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

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Transport through any electronic system is determined by the properties of its electronic states which serve as transport channels; their amplitude at the contacts is essential to the coupling to external leads. 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. They 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 nonlinear conductance on the magnetic field. Our results shed new light on the impact of magnetic fields on quantum systems with nontrivial lattices 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 complex phase whose magnitude depends on the travelled path. For particles with electric charge q moving along a closed path, the phase shift \( \varphi_{AB} = q\Phi_B/h \), known as Aharonov-Bohm shift, is expressed in terms of the magnetic flux \( \Phi_B \) across the area enclosed by the path. Because \( \Phi_B \) 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. 1a. The inclusion of this phase in mesoscopic rings or tubular structures pierced by a magnetic field changes the
Figure 1. **Aharonov-Bohm phase in carbon nanotubes.**

- **a**, Electrons circulating in closed orbits acquire an Aharonov-Bohm (AB) phase proportional to the enclosed magnetic flux.
- **b**, Schematic of a suspended CNT device with its embedded quantum dot. When a magnetic field is applied in parallel to the nanotube, electrons circulating clockwise or anticlockwise around the CNT waist acquire an AB-phase.
- **c**, Dirac cones of the graphene dispersion relation, with the four lowermost transverse subbands forming in a carbon nanotube indicated. In each valley ($K$ or $K'$) spin degeneracy is lifted by the spin-orbit coupling. Here $B_\parallel = 0$. Quantized $k_\parallel$ values due to a finite nanotube length are marked with dots.
- **d**, Sweeping a parallel magnetic field $B_\parallel$ changes $k_\perp$ via the Aharonov-Bohm effect. This modifies the subband energy, effectively scanning the Dirac cones.

For the tangential part of the wave vector, the quantization condition is $k_\perp \rightarrow k_\perp + \varphi_{AB}/r$, where $r$ is the radius of the ring or of the tubulus, and is at the basis of remarkable interference phenomena.\(^2\)

Also in carbon nanotubes (CNTs), hollow cylinders of carbon atoms, the electronic wave function acquires an Aharonov-Bohm phase when a magnetic field is applied along the...
nanotube axis, see Fig. 1a.\textsuperscript{3} The phase gives rise to interference effects visible in resistance oscillations in a varying magnetic flux.\textsuperscript{4} Since the Aharonov-Bohm phase changes $k_\perp$, this also changes the energy $E = E(k)$ of an electronic state, through its dependence on the wave vector $k = (k_\parallel, k_\perp(B_\parallel))$. In CNTs such a magnetic field dependence of the energies has been observed through beatings in Fabry-Perot patterns,\textsuperscript{5} or in the characteristic evolution of excitation spectra of CNT quantum dots (see Fig. 1b) in the sequential tunneling\textsuperscript{6–9} and the Kondo\textsuperscript{10–15} regimes.

In this work we show that, counter-intuitively, also the longitudinal part $k_\parallel$ of the wave vector can be affected by an axial magnetic field in CNT quantum dots. We are aware of only one other system in which such coupling has been achieved and measured, a semiconducting quantum dot with pyramid shape.\textsuperscript{16} In CNTs the bipartite nature of the honeycomb lattice, combined with finite size effects, is what intertwines the parallel and transverse components of the wave vector, $k_\parallel$ and $k_\perp$.\textsuperscript{17} As a result, the shape of the axial part of the wavefunction becomes tunable by the magnetic field.

Experimentally, this effect can be detected via differential conductance measurements. Similar to graphene, in CNTs the honeycomb lattice gives rise to two non-equivalent Dirac points $K$ and $K'$ (also known as valleys) with associated Dirac cones. The valley and spin degrees of freedom thus characterize the four lowermost longitudinal CNT subbands shown in Fig. 1c. As discussed below, the $k_\perp(B_\parallel)$ evolution is different for a $K$ or $K'$ valley, see Fig. 1d. This in turn affects $k_\parallel(B_\parallel)$ and thus the magnetoconductance. Our measurements reveal unexpectedly: i) a very low magnetoconductance for transitions associated to $K$-valley transport; ii) an increase and then a decrease of the magnetoconductance for $K'$-valley transitions as the axial field is varied from 0 up to 17 T. Such behavior can also be seen in results on other CNT quantum dots in the single electron conduction regime.\textsuperscript{7,9} To our knowledge, no microscopic model explaining this peculiar observation has yet been proposed. To appreciate the experimental findings and their interpretation, we recall in the following relevant features of the spectrum of a CNT in an axial magnetic field.

