Interaction-induced harmonic frequency mixing in quantum dots

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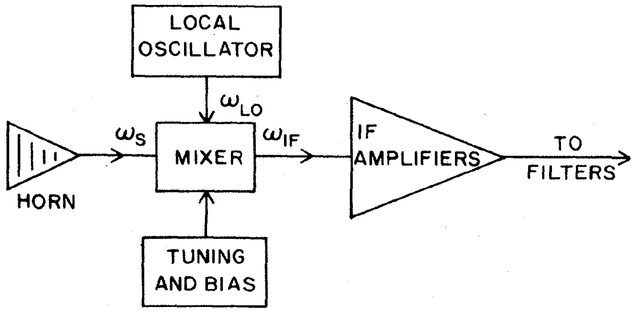
Outline

- Introduction: general remarks on mixing, definition
- No mixing in non-interacting quantum dots
- Add interaction perturbatively: mixing occurs
- Mixing in the Kondo regime:
  exact results at some Toulouse point
- Mixing in the sequential tunneling regime: master equation
- Phonon-mediated interaction ⇒ mixing

Thorwart, Egger, Gogolin, preprint

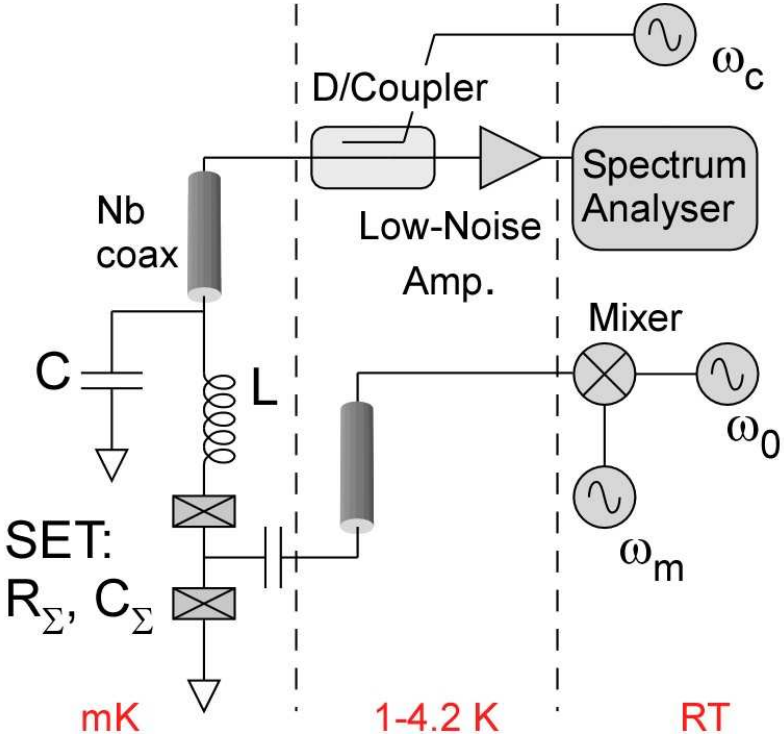
Nonlinear frequency mixing

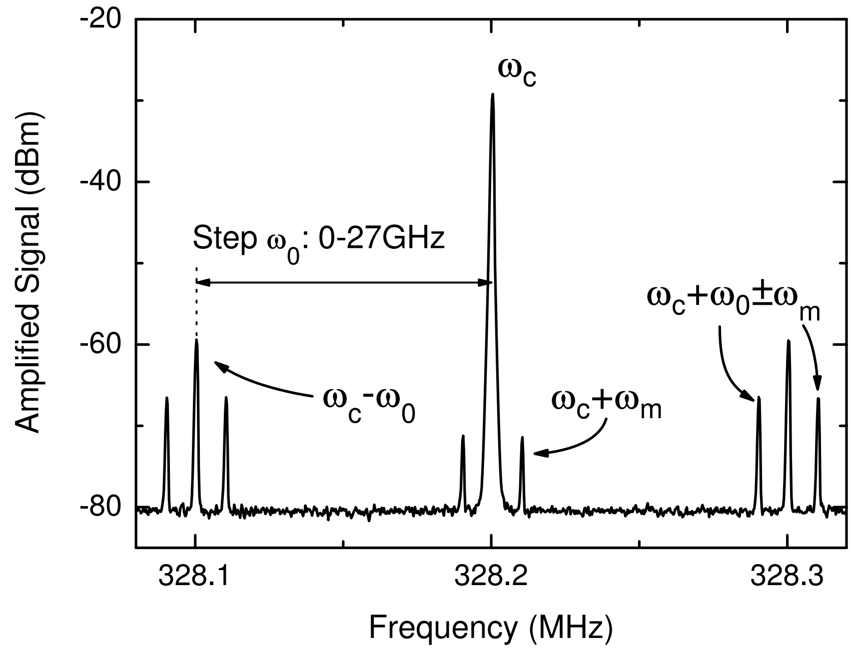
- Input: Two time-dependent signals with frequencies $\omega_{L/R}$
- often, one frequency is unknown, the other serves as reference



- Output: Nonlinear mixer generates signal with mixing frequencies $\omega_{nm} = n\omega_L + m\omega_R$ $(n, m$ integer)
- Many applications: microelectronics, nonlinear optics, telecommunications, radioastronomy: sub-mm RF signal detection using supercond. tunneling junctions Tucker and Feldman, Rev. Mod. Phys. 1985
- Mixing in quantum coherent nanoscale conductors? No theory for quantum dots exists.

Experiments: adiabatic RF-SET





low frequencies, no photon-assisted effects

