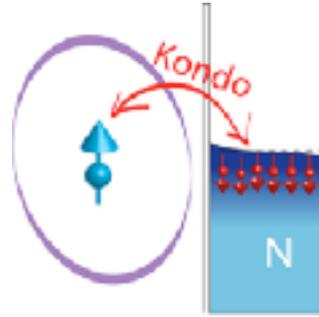


# Thermally driven out-of-equilibrium Kondo system

**Rosa López IFISC**  
Jong Soo Lim KIAS  
Miguel A. Sierra IFISC



**Phys. Rev. Lett. 121, 096801**

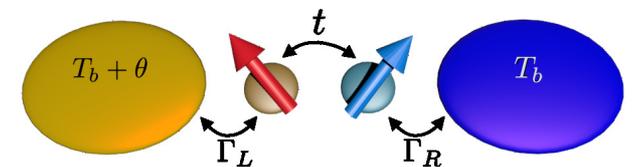
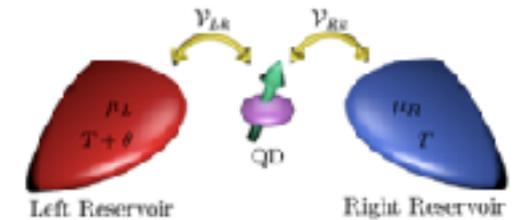


UNIT OF  
EXCELLENCE  
MARÍA  
DE MAEZTU





- Thermoelectric transport
- Kondo effect in quantum dots
- Thermally driven single dot
- Theoretical approaches
- Results(I)
- Thermally driven two impurity Kondo system
- Theoretical model
- Results (II)
- Conclusions



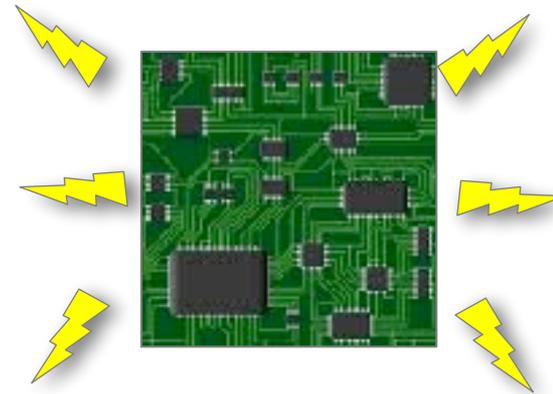
First experimental evidences of Kondo effect

D. Goldhaber-Gordon *et al.*, *Nature* 391, 156 (1998).

S. M. Cronenwett, T. H. Oosterkamp, L. P. Kouwenhoven, *Science* 281, 540 (1998).

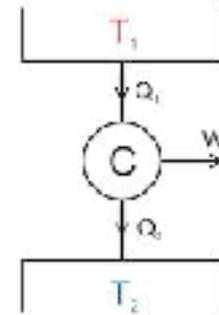
J. Schmid, J. Weis, K. Eberl, K. v. Klitzing, *Physica B* 256, 182 (1998).

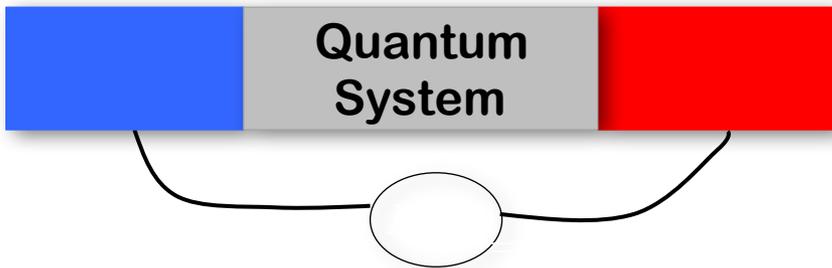
Circuit heat generation is one key limiting factor for scaling device speed



Waste heat recovery: typically 30-40 % efficiency for heatengines, waste heat 60%

THERMOELECTRICS is recognized as a potentially transformative energy conversion technology: heat is directly converted into electricity and vice versa



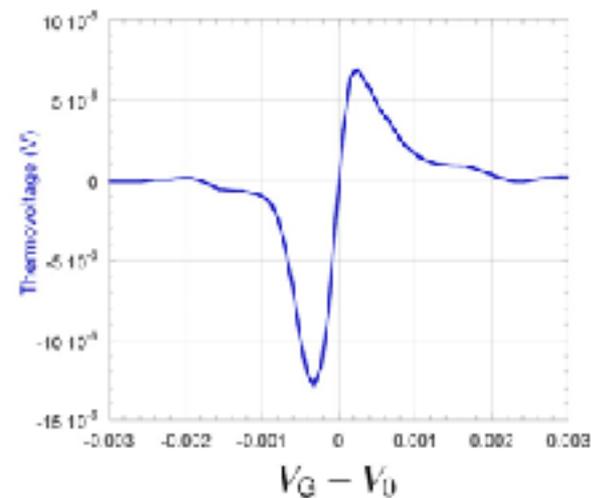
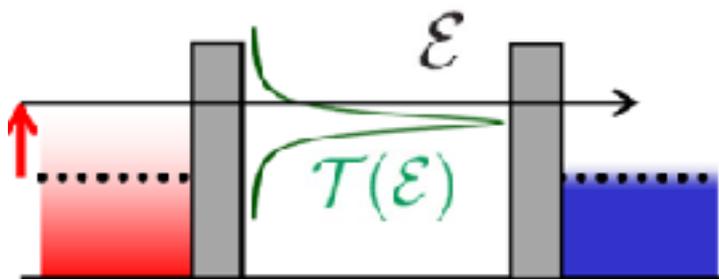


Large thermal gradients large as 13K/mV

R. Venkatasubramanian et al., Nature 413, 597 (2001)

Rectification effects due to interactions

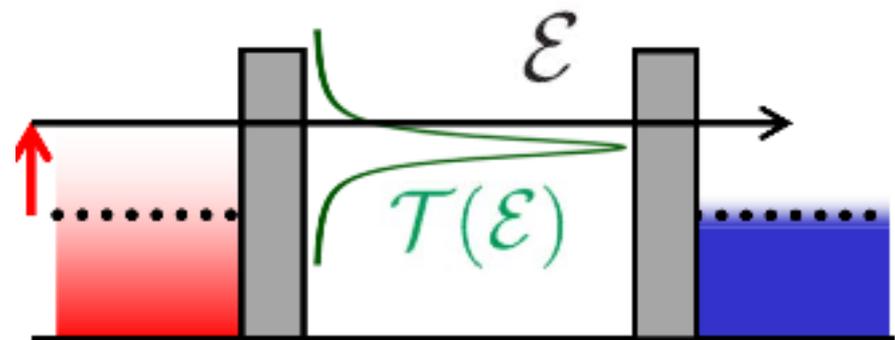
Thermopower changes sign, A.A.M. Staring et al., EPL 22, 57 (1993)





Nanosystems exhibit **REMARKABLE THERMOELECTRIC** properties

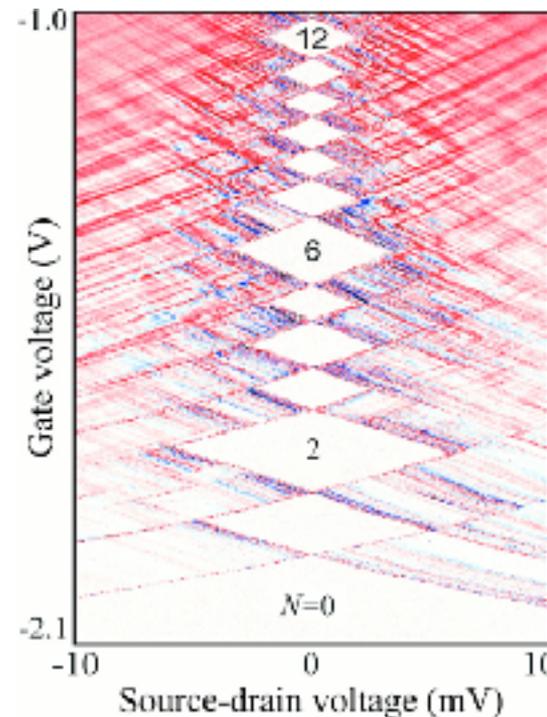
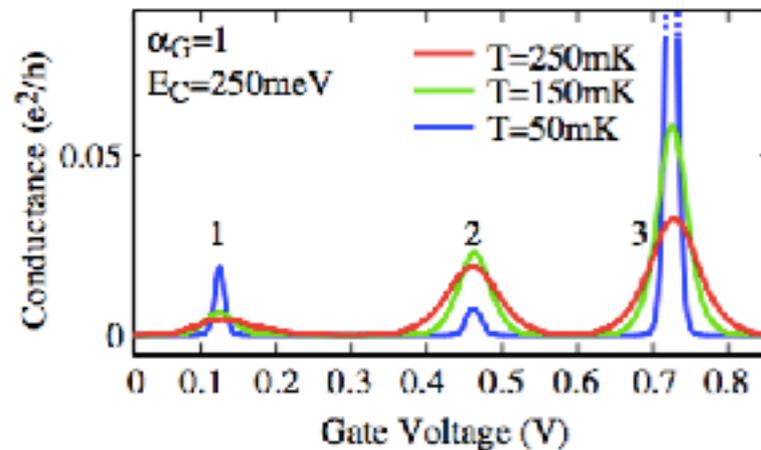
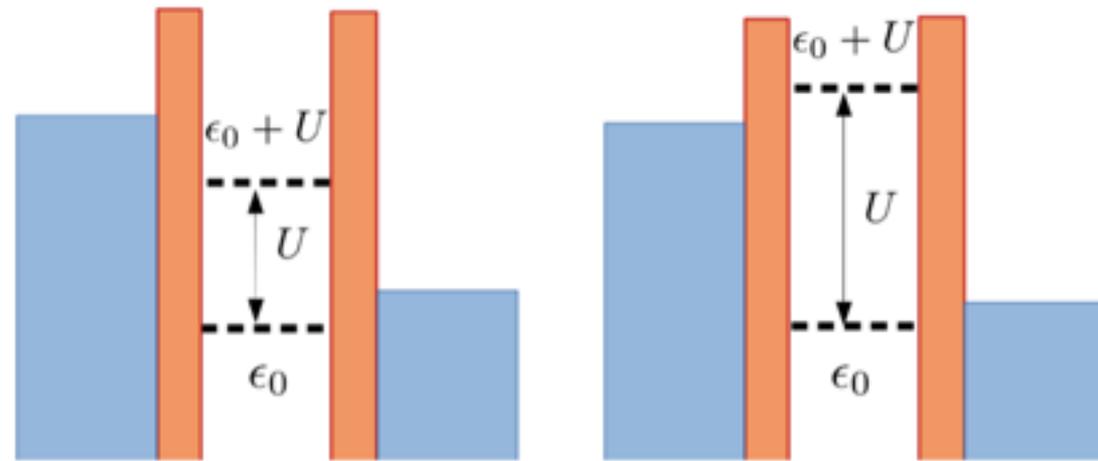
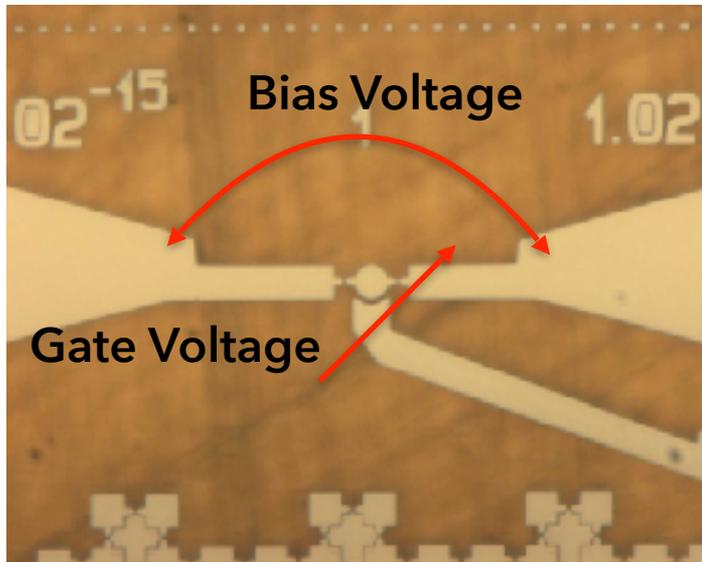
Specially when transport properties depend very much on **energy** as in the case of **quantum dots**



