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Towards a consistent picture for quasi-1D organic superconductors

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ABSTRACT

The electrical resistivity of the quasi-1D organic superconductor $(TMTSF)_2PF_6$ was recently measured at low temperature from the critical pressure needed to suppress the spin-density-wave state up to a pressure where superconductivity has almost disappeared [1]. This data revealed a direct correlation between the onset of superconductivity at T_c and the strength of a non-Fermi-liquid linear term in the normal-state resistivity, going as $\rho(T)=\rho_0+AT+BT^2$ at low temperature, so that $A\to 0$ as $T_c\to 0$. Here we show that the contribution of low-frequency antiferromagnetic fluctuations to the spin–lattice relaxation rate is also correlated with this non-Fermi-liquid term AT in the resistivity. These correlations suggest that anomalous scattering and pairing have a common origin, both rooted in the low-frequency antiferromagnetic fluctuations measured by NMR. A similar situation may also prevail in the recently discovered iron-pnictide superconductors.

Organic superconductivity has been discovered in the organic salt $(TMTSF)_2PF_6$ which belongs to the much broader isostructural family of $(TM)_2X$ compounds where TM stands for the molecules TMTSF or TMTTF. This diagram has been studied extensively over the last 30 years with detailed reports in the literature [2]. However, the electronic transport of the metallic state has attracted much attention since the finding of a strongly temperature dependent resistance prior to the SC transition around 1 K was unusual for a metal as the resistance is expected to saturate at low temperature following a power law with an exponent larger than unity. In the initial discovery a sublinear temperature profile of the resistivity in the vicinity of the superconducting transition had been noticed [3] and attributed to precursors of the SC transition itself [4]. Above this precursor regime, the resistivity remained quasi-linear up to about 10 K, a temperature above which a more traditional quadratic Fermi liquid-like regime was recovered. This behaviour proportional to T in the low temperature limit has also been recognized by other groups [5] and considered as an important experimental feature in these materials. The fact that a strong temperature dependence of the resistivity prevails at low temperature had thus been considered as a piece of evidence for the existence of paraconductivity in a quasi-1D conductor above T_c [4]. The calculation of Azlamazov–Larkin diagrams based on a time dependent Ginzburg–Landau theory provided, (i) a 3D domain restricted to the very vicinity of T_c where the transverse coherence extends over several interchain distances d, i.e., $\xi_\perp/d>1$ with $\rho_\parallel^{3D}\alpha T^{-1/2}(\ln T/T_c)^{1/2}$ leading in turn to the classical $1/(T\text{-}T_c)^{1/2}$ divergence of the paraconductivity and, (ii) a 1D regime where $\xi_\perp/d<1$ with $\rho_\parallel^{1D}\alpha T^{1/2}(\ln T/T_o)^{-3/2}$ where T_o represents a renormalized mean field temperature, about one-third of the actual mean field temperature [6].

While the comparison between experiments and the theory for paraconducting fluctuations in Ref. [4] revealed a reasonable agreement in the 3D regime, the fluctuation model failed to reproduce the correct temperature dependence in the range 4–15 K and revisiting this problem appeared to be required.

Furthermore, magnetism is also an important feature in the generic phase diagram of $(TM)_2X$ compounds since a good nesting of the quasi-1D Fermi surface $(TMTSF)_2PF_6$ leads to a spin density wave transition at 12 K accompanied by the opening of a gap over most of the Fermi surface. Increasing pressure degrades nesting and gives way to SC, first inhomogenous at 6 kbar [7] and subsequently homogenous above $P_c\approx 9$ kbar [8] (Fig. 1). The SDW instability can be revived under magnetic field applied along the direction of weakest interchain coupling and fairly well understood in terms of weak coupling theories [9,10]. An additional evidence for the role of repulsive interaction leading to magnetic ordered ground states came from the strong enhancement of the NMR relaxation rate, $1/T_1$ [11–13] revealing the existence of strong antiferromagnetic spin

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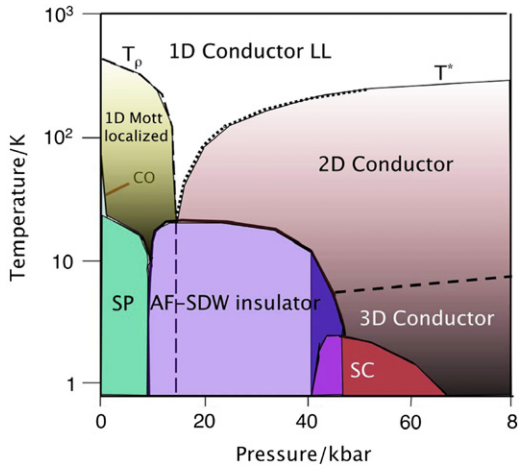

Fig. 1. Generic temperature–pressure phase diagram of the $(TM)_2X$ family of organic conductors. The diagram is drawn for the compound $(TMTTF)_2PF_6$ taken for the origin of the pressure scale. The location of the compounds $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ under ambient pressure are $\approx$37 and 47 kbar respectively in this diagram.

