

Universality of the Kondo Effect in Quantum Dots with Ferromagnetic Leads

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We investigate quantum dots in clean single-wall carbon nanotubes with ferromagnetic PdNi-leads in the Kondo regime. Most of the Kondo resonances exhibit a splitting, which depends on the tunnel coupling to the leads and an external magnetic field B , but only weakly on the gate voltage. Using numerical renormalization group calculations, we demonstrate that all salient features of the data can be understood using a simple model for the magnetic properties of the leads. The magnetoconductance at zero bias and low temperature depends in a universal way on $g\mu_B(B-B_c)/k_B T_K$, where T_K is the Kondo temperature and B_c the external field compensating the splitting.

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The Kondo effect resulting from the exchange interaction of a single spin with a bath of conduction electrons [1] is one of the archetypical phenomena of many-body physics. Its competition with ferromagnetism and possible applications in spintronics [2] have raised wide interest in the past few years. The Kondo effect in quantum dots [3–5] has, in recent experiments, been investigated in the presence of ferromagnetic (FM) leads [6–8]. It was found that the Kondo resonance, usually observed at zero bias in the odd Coulomb blockade (CB) valleys, is split into two peaks at finite bias [6]. The splitting consists of a term depending logarithmically on gate voltage [7,8], and, as demonstrated here, a second term nearly independent of gate voltage. These phenomena were predicted theoretically [9–12], attributing the splitting of the Kondo resonance to a tunneling-induced exchange field, which results from the magnetic polarization of the leads. So far no detailed and quantitative comparison of the measured conductance with the theory has been undertaken to verify whether the simplistic description of FM leads used in Refs. [9–12] has quantitative predictive power. The lead-induced spin polarization of the Kondo resonance may be used for spin filtering at the single channel level.

In this Letter we present low-temperature transport measurements of a single-wall carbon nanotube quantum dot with PdNi leads. We concentrate on the less studied gate-independent contribution of the exchange splitting of the Kondo resonance and attribute it to the saturation magnetization of the contact material. We show that the evolution of the conductance with magnetic field and gate voltage can be understood within a simple model for the magnetization and polarization in the FM leads, by presenting numerical renormalization group (NRG) calculations for this model, using parameters extracted from experiment. Moreover, by comparing resonances of different transparency, we demonstrate a universal scaling of the magnetic

field dependence of the Kondo conductance. This proves that the magnetization of the leads can indeed be viewed as an exchange field, which acts analogously to an external magnetic field.

Experimental setup.—The nanotubes are grown by chemical vapor deposition on an oxidized silicon substrate acting as a back gate [13]. The contact electrodes with a thickness of roughly 45 nm are subsequently structured by electron beam lithography and consist of Pd_{0.3}Ni_{0.7}, known to generate highly transparent contacts [14]. The current I is measured in a two point geometry with a voltage bias V_{sd} applied to the source contact.

The differential conductance $G = dI/dV_{sd}$ is plotted in Fig. 1 in color scale, providing the typical charging diagram of a quantum dot [15]. Our device exhibits regular CB oscillations and a clear fourfold symmetry characteristic for carbon nanotubes [16]. Coupling to the contacts is strong, leading to broad resonance lines and a variety of

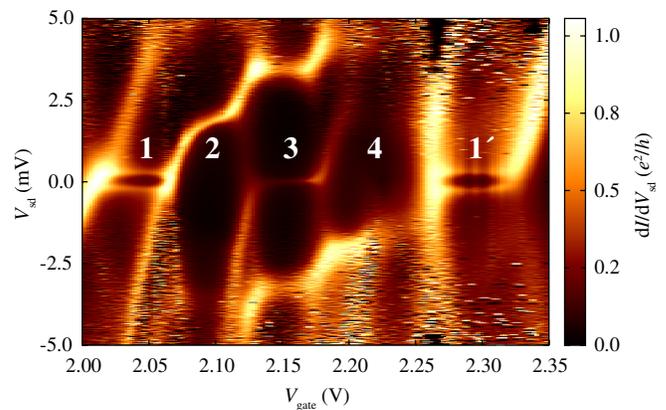


FIG. 1 (color online). Differential conductance dI/dV_{sd} versus bias voltage V_{sd} and gate voltage V_{gate} at $T = 25$ mK and $B = 0$ T. The CB regions are numbered for future reference.

higher-order processes. From Fig. 1 we extract a charging energy $U \approx 5$ meV, a level separation $\Delta \approx 9.5$ meV, and a subband mismatch of about $\delta \approx 1$ meV [16,17]. The tunnel coupling Γ between leads and quantum dot can be inferred from the linewidth of the conductance peak. Between valleys 1 and 2 in Fig. 1 we obtain a full width at half maximum (FWHM) of $\Gamma = 1.1$ meV $\gg k_B T$.

