

Classical phase fluctuations in incommensurate Peierls chains

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In the pseudogap regime of one-dimensional incommensurate Peierls systems, fluctuations of the phase of the order parameter prohibit the emergence of long-range order and generate a finite correlation length. For classical phase fluctuations, we present exact results for the average electronic density of states, the mean localization length, the electronic specific heat, and the spin susceptibility at low temperatures. Our results for the susceptibility give a good fit to experimental data.

Continuous symmetries in one-dimensional electronic systems are not spontaneously broken at any finite temperature T . The mean-field prediction of a finite critical temperature T_c^{MF} is incorrect in this case. The experimentally observed Peierls transition at a finite temperature $T_c^{\text{3D}} \ll T_c^{\text{MF}}$ in many quasi-one-dimensional conductors is due to weak interchain coupling which triggers a crossover to three dimensionality. In the intermediate temperature regime $T_c^{\text{3D}} \leq T \ll T_c^{\text{MF}}$, the physical properties of Peierls chains are dominated by one-dimensional order-parameter fluctuations.¹ This is the so-called pseudogap regime where mean-field theory is not even qualitatively correct.

In this work, we shall present an exact solution of an effective model for the low-temperature thermodynamics of incommensurate Peierls chains. For incommensurate chains, the order parameter $\Delta(x)$ is complex, so that in the pseudogap regime the generalized Ginzburg-Landau potential has the form of a ‘‘Mexican hat.’’¹ It is then a good approximation to ignore amplitude fluctuations of $\Delta(x) = |\Delta(x)|e^{i\vartheta(x)}$ and focus on the gapless fluctuations of the phase $\vartheta(x)$. At long wavelengths and low energies, the thermodynamics of phase fluctuations can be described by a classical Hamiltonian which is formally identical with the kinetic energy of a superfluid¹

$$H_{\vartheta} = \frac{1}{2} m^* n_s \int_0^L dx v_s^2(x), \quad v_s(x) = \frac{\partial_x \vartheta(x)}{2m^*}. \quad (1)$$

Here, m^* is the effective mass of the electrons, L is the length of the chain, and the one-dimensional density n_s measures the stiffness of the system with respect to long-wavelength distortions of the phase of the order parameter.² A two-dimensional analog of Eq. (1) has been used by Emery and Kivelson³ to explain the pseudogap behavior of underdoped high-temperature superconductors. In one dimension, the problem is much simpler, because there is no Kosterlitz-Thouless transition and the thermodynamics of the phase variable is trivial. However, the calculation of the electronic properties amounts to solving a one-dimensional random problem with colored noise. Usually, problems of this type cannot be solved exactly.⁴ At low energies, the electronic degrees of freedom can be described by the Hamiltonian of the so-called fluctuating gap model^{5,6} (FGM)

$$H_{\text{el}} = -iv_F \partial_x \sigma_3 + \Delta(x) \sigma_+ + \Delta^*(x) \sigma_-, \quad (2)$$

where v_F is the Fermi velocity, σ_i are the usual Pauli matrices, with $\sigma_{\pm} = \frac{1}{2}(\sigma_1 \pm i\sigma_2)$. In the pseudogap regime, the gapped amplitude fluctuations are frozen out, so that we may set $\Delta(x) = \Delta_s e^{i\vartheta(x)}$, where Δ_s is determined by the local minimum of the generalized Ginzburg-Landau functional. From Eq. (1), it is then easy to show that

$$\langle \Delta(x) \Delta^*(x') \rangle = \Delta_s^2 e^{-|x-x'|/\xi}, \quad (3)$$

where $\langle \dots \rangle$ denotes the thermodynamic average with respect to the Hamiltonian H_{ϑ} given in Eq. (1), and the order-parameter correlation length is $\xi = n_s / (2m^*T)$. In this work, we calculate the average electronic density of states (DOS) $\rho(\omega) = \langle \text{Tr} \delta(\omega - H_{\text{el}}) \rangle$ of the model defined via Eqs. (1)–(3) exactly for arbitrary ξ .

