Cotunneling current through magnetic atoms with phonon-assisted spin-flip processes

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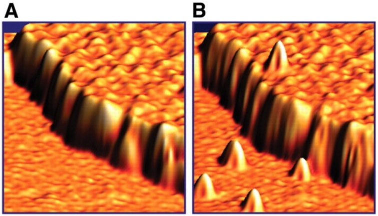
Outline

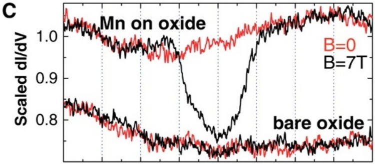
Quantum computing: DiVincenzo criteria

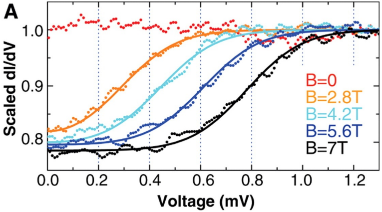
- scalable physical system with well-defined qubits
- initializable to a specific state
- universal set of quantum gates
- long decoherence times
- high quantum efficiency, qubit-specific measurements

Current through single surface-adsorbed Mn atoms

- A.J. Heinrich et al., Science **306**, 466 (2004).

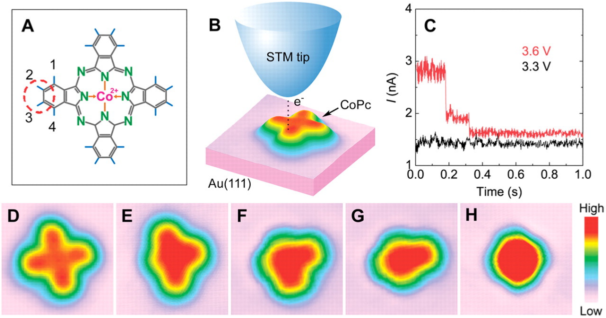




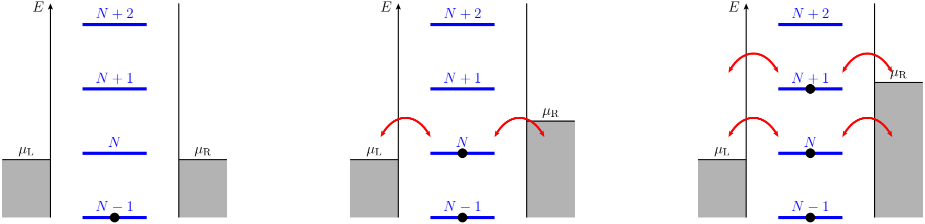


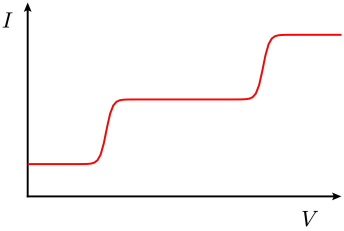
Control of coupling strength via chemical bonding

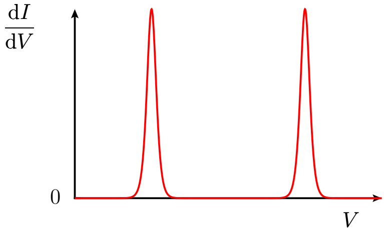
- A. Zhao et al., Science **309**, 1542 (2005).



Sequential tunneling

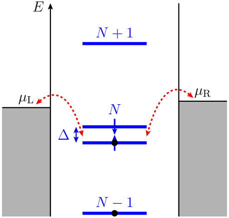




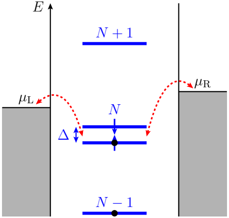


Cotunneling

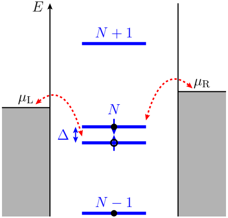
in between conductance peaks: transport via virtual changes of N



