Fluctuation Effects in Low-Dimensional Spin-Peierls Systems: Theory and Experiment

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The influence of one-dimensional spin-Peierls fluctuations on the temperature dependent magnetic susceptibility of an antiferromagnetic chain is calculated using the renormalization group and the functional-integral methods. The results are shown to give an accurate description of fluctuation effects found in recently synthesized organic spin-Peierls compounds.

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A distinctive feature of a large variety of phase transitions in quasi-one-dimensional compounds is their broad regime of 1D fluctuations precursor to the true critical point. Peierls or charge-density-wave systems stand as classical examples for which 1D lattice softening found by either x-ray or neutron scattering experiments has a marked influence on electronic spin degrees of freedom well above the transition [1]. Surprisingly, it is only very recently that similar effects were observed in insulating quasi-1D spin-Peierls (SP) systems. The organic series [2] $(\mathrm{BCPTTF})_2X$ (BCPTTF stands as benzocyclopentyltetrathiafulvalene, $X = \mathrm{AsF}_6, \mathrm{PF}_6$) and the cuprate compound [3] CuGeO_3, for example, can be considered among the first few spin-Peierls systems for which 1D fluctuations effects are clearly visible in x-ray diffraction and magnetic susceptibility measurements. Correspondingly, despite an apparent similarity existing between the Peierls and spin-Peierls instabilities, there is so far no theoretical description of coupled lattice and spin fluctuations in the spin-Peierls case [4]. In this Letter, we propose a microscopic treatment for such fluctuation effects combining the renormalization group (RG) and functional-integral methods. In the Jordan-Wigner (JW) fermion representation of spins, we first show that a one-loop approximation including lattice and thermal RG transients allows a precise determination of the temperature dependent spin susceptibility $\chi(T)$, which is comparable to recent joined together Bethe ansatz and conformal field theory calculations for the Heisenberg model [5]. The RG method is subsequently used to generate an effective field theory for the adiabatic coupling of JW fermions with soft lattice degrees of freedom. Fluctuation effects are then treated by the standard functional-integral method, and the results for $\chi(T)$ are shown to give a controlled description of precursor effects in the SP organic compound $(\mathrm{BCPTTF})_2\mathrm{AsF}_6$. The formalism applies equally well to the Peierls instability in which case the results reduce to those of Lee *et al.* [6] in the absence of electron-electron correlations, setting in turn these previous results within a more formal framework.

Let us consider a 1D array of N spins decribed by the 1D Heisenberg-Ising Hamiltonian $H = \sum_{i,j} J_{ij}(S_i^x S_j^x + S_i^y S_j^y) + J_{z,ij} S_i^z S_j^z$, where the transverse (longitudinal) exchange $J_{(z),ij} \simeq J_{(z)} + J'_{(z)}(\phi_i - \phi_j)$ between spins is modulated by the longitudinal lattice displacement ϕ. Using the JW fermion representation for spins and considering a linear fermion-lattice coupling, the partition function can then be expressed as a functional integral

$$
\begin{aligned}
Z &= \int\int \mathcal{D}\psi^*\mathcal{D}\psi\mathcal{D}\phi\, e^{S_0^e[\psi^*,\psi]+S_0[\phi]+S_\lambda[\psi^*,\psi,\phi]+S_I^e[\psi^*,\psi]} \\
&= \int\int \mathcal{D}\psi^*\mathcal{D}\psi\mathcal{D}\phi\, \exp\Bigg\{\sum_{\tilde{k}}[G^0(\tilde{k})]^{-1}\psi^*(\tilde{k})\psi(\tilde{k}) + \sum_{\tilde{q}}[D^0(\tilde{q})]^{-1}|\phi(\tilde{q})|^2 \\
&\qquad - (T/N)^{1/2}\sum_{\tilde{k},\tilde{q}}\lambda(k,q)\psi^*(\tilde{k}-\tilde{q})\psi(\tilde{k})\phi(\tilde{q}) \\
&\qquad - T/N\sum_{\{p\tilde{q}\}} g(q)\psi^*(\tilde{k}_1+\tilde{q})\psi^*(\tilde{k}_2-\tilde{q})\psi(\tilde{k}_2)\psi(\tilde{k}_1)\Bigg\}
\end{aligned}
\tag{1}
$$

