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Transfer-matrix method for non-Hermitian pseudo-Hamiltonians and the commensurate-incommensurate charge-density-wave transition

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A microscopic commensurability pinning Hamiltonian with charge conservation is presented and analyzed as a model for the commensurate-incommensurate charge-density-wave transition. The ground state is always a soliton lattice with its character changing continuously between commensurate and incommensurate states depending on the commensurability strength. The existing transfer-matrix methods are criticized and extended to non-Hermitian pseudo-Hamiltonian systems. Charge conservation leads to results for the charge-density correlation length and the structure factor that are consistent with the x-ray-scattering experiment and the NMR Knight-shift experiment on 2H-TaSe$_2$.

I. INTRODUCTION

The incommensurate (IC) to commensurate (C) or "lock-in" transition of charge-density waves (CDW's) in systems of reduced dimensionality such as TTF-TCNQ and 2H-TaSe$_2$ has been studied theoretically along the two directions: One is the Landau phenomenology and the other is the transfer-matrix (TM) path-integral method. The Landau theory was initiated by McMillan[1] and developed further by a number of authors.[2] In order to reproduce rather rich experimental observations, the direction in the Landau theory is inevitably toward more complex Ginzburg-Landau (GL) free-energy functionals. On the other hand, the TM method[3-5] approach was initiated by Brazovskii, Dzyaloshinskii, and Obukhov[6] and further developed by Okwamoto, Takayama, and Shiba[7] and Turkevich and Doniach.[8] The direction here is toward some exact results on the C-IC crossover in one-dimensional phase-only GL fields.

In this paper, we shall revisit the latter TM approach with the following improvements: (i) Instead of GL free-energy functionals, we start with a microscopic commensurability pinning Hamiltonian developed by Lee, Rice, and Anderson and others.[9-11] We thus eliminate certain parametric ambiguities, and the temperature range to be explored in comparison with experiments can be identified. (ii) We take into account charge conservation, which was totally ignored previously in both the Landau phenomenology and the TM method approach. As we will see, charge conservation leads to results qualitatively different from the previous findings in the TM approach. (iii) The non-Hermiticity of pseudo-Hamiltonians in the TM formulations was also overlooked by the previous authors, casting some doubts on their results. We will develop a correct procedure for non-Hermitian pseudo-Hamiltonians. Through the exact evaluation of the charge-density–charge-density correlation function leading to the CDW coherence length and the structure factor, the present TM theory makes an experimental implication: At the C-IC transition, the CDW wavelength should experience a sudden change, whereas the Knight shift of the NMR absorption lines would not show any sudden changes. This is consistent with experiments.[12,13,16]

We have organized the paper as follows: In Sec. II the model Hamiltonian with the charge conservation is discussed. Section III discusses the ground state and Secs. IV and V discuss thermal properties, i.e., the grand partition function and the charge-density–charge-density correlation function with an emphasis on the improved TM method for the non-Hermitian pseudo-Hamiltonian systems and the charge conservation effect. Section VI presents numerical results for the charge-density correlation length and the structure factor in comparison with the experimental results. Finally a conclusion is given in Sec. VII.

II. THE MODEL HAMILTONIAN

The IC CDW state is characterized by the charge density

$$\rho(x)=\bar{\rho}+\rho_0\cos(qx+\Phi) , \tag{1}$$

where $\bar{\rho}$ is the uniform part of the density, the second term is the CDW order parameter, and $q=2k_F$ with k_F representing the Fermi wave number associated with the conduction electrons which condense into a CDW state at low temperatures. The effective Hamiltonian for the phase $\Phi(x,t)$ for describing the low-energy fluctuations of an IC CDW can be derived as follows.[10,11] Since we are interested in a weak spatial modulation, we may write $\varphi(x,t)=qx+\Phi(x,t)\approx q(x-vt)$ to get a local speed of electrons associated with the phase modulation,

$$v=-\frac{\Phi_t}{q} . \tag{2}$$

Now let m^* be an effective mass of electrons in CDW condensates and n be the number density of electrons per spin. The kinetic energy per length is then given by

$$2\frac{nm^*}{2}\left[\frac{\Phi_t}{q}\right]^2 \tag{3}$$

where the factor 2 accounts for up and down spins of electrons. Note that the total number N of electrons is related to the IC CDW wave number q as $N=2k_F/(2\pi/L)$ where L is the system size, and therefore $n=q/2\pi$. Next the elastic energy per unit length should have the form

$$\frac{v_F}{4\pi}\Phi_x^2 , \tag{4}$$

where v_F is the Fermi velocity, in order that the resulting linear dispersion relation is consistent with the microscopic theory of Lee, Rice, and Anderson.[9] Finally an interaction potential per unit length describing the coupling of CDW's to the underlying lattice, the so-called commensurability-pinning potential, can generally be written as

$$d\cos(n_0\varphi-Gx)=d\cos(\Delta qx+n_0\Phi) , \tag{5}$$

where $\Delta q\equiv n_0q-G$ and G is a reciprocal-lattice vector. Thus the model Hamiltonian reads

$$H=\int_0^L dx\left\{\frac{m^*}{2\pi q}\Phi_t^2+\frac{v_F}{4\pi}\Phi_x^2+d\cos(\Delta qx+n_0\Phi)\right\} . \tag{6}$$

The Hamiltonian (6) and its impurity-pinning variant have been used before to study various properties of the IC phase.[9-11]

