Spectral function of a quarter-filled one-dimensional CDW insulator

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We consider a one-dimensional charge density wave (CDW) insulator formed by Umklapp processes in a quarter-filled band. The spectrum of the model consists of gapless, uncharged excitations carrying spin \( \pm 1/2 \) (spinons) and gapped, spinless excitations carrying charge \( \mp e/2 \) (solitons and antisolitons). We calculate the low-energy behaviour of the single-electron Green’s function at zero temperature. The spectral function exhibits a featureless scattering continuum of two solitons and many spinons. The theory predicts that the gap observed by Angle Resolved Photoemission (ARPES) is twice the activation gap in the dc conductivity. We comment on possible applications to PrBa\(_2\)Cu\(_3\)O\(_7\) and to the Bechgaard salts.

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The TMTSF and TMTTF families of quasi one dimensional (1D) organic conductors have attracted much attention over the last 30 years. They feature a rich variety of 3D ordered ground states, including spin and charge density waves, spin-Peierls insulators and superconductors. Recently much of the attention has focussed on the 1D phases \[1–3\] at low temperatures, which are believed to realize exotic strongly correlated Mott or CDW insulating states. With regard to a theoretical description of these phases there are two main points of controversy. Firstly, it is not clear how “one-dimensional” in particular the TMTSF salts are in the relevant regime of energies/temperatures, and secondly it is unknown how strong the effective electron-electron interaction at low energies is. Persuasive arguments in favour of strong interactions, giving rise to a small value of the Luttinger liquid parameter \( K_c \), are given in \[1–3\]. The authors of \[2\] quote an estimate of \( K_c \approx 0.22 \). At such values of \( K_c \) the single-particle tunneling between the chains should be strongly suppressed, leading to the enhancement of 1D effects. Perhaps, the strongest evidence in favour of such a scenario comes from ARPES measurements \[4–6\]. Experiments done by different groups not only show the absence of quasi-particle peaks, but find no evidence of any dispersing features in the single-electron spectral function. Quite similar behaviour has recently been observed in the quarter-filled Cu – O chain material PrBa\(_2\)Cu\(_3\)O\(_7\) (“P123”) \[6–8\].

The great difficulty in determining dynamical response functions theoretically is the presence of Umklapp scattering processes, which dynamically generate a spectral gap. In this work we use a method based on the integrability of the low-energy theory to calculate the single-particle Green’s function for the first time. In the Bechgaard salts there are two separate mechanisms that lead to Umklapp scattering: (1) “double” Umklapp processes due to the commensurate band filling 1/4 \[\beta = 1\]. These generate a gap only for strong interactions \( (K_c < 0.25) \). (2) a small dimerization, which halves the Brillouin zone and gives rise to “single” Umklapp processes \[\beta = 0.5\]. These open a gap already for weak interactions \( K_c < 1 \) but their coupling constant is proportional to the dimerization and thus small. It is an open question which of these two mechanisms dominates in the Bechgaard salts. Here we develop a theory for ARPES in the case where one of the processes can be neglected. We consider the case with only double Umklapp scattering in detail and present some results for the dimerized case at the end. Following \[\beta = 1\] we adopt a field theory description of the quarter-filled CDW insulator at low energies. The small parameter is this approach is the ratio of the gap to the bandwidth \( W \). The underlying lattice model may be thought of as a quarter-filled Hubbard model with additional density-density interactions \[\beta \leq 1\]. The Lagrangian density is the sum of two terms describing the spin and charge degrees of freedom respectively (“spin-charge separation”) \( \mathcal{L} = \mathcal{L}_s + \mathcal{L}_c \), where

\[
\mathcal{L}_s = \frac{1}{16\pi} \left[ v_s^{-1} (\partial_x \Phi_s)^2 + v_s (\partial_x \Phi_s)^2 \right],
\]

\[
\mathcal{L}_c = \frac{1}{16\pi} \left[ v_c^{-1} (\partial_x \Phi_c)^2 + v_c (\partial_x \Phi_c)^2 \right] + \lambda \cos(\beta \Phi_c). \tag{1}
\]

Here \( \beta = \sqrt{4K_c} \) and the spin sector is gapless \[1\] and spin rotationally invariant, which fixes the corresponding Luttinger liquid parameter. The cosine term in the charge sector is generated by double Umklapp processes and we assume it to be relevant, which implies \( \beta < 1 \). We will take \( \beta^2 \approx 0.9 \) which best fits the estimates given in \[2\] for the case of Bechgaard salts. The spin and charge velocities \( v_s \) and \( v_c \) are parameters of the theory.

