Optical absorption edge in one-dimensional conductors

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The frequency-dependent conductivity is studied for both the one-dimensional Hubbard model and a model of spinless fermions, using a selection rule, the Bethe ansatz energy eigenstates, and conformal invariance. For densities where the system is metallic the absorption spectrum has two contributions, a Drude peak at \( \omega = 0 \) separated by a pseudo-gap from a broad absorption band whose lower edge is characterized by a non-classical critical exponent. Our findings are expected to shed new light on the “far infrared puzzle” of metallic organic chain compounds.

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Quasi-one-dimensional materials with partially filled electronic bands, i.e. “metals” from a naive point of view, usually undergo a transition to a low-temperature phase with a gap in the charge excitation spectrum. These transitions are driven by electron-electron and/or electron-phonon interactions. Depending on the coupling constants and the band fillings the broken symmetry ground states are of different kinds: spin- or charge-density waves, bond alternation, superconductivity. Simple mean-field theory is able to describe these transitions, at least qualitatively. The broken symmetry induces a gap at the Fermi energy which may be displayed in the optical absorption spectrum. In the case of charge-density waves accompanied by a lattice distortion, optical gaps have indeed been clearly observed, and beautiful examples have been discovered among organic molecular chain compounds [1], conjugated polymers [2], and inorganic chain-like materials [3]. In the case of a moving spin-density wave, the sliding collective mode removes all the finite-frequency transitions and produces a single peak at \( \omega = 0 \) [4]. Pinning by impurities not only shifts this peak to a finite frequency but also restores optical transitions above the gap [5].

The mean-field scenario makes sense at very low temperatures, where interchain coupling stabilizes three-dimensional long-range order. In contrast, above the transition temperature the system may enter an essentially one-dimensional regime, which requires a more refined treatment of interactions. The real part of the optical conductivity can be written as \( \sigma_1(\omega) = 2\pi D\delta(\omega) + \sigma_{1}^{\text{reg}}(\omega) \), where the frequency-dependent conductivity \( \sigma_1^{\text{reg}}(\omega) \) is defined in Eq. [6] below. Giamarchi and Millis have used the bosonization technique and found \( \sigma_1^{\text{reg}}(\omega) \sim \omega^3 \) in the limit \( \omega \to 0 \) for incommensurate band fillings [8]. Unfortunately, it is at present not clear to what extent their treatment is a conserving approximation and, therefore, it is important to compare their results with those obtained using other techniques. Several authors have used exact diagonalization for the one-dimensional Hubbard model. In addition to finding no low-frequency absorption at half-filling, where the system is an insulator, these authors have found practically no low-frequency (\( \omega \neq 0 \)) absorption away from half-filling, where a Drude peak at \( \omega = 0 \) confirms that the system is metallic [7]. This is not only in contrast to the bosonization results, but also to the two-dimensional case where doping away from half filling induces mid-infrared absorption [15]. Analytical calculations in the large \( U \) limit of the one-dimensional Hubbard model have so far been restricted to the electronic density \( n = 1/2 \) [9]. In this limit the optical gap occurs at \( E_{\text{opt}} = U - 4t \), followed by an absorption band extending up to \( U + 4t \) [12].

In this paper the optical absorption of the one-dimensional Hubbard model is discussed in the framework of the Bethe ansatz, which has been used previously by Schulz [12] to calculate \( D \). Together with the optical sum rule, this allowed him to derive the total intensity of finite-frequency transitions. This intensity is generally small, except close to half filling for large enough (but not too large) values of \( U \). We have found that these finite-frequency transitions are likely to be limited to a well-defined band above an effective optical gap, which is actually smallest at half filling and have also been able to derive the exponent for the frequency-dependence above the absorption edge.