We now take (6) as a model Hamiltonian for the C-IC transition. In fact, we can put (6) in the form (13) below, which is identical to the GL free-energy functional introduced by McMillan.[1] An essential difference is that we take into account the charge conservation. The charge is related to the phase Φ as follows. First note that the current density is given by

$$j=2nev=-2ne\frac{\Phi_t}{q} . \tag{7}$$

Associated with the current density is the charge density such that

$$\rho_t+\nabla j=0 . \tag{8}$$

From (7) and (8) we have

$$\rho=\frac{2ne}{q}\Phi_x=\frac{2e}{2\pi}\Phi_x \tag{9}$$

and the net charge associated with the phase dynamics is given by

$$Q=\frac{2e}{2\pi}[\Phi(L)-\Phi(0)] . \tag{10}$$

Taking the IC ground state as a reference state, the charge conservation requires that the phase dynamics is restricted to

$$\Phi(L)-\Phi(0)=0 . \tag{11}$$

Now making the transformation

$$n_0\Phi+\Delta qx=\phi , \tag{12}$$

the model Hamiltonian (6) and the boundary condition (11) becomes

$$H=\int_0^L dx\left[\frac{m^*}{2\pi qn_0^2}\Phi_t^2+\frac{v_F}{4\pi n_0^2}\{\phi_x-\Delta q\}^2+d(1-\cos\phi)\right] , \tag{13}$$

$$\phi(L)-\phi(0)=\Delta qL=n_0\delta qL=2\pi n_0\delta N \tag{14}$$

where we have introduced a parameter

$$\delta\equiv 1-\frac{G}{n_0q} . \tag{15}$$

It is noted that the Hamiltonian (13) is, apart from coefficients, identical to the GL free-energy functional of McMillan as analyzed previously by the TM method[4-8] *without* the charge conservation constraint (14). TM analyses of the systems identical to (13) *with* (14) have been done before by Gupta and Sutherland (GS) ($\Delta q=0$ case) (Ref. 4) and by Guyer and Miller (GM) on general ground[5] and in the context of the physisorbed one-dimensional (1D) monolayer problem.[14] For a more general approach to the 1D commensurability problem see Sacco, Widom, and Sokoloff.[15] The ground-state analysis in the next section is thus simply a translation of their results to our language, but the calculation of the grand partition function requires some mathematical clarifications: First, the GS approach is based on a trick of analytic continuation of the chemical potential to the complex space, whereas the GM approach can avoid the trick. A legitimate question is if this remarkable trick is correct. We will give an affirmative answer to this question by providing an equivalence proof for the two approaches. Second, and more significant, both formulations overlooked the necessity of modifying the existing TM formulation for non-Hermitian pseudo-Hamiltonians. We will develop a correct procedure for the non-Hermitian pseudo-Hamiltonian systems.

III. THE GROUND STATE

Let us first discuss the ground state. The equation of motion for ϕ is the sine-Gordon equation

$$\frac{m^*}{\pi qn_0^2}\phi_{tt}-\frac{v_F}{2\pi n_0^2}\phi_{xx}+d\sin\phi=0 . \tag{16}$$

The degenerate vacua $\phi=0,\pm2\pi,\pm4\pi,\ldots$ are connected by soliton (+) and antisoliton (−) excitations

$$\phi_\pm=4\tan^{-1}\left\{\exp\left[\pm\left[\frac{2\pi d}{v_F}\right]^{1/2}n_0x\right]\right\} , \tag{17}$$

which provide a characteristic length (soliton size)

$$s=\left[\frac{v_F}{2\pi d}\right]^{1/2}\frac{1}{n_0} \tag{18}$$

and a characteristic energy (soliton energy for $\Delta q=0$)

$$E=8sd . \tag{19}$$

Since these excitations accompany $\pm2\pi$ phase shifts, the charges carried by these particles are found from (9) and (12) to be $\pm 2e/n_0$. Note that the $n_0=2$ case is the φ particle of Rice *et al.*[11]

It is clear that the ground state, characterized by (16) with $\phi_t=0$ and the boundary condition (14), is a lattice of $n_0\delta N$ solitons. Note that from (12),

$$\varphi = qx + \Phi = \frac{G}{n_0}x + \frac{\phi}{n_0} , \tag{20}$$

and therefore the vacua $\phi = 0(\mathrm{mod}\,2\pi)$ are of a commensurate nature. Thus the ground state may be viewed as commensurate regions separated by solitons or McMillan's "discommensurations." In contrast, McMillan regarded a soliton lattice as an incommensurate state and a flat vacuum as a commensurate state. This is because in his theory the charge is not conserved. Although the ground state is always a soliton lattice in our charge conserved analysis, it is clearly incommensurate when $d=0$, commensurate when d is large, and continuously changes its character between commensurate and incommensurate with d.

A more appropriate parameter to measure the degree of commensurability is

$$\eta = \frac{L - n_0\delta N s}{n_0 \delta N s} , \tag{21}$$

which describes the fraction of commensurate areas in the total length L; the larger η means the stronger commensurability. In Fig. 1, we have plotted the ground-state configurations for various η.

Here we get a rough idea for the size of the system parameters n_0 in (5), δ in (15), the soliton size s in (18), the soliton energy E in (19), and the commensurability strength η in (21). For TTF-TCNQ,[11,12] $n_0=4$, $\delta \sim 0.17$, $s \sim 130$ Å, $E \sim 280$ K, and $\eta \sim 10$. For 2H-TaSe$_2$,[13,16] $n_0=3$, $\delta \sim 0.02$, $s/a \sim 5$ where a is the lattice constant, $E \sim 50$ K, and $\eta \sim 10$. In the latter estimate, we have assumed the same value $v_F/a \sim 1\ \mathrm{sec}^{-1}$ as in TTF-TCNQ and used the relationship between our η and the parameter γ of Suits, Couturié, and Slichter,[13]

$$\eta = 2\int_0^{\pi/2} \frac{dz}{\sqrt{\gamma^2 + \cos^2 z}} - 1 . \tag{22}$$

Their values $0.0008 \sim 0.08$ correspond to our $\eta = 7 \sim 16$, $\eta = 10$ being an average. In our numerical calculations below, we will use the parameters for 2H-TaSe$_2$. In this case, the IC-CDW locks in to the underlying lattice and becomes a C-CDW by decreasing its wavelength about 2%. Note also that small δ implies $|\phi_x| \ll q$, consistent with our assumption of weak spatial modulation.

