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$(TM)_2X$ ORGANIC SUPERCONDUCTORS AND RELATED PHYSICS

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Organic Superconductivity has been discovered in 1979 in the one dimensional organic conductor $(TMTSF)_2PF_6$. This discovery has to be considered in the framework of a long quest for new superconductors which started fifteen years earlier with the suggestion made by W. Little [1] for a new pairing mechanism in organic conductors. The extensive studies carried on over the last 20 years have revealed the existence of a vast series of isostructural compounds, $(TM)_2X$, exhibiting signatures of a non-Fermi behavior at high temperature and a variety of ground states at low temperature. Besides superconductivity, low dimensional organic conductors have brought a wealth of new concepts in physics.

Keywords: $(TMTSF)_2PF_6$; organic superconductivity; Luttinger liquids; 1-D conductors

1. CHARGE TRANSFER CONDUCTORS IN A NUTSHELL

The discovery of the first organic molecular compound exhibiting a conductivity approaching that of good metals at room temperature created a tremendous enthusiasm in the early seventies although reports of high conductivity in molecular crystals had been made before [2]. This was the time where the first organic molecular compound, tetrathiafulvalene-tetracyanoquinodimethane TTF-TCNQ exhibiting a large and truly metal-like conductivity had been discovered [3,4]. To our knowledge, the finding of high conductivity in TTF-TCNQ has led for the first time to the suggestion made by experimentalists of superconductivity in organic matter.

This anniversary article is a unique opportunity to thank very warmly all my co-workers who have contributed at Orsay and Copenhagen to the development of the physics and chemistry of organic conductors. The development of this field has also greatly benefited from a close cooperation with theoreticians at Orsay and Sherbrooke. Organic conductors would not have developed so successfully at Orsay without the cooperation of J. Friedel. H.J. Schulz who had been a very close friend and colleague left us much too early. His contribution has had a great impact on the development of low dimensional physics.

Although the subsequent experimental studies had shown that this claim happened not to be justified, in certainly gave us the necessary motivation to go ahead and find the first organic superconductor six years latter [5].

The prerequisite for the formation of molecular conductor is first the requirement for having charged molecules in a solid and second to allow their charge to delocalize between molecular entities. Charging the molecules is achieved in TTF-TCNQ by a transfer of charge between the donor molecule TTF and the acceptor molecule TNCQ. TTF-TCNQ is a material comprising segregated parallel stacks of TTF and TCNQ molecules. The intermolecular overlap is thus optimized along the stacks of molecules and the π orbitals of the HOMO (LUMO) molecular orbitals (for TTF and TCNQ respectively) give rise to two conduction bands. The overlap of molecular orbitals being largest along the stacks (bandwidth of about 0.5 to 1 eV) and much weaker between them makes the electron dispersion one dimensional. The fact that the charge can delocalize in TTF-TCNQ shows that the on-site Coulomb-Hubbard repulsion U does not overcome the energy $4t_{\|}$ which is gained by the formation of energy bands in the solid, i.e., the bare molecular Coulomb repulsion (≈ 5 eV) experiences a strong dielectric screening by the environment which reduces it by an order of magnitude, making it about 0.3–0.5 eV [6]. It is the announcement in 1973 of a conductivity peaking at a value of 5.10^4–$10^5(\Omega\,\mathrm{cm})^{-1}$ at $T = 60$ K [3] just above the transition towards an insulating ground state which triggered a tremendous interest although the precise amplitude of the conductivity peak has been seriously questioned by several authors [7]. This early work suggested that the metal-insulator transition could be the manifestation of the instability due to the spontaneous distortion of the lattice with a wave vector $2k_F$ predicted by Peierls in 1957 for a one dimensional conductor [8]. Since, electron-electron and electron-hole pairings are known to compete in a 1-D conductor on the same footing and could possibly coexist the enormous conductivity at 60 K was ascribed to precursors of a superconducting transition. It was claimed that the softening of a phonon mode at the wave vector $2k_F$ could possibly enhance the superconducting fluctuations in the high temperature metallic regime [3]. Making the conductor less one dimensional via the increase of the overlap between stacks (applying an hydrostatic for example) was considered as one possible remedy to suppress the one-dimensional Peierls instability [9,10] and enable the superconducting instability (which should be less sensitive to the 1-D character) to develop at high temperature.

