Kondo Tunneling through Real and Artificial Molecules

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When an asymmetric double dot is hybridized with itinerant electrons, its singlet ground state and lowly excited triplet state cross, leading to a competition between the Zhang-Rice mechanism of singlet-triplet splitting in a confined cluster and the Kondo effect (which accompanies the tunneling through quantum dot under a Coulomb blockade restriction). The rich physics of an underscreened $S = 1$ Kondo impurity in the presence of low-lying triplet-singlet excitations is exposed and estimates of the magnetic susceptibility and the electric conductance are presented, together with applications for molecule chemisorption on metallic substrates.

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Recent experimental observations [1] of Kondo-type resonance tunneling in quantum dots (QDs) agree with theoretical predictions [2]. In most pertinent experiments the contribution of Kondo tunneling to the conductance in Coulomb blockade windows is observed on QDs with odd occupation and spin $1/2$ ground state (see, however, [3]). Yet, it was pointed out that Kondo tunneling in QD with even occupation is feasible, provided it has soft triplet excitations. These occur both in QD formed by orbitally nondegenerate electrons [4] and in vertical QDs with shell-like structure of electronic states [5–7]. Transition to the Kondo regime must then be controlled by an *external parameter* (here a magnetic field). Indeed, both types of field-induced Kondo tunneling were observed [8].

In this Letter, a novel mechanism is proposed, according to which the transition from a singlet state in a weak coupling regime to a triplet state in a strong coupling regime is shown to be an *intrinsic* property of nanoobjects with even occupation. It is manifested in tunneling through real and artificial molecules in which the electrons are spatially separated into two groups with a different degree of localization. Electrons in the first group are responsible for strong correlation effects (Coulomb blockade), whereas those in the second group are coupled to a metallic reservoir [9]. The closest analog of such "double-shell dot" (DSD) is the Zhang-Rice (ZR) singlet-triplet (S-T) pair which is formed by two holes in Cu-O planes of high-T_c perovskites [10]. The rich physics is then determined by the competition between the ZR mechanism in a confined cluster and the Kondo effect which accompanies the tunneling through the quantum dot under Coulomb blockade confinement [2]. The necessary preconditions under which the singlet ground state changes into a partially screened $S = 1$ Kondo state due to hybridization with metallic leads is the existence of charge-transfer singlet exciton in a DSD. Moreover, unlike QD with odd occupation whose Hamiltonian is mapped on the Kondo-type sd-exchange Hamiltonian with a localized spin $S = 1/2$, DSD in contact with metallic leads can be treated as a *quantum spin rotator* with $S = 1$. Possible realizations of the model are suggested.

A simple model which describes DSD was introduced in [11], hereafter referred to as a "Fulde molecule" (FM). It contains two electrons occupying a potential well which is formed by deep and shallow valleys. The Hamiltonian of an isolated FM is

$$H_d = \sum_i \sum_\sigma \epsilon_i n_{i\sigma} + V \sum_{i\neq j} d^\dagger_{i\sigma} d_{j\sigma} + H_{\rm corr}. \quad (1)$$

Here $d^\dagger_{i\sigma}$ creates a dot electron with spin σ at valley $i = f, l$ while the coupling constant $V = \langle d_l|V|d_f\rangle$ is the interwell tunneling integral. There are two electrons in a neutral ground state. $H_{\rm corr} = Qn_f(n_f - 1)/2$ is the interaction term responsible for the Coulomb blockade of charged states (here $n_f = \sum_\sigma d^\dagger_{f\sigma} d_{f\sigma}$). The energy difference $\Delta = \epsilon_l - \epsilon_f$ is postulated to exceed the overlap integral, $\beta = V/\Delta \ll 1$. Two-electron states $|\Lambda\rangle$ of the FM are classified as a ground state singlet $|S\rangle$, low-lying triplet exciton $|T0\rangle$, $|T\pm\rangle$ and high-energy singlet charge-transfer exciton $|L\rangle$. To order β^2 they are