# Quantum Dots

## Coulomb blockade phenomena

Quantum dot: 0D system

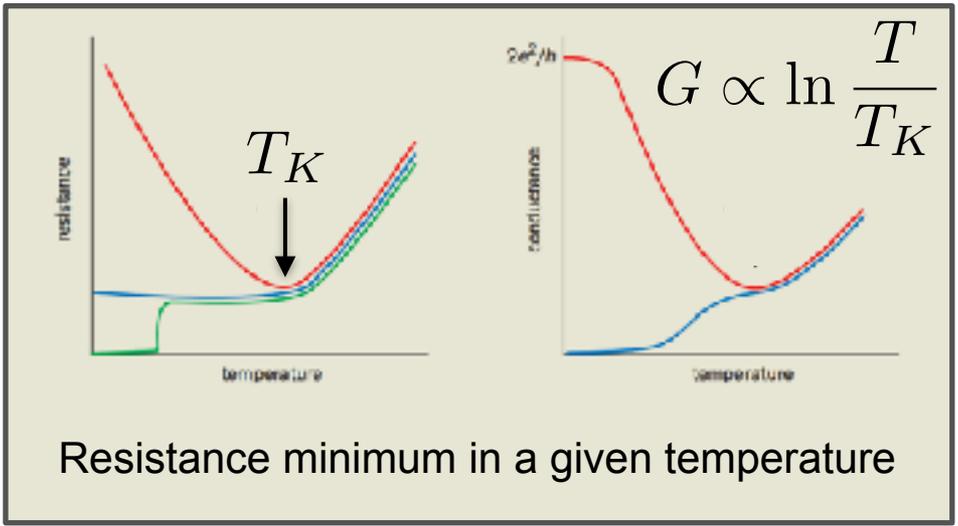
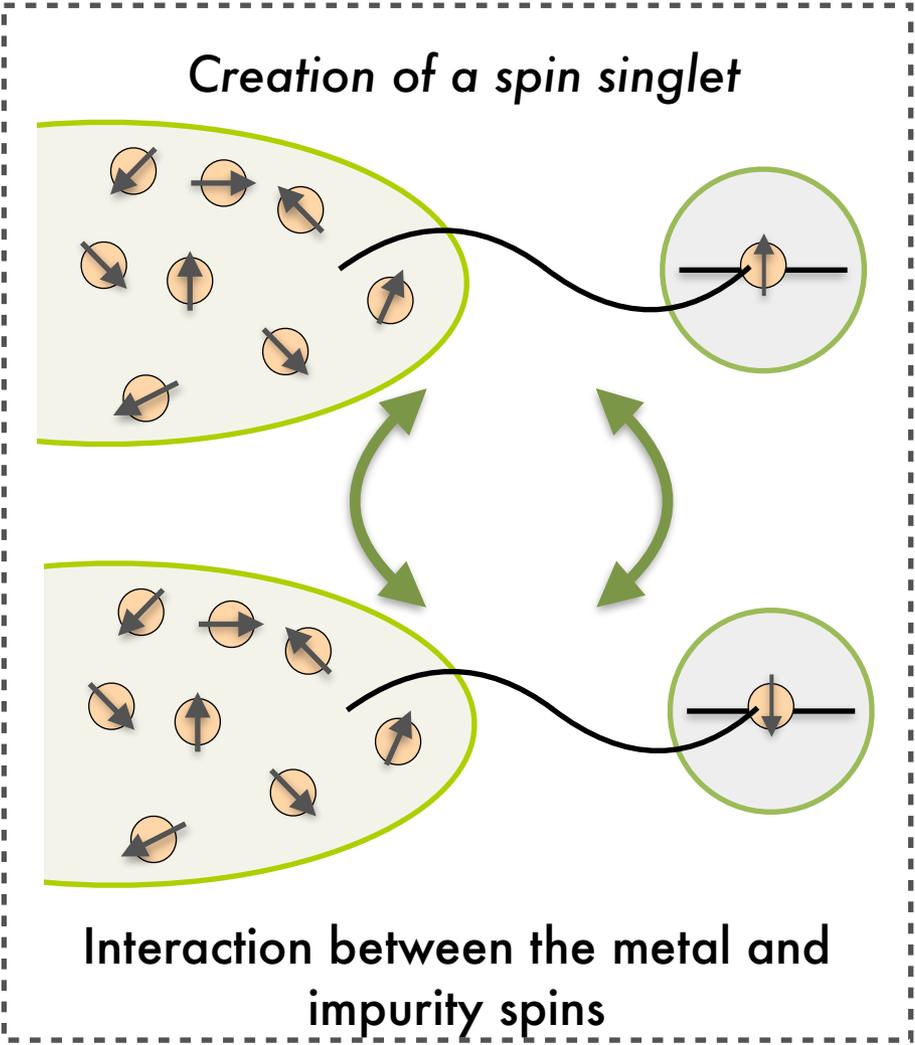


Energy levels  
Quantized

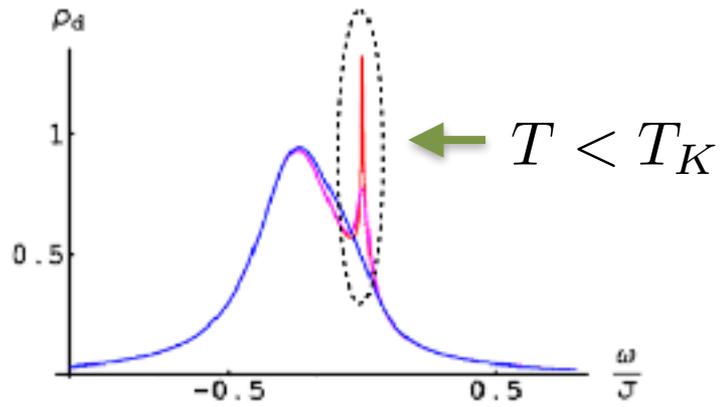
Coulomb energy  
 $E_c = q^2 / 2C$



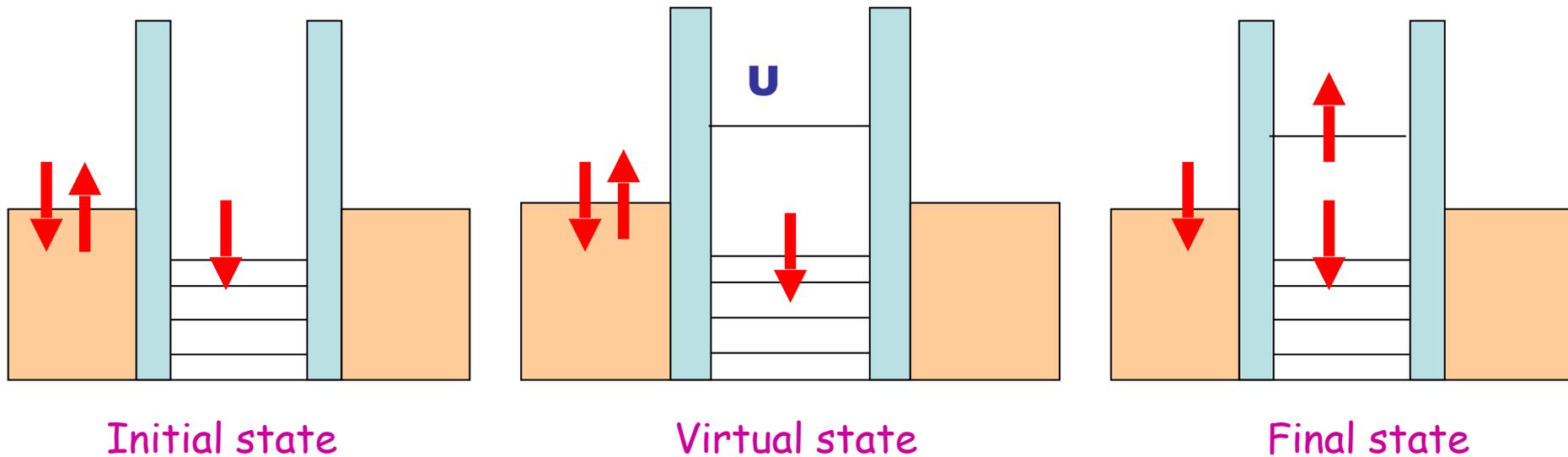
Resistance Minimum in Dilute Magnetic Alloys" Progress of Theoretical Physics 32 (1964) 37  
 J. Kondo



Temperature Kondo: characteristic energy scale

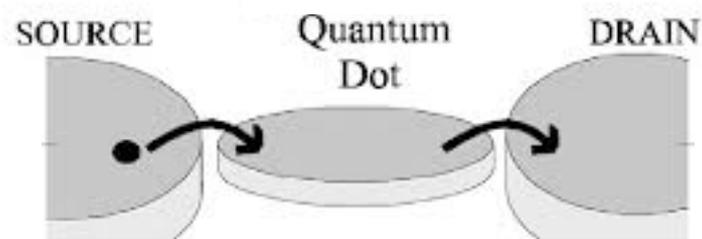


L.I. Glazman and M.E. Raikh JETP Lett. 47, 452(1988),  
 T.K..Ng and P.A. Lee, Phys. Rev. Lett. 61, 1768 (1988)



The resulting correlated motion gives rise to a Kondo resonance in the quasiparticle density of states at the Fermi energy

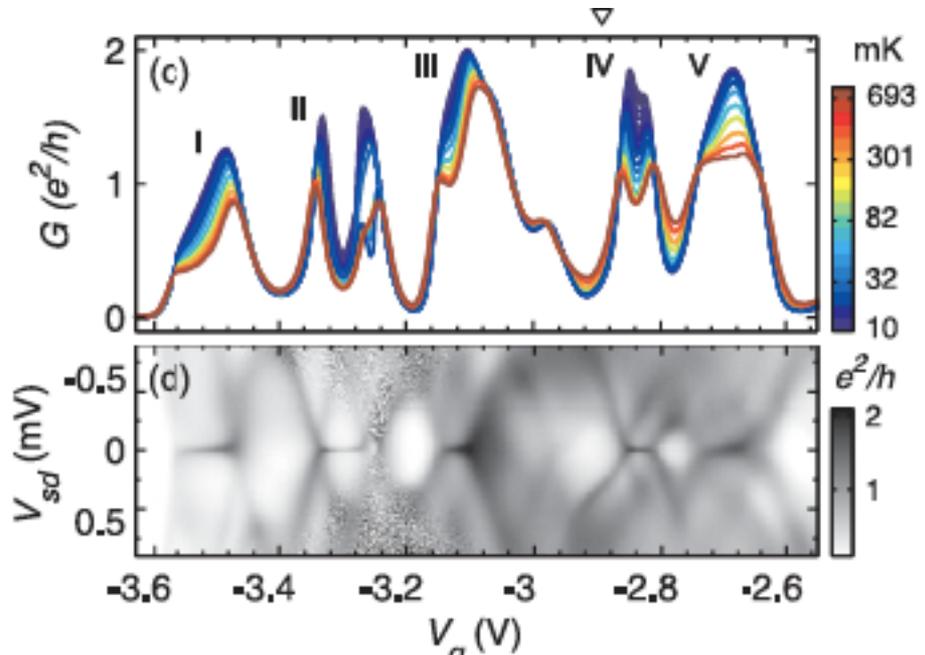
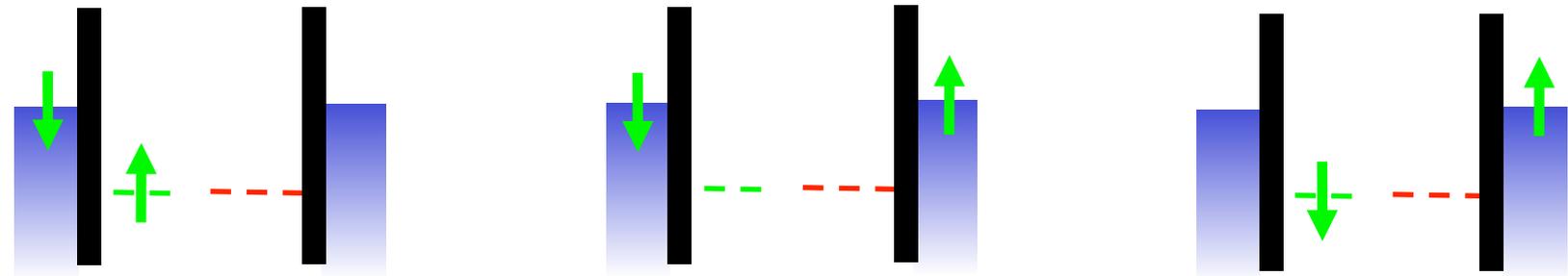
Due to this resonance the transmission through the quantum dot is perfect, the so-called "unitary limit"



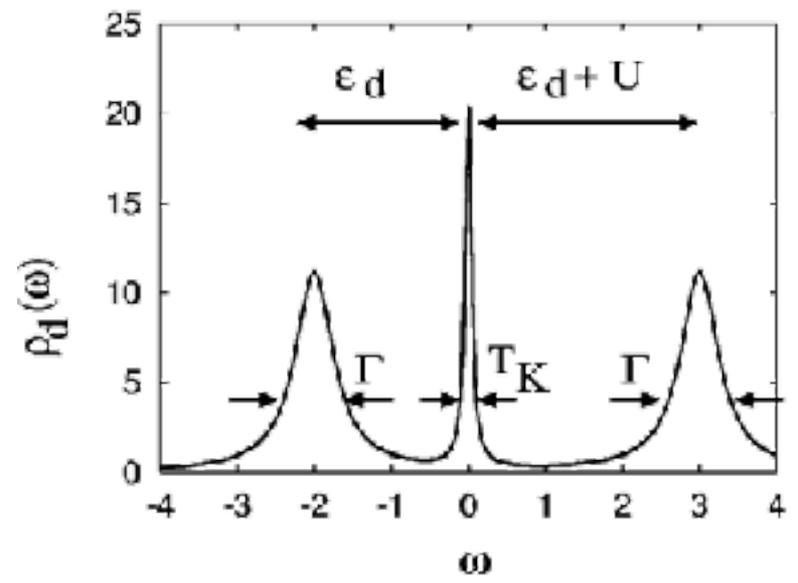
Unitary limit in the even valleys when the temperature is lowered

