Link between antiferromagnetism and superconductivity probed by nuclear spin relaxation in organic conductors

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The interdependence of antiferromagnetism and superconductivity in the Bechgaard salts series of organic conductors is examined in the light of the anomalous temperature dependence of the nuclear spin-lattice relaxation rate. We apply the renormalization group approach to the electron gas model to show that the crossover from antiferromagnetism to superconductivity along with the anomalous nuclear relaxation rate of the Bechgaard salts can be well described within a unified microscopic framework. For sizable nesting deviations of the Fermi surface, scaling theory reveals how pairing correlations enhance short-range antiferromagnetic correlations via magnetic Umklapp scattering over a large part of the metallic phase that precedes superconductivity. These enhanced magnetic correlations are responsible for the Curie-Weiss behavior observed in the NMR relaxation rate.

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I. INTRODUCTION

In the attempt to understand how itinerant antiferromagnetism can give rise to superconductivity in correlated electron systems, one faces the difficulty of linking the behavior of spin fluctuations that can be extracted from experiments to the mechanism of pairing that leads to superconductivity. This work is about the quest of such a connection in the Bechgaard salts [(TMTSF)$_2$X] series of organic conductors. We ground our analysis on scaling theory, which allows a reexamination of the nuclear spin relaxation in the metallic phase of these low dimensional molecular systems.

The (TMTSF)$_2$X compounds are quasi-one-dimensional (quasi-1D) conductors known to exhibit a spin-density-wave (SDW) state adjacent to superconductivity (SC) in their phase diagrams\textsuperscript{1,2}. This particular sequence of states is achieved by the application of hydrostatic pressure or by chemical means from anion X substitution. Among the host of experimental tools used to study this pattern of phases, the Nuclear Magnetic Resonance (NMR) technique takes on particular importance due to its sensitivity to spin correlations\textsuperscript{3,4,5,6}. In the metallic state of the SC side of the phase diagram, NMR measurements have revealed the existence of an anomalous enhancement of the nuclear spin-lattice relaxation rate $T_{1}^{-1}$. The enhancement was first observed for the ambient pressure superconductor (TMTSF)$_2$ClO$_4$\textsuperscript{7,8,9} and subsequently found by different groups to be a common characteristic of the series in the metallic state above the critical pressure for superconductivity\textsuperscript{10,11,12,13,14}. The $T_{1}^{-1}$ temperature profile shows pronounced deviations from the Korringa law, $T_{1}^{-1} \propto T$, which is normally expected in conventional metals. These deviations were originally ascribed to the presence of short-range antiferromagnetic spin fluctuations extending in temperature dozens of times the superconducting $T_c \sim 1$K in the metallic state\textsuperscript{4}. The amplitude of the deviations are strongly pressure dependent and seemingly tied to the amplitude of $T_{K}^{-1}$, suggesting that antiferromagnetism and Cooper pairing are closely related.

It was initially proposed that one-dimensional short-range antiferromagnetic correlations are a key determinant in the enhancement of $T_{1}^{-1}$. This low dimensional response was shown to ultimately cross over into a higher dimensional metallic phase around 10K, below which a Korringa law was predicted to be recovered\textsuperscript{13}. However, the dimensional aspects of such an interpretation run into difficulties when one compares the temperature scale for the crossover to the range of values otherwise extracted from experiments\textsuperscript{14,15} and which would rather place this scale an order of magnitude higher in temperature. An additional defect comes from an important observation made by Brown et al.,\textsuperscript{3} concerning the temperature profile of the relaxation. Based on the analysis of recent measurements\textsuperscript{16,17} it was shown that the Korringa behavior is in effect not recovered down to the lowest temperature preceding superconductivity. Deviations actually persist indicating that staggered spin fluctuations, though non singular, keep growing as the temperature is lowered, imposing a Curie-Weiss (C-W) temperature dependence for the relaxation rate. Noticeably enough, however, the characteristics of the C-W response persists down to $T_c$ and the amplitude of the anomaly as a whole evolves rapidly under pressure like the temperature scale for the onset of superconductivity. The question then arises if, in accordance with the customary view, a sharp distinction can be drawn between both phenomena, or if they are in effect dynamically linked, a possibility that would not only connect magnetism to superconductivity, but also involve superconductive pairing in the enhancement of spin correlations.

It is from the latter perspective that we shall reconsider
the $T_{1}^{-1}$ problem for the Bechgaard salts. This will be achieved with the help of the weak coupling renormalization group (RG) theory. Recent developments along these lines have demonstrated how the one-loop RG can take into account density-wave and Cooper pairings on equal footing in correlated quasi-1D metals. In the framework of the repulsive quasi-1D electron gas model, it was found that unconventional singlet ‘d-wave’ (SCd) or equal footing in correlated quasi-1D metals. In the framework of the repulsive quasi-1D electron gas model, it was found that unconventional singlet ‘d-wave’ (SCd) or in certain conditions triplet ‘f-wave’ (SCf) superconductivity can be dynamically generated next to a SDW state as alterations of the nesting of the Fermi surface – which mimic pressure effect – are made sufficiently large. From a similar approach to be brought forward here, a C-W type behavior for the spin fluctuation response is shown to take place over a large temperature interval above $T_c$ within a SCd scenario. It originates from magnetic Umklapp scattering whose amplitude is apparently strengthened by constructive interference with superconducting pairing. The amplitude of spin correlations rapidly decline under ‘pressure’, in line with the decrease of $T_c$. When transposed into a $T_{1}^{-1}$ calculation, the RG results can give a satisfactory account of the key features shown by the nuclear relaxation rate, establishing a direct connection between spin fluctuations and the mechanism of superconductivity in the Bechgaard salts.