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

The cylindrical geometry restricts the allowed values of the transverse momentum $k_\perp$
through the boundary condition \( \Psi(R + C) = \Psi(R) \), with \( C \) the wrapping vector of the CNT, giving rise to transverse subbands, see Fig. 1c. Furthermore, curvature effects are responsible for a chirality-dependent offset \( \tau \Delta k^c \) of the Dirac points which opens a small gap in nominally metallic CNTs with \( \kappa_\perp = 0 \), as well as for a spin-orbit coupling induced shift \( \sigma k_{SO} \) of the transverse momentum (here \( \sigma = \pm 1 \) denotes the projection of the spin along the nanotube axis).\(^{18}\) As shown in Fig. 1c, the latter removes the spin-degeneracy of the transverse subbands. When a magnetic field \( B_\parallel \) is applied along the CNT axis, the Aharonov-Bohm phase further modifies the transverse momentum component. The energy \( E(k_\parallel, k_\perp(B_\parallel)) \) of an infinite CNT then follows again from the Dirac equation under the replacements

\[
\begin{align*}
\kappa_\perp &\to \kappa_\perp + \frac{\varphi_{AB}}{r} + \sigma \Delta k_{SO} + \tau \Delta k^c_\perp, \\
\kappa_\parallel &\to \kappa_\parallel + \tau \Delta k^c_\parallel,
\end{align*}
\]

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

**Magnetospectrum of a CNT-based quantum dot.** Differential conductance measurements of quantum dots as a function of the applied bias voltage \( V_{bias} \) and gate voltage \( V_{gate} \) (known as stability diagrams) give access to the quantum dot excitation spectrum via transport experiments.\(^{18}\) Figure 1b shows a schematic of our device: a suspended clean carbon nanotube grown \textit{in situ} over rhenium leads. By tuning the back gate voltage we can explore both the transparent hole conduction regime and the electron conduction regime, the latter displaying clear Coulomb oscillations near the band gap (see Section III of the Supplementary Material). This way it was possible to clearly identify the gate voltage region corresponding to the quantum dot occupation of \( N = 0 \) or \( N = 1 \) electrons in the conduction band.

Fig. 2a 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. Our conductance measurement reveals two closely spaced sets of two Kramers doublets
Figure 2. Magnetospectrum of a single electron at high magnetic field. a, Zero magnetic field differential conductance \( \frac{dI}{dV_{\text{bias}}} \) of a CNT quantum dot in the gate voltage region where \( 0 \leq N \leq 1 \) conduction band electrons are on the quantum dot. Two pairs \( \alpha \) and \( \beta \) of differential conductance lines are visible. Each line represents a Kramers doublet; the line separation in each pair is caused by the spin-orbit coupling. b, Differential conductance for constant gate voltage \( V_{\text{gate}} = 0.675 \text{ V} \) and varying magnetic field \( |B_\parallel| \leq 1.5 \text{ T} \). The Kramers doublets split further at finite field yielding four states for each set \( \alpha \) and \( \beta \). Spin and valley of the \( \alpha \) states for \( B_\parallel \gg 0.5 \text{ T} \) are indicated. c, Differential conductance for the same gate voltage, now for \( B_\parallel \) up to 17 T. The four visible conductance lines correspond to \( K' \) states in shells \( \alpha \) and \( \beta \); as already visible in b, the \( K \) lines fade out very fast. Between 4 T and 8 T the quantum dot hosts \( N = 1 \) electron at \( V_{\text{bias}} \approx 0 \), see Section I of the Supplementary Material. 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 are clearly visible.
each, which we henceforth will call sets $\alpha$ and $\beta$. By fixing the gate voltage and sweeping a magnetic field, the evolution of the states in the field can be recorded, as shown in Figs. 2b,c. The Kramers degeneracy is then lifted, revealing four states in each of the two sets $\alpha$ and $\beta$.