Appearance of the Zero Bias Anomaly in the Coulomb diamond

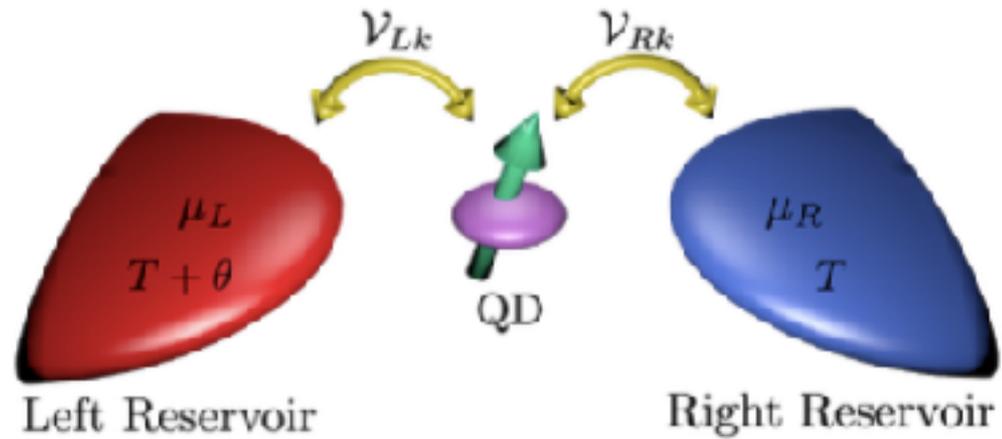
# SPIN KONDO

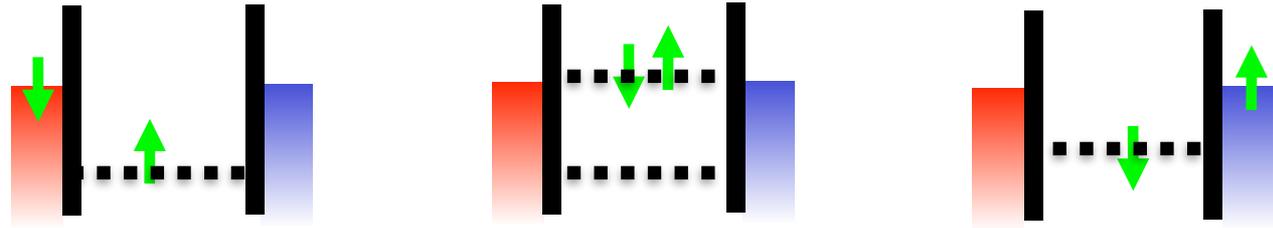
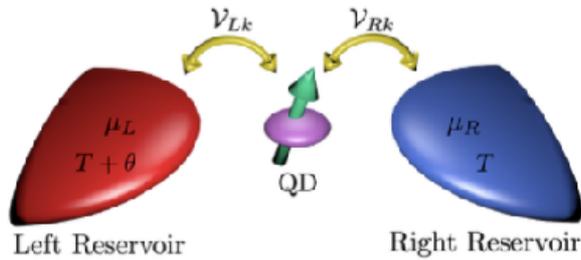


Impurity DOS in Kondo Regime



## Single Quantum Dot case





Kondo Hamiltonian  $\mathcal{H}_K = \mathcal{H}_0 + \mathcal{H}_1$

$$\mathcal{H}_0 = \sum_{\alpha k \sigma} \varepsilon_{\alpha k} C_{\alpha k \sigma}^\dagger C_{\alpha k \sigma} \quad \mathcal{H}_1 = \sum_{\alpha k \sigma \beta q s} \mathcal{J}_{\alpha \beta}(t) x_{\sigma s} C_{\alpha k \sigma}^\dagger C_{\beta q s}$$

$$x_{\sigma s} = \delta_{\sigma s} / 4 + \hat{S}_l^l s_{\sigma s}^l$$

$$\mathcal{J}_{\alpha \beta}(t) = \mathcal{J}_{\alpha \beta}^{(0)} \exp\left(-\frac{ie}{\hbar} [V_\alpha - V_\beta] t\right),$$

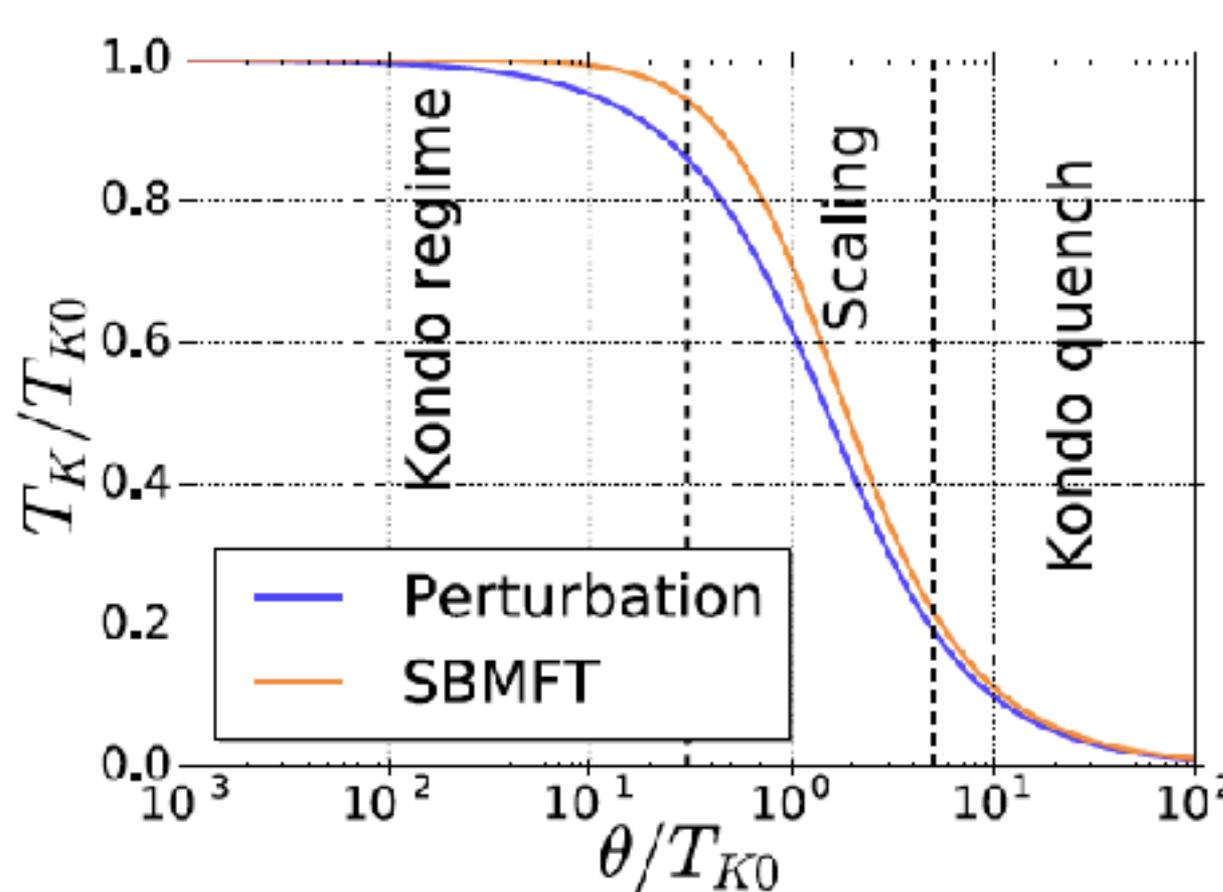
Localized and delocalized electrons:  
spin operators

$$\mathcal{J}_{\alpha \beta}^{(0)} = -\nu_\alpha \nu_\beta U / [\varepsilon_d (U + \varepsilon_d)]$$

$$S_l, l = x, y, z$$

Poor man scaling procedure

$$G = -\frac{3e^2\pi}{4\hbar}v^2[\mathcal{J}_{LR}^{(0)}]^2 \left( 1 - \frac{\nu}{2}(\mathcal{J}_{LL}^{(0)} + \mathcal{J}_{RR}^{(0)}) \ln \left| \frac{k_B^2 T_L T_R}{D_0^2} \right| \right) - \frac{e^2\pi}{4\hbar}v^2[\mathcal{J}_{LR}]^2. \quad (11)$$



$$k_B T_{K0} = D_0 \exp \left[ \frac{\pi \varepsilon_d (U + \varepsilon_d)}{U \Gamma} \right]$$

$$\Gamma_\alpha = \pi \nu |\mathcal{V}_\alpha|^2$$

$$\Gamma = \Gamma_\cdot + \Gamma_\bullet$$

$$T_K(\theta) = \sqrt{\left(\frac{\theta}{2}\right)^2 + T_{K0}^2} - \frac{\theta}{2}$$

The Kondo scale is determined when the second order contribution in perturbation theory prevails on the first order

## Anderson Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{leads}} + \mathcal{H}_{\text{dot}} + \mathcal{H}_{\text{tun}}$$

$$\mathcal{H}_{\text{leads}} = \sum_{\alpha k \sigma} \varepsilon_{\alpha k} C_{\alpha k \sigma}^\dagger C_{\alpha k \sigma}$$

$$\mathcal{H}_{\text{dot}} = \sum_{\sigma} \varepsilon_d d_{\sigma}^\dagger d_{\sigma} + U d_{\uparrow}^\dagger d_{\uparrow} d_{\downarrow}^\dagger d_{\downarrow},$$

$$\mathcal{H}_{\text{tun}} = \sum_{\alpha k \sigma} v_{\alpha k} C_{\alpha k \sigma}^\dagger d_{\sigma} + \text{H.c.}$$

$$U \rightarrow \infty$$

$$d_{\sigma} = b^\dagger f_{\sigma}$$

pseudofermion operator  $f_{\sigma}$

boson field operator  $b^\dagger$

$$\mathcal{H}_{\text{Lag}} = \lambda \left( b^\dagger b + \sum_{\sigma} f_{\sigma}^\dagger f_{\sigma} - 1 \right)$$

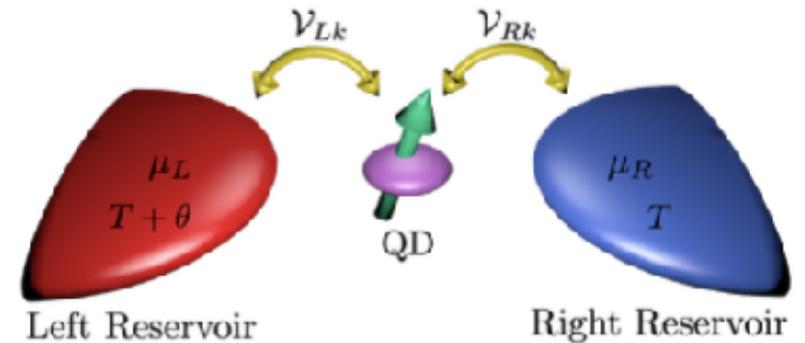
$$\mathcal{H}_{\text{tun}} = \sum_{\alpha k \sigma} v_{\alpha k} C_{\alpha k \sigma}^\dagger b^\dagger f_{\sigma} + \text{H.c.}$$

## Mean-field approach

$$\langle b \rangle = \tilde{b}$$

$$\sum_{\alpha k \sigma} \tilde{v}_{\alpha k} G_{f_{\sigma}, \alpha k \sigma}^{\leq}(t, t) = -i N \lambda |\tilde{b}|^2 / \hbar$$

$$\sum G_{f_{\sigma}, f_{\sigma}}^{\leq}(t, t) = i(1 - N |\tilde{b}|^2) / \hbar$$



$$G_{f_{\sigma}, f_{\sigma}}^{\leq}(t, t') = -(i/\hbar) \langle f_{\sigma}^\dagger(t') f_{\sigma}(t) \rangle$$

$$G_{f_{\sigma}, \alpha k \sigma}^{\leq}(t, t') = -(i/\hbar) \langle C_{\alpha k \sigma}^\dagger(t') f_{\sigma}(t) \rangle$$

## Mean field parameters

$$\tilde{\Gamma} = |\tilde{b}|^2 \Gamma \quad \text{Kondo width, Kondo temperature}$$

$$\tilde{\varepsilon}_d = \varepsilon_d + \lambda \quad \text{Kondo peak position}$$

## Mean field equations

$$\sum_{\alpha} \frac{\Gamma_{\alpha}}{\Gamma} \left[ \log \left| \frac{2\pi k_B T_{\alpha}}{D} \right| + \psi \left( \frac{1}{2} + \frac{i\tilde{\varepsilon}_d + \tilde{\Gamma}}{2\pi k_B T_{\alpha}} \right) \right] = \frac{\pi N \varepsilon_d}{2\Gamma}$$

