Optical conductivity of the quasi-one-dimensional organic conductors: The role of forward scattering by impurities

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We calculate the average conductivity $\sigma(\omega)$ of interacting electrons in one dimension in the presence of a long-range random potential (forward scattering disorder). Taking the curvature of the energy dispersion into account, we show that weak disorder leads to a transport scattering rate that vanishes as ω^2 for small frequency ω. This implies Im $\sigma(\omega) \sim \tilde{D}_c / \omega$ and Re $\sigma(\omega) \sim \tilde{D}_c \tau$ for $\omega \to 0$, where $\tilde{D}_c$ is the renormalized charge stiffness and the time τ is proportional to the strength of the impurity potential. These nontrivial effects due to forward scattering disorder are lost within the usual bosonization approach, which relies on the linearization of the energy dispersion. We discuss our result in the light of a recent experiment. [S0163-1829(99)02315-2]

In a recent measurement of the frequency-dependent conductivity $\sigma(\omega)$ of quasi-one-dimensional organic conductors of the $(TMTSF)_2X$-series, Schwartz *et al.*[1] found that for small frequencies ω the data could be fitted by

$$\sigma(\omega) = \frac{D_c}{\Gamma(\omega) - i\omega m^*(\omega)/m_b}, \quad (1)$$

where D_c is the bare charge stiffness, and where the transport scattering rate $\Gamma(\omega)$ and the effective mass enhancement $m^*(\omega)/m_b$ are given by

$$\Gamma(\omega) = \Gamma_0 + \frac{\lambda_0 \alpha \omega^2}{1 + \alpha^2 \omega^2}, \quad (2)$$

$$\frac{m^*(\omega)}{m_b} = 1 + \frac{\lambda_0}{1 + \alpha^2 \omega^2}. \quad (3)$$

The quadratic frequency dependence in Eqs.(2) and (3) is characteristic of Fermi liquids in three dimensions. Expressions of this type were used by Sulewski *et al.*[2] in their study of the compound UPt_3, which is a three-dimensional Fermi liquid with heavy mass. The experimentally obtained values[1,3] for a sample consisting of $(TMTSF)_2PF_6$ are $\Gamma_0/(2\pi c) = 0.56$ cm^{-1}, $\lambda_0 = 1$, and $1/(2\pi c\alpha) = 1$ cm^{-1}. Schwartz *et al.*[1] speculated that the physical origin of the quadratic frequency dependence of the second term in Eq. (2) is inelastic electron-electron scattering in a clean three-dimensional Fermi liquid. The physical picture is that at sufficiently low temperatures, there is a crossover from one-dimensional behavior at higher energies to a regime characterized by three-dimensional phase coherence: this is a consequence of the finite coupling between the chains.[4] Experimental evidence for the existence of such a crossover in the organic conductors has also been given by Moser *et al.*[5] via dc transport measurements. However, the authors of Ref. 1 emphasized that the interpretation of the frequency dependence in Eqs. (2) and (3) in terms of Fermi-liquid theory

leads to an anomalously small value of $1/\alpha$. Moreover, the Fermi surface of the organic conductors is nested, so that one should expect a scattering rate linear in ω,[6] in disagreement with the experimental result.[1] In this paper we would like to point out that there exists an alternative (and in our opinion physically more plausible) explanation for the quadratic frequency dependence of the scattering rate and the finite effective mass renormalization in the organic conductors, namely *forward scattering by impurities*. As we shall show below, this interpretation of the data also leads to a natural explanation of the anomalously small value of $1/\alpha$ seen in the experiment.[1]

As discussed in Ref. 7, in the quasi-one-dimensional organic conductors it is natural to expect that the disorder potential seen by the electrons on the chains is weak and slowly varying along the chain. Such a potential can be modelled by a Gaussian random potential $U(x)$ with zero average and long-range correlator

$$\overline{U(x)U(x')} = \gamma_0 C(x - x'), \quad (4)$$

where the overline denotes averaging over the disorder. Here, γ_0 is a measure of the strength of the disorder, and $C(x)$ is assumed to have a maximal range ξ that is large compared with $(2k_F)^{-1}$, where k_F is the Fermi wave vector. In other words, we assume that $C(x)$ is a finite positive constant for $|x| \lesssim \xi$, and vanishes for $|x| \gg \xi$. For convenience we normalize $C(x)$ such that its Fourier transform

