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## Nonlinear optical responses induced by Auger ionization in single-walled carbon nanotubes

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*New Journal of Physics* **14** (2012) 023053 (10pp)

Received 8 September 2011

Published 24 February 2012

Online at <http://www.njp.org/>

doi:10.1088/1367-2630/14/2/023053

**Abstract.** We theoretically study the optical response of single-walled carbon nanotubes (SWNTs) under high-intensity laser irradiation. Due to the quasi-one-dimensional structure of SWNTs, the Coulomb interaction between excitons is enhanced relative to the bulk. As a result, excitons can be ionized to electrons and holes by the Auger ionization process under high-intensity laser irradiation. Taking into account the effect of Auger-ionized carriers, we calculate absorption spectra by solving the Bethe–Salpeter equation in the tight-binding approximation. We found that Auger-ionized carriers induce a band-gap renormalization and a bleaching of excitons, leading to a nonlinear optical response. Our calculation reveals the strong influence of Auger-ionized carriers on excitons under high-intensity laser irradiation, which may unravel the recently observed nonlinear behavior of photoluminescence emission spectra.

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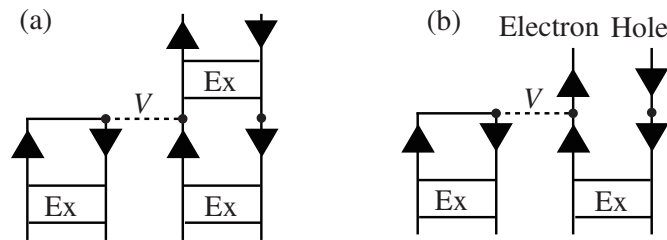
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**1. Introduction**

Semiconducting single-walled carbon nanotubes (SWNTs) provide an excellent experimental platform for investigating exciton physics by probing unusual optical properties [1] that originate from the strong Coulomb interaction between photo-excited carriers that is inherent to their quasi-one-dimensional structure. Excitons have binding energies of up to a few hundreds of meV [2–4] in SWNTs and thus are stable even at room temperature. Consequently, excitons strongly affect the optical properties of SWNTs and can be used to investigate nonlinear optical effects that originate from many-body interactions among excitons [5, 6]. Several experimental studies have recently investigated the optical properties of SWNTs under high-intensity laser irradiation [7–11] with the expectation of observing nonlinear optical effects in SWNTs. However, despite considerable effort to investigate the nonlinearity of spectra, no experiments have so far revealed phenomena such as band-gap renormalization [12, 13], a Fermi-edge singularity [14–17], an exciton Mott transition or an exciton Bose condensation [18], which have been the main objects of study in the nonlinear many-body physics of semiconductors.

Nevertheless, recent experimental studies have revealed some evidence of nonlinear optical phenomena in SWNTs caused by the many-body effects among interacting excitons. In particular, photoluminescence (PL) emission spectra of SWNTs have been shown to saturate under high-intensity laser irradiation. This PL saturation has been attributed to nonradiative Auger recombination of excitons [19]. Since the Coulomb interaction is enhanced not only between electrons and holes but also between excitons in SWNTs, efficient scattering occurs between excitons, leading to the fast nonradiative recombination of excitons. In this sense, the Auger process is classified as a nonlinear process induced by many-body exciton effects in SWNTs. The Auger recombination rate exceeds the rates of other recombination processes under high-density excitation. Thus, the Auger recombination process dominates over other relaxation processes, such as the one mediated by phonons [7, 19–21]. In addition to the above-mentioned PL saturation, Xiao *et al* [10] have observed declining behavior at pump fluences above the saturation region. This peculiar optical response cannot be explained in terms of the Auger process of excitons; other mechanisms are required to explain such nonlinear optical responses.



**Figure 1.** Auger processes. Inelastic scattering between excitons produces (a) an excited exciton or (b) an ionized electron and hole.