In Sec. II we introduce the quasi-1D electron gas model in the presence of weak Umklapp scattering and alteration of nesting. We review the results of the three-variables RG method obtained at the one-loop level within a SDW-SCd scenario. The temperature profile of the antiferromagnetic and superconducting responses are given and scrutinized as a function of nesting deviations of the Fermi surface. The temperature scales are extracted and used to construct the phase diagram of the model. In Sec. III, the explicit form for $T_{1}^{-1}$ is calculated from the RG results for both the antiferromagnetic and uniform components of spin fluctuations. In Sec. IV we discuss the results and conclude.

II. ITINERANT ANTFERROMAGNETISM AND SUPERCONDUCTIVITY: RENORMALIZATION GROUP RESULTS

We consider the electron gas model whose bare Hamiltonian for a square lattice of $N_{\perp} \times N_{\perp}$ chains of length $L$ is given by

$$H = \sum_{p,k,\sigma} E_p(k) \hat{c}_{p,k,\sigma}^\dagger \hat{c}_{p,k,\sigma} + \frac{1}{LN_{\perp}^{2}} \sum_{\{k,\sigma\}} \left[ g_1 \hat{c}_{1,+k,\sigma}^\dagger \hat{c}_{1,-k,\sigma} \hat{c}_{2,+k,\sigma} \hat{c}_{2,-k,\sigma} \hat{c}_{1,\sigma} \right] + g_2 \left( \hat{c}_{1,+k,\sigma}^\dagger \hat{c}_{1,-k,\sigma} \hat{c}_{2,+k,\sigma} \hat{c}_{2,-k,\sigma} \hat{c}_{1,\sigma} \right) + g_3 \left( \hat{c}_{1,+k,\sigma}^\dagger \hat{c}_{1,-k,\sigma} \hat{c}_{2,+k,\sigma} \hat{c}_{2,-k,\sigma} \hat{c}_{1,\sigma} \right) + \text{H.c.} \delta_{k_1+k_2=k_1'+k_2'(\pm G)},$$

where the operator $\hat{c}_{p,k,\sigma}$ ($\hat{c}_{p,k,\sigma}$) creates (destroys) a right ($p = +$) and left ($p = -$) moving electron of wave vector $k = (k_{\perp}, k_{\parallel})$ and spin $\sigma$. The free part is modeled by the one-electron energy spectrum

$$E_p(k) = v_F |p - k_{\perp}| - 2t_{\perp} \cos k_{\perp} - 2t_{\perp}' \cos 2k_{\perp} - 2t_{\parallel} \cos k_{\parallel},$$

where $v_F$ and $k_{\perp}$ are the longitudinal Fermi velocity and wave vector; $t_{\perp}$ and $t_{\parallel}$ are the nearest-neighbor hopping integrals in the two perpendicular directions. The small transverse second nearest-neighbor hopping $t_{\perp}' \ll t_{\perp}$ parameterizes the alteration of nesting of the open Fermi surface, which simulates the most important effect of pressure in our model. The quasi-1D anisotropy of the spectrum is $E_F \approx 15t_{\perp} \approx 3000K$, and $E_F = v_F k_{\perp} \approx 3000K$ is the longitudinal Fermi energy conguent with the range found in the Bechgaard salts. $E_F$ is half the bandwidth $E_0 = 2E_F$ in the model. The interacting part of the Hamiltonian is described by the bare backward ($g_1$) and forward ($g_2$) scattering amplitudes between right and left moving electrons. In terms of the extended-Hubbard model parameters, $g_1 = U - 2V$ and $g_2 = U + 2V$, where $U$ and $V$ are the on-site and nearest-neighbor repulsion. The half-filling character of the band – due to the small dimerization of the chains – gives rise to Umklapp scattering of bare amplitude $g_3$ for which momentum conservation is satisfied modulo the longitudinal reciprocal lattice vector $G = (4k_F, 0, 0).

In the repulsive sector, the couplings satisfy $g_1 - 2g_2 < g_3$, a condition that promotes antiferromagnetic spin fluctuations in the presence of nesting. In spite of a variety of possibilities for the couplings that would be generic for the phenomena we want to discuss, one can call upon experiments and band calculations to delimit their range and make a choice for the amplitude of normalized couplings $g_i \equiv g_i / \pi v_F$ for the calculations that will follow. We first note that – half-filling – Umklapp term $\tilde{g}_3 \approx \Delta_D / 4 \Delta_D$ is proportional to a small dimerization gap $\Delta_D$ due to the modulation of the electron transfer integral along the stacks. For (TMTSF)$_2$X compounds, the modulation is relatively small and one finds $\Delta_D / 4 \Delta_D \lesssim 0.1$. The backscattering coupling $g_1$ governs spin fluctuations.
Excitations and is involved in the enhancement of static spin susceptibility.\textsuperscript{27,29} Experiments in the Bechgaard salts indicate that this enhancement is around 20\% the non interacting band value at low temperature.\textsuperscript{27,29,31}

This is compatible with the use of a bare backscattering amplitude in the interval $\bar{g}_1 \simeq 0.3\ldots0.5$, giving in turn the range $\bar{g}_3 \simeq 0.02\ldots0.04$ for Umklapp. As for the forward scattering, its bare amplitude can be adjusted in order for the calculated temperature scale for SDW ordering to fall in the range of observed values 10...20 K at moderate nesting frustration. This leads to $\bar{g}_2 \simeq 0.5\ldots0.7$. Though non exhaustive, this range of parameters is found to be generic of the interdependence between magnetism and superconductivity. The RG calculations that follow have been carried out for $\bar{g}_1 = 0.32$, $\bar{g}_2 = 0.64$ and $\bar{g}_3 = 0.02$.

