A model dielectric response function for metallic nanotube ropes

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Abstract

We propose a model dielectric function for ropes of single-walled nanotubes distributed in a glassy graphite host medium. We study the significance of the bosonic charge excitations arising in interacting quasi-one-dimensional systems in the screening processes. We also pay special attention to the role of the intertube Coulomb interactions. In order to compare with experiments, weak relaxation processes are also considered in the relaxation-time approximation.

Key words: nanotube ropes, dielectric response
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1 Introduction

Carbon nanotubes are being considered as substitutes of silicon in many electronic devices (see [1] for a review). Besides their extraordinary chemical stability and strength, they support ballistic electronic transport in a large range of temperatures. On the basis of these properties and several experimental demonstrations of performance in specific devices [2], there is an ongoing effort to find more controllable and efficient methods of synthesis, which are necessary in mass-production electronics. Simultaneously, the importance of the one-dimensional nature of the electronic structure and the role of the electronic interactions in these materials has been recognized. The basic electronic properties of single-walled nanotubes (SWNT) are determined by the helicity of the graphene sheet that forms the nanotube, \textit{i.e.} the chirality. Nanotubes
with \((n,n)\) or \((0,n)\) chiral vectors are metallic. In these systems the interactions, either Coulomb [3] or phonon-assisted [4], play a very important role in the transport properties of nanotubes at low temperatures. The observation of Luttinger Liquid (LL) phenomenology [5,6] has been attributed to the existence of strong interactions in the forward scattering channel [7,8], along with the one-dimensional character of the electronic structure. In addition to clear signals of non-BCS superconducting behavior both in ropes [9] and in small-radius single nanotubes [10], other types of charge instabilities have been predicted theoretically [11,12,13], deriving from the strong renormalization of the electron-electron couplings typical of quasi-one-dimensional systems at very low frequencies.

In this paper we study the screening processes involved in a rope of nanotubes and we compute the dielectric response. Our calculations are valid in the frequency range \(\Delta < \omega \leq E_c\), where \(\Delta\) is the single-particle gap associated with the low temperature instabilities in the rope, and \(E_c\) is an energy scale, to be established later, below which the ropes behave as one-dimensional interacting electron systems. Here, we will be mainly concerned with the longitudinal response. The real part of the conductivity can be expressed in terms of the imaginary part of the dielectric function via Hopfield-like formulas.

2 Dielectric response

We start the discussion giving precise meaning to the range of the Coulomb interaction in nanotubes. Unlike in three dimensional metals, in strictly one-dimensional systems there is no conventional plasmon screening mechanism of the Coulomb interaction that remains long-ranged [14,15,16]. On the other hand, SWNT’s have an extra dimension perpendicular to the main axis, namely the radius of the tube \(R\), and therefore the electronic wave function has an extension in this direction which cuts off the otherwise diverging Fourier transform \(U(k)\) of the interaction. There are several ways to parametrize this partially screened Coulomb interaction. Here, we use one that, despite its simplicity, captures the physical mechanism of the screening at small momentum transfer using just two parameters, namely \(U(k) = \frac{2\pi v_F U_0}{k} \log \left( \frac{k + k}{k} \right)\), and it is well motivated from the physical point of view [17]. \(U_0\) encodes the intensity of the interaction and \(k_c\) is a momentum cut-off determined by a length scale associated to the transverse direction.

The role of the Coulomb interaction in an isolated metallic nanotube has been the subject of intense study, especially in relation to the experimental observation of Luttinger Liquid phenomenology in these systems. Actually, it has been shown [7,8] that the strength of the backscattering and Umklapp processes mediated by the Coulomb potential is reduced by a relative factor
\( \sim 0.1(a/R) \), where \( a \) is the lattice spacing, with respect to forward scattering processes. In addition, the intertube single particle hopping is exponentially suppressed in typical ropes which are made of nanotubes with different diameters and chiralities [18]. Under these conditions the polarization effects can be computed within the RPA approximation. Let us consider then the generic non-interacting polarizability:

\[
\Pi^{(0)} = 2 \int \frac{dp}{2\pi} \frac{f[\varepsilon_s(p)] - f[\varepsilon_{s'}(p+k)]}{\omega + \varepsilon_s(p) - \varepsilon_{s'}(p+k)},
\]

where \( s \) and \( s' \) are arbitrary band indices, and first we estimate the interband polarization effects. We recall some elementary facts about the electronic band structure of \((n,n)\) nanotubes. For nanotubes with typical diameters of \( \sim 1.5 \) nm, curvature effects are negligible and the band structure can be obtained by wrapping the graphene sheet and finding quantization conditions for the wave vector in the radial direction. One finds

