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## Molecular Crystals and Liquid Crystals

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## One Dimensional Organic Superconductivity in (TMTSF)<sub>2</sub>PF<sub>6</sub> and (TMTSF)<sub>2</sub>C10<sub>4</sub> Detected Via Tunnel Spectroscopy

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ONE DIMENSIONAL ORGANIC SUPERCONDUCTIVITY IN (TMTSF) 2PF AND (TMTSF) 2C104DETECTED VIA TUNNEL SPECTROSCOPY.

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Electron tunnelling data in (TMTSF)<sub>2</sub>X-GaSb Schottky barriers support the idea that strong superconducting fluctuations exist in quasi-one-dimensional (TMTSF)<sub>2</sub>X up to temperatures which are about one order of magnitude higher than the 3-D ordering critical temperature. Other tunnelling data tend to indicate that stabilization of superconductivity is possible at high temperature via chain cross-linking.

ELECTRON TUNNELLING IN (TMTSF)  $2^{PF}6^{-N/GaSb}$  SCHOTTKY JUNCTIONS UNDER PRESSURE

In a quasi-one-dimensional conductor the transition towards a zero-resistance superconducting state occurs at a temperature  $T_3$  (the index means the onset of three-dimensional ordering) which can be significantly lower than the temperature T<sub>1</sub> at which long range order develops along each chain within the mean-field theory approximation. In 1-D conductors any amount of defects (or domain walls) suppresses the onset of 3-D ordering. However, as the defect concentration is low in the range T3<T<T1the meansquare amplitude of the superconducting order parameter can still be large, i.e.  $\Delta = \Delta \ln \Delta$  (mean field) on nearly the whole volume. In the same temperature range, even without long range order a pseudo-gap exists in the density of quasiparticles of the 1-D fluctuating superconductor. The pseudo-gap is large  $2\Delta = 2\Delta (m-f)$  at low temperature  $T \gtrsim T_3$  and decreases smoothly to zero in the vicinity of T1.

Electron tunnelling through the Schottky barrier at the interface between the organic superconductor(TMTSF), PF6(1) and the evaporated N-type degenerate semiconductor GaSb provides a dV/dI characteristics which can be interpreted by the existence of a pseudo gap in the quasi particle density of states at the Fermi level of the organic superconductor under a pressure of 11 Kbar(2), figure 1. The energy splitting between the two dV/dI minima on either sides of the zero-bias provides an estimate for the pairing energy, namely  $2\Delta = 3.6 \text{ meV}$  (40 K) at T = 0.1 K under 11 Kbar. The tunnelling data of figure 1 are only weakly affected by a warming through the three dimensional ordering temperature (i.e.T $_{q}$ =1K)figure 2, suggesting therefore that the pseudo-gap characterized by the energy of 40 K is related to the development at high temperatures of superconducting order along the chains without interchain coherence. Although decreasing significantly at increasing temperature, the amplitude of the zero bias resistance maximum, remains visible up to 15 K or so in a large number of junctions with relatively low background resistances. Moreover, the application of a magnetic field smears out the zero-bias resistance maximum, figure 2. The previous tunnelling data are consistent with a 1-D mean field transition temperature of the order of 12 K in fair agreement with the value derived from the knowledge of a pairing energy of 40 K, with the BCS relation  $2\Delta/kT_c = 3.5$ .

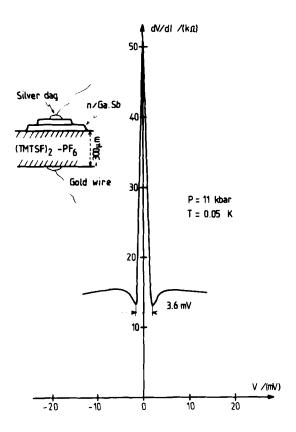


FIGURE 1 Differential resistance of a (TMTSF)<sub>2</sub>PF<sub>6</sub>-GaSb Schottky junction. The voltage difference between the two resistance minima is taken as a mesure of  $2\Delta$ .

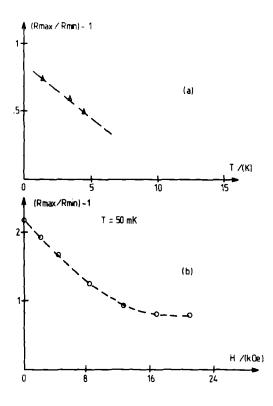


FIGURE 2 The temperature dependence (a) and field dependence (b) of the differential resistance characteristics of a (TMTSF)  $_2^{\rm PF}$   $_6^{\rm -GaSb}$  junction. The modulation current is  $\Delta I = 10^{-7}$  A (a) and 5 x  $10^{-9}$  (b).

