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Effect of periodic potential on exciton states in semiconductor carbon nanotubes

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ABSTRACT

We develop a theoretical background to treat exciton states in semiconductor single-walled carbon nanotubes (SWCNTs) in the presence of a periodic potential induced by a surface acoustic wave (SAW) propagating along SWCNT. The formalism accounts for the electronic band splitting into the Floquet subbands induced by the Bragg scattering on the SAW potential. Optical transitions between the Floquet states and correlated electron–hole pairs (excitons) are numerically examined. Formation of new van Hove singularities within the edges of Floquet sub-bands and associated transfer of the exciton oscillator strengths resulting in the photoluminescence quenching are predicted. The simulations demonstrate the exciton energy red Stark shift and reduction in the exciton binding energy. Comparison of our results with reported theoretical and experimental studies is provided.

1. Introduction

Single-walled carbon nanotubes (SWCNTs) are perfect quasi-1D structures demonstrating reach variety of transport and optical properties. [1,2] They are promising candidates for building blocks in nanoelectronics, quantum information processing, opto-electronics, and light emitting devices. In contrast to semiconductor low-dimensional materials, SWCNTs electronic structure is described by the Dirac equation for right and left propagating quasi-spinors associated with the chiral Bloch states. The quantum interference and tunneling of these states can result in new effects that are not feasible in semiconductor nanostructures. [3] Coulomb interactions in SWCNTs are substatial making many-body electronic effects important in understanding their photophysical properties [4].

Common approach to control the transport and optical properties of semiconductor nanostructures, photonic materials, and SWCNTs is based on modifications of their chemical composition and geometry. However, in many situations (e.g., tunable sensors and tunable light emitting devices) it is desirable to control optical and transport properties dynamically by applying external electric and magnetic fields. A reliable way to produce and control the external potential is to excite MHz through THz frequency surface acoustic waves (SAW) in a piezoelectric substrate holding a nanostructure. Polarization of the substrate surface produces SAW-modulated electric field that couples to the materials electron gas modulating its wavefunction and as a result affecting charge carriers transport and emission properties, and in particular giving rise to the effect of sonoluminescence [5].

Optical properties of various semiconductor nanostructures such as quantum dots, quantum wires and quantum wells influenced by SAW potential have been extensively examined [6–11]. In light of developing quantum photon sources, an ability to control single-photon emission by applying SAW has been demonstrated in nanowires [12]. In arrays of semiconductor quantum dots and rods, SAW results in the modulation of the emission line and leads to the exciton charging that can be detected via sonoluminescence measurements [13,10,11]. On the other hand, use of SAW has been reported to control properties of photonic materials [14,15]. In semiconductors, SAW-induced fields form dynamically modulated band gap structures. Specifically in quantum wires and quantum wells, this causes phototoluminescence quenching via exciton dissociation into free electron and hole sates and subsequent carriers transport to the emission centers resulting in spatial modulation of emission properties which is the essence of the sonoluminescence effect [7–9]. Description of the quenching by a conventional Stark effect predicts an essential exciton emission line red shift accompanying the reduction in exciton oscillator strength. However such shift has not been experimentally observed in core–shell nanowires [12].

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The Dirac electronic structure of *metallic* SWCNTs can also be influenced by an applied periodic potential. Specifically, such a potential with period much larger than SWCNT diameter causes formation of a Bragg grating that mixes the right and the left propagating Bloch states. This gives rise to the splitting of the valence and conduction band into Floquet sub-bands separated by dynamical small gaps [16,17]. In light of developing new electronic devices including those for quantum information processing and metrology a problem of quantized transport [18] can be addressed using SAW modulation of metallic SWCNTs. It has been demonstrated that the electric potential applied to contacts or impurity states in SWCNT can produce local potential walls forming the so-called quantum dots in which the charges get localized. Application of SAW potential results in the Coulomb-blockade-assisted quantized charge pumping through the quantum dots [19–21].

Recently, SAW-induced photoluminescence quenching in a collection of *semiconductor* SWCNTs has been examined experimentally [22]. From theory point of view the effect of *static* electric field on exciton properties in semiconductor SWCNTs has been considered [23]. However, such approach meets the same difficulties as in the case of nanowires, namely, the absence of a discernible red shift of exciton states. This calls for the development of a self-constant theoretical approach treating the effect of periodic potential on non-interacting electron and hole states in semiconductor SWCNTs and subsequently on the optical properties of the correlated electron–hole pairs (i.e., the excitons). In Section 2, we propose such a theoretical model and apply it in Section 3 to model SAW effect on the lowest exciton states in semiconductor SWCNTs. Conclusions are drawn in Section 4.

2. Model for exciton states modulated by periodic potential

In our calculations we apply band folding approach to a semiconductor SWCNT oriented along the y-axis. Periodical boundary conditions are applied around the circumference direction defined by the x-axis of the lab coordinate system with the period of L_x. The SAW is assumed to be launched by a transducer on the piezoelectric surface along the tube direction as described in Ref. [12]. In the frame of reference moving along the tube direction with the SAW phase velocity, the piezoelectric potential can be expressed as

$$U(y) = U_0 \cos(G_0 y), \tag{1}$$

where the SAW wave vector is $G_0 = 2\pi/L_y$, and L_y is the SAW wave length. The length of SWCNT is set to be multiple of L_y. The effect of SAW transverse component is neglected.

