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Organic Superconductors: A Survey of Low Dimensional Phenomena

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ORGANIC SUPERCONDUCTORS: A SURVEY OF LOW DIMENSIONAL PHENOMENA+

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An experimental investigation of the electronic properties of the conducting cation radical salts $(TMTSF)_2X$, has shown how the development of superconductivity at low temperature is affected by the low dimensional character of the band structure. Electron Schottky tunnel spectroscopy has given evidence that the critical temperature is not an intrinsic property of the intrachain superconducting pairing. The pairing energy in $(TMTSF)_2PF_6$ (P=11 Kbar) derived from the pseudo-gap in the quasi particle density of states amounts to $\sim$40 K i.e about 40 times the critical temperature. Consequently, properties such as conductivity, thermal conductivity, thermopower, etc...of these Quasi-One-Dimensional Superconductors are to a large extent dominated by the onset of 1-D fluctuating superconducting pairing below 30 K or so. Moreover, the stabilization of these fluctuations into a 3-D ordered superconducting state at higher temperature, namely 12 K for $(TMTSF)_2PF_6$ (P=11 Kbar) seems feasible via chemically induced chain bridging. The large pairing energy as well as the presence of a spin density waves state in the (T.P) phase diagram of these organic superconductors suggest the importance of the Coulomb interactions in the mechanism of Cooper pair formation.

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INTRODUCTION

The discovery of superconductivity in condensed organic matter[1] is the outcome of a concerted work initiated by a suggestion of F.London[2] in 1950. According to F.London, superconductivity need not be limited to the class of inorganic conductors but could possibly play an important role in certain large organic molecules of biochemical interest.

Following Bardeen, Cooper and Schrieffer's successful theory of superconductivity in metals[3] based on the coherent motion of electron pairs comprising electrons with equal and opposite momentum and spin and held together by a phonon-mediated attraction, Little proposed an extension of that same pairing idea to electrons moving in media of high electrical polarizability[4]. In Little's model the attractive interaction in a pair of electrons moving along an organic chain occurs through interactions with the electrons of conjugated ring side-molecules; i.e.molecules that have alternating double and single bonds, as in benzene. In this case the electron pair should be more firmly bound than in the usual type of pairing, and should help to stabilize the superconducting state and so allow superconductivity at higher temperatures.

However, despite the stimulating proposal of Little it was not until the early seventies that organic conductors appeared in the guise of radical-cation salts[5] and charge transfer complexes[6]. Both systems contain open shell planar molecules between which large intermolecular π-wave function overlaps are responsible for an electron delocalization. The stacked configuration of the planar molecules in the crystals confines the motion of the carriers to a single direction in space since the π-electrons interaction is weak in the directions perpendicular to the stacking axis. It appeared therefore that the concepts of Physics in One Dimension fitted the description of the electronic properties of such unusually anisotropic conductors [7].

The behaviour at low temperature of a 1-D electron gas in which the Fermi surface consists of two planar sheets intersecting the $k_{//}$ axis at $+k_F$ and $-k_F$ is qualitatively different from what we know about 3-D conductors. A 1-D interacting electron gas is usually decribed in terms of two coupling constants g_1 and g_2 for electron-electron backward and forward scattering respectively. Byschkov et al[8] pointed out that in the framework of the parquet approximation in which only terms like $g \ln E_F/T$ are kept in the perturbation expansion, charge density

fluctuations develop simultaneously with the fluctuations towards superconductivity. Renormalization group calculation[9] indicates that for $g_1>0$, $g_1>2g_2$ the electron gas shows a superconducting instability (at T = 0 strictly speaking for a 1-D electron gas), for $g_1>0$, $g_1<2g_2$ a charge density wave (CDW) or spin density wave (SDW) instability and for $g_1<0$ a superconducting or CDW state for $g_1>2g_2$ and $g_1<2g_2$ respectively. In quasi one dimensional systems however, there are always interactions between neighbouring chains leading to phase transitions at finite temperature. On the one hand, interchain Coulomb interactions between neighbouring chains couple CDW's[10] and therefore lead to a Peierls transition at finite temperature[11,12]. On the other hand, coupling between superconducting order parameters requires the electrons to move from one chain to the next, so that a superconducting transition may only occur for finite interchain tunnelling integral, $t_\perp$[13,14,15,16].

In an attempt to understand the phase transitions of quasi one dimensional (Q-1-D) conductors two scales of temperatures are important. First, the temperature T_3 at which three dimensional long range order occurs and where a phase transition is detected as in 3-D materials. Secondly, the temperature T_1 such as $T_1>T_3$ which characterizes the growth of the expectation value of the order parameter without any interchain coherence. As far as superconductivity is concerned the dimensionality of the order parameter is two (amplitude and phase). The treatment of the superconducting phase transition of a Q-1-D conductor is thus equivalent to the ordering of a Heisenberg two dimensional ferromagnet and the transition temperature is given by[17]

$$T_3 \sim (\alpha_{\parallel} \alpha_\perp)^{1/2} \qquad (1)$$

where $\alpha_{\parallel}$ and $\alpha_\perp$ are respectively the intra and interchain superconducting coupling strenghts. $\alpha_{\parallel}$ is related to the temperature T_1 (or the amplitude of the pseudo-gap at the Fermi level of the quasi particle density of states) and $\alpha_\perp$ is given by the interchain Josephson coupling $t_\perp^2 / t_{\parallel}$ ($t_{\parallel}$ is the intrachain overlap integral). A weak interchain coupling can lead to a sizeable depression of the transition temperature by phase-fluctuations of the order parameter below T_1.

From the experimental stand-point the temperature T_3 which corresponds to a real phase transition of the Q-1-D conductor is relatively easy to determine since

singularities in the transport, magnetic, thermodynamic etc. .. properties are expected to occur there in a way very similar to that of 3-D conductors. The temperature T_1 possesses however a slightly more abstract meaning. No singularities of thermodynamic functions occur at T_1, sometimes refered as the 1-D mean-field transition temperature i.e. the temperature where a real phase-transition would occur within the mean-field approximation for the 1-D electron gas.

EXPERIMENTAL: THE $(TMTSF)_2X$ FAMILY

Effects of low dimensionality on the electronic properties of organic conductors have first been encountered in the extensive studies of the Peierls transition[7]. As far as the CDW instability is concerned both a gap in the density of states around the Fermi level and a contribution of the fluctuating CDW's to the conduction are expected to take place below T_1[18]. This question is by now well established in TTF-TCNQ thanks to unambiguous experiments such as commensurability induced pinning of the fluctuating sliding conduction[19] and far-infrared optical studies at low temperature[20].