Although very brittle, organic samples like TTF-TCNQ and all subsequently discovered materials, $(TM)_2X$, etc., have been studied rather easily under hydrostatic pressure using a teflon cell up to about 35 kbar and Bridgman anvils above. It is now widely recognized that high pressure

measurements have played a decisive role in the study of most molecular conductors. They have shown that the metal-insulator transition (confirmed to be a Peierls instability by X-ray diffuse scattering experiments [11]) cannot be suppressed under pressure [12].

The response of TTF-TCNQ to high pressure can be summarized as follows: 1) It is the transverse Coulomb coupling and not the single particle hopping which stabilizes the Peierls state in TTF-TCNQ. Consequently, stabilizing superconductivity under pressure in TTF-TCNQ and parent compounds appears to be hopeless below 35 kbar. 2) The phase diagram can be understood in terms of a band filling being incommensurate ($\rho = 0.55$ carrier/unit cell at 1 bar) increasing under pressure and becoming commensurate ($\rho = 0.66$ or $2/3$) in the pressure window 15–20 kbar [13,14]. This is a major difference with the subsequently discovered superconductors where the band filling is fixed by the chemistry and thus insensitive to pressure. 3) The conductivity peak at 60 K ($\sigma(60\,\mathrm{K})/\sigma(300\,\mathrm{K}) = 15-20$) is not as large as it was first claimed but still up to 80% of its value can be ascribed to long-lived 1-D charge density wave (CDW) fluctuations becoming large in the proximity of the Peierls instability, (Figure 2). The optical properties do not follow the usual Drude law and instead the far-infrared (FIR) conductivity displays a sharp DC collective mode ($1/\tau = 4\,\mathrm{cm}^{-1}$) with a pseudo-gap of about $1000\,\mathrm{cm}^{-1}$ [15,16], a feature which is also found in superconductors *albeit* for different reasons.

The study of the two-chain charge transfer compounds went on with TMTSF-DMTCNQ (TM-DM) where the donor is the tetramethyl selenide derivative of TTF. The outcome of this study has been decisive for the quest of organic superconductivity [17]. This 1-D conductor undergoes a Peierls transition at 42 K [18] where unlike TTF-TCNQ a distortion occurs simultaneously on both chains [19]. Several other results have triggered our attention. X-ray experiments had shown that the charge transfer is only $\rho = 0.5$ leading to a quarter-filled band situation [19]. Transport and thermopower data emphasized the dominant role played by the TMTSF chain in the mechanism driving the Peierls transition and also in its contribution to the conduction at high temperature [20]. The really new and unexpected finding has been the suppression of the Peierls transition under pressure and the conductivity remaining metal-like reaching $10^5(\Omega\,\mathrm{cm})^{-1}$ under 10 kbar at the temperature of liquid helium [21]. The conducting state of TM-DM was also remarkable in displaying a huge transverse magnetoresistance below 50 K [22].

Since all these phenomena were new and unexpected the effort was put on a structure made of only one organic stack comprising the lucky TMTSF molecule. Such a structure was already known from the early work of the Montpellier chemistry group who synthesized and studied the series of