The RG method consists of integrating successively the degrees of freedom from the high energy cut-off $E_F$ down to the energy $\frac{1}{\ell}E_0(\ell) = \frac{1}{2}E_F e^{-\ell}$ above and below the Fermi sheets at step $\ell$. At the one-loop level, the corrections to the amplitudes $\bar{g}_i$ as a function of $\ell$ come from the electron-electron (Cooper) and electron-hole (Peierls) scattering channels. Both interfere and generate momentum dependence for the scattering amplitudes as $E_0(\ell) = E_0 e^{-\ell}$ is reduced with increasing $\ell$. Here the influence on the RG flow of the smallest transverse hopping integral, $t_{\perp,c}$, is negligible and has been ignored. In the three momentum variables scheme of the renormalization group adopted here\textsuperscript{15}, each sheet of the Fermi surface is divided into 32 pieces or patches whose location defines a particular transverse momentum $k_3$ in the $b$ direction. Only the $k_3$ dependence is retained for the couplings, which becomes $g_i \to g_i(k_{\perp,l}^{'},k_{\perp,l}^{''};k_{b1},k_{b2})$. The explicit form of the corresponding flow equations has been given previously [Eqns.(10-12) of Ref.\textsuperscript{15}] and these need not to be repeated here.

Following their integration up to $\ell \to \infty$, the presence of a singularity in the scattering amplitudes signals an instability of the normal state at a ‘critical’ temperature $T_{\mu}$. To see what kind of order it refers to, we compute the susceptibilities. For the intrachain interactions given above, a singularity has been shown to occur either in the static $\mu$=SDW or the $\mu$=SCd susceptibility $\chi_{\mu}^{SDW}$\textsuperscript{12,15}. In the RG framework, these are expressed as a loop integration

$$\chi_{\mu}(\mathbf{q},\omega) = \frac{1}{\pi v_F} \int_0^{x_{\mu}(\mathbf{q},\omega)} (f_{\mu}(k_{\perp,l}) z_{\mu}^2(k_{b})) d\ell,$$

where $(\ldots)$ is an average over $k_{b}$, $f_{\mu}(k_{b}) = 1(\cos k_{b})$ is a form factor for the $\mu$ =SDW (SCd) order parameters; $z_{\mu}(k_{b})$ is the scaling factor associated to the response function of the channel $\mu$ and which will be defined shortly. In the $T_{\mu}^{-1}$ analysis given in Sec. II A, the dependence on the (real) frequency $\omega$ and the three-dimensional wave vector $\mathbf{q} = (q_{b},q_{b},q_{c})$ of $\chi_{\mu}^{SDW}$ is needed. It can be introduced through the upper bound of the loop integration, which will be taken as the normalized free electron dynamic susceptibility $\chi_{\mu}^{0} = \pi v_F x_{\mu}^{0}$ of the SDW channel

$$\chi_{\mu}^{0}(\mathbf{q}_{0} + \mathbf{q},\omega) = \ln \left[ \frac{E_F + \psi(\frac{1}{2})}{T} \right] - \frac{1}{8\pi^2} \int^{+\pi}_{-\pi} d\mathbf{k}_{b} d\mathbf{k}_{c} \left[ \psi \left( \frac{1}{2} + i \frac{\xi_{\mu}(k_{\perp,l},\mathbf{q},\omega)}{4\pi T} \right) + c.c. \right].$$

$\chi_{\mu}^{0}$ has been expressed in terms of the deviations $\mathbf{q}$ to the best nesting vector $\mathbf{q}_{0} = (2k_{F},\pi,\pi)$. Here $\psi(x)$ is the digamma function and

$$\xi_{\mu}(k_{\perp,l},\mathbf{q},\omega) \simeq v_F q + (2t_{\perp,b} \sin k_{b}) q_{b} - 4t_{\perp,b} \cos 2k_{b} + (2t_{\perp,c} \sin k_{c}) \sin q_{c} - \omega.$$

As for $\chi_{\mu}^{SCd}$ in the Cooper channel, it will be evaluated in the uniform $q = 0$ and static $\omega = 0$ limit, where $\chi_{\mu}^{0} = \ln E_F/T$.

Following Ref.\textsuperscript{13}, the flow equation for the static SDW vertex part at $(2k_{F},\pi)$ is given by

$$\partial_{\ell} z_{\mu}^{SDW}(k_{b}) = \frac{1}{2\pi} \int^{+\pi}_{-\pi} d\bar{k}_{b} B_{\mu}(\bar{k}_{b}) z_{\mu}^{SDW}(\bar{k}_{b}) \times [g_2(\bar{k}_{b} + \pi,k_{b},k_{b} + \pi) + g_3(\bar{k}_{b},\bar{k}_{b},\bar{k}_{b} + \pi,k_{b} + \pi)],$$

whence $q_{\pi}$ is raised and at the approach of $T_{\mu}$. The above expressions depend on the $\ell$-derivative of the Peiers and Cooper loops which read

$$B_{\mu/C}(\bar{k}_{b}) = \sum_{\nu=\pm 1} \theta \left| E_0(\ell)/2 + \nu A_{\mu/C}(\bar{k}_{b}) \right| - E_0(\ell)/2 \times \left[ \frac{1}{2} \left( \tanh \frac{E_0(\ell)/2 + \nu A_{\mu/C}(\bar{k}_{b})}{4T} + \tanh \frac{E_0(\ell)/2}{4T} \right) \right].$$

where $A_{\mu}(\bar{k}_{b}) = 4t_{\perp,b} \cos 2k_{b}$, $A_{C} = 0$, and $\theta(x)$ is the step function with the definition $\theta(0) = \frac{1}{2}$.

