Thermal Symmetry Crossover and Universal Behaviors in Carbon Nanotube Dots

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Motivated by recent experiments on electronic transport through a carbon nanotube dot, we investigate the role of the intra- and inter-orbital Coulomb interactions on the temperature evolution of the conductance. It is shown that small amount ($\lesssim 10\%$) of asymmetry between these Coulomb repulsions substantially deforms the conductance profile at finite temperature, particularly around half-filling. The nature of such thermal symmetry crossover is elucidated.

KEYWORDS: carbon nanotube, Coulomb interaction, quantum dot, Kondo effect, universality, Anderson impurity model

In the last decade, electronic transport through semiconducting nanostructures has been extensively studied for future applications of quantum nano-devices. As for a single quantum dot, we have now understood fairly well that Coulomb interaction on the dot fundamentally affects its transport. It not only constrains electrons to pass through a dot (the Coulomb blockade phenomena[1]) but also gives rise to the singular conductance enhancement at low-temperatures in an odd electron number on the dot (the Kondo effect[2]). A prominent feature of the latter phenomena is a universal temperature dependence scaled by the Kondo temperature, where low-temperature conductance quantitatively agrees with the "universal curve" predicted theoretically by the Anderson impurity model.[3]

Recent experimental observations of the Kondo effect in carbon nanotube (CNT) dots[4–6] have rekindled interest in a role of strong interaction in two important aspects: enlarged symmetry and apparently nonuniversal temperature behavior. While the standard Kondo effect occurs through a spin one-half degenerate single level, the enlarged symmetry may augment the role of the spin degree. Carbon nanotube dots arguably have almost doubly degenerate orbits in the topmost shell, producing a variety of low-temperature phenomena.[7–14] At a quarter filling $N_d \approx 1$, the entanglement between spin and orbital degrees of freedom gives rise to the $SU(4)$ Kondo effect, with the Kondo temperature one order of magnitude higher than the standard case.[15] What is more intriguing is the temperature evolution around an even valley $N_d \approx 2$. While the experiment by Makarovski *et al.* has observed visible low-temperature "Kondo-like enhancement" around $N_d = 2$,[6] such enhancement was absent in others.[4,5] Rather, a characteristic 'dip' is observed in the conductance profile around half-filling ($V_g \sim 3$V) (See Fig. 3(a) or Supplementary Information Fig. SI2 of Jarillo-Herrero *et al.*[5] and compare Fig. 2 (a) of Makarovski *et al.*[6]). A few possibilities of the nature at half-

filling have been argued actively so far, due to the singlet-triplet Kondo effect,[5] the $SU(4)$ Kondo effect[6,16,17] with/without some more complications. Observed temperature dependence of conductance through CNT seems nonuniversal rather than universal.

In this Letter, motivated by the mentioned experiments on low-temperature transport through a CNT dot, we investigate the role of the inter-orbital Coulomb repulsion and the thermal symmetry crossover. Our main concern is on the temperature dependence of the linear conductance profile particularly around half-filling. While the RG flow either at quarter-filling or at half-filling has been shown to flow toward the $SU(4)$ symmetric strong coupling point,[14] the absence of the exact $SU(4)$ symmetry may affect substantially the behavior at finite temperature. Indeed a small amount of interaction difference less than 10% will cause a large deformation to the conductance profile at finite temperature (see Figs. 3 and 4 later). We point out that the thermal crossover by interaction asymmetry provides a systematic understanding to observed thermal evolutions.[4–6]

Relatively large energy scale of the Coulomb interaction allows us to focus on the topmost electron shell. Regarding a CNT dot, a great success of Anders *et al.* explaining Makarovski *et al.* experiments makes us feel certain that the $SU(4)$-symmetric model is a good starting point to model a CNT dot system; the topmost shell of CNT doubly degenerates in orbits $i = 1, 2$ and the view conforms to observations of $SU(4)$-Kondo effect at quarter-filling.[15] The $SU(4)$-symmetric model nevertheless misses something, failing to explain the temperature evolution at half-filling of some experiments.[4,5] Within the universality of the topmost shell, we regard the interaction asymmetry among the orbits a clue.

