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Transverse transport in $(TM)_2X$ organic conductors: possible evidence for a Luttinger liquid

J. Moser[1,a], M. Gabay[1], P. Auban-Senzier[1], D. Jérome[1], K. Bechgaard[2], and J.M. Fabre[3]

[1] Laboratoire de Physique des Solides[b], Université Paris-Sud, 91405 Orsay Cedex, France
[2] Department of Solid State Physics, Risø National Laboratory, 4000 Roskilde, Denmark
[3] Laboratoire de Chimie Structurale Organique, Université des Sciences et Techniques du Languedoc, place E. Bataillon, 34060 Montpellier, France

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Abstract. We present *c*-axis resistivity measurements performed on the organic conductors $(TMTSF)_2PF_6$ and $(TMTTF)_2PF_6$ under pressure. The aim is to probe the density of states of quasi-one dimensional compounds the high temperature properties of which are those of a Luttinger liquid. It is found that the 1-D Luttinger description breaks down below a specific pressure-dependent temperature, giving rise to a transient regime. The Fermi liquid behaviour is however restored at low temperature *i.e.* around 10 K, as evidenced by NMR measurements. Accordingly, two different energy scales 100 K and 10 K are required to get a fair understanding of all observed physical phenomena. Our interpretation supports the picture of a power law exponent K_ρ for the correlation functions of the order of 0.25-0.30.

PACS. 67.55.Hc Transport properties – 71.10.Pm Fermions in reduced dimensions (anyons, composite fermions, Luttinger liquid, etc.) – 74.20.Mn Nonconventional mechanisms (spin fluctuations, polarons and bipolarons, resonating valence bond model, anyon mechanism, marginal Fermi liquid, luttinger liquid, etc.)

1 Introduction

The essence of Fermi liquid (FL) theory is the description of the low energy elementary excitations of interacting electrons in terms of an ensemble of free "quasiparticles" behaving as particles dressed by interactions-related high-energy effects [1]. This picture has been remarkably successful, explaining for example, itinerant magnetism, the physics of heavy fermion compounds and providing the starting point for the classical Bardeen Cooper Schrieffer (BCS) model of superconductivity. However, theoretical limits are known for the applicability of the FL theory. This is the situation encountered in the strong coupling limit (the Mott transition) and also the case of electrons confined in one dimension. In this latter situation, the failure of the model concerns the essential requirement that in a FL the width of the quasiparticle states should be small as compared to their energy. This is no longer fulfilled in dimensionality one. Instead, low energy excitations in 1-D metals are best described by collective excitations which separate into spin and charge modes each with different (interaction dependent) velocities [2–6]. The main features of the 1-D electron gas (also called the Luttinger liquid) are (i) a decoupling between spin and charge degrees of freedom, (ii) the absence of any discontinuity in the distribution function for electron states at the Fermi energy and (iii) a power law decay at long distance of the spin or charge correlations functions which leads to the absence of long range order in a 1-D system.

Confronted with the unusual magnetic, optical and transport properties displayed by high T_c cuprates, several theoreticians have claimed that a non-FL description was also necessary to interpret the physical properties of these new low dimensional conductors [7]. The issue of whether one may extend the 1-D Luttinger liquid approach to the two-dimensional case for the cuprates (as proposed by Anderson [8]) is still hotly debated. In particular, a major theoretical problem deals with the stability of the Luttinger liquid when interchain coupling is gradually switched on.

The purpose of the present article is to contribute to this controversial debate through the study of the DC transport properties displayed by the one-dimensional organic conductors family which has led to the discovery of organic superconductivity, namely the $(TM)_2X$ series where TM stands for TMTSF (tetramethyltetraselenafulvalene, selenide compounds) or TMTTF (tetramethyltetrathiafulvalene, sulfur compounds).

The one-dimensional character of the electronic structure of TM_2X materials is based on the anisotropy of the tight-binding overlap integrals along the three directions, *i.e.* 1/0.1/0.005 for the high (*a*), intermediate (*b*) and

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[a] e-mail: `moser@lps.u-psud.fr`

[b] URA 02 CNRS

weak (c) coupling directions respectively. Orders of magnitude for sulfur and selenide compounds in the TM$_2$X series are $t_a \sim 0.15$ eV and 0.3 eV respectively [9]. The coupling along the c-direction is not known with much accuracy but crudely speaking it does not reveal any significant difference between sulfur and selenide compounds. A typical calculated value of $t_c = 1$ meV suggests that it can be ignored when dealing with the high or intermediate temperature regime of most compounds [10]. Consequently, the 2-D approximation is presumably no longer valid at very low temperature, *i.e.* at $T < 5$ K or so. However, the situation is dramatically different for the b-coupling. The value of the bare coupling $t_b \sim 20$ meV in selenide compounds makes this coupling quite pertinent in the temperature domain 300-100 K [10,11]. The main question raised by such an amplitude for the intermediate coupling is whether Q-1-D conductors can still exhibit properties which are reminiscent of the Luttinger liquid behaviour. There is an on-going controversy on this issue [12,13].