In 1979, Bechgaard et al.[21] found that the family of Q-1-D conducting salts based on the tetramethyltetraselenafulvalene (TMTSF) molecule, namely $(TMTSF)_2X$ with $X = (NO_3, PF_6, AsF_6, SbF_6...)$ exhibits unusually large values of the conductivity at low temperature. Around 15K, conductivities of the order of $10^5 (\Omega cm)^{-1}$ have been reported for $(TMTSF)_2PF_6$ or $(TMTSF)_2NO_3$. The increase of the conductivity at low temperature is however interrupted by the occurence of a sharp transition towards an insulating ground state. As far as $(TMTSF)_2PF_6$ is concerned the metal-insulator transition occurs at $T_{M-I} \simeq 12$ K under ambient pressure. $(TMTSF)_2X$ salts are cation-radical salts of TMTSF with inorganic anions $X = NO_3, BF_4, PF_6, AsF_6, SbF_6, TaF_6, ClO_4, ReO_4$, etc....in which a positive charge is shared between two organic molecules. The three-dimensional structure consists of a triclinic columnar arrangement of TMTSF molecules in the form of sheets of donor stacks separated by anion sheets[22], figure 1. The three dimensional side-view of the structure as shown on figure(1) could imply that the potential created on the TMTSF molecules by the anions on the left and on the right of a given stack cancel each other. However, a consideration of the full three-dimensional triclinic structure indicates that no such cancellation

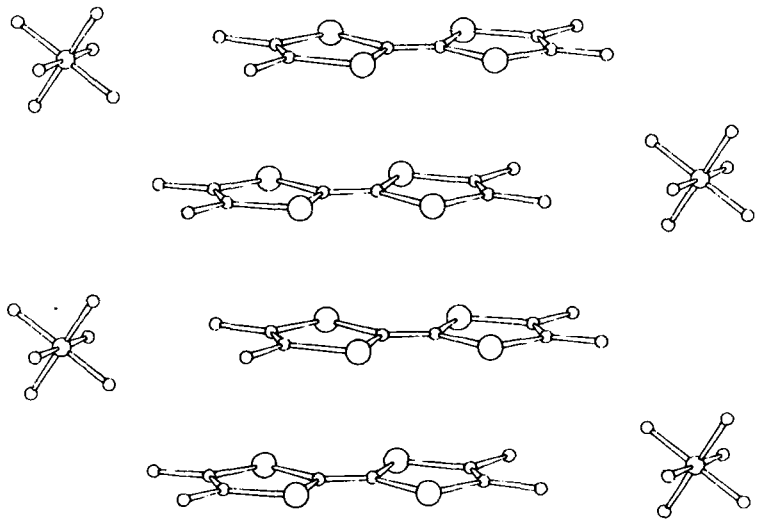

FIGURE 1 A side-view of the $(TMTSF)_2PF_6$ structure. The high conductivity direction coincides with the stacking axis. PF_6 anions are located at inversion centers of the triclinic unit cell.

occurs, and therefore the periodicity of the potential seen by the delocalized charges on the TMTSF stack is fixed by the distance between the anions, namely a. Consequently, the conduction band becomes half-filled (instead of 1/4 filled as suggested by the stoechiometry and figure 1. The existence of a weak dimerization of the average intermolecular distance is the signature of the a lattice periodicity along the stacking direction.

At room temperature, under ambient pressure, the dimerization amounts to 0.08% and 0.027% in $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ respectively [22,23]. It is however interesting to notice that the sulfur analogue series of the

$(TMTSF)_2X$ family exhibits a dimerization which, crudely speaking, is one order of magnitude larger, since it amounts to 0.6% at 300 K in $(TMTTF)_2Br$ [24] (the less dimerized salt of the sulfur series).

The $(TMTSF)_2X$ family exhibits a wide spread of electronic properties, with the perrhenate (ReO_4) and the tetrafluoroborate (BF_4) becoming insulating at 180 K and 40K respectively[21,26], the hexafluorate salts (PF_6, AsF_6, TaF_6 SbF_6) undergoing a metal to insulator transition at low temperature[21,27] ($T_{M-I} \simeq 12$ K), see figure 2, and the perchlorate (ClO_4) remaining in a highly conducting state, with $\sigma > 10^5 (\Omega cm)^{-1}$ down to helium temperature[28].

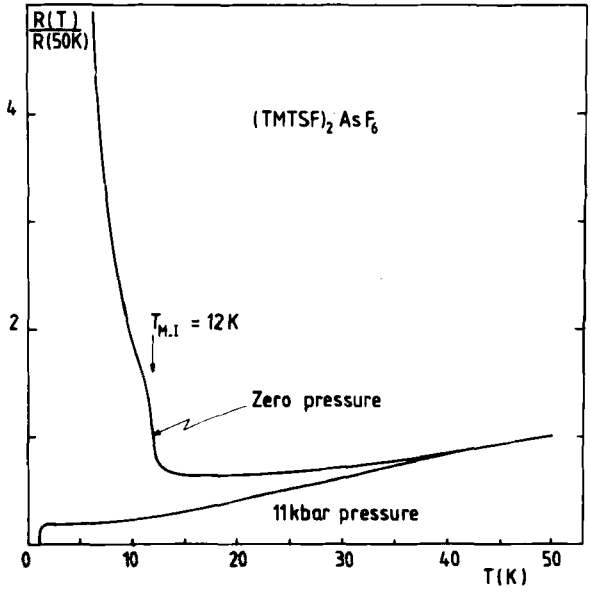

FIGURE 2 Temperature dependence below 50 K of the $(TMTSF)_2AsF_6$ longitudinal resistance normalized to unity at 50 K, showing a metal to insulator transition at 12 K under ambient pressure and the stabilization of a highly conducting state at low temperature under 11 Kbar, becoming superconducting at T = 1.1 K.

The salient feature of the conductivity of a salt such as $(TMTSF)_2PF_6$ remaining highly conducting below 20 K is the discrepancy existing between the temperature dependence of the DC conductivity increasing by x 100 between ambient and 20 K and the optical conductivity increasing by no more than a factor 3.5 in the same temperature interval [29]. A similar discrepancy observed between the temperature dependences of DC and optical conductivities of TTF-TCNQ [30] has been taken as a suggestion for the existence of fluctuating collective conductivity in that material.

However, the fluctuating Fröhlich mechanism cannot contribute in a significant way to the conduction above T_{M-I} since for the half-filled band situation of $(TMTSF)_2X$ the pinning of the CDW's is likely to be especially strong [31]!