$$\tilde{C}(q) = \int_{-\infty}^{\infty} dx\, e^{iqx} C(x) \quad (5)$$

is dimensionless. The above properties of $C(x)$ imply that $\tilde{C}(q)$ vanishes if $|q|\xi \gg 1$. The inverse of ξ can be identified with the maximal possible momentum transfer between two electrons due to scattering by the impurity potential. The requirement $\xi^{-1} \ll 2k_F$ means that impurities do not give rise to backward scattering, i.e., the random potential is dominated by the forward scattering. If this problem is treated by

means of the usual bosonization approach (with linearized energy dispersion), one finds that forward scattering disorder *does not affect the conductivity at all.*[8,9] It is also possible to confirm this result by directly expanding the average conductivity in powers of the impurity potential. *For linearized energy dispersion* one easily verifies that all impurity corrections cancel. This is a consequence of the closed-loop theorem,[10,11] which is well known in the context of the Tomonaga-Luttinger model.[12] The closed-loop theorem implies that at long wave lengths all closed fermion loops with more than two external legs vanish after symmetrization.[10,11] For this cancelation to take place, it is irrelevant whether the external legs represent the dynamic Coulomb interaction or static impurity lines. However, the closed-loop theorem is *only* valid if the energy dispersion $\epsilon(k)$ is linearized close to the Fermi points $\pm k_F$, which amounts to ignoring the quadratic and higher terms in the expansion

$$\epsilon(\pm k_F + q) - \epsilon(\pm k_F) = \pm v_F q + \frac{q^2}{2m_b} + O(q^3). \quad (6)$$

Here v_F is the Fermi velocity and m_b is the band mass. Clearly, in order to calculate the leading effect of forward scattering disorder on the conductivity it is insufficient to work with linearized energy dispersion, as it is done usually in the bosonization approach.

We now present a simple calculation of the effect of forward scattering disorder on the conductivity of an interacting electron gas in one dimension. We assume that the electron gas is metallic and focus on energy scales smaller than possible spin gaps, so that we can ignore backward and umklapp scattering. In principle, one could try to treat the nonlinear terms in the energy dispersion within the framework of bosonization, as it was recently done in a different context in Ref. 13. However, there is a much simpler and physically more transparent solution to our problem. According to Götze and Wölfle,[14] in the perturbative calculation of the conductivity it is often useful to introduce the memory function $M(\omega)$ by setting

$$\sigma(\omega) = \frac{iD_c}{\omega + M(\omega)}, \quad (7)$$

and calculating $M(\omega)$ instead of $\sigma(\omega)$ in powers of the impurity potential. In this way one implicitly takes into account vertex corrections to all orders in perturbation theory. To leading order in the strength of the impurity potential one finds in one dimension[14]

$$M(\omega) = \frac{\gamma_0}{nm_b} \int_{-\infty}^{\infty} \frac{dq}{2\pi} q^2 \widetilde{C}(q) \left[\frac{\Pi(q, \omega + i0^+) - \Pi(q, i0^+)}{\omega + i0^+} \right], \quad (8)$$

where $\Pi(q, \omega)$ is the density-density correlation function and n is the density of the one-dimensional electron gas. The memory function approach has also been used by Giamarchi to study the effect of umklapp scattering on the conductivity of one-dimensional interacting fermions.[15] For the Tomonaga-Luttinger model with short-range disorder Eq. (8) has been evaluated by Luther and Peschel.[16] In this case the anomalous scaling of $\Pi(q, \omega)$ for momenta close to $2k_F$ dominates the conductivity. In contrast, in our case we need

$\Pi(q, \omega)$ only for small momenta, $|q| \lesssim \xi^{-1} \ll 2k_F$. Observe that the leading effect of the nonlinearity in the energy dispersion is already contained in the prefactor $1/m_b$ in Eq. (8); thus, we can calculate $\Pi(q, \omega)$ on the right-hand side of Eq. (8) for linearized energy dispersion. For simplicity we substitute for $\Pi(q, \omega)$ the density-density correlation function of the Tomonaga-Luttinger model[12] with interaction parameters $g_2 = g_4 = \pi v_F F$,