two ac signals to one lead and the gate of SET, use non-linearity of $I_{SD}(V_g)$

Reilly and Buehler, APL 2005
Cleland *et al.*, APL 2005

General considerations

- Two ports: left and right leads, two frequencies ω_L and ω_R:

$$H_{leads}(t) = \sum_{k\sigma\alpha=L/R=\pm} \left[\epsilon_{k,\alpha} + \alpha eV/2 + eV_\alpha(t)\right] c^\dagger_{k\sigma\alpha} c_{k\sigma\alpha}$$

  - dc bias voltage V
  - ac driving: $V_\alpha(t) = V_\alpha^{ac} \cos \omega_\alpha t$
  - ac frequencies $1 - 1000$ GHz
- $H = H_{dot} + H_{leads}(t) + H_T$
- multilevel dot, third electrode: gate voltage $V_g(t)$
  assume energy-independent hybridization matrices $\mathbf{\Gamma}_{L/R}$
- How to quantify frequency mixing?

Mixing amplitudes

- here: fix gate voltage $V_g(t) = V_g$
- Time-dependent current through device

$$I(t) = I_{\text{tun}}(t) + I_{\text{dis}}(t) = \text{Re} \sum_{n,m=-\infty}^{\infty} e^{-i\omega_{nm}t} I_{nm},$$

with frequencies $\omega_{nm} = n\omega_L + m\omega_R$.

- Mixing amplitudes

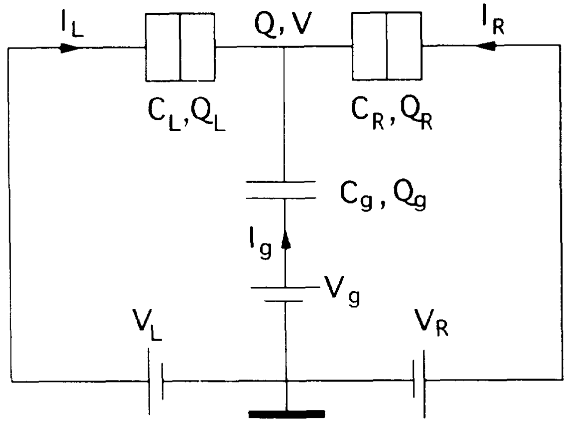
$$J_{nm} \equiv I_{nm} + I^*_{-n,-m} \stackrel{!}{\neq} 0 \qquad n > 0, m \neq 0$$

necessary condition for harmonic frequency mixing to occur: at least one mixing amplitude is finite!

