Exciton absorption of perpendicularly polarized light in carbon nanotubes

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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. The resulting absorption energy is closer to that associated with that of the second gap for light polarized parallel to the axis.

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

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I. INTRODUCTION

A carbon nanotube is a quasi-one-dimensional (1D) system of a rolled-up two-dimensional (2D) graphite sheet. It has characteristic optical properties. For example, 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 known to be important in 1D systems\(^3\)\(^-\)\(^5\) play a crucial role for light polarization parallel to the axis.\(^6\)\(^-\)\(^8\)

The purpose of this paper is to study exciton effects for perpendicularly polarized light in semiconducting single-wall carbon nanotubes.

Various electronic properties of carbon nanotubes have been described successfully in the effective-mass approximation.\(^9\) In fact, the strong anisotropy of inter-band optical absorption due to a depolarization effect was first demonstrated using this scheme.\(^1\)\(^2\) This anisotropy was actually observed in experiments for well-aligned nanotubes.\(^10\)\(^-\)\(^12\)

Important roles of the electron-electron interaction including screening effects were revealed also in the effective-mass scheme.\(^6\)\(^-\)\(^8\)\(^13\)\(^-\)\(^14\) The band gap is substantially enhanced due to the interaction and at the same time an electron in a conduction band and a hole in a valence band form an exciton with a binding energy comparable to the band-gap enhancement due to electron-electron interaction. As a result the exciton energy lies moderately higher than the band gap at exciton levels. Theoretical studies based on first-principles calculations also reported significant interaction effects on optical excitation.\(^15\)\(^-\)\(^17\)

Tight-binding models can be used also for the description of various electronic properties including transport.\(^18\)\(^-\)\(^21\) In particular, effects of a static electric field perpendicular to a tube axis were studied and the static dielectric constant was estimated.\(^22\) Screening effects were investigated.\(^23\) Further, Important roles of electron-electron interaction on optical properties of nanotubes were reported.\(^13\)\(^-\)\(^24\)\(^-\)\(^25\)

Exciton effects on optical absorption spectra were reported in experiments in films of nanotube bundles.\(^26\)\(^-\)\(^27\) Photoluminescence measurements of individual single-wall tubes enabled quantitative comparison between experiments and theories.\(^28\)\(^-\)\(^29\)

Both measured excitation energies and ratio of the exciton peaks of the first and second gaps are well explained by calculations if interaction and exciton effects are properly considered. In these experiments absorption or emission of light parallel to the tube axis are measured usually because of the suppression for the perpendicular polarization due to the strong depolarization effect.

For perpendicular or circular polarizations, calculations of optical absorption spectra including exciton effects for some specific tubes demonstrated that a small exciton peak may appear and its peak position is shifted to the higher energy side.\(^16\)\(^-\)\(^25\) In this paper the exciton absorption for perpendicular light is systematically studied with emphasis on the depolarization effect and dependence on the strength of the Coulomb interaction and the tube diameter.

The paper is organized as follows: In Sec. 2, an effective-mass approximation developed earlier and the method to calculate the dynamical conductivity with exciton effects and depolarization effect are described briefly for the purpose of making this paper self-contained. Numerical results are shown in Sec. 3 and discussed in Sec. 4. Summary and conclusion are given in Sec. 5.

II. EFFECTIVE-MASS APPROXIMATION

A. Energy bands and wave functions

In a 2D graphite sheet schematically shown in Fig. I(a), the conduction and valence bands consisting of \(\pi\)-states cross at the K and \(K'\) points and electrons around these points are described by a \(\mathbf{k} \cdot \mathbf{p}\) equation. Near the K point it is given by

\[
\gamma (\sigma_x \hat{k}_x + \sigma_y \hat{k}_y) \mathbf{F}^K (\mathbf{r}) = \varepsilon \mathbf{F}^K (\mathbf{r}),
\]

where the \(x\) and \(y\) coordinates are chosen in the circumference and the axis directions, respectively, \(\gamma\) is a band
parameter, $\sigma_x$ and $\sigma_y$ are the Pauli spin matrices, and $\textbf{k} = (k_x, k_y)$ is a wave vector operator.

