Fano effect on dynamical conductivity for perpendicular polarization in double-wall carbon nanotubes

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Abstract. The dynamical conductivity of double-wall carbon nanotubes for perpendicularly polarized light to the tube axis is studied by taking into account screening effects, exciton effects and depolarization effects within an effective-mass theory. The exciton peak of the semiconducting inner tube has an asymmetric line shape due to the coupling with a continuum state of the outer tube, indicating the Fano effect. The Fano coupling strength can be tuned by varying the inter-wall distance.

1. Introduction

A carbon nanotube is a quasi-one-dimensional system and thus has characteristic optical properties such as the strong enhancement of exciton effects arising from the electron-hole Coulomb interaction. In fact, exciton effects play a crucial role for absorption of light polarized parallel to the tube axis [1]. For single-wall carbon nanotubes, it is known that the dynamical conductivity for perpendicularly polarized light is reduced considerably by a depolarization effect in comparison with that for parallel polarized light, but still exhibits prominent exciton peaks in semiconducting nanotubes due to the strong Coulomb interaction [2, 3, 4].

For double-wall carbon nanotubes, the Coulomb interaction is suppressed by not only intra-wall screening effects but also inter-wall screening effects. This leads to the reduction of exciton binding energies and band gaps [5]. In this paper, we theoretically clarify perpendicularly polarized optical absorption of double-wall carbon nanotubes by taking into account both exciton effects and depolarization effects within an effective-mass approximation.

2. Model

We consider a double-wall nanotube consisting of an inner tube with radius $R$ and an outer tube with radius $R'$. In usual multi-wall nanotubes where the lattice structure of adjacent walls is incommensurate, inter-wall charge transfer is negligibly small due to cancellation of inter-wall coupling at different sites in the absence of disorder [6, 7]. This fact allows us to discuss screening effects without consideration of inter-wall charge transfer. In the static screened Hartree-Fock
approximation, we can write the screened Coulomb interaction between two electrons on the surface of the tube with radius \( R \) as 
\[
V_{RR}(m, q) = \frac{V_{RR}^0(m, q)}{R} \tilde{\epsilon}_R(m, q) \] 
with 
\[
\tilde{\epsilon}_R(m, q) = \epsilon_R(m, q) + \tau(m(qR, qR'))[\epsilon_R(m, q) - 1],
\]
where 
\[
\epsilon_R(m, q) = \kappa + \frac{V_{RR}^0(m, q)\Pi_R(m, q)}{[V_{RR}^0(m, q)\Pi_R(m, q)]} \] 
and 
\[
\tau(m(qR, qR')) = \frac{V_{RR}^0(m, q)V_{RR}^0(m, q)}{[V_{RR}^0(m, q)\Pi_R(m, q)]}.
\]
Here \( V_{RR}^0(m, q) \) is the bare Coulomb interaction between tubes with radius \( R \) and \( R' \) with the band index \( m \) and the wavenumber \( q \) along the tube axis, and a dielectric constant \( \kappa \) describes screening by electrons in \( \sigma \) bands, core states, and the \( \pi \) bands away from the K and K' points. The polarization function \( \Pi_R(m, q) \) represents contributions of electrons in the vicinity of the K and K' points. Inter-wall screening effects come from the second term of eq. (1) via the presence of the inter-wall Coulomb interaction.

Dynamical conductivity describing absorption spectra is calculated by the Kubo formula.

The conductivity of the tube with radius \( R \) without depolarization effects is written as 
\[
\sigma_R^l(\omega) = \frac{2\epsilon^2}{AL} \sum_{KK'} \sum_{u} \frac{-2i\hbar \omega |\langle u, l; R|v^l_R|g\rangle|^2}{E^u_R(E^u_R - \hbar^2\omega^2 - 2i\hbar\omega\Gamma)},
\]
where \( E^u_R \), \( |u, l; R\rangle \) are the energy and eigen state of an exciton without depolarization effects, respectively, \(|g\rangle \) the ground state, \( L = 2\pi R \) the circumference length of the tube, \( v^l_R \) the velocity operator in the circumference direction for the wavenumber \( 2\pi l/L \) (\( l = \pm 1 \)), \( A \) the length of the tube, and \( \Gamma \) a phenomenological energy broadening.

The absorption spectra can be obtained by performing numerical calculations of perpendicularly polarized exciton states and considering self-consistent electric fields \([4, 8]\). In double-wall nanotubes, the conductivity of each tube with depolarization effects is given by
\[
\tilde{\sigma}_R^l(\omega) = \frac{\sigma_R^l(\omega)}{c_R^l(\omega)}, \quad \tilde{\sigma}_R^r(\omega) = \frac{\sigma_R^r(\omega)}{c_R^r(\omega)},
\]
where for \( R < R' \)
\[
\tilde{\epsilon}_R^l(\omega) = \epsilon_R^l(\omega)c_R^l(\omega) - (R/R')^2[\epsilon_R^l(\omega) - 1]|\epsilon_R^l(\omega) - 1|, \quad \tilde{\epsilon}_R^r(\omega) = \frac{(R/R')^2[\epsilon_R^r(\omega) - 1]}{\epsilon_R^r(\omega) - (R/R')^2[\epsilon_R^r(\omega) - 1]},
\]
with the dielectric functions defined as \( \epsilon_R^l(\omega) = \kappa + \frac{2\pi i l}{\hbar \omega} \sigma_R^l(\omega) \) and \( \epsilon_R^r(\omega) = \kappa + \frac{2\pi i l}{\hbar \omega} \sigma_R^r(\omega) \). The forms of eqs. (4) and (5) are also derived in ref. \([9]\) for static electric fields. Then, we obtain the total conductivity of the system as 
\[
\tilde{\sigma}(\omega) = \frac{R}{2} \sum_l \tilde{\sigma}_R^l(\omega)/2 + R' \sum_l \tilde{\sigma}_R^r(\omega)/2 \] 
\(/(R + R') \).

