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Impurity mediated absorption continuum in single-walled carbon nanotubes

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The authors demonstrate that in single-walled carbon nanotubes, a weak impurity potential can lead to a strong above-gap absorption continuum. The total absorption is enhanced due to the intraband and indirect transitions, as well as plasmon excitations, which are forbidden in perfect nanotubes. Such impurity induced absorption is strongly dependent on the size and chirality of the tube.


Single-walled carbon nanotubes (SWNTs) possess many interesting electronic properties, making them attractive for potential applications in electronic and optoelectronic devices. Their electronic properties display a strong dependence on their size and chirality, as well as a high sensitivity on defects and structural deformations. They have been used as channels in model transistors and have been proposed for building electromechanical sensors. Further, the photon absorption spectra of SWNTs have been exploited to identify their types. There have been considerable interest in using SWNTs to make photodetectors and photovoltaic devices.

The optical properties of SWNTs are expected to be strongly sensitive to defects and impurities, which will play an important role in their use of optoelectronic devices. So far, however, theoretical calculations, especially those based on first-principles methods, have been limited to the case of zero frequency. Our approach takes into account the effects of plasmons and other electronic excitations on the optical absorption. The excitons can affect the absorption characteristics near and below the energy gap, but have a negligible effect on the absorption continuum at high frequencies. Therefore, we shall neglect the exciton effect in this work.

The Hamiltonian of the system can be written as

$$H = H_0 + H_{ee} + H_{ei} + H_{ph},$$

where $H_0$ is the Hamiltonian of noninteracting electrons in a SWNT, $H_{ee}$ is the e-e interaction, $H_{ei}$ is the e-i interaction, given as $\sum U(r-R_i)$, where $R_i$ is the position of the $i$th impurity, and $H_{ph}$ is the coupling of electrons to the electrical field $E(t)=E_0 \exp(-i\omega t)$. The single electron wave functions are given as

$$\psi^{e-i}(k_x,k_y) = (1/\sqrt{2}) \times [\phi_1(k_x) \pm H_{12}(k_x,k_y)]/H_{12}(k_x,k_y)] \phi_2(k_x,k_y),$$

where $\phi_1$ and $\phi_2$ are the TB basis functions, and the nearest-neighbor Hamiltonian matrix element is

$$H_{12} = \gamma_0 \epsilon(k-a)^5 + 2\epsilon^{-2} \alpha^2 \cos(k\alpha/2),$$

where $\alpha=2.46 \text{ Å}$ and $\gamma_0=3.03 \text{ eV}$. The energy dispersion of an $(m,n)$ SWNT (Ref. 1) is

$$E(k) = \epsilon(k_x,k_y).$$

We define $F_{nn'}(k,k+q)$ as the density matrix between the states $(n,k)$ and $(n',k+q)$. Here $a^{+}_n$ ($a_n$) is the creation (annihilation) operator for an electron in the state $|n,k\rangle$. The equation of motion for the density matrix is given as

$$(i\hbar F_{nn'}/\hbar t)(p,p+k) = \left[ H_n(J,p) - E_n(J+L,p) + k_x \right] F_{nn}(p,p+k)$$

$$+ [V(q)n(q,t) - U(q)]$$

$$\times [F_{nn'}(p,p+k) - F_{nn'}(p,p+q)N_{nn'} - N_{nn'}(p+q,p+k)N_{nn'}].$$

(1)

