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Vibrational Sidebands and the Kondo Effect in Molecular Transistors

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Electron transport through molecular quantum dots coupled to a single vibrational mode is studied in the Kondo regime. We apply a generalized Schrieffer-Wolff transformation to determine the effective low-energy spin-spin-vibron interaction. From this model we calculate the nonlinear conductance and find Kondo sidebands located at bias voltages equal to multiples of the vibron frequency. Because of selection rules, the side peaks are found to have strong gate-voltage dependences, which can be tested experimentally. In the limit of weak electron-vibron coupling, we employ a perturbative renormalization group scheme to calculate analytically the nonlinear conductance.

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In recent years, the study of transport in mesoscopic systems has branched into investigations of single-electron devices based on single-molecule transistors [1–5]. Of particular interest is the possibility of combining electronics with mechanics, such that the vibrational or configurational modes of the molecule are coupled to its charge state. A number of interesting issues have already been addressed in this new field of nanoelectromechanics. First, it was shown by Park *et al.* [2] that quantum mechanical behavior of the center of mass oscillation of a C₆₀ can be excited by the tunneling electrons and a series of assisted steps were observed in the current. Similar structures have later been observed in a number of other experiments.

It is well established [6,7] that *single particle* resonance tunneling is not destroyed by the electron-vibron coupling, but instead the resonance breaks up into a number of vibron sidebands. The question remains, though, whether more intricate *many-body* effects, such as the Kondo resonance, also cooperate with the electron-vibron coupling to form “Kondo sidebands.” The usual Kondo resonance has been observed in several molecular devices [3,4] at unusually high temperatures, and recent experiments [5] on C₆₀, and Co based transistors have revealed marked sideband resonances, which were suggested to arise from the interplay of a Kondo resonance with a vibrational mode.

In this Letter we demonstrate that, in contrast to sequential tunneling, which is suppressed by the Franck-Condon overlap factors, the Kondo resonance remains intact well inside the Coulomb-blockade valley. In fact, the electron-vibron coupling is predicted to enhance the exchange coupling and thereby the Kondo temperature. Maintaining the quantum coherence of vibrons, we show that the Kondo resonance breaks up into a series of vibron sidebands. Moreover, we demonstrate that parity selection rules prohibit all sidebands at odd multiples of the oscillator frequency when tuning the gate voltage to the particle-hole symmetric point.

Assuming the energy-level spacing on the molecule to be much larger than the charging energy, the system may

be described by the Anderson-Holstein Hamiltonian

$$H = \sum_{\alpha, \mathbf{k}, \sigma} \xi_{\alpha \mathbf{k}} c_{\alpha \mathbf{k} \sigma}^\dagger c_{\alpha \mathbf{k} \sigma} + \varepsilon_d n_d + \omega_0 b^\dagger b + U n_{d\uparrow} n_{d\downarrow} + \sum_{\alpha, \mathbf{k}, \sigma} (t_{\alpha \mathbf{k}} d_{\alpha}^\dagger c_{\alpha \mathbf{k} \sigma} + \text{H.c.}) + \lambda \omega_0 (b + b^\dagger) n_d, \quad (1)$$

where $c_{\alpha \mathbf{k} \sigma}^\dagger$ and d_{α}^\dagger are creation operators for electrons in the left and right conduction bands ($\alpha = L, R$) and on the molecular quantum dot, respectively, $n_{d\sigma} = d_{\sigma}^\dagger d_{\sigma}$, $n_d = n_{d\uparrow} + n_{d\downarrow}$, and $\xi_{\alpha \mathbf{k}} = \varepsilon_{\mathbf{k}} - \mu_{\alpha}$. The vibrational mode of the molecule is created by the vibron operator b^\dagger and λ denotes the dimensionless coupling strength. Describing the molecule as a quantum dot, we have $\varepsilon_d = (1 - 2\mathcal{N})E_C$ and $U = 2E_C$, in terms of the charging energy E_C and the mean occupation number $\mathcal{N} = C_g V_g / e$, determined by the gate voltage, V_g , and the capacitance to the gate, C_g . Unless explicitly stated otherwise, we shall henceforth work in units where $e = \hbar = k_B = 1$.