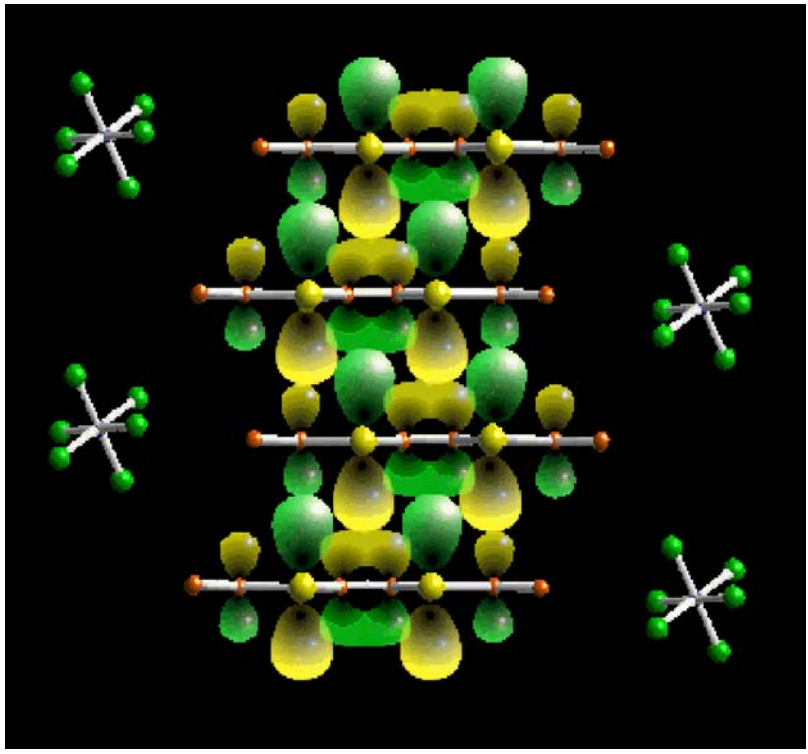

FIGURE 1 View of the $(TM)_2X$ structure, courtesy of IMN, Nantes. (See Color Plate I)

isostructural $(TMTTF)_2X$ organic salts [23] where TMTTF is the sulfur analog of TMTSF and X is a monoanion such as ClO_4^-, BF_4^- or SCN^- etc..., see (Figure 1). All these compounds turns into strong insulators at low temperature. It is the reason why they did not attract much interest until recently when they have been practically rediscovered after twenty years of studies which had been mainly devoted to the selenide series, $(TMTSF)_2X$.

2. THE TM_2X PERIOD

Several exciting properties of $(TMTSF)_2PF_6$, an increase of the metal-like conductivity up to $10^5(\Omega\,\mathrm{cm})^{-1}$ at 12 K where an abrupt metal-insulator transition of SDW nature takes place [24], the absence of any noticeable lattice modulation in X-ray measurements [19,25], stimulated further

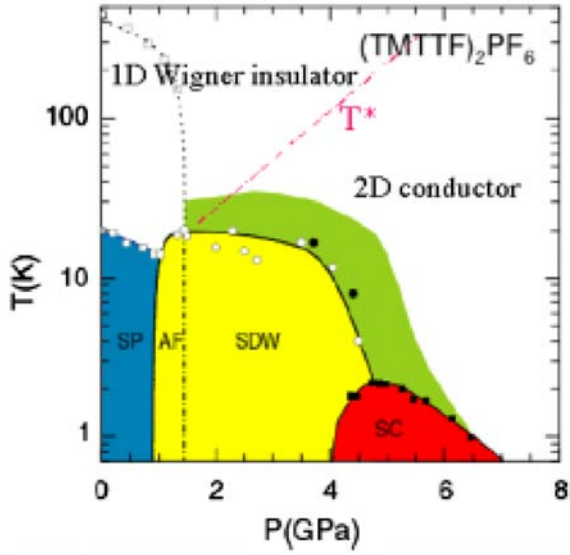

FIGURE 2 The generic phase diagram of the $(TM)_2X$ based on $(TMTTF)_2PF_6$ under pressure after [30]. The normal phase of sulfur compounds can be described as a Luttinger liquid that becomes gapped in the charge sector because of the Wigner localization below T_ρ and can develop either a spin-Peierls (SP) or localized antiferro-magnetic ordered state (AF). Under Pressure, the properties of the sulfur series evolve toward those of the selenides for which the normal state shows a progressive restoration of a Fermi liquid below the cross-over temperature $T^\star$ precursor to the spin density wave state (SDW) and superconductivity (SC). The shaded region outside SDW and SC ground states is dominated by strong AF fluctuations. (See Color Plate II)

investigations under pressure and allowed the stabilization of a metallic state down to liquid helium temperature at a pressure of about 9 kbar.