The present study reveals the strong impact of ionized carriers on excitons under high-intensity laser irradiation and seeks to determine the underlying mechanism for the unusual PL properties of SWNTs by considering many-body effects of excitons induced by the surrounding electron–hole carriers. Even at room temperature, dissociation is unlikely to be induced by thermal agitation because excitons have large binding energies in SWNTs. Instead, we focus on the fact that the Auger process can dissociate excitons into ionized electron–hole pairs. Our calculations demonstrate that Auger-ionized carriers can give rise to a nonlinear optical response, which may be the origin of the interesting behavior observed under intense laser irradiation.

This paper is organized as follows. In section 2, we briefly describe the method used to calculate absorption spectra in the presence of the many-body effects of ionized electron–hole carriers. Section 3 presents the calculation results and discusses them in the light of experimental results. Section 4 summarizes the present findings.

## 2. Theoretical methods

### 2.1. Auger ionization

High-intensity laser irradiation generates multiple excitons in SWNTs and these excitons interact with each other. As a result of scattering, some excitons are excited while others recombine due to energy transfer through the Coulomb interaction. Two types of recombination processes occur in Auger processes [22]: (i) two excitons combine to form a single excited exciton (figure 1(a)) and (ii) two excitons form a single electron–hole pair (i.e. an ionized electron and a hole carrier) (figure 1(b)). Electron–hole pairs are generated by the latter process if the transferred energy is sufficiently larger than the binding energy of the scattered exciton to dissociate the exciton. This inelastic scattering process between excitons is called nonradiative Auger ionization. It has been found to be very efficient in SWNTs [5] due to their quasi-one-dimensional structure. The Auger ionization rate can be calculated by evaluating the diagrams in figure 1 [5, 23]. For example, tight-binding calculations have shown that the Auger ionization rate reaches  $0.15 \text{ ps}^{-1}$  for micrometer-long SWNTs with a diameter of 1.35 nm [24].

However, it is difficult to distinguish both Auger processes in PL experiments, as discussed in [22]. Here, we assume that Auger ionization occurs in SWNTs and that Auger ionized carriers surround the excitons produced during laser irradiation. In the following, we consider how many-body nonlinear effects induced by the surrounding electron–hole carriers on excitons give rise to the unusual properties of PL spectra in the saturation region [7–11].

## 2.2. Many-body exciton theory

When there are ionized carriers, it is essential to include screening and phase-filling effects that originate from the Pauli exclusion principle for ionized electron and hole carriers. The bound state embedded in such ionized carriers can be determined by solving the Bethe–Salpeter equation for the optical susceptibility, which can be used to obtain absorption spectra. In this subsection, we briefly describe how to calculate absorption spectra using the theory of Haug and Schmitt-Rink [12].

In general, absorption spectra can be calculated via the following relation:

$$\begin{aligned}\alpha(\omega) &\propto \sum_{k,k'} M(k)M^*(k')G(k, k', \omega) \\ &= \sum_k M(k)\chi(k, \omega),\end{aligned}\quad (1)$$

where  $\chi(k, \omega) \equiv \sum_{k'} M^*(k')G(k, k', \omega)$  is the optical susceptibility,  $M(k)$  is the matrix element of the dipole transition, and  $G(k, k', \omega)$  is the two-particle Green function for an electron and a hole. The equation of motion for the susceptibility (i.e. the two-particle Green function) is derived as the Bethe–Salpeter equation. In the quasi-static approximation (in which the Coulomb hole is included in the self-energy in addition to the screened exchange, but the dynamic responses of the ionized carriers are ignored) this becomes [12]

$$\chi(k, \omega) = \chi_0(k, \omega) \left[ 1 - \frac{1}{M(k)} \sum_{k'} V_s(k-k')\chi(k', \omega) \right], \quad (2)$$

where  $V_s(k) \equiv V(k)/\kappa\epsilon(k)$  is the statically screened Coulomb interaction and  $V(k)$  denotes the Fourier transform of the Coulomb potential  $V(r)$ . For the Coulomb potential between  $\pi$ -orbitals, we employ the Ohno potential  $V(r) = U/\sqrt{(\frac{4\pi\epsilon_0}{e^2}Ur)^2 + 1}$  (with  $U = 11.3$  eV), which is known to realistically describe optical responses in SWNTs [3, 25, 26]. We further choose a dielectric constant of  $\kappa = 3.3$  to incorporate screening effects due to core states,  $\sigma$  bands and the surrounding environment, while the dielectric function  $\epsilon(k)$  for the polarization of  $\pi$ -electrons is calculated in random phase approximation, i.e.