elastic cotunneling



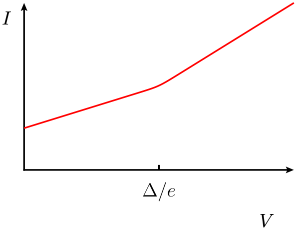
elastic cotunneling

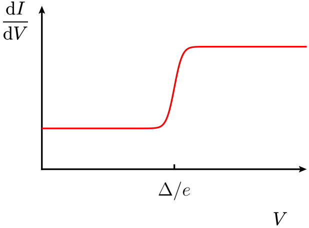


inelastic cotunneling

$eV < \Delta$

$eV > \Delta$

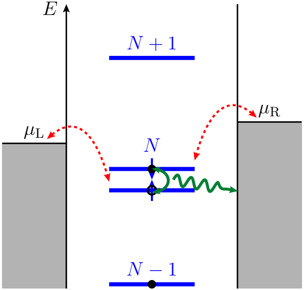




Broadening of cotunneling step

- inelastic cotunneling step broadened by temperature not by molecule-lead coupling

? role of additional inelastic processes leading to spin flips

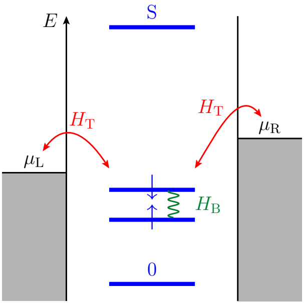


- spin-orbit induced coupling to phonons
- hyperfine coupling
- ...

- molecule as probe for the environment?

Outline

Minimal model



- molecule, metallic leads $\ell = \mathrm{L}, \mathrm{R}$ and phonons

$$H_0 = \sum_{\sigma=\uparrow,\downarrow} E_\sigma n_\sigma + U n_\uparrow n_\downarrow + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} n_{\mathbf{q}} + \sum_{\ell=\mathrm{L,R}} \sum_{\mathbf{k}\sigma} \epsilon_{\ell\mathbf{k}} n_{\ell\mathbf{k}\sigma}$$

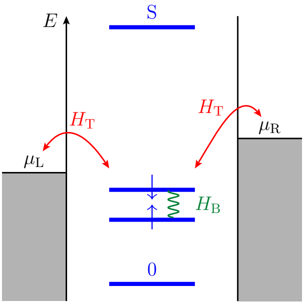
- molecule-lead coupling

$$H_\mathrm{T} = \sum_{\ell\mathbf{k}\sigma} T_{\ell\mathbf{k}}\, c^\dagger_{\ell\mathbf{k}\sigma} d_\sigma + \text{h.c.} \rightarrow \boxed{\Gamma_\ell(\epsilon)}$$

- spin-phonon coupling

$$H_\mathrm{S} = \sum_{\mathbf{q}} (M_{\mathbf{q},x}\,\sigma_x + M_{\mathbf{q},y}\,\sigma_y)(a_{\mathbf{q}} + a^\dagger_{\mathbf{q}}) \rightarrow \boxed{D(\omega)}$$

Minimal model



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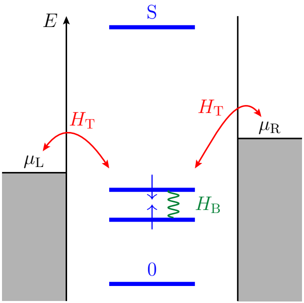

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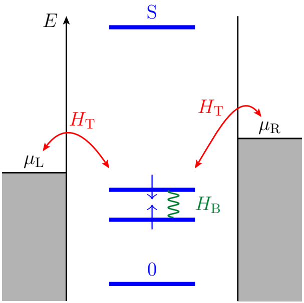
Minimal model



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Schrieffer-Wolff transformation

focus on processes which only virtually change the occupation of the molecule [Schrieffer, Wolff (1966)]

- canonical transformation generated by $S^\dagger = -S$

$$\bar{H} = \mathrm{e}^S H \mathrm{e}^{-S} = H + [S, H] + \frac{1}{2}[S, [S, H]] + \dots$$

