Thermodynamic properties of charge-density waves

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Low-temperature thermodynamic properties of linear-chain compounds exhibiting charge-density waves (CDW) are examined theoretically within a mean-field theory. A result for the spin susceptibility χ is obtained which agrees with the clear-cut available data on $K_{0.3}MoO_3$ for $T<0.9T_P$, where T_P is the Peierls transition temperature. The influence of ordinary impurities on the order parameter Δ, the half-gap Ω_G, and spin susceptibility χ is calculated. Numerical results are obtained for Δ and Ω_G as a function of the impurity concentration x. Substantial difference is found between the lattice distortion parameter Δ and the half-gap Ω_G even for relatively small impurity concentration x, which is directly accessible to experimental verification. Beyond a critical concentration x_c', the excitation spectrum of CDW condensate does not exhibit a gap. The order parameter also yields the transition temperature as a function of x, in agreement with earlier results of Patton and Sham and with recent experiments on TaS_3 doped with Nb and Se impurities. Impurities are found to enhance spin susceptibility. However, the susceptibility at zero temperature remains zero for all concentrations, except in the gapless regime.

I. INTRODUCTION

Inorganic linear-chain compounds which exhibit a Peierls distortion to charge-density-wave (CDW) states have received considerable attention recently. The primary focus has been on the unusual charge transport properties of the sliding CDW condensate such as nonlinear conductivity and narrow-band noise.[1,2] Typical examples of inorganic quasi-one-dimensional (1D) compounds exhibiting charge-density waves are $NbSe_3$, TaS_3, $K_{0.3}MoO_3$, and $(TaSe_4)_2I$. Organic compounds like tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) have also been studied quite extensively; however, the evidences for collective charge transport for these compounds are still controversial.[2,3]

The onset of CDW order has remarkable effects on the thermodynamic properties of quasi-1D conductors as well, since below the Peierls transition temperature T_P a gap opens up in the excitation spectrum. Since the early days of the subject, experimentalists have used thermodynamic measurements to characterize CDW order.[2,3] However, on the theoretical side, sufficient attention has not been given to the calculation of the thermodynamic properties, particularly at low temperatures. The present work is an attempt to narrow this gap between theory and experiments by presenting the calculation of a few thermodynamic properties of the CDW condensate.

Our calculations will be performed within the mean-field theory.[4–7] Strictly one-dimensional systems cannot exhibit any long-range order at any finite temperature, due to the enhanced effect of fluctuations in one dimension.[7] However, even weak coupling between linear chains can lead to long-range correlations below a temperature T_P which is significantly lower than the transition temperature predicted by the mean-field theory. Below this temperature fluctuations are not expected to play an important role. For example, it has been shown recently by Johnston[8,9] that in $K_{0.3}MoO_3$ fluctuations effects die out below $T=0.9T_P$. Thus, our results should be valid at these low temperatures.

The thermodynamic quantity which is of primary interest to us is the spin susceptibility χ. Previous theoretical studies of the spin susceptibility of the CDW condensate was performed by Lee, Rice, and Anderson.[10] The focus of their work was on the effect of 1D fluctuations and the Landau-Ginzburg expansion provided an adequate basis.[11] Their theory is in excellent agreement with the experimental data in the high-temperature fluctuation regime.[8] In our calculations we use thermal Green's functions (mean field) to arrive at a result for χ, which complements the results of Lee *et al.*[10] at low temperatures. Our result for χ is in agreement with susceptibility data[8] in $K_{0.3}MoO_3$ for $T<0.9T_c$. In addition, this formulation of the susceptibility calculation also allows us readily to include the effects of ordinary (nonmagnetic) impurities.

Impurities are known to have important effects on CDW properties and to cause interesting phenomena such as Josephson-type oscillations in the sliding mode.[12,13] It has been pointed out by several authors that impurities should have a pair-breaking effect on the CDW condensate,[4,14,15] since the potential due to ordinary impurities acts with opposite signs on the two members (electron and hole) of the pair. In this connection, Schuster[15] showed that the effective mean-field Hamiltonian describing the CDW condensate with impurities can be cast into a form analogous to that of the effective Hamiltonian appearing in the Abrikosov-Gor'kov (AG) theory[16–18] of superconductors with dilute magnetic impurities, thus suggesting that impurities should cause effects in CDW similar to those obtained by AG. Following his suggestion we shall study the effects of impurities on some of the properties (order parameter, energy gap, and the spin susceptibility)

of the CDW condensate.

One of the major results of the AG theory[16] of superconductors is that one has to make a distinction between the order parameter Δ and the half-gap Ω_G in the presence of magnetic impurities. In fact AG theory predicts that for concentrations exceeding a critical value, one may have a gapless superconductor. However, the prediction of the theory regarding the difference between the gap and the order parameter can only be indirectly tested, since experimentally the superconducting order parameter cannot be obtained directly. In the case of the CDW condensate the order parameter Δ, which describes the periodic lattice distortion, is directly measurable via neutron, x-ray, or electron scattering.[3] Since the experimental value of the energy gap can also be obtained directly either via optical-edge studies[19] or through tunneling experiments,[20] the predictions of the theory regarding the difference in Δ and Ω_G are easily verified experimentally.

The calculation of the energy gap $2\Omega_G$ is also motivated by the fact that not only do thermodynamic properties depend sensitively on the occurrence of the gap in the energy spectrum but other physical properties can also be affected by the changes in the gap brought about by impurities. The effect on the single-particle charge transport is obvious, but more interestingly, the unattenuated collective-charge transport, as first postulated by Fröhlich,[5] is also very much dependent on having a gap in the excitation spectrum. Therefore, our results for the gap may also bear on the recently observed current-carrying sliding CDW state in the presence of electric fields exceeding the threshold electric field E_T required to overcome the impurity pinning.[2]

The present work on the effects of impurities on the CDW state is also inspired by some recent experiments[21] which clearly show the impurity effects similar to the effects caused by magnetic impurities in superconductors. Last, we point out that explicit calculations of the various physical quantities are warranted, since not all the quantities are affected by the impurities in a way one would expect from the AG theory. For example, we show that the expression for the spin susceptibility of the CDW state in the presence of impurities is different from that obtained in the case of a superconductor with paramagnetic impurities.[22]