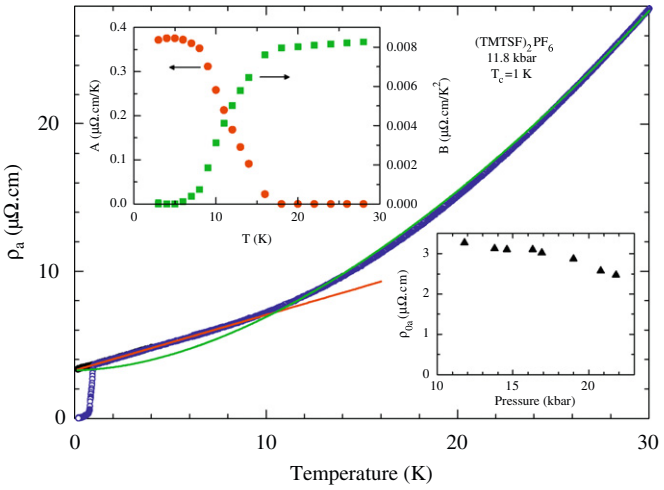

Fig. 2. Temperature dependence of the longitudinal resistivity of $(TMTSF)_2PF_6$ at $P=11.8$ kbar below 30 K, at zero field and under $H=0.05$ T along c^* in order to suppress SC. Left insert: temperature dependence of the A and B coefficients in the polynomial form $\rho(T)=\rho_0+AT+BT^2\ln(t_\perp/T)$ used to fit the resistivity data according to the sliding fit procedure described in the text. The linear and quadratic fits which are displayed in the main panel concern the low and high temperature extremes. Right insert: pressure dependence of the residual resistivity ρ_0 deduced from the low temperature fit at different pressures.

fluctuations in the metallic state above the superconducting instability. These features raise relevant questions about the interplay between pairing and the existence of well-developed repulsive interactions in the neighbourhood of SC.

A reinvestigation of $(TM)_2X$ up to pressures where SC is suppressed, i.e. in the lower right corner of the phase diagram in Fig. 1, became desirable as it might clarify the long standing question about leading parameters governing the existence of organic superconductivity. This work was reported in Ref. [1], where the existence of a close correlation between a linear-in-temperature non-Fermi liquid contribution to the electrical resistivity, at low temperature, AT, and the SC T_c was revealed, showing that both A and T_c vanish at the same pressure.

The data analysis in Ref. [1] focused on the low temperature regime (below 8 K). Here we examine the same data over a larger temperature range, up to 30 K. In Fig. 2 we first show, in the right insert, the value of the residual resistivity ρ_0 determined by a linear extrapolation of the $\rho(T)$ data below 1 K, after suppression of SC by a small magnetic field (see Ref. [1]). The smooth and weak pressure dependence of ρ_0 is a good indication for the quality of the data of the seven pressure runs. The pressure coefficient (-2.5%/kbar) can be attributed to the regular decrease of the effective mass under pressure [17,6].

The resistivity of $(TMTSF)_2PF_6$ at a pressure of 11.8 kbar, close to the critical pressure where $T_{SDW}\to 0$, is displayed in Fig. 2. Below 8 K, it is strictly linear in temperature, with $\rho(T)=\rho_0+AT$. Above 8 K, $\rho(T)$ acquires curvature, which can be captured by a quadratic law $\rho(T)=\rho_0+BT^2$, keeping the same value for ρ_0. A small logarithmic factor actually improves the quality of the quadratic fit [19]. Note that this data just above the SDW critical pressure is very similar to the resistivity of the hole-doped cuprate Nd-LSCO at its quantum critical point [18], where the temperature scale is approximately 10 times higher.

The resistivity can be analysed over a temperature domain from 1 to 30 K, by performing a two parameter fit of the data in Fig. 2 to the polynomial form $\rho(T)=\rho_0+AT+BT^2\ln(t_\perp/T)$ with a sliding temperature window of 4 K and $t_\perp=800$ K. The temperature dependence of the coefficients A and B thus derived is displayed in the left insert of Fig. 2.

