Scaling Relation between Nuclear Relaxation and Magnetic Susceptibility in Organic Conductors: Evidence for 1D Paramagnon Effects

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(Received 18 January 1989)

A scaling relation between nuclear relaxation T_1^{-1} and magnetic susceptibility χ_S is established for $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ organic conductors over a wide domain of temperature at ambient pressure. The analysis in terms of quantum scaling demonstrates the important role played by 1D paramagnon magnetic excitations in the Bechgaard salts. Similar observations made on $(TMTTF)_2PF_6$ clarify the nature and the dimensionality of correlations of the nonordered phases in both $(TMTSF)_2X$ and $(TMTTF)_2X$ series compounds.

PACS numbers: 76.60.Cq, 72.15.Nj, 74.70.Kn

Electronic correlations and anisotropy in quasi-one-dimensional organic conductors are well known to be among the basic ingredients responsible for the remarkable richness of phase transitions found in these materials.[1] For the Bechgaard salts $(TMTSF)_2X$ and the sulphur analog $(TMTTF)_2X$ series (TMTSF stands for tetramethyltetraselenafulvalene and TMTTF stands for tetramethyltetrathiafulvalene), pressure conditions, anion substitution ($X=PF_6$, AsF_6, ClO_4, etc.), applied magnetic field, etc., have demonstrated that antiferromagnetism is the heart of a unified low-temperature phase diagram of both series.[1,2] Despite the established importance of Coulomb interactions, the origin of correlations, their amplitude, dimensionality, and temperature profile in the broad nonordered temperature domain still remain far from being well understood. The nuclear relaxation rate (T_1^{-1}) acting as a probe of electronic correlations for all wave vectors $\mathbf{q}$ and their dimensionality can provide a considerable clarification of these basic questions.[2(b),3] In this Letter, we present and analyze ambient-pressure $^{77}Se(I=\frac{1}{2})$ and $^{13}C(I=\frac{1}{2})$ T_1^{-1} data for the nonordered phases of the three compounds $(TMTSF)_2PF_6$, $(TMTSF)_2ClO_4$, and $(TMTTF)_2PF_6$, which are known to display the essential features of both series.[2,4-6] We correlate the T_1^{-1} data to those of the uniform and static magnetic susceptibility χ_S already measured by Miljak and Cooper,[7] under similar conditions. A clear scaling relation between T_1^{-1} and χ_S^2 is found to exist over quite a large temperature domain. A simple dynamic scaling analysis of T_1^{-1} shows that this remarkable relation results from the important role played by 1D uniform ($q\approx 0$) magnetic excitations (paramagnons) for all these compounds. The low-temperature deviations to this scaling allow in turn the evaluation of the delicate balance between uniform and antiferromagnetic correlations in both series.

Electronic degrees of freedom are well known to influence the nuclear relaxation rate in correlated metals, which according to the Moriya expression[5] reads

$$T_1^{-1}=2\gamma_N^2|A|^2T\int d^dq\,\chi_\perp''(\mathbf{q},\omega_N)/\omega_N \qquad (1)$$

($\hbar=k_B=1$), which relates T_1^{-1} to the imaginary part of the retarded magnetic susceptibility $\chi_\perp''$. Here γ_N and ω_N are the gyromagnetic ratio and the Larmor frequency of the nucleus in the applied magnetic field H_0. The local nature of the hyperfine coupling $|A|$ corresponds to an integration over *all* $\mathbf{q}$. Therefore, T_1^{-1} probes both uniform ($\mathbf{q}\approx 0$) and antiferromagnetic ($\mathbf{q}\sim\mathbf{Q}_0$) correlations. The dynamics, which are quite different for each type of fluctuation, will impose a clear-cut differentiation between the $T_1^{-1}[\mathbf{q}\sim 0]$ and $T_1^{-1}[\mathbf{q}\sim\mathbf{Q}_0]$ contributions, and this will depend on the temperature and also on the *dimensionality* d of the correlations.

