

Phonon-assisted tunneling and two-channel Kondo physics in molecular junctions

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The interplay between vibrational modes and Kondo physics is a fundamental aspect of transport properties of correlated molecular conductors. We present theoretical results for a single molecule in the Kondo regime connected to left and right metallic leads, creating the usual coupling to a conduction channel with left-right parity (even). A center-of-mass vibrational mode introduces an additional phonon-assisted tunneling through the antisymmetric (odd) channel. A non-Fermi-liquid fixed point, reminiscent of the two-channel Kondo effect, appears at a critical value of the phonon-mediated coupling strength. Our numerical renormalization-group calculations for this system reveal non-Fermi-liquid behavior at low temperatures over lines of critical points. Signatures of this strongly correlated state are prominent in the thermodynamic properties and in the linear conductance.

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I. INTRODUCTION

Ground-breaking experimental results in single molecular transistors during the last decade¹ have greatly expanded the field of molecular electronics, opening several possibilities for technological applications and investigations of fundamental aspects of the physics of these devices. By now it is established that strong correlation effects play a key role in the electronic transport through these systems, as evidenced by the observation of the Kondo effect² in both break junctions^{3,4} and scanning tunneling microscope setups.⁵ A clear understanding of the mechanisms involved in the emergence of the Kondo effect in molecular systems is thus of primary importance.

One possibility to advance our knowledge of these devices is by establishing analogies with the well-known transport properties of semiconductor quantum dots in the Kondo regime. This, however, proves to be a challenging approach for several reasons: molecule-lead couplings are very sensitive to the particular configurations, charging energies are significantly larger, and, more importantly, deformations and vibrational modes in the molecule play an active role in transport.⁶

This variety of competing effects also brings theoretical challenges, such as the interesting prospect of investigating the interplay between vibronic states and Kondo physics. Different studies have investigated the effect of electron-phonon couplings in the charge degrees of freedom of the molecule, affecting the exchange correlations leading to the Kondo effect.⁷⁻⁹ This issue has also been highlighted in recent experiments reporting anomalous behavior in the Kondo transport⁴ which have been attributed to the “dressing” of the local energies by Holstein-type phonons.⁷

In addition to these local effects, considerable attention has been given to effects of vibrational modes in the *tunneling* from the molecule to the leads. Phonon-assisted couplings by “breathing”^{10,11} or “center-of-mass”^{10,12-14} molecular modes create additional correlations with the electrons in the leads. Such phonon-mediated tunneling processes will, in general, lead to novel features in the transport properties¹⁰⁻¹⁴

and can be experimentally probed by conductance measurements.

We address this subject in the present work by investigating a two-channel Kondo (2chK) effect^{15,16} in molecular systems with center-of-mass vibrational modes. Two-channel Kondo physics, originally investigated in the context of heavy-fermion materials,^{15,17} has been an active topic in the area of nanostructures. In semiconductor quantum dots, several theoretical predictions¹⁸ and a recent experimental observation¹⁹ of the 2chK have highlighted the renewed interest in such strongly correlated states. In addition, phonon-assisted 2chK behavior has been predicted in effectively non-interacting systems (e.g., metallic carbon nanotubes²⁰ and metallic break junctions²¹) coupled to vibronic states with Kondo-type correlations appearing in orbital (pseudospin) degrees of freedom.^{20,21}

In this paper, we consider the Kondo regime of a singly charged molecular level connected to metallic leads, fully including both electron-electron and electron-phonon interactions, as well as phonon-assisted tunneling processes arising from a center-of-mass vibronic mode. Our numerical renormalization-group (NRG) calculations show that the presence of the extra phonon-mediated conductance channel leads to NFL behavior at low temperatures, with prominent signatures in the thermodynamic properties and in the linear conductance. The 2chK fixed point occurs over lines of critical points covering a wide range of parameters, both away from particle-hole symmetry and in the presence of deformation-induced charge-phonon couplings.

The paper is organized as follows: the model is presented in detail in Sec. II, and a discussion on the 2chK regime and the dependence of the critical parameters is given in Sec. III. In Sec. IV, we discuss the NFL signatures in the transmission phase shift and in the conductance across the junction. We give our concluding remarks in Sec. V.

