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SUPERCONDUCTING TRANSITION OF $(TMTSF)_2ClO_4$ IN MAGNETIC FIELDS

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Superconducting transition of $(TMTSF)_2ClO_4$ was studied by conductivity measurements along the most conductive a-axis in magnetic fields applied along three different principal crystallographic axes. The GL coherence lengths at 0 K along the a-, b-, and c*-axes, $\xi_a(0) \gtrsim 600$ A, $\xi_b(0) \cong 540$ A, and $\xi_c^*(0) \cong 60$ A were obtained from the measurements of the temperature dependence of the upper critical field H_{c2} near the transition temperature. The anisotropy is discussed in terms of the dimensionality arising from the crystal structure and of the conductivity anisotropy in the normal state.

Since the discovery of superconductivity in the quasi-one-dimensional (1D) organic conductors [1-3] of the family di-(tetramethyltetraselenafulvalene)X, $(TMTSF)_2X$, ($X = ClO_4$, PF_6, etc.), these materials attracted interest from the viewpoint of the nature of superconductivity. The purpose of this paper is to report the anisotropic Ginzburg-Landau (GL) coherence lengths along three principal crystallographic axes obtained from the temperature dependence of the upper critical fields, H_{c2}, in $(TMTSF)_2ClO_4$. In this material since the conductivity is highly anisotropic, it is interesting to study the anisotropy of superconducting nature.

The single crystals used in the present study were obtained from the electrochemical oxidation of TMTSF in ultrapure 1,1,2-trichloroethane containing Bu_4NClO_4 in the constant current condition.[4] Typical sizes of the samples used in this study were 5, 0.1, and 0.08 mm for the a-, b-, and c*-directions, respectively.

Four terminal contacts were made with silver paste, DuPont 4922. Contact resistances were typically 20 ohms at room temperature and less than 100 ohms at liquid helium temperatures. Low dc currents of $1 \sim 50\,\mu A$ were supplied along the most conductive a-axis. Samples were placed in a copper chamber which was attached to ^3He evaporator and filled with helium exchange gas.

It was noted that in spite of careful cooling of the samples, e.g. 3 deg/h, and of stability in the measuring systems, sudden increases in resistance along the a-axis were observed frequently. We should note, however, that the phenomenon of the discontinuous change in resistance was not observed when we measured the resistance along the c*-axis.[5] This indicates that the material seems to be very fragile along the a-axis but not along the c*-axis.

Figure 1 shows the superconducting transition in our sample. The transition temperature T_c was around 1.0 ± 0.05K varying slightly from sample to sample. At temperatures

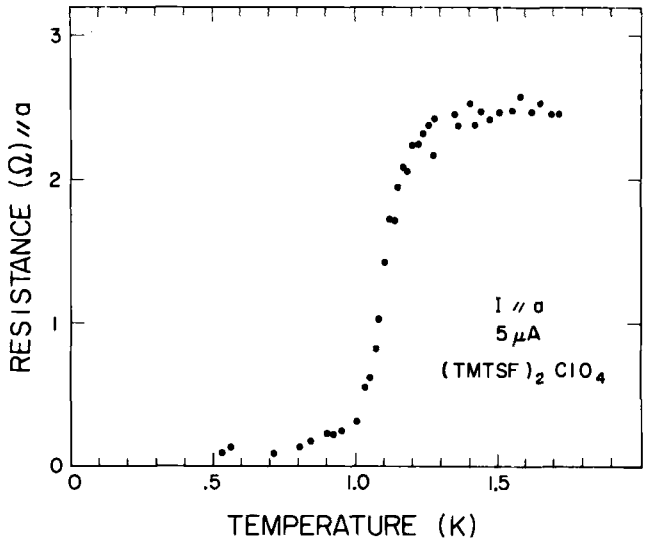

FIGURE 1 Superconducting transition of $(TMTSF)_2ClO_4$. Finite normal resistance can be seen below $T_c \cong 1.05$ K.

below T_c, there remained finite normal resistances showing ohmic behavior, which may be caused by thermally induced microcracks during cooling. The resistance ratio $R(T \ll T_c)/R(T \gtrsim T_c)$ and the variation of T_c seems to be related with the cooling process between 300 K and 1 K.

In Fig. 2 is shown the suppression of the superconductivity by magnetic fields applied parallel to the c*-axis. Because of the fragile nature during cooling we used a new sample in each experiment for different direction of the magnetic field. The temperature dependence of H_{c2} for three different directions is shown in Fig. 3, determined at the half of the difference between the normal state resistance and the finite normal resistance below T_c. By using the relations,

$$dH_{c2_i}/dT\big|_{T=T_c} = \phi_0/[2\pi\xi_j(0)\xi_k(0)T_c], \tag{1}$$

$$\xi(T) = \xi(0)[T_c/(T_c - T)]^{\frac{1}{2}}, \tag{2}$$

where ϕ_0 is a flux quantum and i, j, and k denote different axes, we obtained the GL coherence lengths at 0 K along the a-, b-, and c*-axes, to be $\xi_a(0) \gtrsim 600$ A, $\xi_b(0) \cong 540$ A, and $\xi_{c*}(0) \cong 60$ A, respectively.