Following the standard dynamic scaling hypothesis for the uniform correlations,[8(a),9] there exists a scaling relation $\chi''(\mathbf{q},\omega_N)\approx\chi_S(T)D(\mathbf{q}\xi_F,\omega_N\xi_F^{\bar z})$ between the dynamic and the static susceptibilities for $q\xi_F<1$ and $\omega_N\xi_F^{\bar z}<1$. Here D is a scaling function,[8(b)] ξ_F is the characteristic length scale for $q=0$ (F for ferromagnetic) correlations in the presence of Coulomb interactions, and $\bar z$ stands for the dynamical exponent. From the microscopic paramagnon theory for itinerant-electron magnetic systems, the scaling relies on the characteristic length[9-11] $\xi_F\propto[1-g(T)\chi_S^0]^{-\bar\nu}$ where $\bar\nu>0$, χ_S^0 is the noninteracting static susceptibility at $q=0$, and $g(T)$ is the Coulomb repulsion which is temperature dependent (backscattering) in the quasi-1D case.[10,11] Since q scales as ξ_F^{-1} and ω_N as $\xi_F^{-\bar z}$, the full dependence of T_1^{-1} on χ_S and ξ_F near $q\approx 0$ can be readily extracted:

$$T_1^{-1}[q\sim 0]\propto T\chi_S(T)\xi_F^{\bar z-d}. \qquad (2)$$

The quantum renormalization-group approach for the paramagnon problem[9] allows one to take the *Gaussian*

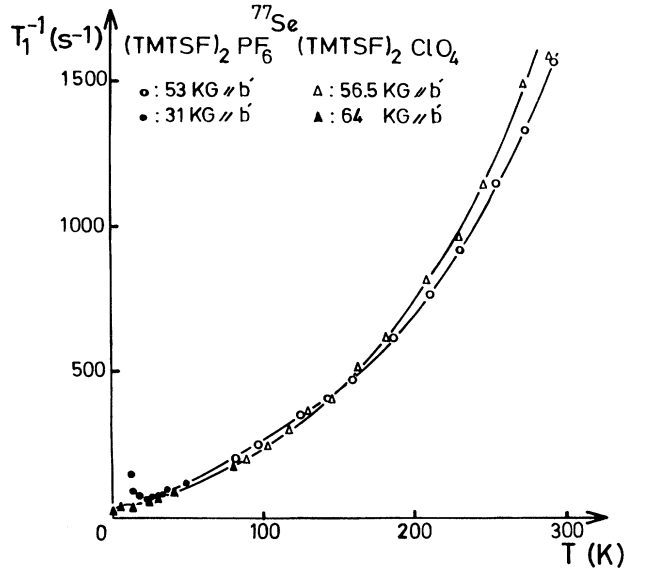

FIG. 1. ^{77}Se T_1^{-1} vs T data of $(TMTSF)_2PF_6$ (circles) and $(TMTSF)_2ClO_4$ (triangles) at $p=1$ bar. The filled circles and triangles correspond to the low-temperature data of Creuzet *et al.*, Refs. 5(a) and 5(c), respectively.

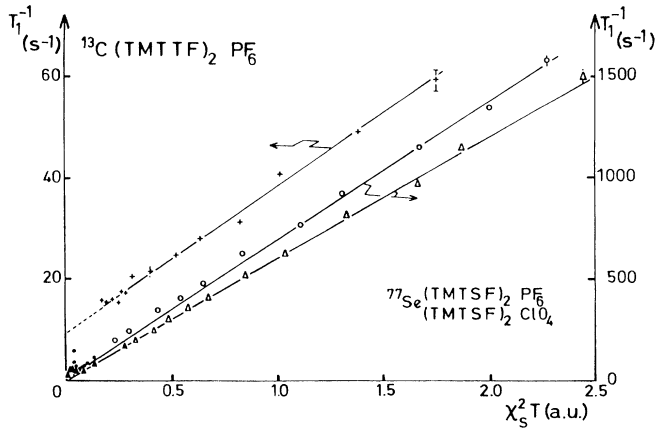

FIG. 2. ^{77}Se data of Fig. 1 vs $\chi_S^2 T$ values of Miljak and Cooper (Ref. 7) for $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ (right scale). On the left scale, ^{13}C T_1^{-1} vs EPR $\chi_S^2 T$ data of Creuzet *et al.* [Ref. 5(b)] for $(TMTTF)_2PF_6$ at $P=1$ bar and $H_0=42$ kG, $\| c^*$.