II. MODEL

We consider a molecular complex (e.g., an organometallic compound, C_{60} , etc.) in the Coulomb blockade (CB) regime

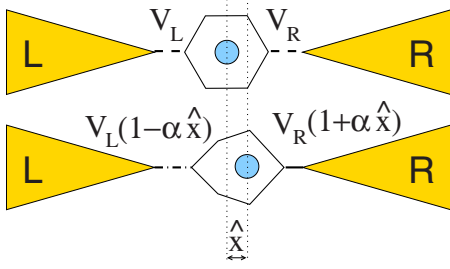


FIG. 1. (Color online) Schematic illustration of the phonon-assisted tunneling process: the coupling of a molecular level to left and right metallic leads is modulated by a center-of-mass vibrational mode.

connected to metallic leads (for instance, in a metallic break junction setup). Kondo correlations appear as the molecule is tuned into a CB valley with an odd number of electrons by a plunger gate voltage. We focus on the low-bias regime, in which electronic transport is dominated by a singly-occupied molecular level of energy ϵ_d (measured from the Fermi energy in the leads and tunable by the gate voltage) with a charging energy U arising from the electron-electron interactions within the molecule.

In our model, the molecule is connected to left (L) and right (R) leads by tunneling couplings proportional to the overlap between the wave functions of the molecular level and the (s -like) metallic states in the leads. More importantly, we consider the effect of phonon-mediated coupling through a center-of-mass (c.m.) vibrational mode, as illustrated in Fig. 1. For small displacements, the molecule-lead tunneling can be effectively written as $V_{L(R)}(1 \pm \alpha \hat{x})$, where \hat{x} is a displacement operator in the direction of the motion and α is a system-specific parameter, being essentially proportional to the ratio between the overlap length and the oscillation amplitude of the vibrational mode. Additionally, deformations in the chemical bonding (also illustrated in Fig. 1) will, in general, introduce an effective coupling of the center-of-mass oscillating mode and the charge state of the molecule. We model this by a Holstein-type electron-phonon term with coupling λ .

The full Hamiltonian is given by

$$H = H_M + H_{\text{Leads}} + H_{\text{Dot-Leads}}, \quad (1)$$

where

$$\begin{aligned} H_M &= \epsilon_d n_{d\sigma} + U n_{d\uparrow} n_{d\downarrow} + \lambda(1 - n_d)(a + a^\dagger) + \omega_0 a^\dagger a, \\ H_{\text{Dot-Leads}} &= \sum_{\mathbf{k}} V_L(1 - \alpha \hat{x}) d_{\sigma}^\dagger c_{L\mathbf{k}\sigma} + \text{H.c.} \\ &\quad + V_R(1 + \alpha \hat{x}) d_{\sigma}^\dagger c_{R\mathbf{k}\sigma} + \text{H.c.}, \\ H_{\text{Leads}} &= \sum_{\mathbf{k}, \ell=L,R} \epsilon_{\ell\mathbf{k}} c_{\ell\mathbf{k}\sigma}^\dagger c_{\ell\mathbf{k}\sigma}. \end{aligned} \quad (2)$$

In the above, d_{σ}^\dagger (d_{σ}) and $c_{\ell\mathbf{k}\sigma}^\dagger$ ($c_{\ell\mathbf{k}\sigma}$) are fermionic operators that create (destroy) electrons with spin σ in the molecule and leads, respectively, ($n_{d\sigma} = d_{\sigma}^\dagger d_{\sigma}$ is the electron number operator), and ω_0 is the frequency of the local center-of-

mass phonon mode, with a^\dagger (a) being the phonon operators ($\hat{x} = a + a^\dagger$). We assume the wide band limit and \mathbf{k} -independent dot-lead couplings ($V_{\ell\mathbf{k}} \equiv V_{\ell}$).

