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Cooperative phenomena in $(\text{TMTSF})_2\text{ClO}_4$: an NMR evidence

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Résumé. — Nous présentons des résultats expérimentaux de relaxation nucléaire pour le composé $(\text{TMTSF})_2\text{ClO}_4$ dans l'état relaxé. Ces résultats montrent clairement l'existence de trois régimes pour le taux de relaxation T_1^{-1} . Dans la région haute température ($T > 25$ K), la loi de Korringa est bien satisfaite, alors qu'une forte déviation est observée en deçà jusqu'à environ 8 K. Cependant, pour $T \lesssim 6$ K, un autre régime de type Korringa, bien que renormalisé et augmenté, apparaît de nouveau. Des résultats préliminaires sur $(\text{TMTSF})_2\text{FSO}_3$ sous pression montrent des caractéristiques de T_1^{-1} tout à fait similaires. Nous proposons une interprétation du comportement de T_1^{-1} basée sur l'existence d'importantes corrélations unidimensionnelles de spins à $2 k_F$ pour une interaction intr chaîne répulsive à courte portée. Le saut cohérent d'électrons de chaîne à chaîne agit comme le principal mécanisme pour la réapparition d'un nouveau régime de Korringa. Cependant, selon les données à faible champ, le schéma d'interprétation proposé ne contredit pas la présence possible d'effets précurseurs supraconducteurs.

Abstract. — We present ^1H and ^{77}Se NMR relaxation data on oriented $(\text{TMTSF})_2\text{ClO}_4$ single crystals in the relaxed state. The data show the existence of three distinct regimes for the nuclear relaxation rate T_1^{-1} . In the high temperature part ($T > 25$ K) the Korringa law seems to be well satisfied whereas in the lower temperature part we have strong deviations down to 8 K or so. At $T \lesssim 6$ K, a different and enhanced Korringa regime appears to be restored. Preliminary results on $(\text{TMTSF})_2\text{FSO}_3$ under pressure also show similar features. We propose an interpretation of the T_1^{-1} behaviour based on the existence of one-dimensional $2 k_F$ spin correlations for repulsive short range intrachain electron-electron interactions. The coherent interchain hopping acts as the main mechanism for the restored but renormalized Korringa behaviour. Moreover, from the low-field data, the proposed scheme of interpretation is not incompatible with the presence of superconducting precursor effects.

1. Introduction.

The organic conductor tetramethyltetraselenafulvalenium-perchlorate $(\text{TMTSF})_2\text{ClO}_4$ is the only organic conductor known so far in which the conducting (metallic-like) state can be maintained down to the helium temperature regime under atmospheric pressure. Moreover, upon

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further cooling, a transition towards a superconducting state is observed at 1.2 K [1], provided great care is taken to cool the system slowly enough, especially below 30 K [2].

The present work deals exclusively with slowly cooled samples (this is the so-called R-state of $(\text{TMTSF})_2\text{ClO}_4$ [3]. The sample was cooled at a constant rate of 10 K per hour). In spite of the clear-cut superconducting properties observed at low temperature, the electronic properties of $(\text{TMTSF})_2\text{X}$ salts have been the source of very intense controversies. On the one hand, several authors [4, 5] pointed out that the moderate band structure anisotropy of the $(\text{TMTSF})_2\text{X}$ series, namely $t_{\parallel}/t_{\perp} \geq 10$ rules out low dimensional effects and supports the application of a mean-field theory for the onset of the superconducting instability. On the other hand, the Orsay group emphasized the need for a careful and self-consistent interpretation of all available experimental data [6, 7]. The latter authors suggested that, even with such moderate anisotropy ratios which are encountered in $(\text{TMTSF})_2\text{X}$ salts, the one-dimensional electron gas theory must be taken as a starting point. The same authors also claimed the existence of precursor effects of the long-range low-temperature order in a domain extending far above the superconducting transition temperature. Such statements were based on the interpretation of low temperature magnetotransport properties (electronic and/or thermal conductivity), far-infrared response at low temperature, electron tunnelling and sensitivity to non-magnetic impurities [8].

