Coulomb versus spin-orbit interaction in few-electron carbon-nanotube quantum dots

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(Dated: May 25, 2009)

Few-electron states in carbon-nanotube quantum dots are studied by means of the configuration-interaction method. The peculiar non-interacting feature of the tunneling spectrum for two electrons, recently measured by Kuenmth et al. [Nature 452, 448 (2008)], is explained by the splitting of a low-lying isospin multiplet due to spin-orbit interaction. Nevertheless, the strongly-interacting ground state forms a “Wigner molecule” made of electrons localized in space. Signatures of the electron molecule may be seen in tunneling spectra by varying the tunable dot confinement potential.

PACS numbers: 73.63.Fg, 73.23.Hk, 73.20.Qt, 73.22.Lp

After almost two decades of research, carbon nanotubes (CNTs)1 still provide a venue for the investigation of fundamental properties of interacting electron systems, such as Luttinger-liquid2 and Wigner-crystal3 behavior, Mott state4, Kondo effect5 and Andreev transport6 in CN quantum dots (QDs). With respect to semiconductor QDs7,8, low-screening, ultra-clean CN QDs appear to be ideal candidates9 for the realization of long-sought “Wigner molecules” (WMs)8 of strongly correlated electrons. These classical geometrical configurations of electrons localized in space are insensitive to the spin state of the system8,9. On the other hand, recently Kuenmth et al.9 showed that orbital and spin degrees of freedom are entangled by strong spin-orbit coupling in few-electron carbon-nanotube quantum dots.

In this Letter we show that non-interacting features of the tunneling spectrum, due to spin-orbit coupling, coexist with the strongly interacting nature of few-electron states, as seen from configuration interaction (CI, also known as exact diagonalization) calculations. Electrons in realistic dots form one-dimensional WMs, which may already have been observed in experiments8,9. We predict that molecular signatures appear in the excitation spectrum by varying the QD confinement potential.

We focus on a QD embedded in a semiconducting CN whose length scale, ℓQD, is smaller that the CN length. Hence, ℓQD is the relevant single-particle (SP) length and the effects of the CN boundaries may be neglected. With respect to previous calculations11,12,13,14, we assume the QD to be defined by an external gate potential, slowly varying on the lattice scale, which we model as a one-dimensional harmonic oscillator (HO) of frequency ω0. The quadratic potential is the low-energy generic form for a soft confinement15, setting ℓQD = (ℏ/m∗ω0)1/2, where m∗ = ℏ2/3Rγ is the effective mass, R is the CN radius, and γ = 0.54 eV·nm is the graphene π-band parameter. SP states ψnτz(r) = N Fνn(r)φnτz(r) are obtained by the envelope-function modulation Fνn(x) of bulk states φnτz(r) at the two non-equivalent minima of the lowest conduction band. The isospin index τz = +1 (−1) labels valley K (K′), Fνn(x) is the wave function of the nth HO excited state, and N is a normalization factor. Here φnτz(r) = exp(−iqτz/3R)|ψX,A(r) + τzψX,B(r)|, where ψX,A(r) and ψX,B(r) are the Bloch tight-binding states for sublattices A and B, respectively, at point X = K (X = K′) in the reciprocal space for τz = +1 (−1). The isospin τz = +1 (−1) points to the (anti)clockwise rotation along the circumference coordinate y, perpendicular to the tube axis x. The interacting Hamiltonian is Ĥ = ĤSP + ĤFW + ĤBW, which includes the SP term ĤSP = nτσ nτσ † nτσ nτσ, as well as the two-body terms for forward (FW) and backward (BW) Coulomb scattering processes ĤFW and ĤBW, respectively12,14,16. Here ĵnτσ destroys an electron occupying the SP orbital ψnτσ(r) with spin σz. The SP energy ĵnτσ includes the dominant term of spin-orbit interaction due to the CN curvature8,17,18 as well as the contribution of an axial magnetic field B17:

\[ ĵnτσ = ĵnHO + ΔSO γ 2 R τz σz + μB(Q′ σz − mRγ 2 R τz). \]