\[
\varepsilon_s(q) = \pm t_{\pi} \sqrt{1 + 4 \cos \left( \frac{\sqrt{3}}{2} qa \right)} \left[ \cos \left( \frac{s\pi}{L_y} \right) + \cos \left( \frac{\sqrt{3}}{2} qa \right) \right],
\]

where \( t_{\pi} \sim 2.5 \) eV is the hopping integral between two carbon \( \pi \) orbitals, \( L_y \) is the perimeter of the tube measured in number of hexagons, and \( s \) is a band index \( (s = 0, \pm 1, \ldots) \). Therefore, there are two bands \( (s = 0) \) crossing the Fermi level at exactly two Dirac points \( q = \pm k_F = \pm \frac{4\pi}{3\sqrt{3}a} \). Linearizing the dispersion relation, Eq. (2), we have \( \varepsilon_{s=0}(p) = \pm v_F |p \pm k_F| \). The next completely empty (and full) bands \( s = 1 \) \( (s = -1) \) have two extrema at \( p \sim \pm k_F \). We can expand \( \varepsilon_s(p) \) as \( \varepsilon_s(p) \sim + E_{\text{int}} + p^2/2m \), where, according to Eq. (2), \( E_{\text{int}} = \pm t_{\pi} \sqrt{2 - 2 \cos(\pi/L_y)} \). For the first empty band \( E_{\text{int}} \) is of the order of 1.5 eV for nanotubes with typical diameters \( (\sim 1.5 \) nm). For momenta close to \( \pm k_F \) the interband polarizability is:

\[
\Pi_{\text{int}}^{(0)}(\omega) \sim \frac{1}{\omega^2 - E_{\text{int}}^2} \int \frac{dp}{2\pi} \left[ f(v_F p) - f \left( \frac{(p+k)^2}{2m} + E_{\text{int}} \right) \right]
\]

and only contributes at large frequencies.

On the other hand, there is a remarkable intertube screening effect. Let us call \( g \) and \( u \) the bare intratube and intertube coupling constants, respectively, and \( D \) and \( V \) the corresponding vertices. Since the intertube polarizability between two metallic nanotubes is of the same order of the intratube one, the effective density-density interaction between two electrons located in the same nanotube \( D_i^{(j)} \) is corrected by the interaction of electrons in different nanotubes \( V_i^{(j)}(a,b) \). We are using the conventional notation for super and subindices [19]. The other pair of indices, \( a \) and \( b \), label different nanotubes. We consider spin independent interactions \( g_{ij}^{(j)} = g_{i\perp}^{(j)} \) and we define our bare
couplings as \( g_i^{(j)} = g_{ij}^{(j)} + g_{i\perp}^{(j)} \) (analogously for the \( u \)'s). The virtual processes that renormalize the interactions can be represented in diagrammatic form (see Fig. 1) and translate into the following Dyson equations for the vertices:

\[
D_i^{(j)} = g_i^{(j)} + \sum_{klmn} \eta_{kmi} \eta_{lnj} g_k^{(l)} \Pi D_m^{(n)} \\
\quad + \sum_{klmn \neq a} \sum \eta_{kmi} \eta_{lnj} u_k^{(l)}(a, c) \Pi V_m^{(n)}(c, a)
\]  (4a)

\[
V_i^{(j)}(a, b) = u_i^{(j)}(a, b) + \sum_{klmn \neq a} \sum \eta_{kmi} \eta_{lnj} u_k^{(l)}(a, c) \Pi V_m^{(n)}(c, b) \\
\quad + \sum \eta_{kmi} \eta_{lnj} \Pi [u_k^{(l)}(a, b) D_m^{(n)} + g_k^{(l)} V_m^{(n)}(a, b)]
\]  (4b)

where \( \eta_{kmi} = \delta_{ii} \delta_{km} + \delta_{2i}(1 - \delta_{km}) \); \( \eta_{lnj} = \delta_{li} \delta_{ln} + \delta_{2i}(1 - \delta_{ln}) \) and \( \Pi^{(0)} = \delta_{j'} \Pi_+^{(0)} + (1 - \delta_{j'}) \Pi_-^{(0)} \), \( \delta_{j'j} \) are Kronecker \( \delta \) functions, and \( \Pi_+^{(0)} \) and \( \Pi_-^{(0)} \) are the respective polarizations for right and left branches with linear dispersion, given by

\[
\Pi_+^{(0)}(k, \omega) = \frac{k}{2\pi(\omega - v_F k)}; \quad \Pi_-^{(0)}(k, \omega) = \frac{-k}{2\pi(\omega + v_F k)}.
\]  (5)

superscript zero refers to the pure case.

