Optical Conductivity of One-Dimensional Mott Insulators

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We calculate the optical conductivity of one-dimensional Mott insulators at low energies using a field theory description. The square root singularity at the optical gap, characteristic of band insulators, is generally absent and appears only at the Luther-Emery point. We also show that only few particle processes contribute significantly to the optical conductivity over a wide range of frequencies and that the bare perturbative regime is recovered only at very large energies. We discuss possible applications of our results to quasi-one-dimensional organic conductors.

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Measurements of dynamical properties and, in particular, the optical conductivity $\sigma(\omega)$ are supposed to provide a stringent test of the existing theories of quasi-one-dimensional (1D) systems. The behavior of $\sigma(\omega)$ in the metallic regime is easily understood in terms of the Tomonaga-Luttinger theory [1]. The situation in the Mott insulating phase [2] is much more complicated as a spectral gap in dynamically generated by interactions. Here $\sigma(\omega)$ has until now only been studied by perturbative methods [3,4], which are expected to work well at high and intermediate frequencies but are not applicable to the most interesting regime of frequencies close to the optical gap. The purpose of the present work is to determine $\sigma(\omega)$ in 1D Mott insulators for all frequencies much smaller than the bandwidth, which is the large scale in the field theory approach to the problem. In particular, we obtain for the first time the true behavior of $\sigma(\omega)$ just above the optical gap.

An important property of one-dimensional systems that significantly simplifies our analysis is spin-charge separation, which occurs at energies much smaller than the bandwidth. In this regime $\sigma(\omega)$ is determined solely by the charge degrees of freedom. The standard description of the charge sector of the 1D Mott insulator is given by the sine-Gordon model (SGM) [4,5]

$$H_{\rm sG} = \int dx \left[4\pi(\Pi)^2 + \frac{1}{16\pi}(\partial_x\phi)^2 + 2\mu\cos(\beta\phi)\right]. \quad (1)$$

Here the momentum and coordinate densities obey the standard commutation relation $[\Pi(x), \phi(y)] = -i\delta(x - y)$. Throughout this Letter we set the charge velocity and $\hbar$ equal to 1.

The cosine term in the Hamiltonian is related to umklapp processes and the value of the sine-Gordon coupling constant β is determined by the interactions. The umklapp processes are relevant for $\beta^2 < 1$ and dynamically generate a spectral gap M, which is related to μ by (12). For $1/2 < \beta^2 < 1$ the spectral gap is related to the optical gap Δ (i.e., the gap seen in the optical absorption) by $\Delta = 2M$, whereas for $\beta^2 < 1/2$ solitonic bound states are formed below $2M$.

Our calculations of $\sigma(\omega)$ are based on the exact solution of the SGM and, in particular, on the work of Smirnov [6]. We confine our analysis to the repulsive regime $1/2 < \beta^2 < 1$, where the excitation spectrum consists of charged particles and holes (solitons and antisolitons), which do not form bound states. At the "Luther-Emery" point $\beta^2 = 1/2$ the SGM is equivalent to the theory of free spinless massive Dirac fermions. In this limit the solitons become noninteracting particles and the Mott insulator turns into a conventional band insulator. In the limit $\beta^2 \to 1$ the SGM acquires an SU(2) symmetry and describes the Hubbard model at half filling in the regime of weak interactions [7,8] and $\sigma(\omega)$ was recently determined in [9].

The optical conductivity is related to the imaginary part of the current-current correlation function, $\chi(\omega, q) = \langle j_{-q} j_q\rangle$, by

$$\sigma(\omega > 0) = \mathrm{Im}\{\chi(\omega, q = 0)\}/\omega\,. \quad (2)$$

The current density operator is proportional to the momentum density

$$j_q = A^{1/2}\Pi_q, \qquad \Pi_q = \int dx\, \Pi(t, x)e^{iqx}. \quad (3)$$

The nonuniversal coefficient $A^{1/2}$ depends on the detailed structure of the underlying microscopic lattice model.