Electronic states near the K point for a nanotube with a sufficiently large diameter are obtained by imposing the boundary conditions around the circumference direction:

$$\textbf{F}^K(r + \textbf{L}) = \textbf{F}^K(r) \exp \left[ 2\pi i \left( \varphi - \frac{\nu}{3} \right) \right],$$

with $\varphi = \phi/\phi_0$, where $\phi$ is an Aharonov-Bohm magnetic flux passing through the cross section of the nanotube as shown in Fig. 1(b) and $\phi_0 = \hbar/e$ the magnetic flux quantum. Further, $\nu$ is an integer determined uniquely as $\nu = 0$ or $\pm 1$ from $n_a + n_b = 3M + \nu$ with integer $M$, where $\textbf{L} = n_a \textbf{a} + n_b \textbf{b}$ is the chiral vector in the circumference direction and $\textbf{a}$ and $\textbf{b}$ are the primitive translation vectors of a 2D graphite sheet shown in Fig. 1(a).

The energy bands are specified by $\alpha = (\pm, n)$ and the wave vector $k$ in the axis direction, where $n$ is an integer, ‘$-$’ denotes the valence band, and ‘$+$’ the conduction band. The wave function for a band associated with the K point is written as

$$\textbf{F}^K_{\alpha k}(x, y) = \frac{1}{\sqrt{AL}} \exp \left[ i\kappa_{\nu \varphi}(n)x + iky \right] \textbf{F}^\nu_{\alpha k},$$

with $A$ being the length of nanotube,

$$\kappa_{\nu \varphi}(n) = \frac{2\pi}{L} (n + \varphi - \frac{\nu}{3}),$$

and

$$\textbf{F}^\nu_{\alpha k} = \frac{1}{\sqrt{2}} \left( b_{\nu \varphi}(n, k) s_\alpha \right),$$

where

$$b_{\nu \varphi}(n, k) = \frac{\kappa_{\nu \varphi}(n) - ik}{\sqrt{\kappa_{\nu \varphi}(n)^2 + k^2}}$$

and

$$s_\alpha = \begin{cases} +1 & (\alpha = +, n), \\ -1 & (\alpha = -, n). \end{cases}$$

The corresponding energy is given by

$$\varepsilon_{\pm \nu}(k) = \pm \gamma \sqrt{\kappa_{\nu \varphi}(n)^2 + k^2}.$$

A $\textbf{k} \cdot \textbf{p}$ equation near the K’ point is given by Eq. (1) in which $\sigma_y$ is replaced by $-\sigma_y$. The wave function and energy are given by Eq. (3) with the complex conjugate of $\textbf{F}^\nu_{\alpha k}$ and Eq. (8), respectively, in which $\nu$ is replaced by $-\nu$.

### B. Electron-electron interaction

Long-range Coulomb potential at $r = (x, y)$ due to a charge $-e$ at the origin is

$$V(x, y) = \frac{\pi e^2}{\kappa L \sqrt{\sin^2(\pi x/L) + (\pi y/L)^2}}.$$

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. Then, a matrix element of electron-electron interaction between an initial state specified by single-particle states $(\beta, k)$ and $(\alpha', k + q)$ and a final state specified by $(\alpha, k + q)$ and $(\beta', k)$ in a band associated with the K point where $\alpha = (\pm, n)$, $\beta = (\pm, m)$, etc. becomes

$$\begin{align*}
\frac{\nu(\alpha, k + q; \beta, k')(\beta', k; \alpha', k + q)}{\sqrt{\varepsilon_{\alpha, k + q} \varepsilon_{\beta', k'}}}
\times & \left[ \textbf{F}^\nu_{\alpha k + q} | \textbf{F}^\nu_{\beta' k} \right] \left[ \textbf{F}^\nu_{\alpha' k + q} | \textbf{F}^\nu_{\beta k} \right] \\
& \times I[n-m](\frac{L|q|}{2\pi}) K[n-m](\frac{L|q|}{2\pi}),
\end{align*}$$

where $I[n](t)$ and $K[n](t)$ are the modified Bessel functions of the first and second kind of the order $|n|$, respectively. For electrons near the K’ point, a matrix element is given by a complex conjugate of Eq. (10) with replacement $\nu \rightarrow -\nu$.