For a single nanotube we can decouple the equations in the form

\[
D_+^{(4)} = g_+^{(4)} + g_+^{(4)} \Pi_+^{(0)} D_+^{(2)} + g_+^{(2)} \Pi_-^{(0)} D_+^{(2)}
\]

\[
D_-^{(2)} = g_-^{(2)} + g_-^{(2)} \Pi_+^{(0)} D_-^{(4)} + g_-^{(4)} \Pi_-^{(0)} D_-^{(2)}.
\]  (6a)

(6b)

where we used the combinations \( ^4_4 \pm ^4_2 = ^4_4 \) and \( ^2_4 \pm ^2_2 = ^2_2 \). Using the Eqs. (5) we obtain the expressions for the vertices. For instance, \( D_+^{(4)} \) in the positive branch is:

\[
D_+^{(4)}(k, \omega) = (\omega - v_F k) \left[ \frac{C_+}{\omega + u_+ k} + \frac{C_-}{\omega + u_- k} \right]
\]

\[
= \frac{1}{4} \left( g_+^{(4)} + g_-^{(4)} \right) \pm \frac{\pi}{2 u_+} \left( v_F^2 - u_+^2 + \frac{v_F}{2\pi} (g_+^{(4)} + g_-^{(4)}) \right)
\]  (9a)

\[
C_+ = \frac{1}{4} \left( g_+^{(4)} - g_-^{(4)} \right) \pm \frac{\pi}{2 u_-} \left( v_F^2 - u_-^2 + \frac{v_F}{2\pi} (g_+^{(4)} - g_-^{(4)}) \right)
\]  (9b)
In the limit of frequencies larger than the “plasma” and free electron energies ($\omega \gg u_\pm k, v_F k$) we recover the value of the bare coupling $D_i^{(j)}(\omega \to \infty, k) = g_i^{(j)}$. If all the bare intratube couplings $g_i^{(j)} = U(k)$, the intratube vertex Eq. (7) takes the form

$$D_i^{(4)}(k, \omega) = U(k) \frac{\omega^2 - (v_F k)^2}{\omega^2 - (u_+ k)^2},$$

from which one can extract the dielectric function as ($D_i^{(4)} \equiv U/\epsilon^{(0)}$):

$$\epsilon^{(0)}(k, \omega) = 1 - \frac{2U(k)}{\pi v_F} \frac{(v_F k)^2}{\omega^2 - (v_F k)^2}.$$  

This result can be obtained by using the alternative route of finding the elementary bosonic excitations using the bosonization technique, and after that computing the density-density correlations.

Let us now consider the problem of a bundle of $n$ nanotubes. We assume that the perpendicular size of the rope is small and therefore, in first approximation, electrons in different nanotubes interact with the same bare strength $u_i^{(j)}(a, b) = u_0$, whereas $g_i^{(j)} = g_0$. Then Eqs. (4) for the vertices can be rearranged in compact matrix form as $\epsilon^{(0)} (D V)^\top = (g_0 - u_0)^\top$, where the $2 \times 2$
matrix
\[
\epsilon^{(0)} = \begin{pmatrix}
1 - 2g_0\Pi^{(0)} & -2(n - 1)u_0\Pi^{(0)} \\
-2u_0\Pi^{(0)} & 1 - 2[g_0 + (n - 2)u_0]\Pi^{(0)}
\end{pmatrix}
\] (12)
generalizes the dielectric function to the case of intratube and intertube screening, and \(\Pi^{(0)} = \Pi^+_0 + \Pi^-_0\).

The two eigenvalues of \(\epsilon^{(0)}\), Eq. (12), give a measure of the screening due to both intratube and intertube scattering. We assume the same functional dependence for both intratube and intertube couplings, \(g_0 = U_1(k)\) and \(u_0 = U_2(k)\), both given by \(U(k)\) but now characterized by different intensities, \(U_{01}\) and \(U_{02}\), and different cutoff momenta, \(k_{c1}\) and \(k_{c2}\), respectively. In particular, different cutoff momenta can be related to the different radii of a single nanotube and of a nanotube rope. In that case, the eigenvalues of Eq. (12) can still be expressed analytically in terms of the total polarizability \(\Pi^{(0)}\), and will be discussed numerically in the presence of impurities and of a host medium below.

