Optical absorption and electron energy loss spectra of single-walled carbon nanotubes

T. Movlaroo y a,b, A. Kompany a, S.M. Hosseini a,*, N. Shahtahmasebi a

Abstract

We have carried out a first principles calculation on the electronic structure, dielectric function and energy loss spectra of single-walled carbon nanotubes (SWCNTs). Calculations of optical spectra have been performed under electric fields polarized both parallel and perpendicular with respect to the nanotube axis. Our results show that the dielectric function is strongly anisotropic and much larger for the applied electric field parallel than perpendicular to the tube axis. We have calculated first, second, and third optical transitions in several SWCNTs with different chiralities, diameters and lengths. It is revealed that the absorption spectra of 4 Å single-walled carbon nanotubes depend strongly on their chiralities, while the absorption spectra of nanotubes with large diameter hardly show any chirality dependence.

The results show that unlike the optical absorption, the energy loss function of SWCNTs does not significantly depend on chirality and show weak anisotropy. It is also found that the energy loss function peaks for electric fields polarized both parallel and perpendicular to the tube axis happen almost at the same energies, but with rather difference amplitudes.

1. Introduction

Over the last two decades there has been an attracted increasing attention on carbon nanotubes (CNTs) and particularly single-walled carbon nanotubes in both fundamental research and industrial applications [1,2]. The electronic and optical properties of SWCNTs are strongly dependent on tube geometry [3]. By imposing periodic boundary conditions along the circumference direction the energy bands of nanotubes having large diameter can be obtained. Scanning tunneling microscopy and spectroscopy as well as the resonant Raman scattering are important tools which have been used to observe one-dimensional electronic structure of CNTs directly [4,5]. Knowing the optical properties of CNTs are very useful keys to determine their structures. Specially the optical absorption and electron energy loss spectra of the nanotubes are very important to be studied. Optical properties of individual single-walled carbon nanotubes have been measured and reported by some researches [6–8]. Theoretically, the effects of many-electron interactions on the optical properties of single-walled carbon nanotubes have been carried out via ab initio by Spataru et al. [9]. The electronic and optical properties of 4 Å single-walled carbon nanotubes contained inside the zeolite channels have been studied with the local-density approximation (LDA) approach by Weng et al. [10]. From the theoretical calculations it has been revealed that the optical absorption spectra depend strongly on the nanotube geometry and the electric field polarization [11–15].

An optical transition in SWCNTs gives information about the structural characterization of a certain nanotube. The energy difference \( E_g \) between corresponding occupied and unoccupied Van Hove singularities (VHSSs) in the one-dimensional electronic density of states (DOS) is approximately inversely proportional to the tube diameter.

In this work we have used the random-phase approximation (RPA) to obtain the dielectric function and the effect of many-electron interactions such as excitonic effects have been neglected. In the other hand, due to the presence of depolarization for light polarization perpendicular to the tube axis, the effect of local field effects (LFE) play an important role. However, in these calculations the effect of LFE is neglected.

In a few interesting papers quasi-particle formalism have been used and predicted the optical transitions of small SWCNTs in agreement with experimental data [16,17]. In [18,19], Marinopoulos et al. further used time-dependent density functional theory (TD-DFT) on small diameter SWCNTs and showed that for the electric field polarized parallel to the tube axis the local field effects and induced exchange correlation components, beyond the random-phase approximation (RPA) are negligible.

Despite the number of papers already published on the first principle study of SWCNTs but the energy loss spectra of carbon
nanotubes have rarely been studied. In this work we have carried out a first principles calculation on the electronic structure, optical absorption and energy loss function of a series of SWCNTs with different chiralities, diameters and lengths. However, our results can be useful for the explanation of data obtained by the recent experimental tools for nanotubes characterization, such as optical and fluorescence spectrosopes, as well as polarized resonant Raman scattering spectroscopy.

