Low temperature upper critical field studies in
organic superconductor
\[ \beta''-(\text{BEDT-TTF})_2\text{SF}_5\text{C}_2\text{C}_2\text{SO}_3 \]

F. Zuo, P. Zhang, X. Su, J. S. Brooks*, J. A. Schlueter†, J.
Mohasham, R. W. Winter, and G. L. Gard†

Department of Physics, University of Miami, Coral Gables, Florida 33124
*National High Magnetic Field Laboratory, Tallahassee, Florida 32306
†Chemistry and Materials Science Divisions, Argonne National Laboratory,
Argonne, Illinois 60439
†Department of Chemistry, Portland State University, Portland, Oregon 97207

Low temperature upper critical field studies have been carried out in a new
organic superconductor \[ \beta''-(\text{BEDT-TTF})_2\text{SF}_5\text{C}_2\text{C}_2\text{SO}_3 \]. For field parallel
to the superconducting layers, the upper critical field determined from trans-
port measurements exceeds the BCS Pauli limit at low temperatures. The
angular dependence of the resistive transition shows that the upper critical
field can be best described by a quasi-two-dimensional model with a cusp near
the field parallel to the plane direction.

PACS numbers: 74.70.Kn, 74.25.Fy, 71.70.Ej, 74.80.Dm

The layered organic molecular crystals (BEDT-TTF)$_2$X (BEDT-TTF
is bis-(ethylenedithia-tetrathiafulvalene) where X is an anion are particu-
larly interesting because they are strongly correlated electron systems with
a number of similarities to the high-$T_c$ cuprate superconductors including
unconventional metallic properties and competition between antiferromag-
netism and superconductivity.\textsuperscript{1,2} Furthermore, they are available in high
purity single crystals and their low superconducting transition temperature
makes experimentally accessible physical characterization of quantities such
as the upper critical field and Shubnikov-de Haas oscillations in steady mag-
netic fields.\textsuperscript{3–5}

In this paper we report the interlayer magnetoresistance measure-
ments on a recently discovered organic superconductor \[ \beta''-(\text{BEDT}-
Single crystals of $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ were synthesized by the electrocrystallization technique described elsewhere. The interlayer resistance was measured with use of the four probe technique. Contact of the gold wires to the sample was made with a Dupont conducting paste or graphite paste. Typical contact resistances between the gold wire and the sample were about 10 $\Omega$. A current of 1 $\mu$A was used to ensure linear $I$-$V$ characteristics. The voltage was detected with a lock-in amplifier at low frequencies of about 20 Hz. To avoid pressure effects due to solidification of grease, the sample was mechanically held by thin gold wires. The data presented in this work were taken in a dilution refrigerator with field up to 18 T at the National High Magnetic Field Laboratory at Tallahassee. Sample can be rotated in the field and the orientation was determined by using a Hall probe at low fields.

Fig. 1 Magnetoresistance as a function of field at different temperatures.

Fig. 2 The temperature dependence of $H_{c2\parallel}$. The dashed line is the BCS Pauli limit.

Shown in Fig. 1 is an overlay of interlayer magnetoresistance as a function of field at different temperatures from 0.72K to 73mK. The field is applied parallel to the planes within the experimental errors of $\pm 0.2^\circ$. With decreasing temperature, the resistive transition shifts toward higher field. The resistive transition in parallel field is typical of the low dimensional organic superconductors with a broad transition width in field and a large positive magnetoresistance in the normal state. The inset shows a semi-log plot at 56 mK. Clearly, the resistance rises exponentially at small field and
Low temperature upper critical field studies ...

quasi-linearly at high field. To analyze the data, the superconducting transition or the upper critical field $H_{c2}$ is defined at 1 Ω level. Critical field defined at higher levels show similar temperature dependence.

Fig. 2 shows the temperature dependence of $H_{c2}$ thus defined. With decreasing temperature, $H_{c2}$ increases and saturates at about 11.3T for temperature below 100 mK. The base line is the BCS Pauli limit $H_p = 1.84 \ T_c = 9.6 \ T$ with $T_c = 5 \ K.$ Clearly, $H_p$ defined this way is well under the measured upper critical fields.

