Vibrational coherence in electron spin resonance in nanoscale oscillators

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We study a scheme for electrical detection, using electron spin resonance, of coherent vibrations in a molecular single electron level trapped near a conduction channel. Both equilibrium spin currents and nonequilibrium spin and charge currents are investigated. Inelastic side-band antiresonances corresponding to the vibrational modes appear in the electron spin resonance spectrum.

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Inelastic effects arising due to coupling between charge carriers and local vibrational modes (vibrons) in nanoscale electronics devices have gained enormous interest recently. Peaks and dips often observed in the differential conductance of molecular electronics devices¹ may indicate strong effects from electron-vibron coupling. Effects from vibrons have been investigated^{2[,3](#page-3-4)} in molecular quantum dots and single electron transistor, in Josephson junctions, 4 and in surfaces using scanning tunneling microscopy.⁵

The interplay between vibrons and charge carrier is expected to generate dynamical signatures also in the spincurrent or spin-dependent transports. Such dynamics should consequently be observable in electron spin resonance (ESR), which will thus allow for electrical detection of both spin and vibron modes. In this Brief Report, we apply the ESR setup 6.7 6.7 to a molecular quantum dot with electron levels coupled to vibrons and we show the emergence of antiresonances in the spin current at frequencies equal to integral numbers of the vibrational mode. The antiresonances can be explained as interference between opposite spin tunneling electron wave functions traversing different molecular excitations. Such information would be useful not only to the conventional semiconductor industry, but also to different research directions such as spintronics and molecular electronics. Distinct from earlier work on ESR setup, $\frac{7}{1}$ we are addressing the different signatures arising from the electronvibron coupling, which will manifest in both equilibrium spin-current and nonequilibrium spin-dependent transports.

We model the resonator to be oscillating with frequency ω_0 , where the vibrational motion is weakly coupled to the electrons with strength λ . We consider the dynamics of a single molecular level ε_0 coupled to external thermal baths. The level is spin split by the external magnetic field B_0 , ε_1 $-e_1 = \omega_r = g \mu_B B_0$, where *g* and μ_B are the gyromagnetic ratio and Bohr magneton, respectively. The spins are coupled by a rotating magnetic field $B_1(\cos \omega_1 t, \sin \omega_1 t)$ applied perpendicular to B_0 and we assume $2g\mu_B B_1 \le \omega_0, \omega_r$. We employ the model $\mathcal{H} = \mathcal{H}_c + \mathcal{H}_d + \mathcal{H}_T$, where

$$
\mathcal{H}_d = \sum_{\sigma} \left[\varepsilon_{\sigma} + \lambda (a^{\dagger} + a) + Un_{\sigma}/2 \right] n_{\sigma}
$$

$$
- g \mu_B B_1 (d_{\uparrow}^{\dagger} d_{\downarrow} e^{i\omega_1 t} + d_{\downarrow}^{\dagger} d_{\uparrow} e^{-i\omega_1 t}) + \omega_0 a^{\dagger} a, \qquad (1)
$$

describes the molecular states, while $\mathcal{H}_c = \sum_{k\sigma} \varepsilon_k n_{k\sigma}$ and \mathcal{H}_T $=\sum_{k\sigma}v_kc_{k\sigma}^{\dagger}d_{\sigma}+$ H.c., are the Hamiltonians for the bath and the

tunneling, respectively. Here $c_{k\sigma}^{\dagger}$ and d_{σ}^{\dagger} create an electron with spin $\sigma = \uparrow, \downarrow$ in the bath and molecule, respectively, $(n_{k\sigma} = c_{k\sigma}^{\dagger} c_{k\sigma}$ and $n_{\sigma} = d_{\sigma}^{\dagger} d_{\sigma}$. The operators a^{\dagger} and *a* denote creation and destruction of the vibrational mode.