By this reasoning, we model the CNT dot by an orbitally degenerate Anderson model with interaction asymmetry among orbits. On the dot, an electron interacts with an electron in the same orbit $i = 1, 2$ by U or in the different orbit by U'. The total Hamiltonian is given by $H = H_D + H_L + H_T$, where the dot H_D, the noninteracting leads H_L, and the coupling between leads and the dot H_T are defined by

$$H_D = \sum_{i\sigma} \left(\varepsilon_d \, \hat{n}_{i\sigma} + U \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}\right) + U' \hat{n}_1 \hat{n}_2, \tag{1}$$

$$H_L = \sum_{\alpha} \sum_{ki\sigma} \varepsilon_k \, c^{\dagger}_{\alpha ki\sigma} c_{\alpha ki\sigma}, \tag{2}$$

$$H_T = \sum_{\alpha} \sum_{ki\sigma} \left(t_k c^{\dagger}_{\alpha ki\sigma} d_{i\sigma} + \text{h.c.} \right). \tag{3}$$

Here the number operator of the dot is defined by $\hat{n}_i = \sum_\sigma \hat{n}_{i\sigma} = \sum_\sigma d^{\dagger}_{i\sigma} d_{i\sigma}$ and the average electron number on the CNT dot $N_d = \langle \sum_i \hat{n}_i \rangle$ is controlled from 0 to 4 by the gate voltage ε_d. In the calculation below, we assume constant density of states ρ_α of the lead α and use $\Gamma = \sum_\alpha \Gamma_\alpha = \sum_\alpha \pi \rho_\alpha |t_k|^2$ as a coupling parameter between the leads and the dot.

In the case of $U' = U$, the total Hamiltonian H retains the full $SU(4)$ symmetry. The states

$(n_1, n_2) = (1, 0)$ and $(0, 1)$ are degenerate at quarter-filling ($N_d = 1$) and so are $(n_1, n_2) = (2, 0)$, $(1, 1)$ and $(0, 2)$ at half-filling ($N_d = 2$). When one breaks the $SU(4)$ symmetry by decreasing U', the degeneracy is broken at half-filling, but unbroken at quarter-filling. This simple argument indicates that the $SU(4)$ symmetry at half-filling is more vulnerable than that at quarter-filling. We will show later that this is indeed the case.

At $T = 0$, the system is fully renormalized into the strong coupling fixed point; the linear conductance is expected to depend only on N_d, *i.e.*, $G(T = 0) = 4G_0 \sin^2\left(\frac{\pi}{4}N_d\right)$ (with $G_0 = e^2/h$) according to the Friedel sum rule. Such behavior has been confirmed in experiments.[6] Since N_d changes only slightly by decreasing the asymmetric parameter $\eta = U'/U$, the zero-temperature conductance is universal and the effect of asymmetry $\eta < 1$ emerges only at finite temperature.

Our analytical approach is based on an extension of the Kotliar-Ruckenstein formulation of slave-boson mean field theory (KR-SBMT),[18] where a bosonic field is attached to each type of local excitations rather than decoupling the charge and spin degrees of freedom. The approach has several advantages that other slave-boson cousins miss: it retains finite Coulomb repulsion effect and reproduces Fermi liquid behavior at $T = 0$ with satisfying the Friedel sum rule. The KR formulation of SBMT is believed to be a powerful non-perturbative method; it gives reliable results not only qualitatively but also quantitatively up to the Kondo temperature, agreeing successfully with numerical renormalization group methods and experiments.[19,20]

