INFLUENCE OF MAGNETIC FIELD AND HIGH PRESSURE ON T_C OF THE ORGANIC SUPERCONDUCTOR $(BEDT-TTF)_4Hg_{2.89}Br_8$

R.N. Lyubovskaya, R.B. Lyubovskii, M.K. Makova, and S.I. Pesotskii

Institute of Chemical Physics, USSR Academy of Sciences, Chernogolovka, 142 432 USSR

Abstract. The upper critical fields H_{C2}^{ab} and H_{C2}^{C*} in magnetic fields up to 150 kOe and the T_c on pressure (up to 35 kbar) in the organic superconductor $(BEDT-TTF)_4Hg_{2.89}Br_8$ have been investigated. The presence of the positive curvature for H_{C2} and the 5-fold exceeding of the Clogstone paramagnetic limit for H_{C2}^{ab} have been observed. The strong electron-phonon coupling is suggested. The positive value for dT_c/dp has been found at pressures below 10 kbar. The sample undergoes a semiconducting transition at pressures above 25 kbar.

Introduction

Bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF) -based organic superconductors are divided into two groups, those with linear (I_3, Br_2, AuI_2, $Cu(NCS)_2$) and bulky (ReO_4, $Hg_{3-\delta}Cl_8$, $Hg_{3-\delta}Br_8$) anions . The main specific feature of superconductors with halide mercurate anions is the existence of two incommensurable sublattices; one of them incorporates BEDT-TTF molecules and Br or Cl anions, the other Hg atoms [1,2].

The lattice-disorder-induced random potential is likely to cause the appearance of a number of peculiar properties, which reveal in the temperature dependence of conductivity [1,3], EPR spectra [4], magnetic susceptibility [4,5], and spin-lattice relaxation [5] and distinguish this family of superconductors among the others. Our recent study of the conductivity of the organic metal $(BEDT-TTF)_4Hg_{2.89}Br_8$ in the magnetic fields up to 50 kOe showed [2] a record value of the dH_{C2}^{a}/dT ~100 kOe/K derivative for organic superconductors and its tendency to the exceeding of the Clogstone paramagnetic limit. The present paper

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reports on the study of upper critical fields of the salt $(BEDT-TTF)_4Hg_{2.89}Br_8$ in magnetic fields up to 150 kOe and the pressure influence up to 35 kbar on the superconducting transition.

Experimental

The measurements were performed by the 4-probe dc-method. Small rhombic crystals were glued to the Pt wires of 10μ thick by the graphite paste. Critical fields were measured in the Bitter magnet in the International Laboratory of Wroclaw (Poland). The values of the second critical field (H_{C2}) were determined from the middle-points of superconducting transitions on the resistivity vs. temperature plots measured along c* in different magnetic fields. The dependence of the superconducting transition in the $(BEDT-TTF)_4Hg_{2.89}Br_8$ single crystals on the hydrostatic pressures up to 35 kbar were measured in the purposely designed high-pressure apparatus [6].

Results

The resistivity along the single crystal plane in the direction of the a axis is ρ^a (300 K) $\approx$ 0.1-0.5 Ohm.cm and becomes (5-10)-fold reduced on cooling down to 5 K. At T_c = 4.3 K the sample transits to the superconducting state (inset in Fig. 1). The resistance measured in the c* direction perpendicular to the crystal plane is (3000-6000)fold higher than ρ^a. It increases with the temperature decrease by 1.5-2 times and reaches its maximum at 20-50 K. Then the resistance decreases sharply till the superconducting transition.

From temperature dependences of upper critical fields it is seen that $H^{a}_{C2} \approx H^{b'}_{C2} \gg H^{c*}_{C2}$ for various directions of magnetic fields (Fig. 1,2). Thus, the anisotropy of critical fields in $(BEDT-TTF)_4Hg_{2.89}Br_8$ is of a well defined quasi-two-dimensional character as it should be expected from the crystal layered structure [1,2].

The resistivity vs. temperature plot in the temperature region below 20 K is depicted for various pressures in Fig. 3. The temperature of the superconducting transition is seen to shift to the region of higher temperatures with the pressure increase and it is ~ 6.8 K at (7±1)kbar. At further pressure increase the T_c decrease and pressures higher than (23±1)kbar suppress superconductivity and the resistivity and pressure increase in parallel. The temperature of the minimum crystal resistivity (T_{min}) increases readily with pressure.

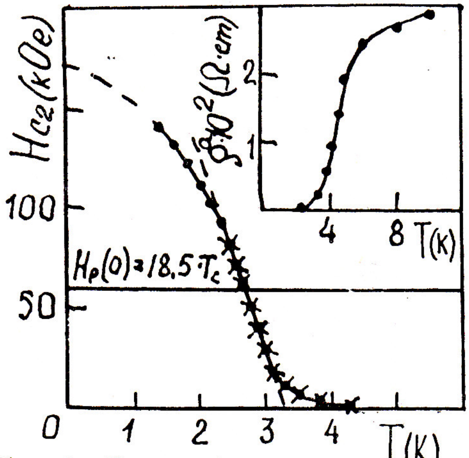

Fig. 1: Temperature dependences of upper critical fields H^{a}_{C2} and $H^{b'}_{C2}$ parallel to the ab plane of the $(BEDT\text{-}TTF)_4Hg_{2.89}Br_8$ single crystal. (•) H∥a; (x) H∥b'. The inset illustrates the resistivity vs. temperature plot at H=0.