![Fig. 3 Magnetoresistance as a function of field at different angles.](image1)

![Fig. 4 The angular dependence of $H_{c2}$. The lines are fits to the data.](image2)

To look at the anisotropy of the upper critical field, a systematic measurements have been taken as a function of angle $\theta$, defined between the field direction and the normal of the plane. Plotted in Fig. 3 is an overlay of resistive transitions as a function of field at different angles at a fixed temperature of 26 mK. The eight curves are representative of the angular dependence from field parallel to the plane ($\theta = 90^\circ$) to nearly normal to the plane ($\theta = 15^\circ$). With decreasing $\theta$, the field dependence of the resistive transition is drastically changed. For $\theta \leq 60^\circ$, a well defined Subnikov de-Hass (SdH) oscillation in the resistance can be observed. Details of the oscillation and its anomalous temperature and field dependence have been published elsewhere.$^{8,9}$

$H_{c2}$ defined in at 1Ω level as a function of angle is shown in Fig. 4 at $T = 26 \ mK$. Clearly, $H_{c2}$ decreases rapidly away from the parallel to the plane direction and is nearly saturated for $\theta \leq 40^\circ$. The two lines are fit to the 3 dimensional anisotropic model and the 2 dimensional thin film model. For 3D anisotropic model, the upper critical field can be described by

\[
\left[ \frac{H_{c2}}{H_{c2\perp}} \right]^2 + \left[ \frac{H_{c2}}{H_{c2\parallel}} \right]^2 = 1, \text{where } H_{c2\perp} \text{ is the upper critical field}
\]
for field perpendicular to the plane and the $H_{c2}$ is for parallel to the plane, $	heta$ is the angle between the field and the normal of the plane. For a 2D thin film Tinkham and Klemm has obtained the following expression:

$$H_{c2} \left[ \frac{\cos(\theta)}{H_{c2,\perp}} \right] + \left[ \frac{\sin(\theta)}{H_{c2,\parallel}} \right]^2 = 1.$$  

The main difference is that at $\theta = 90^\circ$, $H_{c2}(\theta)$ for the 3D model is smooth or bell-shaped with $\frac{dH_{c2}(\theta)}{d\theta} = 0$. On the other hand, $H_{c2}(\theta)$ has a cusp at $\theta = 90^\circ$ for the 2D case.

If the upper critical field is determined solely by coupling of the field to the spins then it will be independent of the field direction. Bulaevskii considered the case where the paramagnetic limit is larger than the upper critical field for fields perpendicular to the layers but smaller than the upper critical field determined by orbital effects for fields parallel to the layers. The angular dependence is then given by

$$\left[ \frac{H_{c2}(\theta) \cos(\theta)}{H_{c2,\perp}} \right] \left[ 1 - \left( \frac{H_{c2,\perp}}{H_{c2,\parallel}} \right)^2 \right] + \left[ \frac{H_{c2}(\theta)}{H_{c2,\parallel}} \right]^2 = 1,$$

where $H_{c2} = H_p$. This also results in an $H_{c2}$ versus $\theta$ curve which has a cusp at $\theta = 90^\circ$. Indeed the angular dependence is difficult to distinguish from 2D model.

While both fits seem reasonable at first sight, clear deviations are seen at near $\theta = 90^\circ$. A cusp-like feature is observed experimentally, as in the 2D fit, while the 3D fit is rounded with a negative curvature at the top. A better agreement with the data for the 2D model at large angles is also evident with the 3D fit lying systematically under the data. At 26 mK, the 2D or the Bulaevskii model gives $H_{c2,\parallel} = 1.4$ T and $H_{c2} = 11.9$ T.

The upper critical field determined from transport measurements has been under a lot of debate in the cuprate superconductors. For field perpendicular to the planes, $H_{c2}(T)$ defined at certain fractional normal state resistance typically gives rise to a positive curvature at low temperatures. Various mechanisms have been proposed for the unconventional temperature dependence. However, it has been suggested that the $H_{c2}$ thus defined corresponds to irreversibility or vortex melting line.

For field parallel to the planes, vortex moving along the plane encounters negligible pinning as there is no normal core associated with Josephson vortices. Magnetization is practically always reversible in this orientation. The resistive onset field is clearly well separated from irreversibility field and reflects the true upper critical field.

The origin for the apparently much larger $H_{c2}$ than the simple BCS $H_p$ is not clear. One possibility is that the Pauli limiting field is enhanced due to strong electron correlations, as suggested in another BEDT-TTF based organic superconductor. Another possibility is that the inhomogeneous state is realized at low temperature and high field, such that the upper critical field in plane for a Josephson coupled system can exceed the Pauli limit by a factor of $\sqrt{2}$.

Careful examination is necessary to distinguish
Low temperature upper critical field studies

the models.

In summary, we have observed an upper critical field determined from resistive transition considerably larger than the BCS Pauli limiting field. The upper critical field is saturated at low temperatures. Angular dependence of the resistive transition is consistent with the highly anisotropic nature of the title compound with a cusp-like angular dependence for field near the plane.

One of us (FZ) acknowledge useful discussion with Drs. R. H. McKenzie and J. Wosnitza. The work is supported in part by NSF grant No. DMR-9623306 and the Petroleum Research Fund ACS-PRF 33812-AC5. Work at the National High Magnetic Field Laboratory was supported by NSF Cooperative Agreement No. DMR-9016241 and the state of Florida. Work performed at Argonne National laboratory was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences, under Contract No. W-31-109-ENG-38.

REFERENCES

15. F. Zuo et al., cond-mat/9904186.