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over Grassman (c-number) $\psi(\phi)$ variables. For the free fermion (S_0^e) and lattice ($S_0[\phi]$) parts of the total action S, $G^0(\tilde{k}) = [i\omega_n - \epsilon(k)]^{-1}$ and $D^0(\tilde{q}) = -M/2[\omega_m^2 + \omega^2(q)]^{-1}$ are the bare fermion and the lattice field propagators with $\tilde{k} = (k, \omega_n = (2n+1)\pi T)$, $\tilde{q} = (q, \omega_m = 2\pi m T)$. Their spectrums are given by $\epsilon(k) = -2J\cos k$ and $\omega(q) = \omega_0|\sin(q/2)|$, respectively, $\omega_0 = 2\sqrt{K/M}$ being the frequency of the lattice mode at $2k_F = \pi$, with K and M the elastic constant and the ionic mass. In the interacting term, S_λ describes the linear coupling between fermions and the lattice field via $\lambda(k,q) = 4iJ'\cos(k + q/2)\sin(q/2)$. Finally, in S_I, fermions on nearest-neighbor lattice sites interact through $g(q) = 2J_z\cos q$.

The problem of low-energy spin degrees of freedom then reduces to the study of interacting lattice JW fermions coupled to the fluctuating field ϕ. Extending a previous RG approach [7], this can be achieved by first integrating high momentum fermion degrees of freedom retaining transients due to fermions on a lattice. We write $\psi^{(*)} \to \psi^{(*)} + \overline{\psi}^{(*)}$ where the $\overline{\psi}^{(*)}$'s describe degrees of freedom to be integrated over in the outer momentum shell of thickness $\frac{1}{2}k_0(\ell)d\ell$ on both sides of the Fermi level at $k_F = \pm\pi/2$ and for all ω_n. Here $k_0(\ell) = k_0 e^{-\ell}$ is the momentum cutoff at the step ℓ and $k_0 = \pi/2$. Keeping the ϕ's fixed, this is formally written as

$$
\begin{aligned}
Z &\propto \int\int_< \mathcal{D}\psi^*\mathcal{D}\psi\mathcal{D}\phi\, e^{S[\psi^*,\psi,\phi]_\ell + \beta\mathcal{F}[\phi]_\ell} \int\int_{\text{o.s.}} \mathcal{D}\overline{\psi}^*\mathcal{D}\overline{\psi}\, e^{S_0^e[\overline{\psi}^*\overline{\psi}]}\left(e^{\overline{S}_\lambda[\overline{\psi}^*,\overline{\psi},\psi^*,\psi,\phi] + \overline{S}_I[\overline{\psi}^*,\overline{\psi},\psi^*,\psi]}\right) \\
&= \int\int_< \mathcal{D}\psi^*\mathcal{D}\psi\mathcal{D}\phi\, e^{S[\psi^*,\psi,\phi]_\ell + \beta\mathcal{F}[\phi]_\ell} \exp\left(\sum_n \frac{1}{n!}\langle(\overline{S}_\lambda + \overline{S}_I)^n\rangle_{\text{o.s.}}\right) \\
&\propto \int\int_< \mathcal{D}\psi^*\mathcal{D}\psi\mathcal{D}\phi\, e^{S[\psi^*,\psi,\phi]_{\ell+d\ell} + \beta\mathcal{F}[\phi]_{\ell+d\ell}},
\end{aligned}
\tag{2}
$$