The RG results at the one-loop level for the temperature dependence of the normalized $\chi_{\mu}^{SDW}$ and $\chi_{\mu}^{SCd}$ ($\bar{\chi}_{\mu} = \pi v_F \chi_{\mu}$) are given in Fig. 1 for different values of the nesting frustration parameter $t_{\perp,l}^{'}$. At small $t_{\perp,l}^{'}$, $\chi_{\mu}^{SDW}$ diverges signaling an instability towards the formation of a SDW state at the temperature $T_{SDW}$. This scale decreases as $t_{\perp,l}^{'}$ is raised and at the approach of the threshold $t_{\perp,l}^{'} \simeq 25.6K$ (for the set of parameters used), it undergoes a rapid drop. However, $T_{SDW}$ does
FIG. 1: (a) Temperature variation of the normalized static susceptibility $\tilde{\chi}_b$ in the SDW and SCd channels at various $t'_{\perp b}$ on either side of the threshold value $t'_{\perp b}^*$; (b) RG phase diagram of the quasi-1D electron gas model (see text). The dashed line stands as the C-W scale $\Theta$ in the superconducting sector. The dotted line defines the temperature domain of the C-W behavior.

As one reaches $t'_{\perp b} = t'_{\perp b}^*$ where the system is superconducting, $\Theta = 0$ and $\tilde{\chi}_{SDW} \sim 1/T$. If superconductivity was absent the system would be then quantum critical in the SDW channel with a singular antiferromagnetic correlation length $\xi_{a,b} \sim T^{-\nu}$, of exponent $\nu = 1/2$ in the $ab$ plane. As for the dynamical exponent $z$, the results of Sec. III give $z = 2$. The C-W form carries on in the superconducting sector where $\Theta$ becomes positive and grows with $t'_{\perp b}$. Since $\tilde{\chi}(T \to 0) \to 1/C\Theta$, the amplitude of $\Theta$ is thus connected to the size of spin fluctuations in the low temperature limit, which decreases rapidly with $t'_{\perp b}$. It follows that $\tilde{\chi}_{SDW}$, though no longer singular above $t'_{\perp b}^*$, is still temperature dependent: despite altered nesting conditions and scaling towards the formation of a singlet superconducting ground state, antiferromagnetic correlations continue to grow down to $T_c$. The C-W behavior extends several times $T_c$ in the metallic state.

The existence of a C-W behavior above $t'_{\perp b}^*$ is intimately linked to the presence of magnetic Umklapp scattering ($g_1$) in the model: without this coupling, $\tilde{\chi}_{SDW}$ is essentially flat at low temperature. This is confirmed by putting $g_1 = 0$ and carrying out the calculation with the remaining coupling constants. Most surprisingly, however, strongly reduced nesting of the Fermi surface alone plays little role in the enhancement of Umklapp. The augmentation turns out to be a consequence of Cooper SCd pairing that reinforces spin fluctuations in the metal state. This can be easily checked by setting $B_C = 0$, which removes all the Cooper pairing terms from the flow equations of the coupling constants. In this superconductive pairing-free scheme, which is equivalent to a ladder diagrammatic (mean-field) summation in the density-wave pairing channel alone, the C-W behavior is negligible for $t_{\perp b}' \geq t'_{\perp b}$.

Interestingly enough, Umklapp scattering is coupled to $g_1$ and $g_2$, which both flow to strong coupling at the approach of the superconductive fixed point at $T_c$. At variance with ordinary s-wave superconductivity,
these scattering amplitudes are momentum dependent in a SCd scenario yielding an overall positive sign for their coupling with Umklapp. It follows that the singular growth of SCd pairing at low temperature (Fig. 1a) interferes positively with Umklapp expanding the temperature range where this coupling and in turn spin fluctuations increase. It is this self-consistency between the two pairing channels that is responsible for the C-W law for the staggered magnetic susceptibility down to $T_c$. It is worth stressing that the reinforcement of the SDW channel is not limited to the SC sector, but is also manifest for $t'_{1b} \lesssim t'_{1b'}$, where the C-W behavior, with a negative $\Theta$, is indicative of a SDW instability driven by Cooper pairing.