Following Lang and Firsov [8], the electron-vibron coupling in the Hamiltonian (1) is eliminated by the unitary transformation $H' = e^{iS_p} H e^{-iS_p}$, with $S_p = i\lambda(b - b^\dagger)n_d$:

$$H' = \sum_{\alpha, \mathbf{k}, \sigma} \xi_{\alpha \mathbf{k}} c_{\alpha \mathbf{k} \sigma}^\dagger c_{\alpha \mathbf{k} \sigma} + \varepsilon'_d n_d + \omega_0 b^\dagger b + U' n_{d\uparrow} n_{d\downarrow} + \sum_{\alpha, \mathbf{k}, \sigma} (t_{\alpha \mathbf{k}} e^{\lambda(b^\dagger - b)} d_{\alpha}^\dagger c_{\alpha \mathbf{k} \sigma} + \text{H.c.}), \quad (2)$$

where $\varepsilon'_d = \varepsilon_d - \lambda^2 \omega_0$ and $U' = U - 2\lambda^2 \omega_0$. We now consider the weak-tunneling limit, $\Gamma_{\alpha \mathbf{k}} = 2\pi N(0) |t_{\alpha \mathbf{k}}|^2 \ll \min(-\varepsilon'_d, \varepsilon'_d + U')$, where $N(0)$ denotes the conduction electron (*ce*) density of states. In this limit, a generalized Schrieffer-Wolff transformation, devised by Schüttler and Fedro [9], may be used to eliminate all first order terms in t_{α} . To this end, we introduce the generator $S_v = i \sum_{\alpha, \mathbf{k}, \sigma, \eta} (t_{\alpha \mathbf{k}} \zeta_{\alpha \mathbf{k} \sigma \eta} n_{d\bar{\sigma}}^\eta d_{\sigma}^\dagger c_{\alpha \mathbf{k} \sigma} - \text{H.c.})$, where $n_{d\bar{\sigma}}^\eta = (1 - \eta)/2 + \eta n_{d\bar{\sigma}}$, with $\eta = \pm 1$, $\bar{\sigma} = -\sigma$, and $\zeta_{\alpha \mathbf{k} \sigma \eta} = i \int_0^\infty dt e^{-i(E_{\alpha \mathbf{k} \eta} - i0_+)t} e^{-A(t)}$, with $E_{\alpha \mathbf{k} \eta} = \xi_{\alpha \mathbf{k}} - \varepsilon'_d - (1 + \eta)U'/2$ and $A(t) = \lambda(e^{-i\omega_0 t} b - e^{i\omega_0 t} b^\dagger)$. Applying the transformation $H'' = e^{iS_v} H' e^{-iS_v}$

and expanding to second order in t_α , one finds that $H'' = H''_0 + H_{\text{spin}} + H_{\text{dir}} + H_{\text{pair}}$ [9]. We neglect the renormalization of the kinetic energy term in H''_0 and, restricting to the regime of single occupancy, i.e., $\mathcal{N} \approx 1$ and $\lambda^2 \omega_0 \ll E_C (U' > 0)$, H_{pair} vanishes. The potential scattering term, H_{dir} , is omitted since it leads to no logarithmic singularities, and altogether we obtain the effective Hamiltonian

$$H'' = \sum_{\alpha, \mathbf{k}, \sigma} \xi_{\alpha \mathbf{k}} c_{\alpha \mathbf{k} \sigma}^\dagger c_{\alpha \mathbf{k} \sigma} + \omega_0 b^\dagger b + \sum_{\alpha, \mathbf{k}, \sigma; \alpha', \mathbf{k}', \sigma'} \mathbb{J}_{\alpha, \mathbf{k}; \alpha', \mathbf{k}'} \mathbf{S} \cdot c_{\alpha' \mathbf{k}' \sigma'}^\dagger \frac{\tau_{\sigma' \sigma}}{2} c_{\alpha \mathbf{k} \sigma}, \quad (3)$$