$$\epsilon(k) = 1 + V(k)\Pi(k). \quad (3)$$

In the tight-binding approximation, the polarization function is given by [26]

$$\Pi(k) = -2 \sum_q \left[ \frac{|\sum_{n=A,B} C_n^{v*}(q)C_n^c(k+q)|^2}{\epsilon_c(k+q) - \epsilon_v(q)} + \frac{|\sum_{n=A,B} C_n^{c*}(q)C_n^v(k+q)|^2}{\epsilon_c(q) - \epsilon_v(k+q)} \right], \quad (4)$$

where  $\epsilon_a(k)$  is the single-particle energy and  $C_n^a$  the coefficient of the wave function for  $\pi$ -electrons. Here,  $a = c(v)$  indicates the conduction (valence) band of  $\pi$  electrons, while  $n = A, B$  labels the two-graphene sublattice.

The bare susceptibility in equation (2) can be obtained from

$$\chi_0(k, \omega) = \sum_{k'} M(k')G^0(k, k', \omega), \quad (5)$$

where  $G^0$  stands for the bare Green function of an electron–hole pair

$$G^0(k, k', \omega) \simeq \frac{F(k)}{\hbar\omega + i\delta - E_c(k) - E_h(k)}\delta_{k,k'}. \quad (6)$$

Here, we have defined the phase-filling factor as

$$F(k) \equiv 1 - f_e(k) - f_h(k), \quad (7)$$

with  $f_i(k)$  standing for the Fermi-distribution function,

$$\begin{aligned} f_i(k) &\equiv f_i(E_i(k)) \\ &= \frac{1}{\exp[\beta(E_i(k) - \mu_i)] + 1}, \end{aligned} \quad (8)$$

and  $\beta$  for the reciprocal temperature. The quasi-particle energy,  $E_i(k)$ , can be calculated by applying the random phase approximation,

$$E_i(k) = \varepsilon_i(k) + \Sigma_i(k) \quad (i = e, h), \quad (9)$$

while the chemical potential  $\mu_i$  is determined by satisfying a given density:

$$n = \int \frac{dk}{\pi} f_i(k). \quad (10)$$

In the quasi-static approximation, the self-energy correction to the single-particle energies  $\varepsilon_e(k)$  or  $\varepsilon_h(k)$  is [12]

$$\Sigma_i(k) = \sum_{k'} \left\{ \frac{1}{2} [V_s(k') - V(k')] - V_s(k - k') f_i(k) \right\}, \quad (11)$$

where the first term expresses the screened exchange and the second term expresses the Coulomb hole that describes the difference in classical Coulomb self-energies with and without an ionized carrier. Using the quasi-particle energy for electrons and holes, the band-gap energy is defined by

$$\Delta(k) \equiv E_e(k) + E_h(k). \quad (12)$$

To solve the Bethe–Salpeter equation (2), we perform a numerical matrix inversion for a given carrier density (equation (10)) [27]. For this, it is useful to introduce the vertex function  $\Gamma(k, \omega)$ , relating bare and dressed susceptibilities,

$$\chi(k, \omega) = \Gamma(k, \omega) \chi_0(k, \omega). \quad (13)$$

As equation (13) shows, all the interaction effects between electrons and holes are contained in the vertex function. Substituting this into equation (2), we obtain

$$\Gamma(k, \omega) = 1 - \frac{1}{M(k)} \sum_{k'} V_s(k - k') \chi_0(k', \omega) \Gamma(k', \omega), \quad (14)$$