We transform the system into the rotating reference frame of the magnetic field through $\mathcal{H}_{\text{rf}} = e^{S_{\text{rf}}} \mathcal{H}e^{-S_{\text{rf}}} + i(\partial_t e^{S_{\text{rf}}})e^{-S_{\text{rf}}},$ with the unitary transformation $S_{\text{rf}} = -i(\omega_1 t/2)[n_1 - n_1]$ $+\sum_{k}(n_{k\downarrow}-n_{k\uparrow})$, in order to eliminate the time dependence from the Hamiltonian at the cost of introducing a shift in the electronic energies, i.e., $\varepsilon_{k\sigma}^{\text{rf}} = \varepsilon_k + \sigma \omega_1 / 2$ and $\varepsilon_{\sigma}^{\text{rf}} = \varepsilon_{\sigma} + \sigma \omega_1 / 2$, where the factor $\sigma = \pm 1$. The spin split of the conduction channel electron energies originates from the magnetic pumping field through the hybridization between the localized level and the conduction band. The pumping propagates energy from the molecule to the conduction channel and generates the spin chemical potentials $\mu_{\sigma} = -\sigma \omega_1 / 2$ (with reference to $\varepsilon_F = 0$) in the conduction channel. The frequency of the oscillating magnetic field can thus be regarded as the (spin) bias applied to the system. Despite the spin imbalance, however, the charge chemical potential is still $\mu = (\mu_{\uparrow})$ $+\mu$ ₁)/2=0.^{[8](#page-3-9)}

Although the system itself is to be considered in equilibrium, the one-photon imbalance between the spin channels generates a nonequilibrium condition for the two spin projections of the electrons. An electron in the spin-down channel can thus tunnel into the local spin-down level $\varepsilon_1^{\text{rf}}$. The rotating magnetic field flips the spin projection of the localized electron and thereby the electron can tunnel into the spin-up channel and a stationary current builds up by repeated tunneling.

The coupling between the vibrational and electronic degrees of freedom is decoupled by the canonical transformation $\widetilde{H} = e^{S_{ph}}H_{rf}e^{-S_{ph}}$ with $S_{ph} = (\lambda/\omega_0)(a^{\dagger}-a)\Sigma_{\sigma}n_{\sigma}$. Through this transformation the energy levels of the localized states are turned into $\tilde{\epsilon}_{\sigma} = \epsilon_{\sigma}^{\text{rf}} - \lambda^2/\omega_0$, while the charging energy $\tilde{U} = U - 2\lambda^2/\omega_0$ and the tunneling Hamiltonian is changed into $\Sigma_{k\sigma}(v_{k\sigma}c_{k\sigma}^{\dagger}d_{\sigma}X + \text{H.c.})$, where $X = \exp[-(\lambda/\omega_0)(a^{\dagger}-a)].$ In the present Brief Report we assume that a weak coupling between the electronic and vibrational degrees of freedom and the spin currents through the system are small. It is then justified to neglect narrowing effects on the tunneling between the conduction channel and the molecular level.⁹

In the atomic limit and $\tilde{U} = 0$, the molecule is reduced to a simple driven two-level system. It is characterized by a coherent weight transfer (Rabi oscillations) between the two

spin states, which is complete at resonant rotating frequency $\omega_1 = \omega_r$. The spin oscillation period of $T = 2\pi/\Omega$, where Ω $=\sqrt{\Delta^2+4(g\mu_B B_1)^2}$ is the Rabi frequency and $\Delta=\omega_1-\omega_r$ denotes the detuning from the resonance. We transform the molecular electron operators by

$$
\begin{pmatrix} d_{\uparrow} \\ d_{\downarrow} \end{pmatrix} = \mathbf{u} \begin{pmatrix} c_{\uparrow} \\ c_{\downarrow} \end{pmatrix}, \quad \mathbf{u} = \begin{pmatrix} \cos \phi & -\sin \phi \\ \sin \phi & \cos \phi \end{pmatrix}, \tag{2}
$$

where $\tan \phi = 2g \mu_B B_1/(\Omega - \Delta)$. The molecular electronic states are diagonal in the new representation, giving the molecular Hamiltonian $\Sigma_{\sigma} E_{\sigma} c_{\sigma}^{\dagger} c_{\sigma} + \tilde{U} c_{\uparrow}^{\dagger} c_{\uparrow} c_{\downarrow}^{\dagger} c_{\downarrow}$ with $E_{\sigma} = (\tilde{\epsilon}_{\uparrow} + \tilde{\epsilon}_{\downarrow} + \tilde{C})$ $-\sigma\Omega$)/2= $\epsilon_0 - \lambda^2/\omega_0 - \sigma\Omega/2$.