Hamiltonian (1) can be written as an Anderson impurity model coupled to two independent fermionic channels. Defining symmetric (even) and antisymmetric (odd) combinations of the electronic operators in the left and right leads $c_{e(o)\mathbf{k}\sigma} \equiv (V_R c_{R\mathbf{k}\sigma} \pm V_L c_{L\mathbf{k}\sigma}) / 2\sqrt{V_L^2 + V_R^2}$, the $H_{\text{Dot-Leads}}$ term in Eq. (1) becomes

$$H_{\text{Dot-Leads}} = \bar{V} \sum_{\mathbf{k}, \sigma} d_{\sigma}^\dagger c_{e\mathbf{k}\sigma} + \alpha(a + a^\dagger) d_{\sigma}^\dagger c_{o\mathbf{k}\sigma} + \text{H.c.}, \quad (3)$$

where $\bar{V} \equiv 2\sqrt{V_L^2 + V_R^2}$. For $\alpha \neq 0$, a phonon-mediated coupling to the odd channel is present. As we shall see, this has important consequences in the physics of the ground state of the system. Notice that the odd-channel coupling is present even for molecules not symmetrically coupled to the leads (i.e., $V_L \neq V_R$), a more likely configuration in experiments. For $\alpha=0$, this term vanishes and Hamiltonian (1) corresponds to the single-channel Anderson-Holstein model, previously investigated with NRG⁸ and analytical renormalization-group methods.⁹

The eigenstates of Hamiltonian (1) can be labeled by total charge and SU(2) spin symmetry (Q, S). At the molecule site, states are also labeled by the number of phonons m [$(a^\dagger)^m |0\rangle = |m\rangle$]. Notice that both the electron-phonon ($\propto \lambda$) and the phonon-assisted tunneling ($\propto \alpha$) terms couple $|m\rangle$ and $|m \pm 1\rangle$ states. This last term couples only states with a difference of one electron in the odd channel and, thus, commutes with a generalized *parity* operator, defined by $\hat{P} = (-1)^{m+Q_o+1}$, where Q_o is the total charge in the odd channel. Therefore, for $\lambda=0$, Hamiltonian (1) has an additional O(1) parity symmetry (which is lost for $\lambda \neq 0$, as the electron-phonon term will couple states with different parity).

We solve the two-channel problem with Wilson's NRG technique²² adapted to include the phonon degrees of freedom.⁸ As it is standard in the NRG method, we map Eq. (1) into a (two-band) tight-binding Hamiltonian by performing logarithmic discretizations of the continuum spectra in the even and odd fermionic channels. In the calculations, we use a discretization parameter $\Lambda=3$ and keep up to 2500 states in the NRG iterations, which proves to be adequate for the calculation of thermodynamic properties. We have used a cutoff of $N_{\text{ph}}=9$ in the maximum number of phonons and checked for convergence of the results with N_{ph} . We assume a constant (metallic) density of states ρ_0 in the leads with bandwidth D .

III. TWO-CHANNEL KONDO PHYSICS

From the NRG spectra, we calculated thermodynamic properties for this system. A particularly revealing quantity is the contribution to the total entropy coming from the ‘‘impurity’’ degrees of freedom (in the present case, the molecule), defined as the difference between the total entropy $S(T)$ and the entropy calculated in the absence of the molecule $S^{(0)}(T)$. As it has been shown by Bethe ansatz²³ and NRG calculations²⁴ in two-channel Kondo models, $S_{\text{imp}}(T) \equiv S(T)$