indices at $d \geq 1$: $\bar{\nu}=\frac{1}{2}$, $\bar{z}=3$, with $\chi_S(T) \propto \xi_F^2$. One then gets

$$T_1^{-1}[q\sim 0] \propto T[\chi_S(T)]^{(5-d)/2}. \quad (3)$$

Therefore, the deviations of $(T_1T)^{-1}[q\sim 0]$ with respect to a normal metal Korringa law $(T_1T)^{-1}[q\sim 0]$ $\sim$const are entirely dictated by the temperature dependence of $\chi_S(T)$ and the dimensionality d. One then recovers the well-known direct RPA (Gaussian) calculations in $d=3$ $(T_1^{-1} \propto T\chi_S)$ [12] and $d=1$ $(T_1^{-1} \propto T\chi_S^2)$. [11]

We now focus our attention on the ^{77}Se T_1^{-1} vs T data for $(TMTSF)_2PF_6$ [slow-cooled $(TMTSF)_2ClO_4$] single crystals, collected for 50 (40) $< T <$ 297 K. The data were obtained on a regular spectrometer at $H_0=53$ kG, $\| b'$, for $(TMTSF)_2PF_6$ and at 56.5 kG, $\| b'$, for $(TMTSF)_2ClO_4$ and for which the decay of magnetization is observed over more than one decade. In order for the analysis to be complete, we have added in Fig. 1 the low-temperature ^{77}Se data of Creuzet *et al.* [5(a),5(c)] obtained on $(TMTSF)_2PF_6$ above the Néel temperature $T_N \approx 12$ K ($T_N < T < 50$ K; $H_0=31.9$ kG, $\| b'$) [Ref. 5(a)] and $(TMTSF)_2ClO_4$ (6 K $\leq T \leq$ 40 K; $H_0=64$ kG, $\| b'$). [5(c)] For both compounds, the T_1^{-1} increase with T is characterized by an upward curvature over quite a large temperature domain ($T > 30$ K), corresponding to an enhancement of $(T_1T)^{-1}$ which increases with T. This behavior is reminiscent of the observed χ_S profile with T for the same compounds under similar pressure conditions. Using the χ_S data of Miljak and Cooper [7(a)] for $(TMTSF)_2ClO_4$, Fig. 2 shows quite clearly that a relation of the form $T_1^{-1} \propto T\chi_S^2(T)$ is remarkably followed to $T \gtrsim 30$ K. For the PF_6 salt also, the χ_S data of Miljak and Cooper [7(b)] show that the same type of relation dominates the relaxation above the critical domain ($T \geq 30$ K) in which T_1^{-1} becomes singular. The comparison with the above quantum scaling prediction for the $d=1$ Gaussian paramagnon theory is rather striking. The results of Figs. 1 and 2 would suggest the remarkable fact that for the Bechgaard salts the temperature dependences of *both* $(T_1T)^{-1}$ and χ_S are essentially dictated by 1D paramagnon effects. So far, similar effects were only established in $d=3$ itinerant-electron ferromagnets (e.g., intermetallic compounds) for which a $(T_1T)^{-1} \propto \chi_S$ law is known to be commonly observed. [13] The organic conductors studied here, however, are the itinerant-electron antiferromagnet [$(TMTSF)_2PF_6$] and the superconductor [$(TMTSF)_2ClO_4$, $T_c \approx 1.2$ K], [1] and one should thus expect deviations at low temperature. Actually, their amplitudes are best seen by reporting the enhancement $(T_1T)^{-1}$ vs χ_S^2 on a logarithmic scale, as shown in Fig. 3. For the $(TMTSF)_2ClO_4$ salt, for example, deviations from the $(T_1T)^{-1} \sim \chi_S^2$ law (solid lines) are only detectable near $\chi_S^2 \approx 3.3$ ($T \lesssim 30$ K), below which χ_S is essentially temperature independent with an $(T_1T)^{-1}$ enhancement that becomes huge due to antiferromagnetic fluctuations. [5(c)] For $(TMTSF)_2PF_6$, the antiferromagnetic enhancement becomes singular as we approach the transition [5(a)] $[(T_1T_N)^{-1} \sim |T-T_N|^{-1/2}$, $T \rightarrow T_N]$. Antiferromagnetic precursors to the transition are, however, detectable up to $\chi_S^2 \approx 3.5$ ($T \approx 100$ K) above which the law $(T_1T)^{-1} \propto \chi_S^2$ holds. In Fig. 1, this corresponds roughly to the temperature domain where $T_1^{-1}(PF_6) > T_1^{-1}(ClO_4)$. Therefore, from Fig. 3, one can have a rather clear idea of the relative weights associated with $q \approx 0$ and $\mathbf{q} \approx \mathbf{Q}_0$ types of correlations as a function of T in the Bechgaard salts.