Excitation energies with depolarization effects are given by zero points of the effective dielectric functions \( \tilde{\epsilon}_R^l(\omega) \) and \( \tilde{\epsilon}_R^r(\omega) \).

The strength of the Coulomb interaction is specified by the dimensionless parameter \( (\epsilon^2/\kappa L)/(2\pi \gamma L) \), where \( \gamma = (\sqrt{3}/2)a\gamma_0 \) is the band parameter with \( a \) the lattice constant and \( \gamma_0 \) the nearest-neighbor hopping integral. In the following, we present results for \( \kappa = 2.5 \) corresponding to \( (\epsilon^2/\kappa L)/(2\pi \gamma L) = 0.16 \) for \( \gamma_0 \approx 2.7 \) eV, and a cut-off energy \( \epsilon_c/(2\pi \gamma L) = 10 \), which is needed to sum over states, corresponding to a typical tube diameter \( \sim 1.4 \) nm.

3. Results

Figure 1 shows the dynamical conductivity \( \tilde{\sigma}(\omega) \) describing the perpendicularly polarized absorption spectra for double-wall nanotubes consisting of (a) semiconducting inner and outer
Figure 1. Calculated absorption spectra of double-wall carbon nanotubes consisting of a semiconducting inner tube and (a) semiconducting outer tube, and (b) metallic outer tube. The dashed (dotted) curves denote the contribution from the inner (outer) tube.

tubes and of (b) semiconducting inner and metallic outer tubes for $R'/R = 1.5$. The vertical arrow located at the higher (lower) energy side indicates the band edge in the inner (outer) tube. Without the depolarization effect, the large two peaks appear in Fig. 1(a). The peak at the higher (lower) energy side is attributed to an exciton in the inner (outer) tube. Because of inter-wall screening, the band gaps and the exciton energies are slightly redshifted from those in single-wall nanotubes. With the depolarization effect, the excitons shift to higher energy sides and their intensities are reduced to about an order of magnitude, similarly to in single-wall nanotubes [4]. The similar intensity reduction can be seen in Fig. 1(b), although in the metallic outer tube there is no exciton due to the absence of backscattering within linear bands [10]. Further, the exciton peak of the inner tube is changed into a dip structure with the depolarization effect.

We find some features of depolarization effects peculiarly in double-wall nanotubes: (i) The inner tube gives a negative absorption in the lower energy region. This means that the direction of the electric field (or induced current) in the inner tube is nearly opposite to that of an external field. (ii) The exciton peak of the inner tube has an asymmetric line shape or even a dip structure due to the coupling with a continuum state of the outer tube, indicating the Fano effect.

The Fano behavior of the exciton peak can be understood qualitatively by a simple model consisting of a single level and a continuum with a constant density of states:

$$\sigma'_R(\omega) = \frac{\kappa R \omega}{2\pi i} \frac{(\hbar \omega_p)^2}{E_R^2 - (\hbar \omega)^2}, \quad \sigma'_l(\omega) = W_e,$$

where $(\hbar \omega_p)^2 = 8h^2 e^2 \langle \langle u, l; R|\psi_R^l|g\rangle \rangle^2 / (\kappa A R^2 E_R^u)$. The zero point of $\epsilon_R^l(\omega)$ is given by

$$(\hbar \omega_0)^2 = E_R^u + \left[ 1 - \frac{(R/R')^2(\kappa - 1)}{\kappa^2 - (R/R')^2(\kappa - 1)^2} \right] (\hbar \omega_p)^2,$$

which is smaller than (and is, for $R/R' = 0$, reduced to) the zero point of $\epsilon_R^l(\omega)$ in single-wall nanotubes, $(\hbar \omega_0)^2 = E_R^u + (\hbar \omega_p)^2$. 

$\kappa = 2.5$, $(\hbar \gamma)/(2\pi\gamma) = 0.16$, $\varepsilon_e/(2\pi\gamma L) = 10 \kappa = 2.5$, and $\Gamma/(2\pi\gamma L) = 0.01$. The dashed (dotted) curves denote the contribution from the inner (outer) tube.
Figure 2. Model calculations of absorption spectra for (a) $W_c/(e^2/h) = 1.5$ and (b) $W_c/(e^2/h) = 4$. The dashed curves denote $\sigma'_R(\omega)$ in eq. (6) and $\hbar\omega_0$ is given by eq. (7).

Figure 2 shows some examples of absorption spectra $\tilde{\sigma}(\omega)$ calculated from the model (6) for various values of $R'/R$. In Fig. 2(a) for a small $W_c$ corresponding to the semiconducting outer tube, the exciton peak maintains its peak structure but becomes asymmetric with decreasing $R'/R$. In Fig. 2(b) for a large $W_c$ corresponding to the metallic outer tube, the exciton peak almost vanishes, while a dip structure appears, exhibiting a crossover from peak- to dip-behavior with decreasing $R'/R$. These describe well the spectra near the exciton peak of the inner tube in Fig. 1. It is clearly reproduced that the Fano line shape is determined by the inter-wall distance and by the intensity of the continuum state of the outer tube. Consequently, the inter-wall distance can change the Fano coupling strength.

In summary, we have found that the Fano effect emerges in dynamical conductivity for perpendicular polarization in double-wall carbon nanotubes. The exciton peak of the semiconducting inner tube becomes asymmetric or changes into a dip structure depending on the coupling with a continuum state of the outer tube. The Fano coupling strength can be tuned by varying the inter-wall distance.

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