$$\begin{aligned} |S\rangle &\approx \alpha|s\rangle + \sqrt{2}\,\beta|ex\rangle, \\ |T0\rangle &= \frac{1}{\sqrt{2}} \sum_\sigma d^\dagger_{f\sigma} d^\dagger_{l-\sigma}|0\rangle, \qquad |T\pm\rangle = d^\dagger_{l\pm} d^\dagger_{f\pm}|0\rangle, \\ |L\rangle &\approx \alpha|ex\rangle - \sqrt{2}\,\beta|s\rangle, \end{aligned} \quad (2)$$

where $|s\rangle = \frac{1}{\sqrt{2}} \sum_\sigma \sigma d^\dagger_{l-\sigma} d^\dagger_{f\sigma}|0\rangle$, $|ex\rangle = d^\dagger_{l\uparrow} d^\dagger_{l\downarrow}|0\rangle$, and $\alpha = 1 - \beta^2$. In this order, the energy levels E_Λ are [11]

$$\begin{aligned} E_S = \epsilon_l + \epsilon_f - 2V\beta, \qquad E_T = \epsilon_l + \epsilon_f, \\ E_L = 2(\epsilon_l + V\beta). \end{aligned} \quad (3)$$

The spin and charge branches of the excitation spectrum of FM are characterized by rather different energy scales $E_T - E_S = \delta$ and $E_L - E_S \sim \Delta$, respectively. An interplay between Kondo triplet excitations (with some characteristic energy Δ_K) and ZR triplet excitations is expected when $\delta \sim \Delta_K$ in the regime of Kondo resonance induced by tunneling to metallic reservoir [12].

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The tunneling problem is encoded in the Anderson Hamiltonian which incorporates H_d, together with the band Hamiltonian $H_b = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma}$ for the electrons in the leads, and the tunneling term $H_t = \sum_{ik\sigma} W_i c^\dagger_{k\sigma} d_{i\sigma}$. Here $c_{k\sigma}$ are operators for lead electrons and $W_{i=l,f}$ are tunneling matrix elements, taken to be k independent. The asymmetry of orbitals is also reflected in tunneling: the strong localization of the f orbitals justifies the assumption $W_l \neq 0$ but $W_f = 0$. Next, the dot operators $d^\dagger_{i\sigma}$ are conveniently expressed in terms of Hubbard operators, $X^{\Lambda\lambda} = |\Lambda\rangle\langle\lambda|$. Here $\Lambda = S, T, L$ stands for the neutral two-electron states (2), and the index $\lambda = 1\sigma, 3\sigma$ is reserved for the *charged* states $|1\sigma\rangle \approx \sqrt{1-\beta^2}\,|f\sigma\rangle + \beta|l\sigma\rangle$, $|3\sigma\rangle \approx d^\dagger_{f\sigma}|ex\rangle - \frac{V}{Q-\Delta} d^\dagger_{f\sigma} d^\dagger_{f\bar\sigma} d^\dagger_{l\sigma}|0\rangle$. The tunnel matrix elements in the Hubbard representation are given as $W^{\Lambda\lambda}_\sigma = \langle k\sigma, \lambda|\hat W|\Lambda\rangle$, where $\hat W$ is the operator responsible for tunneling. The Anderson Hamiltonian then reads

$$H = \sum_\Lambda E_\Lambda X^{\Lambda\Lambda} + \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{\Lambda\lambda} (W^{\Lambda\lambda}_\sigma c^\dagger_{k\sigma} X^{\lambda\Lambda} + \bar W^{\Lambda\lambda}_\sigma X^{\Lambda\lambda} c_{k\sigma}). \quad (4)$$