However, the existence of non-FL features is not disputed in the sulfur compounds. (TMTTF)$_2$X materials behave very much like canonical 1-D conductors. The charge becomes localized below 250 and 100 K in (TMTTF)$_2$ PF$_6$ and Br respectively due to Umklapp scattering of electrons in a commensurate 1-D lattice [14] while the static susceptibility remains unaffected by this localization (spin-charge decoupling) showing only a slight temperature dependence and stays finite at low temperature (above the onset of spin-Peierls or spin density wave long range order) [9,15]. Furthermore, the temperature dependence of the hyperfine-induced nuclear spin relaxation has been found to follow very closely the predictions made for a Luttinger liquid [16], namely

$$T_1^{-1} = C_0 T \chi_{\mathrm{S}}^2(T) + C_1 T^{K_\rho} \tag{1}$$

where $\chi_{\mathrm{S}}(T)$ is the (temperature dependent) static susceptibility or Knight shift, and K_ρ is the exponent characterizing the decay of correlations at long distance. This exponent is also related to the Fermi surface exponent α giving the power law for n_k at k_{F} [2],

$$\alpha = \frac{1}{4}(K_\rho + 1/K_\rho - 2) \tag{2}$$

which also governs the spectral density in the vicinity of the Fermi level in photoemission experiments [17].

Experimental results for (TMTTF)$_2$PF$_6$ interpreted within the 1-D picture lead to $K_\rho = 0$ (the strong coupling limit) in good agreement with the behaviour expected for a commensurate (undoped) 1-D Mott insulator [18]. It seems that the bare t_b coupling, though still of order 100 K in sulfur compounds [11] does not interfere with a purely 1-D behaviour. As long as the correlation gap Δ_ρ related to the charge localization is larger than the bare t_b intrachain electron-hole attraction leads to a 1-D electron confinement with a concomitant suppression of the effective transverse coupling [19] and makes interchain single particle hopping irrelevant in (TMTTF)$_2$PF$_6$. The situation for selenide compounds is more delicate in many respects as the temperature dependence of T_1^{-1} clearly reveals a broad temperature regime above the SDW ordering at 12 K in (TMTSF)$_2$PF$_6$ or above 8 K in (TMTSF)$_2$ClO$_4$, where the relaxation rate deviates from the prediction of the Fermi Liquid picture ($1/T_1 \propto T$), since both $1/T_1$ and the spin susceptibility are found to be T independent in the same temperature regime [18].

Deviations from Korringa law have been identified in selenide compounds (up to 100 K or so) and a value $K_\rho \sim 0.25$ has been derived using the 1-D formalism for NMR [20]. Furthermore, photoemission experiments performed at 50 K [17] do not show the existence of a finite spectral weight at the Fermi level. These data may rule out the possibility of a standard Fermi model in this temperature domain.

The absence of a Fermi edge in photoemission data pertaining to (TMTSF)$_2$PF$_6$ at 50 K [17] is in fair agreement with assigning a value $K_\rho \sim 0.25$ from NMR relaxation. However, the temperature dependence of the (TMTSF)$_2$PF$_6$ resistivity along the most conducting direction reveals an ambient pressure metallic-like behaviour with $\rho_a(T) \sim T^{1.8}$ between 300 and 100 K becoming approximately $\rho_a(T) \sim T^2$ below and down to the very abrupt metal-SDW transition at 11.5 K [15]. This behaviour is at first glance in serious contradiction with the temperature dependence of the resistivity which is expected from Umklapp scattering theory in a 1-D half filled band conductor, namely $\rho_a(T) \sim T^{4K_\rho - 3}$, *i.e.* T^{-2} [21], given $K_\rho \sim 0.25$ from NMR data. Such a paradoxical situation has stimulated new studies, in particular experiments which could provide a closer access to the spectral density in the vicinity of the Fermi level.

Stimulated by the claim that the temperature dependence of the conductivity for the direction of weakest coupling (σ_c) supports the picture of a 2-D non Fermi liquid behaviour for cuprates [22], an extensive study of the T and P dependence of ρ_c in (TMTSF)$_2$PF$_6$ an (TMTTF)$_2$PF$_6$ has been undertaken. A preliminary account of these results together with an improved version of the (TM)$_2$X generic diagram are presented in this article.

Early reflectance studies on (TMTSF)$_2$PF$_6$ have concluded to the existence of a well defined reflectance edge at room temperature for light polarized parallel to the a direction [23,24]. As far as the b' direction is concerned only an overdamped plasmon behaviour is observed at 300 K with no well characterized reflectance edge, and it is only below 100 K or so that such an edge feature can be observed and has been ascribed to a dimensionality cross-over along the b-direction [23,24]. Much less work has been devoted to the c direction, but the absence of any reflectance in the far IR regime for (TMTSF)$_2$AsF$_6$ at $T = 30$ K has led Jacobsen *et al.* [25] to the conclusion that no coherent transport can be established in this compound down to 30 K (at least). The study of static transport ρ_c had also been the matter of some work in the early days of (TMTSF)$_2$X materials. Jacobsen *et al.* [23,24] were the first to report a non-monotonic temperature dependence of ρ_c in (TMTSF)$_2$PF$_6$ at ambient