$$\Pi(q, \omega) = Z_q \frac{2\omega_q}{\omega_q^2 - \omega^2}, \quad (9)$$

$$Z_q = \frac{|q|}{\pi\sqrt{1+F}}, \quad \omega_q = \widetilde{v}_F |q|, \quad (10)$$

where $\widetilde{v}_F = \sqrt{1+F} v_F$. Using Eq. (8) and assuming $\widetilde{C}(q) = \Theta(1 - |q|\xi)$, we obtain after a simple calculation for $|\omega| \ll \widetilde{v}_F/\xi$

$$\operatorname{Im} M(\omega + i0^+) = a\omega^2, \quad (11)$$

$$\operatorname{Re} M(\omega + i0^+) = b\omega[1 + O(\omega^2)], \quad (12)$$

where

$$a = \frac{\gamma_0}{\pi n m_b \widetilde{v}_F^4 \sqrt{1+F}}, \quad (13)$$

$$b = \frac{2\gamma_0}{\pi^2 n m_b \widetilde{v}_F^3 \xi \sqrt{1+F}}. \quad (14)$$

Note that a and b both vanish for $1/m_b \to 0$, corresponding to the linearization of the energy dispersion (6). We conclude that forward scattering disorder leads to a conductivity of the form given in Eq. (1), with

$$\Gamma(\omega) = a\omega^2, \quad (15)$$

$$\frac{m^*(\omega)}{m_b} = 1 + b + O(\omega^2). \quad (16)$$

In the absence of other scattering mechanisms this implies for the real and imaginary part of the conductivity at frequencies $|\omega| \ll \widetilde{v}_F/\xi$,

$$\operatorname{Re} \sigma(\omega) = \operatorname{Re}\left[\frac{D_c}{a\omega^2 - i(1+b)\omega} \right] = \frac{\widetilde{D}_c \tau}{1 + (\omega\tau)^2}, \quad (17)$$

$$\operatorname{Im} \sigma(\omega) = \frac{\widetilde{D}_c}{\omega[1 + (\omega\tau)^2]}, \quad (18)$$

where the renormalized charge stiffness $\widetilde{D}_c$ and the time τ are given by

$$\widetilde{D}_c = \frac{D_c}{1+b}, \quad \tau = \frac{a}{1+b}. \quad (19)$$

Keeping in mind that for the derivation of Eqs. (17) and (18) we have assumed $|\omega| \ll \widetilde{v}_F/\xi$, it is easy to see that in the regime where Eqs. (17) and (18) are valid the parameter $|\omega|\tau$

is always smaller than unity. Hence, for $|\omega| \ll \tilde{v}_F/\xi$ we may approximate $\mathrm{Re}\,\sigma(\omega) \approx \tilde{D}_c \tau$ and $\mathrm{Im}\,\sigma(\omega) \approx \tilde{D}_c/\omega$. Note that the imaginary part of the conductivity exhibits at small frequencies the usual $1/\omega$ behavior of a clean system (but with renormalized charge stiffness), whereas the Drude peak $\pi D_c \delta(\omega)$ in the real part of the conductivity of the clean system is completely destroyed by the disorder, and is replaced by a constant $\tilde{D}_c \tau$. In this respect the effect of forward scattering disorder in one dimension is similar to the effect of weak short-range disorder in three dimensions. Note, however, that according to Eqs.(13,19) the effective lifetime τ is proportional to the strength γ_0 of the impurity potential. In contrast, for short-range impurity scattering one finds within the Born approximation that the *inverse* lifetime is proportional to the impurity strength. These nontrivial effects associated with long-range disorder in one dimension are missed within the usual bosonization approach, which predicts a Drude peak $\pi D_c \delta(\omega)$ in the real part of the conductivity, even in the presence of long-range disorder.[8,9]