The response of the electron gas at low temperature is directly accessible through NMR properties. NMR has already provided much insight into the study of $(\text{TMTSF})_2\text{X}$ salts, establishing for instance the magnetic character of the ground state of $(\text{TMTSF})_2\text{PF}_6$ [9] and the stabilization of a magnetic ground state under a large magnetic field in $(\text{TMTSF})_2\text{ClO}_4$ [2]. A recent nuclear spin-lattice relaxation study of $(\text{TMTSF})_2\text{ClO}_4$ [10] has revealed a breakdown of the Korringa-like behaviour ($1/T_1 T \sim \text{const.}$) at low temperature, while another study on $(\text{TMTSF})_2\text{PF}_6$ under pressure has shown a field dependent $1/T_1$ at 4 K [11]. In the present work, we report ^1H and ^{77}Se ($I = 1/2$) T_1 measurements in $(\text{TMTSF})_2\text{ClO}_4$ performed between 1.2 and 40 K in fields extending from 1 kOe up to 64 kOe. The temperature dependence of $1/T_1$ rules out the interpretation of the low temperature conducting state in terms of a regular single-particle conducting state. However, NMR data can be reconciled very nicely with the standard quasi-1D models for organic superconductors. We shall conclude to the existence of well developed cooperative phenomena which are observable below 25 K. We also report preliminary data of ^1H and ^{77}Se relaxation rate in $(\text{TMTSF})_2\text{FSO}_3$ under a pressure of 10 kbar, showing similar features at low temperature.

2. Experimental.

Details concerning the preparation and the structure of the $(\text{TMTSF})_2\text{X}$ compounds can be found elsewhere [12]. ^1H and ^{77}Se NMR measurements were performed on both single crystals and powdered sample. The typical single crystal size was $3 \times 0.5 \times 0.5 \text{ mm}^3$. The powdered sample consists of many small crystals of total weight $\simeq 50 \text{ mg}$.

The pulsed NMR spectrometer was operated at frequencies of 4.9, 17.1, 25.9, 52.1 MHz. All measurements were performed with the single coil geometry. For single crystals, the coil was wound around the a -axis, so that the external field H_0 was oriented either along the c^* -direction (perpendicular to the planes of organic molecules) or along the b' -direction (perpendicular to the $(a-c^*)$ plane). For the orientation $H_0 \parallel b'$, all data were collected with the same sample configuration at different fields.

For both ^1H and ^{77}Se nuclei, T_1 was determined recording the free-induction signal after a π - $\pi/2$ sequence changing the delay between the two pulses. An exponential decay was observed over more than one decade. For ^1H measurements, the free induction decay was digitized and analysed by a microcomputer at fixed field. For ^{77}Se measurements, the signal was stored in an analog boxcar integrator, while sweeping the field through resonance.

3. Results.

3.1 ^{77}Se - $1/T_1$ in $(\text{TMTSF})_2\text{ClO}_4$. — $1/T_1$ data for ^{77}Se are displayed in figure 1. We noticed no observable field dependence of the relaxation rate between 21 and 64 kOe in the temperature domain 40-25 K. In the same temperature domain, the Korringa law ($1/T_1 \propto T$) is fairly well obeyed. Such a behaviour breaks down in the intermediate temperature region 25-6 K as $1/T_1$ is no longer proportional to T and becomes field dependent. Below 6 K or so, the temperature dependence of $1/T_1$ approaches an other regime in which $1/T_1 \propto T$ down to 1.2 K. At low temperature, say 2 K, no field dependence is observed. The most striking effect shown in figure 1 is the typical non-Korringa behaviour of $1/T_1$ at intermediate temperatures and the very large enhancement of the relaxation rate observed at low temperature, with respect to the high temperature Korringa regime. Notice that in the same temperature domain the static and uniform spin susceptibility does not reveal any significant temperature and field dependence [13]. The data of the spin-spin relaxation time T_2 ($H_0 = 21$ and 64 kOe $\parallel b'$) are displayed in the insert of figure 1. The homogeneous T_2 is much larger than the inhomogeneous T_2^* derived from the linewidth. But the linewidth appears to be linear in field (22 G at 30 kG; 44 G at 64 kG) and this feature may be attributed to various Knight-shift at each ^{77}Se site. Furthermore, the weak drop of T_2 observed below 5 K for $H_0 = 21$ kOe could be related to the establishment of the low temperature regime for $1/T_1$. The overall values of T_2 at 64 kOe are slightly smaller and no decrease is noticed at low temperature.

