

Abrupt disappearance and re-emergence of the SU(4) and SU(2) Kondo effects due to population inversion

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The interplay of almost degenerate levels in quantum dots and molecular junctions with possibly different couplings to the reservoirs has led to many observable phenomena, such as the Fano effect, transmission phase slips, and the SU(4) Kondo effect. Here we predict a dramatic repeated disappearance and re-emergence of the SU(4) and anomalous SU(2) Kondo effects with increasing gate voltage. This phenomenon is attributed to the level occupation switching which has been previously invoked to explain the universal transmission phase slips in the conductance through a quantum dot. We use analytical arguments and numerical renormalization group calculations to explain the observations and discuss their experimental relevance and dependence on the physical parameters.

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The coexistence of spin-degenerate levels with different couplings to the leads is ubiquitous in quantum dots (QDs), quantum wires, and molecular junctions. It has been pointed out early on that such coexistence may develop in deformed QDs [1,2], with important consequences on the relation between the conductance and the transmission phase of consecutive Coulomb-blockade (CB) peaks. Later on, it has been demonstrated [3] that such a coexistence is actually a generic effect in interacting QDs. In fact, the interplay between levels with weak coupling to the leads and a strongly coupled one has been invoked [2–9] to explain the intriguing experimental observations [10–12] of sharp drops in the transmission phase through a QD between CB peaks. These drops have been attributed to “level occupation switching” (LOS)—the abrupt emptying of the strongly coupled level and the filling of a corresponding weakly coupled level, or vice versa, as the gate voltage is continuously varied. These studies have been further supported by a direct observation [13] of the Fano effect, resulting from the interference between a wide and a narrow level in a single quantum dot. Simultaneous transport through several molecular levels has also been demonstrated [14] in molecular junctions, and the interplay of weakly and strongly coupled levels has been predicted [15] to lead to observable effects in the CB peak structures.

In a seemingly different context, the coexistence of almost degenerate levels has been argued [16–22] to give rise to SU(4) Kondo physics [23], which has indeed been observed in carbon nanotubes [24–26], in atoms [27], and in single [28] and double [29,30] semiconductor quantum dots. In all these systems, the degenerate levels are not necessarily coupled equally to the leads [31,32]. However, in spite of the plethora of studies of the physics of LOS in QDs on one hand, and of SU(4) Kondo physics in such systems on the other hand, the interplay of these two effects has not been addressed so far. In this paper, we predict dramatic abrupt suppression and re-entrance of the Kondo effect due to LOS. We present numerical renormalization group (NRG) calculations, backed up by analytical arguments, and show that in the presence of two spin-degenerate levels, with very different couplings to the leads, then as the gate voltage is varied (Fig. 1), the enhanced conductance due to the Kondo effect is abruptly

suppressed, only to likewise abruptly re-emerge at higher gate voltages. This disappearance and re-emergence may occur more than once. Below, we elaborate on the physics behind this effect, on its dependence on temperature, the ratio of the couplings of the two levels to the leads, on their energy difference and other physical parameters.

The Hamiltonian that describes the two-level QD is given by

$$H_{\text{QD}} = \sum_{i\sigma} \epsilon_i \hat{n}_{i\sigma} + \sum_i U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + U_{12} \hat{n}_1 \hat{n}_2, \quad (1)$$

where $i = 1, 2$ denotes the level index, $\hat{n}_{i\sigma} = d_{i\sigma}^\dagger d_{i\sigma}$, $\hat{n}_i = \sum_{\sigma} \hat{n}_{i\sigma}$ ($d_{i\sigma}^\dagger$ creates an electron on the dot in level i with spin σ), and spin degeneracy has been assumed (i.e., no magnetic field). We will first concentrate on the fourfold degenerate case, $\epsilon_i = \epsilon$ and $U_1 = U_2 = U_{12} = U$. Each one of the levels couples to a different linear combination of states in the leads, which, for simplicity, we assume to be orthogonal. The resulting Hamiltonian is then given by

$$H = H_{\text{QD}} + \sum_{i\sigma k \in L, R} \epsilon_{ik} c_{i\sigma k}^\dagger c_{i\sigma k} + \sum_{i\sigma k \in L, R} (V_{ik} d_{i\sigma}^\dagger c_{i\sigma k} + \text{H.c.}), \quad (2)$$

where $c_{i\sigma k}^\dagger$ creates an electron with spin σ in the left (L) or right (R) lead in momentum state k that couples to level i in the dot. Again, for simplicity, the tunneling amplitude is chosen to be momentum (and spin) independent, but different between the two levels, $V_{ik} = V_i$. The expectation values and the spectral function, required for the calculation of the conductance [33], are calculated using a density matrix numerical renormalization group (DM-NRG) procedure.¹ We assume equal couplings to the left and right leads, $\Gamma_i = \pi \rho V_i^2$, and equal and constant density of states ρ in the two leads, with a symmetric band of bandwidth D around the Fermi energy. In the following, we set D to be the unit of energy.

