Rotational Quantum Impurities in a Metal: Stability of the 2-Channel Kondo Fixed Point in a Magnetic Field

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A three-level system with partially broken SU(3) symmetry immersed in a metal, comprised of a unique non-interacting ground state and two-fold degenerate excited states, exhibits a stable two-channel Kondo fixed point within a wide range of parameters, as has been shown in previous work. Such systems can, for instance, be realized by protons dissolved in a metal and bound in the interstitial space of the host lattice, where the degeneracy of excited rotational states is guaranteed by the space inversion symmetry of the lattice. We analyze the robustness of the 2CK fixed point with respect to a level splitting of the excited states and discuss how this may explain the behavior of the well-known dI/dV spectra measured by Ralph and Buhrman on ultrasmall quantum point contacts in a magnetic field.

1 Introduction

New, exotic quantum states of matter can arise in electronic systems, when two degenerate ground states compete with each other and get entangled, leading to nontrivial behavior of the entropy and of the thermodynamic, magnetic and electric response. In the case of the spin-1/2 two-channel Kondo (2CK) effect, the spin-screening of a spin-1/2 impurity by the first and by the second of two identical, conserved conduction electron continua (channels), respectively, are in competition for each of these screening channels to form a spin singlet [1]. However, in this system both the weak coupling fixed point (decoupled spin-1/2 impurity) and the strong-coupling fixed point (a three-body bound state comprised of the spin-1/2 impurity and two conduction electrons) are doubly degenerate and, hence, are unstable with respect to a small coupling to the conduction band [1, 2]. As a result, a stable intermediate coupling fixed point is formed, where an intricate, quantum-frustrated many-body ground state with a non-vanishing zero-point entropy of $S(T=0) = k_B \ln \sqrt{2}$ is realized, k_B denoting the Boltzmann constant [3, 4]. The spin degree of freedom need not be magnetic spin, but may be any two-dimensional representation of SU(2), i.e. a pseudospin 1/2.

Signatures of the 2CK effect have been observed experimentally in certain heavy-fermion compounds [5, 6], where the Kondo degree of freedom might arise from orbital degeneracy [7, 8]. However, not all measured response quantities have been found to be in accordance with 2CK behavior in these systems. More recently, the 2CK fixed point has been predicted to exist [9] and then realized [10] in an an ingeniously designed, fine-tuned semiconductor Qdot system. However, perhaps the most intriguing as well as controversial experiments regarding the 2CK effect remain the dI/dV spectroscopy measurements by Ralph and Buhrman on ultrasmall Cu quantum point contacts [11,12]. They exhibit scaling behavior of the conductance near zero bias, as expected from a 2CK system [13,14,16], without fine-tuning of parameters as well as sharp conductance spikes at elevated bias. Alternative scattering mechanisms [17,18], different from 2CK physics, cannot account for the complete body of experimental observations [15,19]. A review of theoretical and experimental aspects of 2CK physics can be found in Ref. [20].

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It is difficult to design realistic, microscopic models which generically, i.e., without fine-tuning, exhibit a 2CK fixed point, because either the Kondo (pseudo)spin symmetry or the channel degeneracy are easily broken. Two-level systems (TLS), e.g., an ion in a double-well potential, embedded in a metal, have been put forward early-on as 2CK systems and have been intensively studied by Zawadowski and co-workers [21–23]. These studies have lead to a profound understanding of the dynamics of TLS in metals. However, it turned out that in the standard TLS model of a particle in a double-well potential the 2CK regime cannot be realized. This is because the Kondo coupling and the tunneling rate and, hence, the ground state level splitting are not independent parameters in this model. It was shown that the two-channel Kondo temperature T_K is always smaller than the level splitting, because of screening effects [24] and coupling to higher excited states [25], so that the physics is always dominated by the level splitting. Despite extensions of the TLS model [26] it has remained difficult to stabilize a 2CK fixed point.