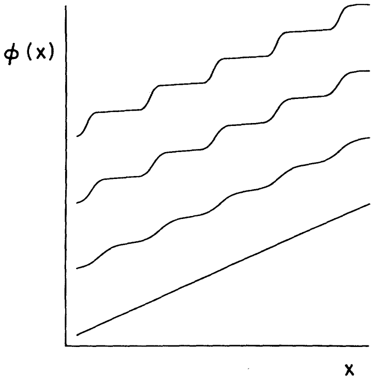

FIG. 1. The ground-state configurations for $\eta =$ 0, 5, 10, and 15 (bottom to top). $\phi(x)$ vs x for five solitons.

IV. THE GRAND PARTITION FUNCTION

We put the system (13) and (14) in a heat bath of temperature T. The classical partition function can be written as a path integral

$$Z = \int \delta\phi \int \delta\pi\, e^{-\beta H} \equiv Z_\pi Z_\phi , \tag{23}$$

where

$$\pi = \frac{m^*\Delta x}{\pi q n_0^2}\phi_t \tag{24}$$

is the canonical momentum in the discretized, $L = M\Delta x$, version of the Hamiltonian (13). We will take $\Delta x \to 0$ and $M \to \infty$ in the end. Z_π is a simple Gaussian integral ($\beta \equiv 1/t$, Boltzmann const$=1$)

$$Z_\pi = \left[\frac{2m^*\Delta x}{\beta q n_0^2} \right]^{M/2} . \tag{25}$$

Taking into account the boundary condition (14), Z_ϕ is written as

$$Z_\phi = \int d\phi_1 \cdots \int d\phi_{M+1}\delta(\phi_{M+1} - \phi_1 - 2\pi N n_0 \delta) \times \prod_{i=1}^{M} K(\phi_i, \phi_{i+1}) , \tag{26}$$

where we have introduced the TM operator

$$K(\phi_i,\phi_{i+1}) \equiv \exp\left[-\beta\Delta x \left\{ \frac{v_F}{4\pi n_0^2} \left[\frac{\phi_{i+1}-\phi_i}{\Delta x} - \Delta q \right]^2 + d(1-\cos\phi_{i+1}) \right\} \right] . \tag{27}$$

Now the standard TM procedure is to expand the δ function in (26) in terms of the eigenfunctions Φ_n of the TM equation

$$\int d\phi_i K(\phi_i,\phi_{i+1})\Phi_n(\phi_i) = \left[\frac{4\pi^2 \Delta x n_0^2}{\beta v_F} \right]^{1/2} e^{-\beta\Delta x \varepsilon_n}\Phi_n(\phi_{i+1}) \tag{28}$$

which, after Taylor expanding $\Phi_n(\phi_i)$ around ϕ_{i+1} and performing the Gaussian integral with respect to ϕ_i, reduces to the pseudo-Schrödinger equation

$$H_{\mathrm{TM}}\Phi_n \equiv \left\{ -\frac{\pi n_0^2}{v_F\beta^2}\frac{d^2}{d\phi^2} + \frac{1}{\beta}\Delta q \frac{d}{d\phi} + d(1-\cos\phi) \right\}\Phi_n(\phi) = \varepsilon_n \Phi_n(\phi) . \tag{29}$$

This does not work here because of the non-Hermiticity of the pseudo-Hamiltonian H_{TM}. Indeed, H_{TM} is not Hermitian,

$$H_{\mathrm{TM}}^{\dagger}-H_{\mathrm{TM}}=-\frac{2\Delta q}{\beta}\frac{d}{d\phi} \tag{30}$$

and

$$\langle \Phi_m|\Phi_n\rangle=\frac{1}{\varepsilon_m^{*}-\varepsilon_n}\frac{-2\Delta q}{\beta}\left\langle \Phi_m\left|\frac{d}{d\phi}\right|\Phi_n\right\rangle . \tag{31}$$

But since H_{TM} and $d/d\phi$ do not commute, Φ_n is not a simultaneous eigenfunction of $d/d\phi$ and H_{TM}. Therefore we have in general

$$\langle \Phi_m|\Phi_n\rangle\neq 0 \tag{32}$$

and the expansion theorem does not hold.

It is noted that due to the periodic potential, the Bloch-Floquet theorem applies, and we can write

$$\Phi_n(\phi)=e^{ik\phi}\psi_{\alpha,k}(\phi) , \tag{33}$$

where $\psi_{\alpha,k}(\phi)$ is 2π periodic. The index is now replaced by a band index α and a wave number k is in the first Brillouin zone

$$k\in[-\tfrac{1}{2},\tfrac{1}{2}] . \tag{34}$$

The correct procedure is the following. We first Schmidt orthogonalize the set $\{\Phi_n\}$ as

$$C_n\overline{\Phi}_n=\Phi_n-\sum_{j=0}^{n-1}\langle\overline{\Phi}_j|\Phi_n\rangle\overline{\Phi}_j ,$$
$$C_n=\left[1-\sum_{j=0}^{n-1}|\langle\overline{\Phi}_j|\Phi_n\rangle|^2\right]^{1/2} . \tag{35}$$

The δ function can now be expanded as

$$\delta(\phi_{M+1}-\phi_1-2\pi Nn_0\delta)=\sum_n\overline{\Phi}_n^{*}(\phi_{M+1}-2\pi Nn_0\delta)\overline{\Phi}_n(\phi_1) . \tag{36}$$

Substituting (35) and (36) into (26) gives

$$Z_\phi=\sum_n(A_n+B_n) , \tag{37}$$

where

$$A_n=\frac{1}{C_n}\int d\phi_1\cdots\int d\phi_{M+1}\overline{\Phi}_n^{*}(\phi_{M+1}-2\pi n_0\delta)\times\prod_{i=1}^{M}K(\phi_i,\phi_{i+1})\Phi_n(\phi_1) , \tag{38}$$

$$B_n=-\frac{1}{C_n}\int d\phi_1\cdots\int d\phi_{M+1}\overline{\Phi}_n^{*}(\Phi_{M+1}-2\pi Nn_0\delta)\times\prod_{i=1}^{M}K(\phi_i,\phi_{i+1})\times\sum_{j=0}^{n-1}\langle\overline{\Phi}_j|\Phi_n\rangle\overline{\Phi}_j(\phi_1) . \tag{39}$$