Previously, the DOS of the FGM has been calculated assuming a Gaussian distribution of $\Delta(x)$ with covariance given by Eq. (3).^{5,7–9} Although in this case the problem is not exactly solvable,^{8,9} a sophisticated algorithm has been developed⁷ which produces an expression for $\rho(\omega)$ which is reasonably close to the exact numerical result for Gaussian disorder.⁹ However, as explained above, the assumption of a Gaussian distribution of $\Delta(x)$ centered at $\Delta = 0$ is rather unphysical in the pseudogap regime. It is therefore not surprising that in this regime the true behavior of $\rho(\omega)$ (to be discussed below) is very different from the DOS for Gaussian disorder.

The electronic contribution to the thermodynamic properties of our system including the coupling to the phase fluctuations can be obtained from the disorder-averaged free energy

$$F = -T \int_{-\infty}^{\infty} d\omega \int_0^L dx \langle \rho(x, \omega) \rangle \ln(1 + e^{-\omega/T}). \quad (4)$$

The local DOS $\rho(x, \omega)$ can be expressed as $\rho(x, \omega) = -\pi^{-1} \text{Im Tr } \mathcal{G}(x, x, \omega + i0^+)$, where the Green function $\mathcal{G}(x, x', \omega + i0^+)$ satisfies

$$[\omega - H_{\text{el}}] \mathcal{G}(x, x', \omega + i0^+) = \delta(x - x') \sigma_0. \quad (5)$$

Here, σ_0 is the 2×2 unit matrix. For periodic boundary conditions, the average $\langle \rho(x, \omega) \rangle$ is independent of x and can be identified with the average DOS $\rho(\omega)$.

Following Ref. 10, we eliminate the phase of the order parameter $\Delta(x) = \Delta_s e^{i\vartheta(x)}$ via a gauge transformation,

$$\mathcal{G}(x, x', \omega) = e^{i/2\sigma_3 \vartheta(x)} \tilde{\mathcal{G}}(x, x', \omega) e^{-i/2\sigma_3 \vartheta(x')}. \quad (6)$$

The transformed Green function $\tilde{\mathcal{G}}$ satisfies an equation of the form (5), but with H_{el} replaced by

$$\tilde{H}_{\text{el}} = -iv_F \partial_x \sigma_3 + V(x) \sigma_0 + \Delta_s \sigma_1, \quad (7)$$

where $V(x) = (v_F/2) \partial_x \vartheta(x)$. Equation (6) is a chiral transformation that eliminates the phase of $\Delta(x)$ in favor of a forward scattering random potential $V(x)$. The local DOS is invariant under this transformation, so that we may replace $\mathcal{G} \rightarrow \tilde{\mathcal{G}}$ in all expressions involving the DOS. The crucial point is now that with H_{ϑ} given by Eq. (1), the probability distribution of $V(x)$ is determined by Gaussian white noise,¹⁰ with zero average and covariance $\langle V(x)V(x') \rangle = v_F^2 (2\xi)^{-1} \delta(x - x')$. This enables us to map well-known results valid in the white-noise regime^{6,11,12} for the integrated Green function $\Gamma(\omega)$ defined via $\partial_\omega \Gamma(\omega) = \text{Tr} \langle \mathcal{G}(x, x, \omega + i0^+) \rangle$ onto results for the problem with a finite correlation length. We find

$$\Gamma(\omega) \equiv l^{-1}(\omega) - i\pi \mathcal{N}(\omega) = \pi \rho_0 \Delta_s I'_{-i\nu}(g) / I_{-i\nu}(g), \quad (8)$$

where $\rho_0 = (\pi v_F)^{-1}$ is the DOS for $\Delta = 0$, and $I_{i\nu}(g)$ is a modified Bessel function with imaginary index $i\nu$. The dimensionless parameters g and ν are

$$g = \frac{4\Delta_s \xi}{v_F} = \frac{2n_s}{m^* v_F} \frac{\Delta_s}{T}, \quad \nu = \frac{4\omega \xi}{v_F} = g \frac{\omega}{\Delta_s}. \quad (9)$$

