Aharonov-Bohm effects on bright and dark excitons in carbon nanotubes

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Abstract. A short-range part of the Coulomb interaction causes splitting and shift of excitons due to exchange interaction and mixing between different valleys in semiconducting carbon nanotubes. In the absence of a magnetic flux only a single exciton is optically active (bright) and all others are inactive (dark). Two bright excitons appear in the presence of an Aharonov-Bohm magnetic flux.

1. Introduction
A single-wall carbon nanotube consists of a rolled two-dimensional (2D) graphite and its electronic states change critically from metallic to semiconducting depending on its circumferential vector. Because of the one-dimensional (1D) structure with cylindrical form, the Coulomb interaction plays an important role in the band structure and the exciton effect is extremely important in optical spectra [1, 2, 3]. The purpose of this paper is to study effects of exchange interaction and mixing between different valleys on the exciton and to determine the energy levels of dark and bright excitons.

Optical absorption spectra reflect the 1D electronic structure directly [4, 5], though excitonic effects are of vital importance [6, 7]. Optical absorption and photoluminescence of individual nanotubes were observed [8, 9]. Splitting of the absorption and emission peaks due to an Aharonov-Bohm (AB) effect associated with magnetic flux passing through the cross section was observed also [10]. Because of the presence of two valleys, known as K and K’ points, and the electron spin, the exciton states have a degeneracy of 16 in the lowest order \( \mathbf{k} \cdot \mathbf{p} \) approximation. If we take into account a weak short-range part of the Coulomb interaction, they are split into several levels, leaving only a single bright level and making all others dark. Their ordering and relative energy splitting are shown to be sensitive to details of the short-range interaction. The presence of AB flux accessible experimentally modifies these energy levels strongly and causes the appearance of two bright excitons.

2. Effective-mass description
A graphite sheet is a zero-gap semiconductor in the sense that the conduction and valence bands consisting of \( \pi \) states cross at K and K’ points of the Brillouin zone [11]. Electronic states near the K point are described by the \( \mathbf{k} \cdot \mathbf{p} \) equation \( \gamma(\hat{\mathbf{\sigma}} \cdot \hat{\mathbf{k}})\mathbf{F}(\mathbf{r}) = \varepsilon \mathbf{F}(\mathbf{r}) \), where \( \gamma \) is the band parameter, \( \hat{\mathbf{k}} = (\hat{k}_x, \hat{k}_y) = -i\nabla \), \( \varepsilon \) is the energy, and \( \sigma_x \) and \( \sigma_y \) are the Pauli spin matrices.
Figure 1. Energy bands of a semiconducting CN with \( \nu = 1 \) and shifts of the band edges as a function of AB flux.

Two components of the wave function \( \mathbf{F}(\mathbf{r}) \) correspond to the amplitude at two sites denoted by A and B in a unit cell. This equation has the form of Weyl’s equation for neutrinos.

The structure of a nanotube is specified by a chiral vector \( \mathbf{L} \) corresponding to the circumference. The boundary conditions are given by \( \mathbf{F}(\mathbf{r} + \mathbf{L}) = \mathbf{F}(\mathbf{r}) \exp[2\pi i(\varphi - \nu/3)] \), with \( \nu = 0, \pm 1 \) depending on the structure and \( \varphi = \phi/\phi_0 \), where \( \phi \) is AB flux passing through the cross section and \( \phi_0 = ch/e \) is the flux quantum. As a result the wave vector in the circumference direction is quantized into \( \kappa \nu \varphi(n) = \left( 2\pi/L \right) \left( n + \varphi - \nu/3 \right) \) with integer \( n \) and \( L = |\mathbf{L}| \). Corresponding results for the K’ point can be obtained by replacing \( \hat{k}_y \) by \( -\hat{k}_y \) and \( \nu \) by \( -\nu \).

Figure 1 shows a schematic illustration of the energy bands of a semiconducting CN with \( \nu = 1 \) and shifts of the band edges in the presence of AB flux.