Using the Wigner-Eckart theorem, one can write $W^{\Lambda\lambda}_\sigma = C^\Lambda_{\sigma\lambda} A_\lambda$, where $C^\Lambda_{\sigma\lambda}$ are Clebsh-Gordan coefficients and A_λ is the reduced matrix element. In a given vector-coupling scheme the tunneling results in the following transitions: $|T\pm\rangle \leftrightarrow |1\pm, p\pm\rangle$ (1); $|T0\rangle \leftrightarrow |1\sigma, p\bar\sigma\rangle$ (2); $|S\rangle \leftrightarrow |1\sigma, p\bar\sigma\rangle$ (3); $|S\rangle, |T0\rangle \leftrightarrow |3\sigma, k\bar\sigma\rangle$ (4); $|T\pm\rangle \leftrightarrow |3\pm, k\mp\rangle$ (5). Here $p\sigma$ and $k\sigma$ are, respectively, the states with an excess electron (and hole) above (below) the Fermi level of the lead. The energy costs of these transitions are

$$\begin{aligned} E_{1p,S} &= \epsilon_p - \epsilon_l + \beta V, \qquad E_{1p,T} = \epsilon_p - \epsilon_l, \\ E_{3k,S} &= \epsilon_l + 4\beta V + \tilde Q - \epsilon_k, \\ E_{3k,T} &= \epsilon_l + 2\beta V + \tilde Q - \epsilon_k, \end{aligned} \quad (5)$$

where $\tilde Q \approx Q[V^2/(Q-\Delta)^2]$. It is assumed that the Coulomb blockade eliminates three-electron states $|3\sigma\rangle$ and the tunnel coupling involves only the states $|1\sigma\rangle$ (processes 1, 2, and 3). The corresponding tunnel matrix elements are

$$W_1 \equiv W, \qquad W_2 = \frac{1}{\sqrt{2}} W, \qquad W_3 = \pm\frac{\alpha}{\sqrt{2}} W. \quad (6)$$

The interplay between the singlet and triplet levels of the DSD is analyzed by the renormalization group (RG) method guided by the "poor man's scaling" approach to the Anderson model [13]. The renormalized levels E_Λ are determined by the equations

$$dE_\Lambda/d\ln D = \Gamma_\Lambda/\pi. \quad (7)$$

Here Γ_Λ are the tunnel coupling constants,

$$\Gamma_T \equiv \Gamma = \pi\rho_0|W|^2, \qquad \Gamma_S = \alpha^2\Gamma, \qquad \rho_0 \sim D^{-1}. \quad (8)$$

Integrating (7) we find the scaling invariants E^*_Λ which determine the scaling trajectories

$$E^*_\Lambda = E_\Lambda - \frac{\Gamma_\Lambda}{\pi} \ln\left(\frac{\pi D}{\Gamma_\Lambda}\right). \quad (9)$$

The level ϵ_f is taken to be close to the bottom of the conduction band [12], so that scaling does not significantly affect it. It is then subtracted from the energies E_T and E_S. Now we see that the energies E_Λ decrease together with D. Since $\Gamma_T > \Gamma_S$, the phase trajectory $E_T(D, \Gamma_T)$ should cross that of $E_S(D, \Gamma_S)$ at a certain point. Thus, quite remarkably, there is a crossover from singlet to triplet ground state of the FM due to tunnel contact with metallic leads. At the crossing point $E_T = E_S$ Eq. (9) implies $\pi(E^*_T - E^*_S) \approx (\Gamma_T - \Gamma_S)\ln(\pi D/\Gamma)$, and the renormalized bandwidth at this crossing point is $\tilde D = D\exp(-\pi\Delta/\Gamma)$. Another important crossing point is the solution of $\bar D \approx \epsilon_l(\bar D)$. Here scaling of the levels $E_{T,S} = \bar E_{T,S}$ (and hence charge fluctuations) becomes irrelevant. This is the Schrieffer-Wolff (SW) limit where only spin fluctuations are responsible for scaling of the Hamiltonian [13]. If $\tilde D > \bar D$, then

$$\bar\delta \equiv \bar E_T - \bar E_S < 0. \quad (10)$$

The SW regime is reached *after* S-T crossover, and a Kondo resonant tunneling is feasible. In the opposite case there is a singlet ground state and a soft triplet excitation at energy $\bar\delta > 0$ (see Fig. 1). The Kondo regime is still accessible if a properly tuned *external* parameter induces the pertinent crossover. This might be a magnetic [4] or an electric field (gate voltage) shifting the dot level ϵ_l upward relative to ε_F (Fig. 1, inset), an experimental aspect not yet explored.