Displacement currents

- important for gauge invariance & current conservation

Büttiker *et al.*, PRB 1985



- In general: $I(t) = I_L(t) - I_R(t)$

Bruder, Schoeller, PRL 1994

$$I_L(t) = \frac{C_R C_L}{C}[\dot{V}_L(t) - \dot{V}_R(t)] + \frac{C_g C_L}{C}[\dot{V}_L(t) - \dot{V}_g(t)] + \frac{C_R + C_g}{C} I_{L,\mathrm{tun}}(t) - \frac{C_L}{C} I_{R,\mathrm{tun}}(t)$$

$$C = C_L + C_R + C_g$$

- $\Rightarrow$ current conservation $I_L(t) + I_R(t) + I_g(t) = 0$
- However: $I_{\mathrm{dis}}(t)$ does not contribute to mixing J_{nm}!

General expression for tunneling current

Jauho, Wingreen & Meir, PRB 1994

$$I_\alpha(t) = -2e\mathrm{Im}\mathrm{Tr}[\mathbf{\Gamma}_\alpha \mathbf{G}^<(t,t)] - 2e\mathrm{Im}\mathrm{Tr}\int dt' \int \frac{d\omega}{2\pi} f_\alpha(\omega) e^{i\omega(t-t')} \times e^{-i[\Delta_\alpha(t)-\Delta_\alpha(t')]} \mathbf{\Gamma}_\alpha \mathbf{G}^r(t,t')$$

$\Delta_\alpha(t) = a_\alpha \sin \omega_\alpha t$ with $a_\alpha = eV_\alpha^{ac}/\omega_\alpha$

- given in terms of the interacting retarded/lesser dot Green's function, valid for $\omega_{L/R} \gg \mathrm{tr}\, \mathbf{\Gamma}_{\mathrm{L/R}}$
- apply wide band limit $\mathrm{tr}\, \mathbf{\Gamma}_{\mathrm{L/R}} \ll \mathrm{D}$:
  - energy independent tunneling amplitudes
  - neglect potential shift, i.e., real part of self-energy of the dot level
- time-dependent gate voltage $\Rightarrow \Delta_\alpha(t) \to \Delta_\alpha(t) - \Delta_g(t)$

Noninteracting dot

For arbitrary noninteracting multi-level dot with the leads in the wide band limit, there is no harmonic frequency mixing of two signals in different leads:

$$J_{n>0,m\neq 0} = 0$$

Proof:

- use Jauho-Wingreen-Meir formula
- notice that $G_0^r(t,t')$ is independent of ac drive
- notice that $G_0^<(t,t)$ depends only additively on ac drive

$\Rightarrow$ Does interaction provide lead-lead mixing?

($\Rightarrow$ non-interacting dot mixes two signals in the same lead or two signals lead-gate, energy dependence of $\Gamma_{L,R}$ also gives mixing)

Notes: Generation of higher harmonics

$I - V -$characteristics for noninteracting dot is already nonlinear
$\Rightarrow$ sufficient for mixing?