Furthermore, contrasting with all other studied charge transfer Q-1-D molecular conductors, X-ray diffuse scattering investigations have failed to detect any appreciable precursor scattering with wave vector $2k_F$ at T larger than T_{M-I} [32] as expected if the insulating state observed below T_{M-I} in $(TMTSF)_2PF_6$ were of a Peierls nature [33]. Instead, magnetic measurements performed in the insulating state of salts such as $(TMTSF)_2PF_6$ and $(TMTSF)_2AsF_6$ have provided clear informations regarding the nature of the insulating phase of these salts. The existence of internal magnetic fields at $T<T_{M-I}$ has been suggested by the inhomogeneous broadening of the NMR methyl-group protons [34,35] and ^{77}Se (I=1/2) [34] signals figure 3, and by an additional spin-lattice relaxation mechanism occuring in the insulating state [34]. $1/T_2$ (the homogeneous line width) decreases significantly below 12 K, figure 4. This has been attributed [34] to the detuning of the proton spins on a microscopic scale i.e the difference in magnetic fields at the site of two neighbouring protons is larger than the inter proton dipolar field.

The ESR line disappears abruptly in the insulating state below T_{M-I} [36], whereas hardly any change of the powder susceptibility is noticed with a Faraday susceptibility technique [37].

Moreover, single crystal susceptibility measurements [38] have revealed a behaviour of the susceptibility of the insulating state analogous to that of an antiferromagnet. Below T_{M-I}, the easy axis of the magnetization [39] lies close to the b"-axis of the crystal structure whereas a and c"-axis are respectively the intermediate and hard axis. Field dependence studies of the magnetization show the existence of a spin-flop transition along b" at fields

exceeding $\gtrsim$4.5 kOe[39].

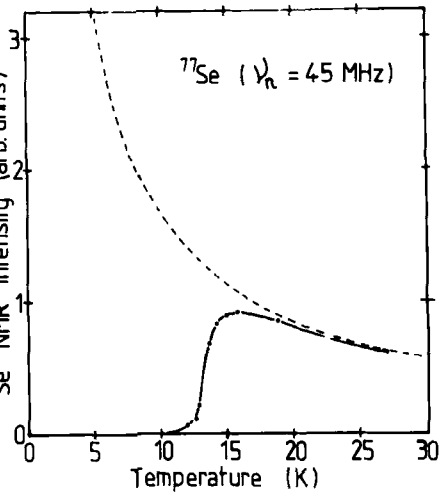

FIGURE 3 The ^{77}Se(I=1/2)NMR signal versus temperature in$(TMTSF)_2PF_6$. The signal vanishing observed below 12 K, instead of a Curie law temperature dependence (dashed curve), is due to the inhomogeneous broadening by internal magnetic fields.

These results point out that unlike TTF-TCNQ and similar compounds exhibiting a Peierls insulating ground state, magnetism remains present in the insulating state of$(TMTSF)_2PF_6$,(AsF_6).

The possibility for the occurence of magnetic ordering i.e.spin density waves(SDW) in low dimensional conductors has been suggested by Slater[40]. Quite generally the stabilization of a SDW state implies the predominance of the repulsive electron-electron over the electron-phonon interactions. Therefore, in a SDW state the exchange part of the electron potential couples the states at $+k_F$ and $-k_F$ and opens an exchange gap at the Fermi level which transforms the Q-1-D conductor at high temperature into an insulator below T_{M-I}. The establishment of a SDW state is very strongly favoured by the quasi planar geometry of the Fermi surface. The new periodicity of the electron

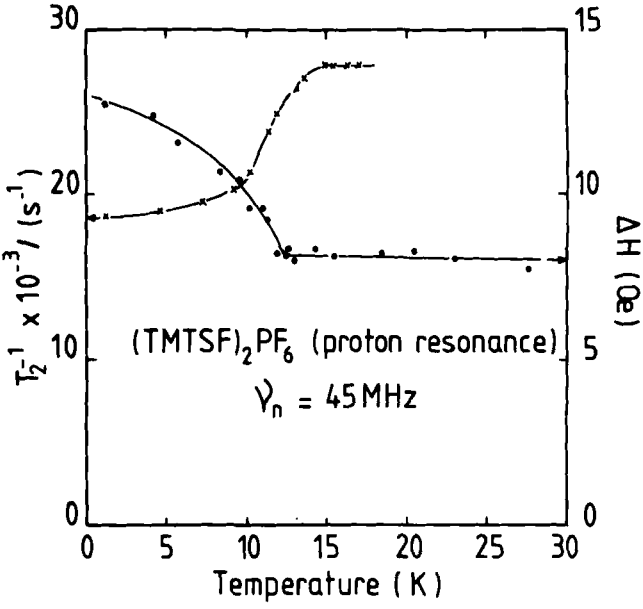

FIGURE 4 The proton NMR signal in $(TMTSF)_2PF_6$ revealing an inhomogeneous line broadening and a homogeneous line narrowing below 12 K.

potential is likely to be 2a along the stacking axis, but neutron diffraction techniques have failed so far to show any evidence of magnetic scattering.

The temperature dependence of the resistivity of $(TMTSF)_2X$ salts comprising non-centrosymmetric anions such as triangular NO_3 [41], or tetrahedral BF_4, ReO_4 [25] and ClO_4 [28] ions reveals some interesting features related to the anion organization. The anions are located on inversion centers of the triclinic unit cell of the 3-D structure. Therefore in absence of magnetic doubling of the unit cell the wavelength of the potential sensed by the electrons moving along the stacking axis is either a or 2a depending on the geometry of the anion. The wavelength is a for symmetrical octahedral anions (PF_6, AsF_6, etc...) or statistically disordered non-centrosymmetric anions but is doubled if non-centrosymmetric ions order. The latter situation is encountered in the NO_3, BF_4 and ReO_4 salts below the order-disorder transition T_{OD} at 35,39 and 180 K respectively [42].

As far as tetrahedral anions are concerned, X-ray diffuse scattering[42] has shown that the superlattice wave vector is ($2k_F$, π/b, π/c). This is also the vector which provides the best nesting of Q-1-D Fermi surfaces. In $(TMTSF)_2NO_3$ however, the observed ($2k_F$, 0, 0) superlattice wave vector[43] does not allow a gap to open on the whole area of the Fermi surface of the Q-1-D conductor. At T_{OD} a small resistance anomaly is recorded, but no gap opens in the density of states, figure 5. The transition towards an insulating state observed below 10 K corresponds probably to the onset of a SDW state as for $(TMTSF)_2AsF_6$ or PF_6.

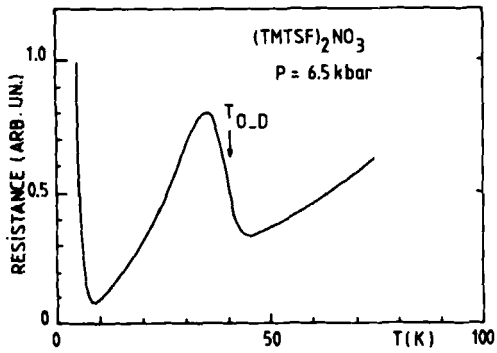

FIGURE 5 Temperature dependence of $(TMTSF)_2NO_3$[41] longitudinal resistance under 6.5 Kbar showing both (i) the metal to semi metal transition occuring around 40 K, with a small reduction of charge carriers as a result of NO_3 ions ordering with wave vector (1/2, 0, 0), (ii) the metal to insulator transition at 6 K, which is very similar to the one observed in $(TMTSF)_2PF_6$ or $(TMTSF)_2AsF_6$ at the same pressure. The low temperature transition is probably driven by magnetic ordering (this awaits further investigations).