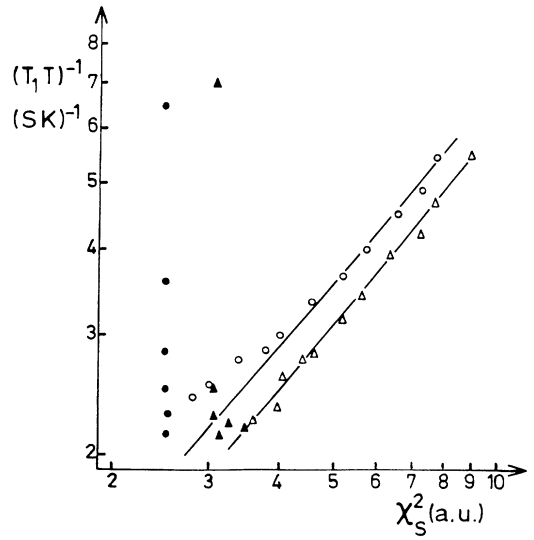

FIG. 3. ^{77}Se enhancement $(T_1T)^{-1}$ data of Fig. 1 as a function of the χ_S^2 data of Miljak and Cooper (Ref. 7) for $(TMTSF)_2PF_6$ (circles) and $(TMTSF)_2ClO_4$ (triangles).

At this point, a comparison with what is observed in the more correlated $(TMTTF)_2X$ series is worthwhile. The $(TMTTF)_2PF_6$ compound at $P=1$ bar is well known to be the most correlated compound of the sulphur series.[2] From resistivity,[4] it is characterized by a Mott-Hubbard charge localization below $T_\rho \approx 230$ K. A 1D $2k_F$ lattice softening and a 3D spin-Peierls distortion have been observed by x ray[14] and by EPR and NMR [5(b)] below 40 and 19 K, respectively. In Fig. 2 (left scale), we have reported the observed ^{13}C T_1^{-1} versus the EPR $\chi_S^2 T$ data of Creuzet *et al.*,[5(b)] obtained on ^{13}C enriched $(TMTTF)_2PF_6$ samples. The data are taken in the localized domain 40 K $\leq T \leq$ 200 K, where lattice softening effects can be considered as irrelevant. Note that the increase of χ_S in this domain is essentially the same as for the Bechgaard salts.[5(b),7] From Fig. 2, it is clear that a linear relation between T_1^{-1} and $\chi_S^2 T$ is again well obeyed over the entire temperature range considered. In contrast to selenide compounds, however, T_1^{-1} does not extrapolate to zero as $\chi_S^2 T \rightarrow 0$. This corresponds to a sizable contribution to T_1^{-1} which is temperature independent. It turns out actually that below T_ρ, 1D antiferromagnetic correlations, well known to be strong for q near $2k_F = Q_0$ in such conditions, give rise precisely to such a contribution. Indeed, from the dynamic scaling hypothesis, one can write near $2k_F$ [Refs. 2(b) and 8],