where $\mathcal{F}[\phi]_\ell$ is a free energy functional of the lattice field to be discussed later. At the one-loop level, the outer shell averages $\frac{1}{2}\langle\overline{S}_I^2\rangle_{\text{o.s.}}$ and $\langle\overline{S}_\lambda\overline{S}_I\rangle_{\text{o.s.}}$ are performed with respect to the free fermion outer shell part in the infrared singular $2k_F$ electron-hole and Cooper channels. They will lead to the renormalization of the forward $\tilde{g}_f = g_f(\pi v)^{-1}$ and umklapp scattering $\tilde{g}_u = g_u(\pi v)^{-1}$ for S_I and the $2k_F$ fermion-lattice vertex part $z\lambda(\pm k_F, \mp 2k_F)$, with $v = 2J$ being the Fermi velocity. If one rescales the momentum $k \to k e^{d\ell}$, the energy, the fields, and the coupling constants transform according to $(\epsilon, \omega_n) \to \zeta(d\ell)(\epsilon, \omega_n)$, $\psi \to \zeta^{-1/2}(d\ell)\psi$, $g_f \to \zeta(d\ell)g_f e^{-d\ell}$, and $g_u \to \zeta(d\ell)g_u e^{-3d\ell}$, respectively. Owing to the curvature of the band the rescaling becomes dependent of ℓ, namely, $\zeta(d\ell) = 1 + \cot(k_0e^{-\ell})k_0e^{-\ell}d\ell$. At the one-loop level the RG flow is then governed by

$$
\begin{aligned}
\frac{d\tilde{g}_f}{d\ell} &= -\tilde{g}_f[1 - \alpha(\ell)] + 4\tilde{g}_u^2 K(\ell), \\
\frac{d\tilde{g}_u}{d\ell} &= -\tilde{g}_u[3 - \alpha(\ell)] + 2\tilde{g}_f\tilde{g}_u K(\ell), \\
\frac{d\ln z}{d\ell} &= \frac{1}{2}(\tilde{g}_f - \tilde{g}_u)K(\ell), \\
\frac{dv}{d\ell} &= 4J^z\pi^{-1}\tanh[\beta J(\ell)]\sin(k_0e^{-\ell})k_0e^{-\ell},
\end{aligned}
\tag{3}
$$

where $\alpha(\ell) = \cot(k_0e^{-\ell})k_0e^{-\ell}$, $K(\ell) = \tanh[\beta J(\ell) \times \sin(k_0e^{-\ell})][1 - \sin^2(k_0e^{-\ell})]^{-1/2}$ with the initial conditions $g_f = -2g_u = 4J^z$ at $\ell = 0$. When $\ell \gg 1$, thermal and lattice transients become negligible and the flow equations for $\tilde{g}_f$ and $\tilde{g}_u$ reduce to those obtained by Black and Emery for the Heisenberg-Ising part of the model in the continuum limit [8]. For the antiferromagnetic case with $J_z/J \leq 1$, the model predicts that $g_u(\ell)$ scales to a nonuniversal value. In one loop the self-energy contribution $\langle\overline{S}_I\rangle_{\text{o.s.}}$ introduces a renormalization of the Fermi velocity $v(\ell) = 2J(\ell)$. In the low temperature limit ($\beta \to \infty$), this reduces to the Hartree-Fock result $v^* = 2J + 4J_z/\pi$ when $\ell \to \infty$, which is close in leading order to the $T = 0$ Heisenberg-Ising exact result [9]. At this point, it is important to establish the accuracy of the above RG procedure from a calculation of $\chi(T)$ for the antiferromagnetic chain. Since a magnetic field h along the z direction acts as a chemical potential, the evaluation of $\chi(T)$ amounts to the calculation of compressibility of JW fermions, namely, $\chi(T) = d\langle N\rangle/dh$. At a given temperature T, the partial RG integration is then conducted down to $\ell_T = \ln[1/\arcsin(T/2J)]$ so that the remaining degrees of freedom to be integrated out are those located inside a thermal width $2T$ around the Fermi level. These give the essential contribution to $\chi(T)$ and, as incoherent states in the nonsingular particle-hole channel, they can be treated by perturbation theory. Thus treating S_I at ℓ_T in RPA, one readily finds

$$
\chi(T) = \frac{\chi_0(T)}{1 + \frac{1}{2}g_f(T)\chi_0(T)}, \tag{4}
$$

where $g_f(T)$ is given by the solution of (3) at ℓ_T and $\chi_0(T) = \int_0^T d\epsilon\, D(\epsilon, T)[2T\cosh^2(\epsilon/2T)]^{-1}$ is the bare compressibility with $D(\epsilon, T) = [2\pi v(T) \times \sqrt{1 - \epsilon^2/4J^2(T)}]^{-1}$ as the effective tight-binding density of states. The resulting temperature profile for $\chi(T)$ is portrayed in Fig. 1(a) for the isotropic case $J = J_z$. From the figure, the above one-loop RG result reproduces the position of the maximum,