The spin- σ current I_{σ} is preferably written as

$$
I_{\sigma} = \frac{ie}{h} \text{tr} \int \Gamma_{\sigma} \{ f_{\sigma}(\omega) \mathbf{G}^{>}(\omega) + [1 - f_{\sigma}(\omega)] \mathbf{G}^{<}(\omega) \} d\omega, \tag{3}
$$

where $\Gamma_{\sigma} = \mathbf{u}_{\sigma} \Gamma$, $\Gamma = 2 \pi \Sigma_k |v_k|^2 \delta(\omega - \varepsilon_k)$, and

$$
\mathbf{u}_{\uparrow} = \tau^{\nu} \mathbf{u}_{\downarrow} \tau^{\nu}, \quad \mathbf{u}_{\downarrow} = \begin{pmatrix} \sin^2 \phi & \sin \phi \cos \phi \\ \sin \phi \cos \phi & \cos^2 \phi \end{pmatrix}, \quad (4)
$$

where τ^y is the *y* component of the Pauli matrix vector whereas $f_{\sigma}(\omega) = f(\omega - \mu_{\sigma})$ is the Fermi function for the spin σ channel.

The current contains the lesser (greater) Green's functions (GFs) $G^{<(>)} = {G^{<(>)}_{\sigma\sigma'}}_{\sigma\sigma'}$. They can be calculated using $\mathbf{G}^{<(>)} = \mathbf{G}^r \mathbf{\Sigma}^{<(>)} \mathbf{G}^a$, where, e.g., the retarded GF is defined through $G'_{\sigma\sigma'}(t) = (-i)\theta(t)\langle\{c_{\sigma}(t), c_{\sigma'}^{\dagger}(0)\}\rangle$ and similarly for the advanced one. The canonical decoupling procedure of the electron-vibron coupling casts the GF into the product of electronic and vibronic parts as

$$
G'_{\sigma\sigma'}(t) = (-i)\theta(t)\langle \{\tilde{c}_{\sigma}(t), \tilde{c}_{\sigma'}^{\dagger}(0)\}\rangle_{\text{electron}}\langle X(t)X^{\dagger}(0)\rangle_{\text{vibron}}
$$

$$
= \tilde{G}'_{\sigma\sigma'}(t)\langle X(t)X^{\dagger}(0)\rangle_{\text{vibron}},
$$
(5)

with $\tilde{c}_{\sigma}(t) = e^{i\tilde{\mathcal{H}}_{\text{electron}}t} c_{\sigma} e^{-i\tilde{\mathcal{H}}_{\text{electron}}t}$ and $X(t) = e^{i\tilde{\mathcal{H}}_{\text{vibron}}t} X e^{-i\tilde{\mathcal{H}}_{\text{vibron}}t}$. The renormalization factor caused by the electron-vibron coupling is calculated as $\langle X(t)X^{\dagger}(0)\rangle_{\text{vibron}} = e^{-\Phi(t)}$, where $\Phi(t) = (\lambda / \omega_0)^2 [n_B(1 - e^{i\omega_0 t}) + (n_B + 1)(1 - e^{-i\omega_0 t})]$ with n_B $=(e^{\beta \omega_0}-1)^{-1}$.^{[10](#page-3-11)} We then calculate the electronic GF $\tilde{G}^r_{\sigma\sigma'}$ in the mean-field approximation, in which the Kondo resonance effect is neglected. For arbitrary on-site charging energy, its Fourier transform is given by $\tilde{G}'_{\sigma\bar{\sigma}}(\omega) = 0$ and $\tilde{G}'_{\sigma\sigma} = \tilde{G}'_{\sigma}$, where

$$
\widetilde{G}_{\sigma}^{r}(\omega) = \frac{\omega - E_{\sigma} - (1 - \langle n_{\overline{\sigma}} \rangle)\widetilde{U}}{(\omega - E_{\sigma} + i\Gamma/2)(\omega - E_{\sigma} - \widetilde{U}) + i\langle n_{\overline{\sigma}} \rangle \widetilde{U}\Gamma/2}
$$
(6)

and $\langle n_{\sigma} \rangle = \text{Im} \int \tilde{G}_{\sigma}^{<}(\omega) d\omega / (2\pi)$. We then find

$$
G'_{\sigma}(\omega) = e^{-(\lambda/\omega_0)^2(2n_B+1)} \sum_n I_n(z) e^{n\beta\omega_0/2} \tilde{G}'_{\sigma}(\omega - n\omega_0), \quad (7)
$$

where $I_n(z)$ is the *n*th modified Bessel function and *z* $=2[\lambda/\omega_0]^2 \sqrt{n_B[n_B+1]}$.