Low field spectra similar to the one shown in Fig. 2b have been reported by several groups\textsuperscript{6–9} and are by now well understood. A quantitative fit can be obtained by a model Hamiltonian for a single longitudinal mode, including a valley mixing on the order of 0.1 meV due to disorder or backscattering at the edge (see Ref. 8 and Section VII of the Supplementary Material). At magnetic field amplitudes larger than 0.5 T, the valley mixing is not relevant, and the Dirac equation, Eq. (2) below, with curvature-induced shifts, fully accounts for the evolution of the spectral lines. This also implies that a valley and spin degree of freedom can be associated to each excitation line at higher fields, as indicated in Fig. 2b for set $\alpha$.

We have traced the evolution of the single particle quantum states from Fig. 2b up to a high magnetic field of $B_\parallel = 17$ T. As can be seen in Fig. 2b and Fig. 2c, the four $K$ conductance lines evolve upwards in energy. They are very weak, fading out already at fields below 1 T. In contrast, the four $K'$ conductance lines evolve initially downwards in energy, gaining in strength, but then turn upwards above 6 T and begin to fade again. The calculation of the conductance in Fig. 2d, which assumes a field independent $k_\parallel$, successfully follows the peak positions but clearly fails to reproduce the intensity variations, especially the suppression of transport for the $K$ lines already at low fields. We will show in the following that this effect results from the $B_\parallel$ dependence of the wave functions’ longitudinal profile.

**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 $R_L/R_R$ are the lattice vectors of the first site beyond the left and right end of the chain, respectively. The linear combinations of Bloch states which satisfy 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 $k_\parallel = n\pi/L$, where $L$ is the length of the chain and $n \in \mathbb{N}$.

The situation is more complex in bipartite lattices, as in the case of the nanotube shown in Fig. 3. The eigenstates of the system are spinors in sublattice space, $\Psi^\dagger = (\psi^\dagger_A, \psi^\dagger_B)$, and
Figure 3. **Boundary conditions for standing waves on a bipartite hexagonal lattice.** a, Bloch function $\psi_k$ in a (6,3) infinite nanotube calculated at the atomic positions (white-rimmed circles for the $A$ sublattice and black-rimmed circles for $B$). The colour map in the background shows the real part of a forward-propagating plane wave with momentum $k_f$. The phase of the Bloch function is advanced by $\eta(k)$ with respect to the plane wave on $A$ sublattice sites and retarded by the same phase on $B$ sites. As an example, in the outlined unit cell at position $R$ the real part of the plane wave is small (very light at $R$), but the Bloch function on the $A$ atom is already large and negative (dark violet circle filling) while on the $B$ atom it is still large and positive (dark brown). b, Standing waves in a finite nanotube are composed of forward (f) and backward (b) propagating Bloch states $\psi_{k_f}$ and $\psi_{k_b}$ from the opposite sides of a Dirac cone and at the same energy, $E(k_f) = E(k_b)$. c, Left end of a (6,3) chiral CNT. The solid-drawn atoms and bonds belong to the CNT quantum dot (cf. Fig. 1b), the faint ones to the tunneling region. In the unrolled lattice the quantum dot area is shaded, while the atoms on pale background belong to the boundary sites $\{R_L\}$. The superposition of $\psi_{k_f}$ and $\psi_{k_b}$, with appropriate coefficients $c_f, c_b \in \mathbb{C}$, forms the wave function of an energy eigenstate in a finite nanotube. The calculated superposition amplitude approaches zero towards the left end on the $A$ atoms only (light atoms in the 3D view or light filling in the 2D view) while remaining strong on the $B$ atoms (dark atoms or dark filling).
near the Dirac points solve the Dirac equation
\[ h v_F \begin{pmatrix} 0 & e^{i\tau \theta} (\tau \kappa_\perp - i \kappa_\parallel) \\ e^{-i\tau \theta} (\tau \kappa_\perp + i \kappa_\parallel) & 0 \end{pmatrix} \begin{pmatrix} \psi_{kA} \\ \psi_{kB} \end{pmatrix} = E \begin{pmatrix} \psi_{kA} \\ \psi_{kB} \end{pmatrix}, \]