- $H = H_0 + H_\mathrm{T}$: choose $[S, H_0] = -H_\mathrm{T}$ to eliminate H_T to first order

$$\boxed{\bar{H} = H_0 + \frac{1}{2}[S, H_\mathrm{T}] + \dots}$$

- Liouvillian of isolated molecule: $L_0 \cdot = [H_0, \cdot] \Rightarrow S = L_0^{-1} H_\mathrm{T}$

Schrieffer-Wolff transformation II

- transformed Hamiltonian

$$\bar{H} = H_0 + H_{\text{exc}} + H_{\text{renorm}} + H_{\text{2e}}$$

- exchange coupling

$$H_{\text{exc}} = -J\, \mathbf{s}_{\text{mol}} \cdot \mathbf{S}_{\text{leads}}$$

- direct (spin-independent) coupling between leads
- renormalization H_{renorm} of molecular Hamiltonian H_0
- two-electron tunneling contribution H_{2e}

Schrieffer-Wolff transformation with H_S

- $H = \underbrace{H_0 + H_\mathsf{S}}_{H_0'} + H_\mathsf{T}$: choose $[S, H_0'] = -H_\mathsf{T}$

$$\bar{H} = H_0' + \frac{1}{2}[S, H_\mathsf{T}] + \dots$$

- Liouvillian of molecule coupled to phonons for weak electron-phonon coupling:

$$L_0'^{-1} = (L_0 + L_\mathsf{S})^{-1} = L_0^{-1} - L_0^{-1} L_\mathsf{S} L_0^{-1} + \mathcal{O}(H_\mathsf{S}^2)$$

- transformed Hamiltonian

$$\boxed{\bar{H} \approx H_0 + H_\mathsf{S} + \frac{1}{2}[L_0^{-1} H_\mathsf{T}, H_\mathsf{T}] + \frac{1}{2}[L_0^{-1}[L_0^{-1} H_\mathsf{T}, H_\mathsf{S}], H_\mathsf{T}]}$$

Current formula

- number of electrons in lead ℓ: $N_\ell = \sum_{\mathbf{k}\sigma} n_{\ell\mathbf{k}\sigma}$
- mean current from left contact into wire

$$I_\ell(t) = e\frac{\mathrm{d}}{\mathrm{d}t}\langle N_\ell\rangle_t$$

- weakly-coupled environment in equilibrium:
  - leads at electro-chemical potential μ_ℓ: Fermi distributions $f_\ell(\epsilon) = f(\epsilon - \mu_\ell)$
  - phonons: Bose distribution $n_\mathrm{B}(\hbar\omega)$
- stationary current $I_\ell(t) \rightarrow$ $I = I_\mathrm{RL} - I_\mathrm{LR}$ with $I_{\ell'\ell} = e\sum_{\sigma\sigma'} W_{\ell'\sigma'\ell\sigma}\, p_\sigma$
  - $W_{\ell'\sigma'\ell\sigma}$: tunneling rate from lead ℓ into lead ℓ' with the molecular spin changing from σ to σ'
  - p_σ: probability for the molecular spin being in state σ

"elastic" cotunneling rate: spin $\sigma \longrightarrow \sigma$

- without phonons

$$W^{(0)}_{\ell'\sigma\ell\sigma} = \frac{\hbar}{2\pi} \int \mathrm{d}\epsilon\, \Gamma_{\ell'}(\epsilon)\, \Gamma_{\ell}(\epsilon)\, \Lambda^{(0),\mathrm{el}}_{\sigma}(\epsilon) f_\ell(\epsilon)\, [1 - f_{\ell'}(\epsilon)]$$

- phonon-assisted rate

$$W^{(1)}_{\ell'\sigma\ell\sigma} = \frac{\hbar}{8\pi^2} \int \mathrm{d}\epsilon'\, \Gamma_{\ell'}(\epsilon') \int \mathrm{d}\epsilon\, \Gamma_{\ell}(\epsilon)\, \Lambda^{(1)}_{\mathrm{el}}(\epsilon, \epsilon') N(\epsilon - \epsilon')\, f_\ell(\epsilon)\, [1 - f_{\ell'}(\epsilon')]$$

