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Gate-Dependent Orbital Magnetic Moments in Carbon Nanotubes

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We investigate how the orbital magnetic moments of electron and hole states in a carbon nanotube quantum dot depend on the number of carriers on the dot. Low temperature transport measurements are carried out in a setup where the device can be rotated in an applied magnetic field, thus enabling accurate alignment with the nanotube axis. The field dependence of the level structure is measured by excited state spectroscopy and excellent correspondence with a single-particle calculation is found. In agreement with band structure calculations we find a decrease of the orbital magnetic moment with increasing electron or hole occupation of the dot, with a scale given by the band gap of the nanotube.

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The response of an electron in a quantum dot to an applied magnetic field is determined by the coupling to the electron spin—the Zeeman effect—and the coupling to the orbital magnetic moment of the electron. In carbon nanotubes the electrons encircle the circumference of the tube with a resulting magnetic moment pointing along the nanotube axis which thus couples to the parallel component of an applied magnetic field. This was first studied by Minot et al. [1], for the first few carriers in small band-gap nanotubes where the resulting orbital g factor, $g_{\text{orb}}$, was shown to reflect the nanotube diameter. Recently, the surprising discovery of a strong spin-orbit interaction in nanotubes [2–10] has spurred renewed interest in the use of carbon nanotubes as templates for spin qubits addressable by electric fields [11,12]. One new possibility is to control the spin-orbit magnetic field and the parallel component of an applied magnetic field by moving the quantum dot along a curved nanotube segment [13].

This calls for a better understanding of the relation between the nanotube band structure and the inherited properties of nanotube quantum dots. Here we extend the work of Ref. [1] by investigating the dependence of $g_{\text{orb}}$ on the electron or hole occupation of a nanotube quantum dot tuned by a potential $V_g$ on a nearby electrostatic gate. We find $g_{\text{orb}}$ decreasing with electron or hole filling as the circumferential velocity of the electrons decrease and we show that while the nanotube diameter determines the value of $g_{\text{orb}}$ for the first carrier, the nanotube band gap $\Delta_g$ sets the scale of its gate dependence.

It is well established that the low energy electron dispersion for nanotubes can be obtained from the graphene dispersion by imposing periodic boundary conditions in the circumferential direction of the nanotube [14]. Ignoring spin-orbit effects this leads to $E(k) = \pm \hbar v_F \sqrt{k^2 + k_{\parallel}^2}$, where $v_F \approx 10^6$ m/s is the Fermi velocity of graphene, $k_{\parallel}$ is the component of the wave vector along the nanotube axis and the offset $k_0$ between the quantization lines and the Dirac points of the graphene dispersion, results in a band gap $\Delta_g = \hbar v_F k_0$ for the nanotube [15]. A magnetic field $B_{||}$ applied parallel to the nanotube axis adds an Aharonov-Bohm phase to the electron wave function and shifts the circumferential quantization lines by $k_{\Phi} = eB_{||} D / 4 \hbar$ where $D$ is the nanotube diameter. Letting $s = \pm 1$ denote the spin $\uparrow, \downarrow$ along the nanotube and $\tau = \pm 1$ the corner points $K, K'$ of the graphene Brillouin zone, the dispersion reads

$$E_{\tau s} = \pm \hbar v_F \sqrt{(\tau k_\Phi - k_0)^2 + k_{\parallel}^2} + \frac{1}{2} g_s \mu_B B,$$

where $g_s = 2$ and the last term accounts for the usual Zeeman effect. Including explicitly the angle $\theta$ between the nanotube axis and the applied field and expanding to linear order in $B$ leads to [16]

$$E_{\tau s} = E_0^\pm + \left( \frac{1}{2} g_s \mu_B \mp g_{\text{orb}} \cos(\theta) \right) \mu_B B,$$

with $E_0^\pm = \pm \sqrt{\Delta_g^2 + \epsilon_N^2}$ and

$$g_{\text{orb}} = \frac{e v_F D}{4 \mu_B \sqrt{1 + (\frac{\epsilon_N}{\Delta_g})^2}}.$$

Here the result has been written for a nanotube quantum dot with electrons confined to a nanotube segment of length $L$ where $\epsilon_N = \hbar v_F N \pi / L$. Experimentally verifying Eq. (3) is the main objective of this work. For the first carriers $\epsilon_N \ll \Delta_g$, yielding $g_{\text{orb}} = e v_F D / 4 \mu_B$ which is the value classically expected for an electron in a circular motion of diameter $D$ and speed $v_F$ [1]. For increasing electron or hole occupation of the dot (larger $\epsilon_N$) $g_{\text{orb}}$ decreases as shown in Fig. 1(a) with a characteristic scale set by the band gap $\Delta_g$. This behavior arises from the graphene dispersion as schematically illustrated in the inset and in Fig. 1(b): Because of the linear dispersion, $v_F$ does not depend on energy (black arrows) and thus the
and spin magnetic moments) and disorder scattering (which favors parallel or antiparallel alignment of orbital split by a combination of spin-orbit coupling $C_1$ SO)

Subsequently palladium/gold ($C_1$ highly doped silicon capped with an insulating oxide. single-wall carbon nanotubes are grown by chemical vapor deposition from catalyst islands predefined on substrates of

device preparation. Figure 3(a) shows the measured transconductance $dI/dV_g$ as a function of applied voltage $V_g$ revealing a series of peaks characteristic of a quantum dot in the Coulomb blockade regime [21]. In the valleys of low conductance the number of electrons on the quantum dot $n$ is fixed and with increasing $V_g$ a peak emerges when the next charge state $(n + 1)$ becomes available for transport. The peak separations, which are extracted in Fig. 2(c), measure the energy, $E_{\text{add}}$, required for adding the next electron. This energy is the combination of the constant electrostatic charging energy and the energy spacing of the quantum levels of the dot. As seen in the figure, $E_{\text{add}}$ is fourfold periodic reflecting the near fourfold degenerate level structure discussed above. In the following, we present the details of the measurements of $g_{\text{orb}}$ in the highlighted quartet.