The main features of our calculation on the effects of impurities are as follows: (i) The order parameter, and hence T_P, is depressed by the impurities in accordance with the earlier result of Patton and Sham[14] and with recent experiments on TaS_3 doped with Nb impurities.[21] (ii) The half energy gap Ω_G and the order parameter differ substantially even for relatively small impurity concentrations. Further, for concentrations exceeding a critical value x_c', the gap goes to zero while the order parameter remains finite, thus giving a gapless CDW state. (iii) We show that the ratio $\Delta(T=0)/T_P$ increases with impurity concentration from 1.76 to $(2\pi)^{1/2}$, which is significant in view of the fact that 1D fluctuations lead to the same effect of comparable magnitude.[8] (iv) Impurities enhance the spin susceptibility χ of the CDW condensate. However, for all concentrations x less than x_c', i.e., except in the gapless region, χ still vanishes at zero temperature. This result is in contrast with that of the AG theory,[16] where for any finite concentration of magnetic impurities one obtains a finite $\chi(T=0)$. (v) Impurities also reduce the slope of the χ-versus-T curve near $T_P(x)$, in much the same way as the 1D fluctuations do. However, the magnitude of the effect seems too small to be of significance.

The remaining paper is organized as follows. In Sec. II we develop the formalism, followed in Sec. III by a discussion of the lattice distortion and the transition temperature. Section IV includes a discussion of the energy gap and its comparison with the other parameter. The spin susceptibility of pure CDW is calculated in Sec. V. The effects of impurities are computed in Sec. VI. Finally, in Sec. VII we summarize the conclusion of our study and various extensions of the present work.

II. FORMALISM

We start with the Frölich-type model Hamiltonian describing noninteracting electrons in a linear chain coupled to phonons:

$$H=\sum_{k,\sigma}\epsilon_k c_{k\sigma}^{\dagger}c_{k\sigma}+\sum_q \omega_q(b_q^{\dagger}b_q+b_{-q}^{\dagger}b_{-q}) + \sum_q \frac{g(q)}{\sqrt{N}}\sum_{k,\sigma}[c_{k+q,\sigma}^{\dagger}c_{k\sigma}(b_q+b_{-q}^{\dagger})+\text{c.c.}]\,, \tag{1}$$

where $c_{k\sigma}^{\dagger}$ is an creation operator for a 1D. Bloch electron, with energy ϵ_k and spin σ. Similarly, $b_q^{\dagger}$ is a creation operator for a longitudinal phonon with energy ω_q. N is the number of atoms in the chain and g is the electron-phonon coupling. For simplicity we shall consider a half-filled band and will set the chemical potential equal to zero.

In the mean-field theory[4-7] one considers only the interaction of the electrons with a single phonon of wave vector $|Q|=2k_F$, where $k_F=\pi N/2L$ is the Fermi wave vector. This phonon condenses below the transition temperature T_P resulting in a static lattice distortion described by the order parameter Δ:

$$\Delta=\frac{2g}{\sqrt{N}}\langle b_Q+b_{-Q}^{\dagger}\rangle_0\,, \tag{2}$$

where $\langle\ \rangle_0$ denotes thermal average with respect to the mean-field Hamiltonian H^0, obtained from H by replacing the phonon operators by their mean values:

$$H^0=\sum_{k,\sigma}\epsilon_k c_{k\sigma}^{\dagger}c_{k\sigma}+\frac{\Delta}{2}\sum_{k,\sigma}(c_{k+Q,\sigma}^{\dagger}c_{k\sigma}+c_{k\sigma}^{\dagger}c_{k+Q,\sigma})\,. \tag{3}$$

In writing Eq. (3) we have neglected a term $\alpha\Delta^2$, which is independent of electron operators. We have also used $c_{k+nQ,\sigma}=c_{k\sigma}$ where n=even integer. Δ satisfies a self-consistent equation, obtained by using Eq. (2) and the equations of motion for phonon operators:

$$\Delta=\frac{2g}{\omega_{2k_F}}\sum_k\langle c_{k+Q,\sigma}^{\dagger}c_{k\sigma}\rangle_0\,. \tag{4}$$

It is convenient for later introducing the effect of nonmagnetic impurities to recast Eqs. (3) and (4) in a matrix representation.[4,15] In this representation one artificially

divides the momentum space into k and $k+Q$ spaces by introducing the spinors[4,15]

$$\phi_{k\sigma}=\begin{bmatrix} c_{k\sigma} \\ c_{k+Q,\sigma} \end{bmatrix}, \tag{5}$$

and the Pauli 2×2 matrices $\hat{\xi}_1,\hat{\xi}_2,\hat{\xi}_3$. Equation (3) then becomes

$$H^0=\sum_{k,\sigma}\epsilon_k\phi^\dagger_{k\sigma}\hat{\xi}_3\phi_{k\sigma}+\Delta\sum_{k,\sigma}\phi^\dagger_{k\sigma}\hat{\xi}_1\phi_{k\phi}\ . \tag{6}$$

The thermal Green's function is defined by

$$G(k,i\omega_n)=-\int_0^\beta d\tau e^{i\omega_n\tau}\langle T_\tau\phi_{k\sigma}(\tau)\phi^\dagger_{k\sigma}(0)\rangle\ , \tag{7}$$

where T_τ denotes time ordering and $\beta=1/T$, $\omega_n=(2n+1)\pi T$. From (6) the inverse of the above matrix is immediately obtained as

$$[G^0(k,i\omega_n)]^{-1}=i\omega_n-\epsilon_k\hat{\xi}_3-\Delta\hat{\xi}_1\ . \tag{8}$$

From now on, we will drop the spin indices since they are irrelevant for most of our discussion. They will be reinserted wherever necessary. Inverting the above matrix, we obtain the thermal Green's function describing the CDW condensate in the absence of impurities:

$$G^0(k,i\omega_n)=\frac{-1}{\omega_n^2+\Delta^2+\epsilon_k^2}(i\omega_n+\epsilon_k\hat{\xi}_3+\Delta\hat{\xi}_1)\ . \tag{9}$$

Next we describe the interaction of the electrons with ordinary impurities by the Hamiltonian

$$H_1=\frac{1}{\sqrt{N}}\sum_i\sum_{k,q}e^{iqr_i}U(q)\phi^\dagger_{k\sigma}\hat{1}\phi_{k\sigma}\ , \tag{10}$$

where we have used the definition Eq. (5) of the spinors. $\hat{1}$ is a unit 2×2 matrix and r_i is the position of an impurity along the chain. This interaction gives rise to a self-energy $\Sigma(k,i\omega_n)$ defined by the Dyson equation

$$G^{-1}(k,i\omega_n)=[G^0(k,i\omega_n)]^{-1}-\Sigma(k,i\omega_n)\ . \tag{11}$$

We calculate $\Sigma(k,i\omega_n)$ in the first Born approximation and assume a random distribution of impurities to obtain

$$\Sigma(k,i\omega_n)=\frac{1}{N}\sum_i\sum_p|U(p-k)|^2G(p,i\omega_n)\ . \tag{12}$$