Electrically driven

$$\tilde{\Gamma}(V) \tilde{\varepsilon}_d = 0$$

$$\tilde{\varepsilon}_d^2 - \left( \frac{eV}{2} \right)^2 - [\tilde{\Gamma}(V)]^2 = -(k_B T_{K0})^2$$

Thermally driven

$$\tilde{\varepsilon}_d = 0$$

$$\tilde{\Gamma} = k_B T_{K0} e^{-\frac{\pi^2}{12} \frac{T_L^2 + T_R^2}{(T_{K0})^2}}$$

Electrically driven

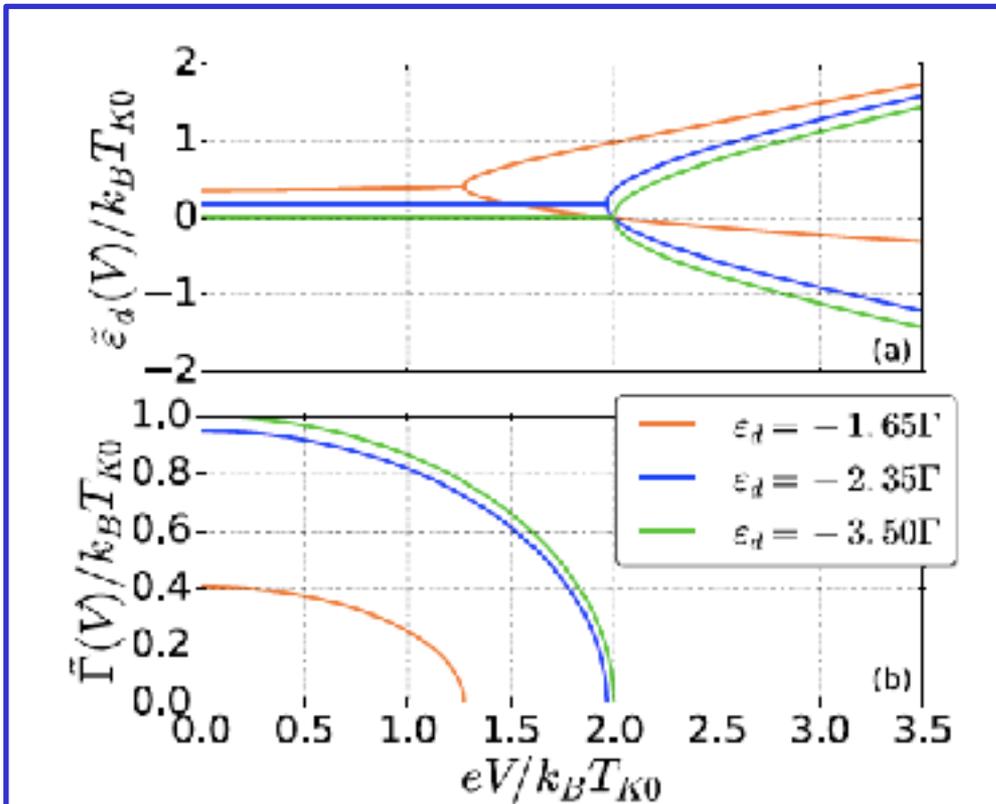


FIG. 3. Position of the SBMFT (a) renormalized energy level  $\tilde{\epsilon}_d$  and (b) width  $\tilde{\Gamma}$  as a function of the applied voltage for different dot level positions. The case  $\epsilon_d = -3.5\Gamma$  agrees with the analytical result given by Eq. (24). Parameters:  $D = 100\Gamma$ ,  $k_B T = 0$ , and  $\Gamma_L = \Gamma_R = \Gamma/2$ .

Bias voltage induced Kondo splitting

Thermally driven

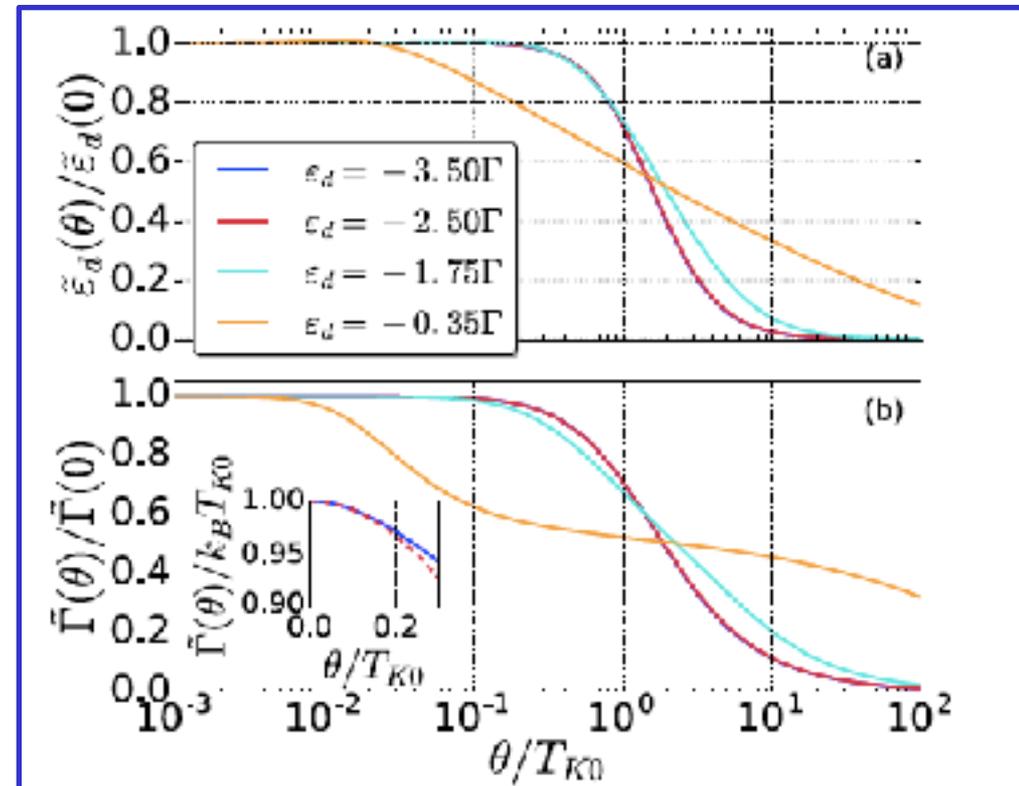


FIG. 4. (a) Renormalized dot gate position  $\tilde{\epsilon}_d$  and (b) resonance width  $\tilde{\Gamma}$  as a function of the thermal bias  $\theta$  for different  $\epsilon_d$  values within SBMFI. (Inset) Resonance width versus the thermal bias from a numerical calculation (solid line) and from the analytical expression given by Eq. (27) (dashed line) for  $\epsilon_d = -3.5\Gamma$ . Parameters:  $D = 100\Gamma$ ,  $k_B T = 0$ , and  $\Gamma_L = \Gamma_R = \Gamma/2$ .

Thermal bias Kondo width quenching

$$I = -\frac{e}{h} \int_{-\infty}^{\infty} d\omega \sum_{\sigma} \frac{4\Gamma_L\Gamma_R}{\Gamma} \text{Im}[G_{\sigma,\sigma}^r(\omega)] [f_L(\omega) - f_R(\omega)]$$

$$L_2 = \frac{4\pi^2 ek_B^2}{3h} \tilde{\Gamma}_L \tilde{\Gamma}_R \frac{\tilde{\epsilon}_d}{\tilde{\epsilon}_d^2 + \tilde{\Gamma}^2}$$

Electrically driven

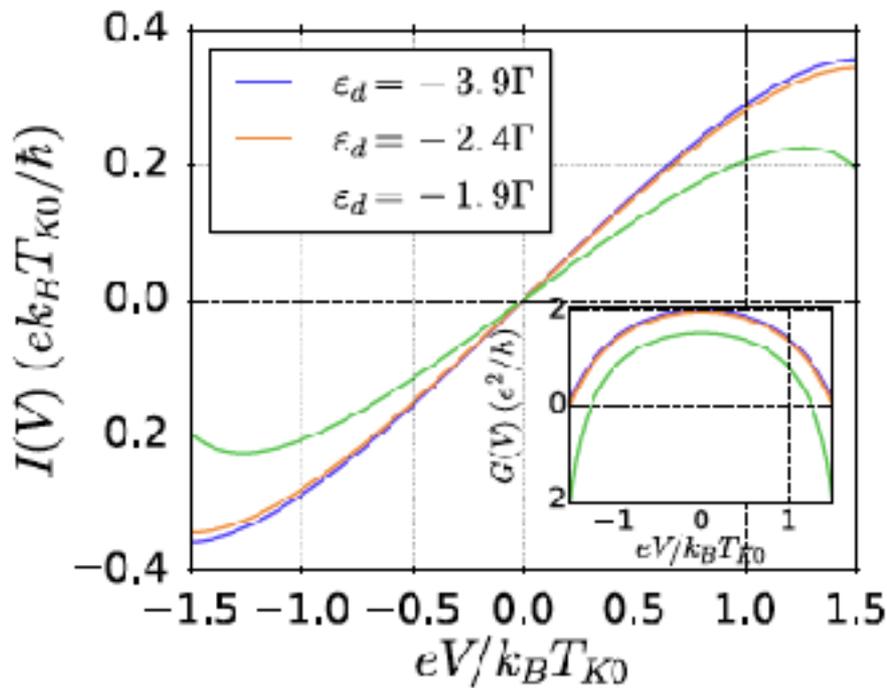


FIG. 5. Current-voltage characteristics of a single level quantum dot in the Kondo regime using slave-boson mean-field theory for different values of the gate voltage (level position). (Inset) Differential conductance of the quantum dot as a function of the applied voltage. Parameters:  $D = 100\Gamma$ ,  $k_B T = 0$ , and  $\Gamma_L = \Gamma_R = \Gamma/2$ .

Unitary limit in the deep Kondo regime

Thermally driven

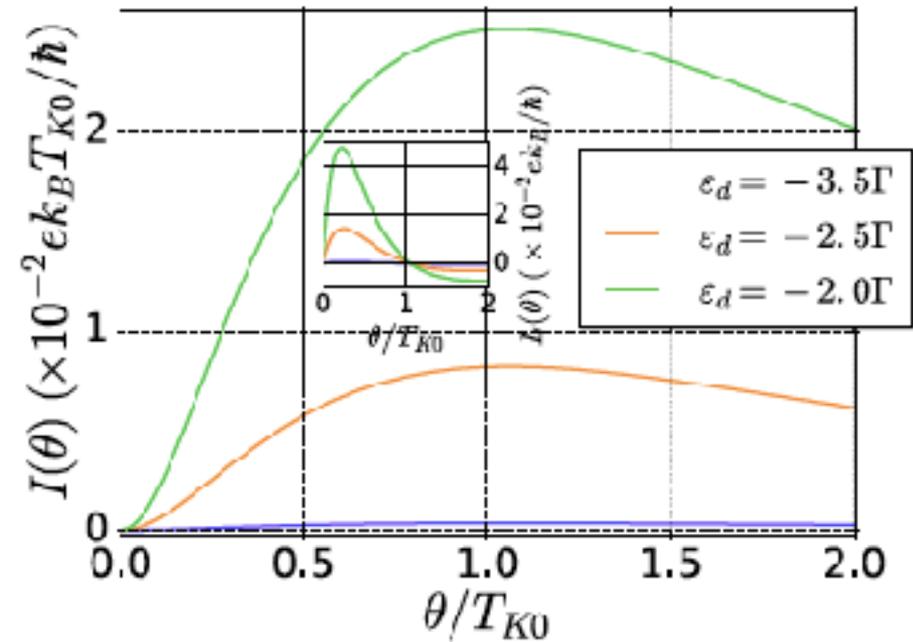


FIG. 6. Thermocurrent as a function the thermal gradient  $\theta$  of a single level quantum dot in the Kondo regime using slave-boson mean-field theory for different values of the dot gate position. Inset: thermoelectric conductance as a function of the thermal bias for the same dot gate positions. Parameters:  $D = 100\Gamma$ ,  $k_B T = 0$ , and  $\Gamma_L = \Gamma_R = \Gamma/2$ .

