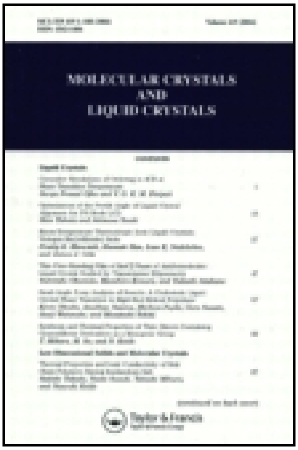

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Evidence for Partial Diamagnetism in $(TMTSF)_2 FSO_3$

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EVIDENCE FOR PARTIAL DIAMAGNETISM IN $(TMTSF)_2FSO_3$

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Abstract We have searched for and have found superconducting diamagnetic screening currents as well as Meissner-effects in $(TMTSF)_2FSO_3$ at different pressures. The largest signals, however, are only 2 % of those expected for a perfect superconductor.

In the family of the Bechgaard salts $(TMTSF)_2X$, the compounds with non-centrosymmetric anions ($X = ClO_4$, ReO_4, FSO_3) exhibit additional phase transitions related to anion orientational order /1,2,3/.

The non-centrosymmetric anion FSO_3 is special because it also contains a small electric dipole moment. In a manner similar to the ReO_4-compound, $(TMTSF)_2FSO_3$ at zero pressure exhibits anion order (causing a doubling of the unit cell in all principal directions) at 86 K, leading to an insulating CDW-state below this temperature /4,5/. Application of pressure again reduces the anion ordering temperature /6,7/. By means of d.c. resistivity measurements, indications of a superconducting (SC) state are detected at temperatures as high as 3 K, and at pressures above ∿5 kbar, while the anion ordering temperature appears to be still around 40 K. No evidence of 3-dimensional SC (e.g. diamagnetic screening or Meissner-effect), however, was reported so far. The resistivity increases below 40 K, but in a manner uncharacteristic of a CDW-state (which would imply the opening of an energy gap). Thermopower measurements in fact show a metalliclike behavior in this regime. Lacoe et al. /6,7/ therefore suggest that the anions condense, below ∿40 K, into a glass-like "frozen" disordered state, thus causing increased disorder-scattering but leading to no gaps on the Fermi surface due to the absence of a superstructure.

In what follows we like to report on new sensitive measurements of the d.c. susceptibility on several crystals in various orientations and at various pressures which show that a small volume fraction (∿2 %) exhibits at least 2-dimensional SC. Our measurements were performed using a SC bridge circuit described before with a SQUID null detector /8/. Two of the bridges 4 inductance coils are mounted inside of a BeCu clamp type pressure cell, one containing the sample and the other a tin reference sample. Magnetization measurements were done by first cooling the samples in nearly zero field (H_{res} <10 mOe), then loading all inductance coils inductively with a supercurrent to provide a magnetic field at the samples and slowly raising the temperature from 80 mK on up. Magnetization changes in the samples cause a change in supercurrent through the SQUID detector which can be recorded. Two such measurements are shown in Fig. 1, where the signals are plotted as fractions of a completely diamagnetic signal. The samples were always cooled from about 90 K to 4.2 K with a similarly slow rate over a period of about 2 1/2 hours. The largest diamagneting shielding signals observed occured in fields oriented transverse to the crystal-axis (a-axis) and at a pressure around 6 kbar and amounted (at 0.3 K) to 2 % of that of a perfect SC. At this pressure the diamagnetic signals vanish at 1.5 K, which corresponds with the temperature where the d.c. resistivity reaches zero at p = 6.5 kbar (Fig. 7 in ref. 7). A remarkable property of the temperature dependence of the shielding signal is the fact that even much below T_c it never saturates, in contrast to what is known from the other organic superconductors /8,9/. Meissner signals are obtained by cooling back below the transition temperature in the same field and observing the magnetization change through flux expulsion. The Meissner signals ranged in magnitude typically from between 50 and 70 % of the shielding signals, independent of the field orientation.

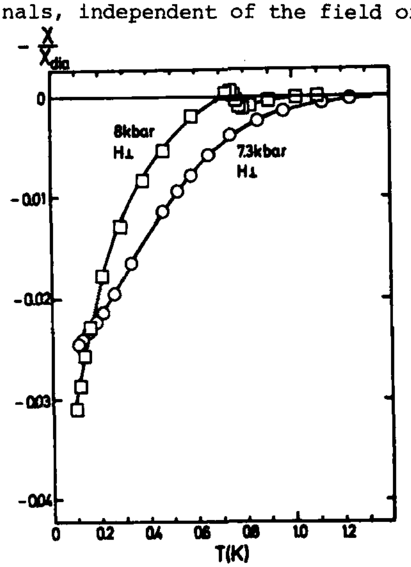

Fig. 1 Diamagnetic susceptibility normalized to a complete superconducting signal in a field of H = 100 mOe transverse to the a-axis