¹We used the open-access Budapest Flexible DM-NRG code, <http://www.phy.bme.hu/~dmnrg/>; O. Legeza, C. P. Moca, A. I. Toth, I. Weymann, and G. Zarand, [arXiv:0809.3143](https://arxiv.org/abs/0809.3143) (unpublished).

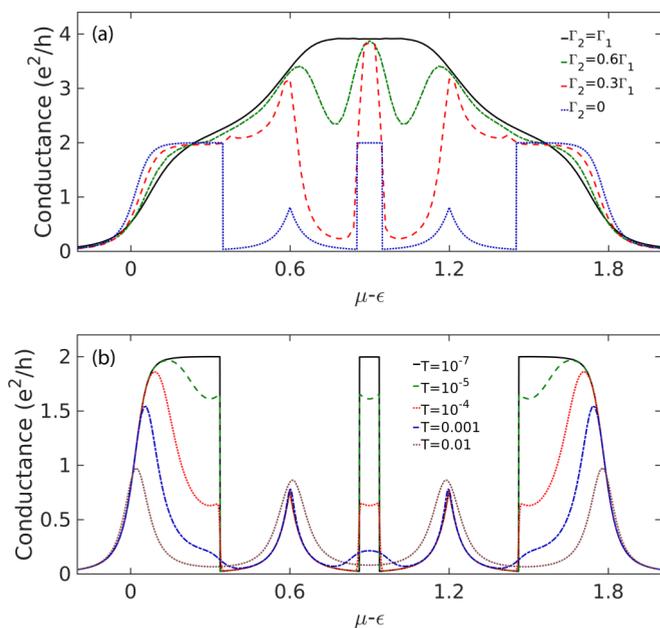


FIG. 1. Conductance through a two-level quantum dot as a function of chemical potential for (a) different ratios of the tunneling amplitudes Γ_2/Γ_1 , with $T = 10^{-7}$ and (b) different temperatures, for $\Gamma_2 = 0$. For both plots, $\Gamma_1 = 0.03$ and $U = 0.6$ (in units of the bandwidth). In both panels, level population switching events lead to abrupt disappearance and re-emergence of the Kondo effect.

Figure 1(a) depicts the conductance as a function of the chemical potential (gate voltage), for different values of Γ_2/Γ_1 . Each Γ_i defines an effective SU(2) Kondo temperature $T_K^{(i)}$. When $\Gamma_2 = \Gamma_1$, one reproduces the standard SU(4)-symmetric Anderson model conductance plot: the conductance G rises from zero to $G = 2e^2/h$ (where e is the electron charge and h the Planck constant), then to $G = 4e^2/h$, in agreement with the Friedel sum rule, $G = e^2/h \sum_{i\sigma} \sin^2(\pi n_{i\sigma})$ (where $n_{i\sigma} = \langle \hat{n}_{i\sigma} \rangle$), which is accurate at such low temperatures. However, when Γ_2 is reduced, LOS starts to take place, resulting in several conductance dips near the mid point, as can be seen for the curves with $\Gamma_2 \neq \Gamma_1$ in Fig. 1(a).

The effect is even more dramatic for smaller Γ_2 where $T_K^{(2)} < T$. In this regime, the conductance rises and plateaus at its Kondo value, $G = 2e^2/h$, only to drop sharply to almost zero at a specific value of the chemical potential, slightly above $\mu = \epsilon + U/2$.² Then the conductance remains at zero, goes through a narrow peak (around $\mu = \epsilon + U$), of a peculiar shape (see below), and eventually rises sharply again to around the Kondo value below the mid-point $\mu = \epsilon + 3U/2$. Since the model is symmetric around that point, the same behavior is reflected around $\mu = \epsilon + 3U/2$.

²It is interesting to note that resemblant conductance plots have been numerically obtained in a different model, where the orbital states in a carbon nanotube are mixed while tunneling to the orbital states in the leads [40]. That model, which is most likely experimentally inaccessible, can be mapped in some regime to the Hamiltonian studied here, but the occupation oscillations found in that paper were differently interpreted.