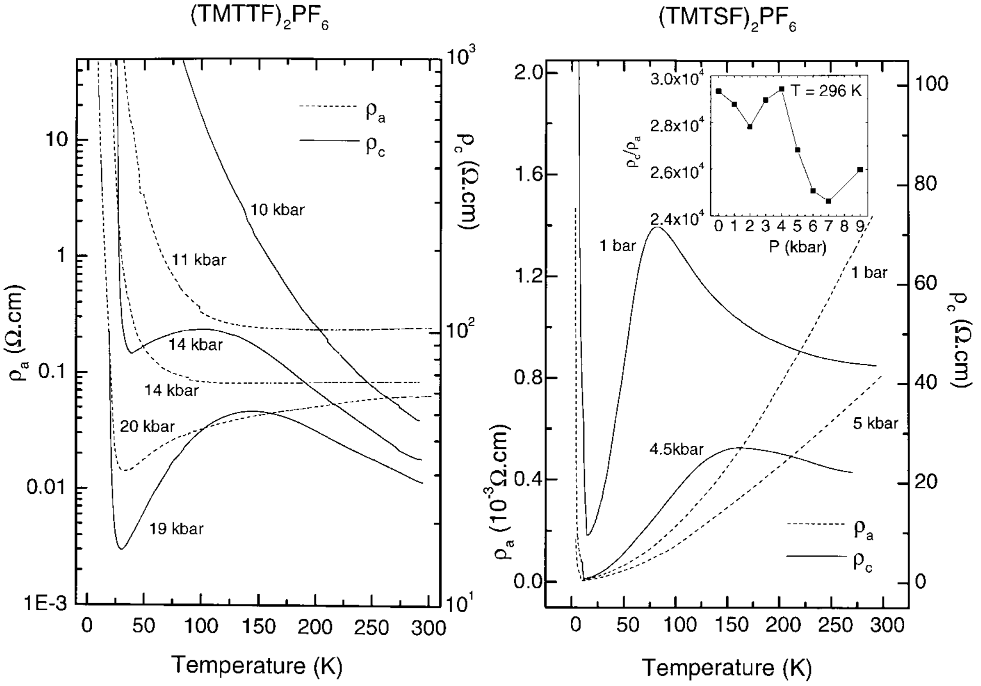

Fig. 1. Typical sets of c-axis and a-axis resistivity curves performed on both $(TMTTF)_2PF_6$ (left panel) and $(TMTSF)_2PF_6$ (right panel) under pressure. Note the well defined ρ_c maximum which shifts toward high temperatures as pressure is further increased. Inset of right panel depicts the pressure dependence of ρ_c/ρ_a anisotropy in $(TMTSF)_2PF_6$ at room temperature.

pressure going through a well characterized maximum at 80 K at variance with the results for ρ_a and ρ_b exhibiting a monotonous metallic-like decrease upon lowering temperature. The c-axis resistivity has also been extensively studied by the Zagreb group [26,27] corroborating Jacobsen's data [23,24] and showing that the c-axis component leads to remarkably reliable measurements.

2 Experimental results

Throughout the study set out below, we used representative $(TMTTF)_2PF_6$ and $(TMTSF)_2PF_6$ single crystals which we cleanly cut off with a sharp razor blade along the a-axis to get 2 mm-long pieces. Their typical size (length ×width × thickness) was about $2000 \times 500 \times 100$ μm^3. Resistivity measurements were performed through a four-probe technique to eliminate contact resistances. In order to follow the anisotropy variation with temperature we measured resistivity along both a and c axis. For the latter, two gold pads were evaporated onto both *(ab)* faces of each sample. In this configuration any contact on one face and the contact on the other one are rigorously opposite to each other. With regard to ρ_a, the two inner pads were used as voltage contacts, and the two outer ones covered both ends to ensure an homogeneous current flow. In both setups, 17.5 μm-diameter gold wires were pasted on each pad with a small amount of silver paint. We mounted two samples on a 5 mm-diameter support that we covered with a teflon cap filled up with silicon oil used as a pressure medium. Measurements were performed using an AC current with 10 μA of peak amplitude. Voltages were measured by two lock-in amplifiers. The samples were slowly cooled down (0.1-0.2 K/min) to avoid cracks and ensure a good thermalization with temperature sensors. Care was taken to ensure that the results are sample-independent, in a sense that two samples of different size and shape measured simultaneously always showed similar ρ_c behaviour with a maximum occurring at the same temperature for a given pressure.

Typical sets of ρ_c *versus* T are displayed in Figure 1 at several pressures up to 9 kbar. These data invariably show a well defined maximum at $T_{\max}$ moving towards higher temperatures as pressure is increased. $T_{\max}$ is found to be particularly pressure sensitive since it increases at a rate of 25% kbar^{-1}. The ρ_c maximum tends to become shallow at 9 kbar and the T dependence approaches the behavior of ρ_c as measured in $(TMTSF)_2ClO_4$ [28]. As shown in the inset of Figure 1, the pressure dependence is about the same for ρ_c and ρ_a with $\partial \ln \sigma/\partial P$ ~22% kbar^{-1}. The existence of a ρ_c maximum is also proved to exist in $(TMTTF)_2PF_6$ when submitted to a pressure of 19 kbar (see Fig. 1). However, releasing pressure from 19 down to 10 kbar, the resistivity maximum smoothly merges into a monotonically activated resistivity and $T_{\max}$ can no longer be defined below 13 kbar.

3 Interpretation

The first point we wish to emphasize is the incoherent character taken on by the transport in $(TMTSF)_2PF_6$ along the c-direction in the whole temperature regime down to ~ 20 K. This argument is supported not only by the absence of any reflectance edge for the light polarized along c [25] but also by the failure of the classical band model to describe the ρ_c magnetoresistance of $(TMTSF)_2PF_6$. Cooper *et al.* [27] have pointed out that the band-like interpretation of the observed Kohler's behaviour of magnetoresistance ($\Delta\rho_c/\rho_{c_0} \propto H^2/\rho_{c_0}^2$) leads