$(TMTSF)_2ClO_4$ is the only salt of a tetrahedral anion known so far in which the anions remain disordered down to low temperature. Correspondingly the resistance of $(TMTSF)_2ClO_4$ exhibits a continuous fall by about two orders of magnitude between room temperature and 5 K[28]. Below 5 K a slight up turn of the resistance is observed in good quality (low resistance) crystals[27]. The resistance at 1.2K

can become two or four times larger than the value at 5 K.

PRESSURE STUDIES

The stabilization of a highly conducting state is possible under pressure for all the members of the $(TMTSF)_2X$ series (X=PF_6 [1], AsF_6 [32], TaF_6 [27], SbF_6 [27], BF_4, ReO_4 [44] and NO_3 [41]. The phase diagram of $(TMTSF)_2PF_6(AsF_6)$ is displayed on figure 6. As shown on fig (6), the SDW state is very quickly suppressed by the hydrostatic pressure above 6 Kbar. Above 8.5 Kbar the conducting state of $(TMTSF)_2PF_6(AsF_6)$ undergoes a transition towards a superconducting state at low temperature, as shown by a zero resistance, diamagnetic state [1,45,46], figure 7. In the pressure regime where SDW and superconductivity coexist with a common phase transition line, the phase diagram is rather complex [47]. In fact, a detailed pressure investigation performed on the $(TMTSF)_2AsF_6$ phase diagram shows that superconductivity is reentrant under the SDW state at low temperature in that pressure regime [48]. No superconductivity has yet been observed in $(TMTSF)_2NO_3$ down to 50 mK either by resistive or inductive techniques [41] above 8Kbar, figure 8. The figure 9 shows that the order-disorder transition of the anion NO_3 remains practically unaffected by pressure [41].

As displayed on figure (6), the superconducting critical temperature is strongly pressure dependent with $\partial \ell n T_c / \partial P \simeq -7\%$ Kbar^{-1} at 11 Kbar [49]. Although the suppression of the superconduction under pressure, a general behaviour in the $(TMTSF)_2X$ series, is not fully understood, we feel it cannot be attributed solely to a band broadening (decrease of $N(E_F)$ or to an increase of the 3-D coupling under pressure). In fact, the Seebeck coefficient of $(TMTSF)_2PF_6$ does not vary significantly at 300 K between 1 atm and 12 Kbar [50] and the pressure dependence of the longitudinal conductivity obtained without sample breaks in $(TMTSF)_2NO_3$ up to 12 Kbar [41] which is analogous to the $\sigma_\perp$ pressure dependence, figure 10 shows that neither $t_{//}$ nor the anisotropy $t_{//}/t_\perp$ can be drastically affected by pressure.

Magnetization curves of the superconducting states of $(TMTSF)_2X$ salts show that field penetration occurs already at very small values of magnetic field, namely $H_{c_1}^\perp$ is lower than the earth magnetic field or so . Superconductivity in $(TMTSF)_2X$ follows a type II behaviour. The upper critical field $H_{c_2}^\perp$ is easily measured by the field at which

the resistivity recovers its value above T_c.

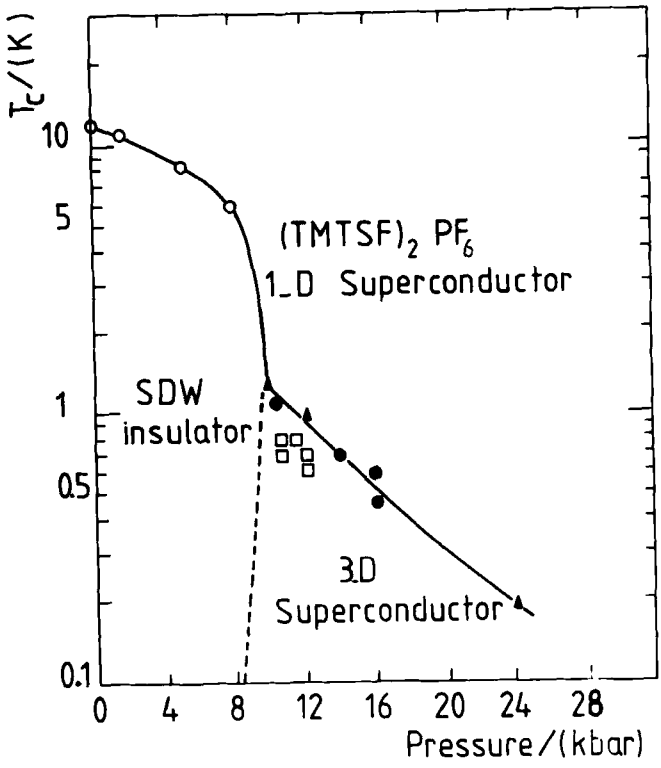

FIGURE 6 The phase diagram of $(TMTSF)_2PF_6$. The various experimental points on the superconducting transition line have been compiled in the literature, Orsay ▲ [49], Bell Labs ● [46] and IBM ◘ [47]. A closer investigation of the similar phase diagram for $(TMTSF)_2AsF_6$ [48] shows a slight reentrance phenomenon near the triple point at $P \approx 8.5$ Kbar, $T \approx 1.2$ K.

The angular dependence of $H_{c_2}^{\perp}$ reveals a very pronounced anisotropy of the critical field in the plane perpendicular to the stacking axis. For example in $(TMTSF)_2AsF_6$ the ratio $H_{c_2}//b^* / H_{c_2}//c^*$ is equal to 15±2 under 9.5 and 11 Kbar [48]. figure 11. This anisotropy of $H_{c_2}^{\perp}$ reveals a band anisotropy along the transverse directions.