- No! This nonlinearity is irrelevant:
  - each lead is only monochromatically driven $\Rightarrow$ Generation of higher harmonics ($n\omega_\alpha$) clearly possible
  - Rectification clearly possible: $I_{0,m}, I_{n,0} \neq 0$ for $V = 0$
  - but: in non-interacting dot, the two leads do not interact $\Rightarrow$ no mixing $I_{n>0,m\neq 0} = 0$
- again: mixing $\Leftrightarrow$ 'cross-talk' between leads

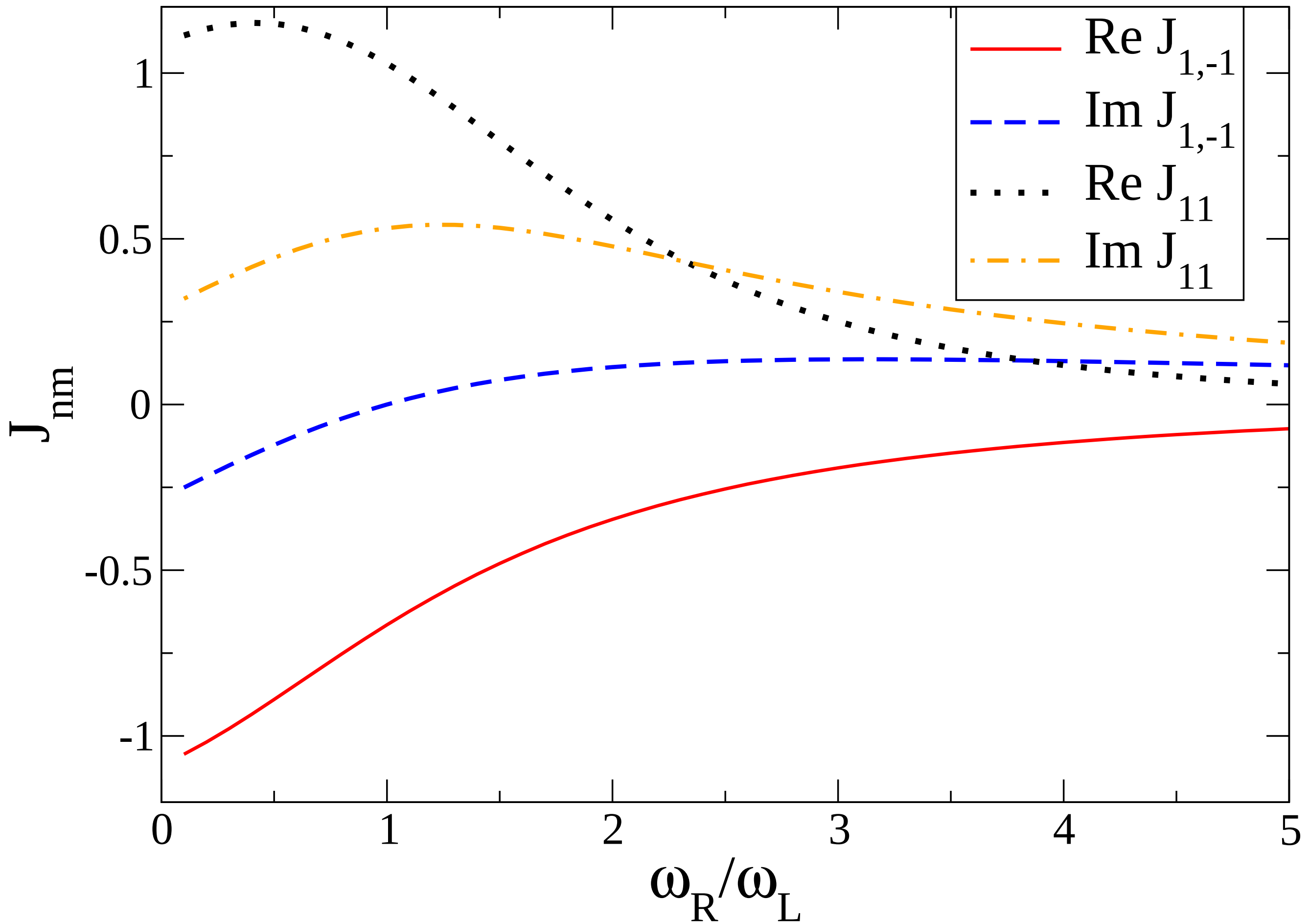
Interacting dot: Anderson model

$$H_{dot} = \epsilon_0(n_\uparrow + n_\downarrow) + U n_\uparrow n_\downarrow$$

$$n_\sigma = d_\sigma^\dagger d_\sigma$$

- perturbation theory to first order in U:
  self energy: $\Sigma^r(t,t') = U n(t)\delta(t-t')$, $n(t) = -iG_0^<(t,t)$
- generates corrections to retarded GF: $\Delta G^r = G_0^r \Sigma^r G_0^r$
- use Jauho-Wingreen-Meir formula $\Rightarrow I_{nm}$
- result has correct symmetry for $V=0$:
  $I_{nm}(\omega_L,\omega_R) = -I_{mn}(\omega_R,\omega_L)$
- $J_{nm} = 0$ for even $n+m$ at e-h symmetric point
  $\epsilon_0 = -U/2, V = 0$
- away from symmetric point: always mixing

Perturbation theory in U, small drive



$$V = 0.2\Gamma, \omega_L = \Gamma/2, T = \epsilon_0 = 0$$

given in units of $(e/8\pi^2\hbar)UV_L^{ac}V_R^{ac}/\Gamma^2$