where \( v_F \) is the Fermi velocity and \( \theta \) is the chiral angle describing the rolling direction of the CNT. For a given momentum 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 \( 2\eta(k) = -\tau \arctan(\kappa_\parallel/\kappa_\perp) + \tau \theta \) between the two sublattice wavefunctions \( \psi_{kA} \) and \( \psi_{kB} \). On the \( A \) 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. 3a for a (6,3) chiral nanotube, where the real part of the plane wave \( e^{i k_f \cdot r} \) is plotted in the background, and the real part of the complete Bloch function \( \Psi_{k_f}(r) \) 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. 3b. 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 which interfere destructively on \( A \) remain finite on \( B \) because they are superposed with different phases, as is shown in Fig. 3c. As a result of the small difference \( k_f - k_b \) of forward and backward propagating waves the modulus \( |\Psi|^2 \) acquires a very slow modulation on the scale of the CNT length \( L \). There is no non-trivial superposition which would have nodes at both ends for both sublattice components. Thus, the appropriate 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.\(^{17,19}\) 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 \( k_\parallel = \pi/L \). The superposition of forward and backward moving Bloch states with \( \pm \kappa_\parallel \) and the same \( \tau \kappa_\perp \), together with the bipartite boundary conditions, leads to the unusual quantization condition\(^{17,19,22}\)
\[ e^{2i\kappa_\parallel L} = e^{-2i\eta(k)} e^{i\tau \theta} = \frac{\tau \kappa_\perp + i \kappa_\parallel}{\tau \kappa_\perp - i \kappa_\parallel}. \]

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 \( \kappa_\parallel \) depends on \( B_\parallel \).
Figure 4. **Tuning electronic wavefunctions in an axial magnetic field.**  

*a*, The first three solutions of Eq. (3) in the $K'$ and $K$ valley for an arbitrarily chosen chiral nanotube. Both real (continuous lines) and imaginary (dashed lines) solutions exist. At large $\kappa_\perp$ we have $\kappa_\parallel = n\pi/L$ (horizontal grey lines) and the wave functions on each sublattice display nodes at both nanotube ends (half-wave configuration). At $\kappa_\perp = 0$, conversely, $\kappa_\parallel = (n + 1/2)\pi/L$ yielding a node at one end and an antinode at the opposite end (quarter-wave configuration). The number $n$ is used as the corresponding index of $\psi_n^{\tau}$. The shaded part of the reciprocal space ($B_\parallel < 0$) is not explored in the experiment. The values of $\kappa_\parallel$ at $B_\parallel = 0$ are marked by dots. As $B_\parallel$ increases, the wave functions evolve from configurations close to quarter-wave to half-wave. The states in the $K'$ valley cross the $\kappa_\perp = 0$ point, adopting there the precise quarter-wave shape.  

*b*, Calculated amplitude of the longitudinal wave functions $\psi_1^{\tau}(x_\parallel, B_\parallel)$ of the $K'$ (left) and $K$ (right) valley states for a (15,3) chiral CNT with $L = 121$ nm, obtained by numerical diagonalization of the tight-binding Hamiltonian in real space. Black arrowheads mark the value of $|\psi_1^{\tau}|$ on the $B$ sublattice at the left end.
The solutions of Eq. (3) are plotted as coloured lines in Fig. 4a. For comparison, the grey lines running parallel to the $\kappa_\bot$ axis correspond to the familiar solutions for a unipartite lattice with $\kappa_\| = n\pi/L$. The envelope wave function on the $A$ sublattice is also sketched; the $B$ sublattice counterpart is its mirror image. When $\kappa_\|$ is close to a multiple of $\pi/L$ (i.e., for large $B_\|$), 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, i.e., an amplitude maximum, at the corresponding unconstrained end.