2. Computational details

The calculations have been performed using full potential linearized augmented plane wave (FP-LAPW) method in the framework of density functional theory (DFT) as implemented in the WIEN2k code [20,21]. The Perdew–Burke–Ernzerhof generalized gradient approximation (GGA) was used for the exchange correlation correction [22,23]. The following parameters were chosen for the self-consistency cycles for all computations: the convergence of the basis set is controlled by a cutoff parameter $R_{\text{m}} \times k_{\text{max}} = 5.81$. The values of other parameters are $G_{\text{max}} = 12$ (magnitude of largest vector in charge density Fourier expansion or the plane wave cutoff), $R_{\text{MT}}(C) = 1.28$ au (muffin-tin radius). The iteration halted when the charge difference was less than 0.000001 e between steps as convergence criterion. The optical spectra were calculated using 100 k-points in the first Brillouin zone and setting the Lorentzian broadening with gamma equal to 0.05 eV.

3. Results and discussion

3.1. Structure

The structure of all carbon nanotubes were studied in this work is tetragonal unit cell with $199 \times 1 \times 1$ k-point set mesh. The lattice constants are chosen as $a = \ell$ and $b = c = d + 8$ Å which $\ell$ and $d$ are the tube length and diameter. By optimization of the lattice constants respect to the total energy we found that 8 Å vacuum (nanotube separation) in the lateral directions would be enough to avoid artificial tube–tube interaction. Therefore, the calculations of all spectra were done for the tubes arranged in array as shown in Fig. 1. This figure shows only a $2 \times 2 \times 2$ repeated of single nanotube.

Since we have tetragonal cell, the optical spectra and thus the dielectric tensor would be anisotropic and dielectric tensor are diagonal and have only two components, $x$-direction parallel to tube length and $z$-direction, perpendicular to the tube axis.

3.2. Electronic band structure

3.2.1. Zigzag nanotubes

The calculated electronic band structure and the density of states (DOS) of (13, 0) and (9, 0) zigzag carbon nanotubes are shown in Fig. 2. The Fermi energy level is set to zero which is between the HOMO and LUMO. The electronic band structure and density of states (DOS) of (13, 0) and (9, 0) nanotubes reveal that
these structures have semiconducting behavior with 0.6 eV and 0.1 eV band gap at \( C \), respectively. The calculated energy band gap for different SWCNTs and the results reported by others are summarized in Table 1. The results indicate that the (15, 0) carbon nanotube is a narrow-gap semiconductor on the contrary of finding out to be metallic according to the tight binding method [24].

Our calculations for other \((n, 0)\) zigzag nanotubes also show that all the tubes with \( n \) multiple of 3 are narrow-gap semiconductors, which is in agreement with the previous reported results [25,26]. The zigzag \((n, 0)\) nanotubes with \( n < 7 \), found to have metallic behavior, due to the strong curvature in small nanotubes. However, this result is also in contrast with the tight binding method which predicts that (5, 0) and (4, 0) nanotubes would be semiconductors.

The DOS of (13, 0) and (9, 0) carbon nanotubes shown in Fig. 2 demonstrate a series of traits, Van Hove singularities (VHSSs) near the Fermi energy, which is due to the one-dimensional nature of the electronic bands in SWCNTs. In Fig. 2 \( E_i \) is the energy difference between the corresponding occupied and unoccupied Van Hove singularities in the one-dimensional electronic density of states (DOS). The arrows in Fig. 2a and b indicate the optically allowed interband transitions between valence and conduction states. These four peaks are labeled \( E_1 \)–\( E_4 \) in the optical spectrum as shown in Fig. 4. The (13, 0) and (9, 0) zigzag nanotubes have \( D_{2h} \) symmetry. Further analysis of selection rules shows that the allowed dipole transitions for \( D_{2h} \) symmetry are:

- \( a_1g \), \( a_1u \), \( b_3g \), \( b_3u \), \( b_{2g} \), \( b_{2u} \), \( b_{1g} \) and \( b_{1u} \). These transitions are indicated in Table 2 for the electric field parallel to the tube axis.