Using (28) M times and (33), (38) becomes

$$A_n=\left[\frac{4\pi^2n_0^2\Delta x}{\beta v_F}\right]^{M/2}e^{-\beta L\varepsilon_{\alpha,k}}e^{ik2\pi Nn_0\delta} . \tag{40}$$

As for B_n, we need to expand

$$\overline{\Phi}_j=\sum_{h=0}^{j}g_h\Phi_h . \tag{41}$$

Substituting (41) into (39) and applying (28) M times gives

$$B_n=-\left[\frac{4\pi^2n_0^2\Delta x}{\beta v_F}\right]^{M/2}\times\frac{1}{C_n}\sum_{j=0}^{n-1}\sum_{h=0}^{j}g_h\langle\overline{\Phi}_j|\Phi_n\rangle e^{-\beta L\varepsilon_h}e^{ik2\pi Nn_0\delta}\langle\overline{\Phi}_n|\Phi_h\rangle. \tag{42}$$

It is now clear that $B_n=0$, because Φ_h is a linear combination of $\overline{\Phi}_m$ with $m\leq h\leq j\leq h-1$ all of which are orthogonal to $\overline{\Phi}_n$. From (23), (25), (37), (40), and $B_n=0$, we obtain the partition function

$$Z=\int_{-1/2}^{1/2}dk\, e^{-\beta L\varepsilon_{0,k}}e^{ik2\pi Nn_0\delta} , \tag{43}$$

where the unimportant factor

$$\left[\frac{\Delta x^2 8m^{*}\pi^2}{\beta^2 qv_F}\right]^{M/2}$$

has been omitted and we have kept only the lowest band $\alpha=0$ in the thermodynamic limit $L\rightarrow\infty$. The result (43) turns out to be identical with the previous result obtained by the unjustified procedure.

The k integral in (43) can be done by the GS analytic continuation of the chemical potential μ to the complex plane, $\beta\mu=-2\pi i\lambda$. The grand partition function is evaluated as

$$Z=\int_{-1/2}^{1/2}dk\, e^{-\beta L\varepsilon_0(k)}\int_{-\infty}^{\infty}d(n_0N\delta)e^{N\delta n_0(\beta\mu+2\pi ki)}$$
$$=e^{-\beta Ld\{\varepsilon_0(\zeta)/d\}} , \tag{44}$$

where

$$\lambda=\frac{\beta\mu}{2\pi}i\equiv\zeta i . \tag{45}$$

Of course the chemical potential is related to the soliton minus antisoliton number fixed by the charge conservation,

$$Nn_0\delta=e^{\beta\mu}\frac{\partial}{\partial e^{\beta\mu}}\ln Z . \tag{46}$$

By solving the coupled equations (29) and (46) for ζ and the lowest band energy $\varepsilon_0(\zeta)$, one can obtain the grand partition function (44).

Let us transform (29) and (46) to the forms suitable for numerical calculations. We measure the temperature in units of the soliton rest energy,

$$t\equiv\frac{T}{8sd} . \tag{47}$$

Using (14), (18), and (47), and substituting (33) with the analytic continuation (45), (29) becomes

$$\left[\frac{d^2}{d\phi^2}+f\frac{d}{d\phi}+\frac{1}{32t^2}\cos\phi\right]\psi_{\alpha,\xi}=b_{\alpha,\xi}\psi_{\alpha,\xi}\ , \tag{48}$$

where

$$f\equiv-2\xi-\frac{\pi}{2t(1+\eta)}\ , \tag{49}$$

$$\frac{\varepsilon_{\alpha,\xi}}{d}=1-32t^2\left\{b_{\alpha,\xi}+\xi^2+\frac{\pi\xi}{2t(1+\eta)}\right\}\ . \tag{50}$$

Since $\psi_{\alpha,k}$ is 2π periodic, we can expand with a large integer J

$$\psi_{\alpha,k}=\sum_{l=-J}^{J}C_l e^{il\phi} \tag{51}$$

and the eigenvalue problem (48) is reduced to a matrix eigenvalue problem

$$(-l^2+ilf)C_l+\frac{1}{64t^2}(C_{l-1}+C_{l+1})=b_{\alpha,\xi}C_l\ . \tag{52}$$

As for (46), with the use of (44), (45), and (50) it simplifies to

$$\frac{\partial}{\partial\xi}b_{0,\xi}+2\xi=0\ . \tag{53}$$

Now the skeptical reader may need to be convinced of the correctness of the GS trick. Fortunately, GM presented yet another formulation which cleverly avoids the GS trick and is certainly correct. We provide here an equivalence proof of the two formulations, thereby giving a firm basis to the remarkable trick.

In the evaluation of the grand partition function

$$\mathcal{Z}=\mathrm{Tr}\exp^{(-\beta[H-(\nu/2\pi)\{\phi(L)-\phi(0)\}])} \tag{54}$$

GM absorbs the chemical potential term into the Hamiltonian by writing it as

$$\frac{\nu}{2\pi}\{\phi(L)-\phi(0)\}=\sum_{i=1}^{M}\Delta x\frac{\nu}{2\pi}(\phi_{i+1}-\phi_i)\ , \tag{55}$$

thereby leaving the *unconditional* phase-space path integral

$$\mathcal{Z}=\int\delta\phi\int\delta\pi\,\delta(\phi_1-c)\prod_{i=1}^{M}R(\phi_i,\phi_{i+1})\ . \tag{56}$$