  - emission of phonons ϵ and absorption of phonon $-\epsilon$

$$N(\epsilon) = D(\epsilon/\hbar)\, [n_\mathrm{B}(\epsilon) + 1] + D(-\epsilon/\hbar)\, n_\mathrm{B}(-\epsilon)$$

  - energy denominators

$$\Lambda^{(0),\mathrm{el}}_{\sigma}(\epsilon) = \left(\frac{1}{E_{\bar\sigma} + U - \epsilon}\right)^2 + \left(\frac{1}{E_\sigma - \epsilon}\right)^2$$

$$\Lambda^{(1)}_{\mathrm{el}}(\epsilon, \epsilon') = \sum_\sigma \left(\frac{1}{E_\sigma + U - \epsilon}\,\frac{1}{E_{\bar\sigma} + U - \epsilon'} + \frac{1}{E_\sigma - \epsilon}\,\frac{1}{E_{\bar\sigma} - \epsilon'}\right)^2$$

“elastic” cotunneling rate: spin $\sigma \longrightarrow \sigma$

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$$\Lambda^{(1)}_{\mathrm{el}}(\epsilon,\epsilon') = \sum_{\sigma} \left(\frac{1}{E_{\sigma} + U - \epsilon}\, \frac{1}{E_{\bar\sigma} + U - \epsilon'} + \frac{1}{E_{\sigma} - \epsilon}\, \frac{1}{E_{\bar\sigma} - \epsilon'}\right)^2$$

Cotunneling rates II

- wide-band limit: $\Gamma_\ell(\epsilon) = \text{const.}$
- density of spin-phonon coupling $D(\omega \geq 0) = \gamma\hbar\,\omega_c^{1-s}\,\omega^s$
  $s \to$ coupling mechanism
- phonon-assisted cotunneling rates: lead $\ell \to \ell'$, molecular spin $\sigma \to \sigma'$

$$W^{(1)}_{\ell'\sigma'\ell\sigma} \approx \gamma\,(\hbar\omega_c)^{1-s}\frac{\hbar\Gamma_\ell\Gamma_{\ell'}}{8\pi^2}\left(\frac{1}{\Delta^2_{\bar{\sigma}'\ell}\,\Delta^2_{\sigma\ell'}} + \frac{1}{\Delta^2_{\sigma'\ell}\,\Delta^2_{\bar{\sigma}\ell'}}\right)$$
$$\times\,(k_B T)^{1+s}\,J_s\left(-\left(\mu_{\ell'\ell} + \Delta_{\sigma'\sigma}\right)/k_B T\right)\Theta\left(-\mu_{\ell'\ell} - \Delta_{\sigma'\sigma}\right)$$

  - with $\mu_{\ell'\ell} = \mu_{\ell'} - \mu_\ell$, and $\Delta_{\sigma'\sigma} = E_{\sigma'} - E_\sigma$,
  - dominant contribution $\Theta(\epsilon) = \dfrac{\epsilon}{1 - \exp(-\epsilon/k_B T)}$
  - voltage and temperature-dependent prefactors

“ohmic” bath: $J_1(a) = \frac{1}{6}\left(4\pi^2 + a^2\right)$

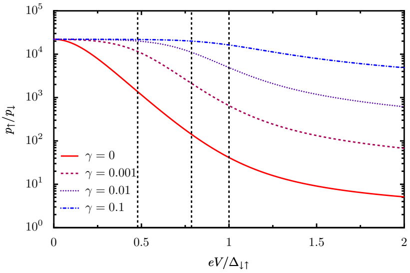
$$J_3(a) = \frac{1}{60}\left(4\pi^2 + a^2\right)\left(8\pi^2 + 3a^2\right)$$

Nonequilibrium effects: heating

- master equation for occupation probabilities p_σ

$$\dot{p}_\sigma = -W_{\bar{\sigma}\sigma} p_\sigma + W_{\sigma\bar{\sigma}} p_{\bar{\sigma}}$$