A striking feature visible in Fig. 1 is lines of enhanced conductance at small, approximately constant bias values in every second CB diamond. We attribute these lines to a spin-1/2 Kondo conductance anomaly, split into two peaks at small opposite bias values due to the presence of FM contacts. Some resonances with very low conductance exhibit no measurable splitting.

Main features of B dependence.—Figure 2(a) displays detailed measurements of valley 1 from Fig. 1 for different

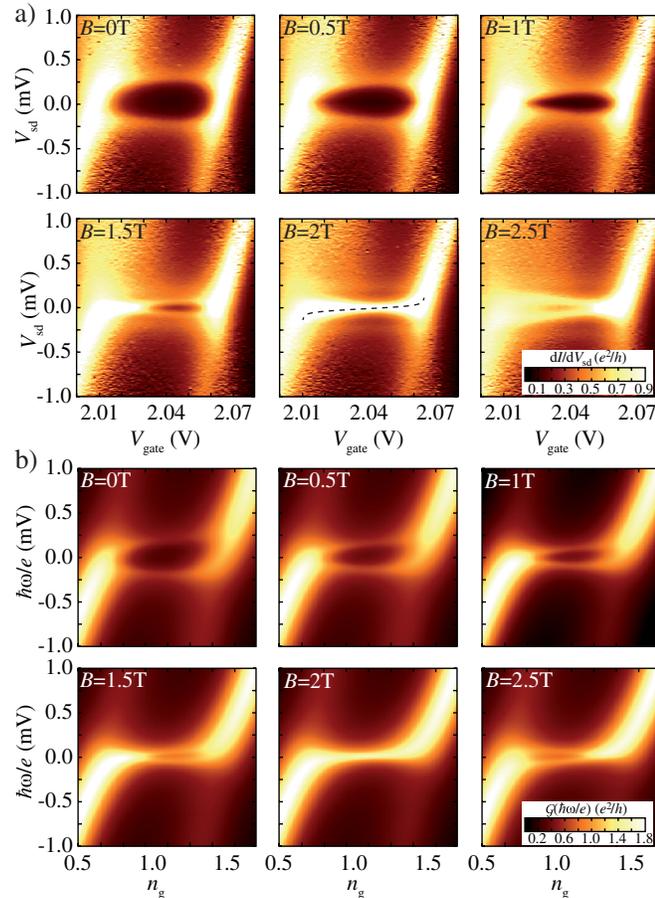


FIG. 2 (color online). Differential conductance versus source-drain and gate voltage, in CB region 1 of Fig. 1, for six values of B . We compare here (a) experimental data with (b) NRG results for the normalized equilibrium zero-temperature spectral function $\mathcal{G} = \sum_{\sigma} \pi \Gamma_{\sigma}(0) A_{\sigma}(\omega)$, obtained using the model and parameters described in the text. $n_g = 1/2 - \varepsilon_d/U$ is the dimensionless gate potential. The dashed line in the panel for $B = 2$ T in (a) indicates the gate-dependent contribution from the polarization for $\mathcal{P} = 10\%$ (see text).

values of external magnetic fields almost parallel to the nanotube axis. The main observations are the following. (i) The dominant contribution to the splitting is independent of V_{gate} . (ii) In all investigated cases the splitting is reduced as the field strength increases until only a single apparent peak remains. This field value is referred to as compensation field (here $B_c \approx 2$ T), since the dominant gate-independent part of the splitting is compensated. At higher fields the peak splits again. (iii) Despite (i), we observe, nevertheless, a slight gate dependence, in particular, near B_c . This is most clearly reflected by the fact that the crossing point of the two resonances moves from the left side of the CB diamond for $B < B_c$ (cf. $B = 1.5$ T) to the right side for $B > B_c$ (cf. $B = 2.5$ T). (iv) The gate dependence increases in strength very close to the edges of the CB diamond.