The static polarization function for electrons near the K point calculated in the random phase approximation becomes

$$\Pi_{n-m}^{K}(q) = -\frac{2}{\pi} \sum_{\alpha', \beta'} \frac{1}{A} \left[ \sum_{k'} \delta_{n-m, n'-m'} |\textbf{F}^\nu_{\alpha' k'} \cdot \textbf{F}^\nu_{\beta' k'}|^2 \right]$$

$$\times g_0(\varepsilon_{\alpha}^{K}(k' + q))g_0(\varepsilon_{\beta'}^{K}(k'))$$

$$\times \frac{f_{\alpha, k'} - f_{\beta', k'}}{\varepsilon_{\alpha}^{K}(k' + q) - \varepsilon_{\beta'}^{K}(k')},$$

where $f_{\alpha, k}$ is the Fermi-Dirac distribution function for $\varepsilon_{\alpha}^{K}(k)$ and $g_0(\varepsilon)$ a cutoff function with a cutoff energy $\varepsilon_c$ defined as

$$g_0(\varepsilon) = \begin{cases} 1 & |\varepsilon| < \varepsilon_c \\ 0 & |\varepsilon| > \varepsilon_c. \end{cases}$$

with $\alpha_c$ being chosen such that $g_0$ decays sufficiently smoothly and the integral converges. The polarization
function near the K' point, $\Pi_{n_m}^\nu(q)$, is given by (11) with replacements $\nu \rightarrow -\nu$ and $\varepsilon^{K'}_\alpha(k) \rightarrow \varepsilon^{K'}_\alpha(k)$. Then, the dielectric function is given by

$$\varepsilon_{n_m}(q) = 1 + \frac{2e^2}{\kappa} I_{|m|} \left( \frac{L|q|}{2\pi} \right) K_{|m|} \left( \frac{L|q|}{2\pi} \right) \times \left[ \Pi_{n_m}^\nu(q) + \Pi_{n_m}^\nu(q) \right].$$

(13)

The screened Coulomb interaction is given by

$$V_{(a,k;\beta,k')\beta',k'+q}(\varepsilon_{n_m}^\nu) = \frac{\varepsilon_{n_m}^\nu}{\varepsilon_{n_m}(q)}.$$  

(14)

The total Hamiltonian consists of the kinetic term in Eq. (1) and the Coulomb interaction with matrix elements given by Eq. (14).

### C. Dynamical conductivity

The dynamical conductivity characterizing optical absorption is calculated in the linear response theory. 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 a static dielectric function. Various approximation schemes were compared and this approximation was shown to be sufficient for single-wall nanotubes. Actual calculations can be performed by solving equation of motion for an electron-hole pair.

In Green’s function formalism this can be achieved by considering the self-energy in the screened Hartree-Fock approximation and by summing up electron-hole ladder diagrams in a current-current correlation function.

An exciton near the K point with momentum $2\pi hl/L$ in the circumference direction is written as

$$|u, l\rangle = \sum_{n,k} \psi_n^l(k) c_{+n+l,k}^\dagger c_{-n,k}|g\rangle,$$

(15)

where $c_{\alpha,k}$ and $c_{\alpha,k}$ are the creation and annihilation operators for electrons, respectively, and $|g\rangle$ is the ground state. The equation of motion for an exciton in the above approximation is given by

$$\varepsilon_u \psi_{n}^l(k) = \left( \Delta \varepsilon^K_{n,k} + \Delta \Sigma_{n,k} \right) \psi_{n}^l(k) \nonumber \nonumber - \sum_{m,q} V_{(+n+l,k;+m+l,k+q)(-m,k+q;+n,k)}(k+q) \psi_{n}^m(k+q),$$