In the case of weak electron-vibron coupling, the contributions to the self-energy Σ from the electron-vibron inter-

action is negligible, hence, the lesser (greater) self-energy can be approximated by

$$
\Sigma^{\leq} = i f_{\uparrow} \Gamma_{\uparrow} + i f_{\downarrow} \Gamma_{\downarrow}, \quad \Sigma^{\geq} = -i \Gamma_{\uparrow} - i \Gamma_{\downarrow} + \Sigma^{\leq}.
$$
 (8)

This leads to the spin- σ current $I_{\sigma} = e\Gamma^2 f \mathcal{I}[f_{\sigma}(\omega)]$ $-f_{\bar{\sigma}}(\omega)$] $d\omega/h$, where the transmission coefficient

$$
T = |G_{\uparrow}^{r}(\omega) - G_{\downarrow}^{r}(\omega)|^{2} \sin^{2} \phi \cos^{2} \phi.
$$
 (9)

We notice that the transmission T is equal for the two spin channels, which is expected in the stationary regime, thus giving the total spin current $I_s = \sum_{\sigma} \tau_{\sigma \sigma}^2 I_{\sigma} = 2I_{\uparrow}$. The form of the transmission coefficient suggests that the spin current can be interpreted as an interference between tunneling electron wave functions in the conduction channel, which are coupled by the molecular level.

The interference interpretation is especially appealing in the context of electron-vibron coupling. For simplicity consider the case of vanishing effective charging energy, \tilde{U} $=0(U=2\lambda^2/\omega_0)$, although the arguments remain true for arbitrary \tilde{U} . Then, the first factor in Eq. (9) (9) (9) can be written as (setting $\omega_{\sigma}^{r/a} = E_{\sigma} \mp i\Gamma/2$)

$$
|G_{\uparrow}^{r} - G_{\downarrow}^{r}|^{2} \sim \left| \sum_{n} \frac{\Omega I_{n}(z) e^{n\beta \omega_{0}/2}}{(\omega - \omega_{\uparrow}^{r} - n\omega_{0})(\omega - \omega_{\downarrow}^{r} - n\omega_{0})} \right|^{2}.
$$
\n(10)

The main ESR peak is given at $\omega_1 = \omega_r$ such that Ω $=2g\mu_B B_1$ and $E_{\sigma} = \varepsilon_0 - \lambda^2/\omega_0 - \sigma g \mu_B B_1$, corresponding to the $n=0$ term in the transmission coefficient. Because of the electron-vibron coupling, additional features in the spin current are expected to occur at frequencies $\omega_1 = \omega_r + n\omega_0$, corresponding to the vibrational side bands. Due to the destructive interference between tunneling electron waves passing through different conduction (spin) channels, these satellites to the main ESR peak appear as dips in the spin current rather than peaks.

In order to illustrate this argument, we consider the first two terms in Eq. (10) (10) (10) , which are the terms with $n=0,1$,

$$
\Omega^2[I_0^2(z)|\tilde{G}_1^r(\omega)\tilde{G}_1^r(\omega)|^2 + I_1^2(z)|\tilde{G}_1^r(\omega - \omega_0)\tilde{G}_1^r(\omega - \omega_0)|^2 e^{\beta \omega_0} + 2 \text{ Re } I_0(z)I_1(z)\tilde{G}_1^r(\omega)\tilde{G}_1^r(\omega)\tilde{G}_1^a(\omega - \omega_0)\tilde{G}_1^a(\omega - \omega_0)e^{\beta \omega_0/2}],
$$
\n(11)

where the first two terms add positively to the transmission and peak at $\omega = E_{\sigma}$ and $\omega = E_{\sigma} + \omega_0$, respectively. The last term, proportional to

$$
-Re \frac{\Omega/(\omega_0 + i\Gamma)}{(\omega - E_{\uparrow} - \omega_0 - i\Gamma/2)(\omega - E_{\downarrow} + i\Gamma/2)}
$$

+
$$
+ Re \frac{\Omega/(\omega_0 + i\Gamma)}{(\omega - E_{\downarrow} - \omega_0 - i\Gamma/2)(\omega - E_{\uparrow} + i\Gamma/2)},
$$
(12)

is negligible at $\omega_1 = \omega_r$ since then $E_\uparrow \approx E_\downarrow$, which leads to the two contributions canceling each other. As $\omega_1 \rightarrow \omega_r + \omega_0$, on the other hand, we have $E_1 + \omega_0 \approx E_1$, since $\Omega \approx \omega_0$. Therefore, the first contribution in Eq. (12) (12) (12) roughly equals

