

Interaction of laser light and electrons with nanotubes

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Abstract

Designer electronic states in confined geometries are attracting considerable attention. In this work, a novel set of electronic states around nanotubes with long lifetime and exotic properties is discussed.

Keywords: Carbon nanotube, image states, ultrafast rotation, Rydberg-like states, long lifetimes

1 Introduction

Technological leaps of the last two decades have opened windows of opportunity for designing and controlling electronic states in novel systems. The fabrication of quantum dots in semiconductor devices has led to the invention of single-electron transistors [1] and controllable single photon emitters [2]. Such designer atoms are ideal settings for the control and manipulation of electronic states. Carbon nanotubes (CNT), first synthesized in 1991 by Iijima [3] as graphitic carbon needles, have remarkable electrical and mechanical properties. Carbon nanotubes are ideal for investigation at the interface of atomic and nanoscopic physics. Our aim has been to bring atomic physics techniques to the study of interaction of light and particles with nanotubes. Below, we will discuss a number of applications of atomic physics in the nanoscopic scale.

2 A brief primer on nanotubes

A carbon nanotube (CNT) is constructed by rolling a graphene sheet, defined by its primitive lattice vectors, \mathbf{a}_1 and \mathbf{a}_2 , along a vector \mathbf{C}_h , the so-called chiral vector, around an axial or translation vector \mathbf{T} (see Fig. 1). The chiral vector,

$$\mathbf{C}_h = n\mathbf{a}_1 + m\mathbf{a}_2 \equiv (n, m),$$

and translation vector,

$$\mathbf{T} = \left[\frac{(2m+n)}{d_R} \mathbf{a}_1 - \frac{(2n+m)}{d_R} \mathbf{a}_2 \right],$$

are determined by two lattice indices, n and m . Here,

$$d_R = \begin{cases} 3d & \text{if } n - m = 3qd \\ d & \text{if } n - m \neq 3qd \end{cases}$$

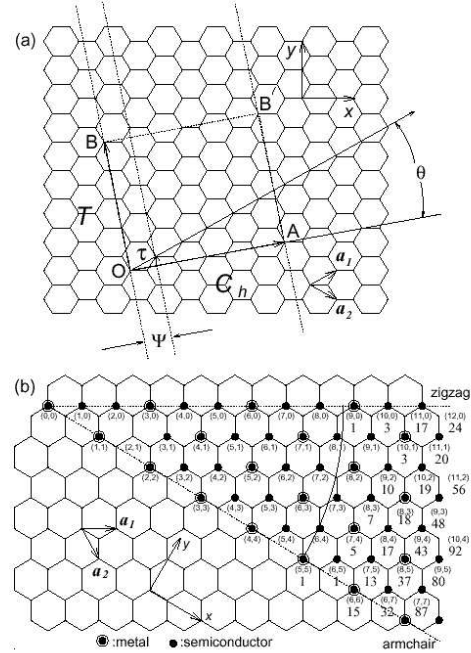


Figure 1: A graphene honeycomb lattice showing the primitive vectors and the chiral and translation vectors. From Ref. 4.

where q is an integer and d_R is the greatest common divisor of n and m .

The diameter of an (n, m) CNT is $d = C_h/\pi$, where $C_h = \sqrt{3}a_{c-c}[m^2 + n^2 - mn]^{1/2}$ is the magnitude of the chiral vector, and $a_{c-c} = 1.42\text{\AA}$ is the carbon-carbon bond length. The chirality of a nanotube is characterized by the chiral angle,

$$\theta = \tan^{-1} \sqrt{3}m/(m + 2n),$$

which is the angle between the vectors \mathbf{C}_h and \mathbf{a}_1 .

Nanotubes having either (but not both) $m = 0$ or $n = 0$ have $\theta = 0^\circ$ and are called zigzag nanotubes due to the zigzag nature of the bonds at the tube ends. Likewise, tubes with $n = m$ have $\theta = 30^\circ$ and are called armchair tubes[4], [5]. All other tubes with $n \neq m$ are called chiral nanotubes. One of the most remarkable properties of carbon nanotubes is that depending on their chirality, conducting, semiconducting or insulating nanotubes are possible[6].