- total spin-flip rate $W_{\bar{\sigma}\sigma} = W^{\text{cot}}_{\bar{\sigma}\sigma} + W^{\text{flip}}_{\bar{\sigma}\sigma}$
  - cotunneling mediated spin-flip rate $W^{\text{cot}}_{\bar{\sigma}\sigma} = \sum_{\ell\ell'} W_{\ell'\bar{\sigma}\ell\sigma}$
  - intrinsic spin-flip rate $W^{\text{flip}}_{\bar{\sigma}\sigma} = \frac{2}{\hbar} N(\Delta_{\sigma\bar{\sigma}})$



with $s = 1$, $T = 0.1\Delta_{\downarrow\uparrow}$, $\hbar\Gamma_L = \hbar\Gamma_R = 0.2\Delta_{\downarrow\uparrow}$

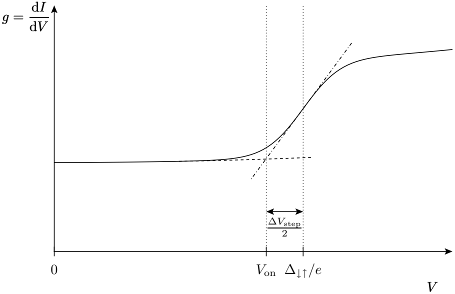
equilibrium $p_\uparrow/p_\downarrow = \exp(-\Delta_{\uparrow\downarrow}/k_B T)$ for

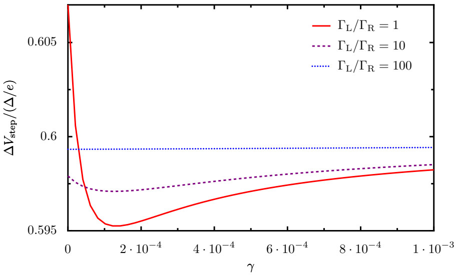
- $eV \ll |\Delta_{\uparrow\downarrow}|, k_B T$

or

- $W^{\text{flip}}_{\uparrow\downarrow} \gg W^{\text{cot}}_{\downarrow\uparrow}$

Width of inelastic cotunneling peak



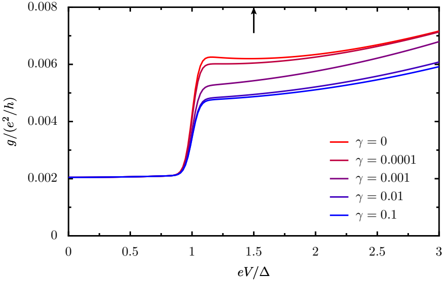


with $s = 1$, $T = 0.1\Delta_{\downarrow\uparrow}$, $\hbar\Gamma_L = \hbar\Gamma_R = 0.2\Delta_{\downarrow\uparrow}$

- leading contribution proportional to temperature T
- only slight change width due to spin-phonon coupling
- mostly due to heating effects (relevant for $\Gamma_L = \Gamma_R$)

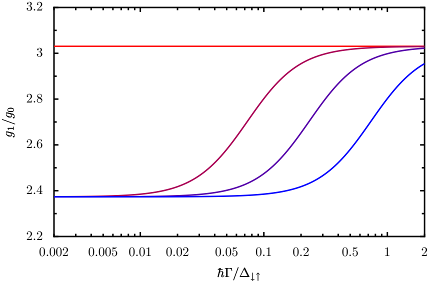
Determination of spin-flip rate

conductance-voltage characteristics



with $s = 1$, $T = 0.02\Delta_{\downarrow\uparrow}$, $\hbar\Gamma_L = \hbar\Gamma_R = 0.2\Delta_{\downarrow\uparrow}$

height of inelastic cotunneling step



- transition between heating and non-heating regime at molecule-lead coupling Γ_{on}

$$W_{\uparrow\downarrow}^{\text{flip}} = \frac{\hbar}{2\pi}\left(\frac{\Gamma_{\text{on}}}{|\Delta_{L\downarrow R\uparrow}|}\right)^2 \Theta(eV - \Delta_{\downarrow\uparrow})$$

Summary

- generalized Schrieffer-Wolff transformation for spin-flip assisted cotunneling
- spin-flip processes modify voltage- and temperature-dependendence of cotunneling rates
- heating effects allow one to determine strength of instrinsic spin-flip rates

Acknowledgements

- EU RTN QuEMolNa
- NoE MAGMANet