The 1D Hubbard model can be written as \( \hat{H} = \hat{T} + U \hat{D} \), where \( \hat{T} = -t \sum_{j,\sigma} [c_j^{\dagger \sigma} c_{j+1\sigma} + h.c.] \) is the “kinetic energy”, \( \hat{D} = \sum_j \hat{n}_{j\uparrow} \hat{n}_{j\downarrow} \) measures the number of doubly occupied sites, \( c_j^{\dagger \sigma} \) and \( c_{j\sigma} \) are electron operators of spin projection \( \sigma \) at site \( j = 1, \ldots, N_a \), \( \hat{n}_{j\sigma} = c_j^{\dagger \sigma} c_{j\sigma} \), \( t \) is the transfer integral, and \( U \) is the on-site Coulomb interaction. We choose a density \( n = N_a/N \) in the interval \( 0 \leq n \leq 1 \) with even \( N \) and zero magnetization. The Fermi momentum is given by \( k_F = \pi n/2 \). We use units such that \( -e = \hbar = 1 \), where \( -e \) is the electronic charge. The regular part of the optical conductivity can be written as
\[
\sigma_1^{reg}(\omega) = \frac{\pi}{N_o} \sum_{\nu \neq 0} \frac{|\langle \nu | \hat{J} | 0 \rangle|^2}{\omega_{\nu,0}} \delta(\omega - \omega_{\nu,0}).
\]

Here \( \hat{J} = -it \sum_{j,\sigma} \left[ c_{j+1,\sigma} c_{j,\sigma} - c_{j,\sigma} c_{j+1,\sigma} \right] \) is the current operator, the summation runs over energy eigenstates, and \( \omega_{\nu,0} = \varepsilon_\nu - \varepsilon_0 \) is the excitation energy above the ground state \( |0\rangle \). The formula \( \text{(1)} \) applies if the ground state is non-degenerate. This is true for an even number of sites \( N_o \) if periodic (anti-periodic) boundary conditions are used for odd (even) values of \( N_o \), respectively. The ground state is then necessarily an eigenstate of the parity operator, \( \hat{P}_\pi \), which moves electrons from sites \( j \) to \( N_o + 1 - j \), \( j = 1, \ldots, N_o \), and has eigenvalues \( \pm 1 \). Importantly, in the present model \( \hat{P}_\pi \) commutes with the Hamiltonian and anticommutes with the current operator. This implies immediately that the states \( |0\rangle \) and \( |\nu\rangle \) can have opposite parities, a key selection rule for optical transitions. The final states \( |\nu\rangle \) can be characterized in terms of holons, antiholons, spinons \( [4,15] \) and a charge-transfer band \( [16] \). The holon and spinon bands describe low-energy charge and spin excitations, whereas the charge transfer band is associated with the upper Hubbard band. We use the labels \( \alpha = c, s, t \) for the holon/antiholon, the spinon, and the charge-transfer bands, respectively \( [16] \), and the quantum number \( \beta \) for distinguishing between holons \( (\beta = -\frac{1}{2}) \) and antiholons \( (\beta = +\frac{1}{2}) \) \([15,16]\). In the present context, the \( s \) band is empty of spinons, the \( c \) band can be populated by holons and by zero or one antiholon, and the \( t \) band can have occupancy zero or one. The momentum variable of the different bands has the form \( q_j = \frac{2\pi}{N_o} I_j \), where \( I_j \) are successive integers or half-odd integers. In contrast to the case of electron bands in a periodic solid, the number of available momenta \( N^c \) can change with band occupancies. These numbers are \( N^c = N_o \), \( N^h = N/2 - N_t \), and \( N^\ell = N_o - N + N_t \), where \( N_t \) is the number of occupied momenta in the charge-transfer band. Therefore, the momentum band widths are \( \Delta q_c = 2\pi \), \( \Delta q_h = 2k_F - 2\pi N_t/N_o \), and \( \Delta q_\ell = 2\pi - 4k_F + 2\pi N_t/N_o \). The numbers \( N^c \) (holons), \( N^h \) (antiholons), and \( N^\ell \) are good quantum numbers which obey the sum rules \( [14] \). \( N_o - N = -2 \sum_{\beta = \pm \frac{1}{2}} \beta N^h_{c,\beta} = N^c - 2N_t \), where \( N^h_c = \sum_{\beta = \pm \frac{1}{2}} N^h_{c,\beta} \). The ground state is characterized by \( N^h_{c,-\frac{1}{2}} = N_t = 0 \) and \( N^h_{c,+\frac{1}{2}} = N_o - N_t \), with a symmetrical holon occupancy in the \( c \) band for momenta \( 2k_F < |q| < \pi \). The energy bands \( \epsilon_\nu(q) \) can be extracted from the Bethe-ansatz solution \( [13] \). Interestingly, we find that within the parameter region of appreciable oscillator strength for optical transitions \( [14] \), the charge transfer band width \( W_t = \epsilon_1(0) - \epsilon_1(\pi - 2k_F) = \epsilon_1(0) \) is very small.