FIG. 1. (Color online) Equilibrium spin-current dependence of λ and \tilde{U} . Here, $\omega_r = 2$, $g\mu_B B_1 = 0.2315$, $\Gamma = 4\sqrt{5}/25$, and $k_B T = 10$, in units of ω_0 .

$$
-\frac{1/[1+(\Gamma/\omega_0)^2]}{(\omega-E_\downarrow)^2+(\Gamma/2)^2},\tag{13}
$$

while the second contribution is negligible. The expression in Eq. ([13](#page-2-0)) peaks around $\omega = E_{\downarrow} \approx E_{\uparrow} + \omega_0$ and contributes destructively to the total transmission coefficient in Eq. (11) (11) (11) . An estimate of the ratios between the third and first and the third and second terms in Eq. ([11](#page-1-3)) at $\omega_1 \approx \omega_r + \omega_0$, yields the lower bounds

$$
\left|\frac{I_1(z)}{2I_0(z)}\right|e^{\beta\omega_0/2}\mathcal{L}(\omega_0), \quad \left|\frac{I_0(z)}{2I_1(z)}\right|e^{-\beta\omega_0/2}\mathcal{L}(\omega_0), \quad (14)
$$

respectively, where ω_0^2 / $[1+(\omega_0/\Gamma)^2] = \Gamma^2/[1$ +(Γ/ω_0)²]. We, thus, find that the transmission is significantly reduced when the detuning Δ equals the first vibrational side band. Including the remaining contributions to the transmission, i.e., summing over all *n*, provides similar reductions in the transmission at all frequencies $\omega_1 = \omega_r + n\omega_0$.

We calculate the spin current by solving Eq. (6) (6) (6) selfconsistently. The equilibrium spin current through the molecular level is plotted in Fig. [1](#page-2-1) as a function of the rotating frequency ω_1 , illustrating the main ESR peak at $\omega_1 = \omega_r$ and the vibrational antiresonances at $\omega_1 = \omega_r + n\omega_0$, where $n \neq 0$ $(\omega_0 / \omega_r = 1/2$ in the plot). At vanishing correlation energy, the spin current decreases for increasing coupling strength λ , which is understood as an effect of the density being distributed among an increasing number of vibrational side bands for increasing electron-vibron coupling [cf. Fig. $2(a)$ $2(a)$]. Increasing spin current for increasing correlation energy can be explained by the same effect [see Fig. $2(b)$ $2(b)$].

It is easy to generalize the above theory to two leads and nonequilibrium conditions. The voltage between the leads is $eV = \mu_L - \mu_R$ and in each lead we have the spin imbalance such that $\mu_{\chi} = (\mu_{\chi\uparrow} + \mu_{\chi\downarrow})/2$, where $\chi = L, R$. The current $I_{L\sigma}$ for the spin- σ current flowing from the left lead into the molecule is written as (with obvious notation)

$$
I_{L\sigma} = \frac{e}{h} \int \Gamma^{L} \{ \Gamma^{R} \mathcal{T}_{c} [f_{L\sigma}(\omega) - f_{R\sigma}(\omega)] + \Gamma^{L} \mathcal{T}_{s} [f_{L\sigma}(\omega) - f_{L\overline{\sigma}}(\omega)]
$$

+
$$
\Gamma^{R} \mathcal{T}_{s} [f_{L\sigma}(\omega) - f_{R\overline{\sigma}}(\omega)] \} d\omega, \qquad (15)
$$

where $\mathcal{T}_c = |G^r|\cos^2 \phi + G^r|\sin^2 \phi|^2$, whereas \mathcal{T}_s is the transmis-sion coefficient given in Eq. ([9](#page-1-0)). Here, also $f_{\chi\sigma}(\omega) = f(\omega)$ $-\mu_{\chi\sigma}$). The expression for the current in Eq. ([15](#page-2-3)) is obtained by the observation that the lesser (greater) self-energy in this case is given by

$$
\Sigma^{\leq} = i \sum_{\chi\sigma} f_{\chi\sigma} \Gamma^{\chi}_{\sigma}, \quad \Sigma^{\geq} = -i \sum_{\chi\sigma} (1 - f_{\chi\sigma}) \Gamma^{\chi}_{\sigma}. \tag{16}
$$

We identify the contributions in Eq. (15) (15) (15) with the first contribution being the usual charge transport as derived by Meir and Wingreen, $\frac{11}{11}$ the second contribution is the one discussed above in Eq. (3) (3) (3) , and the third contribution accounts for the spin current between the leads.