Larger thermocurrent when charge fluctuations are present

By using EOM technique the dot Green function reads

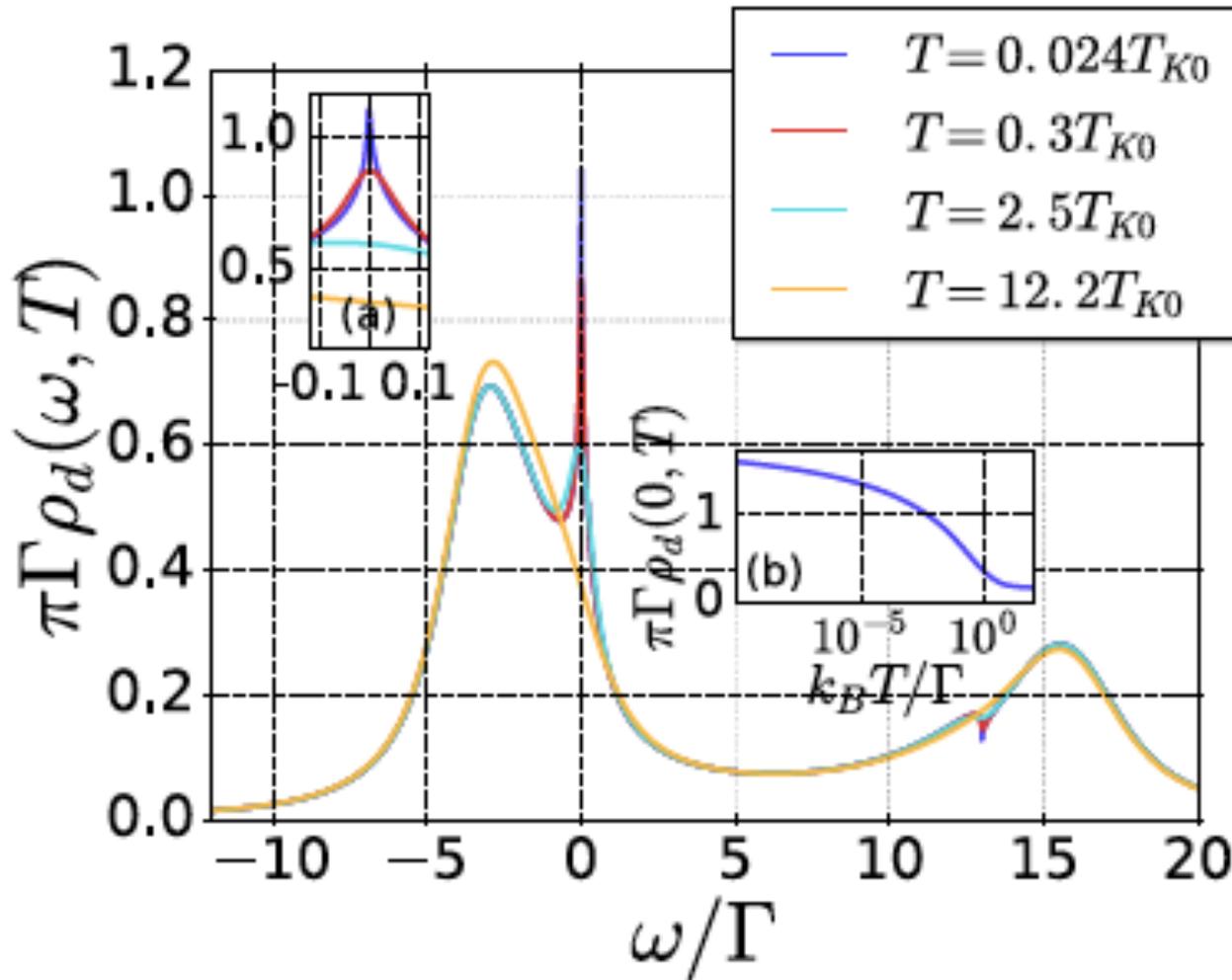
$$G_{\sigma,\sigma}^r(\omega) = \frac{1 - \langle \tilde{n}_{\bar{\sigma}} \rangle}{\omega - \varepsilon_d - \Sigma_0 + U \Sigma_1 / [\omega - \varepsilon_d - U - \Sigma_0 - \Sigma_3]} + \frac{\langle \tilde{n}_{\bar{\sigma}} \rangle}{\omega - \varepsilon_d - \Sigma_0 - U - U \Sigma_2 / [\omega - \varepsilon_d - \Sigma_0 - \Sigma_3]}$$

$$T_{K0} \approx \sqrt{2\Gamma U} \exp \left[ -\frac{\pi |\varepsilon_d| (U + \varepsilon_d)}{2\Gamma U} \right]$$

Self-energies are determined and the dot occupation is computed self-consistently

$$\left[ \Sigma_0 \quad \Sigma_1 \quad \Sigma_2 \quad \Sigma_3 \right] \langle \tilde{n}_{\bar{\sigma}} \rangle$$

## Equilibrium spectral density



Finite U calculation

The Kondo peak height decreases as the background temperature enhances

Here, no dephasing is included

FIG. 8. Finite- $U$  quantum dot spectral density at equilibrium for different background temperatures. Parameters:  $\varepsilon_d = -3.5\Gamma$ ,  $D = 100\Gamma$ , and  $U = 20\Gamma$ . (Insets) (a) Detail of the dot spectral density of states around the Fermi energy. (b) Height of the Kondo peak as a function of the background temperature.

## Electrical and thermal driven cases: spectral density

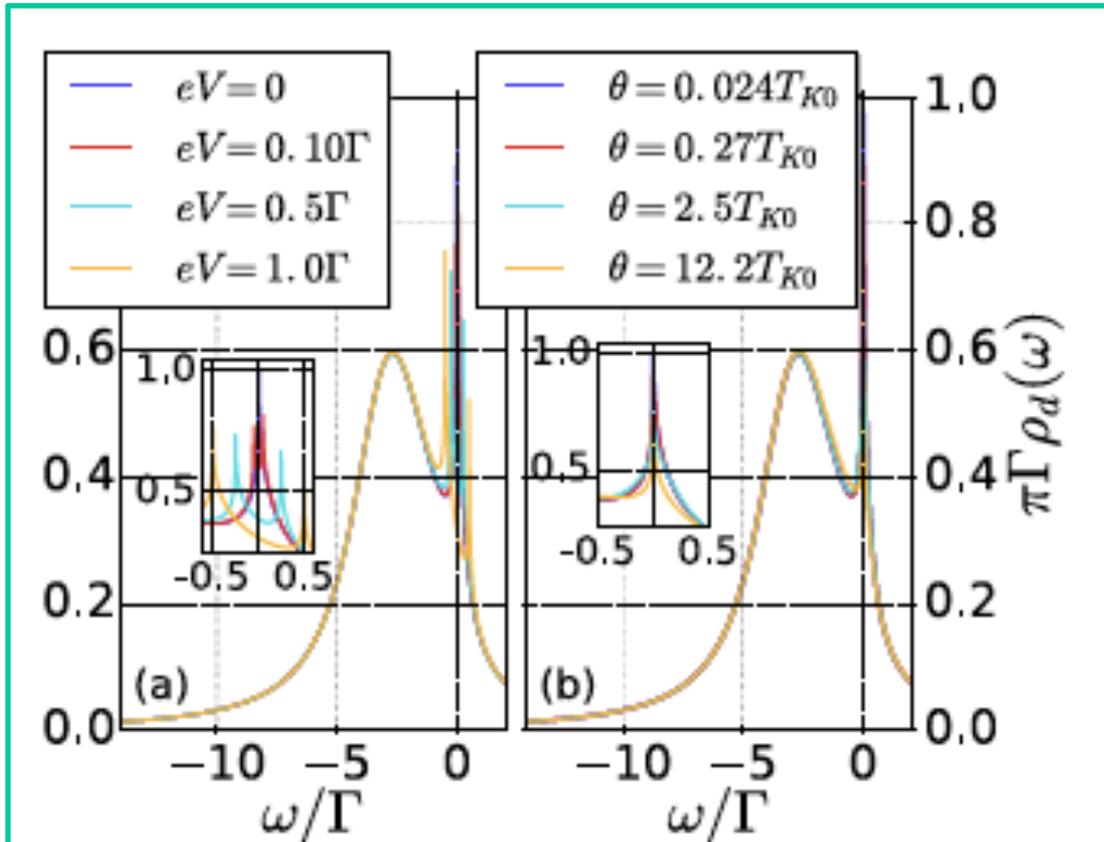


FIG. 7. (a) Nonequilibrium infinite- $U$  quantum dot spectral density of states for different  $eV$  values. (Inset) Detail of the density of states around the Fermi energy ( $\varepsilon_F = 0$ ). (b) Nonequilibrium infinite- $U$  quantum dot spectral density of states for different thermal gradients. The background temperature is set at  $T = 0.024T_{K0}$ . (Inset) Detail of the density of states around the Fermi energy ( $\varepsilon_F = 0$ ). Parameters:  $\varepsilon_d = -3.5\Gamma$ ,  $D = 100\Gamma$ , and  $T = 0.024T_{K0}$ .

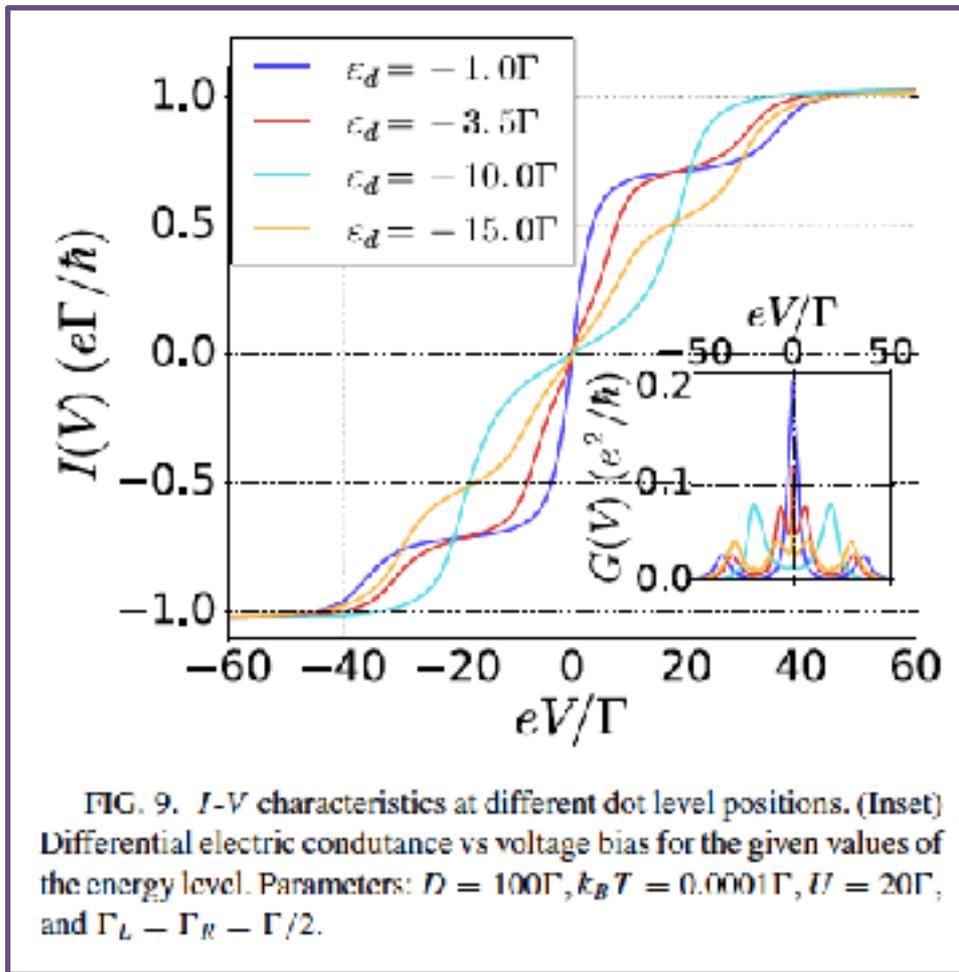
The Kondo peak splits with the bias voltage

Kondo peak is quenched with the thermal bias

Artifact of the model: the cold reservoirs always yields a Kondo singularity even for large thermal gradients