Large U: Kondo-Toulouse point

- Different approach: Kondo regime, $\epsilon_0 \approx -U/2, \Gamma \ll U$
- Mapping to spin$-1/2$ magnetic impurity with exchange couplings J_{RR}, J_{LL}, J_{LR}
- anisotropic generalization $\Rightarrow$ exact solution at special Toulouse point
- Mapping to free fermions: $2\pi\hbar v_F = J^z_{RR} = J^z_{LL}, J^z_{LR} = 0$
- also works for time-dependent case Schiller & Hershfield, PRL 1996

$$I_{nm} = (-)^m \frac{ie\Gamma_1}{\pi\hbar} \sum_{kl} J_k(a_L) J_{k+n}(a_L) J_l(a_R) J_{l+m}(a_R)$$
$$\times \ln\left(\frac{k_B T_K + i[eV + \hbar\omega_{kl}]}{D}\right)$$

$$\Gamma_1 = \Gamma_1(J^{\perp}_{LR})$$
$$T_K = T_K(J^{\perp}_{LL,RR,LR})$$

Master equation approach: arbitrary U

- Sequential tunneling approximation $k_B T \gg \Gamma$
- monochromatic ac drive, see Bruder, Schoeller PRL 1994
- Occupation probabilities $P_s(t)$ of four possible dot states with $E_1 = 0, E_{2,3} = \epsilon_0, E_4 = 2\epsilon_0 + U$
- rate equation:

$$\dot{P}(t) = \sum_{s',\alpha=L/R} \left[K^{\alpha}_{ss'}(t) P_{s'}(t) - K^{\alpha}_{s's}(t) P_s(t)\right]$$

- tunneling current:

$$I_\alpha(t) = \sum_{s \neq s'} \mathrm{sgn}(s - s') K^{\alpha}_{ss'}(t) P_{s'}(t)$$

