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Electrical conductivity of the narrow-half-filled-band Hubbard model with nearest-neighbor interaction

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The recent theoretical work on the one-dimensional Hubbard model showed this to be inadequate to explain the properties of n-methyl phenazinium tetracyanoquino dimethane (NMP-TCNQ) in the regime of narrow bandwidth compared to Coulomb repulsion; however, it also supported the suggestion existing in the literature that it is possible to fit both the magnetic susceptibility and the low-T activation energy of the electrical conductivity by introducing a temperature dependence in the parameters of the Hamiltonian. Since the Hubbard Hamiltonian neglects important interactions (long-range Coulomb repulsion, electron-lattice interaction, etc.), it is reasonable to think that these interactions may be responsible for this temperature dependence. In this paper I add to the Hubbard Hamiltonian a nearest-neighbor Coulomb interaction and calculate the electrical conductivity in the narrow-bandwidth regime.

I. INTRODUCTION

The recent theoretical work[1–4] on the so-called "extended" or "modified" Hubbard model was motivated by a rather wide range of reasons. Before reviewing them briefly, let me first define the model in question. The Hamiltonian in one dimension is written as follows:

$$H=b\sum_{i,\sigma}(c^\dagger_{i\sigma}c_{i+1\sigma}+c^\dagger_{i+1\sigma}c_{i\sigma})+U\sum_i n_{i\uparrow}n_{i\downarrow}+V\sum_i n_i n_{i+1}-\mu\sum_{i,\sigma}n_{i\sigma}\,, \qquad (1)$$

where $c^\dagger_{i\sigma}$, $c_{i\sigma}$ are the creation and destruction operators for electrons in the Wannier function W_i centered at site i and spin σ ($\sigma=\uparrow,\downarrow$); $n_{i\sigma}=c^\dagger_{i\sigma}c_{i\sigma}$ is the corresponding occupation number operator ($n_{i\sigma}=0,1$) and $n_i=n_{i\uparrow}+n_{i\downarrow}$. Periodic boundary conditions are used: $c^\dagger_{N_a+i\sigma}=c^\dagger_{i\sigma}$ (N_a = number of sites of the chain). The first term of Eq. (1) is the noninteracting particle energy in the tight-binding approximation; this term gives rise to a band of width $w=4b$ if the interaction is neglected. The second term represents the Coulomb repulsion between two electrons on the same site and the third term represents the Coulomb repulsion between the electronic charges on nearest-neighbor sites. Finally, the last term is μN ($N=\sum_{i\sigma}n_{i\sigma}$), where μ is the chemical potential and N is the total number operator. b, U and V are the positive parameters of the model Hamiltonian. Throughout this paper μ is chosen in such a way that the grand canonical thermal average

$$\langle N\rangle=\sum_{i\sigma}\langle n_{i\sigma}\rangle=\frac{\mathrm{tr}e^{-\beta H}N}{\mathrm{tr}e^{-\beta H}}=N_a$$

equals the number of sites of the chain (half-filled band). Equation (1) without the third term reduces to the usual Hubbard Hamiltonian[5] in one dimension.

Bari[1] first studied the thermodynamics of Hamiltonian (1) in three dimensions with bandwidth $\equiv 0$, using a Green's-function decoupling technique and starting from the variational principle. His aim was to study the role of electron lattice interactions in a narrow half-filled band, which he can effectively include in the interaction part of Eq. (1). He first pointed out[1] that for large enough V/U the system might undergo a phase transition to a so-called charge-ordered state at low temperature, with the sites of sublattice A doubly occupied and the sites of sublattice B empty (the total lattice is made up of the two equivalent sublattices A and B); on the other hand, for V/U less than a critical value no such transition occurs and the ground state has each site singly occupied. Bari[1] showed that in one dimension there is no phase transition even when the ground state has alternating doubly occupied and unoccupied sites (for V/U greater than the critical value). Ihle and Lorentz[4] performed a study similar to Bari's work, by using a more sophisticated decoupling scheme for the one-particle Green's function, and found similar results.

