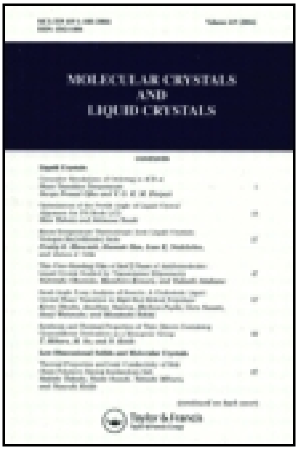

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# MEISSNER ANISOTROPY IN DEUTERATED $(TMTSF)_2CLO_4$

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Both the Meissner-signal (flux expulsion on cooling through $T_c$) and the shielding signal (obtained by turning on a field in the superconducting state) have been observed in deuterated single crystals for fields oriented both along and normal to the chain axis. A possible isotope effect on $T_c$ is discussed. We find complete diamagnetism in normal fields and incomplete (∿ 40 %) diamagnetism in parallel fields. A dramatic drop of the Meissner-effect in parallel fields well below $H_{c1}$ is observed and not yet well explained.

The compound ditetramethyltetraselenafulvalene-perchlorate [$(TMTSF)_2ClO_4$] is the first in the family of organic charge transfer salts which shows superconductivity at zero pressure. K. Beechgaard and coworkers have observed a transition to zero-resistance between 1.2 K to 1.4 K for different samples /1/. To prove that superconductivity in these crystals is a real bulk property it has to be shown that they exhibit the Meissner-effect, i.e. that they expel magnetic flux when cooled below the transition temperature. This effect has already been observed by two of the authors /2/ in $(TMTSF)_2PF_6$, a compound that becomes superconducting under pressure.

Here we report on the observation of a partial Meissner-effect both in the normal as well as in the almost completely deuterated $(TMTSF)_2ClO_4$.

Sample preparation and experimental set-up have been similar to reference /2/.

The diamagnetic measurements have been carried out by means of a superconducting bridge circuit with a SQUID-detector to monitor changes in the sample magnetization. The two sample coils of the bridge contained the organic crystal and a tin calibration sample of similar shape. A Permendur shield around the cryostat reduced the Earth's field to below 2 millioersteds.

The high quality of the deuterated crystals could be demonstrated in a mechanical twinning experiment. A thin, needle-shaped sample was clamped on one side to a glass slide with a dab of glue, and pressure was exerted on the other, free end in the direction of the a-axis (= sample axis). Twinning of the crystal could thus be induced as was obvious by a kink forming at a definite angle. The fact that this kink could be moved freely back and forth over the whole length of the crystal by small lateral forces indicates that the crystal cannot contain a high dislocation density. It is suprising that this effect, normally seen in whisker crystals, can be observed at all in a molecular crystal like $(TMTSF)_2ClO_4$.

By comparing the magnetization and resistive transition of two samples each of the deuterated and undeuterated $(TMTSF)_2ClO_4$ one obtains a slight isotope shift of the transition temperature of $\Delta T_c \simeq 0.09$ K. The compound with the greater mass, the deuterated $(TMTSF)_2ClO_4$ has the lower $T_c$ as one would expect within the framework of the BCS theory. $(D_{12}\text{-TMTSF})_2ClO_4$: $T_c = 0.99$ K.

The result, however, is very preliminary since we observe a slight scatter in $T_c$ of the different samples of the same batch. (Maximum difference in $T_c$ of three samples was 0.04 K. $T_c$ was determined from magnetization versus temperature in a constant field well below $H_{c1}$. $T_c$ is defined as the intercept of the extrapolation of the linear part of the magnetization curve with M = 0.)

For a better determination of the isotope effect due to deuteration more observation with better statistics are necessary.

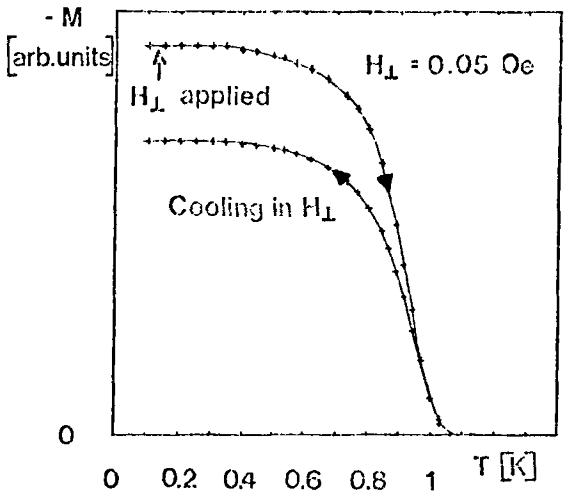


Fig. 1 Observed diamagnetic magnetization of $(D_{12}\text{-TMTSF})_2ClO_4$ for a magnetic field perpendicular to the linear chain axis.

Fig. 1 exhibits a typical transition curve of the magnetization of a $(D_{12}\text{-TMTSF})_2ClO_4$ single crystal in a constant magnetic field perpendicular to the chain axis. First the sample is cooled well below $T_c$ in zero field, then $H_\perp$ is applied (see arrow) and the decrease of the shielding signal upon warming the sample above $T_c$ is observed. On subsequent cooling the Meissner-signal is monitored.