(16)

where

$$\Delta \varepsilon^K_{n,k} = \varepsilon^K_{n+k+1}(k) - \varepsilon^K_{n}(k),$$

(17)

$$\Delta \Sigma_{n,k} = \Sigma_{+n+l,k} - \Sigma_{-n,k},$$

(18)

with $\Sigma_{+n,k}$ being the self-energy. In the conventional screened Hartree-Fock approximation, it is given by

$$\Sigma_{+n,k} = -\sum_{m,q} V_{(+n,k;+m,k+q)(-m,k+q;+n,k)}.$$  

(19)

at zero temperature, where the summation is over the occupied valence bands. The particle-hole symmetry is broken in the self-energy of Eq. (19) although very weakly when the cutoff function is introduced. In order to avoid this weak insufficient feature, therefore, we should redefine the self-energy with the particle-hole symmetry as

$$\Sigma_{\pm,n,k} = \pm \frac{1}{2} (\Sigma_{+n,k} - \Sigma_{-n,k}).$$

(20)

The velocity operator in the circumference direction is independent of the K and K' points and is given by

$$v_{xz}^l = \frac{\gamma}{\hbar} \sigma_x e^{i\theta},$$

(21)

with $\theta = 2\pi x/L$. Its matrix element between the ground state and the exciton state is

$$\langle u, l | v_{xz}^l | g \rangle = \sum_{n,k} \langle v_{n,k}^l | \psi_n^l(k)^*,$$

(22)

where $v_{n,k}^l$ is a matrix element of $v_{xz}^l$ between a one-particle state with $K_u(p+n+l)$ and $k$ at the conduction band and that with $K_u(p)$ and $k$ at the valence band for the K point. For the K' point it is similarly defined for states with $K_u(p+n+l)$ and $k$ at the conduction band and with $K_u(p)$ and $k$ at the valence band. For the K point it becomes

$$v_{xz}^l(n,k) = \frac{\gamma}{\hbar} F_{n+k} \sigma_x F_{n-k},$$

(23)

and for the K' point it is given by complex conjugate of Eq. (23) with $\nu \rightarrow -\nu$.

In order to calculate the dynamical conductivity, we use the Kubo formula

$$\sigma_{xx}^l(\omega) = \frac{i}{\omega} \left[ K_{xx}^l(\omega) - K_{xx}^l(0) \right],$$

(24)

where $K_{xx}^l(\omega)$ is a current-current correlation function and explicitly written as

$$K_{xx}^l(\omega) = -\lim_{\delta \rightarrow 0} \frac{2e^2}{\hbar L} \sum_{K,\nu} \sum_{u} \varepsilon_u \varepsilon_{u+n} \left( \frac{\hbar \omega + i\delta}{\hbar \omega + i\delta} \right) \sum_{m,q} \left| \langle u, l | v_{xz}^l | g \rangle \right|^2.$$  

(25)

The presence of the terms with $+\omega$ and $-\omega$ corresponds to the fact that the external field is real and therefore contains both $e^{-i\omega t}$ and $e^{+i\omega t}$. Then, the dynamical conductivity is given by

$$\sigma_{xx}^l(\omega) = \frac{2he^2}{\hbar L} \sum_{K,\nu} \sum_{u} \varepsilon_u \varepsilon_{u+n} \left( \frac{\hbar \omega + i\delta}{\hbar \omega + i\delta} \right) \sum_{m,q} \left| \langle u, l | v_{xz}^l | g \rangle \right|^2.$$  

(26)

where a factor 2 in front of the sum comes from the spin degeneracy and a phenomenological energy broadening $\Gamma$ has been introduced.

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 $2\pi\gamma/L$, i.e.,

$$\frac{e^2}{\kappa L} \approx \frac{0.35}{\kappa}. \quad (27)$$

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 $0.1 \sim 0.2$.