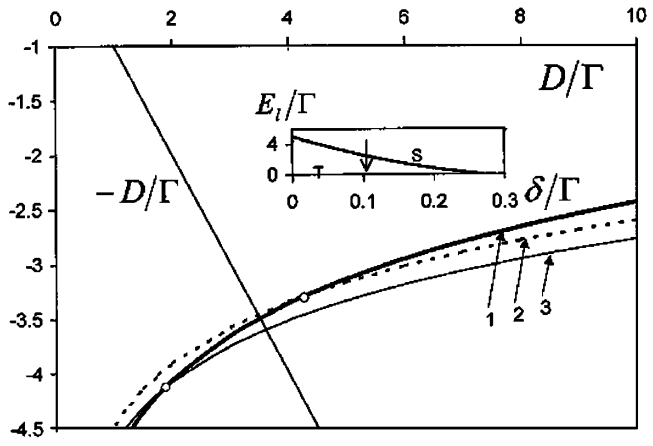

FIG. 1. Scaling trajectories [Eq. (7)] demonstrating crossovers from singlet to triplet ground state: $E_T(D)$ (curve 1), $E_S(D)$ for $\delta/\Gamma = 0.03, 0.1$ (curves 2, 3, respectively) at $\Gamma_S/\Gamma_T = 0.8$, $D_0/\Gamma = 10$. Inset: S-T transition as a function of the level depth $E_l = \varepsilon_F - \epsilon_l$ with changing initial level splitting δ.

The physically richer case of triplet ground state ($\bar{\delta} < 0$) is analyzed below. For a two-electron FM the SW transformation [13] projects out the states $|\lambda, k\sigma\rangle$ and maps H onto an effective Hamiltonian acting in two-electron subspace and reduced conduction band, $H_{\rm eff} = H^S + H^T + H^{ST}$, with

$$\begin{aligned} H^S &= \bar{E}_S X^{SS} + J^S \sum_\sigma X^{SS} c_\sigma^\dagger c_\sigma \,, \\ H^T &= \bar{E}_T \sum_\mu X^{\mu\mu} + J^T \mathbf{S}\cdot\mathbf{s} + \frac{J^T}{2} \sum_{\mu\sigma} X^{\mu\mu} c_\sigma^\dagger c_\sigma \,, \\ H^{ST} &= J^{ST}(\mathbf{P}\cdot\mathbf{s}) \end{aligned} \tag{11}$$

(in X^{SS}, $X^{\mu S}$, and $X^{\mu\mu'}$, S indicates singlet and $\mu = 0, \pm$ stands for triplet spin states). The effective exchange integrals are $J^T = -2|W|/(\varepsilon_F - \bar{\epsilon}_l)$, $J^S \approx \alpha^2 J^T$, and $J^{ST} = \alpha J^T$. The electron operators appearing in (11) are $c_\sigma = \sum_k c_{k\sigma}$; $\mathbf{s} = 2^{-1/2} \sum_{\sigma\sigma'} c_\sigma^\dagger \hat{\tau} c_{\sigma'}$, in which $\hat{\tau}$ are the Pauli matrices. The singlet and triplet states are now intermixed, and the spin properties of FM are characterized by the vector operators $\mathbf{S}$ and $\mathbf{P}$ in accordance with the dynamical symmetry of the spin rotator:

$$\begin{aligned} S^+ &= \sqrt{2}\,(X^{+0} + X^{0-}), \qquad S^- = \sqrt{2}\,(X^{0+} + X^{-0}), \\ S_z &= X^{++} - X^{--}, \qquad P_z = -(X^{0S} + X^{S0}), \\ P^+ &= \sqrt{2}\,(X^{+S} - X^{S-}), \qquad P^- = \sqrt{2}\,(X^{S+} - X^{-S}). \end{aligned} \tag{12}$$

These operators obey the moment algebra ($i = x, y, z$):

$$\begin{aligned} [P^i, P^j] = i\varepsilon_{ijk} S^k, \qquad [P^i, S^j] = i\varepsilon_{ijk} P^k, \\ \mathbf{S}\cdot\mathbf{P} = 0 \,, \end{aligned} \tag{13}$$

and the Casimir operator is $S^2 + P^2 = 3$. Surprisingly, this special representation of $O(4)$ played an important role in particle physics many years ago [14].