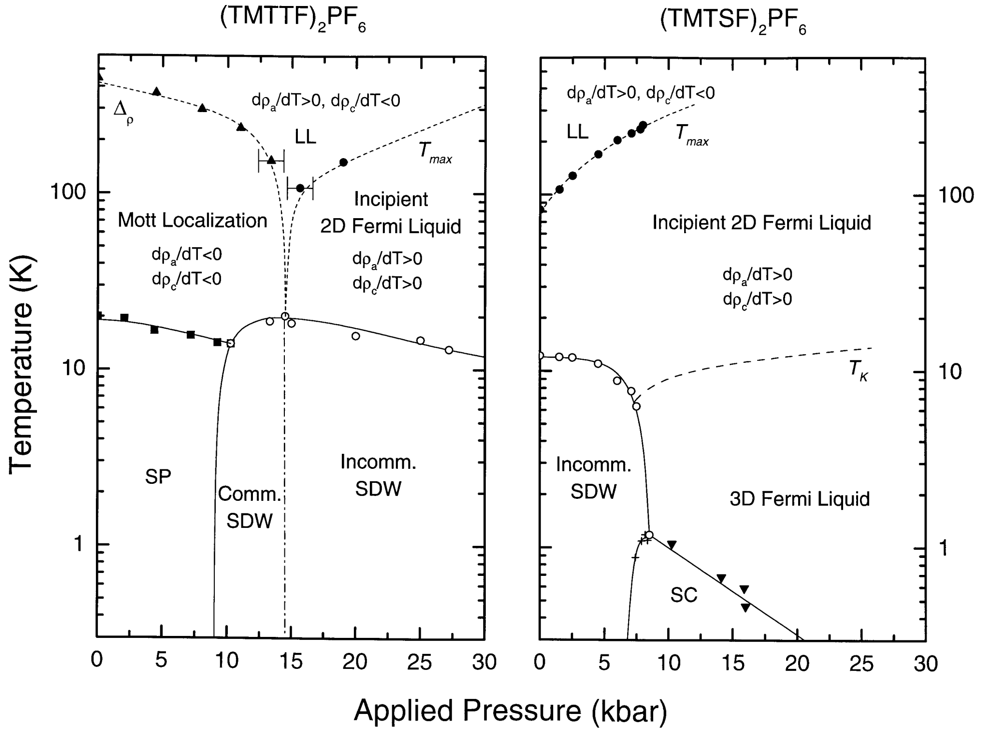

Fig. 2. Phase diagram (T-P) of (TMTTF)$_2$PF$_6$ (left panel) and (TMTSF)$_2$PF$_6$ (right panel). Superimposed on the pre-existing phase diagram for quasi 1-D conductors is displayed the $T_{\rm max}$ *versus* P line (full circles) setting a border between a Luttinger regime (LL) and a transient phase towards a Fermi liquid. Different compressibility and renormalization effects account for the apparent $T_{\rm max}$ line mismatch between the two organic compounds. Below the $T_{\rm K}$ line a 3D Fermi liquid is fully restored. The Mott localization line is also depicted (full triangle) and new SDW transition points have been plotted as additional results of ρ_c measurements (hollow circles). The superconducting (SC) re-entrance (crosses) is in fact that of (TMTSF)$_2$AsF$_6$ (from R. Brusetti *et al.*, J. Phys. France **43**, 801 (1982)). SP stands for Spin Peierls. Commensurate (Comm) and incommensurate (Incomm) SDW states have been singled out.

to $t_c \sim$ 0.10-0.16 meV in (TMTSF)$_2$PF$_6$. This value is about 6 times smaller than what was predicted from the extended Hückel model, namely $t_c \sim 1$ meV [10]. Referring to the structure of (TM)$_2$X salts the heteroatoms of which do not contribute to the interchain coupling along the c direction, a wide spread of t_c amplitudes over various members of the series is unlikely. This claim is corroborated by recent extended Hückel calculations leading to similar values of t_c, namely $\sim$ 0.65 meV in both (TMTSF)$_2$PF$_6$ and (TMTTF)$_2$Br [29]. The mean free path along c when evaluated from magnetoresistance data within a band model leads to the unrealistic value of $l_c \sim c/40$ at 21 K in (TMTSF)$_2$PF$_6$ [27]. However, the situation is apparently different for (TMTSF)$_2$ClO$_4$ since the regime $l_c > c$ is achieved at helium temperature (with $l_c \sim 2.17c$ at 4.5 K [28]).

Furthermore, given the value $t_c \sim 0.75$ meV, the warping of the Fermi surface along c is not pertinent since $\hbar/\tau > t_c$ above 20 K in (TMTSF)$_2$PF$_6$. At ambient pressure, the condition $\hbar/\tau \sim t_c$ is fulfilled at $T = 20$ K only. We may thus conclude that c-axis transport in (TMTSF)$_2$PF$_6$ remains incoherent down to the SDW phase transition temperature in spite of the metal-like temperature dependence of the c-axis resistance displayed between 80 and 12 K. It has already been stressed in the context of the conductivity in Luttinger liquids that a positive temperature dependence of the resistivity does not necessary imply metallic conductivity [30]. Cooper *et al.* [27] have ascribed the deviation to Kohler's law observed above 80 K to an increase of t_c as T increases. This assumption is somewhat speculative as the study of the (TMTSF)$_2$PF$_6$ lattice parameters has failed to show any significant anomalous temperature dependence in the vicinity of 80 K [31,32]. In addition, thermal expansion would lead to a decrease of t_c rather than an increase at increasing temperature.