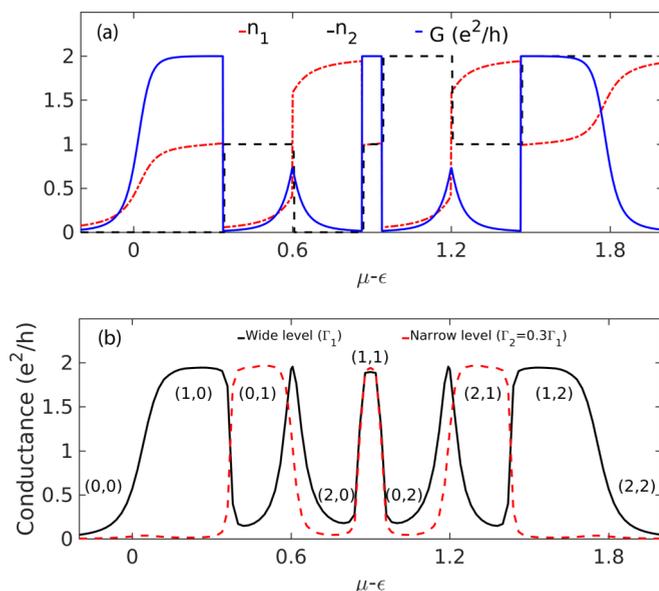


FIG. 2. (a) The occupation in each level (n_1, n_2) and the conductance G , calculated using the Friedel sum rule, as a function of chemical potential. Here, $\Gamma_1 = 0.03$, $\Gamma_2 = 0$, $T = 10^{-7}$, and $U = 0.6$. (b) The separate contributions to the conductance from the wide (with coupling Γ_1) and the narrow (Γ_2) levels for $\Gamma_2/\Gamma_1 = 0.3$, demonstrating a transition between the respective Kondo effects. The occupations for the wide and narrow levels, rounded to a representative integer, are written in parenthesis in each regime.

Figure 1(b) depicts how this effect depends on temperature, for the case $\Gamma_2 = 0$. At high temperature, $T \gg T_K^{(1)}$, one reproduces the CB peak structure. Note that in spite of only one level being coupled to the leads, there are 4 CB peaks, all of similar width, indicating that in each case transport is through the strongly coupled level [3]. However, for smaller temperatures, switching events lead to enhancement of the conductance by the Kondo effect, but only in specific regions of the chemical potential, giving rise to the sharp drops in the conductance mentioned above. This peculiar behavior of the conductance can be understood by combining the Friedel sum rule, with the physics of LOS. Figure 2(a) depicts the occupations of the two levels, for $\Gamma_2 = 0$, at the lowest temperature of Fig. 1(b), and the resulting conductance, using the Friedel sum rule for the $\Gamma_2 = 0$ case: $G = e^2/h \sum_{\sigma} \sin^2(\pi n_{1\sigma})$.

The physics of LOS is relatively well understood [9]. Consider, for example, the limit of $\Gamma_2 = 0$. When μ lies between the first two CB peaks, there is a competition between two configurations: the partially occupied wide level and the fully occupied narrow level. Due to tunneling (Γ_1), the energy of the former is reduced by an electron process (e-process)—tunneling of the electron in that level to the leads, and by a hole process (h-process)—tunneling of an electron from the leads into the dot, making it doubly occupied. For small width, the energy of this configuration is $E_1 = \epsilon - \frac{\Gamma}{2\pi} [\ln(\frac{D}{\epsilon}) + \ln(\frac{D}{\epsilon+U})]$. On the other hand, the second configuration energy is reduced by lead electrons of either spin tunneling into the empty wide level, i.e., twice the h-process, $E_0 = -2 \times \frac{\Gamma}{2\pi} [\ln(\frac{D}{\epsilon+U})]$. As μ crosses the symmetry point $\mu = \epsilon + U/2$, the reduction in energy due to the h-process is larger than that of the e-process.