### 3.2.2. Small diameter (4 Å)

The small diameter carbon nanotubes will show either semiconducting or metallic behavior, depending on the tube chirality and diameter [30,31]. The calculated DOS and energy band structure of (5, 0), (3, 3) and (4, 2) nanotubes with 4 Å diameter are shown in Fig. 3. The results show that both (3, 3) and (5, 0) carbon nanotubes have metallic behavior while (4, 2) is semiconducting with a small gap of 0.24 eV at X-point and the HOMO–LUMO gap is about 1.2 eV. The calculated band structures results differ significantly from simple \( p \)-band tight binding model. This simple \( p \)-band model predicts that (5, 0) nanotube has semiconductor behavior. This difference between our calculations and tight binding model is due to the curvature effects. In fact, the hybridization of \( \pi \) and \( \sigma \) orbital causes the modification of the band structure and hence the behavior of carbon nanotubes.

The larger curvatures give rise to the larger hybridizations. Since the 4 Å nanotubes are the smallest nanotubes studied so far [7] and have the largest curvature, thus having the strongest hybridization. The energy bands and DOS calculated, in this work, for 4 Å carbon nanotubes are very close to the data reported previously by Guo et al. [29]. Our results also show that the DOS of (3, 3) armchair CNT is not symmetric with respect to the Fermi level.

### 3.3. Dielectric function

The imaginary part of the dielectric tensor can be computed from the knowledge of the electronic band structure of a solid. In

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Fig. 2. The density of states and the electronic band structure of zigzag (a) (13, 0) and (b) (9, 0) carbon nanotube.
within the framework of the random-phase approximation we can use the following well-known relations [32]:

\[ \text{Im} \epsilon_{ab}^{\text{int}}(\omega) = \frac{\hbar^2 e^2}{\pi m \omega^2} \sum_{\Delta} \int dk |c_i|^2 |v_k|^2 \langle v_k | p^2 | c_k \rangle \delta(e_k - e_{\Delta k} - \omega) \]

(1)

The absorption spectrum is proportional to the sum over interband transitions from occupied valence \(v_k\) to empty conduction \(c_k\) states over the Brillouin-zone \(k\) points. From the imaginary part of the dielectric tensor component the corresponding real part is obtained by the Kramers–Kronig transformation.

\[ \text{Re} \epsilon_{ab}^{\text{int}}(\omega) = \delta_{ab} + \frac{2}{\pi} \int_0^\infty \frac{\alpha}{\alpha^2 - \omega^2} \text{Im} \epsilon_{ab}^{\text{int}}(\omega) d\omega \]

(2)

The limit of linear optics, in the case of non-spin polarized, and within the framework of the random-phase approximation we can use the following well-known relations [32]:

\[ \frac{\text{Im} \epsilon_{ab}^{\text{int}}(\omega)}{C_2^2} = \frac{\hbar^2 e^2}{\pi m \omega^2} \sum_{\Delta} \int dk |c_i|^2 |v_k|^2 \langle v_k | p^2 | c_k \rangle \delta(e_k - e_{\Delta k} - \omega) \]

(1)

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\[ \text{Re} \epsilon_{ab}^{\text{int}}(\omega) = \delta_{ab} + \frac{2}{\pi} \int_0^\infty \frac{\alpha}{\alpha^2 - \omega^2} \text{Im} \epsilon_{ab}^{\text{int}}(\omega) d\omega \]

(2)

For the metals due to the intraband transitions contribution, we would have [33]:

\[ \text{Im} \epsilon_{ab}^{\text{intr}}(\omega) = \frac{\Gamma \alpha_{pl}^2}{\text{Im} (\omega^2 + \Gamma^2)} \]

(3)

\[ \text{Re} \epsilon_{ab}^{\text{intr}}(\omega) = 1 - \frac{\alpha_{pl}^2}{\text{Im} (\omega^2 + \Gamma^2)} \]

(4)

where \(\Gamma\) is a lifetime broadening and \(\alpha_{pl}\) is the plasma frequency.