Tunneling-induced level shifts.—The presence of the splitting, its dependence on magnetic field, and a potential gate dependence can be consistently explained by the renormalization of the quantum dot level energy due to charge fluctuations between the dot and the leads. Since the density of states (DOS) in the FM contacts is spin dependent, the renormalization results in a spin splitting of the dot level $\Delta\varepsilon \equiv \delta\varepsilon_{\uparrow} - \delta\varepsilon_{\downarrow} + g\mu_B B$. For a single impurity Anderson model, the correction $\delta\varepsilon_{\sigma}$ ($\sigma = \uparrow, \downarrow$) from second order perturbation theory is (see, e.g., Ref. [11]):

$$\delta\varepsilon_{\sigma} \approx -\frac{1}{\pi} \int d\omega \left(\frac{\Gamma_{\sigma}(\omega)[1 - f(\omega)]}{\omega - \varepsilon_{d,\sigma}} + \frac{\Gamma_{-\sigma}(\omega)f(\omega)}{\varepsilon_{d,-\sigma} + U - \omega} \right). \quad (1)$$

Here, $\varepsilon_{d,\sigma} = \varepsilon_d \mp g\mu_B B/2$ is the quantum level energy for spin σ , U the charging energy, $\Gamma_{\sigma}(\omega)$ the spin-dependent tunneling rate, and $f(\omega)$ the Fermi function.

From Eq. (1) one sees that the splitting is not only a consequence of properties at the Fermi surface, but of the full DOS. The first and second terms in Eq. (1) describe electron- or holelike processes, meaning fluctuations between the states $|1, \sigma\rangle$ and $|2\rangle$ or $|0\rangle$, respectively (the numeral denotes the charge occupation of the quantum dot). The spin-dependent energy corrections $\delta\varepsilon_{\sigma}$ are negative, as always in second order perturbation theory. Consequently, the spin direction that allows stronger fluctuations will have lower renormalized energy.

Effect of magnetization.—First we assume a shift between bands of equal and constant DOS for different spin directions, $\rho_{\uparrow} = \rho_{\downarrow} = \rho_0$, described by a constant Stoner splitting Δ_{St} , see Fig. 3. The tunneling-induced splitting $\Delta\varepsilon^{(\mathcal{M})}$ due to Δ_{St} is directly related to the saturation magnetization $\mathcal{M} \equiv (n_{\uparrow} - n_{\downarrow})/N_a = \Delta_{\text{St}}/(2D_0)$. Here $n_{\sigma} = \rho_0(D_0 + \varepsilon_F \pm \Delta_{\text{St}}/2)$ is the number of spin- σ electrons, ε_F the Fermi energy, and N_a the number of states per atom and spin. Starting from Eq. (1) we can deduce the spin orientation of the dot ground state as follows. Figures 3(b) and 3(c) show the phase space available

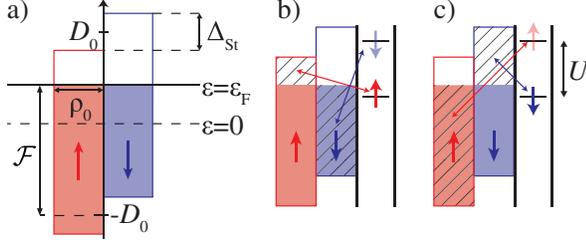


FIG. 3 (color online). Schematic of the level renormalization process. (a) We assume flat bands $\rho_{\sigma}(\omega) = \rho_0$ with bandwidth D_0 , shifted with respect to each other by a constant Stoner splitting Δ_{St} . The filling fraction \mathcal{F} determines the Fermi energy ε_F . (b) Charge fluctuations for a spin-up electron on the dot, involving the empty states of the spin-up band and the occupied states of the spin-down band (hatched areas). (c) Analogous situation for a spin-down electron on the dot; the available phase space (hatched) is larger than in (b).

(hatched) for quantum charge fluctuations for a spin-up or a spin-down electron residing on the quantum dot, respectively. Comparing the total sizes of hatched areas for (b) and (c), the phase space is larger for the latter, thus favoring spin-down. Hence the ground state spin is always oriented opposite to the magnetization of the leads if $B < B_c$. This explains why the splitting is always initially reduced (never increased) when an external field is applied [cf. observation (ii) listed above; the magnetization direction follows that of B in our setup]. The magnitude of this effect depends on the Stoner splitting, i.e., on \mathcal{M} . For $|\varepsilon_d|, \varepsilon_d + U \ll D_0, \Delta_{St}$, which is compatible with the experiment, we obtain, in extension of [12],

$$\Delta\varepsilon^{(\mathcal{M})} \simeq \frac{\Gamma}{2\pi} \ln \left[\frac{(1 - \mathcal{M})^2 - (2\mathcal{F} - 1)^2}{(1 + \mathcal{M})^2 - (2\mathcal{F} - 1)^2} \right], \quad (2)$$

where $\mathcal{F} = (1 + \varepsilon_F/D_0)/2$ is the filling fraction of the band. This shift is independent of the gate voltage [explaining observation (i)] because the position ε_d of the level is very close to ε_F , while the integration in Eq. (1) ranges over a large fraction of the d band.