When KR-SBMT is extended to the dot with doubly degenerate orbits, 16 bose fields are needed associated to each state of the dot: e for the empty, $p_{i\sigma}$ for one electron with orbit i and spin σ, x_i for two electrons on the same orbit i, $y_{s\sigma}$ for two electrons at different orbits with total spin (s, σ), $h_{i\sigma}$ for three electrons with a hole on $i\sigma$, and b for fully occupied state.[21,22] To eliminate unphysical states, the completeness condition and the correspondence condition between boson and fermion number $n_{i\sigma} = Q_{i\sigma} = p^\dagger_{i\sigma}p_{i\sigma} + x^\dagger_i x_i + y^\dagger_{1\sigma}y_{1\sigma} + \frac{1}{2}(y^\dagger_{00}y_{00} + y^\dagger_{10}y_{10}) + h^\dagger_{i\bar{\sigma}}h_{i\bar{\sigma}} + \sum_{\sigma'} h^\dagger_{\bar{i}\sigma'}h_{\bar{i}\sigma'} + b^\dagger b$ are imposed in terms of Lagrange multipliers $\lambda_{i\sigma}$. By applying the mean field approximation, all the boson fields are replaced by the expectation values. On determining these auxiliary parameters self-consistently at each temperature and each gate voltage, the system reduces to the renormalized resonant level model with the effective dot level $\tilde{\varepsilon}_d$ and the effective hopping $\tilde{t}_k = z_{i\sigma}t_k$,

$$H_{\rm eff} = H_L + \sum_i \tilde{\varepsilon}_d\, \hat{n}_i + \sum_{\alpha k i\sigma}\left(\tilde{t}_k\, c^\dagger_{\alpha k i\sigma}d_{i\sigma} + \text{h.c.}\right). \tag{4}$$

Here $z_{i\sigma} = (1 - Q_{i\sigma})^{-\frac{1}{2}}[e^\dagger p_{i\sigma} + p^\dagger_{i\bar{\sigma}}x_i + p^\dagger_{\bar{i}\sigma}y_{1\sigma} + p^\dagger_{\bar{i}\bar{\sigma}}(y_{00} + y_{10})/2 + x^\dagger_{\bar{i}}h_{i\bar{\sigma}} + (y^\dagger_{00} + y^\dagger_{10})h_{\bar{i}\bar{\sigma}}/2 + y^\dagger_{1\bar{\sigma}}h_{\bar{i}\sigma} + h^\dagger_{i\sigma}b]Q^{-\frac{1}{2}}_{i\sigma}$. The effective Hamiltonian eq. (4) conforms to the Fermi liquid description.[23] The form of $H_{\rm eff}$ enables us to find the linear/nonlinear conductance by the Meir-Wingreen formula.[24] We will present the result of the linear conductance for the symmetric coupling between the leads and the dot $\Gamma_L = \Gamma_R$ below.

The effective Hamiltonian eq. (4) defines a natural energy scale, the characteristic temperature

$$T^* = \sqrt{\tilde{\varepsilon}_d^2 + \tilde{\Gamma}^2}\Big|_{T=0} . \tag{5}$$

where $\tilde{\Gamma} = \sum_\alpha \pi\rho_\alpha|\tilde{t}_k|^2$ is the effective coupling between the dot and the leads. Interaction effect is encoded in terms of $\tilde{\varepsilon}_d$ and $\tilde{\Gamma}$. Note that the scale T^* is defined in the entire range of the gate voltage and it reduces to the usual Kondo temperature at the Kondo valley $N_d = 1$. We evaluate renormalized parameters self-consistently at each ε_d and at temperature T.

Figure 1 demonstrates typical temperature evolutions of the conductance profile as a function of the gate voltage with increasing asymmetry (a) $U'/U = 1.0$, (b) 0.8, (c) 0.6, and (d) 0.4, respectively. Even with a small asymmetry $U'/U = 0.8$, a characteristic dip structure at half-filling develops clearly at finite temperature, while only small deformation is seen around quarter and three-quarter fillings. Overall widths of conductance peaks are determined by $U + 2U'$. Here the conductance profile at $T = 0$ can be considered universal, fully recovering $SU(4)$ symmetry in the sense that it is determined by the Friedel sum rule. Interaction asymmetry manifests itself only at finite temperature as a thermal crossover by modifying the conductance profile substantially around half-filling.