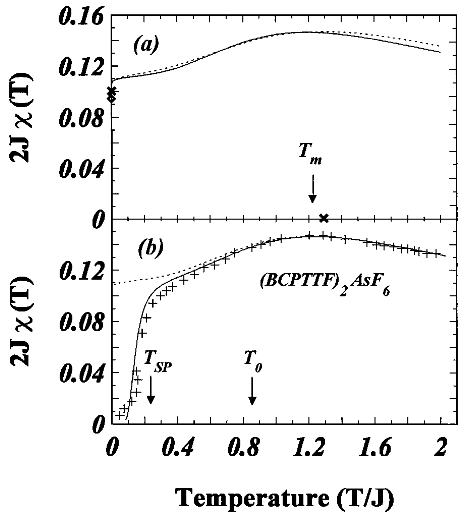

FIG. 1. Magnetic susceptibility vs temperature. (a) Comparison of our results (continuous line and diamond) with those of Ref. [5] (dashed line and crosses) for the AF chain; (b) comparison of our results in the presence (continuous line) and absence (dashed line) of SP fluctuations with experimental data for (BCPTTF)$_2$AsF$_6$ (crosses).

$\chi(T_m) \approx 0.72/J$ at $T_m \simeq 1.22J$, the infinite slope approach to the $\chi(T = 0) = (2\pi v^* + g_f^*/2)^{-1}$ value [due to g_u-induced transients on $g_f(T)$] and the overall temperature dependence of $\chi(T)$ with a very good accuracy compared to the combination of the thermodynamic Bethe ansatz [5] and the Bonner-Fisher [10] numerical results at high temperature and conformal field theory at low temperature [5]. The discrepancy with respect to the Bethe ansatz and Bonner-Fisher calculations for $T > 1.2J$ has been reduced by the inclusion of one-loop mode-mode coupling effects. The latters result from the curvature of the band and gain in importance only in the high temperature domain.

In order to see how lattice fluctuations modify the above picture, one can first look at the closed fermion loops $\langle \overline{S}_\lambda^n \rangle_{\text{o.s.}}/n!$ in $n \geq 2$ powers of the ϕ's and which are generated by the partial integration (2). This will not only give rise to corrections for $S_0[\phi]_\ell$ at $n = 2$, but when combined to the $n > 2$ terms it yields and infinite series in powers of ϕ which is nothing but the quantum Landau-Ginzburg free energy expansion $\mathcal{F}[\phi]_\ell$ of the lattice field ϕ at ℓ [7]. By rescaling the field $\phi \to \lambda\phi$ in the adiabatic limit, one can construct up to the quartic mode-mode coupling term, the following Landau-Ginzburg functional near $2k_F$:

$$\mathcal{F}[\phi]_\ell = \sum_q [a(\ell) + c(\ell)(q - 2k_F)^2] |\phi(q)|^2 + N^{-1} b(\ell) \sum_{\{q\}} \phi(q_1) \cdots \phi(q_4) \delta_{\Sigma_{q_i = \pm 4k_F}}, \quad (5)$$

where the low-lying collective character of the fluctuations has allowed us to take the static limit. This is a generic form of the free energy for the transfer matrix analysis of fluctuations for a one-component order parameter in 1D [11]. As a rule the flow of the SP Ginzburg-Landau parameters $a(\ell)$, $b(\ell)$, and $c(\ell)$ is conducted down to the neighborhood of a characteristic (mean-field) temperature scale T_0 ($\ell_{T_0} = \ln[1/\arcsin(T_0/2J)]$) marking the onset of strong SP fluctuations. Explicitly, assuming a power law form $z(\ell) \approx [E_0(\ell)/E_0]^{-\gamma^*/2}$ for the vertex part at large ℓ where $\gamma^* = \tilde{g}_f^* - \tilde{g}_u^* \simeq 0.72$ is the exponent taken at the fixed point, one finds