In order to provide an explanation for the 2CK physics and at the same time for the conductance spikes observed in the Ralph-Buhrman experiments without fine-tuning, we have earlier proposed and analyzed the model of a dynamical impurity with a *rotational* degree of freedom, immersed in a metal [27]. In this rotational impurity model (RIM) the Kondo SU(2) symmetry is stabilized by the space inversion symmetry of the host material, while the channel degree of freedom is the (magnetic) conduction electron spin, and its degeneracy is guaranteed by time reversal symmetry. Taking the first rotational doublet into account, it was shown by perturbative renormalization group (RG) that this model generically has a stable 2CK fixed point within a wide range of parameter values. In addition, the conductance spikes were naturally explained within the same model as Kondo-like transitions between a rotational doublet and the impurity ground state. In the present paper we extend this study to the behavior in a magnetic field, Zeeman-coupled to the magnetic moment of a rotating, charged particle. In Section 2 we describe the model and its renormalization group treatment in more detail and discuss briefly, why charge screening effects or higher excitations of the rotational impurity will not suppress T_K, in contrast to the case of a double-well impurity [24, 25]. In Section 3 we present the results of the perturbative RG for the RG flow of the energy and the decay rate of the excited rotational doublet and, in particular, the phase diagram of the model in the presence of a magnetic field, lifting the doublet degeneracy. We conclude in Section 4 with a discussion of the implication of the results for the interpretation of the conductance spectroscopy measurements [11, 12].

2 Microscopic model and renormalization group treatment

2.1 Hamiltonian

Hydrogen is easily dissolved in ionic form in noble metals like paladium or copper. The protons occupy the interstitial spaces of the host lattice. If this lattice obeys space inversion symmetry, like the Cu fcc lattice, all excited states of a proton in the lattice potenial are doubly degenerate, while it's ground state is unique. The excited-state doublets may be visualized as roton states with opposite rotational orientation, see Fig. 1 a). Taking only the first excited doublet and the ground state into account and considering that conduction electron scattering can induce transitions between any of these three states, one obtains a three-level model whose SU(3) symmetry is partially broken, due to the bare level splitting Δ_0 between the ground and the excited states [27]. The level scheme is shown in Fig. 1 b), along with the various couplings of conduction electrons to the dynamical impurity. The corresponding Hamiltonian reads,

$$
\begin{aligned}
H \;=\; & {\sum_{\mathbf{k}\sigma m}}' \varepsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma m} c_{\mathbf{k}\sigma m} + \Delta_0 \sum_{m=\pm 1} f^\dagger_m f_m + B_0 \sum_{m=\pm 1} m\, f^\dagger_m f_m \\
& + \sum_\sigma \Big(\Big[\frac{1}{2} \sum_{i,j=\pm 1} ij J^{ij}_z S_{i,i} s^\sigma_{j,j} \Big] + \sum_{\substack{i,j=\pm 1 \\ i\neq j}} J^i_\perp S_{i,j} s^\sigma_{j,i} \Big) \\
& + \sum_\sigma \Big(\sum_{\substack{m,n \\ -1\leq n-m\leq 1}} \Big[g^{(n)}_{m0} S_{m,0} s^\sigma_{n-m,n} + H.c. \Big] + \sum_{m=\pm 1} 2 g^{(0)}_{mm} S_{m,m} s^\sigma_{0,0} \Big) .
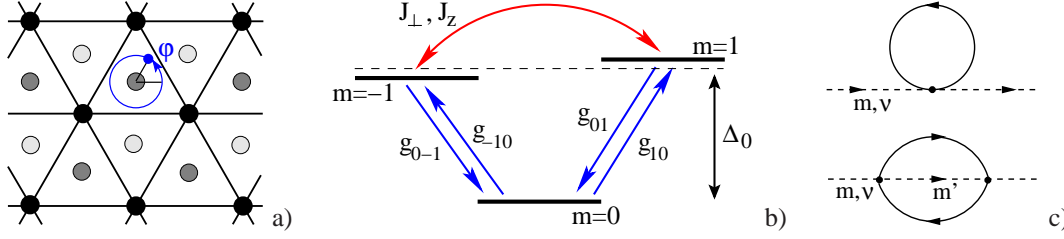\end{aligned}
\tag{1}
$$

Fig. 1 a) Cu lattice with a rotational impurity at interstitial site. Black dots define one layer of the Cu (111) plane, light and dark grey dots define the layers above and below, respectively. The blue dot is the impurity with a two-fold degenerate rotational degree of freedom. **b)** Level scheme of the three-state quantum impurity, defining the coupling constants. The additional superscript carried by the $g_{mn}^{(j)}$ in Eq. 1 indicates the initial conduction electron state before scattering, which is crucial for the RG treatment of excited states [27]. **c)** Impurity self-energy diagrams, where solid lines represent conduction electron propagators and dashed lines pseudo fermion progagators.