The pertinent physics is that of an underscreened Kondo impurity [15] in the presence of potential scattering and low-lying triplet-singlet excitations. A similar model was considered recently in Ref. [6] studying the physics of tunneling through vertical QD in a magnetic field [8]. In that case, the electron orbital motion in a plane perpendicular to the axis of the dot is characterized by the same quantum number both in the dot and in the leads [7], and two orbitals participate in the S-T transitions. The models studied here and in Refs. [6,7] can be mapped onto a two-spin 1/2 Kondo model. The fictitious spin representation of Ref. [7] is alternative to that of (12), but the latter seems more compact as it respects the dynamical symmetry of the Hamiltonian. In particular, in the two-spin representation $H_{\rm eff}$ includes four invariants, whereas Eqs. (11) contain just a couple.

Following [13] the poor man scaling approach [16] is now applied to the Hamiltonian $H_{\rm eff}$ (11). Neglecting the irrelevant potential scattering phase shift [17], a system of scaling equations is obtained (cf. [6])

$$dj_1/d\ln d = -[(j_1)^2 + (j_2)^2], \qquad dj_2/d\ln d = -2j_1 j_2 \tag{14}$$

(here $j_1 = \rho_0 J^T$, $j_2 = \rho_0 J^{ST}$, and $d = \rho_0 D$). The corresponding RG flow diagram has the fixed point $j_1 = \infty$, but the resulting Kondo temperature $T_K(\bar{\delta})$ turns out to be a sharp function of $\bar{\delta}$ [6]. It is maximal when the T, S states are quasidegenerate, $|\bar{\delta}| \ll T_K(\bar{\delta})$. The scaling in this case is governed by the effective integral $j_+ = j_1 + j_2$, and the system (14) is reduced to a single equation

$$dj_+/d\ln d = -(j_+)^2 \tag{15}$$

with $T_{K0} = \bar{D}\exp(-1/j_+)$. In the opposite limit $|\bar{\delta}| \gg T_K(\bar{\delta})$ the scaling of J^{ST} stops at $D \simeq |\bar{\delta}|$. Then $j_{1,2}(\bar{\delta}) = j_{1,2} \ln^{-1}(\frac{|\bar{\delta}|}{T_{k0}})$ and $T_K(\bar{\delta}) = |\bar{\delta}|\exp[-1/j_1(\bar{\delta})] \ll T_{K0}$. The singlet ground state S with zero T_K is realized at positive $\bar{\delta} > T_K(\bar{\delta})$.

Natural candidates for DSD are lanthanocene molecules $\mathrm{Ln(C_8H_8)_2}$ adsorbed on metallic substrate. Here the mixed valent ion Ln = Ce, Yb is sandwiched between two π-bonded carbon rings [18]. In these molecules the electrons in a strongly correlated f shell are coupled with loosely bound π electrons. The ground state of this molecule is a spin singlet combination $^1A_{1g}(f\pi^3)$ of an f electron and π orbitals, and the energy of the first excited triplet state $^3E_{2g}$ is rather small ($\sim$0.5 eV). In the ytterbocene (hole counterpart of cerocene) the ground state with one f hole is a triplet, and the gap for a singlet excitation is tiny, $\sim$0.1 eV. The fullerenelike molecules doped with Ce or Yb form another family with apparently similar properties. In all these systems there is no direct overlap between the strongly correlated f electrons and the metallic reservoir. However, these electrons can influence the tunnel properties of the molecule via covalent bonding with the outer π electrons which are coupled to the reservoir.

Artificial candidates are double-dot structures (say D_1 and D_2) in tunneling contact with each other, but only D_1 is coupled with the metallic leads. The respective gate voltages are such that $V_{g1} < V_{g2}$. Coulomb blockade then prevents double charging of D_2, so it can play the same role as a $4f$ atom in molecular complexes described above. The dot D_1 donates the loosely bound electrons which contribute to the tunnel current [19].