or, in vector notation,

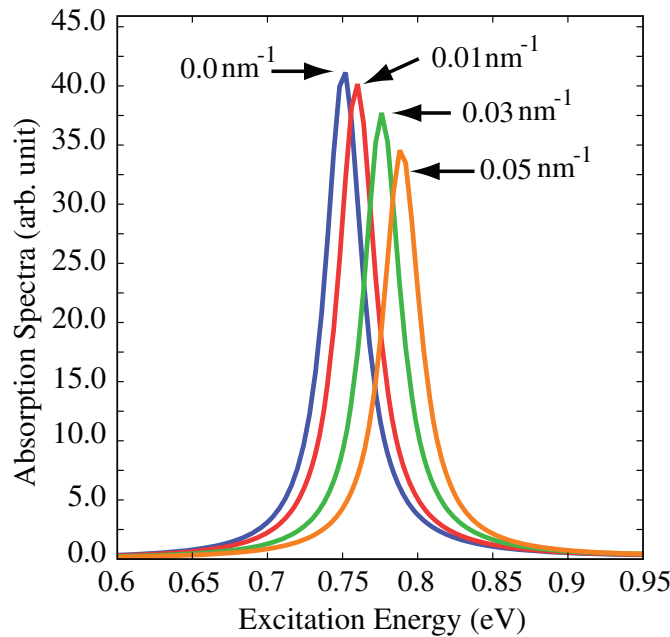
$$\mathbf{\Gamma}(\omega) = \mathbf{1} - \hat{A}(\omega) \cdot \mathbf{\Gamma}(\omega), \quad (15)$$

where  $\mathbf{1}$  is a unit vector and  $\hat{A}$  is given by

$$A_{k,k'}(\omega) = \frac{1}{M(k)} V_s(k - k') \chi_0(k', \omega). \quad (16)$$

We then obtain

$$\mathbf{\Gamma}(\omega) = [\hat{\mathbf{1}} + \hat{A}(\omega)]^{-1} \cdot \mathbf{1}, \quad (17)$$



**Figure 2.** Absorption spectra of (17, 0) SWNTs for electron–hole densities of 0.0, 0.01, 0.03 and 0.05 nm<sup>-1</sup> at  $T = 300$  K.

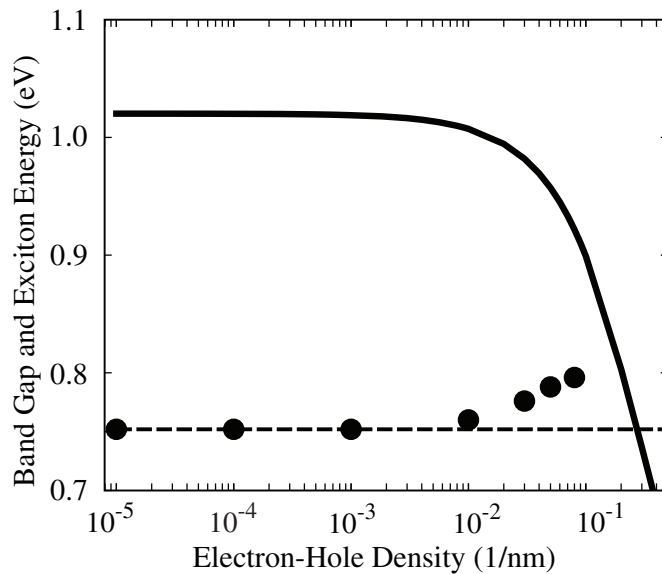
where  $\hat{1}$  is a unit matrix. We can calculate the vertex function from this equation by inverting the matrix  $\hat{1} + \hat{A}(\omega)$ . We obtain the susceptibility by substituting the vertex function into equation (13). Finally, we obtain the absorption spectra from equation (1). To evaluate the above quantities, we used the tight-binding approximation that takes into account nearest-neighbor hopping of 3.0 eV. For the optical response in doped SWCNTs, a calculation was performed recently [28] by using a many-body *ab initio* approach based on the density functional theory [4, 29]. We consider the (17, 0) nanotube as a representative semiconducting SWNT.