The term $\delta(\phi_1-c)$ with a constant c had to be introduced to avoid the divergence arising from the global translational invariance in ϕ space. The TM operator $R(\phi_i,\phi_{i+1})$ is now slightly different from $K(\phi_i,\phi_{i+1})$ and the new pseudo-Schrödinger equation reads

$$\left[\frac{d^2}{d\phi^2}-\left\{2\bar{\xi}+\frac{\pi}{2t(1+\eta)}\right\}\frac{d}{d\phi}+\frac{1}{32t^2}\cos\phi\right]\psi_{\alpha,0}=g_{\alpha,0}\psi_{\alpha,0}\ , \tag{57}$$

where $\bar{\xi}\equiv\beta\nu/2\pi$ and

$$\frac{\bar{\varepsilon}_{\alpha,0}}{d}=1-32t^2\left\{g_{\alpha,0}+\bar{\xi}^2+\frac{\pi\bar{\xi}}{2t(1+\eta)}\right\}\ . \tag{58}$$

The remaining procedure toward the grand partition function is almost the same as in the GS formulation. We have, neglecting an unimportant factor,

$$\mathcal{Z}=e^{-\beta L\bar{\varepsilon}_{0,0}}\ . \tag{59}$$

As for the chemical potential ν, it is determined by

$$2\pi Nn_0\delta=\langle\phi_{M+1}-\phi_1\rangle=-\frac{\partial}{\partial\bar{\xi}}\ln\mathcal{Z}\ , \tag{60}$$

or using (58) and (59),

$$\frac{\partial}{\partial\bar{\xi}}g_{0,0}+2\bar{\xi}=0\ . \tag{61}$$

One can now easily see that the coupled equations (57) and (61) are identical to the coupled equations (48) with (49) and (53) with the identification

$$\xi=\bar{\xi} \tag{62}$$

showing the equivalence of the two formulations.

V. THE CHARGE-DENSITY–CHARGE-DENSITY CORRELATION FUNCTION

As we will see in Sec. VI the charge-density–charge-density correlation function is directly relevant to the x-ray-scattering experiment and the NMR Knight shift experiment. It has been studied before by Turkevich and Doniach[8] without the charge conservation and with the TM procedure which is correct only for Hermitian pseudo-Hamiltonians. Unlike in the grand petition function, the correct TM result for the charge-density–charge-density correlation function is now different from the result of the incorrect TM procedure.

The charge-density–charge-density correlation function is defined by

$$H(x-x')=\langle e^{i\varphi(x)}e^{-i\varphi(x')}\rangle$$
$$=e^{i(G/n_0)(x-x')}\frac{1}{\mathcal{Z}}\sum_{n_0\delta N=-\infty}^{\infty}(e^{\beta\mu})^{n_0\delta N}Z_\pi Y\ , \tag{63}$$

$$Y=\int d\phi_1\cdots\int d\phi_{M+1}\delta(\phi_{M+1}-\phi_1-2\pi n_0\delta N)\times\prod_{i=1}^{M}K(\phi_i,\phi_{i+1})e^{i\phi_l/n_0}e^{-i\phi_m/n_0}\ , \tag{64}$$

where we have written $x=(l-1)\Delta x$, $x'=(m-1)\Delta x$, $\phi(x)=\phi_l$, and $\phi(x')=\phi_m$. Let us consider the case $x<x'$. To evaluate Y we proceed as follows. We first use the expansion theorem (36) for the δ function in (64). Here we use the Einstein convention, i.e., a sum is meant over repeated indices. Expanding

$$\bar{\Phi}_{\alpha,k}=C_{\alpha,j,k}\Phi_{j,k} \tag{65}$$

we can apply the TM equation (28) $l-1$ times. Next we expand

$$e^{i\phi_l/n_0}\Phi_{j,k}(\phi_l)=\langle\overline{\Phi}_{l,k+1/n_0}|e^{i\phi/n_0}|\Phi_{j,k}\rangle C_{l,j',k+1/n_0}\Phi_{j',k+1/n_0}\ , \tag{66}$$

which allows us to perform the next $(m-l)$ TM operations. Repeating the similar steps, we get (again omitting an unimportant factor)

$$\begin{aligned}Y=\int d\phi_{M+1}\overline{\Phi}^{*}_{\alpha,k}(\phi_{M+1}-2\pi Nn_0\delta)\Phi_{j'',k}(\phi_{M+1})C_{\alpha j,k}\\ \times\langle\overline{\Phi}_{l,k+1/n_0}|e^{i\phi/n_0}|\Phi_{j,k}\rangle C_{lj',k+1/n_0}\langle\overline{\Phi}_{l',k}|e^{-i\phi/n_0}|\Phi_{j',k+1/n_0}\rangle\\ \times C_{l'j'',k}e^{-\beta\varepsilon_{j,k}x}e^{-\beta\varepsilon_{j',k+1/n_0}(x'-x)}e^{-\beta\varepsilon_{j'',k}(L-x')}\ .\end{aligned} \tag{67}$$

We now substitute the expansion

$$\Phi_{j'',k}=d_{j''l'',k}\overline{\Phi}_{l'',k} \tag{68}$$

into (67) and perform the integration over ϕ_{M+1}. Using the property

$$d_{jl}C_{lm}=\delta_{jm} \tag{69}$$

Y reduces to

$$\begin{aligned}Y=e^{ik2\pi Nn_0\delta}\langle\overline{\Phi}_{l,k+1/n_0}|e^{i\phi/n_0}|\Phi_{j,k}\rangle C_{lj',k+1/n_0}\\ \times\langle\overline{\Phi}_{l',k}|e^{-i\phi/n_0}|\Phi_{j',k+1/n_0}\rangle C_{l'j,k}\\ \times e^{-\beta(\varepsilon_{j',k+1/n_0}-\varepsilon_{j,k})(x-x')}e^{-\beta\varepsilon_{j,k}L}\ .\end{aligned} \tag{70}$$