The calculated imaginary part of the dielectric function of small diameter (4 Å) carbon nanotubes, zigzag (5, 0), armchair (3, 3) and chiral (4, 2) SWCNTs are shown in Fig. 4a–c respectively. The imaginary part of the dielectric function of larger diameter (10, 0), (13, 0) and (16, 0) carbon nanotubes, for the electric field polarized parallel to the tube axis, are displayed in Fig. 4d–f respectively. In the imaginary part of the dielectric function the observed peaks, labeled with \(E_i\), indicate the direct interband transitions between the Van Hove singularities of the density of states.

Fig. 4 shows that the number of optical transitions (peaks) decreases with reducing the tube diameters, which is due to the larger \(\pi + \sigma\) hybridization in the tubes with smaller diameters. The smaller nanotubes have the larger curvature and so show more hybridization between \(\pi\) and \(\sigma\) orbital. The first peak position for armchair nanotube (3, 3) is seen near energy of 3.0 eV which is in agreement with the reported previous theoretical and experimental measurements [7,18]. This peak is mainly due to the HOMO to LUMO + 1 and HOMO to LUMO transitions. In armchair CNTs, optical transitions between HOMO and LUMO bands are symmetry forbidden. Due to the metallic behavior of (3, 3) carbon
nanotube and according to the Eqs. (3) and (4), the imaginary and the real parts of the dielectric function of \((3, 3)\) carbon nanotube has singularity at zero frequency for the polarization parallel to the tube axis as illustrated in Fig. 4b.

The real and imaginary parts of the dielectric function of \((13, 0)\) and \((10, 0)\) carbon nanotubes for both polarizations, parallel and perpendicular to the tube axis are displayed in Figs. 5 and 6 respectively. The optical spectrum can be divided into two regions, namely, the low-energy range from 0 to 6 eV and the high-energy range from 6 to 20 eV. For the electric field polarized parallel to the tube axis, the imaginary part of the dielectric function in the low-energy region consists of a few distinct peaks (see Fig. 6). These peaks can give information about the chirality of the grown carbon nanotubes. In the low-energy range the absorptive part is larger than in the high-energy range. Fig. 6 also indicates that the optical gap of \((13, 0)\) and \((10, 0)\) CNTs for the electric field polarized perpendicular to the tube axis is larger than that for the electric field polarized parallel to the tube axis.

The first, second, third and fourth optical transitions in different SWCNTs are listed in Table 3. These results show that the absorption spectra of small diameter (4 Å) single-walled carbon nanotubes depend strongly on their chiralities, while the absorption spectra of carbon nanotubes with large diameter are insensitive to the tube chiralities. The calculated electronic dielectric constants and optical gaps of different types of SWCNTs for both polarizations, parallel (x) and perpendicular (z) to the tube axis are listed in Table 4.

The calculated results show that the optical gaps of the large diameter carbon nanotubes for the perpendicular polarization is larger than for the parallel polarization while for the small diameter (4 Å) carbon nanotubes we have found larger optical gaps for the parallel polarization than perpendicular polarization. The value of optical gaps increases by decreasing tube diameters. It is also revealed from Table 4 and also Figs. 5 and 6 that the dielectric function is highly anisotropic and is significantly larger for the external field polarized along the tube axis comparing to the perpendicular polarized field.

For the polarization perpendicular to the tube axis, the measured dielectric functions are small and flat. The comparison of first optical transitions of \((3, 3)\), \((4, 2)\) and \((5, 0)\) small diameter (4 Å) SWCNTs with the theoretical and experimental results reported previously are summarized in Table 5. These results show the importance role of chirality in the optical spectra of small diameter SWCNTs. Table 5 also indicates that the optical gap of small diameter carbon nanotubes is approximately inversely proportional to the tube length.