The first term is the usual conduction electron kinetic energy. The conduction electron operators $c_{\mathbf{k}\sigma m}^{\dagger}$ carry the *conserved* magnetic spin $\sigma = \pm\frac{1}{2}$, acting as the channel degree of freedom, and additionally an SU(3) index $m = 0, \pm 1$, labelling lattice angular momentum states of the conduction electrons which may be changed in a scattering process. The prime on the sum in Eq. (1) indicates a restricted momentum sum such that $\sum_{\mathbf{k}m}{}' \equiv \sum_{\mathbf{k}}$ comprises a summation over a unique and complete basis of single-electron states. The second term describes the two-fold degenerate impurity states, $m = \pm 1$, with bare excitation energy Δ_0 above the impurity ground state, $m = 0$. In the presence of a magnetic field coupling to the rotational motion (magnetic moment) of the charged impurity particle, the excited states acquire an additional Zeeman splitting, $2B_0$, described by the third term in Eq. (1). We use Abrikosov's pseudo-fermion representation to describe the impurity levels, where $f_m^{\dagger}$ is the creation operator for the impurity in state $m = 0, \pm 1$. The defect dynamics are restricted by the constraint $Q = \sum_{m=0,\pm 1} f_m^{\dagger} f_m = 1$. The last two terms represent transitions between the local level induced by conduction electron scattering, including the potential scattering term for completeness, although it is an irrelevant operator. See Fig. 1 b) for the definition of the coupling constants. The impurity operators are defined as $S_{m,n} = f_m^{\dagger} f_n$, and the operators acting on the electronic Fock space are obtained by substituting $f_m \to \sum_{\mathbf{k}} c_{\mathbf{k}\sigma\mathbf{m}}$ in the above expressions.

2.2 Perturbative RG Analysis

We employ the one-loop (second-order perturbation theory) renormalization group to analyze the three-level model, Eq. (1). Transitions between the non-degenerate impurity states ($\Delta m = \pm 1$) necessarily involve resonant, inelastic electron scattering, where an electron initiates from or ends up in an excited state. Therefore, it is not sufficient to calculate renormalized couplings at the Fermi energy, but it is crucial to take the dependence of the renormalized coupling constants on the (initial) energy of the scattering electrons into account [27]. The corresponding formalism has originally been developed for non-equilibrium Kondo systems at finite bias [28–30], but it is suitable for the present situation as well. We take the impurity dynamics on-shell, i.e. the pseudofermion energy ν equal to the respective impurity level energy, $\nu = |m|\Delta(D)$, in all expressions. The one-loop RG equations for the running couplings $g_{mn}^{(j)}$ then read,

$$\frac{dg_{mn}^{(j)}(\omega)}{d\ln D} = 2 \sum_{\substack{j\ell \\ -1 \leq j+n-\ell \leq 1}} g_{m\ell}^{(j+n-\ell)}(\Omega_{n\ell})\, g_{\ell n}^{(j)}(\omega)\, \Theta\Big(D - \sqrt{\Omega_{n\ell}^2 + \Gamma_\ell^2}\Big)\big(1 - \delta_{m\ell}\delta_{n\ell}\big) - (\text{exch.}) \quad (2)$$

with $g_{1,-1}^{(1)} = g_{-1,1}^{(-1)} = J_\perp$ and $g_{mm}^{(j)} = jm\, J_z/2$, for $j = \pm 1$, $m = \pm 1$. The exchange terms (exch.) on the right-hand side (RHS) of Eq. (2) are obtained from the direct ones by interchanging in the g's in- and out-going pseudofermion indices and by interchanging $m \leftrightarrow n$ everywhere. The energy arguments