Keeping in mind that Eqs. (11) and (12) are the leading terms for small ω, Eqs. (15) and (16) are consistent with the frequency-dependent part of the experimentally seen behavior given in Eqs. (2) and (3).[17] The constant part Γ_0 of the scattering rate in Eq. (2) must be due to some other (short-range) impurity scattering mechanism that we have not taken into account in our calculation. A similar assumption concerning the origin of Γ_0 must be made if one interprets the frequency dependence of $\Gamma(\omega)$ to be due to inelastic scattering in a three-dimensional Fermi liquid.[1] Obviously we may identify $\lambda_0 = b$, and

$$\frac{1}{\alpha} = \frac{b}{a} = \frac{2\tilde{v}_F}{\pi \xi}. \qquad (20)$$

Note that α is independent of γ_0 and m_b; it depends only on the renormalized Fermi velocity $\tilde{v}_F$ and on the correlation length ξ of the impurities. Using the experimental value[1] $2\pi c \alpha = 1$ cm we obtain from Eq. (20) $\xi = \pi^{-2}(\tilde{v}_F/c)$ cm. Given a bare interchain hopping $t_\parallel \approx 250$ meV,[1] a reasonable estimate for the Fermi velocity is $\tilde{v}_F \approx 10^7$ cm/s. Then we obtain for the impurity correlation length $\xi \approx 3 \times 10^{-5}$ cm. This is certainly larger than $(2k_F)^{-1}$, so that our interpretation of the low-frequency behavior of the conductivity data[1] in terms of long-range disorder is internally consistent. The large value of ξ explains the anomalously small value of α^{-1} seen in the experiment.[1] The impurity correlation length ξ should not be confused with the transport mean free path l_{tr}, although in the experiment[1] both length scales have the same order of magnitude. A rough estimate for l_{tr} can be obtained from Eqs. (1) and (2) by identifying $l_{\mathrm{tr}} = \tilde{v}_F/\Gamma_0$. Using the measured value $\Gamma_0 \approx 2\pi c \times 0.56$ cm^{-1}, we obtain $l_{\mathrm{tr}} \approx 9 \times 10^{-5}$ cm. The importance of long-range disorder in the organic conductors has been pointed out by Gorkov[7] along time ago: because defects damage only a small fraction of the big planar molecules, it is natural to expect that the imperfection potential of the defects, as seen by the electrons on the conducting chains, is slowly varying along the chains.

An inelastic *single-particle* scattering rate $\Gamma_1(\omega) \propto \omega^2$ due to electron-electron interactions is one of the hallmarks of a three-dimensional Fermi liquid. However, the transport scattering rate $\Gamma(\omega)$ in Eq. (15) is defined in terms of a two-particle Green's function, and it is obviously not related to $\Gamma_1(\omega)$ of a three-dimensional Fermi liquid. This shows that it is crucial to distinguish between transport and single-particle scattering rates in the interpretation of conductivity measurements of quasi-one-dimensional conductors. To understand why the ω^2 behavior of the transport scattering rate $\Gamma(\omega)$ does not imply a similar behavior of the single-particle scattering rate $\Gamma_1(\omega)$, one should keep in mind that the above mentioned cancelations between self-energy and vertex corrections, which are responsible for the ω^2 correction to the transport scattering rate, do not occur in the calculation of the single-particle Green's function. In fact, for non-Fermi liquids in arbitrary dimensions there is no reason to expect that the transport and single-particle scattering rates are simply related.[18] Although Eqs. (15) and (16) look like conventional Fermi liquid behavior, the single-particle Green's function of the system exhibits Luttinger liquid behavior. For example, the single-particle density of states vanishes with a nonuniversal power law.[10,12]

So far we have assumed that the system is strictly one-dimensional. However, realistic experimental systems have a finite hopping $t_\perp$ between the chains, so that it is important to estimate the modification of the above result due to a finite value of $t_\perp$. For simplicity, let us assume that the interchain hopping $t_y = t_\perp$ in one direction transverse to the chains (the y direction) is much larger than in the other transverse direction. To a first approximation we may then ignore the hopping t_z in the z direction. In view of the anisotropy of the interchain hopping in the organic conductors,[19] this approximation is not unreasonable. The transport scattering rate due to forward scattering by impurities can then be estimated from the two-dimensional analog of Eq. (8),

$$M(\omega) = \frac{\gamma_0}{n_2 m_b} \int_{-\infty}^{\infty} \frac{dq_x dq_y}{(2\pi)^2} q_x^2 \tilde{C}(\mathbf{q})$$
$$\times \left[\frac{\Pi(\mathbf{q},\omega + i0^+) - \Pi(\mathbf{q}, i0^+)}{\omega + i0^+} \right], \qquad (21)$$