Even though we do not know the exact significance of $T_{\rm max}$ yet, it sets the cross-over between two different temperature regimes for $\rho_c(T)$. Since a similar behaviour is observed for (TMTTF)$_2$PF$_6$ under pressure we may emphasize that the cross-over at $T_{\rm max}(P)$ is indeed a genuine property of the (TM)$_2$X diagram, see Figure 2. Following the onset below 100 K of a reflectance edge for the light polarized along the b-axis, one may argue that $T_{\rm max}$ could mark the beginning of a dimensional cross-over regime along this direction. However, the very strong pressure dependence of $T_{\rm max}$ rules out a simple relation such as $T_{\rm max} \sim t_b$ since the pressure coefficient of the bare transverse coupling should amount to that of t_a, *i.e.* $\sim$2% kbar^{-1} according to the independence of ρ_a/ρ_b

anisotropy under pressure [33]. Consequently, the strong pressure dependence of $T_{\max}$ suggests that the onset of the cross-over regime is related to a renormalized value.

As far as c-axis tunneling is concerned, the situation above $T_{\max}$ can be described in terms of carriers tunneling incoherently between Luttinger chains. The problem has not yet received a detailed theoretical treatment but it has already been considered in the context of the transverse (c-axis) conductivity between adjacent Luttinger liquids in underdoped cuprate superconductors [34]. Using the Fermi Golden rule for the hopping rate of carriers between two Luttinger states, σ_c follows the power law $\sigma_c \propto (\omega,\ k_{\mathrm{B}}T)^{2\alpha}$ where α is the exponent of the density of states, see equation (2). Since α is expected to be positive and smaller than unity in the Luttinger liquid [2] a negative temperature dependence can be expected for $\rho_c(T)$. A more rigorous derivation will be given in the next section.

4 Theory

In this section we derive an approximate expression for the c-axis conductivity σ_c, for temperatures T such that

$$t_c \ll t_b < T \ll t_a. \tag{3}$$

In this regime, the thermal wavelength of the electron is much less than the interchain distances b and c in directions denoted by y and z respectively. Transport along y and z is totally incoherent and amounts to the tunneling of real electrons in and out of 1D Luttinger chains. As a result, to lowest order in perturbation, thermal averages will be computed over Hamiltonian H_0 which is a sum of uncoupled 1D Luttinger liquids along x. In effect we are setting

$$t_b = t_c = 0 \tag{4}$$

in H_0. Current along z will be a tunneling current and to compute it we may either use the Kubo formula or the tunneling approach followed by Clarke *et al.* [34] and Anderson *et al.* [35]. As we show below both methods give similar results.

Starting with the Kubo formula, we compute $\sigma_c(\omega)$; the T-dependence of the dc-conductivity is obtained by substituting T for ω.

The current operator in the z direction is

$$\mathrm{j}_z = -i\frac{t_c ec}{\hbar}\sum_{ix\sigma}\mathrm{C}^{+}_{i+1,\sigma}(x)\mathrm{C}_{i,\sigma}(x) + H.c \tag{5}$$

where i is the chain index along z and j_z is the current flowing through the (x, y) plane. Computing the current-current correlator $\langle[\mathrm{j}_z(\mathbf{r}, t), \mathrm{j}_z(\mathbf{r}_1, t_1)]\rangle_{\mathrm{H}_0}$ allows to extract the real part of the conductivity [36] and we find

$$\sigma_c(\omega) = \frac{8e^2}{\hbar}c^2t_c^2\int\limits_{-\pi/c}^{\pi/c}\frac{\mathrm{d}k_z}{2\pi}\sin^2 k_z c$$
$$\times\int\mathrm{d}x'\int\frac{\mathrm{d}\varepsilon}{2\pi}\frac{f(\varepsilon)-f(\varepsilon+\hbar\omega)}{\hbar\omega}A_{\mathrm{e}}(x',\varepsilon)A_{\mathrm{h}}(x',\varepsilon+\hbar\omega),$$

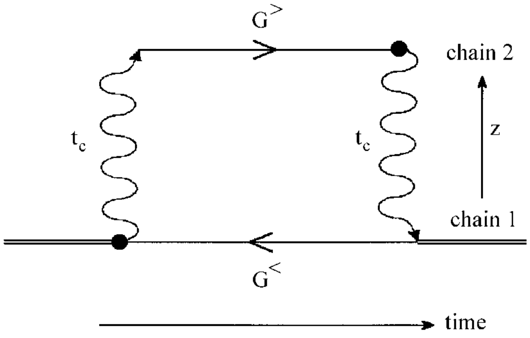

Fig. 3. Transverse electron tunneling scheme between two Luttinger chains. G$^>$(G$^<$) is the Green's function for electrons (holes).

i.e.

$$\sigma_c(\omega) = \frac{4e^2ct_c^2}{\hbar}\int\mathrm{d}x'\int\frac{\mathrm{d}\varepsilon}{2\pi}\frac{f(\varepsilon)-f(\varepsilon+\hbar\omega)}{\hbar\omega}$$
$$\times A_{\mathrm{e}}(x',\varepsilon)A_{\mathrm{h}}(x',\varepsilon+\hbar\omega). \tag{6}$$

A_{e} (A_{h}) is the spectral function of the *real* electron (hole) in the Luttinger liquid, $f(\varepsilon) = \frac{1}{\mathrm{e}^{\beta\epsilon}+1}$ is the Fermi distribution function and $x' = x - x_1$ is the distance travelled by the electron (hole) between scattering events.