Now $G(k,i\omega_n)$ is strongly peaked near the Fermi level. The only important scattering is that in which momentum changes by $2k_F$. The term $q=0$ in H_1 causes only renormalization of the chemical potential. Thus replacing $U(p-k)$ by $U(2k_F)=U$, we have

$$\Sigma(k,i\omega_n)=xU^2\sum_p G(p,i\omega_n)\equiv\bar{G}(i\omega_n)\ , \tag{13}$$

where x is the impurity concentration. Substituting (13) in (11), we obtain the inverse of the matrix Green's function:

$$G^{-1}(k,i\omega_n)=(i\tilde{\omega}_n-\epsilon_k\hat{\xi}_3-\tilde{\Delta}_n\hat{\xi}_1)\ , \tag{14}$$

where

$$i\tilde{\omega}_n=i\omega_n-\bar{G}_{11}(i\omega_n)\ , \tag{15a}$$

$$\tilde{\Delta}_n=\Delta+\bar{G}_{12}(i\omega_n)\ . \tag{15b}$$

Inversion of matrix (14) immediately gives

$$G(k,i\omega_n)=\frac{-1}{\tilde{\omega}_n^2+\tilde{\Delta}_n^2+\epsilon_k^2}(i\tilde{\omega}_n+\epsilon_k\hat{\xi}_3+\tilde{\Delta}_n\hat{\xi}_1)\ . \tag{16}$$

Equations (13), (15a), (15b), and (16) form a closed set of equations. The solution proceeds by integrating (16) over k to obtain $G_{11}(i\omega_n)$ from Eq. (13). The results are substituted in Eqs. (15a) and (15b) to give

$$\tilde{\omega}_n=\omega_n+\frac{1}{2\tau}\frac{\tilde{\omega}_n}{(\tilde{\omega}_n^2+\tilde{\Delta}_n^2)^{1/2}}\ , \tag{17a}$$

$$\tilde{\Delta}_n=\Delta-\frac{1}{2\tau}\frac{\tilde{\Delta}_n}{(\tilde{\omega}_n^2+\tilde{\Delta}_n^2)^{1/2}}\ , \tag{17b}$$

where $\tau^{-1}=xU^2/hv_F$ is the electron lifetime due to impurities. As in other similar problems,[16,18] we find it convenient to introduce the quantity u_n:

$$u_n\equiv\tilde{\omega}_n/\tilde{\Delta}_n\ ,$$
$$\Delta u_n=\omega_n+\frac{1}{\tau}\frac{u_n}{(1+u_n^2)^{1/2}}\ . \tag{18}$$

Use has been made of Eqs. (17a) and (17b) in obtaining the second expression.

Equation (16), along with Eqs. (17a) and (17b) [or Eq. (18)], completely determine the thermal Green's function describing the CDW condensate in the presence of an ordinary impurity. We note in the passing that Eqs. (17a) and (17b) determining $\tilde{\omega}_n$ and $\tilde{\Delta}_n$ in terms of ω_n and Δ are the same relations as those occurring in Abrikosov-Gor'kov[16] theory of superconducting alloys with paramagnetic impurities. The origin of this analogy is due to the fact that in both cases impurity potential acts with opposite signs on the "particles" of the bound pair. Similar relations have also been obtained in the theory of excitonic insulator in the presence of nonmagnetic impurities.[18]

III. LATTICE DISTORTION AND THE TRANSITION TEMPERATURE

We first derive an equation for Δ describing the lattice distortion valid for arbitrary temperatures and concentrations, and then discuss the solution for various limiting cases which also yield an equation for the transition temperature T_P. The complete numerical solution of the equation for Δ is given in the next section in a graphical form.

Combining Eqs. (4) and (7), we have

$$\Delta=\frac{-2g}{w_{2k_F}}T\sum_{\omega_n}\int\frac{dk}{2\pi}G_{12}(k,i\omega_n)\ . \tag{19}$$

Use of Eq. (16) immediately gives

$$1=\frac{2g}{\omega_{2k_F}}2N_1(0)T\sum_{\omega_n}\int_0^{\epsilon_0}d\epsilon_k\frac{\tilde{\Delta}}{\Delta(\tilde{\omega}^2+\tilde{\Delta}_n^2+\epsilon_k^2)}\ , \tag{20}$$

where we have assumed a constant density of states

$N_1(0)=(2\pi\hbar v_F)^{-1}$ and introduced a cutoff $\epsilon_0\sim\omega_{2k_F}$ because the effective interaction between electrons and holes is restricted to a small region around the Fermi level ϵ_F. To proceed further, one adds and subtracts under the integral sign the corresponding expression in the pure ($\tau^{-1}=0$), metallic ($\Delta=0$) state. This allows one to extend the integration over ϵ in the difference to infinity, while the added term can be evaluated in a straightforward manner. The result is

$$1=\frac{2g}{\omega_{2k_F}}N_1(0)\ln\left[\frac{2\gamma}{\pi}\frac{\epsilon_0}{k_BT}\right]+\frac{2g}{\omega_{2k_F}}N_1(0)\pi T\sum_{\omega_n}\left[\frac{1}{\Delta(1+u_n^2)^{1/2}}-\frac{1}{|\omega_n|}\right], \quad (21)$$

where $\gamma=1.78$ is an Euler's constant. The coupling g in the above equation can be eliminated in favor of T_{P_0}, the transition temperature in the pure case, by noting that in the pure case as $\Delta\to0$, the difference terms in Eq. (20) vanishes. Thus one obtains the standard BCS-type result:

$$k_BT_{P_0}=\frac{2\gamma}{\pi}\epsilon_0e^{-1/\lambda}, \quad (22)$$

where $\lambda=2gN_1(0)\omega_{2k_F}$ is the dimensionless electron-electron coupling. Using the above result, we can rewrite Eq. (21) as

$$\ln\left[\frac{T}{T_{P_0}}\right]=\pi T\sum_{\omega_n}\left[\frac{1}{\Delta(1+u_n^2)^{1/2}}-\frac{1}{|\omega_n|}\right]. \quad (23)$$