SQUID measurements allow us to determine a magnetic moment of $\mu = 0.58\mu_B/\text{atom}$ for our PdNi alloy [18], implying an effective magnetization of $\mathcal{M} = 0.116$. *Ab initio* calculations of the band structure provide a filling fraction of $\mathcal{F} = 0.853$ [19]. Thus, we estimate $\Delta\varepsilon^{(\mathcal{M})} \simeq -175 \mu\text{eV}$. In transport spectroscopy this would lead to conductance peaks split at zero external field by $2\Delta\varepsilon^{\text{theo}}/e = 2|\Delta\varepsilon^{(\mathcal{M})}|/e \simeq 350 \mu\text{V}$. Given the simplicity of our model, this agrees reasonably well with the experimentally determined peak distance of $2\Delta\varepsilon^{\text{exp}}/e \simeq 550 \mu\text{V}$. For a more weakly coupled resonance the predicted peak distance of $60 \mu\text{V}$ agrees similarly with the experimentally found value of $105 \mu\text{V}$. Additional contributions to $\Delta\varepsilon^{(\mathcal{M})}$, resulting from other factors as, e.g., discussed in [20,21] cannot be ruled out.

Effect of polarization.—The case of $\rho_{\uparrow} \neq \rho_{\downarrow}$, implying nonzero polarization $\mathcal{P} = (\rho_{\uparrow} - \rho_{\downarrow})/(\rho_{\uparrow} + \rho_{\downarrow})$ at ε_F , has already been discussed in Refs. [7,12] and earlier publications referenced there. Assuming a flat band with a spin-dependent DOS, e.g., $\rho_{\uparrow} > \rho_{\downarrow}$ but zero \mathcal{M} for simplicity, quantum charge fluctuations renormalize the quantum dot level depending on its position relative to ε_F . This contribution shows a logarithmic divergence for $\varepsilon_d \rightarrow 0$ and $\varepsilon_d + U \rightarrow 0$ [7,12], resulting in the upward and downward bending of the compensated conductance peak towards the corners of the diamond [cf. observation (iv)].

Numerical results.—The quality of our model is reflected by the close correspondence of Figs. 2(a) and 2(b) (see also the corresponding line traces in Fig. S1 of the Supplemental Material [22]). Figure 2(b) presents our NRG results for the spectral function $A(\omega)$ [23] versus the normalized gate voltage n_g , calculated for a single-lead Anderson model with the DOS shown in Fig. 3, but with $\rho_{\uparrow} \neq \rho_{\downarrow}$, using *full* density-matrix NRG [24,25]. Using the measured parameters of the quantum dot and modeling the ferromagnetism in the leads by taking $\mathcal{P} = 10\%$ and $\mathcal{M} = 0.116$, very good agreement with experiment is found, except for the background current at high V_{sd} , which results probably from cotunneling processes involving higher levels not included into the model. In the experiment as well as in the numerical data, at $B = 2 \text{ T}$ the gate-independent contribution, cf. Eq. (2), is fully compensated and only the (weak) gate-dependent contribution from \mathcal{P} remains [cf. observation (iii)], indicated by a dashed line in Fig. 2(a) ($B = 2 \text{ T}$). The crossing point of the resonances lies in the center of the CB diamond. By varying \mathcal{P} and comparing the shape of the conductance peak at B_c between experiment and theory, we estimate $\mathcal{P} \simeq 10\%$.

Universality.—For a quantum dot coupled to normal leads, the normalized zero-bias conductance is a universal function (i) of $\tilde{T} = T/T_K$ at zero field, and (ii) of $\tilde{B} = g\mu_B B/k_B T_K$ at zero temperature [26]. We find, quite remarkably, that both these universal features are recovered also for ferromagnetic leads, if B is replaced by the effective field $\delta B = B - B_c$ [12]. Regarding (i), Fig. 4(d) shows that at the compensation field, $B = B_c$, the T/T_K dependence of the normalized conductance $G(T)/G(0)$ agrees with the NRG data, both for normal contacts at $B = 0$ (cf. [5]) and ferromagnetic contacts at $B = B_c$. Although the latter behavior is well established for dots coupled to normal leads, its emergence here is nontrivial: it demonstrates that despite the ferromagnetic environment, local spin symmetry can indeed be fully restored by fine tuning the field to B_c .