Using the spectral representation one can express the optical conductivity at $T = 0$ as a sum over matrix elements of the zero wave vector Fourier component of the momentum operator:

$$\sigma(\omega > 0) = \frac{A}{\omega}\sum_n |\langle 0|\Pi_0|n\rangle|^2 \delta[\omega - (E_n - E_0)]. \quad (4)$$

Here $|0\rangle$ and $|n\rangle$ represent the ground state and excited states with energies E_0 and E_n, respectively. The difficulties in computation of the optical response are related to the fact that one requires not only the knowledge of the spectrum E_n, but also of the matrix elements of the momentum

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operator. The exact expressions for the matrix elements $\langle n|\Pi_0|0\rangle$ are extracted from the exact solution by means of the so-called form factor bootstrap procedure [6]. This approach is particularly efficient for strongly interacting integrable models with spectral gaps, because for a given energy ω the spectral representation for the imaginary part contains only a *finite* number of terms (in the absence of bound states at most $[\omega/\Delta]$ terms). In practice the spectral sum is found to converge extremely rapidly, so that a very good approximate description can be obtained by taking into account intermediate states with at most four particles [10]. The multiparticle matrix elements become essential only at very high energies where the field theory can no longer be used to describe the underlying lattice model anyway.

In order to compute (4) we need to introduce a suitable spectral representation. In the parameter regime we study, the spectrum contains only solitons and antisolitons with relativistic dispersion $e(p) = \sqrt{p^2 + M^2}$. It is useful to parametrize the spectrum in terms of a rapidity variable θ such that $p = M\sinh\theta$, $e = M\cosh\theta$. Solitons and antisolitons are distinguished by the internal index $\varepsilon = \pm$. A state of n solitons/antisolitons with rapidities $\{\theta_k\}$ and internal indices $\{\varepsilon_k\}$ is denoted by $|\theta_n \cdots \theta_1\rangle_{\varepsilon_n\cdots\varepsilon_1}$. Its total energy E, momentum P, and electric charge Q are

$$P = M\sum_{k=1}^{n}\sinh\theta_k, \qquad E = M\sum_{k=1}^{n}\cosh\theta_k, \qquad Q \propto \sum_{k=1}^{n}\varepsilon_k \,. \tag{5}$$

In terms of this basis $\sigma(\omega)$ is expressed as

$$\begin{aligned}\sigma(\omega) = {}& \frac{2\pi^2 A}{\omega}\sum_{n=0}^{\infty}\sum_{\varepsilon_i} \\ &\times \int \frac{d\theta_1\cdots d\theta_n}{(2\pi)^n n!}\left| f^j(\theta_1\cdots\theta_n)_{\varepsilon_1\cdots\varepsilon_n}\right|^2 \\ &\times \delta\Big(M\sum_k \sinh\theta_k\Big)\delta\Big(\omega - M\sum_k\cosh\theta_k\Big) \\ = {}& \sigma_2(\omega) + \sigma_4(\omega) + \cdots . \end{aligned} \tag{6}$$

Here

$$f^j(\theta_1\cdots\theta_n)_{\varepsilon_1\cdots\varepsilon_n} \equiv \langle 0|\, j(0,0)\,|\theta_n\cdots\theta_1\rangle_{\varepsilon_n\cdots\varepsilon_1} \tag{7}$$

are the form factors of the current operator, $\sigma_2(\omega)$ and $\sigma_4(\omega)$ represent the contributions from two- and four-particle processes, and the dots indicate processes involving a higher number of (anti)solitions. We note that as a consequence of symmetry properties only intermediate states with an even number of particles contribute to this correlation function. From (6) it is easy to see that only two-particle processes contribute up to energies $\omega = 4M$, only two- and four-particle processes up to $\omega = 6M$, and so on.