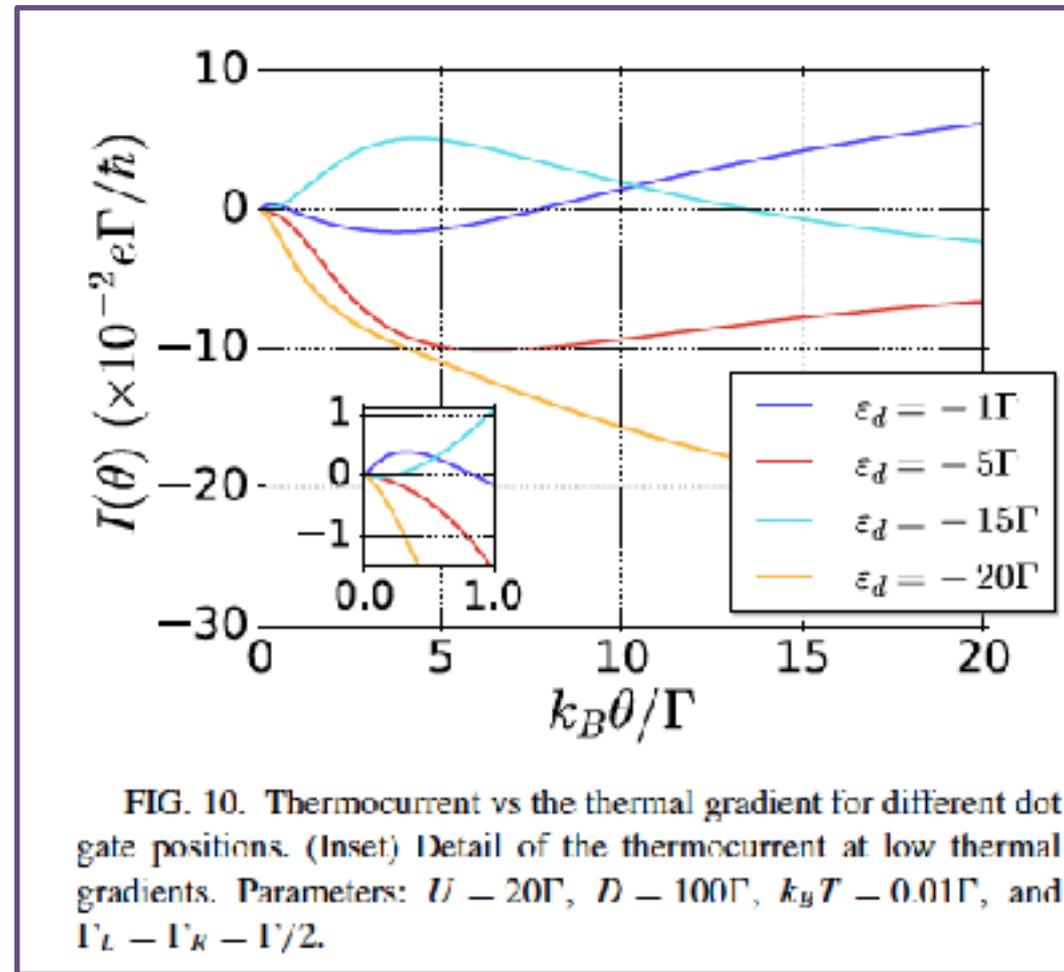
## Electrical current

Electrically driven



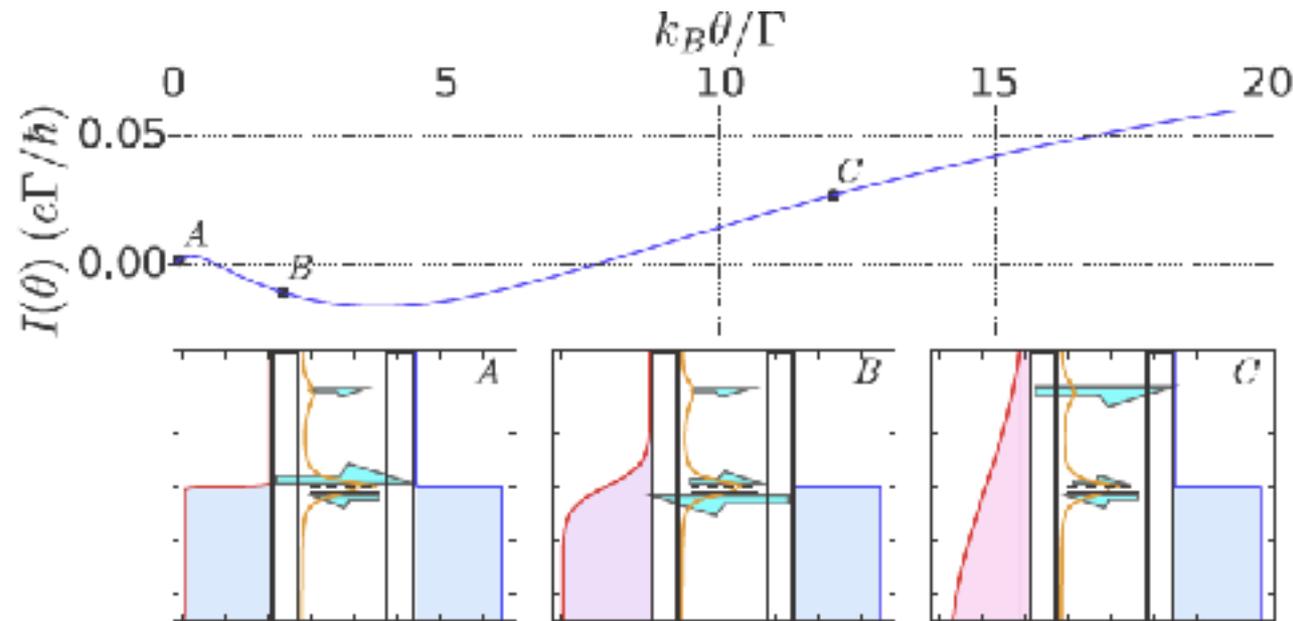
Steps at the dot resonances: mean-field peaks and Kondo singularity

Thermally driven



Non trivial zero at the spin and charge fluctuation energy scales

## Nontrivial zeros at the thermoelectrical current in the finite $U$ case



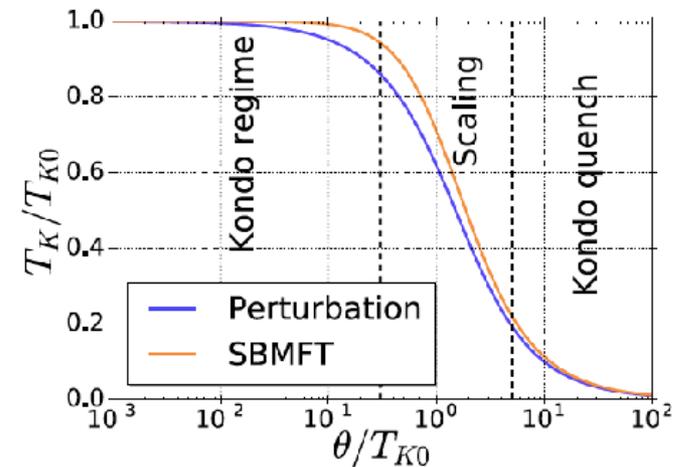
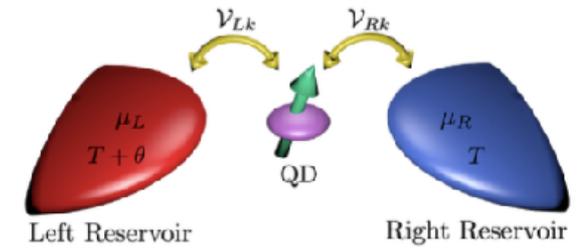
- A** e-like dominant Kondo peak
- B** h-like dominant Kondo peak and e-like contribution from the mean field peak
- C** e-like dominant from the second mean field peak, the others are h-like

The different electron-like and hole-like contributions of the Kondo and mean-field peaks leads to the appearance of the nontrivial zeros in the thermoelectrical current when the thermal bias increases



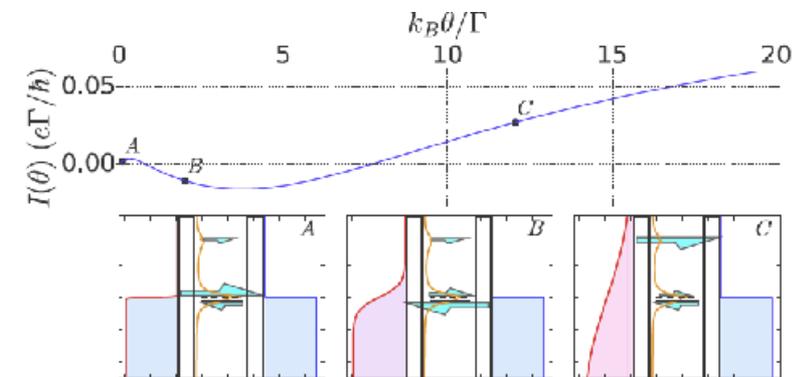
## System

- ➔ A single quantum dot is considered in the Kondo regime.
- ➔ The Kondo effect is quenched with the thermal bias: results from perturbation theory, SBMFT, and EOM

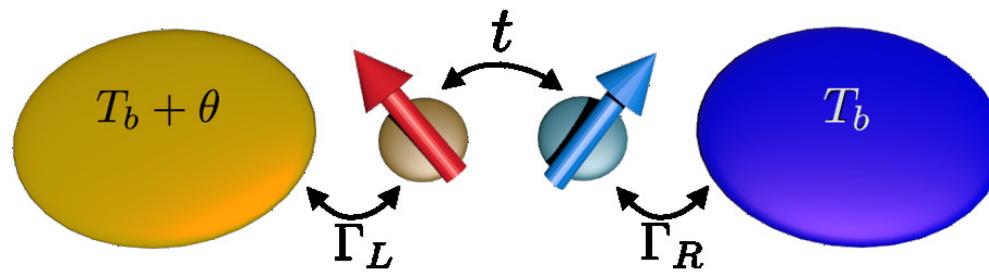


## Transport

- ➔ Thermocurrent
  - ▶ Nonlinear behavior
  - ▶ Appearance of nontrivial zeros



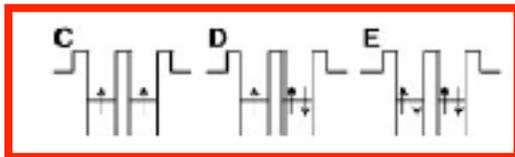
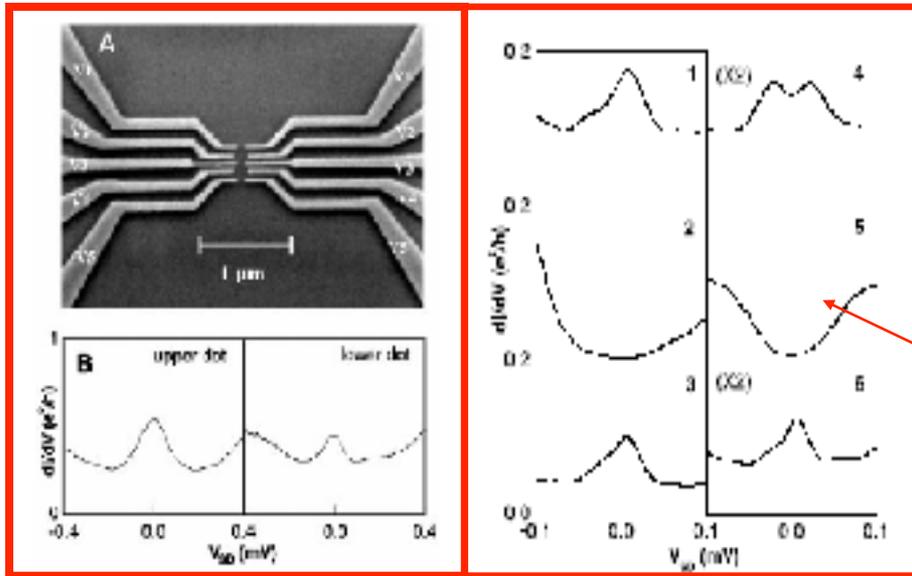
## Double Quantum Dot case



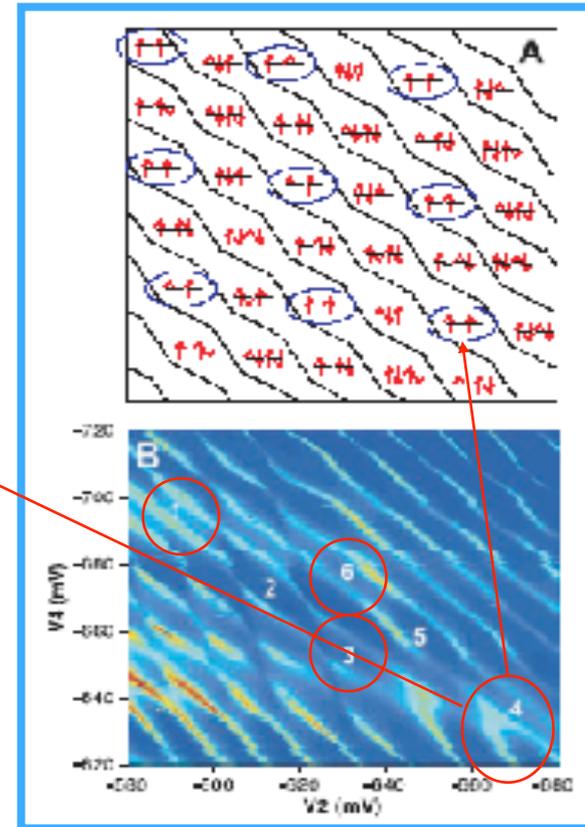
## The Kondo Effect in an Artificial Quantum Dot Molecule