of the g's on the RHS arise from energy conservation at each vertex, and the Kronecker-δ factor excludes non-logarithmic terms which do not alter the impurity state. The Θ step function in Eq. (2) cuts off the RG flow of a particular term when the band cutoff D is reduced below the intermediate-state energy $\Omega_{n\ell}$ of an electron which, before the scattering process, had the energy ω. This energy is $\Omega_{n\ell} = \omega + (|n| - |\ell|)\Delta(D) + (\ell - n)B(D)$. It depends on the renormalized level spacing $\Delta(D)$ and the renormalized Zeeman splitting $B(D)$. The cutoff also involves the decay rates of the intermediate impurity states [31], $\Gamma_m(D)$. Although $\Delta(D)$ and $B(D)$ and $\Gamma_m(D)$, contain no leading logarithmic terms, they acquire an RG flow, since they are calculated from the 2nd-order self-energy diagrams shown in Fig. 1 c). The 1st-order diagram is real, gives the same shift for all impurity states and, hence, does not contribute. Specifically, $\Delta(D)$ and $B(D)$ are obtained during the RG flow as [27],

$$\Delta(D - \delta D) = \Delta(D) - \delta\mathrm{Re}\Sigma_0(\nu = 0) + \frac{1}{2}\big(\delta\mathrm{Re}\Sigma_1(\nu = \Delta + B) + \delta\mathrm{Re}\Sigma_{-1}(\nu = \Delta - B)\big) \quad (3)$$

$$B(D - \delta D) = B(D) + \frac{1}{2}\big(\delta\mathrm{Re}\Sigma_1(\nu = \Delta + B) - \delta\mathrm{Re}\Sigma_{-1}(\nu = \Delta - B)\big), \quad (4)$$

where $\delta\mathrm{Re}\Sigma = \mathrm{Re}\Sigma(D) - \mathrm{Re}\Sigma(D - \delta D)$.

2.3   Two-channel Kondo regime and stability against charge screening

One of us and collaborators showed in Ref. [27] that in zero magnetic field ($B_0 = 0$) the model (1) has generically a stable 2CK fixed point for a wide range of parameters. The 2CK phase occurs, because the excited state doublet of the non-interacting impurity is down-renormalized by Kondo-like interactions below the non-interacting ground state and the system must then flow to a stable 2CK fixed point. The Kondo state is formed due to the unbroken SU(2) subgroup of SU(3) within the (initially excited) doublet of the three-level system. Moreover, it was shown that spikes in the differential conductance at finite bias voltage arise from Kondo-enhanced transitions between the ground and the excited states [27]. These spikes are analogous to the well-known Kondo satellite resonances observed in rare-earth Kondo impurities with several, crystal-field split, local orbitals [32].

It should be emphasized that in the RIM the 2CK Kondo temperature is not reduced by charge screening effects: The charge of any impurity is screened by the conduction electrons. In a TLS in a double-well potential the spatial charge density distribution is coupled to the presumed Kondo pseudospin degree of freedom and is altered by a pseudospinflip process. Therefore, only those low-energy electrons participate in the Kondo scattering which cannot screen the flipping TLS charge distribution instantaneously. As shown in Ref. [24], this reduces the energy range available for Kondo scattering from the bare conduction bandwidth to the TLS tunneling frequency and, thus, suppresses T_K. By contrast, within the RIM Kondo scattering occurs within space-inversion symmetric roton doublets which alters the phase, but not the charge density distribution of the system. Therefore, charge density and Kondo degree of freedom are independent, and T_K is not influenced by charge screening. Moreover, to realize a 2CK regime in the RIM it is not necessary to invoke transitions via higher excited states in order to enhance T_K over the level splitting of the doublet – which can be prevented by the alternating parity of the higher excited states [25]. This is because in the RIM the excited-state doublet is degenerate by space inversion symmetry and because, in any case, a possible level splitting $2B_0$ and the Kondo couplings $J_\perp$, J_z are independent parameters. Hence, the 2CK behavior can be cut off only by breaking the space inversion symmetry, e.g., by lattice distortion, or, in a more controlled way, by a magnetic field.