where $\mathbf{S} = \frac{1}{2} d_{\sigma'}^\dagger \tau_{\sigma' \sigma} d_\sigma$ denotes the local spin-1/2, and $\mathbb{J}_{\alpha, \mathbf{k}; \alpha', \mathbf{k}'} = t_{\alpha' \mathbf{k}'}^* t_{\alpha \mathbf{k}} [(X_{\alpha \mathbf{k}}^- - X_{\alpha \mathbf{k}}^+) + (X_{\alpha' \mathbf{k}'}^- - X_{\alpha' \mathbf{k}'}^+)^\dagger]$ with $X_{\alpha \mathbf{k}}^\eta = i \int_0^\infty dt e^{-i(E_{\alpha \mathbf{k}} \eta - i0_+)t + i\eta \lambda^2 \sin(\omega_0 t)} e^{A(0) - A(t)}$.

In this effective Kondo model, the exchange-coupling \mathbb{J} incorporates the dynamics of the vibron through the displacement operator e^A . In the vibron number-state basis it is therefore convenient to introduce Franck-Condon factors $f_{n'n} = \langle n' | e^{A(0)} | n \rangle$ [10], which allows us to write the matrix elements of \mathbb{J} in the more transparent form:

$$J_{\alpha', \mathbf{k}'; \alpha, \mathbf{k}}^{n'n} \equiv \langle n' | \mathbb{J}_{\alpha', \mathbf{k}'; \alpha, \mathbf{k}} | n \rangle = t_{\alpha' \mathbf{k}'}^* t_{\alpha \mathbf{k}} \sum_{m=0}^{\infty} \left\{ f_{mn'} f_{mn} \left[\frac{1}{\xi_{\alpha \mathbf{k}} - \varepsilon_- + (m - n') \omega_0} + \frac{1}{\xi_{\alpha' \mathbf{k}'} - \varepsilon_- + (m - n) \omega_0} \right] - f_{n'm} f_{nm} \left[\frac{1}{\xi_{\alpha \mathbf{k}} - \varepsilon_+ - (m - n) \omega_0} + \frac{1}{\xi_{\alpha' \mathbf{k}'} - \varepsilon_+ - (m - n') \omega_0} \right] \right\}, \quad (4)$$

valid for $\xi_{\alpha \mathbf{k}}, \xi_{\alpha' \mathbf{k}'}, n \omega_0, n' \omega_0 \ll \min(\varepsilon_+, -\varepsilon_-)$, where

$\varepsilon_- = \varepsilon'_d$ and $\varepsilon_+ = \varepsilon'_d + U'$ are the energies of intermediate, empty, or doubly occupied states. In this sum, the energies of intermediate vibron states $|m\rangle$ shift the energy denominators and the Franck-Condon factors determine the overlap between initial and final vibron states with intermediate states of the oscillator shifted by $\sqrt{2} \lambda \ell_0$, where ℓ_0 is the characteristic oscillator length.