The electronic creation and annihilation operators \( c_{j,\sigma}^\dagger \), \( c_{j,\sigma} \) \( (\sigma = \uparrow, \downarrow) \) of the underlying lattice model are related to the fields appearing in the Lagrangian and their corresponding dual fields

\[
\Theta_{c,\sigma}(x) = \frac{-i}{v_{c,\sigma}} \int_{-\infty}^{x} dy \partial_x \Phi_{c,\sigma}(\tau, y) \tag{2}
\]

by

\[
c_{j,\sigma} \propto \left[ \exp(iK_F x) R_{\sigma}(x) + \exp(-iK_F x) L_{\sigma}(x) \right], \tag{3}
\]
where \( x = j a_0 \) (\( a_0 \) is the lattice spacing) and
\[
L_\sigma = \eta_\sigma e^{\pm \frac{i}{2}(\Phi_\sigma - \frac{\theta}{2} \Theta_\sigma)} e^{\mp \frac{\theta}{2}(\Phi_\sigma - \Theta_\sigma)},
\]
\[
R_\sigma = \eta_\sigma e^{-\frac{i}{2}(\Phi_\sigma + \frac{\theta}{2} \Theta_\sigma)} e^{-\frac{\theta}{2}(\Phi_\sigma + \Theta_\sigma)}.
\]
Here \( \delta = \pm \) for up/down spins and \( \eta_\sigma = \eta_\uparrow \) are Klein factors that fulfill \( \{\eta_\sigma, \eta_\tau\} = 2\delta_{\sigma,\tau} \). In order to determine the spectral function at zero temperature we evaluate the single-electron Green’s function
\[
G^{(\sigma)}_{RR}(\tau, x) = \langle 0| R_\sigma(\tau, x) R^\dagger_\sigma(0)|0\rangle_c.
\]
We do this by exploiting the factorization (4)
\[
G^{(\sigma)}_{RR}(\tau, x) = \epsilon(0) \mathcal{O}_c(\tau, x) \mathcal{O}_c(0) \epsilon(\nu_\tau - i\epsilon) \frac{1}{2}.
\]
The large-distance asymptotics of the correlation function in the charge sector can be analyzed by going to the spectral representation of the corresponding sine-Gordon model (SGM). In the repulsive regime \( \beta > 1/\sqrt{2} \), the only single-particle excitations of the SGM (6) are soliton and antisoliton. A rough physical picture of what solitonic excitations look like may be obtained by considering a quarter filled extended Hubbard model with sufficiently large nearest-neighbour repulsion \( V \), so that we are in the 4kF CDW insulating phase. One naively obtains two ground states as shown in Fig. 1 and the antisoliton is then the kink connecting these two ground states.

![FIG. 1. Ground states and antisolitonic kink in a U-V extended Hubbard model.](image)

It is also clear from Fig. 1, the soliton carries an effective charge of \(-e/2\). Soliton and antisoliton have massive relativistic dispersions, which we as usual parametrize in terms of a rapidity variable \( \theta \)
\[
E(\theta) = M \cosh \theta, \quad P(\theta) = \frac{M}{v_c} \sinh \theta = m \sinh \theta.
\]

A basis of scattering states of solitons and antisolitons can be constructed by means of the Zamolodchikov-Faddeev (ZF) algebra. The ZF algebra is essentially the extension of the algebra of creation and annihilation operators for free fermions or bosons to the case or interacting particles with factorizable scattering. The ZF algebra is based on the knowledge of the exact spectrum and scattering matrix (15). For the SGM the ZF operators (and their hermitian conjugates) satisfy the following algebra
\[
Z^{\uparrow}(\theta_1)Z^{\downarrow}(\theta_2) = Z^{\downarrow}(\theta_1 - \theta_2)Z^{\uparrow}(\theta_2)(\theta_1) , \quad (9a)
\]
\[
Z^{\uparrow}_1(\theta_1)Z^{\downarrow}_2(\theta_2) = Z^{\downarrow}_2(\theta_2)Z^{\uparrow}_1(\theta_1)S^{\uparrow}_1(\theta_1 - \theta_2) , \quad (9b)
\]
\[
Z^{\downarrow}_1(\theta_1)Z^{\uparrow}_2(\theta_2) = Z^{\uparrow}_2(\theta_2)S^{\downarrow}_2(\theta_2 - \theta_1)Z^{\uparrow}_1(\theta_1) + (2\pi)\delta(\theta_1 - \theta_2), \quad (9c)
\]
where \( S^{\uparrow}_{1,2}(\theta) \) are the known factorizable two-particle scattering matrices (13) and \( \varepsilon_j = -, + \) for solitons and antisolitons respectively. Using the ZF operators a Fock space of states can be constructed as follows. The vacuum is defined by
\[
Z_{\uparrow}(\theta)|\Omega\rangle = 0 .
\]