To clarify the nature of the temperature dependence and understand how the conductance is affected by the asymmetric interaction particularly around $N_d \approx 2$, we make a direct comparison between the behaviors at quarter-filling and half-filling. Figure 2 (a,b) shows the temperature dependence of the conductance at (a) $N_d = 2$ and (b) $N_d = 1$ for $U'/U = 1$, 0.8, 0.6 and 0.4. By decreasing U' away from $U' = U$, it is clear that decreasing U' reduces the characteristic energy scale at $N_d = 2$ but increases it at $N_d = 1$. The tendency is elucidated by estimating the characteristic temperature T^* defined in eq. (5); while T^* at $N_d = 2$ is already lower than that at $N_d = 1$ at $U' = U$, the difference of T^* becomes more amplified by the presence of interaction asymmetry (Fig. 2 (c)). This behavior for $N_d = 2$ agrees well with other numerical results.[13,16] Clearly, the conductance enhancement around $N_d \approx 2$ is destroyed more rapidly than at $N_d \approx 1$ at finite temperature, which induces a substantially large deformation in the conductance profile. At the filling corresponding to Coulomb blockade peaks ($N_d \approx 1.5, 2.5$), T^* is found almost unchanged by U'/U.

Another important observation is on the universal temperature dependence. Insets of Fig. 2 (a,b) show the data as a function of scaled temperature T/T^*. As is seen, the curves collapse well up to $T \lesssim T^*$ (T^* is a upper scale restricting the validity of the present analysis). It implies that the universal temperature dependence either at half-filling or at quarter-filling reduces to the $SU(4)$ symmetric case of $U' = U$. Additionally, Fig. 2 (d) shows that such universal dependence differs slightly but significantly between $N_d = 1, 2$.[17] It shows that the thermal symmetry crossover observed at finite temperature is *not* considered a crossover between *different universality classes* (between $SU(4)$ and $SU(2) \times SU(2)$). We can attribute

the phenomena to renormalizing the characteristic temperature T^* by asymmetry U'/U at each gate voltage.

To stress the relevance of asymmetric interaction U and U' to experimentally observed conductance profiles,[4–6] we now present schematic calculations mimicking experimental situation, changing U'/U slightly. In the comparison, we have in mind a rough estimate $U \approx 100$K, but we find the following characteristics pretty generic.

Figure 3 demonstrates the conductance profile of $U'/U = 0.997$ with $U = 30\Gamma$ ($U = 15\Gamma$ for the inset). With this small amount of asymmetry, the profile reproduces all the features of the $SU(4)$ symmetric model.[17] A distinctive feature of the conductance profile in large U/Γ region is that the Kondo enhancement by decreasing the temperature occurs either at $N_d \approx 2$ or at $N_d \approx 1$ similarly. Regarding a smaller value of U/Γ, four peaks merge to form one big peak showing the Kondo enhancement (see the inset). As was claimed already,[17] these temperature evolutions by the $SU(4)$ symmetric Anderson model agree very well with what is observed either at $V_g \sim 3.9$V or at $V_g \sim 5.3$V in Makarovski *et al.*[6] When we resort to the (heuristic) prescription in choosing U/Γ,[25] our results agree very well to those by the NRG results by Anders *et al.*. Detailed comparison will be presented elsewhere.

Figure 4 illustrates the temperature evolution of the conductance profile with all the same parameters with Fig. 3 except for $U'/U = 0.9$, whose value is deduced from the Coulomb blockade peak spacings at $T = 8$K.[5] The conductance profiles regarding a smaller U/Γ (the inset) are almost identical with the symmetric case. To our surprise, however, the conductance profiles with four peaks ($U = 30\Gamma$) is modified considerably by this relatively small asymmetry (less than 10%), having a characteristic dip structure around half-filling. It is noted that in Fig. 3(a) or Fig. SI2 of Jarillo-Herreo *et al.*,[5] the conductance at $V_g \approx 3$V is smaller than that of $V_g \approx 2.8$V or 3.2V at each temperature. We claim that Fig. 4 captures well essential characteristics of experiments[4,5] with a reasonable choice of parameters.

How much asymmetry brings the system away from the $SU(4)$ symmetric behavior of Fig. 3 into that of Fig. 4? The characteristic temperature T^* at half-filling (of the symmetric model) controls it; with $T^* = 1.4 \times 10^{-2}U$ in Figs. 3 and 4, $|U - U'| \lesssim T^*$ is valid in the former but $|U - U'| \gg T^*$ in the latter. This difference affects the conductance profile at finite temperature.