### 3. Results and discussion

#### 3.1. Absorption spectra

Before we present below the calculated absorption spectra, we first briefly describe the implications of the theory described in the previous section. A finite density of electron–hole pairs affects the exciton states by screening the Coulomb interaction and by phase-space filling. The screening expressed by the self-energy correction (equation (11)) reduces the quasi-particle energies (equation (9)), leading to a reduction in the band-gap energy (equation (12)). On the other hand, the phase-space filling factor  $F(k)$  defined by equation (7) weakens the electron–hole interaction, reducing the binding energy of excitons. The interplay between these two effects determines the absorption spectra at finite electron–hole densities.

Figure 2 shows the absorption spectra obtained for electron–hole densities of 0.0, 0.01, 0.03 and 0.05 nm<sup>-1</sup> at  $T = 300$  K. At zero density, the main peak corresponds to the lowest exciton state. Figure 2 reveals two salient features at finite densities: the position of the exciton peak shows a blueshift of 36 meV at 0.05 nm<sup>-1</sup> and the exciton peak is bleached with increasing



**Figure 3.** The electron–hole density dependence of the band gap and the exciton energy of (17, 0) SWNTs at  $T = 300$  K. The solid line represents the band gap and the full circles represent the exciton energy. As a guide to the eyes, the dashed line represents the exciton energy vanishing electron–hole density.

electron–hole density. The following subsection discusses in detail these two features of the absorption spectra.

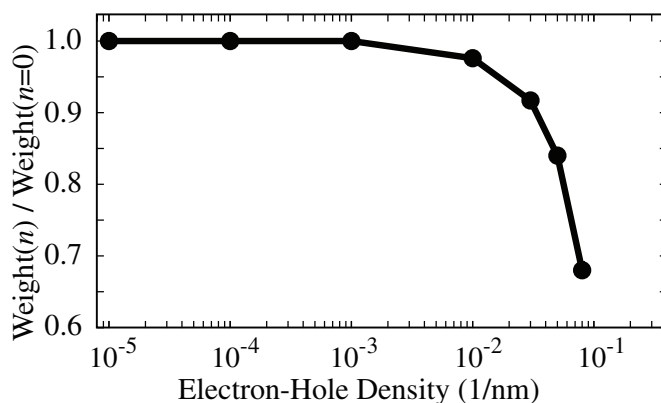
### 3.2. Band-gap energy and exciton energy

Figure 3 shows the calculation results for the band-gap energy defined by equation (12) together with the exciton energy derived from the pole of the susceptibility at  $T = 300$  K. The band-gap energy calculated using equation (12) decreases with increasing carrier density. This is band-gap renormalization. Both the Coulomb hole and the screened exchange terms of equation (11) reduce the quasi-particle energy with increasing carrier density.

On the other hand, the exciton energy (represented by the points in figure 3) is blue-shifted with increasing carrier density. This energy shift in the excitons is ascribed to the quasi-static approximation employed here to solve the Bethe–Salpeter equation (2). The exciton energy should not shift much when dynamic effects are fully included due to the almost complete compensation of changes between the exciton binding energy and the band-gap energy [30, 31]. On further increasing the carrier density, a Mott transition is likely to occur, which is associated with negative absorption (i.e. optical gain) [12]. The gain spectra for SWNTs have been obtained by solving the semiconductor Bloch equation under the quasi-equilibrium approximation [32], although recent first-principles calculations for doped SWNTs imply that the Mott transition will be hardly observed when dynamic screening by acoustic plasmons is properly treated [28]. In this respect, our calculation result is valid up to a carrier density of about  $10^{-1} \text{ nm}^{-1}$  where a Mott transition seems to occur.

Two-photon spectroscopy is a promising technique for measuring the band gap and for directly observing band-gap renormalization. Lee has recently used photocurrent spectroscopy





**Figure 4.** The electron–hole density dependence on the oscillator strength of (17, 0) SWNTs normalized to unity at zero density.

to observe band-gap renormalization in a p–n diode structure of carbon nanotubes [33]. It is thus possible to observe band-gap renormalization induced by Auger ionization by two-photon spectroscopy or photocurrent spectroscopy.

