Shaping electron wave functions in a carbon nanotube with a parallel magnetic field

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A magnetic field, through its vector potential, usually causes measurable changes in the electron wave function only in the direction transverse to the field. Here we demonstrate experimentally and theoretically that in carbon nanotube quantum dots, combining cylindrical topology and bipartite hexagonal lattice, a magnetic field along the nanotube axis impacts also the longitudinal profile of the electronic states. With the high (up to 17 T) magnetic fields in our experiment the wave functions can be tuned all the way from “half-wave resonator” shape, with nodes at both ends, to “quarter-wave resonator” shape, with an antinode at one end. This in turn causes a distinct dependence of the conductance on the magnetic field. Our results demonstrate a new strategy for the control of wave functions using magnetic fields in quantum systems with nontrivial lattice and topology.

As first noticed by Aharonov and Bohm [1], when a charged quantum particle travels in a region of finite electromagnetic potential, its wave function acquires a phase whose magnitude depends on the travelled path. For particles with electric charge $q$ moving along a closed path, the phase shift $ϕ_{AB} = qΦ_B/ℏ$, known as Aharonov-Bohm shift, is expressed in terms of the magnetic flux $Φ_B$ across the enclosed area. Because $Φ_B$ depends only on the magnitude of the magnetic field component normal to this area’s surface, the phase is acquired along directions transverse to the magnetic field, see Fig. 1(a). In mesoscopic rings or tubular structures pierced by a magnetic field, the phase changes the quantization condition for the tangential part of the electronic wave vector by $k_⊥ → k_⊥ + ϕ_{AB}/r$ (with $r$ the radius of the ring or tubus) and is at the basis of remarkable quantum interference phenomena [2]. However, as the perpendicular components of the magnetic vector potential commute with the parallel component of the momentum, a parallel magnetic field is not expected to affect the wave function in the direction of the field.

Also in carbon nanotubes (CNTs), the electronic wave function acquires an Aharonov-Bohm phase when a magnetic field is applied along the nanotube axis [3], see Fig. 1(a). The phase gives rise to resistance oscillations in a varying magnetic flux [4]. Since it changes $k_⊥$, it also changes the energy $E(k)$ of an electronic state, through its dependence on the wave vector $k = (k∥, k_⊥(B∥))$. Such a magnetic field dependence of the energies has been observed through beatings in Fabry-Perot patterns [5], or in the characteristic evolution of excitation spectra of CNT quantum dots in the sequential tunneling [6–9] and Kondo [10–15] regimes.

In this Letter we show that the combination of the bipartite honeycomb lattice, the cylindrical topology of the nanotubes, and the confinement in the quantum dot intertwines the usually separable parallel and transverse components of the wave function. This leads to an unusual tunability of the wave function in the direction parallel to the magnetic field. Experimentally, it manifests in a pronounced variation of the conductance with magnetic field, arising from the changes of the wave function amplitude near the tunnel contacts between the electrostatically defined quantum dot and the rest of the CNT.

Similar to graphene, in CNTs the honeycomb lattice gives rise to two non-equivalent Dirac points $K$ and $K'$ (also known as valleys). The valley and spin degrees of freedom characterize the four lowermost CNT subbands, see Fig. 1(c). Our measurements display i) a conductance rapidly vanishing in a magnetic field for transitions associated to the $K$-valley; ii) an increase and then a decrease

FIG. 1. (a) Electrons circulating in closed orbits acquire an Aharonov-Bohm phase proportional to the enclosed magnetic flux. (b) Schematic of a suspended CNT device with its embedded quantum dot (shaded green) and a magnetic field parallel to the nanotube. (c) Dirac cones of the graphene dispersion relation. Blue and red lines indicate the lowermost transverse subbands forming in a CNT. Spin degeneracy is lifted by the spin-orbit coupling. Quantized $k∥$ values due to a finite CNT length are marked with dots; $B∥ = 0$. (d) An axial magnetic field changes $k_⊥$ via the Aharonov-Bohm effect, shifting the 1-d subbands across the Dirac cones.
of the conductance for $K'$-valley transitions as the axial field is varied from 0 up to $17 \, \text{T}$. Indications of similar behavior can also be found in results from other CNT quantum dots [7, 9]. To our knowledge, no microscopic model explaining it has yet been proposed. Our calculation captures this essential difference between the K and K' valley states.

**Dispersion relation of long CNTs**—In CNTs the eigenstates are spinors in the bipartite honeycomb lattice space, solving the Dirac equation, Eq. (2) below. The resulting dispersion is $E(k) = \pm \hbar v_F \sqrt{k_\perp^2 + k_\parallel^2}$, see Fig. 1(c), where $k_\perp = k_{1/\parallel} - \tau K_{1/\parallel}$ are wave vectors relative to the graphene Dirac points $K$ ($\tau = 1$) and $K' = -K$ ($\tau = -1$).