Figure 4b shows the calculated wave function amplitudes for the lowest mode ($n = 1$), $|\psi_{1\tau}(x_\|)|$ on the $A$ and $B$ sublattices, of a specific (15,3) CNT with $L = 121$ nm. They were obtained by direct diagonalization of a tight-binding Hamiltonian on finite lattice, with four valence orbitals per atom (for clarity without spin dependence). The shapes follow closely the expectations based on our analysis of the solutions of Eq. (3). Although we assumed hard-wall boundary conditions, our analysis holds also for a range of soft confinement potentials, as discussed in Section II of the Supplementary Material.

**Fading of the differential conductance.** To explain the fading magnetoconductance lines in Figs. 2b and 2c, we account for the $B_\|$-dependence of the longitudinal CNT wave function in our transport calculations. This implies a $B_\|$ dependent tunneling amplitude, given by the overlap between the CNT and lead wave functions in the contact region. In the single-electron tunneling regime of the experiment, tunneling is weak and the tunneling amplitude is to a good approximation determined by the value of the CNT wavefunction at its ends. This yields for the tunnel couplings at the left ($L$) contact

$$\Gamma_{L\mu}(B_\|) = |\alpha_L|^2 \frac{2\pi}{\hbar} |\psi_{B\mu}(x_\| = 0, B_\|)|^2,$$

where $\mu = (n, \tau, \sigma)$ is a collective index accounting for the mode, valley, and spin degrees of freedom, and the factor $\alpha_L$ contains both the square modulus of the lead wave function at the left contact and the lead density of states. Similarly, the tunnel coupling at the right ($R$) contact is obtained by replacing $A \leftrightarrow B$ and $L \leftrightarrow R$. The factors $\alpha_l$ ($l = L, R$) encode a possible contact asymmetry. The differential conductance then follows from a reduced density matrix approach to lowest order in $\Gamma_{l\mu}$, as explained in the Methods section. The magnetoconductance calculated assuming $\alpha_L/\alpha_R = 1/4$ is shown in Fig. 5a. The input parameters for Eqs. (1) and (3) (nanotube radius, length, and $\Delta k_\bot^\tau$) were obtained by fitting the measured position of the spectral lines shown in Fig. 2b,c to the spectrum of
the CNT model Hamiltonian described in the Methods section. The fast disappearance of the $K$ lines is in excellent agreement with the experimental data plotted in Fig. 5b. The suppression of $K'$ lines at high field is also clearly reproduced. Neither of these features is present in a transport calculation with field independent couplings $\Gamma_{\mu\nu}$, see Fig. 2d.

In our calculations a CNT quantum dot with hard wall boundary conditions was assumed. In the experiment, though, we expect that confinement is smooth due to electrostatic gating and the resulting pn-junction, as seen by the strong variation in tunneling couplings in the hole and electron transport regimes, see Fig. S-2 of the Supplementary Material. Hence, we have performed numerical calculations of the evolution of the CNT eigenmodes as a function of $B_\parallel$ for the case of soft confinement, see Sec. II of the Supplementary Material. We found qualitative agreement with the findings of the hard wall confinement calculation. Thus, the tunability of the longitudinal wave function with magnetic field occurs for smooth confinement as well.

Our experiment can be regarded as the inverse of an STM measurement. In scanning tunneling microscopy the spatial profile of atomic or molecular orbitals is obtained by scanning...
the tip position over the sample. In CNT quantum dots, information on the spatial wave-function profile can instead be obtained by tuning an axial magnetic field. More generally, the unusual tunability of the wave function shape with parallel magnetic field will influence all phenomena dependent on the full spatial profile of the electronic states, such as, e.g., electron-phonon coupling or electron-electron interaction. Thus, a parallel magnetic field may turn out to be an even more versatile investigative tool than already acknowledged.