The imaginary part of $\Gamma(\omega)$ is proportional to the integrated average DOS $\mathcal{N}(\omega)$ which satisfies $\partial_\omega \mathcal{N}(\omega) = \rho(\omega)$ while, according to Thouless,¹³ the real part of $\Gamma(\omega)$ can be identified with the inverse mean localization length $l^{-1}(\omega)$, i.e., the Lyapunov exponent.⁶ Using a Wronski relation for $I_{-i\nu}(g)$,¹⁴ one gets

$$\mathcal{N}(\omega) = \frac{\rho_0 v_F}{4\pi \xi} \frac{\sinh(\pi \nu)}{|I_{i\nu}(g)|^2}. \quad (10)$$

The inverse mean localization length can be written as

$$l^{-1}(\omega) = \frac{\Delta_s}{v_F} \frac{\partial}{\partial g} \ln |I_{i\nu}(g)|. \quad (11)$$

Recall that Eqs. (8), (10), and (11) determine the DOS and inverse localization length for phase fluctuations with arbitrary correlation lengths ξ and were derived by a reinterpretation of parameters due to a gauge transformation¹⁰ of known results in the white-noise limit $\xi \rightarrow 0$.¹¹

We now discuss the behavior of the average DOS. Because $\rho(\omega)$ is an even function of ω , we restrict ourselves to $\omega \geq 0$. Using¹⁴ $|I_{i\nu}(0)|^2 = (\pi \nu)^{-1} \sinh(\pi \nu)$, one easily verifies that $\mathcal{N}(\omega) \sim \rho_0 \omega$ for $g \rightarrow 0$, so that in this limit we recover the result for free electrons with linearized energy dispersion. While for small g , the leading corrections can be calculated perturbatively in powers of g , in the pseudogap regime $g \gg 1$, the behavior of the average DOS is quite complicated. It is convenient to measure frequencies in units of Δ_s and to express Eq. (10) in terms of the Bessel function $J_{i\nu}(ig)$ with imaginary index and argument, using $I_{i\nu}(g) = e^{\nu\pi/2} J_{i\nu}(ig)$.¹⁴ Defining $\bar{\omega} = \omega/\Delta_s = \nu/g$, we may write

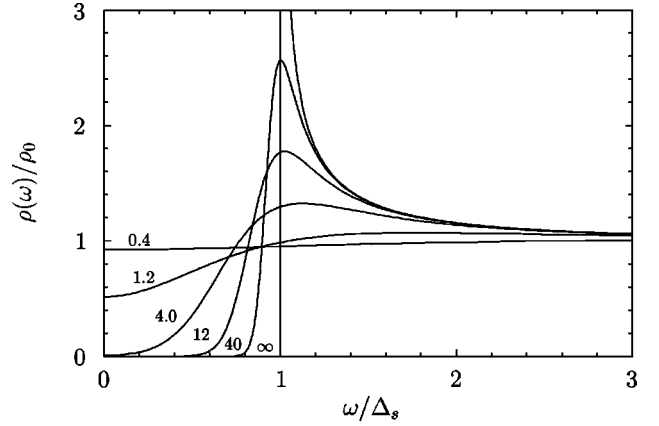


FIG. 1. Frequency dependence of the DOS given in Eq. (12) for $g = 4\Delta_s \xi / v_F = 0.4, 1.2, 4.0, 12, 40$, and ∞ .

$$\rho(\omega) = \frac{\rho_0}{2\pi g} \frac{\partial}{\partial \bar{\omega}} \frac{1 - e^{-2\pi g \bar{\omega}}}{|J_{ig\bar{\omega}}(ig)|^2}. \quad (12)$$