In our calculations of bound exciton states, we follow the formalisms originally proposed by Ando [24,25]. This formalism requires: electronic band structure, screened electron–electron Coulomb interactions, and electron–hole correlations. Those ingredients are combined into a Bethe–Salpeter (BS) equation in the reciprocal space governing bound exciton states well-separated form uncorrelated electron continuum in the semiconductor SWCNTs. Below, we incorporate the effect of the SAW potential (Eq. (1)) into the BS equation. This provides us with the eigenfunctions and eigenenergies of the quasiparticle states that are subsequently used to evaluate basic optical properties using dynamical conductivity function.

2.1. Non-interacting electron states and Floquet sub-bands.

In the vicinity of K and K' points of the graphene Brillouin zone, SWCNT non-interacting electron weave function is sought in the form

$$\mathbf{\Psi}(x,y) = \mathbf{\Phi}(y)\frac{1}{\sqrt{L_x}}\mathrm{e}^{-ik_x(n,v)x}, \tag{2}$$

where

$$k_x(n,v) = \frac{2\pi}{L_x}\left(n - \frac{v}{3}\right), \tag{3}$$

is quantized circumference wave vector depending on integer angular momentum quantum number n and the valley index $v = \pm 1$ associated with the Brillouin zone K (K') point.[1] The envelope function, $\mathbf{\Phi}(y)$, is a two-component spinor satisfying the 1D Dirac equation, gauge transformed to the following form [17,3]

$$\begin{pmatrix} -i\hbar \mathrm{v}_F\partial_y - E & -i\hbar \mathrm{v}_F k_x(n,v)\mathrm{e}^{i\phi(y)} \\ i\hbar \mathrm{v}_F k_x(n,v)\mathrm{e}^{-i\phi(y)} & i\hbar \mathrm{v}_F\partial_y - E \end{pmatrix}\mathbf{\Phi}(y) = 0, \tag{4}$$

where $\hbar \mathrm{v}_F = 6.46$ eVÅ with v_F being the Fermi velocity. In Eq. (4), the SAW periodic potential enters as the phase modulation, $e^{i\phi(y)}$, of the kinetic term. The phase

$$\phi(y) = \frac{2}{\hbar \mathrm{v}_F}\int_0^y d\xi U(\xi) = u_0\int_0^y d\xi \cos\xi, \tag{5}$$

linearly depends on the dimensionless SAW potential amplitude

$$u_0 = \frac{2U_0}{\hbar \mathrm{v}_F G_0}. \tag{6}$$

To account for this phase effect, we expend the exponent in terms of the first kind Bessel functions,

$$e^{i\phi(y)} = \sum_{l=-\infty}^{\infty} J_l(u_o)e^{ilG_0 y},$$

and further seek the envelope function in the form of Floquet series [26,27]

$$\mathbf{\Phi}_k^v(y) = \frac{1}{\sqrt{L_y}}\sum\nolimits_{l=-\infty}^{\infty}\begin{pmatrix} A_{k-lG_0} \\ B_{k-lG_0}\end{pmatrix}e^{-i(k-lG_0)y}. \tag{7}$$

This results in the following set of eigenvalue equations for the expansion coefficients

$$(-\hbar \mathrm{v}_F(k-lG_0) - E)A_{k-lG_0} - i\hbar \mathrm{v}_F k_x(n,v)\sum_{l'=-\infty}^{\infty} J_{l'}(u_0)B_{k-(l-l')G_0} = 0, \tag{8}$$

$$i\hbar \mathrm{v}_F k_x(n,v)\sum_{l'=-\infty}^{\infty} J_{l'}(u_0)A_{k-(l+l')G_0} + (\hbar \mathrm{v}_F(k-lG_0) - E)B_{k-lG_0} = 0. \tag{9}$$

Eqs. (8) and (9) can be solved numerically, resulting in quantized eigenenergies, $E^v_{s,n,k}$, and eigenvector components

$$\boldsymbol{F}^v_{s,n,k-lG_0} = \begin{pmatrix} A^v_{s,n,k-lG_0} \\ B^v_{s,n,k-lG_0}\end{pmatrix}, \tag{10}$$

satisfying the orthonormality relationship $\sum_{l=-\infty}^{\infty}(\boldsymbol{F}^{v*}_{s,n,k-lG_0}\cdot\boldsymbol{F}^v_{s',n,k-lG_0}) = \delta_{s,s'}$. Here, we introduce a new integer quantum number s accounting for the Bragg scattering at the ends of SAW-induced Brillouin sub-zone, $0 \leqslant k \leqslant lG_0$. This quantization results in the splitting of the valence and conduction bands into the so-called Floquet sub-bands. Specifically, s acquiring even (odd) values numerates the valence (conduction) Floquet sub-bands. Finally, spatial distribution of the single particle wavefunction (Eq. (2)) can be written as

$$\mathbf{\Psi}^v_{s,n,k-lG_0}(x,y) = \frac{1}{\sqrt{L_xL_y}}\boldsymbol{F}_{\alpha,k_x,k-lG_0}e^{-i\frac{2\pi}{L_x}(n-\frac{v}{3})x - i(k-lG_0)y}. \tag{11}$$

[1] The case when $v = 0$ corresponds to metallic SWCNT and is not considered below.