Beni and Pincus[2] (BP) and Tu and Kaplan[3] (TK) studied in detail the equilibrium statistical mechanics of Hamiltonian (1) in one dimension with $b\equiv 0$. Both groups of authors were motivated by the possible connection of the theory with experiments on the organic solid n-methyl phenazinium tetracyanoquino dimethane (NMP-TCNQ). Several theoretical works[6–8] had been performed previously to analyze the hypothesis advanced by Heeger and coworkers[9,10] that magnetic susceptibility, specific heat, and electrical conductivity of NMP-TCNQ (Ref. 9) could be explained by the simple half-filled-band Hubbard model. The results of those works,[6–8] suggested, in general, that the Hubbard Hamiltonian is not sufficient to explain the behavior of NMP-TCNQ. Without entering into details, I mention only that the most striking and convincing result is contained in Refs. 6 and 7 where Cabib and Kaplan show the drastic disagreement of the magnetic susceptibility measurements with the theoretical results obtained within the Hubbard model: namely, the existence in the theory, contrary to experiment, of a peak, and the very different magnitude of the moment in the Curie–Weiss region.

The work of BP[2] and of TK[3] go beyond the Hubbard model. The first group[2] argued that the existence of a coupling between the electronic charge on the TCNQ molecules and the induced dipole moments on the NMP molecules can be effectively written in the form of interchain repulsion between the electronic charges on TCNQ in the same way as the third term of Eq. (1) (besides reducing the on-site Coulomb repulsion). The second group[3] argued that the presence of a nearest-neighbor Coulomb repulsion might reduce appreciably the moment in the Curie–Weiss region and therefore might tend to improve the comparison with experiment; however, they found that their results contradicted this expectation.

BP and TK are concerned with the study of the static properties of the Hamiltonian (1) in the $b=0$ limit. They calculated specific heat,[2,3] magnetic susceptibility,[2,3] and density–density correlation functions[2] exactly in one dimension in the grand canonical ensemble, by use of the transfer-matrix method.[11]

In the present work, I am concerned with the calculation of a nonequilibirum quantity, the frequency-dependent electrical conductivity, or linear response to an external electric field. The motivation is twofold: Kaplan and Cabib[8] showed that it is possible to fit simultaneously the magnetic susceptibility and the low-temperature gap of the dc conductivity of NMP–TCNQ with the Hubbard model if a temperature dependence is introduced in the parameters b and U. They also stressed[8] that it is reasonable that interactions neglected by the Hubbard Hamiltonian may be responsible for this temperature dependence of the parameters. A calculation of the dc conductivity by use of Hamiltonian (1) might give better insight about the role of the Coulomb repulsion when its range is longer than just intrasite.

The second motivation is purely theoretical. A calculation of the frequency-dependent conductivity requires the knowledge of the time-dependent two-particle correlation function. BP calculated the time-independent average $\langle n_i n_{i+k}\rangle$ with Hamiltonian (1) in the limit $b=0$ exactly; the calculation of the time-dependent correlation function $\langle c_i^\dagger(t)\, c_j(t) c_k^\dagger c_l\rangle$ can also be performed exactly in the same limit in a manner similar to $\langle n_i n_{i+k}\rangle$, even though it involves more tedious work. In a very recent paper,[12] I showed this type of calculation in a model quite simpler than Eq. (1). Qualitatively, the result was very similar to the one obtained by Bari and Kaplan,[13] who were concerned with the narrow-half-filled-band Hubbard model: in short, (1) existence of optical absorption and emission at ω equals first excited energy minus ground-state energy, and (2) energy-activated dc conductivity.

The more complicated Hamiltonian (1) gives a much more complex structure of the absorption spectrum owing to the presence of the nearest-neighbor Coulomb repulsion. One of my checks of the calculation was to recover Bari and Kaplan's[13] expression in the limit, nearest-neighbor interaction $\rightarrow 0$.

In Sec. II, I show the calculation and discuss the results. In the Appendix, I show some of the details of the calculation.