The salient features of FM stem from the qualitative dependence of its ground state and low-energy spectrum on the coupling constants V and W. The unusual S-T crossing should show up in the magnetic properties of adsorbed molecules and tunnel transparency of asymmetric double quantum dots.

According to quantum chemical calculations of the energy spectrum of the isolated cerocene molecule, the Van Vleck paramagnetic contribution of S-T excitations is too weak to overcome the Larmor diamagnetic contribution of C_8H_8 rings [11,18]. This situation can drastically change for a FM adsorbed on a metallic layer. The

fixed point $j_1 = \infty$ corresponds to the scattering phases $\eta_\sigma(\epsilon_F) = \pi/2$. In the case of adsorbed FM this means that the molecule has a residual spin $1/2$ which interacts ferromagnetically with the conduction electrons [15]. The temperature dependence of magnetic susceptibility $\chi(T)$ is predetermined by the energy parameters $\bar{\delta}$ and $T_K(\bar{\delta})$. In particular, $\chi(T)$ conserves its Curie-like character down to the lowest temperatures when $\bar{\delta} < 0$ and $|\bar{\delta}| \gg T_K(\bar{\delta})$. Then at $T \ll T_K(\bar{\delta})$ the underscreened FM remains paramagnetic, and its susceptibility is

$$\chi(T) = \chi_0(T)[1 - Z(T/T_K)]. \tag{16}$$

Here $\chi_0 = 3C/4T$, $C = (g\mu_b)^2$, and $Z(x)$ is the invariant coupling function (solution of the Gell-Mann–Low equation [20]). At $T > T_K(\bar{\delta})$ the triplet spin state is restored and the Kondo corrections as well as admixture of singlet state can be calculated by perturbation theory,

$$\begin{aligned} \chi(T) &= \frac{2C[3 - \exp(-|\bar{\delta}|/T)]}{9T} \\ &\quad \times \left(1 - \ln^{-1}\frac{T}{T_K(\bar{\delta})} - j_2 \ln\frac{\bar{D}}{|\bar{\delta}|}\right), \\ \chi(T) &= \frac{2C}{T[3 + \exp(-|\bar{\delta}|/T)]}\left(1 - \ln^{-1}\frac{T}{T_{K0}}\right), \end{aligned} \tag{17}$$

respectively, for $|\bar{\delta}| \gg T_{K0}$ and $|\bar{\delta}| \ll T_{K0}$.

In an artificial FM, resonant scattering phase means perfect tunneling transparency of the QD at $T = 0$ and a logarithmic falloff at high temperatures. To calculate the tunneling transparency of FM sandwiched between two leads, one should add an index $n = L, R$ to the operator $c_{nk\sigma}$ and switch to the standing wave basis [2] $\sqrt{2}\, c_{k\sigma\pm} = c_{Lk\sigma} \pm c_{Rk\sigma}$. Only the wave $(+)$ contributes to the current, and the zero bias anomaly in the differential conductance $G(T)$ (due to Kondo cotunneling) in the weak coupling regime $T > T_K(\bar{\delta})$ is found as in (17):

$$\begin{aligned} G/G_0 &= 2\ln^{-2}[T/T_K(\bar{\delta})] + j_1 j_2^2 \ln(\bar{D}/|\bar{\delta}|), \\ G/G_0 &= 3\ln^{-2}[T/T_{K0}], \end{aligned} \tag{18}$$

respectively, for $|\bar{\delta}| \gg T \gg T_K(\bar{\delta})$ and $T \gg T_{K0} \gg |\bar{\delta}|$. Here $G_0 = 4\pi e^2/\hbar$. Again the maximum effect is achieved in a nearly degenerate case. At $T \to 0$ the conductance tends to the unitarity limit.

In conclusion, the central result is that the interplay between ZR-type coupling in real and/or artificial molecules and Kondo coupling between molecules and metallic reservoir may result in a crossover from a singlet spin state in a weak coupling regime to an underscreened $S = 1$ state at zero temperature. It was also pointed out that the onset of Kondo regime in double quantum dot with even occupation can be driven also by a gate voltage. This adds new perspectives beyond earlier suggestions of reaching the Kondo regime using magnetic field.

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