The main transitions contributing to \( \sigma_1^{reg}(\omega) \) are of two types: (a) those leaving the band fillings \( N^h_{c,-\frac{1}{2}}, N^h_{c,+\frac{1}{2}} \), and \( N_t \) unchanged and (b) those changing these numbers by \( \Delta N^h_{c,\pm \frac{1}{2}} = \Delta N^h_{c,\pm \frac{1}{2}} = \Delta N_t = 1 \). The former start, in principle, at \( \omega = 0 \), while the latter have an onset at \( E_{opt} = W_t - 2\epsilon_0(2k_F) > 0 \). We show first that, as a consequence of the selection rule presented above, the transitions of type (a) are likely to have a very small weight. These are excitations within the holon band which can be characterized in terms of single and multiple electron-hole excitations \( [3] \). Single electron-hole excitations give no contributions to the finite-frequency absorption, while multiple excitations are expected to decrease very rapidly in intensity. Double excitations, which would give a contribution \( \sigma_1(\omega) \sim \omega^3 \) \([1] \), are forbidden by the parity selection rule, since these transitions require symmetrical changes of holon occupancies at \( q \) and \(-q \), leaving the parity unchanged. In addition, there may be higher order low-frequency Umklapp processes \( [8] \), but since these require multiple excitations in the holon band, they are expected to have extremely low spectral weight. These considerations explain why numerical studies yield so weak features at low frequencies \( [3,17] \).

The weakness of type (a) transitions implies that the onset of type (b) transitions at \( E_{opt} \) represents a pseudogap which may look like a true gap in an actual experiment. \( E_{opt} \), shown in Fig. 1, increases with increasing \( U \) and decreases with increasing density. For \( n = 1 \) it coincides with the Mott-Hubbard gap \( E_{MH} \) \([16] \), which can be expressed as \( E_{MH} = -2\epsilon_0^e(\pi) \) and is an increasing function of \( U \). In general, the type (b) transitions are also weak, except for \( n \) close to 1 and large \( U \) \([3] \), corresponding to a region of parameters where the bandwidth \( W_t \) is very small. Therefore, we have limited our studies to this parameter region. The important processes are those where in addition to a \( t \) particle a holon-antiholon pair is created in the \( c \) band. This generates an absorption band with a lower edge at \( \omega = E_{opt} \) and a width \( \Delta \omega_{opt} = -2[\epsilon_0^e(0) - \epsilon_0^e(2k_F)] = 8\pi - 2\epsilon_0^e \). [Here \( \epsilon_0^e = \epsilon_0(\pi) - \epsilon_0(2k_F) \), changes between \( \epsilon_0^e \) for \( n \to 0 \) and \( \epsilon_0^e \) for \( n \to 1 \).] The use of the quasi-particle/electron representation of Ref. \([15] \) reveals that these transitions are associated with one zero-momentum electron - hole process. Moreover, creating a \( t \) particle means enhancing double occupancy by 1, at least in the large \( U \) limit. Since the current operator cannot produce more than one doubly occupied site, there will be almost no spectral weight above the upper edge at \( \omega = E_{MH} + 8\pi + W_t \approx E_{MH} + 8\pi \), illustrated by the dashed line in Fig.1.