III. NUCLEAR SPIN LATTICE RELAXATION RATE

A. Theoretical prediction for the staggered and uniform contributions to nuclear relaxation

We now turn to the derivation of the nuclear relaxation rate in the RG scheme. The $T_1^{-1}$ calculation starts from the Moriya expression\(^\text{[11]}\)

$$ T_1^{-1} = T \int |A_q|^2 \frac{\chi''(q, \omega)}{\omega} d^3q, \quad (9) $$

which relates $T_1^{-1}$ to the imaginary part of the retarded spin susceptibility $\chi''$. Here $A_q$ is proportional to the hyperfine matrix element. The integral over all $q$ indicates that $T_1^{-1}$ is sensitive to staggered and uniform electronic spin correlations with respectively large $q \sim q_0$ and small parallel $q \sim 0$. We then consider the following decomposition

$$ T_1^{-1} = T \left( \int_{q=0} + \int_{q \neq 0} \right) |A_q|^2 \frac{\chi''(q, \omega)}{\omega} d^3q, \quad (10) $$

$$ \equiv T_1^{-1}(q \sim 0) + T_1^{-1}(q_0). $$

Let us first examine the staggered component, $T_1^{-1}(q_0)$. Using (11), the expression of $\chi''(q + q_0, \omega)$ at small $q$ and $\omega$ is given by

$$ \text{Im} \chi_{SDW}(q + q_0, \omega) = \frac{\chi_{SDW}(q_0) \Gamma_{\omega}}{(1 + \xi^2_{\perp} q^2 + \xi^2_{\parallel} q^2_0 + \xi^2_{\perp} (\sin q_0)^2 + \Gamma^2_{\omega} \omega^2).} \quad (11) $$

From the results of the Appendix A, $\xi^2_{\perp} = \xi_{0,a}^2(z_{SDW}(k_b))/\bar{\chi}_{SDW}$, is the squared of the correlation length along $i = a, b$, and $c$ directions; $\xi_{0,a} \propto v_F/T_0$ and $\xi_{0,b,c} \propto t_{1, b,c}/T_0$ are the corresponding coherence lengths evaluated at the SDW temperature $T_0 \simeq \frac{12K}{\Gamma}$ obtained at small $t'_{1b}$; $\Gamma = \Gamma_0(z_{SDW}(k_b))/\bar{\chi}_{SDW}$ is the relaxation time for SDW fluctuations and $\Gamma_0 \propto 1/T_0$ is a characteristic short-range time scale. The integration over $q$ is carried out in Appendix A and yields

$$ T_1^{-1}(q_0) = 2\pi^3 |A_{q_0}|^2 [N(E_F)]^2 v_F \Gamma_{\omega}/(\xi_{0,a} \xi_{0,b}) \times T \bar{\chi}_{SDW} \left[ \frac{1}{\sqrt{1 + \xi^2_{\perp}}} - \frac{1}{\sqrt{(1+r)(1+r + \xi^2_{\perp})}} \right], \quad (12) $$

where $r \simeq 1.144(z_{SDW}(k_b))/\bar{\chi}_{SDW}$ and $N(E_F) = 1/\pi v_F$ corresponds to the density of states at the Fermi level. The enhancement of the staggered component as the temperature is lowered is thus connected to the static SDW response which can be obtained by the RG method.

We next consider the uniform component of the relaxation rate which is connected to the imaginary part of the dynamic spin susceptibility at small $\omega$ and $q$. In this limit, $\chi(q, \omega)$ has been shown to be non singularly enhanced by interactions at low temperature\(^\text{[20,21]}\). Within the random phase approximation, the expression of the imaginary part reads

$$ \chi''(q, \omega) \bigg|_{\omega, q = 0} = -\frac{\eta^2}{4\pi^3} \sum_p \int dk d^3k_c [n(E_p(k + q)) - n(E_p(k))] \delta(\omega - E_p(k + q) + E_p(k)), \quad (13) $$

where $n(x)$ is the Fermi distribution. The uniform contribution for the imaginary part is enhanced from electron-electron interaction by the factor $\eta$ (see Appendix A). From previous measurements of the static and uniform spin susceptibility\(^\text{[20,21]}\), its enhancement is about 20% in the low temperature domain so that the factor can be fixed to $\eta \simeq 1.2$ considered as temperature independent in the range of interest\(^\text{[22]}\). Substituting in (10), the remaining integrals are carried out in the Appendix A and lead to the ‘Korringa’ component

$$ T_1^{-1}(q \sim 0) = \pi |A_0|^2 [N(E_F)]^2 \eta^2 T. \quad (14) $$

While the uniform contribution is nonsingular, its amplitude is known to become ultimately larger than the staggered component at high enough temperature.\(^\text{[22]}\):
B. Results and relation to experiments

To compare the sum of \( T_1^{-1} \) and \( T_2^{-1} \), with the experimental findings for \(^{77}\text{Se} T_1^{-1}\) in (TMTSF)$_2$PF$_6$ and (TMTSF)$_2$ClO$_4$ [Refs. 115], we adjust the two unknown constants \(|A_0|\) and \(|A_{\text{g}}|\) so that the amplitude of \( T_1^{-1} \) falls in the range of the observed values (Fig. 4). Since in the high temperature region, the nuclear relaxation rate is dominated by the uniform contribution \[T_1^{-1} \approx |A_0| \] matching with the measured \( T_1^{-1} \) values at 50K. The other constant \(|A_{\text{g}}|\) for the staggered part is tuned such that \( T_1^{-1} \) is congruent with the measured value at 20K. A ratio of \(|A_{\text{g}}|/|A_0| \approx 10^{-2}\) is thus found for the hyperfine matrix elements.\(^{10} \) The total expression of \( T_1^{-1} \) is then plotted in Fig. 5 for various values of the nesting frustration parameter \( t'_{\perp,b} \), namely below and above the threshold for superconductivity in the calculated phase diagram of Fig. 1b.