II. CONDUCTIVITY

The fundamental formula of Kubo[14] for the linear response to an external electric field yields the frequency-dependent electrical conductivity tensor as a Fourier transform of the time-dependent current–current correlation function in thermal equilibrium:

$$\sigma_{\mu\nu}(\omega)=\frac{1}{\mathcal{V}}\int_0^\infty dt\, e^{-i\omega t}\int_0^\beta d\lambda\,\langle J_\nu(-i\lambda)\, J_\mu(t)\rangle\,, \qquad (2)$$

$\hbar=1$. If A is an operator, we define the thermal average

$$\langle A\rangle=\frac{\operatorname{tr} e^{-\beta H}A}{\operatorname{tr} e^{-\beta H}}=\operatorname{tr}\rho A\quad\left(\rho=\frac{e^{-\beta H}}{\operatorname{tr} e^{-\beta H}}\right)\,. \qquad (3)$$

$\mathcal{V}$ is the volume of the system, $\beta=1/kT$, J_ν is the ν component of the current operator,

$$A(t)=e^{iHt}A\,e^{-iHt}\,, \qquad (4)$$

and H is the Hamiltonian of the system.

It is easy to show that the real part of Eq. (2) can be rewritten as[13]

$$R(\sigma_{\mu\nu}(\omega))=\frac{1}{\mathcal{V}}\int_{-\infty}^{\infty} dt\, e^{i\omega t}\int_0^\beta d\lambda\,\operatorname{tr}[\rho J_\nu J_\mu(t+i\lambda)]\,. \qquad (5)$$

[To show Eq. (5) from Eq. (2), one uses $R(\sigma_{\mu\nu})=R(\sigma_{\mu\nu})^*$, the transformation of the integration variable $\lambda'=\beta-\lambda$ and the invariance of the trace under rotational permutation of the operators.]

We define[15]

$$J=-ieab\sum_{\substack{i=1\\ \sigma=\uparrow,\downarrow}}^{N_a}(c_{i\sigma}^\dagger c_{i+1\sigma}-c_{i+1\sigma}^\dagger c_{i\sigma})\,. \qquad (6)$$

Clearly, in Eq. (6) we consider only the component of the vector $\vec{J}$ along the chain, whose lattice parameter is a. If we use Eq. (6) in Eq. (5) with $x=t+i\lambda$, we obtain

$$\sigma(\omega) = -\frac{e^2 b^2 a^2}{\mathcal{V}} \sum_{\substack{ij\\ \sigma\sigma'}} \int_{-\infty}^{\infty} dt\, e^{i\omega t} \times \int_0^\beta d\lambda\, \mathrm{tr}\left[\rho(c^\dagger_{i\sigma} c_{i+1\sigma} - c^\dagger_{i+1\sigma} c_{j\sigma})\, e^{iHx} \times (c^\dagger_{j\sigma'} c_{j+1\sigma'} - c^\dagger_{j+1\sigma'} c_{j\sigma'})\, e^{-iHx}\right] . \tag{7}$$

Now we note the following:

(i) In order for (7) to be different from zero, b must not be 0. This is because the band term of the Hamiltonian provides the mechanism for the hopping of an electron from site to site,

(ii) It is very difficult to calculate the trace in Eq. (7) exactly for any b/U and b/V,

(iii) In lowest order of b/U and b/V one can neglect entirely the band term of the Hamiltonian for the purpose of calculating the trace inside the double integral of (7), thus making the problem exactly soluble, but of course limiting the validity of the calculation to the regime $b/U,\ b/V \ll 1$, $kT \gg b^2/U$.[13]

To calculate the trace of (7) with $b=0$, we can simplify the double sum into a single sum, by noting that now $[n_{i\sigma}, H] = 0$ and the eigenstates of H are single Slater determinants with Wannier functions occupied[13]:

$$\sigma(\omega) = \frac{e^2 b^2 a^2}{\mathcal{V}} \sum_{i\sigma} \int_{-\infty}^{\infty} dt\, e^{i\omega t} \times \int_0^\beta d\lambda\, \mathrm{tr}\left[\rho(c^\dagger_{i\sigma} c_{i+1\sigma} c^\dagger_{i+1\sigma}(x)\, c_{i\sigma}(x) + c^\dagger_{i+1\sigma} c_{i\sigma} c^\dagger_{i\sigma}(x)\, c_{i+1\sigma}(x))\right] . \tag{8}$$