The spectral range of optical absorption deduced above on the basis of the holon and charge transfer excitation spectra fully agrees with large \( U \) expansions \([10] \) and numerical calculations \([7,11] \). A much more difficult problem is the spectral line shape. Here, we limit ourselves to the region immediately above threshold, using the hypothesis of conformal invariance. In this region the transition energies have a finite-size depen-
ence \( E_{\nu} - E_0 - E_{\text{opt}} = \frac{2\pi}{N_\alpha} \sum_{\alpha, \pm 1 \beta} v_{\alpha \beta} \Delta_{\alpha} + O(1/N_\alpha) \) where \( \Delta_{\alpha} \) are \( U \) and \( n \) dependent positive parameters. The momentum is the same as in the ground state, i.e., \( P_\nu = \frac{2\pi}{N_\alpha} \sum_{\alpha, \pm 1} t \Delta_{\alpha} = 0 \). By analogy to the case of the two-velocity spectrum of Ref. [13, we interpret this as a low-energy \((\omega - E_{\text{opt}})\) three-velocity conformal-field theory, whose vacuum is \( |0\rangle \) with energy measured from \( E_0 + E_{\text{opt}} \). We have verified that the exponents for the conductivity do not depend on whether they are calculated for \( v_1 \to 0 \) or for \( v_1 = 0 \). The physical fields are of the general form \( \phi(x, t) e^{i E_{\text{opt}} t} \). The anomalous dimensions of the field \( \phi(x, t) \) fields involve the parameters, \( \xi_{\alpha, \alpha'}^j = \delta_{\alpha, \alpha'} + \sum_{i = \pm 1} [ U \Phi_{\alpha, \alpha'}(q_{F \alpha}, l q_{F \alpha'}) \) (with \( q_{F \alpha} = k_F \) and \( q_{F \beta} = 0 \)). Here \( j = 0, 1 \) and the two-particle forward-scattering phase shifts, \( \Phi_{\alpha, \alpha'}(q, \hat{q}) \), can be extracted from the Bethe-ansatz solution [10]. The critical behavior of the conductivity [4] can then be calculated, with the result \( \sigma_1^{\text{reg}}(\omega) = C \left( \omega - E_{\text{opt}} \right)^{\zeta} \) at the onset and a critical exponent

\[
\zeta = -\frac{3}{2} + \frac{1}{2} \left( 2\xi_{00}^\nu - \xi_{00}^t \right)^2 + \frac{1}{2} \left( \xi_{00}^t \right)^2. \tag{2}
\]

The exponent \( \zeta \), shown in Fig. 2, approaches \( \frac{1}{2} \) as \( U \to \infty \), and for densities \( n < 1 \) it increases with decreasing values of \( U \). For \( n \to 1 \) we find \( \zeta = \frac{1}{2} \) and \( W_\ell = 0 \) for all finite values of \( U \). Putting \( v_1 = 0 \) and \( n = 1 \) in the \( x \) and \( t \) dependent current - current (or charge - charge) correlation function and taking the Fourier transform, we again obtain \( \zeta = \frac{1}{2} \) at the threshold of optical absorption. Therefore, we believe that at \( n = 1 \) the expression \( \sigma_1^{\text{reg}}(\omega) = C \left( \omega - E_{\text{MH}} \right)^{\frac{1}{2}} \) is valid for all finite values of \( U \). For the particular case of \( n = 1 \) the same method allows us to evaluate the leading frequency dependence close to the upper edge of the absorption band. We find

\[
\sigma_1^{\text{reg}}(\omega) = C \left( E_{\text{MH}} + 8t - \omega \right)^{\frac{1}{2}}. \tag{3}
\]

This symmetry between the lower and upper absorption bands for \( n = 1 \) was also found in Refs. [10,12]. Moreover, expanding the \( n = 1 \) expression (28) of Ref. [10] around the edge energies leads precisely to the same \( \omega \) power-law dependences with exponents \( \frac{1}{2} \) and \( \frac{1}{2} \). We notice that the constant \( C \) has to vanish for all densities as \( U \to 0 \) or \( U \to \infty \), in these limiting cases the Drude peak exhausts the conductivity sum rule [13].