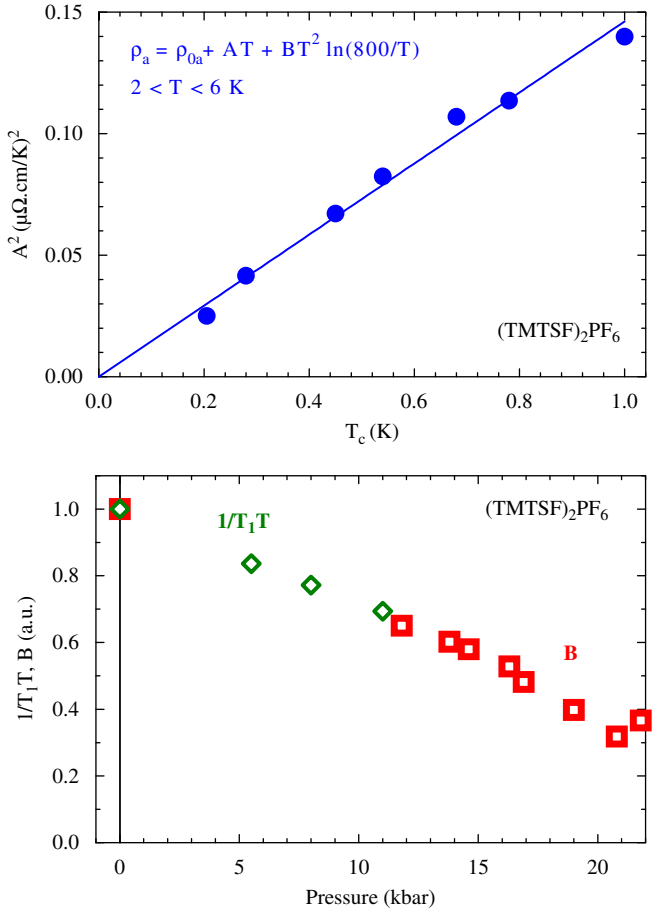

Fig. 3. (Top) Square of the A coefficient, corresponding to the value obtained at low temperature ($2<T<6$ K) for the polynomial fit and the sliding fit procedure described in the text, against T_c onset. The line is a linear fit through the origin with the seven pressure points. (Bottom) Pressure dependence of the B coefficient, corresponding to the value obtained at high temperature ($26<T<30$ K) for the same fit, and of the spin susceptibility measured under pressure via NMR relaxation experiments [12]. Transport data at ambient pressure have been taken from Ref. [15]. The relation $B\propto\chi^2$ is indicative of the Kadowaki–Woods relation encountered in strongly correlated metals [16].

We have applied this fitting procedure to our resistivity curves at all pressures and in Fig. 3(top) we plot A, obtained in the 2–6 K window where B is negligible, as a function of T_c , which is determined by the onset of SC in zero field. It is interesting to see that A^2 and T_c correlate fairly well linearly and quite remarkably, that both vanish at the origin. In addition, the prefactor B of the T^2 law determined in the 26–30 K window, and the square of the electronic susceptibility $(\chi^2(q=0)\propto 1/T_1T)$ follow the same pressure dependence, as shown in the bottom of Fig. 3, which is reminiscent of the Kadowaki–Woods law observed in various strongly correlated metals [16].

In summary, a careful measurement of transport in $(TMTSF)_2PF_6$ up to 21.8 kbar has shown that a reasonable description of the longitudinal resistance up to 30 K or so can be obtained with a second order polynomial fit, provided A and B are allowed to evolve in temperature in a complementary way. This study reveals clearly two extreme regimes. First, the high temperature regime where the T^2 scattering is dominant, with a logarithmic correction due to the 2D character of the electron gas which can be taken as the signature of strong electron–electron Umklapp scattering [20] in a 2D conductor [19]. The amplitude of this scattering is related to the spin susceptibility by a Kadowaki–Woods relation. Second, a low temperature regime where linearity prevails in the temperature dependence of the resistivity, departing from the conventional Fermi liquid behaviour (even at one dimension) and hinting at a tight correlation between such a scattering and the stability of SC. This behaviour can be regarded as a balance between two electron scattering processes adding according to the Mathiessen rule [20]; one channel strictly linear up to 8 K and vanishing above 15 K or so and one growing from zero around 8 K and leveling off above 20 K.