In Fig. 1, we show a graph of Eq. (12) for several values of g . For a more quantitative analysis, we use the uniform asymptotic expansion of $J_{ig\bar{\omega}}(ig)$ for large g and fixed $\bar{\omega}$,¹⁴ which reveals three different regimes: First of all, for $1 - \bar{\omega} \gtrsim g^{-2/3}$ (i.e. for frequencies sufficiently far below Δ_s), the average DOS in the pseudogap regime $g \gg 1$ can be approximated by

$$\rho(\omega)/\rho_0 \approx 2g(1 - \bar{\omega}^2)^{1/2} \exp[-2gQ(\bar{\omega})] \times [1 + e^{-2\pi g \bar{\omega}}] \arccos(\bar{\omega}), \quad (13)$$

where $Q(\bar{\omega}) = (1 - \bar{\omega}^2)^{1/2} - \bar{\omega} \arccos(\bar{\omega})$. In particular, for small $\bar{\omega}$, we may expand $Q(\bar{\omega}) \approx 1 - \pi/2 \bar{\omega} + \frac{1}{2} \bar{\omega}^2$, so that

$$\rho(\omega)/\rho_0 \approx 2\pi g e^{-2g} \cosh[\pi g \bar{\omega}] e^{-g\bar{\omega}^2}, \quad g\bar{\omega}^3 \ll 1. \quad (14)$$

Hence, for $\omega = 0$, the DOS is exponentially small, $\rho(0)/\rho_0 \sim 2\pi g e^{-2g}$. As shown in Fig. 2, such a strong suppression

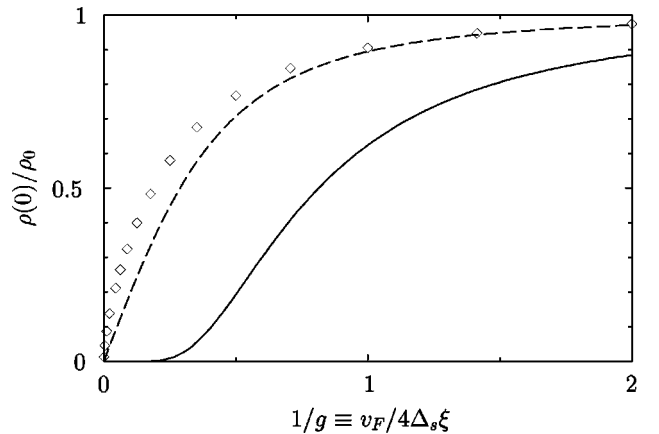


FIG. 2. The solid line is a graph of the DOS $\rho(0)$ at the Fermi energy for classical phase fluctuations as a function of $1/g \equiv v_F/4\Delta_s \xi$. For a comparison, the dashed line shows the result found in the leading-order Born approximation (Ref. 5) and the diamonds give the DOS for Gaussian statistics (Refs. 9 and 12).

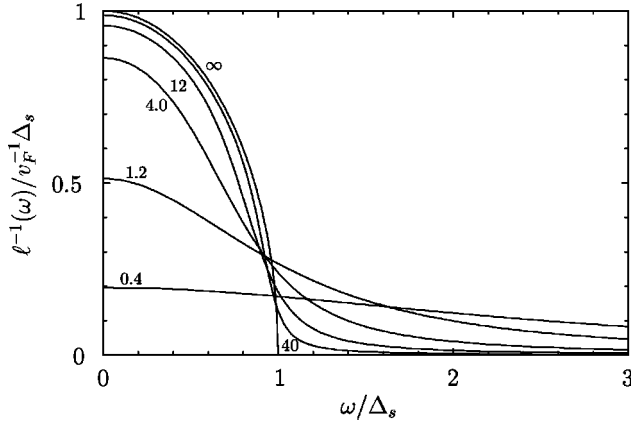


FIG. 3. Graph of the inverse localization length for $g = 4\Delta_s\xi/v_F = 0.4, 1.2, 4.0, 12, 40,$ and ∞ .

