Prominent exciton absorption of perpendicularly polarized light in carbon nanotubes

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Abstract. Optical absorption in carbon nanotubes for polarization perpendicular to the tube axis is studied taking account of exciton effects. Although a strong depolarization effect tends to shift the position to the higher energy side and suppress the intensity, excitons manifest themselves as prominent peaks because of their large binding energy.

Keywords: carbon nanotube, exciton, depolarization effect, optical absorption, effective-mass approximation

PACS: 78.67.Ch, 73.22.Lp

A carbon nanotube has characteristic optical properties. For example, the absorption of light polarized perpendicular to the axis is known to be suppressed considerably because of strong depolarization effect in comparison with that of light polarized parallel to the axis [1, 2]. Further, exciton effects play a crucial role for light polarization parallel to the axis [3, 4, 5]. In this paper we present preliminary results of exciton effects for perpendicularly polarized light in semiconducting single-wall carbon nanotubes.

In the effective-mass approximation [6, 7] we shall use a screened Hartree-Fock approximation to calculate interaction effect on the band structure and introduce an attractive interaction between a photo-excited electron and a remaining hole using the Coulomb interaction screened by electrons in the system. Actual calculations can be performed by solving equation of motion for an electron-hole pair [3, 4, 5].

Dynamical conductivity characterizing light absorption is calculated in the linear response theory. The conductivity without depolarization effects is written as

$$\sigma_{xx}^{\prime}(\omega) = \frac{2\hbar e^2}{A L} \sum_{k,k'} \sum_{u,v} \frac{-2i\hbar\omega \langle u,l | v_{\Gamma}^i | g \rangle^2}{\epsilon_u - (\hbar\omega)^2 - 2i\hbar\omega \Gamma}$$

where \( \epsilon_u \) and \( |u,l\rangle \) are an eigen energy and eigen function of an electron-hole pair without depolarization effects, respectively, \( |g\rangle \) the ground state, \( v_{\Gamma}^i \) a Fourier transform of the velocity in the circumferential direction for the wave number \( 2\pi/L \) with \( L \) being the circumference length and \( l = \pm 1 \), and \( \Gamma \) a phenomenological energy broadening.

Depolarization effects are taken into account by considering a self-consistent electric field [1, 2]. Then, the conductivity is given by

$$\sigma_{xx}(\omega) = \frac{\sigma_{xx}^{\prime}(\omega)}{\epsilon_{xx}^{\prime}(\omega)}$$

with a dielectric function defined as

$$\epsilon_{xx}^{\prime}(\omega) = 1 + \frac{4\pi^2|l|}{\kappa L\omega} \sigma_{xx}^{\prime}(\omega),$$

where \( \kappa \) is an effective dielectric constant describing screening by electrons in \( \sigma \) bands, core states, and the \( \pi \) bands away from the \( K \) and \( K' \) points, and by surrounding materials if any. Excitation energy \( \hbar\omega_0 \) with depolarization effects is given by zero points of the dielectric function, that is,

$$\epsilon_{xx}^{\prime}(\omega_0) = 1 + \frac{4\pi^2i}{\kappa L\omega_0} \sigma_{xx}^{\prime}(\omega_0) = 0.$$ 

The strength of the Coulomb interaction in nanotubes is characterized by the dimensionless quantity given by the ratio of the typical Coulomb energy \( e^2/\kappa L \) and the typical kinetic energy \( 2\pi\gamma/L \) with \( \gamma \) being the band parameter, i.e.,

$$\frac{e^2}{\kappa L 2\pi\gamma} \approx \frac{0.35}{\kappa}.$$ 

Since \( \kappa \) is considered to be of the order of unity, for example, \( \kappa = 2.4 \) for graphite, the typical strength of the Coulomb interaction is of the order of 0.1~0.2.

The summation over states must be cut off by a cutoff energy \( \epsilon_c \) in the effective-mass approximation. It should be of the order of the half of the \( \pi \)-band width \( 3\gamma_0 \), where \( \gamma_0 \) is the resonance integral between nearest neighbor sites and related to the band parameter through \( \gamma = \sqrt{3}a\gamma_0/2 \) with \( a \) being the lattice constant. Therefore, \( \epsilon_c(2\pi\gamma/L)^{-1} \approx (\sqrt{3}/\pi)(L/a) = \sqrt{3}d/a \), with \( d \) being the diameter of the nanotube. In the following, \( \epsilon_c(2\pi\gamma/L)^{-1} = 10 \) is used which corresponds to a diameter of typical single-wall nanotubes \( \sim 1.4 \) nm.

Figures 1(a) and (b) show energy dependence of dynamical conductivity for typical Coulomb interaction \( (e^2/\kappa L) (2\pi\gamma/L)^{-1} = 0.05 \) and 0.2, respectively. The
dashed lines show conductivity without depolarization effect which is denoted by ‘Perturbation’ and solid lines that with depolarization effect which is denoted by ‘Self-Consistent’. The vertical arrows indicate band edges. When the depolarization effect is not considered, the largest peak appears below the lowest band edge. When the depolarization effect is taken into account, the peak is shifted toward the higher energy side and its intensity is reduced. For the stronger interaction \((e^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.2\) shown in Fig. 1(b) there is another small peak below the higher band edge and another peak appears below the lowest band edge, corresponding to an excited exciton state.

The results clearly show that exciton effects are important to optical absorption in single-wall carbon nanotubes. In the case of inter-band optical absorption, there are no peaks in the spectra because of depolarization effects [1, 2].

Recently, photoluminescence spectra of single-wall nanotubes were decomposed into those associated with absorption of parallel and perpendicular light [8, 9]. The obtained spectra for perpendicular polarization showed that a peak with intensity about an order-of-magnitude smaller than that for parallel polarization appears at a position closer to that of the second lowest peak for parallel polarization. This result seems to be consistent with our theoretical prediction of the exciton absorption and emission of perpendicularly polarized light.

**FIGURE 1.** Calculated \(\tilde{\sigma}_{xx}(\omega)\) and \(\sigma_{xx}(\omega)\) for (a) \((e^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.05\) and (b) \((e^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.2\). Solid and dashed lines denote \(\tilde{\sigma}_{xx}(\omega)\) and \(\sigma_{xx}(\omega)\), respectively. The vertical arrows indicate band edges. The energy broadening \(\Gamma(2\pi\gamma/L)^{-1} = 0.01\) is used.

**ACKNOWLEDGMENTS**

This work was supported in part by a 21st Century COE Program at Tokyo Tech “Nanometer-Scale Quantum Physics” and by Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

**REFERENCES**