This equation determines Δ for all temperatures T and concentration x. However, in the following we consider only the various limits of the above equation. In the region near the transition temperature T_P, where Δ is small, we can expand Eq. (17) for small u_n^{-1} to obtain

$$\frac{1}{\Delta(1+u_n^2)^{1/2}}\cong\frac{1}{\omega_n+\Gamma}-\frac{\Delta^2}{2}\frac{\omega_n}{(\omega_n+\Gamma)^4}+O(\Delta^4), \quad (24)$$

where $\Gamma=\tau^{-1}$ is the electron scattering rate from impurities. In this limit, Eq. (22) becomes

$$\ln\left[\frac{T}{T_{P_0}}\right]=\psi(\tfrac{1}{2}+\rho)-\psi(\tfrac{1}{2})-\frac{\Delta^2}{(2\pi T)^2}[\psi^{(2)}(\tfrac{1}{2}+\rho)+\psi^{(3)}(\tfrac{1}{2}+\rho)]. \quad (25)$$

Here $\psi(z)$ is the digamma function and $\psi^{(n)}(z)$ its nth derivative. $\rho=\Gamma/2\pi T_P$ is the pair-breaking parameter. The equation of the transition temperature $T_P(x)$ is obtained by setting $\Delta=0$ in Eq. (25):

$$\ln\left[\frac{T_P}{T_{P_0}}\right]=\psi(\tfrac{1}{2}+\rho)-\psi(\tfrac{1}{2}). \quad (26)$$

This describes the suppression of the transition temperature by nonmagnetic impurities in the same manner as for the excitonic insulator,[18] and is in agreement with earlier results.[14] Equation (26) shows that initially T_c will decrease linearly with the impurity concentration x. This prediction has recently been confirmed by experiments[21] on TaS_3, with Nb or Se added as an impurity. Similar suppression of T_P has also been observed in $NbSe_3$, doped with Ti (Ref. 23) and Ta (Ref. 24), and with irradiation-induced defects.

Equation (26) also predicts that at a critical concentration x_c, given by

$$\tau_c^{-1}=\frac{x_cU^2}{\hbar k_F}=\frac{\gamma}{\pi T_{P_0}}, \quad (27)$$

T_P vanishes. Unfortunately, in addition to the effects considered here impurities also tend to smear out the Peierls transition.[25] Thus one may be not able to experimentally determine x_c precisely. However, a 5% Ta doping in $NbSe_3$ depresses the lower transition ($T_{P_0}=59$ K) so much that it is no longer detectable[23] and thus provides credibility to the above result.

Next we consider the $T=0$ limit of the lattice distortion parameter $\Delta(0)$. In this case one can replace the summation over ω_n by an integration

$$T\sum_{\omega_n}\to\frac{1}{2\pi}\int_{-\infty}^{\infty}d\omega,$$

and proceed as before, to obtain from Eq. (19)

$$1=\frac{2g}{\omega_{2k_F}}N_1(0)\ln\left[\frac{2\epsilon_0}{\Delta(0)}\right]+\frac{2g}{\omega_{2k_F}}N_1(0)\tfrac{1}{2}\int_{-\infty}^{\infty}d\omega\left[\frac{1}{\Delta(0)(1+u^2)^{1/2}}-\frac{1}{[\omega^2+\Delta^2(0)]^{1/2}}\right]. \quad (28)$$

The order parameter $\Delta_0(0)$ for the pure case is immediately obtained by setting the second term equal to zero:

$$\Delta_0(0)=2\epsilon_0e^{-1/\lambda}. \quad (29)$$

The integration in Eq. (27) for the second term is easily performed by changing the variable: $\omega\to u$, where u is given by Eq. (18). The result after some algebra and use of Eq. (29), can be written as

$$\ln\left[\frac{\Delta(0)}{\Delta_0(0)}\right]=\begin{cases}-\frac{\pi}{4}\alpha, & \alpha\equiv(\tau\Delta)^{-1}\le1\\ -\ln[\alpha+(\alpha^2-1)^{1/2}]-\frac{\alpha}{2}\left[\frac{\pi}{2}-\frac{(\alpha^2-1)^{1/2}}{\alpha^2}-\tan^{-1}[(\alpha^2-1)^{1/2}]\right], & \alpha>1.\end{cases} \quad (30)$$

IV. ENERGY GAP

In the following we consider density of states in order to calculate the energy gap $2\Omega_G$ and show that there is a substantial difference between Ω_G and the order parameter Δ even for small impurity concentration. Furthermore, beyond a critical concentration x_c' the CDW condensate does not exhibit a gap in the energy spectrum. It is also pointed out that impurities cause strong deviations from the relation $\Delta(T=0)=1.76k_BT_{P_0}$. The density of states is given by

$$N(\omega)=-\frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{dk}{2\pi}\mathrm{Im}\,\mathrm{Tr}G(k,i\omega_n)\,|_{i\omega_n=\omega}\,, \quad (31)$$

where G is given by Eq. (16). A factor of $\frac{1}{2}$ is inserted to avoid double counting in k space. The integration over k is easily performed to give

$$N(\omega)=N_1(0)\,\mathrm{Re}\left[\frac{u}{(u^2-1)^{1/2}}\right], \quad (32)$$

where $u=\omega/\Delta$ is now given by

$$u\Delta=\omega+\frac{1}{\tau}\frac{u}{(u^2-1)^{1/2}}\,. \quad (33)$$

In the absence of impurities ($\tau^{-1}=0$), Eq. (31) reduces to

$$\frac{N(\omega)}{N_1(0)}=\begin{cases}0, & |\omega|<\Delta_0\\ \dfrac{|\omega|}{(\omega^2-\Delta_0^2)^{1/2}}, & |\omega|<\Delta_0\,.\end{cases} \quad (34)$$

As expected the pure system exhibits a gap $2\Delta_0$ in the excitation spectrum.

In the presence of impurities, u is, in general, a complex function of $\omega'=\omega/\Delta$. However, for small values of ω' (and α), u is real and less than one. Thus $N(\omega)=0$ for small ω. The half-gap Ω_G is determined by the maximum value of ω, such that Eq. (33) has a real solution, with $u<1$. Thus maximizing (33), we have

$$\Omega_G=\Delta(1-\alpha^{2/3})^{3/2} \quad (35)$$

for $\alpha\leq 1$, while $\Omega_G=0$ for $\alpha<1$.