THREE DIMENSIONAL STABILIZATION OF SUPERCONDUCTIVITY AT 12 K IN (TMTSF)<sub>2</sub>PF<sub>6</sub>

Figure 3 displays two different behaviours observed with (TMTSF)  $_2$ PF $_6$ -GaSb junctions (3). The state I corresponds to the tunneling characteristics of electrons through a Normal Insulator-Superconductor junction as discussed in the previous section. The state 2, exhibiting resistance anomalies at bias voltages of  $\pm \Delta$  and  $\bullet$   $2\Delta$  can be understood in terms of single electron tunnelling (N-I-S junctions) and S.I.S. tunnelling with the existence of tunnelling between two superconducting spots in (TMTSF)  $_2$ PF $_6$  separated by a thin insulating region. It is also conceivable

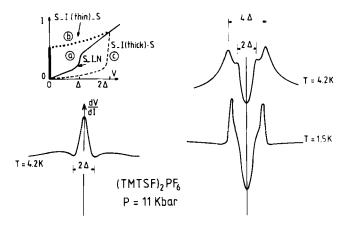


FIGURE 3 Differential resistance showing the two states observed in a junction. State I (left) displays N-I-S electron tunnelling. State 2 displays electron tunnelling with resistance anomalies separated by the voltage  $2\Delta$  and pair tunnelling (S-I-S) characterized by resistance peaks at  $\pm$  2 $\Delta$ . On cooling the value of the resistance at zero bias decreases, as shown by the 4.2 and 1.5 K curves. The different I/V characteristics are sketched in the insert. The dotted curve (b) refers to the situation of a (S-I-S) weak link between two superconducting spots.

that one of the superconducting electrodes is due to proximity effects of the (TMTSF) 2PF superconductivity onto GaSb. It is very likely that in the process of junction preparation, foreign atoms diffuse away from the surface between microcracks or even between the individual chains at a microscopic level. As a result interchain coupling becomes locally increased. In one junction, presenting a low resistance ( $>10\Omega$ ) at elevated temperature, the zerobias resistance (i.e.the resistance in the center of the dip in state 2 on figure 3) drops abruptly to zero below 12 K, figure 4. This result, reversible in temperature, can be interpreted by a contact between (TMTSF), PF, and N/GaSb becoming superconducting below 12 K. Chain crosslinking via diffused foreign atoms may possibly stabilize locally the 1-D superconducting fluctuations into superconducting spots exhibiting 3-D ordering at a temperature close to the single-chain mean-field transition temperature (4).

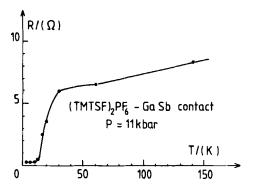


FIGURE 4 The zero-bias differential resistance of a low resistance (TMTSF)  $_2$ PF $_6$ -GaSb contact. The contact becomes superconducting below 12 K. The residual resistance of 0.2 $\Omega$  corresponds to the series resistance of the silver dag contacts and the remaining bulk organic sample. Under the same conditions bulk (TMTSF) $_2$ PF $_6$  is superconducting below 1 K only.

TUNNELLING PROPERTIES OF (TMTSF)<sub>2</sub>C10<sub>4</sub>-GaSb JUNCTIONS AT AMBIENT PRESSURE

With (TMTSF) ClO<sub>4</sub>-GaSb junctions at ambient pressure we have obtained dV/dI characteristics corresponding to state  $2^{(5)}$ . In some cases a deep resistance minimum is observed at zero bias together with two sharp maxima splitted by  $4\Delta=16\text{mV}$ , figure 5. Following the discussion of the previous section the corresponding 1-D mean-field temperature would therefore amount to  $T_1=2\Delta/3.5 \gtrsim 26\text{K}$ . This temperature is thus significantly larger than the 3-D ordering temperature  $T_3 \gtrsim 1.2 \text{ K}(6)$ .

As shown on figure (6), the characteristics of such (TMTSF)  $_2$ ClO $_4$ -GaSb contacts happen to be extremely sensitive to a microwave ( $\nu$ =9.6 GHz) irradiation. We take this extreme sensitivity as an evidence for the existence of a Josephson effect occuring in the S-I-S regions of the junction. However no Josephson current can be observed at zero bias, as a consequence of strong thermal fluctuations suppressing the dc Josephson current in high resistance ( $\nu$  20k $\Omega$ ) junctions (7). Finally a test junction performed with TTF-TCNQ and exhibiting similar electrical

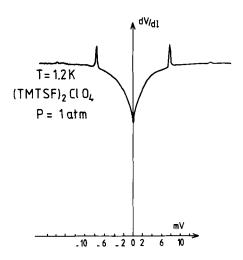


FIGURE 5 Differential resistance of a  $(TMTSF)_2C10_4GaSb$  contact showing sharp resistance maxima splitted by  $4\Delta = 16$  mV and a zero-bias resistance dip, according to the state 2 on figure 3.

specifications in the temperature domain 35-55K has failed to show any sensitivity to microwave irradiation up to a power of 10 mW.

In conclusion the tunnel spectroscopy technique has revealed the existence of a very large pseudo-gap of superconducting origin in the quasi particle density of states of the organic superconductors (TMTSF).,X. These results are in fair agreement with the recent determination in the same series of a far infrared pseudo-gap by optical spectroscopy (8).

Several experimental observations suggest that stabilization of superconductivity at high temperature is indeed possible via chain cross-linking.

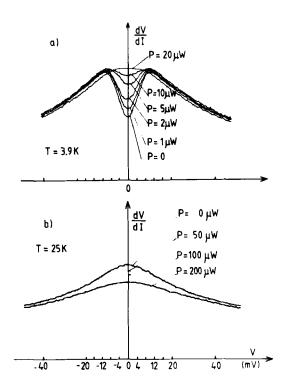


FIGURE 6 Differential resistance of a (TMTSF)  $_2$ ClO $_4$ -chrysocal junction put in a microwave cavity with different level of microwave ( $\nu$  = 9.6 GHz) irradiation, at T = 3.9 K (a) and 25 K (b). The absolute value of dV/dI is approximately 20  $\Omega$ .

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