Exceeding the paramagnetic limit of H_{c2} in an organic superconductor β-$(ET)_2I_3$ with $T_c = 7.1$ K

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The temperature dependences of the critical field $H_{c2}(T)$ of an organic superconductor β-$(ET)_2I_3$ with $T_c = 7.1$ K is studied experimentally at temperatures 1.8–7.1 K and normal pressure in two orientations of the magnetic field, $H \| b'$ and $H \| c^*$, in fields ranging from 0 to 140 kOe. The paramagnetic effect in this superconductor is found to be strongly suppressed. The reasons for this suppression are discussed.

Superconducting state with $T_c = 7$–8 K in an organic metal[1)] β-$(ET)_2I_3$ can occur either at a relatively low pressure[1,2] or at a normal pressure but in β-phase samples obtained as a result of thermolysis of the ϵ-phase samples of the composition[3,4] $(ET)_2I_7$. It was shown in Ref. 4 that β-$(ET)_2I_3$ samples obtained as a result of an $\epsilon \to \beta$ transition exhibit near T_c a typical two-dimensional anisotropy of the critical fields, at which the derivatives $(dH_{c2}/dT)_{T_c}$ in the directions a, b', and c^* are equal to

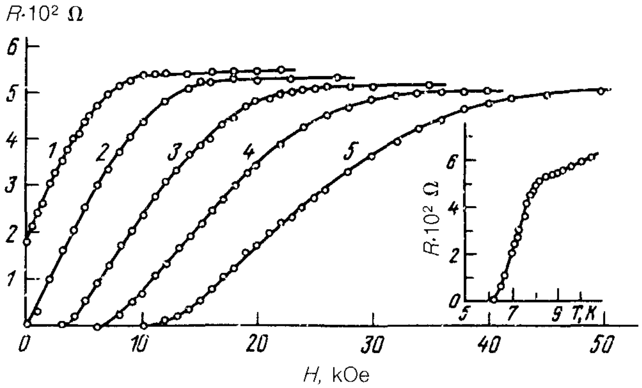

FIG. 1. Curves for the destruction of the superconducting state by the magnetic field $H||c^*$ at various temperatures: 1—6.5 K; 2—5.5 K; 3—4.2 K; 4—3.0 K; 5—1.9 K. Inset—the superconducting transition of the test sample at $H = 0$.

27.5, 25, and 3.4 kOe/K, respectively. With derivatives of this magnitude, when the magnetic field is directed along the conducting layers (the *ab* plane[5]), the $H_{c2}(T)$ curves are expected to exhibit some curious structural features as $T \to 0$, specifically, a situation in which the paramagnetic limit is surpassed. In the present letter we present data on the properties of such organic superconductors in magnetic fields up to 140 kOe and temperatures2) up to 1.8 K.

Figures 1 and 2 show typical curves for the destruction of the superconducting state by a magnetic field of different orientations in a β-phase sample with $T_c = 7.1$ K obtained as a result of a $\epsilon \to \beta$ transition. H_{c2} was taken to be the center of these curves. Figure 3 is a plot of the $H_{c2}(T)$ curves for two directions of the magnetic field: $H||c^*$ and $H||b'$. It is interesting to note that the superconducting transitions in the

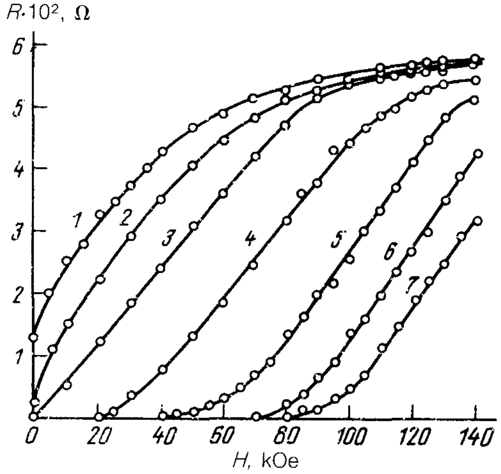

FIG. 2. Curves for the destruction of the superconducting state by the magnetic field $H||b'$ at various temperatures: 1—6.7 K; 2—6.2 K; 3—5.7 K; 4—4.8 K; 5—3.5 K; 6—2.5 K; 7—1.85 K.

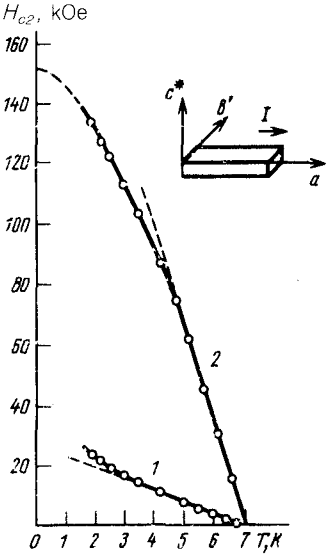

FIG. 3. Plots of the critical field H_{c2} for $H||c^*$ (curve 1) and $H||b'$ (curve 2) versus the temperature.

magnetic field are very broad: $\Delta H > (dH_{c2}/dT)_{T_c} \cdot \Delta T$, where ΔT is the transition width at $H = 0$ (see the inset in Fig. 1). If for $H||b'$ this width remains roughly constant at all temperatures, then for $H||c^*$ it will increase with decreasing temperature.