Multiparticle states are then obtained by acting with strings of creation operators \( Z^\dagger_\sigma(\theta) \) on the vacuum
\[
|\theta_n \ldots \theta_1\rangle_{\epsilon_1 \ldots \epsilon_n} = Z_{\epsilon_n}(\theta_n) \ldots Z_{\epsilon_1}(\theta_1)|\Omega\rangle . \quad (11)
\]

In term of this basis the resolution of the identity is given by
\[
\mathbb{1} = |\Omega\rangle\langle\Omega| + \sum_{n=1}^{\infty} \sum_{\epsilon_1 \ldots \epsilon_n} \int_{-\infty}^{\infty} d\theta_1 \ldots d\theta_n |\theta_n \ldots \theta_1\rangle_{\epsilon_1 \ldots \epsilon_n} \langle\theta_1 \ldots \theta_n| .
\]

Using this basis of states in the charge sector SGM we obtain the following spectral representation
\[
c(0)\mathcal{O}_c(\tau, x) \mathcal{O}_c(0) \epsilon_c = \sum_{n=1}^{\infty} \sum_{\epsilon_1 \ldots \epsilon_n} \int \frac{d\theta_1 \ldots d\theta_n}{(2\pi)^n n!} \\
\times e^{-\frac{1}{2} \sum_{\epsilon_1 \ldots \epsilon_n} i[P(\theta_{\epsilon_1} + E(\theta_{\epsilon_1})\tau)]} |\Omega\rangle\langle\Omega|_c(0, 0)|\theta_n \ldots \theta_1\rangle_{\epsilon_1 \ldots \epsilon_n} \langle\theta_1 \ldots \theta_n| . \quad (12)
\]

The first non-vanishing matrix element (formfactor) corresponds to the emission of two solitons. According to Eq.(2.12) of (16) we have
\[
|\Omega\rangle\langle\Omega|_c(\theta_1, \theta_2) = Z e^{-\frac{\theta_1 + \theta_2}{2} |G(\theta_1 - \theta_2)|^2} \times \exp \left( \frac{\int_{0}^{\infty} dt \sinh^2(t) [t(1 - \theta_2/\pi)] \sinh(t[\xi - 1])}{\sinh 2\pi \sinh \xi \cosh t} \right), \quad C_1 = \exp \left( -\int_{0}^{\infty} \frac{dt \sinh^2(t/2) \sinh(t[\xi - 1])}{\sinh 2\pi \sinh \xi \cosh t} \right), \quad (13)
\]
The overall constant \( Z \) is calculated in (16) and is fixed by the following short-distance normalization for \( r = \sqrt{v_c^2 t^2 + x^2} \rightarrow 0 \).
where \( b \) is the single-electron Green's function is therefore given by
\[
G_{RR}(\tau, x) \approx \frac{Z}{(v_s \tau - ix)} \int_{-\infty}^{\infty} \frac{d\theta}{2\pi} \frac{G(2\theta_+)}{2m \cosh \theta (v_s \tau - ix)} + \frac{G(2\theta_-)}{2m \cosh \theta (v_s \tau - ix)}.
\]

In order to restore proper units one needs to multiply the correlation function by an appropriate power of the lattice spacing. The first subleading contribution to the correlation function involves three solitons and one antisoliton and is expected to be small at low energies, so we do not consider it here. The contribution of intermediate states involving two solitons to the single-electron Green's function is therefore given by
\[
G_{RR}(\tau, x) \approx \frac{Z}{(v_s \tau - ix)} \int_{-\infty}^{\infty} \frac{d\theta}{2\pi} \frac{G(2\theta_+)}{2m \cosh \theta (v_s \tau - ix)} \times \frac{(\omega + v_s q \sqrt{\cosh \theta - s^2})^2}{\sqrt{\cosh \theta}}.
\]