The two terms within the square brackets of (8) give the same contribution to σ. To see this, I introduce the operator θR, where R reverses the spin and θ is the time-reversal operator. Obviously $[\theta R, H] = 0$, and it is easy to show the equality of the two terms by performing the transformation of integration variable $\lambda' = \beta - \lambda$ in the expression:

$$\int_{-\infty}^{\infty} dt\, e^{i\omega t} \int_0^\beta d\lambda\, \mathrm{tr}[\rho\theta R c^\dagger_{i\sigma} c_{i+1\sigma} c_{i+1\sigma}(x)\, c_{i\sigma}(x) (\theta R)^{-1}] , \tag{9}$$

$$\theta R c^\dagger_{i\sigma} (\theta R)^{-1} = c^\dagger_{i\sigma}, \quad \theta i \theta^{-1} = -i .$$

The same argument applies to the sum on i, and $\sigma = \uparrow, \downarrow$ (one has to choose the appropriate symmetries of the Hamiltonian) so that

$$\sigma(\omega) = \frac{4e^2 b^2 a^2 N_a}{\mathcal{V}} \int_{-\infty}^{\infty} dt\, e^{i\omega t} \times \int_0^\beta d\lambda\, \mathrm{tr}\left[\rho\, c^\dagger_{i+1\uparrow}(-x)\, c_{i\uparrow}(-x)\, c^\dagger_{i\uparrow} c_{i+1\uparrow}\right] . \tag{10}$$

From the equation of motion,

$$\frac{d}{dx} c^\dagger_{i\uparrow}(-x) = -i\,(U n_{i\downarrow} + V(n_{i+1} + n_{i-1}))\, c^\dagger_{i\uparrow}(-x) , \tag{11}$$

we have [with $c^\dagger_{i\uparrow}(0) = c^\dagger_{i\uparrow}$]

$$c^\dagger_{i\uparrow}(-x) = \exp[-i(U n_{i\downarrow} + V(n_{i-1} + n_{i+1}))\,x]\, c^\dagger_{i\uparrow} . \tag{12}$$

It is now straightforward to see that

$$c^\dagger_{i+1\uparrow}(-x)\, c_{i\uparrow}(-x)\, c^\dagger_{i\uparrow} c_{i+1\uparrow} = \exp[i(U - V)(n_{i\downarrow} - n_{i+1\downarrow})\,x + iV(n_{i-1} - n_{i+2})\,x] \times n_{i+1\uparrow}(1 - n_{i\uparrow}) . \tag{13}$$

Here I have used the identities

$$e^{-iVn_{i\uparrow}x} c_{i\uparrow} = c_{i\uparrow} , \tag{14}$$

$$c^\dagger_{i+1\uparrow} e^{iVn_{i+1\uparrow}x} = c^\dagger_{i+1\uparrow} . \tag{15}$$

After substituting Eq. (13) into Eq. (10), we can use the transfer-matrix method[11] to calculate the trace. The procedure is the same as the one used by Cabib[12] with the Hamiltonian of Cullen and Callen[16]; BP[2] used it in the easier case of the time-independent correlation function, which does not include the exponential factor in (13).

In the Appendix, I will show the details of the calculation of the trace in (10), whereas here I display and discuss the results.

