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Su(4) mixed valence regime in carbon nanotube quantum dots

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Abstract

We study the evolution of conductance regimes in carbon nanotubes with doubly degenerate orbitals (“shells”) by controlling the contact transparency within the same sample. For sufficiently open contacts, Kondo behavior is observed for 1, 2, and 3 electrons in the topmost shell. As the contacts are opened more, the sample enters the “mixed valence” regime, where different charge states are strongly hybridized by electron tunneling. Here, the conductance as a function of gate voltage shows pronounced modulations with a period of four electrons, and all single-electron features are washed away at low temperature. We successfully describe this behavior by a simple formula with no fitting parameters. Finally, we find a surprisingly small energy scale that controls the temperature evolution of conductance and the tunneling density of states in the mixed valence regime.

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We study the evolution from the Kondo to the mixed valence regime by controlling the contact transparency within the same semiconducting carbon nanotube. The quantized orbitals originating in two electronic subbands of nanotubes are doubly degenerate, forming four-electron “shells” (see Ref. [1]). At low enough temperatures and sufficiently open contacts, the Kondo behavior develops in the valleys with 1, 2, and 3 electrons in the topmost shell [2,3]. The Kondo effect in this situation is expected to obey the SU(4) symmetry [4–6]. As the contacts are opened even more, so that the individual charge states are no longer well defined, we observe the mixed valence behavior throughout the entire shell. In this regime, the four single-electron conduction peaks in a shell, visible at high temperature, merge at low temperatures into a single broad maximum. We successfully describe the conductance dependence on gate voltage (V_{gate}) by a simple formula (1), which we argue has deep implications. The characteristic temperature and energy scale in the mixed valence regime are found to be surprisingly small.

The nanotubes are grown by the recipe of Ref. [7] and contacted by Cr/Au electrodes. The middle section of small-gap semiconducting nanotubes fills with electrons at positive V_{gate} (Fig. 1a). The parts of the nanotube adjacent to the electrodes stay p-type (forming “leads”). Thereby, a quantum dot is formed *within* the nanotube, defined by p–n and n–p junctions. It is important for the observation of the SU(4) symmetry that the “leads” to the dot are formed within the same nanotube, and thus have the same orbital symmetry [5]. The width of the p–n junctions in the nanotube depends on V_{gate}: larger V_{gate} make the junctions narrower (Fig. 1a). We therefore can vary the junctions transparency by changing V_{gate}. Indeed, the widths of the single-electron peaks in Fig. 1 grows with V_{gate} (from shell I to shell IV), indicating the growing lifetime broadening of the levels, Γ.

The conductance in the 1, 2, and 3e valleys (shells I an II, Fig. 1b) grows at lower temperatures, resulting in the gradual disappearance of the single-electron peaks. In particular, this Kondo behavior is observed in the 2e valleys, indicating a degenerate ground state. The energy splittings between the six possible states of two electrons in a shell are small [1]. Hence even for a moderate broadening

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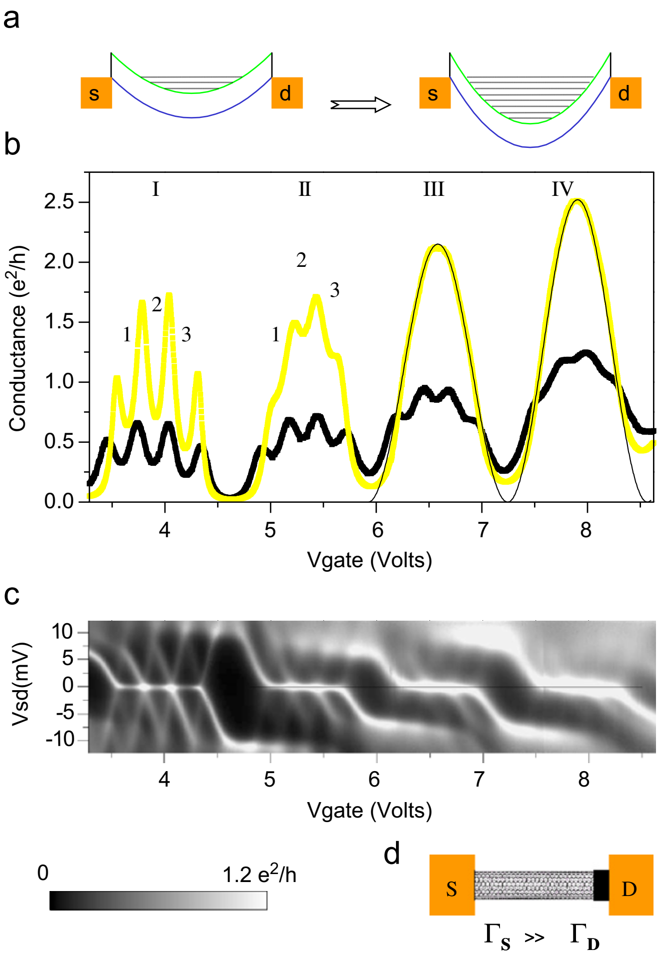

Fig. 1. (a) Band structure of a semiconducting nanotube, which can sustain either electron or hole populations, depending on V_{gate}. In the n-doped regime, p–n junctions are formed near the contacts, creating in a nanotube. As V_{gate} grows, the tunneling barriers become narrower and more transparent, increasing the level broadening Γ. (b) Differential conductance measured as a function of V_{gate} at 1.3 K (top, yellow/gray symbols) and 15.0 K (bottom, black symbols). Four shells are shown (I–IV). The numbers (1–3) indicate the number of electrons in a shell. The Kondo effect enhances the conductance in the valleys in each shell at low temperatures. At 1.3 K, the single-electron conductance peaks in shells III and IV merge to form smooth oscillations with a four-electron periodicity. Solid line: formula (1). (c) Differential conductance map (same range as in (b) as a function of V_{gate} and source–drain voltage (scale of the colormap: $0-1.2e^2/h$; $T = 1.3$ K. (b) can be viewed as a horizontal cross-section of (c). The outlines of the "Coulomb diamonds" are visible, most clearly in shell I. The Kondo ridge is formed close to zero source–drain voltage in all three diamonds. Going from shell I to shell IV, the Kondo ridge becomes the most prominent feature of the data. (d) One of the contacts is better coupled to the nanotube than the other, which results in up–down asymmetry of the image in (c). One can consider the weakly coupled contact as a tunneling probe, which measures the density of states in the system made of the nanotube and the strongly coupled contact.