Equation (35) clearly shows that in the presence of impurities a distinction has to be made between the order parameter Δ and the half-gap Ω_G. For example, the half-gap Ω_G and the order parameter exhibit quite different concentration dependences as shown in Fig. 1 at $T=0$ K. The order parameter has been calculated from Eq. (30). Even at small concentrations, for example, 10% of x_c, Ω_G and Δ differ significantly. Δ can be measured directly by neutron scattering, electron diffraction, or x-ray scattering while the half-gap Ω_G can be obtained directly by optical-edge studies[19] or tunneling experiments.[20] Ω_G can also be deduced from low-temperature spin susceptibility (see Sec. VI) or specific-heat measurements. Thus the predictions of the theory shown in Fig. 1 are easily verifiable.

Figure 1 also shows that the difference between Ω_G and Δ increases with the concentration, until at another critical concentration x_c', Ω_G vanishes, though the value of the order parameter is still about 50% of its pure value

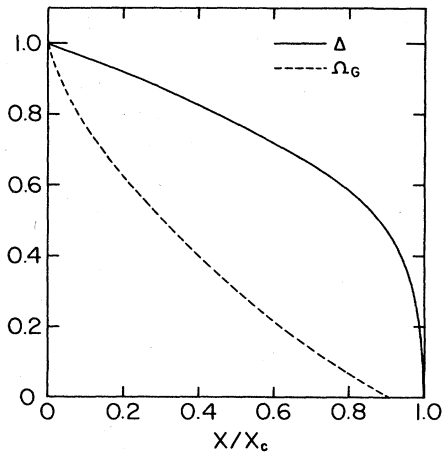

FIG. 1. Comparison of the concentration (x) dependence of the order parameter Δ and the half-energy gap Ω_G at $T=0$ K. The gap vanishes at $x_c'=0.912x_c$, where x_c is the critical concentration at which CDW order disappears.

$\Delta_0(0)$. Thus for the concentrations x such that $x_c'<x<x_c$, there is no gap in the energy spectrum, though we are in a Peierls state, as was first suggested by Schuster.[15] The new critical concentration x_c' at which the gap vanishes is determined by the condition $\alpha=1$. At $T=0$ K we can use Eqs. (30) and (27) to obtain

$$x_c'=\frac{hv_F}{U^2}\exp(-\rho/4)\Delta_0(0)\cong 0.918x_c\,. \quad (36)$$

The absence of the gap in the energy spectrum for $x_c'<x<x_c$ has important implications for spin susceptibility and specific heat. For example, in the gapless region one expects $\chi(T=0)\neq 0$, as will be confirmed in Sec. VI. The absence of the gap obviously effect the single-particle charge transport. The unattentuated collective-charge transport as first suggested by Fröhlich[5] hinges on having a gap in the single-particle excitation. Thus the collective charge transport is also expected to be affected by having a gapless CDW condensate.

It should be noted that for any finite concentration x of impurities, there is always a temperature region where the gap vanishes. The curve that separates the boundary of the gapless region from the region with a gap is given by $\tau^{-1}=\Delta(T)$. The gapless region is shown in Fig. 2 as the

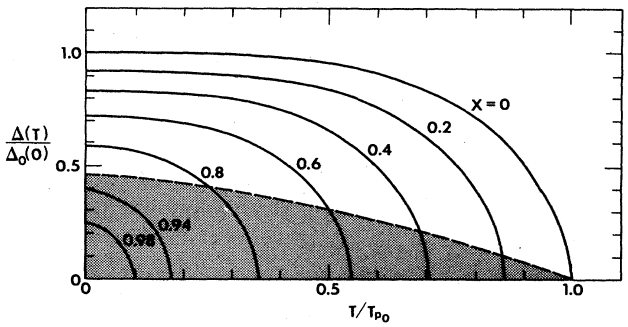

FIG. 2. Temperature dependence of the order parameter for various values of the concentration. The dotted area shows the gapless regime.

dotted area, where $\Delta(T)$ has been evaluated numerically using Eqs. (23) and (18). The calculational details of the numerical solution, which requires solution of the complex Eq. (18) can be found in Ref. 26.

Another important effect is that the BCS relation for the pure case $\Delta(0)\cong 1.76T_{P_0}$ between the order parameter at $T=0$ K and the transition temperature does not hold in the presence of impurities, as can be inferred from Fig. 2. The ratio $\Delta(T=0)/T_P$ increases with the impurity concentration from 1.76 to its limiting value of $(2\pi)^{1/2}$ at $x=x_c$. We feel this result is especially important in CDW systems since the observed T_P is always less than the T_P one obtains from the BCS relation $\Delta(0)=1.76T_P$. For example, in $K_{0.3}MoO_3$, the experimental value of $\Delta(0)$ gives a $T_P=320$ K, which is about 75% larger than the observed T_P.[9] This reduction in the critical temperature is usually attributable to the enhanced fluctuation effects in one dimension.[9,11] However, we see here that the impurities can cause a very similar effect, of comparable magnitude, and therefore, one must be very careful in identifying the source of the discrepancy between the measured T_P and the value of T_P deduced from $\Delta(0)$.

V. SPIN SUSCEPTIBILITY (PURE CASE)

We now derive an expression for the spin susceptibility χ using the thermal Green's function G^0, discussed in Sec. III. This formulation of the susceptibility calculation also allows one readily to introduce the effects of impurities, which will be the subject matter of the next section. The result derived below is in agreement with experimental data on $K_{0.3}MoO_3$ up to $T=0.9T_P$.

We first obtain a general expression for the wave-vector-dependent susceptibility $\chi(q)$, which easily yields the desired result for χ in the limit $q\to 0$.

The wave-vector-dependent spin susceptibility is given by[27]

$$\chi(q)=\mu_B^2\sum_{\substack{p,p'\\ \sigma,\sigma'}}\sigma\sigma'\int_0^\beta d\tau\langle T_\tau c^\dagger_{p+q,\sigma}(\tau)c_{p\sigma}(\tau)c^\dagger_{p'-q,\sigma}(0)c_{p',\sigma'}(0)\rangle\ , \tag{37}$$

where σ,σ' (± 1) are the spin indices. To perform the sum over σ and σ', we note that in the charge-density-wave condensate there is no correlation between electrons of opposite spins and hence only $\sigma=\sigma'$ terms contribute. This situation is in contrast with a BCS superconductor where strong correlation exists between electrons of opposite spins and $\sigma\neq\sigma'$ terms must be kept. Despite this difference, we show below that the final expressions of $\chi(q)$ are exactly of the same form in the two cases. However, this difference is important in considering the effects of impurities on χ as will be seen in Sec. VI.