Methods

Device fabrication and measurement. On a highly p-doped silicon substrate with 300 nm thermal surface oxide, 40 nm thick rhenium contact electrodes were deposited via standard electron beam lithography, dc-sputtering and lift-off. Following anisotropic dry etching to deepen the trenches between the electrodes, in an additional lithography step growth catalyst was locally deposited close to the electrodes and CNTs were grown in situ via chemical vapour deposition.23 After the growth, no further wet processing or imaging was performed to fully take advantage of the clean, as-grown macromolecules.24,25

Measurements were performed in an Oxford Instruments TLM dilution refrigerator with rotatable sample holder, at a base temperature of ∼ 30 mK. The gate voltage was applied to the chip substrate, the bias voltage to a source contact; the resulting dc current at the corresponding drain contact was amplified and recorded. The magnetic field direction with respect to the nanotube was calibrated by recording transport spectra at varying angle and identifying the symmetry points.14 The data was recorded using the Lab::Measurement software package.26

Modelling the CNT spectrum. As the basis for our calculation we use a standard tight-binding model of the CNT, taking into account all four valence electrons of the carbon atoms,20,21 with three modifications. This effective Hamiltonian in the neighbourhood of the Dirac points is then given by

$$H_{\text{CNT}}(B_\parallel) = \sum_{m=0,1} \sum_{\tau,\sigma,\kappa_\|} \hbar v_F \left[ \tau \kappa_\perp(B_\parallel) s_x + \kappa_\parallel(B_\parallel) s_y \right]$$

$$+ \tau \sigma \varepsilon_{SO} + \sigma \mu_B B_\parallel + m \Delta_{\alpha\beta} + \tau \mu_x B_\parallel, \quad (5)$$

where $\kappa_\perp/\parallel$ correspond to the momentum measured from the Dirac point in transverse and longitudinal directions, modified by the curvature effects and the spin-orbit coupling according to Eq. (1). The Pauli matrices $s_x, s_y$ act in the sublattice space. The valley index is $\tau = \pm 1$ for the $K/K'$
valley, the spin index $\sigma = \pm 1$ for the spin projection onto the direction of the CNT axis. Spin-orbit coupling contributes to the Hamiltonian through two terms, $\Delta k_{SO}$ (orbital-like in the language of Ref. 18) and $\varepsilon_{SO}$ (Zeeman-like in the same terminology).

Our first modification consists of taking into account the dependence of $\kappa_\parallel$ on the magnetic field through the cross-quantization condition, Eq. (3). The remaining two modifications are the last two terms of the Hamiltonian, Eq. (5), which reflect the particular nature of our device. We are able to consistently fit the positions of the energy levels only by adding a constant contribution $\mu_x$ to the orbital magnetic moment corresponding to 7% of the overall variation of $\mu$ with magnetic field. A more detailed discussion can be found in Section IV of the Supplementary Material.

Finally, a high bias and low resolution measurement of the region near the band gap, shown in Section V of the Supplementary Material, yields a series of excited states with energy spacing of the order of 8 meV. This value is consistent with the finite size quantization in a nanotube with the length of a few hundred nm. Conversely, the sets $\alpha$ and $\beta$ are very closely spaced in energy and very similar in their $B_\parallel$ dependence.

An explanation for this may be that our device is a bundle of two CNTs (which occur in our growth process and have been observed frequently in other processes as well)\textsuperscript{27} with very similar chiralities. The tunnel coupling between the bundle members would then cause the energy states in individual CNTs to hybridize, turning each longitudinal shell into an effective octuplet spread over two tubes. We assume this hybridization to be constant with magnetic field and denote it by $\Delta_{\alpha\beta}$. The index $m$ is $m = 0$ for set $\alpha$ and $m = 1$ for set $\beta$. The breaking of rotational symmetry in both nanotubes, caused by the presence of the neighbour tube, contributes to a valley mixing visible at low magnetic field. At high fields though, where the valleys are already well separated, this effect is negligible. Supporting numerical calculations are presented in Section V of the Supplementary Material.

The numerical values of all parameters in Eq. (5) used in our calculation are listed in Table I. Together with the cross-quantization condition, Eq. (3), they yield the field-dependent electronic wave functions and energy levels.

**Transport calculation.** The amplitudes of the wave functions at the nanotube ends were used to determine the field and valley-dependent tunneling couplings to the leads, $\Gamma_{\mu}(B_\parallel)$. Then a generalized master equation for the reduced density matrix\textsuperscript{28} was set up in a sequential tunneling regime. In the range of gate and bias voltages shown in the main text only states with $N = 0$ and
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**TABLE I. Model parameters.** Numerical values of the parameters used in Eq. (5) in order to reproduce the experimental data.