To understand experimental data fully, one more complication seems to remain. By decreasing the temperature below 8K, one conductance profile at half-filling ($V_g \approx 3$V) begins to enhance, conforming to the Kondo-like effect, but it eventually reduces below 2K.[5] We regard such behavior beyond our scope of description with a possible extra mechanism at an energy scale much smaller than the Kondo temperature; some interesting possibilities such as a slight difference of degenerate levels, exchange coupling and orbital-dependent coupling have been argued, but it still remains to be seen.

In conclusion, we have investigated the role of the inter- and intra-orbital interactions in transport through a carbon nanotube dot. By using the KR formulation of slave-boson mean field theory, we have shown that a small amount of asymmetry between the intra-orbital and inter-orbital interactions can give rise to a substantial effect on the conductance profile at finite temperature by renormalizing T^*. It is suggested that interaction asymmetry at finite temperature enables us to understand systematically the existing experimental data.[4–6] We also anticipate to observe a similar crossover phenomena by applying small amount of finite bias voltage because it should serve as an energy cut-off similarly to finite temperature.

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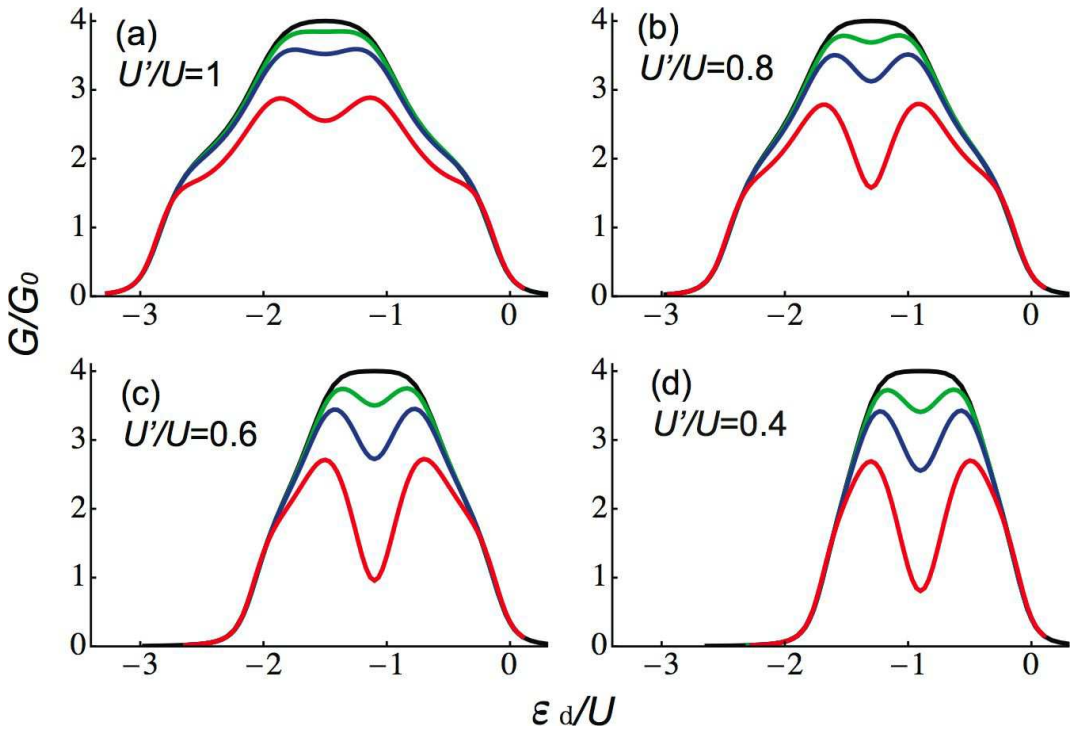

Fig. 1. (Color online) Temperature evolution of the conductance as a function of the gate voltage with varying asymmetry $U'/U = 1.0$, 0.8, 0.6, and 0.4. Temperatures are $T/U = 0$ (black), 1.67×10^{-3} (green), 3.33×10^{-3} (blue), and 8.33×10^{-3} (red). U is set to $U = 30\Gamma$.