In parallel fields the diamagnetic shielding-signals are quite incomplete, only ∿ 40 % (as determined by comparison with a Sn reference. The uncertainty of this estimation is about 15 %). The Meissner-effect (here determined as the ratio of the Meissner-signal and the shielding-signal) drops from about 50 % in very low fields (∿ 0.02 Oe) to about 15 % at 0.1 Oe, a field still belcw $H_{c1}$. At higher fields this value is slowly decreasing below 10 %. (Fig. 2). On the other hand the shielding-signal in a field transverse to the chain axis (but without specified orientation with respect to the b and c axis) shows the full diamagnetic value. The

Meissner-effect is about 80 % in the lowest field and decreases only slightly to about 70 % in the higher fields.

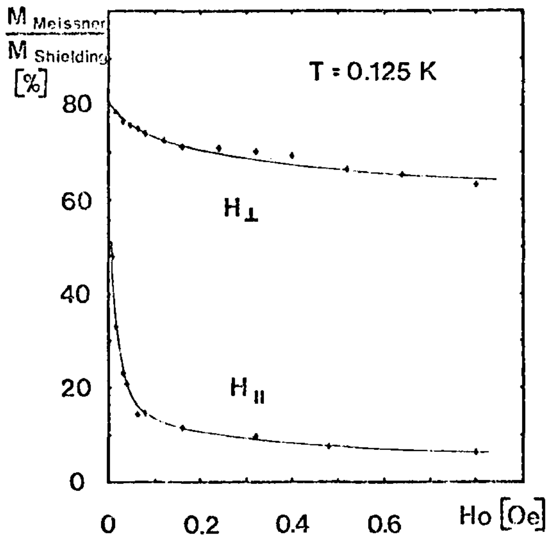


Fig. 2 Meissner-effect of $(D_{12}\text{-TMTSF})_2ClO_4$ vs. applied field at T = 0.125 K for fields oriented parallel and perpendicular to the linear chain axis of the crystal.

In Fig. 3 the shielding magnetization is plotted versus the applied field both for $H_\perp$ and $H_\parallel$ and the Meissner magnetization for $H_\parallel$ at T = 0.15 K. The field at which the first clear departure from linearity of the shielding magnetization occurs is taken to be $H_{c1}$.

At T = 0.2 K we find $H_{c1\perp}$ = 5.6 Oe (after correction due to demagnetizing effects) and $H_{c1\parallel}$ = 1 Oe.

$H_{c1\perp}$ and $H_{c2\perp}$ versus temperature are plotted in Fig. 4; $H_{c2\perp}$ (O) is estimated to about 1.4 kOe.

The behaviour of the Meissner-effect in parallel fields, especially the dramatic drop in fields well below $H_{c1}$ is not yet well understood.

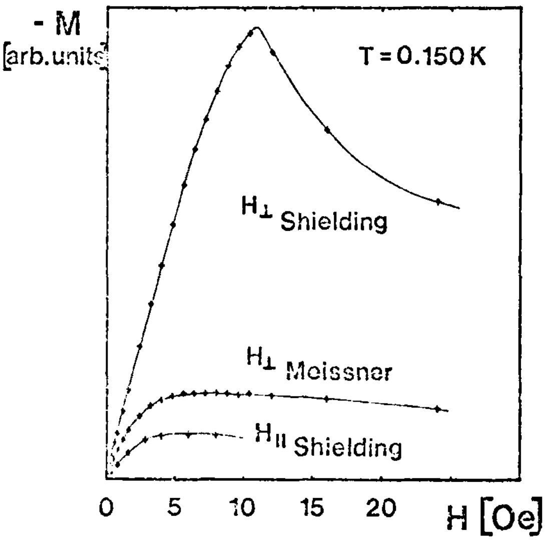


Fig. 3 Diamagnetic shielding and Meissner-magnetization for $H_\perp$ and $H_\parallel$ at T = 0.150 K of $(D_{12}\text{-TMTSF})_2ClO_4$

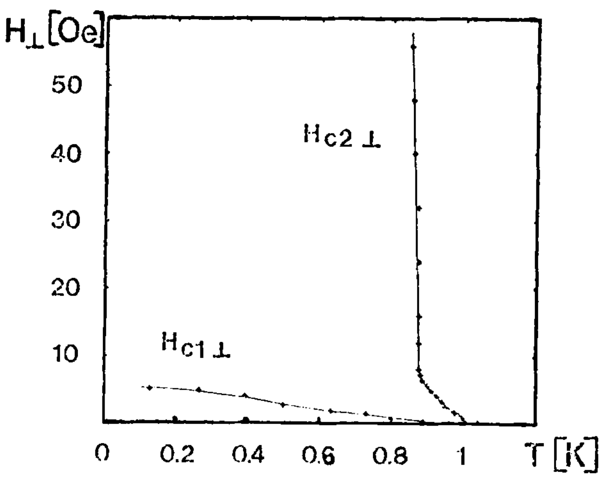


Fig. 4 $H_c$ - $T_c$ phase diagram for perpendicular field. $H_{c2\perp}$ (O) $\sim$ 1.4 kOe

We speculate that both this effect as well as the reduced shielding-signal in parallel fields have to do with anisotropies of both coherence length and London penetration depth in the b-c plane normal to the chain axis. High effective masses in the c-direction might lead to a macroscopically large field penetration in the b-direction, which might stabilize a sort of intermediate state, except for the very lowest fields.

In conclusion, we have shown that the deuterated $(TMTSF)_2ClO_4$ is a bulk superconductor with perfect shielding behaviour in magnetic fields applied transverse to the chain axis and a rather incomplete one in parallel fields. The drop of the Meissner-effect in low fields and the possibility of macroscopically large penetration depth will be studied in further investigations.

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