Spin-orbit interaction in chiral carbon nanotubes probed in pulsed magnetic fields

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The magneto-conductance of an open carbon nanotube (CNT)-quantum wire was measured in pulsed magnetic fields. At low temperatures we find a peculiar split magneto-conductance peak close to the charge neutrality point. Our analysis of the data reveals that this splitting is intimately connected to the spin-orbit interaction and the tube chirality. Band structure calculations suggest that the current in the peak regions is highly spin-polarized, which calls for application in future CNT-based spintronic devices.

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A source of spin-polarized electrons is one of the important building blocks of a future spin-based electronics [1]. Very high degrees of polarization can potentially be achieved by exploiting spin-orbit interaction (SOI) [2, 3]. Based on the low atomic number Z = 6 of carbon the spin-orbit interaction in carbon nanotubes (CNTs) was mostly believed to be very weak, until a recent experiment [4] has demonstrated the effect of spin-orbit interaction in clean CNT quantum dots. Evidence for the spin-orbit splitting in simple magneto-conductance (MC) measurements has not yet been reported.

In this Letter, we present MC data for the complementary situation of an open CNT-quantum wire obtained in pulsed magnetic fields. In a parallel magnetic field \(B_\parallel\), a small band-gap CNT evolves via a metallic state into a semiconducting one, resulting in a typical peak in the MC [5–7]. In one of our tubes, however, we observed a splitting of this MC-peak into two peaks at low temperature. Recording MC-traces at different \(V_g\) shows that the splitting vanishes when moving away from the charge neutrality point (CNP). A thorough comparison to band structure calculations reveals that the splitting is explained by the SOI, which becomes strong for small tube diameters. An interesting implication of our analysis is the prediction of a highly spin-polarized current in the peak regions.

The experiments have been performed on devices made of individual CNTs prepared on Si/SiO\(_2\)/Si\(_3\)N\(_4\) substrates. The heavily p-doped Si was used as a back gate and the thickness of the insulating layer was 350 nm. CNTs were grown by means of a chemical vapor deposition method [7] and Pd (50 nm) electrodes were defined on top of the tubes by e-beam lithography. In order to exclude strain effects on the band structure [8], only straight and long (\(\sim 50 \mu m\)) CNTs were selected for devices and the distance between two Pd electrodes was \(\sim 500 \mu m\). The dc magneto-conductance was studied in pulsed magnetic fields of up to 60 T, applied parallel to the tube axis. The accuracy of the alignment was \(\sim \pm 5^\circ\) (See EPAPS for further experimental details).

Figure 1 shows the magneto-conductance \(G(B_\parallel)\) of a small-bandgap CNT device located near the CNP (diameter \(d \sim 1.5 \text{ nm}\)). At 82 K, the conductance \(G\) of the tube initially increases to reach a maximum at \(B_0 = 5.9\ T\), then it exponentially drops to zero at higher fields due to the Aharonov-Bohm (AB) effect [5–9]. Interestingly, when the device was cooled down to 4.2 K, the conductance maximum \(G_{\text{max}}\) at \(B_0 = 3.1\ T\) and \(B = 11.1\ T\). We note that these peaks are symmetric with respect to the conductance dip at \(B_0^* \approx 7\ T\), and \(G(B_0^*)\) is similar in magnitude to \(G(B_\parallel = 0)\). The key to the explanation of the data lies in the magnetic field dependence of the one-dimensional band structure.

A specific CNT is uniquely labeled by the chiral indices \((n, m)\), which define the chiral angle \(\theta\) and the quantized values of the transversal wavevector \(k_\perp\) [10]. The values of \(k_\perp\), combined with the graphene dispersion cones, determine the quasi-one-dimensional band structure of the CNTs. A given CNT is metallic if the lines of allowed energies cross the Dirac points \(K, K'\); otherwise it is semiconducting. For nominally metallic CNTs \((n – m = 3l,\) with \(l\) an integer), the dispersion relation \(E(k_\parallel)\) near the Dirac points reads [4–11, 14]:

\[
E(k_\parallel) = \pm \hbar v_F \sqrt{k_\parallel^2 + k_\perp^2} + \left( \frac{q}{2} \mu_B B_\parallel + \tau \varepsilon_{SO} \right) \sigma,
\]

where \(k_\parallel\) is the wave vector parallel to the tube axis, \(v_F\) the Fermi velocity, \(\frac{q}{2} \mu_B B_\parallel \sigma\) being the Zeeman term with \(\sigma = \pm 1\) for spin parallel/antiparallel to the tube axis, and \(\tau = \pm 1\) for the \(K\) and \(K'\) Dirac points. The transversal wave vector \(k_\perp\) contains three distinct contributions, which are discussed below.

The Aharonov-Bohm flux \(\phi_{AB} = B_\parallel \pi d^2 / 4\) results in a shift \(k_\parallel = (2/d)(\phi_{AB}/\phi_0)\) of \(k_\parallel\), where \(\phi_0 = \hbar/e\) is the flux quantum. Therefore, one can convert a metallic CNT into a semiconducting one, or vice versa, by tuning
the allowed values of $k_\perp$ with a magnetic field parallel to the tube axis [15,16,17].