Master equation approach: arbitrary U

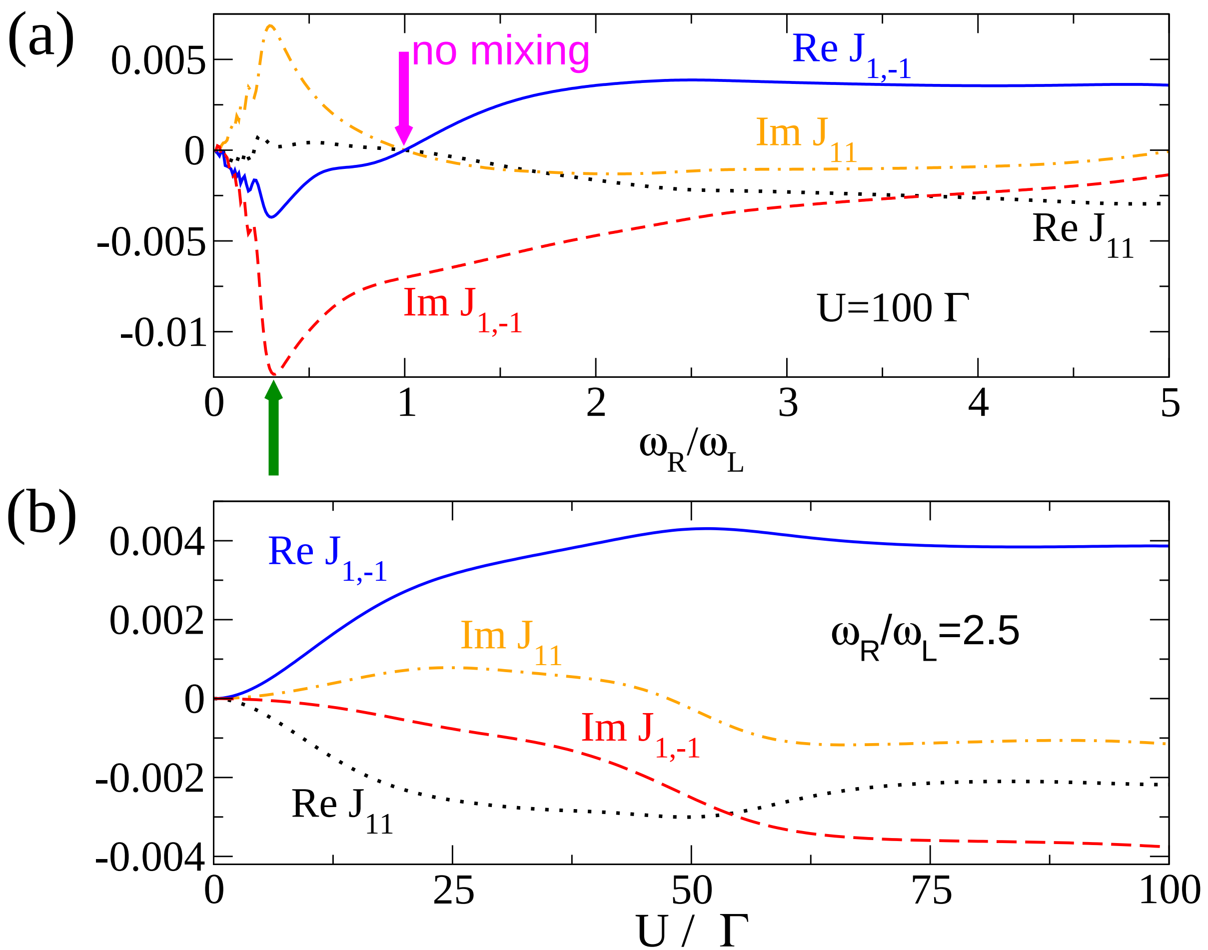
Transition rates:

$$K^{\alpha}_{ss'}(t) = \frac{\Gamma}{2}\mathrm{Re}\sum_{k,q} e^{iq\omega_\alpha t} J_k(a_\alpha) J_{k+q}(a_\alpha) \times \sum_{\sigma,\pm} |\langle s|d_\sigma^{(\dagger)}|s'\rangle|^2 g_\pm(\pm[E_s - E_{s'}] + k\omega_\alpha - \alpha eV/2)$$

with $g_\pm(\epsilon) = f(\epsilon) \pm \frac{i}{\pi}\ln\frac{D}{2\pi k_B T} \mp \frac{i}{\pi}\mathrm{Re}\,\psi\left(\frac{1}{2} + \frac{i\epsilon}{2\pi T}\right)$