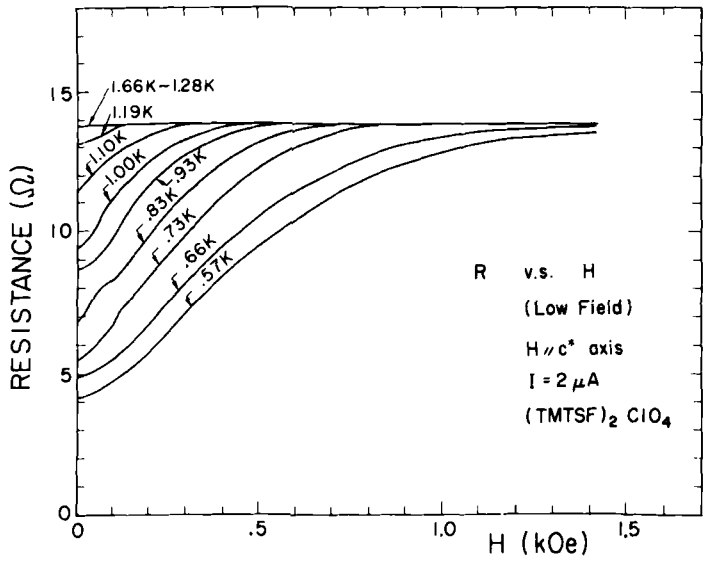

FIGURE 2 Suppression of superconductivity under magnetic field applied parallel to the c*-axis.

These values are smaller than those of $(TMTSF)_2PF_6$; $\xi_{//}(0) = 3750$ A and $\xi_{\perp}(0) = 250$ A.[6] (In $(TMTSF)_2PF_6$ two coherence lengths perpendicular to the 1D axis were not distinguished yet.) In the particular samples examined here the GL coherence lengths are nearly equal within the ab-plane in which TMTSF molecules are positioned within a plane, whereas the coherence length along the c*-axis is an order of magnitude smaller. Along the c*-axis the sheet of the TMTSF molecules are separated by counter ions, ClO_4.

If the BCS coherence length ξ_0 is assumed to be the same for all directions, the GL coherence length is given by,

$$\xi_i(0) = 0.87(\xi_0 \ell_i)^{\frac{1}{2}}, \qquad (3)$$

where ℓ_i is the electron mean-free-path in the i direction.

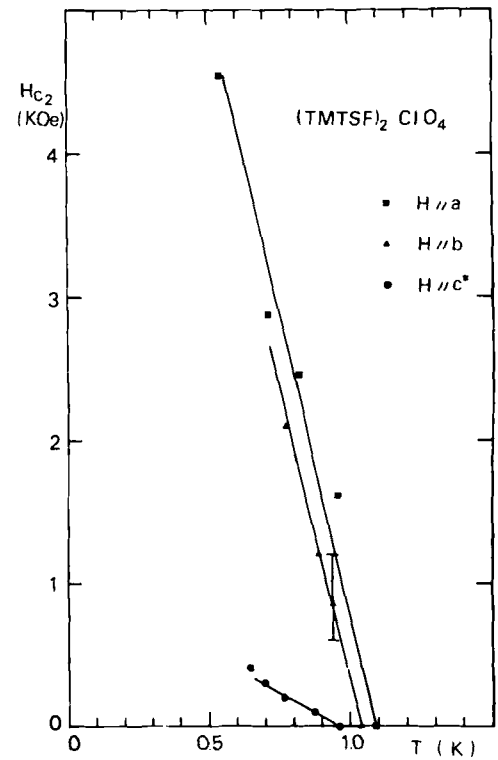

FIGURE 3 Upper critical field, H_{c2}, as a function of temperature. The GL coherence lengths, $\xi_a(0)$, $\xi_b(0)$, and $\xi_{c*}(0)$, are estimated from the curves using Eqs. (1) and (2).

The ratio of $\xi_1(0)$ along the a-, b-, and c*-directions is given by,

$$\xi_a(0):\xi_b(0):\xi_{c*}(0) = \ell_a^{\frac{1}{2}}:\ell_b^{\frac{1}{2}}:\ell_{c*}^{\frac{1}{2}}$$

$$= \sigma_a^{\frac{1}{2}}:\sigma_b^{\frac{1}{2}}:\sigma_{c*}^{\frac{1}{2}}. \qquad (4)$$

The conductivity anisotropy ratios at room temperature obtained by Montgomery method[7] were $\sigma_a/\sigma_b \cong 25$, and $\sigma_a/\sigma_{c*} \cong 940$. The ratio tended to increase with decreasing temperature, e.g. the ratio σ_a/σ_b increases from 25 at 300 K to 66 at 90 K. It is expected that $\xi_a(0)$ is much larger than $\xi_b(0)$, contrary to the experimental results that $\xi_a(0)$ and $\xi_b(0)$ are similar in magnitude. This may be ascribed to the induced microcracks which limit the electron mean-free-path along the a-axis at low temperatures, reducing the GL coherence length. Another possibility is a misalignment of the relative directions of magnetic fields, 5 deg, against the crystallographic axes.

We should briefly mention about the experiment on superconductivity along the c*-axis,[5] which is the least conductive in the normal state. We obtained voltage vs. current curve typical of zero resistance superconductor along the c*-axis, showing evidently the three dimensional (3D) ordering of superconductivity in detectable strength in $(TMTSF)_2ClO_4$. This is consistent with the fact that the coherence length $\xi_{c*}(T) \geq \xi_{c*}(0) \cong 60$ A exceeds the spacing, 13 A, between sheets of TMTSF molecules, which are isolated by counter ions ClO_4. The 3D ordering may suppress the superconducting fluctuations along the a-axis to some extent or induce dimensional cross over since the spacing and the coherence length are the same order of magnitude.

In conclusion we evaluated the coherence length along three principal axes of $(TMTSF)_2ClO_4$ with T_c around 1 K. The coherence lengths along the most and next conductive axes were of comparable magnitude, which is ascribed to the limitation in the electron mean-free-path caused by thermally induced microcracks. The coherence length along the least conductive c*-axis was an order of magnitude smaller, indicating the anisotropic nature of superconductivity in the material.

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