The final expression for the dc conductivity in the general case $U,\ V \neq 0$ is

$$\sigma(\omega = 0) = \frac{4e^2 b^2 a^2 N_a}{\mathcal{V}}\, 2\pi\beta\, \frac{2x_1^2 x_0^2}{\lambda_1^2} \times \{1 + y^{-2} + y(\lambda_1 - 1 - y^{-2})^2\}\, \delta(\omega) , \tag{16}$$

where

$$x_0 = e^{\beta(U/4+V)} , \tag{17}$$

$$y = e^{-\beta V} , \tag{18}$$

$$x_1 = \frac{1}{\sqrt{2}} \frac{\lambda_1 - 2x_0^2 y}{(4x_0^2 + (\lambda_1 - 2x_0^2 y)^2)^{1/2}} , \tag{19}$$

$$\lambda_{1,2} = \tfrac{1}{2}\{p \pm (p^2 - q)^{1/2}\} , \tag{20}$$

$$p = 1 + e^{2\beta V} + 2\, e^{\beta(V+U/2)} , \tag{21}$$

$$q = 8\, e^{\beta(V+U/2)} (1 - e^{\beta V})^2 \tag{22}$$

(I have followed the notation of BP[2]).

One also gets emission and absorption lines at the frequencies $\omega = U,\ V,\ 2V,\ U - V,\ U + V,\ U - 2V,\ U - 3V,\ U - 3V$ each of them with different temperature dependence. These energies (and the ones with opposite sign $-U,\ -V,\ -2V, \ldots$) correspond to all the possible changes in energy due to the hopping of one electron from a site to a neighbor one. Of course, if one wants to check the limits $U \to 0$ and $V \to 0$, of the dc contribution to σ, one cannot simply put $U=0$ or $V=0$ in Eq. (16) because some of the absorption and emission lines contribute to the dc conductivity in either of the two limits. We checked that when $V \to 0$ we recover the result obtained in the Hubbard model.[13]

Let us examine briefly the low-temperature behavior of Eq. (16). One has to consider separately the two cases $U/2 > V$ and $U/2 < V$.

A. $U/2 > V$

The ground state has one electron on each site,[1] with energy $E_0 = (N_a - 1)V$, and the lowest excited state has one doubly occupied site and one unoccupied site, with energy $E_0 + U - V$. At very high temperature σ decreases as $1/kT$. The low-temperature behavior is of the form

$$\sigma \propto e^{-\beta U/2}, \tag{23}$$

with an activation energy $\Delta = U/2$. Δ is therefore different from gap/2, as one would expect in an ordinary semiconductor, and as one finds in the Hubbard[13] and in the Cullen and Callen model.[12] Equation (23) shows also the rather surprising result that V does not affect the activation energy, which is equal to $U/2$ and is independent of V.

B. $U/2 < V$

Now the ground state has alternate unoccupied and doubly occupied sites,[1] with an energy $E_0 = N_a U/2$ and a first excited energy $= E_0 - U + 3V$ that corresponds to breaking one pair on one site by transferring one electron to a nearest-neighbor site. The existence of this charge density wave ground state has been of interest[1,17] recently, and it occurs also for $U < 0$, $V = 0$.

The dc contribution to the conductivity is asymptotically $\propto e^{-\beta(2V - U/2)}$ at low temperature. Also in this case the activation energy is different from gap/2, and at high temperature is $\propto 1/kT$. The reason for this dependence of the activation energy on the parameters U and V is not clear, although it is interesting to note the following: in both cases $U/2 > V$ and $U/2 < V$, the activation energy Δ is equal (within the canonical ensemble) to $(E_2 - E_0)/2$, where E_0 is the ground-state energy and E_2 is the energy of the lowest state, which is $\sim N^2$-fold degenerate (the first excited state E_1 is $\sim N$-fold degenerate). This is true also for the activation energy of the conductivity of the Cullen and Callen Hamiltonian[12]: there E_1 is already $\sim N^2$-fold degenerate (as is E_2) and $\Delta = (E_1 - E_0)/2$. Furthermore within the grand canonical ensemble, again for both $U/2 > V$ and $U/2 < V$, $\Delta = E_1 - E_0$, where E_0 and E_1 are the lowest and next-to-lowest eigenvalues of $H - \mu N$. This latter remark seems to be less general than the former, because it does not apply to the Cullen and Callen model.