Fourier transforming and analytically continuing we find
\[
G_{RR}(\omega, k_F + q) = -Z \sqrt{\frac{2v_F^2}{v_c + v_s}} \int_{-\infty}^{\infty} \frac{d\theta}{2\pi} \frac{|G(2\theta_+)|}{\sqrt{\cosh \theta}} \times \frac{\omega + v_s q \sqrt{\cosh \theta - s^2}}{\sqrt{\cosh \theta}}.
\]

Thus the spectral weight increases linearly with \( s - 2M \) above the threshold. There is a remarkable relation between the spectral function and the real part of the optical conductivity \( \sigma(\omega) \), which for \( \beta \to 1 \) and \( v_c = v_s \) takes the simple form
\[
A_{RR}(\omega, q) \approx \frac{\text{const}}{\omega - q} \int_{-\infty}^{\infty} \frac{dx}{\omega^2 - v_c^2 q^2 - x^2}.
\]

The tunneling density of states for \( v_c = v_s \) is
\[
\rho(\omega) = 2 \int_{0}^{\text{arccosh}(\omega/2M)} \frac{d\theta}{2\pi} \frac{|G(2\theta_+)|}{\sqrt{\cosh \theta}}.
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\]
negative, the dimerization leads to the confinement of the kinks and we expect the physics to be more similar to the case without double Umklapp described below. At present no definite estimates for $U$ and $V$ for the Bechgaard salts are available. Having said this, let us try to compare our results to the experimental findings on (TMTTF)$_2$PF$_6$. The experimental estimates for the gaps are $\Delta_{\text{PF}} = 100 \pm 20$ meV, $\Delta_{\text{opt}} \approx 100$ meV and $\Delta_{\text{T}} \approx 43$ meV [15,16]. This fits well into the small-$K_c$ scenario discussed above: $\Delta_{\text{opt}}$ is twice the single-particle gap because the lowest-lying intermediate states that couple to the current operator are soliton-antisoliton states [2]. In the quarter-filled case, the gap observed in photoemission corresponds to the emission of two solitons and therefore also occurs at twice the single-particle gap. This is a distinct feature of the quarter-filled 4$k_F$-CDW insulator and it looks like there is qualitative agreement with the experiments here.

Let us now turn to the case where we have dimerization, but the interactions are weak so that the double Umklapp is irrelevant. The relevant Lagrangian is again of the form (1), but now $\beta^2 = K_c$. The main difference to the double-Umklapp case is the expression for the electron creation operator, which is obtained by replacing $\beta/2$ by $\beta$ in (1). The spectral function can then be calculated by the same methods as before [17], but now solitons carry charge $e$ and only a single soliton couples to the electron creation operator at low energies. This means that now the gap $M$ seen in ARPES is the same as the thermal activation gap and half the optical gap. In Fig. 3 we show the spectral function for $v_s = 0.8v_c$ and 10 different values of $q$.

![Figure 3](https://example.com/figure3.png)

**FIG. 3.** $A_{RR}(\omega, k_F + q)$ as a function of $\omega/M$ with dimerization but no double Umklapp and $v_s = 0.8v_c$. The curves for different $q$ are offset and have been smoothed to remove the square root threshold singularities.

For $v_s q \leq Q = Mv_s/\sqrt{v_s^2 - v_c^2}$ the spectral function exhibits square root singularities above the threshold at $\omega = \sqrt{M^2 + v_s^2 q^2}$. For $v_s q > Q$ there are two square-root singularities: one above the threshold at $\omega = v_s q + M\sqrt{1 - (v_s/v_c)^2}$ and a second one when $\omega$ approaches $\sqrt{M^2 + v_s^2 q^2}$ from below. Their interpretation is the same as for the half-filled Mott insulator [9]: the first singularity corresponds to states where the holon does not carry momentum, and the second one to states where the spinon does not carry momentum.

This scenario appears to be much less compatible with the available ARPES data than the double Umklapp scenario in the sense that it predicts a rather sharp, dispersing feature with significant spectral weight around the threshold. In the data essentially no dispersing feature has been observed, which would appear to be more compatible with the very broad signal shown in Fig. 2 than with the one shown in Fig. 3.

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