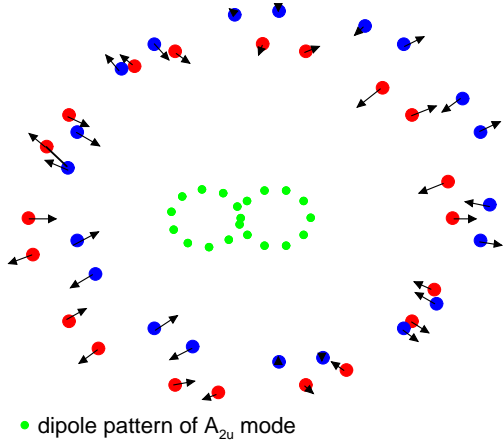


Figure 2: Ir-active A_{2u} phonon mode in (10,10) carbon nanotube at $\omega_{A_{2u}} = 870 \text{ cm}^{-1}$.

3 Photoabsorption in carbon nanotubes: mechanical rotation

In one such application, we calculated the absorption of circularly-polarized infrared photons by the optically-active resonant phonon modes in CNT[7]. The vibrational modes of carbon nanotubes have been investigated by several workers and the phonon dispersion energy curves have been calculated for a large number of CNTs using the zone-folding method [5], [8]. Of particular interest are two optically-active infrared (IR) phonon modes at $\omega_{A_{2u}} = 870 \text{ cm}^{-1}$ and $\omega_{E_{1u}} = 1580 \text{ cm}^{-1}$. Both of the IR-active modes absorb photons and carry angular momentum.

Fig. 2, gives the dipolar pattern of the carbon atom displacements [9] in the doubly-degenerate A_{2u} phonon mode for a (10,10) CNT. We found that through the transfer of photon angular momentum, at a rate of $\dot{L}_{A_{2u}} = \hbar \dot{n}_{A_{2u}}^+ \approx 2.5 \times 10^{-29} \text{ Nm}$, to the phonon modes and their eventual decay into two acoustical phonons- the observed decay lifetime of the A_{2u} mode is roughly 2.2 ps- CNTs would rotate with a frequency of $\omega_{rot} \approx 28 \text{ GHz}$. The steady-state angular momentum of $L_{A_{2u}} = \hbar \Delta n_{A_{2u}}^+ = \dot{L}_{A_{2u}} \tau_{A_{2u}} \approx 5.2 \times 10^{-41} \text{ Js}$. This angular momentum is transferred to the tube following the two-acoustical phonon decay of the A_{2u} mode near the $k = 0$ branch.

This proof-of-concept study demonstrated that nanotubes with their particular mechanical and electrical properties could form parts of nanoscopic motors, centrifuges or stabilizers.

4 Rydberg-like electronic states near nanotubes

Rydberon states are ubiquitous in atomic and molecular physics [10]. Collisions between ultracold neutral and Rydberg atoms in magneto-optical traps have been predicted to form exotic classes of Rydberg molecules

dubbed “trilobite” and “butterfly” molecules [11].

Rydberg electronic states have also been observed above conducting surfaces. This is possible due to the attractive interaction of an electron with its image charge. The resulting interaction potential for an electron at a distance z above a flat surface having a dielectric constant ϵ , is: $V(z) = -\frac{e^2}{4z} \left(\frac{\epsilon-1}{\epsilon+1} \right)$. Because of the Coulomb interaction, this potential supports an infinite number of “image states” that form the familiar Rydberg-series, $E_n = -\frac{13.60}{16n^2} \left(\frac{\epsilon-1}{\epsilon+1} \right)^2 \text{ eV}$, where n is the principal quantum number. Femtosecond two-photon pump-probe spectroscopy of electronic image states near Cu(100) surfaces has revealed quantum beats due to the interference of different eigencomponents in the resulting electronic wavepackets above the surface [12]. The collapse of these states into the surface is the primary reason for their short lifetimes.

In a recent work, we showed that conducting CNTs do indeed have Rydberg-like excitations[13]. However, because an electron can have angular momentum about the nanotube axis, a centrifugal barrier forms, dramatically increasing the lifetimes of the states compared to their counterparts above flat surfaces, see Fig. 3. We take the charge (the electron) to be located outside the tube at the position $(\rho_0, 0, 0)$ and $(I_m(x), K_m(x))$ are the regular and irregular modified Bessel functions. The electrostatic force between the electron and the conducting cylinder is calculated by differentiating the induced scalar potential Φ_{ind} with respect to ρ :

$$\begin{aligned} F(\rho_0) &= -q \partial \Phi_{ind} / \partial \rho |_{(\rho_0, 0, 0)} \\ &= \frac{2q^2}{\pi a^2} \int_0^\infty dx \left[A_0(x) + 2 \sum_{m=1}^\infty A_m(x) \right], \\ A_m(x) &= \frac{I_m(x)}{K_m(x)} K_m(x \rho_0/a) x K'_m(x \rho_0/a). \end{aligned} \quad (1)$$

The potential energy $V(\rho_0) = -\int^{\rho_0} F(\rho) d\rho$ can be calculated numerically from these expressions. Alternatively, physical intuition can be gained by an asymptotic analysis of the potential. The result,

$$V(\rho_0) \sim \frac{q^2}{a} \text{li} \left(\frac{a}{\rho_0} \right) \approx -\frac{q^2}{a} \frac{1}{(\rho_0/a) \ln(\rho_0/a)}, \quad (2)$$

is dominated by the $m = 0$ term in Eq. 1 and is given in terms of the logarithmic integral $\text{li}(x) \equiv \int_0^x dt / \ln(t)$.