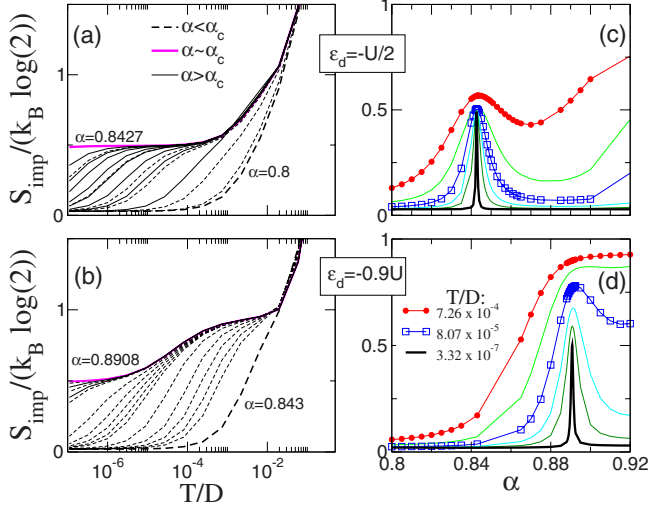


FIG. 2. (Color online) [(a) and (c)] Impurity contribution to the entropy S_{imp} at the particle-hole symmetric point $\epsilon_d = -U/2$ and [(b) and (d)] at $\epsilon_d = -0.9U$. In both cases, an unstable 2chK fixed point for which $S_{\text{imp}} \rightarrow k_B \log 2^{1/2}$ as $T \rightarrow 0$ is reached at $\alpha = \alpha_c$. Panels (a) and (b) show S_{imp} versus T/D for fixed α while (c) and (d) show S_{imp} versus α for fixed temperatures.

$-S^{(0)}(T)$ reaches a universal low-temperature plateau at $S_{\text{imp}}/k_B = 1/2 \ln(2)$ (k_B is Boltzmann's constant) when both channels are equally coupled to the impurity.

Figure 2 shows S_{imp} versus temperature T/D and α for two values of the molecular level energy: $\epsilon_d = -U/2$, corresponding to the particle-hole symmetric point (top panels), and $\epsilon_d = -0.9U$ (bottom panels).²⁵ In both cases, an entropy plateau $S_{\text{imp}}/k_B = 1/2 \ln(2)$ is reached at low temperatures [Figs. 2(a) and 2(b)], signaling the presence of a NFL fixed point.

This NFL fixed point is reached as α approaches a critical value α_c and at temperatures below a characteristic crossover energy scale T^* . Deviations from $\alpha = \alpha_c$ drive the system away from this state to the more conventional Kondo-screened state (characterized by $S_{\text{imp}} = 0$), illustrating the unstable nature of the fixed point. This is depicted in Fig. 2(c), showing S_{imp} versus α at fixed temperatures. At lower temperatures, a narrow peak of height $S_{\text{imp}} = 1/2 \ln 2$ pinpoints the critical value $\alpha = \alpha_c$. This marks the position of the fixed point in parameter space. At higher temperatures, the broadening of these peaks indicate that signatures of NFL behavior extend over a finite range of α .

This behavior persists away from particle-hole symmetry, as shown in Figs. 2(b) and 2(d). Interestingly, the critical value α_c increases as the system approaches the mixed-valence regime ($\epsilon_d \rightarrow 0, -U$; $\langle n_d \rangle \rightarrow 0, 2$) as compared with the $\epsilon_d = -U/2$ case ($\langle n_d \rangle = 1$). This contrasts with the (phonon-independent) two-channel Anderson model (2chAM), for which the critical couplings are ϵ_d independent.²⁴ The crossover energy scale to the NFL fixed point, related to the Kondo temperature, decreases exponentially as the system enters the mixed-valence regime, as in the 2chAM.

In the mixed-valence regime, the system flows into a Fermi-liquid (FL) fixed point with $S_{\text{imp}} = k_B \ln(2)$ at higher

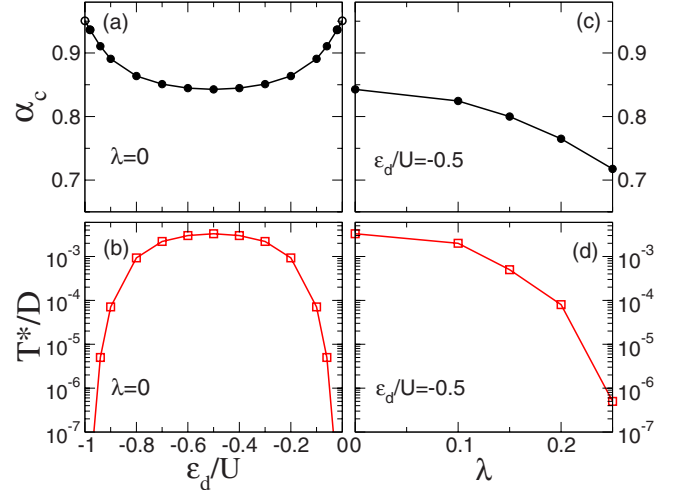


FIG. 3. (Color online) (a) Critical dot-lead coupling α_c and (b) crossover temperature T^* vs molecule level position ϵ_d . [(c) and (d)] Same quantities but now as a function of the electron-phonon coupling λ at $\epsilon_d = -U/2$.

temperatures before entering the NFL regime [as seen, e.g., in Fig. 2(b)]. This fixed point is nonmagnetic and characterized by a *parity* degeneracy in the ground state (rather than the usual spin degeneracy in similar models).