In addition, curvature [15] affects the allowed values of $k_\perp$ and induces small band gaps in nominally metallic CNTs. The curvature-induced shift [15,16] $k_\perp^0 = -\sigma a_0 \cos(3\theta)/(2d)^2$ of the allowed $k$-states results in a band gap $E_{\text{curv}} = 2\hbar v_F |k_\perp^0|$ at $B || = 0$, where $a_0$ is the C–C bond length.

A second consequence of curvature is a spin-dependent shift

$$k_{SO} = -\sigma (2/d) (\phi_{SO}/\phi_0),$$

of $k_\perp$ by the spin orbit interaction [4,11,4,20,21] which removes the four-fold spin and $K, K'\perp$-degeneracy in favor of two Kramers doublets corresponding to parallel and antiparallel alignment of orbital and spin magnetic moments. This SOI-induced shift in $k_\perp$ is equivalent to the presence of an AB flux $\phi_{SO} \approx 10^{-3} \phi_0$ [4,11], and produces a spin-orbit energy splitting $\Delta_{SO} = 2\hbar v_F |\delta_{SO}|$. For a CNT with $d \sim 1$ nm, $\phi_{SO}$ corresponds to $\approx 5$ T, while $\phi_0$ is $\approx 5000$ T.

In contrast, the term with $\varepsilon_{SO} = -\delta \cos(3\theta)/d$, added to the root in the Eq. [4] (like the Zeeman term), solely shifts the energy but not $k_\perp$, leading to an asymmetric spin-orbit energy splitting for the hole ($\Delta_{SO} + 2\varepsilon_{SO}$) and the electron band ($\Delta_{SO} - 2\varepsilon_{SO}$) of chiral metallic tubes [13,14]. As $\varepsilon_{SO}$ contains the factor $\cos(3\theta)$, it is small for near armchair tubes. The parameter $\delta$ ranges from 0.3-0.7 nm meV [13,14].

The resulting evolution of the band structure in magnetic field is visualized in Fig. 1. At zero field the band gap $E_g^0 = E_{\text{curv}} - \Delta_{SO}$ is reduced by the SOI. With the application of $B_{\parallel}$, the two spin sub-bands separated by the SOI cross the corner point of the Brillouin zone (either at $K$ or $K'$), thus explaining two subsequent MC-peaks at $B_1$ and $B_2$. In between, a conductance dip appears at $B_0$ when the spin sub-bands are located symmetrically around the corner point. If the Zeeman-like terms in Eq. [4] are neglected the energy gap has a local maximum at $k_{AB} = -k_\perp^0$ corresponding to $E_g(B_0^*) \approx \Delta_{SO}$. The distance between the two peaks, $\Delta B = (4/\pi d^2) \Delta \phi_{AB}$, is determined by $\Delta \phi_{AB} = 2 \phi_{SO}$ (the factor 2 comes from $\sigma = \pm 1$). For the observed values of $\Delta B = 8$ T, and $d = 1.5$ nm we find

$$\phi_{SO} = \frac{\pi d^2 \Delta B}{8} \approx 1.7 \times 10^{-3} \phi_0. \quad (3)$$

For a conservative confidence interval of $\pm 0.5$ nm for $d$ determined with an atomic force microscope one obtains $0.76 < 10^3 \phi_{SO}/\phi_0 < 3$ compatible with previous studies [4,11,20]. Eqs. [1], [2] and [3] result in the energy splitting $\Delta_{SO}$ at $B_1 = 0$ (assuming $k_\parallel = 0$)

$$\Delta_{SO} = \frac{4\hbar v_F \phi_{SO}}{d} \approx 2.5 \pm 0.8 \text{ meV}. \quad (4)$$

This value corresponds to $\sim 30$ K and explains the disappearance of the double-peak structure and the single conductance maximum at $B_{\parallel} = 0 \approx B_0^*$ for the 82 K trace of Fig. 1. Because $\Delta_{SO}$ is inversely proportional to the diameter, it becomes large for small-diameter tubes [22].

With further increase of $\phi_{AB}$ the energy gap $E_g$ linearly opens again as both orbital sub-bands gradually move away from the corner points of the Brillouin zone. The exponential decrease of $G$ at high fields (the inset of Fig. 1b) is thus explained by charge carriers thermally activated over the magnetic-field-induced band gap, as described by previous authors [5,23]. Since the inset in Fig. 1 suggests that the conductance depends exponentially on the band gaps $E_g^0$ and $E_g(B_0^*)$, they dominate the conductance at $B_1 = 0$ and $B_{\parallel} = B_0^*$. Hence, the approximate equality $G(0) \approx G(B_0^*)$ inferred from Fig. 1a.
suggests the following relation between the two energy scales $E_{\text{curv}}$ and $\Delta_{\text{SO}}$:

$$\frac{E^0_0}{E^0_0(B_0')} = \frac{E_{\text{curv}} - \Delta_{\text{SO}}}{\Delta_{\text{SO}}} \approx 1. \quad (5)$$

We now turn to the discussion of the effect of tube chirality. The strong dependence of $B_0$ and $B_0'$ on the chirality can be used to identify the chiral indices of small-bandgap CNTs [5]. Out of 53 small-bandgap CNTs with $d = 1.5\pm0.5$ nm, only five tubes [(12,9), (13,10), (16,10), (18,9) and (19,10)] display values of $B_0 \approx 5-8$ T compatible with our data, while $B_0$ can take much larger values for other CNTs, e.g., the (12,3) and (17,2) tubes.