The finding of a very small and still non-saturating resistivity at 1.3 K extrapolating linearly to a practically zero value at $T = 0$ K was a sufficient motivation to trigger its study in a dilution refrigerator and a zero resistance state was stabilized below 1 K [5]. Shortly after the discovery of superconductivity in $(TMTSF)_2PF_6$ many other members of the same series with a variety of anions have also been found superconducting in the vicinity of 1 K in the 10 kbar pressure domain [26]. $(TMTSF)_2ClO_4$ is the only member of the $(TMTSF)_2X$ series to show superconductivity under atmospheric pressure [27]. The discovery of superconductivity in the $(TMTSF)_2X$ family was a very exciting phenomenon since that was the first time such an instability could be stabilized in an organic compound. This happened about 15 years after the publication of Little's

suggestion and 10 years after the holding of an international symposium organized by W.A. Little at Hawaii on the Physical and Chemical Problems of Possible Organic Superconductors [28]. Probably the most surprizing feature of superconductivity in $(TMTSF)_2X$ is the common border existing between antiferromagnetic (SDW) and superconducting phases with region of maximum T_c for superconductivity located right at the border.

It is this interesting behavior which initiated a reinvestigation of the sulfur-analog series, $(TMTTF)_2X$, with the hope to stabilize superconductivity in this series as well. This has been successfully achieved only recently with $(TMTTF)_2Br$ under 26 kbar [29] and $(TMTTF)_2PF_6$ at 47 kbar [30,31]. $(TMTTF)_2PF_6$ is the only member of the TM_2X series to exhibit all three different ground states allowing therefore to render the phase diagram a genuine property of these Q-1-D conducting salts.

It is clear that we have now reached a certain level of understanding in the physical properties of these 1-D conductors namely, $(TMTTF)_2X$ and $(TMTSF)_2X$ salts which both belong to the same $(TM)_2X$ family [32], (Figure 2).

The salient feature of the $(TM)_2X$ diagram is the existence of a wide variety of ground states occurring below 20 K or so. Moving towards the right across the $(TM)_2X$ phase diagram a succession of ground states is revealed either changing compounds or changing pressure on a given compound. The compound at the extreme left in the $(TM)_2X$ diagram, $(TMTTF)_2PF_6$ is the only one which can be taken through the entire series of ground states under high pressure, Spin-Peierls, Néel antiferromagnetism, SDW phase with an incommensurate magnetic modulation and ultimately superconductivity [33].

As far as the non ordered high temperature phase is concerned, its character in the phase diagram depends crucially on the nature of the corresponding ground state. Crudely speaking, all compounds left of $(TMTTF)_2Br$ i.e., 1.2 GPa in (Figure 2) are insulators whereas those on the right are highly conducting and display a metal-like conductivity down to the transition into the SDW insulating phase.

It is also most illuminating to have a look at the conductivity in the far infrared regime. A large gap of order 2000 K is observed in the frequency dependence of the FIR conductivity of sulfur compounds. This is in line with the activation energy of the DC conductivity for those compounds. However, the surprise arose for selenium compounds which behave apparently as normal metals in spite of a marked gap observed in the FIR regime. This feature suggests that all $(TM)_2X$ compounds exhibit a FIR gap, the amplitude of which depending on their actual location in the phase diagram. Given the unified phase diagram established experimentally and the recent proposal of theoretical ideas in 1-D physics applying specifically

to 1-D conductors with a commensurate band filing we have reached a plausible theoretical description which we intend to summarize very briefly in the rest of this article [34].