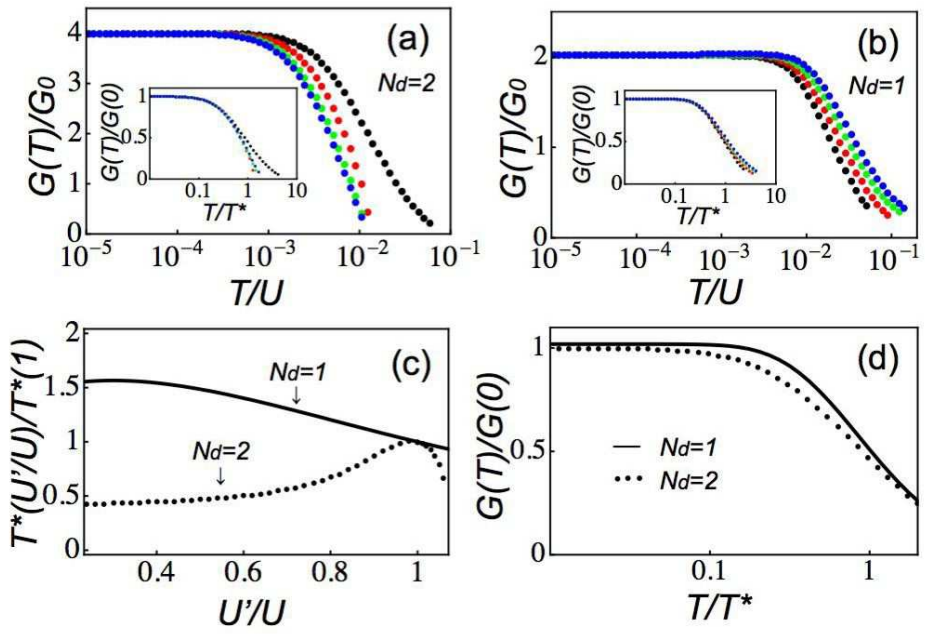

Fig. 2. (Color online) Temperature dependence of the conductance (a) at $N_d = 2$ and (b) $N_d = 1$ by varying $U'/U = 1$ (black), 0.8 (red), 0.6 (green) and 0.4 (blue). Other parameters are the same with Fig. 1. (c) T^* as a function of interaction asymmetry U'/U. (d) Universal temperature dependence at $N_d = 1$ (solid) and at $N_d = 2$ (dotted) for $U'/U = 1.0$.