Since $\sum_{m=0}^{\infty} f_{n'm} f_{nm} = \delta_{n'n}$ and $f_{n'n} \rightarrow \delta_{n'n}$ for $\lambda \rightarrow 0$, the usual exchange coupling, $J_{\alpha' \alpha} = 4 t_{\alpha'}^* t_\alpha / E_C$, is recovered in either of the limits $\omega_0 \rightarrow 0$ or $\lambda \rightarrow 0$. More generally, $J^{n'n}$ may be represented as an asymptotic power series as $E_C / \omega_0 \rightarrow \infty$, with leading terms $J_{\alpha' \alpha}^{n'n} \propto J_{\alpha' \alpha} (\lambda \omega_0 / E_C)^{|n' - n|}$. In terms of the incomplete Gamma function, $\gamma(\alpha, x)$, one has $J_{\alpha' \alpha}^{00} = J_{\alpha' \alpha} e^{-\lambda^2 (E_C / \omega_0)} \sum_{\eta=\pm} (-\lambda^2)^{\eta \varepsilon_\eta / \omega_0} \gamma(\eta \varepsilon_\eta / \omega_0, -\lambda^2)$, or simply $J^{00} \approx J_{\alpha' \alpha} [1 + (\lambda \omega_0 / E_C)^2]$ for $\lambda \omega_0 \ll E_C$ and $\mathcal{N} = 1$, as found earlier in Ref. [9]. Staying well inside the Kondo regime, any finite λ thus leads to a slight *enhancement* of J^{00} , and thereby of the associated Kondo temperature, $T_K \sim D e^{-1/N(0)J^{00}}$ ($2D$ being the *ce* bandwidth). In contrast to the resonant (sequential) tunneling amplitude involving *real* excitations of the oscillator [2,7], the (cotunneling) amplitude J^{00} is not suppressed by a Franck-Condon overlap since it involves only *virtual* shifts of the oscillator.

We now consider the case of strongly asymmetric and momentum independent tunneling amplitudes, $\Gamma_L \gg \Gamma_R$. The current traversing the molecule from left to right is then given simply as [11] $I = -\frac{2e}{h} \Gamma_R \sum_\sigma \int d\varepsilon [f_L(\varepsilon) - f_R(\varepsilon)] \text{Im} \mathcal{G}_{\sigma\sigma}^R(\varepsilon)$. From the equations of motion for the Hamiltonian (1), the local density of states is found to be related to the *ce* T-matrix as $\text{Im} [\mathcal{G}_\sigma^{d,R}(\omega)] = |t_\alpha|^{-2} \text{Im} [T_{\alpha\alpha}^{\sigma\sigma}(\omega)]$, and the latter can now be obtained using the effective Hamiltonian (3). To third order in $J_{\alpha, \alpha'}$, we find that

$$\text{Im} [T_{\alpha' \alpha}^{\sigma' \sigma}(\Omega)] = -\delta_{\sigma' \sigma} \frac{3\pi}{16} N(0) (1 - e^{-\omega_0/T}) (1 + e^{-\Omega/T}) \sum_{lmn\alpha_1\alpha_2} J_{\alpha' \alpha_1}^{nm} J_{\alpha_2 \alpha}^{ln} e^{-n\omega_0/T} [1 - f(\Omega + (n-l)\omega_0)] \times \theta(D - |\Omega + (n-l)\omega_0|) \left\{ \delta_{ml} \delta_{\alpha_1\alpha_2} + N(0) J_{\alpha_1\alpha_2}^{ml} \left[\ln \left| \frac{D}{\Omega + (n-m)\omega_0} \right| + \ln \left| \frac{D}{\Omega + (m-l)\omega_0} \right| \right] \right\}, \quad (5)$$

with the shorthand notation $\ln|D/x| = \ln[D/\sqrt{x^2 + T^2}]$. In the asymmetric limit considered here, we take $\mu_L = 0$ and bias the right lead to $\mu_R = -V$, leaving the position of the molecular energy levels unaffected, and for $T \ll \omega_0 \ll D$ the differential conductance is then obtained from the current as $G(V) = -\frac{2e^2}{h} (\Gamma_R / \Gamma_L) N(0) \sum_\sigma \text{Im} [T_{LL}^{\sigma\sigma}(eV)]$. From Eq. (5), the differential conductance appears to diverge as $\ln(D/T)$ at voltages corresponding to multiples of the oscillator frequency, reflecting the onset of a Kondo effect assisted by coherent vibron exchange. In Fig. 1, the upper panel shows a gray-scale plot of $\partial^2 I / \partial V^2$ as a function of bias voltage and mean occupation number (gate voltage). The lower panel shows three cuts revealing

the sideband resonances on the flanks of the central zero-bias resonance.