- Fourier expansion $\Rightarrow$ algebraic matrix equation
- SVD $\Rightarrow$ solution $P_s(t)$
- $U = 0 \Rightarrow$ ‘no mixing’ recovered
- perturbative-in-U results at high T and small U recovered

Master equation approach: Results



$$V = \epsilon_0 = 0, \omega_L = 20\Gamma, k_B T = 5\Gamma, V_\alpha^{ac} = 50\Gamma$$

J_{nm} given in $e\Gamma/h$

Features:

- peaks
- steps

Photon-assisted tunneling picture

- ac voltages effectively induce sidepeaks in the density of states of the dot at energies $E_s + \hbar\omega_{nm}$
- mixing becomes efficient when Fermi energy matches with one of these quasi-energies
  - Example: Peak at $\omega_R/\omega_L = 1/3$ caused by the DOS sidepeaks $(n, m) = (4, 3)$
- detailed transport spectroscopy possible, depending on driving strengths and frequencies
- mixing disappears for $\omega_R/\omega_L \to 0, \infty$
- mixing disappears for $n + m$ even and e-h symmetry

Phonon-mediated interaction

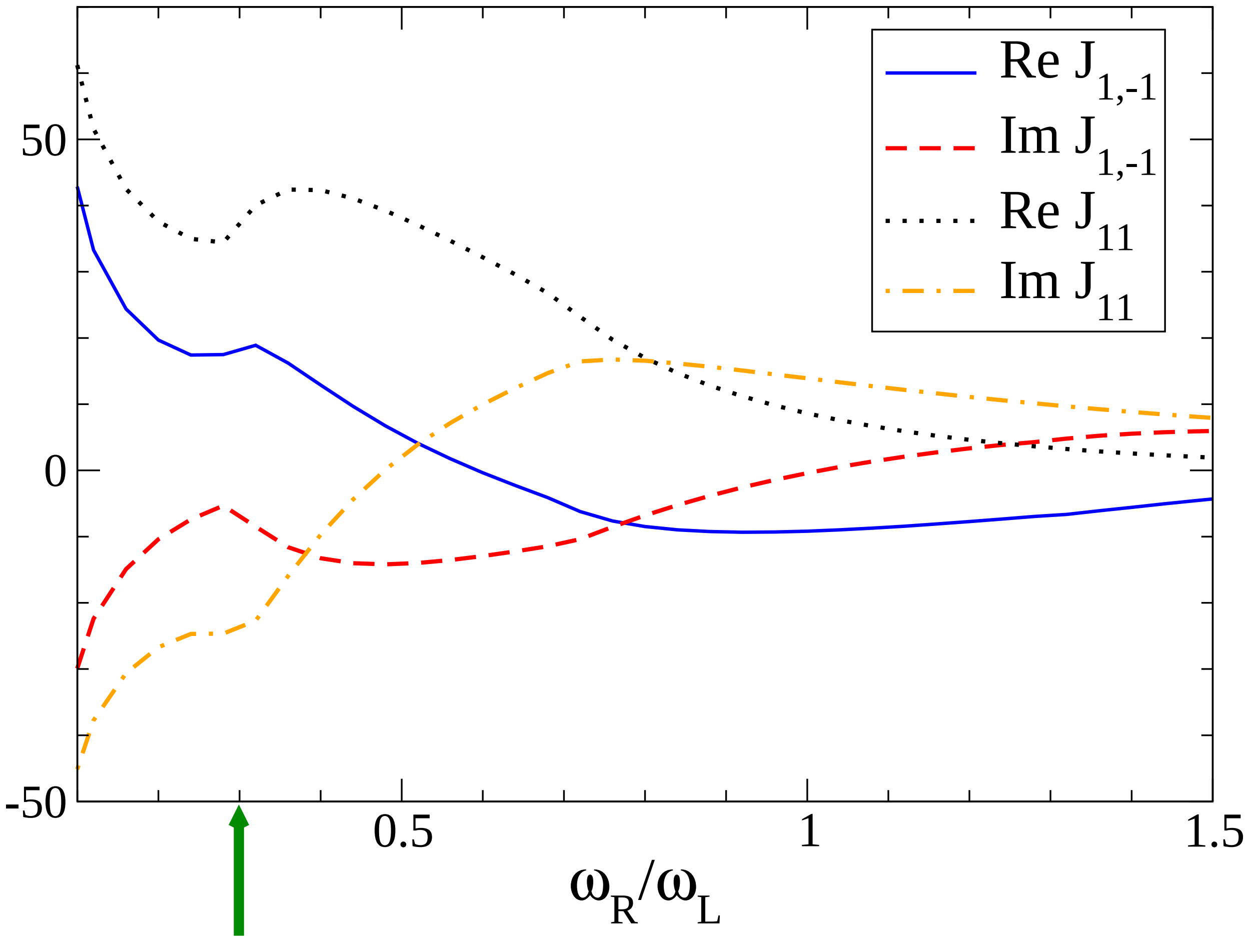
Local Holstein model (vibrating molecule, one spinless level d):