Here $V(q,J)=4\pi e^2 I_0(q,J)K_0(q,J)$ is the Fourier transform of the e-e interaction, $I_0(K_0)$ is the modified Bessel function of the first (second) kind, and $U(q)$ is the Fourier component of $U(r-R_i)$. The electron density operator is written as $n(q) = \Sigma_{k,n} F_{nn'}(k,k+q) N_{nn'}(k_x,q_y,J)$, where $N_{nn'}(k_x,q_y,J)$ is the matrix element between the two TB states.
intraband impurity scattering is inversely proportional to the
tering after the first peak is mainly due to the fact that the
intraband plasmon excitation. The rapid decrease of the scat-
citation is strongest at
written as
semiconducting
the dynamic structure factor of three zigzag SWNTs. For
metallic
R
sorption coefficient
the information on the e-e and e-i interactions; it can be
level excitations. If the tube is metallic, intraband plasmon
the tube. If the tube is semiconducting, the main contribution
SWNTs are strongly dependent on the size and chirality of
phase approximation. 22 By comparing Eq. 
memory function 
is solved in two steps. 27 We first calculate the
static deviation of
ne
a
Real part, (b) imaginary part, and (c) dynamic structural factor.
Figures 2 and 3 demonstrate that the scattering exhibits strong size dependence. For the same type of SWNTs con-
taining the same impurity density, a tube with a large diam-
eter has less scattering, which is true for both semiconduct-
ing and metallic tubes. Our result is a clear demonstration of
the size dependence of electron correlation and dynamic scattering in carbon nanotubes. The probability of forward
is close to the energy of the next interband transition, the
scattering starts to increase again. For semiconducting tubes, the
scattering due to the intraband excitation is absent at low
frequencies. There is a threshold frequency, defined by the
band gap, above which the scattering starts to increase. Fig-
ure 3 shows the rate in three metallic armchair SWNTs. The
overall frequency dependence of the absorption is very simi-
lar to that in the metallic zigzag (15,0) tube, as shown in
Fig. 2.

In Fig. 2 we plot the scattering rate of the same three
zigzag SWNTs, as a function of photon energy, for 
n_i/n_e
= 1%. There is a striking difference between the rate of the
metallic (15,0) tube and semiconducting (10,0) and (20,0)
tubes. For metallic tubes, the main contribution at low fre-
quency is from the single-particle excitations. The scattering
in the region of \hbar \omega < 0.5 \gamma_0 \sim 1.5 \text{ eV} is very similar to that of
a normal metal. The scattering due to the single-particle ex-
citation is strongest at \sim 0.6 \text{ eV}, the characteristic energy of
intraband plasmon excitation. The rapid decrease of the
scattering after the first peak is mainly due to the fact that the
intraband impurity scattering is inversely proportional to the
frequency. Further increase of frequency will open up more
channels through interband excitations. When the frequency

\text{FIG. 2.} Electron scattering rate of three zigzag (10,0),
(15,0), and (20) SWNTs as a function of photon energy.
The plasmon frequency of SWNT is taken to be 2.4/9253 and 0.1 eV for nI
port scattering rate. This is taken to be 0.05 eV for lifetime broadening is usually much smaller than the trans-
the band gap and a large continuum beyond the gap. The

1–3/H20849
10,0
0
4

FIG. 3. Same as Fig. 2 for three armchair (10,10), (12,12), and (20,20)
SWNTs.

scattering is smaller in small tubes than that in large tubes,
because the smaller tubes behave closer to a one-dimensional
system.

For perfect SWNTs, the absorption spectra contain dis-
crete peaks separated by small tails [Fig. 1(b)], as shown by
previous theories with or without many-body effect.10,18–21
The total absorption is the sum of the direct absorption and
the impurity mediated absorption. The total photoresponse (PR) 
\(a(\omega)=\omega\epsilon(\omega)=\omega\epsilon_1(\omega)+4\pi\sigma(\omega)\). Our results (Figs.
1–3) are based on microscopic models that do not contain
any adjustable parameters. The total PR will be dependent on
additional parameters such as the impurity concentration.

In Fig. 4, we plot the PR of a (10,0) tube for two different
impurity concentrations, \(n_I=5\%\) and 10% relative to \(n_e\).
The plasmon frequency of SWNT is taken to be 2.4\(\gamma_0\). The
lifetime broadening is usually much smaller than the trans-
port scattering rate. This is taken to be 0.05 eV for \(n_I=5\%
and 0.1 eV for \(n_I=10\%\). The PR shows an absorption peak at
the band gap and a large continuum beyond the gap. The

peak of the direct absorption decreases while the intensity of
the continuum increases with increasing potential strength.
To experimentally test our result, we suggest to perform optical
absorption measurements in tubes with random defects.
The other way to test this is to measure PR for tubes at
helium temperature and at room temperature. We would ex-
pect the room temperature absorption to show a strong
above-gap continuum due to random potentials (electron-
phonon interaction in this case).

In conclusion, we have formulated a quantum transport
equation to account for the impurity-scattering-induced pho-
ton absorption in SWNTs. We demonstrate that even a very
weak random impurity potential can add a significant con-
tinuum spectrum to the photon absorption. Our result indi-
cates a universal nature of impurity mediated absorption.

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FIG. 4. Photoresponse of a (10,0) tube for two different strengths of the
random potentials.