$$\chi_\perp''(q-2k_F,\omega) \approx \chi(2k_F,T) D_{AF}(q\xi_{AF}, \omega\tilde{\xi}_{AF}^z)$$

for $q\xi_{AF} < 1$ and $\omega\tilde{\xi}_{AF}^z < 1$. Here $\xi_{AF} \sim T^{-\nu}$ is the antiferromagnetic correlation length while the $2k_F$ magnetic susceptibility is written as a power-law singularity,[15] $\chi(2k_F,T) \propto (T/T_\rho)^{-\gamma}$. From microscopic calculations, the correlation length (ν) and the dynamical (z) exponents are equal in 1D and are given by $\nu = z = 1$. This is so since T, ω, and q always enter on the same footing in the calculations.[15] Commensurability effects in the electronic degrees of freedom below T_ρ lead to the well-established result $\gamma = 1$ for repulsive interactions.[2,15] In $d=1$, one then has $T^{-1}[q \sim 2k_F] \propto T\xi_{AF}^{\gamma/\nu - 1 + z} \sim$ const, which is temperature independent, a result which agrees with the direct calculation of Ref. 5(c). The total T_1^{-1} profile below T_ρ will then be given by

$$T_1^{-1} \approx C_0 T \chi_S^2 + C_1 , \tag{4}$$

where C_0 and C_1 are positive constants. Therefore this T_1^{-1} prediction for a 1D quantum antiferromagnet fully agrees with the data of Fig. 2 for $(TMTTF)_2PF_6$. This result also supports the spin-Peierls character of the lattice instability at lower temperature.[5(b)] More generally, Fig. 2 illustrates rather nicely how the $(TMTTF)_2X$ series can be more correlated than the $(TMTSF)_2X$ series with respect to *both* types of correlations. If we now look at the effects of pressure, resistivity measurements[6] show that T_ρ decreases under pressure, and when T_ρ is no longer observed, the phase diagram presents the characteristics of the $(TMTSF)_2X$ series. Here, a decreasing T_ρ means that $\chi(2k_F,T)$ is depressed and C_1 will then become smaller. This will correspond in Fig. 2 to T_1^{-1} profiles that will naturally evolve smoothly toward those of the $(TMTSF)_2X$ series.

It is important to emphasize at this point that it is the quantum nature of paramagnon excitations that justifies the use of Gaussian exponents for $T_1^{-1}(q \sim 0)$ in (3). According to the standard quantum renormalization-group analysis of Hertz,[9] the upper critical dimension for paramagnon systems is $d_c = 1$ [$\epsilon = 4 - (d + \tilde{z})$]. Therefore, at d_c ($\epsilon = 0$) only logarithmic corrections are expected for T_1^{-1} in (3). As the effective interaction $g(T)$ (backward scattering), which is responsible for these corrections, scales to zero for $T \rightarrow 0$ for a 1D metal,[10,11,15] these corrections are thus very weak and will not be easily detectable. Another important remark is that for a 1D interacting electron gas in the continuum limit, paramagnons correspond to the spin bosons of the bosonization transformation of the fermion Hamiltonian.[15] For metallic conditions, their harmonic (Gaussian) character is well known to be relevant. In turn, it is precisely this harmonicity that characterizes the phase fluctuations associated with antiferromagnetic correlations and which leads to the powerlike singularity of $\chi(2k_F,T)$ [15,16] used in (4) for $T_1^{-1}[q \sim 2k_F]$.

In conclusion, we have shown that from a 1D quantum-scaling analysis of the observed relation between T_1^{-1} and χ_S, one can evaluate the respective weights associated with uniform and antiferromagnetic correlations in both the $(TMTSF)_2X$ and $(TMTTF)_2X$ series. The evidence for one-dimensional paramagnon effects in $(TMTSF)_2ClO_4$ clearly emphasized that, as far

as the $(TMTTF)_2X$ series is concerned, 1D features should not be excluded from the microscopic description of low-temperature short- and long-range correlations of the Bechgaard salts.

C.B. thanks the Centre National de la Recherche Scientifique for support. We thank C. Lenoir for contribution in the sample preparation.

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