In the tunneling formalism approach, the tunneling Hamiltonian reads

$$t_c\sum_{ix\sigma}\mathrm{C}^{+}_{i+1,\sigma}(x)\mathrm{C}_{i,\sigma}(x) + H.c. \tag{7}$$

and leads to a tunneling current [36]

$$I = \frac{4ect_c^2}{\hbar}\int\mathrm{d}x'\int\frac{\mathrm{d}\varepsilon}{2\pi}\left(f(\varepsilon)-f(\varepsilon+\hbar\omega)\right)$$
$$\times A_{\mathrm{e}}(x',\varepsilon)A_{\mathrm{h}}(x',\varepsilon+\hbar\omega). \tag{8}$$

The conductance g in the z direction is then

$$g \sim \frac{4e^2ct_c^2}{\hbar}\int\mathrm{d}x'\int\frac{\mathrm{d}\varepsilon}{2\pi}\frac{f(\varepsilon)-f(\varepsilon+\hbar\omega)}{\hbar\omega}$$
$$\times A_{\mathrm{e}}(x',\varepsilon)A_{\mathrm{h}}(x',\varepsilon+\hbar\omega) \tag{9}$$

in agreement with (6) for σ_c.

The physical process associated with (6, 9) is depicted in Figure 3. An electron is scattered out of the Luttinger chain 1, propagates on chain 2 and is scattered back at a later time. So A_{e} refers to chain 2 while A_{h} concerns chain 1, and by definition (see (4)) subsystems 1 and 2 are decoupled in equilibrium.

The only difference between (6) and (9) and the standard expression for the tunneling current comes from the integral over x', which we get in our case. This integral expresses momentum conservation for the hole and the electron propagating in chains 1 and 2 respectively. In other words there is a coherent motion of electrons and holes in the two chains.

If subsystems 1 and 2 were ordinary metals, a natural cut-off for the integral over x' would be the mean free path

$l \sim v_{\mathrm{F}}\tau_a$. So we would expect (see for instance [36])

$$\sigma_c = N_1(0)N_2(0)\frac{\tau_a t_a}{\hbar} \tag{10}$$

where $N_1(0)$ is the density of states of subsystem 1 and $N_2(0)$ that of subsystem 2.

For "1D Fermi liquids" $\tau \sim 1/\omega$ or $\tau \sim 1/T$ (neglecting logarithmic corrections). If, instead of decoupled chains we had decoupled planes, and if these planes behaved as 2D Fermi liquids, we would find $\sigma_c \sim 1/T^2$. In the present situation however, electron (hole) states are not eigenstates of the chain; we should not expect to get a coherent motion of the electron and the hole if $x' \geq a$, since these entities decay into spin and charge collective excitations beyond that distance. Then

$$\sigma_c(\omega) \sim \frac{4e^2}{\hbar} act_c^2 \int \frac{\mathrm{d}\varepsilon}{2\pi} \frac{f(\varepsilon) - f(\varepsilon + \hbar\omega)}{\hbar\omega} \times A_{\mathrm{e}}(0,\varepsilon)A_{\mathrm{h}}(0,\varepsilon+\hbar\omega). \tag{11}$$

In the $(\mathrm{TMTSF})_2\mathrm{PF}_6$ compound, susceptibility data indicate that the Hubbard U is comparable to the bandwidth, so that the charge velocity of the chain v_c is much larger than the spin velocity. In the Luttinger regime, we then write,

$$A(x,t) = \frac{1}{t^{\alpha+1}} h\left(\frac{x}{v_c t}\right)$$

where h is a scaling function and leads to

$$A(x,\varepsilon) = \frac{1}{v_c}\left(\frac{\varepsilon}{\hbar v_c}\right)^{\alpha} \tilde{h}\left(\frac{\omega x}{v_c}\right)\cdot \tag{12}$$

Plugging (12) into (11) gives the expression for σ_c:

$$\sigma_c(\omega) \sim t_c^2 \frac{e^2}{\hbar}\frac{ac}{v_c^2}\left(\frac{\omega}{v_c}\right)^{2\alpha}\cdot \tag{13}$$

Accordingly, the dc-conductivity $\sigma_c(T)$ is such that

$$\sigma_c(T) \sim T^{2\alpha}\,. \tag{14}$$

In essence, this expression is the result of the Fermi Golden rule for a 1D system with a density of states $N(\varepsilon) \sim \varepsilon^{\alpha}$. σ_c directly probes the density of states of the Luttinger liquid.

When $t_c \ll \omega, T < t_b < t_a$, we may repeat the tunneling argument for planes, instead of chains. For an anisotropic system with mean free paths of order $\tau_a t_a/\hbar$ and $\tau_b t_b/\hbar$ along a and b respectively, the integral over x' yields a contribution of order $\sqrt{\tau_a t_a \tau_b t_b/\hbar^2}$, provided we may define quasiparticle states for the *(ab)* planes. In that limit, $\sigma_c \sim \sqrt{\sigma_a\sigma_b}$. Furthermore, if the planes behave like 2D Fermi liquids we set $\alpha = 0$ in (13). Since $\tau \sim 1/T^2$, $\sigma_c(T) \sim 1/T^2$ so that $\sigma_c(T)/\sigma_a(T)$ is a constant in that regime.