We note that the anisotropy of H_{c2} is strong over the entire temperature range (Fig. 3). Near T_c the derivatives $(dH_{c2}/dT)_{T_c}$ in the b' and c^* directions are equal to 30 and 3.8 kOe/K, respectively, for the given sample. We see that the $H_{c2}(T)$ curves are not the usual kind. The $H_{c2}(T)$ curve for $H||c^*$ has a positive curvature which is not seen in the β phase with[6] $T_c \simeq 1.5$ K. Note that the sign of the curvature does not depend on the method used to determine H_{c2} (from the onset, middle, or end of the transition in a magnetic field). This curvature for the β-phase samples with $T_c \simeq 8$ K, which were obtained under a pressure, was also observed in Ref. 7. Extrapolation of the experimentally measured $H_{c2}(T)$ curve for $H||b'$ to $T = 0$ K yields a value $H_{c2} \simeq 150$ kOe. For a purely diamagnetic effect the value of H_{c2} at $T = 0$ is $H_{c2}^{(d)}(0) \simeq 0.7 \times (dH_{c2}/dT)_{T_c} = 150$ kOe. It would seem that the paramagnetic effect does not influence the critical field in any way in the ab plane. At the same time, in the approximation of a weak interaction in a pair the Clogston[8] paramagnetic limit is $H_p(0) \simeq 18.5 \cdot T_c = 130$ kOe for the sample in question. In other words, the paramagnetic effect is indeed suppressed appreciably.

The spin-orbit scattering might be one of the reasons for such a suppression. The spin-orbit scattering, however, influences $H_p(0)$ substantially in the case of very "dirty" superconductors, i.e., when the condition $\xi_0 > l_{s0}$ holds, where l_{s0} is the mean free path involving a spin flip. In general, it is $l_{s0} \gg l$. Since the high-pressure phase is probably responsible, as was noted in Ref. 4, for the superconducting transition in the samples obtained through the conversion[1] $\epsilon \rightarrow \beta$, we can use for the samples in question the estimates of the mean free path l for high-pressure-phase samples. From the simple relations for the model of nearly free electrons, we find $l=\sigma\hbar k_F/ne^2\simeq 200$ Å, where $\sigma\simeq 3\times 10^4$ S/cm is the conductivity of the high-pressure phase before the superconducting transition,[1,2] and $k_F\simeq 1.5\times 10^7$ cm^{-1} is the reciprocal of the mean size of the lattice in the ab plane.[5] At the same time, the correlation length in the ab plane in the superconductor at hand is

$$\xi_{ab}(0) = \sqrt{\frac{\phi_0}{2\pi(dH_{c2}^{(c^*)}/dT)_{T_c}T_c}} \simeq 100\,\text{Å}\,.$$

We thus have $\xi(0)\ll l$; i.e., the samples with T_c = 7–8 K are "pure" superconductors or at least "intermediate" superconductors. Consequently, the paramagnetic limit cannot be increased markedly in these superconductors by the spin-orbit scattering.

The paramagnetic limit can be expected to increase in superconductors with a strong electron coupling. The possibility for the existence of such a mechanism in the superconductor $(\mathrm{ET})_2\mathrm{AuI}_2$ was indicated in Ref. 9.

It is also conceivable that there might be a triplet coupling in these samples, as was noted in Ref. 2. The necessary condition for such a coupling is $\xi_0<l$. The paramagnetic effect is known to be completely absent in the case of triplet coupling.

In conclusion we note that despite the rather strong 2D-type anisotropy of H_{c2} and the layered crystal structure,[5] the β-$(\mathrm{ET})_2\mathrm{I}_3$ crystals nevertheless should not be regarded as 2D superconductors but rather as strongly anisotropic 3D superconductors with a 2D anisotropy. A Josephson coupling between the layers can be achieved by satisfying the condition[10] $r=(16/\pi)[\xi_{c^*}(0)/d]^2<1$, where d is the distance between the levels, whereas the parameter $r\simeq 3$ for the β-$(\mathrm{ET})_2\mathrm{I}_3$ samples with T_c = 7–8 K.

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[1)] $(\mathrm{ET})_2\mathrm{I}_3$—bis(ethylenedithio) tetrathiofulvalene triiodide.

[2)] The experimental part of this study was carried out at the International Laboratory of Strong Magnetic Fields and Low Temperatures, Vrotzlaw, Polish People's Republic.

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