Thus, using $\sigma^2=1$ and dropping the spin indices, the above correlation function can be written as a product of pairwise time-ordered averages:

$$\chi(q)=-2\mu_B^2\int_0^\beta d\tau\sum_{p,p'}\langle T_\tau c_{p'}(0)c^\dagger_{p+q}(\tau)\rangle\langle T_\tau c_p(\tau)c^\dagger_{p'-q}(0)\rangle\ . \tag{38}$$

One can express the right-hand side of the above equation in the terms of the various components of the Green's function G^0 describing the CDW in the absence of impurities. Using Eq. (7) and the usual definition of the Fourier transform to ω_n space,[27] we obtain

$$\begin{aligned}\chi(q)=-2\mu_B^2T\sum_{\omega_n}\sum_k[&G^0_{11}(k+q,\omega_n)G^0_{11}(k,\omega_n)\\ &+G^0_{22}(k+q,\omega_n)G^0_{22}(k,\omega_n)\\ &+G^0_{12}(k+q,\omega_0)G^0_{21}(k,\omega_n)\\ &+G^0_{21}(k+q,\omega_n)G^0_{12}(k,\omega_n)]\ .\end{aligned} \tag{39}$$

The matrix G^0 is given by Eq. (9). To perform the sum over ω_n, we find it convenient to express various components of G^0 in the following form:

$$G^0_{11}(k,\omega_n)=\frac{u_k^2}{i\omega_n-E_k}+\frac{v_k^2}{i\omega_n+E_k}\ , \tag{40a}$$

$$G^0_{22}(k,\omega_n)=\frac{v_k^2}{i\omega_n-E_k}+\frac{u_k^2}{i\omega_n+E_k}\ , \tag{40b}$$

$$G^0_{12}(k,\omega_n)=G^0_{21}(k,\omega_n)=u_kv_k\left[\frac{1}{i\omega_n-E_k}-\frac{1}{i\omega_n+E_k}\right]\ , \tag{40c}$$

where

$$E_k^2=+(\Delta^2+\epsilon_k^2)^{1/2} \tag{41}$$

and the coherence factors are given by

$$u_kv_k=\frac{\Delta}{2E_k}\ , \tag{42a}$$

$$u_k^2=1-v_k^2=1+\epsilon_k/E_k\ . \tag{42b}$$

ϵ_k is the free-electron energy measured from the Fermi level. With this form, the sum over ω_n in Eq. (39) is easily evaluated by conventional contour integration to give

$$\chi(q)=-2\mu_B\left[\sum_k(u_ku_{k+q}+v_kv_{k+q})^2\frac{f(E_k)-f(E_{k+q})}{E_k-E_{k+q}}-(u_{k+q}v_k-u_kv_{k+q})^2\frac{1-f(E_k)-f(E_{k+q})}{E_k+E_{k+q}}\right]\ . \tag{43}$$

This is our final expression for the wave-vector-dependent spin susceptibility $\chi(q)$ of the CDW condensate valid for arbitrary temperature T. As promised, it has the same form as that of the spin susceptibility $\chi(q)$ of a BCS superconductor.[28] The wave-vector dependence of $\chi(q)$ is crucial to the question of coexistence of the CDW state and magnetic order and to the related problem of indirect magnetic interactions in the CDW condensate. These issues will be addressed elsewhere.[29] Here we focus on the $q=0$ limit.

In the limiting case of $q=0$, the second term in the above equation vanishes, while the first term yields $[\chi \equiv \chi(q=0)]$

$$\chi = -2\mu_B^2 \sum_k \frac{\partial f(E_k)}{\partial E_k} . \tag{44}$$

This is our final result for the spin susceptibility in the CDW condensate. The corresponding result for the BCS superconductor, which again has the same form as above, was first obtained by Yoshida.[30] This result can be written in a more useful form, by using the variable $y=\beta E_k$, where E_k is given by Eq. (41):

$$\frac{\chi}{\chi^p} = 2\int_\delta^\infty dy \frac{e^y}{(e^y+1)^2} \frac{y}{(y^2-\alpha^2)^{1/2}} , \tag{45}$$

where $\delta=\beta\Delta$ and $\chi^p=2\mu_B^2 N_1(0)$ is the Pauli susceptibility in the metallic state.

As $\Delta\to 0$, the right-hand side of Eq. (45) becomes unity. Thus the paramagnetic susceptibility decreases from its metallic state value χ^p at the transition temperature T_P with decreasing temperature and vanishes exponentially as $T\to 0$ K. The complete temperature dependence of χ is shown in Fig. 3.

The above result can also be obtained in a phenomenological manner by assuming as 1D semiconductor model, with a gap $2\Delta_0$. Johnston[8] uses such an approach to analyze his experimental data on $K_{0.3}MoO_3$ which are the only clear-cut available data. He employs $\Delta_0(T)$ measured by Sato *et al.*[31] via neutron scattering and shows that the above result is in precise agreement with experiments up to about $0.9T_P$, where $T_P=183$ K. At higher temperatures strong deviations from the theory are found due to the effects of 1D fluctuations.[11] For high temperatures the theory of Lee *et al.*,[10] which incorporates the effects of 1D fluctuations, but is based on Landau-Ginzburg expansion,[11] provides an excellent fit to the experimental data.[8] It should be noted that although the experimental order-parameter results of Sato *et al.*[31] follow closely the BCS-type temperature dependence, the data probably included some three- or two-dimensional effects. The spin

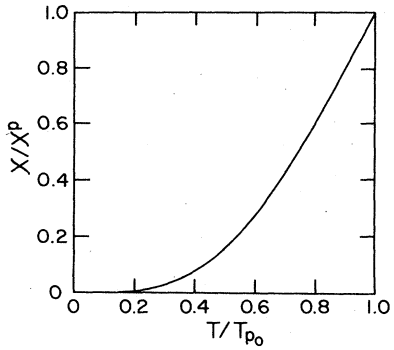

FIG. 3. Spin susceptibility χ_{CDW} in the absence of impurities as a function of reduced temperature T/T_{P_0}. χ^p is the Pauli susceptibility.

susceptibility of other quasi-1D systems, e.g., TaS_3 and $(TaSe_4)_2I$, show similar temperature dependences.[32] $NbSe_3$ also shows a decrease in susceptibility at both transitions.[33] However, the decrease in χ is much less than expected from our result. This could be due to an increase in the Van Vleck magnetism at low temperatures.[33]

VI. EFFECT OF IMPURITIES ON SPIN SUSCEPTIBILITY

In the following we discuss the effect of nonmagnetic impurities on the spin susceptibility of the CDW condensate. From Eq. (39) it is clear that in the presence of impurities, one needs to calculate the average over the random positions of impurities of the product of two Green's function. This requires calculating (i) the averaged Green's function, i.e., the self-energy correction and (ii) the vertex corrections, since the average of the product of two Green's function is not equal to the product of averaged Green's functions. The averaged Green's functions were obtained in Sec. II. Thus, in the following we mainly focus on the vertex correction.