We point out finally that some of the ac contributions to σ may not vanish at 0 temperature, while most of them do. For $U/2 > V$ the transition of energy $U - V$ is finite at $T = 0$, and for $U/2 < V$, the one of energy $3V - U$ is finite at $T = 0$. This is easily understood physically if one considers the difference between the energies of the ground state and the first excited state in the two cases $U/2 > V$ and $U/2 < V$.

APPENDIX

To calculate the trace in Eq. (10) in the thermodynamic limit ($N_a \to \infty$) with $\langle N \rangle = N_a$ (half-filled band), one needs the transfer matrix[2]

$$P = \begin{pmatrix} 1 & x_0 & x_0 & y^{-2} \\ x_0 & x_0^2 y & x_0^2 y & x_0 \\ x_0 & x_0^2 y & x_0^2 y & x_0 \\ y^{-2} & x_0 & x_0 & 1 \end{pmatrix}. \tag{A1}$$

(In the half-filled band w_0 of Ref. 2 is equal to $x_0^{-2} y^{-2}$). x_0 and y are defined by Eqs. (17) and (18). One also needs the following five matrices:

$$P_1 = PT_1, \tag{A2}$$

$$P_2 = T_1 P T_2, \tag{A3}$$

$$P_3 = T_3 P T_4, \tag{A4}$$

$$P_4 = P_2^\dagger, \tag{A5}$$

$$P_5 = P_1^\dagger, \tag{A6}$$

where

$$T_1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & e^{iVx/2} & 0 & 0 \\ 0 & 0 & e^{iVx/2} & 0 \\ 0 & 0 & 0 & e^{iVx} \end{pmatrix}, \tag{A7}$$

$$T_2 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & e^{i(U-V)x/2} & 0 \\ 0 & 0 & 0 & e^{i(U-V)x/2} \end{pmatrix}, \tag{A8}$$

$$T_3 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & e^{i(U-V)x/2} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \tag{A9}$$

$$T_4 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & e^{-i(U-V)x/2} \end{pmatrix}. \tag{A10}$$

So we have

$$W = \mathrm{tr}\,[\rho\, c^\dagger_{i+1\uparrow}(-x)\, c_{i\uparrow}(-x)\, c^\dagger_{i\uparrow}\, c_{i+1\uparrow}]$$

$$= \frac{1}{Z}\,\mathrm{tr}\,(P^{i-3} P_1 P_2 P_3 P_4 P_5 P^{N_a - i - 2}) \;. \tag{A11}$$

Z is the partition function

$$Z = \mathrm{tr}\, e^{-\beta H} = \mathrm{tr}\, P^{N_a} = \lambda_1^{N_a} \;. \tag{A12}$$

Equation (A12) is valid when $N_a \to \infty$, and λ_1 is the maximum eigenvalue of P defined by Eqs. (20)–(22). Using the rotational invariance of the trace, we can write

$$W = \frac{1}{Z}\,\mathrm{tr}\,(S P^{N_a - 5} S^{-1} S P_1 P_2 P_3 P_4 P_5 S^{-1}) \;, \tag{A13}$$

where S diagonalizes P. S is easily written, once the eigenvectors of P are known:

$$S = \begin{pmatrix} x_1 & y_1 & y_1 & x_1 \\ x_2 & y_2 & y_2 & x_2 \\ 1/\sqrt{2} & 0 & 0 & -1/\sqrt{2} \\ 0 & 1/\sqrt{2} & -1/\sqrt{2} & 0 \end{pmatrix} , \tag{A14}$$

where x_1 is defined in Eq. (19), and

$$y_1 = \frac{2 x_0 x_1}{\lambda_1 - 2 x_0^2 y} \;. \tag{A15}$$

x_2 and y_2 are obtained from x_1, y_1, by replacing λ_1, with λ_2 [Eq. (20)]. After performing the multiplication of the matrices in (A13), one has to use W in Eq. (10) where the integrations are trivial.

The final complete expression for Eq. (A11) is not of great interest and it is quite complicated. The dc part in the general case U, $V \neq 0$ is given in Eq. (16).

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