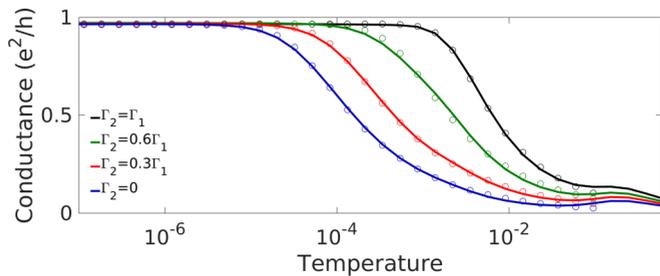


FIG. 3. Logarithmic temperature dependence of the conductance for the curves in Fig. 1(a), at $\mu - \epsilon = 0.2264$, depicting the change in the Kondo temperature from its SU(4) value to its SU(2) one. The circles on each plot are the corresponding phenomenological fit functions (see text). For the curves from right to left, the fitting parameters were $(n, s) = \{(3, 0.2), (2, 0.25), (2, 0.22), (2, 0.23)\}$ and the Kondo scales $T_k = \{68, 21, 5, 2\} \times 10^{-4}$.

As a result of this, it is eventually energetically favorable to occupy the narrow level instead of the wide level, and there is a LOS event. As can be seen in Fig. 2(a), there are several more switching events for similar reasons. As electrical current flows mainly through the strongly coupled level, a sudden switch in its occupation will lead to an abrupt change in the conductance, in accordance with the Friedel sum rule. Indeed, given the occupations, the conductance calculated using the Friedel sum rule [Fig. 2(a)] agrees perfectly with the direct calculation of the conductance (Fig. 1). Similar arguments can be applied to the regime where transport occurs through both levels, e.g., the curve $\Gamma_2 = 0.3\Gamma_1$ in Fig. 1, where both Kondo temperatures obey $T_K^{(i)} \gg T$. In this case, when the occupations switch from around $(n_1, n_2) = (1, 0)$ to around $(0, 1)$, there is a switch from the Kondo effect due to level 1 to that due to level 2 [Fig. 2(b)], resulting in a small hump in the conductance, visible in Fig. 1(a). On the other hand, when the occupations switch from $(2, 0)$ to $(1, 1)$, there is an abrupt jump in the conductance from almost zero to the coexisting Kondo value of $G = 4e^2/h$.

Interestingly, Fig. 1(a) demonstrates that one can induce, by changing the ratio Γ_2/Γ_1 , a crossover from SU(4) to SU(2) Kondo physics. Figure 3 depicts the conductance as a function of temperature for the values of Γ_2 , which were used in Fig. 1(a). In this figure, the crossover between SU(4) physics to SU(2) physics with decreasing Γ_2/Γ_1 is manifested in the reduction of T_K from $T_K^{\text{SU}(4)}$ to $T_K^{\text{SU}(2)}$ and the corresponding change in the slope. To study this crossover in more detail, we employ a phenomenological fitting function $G = G_0[1 + (2^{1/s} - 1)(\frac{T}{T_K})^n]^{-s}$, where T_K , s and n are phenomenological fitting parameters. This function has been shown [30,34] to accurately approximate the numerical renormalization calculations, with $s \simeq 0.22$ and $n = 2$ for the SU(2) case, and $s \simeq 0.20$ and $n = 3$ for the SU(4) case. Thus fitting the numerical data to this function (Fig. 3) allows direct estimate of the Kondo temperature T_K and the slope n . Employing this procedure, we indeed find that the parameter n changes from $n = 3$ to $n = 2$ as Γ_2/Γ_1 decreases. Moreover, we can infer the Kondo scales to be $T_K = (68, 21, 5, 2) \times 10^{-4}$, respectively, for the four curves of decreasing Γ_2/Γ_1 . Equating these values to the expression for the Kondo temperature

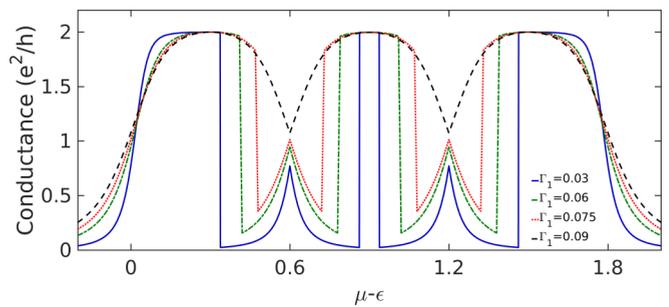


FIG. 4. Conductance as a function of chemical potential for various Γ_1 , with $\Gamma_2 = 0$, $T = 10^{-7}$ and $U = 0.6$. The larger $\Gamma_1 = 0.09$ curve exhibit three wide Kondo peaks.

for the SU(N) symmetric case $T_K^{\text{SU}(N)} \propto D(N\rho J)^{1/N} e^{-1/N\rho J}$ [35], with $J = UV^2/[(\epsilon - \mu)(\epsilon - \mu + U)]$ we can deduce the effective N , and verify the transition from $N = 4$ to $N_{\text{eff}} = 3, 1, 2, 4$ and finally to $N = 2$. Thus the numerical results confirm the possibility to tune the symmetry by the modifying the ratio of Γ_2/Γ_1 .