The form factors (7) have been determined in [6] and can be used to calculate the first few terms in the expansion (6). Here we give explicitly only the two-particle contribution and refer to [11] for details on the much more complicated four-particle contribution. We find

$$\sigma_2(\omega) = \frac{2A\Theta(\omega - 2M)}{\omega^2\sqrt{\omega^2 - 4M^2}}\,|f(\theta)|^2, \tag{8}$$

where $\Theta(x)$ is the Heaviside function,

$$\begin{aligned} f(\theta) = f^j(\theta)_{+-} = f^j(\theta)_{-+} = {}& \frac{2\pi M}{i\beta}\,\frac{\sinh\theta/2}{\cosh(\frac{\theta+i\pi}{2\xi})} \\ & \times \exp\left\{\int_0^\infty dt\,\frac{\sinh^2 t(1 - i\theta/\pi)\sinh t(\xi - 1)}{t\sinh 2t\cosh t\sinh t\xi}\right\}, \end{aligned} \tag{9}$$

and

$$\theta = 2\mathrm{arccosh}(\tilde{\omega}), \qquad \xi = \beta^2/(1 - \beta^2), \qquad \tilde{\omega} = \omega/2M \,. \tag{10}$$

The two- and (100 times the) four-particle contributions to $\sigma(\omega)$ for $\beta^2 = 0.9$ are presented in Fig. 1. Most importantly, the square root singularity, being a characteristic feature of band insulators, is suppressed by the momentum dependence of the soliton-antisoliton form factor and reappears only for the Luther-Emery point $\beta^2 = 1/2$. This effect was noted previously for the Hubbard model at half filling [9] which corresponds to the special SU(2) symmetric point $\beta^2 = 1$. We find that for any $\beta^2 \neq 1/2$ there is a square root "shoulder" $\sigma(\omega) \propto \sqrt{\omega - \Delta}$ for $\omega/\Delta - 1 \ll 1$ as shown in the inset of Fig. 1. In the vicinity of the Luther-Emery point $\beta^2 = 1/2$ we obtain the following analytical expression valid for $\tilde{\omega} - 1 \ll 1$:

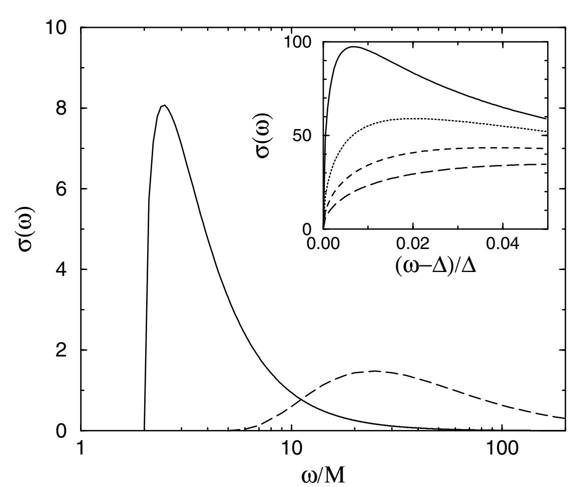

FIG. 1. Two-particle (solid line) and 100 times the four-particle (dashed line) contributions to the optical conductivity as a function of (ω/M) for $\beta^2 = 0.9$. Inset: threshold behavior of $\sigma(\omega)$ close to the Luther-Emery point for four different values of β; $\beta = 0.72$ (solid line), $\beta = 0.73$ (dotted line), $\beta = 0.74$ (dashed line), and $\beta = 0.75$ (long dashed line).

$$\sigma(\omega) \propto \frac{\sqrt{\tilde{\omega}^2 - 1}}{[\tilde{\omega}^2 - 1] + \xi^2 \sin^2 \gamma}, \quad \gamma = \pi\left(\frac{1}{2\beta^2} - 1\right). \tag{11}$$

The square root singularity above $\omega = \Delta$ for $\beta^2 = 1/2$ is replaced by a maximum occurring at $\omega/\Delta - 1 \propto \gamma^2$.

The four-particle contribution to σ is seen to be insignificant at low energies and becomes larger than the two-particle contribution only at $\omega \approx 180M$ for $\beta^2 = 0.9$. This suggests that the optical conductivity is well described by the combination of two- and four-particle contributions up to several hundred times the mass gap. Computation of higher order terms in Eq. (6) becomes cumbersome and probably of no physical interest, since the previous analysis suggests that they become important outside the region of applicability of the field theory approach to physical systems.