$$\begin{aligned} a(T) &\simeq a'(T/T_0 - 1), \\ c(T_0) &\simeq a' 7\zeta(3) v^*/16\pi^2 T_0^2, \\ b(T_0) &\simeq 7\zeta(3)/(32\pi^2 v^{*2}) [2\pi v^*(2 + 2\gamma^*)]^{-1} \\ &\quad \times (2v^*/T_0)^{2+2\gamma^*} \end{aligned} \quad (6)$$

for $2J \gg T_0$ where $a' = z^2(T_0)(2\pi v^*)^{-1}$ and $\zeta(3) = 1.20\ldots$. Here, the power law expression for the mean-field temperature $T_0 \simeq v^*[\tilde{g}_\lambda/(\gamma^* + \tilde{g}_\lambda)]^{1/\gamma^*}$ with $\tilde{g}_\lambda = |\lambda(2k_F)|^2(4\pi K v^*)^{-1}$ agrees to leading order with previous mean-field results [12]. According to (2), our effective low-energy theory then describes the coupling of the remaining thermal fermion states around the Fermi level to the static fluctuations of the SP field governed by (5). This can be seen as the SP analog of the analysis made by Lee *et al.* [6] for the Peierls instability in the absence of electronic correlation effects. In the present approach, however, fermion degrees of freedom being not entirely integrated out, the interplay between lattice fluctuations and fermions at ℓ_T can be formally incorporated. The contribution of SP fluctuations to the fermion self-energy Σ of the propagator $G = ([G^0]^{-1} - \Sigma)^{-1}$ at ℓ_T being essentially static, one has in leading order

$$\Sigma(\tilde{k}, \{\phi\}) = -TN^{-1}z^2(T_0) \times \sum_q G^0(k \pm 2k_F - q, i\omega_n)\chi_\phi(2k_F + q), \quad (7)$$

where $\chi_\phi(2k_F + q) = 2\xi(T)\langle|\Phi|^2\rangle[1 + \xi^2(T)q^2]^{-1}$ is the Fourier transform of the correlation function of the SP field with $\langle|\Phi|^2\rangle = z^2(T_0)\langle|\phi(2k_F)|^2\rangle$. Here $\langle|\phi(2k_F)|^2\rangle$ and $\xi(T)$ are the amplitude of mean-square fluctuations and the correlation length, respectively. Both can be calculated exactly from the functional integration over the ϕ's described by (5) [11]. As T_0 is approached from above, fluctuations become large and $\xi(T)$ increases rapidly and marks the formation of a pseudogap in the fermion density of states. Actually, it follows that a self-consistent use of the dressed propagator G below T_0 will considerably slow down the RG flow of parameters given in (3) as well as the closed fermion loops of $\mathcal{F}[\phi]$ [consistently with the sharp cutoff procedure used in (6)]. The pseudogap also affects the particle-hole compressibility bubble $\chi_0(T)$. Following Lee *et al.* [6], one substitutes in leading order the dressed G for one of the propagators entering in the formal expression for χ_0, namely, $TN^{-1}\sum_{\tilde{k}} GG^0$, and one readily finds

$$\chi_0(T) = \int_0^T d\epsilon\, \overline{D}(\epsilon, T)\,[2T\cosh^2(\epsilon/2T)]^{-1}, \qquad (8)$$

where

$$\overline{D}(\epsilon, T) = D(\epsilon, T)\frac{\alpha[2(y + x)]^{1/2}}{[2(y + x) - \alpha^2]y} \qquad (9)$$

is the density of states in the presence of a pseudogap. Following Ref. [6], we have $\alpha = v^*\xi^{-1}(T)\langle|\Phi|^2\rangle^{1/2}$, $x = 1 + \frac{1}{4}\alpha^2 - \overline{\epsilon}^2$, and $y = (x^2 + \overline{\epsilon}^2\alpha^2)^{1/2}$, with $\overline{\epsilon} = \epsilon/\langle|\Phi|^2\rangle^{1/2}$. Since $\chi_0(T)$ goes essentially like $\xi^{-1}(T)$ at low temperature, the results of the transfer matrix method show that the fluctuation-induced depression of $\chi(T)$ evolves towards a thermally activated behavior of the form $\chi_0(T) \sim e^{-\rho T_0/T}$ below $T^* \simeq T_0/3$ with $\rho \simeq 0.92$ using the above Ginzburg-Landau parameters [11]. It is worth pointing out here that T^* marks the temperature scale for true long range order when a small but finite interchain coupling is taken into account.