Substituting (70) into (63) and performing the series sum with the GS trick gives

$$\begin{aligned}H(x-x')=e^{i(G/n_0)(x-x')}\\ \times\sum_{\alpha}W(\alpha)e^{-\beta(x'-x)(\varepsilon_{\alpha,\zeta-i/n_0}-\varepsilon_{0,\zeta})}\ ,\end{aligned} \tag{71}$$

$$\begin{aligned}W(\alpha)=\sum_{m}C_{m0,\zeta}\langle\overline{\psi}_{m,\zeta}|\psi_{\alpha,\zeta-i/n_0}\rangle\\ \times\sum_{l}C_{l\alpha,\zeta-i/n_0}\langle\overline{\psi}_{l,\zeta-i/n_0}|\psi_{0,\zeta}\rangle\ .\end{aligned} \tag{72}$$

Our numerical calculation shows that $W(\alpha)$ is a real function of α and the lowest band $\alpha=0$ contribution is overwhelmingly large with the higher bands contribution being at most a few percent for the temperature ranges and commensurability strengths considered (see below). The nonunitarity of the coefficient matrix C_{ij} prevented us from finding an analytical proof of the reality of $W(\alpha)$, but we could prove a sum rule as required by putting $x=x'$ in (63) and (71),

$$\begin{aligned}\sum_{\alpha}W(\alpha)=\sum_{m}C_{m0,\zeta}\langle\overline{\psi}_{m,\zeta}|\psi_{0,\zeta}\rangle\\ =C_{00,\zeta}=1\ .\end{aligned} \tag{73}$$

The sum rule provides a good test of the numerical accuracy. For example, for $\eta=5$ and $t=0.01$ we obtain

$$\begin{aligned}\sum_{\alpha}W(\alpha)=0.9736+0.014\,83+0.011\,61+\cdots\\ =1.0000+O(10^{-5})\ ,\end{aligned} \tag{74}$$

which simply demonstrates a very high precision of our numerical calculations.

The results (71) and (72) are for $x<x'$. For $x>x'$,

$$H(x-x')=\{\text{right-hand side of (71) with } x\leftrightarrow x'\}^{*}\ . \tag{75}$$

From (71) and (75), we finally have for general x and x'

$$\begin{aligned}\langle e^{i\varphi(x)}e^{-i\varphi(x')}\rangle\\ =e^{i(G/n_0)(x-x')}\sum_{\alpha}W(\alpha)e^{-\beta|x-x'|(\mathrm{Re}\varepsilon_{\alpha,\zeta-i/n_0}-\varepsilon_{0,\zeta})}\\ \times e^{i\beta(x-x')\mathrm{Im}\varepsilon_{\alpha,\zeta-i/n_0}}\ .\end{aligned} \tag{76}$$

It is noted that the lowest band energy $\varepsilon_{0,\zeta}$ is real.

It is interesting to compare (76) with what would be obtained by the erroneous TM procedure. By letting the coefficient matrix be a unit matrix, we get the same form (76) but $W(\alpha)$ is now replaced by

$$\overline{W}(\alpha)=|\langle\psi_{\alpha,\zeta-i/n_0}|\psi_{0,\zeta}\rangle|^2\ . \tag{77}$$

Our numerical result for the case $\eta=5$ and $t=0.01$ is that

$$\begin{aligned}\sum_{\alpha}\overline{W}(\alpha)=0.9992+0.9243+0.9392+0.7285\\ +0.7351+0.4651+0.4582+0.2314\\ +0.2197+0.081+\cdots\ ,\end{aligned} \tag{78}$$

which not only violates the sum rule but totally denies the naive argument that the lowest band contribution would be dominant. It would be fair to point out that the previous TM analyses made two serious mistakes to reach the approximately correct result [put $W(\alpha)=\delta_{\alpha,0}$ in (76)] for the charge-density–charge-density correlation function: one in treating the non-Hermitian pseudo-Hamiltonian and one in neglecting the higher band contribution in (78).

VI. NUMERICAL RESULTS AND EXPERIMENTAL IMPLICATIONS

Based on the charge-density–charge-density correlation function (76), we can calculate some quantities of experimental relevance. The x-ray-scattering structure factor due to the CDW condensed electrons is given by the Fourier transform of the correlation function,

$$S(Q)=\rho_0^2\int dz\, e^{-izQ}\langle e^{i\varphi(z)}e^{-i\varphi(0)}\rangle \; . \tag{79}$$

Substituting (76) into (79) and keeping only the dominant contribution from the lowest band gives a Lorentzian line shape

$$S(Q)=\frac{\xi^{-1}}{\xi^{-2}+(Q-Q_p)^2} \; , \tag{80}$$

where the linewidth is given by

$$\xi^{-1}(\eta,t)=\beta(\mathrm{Re}\varepsilon_{0,\zeta-i/n_0}-\varepsilon_{0,\zeta}) \; , \tag{81}$$

which also describes the charge-density inverse correlation length and the peak position is given by

$$Q_p(\eta,t)=\frac{G}{n_0}+\beta\,\mathrm{Im}\varepsilon_{0,\zeta-i/n_0} \; . \tag{82}$$

The coupled equations (52) and (53) have been solved by an iteration method. We find that the 51×51 matrix size is sufficient to make the calculation convergent for the parameter ranges considered. Figure 2 shows the chemical potential in units of the rest soliton energy $\mu/(8sd)$ versus the reduced temperature t_n, which is related to t as

$$t=\frac{11}{1+\eta}t_n \; . \tag{83}$$

The reduced temperature t_n is simply the temperature measured in units of the rest soliton energy at $\eta=10$. The result Fig. 2 is physically reasonable. For larger η, the system has a stronger tendency toward a less charged state, and to conserve the net charge, one needs to apply a stronger external force leading to a larger μ. Since this tendency is thermally weakened, μ must decrease with temperature.