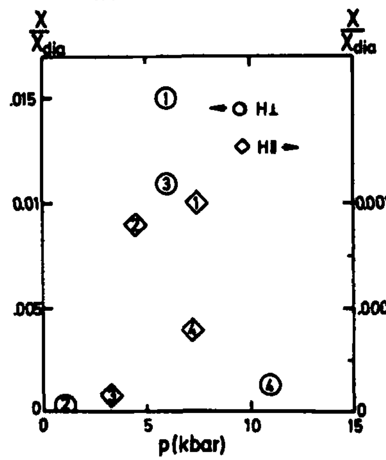

Fig. 2 Pressure sequences of two different samples: Observed diamagnetic susceptibility at T = 0.3K (measured in a field of 1 Oe), normalized to that of a perfect superconductor, plotted vs pressure. Circles correspond to H ⊥ a-axis, canted squares to H ∥ a-axis. The numbers in the symbols indicate the consecutive cooling cycles.

In order to demonstrate the dependence of this clear trace of superconductivity on pressure, we show in Fig. 2 the shielding signals of two samples, one with the measuring field oriented along (canted squares), the other transverse (circles) to the a-axis, as a function of pressure. The pressure sequence for each sample is indicated by the numbers in the symbols. It can first be seen that the diamagnetic signal appears, with limited reproducibility, in the pressure range between about 5 and 12 kbar, with largest magnitude as well as highest (diamagnetic) transition temperatures around 6 kbar (T_c is defined as the onset of the shielding magnetization). This latter fact is shown in Fig. 3 which also contains results of other samples. The slope $dT_c/dp \sim 0.11$ K/bar is about the same as that observed in $(TMTSF)_2PF_6$ /8,10/. Secondly, the shielding signals for the longitudinal field orientation are almost an order of magnitude smaller than those for the transverse orientation.

A typical magnetization curve for the transverse field orientation, constructed from temperature sweeps at various fields, is shown in Fig. 4.

The main results then are that the diamagnetism in $(TMTSF)_2FSO_3$ is very anisotropic and small and that the Meissner signals are always as large as 50 to 70 % of the shielding signals. This latter fact would seem to exclude a state where 3-dimensionally connected

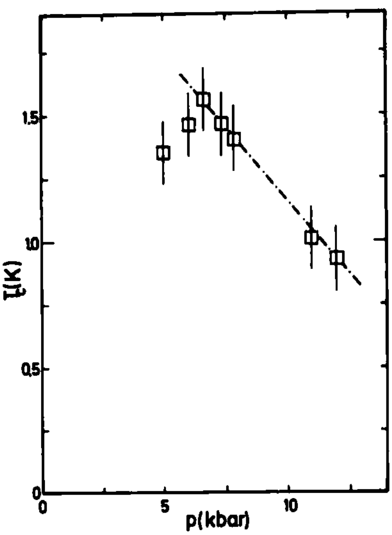

Fig. 3 Critical temperature (see text) versus pressure. The dash-dotted line corresponds to a pressure coefficient of -0.114 K/kbar.

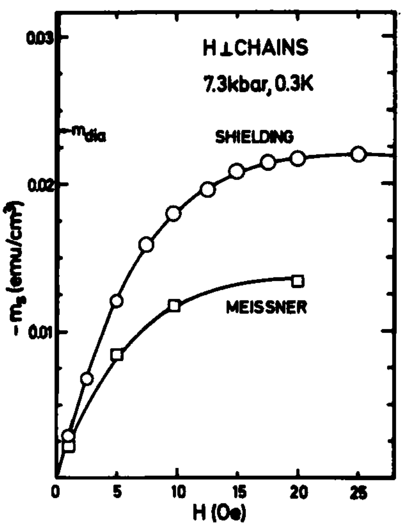

Fig. 4 Diamagnetic, shielding- and Meissner signals in transverse fields normalized to $m_{dia}=-H/2\pi$, versus applied field, measured at 0.3 K and 7.3 kbar.

SC filaments exist, as such a state always shows much larger shielding- than Meissner signals. The relative sizes of the shielding- and Meissner signals suggest that SC develops in (at least 2-dimensional) regions that are larger than a penetration depth, and in total amount to only about 2 % of the sample volume. On the other hand, there seems to be a high probability that those regions are connected along the a-direction of the crystal, leading to the observed resistive transition in that direction. Guided by the properties of $(TMTSF)_2ClO_4$, where SC only develops when the anion orientational order shows a superstructure along b, but not along a or c, and where states with a high degree of frozen-in anion disorder are also observed to show small fractions of volume superconductivity, one might speculate that it is regions with similar order among the FSO_3 anions which cause superconductivity in this salt. The essential difference between the two salts is that the order among the ClO_4 ions is controlled only by lattice interactions, whereas in the FSO_3 salt it is controlled in addition by the competing dipolar interactions and by pressure. This would seem to make it more difficult, if not impossible, to prepare that pure state of anion order in the ground state which would cause complete volume superconductivity.

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