As previously discussed, the S_{imp} vs T/D curves display the NFL plateau at low temperatures for $-U < \epsilon_d < 0$ at a critical $\alpha = \alpha_c$. The critical value α_c increases as the system is moved away from particle-hole symmetry, reaching its highest values at $\epsilon_d = 0^-, -U^+$, as depicted in Fig. 3(a). In addition, at $\alpha = \alpha_c$ the crossover temperature sharply decreases as the systems are driven away from the particle-hole symmetric point. This is illustrated in Fig. 3(b), where we define the crossover temperature T^* as $S_{\text{imp}}(T = T^*) = 3/4 \ln 2$. This panel shows that T^* decreases exponentially as $\epsilon_d \rightarrow -U, 0$, indicating that the 2chK is only reached within the range of gate voltages for which the molecular level is singly occupied. In fact, we find the NFL fixed point in this “local-moment” range ($-U < \epsilon_d < 0$) only, indicating that, along with susceptibility calculations, the Kondo screening occurs in the “real spin” as opposed to a “pseudospin” degree of freedom (e.g., those connected to charge states, or $|m\rangle$ and $|m \pm 1\rangle$ phonon states). In this range, the value α_c varies nearly quadratic with ϵ_d , as shown in Fig. 3(a).

Furthermore, for fixed ϵ_d , both α_c and T^* decrease as the electron-phonon coupling λ increases [Figs. 3(c) and 3(d)]. The decrease in T^* is consistent with the fact that the electron-phonon coupling renormalizes the electron-electron interaction as $U_{\text{eff}} \approx U - 2\lambda^2/\omega_0$,⁸ effectively driving the system into the mixed-valence regime. The decrease in α_c with λ indicates that not only the molecule-lead couplings but also the coupling of the vibrational mode with the *charge state* of the molecule plays a role in the mechanism leading to the two-channel Kondo effect.

IV. PHASE SHIFTS AND CONDUCTANCE

We now turn to the transport properties across the molecule. We first point out that the presence of the c.m. phonon

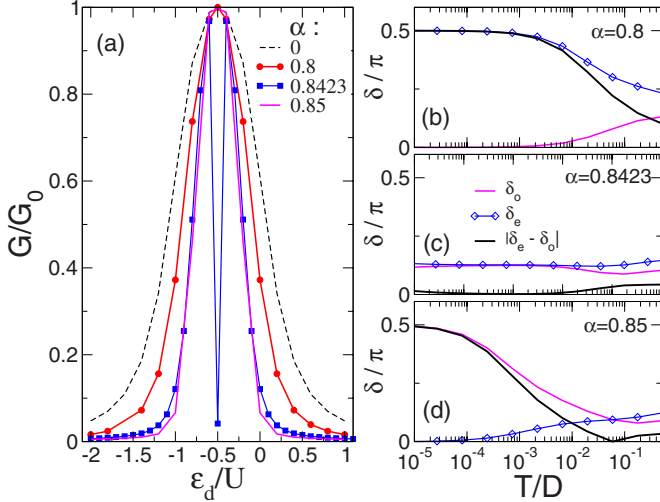


FIG. 4. (Color online) (a) Conductance G versus ε_d for different values of α . A dip appears for $\alpha = \alpha_c$ at $\varepsilon_d = -U/2$, signaling the 2chK state. [(b)–(d)] Even (δ_e), odd (δ_o), and relative $|\delta_e - \delta_o|$ phase shifts vs temperature at $\varepsilon_d = -U/2$ and different values of α .

term in Eq. (1) breaks the “proportionate coupling” condition²⁶ between left and right leads (even in a rather unrealistic symmetric coupling configuration $V_L = V_R$). For this reason, a calculation of the linear conductance via a Landauer-type formula²⁶ would involve not only the local interacting retarded Green’s function (obtainable from NRG) but also Keldysh Green’s functions.