What 1-D physics means is that instead of the usual description of low lying excitations in terms of quasi particle states in the Landau–Fermi liquid model, a collective mode description with decoupled spin and charge modes is a more appropriate starting point [35,36]. Such a model for 1D conductors has been proposed starting from a linearized energy spectrum for excitations close to the Fermi level and adding the relevant Coulomb repulsions which are responsible for electron scattering with momentum transfer $2k_F$ and 0. This is the popular Luttinger model for a 1-D conductor in which the spatial variation of all correlation functions exhibit a power low decay at large distance, characterized by a non-universal exponent K_ρ (which is a function of the microscopic coupling constants) [37]. However, (TM)$_2$X conductors are rather peculiar systems since the stoichiometry imposes half a carrier (hole) per TM molecule independent of the applied pressure. If the molecules are uniformly spaced along the stacking axis (a situation which is actually met in (TMTSF)$_2$ClO$_4$), the unit cell contains 1/2 carrier, i.e., the conduction band is quarter-filled although the existence of a slight dimerization of the molecules along the stacks in sulfur compounds could contribute to make them half-filled band compounds (*vide infra*). The commensurate band filling opens a new scattering channel (Umklapp scattering) of the carriers between both sides of the Fermi surface. This leads to important modifications in the model of the gapless Luttinger liquid which in turn becomes a Wigner type insulator with a gap in the charge sector [38] subjected to the condition $K_\rho < 0.25$ (for 1/4 filling) but still no gap in the spin channel. The gap in the density of states at the Fermi energy reads: $2\Delta_\rho \approx W(\frac{gU}{W})^{1/(2-2n^2K_\rho)}$ and gU is the coupling constant $W(\frac{U}{W})^3$ where $n=2$ in the Hubbard limit.

According to band calculations, the transverse overlap $t_\perp$ along the b-direction is of order 120 K and 200 K for sulfur and selenium compounds respectively [34]. Therefore, it is quite natural to expect first 1-D theory to govern the physics of these quasi 1-D conductors, at least in the high temperature regime when $T > t_\perp$ and second, to observe a cross-over towards higher dimensionality physics below room temperature. As observed very early in the study of (TMTSF)$_2$PF$_6$ a plasma edge for a light polarized along the b-axis becomes observable below 100 K [39].

The striking behavior of the transport properties emerges from a comparison between the temperature dependence of longitudinal and transverse components of the resistivity [40]. The longitudinal resistivity of (TMTSF)$_2$PF$_6$ is metal-like down to the SDW transition at 12 K, varying like $T^{0.93}$ from 300 to 150 K once the constant volume correction is taken into account and more like T^2 below 150 K. However, the behavior of the

resistance along the direction of weakest coupling i.e., along the c-axis displays an insulating character with a maximum around 120 K and becomes metallic below although still orders of magnitude above the Mott critical value. Similarly, the Hall constant displays a marked temperature dependence below room temperature, passing through a minimum at the same temperature $T^\star$ where the c-axis resistivity displays a maximum [41].

The longitudinal transport is expected to vary according to a power law $\rho_{\|} \approx T^{4n^2K_\rho - 3}$ at $T > \Delta_\rho$ [38], whereas the transport along c being incoherent should probe the density of quasiparticle states in the (a-b) planes. In the high T regime, the picture of non-coupled chains is approached. Therefore, the density of quasiparticle states should resemble the situation which is expected to prevail in a Luttinger liquid namely, $N(E) \approx |\omega|^\alpha$ where α is related to K_ρ by $\alpha = \frac{1}{4}(K_\rho + 1/K_\rho - 2)$. At energies $\omega > \Delta_\rho$, the actual density of states of the Wigner state should be reminiscent of the density of states of the gapless Luttinger liquid. Therefore, a temperature dependence of the transport $\rho_c(T)T^{-2\alpha}$ is expected in the 1-D regime ($T > T^\star$). The far infrared (FIR) conduction of $(TMTSF)_2PF_6$ exhibits also very unusual properties. There exists as FIR gap of about $\Delta_\rho = 200\,\mathrm{cm}^{-1}$ in $(TMTSF)_2PF_6$ which has been taken as the signature of the Wigner gap [42]. The experimental data and the theoretical power law dependence $\sigma(\omega) \approx \omega^{4n^2K_\rho - 5}$ at $\omega > 2\Delta_\rho$ also lead to $K_\rho = 0.23$ [42]. This value for K_ρ would fit the 1/4 filled scenario with W = 12000 K (from band structure calculations and plasma edge measurements), $\Delta_\rho = 200\,\mathrm{cm}^{-1}$ (from FIR data) and U/W = 0.7 (which is in fair agreement with the enhancement of the spin susceptibility at low temperature) [34]. Since the Wigner gap varies exponentially with K_ρ even a small variation of the ratio between the Coulomb interaction and the bandwidth under pressure can explain a decrease of K_ρ moving from the left to the right in the phase diagram. Both optical and transport data give $2\Delta_\rho = 1000\,\mathrm{K}$ in $(TMTTF)_2PF_6$ at ambient pressure [43]. The difference between K_ρ for selenium and sulfur compounds in which $K_\rho = 0.18$ can be afforded by the difference of their bare bandwidths. The temperature $T^\star$ corresponding to the c-resistance maximum moves up under pressure and reaches room temperature under 10 kbar. This temperature can be attributed to the beginning of a cross-over between 1-D and 2-D regimes.