3.2 ^1H - $1/T_1$ in $(\text{TMTSF})_2\text{ClO}_4$. Data of ^1H spin-lattice relaxation rates at various fields (1, 4 and 12 kOe) are summarized in figure 2. As far as proton resonance is concerned, it is important to distinguish between two contributions; one provided by the hyperfine interaction between electrons spins and methyl-protons nuclear spins, and an other due to the quantum tunnelling

Fig. 1. — ^{77}Se spin-lattice relaxation rate $1/T_1$ in relaxed $(\text{TMTSF})_2\text{ClO}_4$ at various fields. $\circ$: $H_0 = 64.2$ kG $\parallel b'$; $\bullet$: $H_0 = 31.9$ kG $\parallel b'$; $\times$: $H_0 = 21.0$ kG $\parallel b'$; $\triangle$: $H_0 = 6.0$ kG powder; $\square$: $H_0 = 31.9$ kG $\parallel c^*$. (The line is a guide for the eyes.) At the lowest temperature (1.25 K), the arrows mean that the points are undistinguishable. Insert shows spin-spin relaxation time T_2 data.

Fig. 2. — ^1H spin-lattice relaxation rate $1/T_1$ in relaxed $(\text{TMTSF})_2\text{ClO}_4$ at various fields. $\square$: $H_0 = 1.1$ kG powder; $\bullet$: $H_0 = 4.0$ kG $\parallel b'$; $\circ$: $H_0 = 12.2$ kG $\parallel b'$.

of methyl groups at low temperature already extensively studied in several tetramethylated molecules [14, 15, 10]. At high enough temperature, the tunnelling contribution is dominant and therefore can be extrapolated at low temperature with the assumption of an activated behaviour.

The temperature dependence of both protons and selenium relaxation rates scale together. The scaling factor is given by the ratio of hyperfine interactions, which reads (according to Fig. 3) $[a(\text{Se})/a(\text{H})]^2 \approx 2 \times 10^3$. Hence, ^1H low temperature relaxation data may provide some information concerning the electronic properties in low fields. However, it must be kept in mind that in the limit of very low fields, possible parasitic relaxation through paramagnetic impurities could become a dominant effect. The reliability of the low-field data ($H \lesssim 1$ kOe) must therefore be taken with a « grain of salt » although no strong effects of paramagnetic impurities have been reported in $(\text{TMTSF})_2\text{X}$ conductors [13].

The ^1H spin-spin relaxation time T_2 has been found temperature independent and equal to ≈ 16 μs in a field of 4 kOe.

3.3 $(\text{TMTSF})_2\text{FSO}_3$ results. — Similar NMR measurements have been performed on a $(\text{TMTSF})_2\text{FSO}_3$ single crystal under a pressure of 10 kbar in the magnetic field of 55 kOe $\parallel b'$ for ^{77}Se . Under pressure the conducting state of $(\text{TMTSF})_2\text{FSO}_3$ remains stable down to the helium temperature domain, with a possible onset of superconductivity below 3 K at 7 kbar [16]. The nature of the superconducting state of $(\text{TMTSF})_2\text{FSO}_3$ is still under controversy since magnetization measurements have failed to detect any sizeable flux expulsion [17].

The ^{77}Se - $1/T_1$ data are presented in figure 4. They display a broad but well characterized peak of the relaxation rate around 9 K. The temperature at the maximum of $1/T_1$ is about twice higher than the temperature for the maximum for $(\text{TMTSF})_2\text{ClO}_4$ at a similar field. The ^1H - $1/T_1$ data obtained at 10.6 kOe $\parallel b'$ do exhibit the same peaking around 8 K (Fig. 4). Apparently, the temperature at maximum $1/T_1$ scales with the enhancement of the superconducting transition in (2.5 - 3 K) instead of 1.2 K in $(\text{TMTSF})_2\text{ClO}_4$. The interpretation of ^1H relaxation data at low temperature is less ambiguous than in $(\text{TMTSF})_2\text{ClO}_4$ since pressure tends to narrow the CH_3 -tunnelling peak [15].