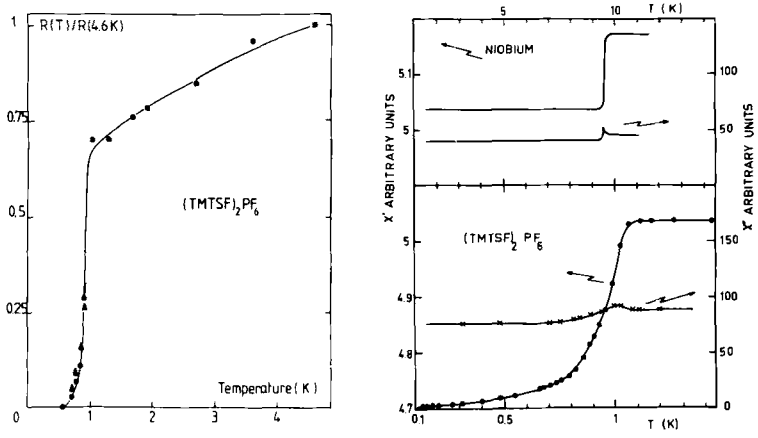

FIGURE 7 The superconducting transition of $(TMTSF)_2PF_6$ under 12 K at T_c=0.9 K detected by resistance measurements (left) and AC-susceptibility methods (right). A marker sample (niobium metal) located in the pressure cell, $T_c \simeq 9.5$ K, is used for the purpose of signal calibration.

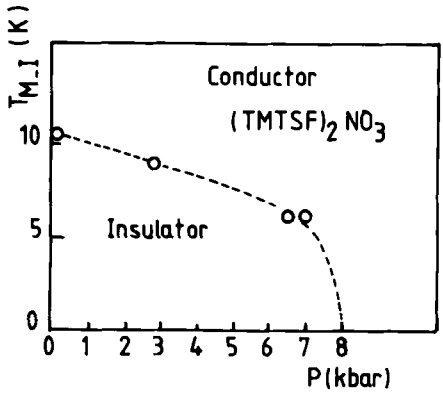

FIGURE 8 The phase diagram of $(TMTSF)_2NO_3$. Above 8 Kbar a highly conducting phase is stable at low temperature, but no superconductivity has yet been found either resistively or inductively down to 50 mK.

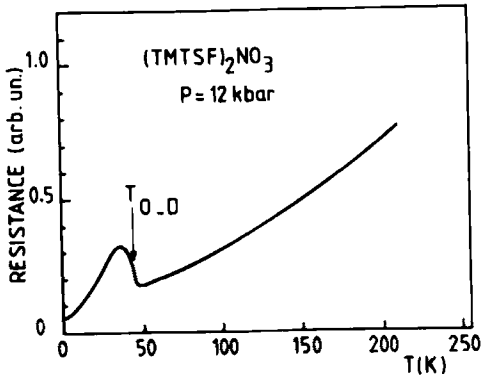

FIGURE 9 The temperature dependence of the $(TMTSF)_2NO_3$ longitudinal resistance at 12 Kbar showing (i) the stabilization of the conducting state at low temperature and (ii) the order-disorder transition around 40 K which is hardly affected by pressure between 1 atm and 12 Kbar.

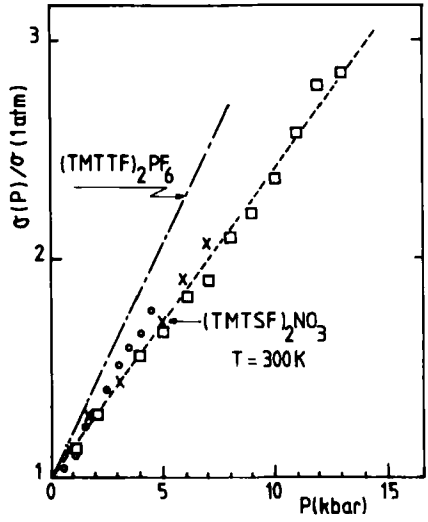

FIGURE 10 The pressure dependence of the longitudinal conductivity of $(TMTSF)_2NO_3$ at T=300K, increasing at a rate of 15% $Kbar^{-1}$. Various symbols correspond to different samples. The data displayed on this figure have been obtained without sample breaking. The transverse conductivity exhibits the same pressure coefficient. The dash-dot line shows the P-dependence of the longitudinal conduction of the sulfur analogue salt $(TMTTF)_2PF_6$ [41].

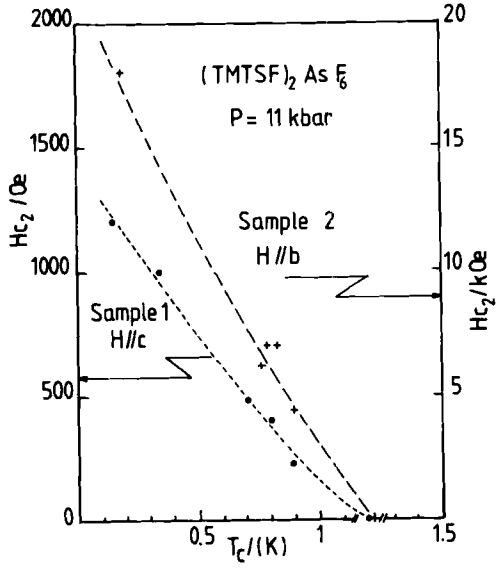

FIGURE 11 Anisotropy of the upper critical field $H_{c_2}^{\perp}$ of $(TMTSF)_2AsF_6$ under 11 Kbar in the a-b plane.

Within the frame work of the Landau-Ginzburg theory for the superconducting state below T_c with open Fermi surfaces, the anisotropy of coherence length and band parameter are equal[51]. Therefore, as far as $(TMTSF)_2AsF_6$ is concerned $t_{\perp}^{b''}/t_{\perp}^{c''} = 15\pm2$. The accurate measure of the anisotropy between the stacking axis direction and the transverse directions is hard to reach via critical fields experiments since it requires an alignment of the sample with the external field which is very difficult to achieve with small crystals.

The band structure anisotropy can also be derived from DC and microwave conductivity anisotropy. The DC and 35 GHz a to b conduction anisotropy amounts to 450 and 90 respectively for $(TMTSF)_2PF_6$ at ambient temperature[21]. These values correspond to a band structure anisotropy $t_{//}/t_{\perp}^{b''}$ in the range of 10 to 20 following the relation $\sigma_{//}/\sigma_{\perp} \simeq (t_{//}/t_{\perp})^2$ which is obeyed when the transverse conductivity is diffusive[52]. Plasma edge studies on $(TMTSF)_2PF_6$ at 25 K lead to transfer integral of 250 meV and 3 meV for a and b'' axis respectively[29]. Therefore, order of

magnitude wise it seems reasonable to take $t_{//}/t_{\perp}^{b^*} \simeq 30$ or so. Consequently, the band parameters of the $(TMTSF)_2PF_6$ and $(TMTSF)_2AsF_6$ structures are within the ratios $t_{//}^{a} : t_{\perp}^{b^*} : t_{\perp}^{c^*} = 1, 1/30, 1/450$ respectively, with an anisotropy which is independent of pressure.