The magnetic field dependence (ii) has so far attracted much less attention [27]. To explore it, Figs. 4(a)–4(c) show $G(B)$ at fixed $T \ll T_K$ for several charge states differing in T_K , B_c , and $\Delta\varepsilon(B = 0)$. The position of the conductance peak roughly follows the Zeeman law

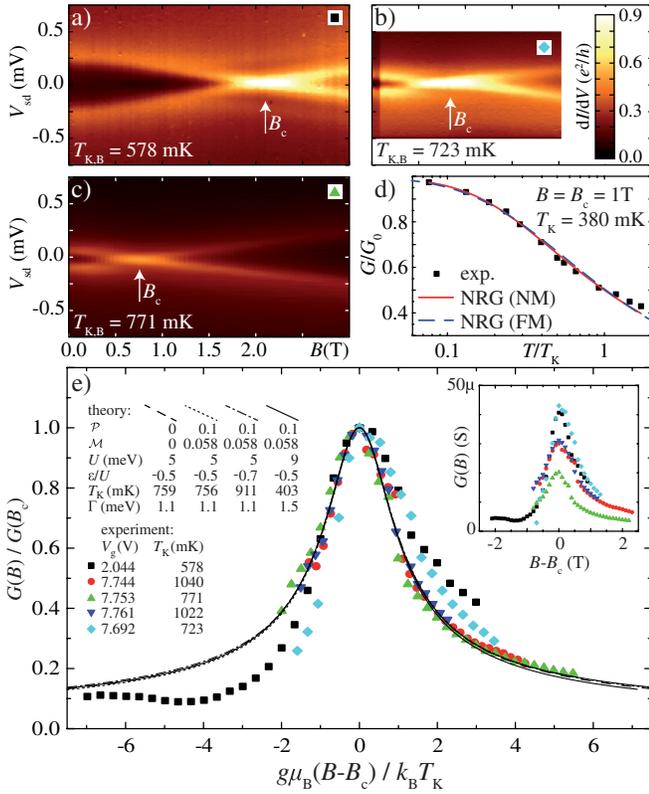


FIG. 4 (color online). (a)–(c) Magnetic field dependence of the splitting for different charge states. Vertical arrows denote the compensation field B_c . (d) Scaling plot of $G(T)/G(T = 0)$ vs T/T_K (symbols), at $B = B_c$, for the charge state shown in Fig. 4(c); the solid line displays standard NRG data for normal leads (NM) at $B = 0$ while the dashed line displays the results of our NRG calculations with ferromagnetic leads (FM) at $B = B_c$. (e) Scaled zero-bias conductance $G(B)/G(B_c)$ plotted against the effective normalized field $\delta\tilde{B}$, at fixed $T = 50$ mK $\ll T_K$ (100 mK for \blacksquare). Lines represent NRG calculations for several parameter sets. Inset: $G(B)$ vs $(B - B_c)$ before scaling.

for $g = 2$, with slight deviations in the vicinity of the compensation field B_c [27]. We find that B_c and T_K vary independently for different charge states, implying different couplings Γ .

Now we check whether, according to (ii), $G(B)/G(B_c)$ even in the presence of ferromagnetic contacts is a universal function of $\delta\tilde{B} = g\mu_B(B - B_c)/k_B T_K$. The lines in Fig. 4(e) show this curve, calculated by NRG for four different sets of model parameters, yielding a good scaling collapse. Symbols show experimental data for $G(B)/G(B_c)$ vs $\delta\tilde{B}$, for several different gate voltages, with T_K extracted by numerical fitting to the NRG results. For three data sets taken on the same charge state (circles and triangles), scaling works very well and agreement with theory is excellent; for the other two sets (squares, diamonds) where the conductance is very high, the quality of scaling is reduced at higher $\delta\tilde{B}$ by a background contribution to the magnetoconductance which is asymmetric with respect to B_c . Nevertheless, the overall agreement between theory

and experiment shows that the model correctly captures the universal, sample-independent features of $G(B)$ as a function of $\delta\tilde{B}$.

Conclusions.—We have performed a quantitative comparison of the conductance of quantum dots with FM contacts, in the Kondo regime, with model NRG calculations. The quantitative agreement between experimental and numerical data lends strong support to the scenario proposed in Refs. [9,12]: the exchange field induced by magnetic contacts causes the local level to be split by an amount $\Delta\epsilon$, which adds a constant offset to the Zeeman splitting induced by an external magnetic field. At the compensation field B_c , universal scaling features of the Kondo effect are recovered. With a proper choice of the contact material the spin splitting of the Kondo resonance in our samples provides the basis for a source of highly spin-polarized electrons, which may prove useful for future application in spintronics research.

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Note added.—Recently we became aware of a similar study in quantum dots with nonmagnetic contacts [28].

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