3   Results in finite magnetic field

We have investigated the appearance of 2CK physics in the three-level model (1) in the presence of a finite doublet splitting or magnetic field, $B_0 > 0$, according to Eqs. (2), (3) and (4)

Fig. 2 a) shows an example of the RG flow of the excited doublet levels ($m = \pm 1$) relative to the initial ground state level ($m = 0$) for a finite magnetic field B_0. B_0 was chosen of the order of the Kondo
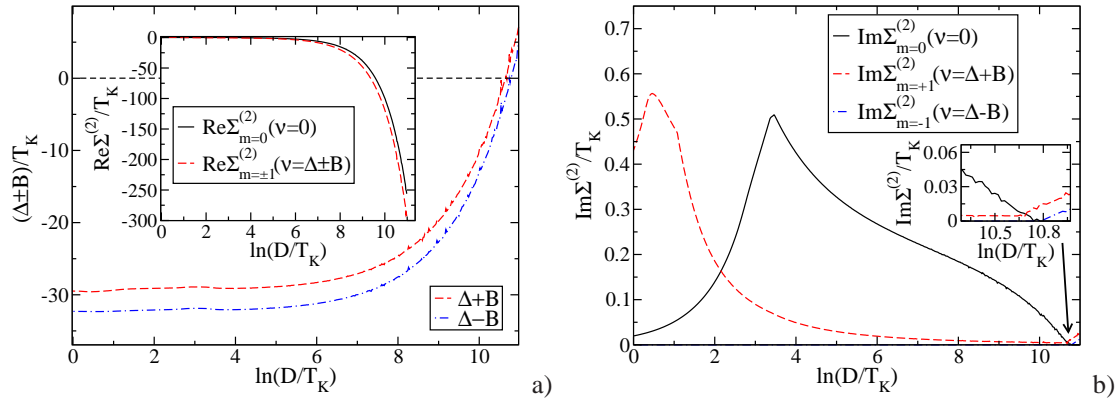
Fig. 2 **a)** Perturbative RG flow of the level spacings between the initially excited $m = \pm 1$ states and the $m = 0$ state for $B_0 = 1.45 \cdot T_K$, $T_K = 1.7 \cdot 10^{-5} D_0$, $\Delta_0 = 5.9 \cdot T_K$ and $J_\perp = J_{||} = 0.015 \, N(0)$, $g_{m0}^{(j)} = g_{0m}^{(j)} = J_\perp/2$. D_0 is the bare high-energy cutoff. The inset shows the RG flow of the real parts of the self-energies for $m = 0$ and $m = \pm 1$. **b)** RG flow of the imaginary part of the 2nd-order impurity level self-energies for the same parameters. The inset is a blow-up of the region where the level crossing occurs.

temperature. T_K was determined here and throughout as the cutoff value D for which the dimensionless Kondo coupling reaches $N(0)J_\perp(\omega = 0) = 1$ in zero magnetic field, with $N(0)$ the density of states at the Fermi level. The inset of Fig. 2 a) shows the flow of the on-shell real parts of the impurity self-energies. The differences $\mathrm{Re}\Sigma_{m=1}(\Delta+B) - \mathrm{Re}\Sigma_{m=0}(0)$ and $\mathrm{Re}\Sigma_{m=-1}(\Delta-B) - \mathrm{Re}\Sigma_{m=0}(0)$ provide the renormalization of the level spacings $\Delta(D) \pm B(D)$, respectively. As the inset shows, this renormalization is negative and initially stronger than the bare spacings $\Delta_0 \pm B_0$, thus causing a level crossing with the $m = 0$ state. If this level crossing occurs for both states, $m = \pm 1$, as seen in Fig. 2 a), 2CK behavior is realized at the lowest energies, involving the two nearly degenerate local levels $m = \pm 1$ and the two conduction electron channels with magnetic spin $s = \pm 1/2$, similar to the $B_0 = 0$ case [27]. Although the perturbative RG calculations cannot access this strongly correlated regime, the level crossing occurs in general at an early state of the renormalization, where the perturbative RG calculations are well controlled, so that the occurence of a 2CK fixed point can be safely predicted. Towards low energies, the 2CK behavior will be cut off only at the scale of the Zeeman splitting $2B(D \to 0)$.