### 3.3. Oscillator strength

Finally, we discuss the dependence of the oscillator strength on the carrier density. Figure 4 shows the normalized oscillator strength at finite carrier densities at  $T = 300$  K. Here, the normalized oscillator strength is expressed as the ratio between the oscillator strength at finite density and at zero density. The normalized oscillator strength begins to decrease when the carrier density exceeds about  $1.0 \times 10^{-3} \text{ nm}^{-1}$ . This reduction in the oscillator strength is explained by the loss of excitonic character. Both screening and phase-space filling weaken the bond between the electron and the hole of an exciton. Consequently, the exciton Bohr radius increases, reducing the oscillator strength.

### 3.4. Discussion

Here, we propose an explanation for the decrease in PL emission reported in [10] based on the reduction of the oscillator strength discussed in the previous sections. For this purpose, we estimate the density of Auger-ionized carriers using the experimental data in [10]. From the experimental data, the nonlinear PL saturation behavior begins at a pump fluence of, for instance,  $30 \times 10^{16} \text{ photons cm}^{-2} \text{ pulse}^{-1} \text{ atom}$ , which gives an exciton number of 2 by using the absorption cross section  $\sigma_{ab} = 0.6 \times 10^{-17} \text{ cm}^2 \text{ atom}^{-1}$  that is derived in the same experiment. Since the decrease in PL starts at about  $100 \times 10^{16} \text{ photon cm}^{-2} \text{ pulse}^{-1} \text{ atom}$ , the number of excitons would become 6 at this pump fluence using the same absorption cross section if there were no Auger recombination process. However, when we consider the effect of the Auger process, the produced excitons are ionized at pump fluence where exciton–exciton scattering takes place. Therefore, assuming that all the excitons lost in the Auger process are ionized to form electron–hole carriers, we can roughly estimate the ionized carrier density to be of order of the  $10^{-3} \text{ nm}^{-1}$  for a  $1 \mu\text{m}$  long SWNT, which is derived by subtracting the exciton number at the pump fluence where the saturation starts from the one at the pump fluence where

the decline starts. This estimated density is in good agreement with our calculated result that the oscillator strength begins to decrease at a carrier density of  $1.0 \times 10^{-3} \text{ nm}^{-3}$ . Hence, it is reasonable to relate the experimentally observed reduction of the PL emission in [10] to the reduction in oscillator strength as shown in figure 4. We thus conclude that the observed nonlinear behavior of the PL emission originates from a reduction in the oscillator strength induced by the interaction between excitons and Auger ionized carriers.

#### 4. Summary

We have theoretically investigated many-body nonlinear effects induced by Auger ionization in the optical response of SWNTs by analyzing the Bethe–Salpeter equation for the optical susceptibility that includes screening of the Coulomb interaction and phase-filling effects. Due to efficient exciton ionization, excitons can dissociate into electron and hole carriers that interact with excitons under high-intensity laser irradiation. We found that Auger-ionized carriers cause band-gap renormalization and bleaching of excitons. Our calculations imply that the recently observed nonlinear behavior of PL spectra [10] can be attributed to the nonlinear many-body effects of Auger-ionized carriers.

Based on the present results, many-body exciton processes (e.g., the Fermi-edge singularity) are expected to occur in optically excited SWNTs with high densities of excitons and dissociated carriers. Indeed, it was experimentally shown recently that charged carrier–exciton bound states (i.e. trions) can be generated in undoped carbon nanotubes by controlling the photo-excitation intensity. The generation mechanism of trions was ascribed to Auger ionization [34]. Therefore, the study of ionized carriers interacting with excitons is expected to reveal further fascinating nonlinear optical phenomena in SWNTs.

#### Acknowledgments

This work was supported by CREST of the Japan Science and Technology Agency and a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan.

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