In Fig. 3, we have plotted the inverse correlation length in units of the inverse lattice constant

$$a\xi^{-1}=\frac{(1+\eta)\delta}{8t}\left[\mathrm{Re}\frac{\varepsilon_{0,\zeta-i/n_0}}{d}-\frac{\varepsilon_{0,\zeta}}{d}\right] \; . \tag{84}$$

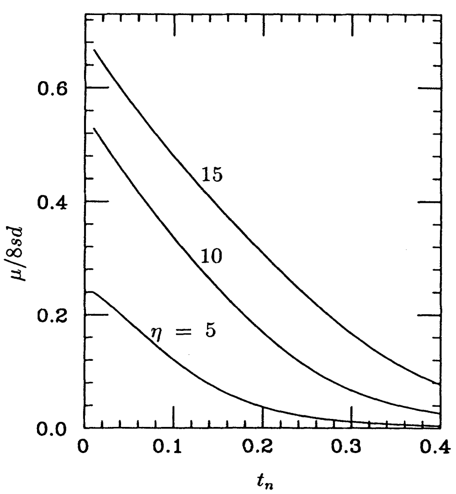

FIG. 2. Chemical potential vs temperature.

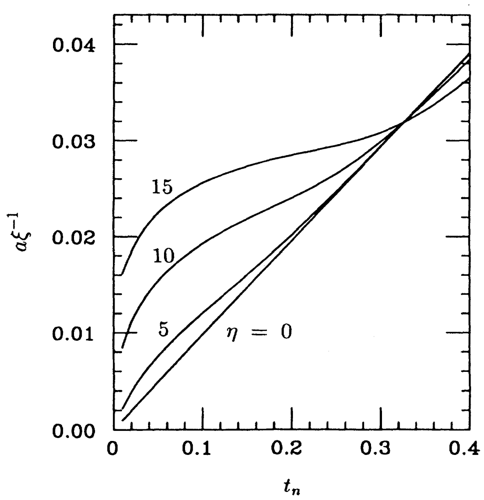

FIG. 3. Inverse correlation length vs temperature for the charge conserved case.

In Fig. 4, we have plotted the peak position measured from G/n_0 again in units of the inverse lattice constant

$$a\Delta Q\equiv a\left[Q_p-\frac{G}{n_0}\right]=\frac{(1+\eta)\delta}{8t}\mathrm{Im}\frac{\varepsilon_{0,\zeta-i/n_0}}{d} \; . \tag{85}$$

The corresponding results, Fig. 5 for $a\xi_{-1}$ and Fig. 6 for $a\Delta Q$, of Turkevich and Doniach[8] can be obtained by simply letting $\zeta=0$, thereby removing the charge conservation constraint.

In the charge nonconserved case Fig. 5, the decrease of correlation length starts at higher temperature for larger η. This is simply because the soliton energy is larger for larger η and a higher temperature is necessary to thermally create solitons and antisolitons. In contrast, the charge conserved case Fig. 3 shows an opposite ten-

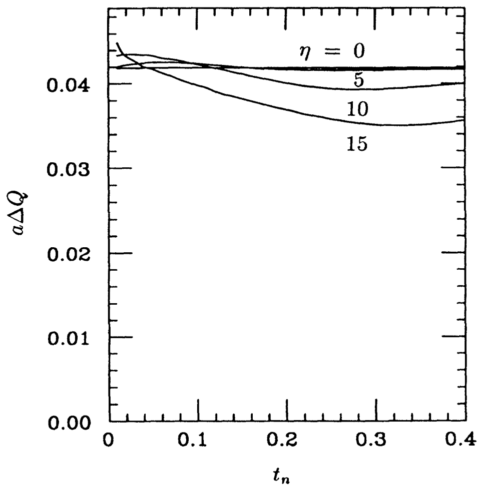

FIG. 4. Scattering peak position vs temperature for the charge conserved case.

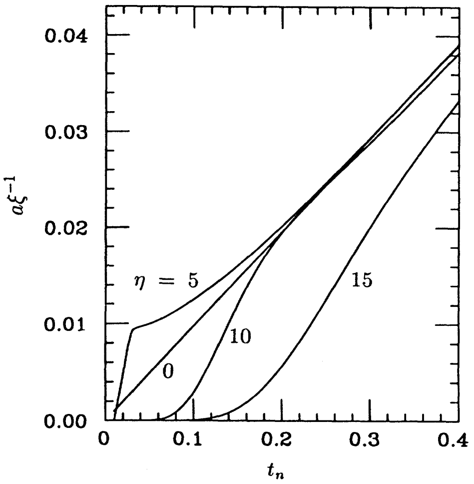

FIG. 5. Inverse correlation length vs temperature for the charge nonconserved case.

dency; the correlation length decreases more rapidly with temperature for large η. The fragility of the $T=0$ soliton lattice states was pointed out before by GS (Ref. 4) and GM.[5] Our result shows that the stronger the commensurability, the more fragile the soliton lattice. As for the scattering peak position, in the charge nonconserved case Fig. 6, the deviation from the commensurate wave number starts at higher temperatures for larger η. This is reasonable because for stronger commensurability the soliton energy is larger, and therefore the ground state is less charged and more stable against temperature. The result for the charge conserved case Fig. 4 is again in sharp contrast with the $\mu=0$ case. The scattering peak does not converge toward G/n_0 in the same temperature range as in Fig. 6. However, since at $T=0$ and for large η the system is predominantly near the commensurate state, the peak must be located near G/n_0 at $T=0$. As in the case of correlation length, the result can be naturally understood in terms of the fragility of the soliton lattice states. It is noted that $\mu \sim 0$ for all t at $\eta=0$ and thus the two cases give similar results.

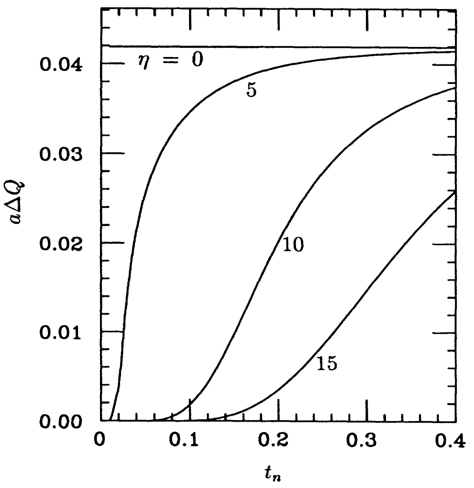

FIG. 6. Scattering peak position vs temperature for the charge nonconserved case.