The charge current between the leads, $I_c = \sum_{\sigma} I_{L\sigma}$, becomes

$$
I_c = \frac{e}{h} \sum_{\sigma} \int \Gamma^L \Gamma^R (T_s + T_c) [f_{L\sigma}(\omega) - f_{R\sigma}(\omega)] d\omega, \quad (17)
$$

which is just the sum of the different transmission contributions between the leads. As one would expect, I_c lacks the interference effects that occur in the spin current, which becomes clear by noticing that $T_c + T_s = |G_f|^2 \cos^2 \phi$ $+ |G_{\perp}^r|^2 \sin^2 \phi$. The spin current $I_s = \sum_{\sigma} \tau_{\sigma \sigma}^z I_{L\sigma}$ is given by

$$
I_s = \frac{e}{h} \int \Gamma^L \{ 2\Gamma^L T_s[f_{L\uparrow}(\omega) - f_{L\downarrow}(\omega)]
$$

+ $\Gamma^R T_s[f_{L\uparrow}(\omega) - f_{R\downarrow}(\omega) + f_{R\uparrow}(\omega) - f_{L\downarrow}(\omega)]$
+ $\Gamma^R T_c[f_{L\uparrow}(\omega) - f_{R\uparrow}(\omega) + f_{R\downarrow}(\omega) - f_{L\downarrow}(\omega)] \} d\omega,$ (18)

which contains three contributions. The first contribution has the same origin as discussed above in the single medium case; the second contribution accounts for the spin current between the leads; the third contribution stems from the spin imbalance in the charge current, which arises from the spinbiased leads.

The nonequilibrium spin current is plotted in Fig. [3,](#page-3-13)

FIG. 2. Local molecular DOS for λ/ω_0 =0.35 and (a) \tilde{U} =0 and (b) \tilde{U}/ω_0 =10. Other parameters are as in Fig. [1.](#page-2-1)

FIG. 3. (Color online) Bias voltage dependent spin current for λ/ω_0 =0.35 and \tilde{U} =0. Other parameters are as in Fig. [1.](#page-2-1)

showing its dependence on the bias voltage. For low rotating frequency, the spin current is dominated by transport that is assisted by the rotating magnetic field, cf. first and second terms in Eq. (18) (18) (18) , which provides the main ESR peak and vibrational antiresonances analogous to the equilibrium case. Increasing frequency ω_1 increases the potential barrier for a molecular level spin flip. Hence, ac magnetic-field-assisted transport becomes suppressed, in analogy with the equilibrium situation. The nonequilibrium conditions do, however, enhance tunneling between the leads of electrons that do not undergo spin flips when in the molecule, i.e., the contribution from the third term in Eq. (18) (18) (18) increases.

We have, for simplicity, neglected effects on the vibrational coherence from the environment, which is justified whenever the dwell time of the localized electrons $\tau_d^{-1} = \Gamma$ $\ll \omega_0$.^{[12](#page-3-14)} By studying the vibron mode lifetime τ_v to the second order in the electron-vibron coupling λ , we find τ_v^{-1} $\sim \lambda^2 \Gamma / [\pi(\omega_0^2 + \Gamma^2)]$. In the present Brief Report we thus have $\omega_0 \tau_v$ ~ 10² for typical electron-coupling strengths and cou-

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plings between the local electrons with the conduction channel, which justifies the introduced approximations. Assuming a vibron mode $\omega_0 \sim 1 \mu$ eV provides a vibron lifetime at least in the order of tens of nanosecond, which should be sufficient for measurements.

The herein reported antiresonances are expected to occur more generally within the ESR setup. In fact, we have studied the occurrence of the antiresonances in systems where the local level is coupled to a general system with two or more levels and we find that the antiresonances will occur whenever the subsystems have direct interactions with one another. The nature of the interactions may be, e.g., tunneling, Coulomb, spin-spin exchange interactions between electrons, or (as discussed in the present Brief Report) fermionboson interactions.

In summary, we have studied a scheme for electrical detection, using ESR, of vibrational coherence in molecular single electron level trapped near conduction channel. We have shown that the electron-vibron coupling generates antiresonances in the spin current at frequencies equal to integral numbers of the vibrational mode. The antiresonances can be explained as interference between opposite spin tunneling electron wave functions traversing different molecular excitations. Observations of the vibrational antiresonances do not require extremely low temperatures and, since we are using realistic parameters in the study, we believe that our findings should be within experimental reach.

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