The cylindrical geometry restricts the values of the transverse momentum $k_\perp$ through the boundary condition $\psi(R + C) = \psi(R)$, with $C$ the wrapping vector of the CNT, generating transverse subbands, see Fig. 1(c). Furthermore, curvature causes a chirality-dependent offset $\tau \Delta k_\perp$ of the Dirac points, opening a small gap in metallic nanotubes with $k_\perp = 0$, as well as a spin-orbit coupling induced shift $\sigma \kappa_{SO}$ of the transverse momentum [16–18] ($\sigma = \pm 1$ denotes the projection of the spin along the CNT axis). As shown in Fig. 1(c), the latter removes spin-degeneracy of the transverse subbands. When an axial magnetic field is applied, the Aharonov-Bohm phase further modifies $k_\perp$. The energy $E(k_\parallel, k_\perp)$ of an infinite CNT then follows again from the Dirac equation under the replacements

$$k_\perp \rightarrow k_\perp + \frac{\varphi B}{R} + \sigma \Delta k_{SO} + \tau \Delta k_\perp,$$

$$k_\parallel \rightarrow k_\parallel + \tau \Delta k_\parallel,$$

the addition of a Zeeman term $\mu_B \sigma B_\parallel$, and a field-independent energy shift due to the spin-orbit coupling [16–18]. In CNT quantum dots with lengths of few hundreds of nanometers the longitudinal wave vector becomes quantized, leading to discrete bound states (dots in Fig. 1(c)). The magnetic field dependence of $E$ for two bound states belonging to different valleys is shown in Fig. 1(d) for fixed $k_\parallel$. A characteristic evolution, distinct for the two valleys, is observed.

**Magnetospectrum of a CNT quantum dot**—The non-linear differential conductance of a quantum dot gives access to its excitation spectrum [18]. Fig. 1(b) shows a schematic of our device: a suspended CNT grown in situ over rhenium leads [19, 20]. By tuning the back gate voltage we can explore both hole and electron conduction regimes, the latter displaying clear Coulomb oscillations near the band gap (see also Sec. VII of the Supplement [21]). This way it was possible to clearly identify the gate voltage region corresponding to $0 \leq N \leq 1$ trapped conduction band electrons.

Figure 2(a) shows the stability diagram of the CNT in this gate voltage region. The resonance lines correspond to the single particle energies of the lowest discrete states of the quantum dot. Two closely spaced sets $\alpha$ and $\beta$ of two Kramers doublets each are visible. By fixing $V_{\text{gate}}$ and sweeping a magnetic field, the evolution of the states in the field can be recorded, see Figs. 2(b,c). The Kramers degeneracy is then lifted, revealing four states in each set $\alpha$ and $\beta$.

**Low field spectra**— FIG. 2. (a) Zero magnetic field differential conductance $dI/dV_{\text{bias}}$ of a CNT quantum dot with $0 \leq N \leq 1$ conduction band electrons. Two pairs $\alpha$ and $\beta$ of conductance lines, all four representing Kramers doublets, are visible. (b) Differential conductance for constant $V_{\text{gate}} = 0.675 \, \text{V}$ and varying magnetic field $|B_\parallel| \leq 1.5 \, \text{T}$. The Kramers doublets split at finite field into four states for both $\alpha$ and $\beta$. Spin and valley of the $\alpha$ states for $B_\parallel \gg 0.5 \, \text{T}$ are indicated. (c) Differential conductance at the same $V_{\text{gate}}$, now for $B_\parallel \leq 17 \, \text{T}$. The four visible lines correspond to $K'$ valley states in shells $\alpha$ and $\beta$; the $K$ lines fade out fast. (d) Calculated conductance, using the reduced density matrix technique and assuming field-independent tunneling coupling of all states to the leads. In contrast to the measurement, both $K$ and $K'$ valley lines clearly persist at high magnetic field.

We have traced the single particle states from Fig. 2(b) up to a high magnetic field of $B_\parallel = 17 \, \text{T}$. As visible in Fig. 2(b,c), the four $K$ lines evolve upwards in energy. They are comparatively weak, fading out already below
In contrast, the four $K'$ conductance lines evolve initially downwards, gaining in strength, but then turn upwards above 6 T and fade too. The model calculation of the conductance in Fig. 2(d), assuming a field independent $k_{||}$, successfully follows the peak positions but clearly fails to reproduce the intensity variations, especially the suppression of transport for $K$ lines already at low fields. The presence of both weak $K$ and strong $K'$ transitions at the same bias excludes the possibility of a trivial dependence of tunneling rates on the bias voltage.

We show in the following that this effect results from the $B_{||}$ dependence of the wave functions’ longitudinal profile. When the field is applied perpendicular to the CNT axis no such effect occurs and all excitation lines are present at almost constant strength [21].

Boundary conditions on bipartite lattices—The spatial profile of the wave functions $\psi(r)$ of a finite quantum system is determined by the boundary conditions and the resulting quantization of the wave vector. In unipartite lattices, e.g., monoatomic chains, hard-wall boundary conditions are $\psi(R_L) = 0 = \psi(R_R)$, where $R_{L/R}$ are the lattice vectors of the first site beyond the left and right end of the chain, respectively. The linear combinations of Bloch states satisfying these conditions create standing waves with nodes at $R_L$ and $R_R$, as those of a half-wave resonator. Their wave vectors are quantized according to the familiar condition $k_{||} = n\pi/L$, where $L$ is the length of the chain and $n \in \mathbb{N}$.