The low energy sector of the theory is integrable and therefore we expect finite weight for the Drude delta peak in the conductivity at zero frequency. In order to make contact with experiments, we need to introduce weak relaxation effects, due to collisions with defects or impurities. We can introduce these effects in the non-interacting polarizability within the relaxation time approximation, which is equivalent to carry out the substitution [20,21]:

\[
\Pi^{(0)}(k, \omega) \rightarrow \frac{(1 + i/\omega\tau)\Pi^{(0)}(k, \omega + i/\tau)}{1 + (i/\omega\tau)\Pi^{(0)}(k, \omega + i/\tau)/\Pi^{(0)}(k, 0)},
\] (13)
to the total polarizability \(\Pi^{(0)} = \Pi^+_0 + \Pi^-_0\). Here, \(\tau\) is an external relaxation time which can be extracted phenomenologically by comparison and fitting of experimental results [22,24]. In the case of a single nanotube, Eq. (11), or for a rope of \(n\) nanotubes with intratube interaction simply proportional to the intertube one \(U_{int}(k) = \eta U(k)\), we explicitly find

\[
\epsilon(k, \omega) = 1 - \tilde{U}_0 \log \left(\frac{k_x + k}{k}\right) \frac{k^2}{\omega^2 - k^2 + i\omega},
\] (14)
where momenta are measured in units of \((v_F \tau)^{-1}\) and frequencies in units of \(\tau^{-1}\), and \(\tilde{U}_0 = [1 + \eta(n - 1)]U_0\). In the case of a rope of \(n\) nanotubes, with intratube and intertube interactions parametrized by \(U_1(k)\) and \(U_2(k)\), respectively, one has to perform Mermin’s substitution for the total polarizability, Eq. (13), in the elements of the dielectric matrix, Eq. (12), before solving the eigenvalue equation. Fig. 2 shows the real and imaginary parts of the larger dielectric eigenvalue, as a function of \(k\) (left panels), and of \(\omega\) (right panels).

Our results are consistent with those found in semiconductor quantum-wire
Fig. 2. Momentum (left panels) and frequency (right panels) dependence of the real (upper panels) and imaginary (lower panels) parts of the larger dielectric eigenvalue of Eq. (12), including weak relaxation effects. Momentum and frequency are rescaled as $v_F \tau k$ and $\omega \tau$, respectively. We take $n = 10$, with $U_{01} = 7$, $U_{02} = 2$, $v_F \tau k_{c1} = 0.25$, and $v_F \tau k_{c2} = 0.05$.

nanostructures [14,23]. Specifically, a system of two arbitrarily close wires with equal densities and effective masses is formally equivalent to the single nanotube we have considered. In our case, $g_i^{(j)} = U(k)$ and the acoustic plasmon is degenerated with the continuum. However, strong renormalization effects are expected at low frequencies [11,12,13] and it would be interesting (but beyond the scope of this communication) to study the impact of such effect in the plasmon peaks, as observed in far infrared spectroscopy or Raman scattering. Likewise, small-radius nanotubes seem good candidates for the observation of three plasmons and an analysis of plasmon stability and spectral weight. Following Ref. [23], one can define the dispersion relations associated with the RPA collective modes as the zeroes of $\det \varepsilon$ in Eq. (12), now taking into account also the effects of impurities. One finds two plasmon-like modes, linearly dispersing in the long-wavelength limit. Specifically, the lower-frequency mode becomes more damped with increasing number $n$ of nanotubes in a rope, which is consistent with the results of Ref. [23], where the lower (acoustic) mode approached the Landau damping region with decreasing spatial separation between the two wires.

So far, we have considered the intrinsic properties of a nanotube rope. In realistic samples the nanotubes are scattered in a medium of glassy graphite produced during the synthesis. Following [24] we will now assume that the nanotubes are placed in a host medium of insulating glassy graphite with a
dielectric constant $\epsilon_h$. The dielectric response of such a composite is given by a generalized Maxwell-Garnett expression [25]

$$\frac{\epsilon_T}{\epsilon_h} = \frac{2(1 - f) + (1 + 2f)(\epsilon/\epsilon_h)}{(2 + f) + (1 - f)(\epsilon/\epsilon_h)},$$

(15)

where $f$ is the filling fraction of the nanotubes. Expression (15) interpolates between the the host dielectric function at $f = 0$ and the intrinsic value $\epsilon(\omega)$, given by Eq. (14), at $f = 1$. The value of the host medium $\epsilon_h$ can be computed within a classical dispersion theory [24]. We assume that $\epsilon_h$ is nearly constant in the frequency range we are interested. The most valuable information that can be extracted when comparing with real experiments is the position and width of the maximum in the conductivity at low frequencies (which is simply related to the dielectric constant). In Fig. 3 we show the total dielectric response in a medium with $\epsilon_h = 1$ as a function of $\omega\tau$ for different values of the filling fraction $f$.

In conclusion, we have studied the nature of the screening processes of metallic single-walled nanotube ropes. We have considered different factors contributing to the longitudinal dielectric response of these systems: intratube and intertube Coulomb interactions, the presence of a glassy graphite environment and the influence of weak relaxation effects produced by impurities or defects.
Our results suggest that metallic nanotubes may be a new playground for the study of collective charge excitations.

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