THE FLUCTUATIONS ABOVE T_3

So far, this article has been concerned with the phenomenon of superconduction occuring at very low temperature ($T < 1.2$ K), whenever the resistance drops to zero. However, above T_c the electronic properties of $(TMTSF)_2X$ are remarkably different from those of regular metallic conductors. We shall list now some of these remarkable properties.

a) The longitudinal conductivity overcomes 10^4 $(\Omega cm)^{-1}$ below 40 K or so, and reaches $\simeq 10^5 (\Omega cm)^{-1}$ around 4.2 K (21, 1) i.e. a value which is about 10 times larger than the maximum conductivity observable in other conducting charge transfer compounds at low temperature. Given the electron density of 10^{22} charges/cm^3, a mobility of $\simeq 2.10^3$ cm^2/V.S is derived at 1.3 K (for $\sigma_{//} = 5.10^5$ $(\Omega cm)^{-1}$) leading to an approximate mean-free path of 700 intermolecular spacings along the a axis (m^* = free-electron mass).

b) The transverse conductivity displays about the same temperature dependence as the longitudinal component down to 50 K and increases somewhat faster at low temperatures (51, 53), see figure 12.

c) The longitudinal low temperature ($1.2 < T < 30$ K) conductivity is extraordinarily affected by the application of a magnetic field along the transverse c^*-direction (53) but the magnetoresistance is reduced for $H // b^*$ (48) and even practically inexistant up to 75 kOe as $H // a$.

d) The temperature dependence of the electrical resistivity is still large below 4.2K, in a temperature domain where the resistance of all 3-D metallic conductors is limited by residual impurities (1) figure (7).

Admittedly the picture of especially light and anisotropic carriers in motion along stacks of great perfection could probably explain these peculiar transport properties. However, we find such an interpretation rather unlikely considering (i) the optical effective mass along the chain axis close to the free electron mass (29) (ii) the extraordinary sensitivity of the resistance at low temperature in the conducting regime to the presence of defects since $\Delta\rho \simeq 3000$ $\mu\Omega$cm per % of defects (54). When a similar highly

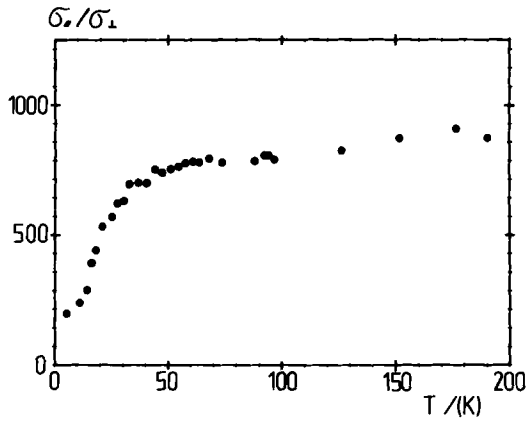

FIGURE 12 Temperature dependence of the conductivity anisotropy for $(TMTSF)_2PF_6$ at 12 Kbar. The transverse direction has not been determined in this experiment.

conducting state had been stabilized under pressure in the charge-transfer compound TMTSF-DMTCNQ, with $\sigma_{//}>10^5\,(\Omega cm)^{-1}$ at 1.2 K and P>10 Kbar(55)we suggested the existence of a gradual growth of a fluctuating superconducting pairing arising below 30 K(56)figure 13.

In order to support a similar interpretation for the unusual properties of conducting $(TMTSF)_2X$ at low temperature, experiments probing the density of states around the Fermi level have been carried out.

As a matter of facts, as short range superconducting order develops on each chain below the temperature T_1 or so, the growth of the average amplitude of the order parameter must be accompanied by the occurence of a pseudo-gap in the density of quasiparticle states at the Fermi level (57,58). This pseudo-gap could be visible at $T<T_3$ but also at $T_3<T<T_1$ since it characterizes the onset of 1-D fluctuations. This is the reason which triggered the investigation of the superconductivity by electron tunneling spectroscopy, a technique which has been widely used to derive the quasiparticle density of states in metallic superconductors(59).

As far as $(TMTSF)_2PF_6$ is concerned the Schottky barrier

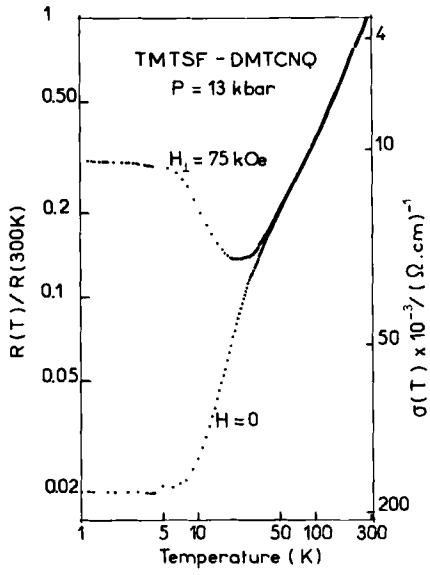

FIGURE 13 Temperature dependence of the longitudinal resistance of TMTSF-DMTCNQ[55] at 13 Kbar (i) without magnetic field $\sigma_{\parallel}$ reaches $2\times10^{5}(\Omega cm)^{-1}$ at low temperature, (ii) a very large magnetoresistance is observed up to 30 K for a transversely applied magnetic field.

technique has been used with some success[60]. Schottky junctions are prepared by evaporating tellurium doped GaSb($N_D>3.10^{18}$ cm^{-3}) on a natural face of a$(TMTSF)_2PF_6$ single crystal. After pressurization and cooling, the differential resistance of the junction is studied versus the bias voltage. The large resistance maximum at zero bias, together with the two minima observed on each sides of the characteristics, point out the existence of a gap in the quasiparticle density of states, figure 14. The recording on figure (14) has been performed under 11 Kbar, at T=50 mK. Following the traditional interpretation of the tunneling data[59] the pairing energy 2Δ on figure 14 amounts to 3.6 mV (40 K) i.e. about 40 times the critical temperature for $(TMTSF)_2PF_6$ at 11 Kbar.