The cutoff energy 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 $\gamma = \sqrt{3}\gamma_0/2$ with $a = 2.46$ Å being the lattice constant. Therefore, $\varepsilon_c(2\pi\gamma/L)^{-1} \approx (\sqrt{3}/\pi)(L/a) = \sqrt{3}d/2a$, with $d$ being the diameter of the nanotube. For example, $\varepsilon_c(2\pi\gamma/L)^{-1} = 10$ corresponds to a diameter $\sim 1.4$ nm.

D. Depolarization Effect

A spatially inhomogeneous current $j$ induces a charge distribution $\rho$. They are related through the continuity equation

$$\frac{\partial \rho}{\partial t} + \vec{\nabla} \cdot \vec{j} = 0. \quad (28)$$

For an electric field perpendicular to the tube axis, they are written as

$$\rho = \sum_l \rho_l e^{il\theta - i\omega t}, \quad (29)$$

Then, the charge density is related to the current as

$$\rho_l = \frac{2\pi l}{L\omega} j_l. \quad (30)$$

It gives polarization in the circumference direction

$$P_x = \sum_l P^l_x e^{il\theta - i\omega t}, \quad (32)$$

where

$$P^l_x = \frac{i\rho^l}{2\kappa|l|}. \quad (33)$$

In this case total and external electric fields in the circumference direction, $E_x$ and $D_x$, respectively, are written as

$$E_x = \sum_l E^l_x e^{il\theta - i\omega t}, \quad (34)$$

$$D_x = \sum_l D^l_x e^{il\theta - i\omega t}, \quad (35)$$

where $E^l_x$ and $D^l_x$ are a Fourier transform of total and external electric field, respectively, with momentum $2\pi hl/L$. Then, there is a relation

$$E^l_x = D^l_x - 4\pi P^l_x. \quad (36)$$

Thus, $E^l_x$ is given by

$$E^l_x = \frac{D^l_x}{\varepsilon^l_{xx}(\omega)}. \quad (37)$$
where Eqs. (31) and (33) and $j_x^l = \sigma_{xx}^l(\omega)E_x^l$ are used and $\varepsilon_{xx}(\omega)$ is a dielectric function

$$\varepsilon_{xx}(\omega) = 1 + \frac{4\pi^2|l|}{\kappa L\omega}\sigma_{xx}(\omega). \quad (38)$$

The relation between the current and the external field is written as

$$j_x^l = \tilde{\sigma}_{xx}(\omega)D_x^l, \quad (39)$$

where

$$\tilde{\sigma}_{xx}(\omega) = \frac{\sigma_{xx}(\omega)}{\varepsilon_{xx}(\omega)}. \quad (40)$$

For perpendicularly polarized light external electric field $D$ in the $xy$ plane is written as

$$D = (D \sin \theta, 0). \quad (41)$$

Then,

$$D_x^l = \frac{D}{2i} \tilde{b}_{x1} - \frac{D}{2i} \tilde{b}_{x-1}. \quad (42)$$

Therefore, the electric field has momentum $\pm 2\pi \hbar / L$ in the circumference direction. It excites excitons with the same momentum. The absorption is given by

$$P(\omega) = \frac{1}{2\pi} \int_0^{2\pi} d\theta \text{Re}[j_x^l E_x^l]$$

$$= \frac{1}{4} \text{Re}[\tilde{\sigma}_{xx}(\omega)] D^2, \quad (43)$$

where

$$\tilde{\sigma}_{xx}(\omega) = \frac{1}{2} [\sigma_{xx}^{l+1}(\omega) + \sigma_{xx}^{l-1}(\omega)]. \quad (44)$$

When the depolarization effect is neglected completely, the absorption is proportional to the real part of the dynamical conductivity $\sigma_{xx}(\omega)$ given by

$$\sigma_{xx}(\omega) = \frac{1}{2} [\sigma_{xx}^{l+1}(\omega) + \sigma_{xx}^{l-1}(\omega)]. \quad (45)$$