As the scattering from ordinary impurities does not mix the up- and down-spin spaces, it is sufficient to consider only one spin direction. Then the generalization of Eq. (39) in the presence of impurities is

$$\chi = 2\mu_B^2 T \sum_{\omega_n} \frac{dk}{2\pi} \mathrm{Tr}[G(k,\omega_n)\Lambda(k,\omega_n)G(k,\omega_n)] , \tag{46}$$

where G is the matrix Green's function given by Eq. (16) and includes the self-energy corrections due to impurities. $\Lambda(p,\omega_n)$ is the vertex function and in the standard ladder approximation is determined by the integral equation:

$$\Lambda(p,\omega_n) = \hat{1} + \frac{x}{2\pi}\int dp'[U(p-p')G(p',\omega_n)\Lambda(p',\omega_n)G(p',\omega_n)U(p'-p)] . \tag{47}$$

The solution of the above equation is obtained by noting that since the integrand decreases rapidly on going away from the Fermi level, one can assume $\Lambda(p,\omega_n)=\Lambda(\omega_n)$. Further, we make the following ansatz regarding the form of $\Lambda(\omega_n)$:

$$\Lambda(\omega_n) = \begin{bmatrix} \Lambda^1 & \Lambda^2 \\ \Lambda^2 & \Lambda^1 \end{bmatrix} . \tag{48}$$

We note that this ansatz is slightly different than the one used by Gor'kov and Rusinov[22] in their calculation of the

spin susceptibility of a superconductor with magnetic impurities. This is due to the fact in our calculation electronic spin does not play any important role.

With this ansatz, integration in Eq. (47) is easily performed to give

$$\Lambda^1=1+I_1\Lambda^1+I_2\Lambda^2 , \tag{49a}$$

$$\Lambda^2=\Lambda^1 I_3+\Lambda^2 I_4 , \tag{49b}$$

where

$$I_1=\frac{1}{D} , \tag{50a}$$

$$I_2=I_3=\frac{iu_n}{D} , \tag{50b}$$

$$I_4=-\frac{u_n^2}{D} \tag{50c}$$

with

$$D=2\tau\tilde{\Delta}_n(1+u_n^2)^{3/2} . \tag{51}$$

The solution of the above coupled equations is

$$\Lambda^1=\frac{1+u_n^2/D}{1+(u_n^2-1)/D} , \tag{52a}$$

$$\Lambda^2=\frac{iu_n D}{1+(u_n^2-1)/D} . \tag{52b}$$

The integration over k in Eq. (46) is formally divergent and one must sum over ω_n first. However, this formal difficulty can be overcome by adding and subtracting under the integral sign the corresponding expression in the metallic state. Then it is legitimate first to integrate over k in the difference term to obtain

$$\frac{\chi}{\chi^p}=1-\pi T\sum_{\omega_n}[(I_1\Lambda^1+I_2\Lambda^2)2\tau] , \tag{53}$$

which upon using Eqs. (49a) and (52a) immediately yields

$$\frac{\chi}{\chi^p}=1-\pi T\sum_{\omega_n}\frac{1}{(1+u_n^2)\left[\tilde{\Delta}_n(1+u_n^2)^{1/2}+\frac{1}{2\tau}\frac{u_n^2-1}{1+u_n^2}\right]} . \tag{54}$$

This result can be written in a more convenient way by the use of Eq. (17b) for $\tilde{\Delta}_n$:

$$\frac{\chi}{\chi^p}=1-\frac{\pi T}{\Delta}\sum_{\omega_n}\frac{1}{(1+u_n^2)\left[(1+u_n^2)^{1/2}-\frac{1}{\tau\Delta}\frac{1}{1+u_n^2}\right]} . \tag{55}$$

This is our final result of the spin susceptibility describing the effect of impurities. Note that this expression is different than the corresponding expression of the spin susceptibility in the AG theory of superconductors with dilute magnetic impurities.[22]

To discuss the concentration dependence of χ, we consider the $T=0$ K case. In this case the sum over ω_n can be converted to an integral over ω which in turn can be written as an integral over u by the use of Eq. (18) and we get

$$\frac{\chi}{\chi^p}=1-\int_{u_0}^{\infty}du\left[1-\alpha\frac{1}{(1+u^2)^{3/2}}\right]\left[\frac{1}{(1+u^2)^{3/2}-\alpha}\right] , \tag{56}$$

where $\alpha=(\tau\Delta)^{-1}$. $u_0=0$ for $\alpha\leq 1$ and $u_0=(\alpha^2-1)^{1/2}$ for $\alpha>1$. We have evaluated the integral numerically and the results for the susceptibility χ as a function of impurity concentration x are shown in Fig. 4. For $x<x_c'$ we find $\chi(T=0)=0$ due to the fact that for $x<x_c'$, the excitation spectrum has a finite gap. Note that this result is qualitatively different than that in the AG theory, where any finite concentration of magnetic impurities gives rise to a finite value of the spin susceptibility. For $x>x_c'$, χ first increases rapidly with the concentration and then goes over smoothly to its metallic state value χ^p as $x\rightarrow x_c$. Indirect experimental verification of our results shown in Fig. 4 comes from recent experiments[34] in a similar system (heavily doped polyacetylene), which also shows a transition to a gapless Peierls state.