5 Discussion

Attempting to process experimental data in Figure 1 much attention should be paid to the large volume dependence of ρ_c at fixed temperature. Therefore, the ambient pressure T-dependence of ρ_c cannot be straightforwardly used to extract the exponent α (which is a constant-volume quantity). Instead, the conversion from constant pressure to constant volume must be performed before any comparison with theory is made. This problem has already been encountered in the study of charge transfer compounds [37,38]. We may briefly sketch the procedure here. Taking the unit cell at 50 K as the reference unit cell (this approximation being justified by the lack of thermal expansion at low temperature) and increasing temperature up to T, a pressure P must be applied at T in order to restore the reference volume, taking into account thermal expansion and compressibility data [31,39]. The results of this procedure are displayed in Figure 4 using the measured isobaric temperature dependences at 1.5, 2.5, 4.5 and 7.0 kbar. This procedure leads to the dots plotted in Figure 4 which can be interpolated by the dashed line in order to derive the T-dependence at the volume of 50 K and 1 bar. The high temperature regime suggests $\rho_c \sim T^{-1.4}$ *i.e.* $\alpha = 0.7$.

The situation is even more subtle below $T_{\max}$ as the motion remains incoherent along the c-axis but coherence begins to build up between chains in the (ab) planes. A somewhat similar situation has been encountered for the interpretation of transport properties in charge transfer salts. In the diffusive regime for c-axis transport in 1-D conductors the transverse conduction remains proportional to the longitudinal a-conductivity, namely $\sigma_c \propto \sigma_a$ [40,41]. The calculation in the previous section can be extended to the situation where the subsystem is a 2-D liquid although not necessarily Fermi-like with an anisotropic behaviour. As long as quasiparticle states can be defined with life times τ_a and τ_b along the directions a and b respectively, the resistivity along c should read

$$\rho_c(T) \propto \sqrt{\rho_a(T)\rho_b(T)}. \tag{15}$$

Provided the quasiparticle life-times along the a and b directions exhibit the same temperature dependences the ρ_c/ρ_a ratio turns out to be temperature independent. Such a behaviour is definitely not observed in the temperature domain of the incipient 2D Fermi liquid under ambient pressure and also under 9 kbar, see the inset of Figure 4. Since the laws $\rho_a \sim T^2$ and $\rho_c \sim T^{1.5}$ are followed under 1 bar and 9 kbar, we infer from equation (15) that ρ_b should linearly depend on temperature. This is in fairly good agreement with early data for ρ_b below 80 K [25]. We consider the temperature domain below $T_{\max}$ as a broad cross-over regime for the onset of coherence along the b-direction, with a concomitant evolution towards the establishment of a 2D Fermi liquid at low temperature. Hence, it seems to be clear that even if a typical Fermi-like behavior happened to be applicable to the a-direction it would not be the case for the b-direction.

Let us point out at this stage the important difference between the behaviour of $(\mathrm{TMTSF})_2\mathrm{PF}_6$ at ambient pressure on the one hand and that of $(\mathrm{TMTSF})_2\mathrm{PF}_6$ under

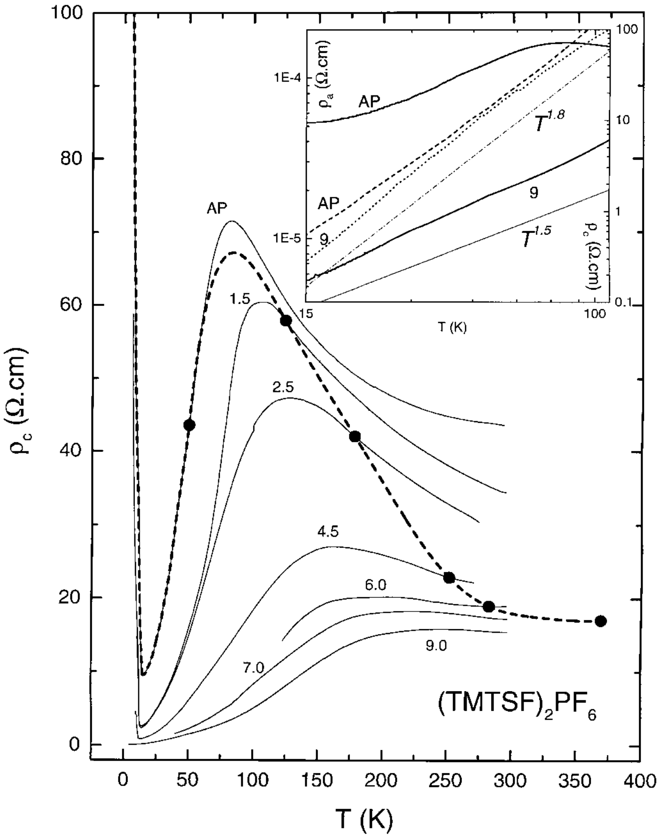

Fig. 4. c-axis resistivity of (TMTSF)$_2$PF$_6$ under various hydrostatic pressures (P in kbar). The bold, dotted curve depicts the ρ_c curve for a would-be (TMTSF)$_2$PF$_6$ compound whose lattice parameters would remain constant all the way from 50 K up to room temperature. The constant volume correction procedure is set out in the text. Inset shows in a log-log scale ρ_a (dashed and dotted lines) and ρ_c (solid lines) at ambient pressure (AP) and under 9 kbar. Obviously the anisotropy does depend on temperature in the transient regime, even under 9 kbar where the metallic phase is fully stabilized, this being strongly suggestive of a non-Fermi liquid behaviour.

pressure (9 kbar) or (TMTSF)$_2$ClO$_4$ on the other hand at $T = 20$ K; ρ_c differs by a factor 100 in the two situations. This feature can be attributed to the fact that an incoherent to coherent transition along c is induced under pressure in (TMTSF)$_2$PF$_6$, the signature of which being a tremendous suppression of ρ_c (at low temperature) under pressure, see Figure 1 (right panel). It is realistic to expect a 3-D coherent state to be achieved at low temperature for either Se$-$ClO$_4$ or Se$-$PF$_6$ under pressure, while the ambient pressure phase remains in a 2-D (possibly non Fermi) liquid state for Se$-$PF$_6$ down to the SDW phase transition. Consequently, the recovery of the T_1^{-1} Korringa behaviour (with a strongly renormalized density of spin fluctuations) at $T_K \sim 10$ K which is observed in Se$-$ClO$_4$ or in Se$-$PF$_6$ at 8 kbar (and is also very much pressure dependent) could be ascribed to the onset of 3-D coherence, namely $t_c \sim 8$ K [15].