Consider first interband optical transitions neglecting exciton effects. The dynamical conductivity $\sigma_{xx}^{l+1}(\omega)$ exhibits a divergence at the band edges corresponding to the 1D joint density of states. This divergence cancels between the numerator and the denominator for $\tilde{\sigma}_{xx}(\omega)$ and therefore $\tilde{\sigma}_{xx}(\omega)$ does not have any peak structure at the band edges. Figure 2 shows an example of calculated $\sigma_{xx}(\omega)$ and $\tilde{\sigma}_{xx}(\omega)$ in a semiconducting nanotube with $\phi = 0$ for $(e^2/\kappa L)(2\pi \gamma / L)^{-1} = 0.1$. There is a clear van Hove singularity in $\sigma_{xx}(\omega)$, but it disappears completely in $\tilde{\sigma}_{xx}(\omega)$.

The situation changes when exciton effects are considered. Now $\sigma_{xx}(\omega)$ has bound states with a delta-function peak below the interband continuum because of multiple scattering of an excited electron in the conduction band and a remaining hole in the valence band. The real part of the dielectric function $\varepsilon_{xx}(\omega)$ is schematically shown in Fig. 3 near the lowest band edge. The dielectric function diverges at $\hbar \omega = \varepsilon_u$ and monotonically increases between the neighboring $\varepsilon_u$'s. The excitation energy is given by

FIG. 4: Calculated $\tilde{\sigma}_{xx}(\omega)$ and $\sigma_{xx}(\omega)$ for $(e^2/\kappa L)(2\pi \gamma / L)^{-1} = (a) 0.1$ and (b) 0.25. Solid and dashed lines denote $\tilde{\sigma}_{xx}(\omega)$ and $\sigma_{xx}(\omega)$. The vertical arrows indicate band edges. The cutoff energy is chosen as $\varepsilon_c(2\pi \gamma / L)^{-1} = 10$ and the energy broadening $\Gamma(2\pi \gamma / L)^{-1} = 0.01$ is used.
FIG. 5: Coulomb interaction dependence of the lowest excitation energy (solid lines), energy gap (dashed lines), and absorption peak energy without depolarization effect (dotted lines). Results for the cutoff energies $\varepsilon_c(2\pi\gamma/L)^{-1} = 2, 5, \text{and } 10$ are shown.

FIG. 6: Coulomb interaction dependence of the lowest excitation energy for perpendicular (solid lines) and parallel polarizations (dashed lines). For parallel polarization the energies associated with the first and second lowest subbands are plotted.

FIG. 7: Dependence of dimensionless oscillator strength on the Coulomb interaction. The solid lines represent the strength for the perpendicular polarization and the dashed lines for the first gap in the case of the parallel polarization.

A dimensionless oscillator strength of the lowest exciton absorption peak for parallel polarization has been defined as

$$f_u = 2m^* \frac{|\langle u | v_y | g \rangle|^2}{\varepsilon_u},$$

where $|u\rangle$ is a wave function of exciton with zero momentum given by solutions of Eq. (16) for $l = 0$, $v_y$ the velocity operator in the $y$ direction, and the effective mass of the first conduction band is defined by

$$m^* = \frac{2\pi \hbar^2}{3\gamma L}.$$
The same definition gives the following expression for the perpendicular polarization:

\[ f_u = \frac{2m^*}{4\pi^2\hbar^2} \frac{\kappa L^3\omega_0}{\omega^2} \left[ \frac{\partial^2 e_{xx}(\omega)}{\partial \omega^2} (\omega_0) \right]^{-1} \].

The dielectric function (38) with Eq. (26) shows that the static dielectric function \( e_{xx}(0) \) has a value larger than unity. This dielectric function corresponds to that considered previously in a tight-binding model \(^{22}\) and in an effective-mass approximation. \(^{14}\) The present calculation including interband transitions without exciton effects gives \( 4.2 < e_{xx}(0) < 5 \) in agreement with the previous results \(^{14,22}\) when the interaction strength is correspondingly chosen.