The total effective potential that an electron would “see” in front of a conducting nanotube is

$$V_{eff}(\rho) = V(\rho) + \frac{(l^2 - \frac{1}{4})}{2m\rho^2}, \quad (3)$$

where m is the mass of the electron. This potential for different values of the angular momentum, is shown in

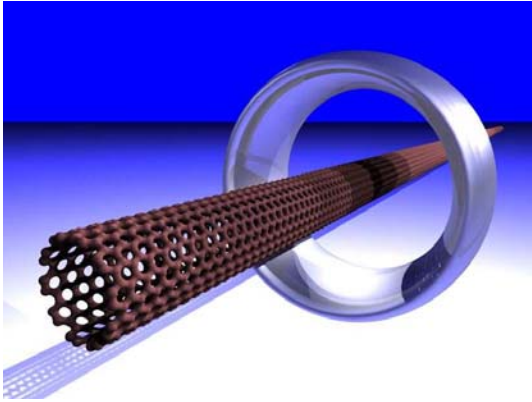


Figure 3: Visualization of an electron in a tubular image state around a (10,10) metallic carbon nanotube. From Ref. 13

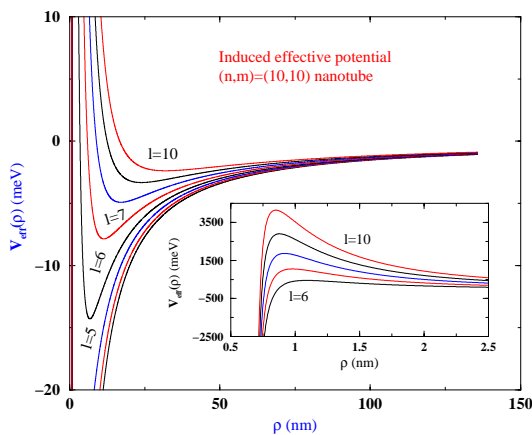


Figure 4: The effective potential between an electron and a conducting nanotube as a function of the angular momentum, l . From Ref. 13.

Fig. (4) for a (10,10) carbon nanotube of radius $a = 0.68$ nm. For moderate angular momenta ($l \geq 6$), the effective potential possesses extremely long-range wells that support bound states. The inset of Fig. 4 shows that high (1 – 2 eV) potential barriers separate these wells from the tube surface. Indeed, for angular momentum values of $l < 6$, no long-range wells exist so that low angular momentum states will exist close to the surface and have short lifetimes. However, because the radial overlaps between the high angular momentum and lower angular momentum states are small (they are localized at different radial distances) the decay of the high angular momentum states to ones with lower angular momentum through spontaneous radiative decay will be greatly suppressed.

The lifetimes due to tunneling increase exponentially with l and are substantially longer than those of image states above planar surfaces. Interaction with holes in the nanotube and the coupling of these states to the phonon modes in the tube can reduce their lifetimes.

These decay mechanisms can be suppressed by operating at temperatures ($T < 10$ K) lower than the transition energy between different image states. Spontaneous radiative transitions between different states also limit their lifetimes. The calculated lifetimes between $l_f = l_i \pm 1$ are found to be 5-10 ms and stimulated transitions due to blackbody radiation cut these lifetimes to probably hundreds of microseconds.

Two likely mechanisms for forming these states are inverse photoemission and charge exchange with ultra-cold Rydberg atoms. The former process favors low-energy recombination with rate coefficients of $\alpha^{rr} \sim 10^{-13} - 10^{-12} \text{ cm}^3 \text{ s}^{-1}$. In the latter approach, the cross section to capture electrons from Rydberg atoms scales geometrically as n^4 , where n is the Rydberg atom's principle quantum number.

Recent experimental realization of suspended CNT network[14] provides the necessary ground for observing the proposed states. We are extending our treatment to a periodic array of carbon nanotubes. The preliminary calculations show rich electronic structures with band gaps for the propagation inside the array.

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