Galperin, Ratner, Nitzan, J. Phys. Cond.Matt. 2007

$$H_{\text{dot}} = [\epsilon_0 + \lambda(b + b^\dagger)]d^\dagger d + \Omega b^\dagger b + H_{leads}(t) + H_T$$

- compute mixing amplitude perturbatively up to λ^2
- closed but lengthy expressions...
- result effectively reduces to Anderson dot result for anti-adiabatic limit $\Omega \gg \Gamma$
- again, sharp features appear in frequency mixing spectroscopy

Phonon-mediated interaction



$$T = \epsilon_0 = 0, eV = 0.2\Gamma, \Omega = 0.25\Gamma,$$
$$\omega_L = 2\Omega, a_\alpha \ll 1$$

J_{nm} given in $\lambda^2 a_L a_R \omega_L \omega_R/(2\pi\Gamma)^3$

Features:

- kinks at $\Omega = \omega_{nm}$
- example: $\omega_R/\omega_L = 1/4$ $\Leftrightarrow$ $(n, m) = (1, -2)$
- measure Ω by mixing spectroscopy

Experimental viability

Estimate:

- Parameters of Cleland *et al.*, APL (2005):
  - dot capacitance $C_\Sigma = 550$ aF $\Rightarrow U = e^2/2C_\Sigma = 35$ GHz
  - resistance $R = 850$ k$\Omega \Rightarrow \Gamma = 2\pi/RC_\Sigma = 13$ GHz
  - $\Rightarrow U/\Gamma = 2.7$
- mixing achieved for $U/\Gamma = 50$, therefore
  - choose same dot capacitance $C_\Sigma = 550$ aF
  - increase resistance to $R = 20$ M$\Omega \Rightarrow \Gamma = 500$ MHz
  - $\Rightarrow \omega_{L/R} = 1$ GHz

Conclusions

- Quantum theory of harmonic frequency mixing in quantum dots
- No lead-lead mixing in non-interacting quantum dots
- Interaction also generates frequency mixing!
- Mixing provides useful tool to extract information on interaction effects
- Mixing can be used to detect frequency of local phonon

Thorwart, Egger, Gogolin, preprint

Discussion

Gauge invariance

- consider multi-terminal chaotic quantum dot with monochromatic ac voltages (freq. ω) at all terminals:

  Polianski, Samuelsson, Büttiker, PRB **72**, 161302 (R) (2005)

- Gauge invariance requires to include time-dependent internal dot potential $U(t)$
- Result: Time-averaged potential:

$$\overline{U} = V^{ac} + \frac{\text{const.}}{1 - i\omega/\Gamma}$$

- for $\omega \gg \Gamma$ second term small
- expected to be similar for bichromatic case
- first terms additive $\Rightarrow$ no influence on mixing