of the DOS at the Fermi energy is a unique feature for classical phase fluctuations, which is neither reproduced within the Born approximation⁵ [which predicts $\rho(0) \propto g^{-1}$], nor for Gaussian disorder⁹ [where $\rho(0) \propto g^{-\mu}$, with $\mu \approx 0.64$]. The approximation (13) breaks down when $1 - \bar{\omega}$ becomes comparable with $g^{-2/3}$. Note that $Q(1 - \epsilon) \sim (2^{3/2}/3)\epsilon^{3/2}$ for $\epsilon \ll 1$, so that $gQ(\bar{\omega}) = O(1)$ when Eq. (13) ceases to be valid. In this case, we have to go back to our exact result (12) which implies for $|\bar{\omega} - 1| \leq g^{-2/3} \ll 1$

$$\rho(\omega)/\rho_0 \approx a_1 g^{1/3} [1 - a_2 g^{4/3} (\bar{\omega} - 1)^2]. \quad (15)$$

Here, $a_1 = 2^{-4/3} \pi^{-1} c_2/c_1^3 \approx 0.7306$ and $a_2 = 2^{2/3} [3(c_2/c_1)^2 - c_1/c_2] \approx 0.3534$, with $c_1 = \text{Ai}(0) = [3^{2/3} \Gamma(2/3)]^{-1}$ and $c_2 = -\text{Ai}'(0) = [3^{1/3} \Gamma(1/3)]^{-1}$, where $\text{Ai}(x)$ is the Airy function. From Eq. (15), we conclude that, to leading order in $g \gg 1$, the average DOS exhibits a maximum at $\omega = \Delta_s$, with a height that diverges as $g^{1/3} \propto \xi^{1/3} \propto T^{-1/3}$ for $T \rightarrow 0$. Finally, for $\bar{\omega} - 1 \geq g^{-2/3}$, our exact result (12) reduces to the well-known expression for the DOS in the presence of a static gap, $\rho(\omega)/\rho_0 \approx \bar{\omega}/\sqrt{\bar{\omega}^2 - 1}$. At $\bar{\omega} - 1 \approx g^{-2/3}$, this expression smoothly matches with the parabola (15).

In Fig. 3, we show the exact inverse localization length $l^{-1}(\omega)$ given in Eq. (11) for several values of g . For $g \gg 1$, we obtain the following approximations: $l^{-1}(\omega) \approx (\Delta_s/v_F)(1 - \bar{\omega}^2)^{1/2}$ for $1 - \bar{\omega} \geq g^{-2/3}$; for $|\bar{\omega} - 1| \leq g^{-2/3}$ we find $l^{-1}(\omega) \approx (\Delta_s/v_F)[a_3 g^{-1/3} - (3g)^{-1}]$, with $a_3 = 2^{-2/3} c_2/c_1 \approx 0.4592$; finally, for $\bar{\omega} - 1 \geq g^{-2/3}$ the leading behavior is $l^{-1}(\omega) \approx (\Delta_s/v_F)[2g(\bar{\omega}^2 - 1)]^{-1}$.

Let us now consider the free energy F defined in Eq. (4) which describes the electronic system coupled to phase fluctuations. For the FGM with a linearized energy dispersion, Eq. (4) is ultraviolet divergent, because then the DOS approaches a constant for $|\omega| \rightarrow \infty$. However, physical quantities involve derivatives of F , which at low temperatures depend only on the low-energy part of the spectrum and are finite. For convenience, we regularize Eq. (4) by subtracting from F the free energy $F^{\xi=\infty}$ for an infinite correlation length, where the gap is static. After an integration by parts, we express the integral in Eq. (4) in terms of a fermionic Matsubara sum and obtain

$$F - F^{\xi=\infty} = \frac{2L\Delta_s T}{v_F} \sum_{n=0}^{\infty} \left[\sqrt{1 + \bar{\omega}_n^2} - \frac{I'_{g\bar{\omega}_n}(g)}{I_{g\bar{\omega}_n}(g)} \right], \quad (16)$$