H. Jeong, A. M. Chang and M. R. Melloch

*Science* 293 (5538), 2221-2223.  
DOI: 10.1126/science.1063182



$\Delta = 45 \mu\text{eV}$

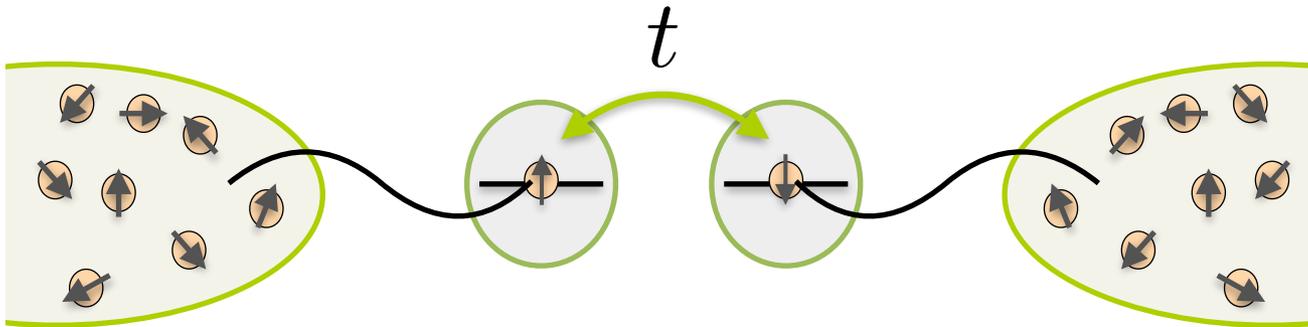


Kondo

- 1
- 3
- 4
- 6

Hybridization of Kondo singlets into a coherent superposition of Kondo states

**Bonding-Antibonding Kondo states!**



Two magnetic impurities in the Kondo regime

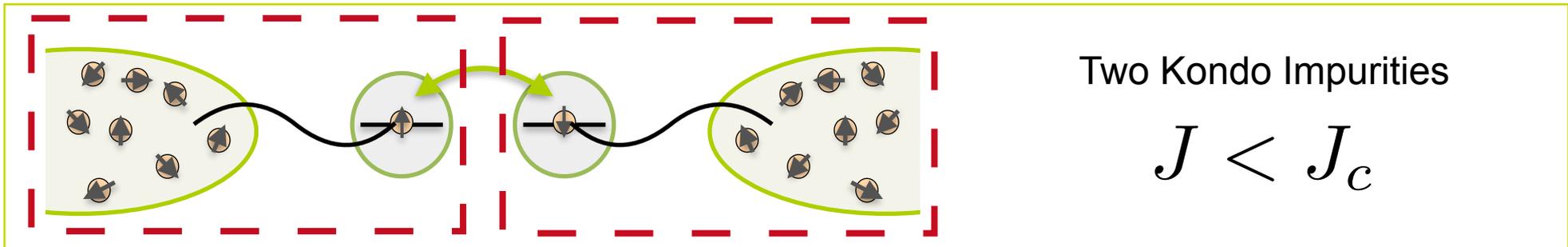
Crossover at  $J = J_c$

$$J = \frac{4t^2}{U}$$

Superexchange

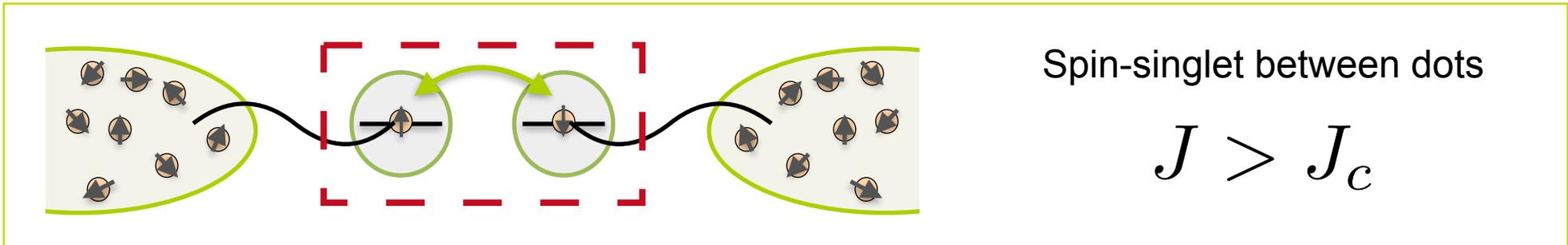
$t$  → Interdot tunneling amplitude

$U$  → Intradot Coulomb interaction



Two Kondo Impurities

$$J < J_c$$

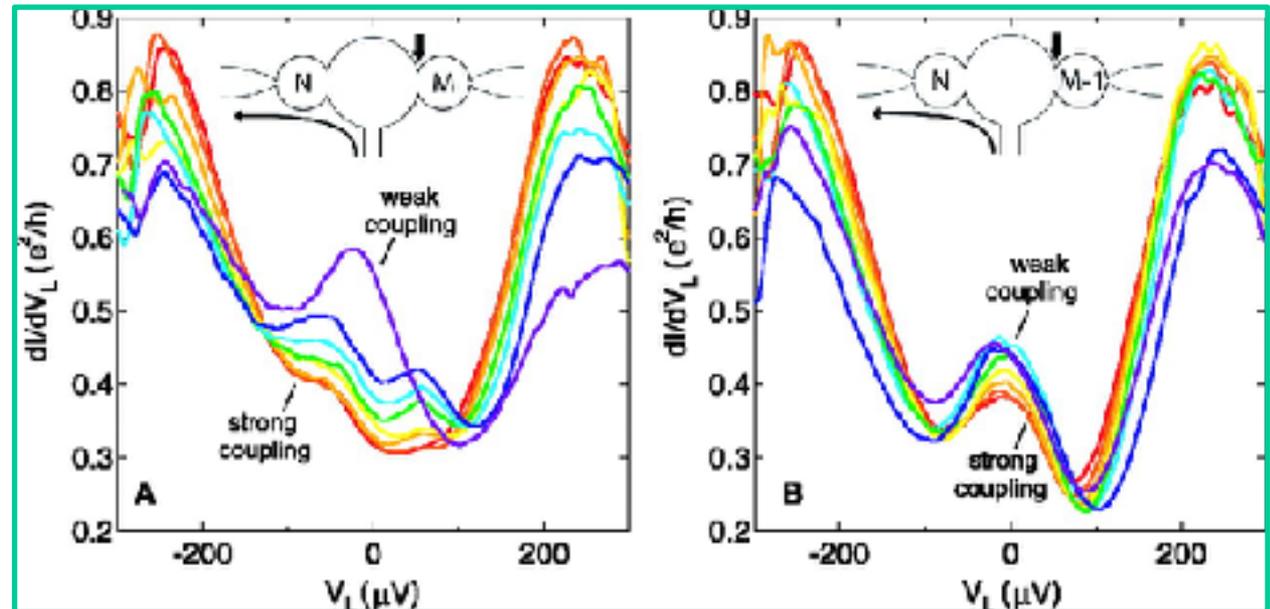
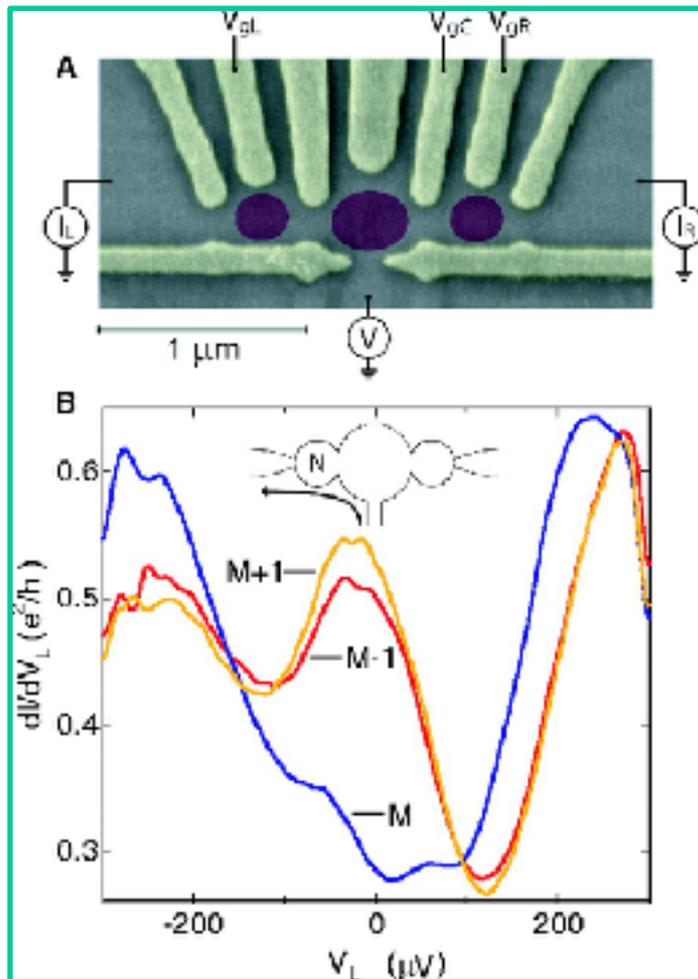


Spin-singlet between dots

$$J > J_c$$

# Tunable Nonlocal Spin Control in a Coupled-Quantum Dot System

N. J. Craig, et al., Science 304, 565 (2004)



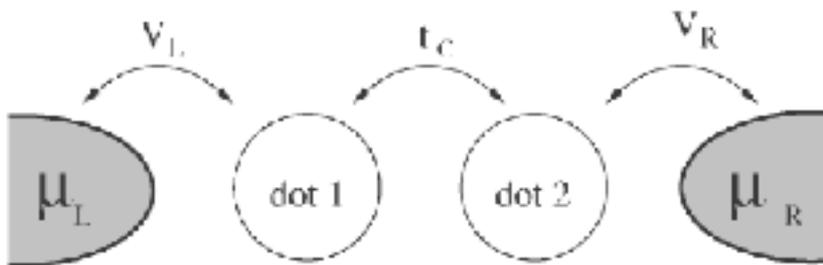
Coherent control of quantum dot spins by a nonlocal RKKY-like interaction

$$\mathcal{H}_J = J \hat{S}_L \cdot \hat{S}_R$$

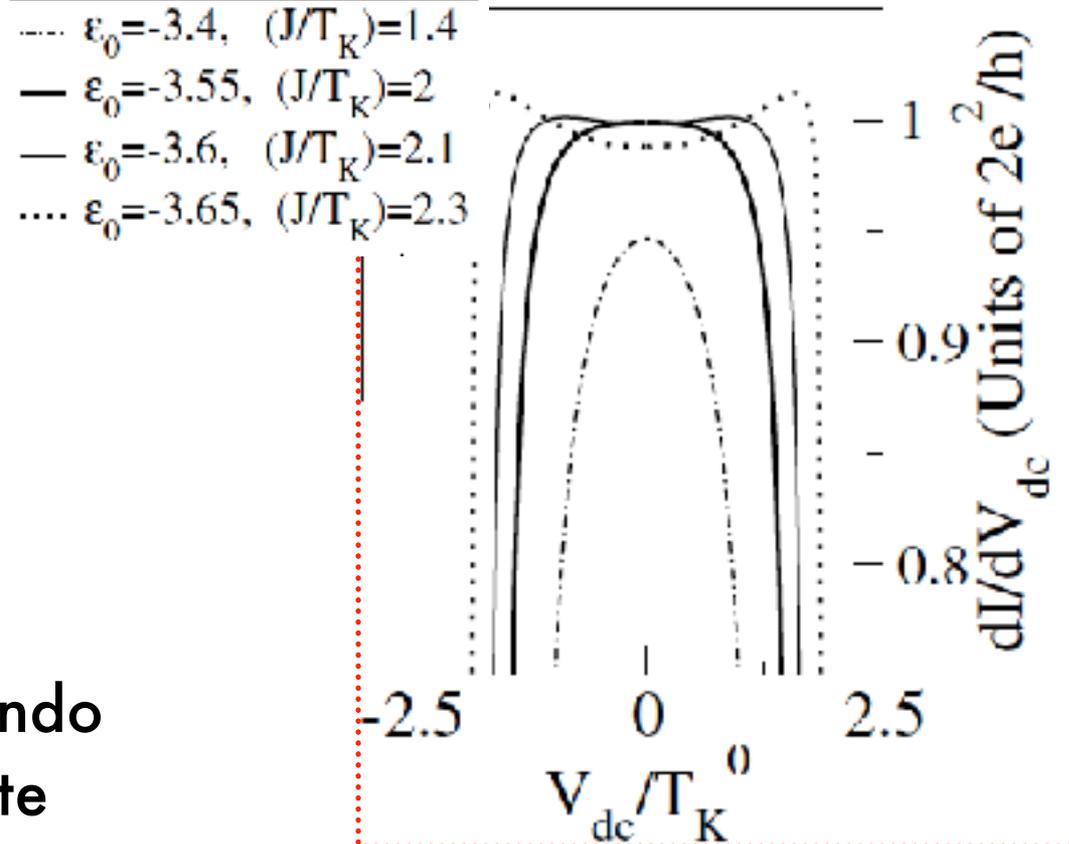
R. López et al., Physical review letters 89, 136802 (2003)

Electrically driven

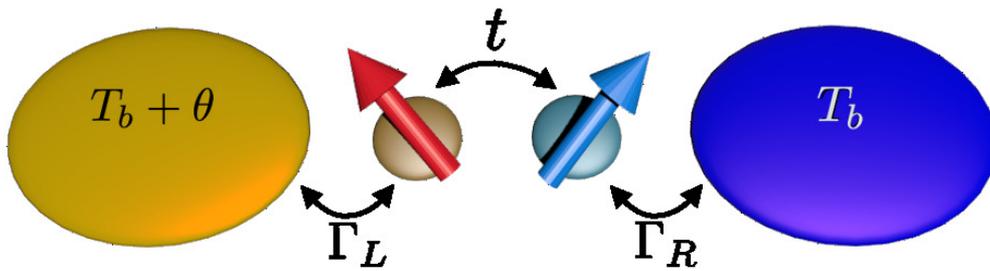
$$\mathcal{H}_J = J \hat{S}_L \cdot \hat{S}_R$$