Fig. 3. — Calculation of the ^{77}Se spin-lattice relaxation rate $1/T_1$ in relaxed $(\text{TMTSF})_2\text{ClO}_4$ from ^1H data (see text). $\circ$: $H_0 = 1.1$ kG powder ; $\bullet$: $H_0 = 4.0$ kG $\parallel b'$; $\square$: $H_0 = 12.2$ kG $\parallel b'$.

Fig. 4. — Spin-lattice relaxation rates $1/T_1$ in $(\text{TMTSF})_2\text{FSO}_3$ under pressure (10 kbar). Right side : ^1H in $H_0 = 10.6$ kG $\parallel b'$; left side : ^{77}Se in $H_0 = 55.3$ kG $\parallel b'$. (The line is a guide for the eyes.)

Because of a weak signal-to-noise ratio, we were unable to extend measurements to higher temperature and thus to recover a Korringa behaviour for the ^{77}Se relaxation rate. However, the data of figure 4 indicate that the relaxation rate at high temperature ($T \gtrsim 20$ K) is about 4 times smaller than in $(\text{TMTSF})_2\text{ClO}_4$. A decrease of the susceptibility by a factor of two in 10 kbar would be large enough to account for the smaller relaxation rate of $(\text{TMTSF})_2\text{FSO}_3$ under pressure [18].

4. Theory.

In what follows, we will try to reconcile the observed experimental data with the predictions of a quasi-1D or « g -ology » model for organic conductors. The g -ology picture has proved to be relatively well suited to treat electron-electron correlations which are well established in organic compounds. Our program is to look at the contribution of these correlations to the nuclear spin relaxation rate T_1^{-1} via hyperfine coupling. The general expression of T_1^{-1} is given by [19] :

$$T_1^{-1} = \gamma_N^2 |A|^2 \sum_{\mathbf{q}} \frac{\chi''_{\perp}(\mathbf{q}, \omega_N)}{\tanh\left(\frac{\omega_N}{2T}\right)}, \quad (1)$$

where A is the contact hyperfine matrix element taken as q -independent, $\omega_N = \gamma_N H_0$ is the NMR frequency with γ_N as the gyromagnetic ratio of the nucleus and H_0 is the applied static magnetic field. $\chi''_{\perp}$ stands for the imaginary part of the retarded spin density wave (SDW) correlation function. For the present theoretical approach, the main contribution to T_1^{-1} will come from the 1D-2 k_F component of $\chi''_{\perp}$. This is supported by the uniform magnetic susceptibility data which do not show any significant variation for $T < 50$ K [13]. The possibility of low temperature SDW enhancement can be predicted in a g -ology picture of the electronic interaction along the chain [20]. This occurs when we assume a repulsive regime for the backward scattering process ($g_1 > 0$). For the forward scattering (g_2), the effect of long-range 3D Coulomb interaction has to be considered. Umklapp scattering (g_3) is not strong since no activation gap for the conductivity is observed for the perchlorate compound [1]. However, at low temperature g_3 may be relevant for the absence of 2 k_F charge density wave effect and the enhanced magnetic behaviour [21-23]. Because of the presence of selenium atoms in TMTSF molecules, some magnetic anisotropy induced by spin-orbit couplings may also be important [21, 24]. The possible nature of the correlations related to these couplings is well known [20, 21, 24] and both divergent SDW and triplet pairing fluctuations can occur. Three-dimensional electronic motion will be assured by inter-chain hopping of electrons with a strength characterized by the transverse hopping integral $t_{\perp}$. The relevance of these and related choices of couplings in connection to the proximity of superconductive and antiferromagnetic states in organic materials have already received considerable attention [21, 23-26].