Here ĵnHO = γ/3R + hω0(n + 1/2), ΔSO is the spin-orbit coupling term, μB is the Bohr magneton, g* is the giro-magnetic factor. The two-body terms ĤFW and ĤBW are of the type Ĥ = 1/2 \[ \sum_{α,βσ} ĵαβσ ĵ†αβσ ĵβστ′ × δαστδστ′, \] where α = (n, τz) and Vαβσδ is the matrix element of the Ohno potential V(r − r′) = U0(1 + c2 |r − r′|2/U0/e)−1/2, which interpolates the two limits of Coulomb-like long-range and Hubbard-like short-range interactions (e is the relative dielectric constant, and U0 = 15 eV)14. In the envelope function approach19 the underlying graphene physics is buried into the precise form of Vαβσδ’s, which depend on both Bloch states φnτz(r) and envelopes Fνn(x). We evaluate Vαβσδ by considering explicitly the tight-binding expansion of φnτz(r) (cf.14). FW and BW terms correspond to direct [(τz, τz′) → (τz, τz′)] and exchange interactions.
Note that Fig. 2(a) perfectly matches Fig. 3(c) of [9]. The translation, i.e., converting into the gate voltages at which electrons tunnel into the QD [8]. We infer from [9] the inputs of CI calculation of the CI expansion of the wave function.

\[
|\tau_z, \tau'_z\rangle \rightarrow |\tau'_z, \tau_z\rangle, \tau_z \neq \tau'_z \]
is isospin scattering processes, respectively.

The few-body problem is solved by means of the CI method [8, 20]. We diagonalize \( H \), which is a matrix in the basis of the Slater determinants \( |\Phi_i\rangle \) obtained by filling with \( N \) electrons in all possible ways the thirty lowest-energy SP orbitals \( \psi_i(r) \). We obtain energies and wave functions of the many-body ground- and excited-states \( |\Psi(n)\rangle \), written as linear combinations of \( |\Phi_i\rangle \)'s, \( |\Psi(n)\rangle = \sum c_i^{(n)} |\Phi_i\rangle \), in each sector of the Fock space labeled by \( N \), the \( z \)-component of the total spin \( S_z \), and the total parity under spatial inversion \( x \rightarrow -x \).

To compare with tunneling spectra [8], we compute the chemical potential \( \mu(N) \) for a given value of \( B \), \( \mu(n) = E(n)(N) - E_0(N - 1) \), where \( E_0(N) (E_i(N)) \) is the energy of the many-body ground (ith excited) state with \( N \) electrons. The predicted \( \mu(N) \)'s may be converted into the gate voltages at which electrons tunnel into the QD [8]. We infer from [8] the inputs of CI calculation, i.e., \( h\hbar \omega_0 = 8 \text{ meV}, R = 3.6 \text{ nm}, \Delta_{SO} = 1.24 \times 10^{-3} \), \( \eta^* = 2.14 \). The unknown value of \( \epsilon \) is a fit parameter for a small-gap semiconductor which may be strongly affected by the leads. By choosing \( \epsilon = 3.5 \) we obtain the curves of Fig. 1 \( \mu(N) \) vs \( B \) for \( 1 \leq N \leq 3 \), which compare well with those of Fig. 3(a) in [9]. The plot quantitatively reproduces the dependence of \( \mu(N) \) on \( B \), specifically the isospin scattering processes, respectively.

The kink of \( \mu(2) \) at \( B_c \approx 0.125 \text{ T} \) and \( \mu(3) \) at \( \approx 0.15 \text{ T} \) [21]. As a check of consistency with the experiment, we remark that the CI spacing between \( \mu(2) \) and \( \mu(1) \) at \( B = 0 \), 17.8 meV, agrees within 6% with the value estimated in [9].