After the level crossing has occured in the RG flow, the level renormalizations $\mathrm{Re}\Sigma_{0,\pm1}$ become gradually small, and the level spacings $\Delta(D) \pm B(D)$ become nearly constant. The Zeeman splitting, i.e., the spacing between the $m = +1$ and the $m = -1$ states, remains nearly constant during the entire RG flow. This level flow can be understood from the behavior of the imaginary parts of the impurity self-energies, shown in Fig. 2 b) for the same set of parameters, since real and imaginary parts are related by Kramers-Kronig relations: Before the level crossing occures, the two $m = \pm 1$ states are excited states and, thus, have a large decay rate, proportional to the imaginary parts of their selfenergies, $\mathrm{Im}\Sigma_{\pm1}$, while the on-shell decay rate of the impurity ground state, $m = 0$ is zero (Fig. 2 b), inset). Via Kramers-Kronig, this causes a strong, initial down-renormalization of the $\Delta(D) \pm B(D)$ and, hence, a level crossing. After the level crossing, the $m = -1$ state is the lowest-lying level, and its on-shell decay rate vanishes [blue, dashed line in Fig. 2 b)]. The decay rate of the $m = 0$ state, in turn, starts to grow as the band cutoff D is further reduced, due to the increasing coupling constants $g_{0m}^{(j)}(D)$. This counter-acts the increase of $\Delta(D)$ and causes it to level-off at low energies. Eventually, transitions to the $m = 0$ state are frozen out when the cutoff is reduced below its (renormalized) excitation energy. Below this stage of the renormalization the $m = 0$ decay rate decreases again [black line in Fig. 2 b)]. The decay rate of the Zeeman-split $m = 1$ level [red, dashed line in Fig. 2 b)] follows that of the $m = 0$ level, however shifted towards lower energies, because of its lower excitation energy, $2B(D)$, above the $m = -1$ state. Since for the parameter values of Fig. 2 the initial Zeeman splitting is relatively small compared to Δ, level renormalizations of the

$m = +1$ and the $m = -1$ state are nearly the same, so that the Zeeman splitting $2B$ remains essentially unrenormalized.

The $m = \pm 1$ level renormalization is shown in Fig. 3 a) for various magnetic fields B_0. For stronger magnetic fields the level renormalizations, as well as the decay rates become slightly smaller, because resonant scattering within the $m = \pm 1$ doublet is reduced. For magnetic fields up to the order of the Kondo temperature T_K and not too large initial level spacing Δ_0, both $m = \pm 1$ doublet states cross the $m = 0$ level. This implies 2CK behavior in the energy range $B_0 \lesssim E \lesssim T_K$. However, if $B_0 \gtrsim 10 \cdot T_K$, the level crossing occurs only for one or none of the $m = \pm 1$ states. In this case, the fixed point is a weak coupling potential scattering impurity. These results can be summarized in a phase diagram in terms of the coupling constants J, g and the level spacings Δ_0 and B_0, see Fig. 3 b). The 2CK phase of the system is defined as the regime where both $m = \pm 1$ levels cross the $m = 0$ level. It occurs to the upper left of the phase boundary lines shown in Fig. 3 b) for various B_0. For each value of B_0 the critical Kondo coupling J for realizing the 2CK phase is in good approximation a quadratic function of the level spacing δ_0 [phase boundary lines in Fig. 3 b)]. This is a consequence of the fact that for not too large B_0 the level crossing occurs at an early stage of the RG flow and, hence, may well be described in 2nd-order perturbation theory. Deviations from the quadratic behavior occur for strong magnetic field [blue triangles in Fig. 3 b)], where the level crossing occurs later in the RG flow [see Fig. 3 a)] and 2nd-order perturbation theory is not sufficient. For finite B_0 the critical Kondo coupling is non-zero even for vanishing Δ_0, as expected. However, the 2CK phase is still realized in a wide range of parameters.