By tuning the gate voltage to $\mathcal{N} = 1 - \lambda^2 \omega_0 / E_C$, one reaches the particle-hole (PH) symmetric point where $\varepsilon_+ = -\varepsilon_-$, and using the general symmetry $f_{n'n} = (-1)^{|n' - n|} f_{n'n'}$ one finds from Eq. (4) that $J^{n'n} \propto [1 + (-1)^{|n' - n|}]$, implying that all spin-exchange processes involving the emission or absorption of an *odd* number of vibrons are prohibited at this particular gate voltage. This parity selection rule, reflecting the inversion symmetry of the Kondo Hamiltonian (3) at low energies, has important experimental bearings, since it predicts that all Kondo

sidebands in the differential conductance located at voltages equal to *odd* multiples of ω_0 must vanish when tuning the gate voltage to the symmetric point, corresponding to the midpoint of the Coulomb-blockade valley. This is apparent in Fig. 1, where the conductance peak at $V = \omega_0$ disappears as \mathcal{N} approaches the symmetric point (cf. curve *a*). Note, however, that any appreciable vibron modulation of the tunneling amplitudes will break the inversion symmetry [12] and thereby destroy this selection rule.

The logarithmic divergences appearing in third order perturbation theory call for a resummation of leading logarithmic contributions to all orders. This is done using the perturbative renormalization group (RG) method for frequency dependent couplings developed in Refs. [13,14]. Parametrizing the dimensionless couplings, $g_{n'n} = N(0)J_{LL}^{n'n}$, by the total energy of the ingoing conduction electron and vibron state, we arrive at the infinite hierarchy of coupled (1-loop) RG equations:

$$\frac{\partial g_{n'n}(\omega)}{\partial \ln D} = -\frac{1}{2} \sum_{m=0}^{\infty} \{g_{n'm}(0)g_{mn}[(m-n)\omega_0]\Theta_{\omega+(n-m)\omega_0} + g_{n'm}[(n'-m)\omega_0]g_{mn}(0)\Theta_{\omega+(m-n')\omega_0}\}, \quad (6)$$

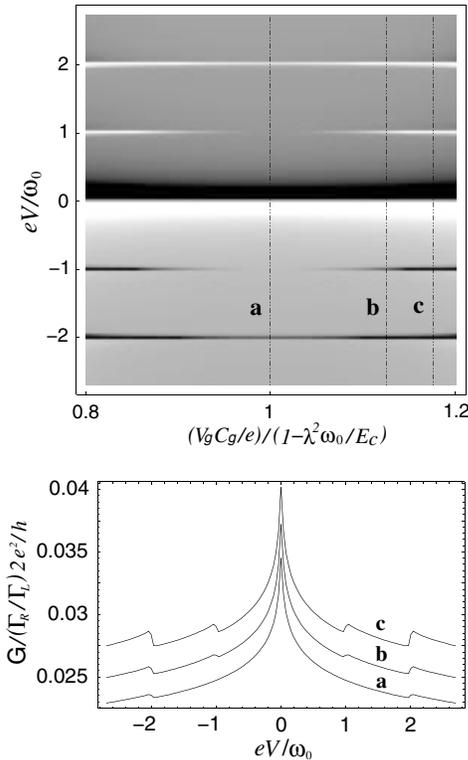


FIG. 1. Upper panel: $\partial^2 I / \partial V^2$ vs bias and gate voltage, for $\lambda^2 = 3$, $N(0)|t_L|^2 = 0.1\omega_0$, $D = E_C = 8\omega_0$, and $T = 0.01\omega_0$. Black (white) indicates large negative (positive) values. Lower panel: conductance vs bias voltage for three values of V_g corresponding to the vertical black lines (*a*, *b*, *c*) in the upper panel. The lower curve (*a*) corresponds to the PH-symmetric point $\mathcal{N} = 1 - \lambda^2\omega_0/E_C$.