At frequencies much larger than the gap it is possible to determine $\sigma(\omega)$ by perturbative methods. The leading asymptotics can be calculated by "conformal perturbation theory" [12]. Here the cosine interaction in (1) is considered as a (relevant) perturbation of the Gaussian model and correlation functions are calculated in an expansion in powers of the scale μ, which can then be expressed in terms of the physical gap M as [13]

$$\mu = \frac{\Gamma(\beta^2)}{\pi\Gamma(1-\beta^2)}\left[M\frac{\sqrt{\pi}\,\Gamma(1/2 + \xi/2)}{2\Gamma(\xi/2)}\right]^{2-2\beta^2}. \tag{12}$$

We find to leading order

$$\sigma(\omega) = 2^{9-4\beta^2}\left(\frac{\pi^2\beta}{\Gamma(2\beta^2)}\right)^2 \mu^2 \omega^{(4\beta^2-5)} = \frac{8\pi^3\beta^2}{\omega\Gamma^2(1-\beta^2)\Gamma^2(\frac{1}{2}+\beta^2)} \times \left[\frac{\Gamma(\frac{\xi}{2})}{2\sqrt{\pi}\,\Gamma(\frac{1+\xi}{2})}\,\frac{\omega}{M}\right]^{4\beta^2-4}. \tag{13}$$

We emphasize that the ratio of the coefficients of the high- and low-energy asymptotics (13), (8) is *fixed* [6,14], In other words, the amplitude of the power law in (13) is tied to the overall factor in (8) and the form factor expansion must approach the perturbative result in the large-ω limit. A comparison between the form factor results and (13) is shown in Fig. 2. We see that the asymptotic regime is not yet reached at energies as high as $\omega \sim 1000M$ [in practical terms this implies that perturbation theory (PT) cannot be used to make contact with experiment]. We note that the contributions due to intermediate states with $6, 8, \ldots$ particles are all positive and will make the agreement of the form factor sum with PT in the region $\omega \approx 1000M$ only worse. A good way to overcome these deficiencies of bare PT is to carry out a renormalization-group (RG) improvement as performed in [4]. In Zamolodchikov's scheme [13] the RG equations for the sine-Gordon model are given by

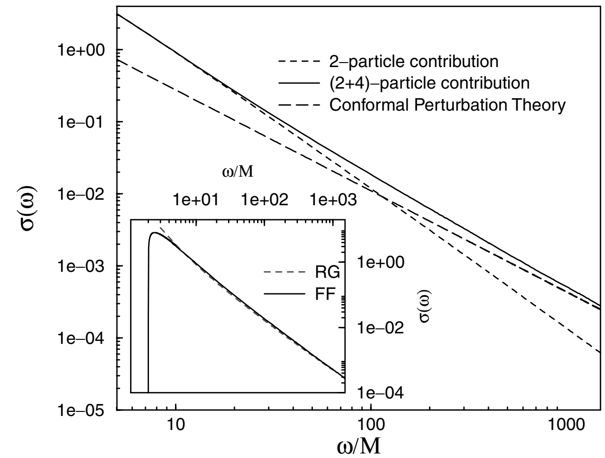

FIG. 2. Comparison between the 2- and 2 + 4-particle contributions to the optical conductivity and the perturbative result, for $\beta^2 = 0.9$. Inset: comparison between the form factor result and RG improved PT.

$$\frac{dg_\perp}{dt} = \frac{g_\parallel g_\perp}{1 + \frac{g_\parallel}{2}}, \qquad \frac{dg_\parallel}{dt} = \frac{g_\perp^2}{1 + \frac{g_\parallel}{2}}. \tag{14}$$

The solution of (14) is

$$g_\perp = 4\,\frac{1-\beta^2}{\beta^2}\,\frac{\sqrt{q}}{1-q}, \qquad g_\parallel = 2\,\frac{1-\beta^2}{\beta^2}\,\frac{1+q}{1-q}, \tag{15}$$

where

$$q\left(\frac{(1-q)\beta^2}{4(1-\beta^2)}\right)^{2\beta^2-2} = e^{(4-4\beta^2)(t-t_0)}. \tag{16}$$

Using $t - t_0 = \ln[(\sqrt{\pi}\,e^{3/4}M/2^{3/2}\omega)]$ we can reexpress (13) up to higher order terms as

$$\sigma(\omega) = \frac{\pi^3\beta^6 g_\perp^2}{2\omega\Gamma^2(2-\beta^2)\Gamma^2(\frac{1}{2}+\beta^2)} \times \left[\frac{\Gamma(\frac{\xi}{2})e^{3/4}\sqrt{\xi}}{2^{7/2}\Gamma(\frac{1+\xi}{2})}\right]^{4\beta^2-4}. \tag{17}$$

The RG improved result (17) for $\sigma(\omega)$ is compared to the form factor result (sum of the two- and four-particle contributions) in the inset of Fig. 2. The agreement is rather good down to energies of the order of $5M$.