It is worthwhile to compare our predictions with experiments on one-dimensional conductors. Unfortunately, as a general rule these materials cannot be represented in terms of the Hubbard model alone, but additional interactions have to be taken into account. An exception seems to be the organic chain compound \((TTM - TTP)I_5\), which displays the properties of the one-dimensional Hubbard model at half filling with a relatively small value of \( U \) \((U \approx 3.5t)\); an optical gap of the expected size \( 15 \) and a Pauli-like susceptibility [20]. The optical absorption is consistent with a smooth onset at threshold, but it seems difficult to extract a critical exponent from the data. In the past there has been considerable discussion concerning the phenomenon of suppressed far infrared conductivity observed in organic chain compounds, despite their large dc conductivity [21]. Very recently, the optical absorption spectrum of large single crystals of Bechgaard salts has been measured over a huge frequency range, confirming the far infrared problem [22]. A Drude peak corresponding to a very small carrier density is separated by a gap of about 0.01 eV from an absorption band of a width of about 0.6 eV. The onset of absorption above the gap is very sharp at low temperatures. In the framework of the one-dimensional Hubbard model these results can be understood if the system has a nearly half-filled band, in which case the optical threshold is essentially given by the Mott-Hubbard gap. The parameters \( t \approx 0.25eV \) and \( U/t \approx 1.5 \) allow to reproduce the gross features of the experimental spectra.

While the simple Hubbard model may be sufficient for reproducing the overall absorption spectrum of some of the one-dimensional conductors, additional terms such as the Coulomb interaction between nearest neighbor sites would have to be considered for a more detailed description of the spectrum, for instance for the excitonic enhancement at the absorption edge [4]. Since the extended Hubbard model is not solvable by the Bethe ansatz, we have considered the model Hamiltonian \( H = -t \sum_{j \neq k} \epsilon_j \hat{c}_j \hat{c}_k + V \sum_{j} \hat{c}_j \hat{n}_j \) where \( \epsilon_j \) creates a spinless fermion at site \( j \), \( \hat{c}_j \) and \( \hat{n}_j \) is the nearest-neighbor Coulomb interaction. For a density \( n = \frac{N_\alpha}{N} = \frac{1}{2} \) this model has a metal - insulator transition for \( V > 2t \). As for the Hubbard model, in the region \( V > 2t \) we find almost no spectral weight in \( \sigma_1(\omega) \) for \( \omega < E_{\text{opt}} \), where \( E_{\text{opt}} \) is a \( V \)-dependent optical gap. We again obtain a critical behavior \( \sigma_1^{\text{reg}}(\omega) = C \left( \omega - E_{\text{opt}} \right)^{\vartheta} \) for low values of \( (\omega - E_{\text{opt}}) \) with \( \vartheta \to \frac{1}{2} \) both as \( n \to \frac{1}{2} \) and \( V > 2t \) and for all densities as \( V \to \infty \). For \( n = \frac{1}{2} \) and large \( V \) the corresponding absorption extends from \( \omega = V - 4t \) to \( \omega = V + 4t \).

In summary, we have discussed the frequency-dependent conductivity of the one-dimensional Hubbard model (and of a model of spinless fermions with nearest neighbor interaction), using a parity selection rule and known properties of the exact eigenstates. In the insulator phases the optical absorption has a square-root behavior at the onset. For \( n \neq 1 \) \((n \neq 1/2) \) and \( V > 2t \) the spectrum consists of a Drude peak at \( \omega = 0 \) and an absorption band above a pseudo-gap, which increases as the system is doped away from the insulating state. The behavior at the onset of the finite-frequency transitions is described by a critical exponent that depends both on the interaction strength and the density. The intensity of the transition across the gap is appreciable only when both the charge-transfer bandwidth is very narrow. This is reminiscent of the X-ray edge singularity which depends very
sensitively on the recoil of the hole \[23\]. Our results are expected to shed new light on the old problem of optical absorption in one-dimensional conductors.

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FIG. 1. The gap \( E_{opt} \), (a) as function of \( U \) for different values of \( n \) and (b) as function of \( n \) for different values of \( U \).

FIG. 2. The exponent \( \zeta \), (a) as function of \( U \) for different values of \( n \) and (b) as function of \( n \) for different values of \( U \). The straight lines at \( \zeta = 1/2 \) refer to the (a) \( n \to 1 \) and (b) \( U \to \infty \) limits.