At high temperatures, namely $T>T_3$, the amplitude of the zero bias resistance maximum decreases steadily, but remains clearly visible up to about 15 K. A large magnetic

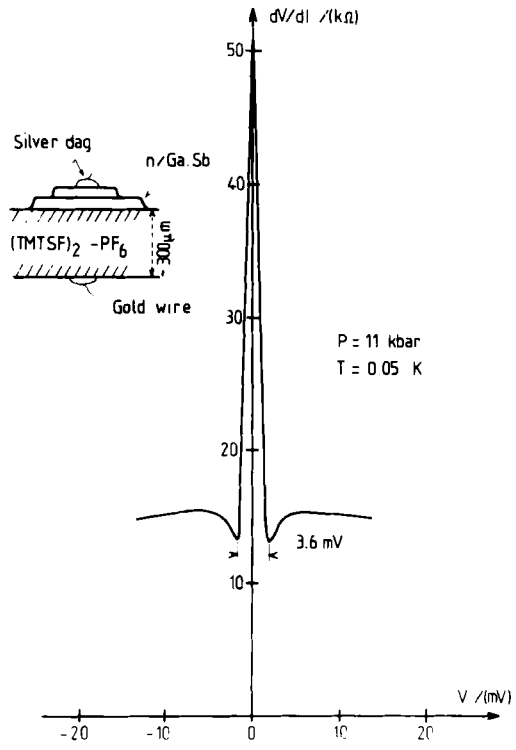

FIGURE 14 Tunneling characteristics of a $(TMTSF)_2PF_6$-N/Ga-Sb Schottky junction under 11 Kbar at 50 mK. The energy difference between the two sharp resistance minima is taken as a measure of the pseudo-gap in the density of quasiparticle states i.e.2Δ = 3.6 meV (40K).

field applied in a direction perpendicular to the stacking axis removes the large zero-bias resistance peak. Results such as those displayed on figure (14) have been interpreted by the opening of a gap in the quasiparticle density of states due to the formation of superconducting pairs in $(TMTSF)_2PF_6$. Electron tunneling data suggest that the gap of $2\Delta = 40$ K is not related to the establishment of a 3-D superconducting state below T_3 but instead to the onset of a very large intrachain pairing up to T_1.

A similar behaviour has been observed under ambient pressure with a Schottky barrier performed on $(TMTSF)_2ClO_4$

(61). There, a pseudo-gap $2\Delta \sim 4$mV in the tunneling characteristics is visible at low temperature.

However in a large number of junctions the tunneling data display a behaviour different from that shown on figure 14. Often a zero bias resistance minimum develops at low temperature and once in a junction of relatively low resistance ($\sim 10\Omega$) at high temperature, the zero bias contact resistance dropped sharply to zero below 12 K, figure 15. Such a behaviour can be understood in terms of a superconducting short-circuit at the barrier(62). That a zero resistance superconducting state can be stabilized at 12 K in the vicinity of the barrier is not too surprising within the model of Q-1-D superconductivity.

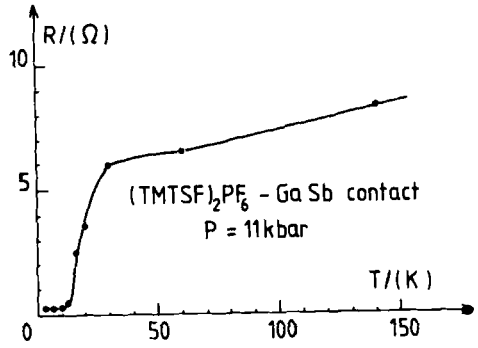

FIGURE 15 Temperature dependence of a $(TMTSF)_2PF_6$-GaSb contact under 11 Kbar i.e. the resistance at zero bias (according to figure 14). The superconducting short circuit below 12 K suggests the presence of 3-dimensionally stabilized superconducting domains in the vicinity of the interface between the organic conductor and the evaporated semiconductor.

Any mechanism establishing interchain coherence for the phase of the order parameter should have the tendency to raise T_3. Randomly distributed interchain cross-links via foreign atoms allowing electron pairs to jump from one chain to its neighbour through local interchain bridges could lead to a significant T_c enhancement(62). The

calculation[63] shows that a concentration of 4% per molecule of cross-links (with $t_\perp$ (local) = $t_{//}$) is large enough to raise T_3 given by eq.1 up to the neighbourhood of T_1.

The temperature of 12 K, below which the $(TMTSF)_2PF_6$-GaSb contact becomes superconducting, is very close to the 1-D mean-field temperature derived from the weak coupling limit BCS relation, namely $2\Delta/kT_c = 3.5$. This agreement is probably not very significant, but order of magnitude wise the tunneling data show that the onset of superconducting pairing along each chain occurs in a temperature domain which is at least 10 times larger than the temperature where 3-D ordering takes place in the bulk of a crystal.

Cross-linking can be achieved via the diffusion of foreign atoms (for example antimony) between organic stacks in the vicinity of the interface between the $(TMTSF)_2PF_6$ crystal and the evaporated Ga-Sb layer[62].

Several other interesting observations have been made with Schottky barriers and point-contact junctions on $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$. They are reported in other publications[64].

Other studies of the electronic properties of $(TMTSF)_2PF_6$ between 4.2 and 77 K have led to an interpretation which corroborates the tunneling data. Such is the case of thermal conductivity experiments performed on $(TMTSF)_2PF_6$ under 12 Kbar[65]. The thermal conductance displays a very large drop at low temperature, beginning around 40 K. Since the Wiedeman-Franz law is fairly well obeyed at high temperature, namely, σ and K increase both with the same rate at 300 K between 1 atm and 12 Kbar, we feel fairly confident that the electronic contribution is dominant in the total thermal conductivity at 40 K. The drop of the thermal conductivity below 40 K can therefore be related to the opening of a pseudo-gap at the Fermi level . Spin susceptibility measurements in $(TMTSF)_2ClO_4$ using the EPR technique[28] reveal a significant drop of the density of states below 7 K, whereas the transition towards the superconducting state occurs at best below 1.2 K (under zero magnetic field). Similar precursor effects have been recently observed with $(TMTSF)_2PF_6$ under pressure[66]. We consider that they bring additional support to the existence of pseudo-gap far above the superconducting critical temperature.

Finally, far infrared reflectance properties of $(TMTSF)_2PF_6$ at 25 K under ambient pressure[67] support the existence of a pseudo-gap around 50 cm^{-1} and a very large contribution to the frequency dependent conductivity which is sharply peaked around zero frequency.

In the presence of a gap(or pseudo-gap)at the Fermi level the density of particles able to contribute to the conduction is likely to decrease with decreasing temperatures. This is however in striking contrast with the huge enhancement of the conduction observed experimentally at low temperature either in $(TMTSF)_2PF_6$ under pressure[1] or even in $(TMTSF)_2ClO_4$ at ambient pressure[78].

As it was suggested following the study of TMTSF-DMTCNQ the formation of fluctuating pairs can contribute to the conduction even at temperatures much larger than T_3 in Q-1-D conductors[56,51]. Since the excess conduction(or paraconduction) is proportional to $\langle|\Delta|^2\rangle$, it becomes negligible above T_c in most 3-D superconductors. But the problem is quite different whenever the volume of a fluctuating domain is reduced by a factor $(d_\perp/\xi_{//})^2$, as in the 1-D fluctuation regime($\xi_\perp < d_\perp$) of a Q-1-D superconductor[51].