In the SDW domain for \( t'_{\perp,b} < t'_{\perp,b}^* \), the relaxation rate \[T_1^{-1} \approx T_2^{-1} \approx |\tilde{\chi}_{\text{SDW}}|/\sqrt{1 + \zeta_c^2} \] at large \( r \), namely close to \( T_\text{SDW} \), where it is dominated by the staggered contribution. The latter then develops a three-dimensional singularity of the form \[T_1^{-1} \approx (T - T_\text{SDW}^{-1})^{-\nu} \] with \( \nu = 1/2 \) when \( \zeta_c \) becomes large as \( T \to T_\text{SDW} \). The power law exponent conforms to the one of mean-field theory in three dimensions.\(^{27} \) This has been shown long ago to agree with the (TMTSF)$_2$PF$_6$ data of Fig. 4.\(^{11} \) As one moves away from \( T_\text{SDW} \) in temperature, \( \zeta_c \) becomes smaller and the system evolves towards a two-dimensional behavior where \[T_1^{-1} \approx \tilde{\chi}_{\text{SDW}} \], corresponding to \( \nu = 1 \).

Now if one moves along the \( t'_{\perp,b} \) scale, by approaching \( t'_{\perp,b}^* \) from below, one enters into a transitional regime where \( T_\text{SDW} \) is relatively small; \( \zeta_c \) then becomes large and three-dimensional order, with \( \nu = 1/2 \), develops only in very close proximity to the critical point. For \( t'_{\perp,b} > t'_{\perp,b}^* \) on the SCd side, \( T_1^{-1} \) is no longer singular but shows a pronounced anomaly due to short-range spin fluctuations. These extend deeply in the normal state up to 20 K or so, above which the uniform component of the relaxation takes over and \( T_1^{-1} \propto T \). To the anomaly of \( T_1^{-1} \) found down to about 10K (Fig. 3) corresponds a distinct region of increase of \( \chi_{\text{SDW}} \) (Fig. 4a) due to the growth of antiferromagnetic correlations close to SDW ordering. \( T_1^{-1} \) passes through maximum near 10K, whose amplitude, and to a lesser extent its location, are \( t'_{\perp,b} \) dependent; \( T_1^{-1} \) then finally starts to go down at lower temperature until one reaches \( T_c \).

The overall structure of the calculated \( T_1^{-1} \) anomaly compares fairly well with the data of Creuzet et al.\(^{10} \) (Fig. 1) and Brown et al.\(^{2} \) on (TMTSF)$_2$PF$_6$ above the critical pressure \( P_c \) for superconductivity and on (TMTSF)$_2$ClO$_4$ at ambient pressure (> \( P_c \), inset of Fig. 4).

If one looks more closely at the decrease of the calculated \( T_1^{-1} \) below 10K in Fig. 3 it is found that it deviates from a linear Korrinaga law as a consequence of the growth of spin fluctuations, responsible for the C-W behavior for \( \chi_{\text{SDW}} \) in this temperature range (Fig. 1). To see how these fluctuations mark the relaxation rate, it is instructive to look at the temperature dependence of \( T_1 T \) shown in Fig. 5. In the left panel of this Figure, the three different regimes of the calculated relaxation rate can be identified. In the high temperature regime \( T_1 T \) tends to level off, dominated by the uniform component (14). At lower temperatures, \( T_1 T \) becomes controlled by the staggered component, two 2D linear regimes, governed by \( \xi_{a,b} \), can be singled out and related to those found previously for \( 1/\tilde{\chi}_{\text{SDW}} \) in Fig. 2. Indeed, in the intermediate temperature range, between 20 and 10K or so, \( T_1 T \) is weakly affected by nesting alterations and evolves with a...
steep slope that would extrapolate to a finite critical temperature. However, approaching 10K, these alterations become more perceptible and the slope of $T_1 T$ is reduced and enters in the low temperature C-W regime of the form $T_1 T = C(T + \Theta)$. Following the example of 1/\tilde{\chi}_{SDW}$ when $t'_{\perp b} \gtrsim t''_{\perp b}$, $\Theta = -T_{sdw}$, and $T_1 T$ is found to be linear, except in the very close vicinity of $T_{sdw}$, where $\xi_c$ becomes large. At $t''_{\perp b}$, $\Theta$ vanishes and finally grows positively for $t'_{\perp b} > t''_{\perp b}$ as $T_c$ decreases. The slope $\xi$ diminishes as $t'_{\perp b}$ grows, whereas the product $C \Theta$, corresponding to the extrapolated zero temperature intercept of $T_1 T$, increases.

From the NMR work of Creuzet et al.\cite{creuzet} (inset of Fig. 4) and from of Shinagawa et al.\cite{shinagawa} (Fig. 5) lower panel on the right) on (TMTSF)$_2$ClO$_4$ at ambient pressure, the three regimes of $T_1 T$ can be discerned in the data. On the same panel the continuous line corresponds to a fit using the sum of (12) and (14), where $|A_{q_0}| / |A_0| \approx 0.65 \times 10^{-2}$, with a C-W regime that corresponds to $\Theta \approx 1.8K$. In the upper panel of the same Figure, the $T_1 T$ data of Wu et al.\cite{wu} and Creuzet et al.\cite{creuzet} (Fig 4) for (TMTSF)$_2$PF$_6$ at $P \approx 10$ kbar are shown. The fit (blue curve) is obtained for $|A_{q_0}| \approx 12.8$ (sec$^{-1}$) and $|A_{q_0}| / |A_0| \approx 0.65 \times 10^{-2}$ for which $\Theta \approx 11.3K$. For the data on the same compound at 8 kbar, we have $|A_{q_0}| \approx 18$ (sec$^{-1}$) and $|A_{q_0}| / |A_0| \approx 0.9 \times 10^{-2}$, with $\Theta \approx 1.7K$. The three temperature regimes are revealed from the data at small $\Theta$. However, as pressure increases, in (TMTSF)$_2$PF$_6$, for example, the temperature interval over which the C-W takes place experimentally apparently increases in size. This pressure effect is not captured by the present calculations for which the C-W temperature interval is essentially constant as a function of $t'_{\perp b}$ (Fig. 4b). As for the rapid increase of $\Theta$ as the ratio $T_c/T_1 (t''_{\perp b})$ falls off in Fig. 4b, it is found to be in fair agreement with the experimental findings in (TMTSF)$_2$PF$_6$, at least up to moderate pressure where data are available.\cite{creuzet}