The strong pressure dependence of the cross-over temperature $T^\star$ is a remarkable phenomenon of the TM_2X physics. According to the pressure data of ρ_c, the pressure dependence of $T^\star$ is about ten times larger than that of $t_\perp$ which is typically $2\%\,\mathrm{kbar}^{-1}$. This feature suggests that $T^\star$ is actually a renormalized version of $t_\perp$ due to the 1-D confinement via intrastacks electron-hole interactions [44].

3. DISCUSSION

In the early days of this field it was believed that the stack dimerization was an important concept to understand the properties of the TM_2X salts. The recent transport and optical studies tend to imply that it may be not so. Apparently, the finite dimerization of the intrastack bonds is not large enough to make the 1/2-filled Umklapp scattering a pertinent mechanism in the formation of the 1-D Wigner phase. As far as the selenium compounds are concerned this feature could be related to the transverse b-direction bandwidth ($W_b \approx 1500$ K) being larger than the dimerization gap (900 K) and thus smearing out the role of the 1-D dimerization. This argument may still be valid for the sulfur series in which the dimerization gap and the transverse bandwidth are both in the same energy range.

The picture emerging for the TM_2X phase diagram is that of a strong coupling Wigner insulating phase with a correlation gap decreasing from sulfur to selenium compounds, (Figure 2). As long as the Wigner gap is large (say, larger than 300 K) the 1-D confinement is quite active and the single particle interstack hopping is meaningless despite an unrenormalized value of the transverse coupling which is still of the order of 100 K. Moving towards the right in the phase diagram, the Wigner gap decreases (because of the band broadening) and the renormalization of the transverse hopping integral becomes less pronounced. $(TMTSF)_2PF_6$ is a compound where T^* and $2\Delta_\rho$ are both of the same order of magnitude ($\approx$150–200 K), (Figure 2). The Fermi liquid picture is recovered eventually at the bottom right of the diagram as supported by the success of the weak coupling Fermi liquid theory explaining the formation of field induced spin density wave phases under large magnetic field [45].

4. CONCLUSION

In conclusion, the discovery of superconductivity in $(TMTSF)_2PF_6$ twenty years ago has had an impact which went far beyond the finding of one additional compound in the already long list of existing superconducting materials. This series of compounds has played the role of prototype for Q-1-D conductors. Evidences of new concepts in physics have been obtained. Its experimental investigation has led over the last 20 years to the discovery of a wealth of new phenomena in the physics of low dimensional conductors such as the role of commensurate band filling in promoting a strong coupling Wigner-Mott-Hubbard insulating phase, the cross-over between the Wigner-Luttinger limit and the Fermi liquid theory at low temperature, the interplay between a magnetic field and the quasi-one dimensionality and finally the possibility for a new exotic mechanism for

superconductivity, both topics which I could not approach in such a short article, see [34,46] for further reading.

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