and ~2.8 kOe/K, respectively. The data available can be used to estimate the electronic state density at the Fermi surface. For an anisotropic superconduc-

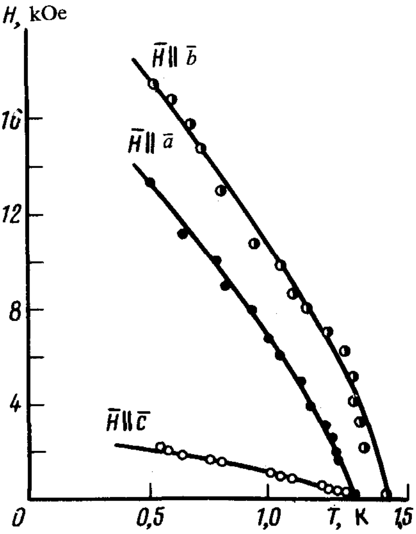

FIG. 4. The dependences $H_{c2}(T)$ obtained for three orientations of the sample with respect to the magnetic field: $\bar{H} \parallel \bar{a}$, $\bar{H} \parallel \bar{b}$, and $\bar{H} \parallel \bar{c}$.

tor, an expression which establishes a relationship between the derivatives $\left.\frac{dH_{c_2}}{dT}\right|_{T_c}$, measured along the corresponding crystallographic axes, the components of the conductivity tensor and the electronic state density is given by[5]

$$\frac{H'_b H'_c}{H'_a} = \frac{1{,}9\,K_b\ \Phi_0}{\hbar D_a}\ ,\quad \sigma_i = 2e^2 D_i N(0)\,, \tag{1}$$

where $H'_i = \left.\frac{dH_{c2}}{dT}\right|_{T_c}$ is directed along the $i = \bar{a}, \bar{b}, \bar{c}$ axis; K_b is the Boltzmann constant; Φ_0 is a fluxoid; and D_i is the coefficient of diffusion along the i axis. Assuming that $H'_a \simeq H'_b$ and substituting into (1) the values $H'_c \cong 2.8$ kOe/K and $\sigma_a \cong 2\times 10^4$ mho/cm, we find $N(0) \cong 0.25\times 10^{34}$ erg$^{-1}\cdot$cm^{-3}. This value is in order-of-magnitude agreement with a corresponding value for $(TMTSF)_2ClO_4$ (Ref. 4).

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[1] É. B. Yagubskiĭ, I. F. Shchegolev, V. N. Laukhin, P. A. Kononovich, M. V. Kartsovnik, A. V. Zvarykina, and L. I. Buravov, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 12 (1984) [JETP Lett. **39**, 12 (1984)].

[2] V. F. Kaminskiĭ, T. G. Prokhorova, R. P. Shibaeva, and E. B. Yagubskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 15 (1984) [JETP Lett. **39**, 17 (1984)].

[3] É. B. Yagubskiĭ, I. F. Shchegolev, S. I. Pesotskiĭ, V. N. Laukhin, P. A. Kononovich, M. V. Kartsovnik, and A. V. Zvarykina, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 275 (1984) [JETP Lett. **39**, 328 (1984)].

[4] P. Garoche, R. Brusetti, and K. Bechgaard, Phys. Rev. Lett. **49**, 1346 (1982).

[5] A. I. Buzdin and L. N. Bulaevskiĭ, Usp. Fiz. Nauk **144**, 3 (1984) [Sov. Phys. Usp. **27**, No. 11 (1984)].

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