Figure 5 shows the susceptibility χ as a function of reduced temperature $T/T_P(x)$ for four different values of the concentration. There is a general enhancement of the spin susceptibility due to the pair-breaking effect of impurities. However, all the curves still start from zero, except the curve $x=0.94x_c$ which represents the gapless regime. Even in this latter case, the initial value of χ is less than χ^p since the density of states at $\omega=0$, though finite, is still less than $N_1(0)$, the density of states in the metallic state. It is interesting to note that near the transition temperatures, impurities cause a decrease in the slope of the χ-versus-T curve. This is qualitatively similar to the effect of 1D fluctuations.[10] Unfortunately the size of the decrease in the slope seems too small to be of significance in relation to the experimental data.

Lastly we consider the low temperature ($T\ll T_P$) regime to show that qualitatively different temperature dependences are obtained in the gapless regime and the re-

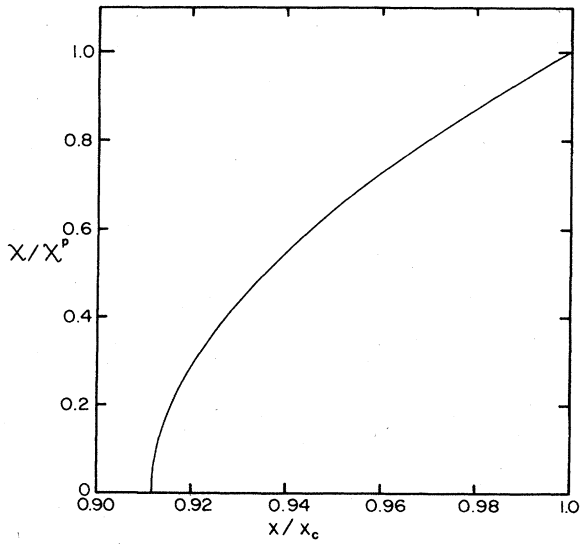

FIG. 4. Concentration dependence of the spin susceptibility at $T=0$ K. Note that χ first become nonzero at $x=0.912x_c$, where the gapless regime begins.

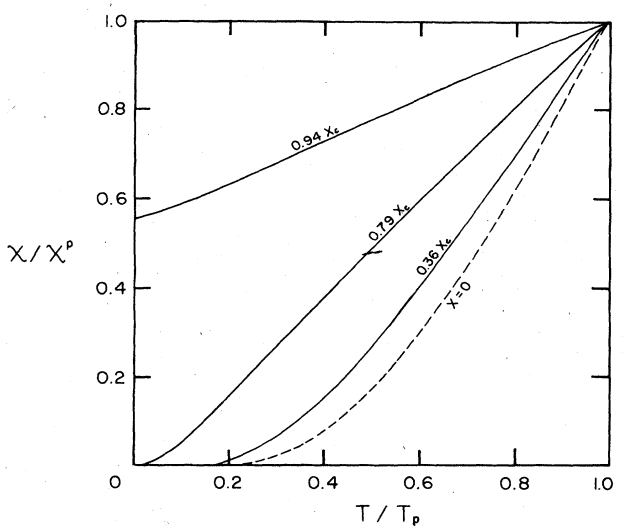

FIG. 5. Temperature dependence of the spin susceptibility for several values of the impurity concentration. Note the decrease in the slope near $T_P(x)$.

gime with a gap. For temperatures slightly above $T=0$ K, one can write Eq. (56) as[17]

$$\frac{\chi}{\chi^p}=1-\frac{1}{\Delta}\int_0^\infty d\omega\frac{1}{(1+u^2)^{3/2}-\alpha} -\frac{2}{\Delta}\int_0^\infty d\omega f(\omega)\mathrm{Im}\left[\frac{1}{(1-u^2)^{2/3}-\alpha}\right], \tag{57}$$

where $f(\omega)$ is a Fermi function. Evaluation of the right-hand side gives

$$\frac{\chi}{\chi^p}=\begin{cases}(1-\alpha^{3/2})^{-1/4}\alpha^{-3/2}\left[\dfrac{2\pi T}{3\Delta}\right]^{1/2}e^{-\Omega_G/T}, & \alpha<1\\ \alpha(\alpha^2-1)^{1/2}+\frac{1}{2}\alpha^{-4}(\alpha^2-1)^{-5/2}\left[\dfrac{\pi T}{\Delta}\right]^2, & \alpha<1\,.\end{cases}$$

Thus in the concentration regime with a finite gap ($\alpha<1$), χ shows an exponential variation with the temperature. On the other hand, in the gapless regime, the initial temperature dependence is quadratic.

VII. CONCLUSIONS

We have derived a low-temperature result for the spin susceptibility, which agrees with the experiments. We have shown impurities enhance spin susceptibility, due to their pair-breaking effect on CDW pairing. The finite-impurity results follow closely the energy-gap results and hence are in basic agreement with a semiconductor model commonly used to describe electronic properties of the CDW condensate. Furthermore, our result for χ in the presence of impurities is different than the corresponding result in the AG theory.[22] Similar differences should also be expected in the other physical quantities such as nuclear spin-relaxation rate, where conduction-electron spin enters explicitly.

Perhaps the most interesting result is the significant difference between the lattice-distortion parameter and the half-gap Ω_G caused by the impurities, which can be tested directly by experiments. In this regard, a recent calculation on the effect of localization on the Peierls transition by Abrikosov and Dorotheyev[35] should be noted. These localization effects must be considered for strictly one-dimensional conductors. They found the energy gap is absent at any impurity concentration due to the localization effects of random impurity potential. Thus careful experimental determination of the concentration dependence of the energy gap in the materials of current interest ($NbSe_3$, $K_{0.3}MoO_3$, etc.) would be very useful in distinguishing the two effects: the lifetime effects considered in the present work and the localization effect.[35] This in turn can help us in understanding the novel transport properties exhibited by these materials.

Some of the impurity effects are found to be similar to those stemming from 1D fluctuations. Notable among these is the substantial increase in the ratio $\Delta(T=0\ \mathrm{K})/k_BT_P$ with impurity concentration.

The specific heat is also a basic thermodynamic property. However in this case, in addition to electronic specific heat, one must calculate the lattice contribution to heat capacity as well. The electronic part is expected to be that of a 1D semiconductor and should show a jump at the transition temperature. Further impurities should reduce the size of the jump. The calculation of specific heat, which requires evaluation of the free energy are underway and will be published elsewhere.

Our theory does not take into account explicitly the weak interchain coupling, which gives rise to the transition to the Peierls state at a finite temperature. A more complete theory of quasi-1D compounds should take into account this weak interchain coupling and other three-dimensional effects. This problem is under investigation.

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