where n_2 is the two-dimensional density, m_b is the effective band mass in the x direction, $\tilde{C}(\mathbf{q})$ is the two-dimensional Fourier transform of the disorder correlator, and $\Pi(\mathbf{q},\omega)$ is the two-dimensional density-density correlation function. Keeping in mind that by assumption $\tilde{C}(\mathbf{q})$ is dominated by small wave vectors, we may use the random-phase approximation to calculate the density-density correlation function. For small $t_\perp$ it can be shown[20] that $\Pi(\mathbf{q},\omega)$ is dominated by the plasmon pole, so that $\Pi(\mathbf{q},\omega)$ can be written as in Eq. (9). Defining the dimensionless parameter

$$\theta = \frac{|t_\perp|}{E_F}, \qquad (22)$$

where E_F is the Fermi energy, the residue, and the plasmon dispersion are for small θ given by[20]

$$Z_{\mathbf{q}} = \frac{\nu_2 v_F \sqrt{q_x^2 + \theta^2 q_y^2}}{2\sqrt{1+F}}, \quad \omega_{\mathbf{q}} = \tilde{v}_F \sqrt{q_x^2 + \theta^2 q_y^2}, \quad (23)$$

where ν_2 is the two-dimensional density of states. Assuming for simplicity $\tilde{C}(\mathbf{q}) = \Theta(1 - |q_x|\xi)\Theta(1 - |q_y|\xi)$, the low-frequency behavior of $M(\omega)$ is easily calculated. We find that the strictly one-dimensional result for $M(\omega)$ given in Eqs. (11) and (12) remains valid as long as

$$|\omega| \gtrsim \frac{\tilde{v}_F \theta}{\xi} \equiv \omega_\perp. \quad (24)$$

For $|\omega| \ll \omega_\perp$ we find that Eqs. (11) and (12) should be multiplied by an extra factor of order $|\omega|/\omega_\perp \ll 1$. Hence, the finiteness of the interchain hopping induces a crossover in the transport scattering rate due to forward scattering by disorder from a ω^2 behavior at higher frequencies to a $|\omega|^3$ law at frequencies $|\omega| \ll \omega_\perp$. Note, however, that the cross-over frequency $\omega_\perp$ is of order $|t_\perp|/(k_F \xi)$, so that for large ξ and small $t_\perp$ the regime where the $|\omega|^3$ behavior is visible is very small and probably experimentally irrelevant.

Some cautionary remarks are in order: In this work we have assumed that the scattering due to the impurities is forward. It is clear, however, that in the organic conductors also backward scattering by impurities is present, which, in a purely one-dimensional system, should lead to localization. Because this implies a vanishing conductivity, at sufficiently small frequencies Re $\sigma(\omega)$ should become smaller than $\tilde{D}_c \tau$ and eventually vanish. However, in the experimental systems the finite interchain coupling might stabilize a metallic phase. Moreover, from the above calculation it is clear that the constant part Γ_0 of the scattering rate in Eq. (2) cannot be explained by invoking any kind of forward scattering disorder. We are aware of the fact that our explanation of the

constant Γ_0 in Eq. (2) is based on a plausible argument rather than on a microscopic calculation. Such a calculation should treat both forward and backward scattering by impurities and take into account also the weak coupling between the chains.

In summary, we have shown that the frequency-dependent part of the scattering rate seen in the low-frequency conductivity data[1] of the quasi-one-dimensional organic conductors $(TMTSF)_2 X$ can be explained by impurity scattering with small momentum transfers. The data therefore do not necessarily imply that at small energy scales the organic conductors become conventional three-dimensional Fermi liquids. Note that the impurity scattering mechanism proposed here leads for temperatures $T \ll E_F$ to a *temperature-independent* scattering rate. This can be easily verified from Eq. (8), which is also valid at finite temperatures if we use the finite temperature density-density correlation function. Keeping in mind that electron-electron or electron-phonon scattering in general lead to temperature-dependent scattering rates, it should be possible to distinguish the impurity scattering mechanism proposed here from other mechanisms by measuring the temperature-dependence of the scattering rates.[21] Note that in several experiments on organic conductors a T^2 behavior of the resistivity has been observed.[22] This cannot be explained by invoking forward scattering by impurities. If the quadratic frequency dependence of the scattering rate seen in the experiment[1] is really due to impurities, then Eqs. (15), (16), and (20) provide a simple way to estimate the impurity correlation length ξ from the low-frequency data for the conductivity.

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