Instead, we turn to the equivalent approach of calculating the $T=0$ conductance using the scattering phase shifts,^{14,21,27} as $G = G_0 \sin^2(\delta_e - \delta_o)$, where δ_e and δ_o are the phase shifts in the even and odd channels, respectively, and $G_0 = (2e^2/h) \sin^2 \theta$ with θ being an overall phase that depends on the microscopic details of the molecule-lead junction (we henceforth consider $\theta = \pi/2$).

The phase shifts $\delta_{e(o)}$ can be obtained from the NRG spectra^{28,29} using the (Q, S, P) quantum numbers to label the states.²¹ We note that the parity quantum number is strictly conserved only for $\lambda=0$. Although it is possible, in some cases, to calculate the difference $\delta_e - \delta_o$ from the (Q, S) NRG spectra,^{14,29} in the following we use $\lambda=0$ as it retains most of the interesting physics. We should note that, away from FL fixed points, the correspondence between the excitations in the NRG spectra and the phase shifts entering the conductance formula is only approximate. Nevertheless, we expect the conductance obtained with this prescription to give a qualitatively accurate picture in the NFL state as well, as discussed below.

Results for the conductance are shown in Fig. 4(a). For $\alpha=0$, the familiar shape is recovered: $G = G_0$ at the particle-hole symmetric point $\varepsilon_d = -U/2$ and $G \rightarrow 0$ as $|\varepsilon_d|$ increases. As α increases, the peak narrows with $G = G_0$ at the p - h symmetric point for $\alpha < \alpha_c$ and $\alpha > \alpha_c$. Interestingly, as α approaches the critical value α_c at $\varepsilon_d = -U/2$, a dip appears in the conductance curve.

This is an indication of the NFL behavior and a signature of the two-channel fixed point. The behavior of individual

phase shifts at the particle-hole symmetric point is illustrated in Figs. 4(b)–4(d). For $\alpha < \alpha_c$ [Fig. 4(b)], $\delta_e \rightarrow \pi/2$ and $\delta_o \rightarrow 0$ at low temperatures, indicating a decoupling of the odd channel while for $\alpha > \alpha_c$ the odd channel becomes strongly coupled at low temperatures [$\delta_o \rightarrow \pi/2$ and $\delta_e \rightarrow 0$ in Fig. 4(d)]. In both cases, one expects a peak in the conductance.²⁷

At the critical point $\alpha = \alpha_c$ (NFL regime), the NRG spectra is identical for both even and odd parities, as predicted by conformal field theory.²⁸ In this case, our prescription for obtaining the phase shifts gives $\delta_e = \delta_o$ at low temperatures, as depicted in Fig. 4(b), causing a destructive interference and suppressing the transmission. We thus expect this result to hold, even though the individual values of $\delta_{e(o)}$ obtained from the NRG spectra in the NFL regime are only approximate.

These signatures in the low-temperature conductance versus gate voltage curves can, in principle, identify the 2chK regime in molecular junctions. Experimentally, a fine tuning of the microscopic parameter α to the critical value is nonetheless a challenging task. In general, the value of α will be determined by specific details of the junction, such as the ratio of amplitude of the center-of-mass vibration and the length of the molecule-lead overlap. It is interesting, however, to notice that, at higher temperatures, one might obtain $\delta_e \approx \delta_o$ over a wider range of α and ε_d near the critical values, as illustrated in Figs. 2(c) and 2(d). In this case, one expects a finite-temperature signature of the $T=0$ non-Fermi-liquid point.

V. SUMMARY

In summary, we have studied center-of-mass vibrational effects and phonon-assisted processes in the transport properties of a molecular junction in the Kondo regime. The interplay between electron-electron and electron-phonon interactions in this system can be described by an effective two-channel Anderson model with phonon-assisted tunnel couplings. Our numerical renormalization-group calculations for the thermodynamic properties of the effective model show non-Fermi-liquid effects below a characteristic crossover temperature over critical lines in parameter space.

We find that the crossover temperature is at a maximum at the particle-hole symmetric point and rapidly approaches zero as the system enters the mixed-valence regime. Furthermore, we find distinct signatures of the non-Fermi-liquid phase in the linear conductance.

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