We consider here the two experimental results on the transition-metal dichalcogenide layer compound 2H-TaSe$_2$. One is the x-ray-scattering experiment[16] reporting an abrupt change, about 2%, of the CDW wave number at the C-IC transition. The other is the NMR Knight shift experiment[13] reporting no abrupt change in the signal at the C-IC transition. The consistency of our result for the temperature dependence of the scattering peak position, Fig. 4, with the x-ray-scattering experiment is obvious. As for the Knight shift experiment, it was regarded as a strong support to McMillan's argument that an IC-CDW state is understood as commensurate regions separated by discommensurations. This is nothing but our soliton lattice at $T=0$. Clearly, the argument of Suits, Couturié, and Slichter[13] is missing an important point. That is, in order for their argument to remain correct, the charge-density correlation length must be much larger than the unit cell size of the C-CDW state, i.e., $n_0 a$. Our result for the temperature dependence of the correlation length shows that, although the soliton lattice state is certainly fragile, the correlation length is still much larger than $n_0 a$, implying a smooth change in the NMR spectra at the C-IC transition.

VII. SUMMARY

We have started with the commensurability pinning Hamiltonian for the phase mode of the charge-density wave with the charge conservation as a microscopic model Hamiltonian for the C-IC CDW transition. As a result, unlike in the GL free-energy functional approaches, we have an idea about the size of the system parameters and the experimentally relevant temperature range, although as a common drawback of simplified one-dimensional models, we must reinterpret $T=0$ as the transition temperature T_c. Charge conservation plays an important role both in the ground state and in thermal fluctuations. McMillan interpreted a soliton lattice as an IC-CDW state and a flat vacuum a C-CDW state. Due to the charge conservation, the ground state is always a soliton lattice, but its character continuously changes between commensurate and incommensurate according to the commensurability strength.

Our study of thermal fluctuations consists of two parts. One is technical, extending the existing TM formulation to the non-Hermitian pseudo-Hamiltonian systems. We have also proved the equivalence of the GS formalism and the GM formalism, thereby giving a firm basis to the remarkable trick of analytic continuation of the chemical potential to the complex space. The other is physical. We have seen that the charge conservation leads to results qualitatively different from the previous analyses without the charge conservation. Although one cannot disprove the possible existence of a charge reservoir in certain CDW systems which might support the latter approach, the x-ray-scattering experiment reporting an abrupt jump in the CDW wavelength at the C-IC transition is consistent with our charge conserved analysis. We have also made an important point that the smooth change in the NMR spectra at the C-IC transition requires a long-enough spatial correlation in the charge density, again in consistency with our results for the correlation length.

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[1] W. L. McMillan, Phys. Rev. B **12**, 1187 (1975); **12**, 1197 (1975).

[2] M. B. Walker and A. E. Jacobs, Phys. Rev. B **24**, 6770 (1981); M. Yamashita, Prog. Theor. Phys. **74**, 622 (1985); H. Yamamoto, I. Nakayama, and T. Ohmi, *ibid.* **59**, 351 (1978); A. D. Bruce, R. A. Cowley, and A. F. Murray, J. Phys. C **11**, 3591 (1978); P. Bak and V. J. Emery, Phys. Rev. Lett. **36**, 978 (1976).

[3] D. J. Scalapino, M. Sears, and R. A. Ferrell, Phys. Rev. B **6**, 3409 (1972); J. A. Krumhansl and J. R. Schrieffer, *ibid.* **11**, 3535 (1975); J. F. Currie, J. A. Krumhansl, A. R. Bishop, and S. E. Trullinger, *ibid.* **22**, 477 (1980); T. Schneider and E. Stoll, *ibid.* **22**, 5317 (1980).

[4] N. Gupta and B. Sutherland, Phys. Rev. A **14**, 1790 (1976).

[5] R. A. Guyer and M. D. Miller, Phys. Rev. A **17**, 1205 (1978).

[6] S. A. Brazovskii, I. E. Dzyaloshinskii, and S. A. Obukhov, Zh. Eksp. Teor. Fiz. **72**, 1550 (1977) [Sov. Phys.—JETP **45**, 814 (1977)].

[7] Y. Okwamoto, H. Takayama, and H. Shiba, J. Phys. Soc. Jpn. **46**, 1420 (1979).

[8] L. A. Turkevich and S. Doniach, Ann. Phys. (N.Y.) **139**, 343 (1982).

[9] P. A. Lee, T. M. Rice, and P. W. Anderson, Solid State Commun. **14**, 703 (1974).

[10] H. Fukuyama, J. Phys. Soc. Jpn. **41**, 513 (1976); S. Kurihara, *ibid.* **41**, 1488 (1976).

[11] M. J. Rice, A. R. Bishop, J. A. Krumhansl, and S. E. Trullinger, Phys. Rev. Lett. **23**, 432 (1976).

[12] F. Denoyer, R. Comés, A. F. Garito, and A. J. Heeger, Phys. Rev. Lett. **35**, 445 (1975).

[13] B. H. Suits, S. Couturié, and C. P. Slichter, Phys. Rev. Lett. **45**, 194 (1980).

[14] R. A. Guyer and M. D. Miller, Phys. Rev. Lett. **42**, 718 (1979); see Y. Okwamoto, J. Phys. Soc. Jpn. **48**, 1836 (1980) for a charge nonconserved analysis.

[15] J. E. Sacco, A. Widom, and J. B. Sokoloff, J. Stat. Phys. **21**, 497 (1979).

[16] I. A. Holy, M. V. Klein, W. L. McMillan, and S. F. Mey, Phys. Rev. Lett. **37**, 1145 (1976).