IV. DISCUSSION AND CONCLUSION

The explanation put forward for the enhancement of spin fluctuations in the metallic state of the (TMTSF)$_2X$ compounds modifies an earlier scheme of interpretation proposed long ago, in which an effective – strongly renormalized – scale for interchain hopping played the dominant part in the temperature profile of the enhancement that was considered one-dimensional in character. In the quasi-one-dimensional view adopted here, the coherent wrapping of the open Fermi surface takes place in the temperature domain $\sim t_{\perp b}$ ($\sim 100K$), that is far above the range where the anomalous features of the relaxation rate are found. It is rather the small parameter $t'_{\perp b}$ for nesting deviations of the whole Fermi surface that acts as the critical parameter and triggers the modification of the relaxation rate in temperature. For repulsive couplings, the increase of $t'_{\perp b}$ alters, as expected, the stability of the SDW fixed point, simulating the effect of pressure. At some threshold value $t'_{\perp b}$, the SDW fixed point is unstable. However, because of a finite mixing between the weakened density-wave and unaltered Cooper pairing singularities in the scattering amplitudes, the electron system is not a Fermi liquid, but is rather characterized by superconducting order, which takes place in the SCd channel for intrachain repulsive interactions.

We have first seen how this crossover between fixed points operates as a function of temperature above $t'_{\perp b}$ for the SDW response function. At high temperature, thermal broadening of the Fermi surface makes nesting deviations less perceptible and the electron system is still attracted by the SDW primary fixed point. As the temperature decreases and the fine details of the Fermi surface in the $ab$ plane become progressively coherent, the singularity of the SDW response is suppressed. This coincides with the emergence of a secondary SCd fixed point, whose influence stretches in temperature about ten times the maximum $T_c$ value reached at $t''_{\perp b}$. Throughout the flow toward $T_c$, SDW correlations, albeit non singular, persist to increase in the $ab$ plane, thanks to the strengthening of Umklapp by Cooper SCd pairing. The increase of the SDW susceptibility can be fitted with a Curie-Weiss law in temperature.

The above features found for the susceptibility are also encountered in the antiferromagnetic component of the nuclear relaxation rate as an anomalous enhancement that emerges out of a ‘Korringa’ or Fermi liquid like behavior at low temperature. These characteristics of the nuclear relaxation rate adhere to a large extent to the experimental facts found by NMR in the Bechgaard salts. The relatively rapid evolution of the relaxation rate en-
hancement seen under pressure, in particular concerning the characteristics of the Curie-Weiss law, can find an explanation in line with the strong reduction of $T_C$ under pressure. This connection between and experiment gives significant support to a mechanism of superconductive pairing mediated by spin fluctuations.

As stressed before, at the core of the weak coupling scaling theory resides the finite quantum interference between density-wave and Cooper pairing channels. It gives rise to d-wave superconductivity from the exchange of spin correlations, and conversely to the enhancement of spin correlations from superconductive pairing. This interference is manifest in the one-loop perturbation theory. Higher order effects like the interaction between SDW modes of fluctuations are neglected. In self-consistent renormalized theory of spin fluctuations where such mode-mode interactions are included, a Curie-Weiss enhancement can be found in the high temperature part of the normal phase. In such an approach, the SDW channel is singled out and interference with Cooper pairing absent. It follows that C-W enhancement does not persist down to the lowest temperature, where instead a Fermi liquid behavior takes place. Despite this fundamental difference, one cannot exclude that the observed Curie-Weiss enhancement of the nuclear relaxation rate superimposes to some extent both contributions.

In conclusion the above results have highlighted that from a renormalization group approach to the quasi-one-dimensional electron gas model, it is possible to obtain a microscopic description of the spin fluctuations, as extracted from nuclear relaxation in (TMTSF)$_2$X. It is through the same approach that the mechanism of interplay between itinerant antiferromagnetism and superconductivity has been worked out for the phase diagram of these compounds. In parallel with the work presented here for the relaxation rate, the same approach has been applied to electron transport in the Bechgaard salts, which also shows an anomalous temperature dependence in the metallic state above superconductivity. Calculations of the transport scattering rate are compared to the resistivity in a separate paper.