Γ, these states are effectively degenerate and can all participate in the formation of the Kondo resonance.

The lifetime broadening Γ ranges from ~7 meV in shell I to ~15 meV in shell IV. This broadening should be compared to the charging energy of $E_C \approx 10$ meV and the shell spacing of $\Delta \approx 10$ meV (extracted from Fig. 1c). Since $\Gamma \sim E_C$, the charge fluctuations should be significant, in particular in shells III and IV. Thus, the mixed valence regime should cover the entire V_{gate} range in these shells. Experimentally, the growth of conductance in the valleys completely washes away the single-electron peaks at the lower temperatures in shells III and IV (Fig. 1b). However, the unitary limit plateaus, observed for a well-developed Kondo effect [8], are absent. Instead, we find deep oscillations with a periodicity of four electrons. No single-electron features are visible in this regime.

We have successfully described the low temperature conductance in shells III and IV as (dotted line in Fig. 1b):

$$G \sim \sin^2[\pi C_{\text{gate}}(V_{\text{gate}} - V_{\text{gate}}^{(0)})/4]. \quad (1)$$

Here $N = C_{\text{gate}}(V_{\text{gate}} - V_{\text{gate}}^{(0)})$ is the number of electrons on the topmost shell, which changes linearly with V_{gate}. Kubo's formula predicts conductance in the form of $G = G_0\sum_i \sin^2(\delta_i)$ [9], where δ_i are the scattering phase of the four modes ($i = 1-4$, corresponding to two spin projections and two subbands). Friedel's sum rule yields $\sum_i \delta_i = \pi N$. Assuming that all δ_i are equal to each other (which is not obvious *a priori*), we obtain $\delta_i = \pi N/4$. The conductance then becomes $G = 4G_0 \sin^2(\pi N/4)$. From the nonlinear transport measurement (Fig. 1c), we can estimate [3] the ratio of the contact transparencies $\Gamma_S/\Gamma_D \sim 4$ for shell IV. We then expect the maximal conductance of $4e^2/h 4\Gamma_S\Gamma_D/(\Gamma_S + \Gamma_D)^2 \approx 2.5e^2/h$, in accord with the experiment.

The map of conductance as a function of the source–drain voltage (V_{SD}) and V_{gate} (same range as in Fig. 1b) is shown in Fig. 1c. The zero bias Kondo ridge [10], forms across the three valleys of shell I [2,3], and further grows in shells II–IV. In this sample, one of the contacts is weaker coupled to the nanotube than the other, thus forming a tunneling probe. The conductance is enhanced whenever the Fermi level of the probe is aligned with a resonance in the density of states of the nanotube strongly hybridized with the other contact. In shells III and IV, as V_{gate} increases, a single resonance descends to the Fermi energy, dwells there as the zero-bias ridge and then continues its descent. For one electron in a shell, the SU(4) Kondo peak in the spectral density of states is expected to be shifted above the Fermi level by an energy of the order of the Kondo temperature (i.e. by about its width). For two electrons (two holes), the resonance should be centered at the Fermi energy, and for three electrons (one hole) the resonance should move below the Fermi energy. As a result, the zero bias ridge acquires a small, but noticeable, slope (Fig. 1c).

The half-width of the Kondo ridge (2 meV) measured across the center of shell IV (Fig. 1c, $V_{\text{gate}} = 7.8$ V) agrees well with the characteristic temperature ($T_0 \sim 10$ K) estimated from Fig. 1b. The relation between T_0 and other energy parameters is not precisely known in the SU(4) mixed valence regime; however, it seems surprising that the measured T_0 is ~10 times smaller than Γ, Δ and E_C. Explaining the small energy scale T_0 in the mixed valence regime remains a challenge.

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References

[1] A. Makarovski, et al., Phys. Rev. B 74 (2006) 155431.
[2] W.J. Liang, et al., Phys. Rev. Lett. 88 (2002) 126801.
[3] B. Babic, et al., Phys. Rev. B 70 (2004) 235419;
B. Babic, et al., Phys. Rev. B 70 (2004) 195408.
[4] L. Borda, et al., Phys. Rev. Lett. 90 (2003) 026602;
G. Zarand, et al., Solid State Commun. 126 (2003) 463;
R. Lopez, et al., Phys. Rev. B 71 (2005) 115312.
[5] M.S. Choi, et al., Phys. Rev. Lett. 95 (2005) 067204.
[6] P. Jarillo-Herrero, et al., Nature 434 (2005) 484;
P. Jarillo-Herrero, et al., Phys. Rev. Lett. 94 (2005) 156802.
[7] B. Zheng, et al., Nano Letters 2 (2002) 895.
[8] W.G. van der Wiel, et al., Science 289 (2000) 2105.
[9] M. Pustilnik, L. Glazman, J. Phys. Cond. Matt. 16 (2004) R513.
[10] D. Goldhaber-Gordon, et al., Nature 391 (1998) 156;
S.M. Cronenwett, et al., Science 281 (1998) 540;
J. Schmid, et al., Physica B 258 (1998) 182.