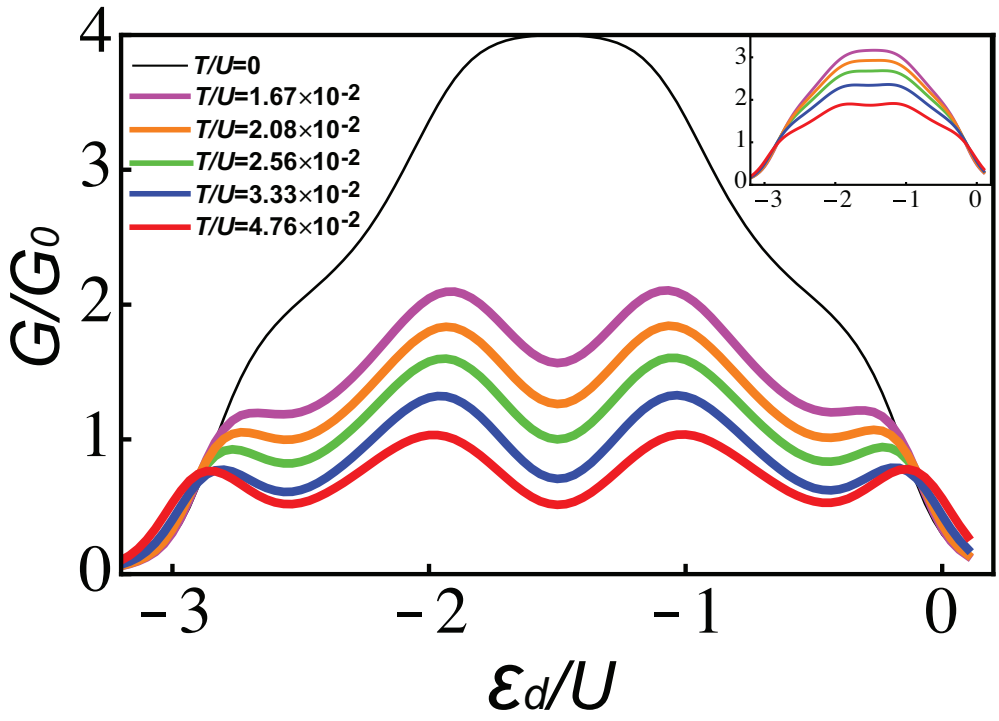

Fig. 3. (Color online) Schematic plotting for the comparison with CNT experiments:[6] Conductance as a function of the gate voltage for $|U - U'| \ll T^*$, where $U = 30\Gamma$ and $U'/U = 0.997$. Inset shows larger coupling parameter regime $U = 15\Gamma$.

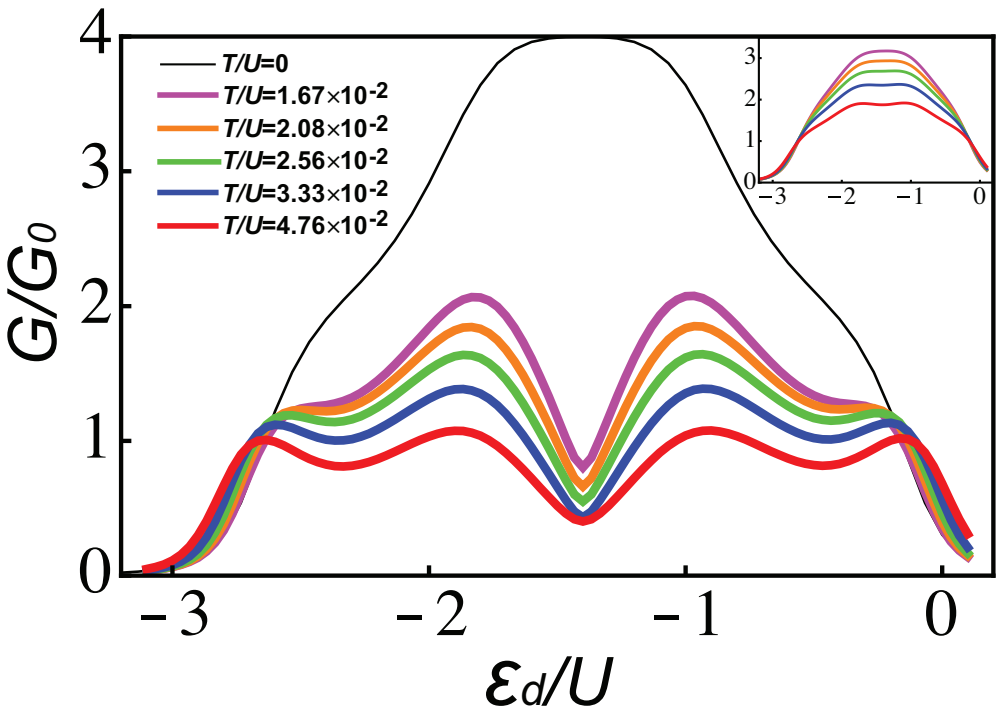

Fig. 4. (Color online) Schematic plotting for the comparison with CNT experiments:[5] Conductance as a function of the gate voltage for $|U - U'| \gg T^*$. Parameters are the same with Fig. 3 except for $U'/U = 0.9$.

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22) We use different symbols from Dong *et al.*[21] to emphasize the importance of the interaction asymmetry: $(e, p_{i\sigma}, d_{0i}, d_{1s}, t_{i\sigma}, f) \to (e, p_{i\sigma}, x_i, y_{s\sigma}, h_{i\sigma}, b)$.
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25) In the single-impurity Anderson model, it has been known[20] that quantitative agreement can be attained between effective theories (the KR-SBMT or the functional RG) and the NRG method only if we choose the twice as large value of U/Γ for the former. It is a heuristic prescription.