### III. NUMERICAL RESULTS

In the following, semiconducting carbon nanotubes with \( \nu = \pm 1 \) in the absence of magnetic flux \( \varphi = 0 \) are studied. Figures 4(a) and (b) show energy dependence of dynamical conductivity for typical Coulomb interaction \( (\epsilon^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.1 \) and 0.25, respectively, and for the cutoff energy \( \epsilon_\nu(2\pi\gamma/L)^{-1} = 10 \). 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 at the lowest energy of Eq. (16) 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. There are other small peaks below each of the higher band edges in Fig. 4(a) for \( (\epsilon^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.1 \). For the stronger interaction \( (\epsilon^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.25 \) shown in Fig. 4(b) another peak appears below the lowest band edge, corresponding to an excited exciton state.

In Fig. 5 the solid lines show the Coulomb interaction dependence of the lowest excitation energy, which corresponds to position of the largest peaks for solid lines in Fig. 4. The lowest band edge is plotted by dashed lines and the largest peak position for conductivity without depolarization effect is also plotted by dotted lines. A weak cutoff-energy dependence arises mainly from that of band gap. \(^{31}\)

Figure 6 shows the Coulomb-interaction dependence of the excitation energy for perpendicular (solid lines) and parallel (dashed lines) polarization. For parallel polarization the lowest energies associated with the first and second lowest subbands are plotted. In the absence of interaction, the lowest band edge for perpendicular polarization is located at the middle between the first and second lowest band edges for parallel polarization. With increase of the interaction the energy for perpendicular polarization becomes close to the second gap for the parallel polarization due to the depolarization effect. For \( (\epsilon^2/\kappa L)(2\pi\gamma/L)^{-1} \sim 0.25 \) these two energies are approximately the same.

Figure 7 shows the oscillator strength of the lowest exciton absorption peaks as a function of the Coulomb interaction. The oscillator strength for perpendicular light has a tendency to decrease with the increase of the Coulomb interaction in contrast to the opposite behavior for parallel polarization. The oscillator strength for the perpendicular polarization lies typically in the range from ten to thirty percent of that for the parallel polarization.

In Fig. 8 a typical example of the dependence on energy broadening \( \Gamma \) is shown. The peak becomes unclear with increase of the broadening and vanishes for the broadening comparable to the binding energy, indicating possibility of diminishing exciton effect in strongly inhomogeneous systems.

### IV. DISCUSSIONS

In a diagrammatic representation, the dynamical conductivity \( \sigma_{xx}^l(\omega) \) with \( l = \pm 1 \) is given by the bubble diagram (a) in Fig. 9. The ladder diagram represents the attractive interaction between an electron in the conduction band and a hole in the valence band. For an electron and a hole excited optically, they have the same spin and belong to the same valley (K or K’ point). In this case, however, the processes shown in (b) become allowed due to the Coulomb interaction and the summation of these diagrams leads to \( \sigma_{xx}^l(\omega) \). The poles of \( \sigma_{xx}^l(\omega) \) give the energies of excitons optically active.
FIG. 9: (a) The diagrammatic representation of the dynamical conductivity \( \sigma'_{xx}(\omega) \) with \( l = \pm 1 \). (b) A diagram contributing to the depolarization effect. For an electron and a hole with the same spin and valley the diagrams like (b) become allowed and lead to the depolarization effect. When they have different spins or valleys, the diagrams like (b) do not appear and therefore their energies are given by the poles of \( \sigma'_{xx}(\omega) \).

When an electron and a hole have different spins as in triplet excitons, on the other hand, the processes (b) are not possible. This shows that the depolarization effect is absent for these states and that the spin triplet excitons have energies determined by the poles of \( \sigma'_{xx}(\omega) \), i.e., \( \varepsilon_u \) obtained from Eq. (16). These triplet excitons are not excited optically, however. The same is applicable to the degree of freedom for the K and K’ points. An exciton consisting of an electron at the K point and a hole at the K’ point has the energy levels \( \varepsilon_u \) unaffected by the depolarization effect. The same is applicable to an exciton consisting of an electron at the K’ point and a hole at the K point. These excitons, independent of spin configurations, do not couple with perpendicularly polarized light, either.