A Kramers-Kronig analysis of the ratio $\frac{\chi''_{\perp}(q + 2k_F, \omega_N)}{\omega_N}$ in equation (1) for $\omega_N \ll T$ and $|V_F q| < T$ leads to the relation :

$$\frac{\chi''_{\perp}(q + 2k_F, \omega_N)}{\omega_N} \simeq \frac{2}{(\eta + 1)} \frac{\bar{\chi}'_{\perp}(T)}{\pi T} \quad (2)$$

$\bar{\chi}'_{\perp}(T) = \frac{\partial \chi'_{\perp}(T)}{\partial \ln\left(\frac{E_0}{T}\right)}$ and $\chi'_{\perp}(T)$ is the real part of the retarded SDW correlation function. The

sum over q in (1) is made over the 1D momentum interval $|V_F q| < T$ and leads to :

$$T_1^{-1} = \frac{4 \gamma_N^2 |A|^2}{\pi^2 V_F (\eta + 1)} T \bar{\chi}'_{\perp}(T) \quad (3)$$

for the dominant contribution to T_1^{-1} in 1D. The expression of T_1^{-1} is readily derived [27] by making use of the asymptotic properties of the correlation function [20, 28] namely :

$$\chi'_{\perp}(q, \omega, T) = a \left[\text{MAX} \left(\frac{V_F q, \omega, T}{E_0} \right) \right]^{-\eta} \quad (4a)$$

a is a constant which depends on the interaction through η . For example, from reference [29] :

$$\chi'_{\perp}(T) = \frac{1}{2 \pi^2 V_F} \sin \left(\frac{\pi}{2} \eta \right) B^2 \left(\frac{1}{2} - \frac{\eta}{4}, \frac{\eta}{2} \right) (T/E_0)^{-\eta} \quad (4b)$$

with B as the Beta function. The power law exponent η depends on the strength of the various couplings present in the model and may be also temperature dependent. Formally, for divergent SDW correlation and electron-electron couplings smaller than the bandwidth, we have $0 < \eta < 2$ [20, 28]. E_0 is the characteristic energy for strong SDW enhancement ($\eta > 0$) and can take any value between 0 and E_F [20, 21]. It can only be calculated by an accurate knowledge of all temperature dependent effective couplings relevant to SDW correlations. In practice, for organic compounds E_0 is very difficult and indeed impossible to predict. However, from the 1D scaling [20] properties of $\bar{\chi}'(T)$ with E_0 , namely that $\bar{\chi}'(T) = \bar{\chi}'_0 \left(\frac{E_0}{E_F} \right) \bar{\chi}'(T/E_0)$ where $\bar{\chi}'_0$ is considered as the sum of all contributions to $\bar{\chi}'$ for $E_F > T > E_0$, the temperature at which we experimentally observe a clear departure from the Korringa law can serve as a reasonable estimate of E_0 . In presence of interchain hopping, such a power law enhancement of SDW saturates at the 1D to 3D crossover temperature [20, 23, 26, 30] and a low temperature ($T < T_x$) Korringa behaviour can be restored. At T_x , 3D coherent motion of quasi-particles is sufficiently important to rule out the interference between the $2 k_F$ electron-hole and pairing correlations existing in the 1D regime. From the work of reference [31], T_x is given by :

$$T_x \approx T_x^0 (t_{\perp}/E_F)^{\alpha/1-\alpha} \quad (5)$$

where $T_x^0 = t_{\perp}/\pi$ is the crossover temperature for a free quasi-1D electron gas [25, 26]. In presence of interaction ($\alpha \neq 0$), the interference of 1D-correlations lowers the density of states at the Fermi level [32] (pseudo-gap effect) which in turn spreads out the interference effect to lower temperature ($T_x < T_x^0$). This effect is therefore naturally connected to the one-electron 1D-correlation function exponent α [31, 28] and the anisotropy ratio $t_{\perp}/E_F$ of the electronic spectrum [31]. For $g_1 > 0$ and in absence of any gaps, we have : $0 < \alpha < 1$ [28]. In contrast to SDW, this effect is always cut off at E_F for non-retarded local interaction [32, 29].

The prediction of the theoretical temperature profile of $1/T_1$ for quasi-1D metals with SDW enhancement is shown in figure 5a for two values of η . For $T \gg E_0$, SDW fluctuations are small and we have a linear Korringa behaviour of T_1^{-1} . From above, the region $T_x < T < E_0$ for T_1^{-1} can be put in the power law form :

$$1/T_1 = CT(T/E_0)^{-\eta}, \quad (6)$$

with

$$C = \bar{\chi}_0 \frac{2 \gamma_N^2 |A|^2 \eta}{\pi^4 V_F^2 (\eta + 1)} \sin \left(\frac{\pi}{2} \eta \right) B^2 \left(\frac{1}{2} - \frac{\eta}{4}, \frac{\eta}{2} \right)$$