with $\Theta_\omega = \Theta(D - |\omega|)$. We shall restrict our attention to the PH-symmetric point and assuming that $\lambda\omega_0 \ll E_C$, we may truncate this hierarchy and consider merely the lowest four coupled equations involving g_{00} , $g_{02} = g_{20}$, and g_{22} . The solution to this reduced set of equations is characterized by the parameters

$$\delta = \frac{g_{22} - g_{00}}{g_{00}g_{22} - g_{20}^2}, \quad \alpha = \frac{2g_{20}}{g_{00}g_{22} - g_{20}^2}, \quad (7)$$

and $T_K = De^{-2/(g_{00}+g_{22}+\sqrt{(g_{22}-g_{00})^2+4g_{20}^2})}$, where $g_{n'n} \equiv g_{n'n}(D; 2n\omega_0)$. All three parameters are invariant under the perturbative RG flow from the initial cutoff D_0 down to $D = 2\omega_0$. At scale D_0 , we have $\alpha, \delta \sim (\lambda\omega_0/E_C)^2/g_{00}$, and therefore our truncation of Eq. (6) remains valid throughout the RG flow roughly when $\max(\alpha, \delta)/\ln(T/T^*) \ll 1$ (see below). Staying within the perturbative regime, we assume that $\omega_0 \gg T \gg T_K$.

We first solve the RG equations for the constant coefficients $g_{n'n}$ and the frequency dependent renormalized couplings are then obtained simply by integrating Eqs. (6):

$$g_{00}(\omega) = \frac{1}{\ln(|\omega|/T^*)} + \frac{\alpha^2}{8\ln^2(2\omega_0/T^*)} \sum_{\nu=0,1} \Theta_\nu(\omega) \times \left(\frac{1}{\ln(|\omega - 2\nu\omega_0|/T^*)} - \frac{1}{\ln(2\omega_0/T^*)} \right) \quad (8)$$

$$g_{20}(\omega) = \frac{\alpha}{4} \sum_{\nu=0,1} \left[\frac{1}{\ln^2(\max(2\omega_0, |\omega - 2\nu\omega_0|)/T^*)} + \frac{2\Theta_\nu(\omega)}{\ln(2\omega_0/T^*)} \left(\frac{1}{\ln(|\omega - 2\nu\omega_0|/T^*)} - \frac{1}{\ln(2\omega_0/T^*)} \right) \right], \quad (9)$$

with $T^* = T_K(T_K/\omega_0)^{(\sqrt{\alpha^2+\delta^2}+\delta)/(2\ln(2\omega_0/T_K)+\sqrt{\alpha^2+\delta^2}-\delta)}$ and $\Theta_\nu(\omega) = \Theta(2\omega_0 - |\omega - 2\nu\omega_0|)$. Note that in Eqs. (8) and (9) we retain only terms which contribute to order $\max(\alpha, \delta)^2/\ln^2(T/T^*)$ in the conductance.

While the logarithmic singularities at $\omega = 0$ are cut off by temperature, those at $\omega = 2\omega_0$ will instead be contained roughly by $\sqrt{T^2 + \gamma^2}$, with γ given by the transition rate from vibron state $|2\rangle$ to $|0\rangle$. Using the golden rule with the renormalized coupling $g_{20}(\omega)$, we find $\gamma \approx \pi\omega_0\alpha^2/[4\ln^2(2\omega_0/T_K)]$. Similarly, the broadening of the vibron states induces a broadening of the step functions in Eqs. (8) and (9) by T or $\sqrt{T^2 + \gamma^2}$ for steps near $\omega = 0$ or $\omega = 2\omega_0$, respectively. (cf. also Ref. [14]).