The paraconductivity of a Q-1-D superconductor worked out by Schulz[51] leads to the following contribution:

$$\sigma'_{//} = \frac{\pi e^2}{16\, d_\perp^2}\, \xi_{//}(0)\, \varepsilon^{-3/2} \qquad (2)$$

where $\varepsilon = \frac{T-T_1}{T_1}$ and $\xi_{//}(0)$ is the zero temperature Landau-Ginzburg coherence length. Besides supercurrents along the chains in the fluctuating regime, transverse supercurrents are likely to occur between adjacent chains with well developed order parameters. As the likelihood of this occurence decreases with increasing temperature $\sigma'_\perp$ decreases in temperature faster than $\sigma'_{//}$, i.e.$\sigma'_\perp \underset{\sim}{\propto} \varepsilon^{-5/2}$.Consequently the anisotropy of paraconductivity varies linearly with temperature above T_1 and is likely to explain the behaviour of the experimental data below 50 K in $(TMTSF)_2PF_6$ under 12 Kbar, figure(12). A numerical estimate of the paraconduction, eq(2), leads to $\sigma'_{//} > 10^4\,(\Omega cm)^{-1}$ at T=25 K, given $\xi_{//}(0)/d_\perp \gtrsim 50$ and $T_1 \lesssim 12$ K according to the tunneling data in $(TMTSF)_2PF_6$ under pressure[60].

Following the previous estimate, the paraconductivity is largely dominant at helium temperature and consequently is likely to be suppressed by (i) the application of a magnetic field along a transverse direction and (ii) a low concentration of magnetic defects created by irradiation (54).

CONCLUSION

In this conclusion section we shall summarize the most significant experimental results of Organic Superconductivity and attempt to suggest some guidelines towards an interpretation of the phenomena.

The properties of the superconductivity which occurs in the conducting cation-radical salts $(TMTSF)_2X$ is strongly influenced by Low Dimensionality. In this respect the tunneling data have answered key questions. Short range superconducting pairing takes place along the chains (without interchain coherence) at temperatures as high as 15 K. The impact of this short range pairing manifests itself on the quasi particle density of states by the observation of a pseudo-gap at the Fermi level below 15 K or so in $(TMTSF)_2PF_6$ under pressure. Moreover the ratio $2\Delta/kT_c$ of about 40 for $(TMTSF)_2PF_6$ under 11 Kbar indicates that the actual critical temperature T_3 below which both a zero resistance state and magnetic flux expulsion are observed is not determined solely by the strength of the superconducting pairing within each chain. The superconducting phase transition occurs when coherence establishes between the phases of the order parameters on adjacent chains. As far as superconductivity is concerned, the transverse couplings which matter most are on the one hand the so-called transverse tunneling coupling $t_\perp$ which allows an electron to travel from one chain to its neighbour, and on the other hand the Josephson coupling $t_\perp^2/t_{//}$ which can transfer an electron pair between the chains. There exists a broad temperature domain extending up to 30 K or so in which the electronic properties of conductors like $(TMTSF)_2ClO_4$ or $(TMTSF)_2PF_6$ are to a very large extent dominated by the onset of interchain superconducting order, (i) the pseudo-gap in the density of states observed by tunneling and FIR reflectance, (ii) the large paraconductivity rising below 40 K which can be removed by the application of a magnetic field in the appropriate transverse direction or a very low concentration of defects (iii) the drop of thermal conductivity and Seebeck coefficient below 40 K.

Moreover, one can probably take advantage of the 1-D fluctuation regime and stabilize the 3-D ordered superconducting state at a temperature related to the amplitude of the intra-chain pairing energy (i.e. 12 K, for $(TMTSF)_2PF_6$ under 11 Kbar) provided the interchain coupling is enhanced via a small concentration of intercalated foreign atoms, establishing bridges between the chains. Preliminary results on $(TMTSF)_2PF_6$ tend to support that T_c

enhancement is possible following this approach. Besides, One Dimensionality of organic conductors is likely to control the nature of the coupling leading to an electron pairing. We consider that the existence of a common phase limit in the phase diagram of the $(TMTSF)_2X$ series between superconductivity and a magnetic state[68] is the crucial experimental evidence which should guide the interpretation. The key question is the following: which of the two interactions (electron-phonon coupling or electron-electron Coulomb repulsion)[69] are dominant in the pairing mechanism?..

Within the framework of the 1-D theory[70] which reduces the interaction to two coupling constants g_1 and g_2 some arguments support the idea of a non-phonon mediated pairing, namely (i) the interplay between superconductivity and a magnetic state which suggests the presence of dominant Coulomb interactions, (ii) the enhancement of the spin susceptibility at high temperature and its non activated temperature dependence which agrees with a positive g_1[71] (iii) the observation of (weak) 1-D $4k_F$ X-ray diffuse scattering at high temperature in $(TMTSF)_2PF_6$, $(TMTSF)_2ClO_4$ and ReO_4[72]. Admittedly 1-D theory is not appropriate for the determination of phase transition temperatures, but a refinement of the theory shows that the nature of low temperature phases (depending on the respective values of g_1 and g_2) is not significantly modified by the inclusion of an interchain tunneling coupling[15]. However, the role played by the retarded electron-phonon interactions versus the non retarded electron-electron interactions awaits further investigations. It is very likely that the stability of the dielectric SDW state is favoured by the presence of Umklapp scattering of two electrons from one side of the Fermi surface to the other. All theories seem to agree on the important role played by Umklapp scattering on the nature of the ground state[68,73]. Under ambient pressure, the dimerization along the a-axis (and consequently the amplitude of the Umklapp scattering) appears to be smaller in $(TMTSF)_2ClO_4$ where superconductivity is observed than in $(TMTSF)_2PF_6$ where a dielectric SDW state is the stable ground state. However it is only with a structure determination of $(TMTSF)_2PF_6$ under pressure that the role of Umklapp scattering will be more firmely established.

In a last (but not least) remark we wish to reemphasize that probably the most original aspect of organic superconductivity in the Q-1-D series $(TMTSF)_2X$ is the existence of two energy scales governing the onset of the superconductivity instability. (i) A small energy scale (of the order of a few degrees Kelvin) which sets the establishment of 3-D order.

This energy fixes the "true" critical temperature ($T_c < 1.2K$) and the values of the critical fields.

(ii) A much larger energy scale ($\tilde{\sim}$ 40 K) which is the characteristic of isolated chains and consequently not in direct correspondance with the T_c and critical fields values for the 3-D ordered state.

Furthermore, the recent observation of phenomena related to superconductivity with organic conductors built from molecules others than TMTSF, namely $(TMTTF)_2Br$ and $(TMTTF)_2PF_6$ [74] suggests that Superconduction might be a general property of Q-1-Dorganic conducting salts with stoechiometry 2:1. Besides the interest of these fascinating materials from the point of view of their new properties, we feel they represent a possible way towards the stabilization of superconductivity at high temperature.

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