Fig. 5. — (a) Theoretical profile of T_1^{-1} in quasi-1D conductors with SDW enhancement as a function of η . The one-electron dimensionality crossover occurs at T_x . (b) Single crystals low-field ($H_0 < 32$ kG) ^{77}Se data (●) together with the theoretical shape of T_1^{-1} (full line). The dashed line gives the linear regime at low-temperature and the open circles are from the 64 kG $\parallel b'$ data.

which can deviate significantly from the linear regime for allowed values of η . Here η is assumed temperature independent and C will coincide with the slope of the high temperature linear regime ($T \gg E_0$). At $T \rightarrow T_x$, we have a saturation of the enhancement which is of the order of $(T_x/E_0)^{-\eta}$, and for $T < T_x$ a three-dimensional analysis of T_1^{-1} becomes valid [19]. Therefore, in absence of any phase transition in the 3D-regime, the cusp of T_1^{-1} at T_x is followed by a renormalized Korringa behaviour down to $T = 0$ [33]. The ratio between the high and low temperature slopes of T_1^{-1} is estimated to be of the order of the enhancement factor at T_x : $(T_x/E_0)^{-\eta}$. Here we have neglected the possible effect of transient terms that can give a smooth behaviour of T_1^{-1} near T_x and E_0 .

We are now in a position to apply our model to the NMR data of relaxed $(\text{TMTSF})_2\text{ClO}_4$ presented in the previous sections. The selenium low-field data ($H_0 < 32$ kG) of figure 1 obtained on single crystals show a clear departure from the Korringa behaviour at $T \approx 25$ K. This gives rise to a power law regime down to 8 K or so and is followed by an enhanced linear regime for $T \lesssim 6$ K. This suggests to take $E_0 \approx 25$ K. The fit of figure 5b of the power law regime gives $\eta \approx 1.1$, that may be a temperature averaged value. $\eta \sim 1$ is therefore only indicative of the presence of strong SDW correlations. Moreover, their strength is compatible with a drop of T_x down to $T_x \approx 8$ K. For example, if we take the values typically found in the literature for the transverse hopping integrals [34, 35] $t_{\perp} \approx 100 \dots 300$ K and for the anisotropy ratio [34] $t_{\perp}/E_F \approx 1/10 \dots 1/30$. It follows from equation (5) that for $T_x \approx 8$ K we must have $\alpha \approx 1/4 \dots 1/2$,

that is a range of values compatible with η and quite acceptable for organic conductors [21, 24-27, 29, 36]. It implies a sizeable decrease of the density of states at the Fermi level due to 1D effects and this clearly correlates with the interpretation of many other experimental results [6-8, 34]. It is worth mentioning that the enhancement of the slope of the Korringa law in the 3D-regime predicted by the theory $(T_x/E_0)^{-\eta} \approx 3.5$ agrees with that observed experimentally for $T \lesssim 6$ K which is equal to 3 or so. Beside the apparent residual component of T_1^{-1} as $T \rightarrow 0$ the relevant linear shape is essentially parallel to that predicted (Fig. 5b).

5. Discussion.

A field dependence of the nuclear spin-lattice relaxation has been observed in other 1D conductors such as TTF-TCNQ in the temperature domain 300-110 K [37]. The field dependence, $1/T_1 \propto H_0^{-1/2}$, which arises in the latter compound above ≈ 10 kOe has been attributed to electron spins diffusing in one dimension. The extra-contribution coming from the 1D spin diffusion follows approximately the law $1/T_1 \propto \frac{H_0^{-1/2}}{t_\perp \tau_\parallel}$ where $t_\perp$ and $\tau_\parallel$ are the transverse overlap and the intrachain electron single-particle scattering time respectively. Furthermore, the 1D contribution to the relaxation vanishes at low temperature when $\tau_\parallel$ increases strongly. This is verified for TTF-TCNQ as $\tau_\parallel$ increases only by a factor ≈ 3 from 300 to 100 K [37]. As far as (TMTSF)₂X is concerned $\tau_\parallel$ is likely to increase by a factor $\geq 10^3$ going from room to low temperature if the very large value of the low temperature conductivity ($\sigma(4.2 \text{ K}) \approx 10^5 (\Omega \text{ cm})^{-1}$) is attributed to single-particle conduction. With such conditions (i.e., $\tau_\parallel(4.2 \text{ K})/\tau_\parallel(300 \text{ K}) \approx 10^3$) the possibility of low dimensional spin diffusion contributing to the nuclear relaxation may be ruled out. Consequently, we do not believe that the T_1^{-1} field dependence of various nuclei (¹H, ¹³C, ⁷⁷Se) which have been reported in the parent compound (TMTSF)₂PF₆ at 6.9 kbar [11, 38, 39] can reasonably be attributed to a low dimensional spin dynamics. This point of view is also supported by preliminary data of the T_1 field dependence obtained in our laboratory at room temperature up to 65 kOe [40].