$N = 1$ are populated. Since tunneling through $N > 1$ states can be neglected, the rate equations for the stationary density matrix $\rho$ acquire a simple form,

$$0 = \sum_l \Gamma^+_{l\mu} \rho_0 - \sum_l \Gamma^-_{l\mu} \rho_\mu,$$

$$0 = -\sum_{l,\mu} \Gamma^+_{l\mu} \rho_0 + \sum_{l,\mu} \Gamma^-_{l\mu} \rho_\mu,$$

where $\rho_0$ is the population of the $N = 0$ state and $\rho_\mu$ are populations of the $N = 1$ states, each with the collective quantum number $\mu = \{n, \tau, \sigma\}$. The rates $\Gamma^\pm_{l\mu}$ are the rates for tunneling into (+) or out of (−) the state $\mu$. They are given by the product of the tunneling couplings defined in Eq. (4) and the Fermi-Dirac distribution at the lead $l$, $\Gamma^+_{l\mu} = \Gamma_{l\mu} f_l(\varepsilon_\mu)$ and $\Gamma^-_{l\mu} = \Gamma_{l\mu} (1 - f_l(\varepsilon_\mu))$, with $\varepsilon_\mu$ the single particle CNT energy. In our case there are eight single particle states to be occupied, with $\tau = \pm 1, \sigma = \pm 1$ and $n = 1$ in the $\alpha$ or $\beta$ shell. The rate equations, completed with the normalization requirement $\rho_0 + \sum_\mu \rho_\mu = 1$, are then solved numerically, and the resulting current in the stationary limit is calculated as

$$I_l = e \sum_\mu \Gamma^+_{l\mu} \rho_0 - \sum_\mu \Gamma^-_{l\mu} \rho_\mu.$$

The derivative of this current with respect to the bias voltage yields the differential conductance. The steady decrease of $\Gamma_{llK\sigma}$ with magnetic field suppresses the $K$ excitation lines, at some point turning them into blocking states with the associated negative differential conductance (NDC),
faintly visible in Fig. 5a though in the experimental data they are drowned by a noisy background. We discuss those NDC features in Section VI of the Supplementary Material, using a minimal model which neglects the shell and spin degrees of freedom and keeps only the valley, together with its wave functions tunable with the magnetic field. In such a model the differential conductance at the bias corresponding to the alignment between the chemical potential of the lead $R$ and the energy of the $K$ state takes the form

$$\frac{dI_R}{dV_{\text{bias}}} \bigg|_{\epsilon_{\text{bias}}=\epsilon_K} = \frac{e}{k_B T} \frac{\alpha_L \alpha_R^2 (\alpha_L + \alpha_R)}{\left[(\alpha_L + 2\alpha_R)^2 - 2\alpha_R^2\right]} \times \left(- (\alpha_L + \alpha_R)|\psi_K| + \alpha_L|\psi_K'|\right),$$

where the dependence of the differential conductance on $|\psi_K|$ and $|\psi_K'|$ at the contacts is explicit. As already stated above, the factors $\alpha_l$ contain the density of the lead states at the contacts and encode possible asymmetry between the coupling of the quantum dot to the left and right lead.

Numerical calculations were performed using the Armadillo library.\textsuperscript{29}

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**Author contributions**

D. R. S. fabricated the device with support by P. L. S., and performed the measurements with support by P. L. S. and A. D. The theoretical analysis and modelling was implemented by M. M. The experimental side was supervised by A. K. H. and C. S., the theoretical side by M. G. The manuscript was written jointly by M. M., A. K. H., M. G., and C. S.

**Additional information**

Supplementary information is available in the online version of the paper. It includes a comparison with measurements in a perpendicular magnetic field, a more detailed discussion of the phenomenological $\mu_x$ correction, the calculation of wave functions in soft confinement as well as a discussion of the model for shells $\alpha$ and $\beta$. 
Competing financial interests

The authors declare no competing financial interests.

Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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J. Cao, Q. Wang, and H. Dai. Electron transport in very clean, as-grown suspended carbon