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**APPENDIX A: NUCLEAR RELAXATION RATE**

1. **Antiferromagnetic part**

The wave vector and frequency dependence of the dynamic susceptibility is in general not given by the RG method used here. However, an expression for the imaginary part of the SDW susceptibility can be obtained by restoring the $q$ and $\omega$ dependence through the boundary conditions of the flow equation for the susceptibility $\chi_{SDW}$. Using $[3]$ at small $q$ and $\omega$, one finds

$$\text{Im} \chi_{SDW}(q + q_0, \omega) = \frac{1}{\pi v_F} \text{Im} \left[ \int_0^\infty \frac{\omega}{\omega^2 + \Gamma_0^2} \frac{d\omega}{\omega} \chi_{SDW}(q_0) \right]$$

$$= \chi_{SDW}(q_0) \text{Im} \left[ 1 - \xi_0^2 \frac{q^2}{q_0^2} - \xi_b^2 \frac{q^2}{q_0^2} - \xi_c^2 \frac{q^2}{q_0^2} \right]$$

This expression will be equated with the expansion of

$$\text{Im} \chi_{SDW}(q + q_0, \omega) \approx \text{Im} \left[ \frac{\chi_{SDW}(q_0)}{1 + \xi_0^2 \frac{q^2}{q_0^2} + \xi_b^2 \frac{q^2}{q_0^2} + \xi_c^2 \frac{q^2}{q_0^2} - i \Gamma_0 \omega} \right]$$

which we shall use in the following. Here $\xi_i^2 = \xi_{0,i}^2 \langle \tilde{z}_{SDW}^2(k_0) \rangle / \tilde{\chi}_{SDW}$, is the square of correlation length along $i = a, b, c$ directions; $\xi_{0,i}^2$ are the corresponding coherence lengths evaluated at the SDW temperature $T_0 \approx 12K$ obtained for small $t_{lib}^2$, that is

$$\xi_{0,i}^2 = -\left( \frac{v_i}{2\pi T_0} \right)^2 \text{Re} \left[ \int_0^\pi \frac{dk_b}{2\pi} \psi'(1 - \frac{t_{lib}^2}{\pi T_0} \cos(2k_0)) \right]$$

where $v_a = v_F/\sqrt{2}$ and $v_{b,c} = t_{lib}$. The relaxation time for SDW fluctuations is $\Gamma = \Gamma_0 (\tilde{z}_{SDW}^2(k_0) / \tilde{\chi}_{SDW}$ where $\Gamma_0$ is a characteristic short-range time scale at short distance, which is given by:

$$\Gamma_0 = \frac{1}{\pi T_0} \text{Re} \left[ \int_0^\pi \frac{dk_b}{2\pi} \psi'(1 - \frac{t_{lib}^2}{\pi T_0} \cos(2k_0)) \right]$$

Substituting the imaginary part $[A1]$ in the expression for the antiferromagnetic component of the nuclear relaxation rate $[10]$, we get in the limit $\omega \to 0$:
\[ T_1^{-1}(q \sim q_0) = 8T|A_0|^2 \int_{q_0}^{\xi_{ab}} \int_{0}^{\xi_{ab}} \int_{0}^{\pi} \frac{\text{Im} \chi(q, \omega)}{\omega} dq dq dq_c \]
\[ = 8\pi|A_0|^2 T N(E_F) \chi_{SDW}(q_0) \frac{\nu_F \Gamma_0}{\xi_{ab}\xi_{ab}} \int_{0}^{\sqrt{\pi}} \int_{0}^{\sqrt{\pi}} \int_{0}^{\pi} \frac{dxdyd\xi_c}{(1 + x^2 + y^2 + \xi_c^2 (\sin \varphi)^2)^2} \]
\[ = 2\pi^3|A_0|^2 T N(E_F) \chi_{SDW}(q_0) \frac{\nu_F \Gamma_0}{\xi_{ab}\xi_{ab}} \left[ \frac{1}{\sqrt{1 + \xi_c^2}} - \frac{1}{\sqrt{(1 + r)(1 + r + \xi_c^2)}} \right] \]

(A5)

Here \( r = \alpha (\xi_{SDW}^2(k_b))/\chi_{SDW} \) and \( \alpha \approx 1.114 \) is a constant introduced to adjust the upper bound cut-off in order to go through a polar integration in the \( ab \) plane. This allows an analytical expression to be found that deviates from the numerical (rectangular) integration by less than 0.1%.

2. Uniform part

Following Eq. (10), the uniform component of the relaxation rate is given by

\[ T_1^{-1}(q \sim 0) = T|A_0|^2 \int_{q \sim 0} \frac{\text{Im} \chi(q, \omega)}{\omega} d^3q / (2\pi)^3, \quad (A6) \]

Here \( \text{Im} \chi(q, \omega) \mid_{\omega \sim 0} = -\eta^2 \frac{1}{4\pi^2} \sum_p \iint dE_p |\nabla E_p(k)| \left[ n(E_p(k) + \omega) - n(E_p(k)) \right] \delta(\omega - E_p(k + q) + E_p(k)), \]

(A8)

Only a constant (Fermi) surface integral remains. The substitution in (A6), allows one to write

\[ T_1^{-1}(q \sim 0) = T (2\pi)^3 |A_0|^2 \eta^2 \sum_p \int d^3q \int \frac{dS_F}{|\nabla E_p(k_F)|} \delta(E_p(k_F + q) - E_p(k_F)) \]

(A9)

Using the approximation \( |\nabla E_p(k_F)| \approx v_F \) for \( t_{\perp b} \ll t_{\perp b} \ll v_F \), and \( \int dS_F = 4\pi^2 \), one obtains the ‘Korringa’ component

\[ T_1^{-1}(q \sim 0) = \pi |A_0|^2 |N(E_F)|^2 \eta^2 T. \quad (A10) \]
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