For parallel polarization, the exciton states have a degeneracy of 16 in the lowest order \( \mathbf{k} \cdot \mathbf{p} \) approximation due to spins and valleys. When a weak short-range part of the Coulomb interaction is considered, they are split into several levels, leaving only a single optically-active bright level and making all others optically-inactive or dark.\(^8,17,25,36,37\) The same is expected to applicable to the present perpendicular polarization although the exciton states have been split into two by the depolarization effect. Due to short-range Coulomb interaction, two optically active excitons are expected to split further into optically active and inactive states. Optically inactive excitons also split into several levels due to exchange and inter-valley mixing. This problem is out of the scope of the present work and left for the future.

Because of the logarithmic divergence of the self-energy without the cutoff energy \( \varepsilon_c \), the tube-diameter dependence of the excitation energy is not completely scaled by the typical kinetic energy \( 2\pi\gamma/L \), but has an extra weak logarithmic dependence on \( \varepsilon_c (2\pi\gamma/L)^{-1} \) with \( \varepsilon_c \sim 3\gamma_0 \). Similarly to the case for parallel polarization,\(^7\) the excitation energy \( \hbar\omega_0 \) for perpendicular polarization is represented well by the form

\[
\frac{\hbar\omega_0}{\gamma_0} = \frac{a}{L} \left[ C_1 + C_2 \ln \left( \frac{L}{a} \right) \right],
\]

(51)

where the parameters \( C_1 \) and \( C_2 \) depend on the normalized Coulomb interaction \( (e^2/\kappa L)(2\pi\gamma/L)^{-1} \) independent of \( L \). The parameters \( C_1 \) and \( C_2 \) are \( \sqrt{3\pi} \) and zero, respectively, without the interaction and monotonically increase with the increase of \( (e^2/\kappa L)(2\pi\gamma/L)^{-1} \). For \( (e^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.1 \) and 0.2, for example, we have \( (C_1, C_2) \approx (6.3, 0.4) \) and \( (6.8, 0.5) \), respectively.

There is no dependence of excitation energies on the chirality and the type \( \nu = \pm 1 \) of semiconducting tubes within the present lowest order of \( \mathbf{k} \cdot \mathbf{p} \) scheme. When higher order terms are taken into account, a family effect, i.e., characteristic dependence on the chirality and \( \nu \), can appear also in the case of perpendicular polarization. Higher order terms can arise from trigonal warping,\(^38\) nonzero curvature,\(^39,40\) lattice distortion,\(^41,42\) and possible asymmetry between conduction and valence bands.\(^38,43\) All of these can shift band edges leading to change of excitation energy. In addition, a modulation of the band-edge effective mass possibly affects binding energy of excitons.\(^7\) In order to investigate such a family effect, however, it is necessary to determine all the parameters describing these effects. This is beyond the scope of the present work and remains as a future issue.

For circularly polarized light propagating along the tube axis, the electric field has either positive or negative momentum in the circumference direction. Then, the dynamical conductivity is given by \( \tilde{\sigma}'_{xx}(\omega) \) with \( l = 1 \) or \( -1 \) and the absorption spectrum is the same as that for perpendicular light. Similar absorption spectra of a small tube in this case was demonstrated in a first-principles calculation with depolarization effect being taken into account in a manner different from the present method.\(^16\)

Recently, photoluminescence spectra of single-wall nanotubes were decomposed into those associated with absorption of parallel and perpendicular light.\(^45,46\) 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.
V. SUMMARY AND CONCLUSION

The optical absorption due to excitons for perpendicularly polarized light has been studied in carbon nanotubes within the effective mass approximation. It has been revealed that prominent exciton absorption peaks appear in spectra in spite of the existence of a strong depolarization effect. The depolarization effect shifts the excitation energy to higher energy side and suppresses intensity of the absorption peaks. As a result excitation energy becomes closer to that associated with that of the second gap for parallel polarization. The intensity of the absorption peaks is weakly dependent on the Coulomb interaction and the tube diameter through the cutoff energy and lies between ten and thirty percent of the peak intensity for parallel polarized light.

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