The kink of \( \mu(2) \) at \( B_c \) is due to the crossing between different ground states of two electrons, labeled A (B) for \( B < B_c \) (\( B > B_c \)) [cf. Fig. 1]. To gain insight into their nature, let us briefly consider also excited-state contributions to \( \mu_i(N) \), which are shown in Fig. 2(a) for \( N = 1, 2 \). Note that Fig. 2(a) perfectly matches Fig. 3(c) of [8]. The four lowest-energy SP levels \( \mu_i(1) \) are explicitly shown. Blue (green) lines indicate states with \( \tau_z = 1 (1) \), whose orbital magnetic moment is (anti)parallel to the CN axis, decreasing (increasing) its energy with \( B \). The fourfold degeneracy of the levels \( \langle \tau_z = \pm 1, \sigma_z = \pm 1 \mid \rangle \) at \( B = 0 \) is split by spin-orbit interaction, which entangles orbital and spin degrees of freedom [9]. This induces an isospin transition \( \langle \tau_z = -1 \mid \tau_z = 1 \rangle \) for the first excited non-interacting level \( \mu(1) \) at \( B \approx 0.125 \text{ T} \). Remarkably, this field is exactly the same as the critical value \( B_c \) at which the A \( \rightarrow \) B transition occurs [cf. the vertical dashed line in Fig. 2(a)]. Because of this feature, it was argued in [9] that the A \( \rightarrow \) B transition may be solely explained in terms of spin-orbit interaction. The state A was supposed to be a single Slater determinant with the electrons in the two lowest spin-orbitals, \( \langle \tau_z = 1, \sigma_z = -1 \rangle \) and \( \langle \tau_z = -1, \sigma_z = 1 \rangle \), whereas B is obtained by moving the electron from \( \langle \tau_z = -1, \sigma_z = 1 \rangle \) to \( \langle \tau_z = 1, \sigma_z = 1 \rangle \). In this picture correlation effects are absent. We next show that A and B are instead strongly interacting states.

The right panel of Fig. 1 shows the Slater determinants with the largest weights in the CI expansion of \( \langle A \rangle \). The blue (green) ladders of levels depict the HO states for \( \tau_z = +1 (1) \), whereas arrows represent spins. The main configuration, whose weight is 53\%, is the Slater determinant proposed in [9] for the ground state. However, CI calculation shows that there are other three relevant determinants where also the excited states of the HO are populated (Fig. 1). This is due to the correlated character of \( \langle A \rangle \), the strongest the impact of Coulomb interaction, the largest the mixing of determinants. Similarly, state B shown in the right panel of Fig. 1 is correlated as well. Besides, state B may be obtained from A by replacing the levels with \( \tau_z = -1 \) with those with \( \tau_z = 1 \) (Fig. 1). Hence, A and B belong to a isospin multiplet, only differing in the projections \( T_z \) of the isospin.
where the components of $\sigma$ are the Pauli matrices [e.g., $T_z = (\hat{n}_+ - \hat{n}_-) / 2$ with $\hat{n}_\mu = \sum_{\alpha} \hat{c}^\dagger_{\mu\alpha} \hat{c}_{\mu\alpha}$]. This is confirmed by extrapolating the behavior of A and B to the limit $\Delta_{SO} \rightarrow 0$ (not shown). In this limit, A and B and other states at higher energies collapse into a six-fold multiplet, which includes three spin triplet ($T = 0$, $S = 1$, $S_z = \pm 1, 0$) plus three isospin triplet ($S = 0$, $T = 1$, $T_z = \pm 1, 0$) states. In fact, the total wave functions must be odd whereas their (unique) orbital part is even under particle exchange. Therefore, except for a tiny residual splitting of $\approx 2 \mu eV$ due to BW interaction, the A-B energy separation depends on spin-orbit interaction only.