The fair agreement between theory and experiment strongly suggests that one-dimensional physics is a relevant and even a necessary ingredient in the understanding of organic compound properties at least for $T \gtrsim 10$ K. It appears then that the usual statement for the vanishing of any 1D effects at $T < T_x^0 \sim t_\perp$ is probably somewhat off for organic conductors studied here. As the renormalized value of T_x decreases with $t_\perp$ one may expect that under high transverse field H_0 , orbital effects begin to play an important role restricting the electronic motion perpendicular to the chains. This decreases effectively $t_\perp$ [41] and can therefore explain the downward shift of the maximum of T_1^{-1} at 64 kOe (Fig. 1). However, one must note that for high fields, the spin-flip part of the short-range electron-electron coupling (g_1) can be strongly affected at low temperature ($T \gtrsim T_x$) [20, 42]. This may also be relevant for the perceptible change in the temperature variation of T_1^{-1} compared to low fields. Nevertheless, the combined effects of orbital motion and field on T_1^{-1} deserve a more detailed study in the future.

As in the 1D regime, electron-hole correlations are generally coupled to the pairing ones, the existence of T_1^{-1} enhancement for protons at field lower than the 3D-superconducting critical field (e.g. $H_0 \sim 4 \text{ kG} \ll b'$) is compatible with coupled antiferromagnetic and superconducting precursor effects at $T \gtrsim 8$ K. As mentioned before, 1D theory states that the triplet pairing can be coupled to $2 k_F$ electron-hole (SDW) pairing for a large range of couplings [24, 20, 21]. CDW effects are also possible but they have no experimental support and can then be discarded. Using such predictions of the 1D theory, many authors have suggested the possibility of triplet ground state for organic superconductors [21, 23, 24, 34, 43]. Here the given interpretation of the NMR data supports, though indirectly, the view that the proximity of antiferromagnetism and superconductivity in organic metals may have a one-dimensional origin.

In conclusion, the behaviour of the ^{77}Se and ^1H nuclear spin-lattice relaxation rate in the R-state of $(\text{TMTSF})_2\text{ClO}_4$ reveals a striking violation of the relaxation theory usually valid in regular metallic conductors (Korringa relation) in the low temperature domain, below about 25 K. We wish to emphasize that the enhancement of the relaxation which we have observed in $(\text{TMTSF})_2\text{ClO}_4$ should not be confused with the $1/T_1$ singularity arising from the establishment of a 3D long-range ordered SDW state in $(\text{TMTSF})_2\text{PF}_6$ at 11 K ($P = 1$ atm) [9, 14]. The establishment of the SDW state in $(\text{TMTSF})_2\text{PF}_6$ induces a spectacular broadening of the ^{77}Se NMR line which has *not been detected* in the present study of $(\text{TMTSF})_2\text{ClO}_4$. A large enhancement of the relaxation in the conducting state of $(\text{TMTSF})_2\text{PF}_6$ at low temperature (under 6.9 kbar) has also been pointed out by Azevedo *et al.* [39].

We have presented an interpretation for the NMR behaviour which derives in a very natural way from the existence of quasi-1D cooperative phenomena in $(\text{TMTSF})_2\text{X}$ using universally accepted band structure parameters.

Furthermore, these new NMR data present more experimental results which are inconsistent with a single-particle interpretation of $(\text{TMTSF})_2\text{ClO}_4$ at low temperature [7].

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