The situation is more complex in bipartite lattices, as in the CNT shown in Fig. 3. The eigenstates are spinors in sublattice space, $\Psi = (\psi_A, \psi_B)$, and near the Dirac points obey the Dirac equation

$$h v_F \begin{pmatrix} 0 & e^{i\tau\theta}(\tau_{K_{||}} - i\kappa_{||}) \\ e^{-i\tau\theta}(\tau_{K_{||}} + i\kappa_{||}) & 0 \end{pmatrix} \begin{pmatrix} \psi_k A \\ \psi_k B \end{pmatrix} = E \begin{pmatrix} \psi_k A \\ \psi_k B \end{pmatrix},$$

where $v_F$ is the Fermi velocity and $\theta$ is the CNT chiral angle. They have the form $\psi_k = w(e^{i\eta(k)}\psi_{kA} + e^{-i\eta(k)}\psi_{kB})$, with $w$ a normalization factor, meaning that there is a phase shift $2\eta(k) = -\pi \arctan(\kappa_{||}/\kappa_{\perp}) + \tau \theta$ between the two sublattice wavefunctions $\psi_{kA}$ and $\psi_{kB}$. On the B atoms the phase is advanced by $\eta(k)$ with respect to the plane wave part of the Bloch state, on the B atoms it is retarded. This is shown in Fig. 3(a) for a $(6,3)$ chiral nanotube, where the real part of the plane wave $e^{i\kappa_{||}r}$ is plotted in the background, and the real part of the complete Bloch function $\Psi_k(r)$ at each atomic position is shown as the filling of the white (sublattice A) and black (sublattice B) circles.

Standing waves in a finite CNT are formed by appropriate linear combinations of forward [$f$] and backward [$b$] propagating waves of the same energy, see Fig. 3(b). A specific combination of Bloch states $\Psi = c_f \Psi_k f + c_b \Psi_k b$ may satisfy the boundary condition $\psi_A(R_L) = 0$, but then in general $\psi_B(R_L) \neq 0$. The counterpropagating Bloch waves interfering destructively on A remain finite on B because they are superposed with different phases, see Fig. 3(c). There is no non-trivial superposition with nodes at both ends for both sublattice components. Thus, the boundary conditions for bipartite lattices are either $\psi_A(R_L) = 0 = \psi_B(R_R)$ or $\psi_A(R_R) = 0 = \psi_B(R_L)$, depending on the sublattice to which the majority of the relevant edge atoms belongs [22, 23]. The only exception is an armchair nanotube, which has equal numbers of A and B atoms at the edges and obeys the usual half-wave quantization condition $k_{||} = \pi/L$. The superposition of forward and backward moving Bloch states with $\pm\kappa_{||}$ and the same $\tau\kappa_{\perp}$, together with the bipartite boundary conditions, leads to the unusual quantization condition [22–24]

$$e^{2i\kappa_{||}L} \equiv e^{-2i\eta(k)} e^{i\tau\theta} = \frac{\tau_{K_{||}} + i\kappa_{||}}{\tau_{K_{||}} - i\kappa_{||}}.$$  

Since Eq. (3) couples the transverse and the longitudinal direction, it can be seen as a cross-quantization condition. It implies that in an axial field also $k_{||}$ depends on $B_{||}$.

The solutions of Eq. (3) are plotted as coloured lines in Fig. 4(a). For comparison, the grey lines parallel to the $k_{||}$ axis correspond to the familiar half-wave solutions. The envelope wave function on the A sublattice is also...
sketched; the $B$ counterpart is its mirror image. When $k_\parallel$ is close to a multiple of $\pi/L$ (for large $B_\parallel$), the wave function has the standard half-wave shape with a node at each end. When the magnetic field is low, the profile on each sublattice is close to a quarter-wave, with an antinode at the corresponding unconstrained end.

Figure 4(b) shows the calculated wave function amplitudes for the lowest mode ($n = 1$), $|\psi_{1\tau}(x_\parallel)|^2$ on the $A$ and $B$ sublattices, of a (15,3) CNT with $L = 121 \text{ nm}$. They were obtained by direct diagonalization of a tight-binding Hamiltonian on finite lattice, with four valence molecular orbitals is obtained by scanning the tip position over the sample. In CNT quantum dots, the contact confinement as well.

In conclusion, our experiment can be regarded as the complement of a scanning tunneling microscopy (STM) measurement. In STM the spatial profile of atomic or molecular orbitals is obtained by scanning the tip position over the sample. In CNT quantum dots, the contact position is fixed, but the wavefunction, and thus the tunnel current, is tuned by an axial magnetic field. We are aware of only one other system in which such coupling has been found, a semiconducting quantum dot with pyramid shape [26]. The unusual tunability of the wave function shape with a parallel magnetic field will influence all phenomena dependent on the full spatial profile of the CNT wave function.
the electronic states, such as, e.g., electron-phonon coupling or electron-electron interaction. Thus the parallel magnetic field is an even more versatile tool to investigate and control complex quantum systems than already acknowledged.

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