Table I lists values of $\phi_{\text{SO}}$ and $\Delta_{\text{SO}}$ for these chiralities, calculated from Eqs. [3] and [4] and the observed $\Delta B = 8$ T. The $\phi_{\text{SO}}$ of the (12,9) tube is closest to $\phi_{\text{SO}} \approx 10^{-3}\phi_0$, predicted in Ref. [11] and measured in Ref. [4]. When we further take into account the condition $E_{\text{curv}} \approx 2\Delta_{\text{SO}}$ for the CNT measured (Eq. [3]), we realize that the (12,9) and (18,9) tubes satisfy this constraint best.

Taking the (12,9) tube with the chiral angle $\theta = 25.3^\circ$ (close to the armchair configuration) as the most probable candidate, we calculated the density of states (DOS) in a parallel magnetic field. For comparison, we show the DOS of a (17,2) tube, which has almost the same diameter but a very different chiral angle $\theta = 5.5^\circ$ close to the zigzag-configuration.

From Fig. 2 it becomes apparent that in an applied magnetic field the band edges change with four distinct slopes away from the two Kramers doublets both in the electron and hole bands, reflecting the orbital and Zeeman splitting. The DOS calculated for the (12,9) tube explains the evolution of the magneto-conductance very well. The band gap is closed at $B_1$ by the spin-down and subsequently at $B_2$ by the spin-up sub-band, in very good agreement with the observed double-peaks at the CNP. The calculated energy gaps at zero-field and at $B_0'$ agree with Eqs. [4] and [5]. On the other hand, the DOS calculated for the (17,2) CNT predicts a significantly reduced intermediate gap region in Fig. 2c, in spite of almost the same $d$ and $\Delta_{\text{SO}}$, when compared with the (12,9) tube. The (17,2) tube has a much larger curvature-induced gap $E_{\text{curv}} \approx 18.5$ meV, resulting in a much higher $B_0 \approx 31.6$ T. Because the spin-orbit gap competes with the Zeeman splitting, the peak splitting in

| $(n,m)$ | $d$ (nm) | $\theta$ ($^\circ$) | $E_{\text{curv}}$ (meV) | $B_0, B_0'$ (T) | $\phi_{\text{SO}}$ ($10^{-3}\phi_0$) | $\Delta_{\text{SO}}$ (meV) |
|-------|--------|-------|----------------|-------------|----------------|----------------|
| (12,9) | 1.43 | 25.3 | 4.6 | 7.8 | 1.54 | 2.36 |
| (13,10) | 1.56 | 25.7 | 3.5 | 5.5 | 1.84 | 2.58 |
| (16,10) | 1.78 | 22.4 | 4.7 | 6.4 | 2.39 | 2.94 |
| (18,9) | 1.86 | 19.1 | 6.0 | 7.8 | 2.63 | 3.08 |
| (19,10) | 2.00 | 19.8 | 4.9 | 5.9 | 3.02 | 3.30 |
| (12,3) | 1.08 | 10.9 | 28.1 | 63.1 | 0.88 | 1.78 |
| (17,2) | 1.42 | 5.5 | 18.5 | 31.6 | 1.51 | 2.33 |
orders of magnitude. The insert shows field-effect-transistor with on-off conductance ratio of several 103. Conductance is around 3 e2/h, close to the theoretical limit of 4e2/h. This shows that our device is in the ballistic regime, where the conductance is determined by the number of available sub-bands with an average transmission probability of ~ 0.8. At B∥ = 0, the hole conductance is around 3e2/h and diminishes down to ~ 0.5e2/h as EF is tuned towards the CNP (Vg ∼ +6 V). While the magneto-conductance is initially positive at low fields, it becomes negative at high fields (B∥ ≫ B0) for all gate voltages, indicating the growth of E∥ due to the AB effect. At B∥ > 30 T, the gate characteristic G(Vg) exhibits the behavior of a p-type CNT field-effect-transistor with an on-off conductance ratio > 104. The double-peak structure is pronounced only in the vicinity of the CNP (+5.6 V ≤ Vg∗ ≤ +6.4 V). Two additional G(B∥) curves, presented in Fig. 3b, show that the two peaks merge again into one as EF is shifted to the electron side across the CNP.

In conclusion, we have investigated single walled carbon nanotubes up to very high magnetic fields. The magneto-conductance of a quasi-metallic tube shows a peculiar double peak, which can be explained in terms of spin split conduction bands, separated by a strong spin-orbit interaction, which exceeds the Zeeman splitting. Our finding may open the path towards the application of CNTs as highly efficient ballistic spin filters.

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