From Eq. 1 and the inspection of CI wave functions of Fig. 2(b), it is clear that the energy splitting between A and B at zero field, $\mu_1(2) - \mu(2) = 2\Delta_{SO}/R$, is the same as that between one-electron levels, $\mu_1(1) - \mu(1)$ [cf. Fig. 2(a)]. Besides, $\mu(N)$ depends on $B$ through the spin and orbital magnetic dipole moments [cf. Eq. (1)], which are linear in $\Delta S_z = S_z(N) - S_z(N-1)$ and $\Delta T_z = T_z(N) - T_z(N-1)$, respectively. It is immediate to verify that $\Delta S_z$ and $\Delta T_z$ are the same for both $\mu(2)$ and $\mu(1)$, for $B < B_c$ ($\Delta S_z = 1/2, \Delta T_z = -1/2$) and $B > B_c$ ($\Delta S_z = 1/2, \Delta T_z = 1/2$). This explains why the critical value of the field for both $\mu(2)$ and $\mu(1)$ is the same despite the correlated nature of A and B, in agreement with the key experimental observation [9].

The non-interacting feature of $\mu(2)$ discussed above is not universal and may be affected by electron correlation. This is the case as one changes e.g. the QD potential, which is controlled in the laboratory by a capacitively coupled gate. This in turn changes the ratio of Coulomb matrix elements $V$ to the SP spacing $\hbar \omega_0$. Figure 2(b) shows the analogous plot of Fig. 2(a) for half the value of the confinement energy, $\hbar \omega_0 = 4$ meV. The pattern of $\mu(2)$ vs $B$ has now changed with respect to Fig. 2(a), due to the crossing between A and a new state, labeled C in the plot, occurring close to the origin, at $B \approx 0.02$ T. This critical value depends on the splitting $\Delta_{CA}$ between C and A at zero field, which in turn is sensitive to Coulomb correlation.

The crucial role of Coulomb interaction in the QD is fully appreciated by considering the excitation spectrum. Figure 3 shows the two-electron excitation spectrum vs the dielectric constant $\epsilon$, which affects the relevance of correlation effects. The lower bound on the horizontal axis ($\epsilon = 1.5$) mimics low screening, typical of large-gate semiconducting CNs, whereas the upper bound ($\epsilon = 5000$) may be regarded as the non-interacting limit. In the latter, all levels bunch into the HO levels, uniformly spaced by $\hbar \omega_0 (= 8$ meV). In the lowest set of levels (red lines) the two electrons occupy the nodeless orbitals in one of the two valleys, whereas the next set (black lines) is obtained by promoting one electron into the first HO excited state. The states of the first (second) set have even (odd) parity. Within each set, a residual fine structure survives, entirely due to spin-orbit interaction. As indicated in Fig. 3 the total number of levels in the first (second) set is 6 (16), given by the possible ways to arrange the two electrons either in the same or in different valleys compatibly with Pauli’s exclusion principle.

As $\epsilon$ is reduced in Fig. 3 Coulomb interaction alters the energy spectrum. In fact, the sixteen odd levels belonging to the second set separate into two multiplets. A first sixfold multiplet is insensitive to $\epsilon$, whereas a second tenfold multiplet experiences a sudden energy drop. The former multiplet is associated to the collective motion of the center of mass (Kohn mode), which is decoupled from the relative motion and hence unaffected by Coulomb interaction [8]. The sixfold degeneracy of this odd multiplet, lifted only by spin-orbit interaction, is the same as that of the lowest even multiplet, since they differ only in the excitation of the center of mass coordinate. The second odd multiplet is instead sensitive to $\epsilon$, i.e., Coulomb interaction. In fact, $\Delta_{CA}$ becomes a small fraction of $\hbar \omega_0$ at the experimental value of $\epsilon = 3.5$ ($\Delta_{CA}/\hbar \omega_0 \approx 0.06$), whereas it vanishes at $\epsilon = 1.5$. As discussed below, such vanishing points to the formation of a WM, the state where Coulomb correlation localizes electrons in space to minimize their electrostatic energy [3, 8].