4   Discussion and summary

To conclude, we have proposed and studied a three-level quantum impurity model with partially broken SU(3) symmetry, comprised of a degenerate excited-state doublet and a unique ground state, which exhibits a generic two-channel Kondo (2CK) fixed point. This model may be realized by the rotational states of a hydrogen ion (proton) in the interstitial space of the host lattice of a noble metal. It qualitatively explains two seemingly distinct features of the differential conductance experiments by Ralph and Buhrman [11, 12] on the same footing, namely the zero-bias anomaly (ZBA) characteristic of 2CK behavior and the conductance spikes at elevated bias. The ZBA is due to a down-renormalization of the doublet below the non-interacting impurity ground state and Kondo scattering from this doublet, while the spikes at elevated
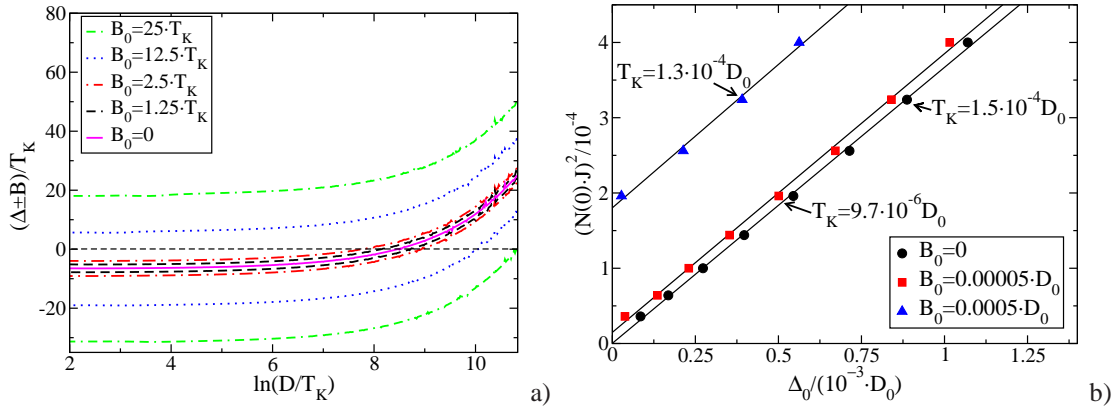

Fig. 3 **a)** Level renormalization for different magnetic fields B_0 for $T_K = 2 \cdot 10^{-5} D_0$, $\Delta_0 = 25 \cdot T_K$ and $J_\perp = J_{||} = 0.015\, N(0)$, $g_{m0}^{(j)} = g_{0m}^{(j)} = J_\perp/2$. The curves of Zeeman-split doublets $m = \pm 1$ for the same magnetic field B_0 are plotted in the same color. **b)** Phase diagram in the plane of $J = J_\perp = J_{||} = g_{m0}^{(j)}$ and Δ_0 for different values of B_0. The symbols mark the boundary between the 2CK phase (upper left) and the potential scattering phase (lower right). The values of T_K given in the figure are for the respective parameter values (Δ_0, J) marked by the arrows. The solid lines are straight line fits to the data points.

bias are due to Kondo-enhanced transitions between impurity ground and excited states [27]. We have discussed that charge screening effects do not reduce the Kondo temperature T_K of the present rotational impurity model, in contrast to the case of two-level systems [24]. Since Kondo coupling and splitting of the doublet are independently controllable parameters of the model, Kondo transitions via virtual excitations of higher excited states are not required in order to enhance the Kondo tmeperature. Rather, real transitions to excited states, induced by finite bias, are expected to lead to multiple conductance spikes [27], which are also observed experimentally [11, 12]. We have also analyzed the effect of a Zeeman splitting of the rotational doublet by an external magnetic field B. A moderately strong field $B < T_K$ does not destroy the 2CK phase of the model, although the 2CK zero-bias conducance anomaly will be cut off at the lowest energies by the Zeeman splitting. This behavior of the model is also observed experimentally [12]. In the three-level model the Zeeman splitting of the $m = \pm 1$ doublet also splits the transition energies $\Delta \pm B$ between the $m = 0$ and the $m = +1$ or $m = -1$ states, respectively, which mark the positions of the finite-bias conductance spikes. A splitting of the conductance spikes in a magnetic field is, therefore, expected if their width is smaller than the Zeeman energy. This will be investigated in forthcoming work [33].

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