One possible realization of a 1D Mott insulator is the $(TMTSF)_2X$ Bechgaard salts [15]. These materials are highly anisotropic and can be modeled as weakly coupled, quarter-filled chains. At energies or temperatures above the 1D–3D crossover scale E_{cr} the interchain coupling becomes ineffective and a description in terms of a purely 1D model with charge sector (1) should be possible [5]. At present there is some uncertainty regarding the value of E_{cr} because interactions can renormalize its bare value, set by the interchain coupling, downwards [16]. There is a lot of ambiguity in fitting our results to the data. The value of the optical gap $2M$ is not known and, as discussed above,

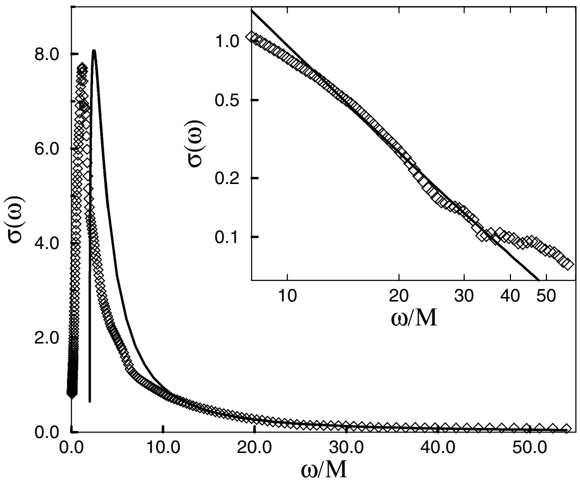

FIG. 3. Comparison between the optical conductivity calculated in the SGM for $\beta^2 = 0.9$ (solid lines) and measured optical conductivity for $(TMTSF)_2PF_6$ from Ref. [15] (diamonds). The inset shows the same comparison on a logarithmic scale.

we cannot calculate the overall normalization of $\sigma(\omega)$. We therefore use these as parameters in order to obtain a good fit at large ω (where the theory is expected to work best as 3D effects are unimportant) to the data [15] for any given value of β. We obtain reasonable agreement between the form factor result $\sigma_2(\omega) + \sigma_4(\omega)$ and the data for $\beta^2 \approx 0.9$, which corresponds to a Luttinger liquid parameter of $K_\rho = \beta^2/4 \approx 0.225$. This value is consistent with previous estimates (see the discussion in [15]) [17].

As is clear from Fig. 3, the model (1) seems to apply well at high energies, but becomes inadequate at energies of the order of about 10 times the Mott gap [≈ 1600/cm in $(TMTSF)_2PF_6$]. Spectral weight is transferred to lower energies beyond that of a pure 1D Mott insulator emerges. There are at least two mechanisms that should be taken into account in this range of energies. First, a small dimerization occurs in the 1D chains and will almost certainly affect the structure of $\sigma(\omega)$ around its maximum. Second, the interchain hopping is no longer negligible [18] and ought to be taken into account.

In summary, we have exactly calculated $\sigma(\omega)$ for a pure 1D Mott insulator in a field theory approach. We have determined the threshold behavior for the first time and found it to exhibit a universal square root increase for any $\beta^2 > 1/2$. This is in contrast to the well-known square root singularity that appears at the Luther-Emery point $\beta^2 = 1/2$. In the "low"-energy region ($\omega/\Delta < 50$) the optical conductivity is dominated by the two-particle contribution with a small correction from four-particle processes. We also have shown that the leading asymptotic behavior obtained in PT is a good approximation only at extremely large frequencies, whereas RG-improved PT works well over a large region of energies.

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