### 3. Exciton mixing and splitting

Exciton energy levels and corresponding absorption spectra were calculated previously in the effective-mass scheme with the conventional Coulomb interaction being taken into account [1, 2]. The results show that the exciton is dominant and interband continuum is negligible in optical properties. The exciton binding energy is characterized by the dimensionless parameter \( (e^2/\kappa L)(2\pi \gamma/L)^{-1} \), where \( \kappa \) is a static dielectric constant describing effects of polarization of electrons of core states, \( \sigma \) bands, \( \pi \) bands away from the K and K’ points, and a surrounding material. This parameter is independent of \( L \) and satisfies \( (e^2/\kappa L)(2\pi \gamma/L)^{-1} < 0.3 \), but its exact value is not known.

Because of the electron spin and the presence of K and K’ points, there are 16 exciton states. They can be written for example as \( |KK’⟩ \), where an electron is in the conduction band at the K point and a hole in the valence band at the K’ point. They are first classified into singlet and triplet depending on the total spin. Among singlet excitons, \( |KK’⟩ \) and \( |K’K⟩ \) remain uncoupled with others and \( |KK⟩ \) and \( |K’K’⟩ \) are coupled into bonding and anti-bonding states in the absence of AB flux. Because the matrix element for the optical transition is same between K and K’ point, only the bonding state is optically active and anti-bonding state becomes inactive. The same is applicable to optically-inactive triplet excitons except that each state is three-fold degenerate corresponding to the spin direction.

These excitons split when a weak short-range part of the Coulomb interaction is considered. The short-range part is characterized by two parameters \( w_1 \) and \( w_2 \) which are comparable to the hopping integral \( \gamma_0 = \gamma(\sqrt{3}a/2)^{-1} \) of nearest-neighbor \( \pi \) orbitals in 2D graphite [14]. The relative ordering of bright and dark excitons changes as a function of the ratio \( w_1/w_2 \), which is quite sensitive to the detailed form of the Coulomb interaction and the wave function of \( \pi \).
Figure 2. Calculated energy levels of the singlet and triplet excitons as a function of the flux for the parameter $w_2/\gamma_0 = 1.5$ in semiconducting nanotubes with $\nu = 1$ and circumference $L/a = 10$. $(e^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.2$. (a) $w_1/w_2 = 0.4$ and (b) 0.2.

The amount of the splitting and shift is of the order of $(a/L)^2 w_2$, where $a$ is the lattice constant of 2D graphite, and therefore becomes rapidly smaller with $L$.

Figure 2 shows some examples of the energy levels of the singlet and triplet excitons as a function of the flux for the parameter $w_2/\gamma_0 = 1.5$ in nanotubes with circumference $L/a = 10$. For $w_1/w_2 = 0.4$ the highest-energy exciton is bright, while for $w_1/w_2 = 0.2$ the bright exciton has a lower energy than anti-bonding states of singlet KK and K’K’ excitons. Mixing between the K and K’ points diminishes rapidly with the increase of the flux and excitons for the K and K’ points become essentially independent of each other for a sufficiently large AB flux. Figure 3 shows some examples of the dynamical conductivity describing the absorption spectra. With the increase of the flux, clear double peaks appear corresponding to the KK and K’K’ exciton, split by AB flux.

The Aharonov-Bohm splitting of the excitons was observed recently [10, 15, 16, 17]. An important characteristic feature of the experimental results is that the splitting does not seem to become observable until the magnetic flux reaches a certain critical value and then starts to increase with the flux. This is consistent with the above result that two peaks become observable only when the Aharonov-Bohm splitting exceeds the mixing between the KK and K’K’ exciton due to the short-range Coulomb interaction. A detailed and careful experimental study of the Aharonov-Bohm effect on the exciton may be used to determine the relative ordering of bright and dark excitons.

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Figure 3. Calculated dynamical conductivity describing the absorption spectra. The solid lines represent the conductivity and the dotted lines contributions of two singlet KK-K’K’ excitons. The flux $\phi$ is varied with interval $\Delta \phi/\phi_0 = 0.001$ up to $\phi_{\text{max}}/\phi_0 = 0.02$. Curves of different $\phi$ are shifted in the vertical direction. The peak positions in the presence and absence of the short-range interaction are shown also. A phenomenological broadening $\Gamma(2\pi\gamma/L)^{-1} = 0.01$ is introduced. $L/a=10$. $(e^2/\kappa L)(2\pi\gamma/L)^{-1} = 0.2$. (a) $w_1/w_2 = 0.4$ and (b) 0.2.

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