### 3.4. Electron energy loss spectroscopy (EELS)

EELS is a well known method for investigation various aspects of materials [35]. The advantage of this method is that it covers a vast energy range which includes the non-scattered, elastically scattered electrons (zero loss) and valence interband transitions (valence loss). Also the fast electrons excite the inner shell electrons (core loss) or induce core level excitation of near edge

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**Table 3**

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<th>Chirality ((n, m))</th>
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<th>Second (E_2) (eV)</th>
<th>Third (E_3) (eV)</th>
<th>Fourth (E_4) (eV)</th>
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**Table 4**

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nanotubes will show metallic or semiconducting behavior, which depends on their chiralities.

The calculated results show that the number of optical transitions decreases with reducing the tube diameters due to the larger $\pi + \sigma$ hybridization in the tubes with smaller diameters. The calculated optical gap of the large diameter carbon nanotubes is found larger for polarization perpendicular than polarization parallel to tube axis. While for the small diameter (4 Å) carbon nanotubes we have found larger optical gaps for parallel than perpendicular polarization.

Our results show that the dielectric function is anisotropic and is much larger for the electric field applied along the tube axis than parallel to the tube axis. The calculated absorption spectra of 4 Å single-walled carbon nanotubes depend strongly on their chiralities, while the absorption spectra of nanotubes with large diameters hardly show any chirality dependence. In 4 Å diameter nanotubes it was found that their optical gaps are inversely proportional to the tube length.

The calculated energy loss spectra, for all SWCNTs studied in this work, have a sharp $\pi$ electron plasmon peak at about 5 eV and a broad $\pi + \sigma$ electron plasmon peak around 15–22 eV. It is also found that the energy loss function peaks for both electric fields polarized parallel and perpendicular to the tube axis happen almost at the same energies, but with rather difference in their amplitudes. Unlike the optical absorption, the energy loss function of SWCNTs does not significantly depend on chirality and also show weak anisotropy.

### 4. Conclusions

We have calculated the electronic structure, dielectric function and energy loss spectra of different single-walled carbon nanotubes by using density functional theory. Our calculations for (n, 0) zigzag nanotubes show that all the tubes with n multiple of 3 are narrow-gap semiconductors, which is in contrast with the tight binding method which predicts to have metallic behavior.

The calculated energy gap for zigzag nanotubes show that tubes with n less than 7, have metallic behavior due to the strong curvature in small nanotubes. It is also found that small diameter carbon nanotubes will show metallic or semiconducting behavior, which depends on their chiralities.

The calculated results show that the number of optical transitions decreases with reducing the tube diameters due to the larger $\pi + \sigma$ hybridization in the tubes with smaller diameters. The calculated optical gap of the large diameter carbon nanotubes is found larger for polarization perpendicular than polarization parallel to tube axis. While for the small diameter (4 Å) carbon nanotubes we have found larger optical gaps for parallel than perpendicular polarization.

Our results show that the dielectric function is anisotropic and is much larger for the electric field applied along the tube axis than parallel to the tube axis. The calculated absorption spectra of 4 Å single-walled carbon nanotubes depend strongly on their chiralities, while the absorption spectra of nanotubes with large diameters hardly show any chirality dependence. In 4 Å diameter nanotubes it was found that their optical gaps are inversely proportional to the tube length.

The calculated energy loss spectra, for all SWCNTs studied in this work, have a sharp $\pi$ electron plasmon peak at about 5 eV and a broad $\pi + \sigma$ electron plasmon peak also seen around 15–22 eV. It is also found that the energy loss function peaks for both electric fields polarized parallel and perpendicular to the tube axis happen almost at the same energies, but with rather difference in their amplitudes. Unlike the optical absorption, the energy loss function of SWCNTs does not significantly depend on chirality and also show weak anisotropy.

### References