It is interesting to note the unusual shape of the conductance peak at $\mu = \epsilon + U$ ($\mu \simeq 0.6$ in Fig. 1). Consider first the case $\Gamma_2 = 0$. As the μ approaches $\epsilon + U$, the wide level starts to be gradually filled, its occupation, and as a result, the total conductance, rises as a tail of a Lorentzian of width Γ_1 . At the switching event its occupation jumps to almost 2, and the conductance start decreasing, again as a tail of a Lorentzian of width Γ_1 . Thus the line shape of this peak will consist of a cusp formed by the intersection of the tails of two shifted Lorentzians. For a finite Γ_2 , the LOS events will occur on this scale, and we expect an additional narrow Lorentzian of width Γ_2 on top of the line shape described above.

Unlike the perturbative calculation [9], which predicts the LOS event exactly at the midpoint between the CB peaks, $\mu = \epsilon + U/2$, in the NRG calculation the switching occurs at a higher chemical potential, which shifts to even higher μ as Γ_1 increases, as can be seen in Fig. 4. In fact, for values of Γ_1 larger than $\simeq U/10$, the anomalous CB peak at $\mu = \epsilon + U$ turns into a dip. For such large Γ_1 the LOS events are at $\mu = \epsilon + U$ and $\mu = \epsilon + 2U$, where the narrow level becomes occupied by one and two electrons, respectively.

Between these points the occupation of the wide level rises continuously from $n_1 = 1/2$ to $n_1 = 3/2$, and drops sharply back to $n_1 = 1/2$ at the switching point. Thus, in this regime, we find another atypical situation: three consecutive wide Kondo peaks, corresponding to the three possible occupation states of the narrow level.

The results shown so far were for the fully degenerate case, $\epsilon_1 = \epsilon_2$ and $U_{12} = U_1 = U_2$. Figure 5 depicts the conductance as one varies $\Delta\epsilon \equiv \epsilon_2 - \epsilon_1$, or U_{12}/U (where $U_1 = U_2 = U$ were still equal). As $\Delta\epsilon$ increases from zero [Fig. 5(a)], the switching point between the first two CB peaks shifts to higher chemical potential, until it reaches the second CB peak and disappears, producing a seemingly standard single-level conductance plot in the Kondo regime. As one expects, at this value of $\Delta\epsilon$, as the dot becomes doubly occupied (both electrons occupy the wide level), there should be no Kondo effect. However, as can be seen in Fig. 5(a),

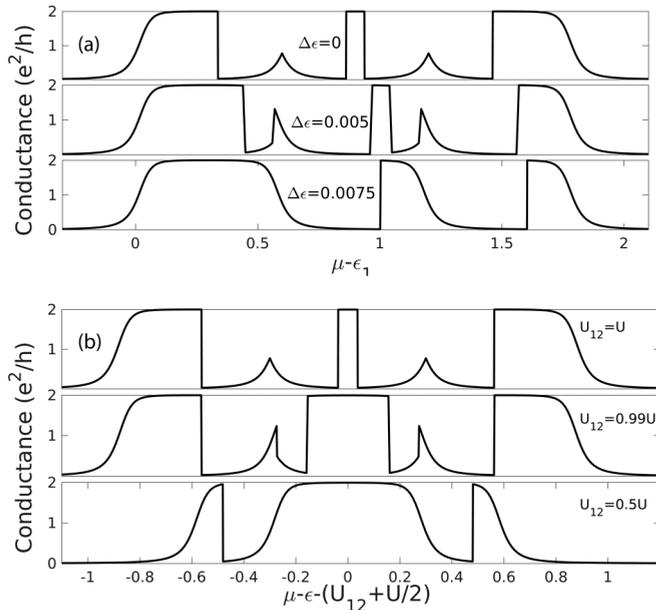


FIG. 5. Conductance as a function of chemical potential for (a) various differences in level energies $\Delta\epsilon = \epsilon_2 - \epsilon_1$ and (b) various ratios between inter and intra dot interaction strengths U_{12}/U . Curves in (b) are centered around the particle-hole symmetry point $U_{12} + U/2$. Plots were calculated with $U = 0.6$, $\Gamma_1 = 0.03$, and $T = 10^{-7}$.