The insets of Fig. 4 show the correlation function $g(x)$ vs $x$ for states A and C. $g(x)$ is the probability of finding the two electrons at relative distance $x = x_1 - x_2$ [normalized as $\int_0^\infty g(x)dx = 1$, with $g(x) = g(-x)$]. As $\epsilon$ decreases from $\epsilon = 25$ (bottom right inset) to the experimental value $\epsilon = 3.5$ (top left inset), the ground-state probability distribution $g_A(x)$ (red curve) develops a well defined peak at $x_0 = 1.7$, showing that electrons localize in space and freeze their mutual distance. $x_0$ compares well with the equilibrium value $x_{0,cl} = 1.6$ of two point-like classical particles in the HO trap interacting via the Coulomb potential $\epsilon^2/|x|$. Note that crystallization proceeds by removing probability weight from
correlation hole rapidly forms in A as \( \epsilon \) decreases (cf. Fig. 3). Similarly, as the overlap between the probability weights of localized electrons is suppressed, exchange interaction is negligible and different values of \( S_z \) (\( S_z = 0 \) for A and \( S_z = -1 \) for C) are admissible. In this limit states A and C, which only differ for now irrelevant quantum numbers like parity and spin, represent the same “classical” configuration.

We exploit the progressive overlap between \( g_A(x) \) and \( g_C(x) \) as \( \epsilon \) is reduced to characterize the transition to the WM state. In Fig. 4 we plot vs \( \epsilon \) the functional distance \( d_{CA} \) between states A and C, defined as \( d_{CA} = \int_0^\infty dx |g_A(x) - g_C(x)| \). The semilog plot allows to identify three separate regions, where \( d_{CA} \) scales differently with \( \epsilon \) (there are no sharp transitions in finite-size systems). For \( \epsilon > 20 \), \( d_{CA} \) slowly tends to the upper bound 1, since the location of the maximum of \( g_A(x) \) approaches the origin whereas \( g_C(x) \) has a node there. The large probability of finding two particles close to each other \( (x \approx 0) \) highlights the absence of a correlation hole in the ground state (cf. the magenta dot and related plot). In the crossover region, \( 5 < \epsilon < 20 \), \( d_{CA} \propto \log \epsilon \) as shown by the linear fit in Fig. 4 (dashed line). Here a significant correlation hole rapidly forms in A as \( \epsilon \) decreases. The WM corresponds to \( \epsilon < 5 \), where \( d_{CA} < 0.1 \) and it slowly decreases with \( \epsilon \), as \( g_A(x) \) and \( g_C(x) \) overlap almost perfectly. Remarkably, the observed case \( \epsilon = 3.5 \) (blue dot in Fig. 4) occurs in this region.

The squeezing of the QD confinement potential via an external gate is a handle to drive Wigner crystallization, since the effect of lowering \( \hbar \omega_0 \) [Fig. 2(b)] is similar to that of decreasing \( \epsilon \) (Figs. 2 and 3). In fact, the critical \( B \)-value of the \( A \to C \) transition reported in Fig. 2(b) is a measure of the vanishing of \( \Delta_{CA} \). As \( \hbar \omega_0 \) is reduced, this critical field approaches zero, implying that the spin-polarized phase C may be induced with no energy cost. The latter behavior has been observed for hole WMs [3].

We have shown that spin-orbit and strong Coulomb interaction coexist in CN QDs, leading to the formation of WMs at experimentally attainable regimes. This insight into the entangled orbital and spin degrees of freedom is relevant for the all-electrical [9] and -optical [22] manipulation of electron spins in CN-based devices. \note{added}{After the submission of this work, Wunsch reported similar results for a square-well QD [23].}

We thank F. Manghi, E. Molinari, E. Andrei, G. Steele for stimulating discussions. This work is supported by INFM-CINECA Supercomputing Project 2008-2009.

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