We are in a position to apply the above results to experimental findings for the organic SP compound $(BCPTTF)_2AsF_6$ previously investigated [2]. The thermal dependence of the spin susceptibility has been obtained from ESR measurements [crosses, Fig. 1(b)]. $\chi(T)$ behaves as predicted for an antiferromagnetic chain (Fig. 1) above 120 K or so. X-ray measurements combining photographic and diffractometric methods reveal the presence of superlattice reflections at the reduced wave vector $\mathbf{q}_{SP} = (1/2, 1/2, 1/2)$ below $T_{SP} = 32.5$ K. The chain dimerization ("$2k_F$" $= 1/2$) occurs at the same temperature at which an anomaly is observed in the drop of $\chi(T)$. Above T_{SP} quite anisotropic structural fluctuations are detected. They exhibit no interchain correlations above 60 K. The fluctuations have been measured until about $T_0 = 120$ K, the temperature at which the correlation length (divided by lattice constant 7.16 Å) along the chain $\xi \simeq 1.12$ grows away from the interspin distance. Figure 2 shows the inverse of the thermal dependence of the x-ray diffuse scattering intensity I (corrected by the thermal population factor) and of ξ^{-1}. Both I and $\xi(T)$ increase rapidly below T_0, whereas I^{-1} and ξ^{-1} vanish at T_{SP} showing the second order nature of the SP transition of $(BCPTTF)_2AsF_6$.

From the position of the maximum of the measured $\chi(T)$ [2] around 168 K, Fig. 1(a) first gives $J \simeq 140$ K for the purely Heisenberg part of the model. The subsequent identification of T_0 with the x-ray temperature scale for strong SP lattice fluctuations (Fig. 2), namely, $T_0 \simeq 120$ K, allows us to fix all the SP parameters in (6) and calculate all quantities of interest. As one can see from Fig. 2, the transfer matrix result for $\xi(T)$ gives a good description of the correlation effect down to the critical domain around $T^* \simeq 40$ K. Correspondingly, the calculated $\chi(T)$ reported in Fig. 1(b) is slightly more depressed when entering in the critical domain. Straightforward but lengthy calculations performed in the presence of interchain coupling (e.g., via three-dimensional phonons) can be shown to improve the accuracy in the critical domain close to T_{SP} but lead to a similar activated behavior well below T_{SP}. Quite similar results have been obtained for the $(BCPTTF)_2PF_6$ analog with $J \simeq 175$ K, $T_0 \simeq 100$ K, and $T_{SP} = 37$ K [2]. The application to existing data for the cuprate compound $CuGeO_3$ [3] is straightforward if one includes second-nearest-neighbor exchange to the spin part of the model, which is known to be relevant for this system [13]. A more detailed account of the present work will be given elsewhere.

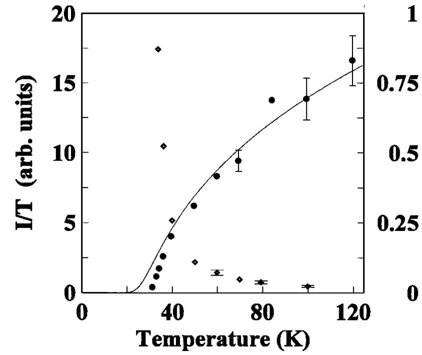

FIG. 2. Measured x-ray diffuse scattering intensity I/T (diamonds) and inverse longitudinal correlation length times the interspin distance ξ^{-1} (dots) vs temperature for $(BCPTTF)_2AsF_6$. The continuous line for ξ^{-1} corresponds to the 1D results of the transfer matrix method.

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