where $\bar{\omega}_n = (2n + 1)\pi T/\Delta_s$. For large $\bar{\omega}_n$, the term in the square bracket vanishes as $\bar{\omega}_n^{-2}$, so that the sum converges. In the pseudogap regime $g \gg 1$, we may use the uniform asymptotic expansion of $I_{g\bar{\omega}_n}(g)$ for large g (Ref. 14) to obtain an expansion of Eq. (16) in powers of $g^{-1} \propto \xi^{-1} \propto T$. For $T \ll \Delta_s$ the leading terms are

$$F - F^{\xi=\infty} = \frac{L}{16\xi} \left[\Delta_s - \frac{v_F}{12\pi\xi} + O(\xi^{-2}) \right]. \quad (17)$$

The physical interpretation of this result is simple: Because ξ is roughly the size of domains where the order parameter is spatially constant, the prefactor L/ξ in Eq. (17) can be identified with the number of locally ordered domains in a system of size L . At distances of the order of ξ , the phase fluctuations distort the order parameter, which leads to an increase of the energy. In the limit $\Delta_s \xi/v_F \rightarrow \infty$, the energy scale associated with a twist in the order parameter is set by Δ_s . For finite ξ this energy scale decreases, because the time ξ/v_F it takes for electrons to propagate over the distance ξ is finite. This gives rise to the second term in Eq. (17). We emphasize that our exact result (16) gives the change in the free energy due to phase fluctuations for arbitrary ξ .

The low-temperature behavior of the specific heat $C = -T \partial^2 F / \partial T^2$ can be calculated analytically. Keeping in mind that $\xi = n_s / (2m^* T)$, we see that the leading contribution to C is due to the first correction term (involving the energy v_F/ξ) in Eq. (17),

$$C \sim (\pi^2/24)(n_0/n_s)^2 \rho_0 L T, \quad (18)$$

where $n_0 = m^* v_F / \pi$, and we have used the fact that the contribution from $F^{\xi=\infty}$ is exponentially small due to the static gap. Thus, in the pseudogap regime, the specific heat considered here is linear in T , just as the specific heat for noninteracting electrons in one dimension, $C_{\text{el}}^{(0)} \sim (\pi^2/3)\rho_0 L T$. Note that $C/C_{\text{el}}^{(0)} = \frac{1}{8}(n_0/n_s)^2$ for $T \rightarrow 0$. In general, we expect that n_s/n_0 is a number of the order of unity for $T \ll T_c^{\text{MF}}$,¹⁵ so that $C/C_{\text{el}}^{(0)} = O(1)$. In the same regime, we find from Eq. (14) that

$$\rho(0)/\rho_0 \sim 4 \frac{n_s}{n_0} \frac{\Delta_s}{T} \exp \left[- \frac{4}{\pi} \frac{n_s}{n_0} \frac{\Delta_s}{T} \right],$$

i.e., the average DOS at the Fermi energy is exponentially small (see the dashed line in Fig. 4). It should be recalled that the linear dependence of C on T is due to the T dependence of the correlation length. This term expresses the coupling of electronic degrees of freedom to phase fluctuations. The dynamics of classical phase fluctuations, which were not considered here, should also lead to a contribution of order LT/v_F , so that the total specific heat is expected to be of order $C_{\text{el}}^{(0)}$.

Given $\rho(\omega)$, we may also calculate the spin susceptibility $\chi = T^{-1} \int_0^\infty d\omega \rho(\omega) \cosh^{-2}(\omega/2T)$. A graph of χ as a function of T/T_c^{MF} is shown in Fig. 4 (solid line). The low-temperature behavior can again be calculated analytically. If