The renormalized conductance is now obtained by inserting in the formula $G(V) = (2e^2/h)(\Gamma_R/\Gamma_L)(3\pi^2/4) \times [g_{00}(eV)^2 + \sum_{\nu=\pm} \Theta(\nu eV - 2\omega_0)g_{20}(\nu eV)^2]$, and indeed when expanding this result in bare couplings, we recover the result obtained by expanding Eq. (5) to order $(\lambda\omega_0/E_C)^4$. Including the broadening in both couplings and Θ , the result is plotted in Fig. 2. Notice the slight resemblance to Fig. 3 of Ref. [15], obtained using a dia-

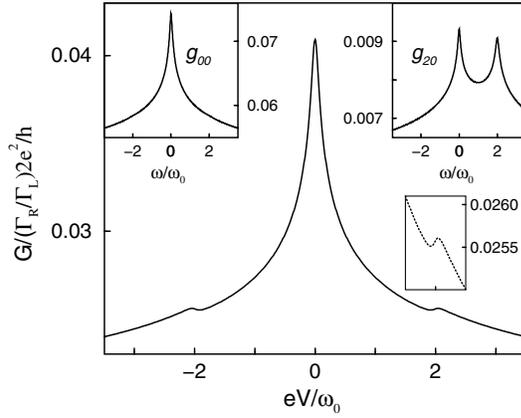


FIG. 2. Conductance vs bias voltage using *perturbatively* renormalized couplings. Gate voltage is tuned to the PH-symmetric point and $\alpha = 4.30$, $\delta = 7.54$, $T_K = 1.51 \times 10^{-4} \omega_0$, $T^* = 7.18 \times 10^{-8} \omega_0$, and $T = 0.05 \omega_0$, i.e., $\alpha / \ln(T/T^*) = 0.32$ and $\delta / \ln(T/T^*) = 0.56$, corresponding to bare parameters: $\lambda^2 = 4.5$, $N(0)|t_L|^2 = 0.1 \omega_0$, and $D = E_C = 8 \omega_0$. Insets show the renormalized couplings $g_{00}(\omega)$ and $g_{20}(\omega)$, as well as a zoom in the conductance curve showing the satellite peak on a separate conductance scale but on the same voltage scale.

grammatic real-time technique in the case where $\Gamma_L = \Gamma_R$ and $\omega_0 \ll T_K$. Since the nonperturbative approach used in Ref. [15] omits certain classes of diagrams, which are included to each order in our systematic perturbative approach, a detailed correspondence is not necessarily to be expected even for $\Gamma_L \gg \Gamma_R$ and $T_K \ll \omega_0$.

The possible decoherence effects arising when coupling the oscillator to phonons within the leads remain an open question. It is straightforward to generalize the Schrieffer-Wolff transformation applied here to a system where the molecule-oscillator is coupled to a separate bath of oscillators. However, even determining the effects on the leading logarithms for a given Q factor involves a rather involved cumulant expansion. We expect the Kondo effect to be more pronounced when dealing solely with *intra*-molecular vibrations, since these have been demonstrated to have particularly large Q factors [16].

In the case of nearly symmetric couplings, we can no longer assume the oscillator to be in equilibrium with the conduction electrons of one specific side of the junction [17]. In line with the findings of Refs. [15,18], we expect that nonequilibrium effects may in fact serve to enhance the Kondo side peaks.

In conclusion, we have demonstrated the viability of an inelastic Kondo effect carried by coherent vibron-assisted exchange tunneling, which can be observed as Kondo sidebands in the nonlinear conductance. In contrast to the case of an applied microwave field [19], the zero-bias resonance is *not* suppressed by the vibronic coupling, and it may therefore be difficult to discern the satellites from the background conductance. Nevertheless, even with very

weak satellites, it should be possible to track their dependence on V_g (possibly in a plot of $\partial^2 I / \partial V^2$) and thereby test our prediction that satellites at odd multiples of ω_0 are strongly reduced at the PH-symmetric point. Faint sidebands to a zero-bias Kondo peak have indeed been observed in the recent experiment by Yu *et al.* [5]. In contrast to the findings reported here, however, both satellite *peaks* and *dips* were observed in the nonlinear conductance. This may hint at interference effects between Kondo sidebands and other tunneling channels not included in the present model.

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