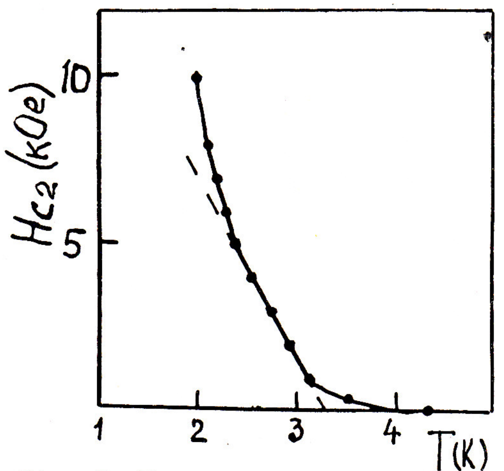

Fig. 2: Temperature dependence of the upper critical field H^{c*}_{C2} perpendicular to the ab plane, H∥c*.

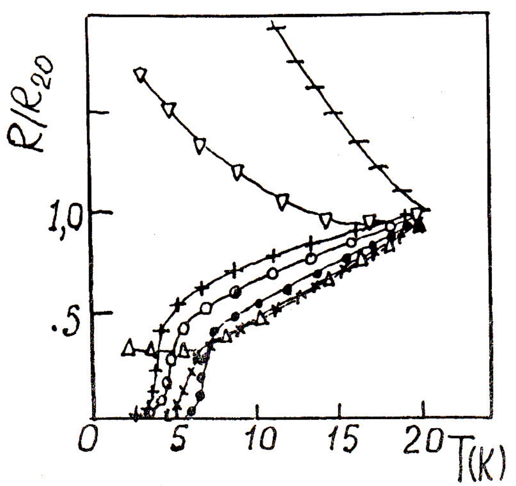

Fig. 3: Temperature dependence of resistivity of the single crystals $(BEDT\text{-}TTF)_4Hg_{2.89}Br_8$ at various pressures.

(+) 0 kbar; (o) 4 kbar;
(•) 7 kbar; (x) 13 kbar;
(Δ) 23 kbar; (∇) 30 kbar;
(−) 34 kbar

Discussion

Fig. 1 and 2 show that near the T_c (4.3 K) a site of the positive curvature is characteristic of both H^a_{C2} and H^{c*}_{C2} and the superconducting state is easily destroyed by low magnetic fields. Such state is probably caused by the destruction of weak coupling between the bulk elements with higher critical temperatures. At lower temperatures both curves are characterized by a straight site, which probably corresponds to the main part of the sample with $T_c \approx 3.3$ K. The dH^a_{C2}/dT and dH^{c*}_{C2}/dT are found to be 110 and 5 kOe/K, respectively, from the slope of the straight-line sites and according to the Ginzburg-Landau theory the correlation lengths can be estimated as $\xi^a(0) \approx \xi^b(0) \approx 170$ Å and $\xi^{c*}(0) \approx 8$Å

The critical field anisotropy is $H^a_{C2}/H^{c*}_{C2} = \xi^a/\xi^{c*} \approx 21$. It should be noted that the transverse correlation length $\xi^{c*}(0)$ is approximately twice as less than the interlayer distance.

The extrapolation of the $H^a_{C2}(T)$ dependence to the ordinate axis gives $H^a_{C2}(0) \approx 170$ kOe, which almost thrice as exceeds the Clogstone paramagnetic limit determined within the BCS model as $H_p = 18.5\ T_c \approx 60$ kOe. The evaluation of the orbital contribution to the superconductivity at 0 K gives $H^{a(dia)}_{C2}(0) = 0.7|dH^a_{C2}/dT|_{Tc} \cdot T_c \approx 260$ kOe, which is much higher than the experimental value of $H^a_{C2}(0)$. On considering the latter value as the result of the joint orbital and paramagnetic effects it is possible to estimate that the value of the paramagnetic limit H_p characteristic of the $(BEDT\text{-}TTF)_4Hg_{2.89}Br_8$ salt equals to ~ 310 kOe and is 5-fold higher than the Clogstone paramagnetic limit in the weak coupling approximation.

The paramagnetic limit exceeding in superconductors may be explained by one of the following reasons:

(1) triplet coupling of electrons;

(2) strong spin-orbital interaction;

(3) strong electron-phonon interaction with $\lambda > 1$.

Apparently the (1) and (2) cases cannot be applied to the superconductor $(BEDT\text{-}TTF)_4Hg_{2.89}Br_8$. Triplet superconductivity is to be expected only in pure superconductors [8], which is not the case of the salt under study because of the incommensurability in its structure. Strong spin-orbital interaction cannot take place either , which is confirmed by the study of the g-factor in $(BEDT-TTF)_4Hg_{2.89}Br_8$ [9]. The supposition on strong electron-phonon interaction considered within the extremely strong coupling model [10] allows provides both understanding of the considerable exceeding in paramagnetic limit and qualitative explanation to the positive curvature of $H^{c*}_{C2}(T)$ at low temperatures. However, qualitative comparison of the experimental results obtained with theoretical ones [10] gives far from the real value of the electron-phonon interaction, $\lambda > 10$.

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