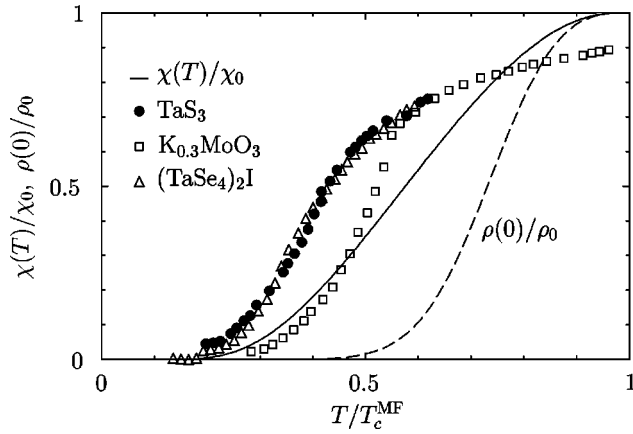


FIG. 4. Graph of the susceptibility $\chi(T)$ calculated for $\xi(T) = n_s(T)/2m^*T$ with $n_s(T)$ given in Ref. 15 and $\Delta_s(T)$ determined by minimizing a generalized Ginzburg-Landau functional. The symbols represent susceptibility data from Ref. 16. The dashed line is the DOS at the Fermi energy.

$g \gg 1$ but $n_s/n_0 < \frac{1}{4}$, we find $\chi \sim \frac{1}{8}\rho(0)$ (assuming now $s = 2$ for spin degeneracy²). On the other hand, for $n_s/n_0 > \frac{1}{4}$ the frequency integral is dominated by a new saddle point at $\omega = \cos r$, where $r = (\pi/8)(n_0/n_s)$. Using Eq. (13), we obtain

$$\chi/\chi_0 \sim 2(2\pi)^{1/2} r^2 (\Delta_r/T)^{3/2} \exp[-\Delta_r/T], \quad (19)$$

where $\Delta_r = (\sin r/r)\Delta_s$ and $\chi_0 = 2\rho_0$ is the susceptibility of free electrons. $n_s/n_0 > \frac{1}{4}$ implies $r < \pi/2$, so that at low temperatures the ratio $\chi/2\rho(0)$ is exponentially large, $\chi/2\rho(0) \propto \exp[(1 - \sin r)\Delta_s/(rT)]$. Our graph of $\chi(T)$ in Fig. 4 is quite

similar to the corresponding graph given by Lee, Rice, and Anderson.⁵ Note, however, that these authors assumed a *real* order parameter and an exponentially large correlation length at low temperatures. Because incommensurate Peierls chains are characterized by a *complex* order parameter and a correlation length that diverges only as a power law, $\xi \propto T^{-1}$, the agreement between the theory of Ref. 5 and experiments for incommensurate chains¹⁶ seems to be accidental. Here, we have shown that the susceptibility data for incommensurate Peierls chains can be explained by a nonperturbative treatment of classical phase fluctuations. Keeping in mind that our model is strictly one-dimensional and ignores amplitude fluctuations (which become important at temperatures of order T_c^{MF}), our theoretical curve for $\chi(T)$ shown in Fig. 4 agrees reasonably well with the susceptibility data.¹⁶

In summary, we have presented exact results for the average DOS, the mean localization length, the susceptibility and the low-temperature thermodynamics of disordered incommensurate Peierls chains in the pseudogap regime, where only phase fluctuations are important. In particular, we have derived the exact frequency dependence of $\rho(\omega)$ which can be measured by means of angular integrated photoemission; we predict that at low temperatures $\rho(\omega)$ exhibits a maximum at $\omega = \Delta_s$, the height of which scales as $T^{-1/3}$. Since this result only applies to the pseudogap regime $T_c^{3D} \leq T \leq T_c^{\text{MF}}$, where $T_c^{3D} \approx \frac{1}{4}T_c^{\text{MF}}$, the scaling $\rho(\Delta_s) \propto T^{-1/3}$ will probably be very difficult to observe experimentally.

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²With the exception of Eq. (19), all equations are given for spinless fermions. Spin degeneracy s can be taken into account by replacing $n_s \rightarrow sn_s$ and multiplying all expressions for F and C by a factor of s .

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