Note that the precise power with which A scales with T_c depends on the form of the fit and the fitting interval. In the fit performed here, where $\rho(T)=\rho_0+AT+BT^2\ln(800/T)$ over an interval from 2 to 6 K, we find $A\propto T_c^{0.5}$ (Fig. 3). In Ref. [1], a fit of the form $\rho(T)=\rho_0+AT+BT^2$ over an interval from T_c to 8 K yields $A\propto T_c^{0.7}$. Performing the same fit over an interval from 0.1 to 4 K yields $A\propto T_c^{1.0}$ [1]. But in all cases, crucially, the onset of the linear term in the resistivity matches with the onset of SC with decreasing pressure, in other words, a pure T^2 Fermi liquid resistivity is only seen when T_c has been completely suppressed with increasing pressure.

The relation between the non-Fermi liquid-like scattering A and the behaviour of the ^{77}Se spin–lattice relaxation rate measured previously in $(TMTSF)_2PF_6$ under pressure [12,14] is also enlightning.

When the contribution to the relaxation provided by AF fluctuations is extracted from the total relaxation rate one can compare its temperature dependence with that of the "linear" scattering. Such a comparison is shown in Fig. 4 where the additional contribution to the $(q=0)$ Korringa relaxation, $\Delta(1/T_1T)$, and A are both seen to decrease with temperature. Since transport and NMR experiments have not been conducted at exactly the same pressure a second best approach consists in using the transport data of the present measurements at 11.8 kbar and the 11 kbar NMR data available in the literature [12] (insert, Fig. 4). Consequently, the comparison between temperature dependencies of A and $\Delta(1/T_1T)$ should not be taken at face value. This is only indicative of a common origin.

In conclusion, there exists in the low-temperature metallic phase of the salt $(TMTSF)_2PF_6$ close to the SDW order a correlation between SC and two properties of the metallic phase: (i) the non-Fermi liquid linear-T resistivity as $T\to 0$ and (ii) the contribution to the spin-lattice relaxation coming from the presence of antiferromagnetic fluctuations close to the SDW instability. Away from the SDW phase at elevated temperatures, the resistivity

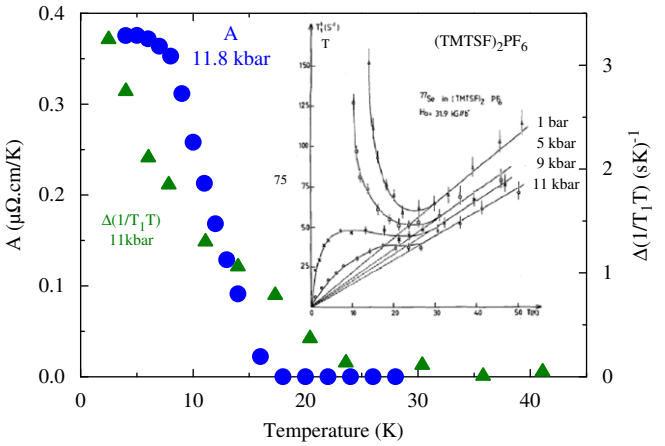

Fig. 4. Temperature dependence of the fluctuation-induced relaxation $\Delta(1/T_1T)$ at 11 kbar from Ref. [12] and of the coefficient of the linear resistivity (A) at 11.8 kbar. The insert shows the ^{77}Se relaxation data at four different pressures where the bump of extra relaxation coming form AF fluctuations is clearly seen [12]. Similar results have been obtained in Ref. [13].

acquires a T^2 curvature whose pressure dependence appear to correlate well with the Korringa law observed in NMR experiments when $T_c\to 0$. The correlation between non-Fermi-liquid resistivity and superconducting T_c further suggests that anomalous scattering and pairing have a common origin [1,22,23].

A similar correlation has been observed in $(TMTSF)_2ClO_4$ under pressure [21] where NMR studies have also been performed. Furthermore, the study of $(TMTSF)_2ClO_4$ has shown that organic superconductivity is governed by two control parameters: (i) the intrinsic control parameter monitored by the intrinsic strength of interactions (pressure) and (ii) the pair breaking mechanism governed by the elastic electron life time which is quite influential in these spin-singlet superconductors [25] with line nodes in the gap [24].

We note that the phase diagram of the iron-pnictide superconductors $Ba(Fe_{1-x}Co_x)_2As_2$, with its adjacent semi-metallic SDW and superconducting phases [26–28], closely resembles that of $(TM)_2X$. The present data analysis supports the suggestion made in Ref. [1] regarding a similar interpretation holding for both families of materials.

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