The region above $T_{\max}$ in the generic (TM)$_2$X phase diagram (Fig. 2) represents isolated chains with a Luttinger liquid behaviour. The intermediate region in between $T_{\max}$ and T_{K} is that of an incipient 2-D Fermi liquid, whereas this is only below T_{K} that a full 3-D Fermi liquid behaviour is restored.

In the high temperature phase $\alpha = 0.7$ as derived from the T-dependence of ρ_c (at the fixed unit cell volume of 50 K) leads to $K_\rho = 0.22$ which is very close to the derivation from NMR data [18].

The value of K_ρ derived from ρ_c allows in turn a prediction for the constant volume T-dependence for ρ_a at $T > T_{\max}$. At high temperature the resistivity of a 1-D commensurate and interacting electron gas is governed by Umklapp scattering. Both half-filled and quarter-filled scattering can be active in the (TM)$_2$X series. When the lattice dimerization is strong *e.g.* in the sulfur series, half-filled U-scattering is expected to dominate and $\rho_a(T) \sim g_{1/2}^2 T^{4K_\rho - 3}$. However, the lattice dimerization is much weaker for selenium compounds (dimerization gap smaller than transverse coupling along b) but we are left with the remaining quarter-filled U-scattering. Thus, the resistivity reads [42], $\rho_a(T) \sim g_{1/4}^2 T^{16K_\rho - 3}$. Given $K_\rho \sim 0.20-0.25$ from ρ_c in the Luttinger regime the expected constant volume temperature dependence of ρ_a becomes $\rho_a \sim T^{-1}$ or $\rho_a \sim T$ for half-filled and quarter-filled Umklapp scattering respectively. The conversion of ρ_a data from constant P to constant V *versus* T [15] supports the claim of quarter-filled Umklapp scattering for the T-dependence of ρ_a.

The observation of a Kohler's law for ρ_c [27] in the intermediate temperature regime is in agreement with our description and equation (15) but implies neither a coherent motion along c nor a Fermi liquid behavior in the (ab) planes. The breakdown of this law above $T_{\max}$ agrees with the quasiparticle states being no longer the eigenstates in the Luttinger liquid.

The finding of a small exponent K_ρ in the intermediate state at $T < T_{\max}$ suggests that some 1-D features remain even though the 2D anisotropic electron gas evolves towards a Fermi liquid. Actually, this claim is supported by the existence of an infrared absorption peak of $\sigma(\omega)$ which is well defined at 170 cm^{-1} in the PF$_6$ and also in ClO$_4$ salts [43–46] and contains most of the spectral weight.

Finally we must comment on the strong pressure dependence of $T_{\max}$ which rules out a linear relation between t_b and $T_{\max}$. Bourbonnais *et al.* [13] have derived a strong renormalization of the intra-chain hopping rate by the intra-chain electron hole interaction. The 2-D crossover thus reads

$$T_{\max} \sim t_b \left(\frac{t_b}{t_a}\right)^{\frac{1-K_\rho}{K_\rho}} \cdot \tag{16}$$

Following the data for (TMTSF)$_2$PF$_6$, equation (16) implies a pressure dependence for K_ρ of 2% kbar^{-1} in line with the lattice dimerization decreasing from left to right in the diagram of Figure 2. However, the most serious problem with this model lies in the amount of renormalization which is far too strong for the selenide compounds.

Equation (16) might be only valid in the case of strong renormalization, *i.e.* for the sulfur compounds.

6 Conclusion

A striking behaviour of ρ_c has emerged from the study of its temperature and pressure dependence in $(\mathrm{TMTSF})_2\mathrm{PF}_6$ and $(\mathrm{TMTTF})_2\mathrm{PF}_6$. We have identified a new strongly pressure dependent line in the $(\mathrm{TM})_2\mathrm{X}$ phase diagram which is defined by the temperature at each pressure where $\mathrm{d}\rho_c/\mathrm{d}T = 0$ and is related to the onset of a 2-D cross-over. We infer that the low lying excitations behaviour of the 1-D electron system is that of a 1-D Luttinger liquid above this line. The intermediate phase is still not behaving like a Fermi liquid since $\rho_a \sim T^2$ while $\rho_b \sim T$. A Fermi liquid behaviour is restored below 10 K or so, probably of a 3-D nature in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ or in $(\mathrm{TMTSF})_2\mathrm{PF}_6$ under pressure. The intermediate temperature regime is characterized by different temperature dependences of the resisitivity along b and a with a temperature dependence of ρ_c governed by the geometrical mean between the a and b temperature-dependences. The b-transverse coupling leading to a deviation from perfect 2-D nesting governs the DC and very low frequency transport while 1-D features can still be observed in the far infrared properties. As far as $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ is concerned, the intermediate state extends up to room temperature according to the behaviour of ρ_c [47] and optical data displaying a transverse plasma edge [25].

The picture of the intermediate temperature range can be viewed as the coexistence of Mott insulating properties at high energy with those of a very good metal at low energy.

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