there is re-entrance into the Kondo regime at higher chemical potential (e.g., $\mu = 1$ for $\Delta\epsilon = 0.0075$), again to disappear and to re-emerge again (e.g., at $\mu = 1.6$ for same $\Delta\epsilon$). These re-entrances into the Kondo regime occur due to LOS: whenever the narrow level gets occupied by an additional electron, the energy of the strongly coupled level shifts up, and its occupation is reduced to below double occupation, leading to reappearance of the Kondo effect [9]. When $\Delta\epsilon$ is negative (not shown) one finds the mirror image of the positive- $\Delta\epsilon$ chemical-potential dependence, as now the narrow level is preferentially filled. Thus, even when breaking the energy degeneracy, one still finds the abrupt transitions and the re-entrances that one observes in the degenerate case. Similarly, when U_{12}/U is reduced from unity the LOS still persists, though the first switching event shifts to lower chemical potential.

This makes the first plateau (where $n_2 = 0$) narrower and the regions where $n_2 = 1$ wider. For smaller U_{12}/U , when the second plateau becomes wide enough and the second $n_2 = 0$ region disappears entirely, the conductance exhibits Kondo peaks that are again abruptly suppressed and then re-emerge as the narrow level becomes occupied, with each peak corresponding to a different value of $n_2 = 0, 1, 2$.

The observation of the various predictions made in this paper can be checked in different physical setups. One such system would be a single quantum dot, where the physics of level occupation switching has already been demonstrated by the universality in the transmission phase and its abrupt drop between Coulomb-blockade peaks [10–12]. In such a system, since both levels occupy the same dot, one expects $U_1 \simeq$

$U_2 \simeq U_{12}$. Thus if one can reach a regime where the Kondo temperatures associated with the two levels obey $T_K^{(1)} \gg T \gg T_K^{(2)}$, then one should observe the physics described in this paper. Experimentally, by increasing the coupling of the quantum dot to the leads, the $\pi/2$ phase shift associated with Kondo effect has indeed been observed [36,37], indicating that $T_K^{(1)} > T$. However, in that regime, no abrupt phase drops have been observed, indicating that in this regime the condition $T > T_K^{(2)}$ has not been met. In order to fulfill this latter condition, one may either tune the temperature to that regime, or select a narrow level of a smaller Γ_2 . Another relevant example is transport through nanotube quantum dots [38,39], where each orbital level is fourfold degenerate. Again in this setup one expects, for the same reason, that the Coulomb energies will be of the same order of magnitude. By appropriate application of a magnetic field and gate voltage one can tune the system to have simultaneous transport through two different orbital states, with different coupling to the leads. If the Zeeman splitting is much larger than the coupling of the levels to the leads, then this system will display the orbital SU(2) Kondo effect, as has been observed in Ref. [38]. On the other hand, if the width of the strongly coupled level is larger than the Zeeman splitting, one may observe the abrupt disappearance and re-emergence of the anomalous SU(4) physics detailed in this paper. Another relevant physical system is the double quantum dot system that was utilized to observe the SU(4) Kondo effect [29,30]. In this system, the two separate quantum dots play the role of the two levels in our theory. Experimentally, one can use gate voltages to tune the QDs energies and couplings to the leads, i.e., the parameters Γ_i and ϵ_i . Thus, if one tunes to the already observed SU(4) fixed point, and then gradually reduce the ratio Γ_2/Γ_1 , we predict a gradual transition from SU(4) Kondo to SU(2) Kondo behavior and the eventual emergence of the abrupt suppression and re-emergence of the Kondo peak, as detailed in Fig. 1. One problem that may be relevant to the two-dot setup is that the interdot Coulomb energy U_{12} is typically smaller than the U_i , the intradot one. In principle, in order to achieve degeneracy, one may tune the difference in energies of the two dots, to compensate for the difference in the Coulomb energy. This physics will be explored elsewhere.

To conclude, we have presented physical arguments and numerical renormalization-group calculations that demonstrate a dramatic suppression and re-emergence of the SU(4)/SU(2) Kondo effect in quantum dots that contain two spin-degenerate levels, with different couplings to the leads. Since this has been claimed to be a generic phenomenon in quantum dots and molecular junctions, we expect our results to have a wide range of applicability. In particular, the experiments that have already observed SU(4) Kondo effect, either in carbon nanotube quantum dots, or in semiconductor quantum dots, could be employed to study the physical regime discussed in this paper, and to critically check our predictions.

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