Figure 3(a) shows the measured transconductance $dI/dV_g$ as a function of applied bias $V_{\text{sd}}$ and $V_g$ of the shaded quartet in Figs. 2(c) and 2(d) corresponding to $n_0 \approx 120$ electrons occupying the quantum dot. The diamond shaped zero conductance regions are characteristic for a quantum dot in the Coulomb blockade regime.
with Fig. 2(a) the diamond edge in Fig. 3(a) has split into two doublets (see Supplemental Material [23]). Clearly, $g_{\text{orb}}$ decreases as electrons or holes are added to the conduction or valence band confirming the decrease of the orbital magnetic moment for states further away from the band edge as predicted by Eq. (3). This is the main result of our work. The solid line shows a fit to Eq. (3) with the diameter $D$ and $\Delta g$ as the free parameters [24]. The fit is in reasonable agreement with the measurement and yields a band gap of $23 \text{ meV}$ in good agreement with the gap estimate of $\sim 15 \text{ meV}$ from the measured stability diagram (see Supplemental Material [23]). The fit, however, yields a diameter of $D = 5.3 \text{ nm}$, which is unrealistically large for nanotubes grown by chemical vapor deposition which are expected to have $D \lesssim 3 \text{ nm}$. Unexpectedly large diameters are also inferred in other spectroscopic studies [1,2,25,26]; e.g., Kuemmeth et al. found a diameter of $D \approx 5 \text{ nm}$ [2]. Yet other reports find more reasonable
i.e., our model assumes a constant length of the quantum dot, velocity. For flat graphene various explanations for this finding. First, as seen from experimentally determined diameters and band gaps, e.g., combined with Raman spectroscopy [33].

In this work, $g_{orb}$ was measured for all charge states in the region indicated by the horizontal error bar with a spread in values indicated by the vertical error bar. The line is a fit to Eq. (3) assuming a constant dot length and with $\epsilon_N(V_g) \propto (V_g - 0.8 \text{ V})$ yielding $D = 5.3 \text{ nm}$ and $\Delta_{g} = 23 \text{ meV}$.

FIG. 4 (color online). $g_{orb}$ and corresponding orbital magnetic moments $\mu_{orb} = g_{orb} \mu_B$ as a function of gate, showing a decrease with electron or hole filling. Values are extracted from excited state spectroscopy ($\bigcirc$) and Coulomb peak dependence on $B_\parallel$ (□). Close to the band gap a clear fourfold periodicity is absent impeding direct comparison to the model. Instead $g_{orb}$ was measured for all charge states in the region indicated by the horizontal error bar with a spread in values indicated by the vertical error bar. The line is a fit to Eq. (3) assuming a constant dot length and with $\epsilon_N(V_g) \propto (V_g - 0.8 \text{ V})$ yielding $D = 5.3 \text{ nm}$ and $\Delta_{g} = 23 \text{ meV}$.

In conclusion, we have investigated the gate dependence of the orbital magnetic moment of quantum states in high-quality carbon nanotube quantum dots. We present low temperature transport measurements of the dependence of quantum state energies on the angle between the nanotube axis and an applied magnetic field. This allows the determination of the nanotube axis from transport measurements alone and an accurate value for the orbital $g$ factor is found by comparison with a single-particle model taking into account both disorder-scattering and spin-orbit interaction. Repeating such measurement over a wide range of gate voltages we find that the orbital magnetic moment decreases with dot occupation in agreement with the expectations from band structure considerations.

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15. In this work, $\Delta_{g}$ refers to a curvature induced band gap in a nominally metallic nanotube, but in the case of a semiconducting nanotube, the dominating band gap $E_{g}$ due to a periodic boundary condition around the nanotube circumference should be taken into account.
16. Including the spin-orbit effect adds a term $s \tau \Delta_{SO}/2$ with the effective spin-orbit coupling $\Delta_{SO} = 2(\Delta_{SO}^0 + \frac{\hbar \Delta_{g}}{\sqrt{1+(2\tau)^2}})$, where $\Delta_{g}$ and $\Delta_{SO}$ are spin-orbit parameters related to the band structure.
[22] In Figs. 3(b)–3(d), an overall shift of the gate-position of the diamond has been subtracted to reveal the relative shifts of the excited states with respect to the ground state.
[24] From the measured 8 shells per 1 V gate voltage and an average level spacing of 3.1 meV measured at large filling when \( \Delta E = h v_F \pi / L = \epsilon_N / N \) we estimate \( \epsilon_N \approx (25 \text{ meV}/V) V_g \).