Smooth transition from the Kondo state to the AF spin singlet state around  $(J/T_K) = 2.5$



Serial QDs: Crossover from the Kondo phase to the antiferromagnetic phase



We consider the slave-boson  $U \rightarrow \infty$  Anderson Hamiltonian

### Lead Hamiltonian

$$\mathcal{H}_C = \sum_{\alpha k \sigma} \varepsilon_{\alpha k} C_{\alpha k \sigma}^\dagger C_{\alpha k \sigma}$$

### Quantum dots Hamiltonian

$$\mathcal{H}_d = \sum_{\alpha \sigma} \varepsilon_{\alpha} f_{\alpha \sigma}^\dagger f_{\alpha \sigma} + \sum_{\sigma} t (f_{L \sigma}^\dagger f_{R \sigma} + \text{H.c.})$$

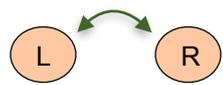
### Tunneling Hamiltonian

$$\mathcal{H}_t = \sum_{\alpha k \sigma} \mathcal{V}_{\alpha k \sigma} C_{\alpha k \sigma}^\dagger b_{\alpha}^\dagger f_{\alpha \sigma} + \text{H.c.}$$

### Lagrange Hamiltonian

$$\mathcal{H}_l = \sum_{\alpha} \lambda_{\alpha} \left( b_{\alpha}^\dagger b_{\alpha} + \sum_{\sigma} f_{\alpha \sigma}^\dagger f_{\alpha \sigma} - 1 \right)$$

$\alpha = \{L, R\}$  Dots and leads  
 $\equiv \equiv \equiv \}$   $\varepsilon_{\alpha}$  Dot Energy level



$t$  Dot-dot Tunnel amplitude

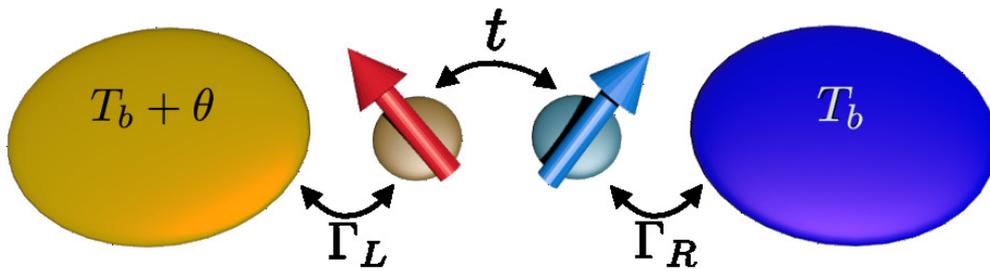


$\mathcal{V}_{\alpha k}$  Lead-dot Tunnel amplitude

$\lambda_{\alpha}$  Lagrange Multiplier

$b_{\alpha}^\dagger$  Creates a vacuum state

Valid in the **Fermi Liquid Regime**



We assume the *slave-boson mean-field theory*  
 $b_\alpha \rightarrow \tilde{b}_\alpha \equiv \langle b_\alpha \rangle$

Two conditions need to be satisfied

**Boson equation of motion**  
 $\partial_t b_\alpha = 0$

**Lagrange condition**  
 $b_\alpha^\dagger b_\alpha + \sum_\sigma f_{\alpha\sigma}^\dagger f_{\alpha\sigma} = 1$

$$\sum_\sigma G_{f_{\alpha\sigma}, f_{\alpha\sigma}}^<(t, t) = \frac{i}{\hbar} (1 - N|\tilde{b}_\alpha|^2)$$

$$\sum_{k\sigma} [g_{\alpha k}^r]^{-1} G_{f_{\alpha\sigma}, f_{\alpha\sigma}}^<(t, t) = -\frac{i}{\hbar} \lambda_\alpha N|\tilde{b}_\alpha|^2$$

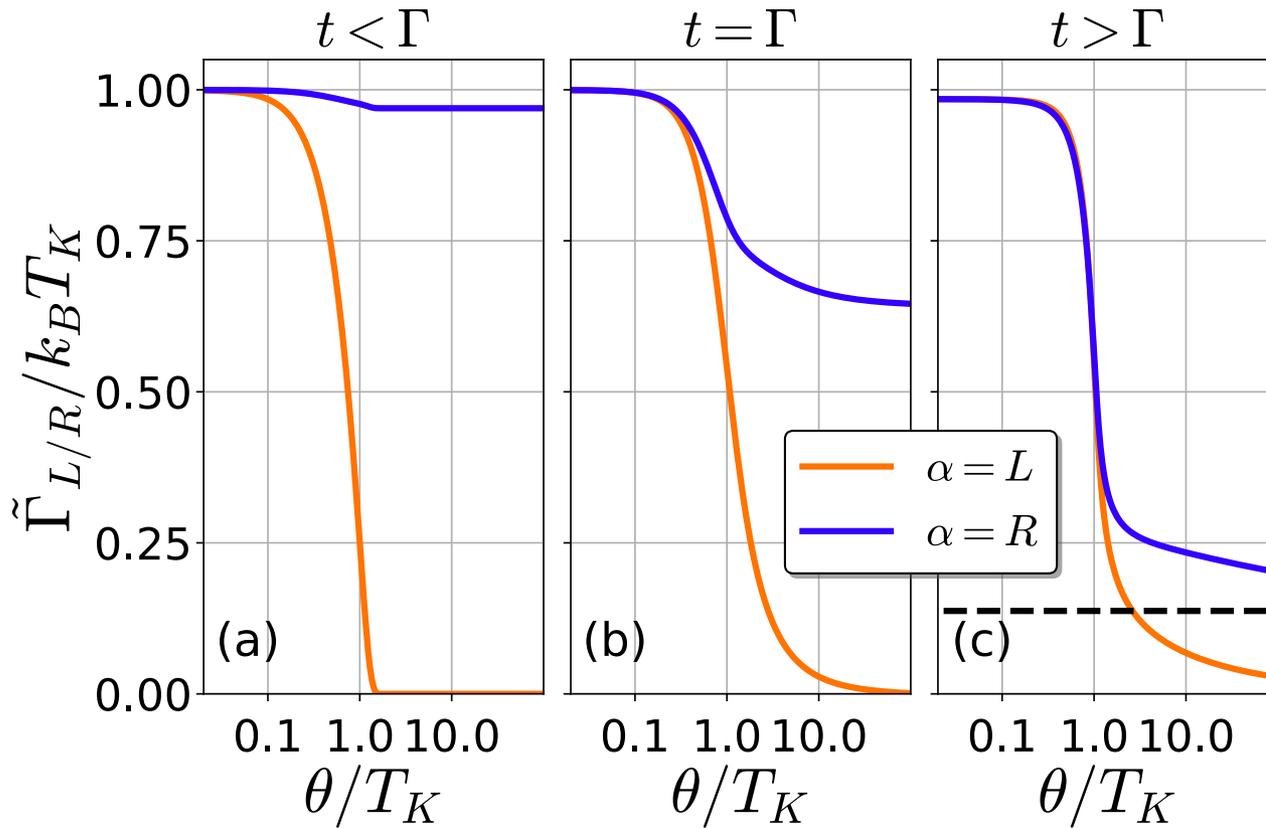
$G_{f_{\alpha\sigma}, f_{\alpha\sigma}}^<(t, t)$   
 Lesser Green's function  
 $N$   
 Spin degeneracy

Two renormalized parameters

$$\tilde{\Gamma}_\alpha = \Gamma_\alpha |\tilde{b}_\alpha|^2 \quad \text{Kondo resonance width}$$

$$\Gamma_\alpha = \pi \rho_\alpha |\mathcal{V}_\alpha|^2$$

$$\tilde{\epsilon}_\alpha = \epsilon_\alpha + \lambda_\alpha \quad \text{Kondo resonance position}$$



$$T_{K\alpha} \equiv \tilde{\Gamma}_\alpha(\theta)$$

Renormalized width determines the role of the Kondo effect

**Three different regimes**

depending on

$t$  and  $\Gamma = \Gamma_L + \Gamma_R$

**Weakly coupling**

$$t < \Gamma$$

$\tilde{\Gamma}_L$  Rapid decrease

$\tilde{\Gamma}_R$  Almost unaffected

**Intermediate**

$$t \approx \Gamma$$

Both  $\tilde{\Gamma}_\alpha$  affected

**Strong coupling**

$$t > \Gamma$$

$\tilde{\Gamma}_\alpha$  behave similarly

Bonding and anti-bonding states

$$Q = \frac{1}{h} \int d\omega [f_L(\omega) - f_R(\omega)] (\omega - \mu_\alpha) \mathcal{T}(\omega)$$

Transmission function

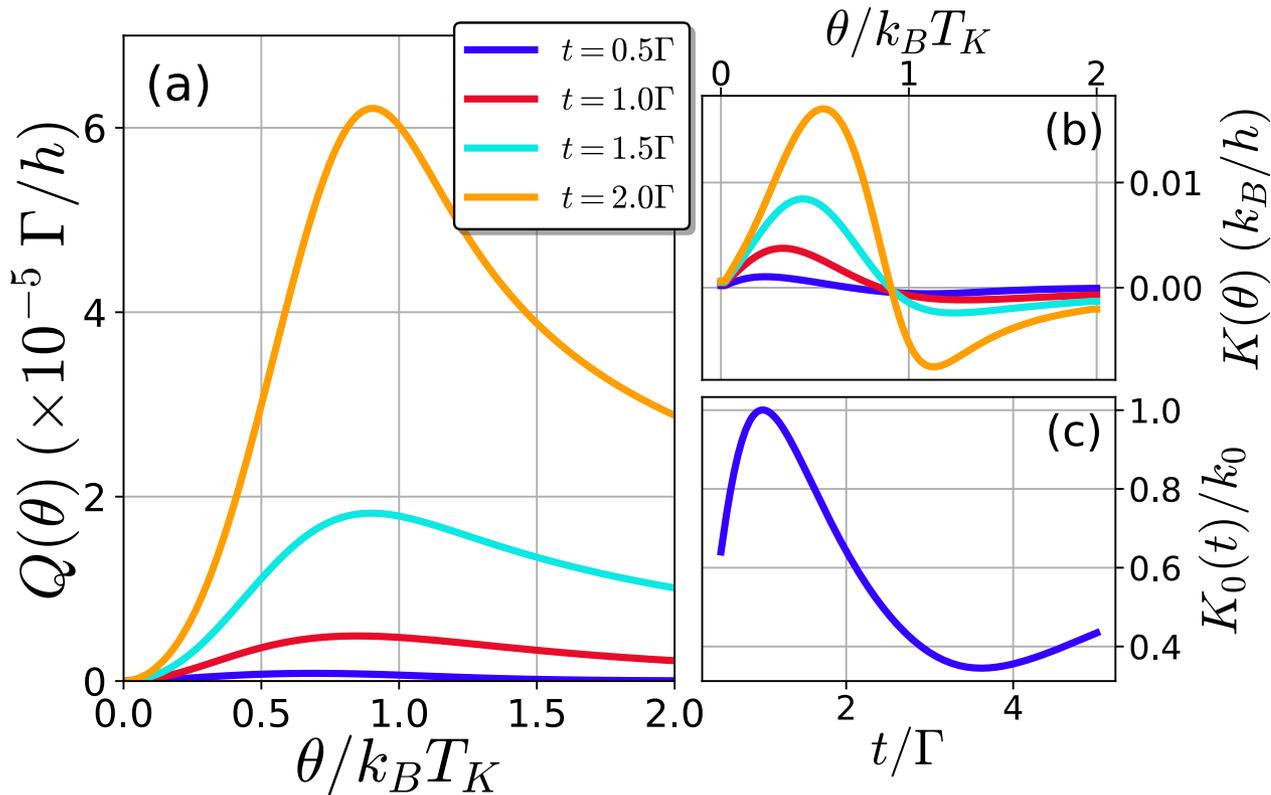
$$\mathcal{T}(\omega) = 4 \sum_{\sigma} \tilde{\Gamma}_L \tilde{\Gamma}_R |G_{fL\sigma, fR\sigma}^r|^2$$

Fermi function

$$f_{\alpha}(\omega) = \frac{1}{1 + \exp \frac{\omega - \mu_{\alpha}}{k_B T_{\alpha}}}$$

$$\mu_{\alpha} = \varepsilon_F$$

$$\theta = T_L - T_R$$



- Heat current more intense in the **strong coupling regime**
  - It presents a maximum and vanishes at large  $\theta$ .
  - Thermal diode** behavior.
  - It obeys Fourier's law.
- $$K_0 \approx \mathcal{T}(\varepsilon_F)$$
- $$Q \approx K_0 \theta$$

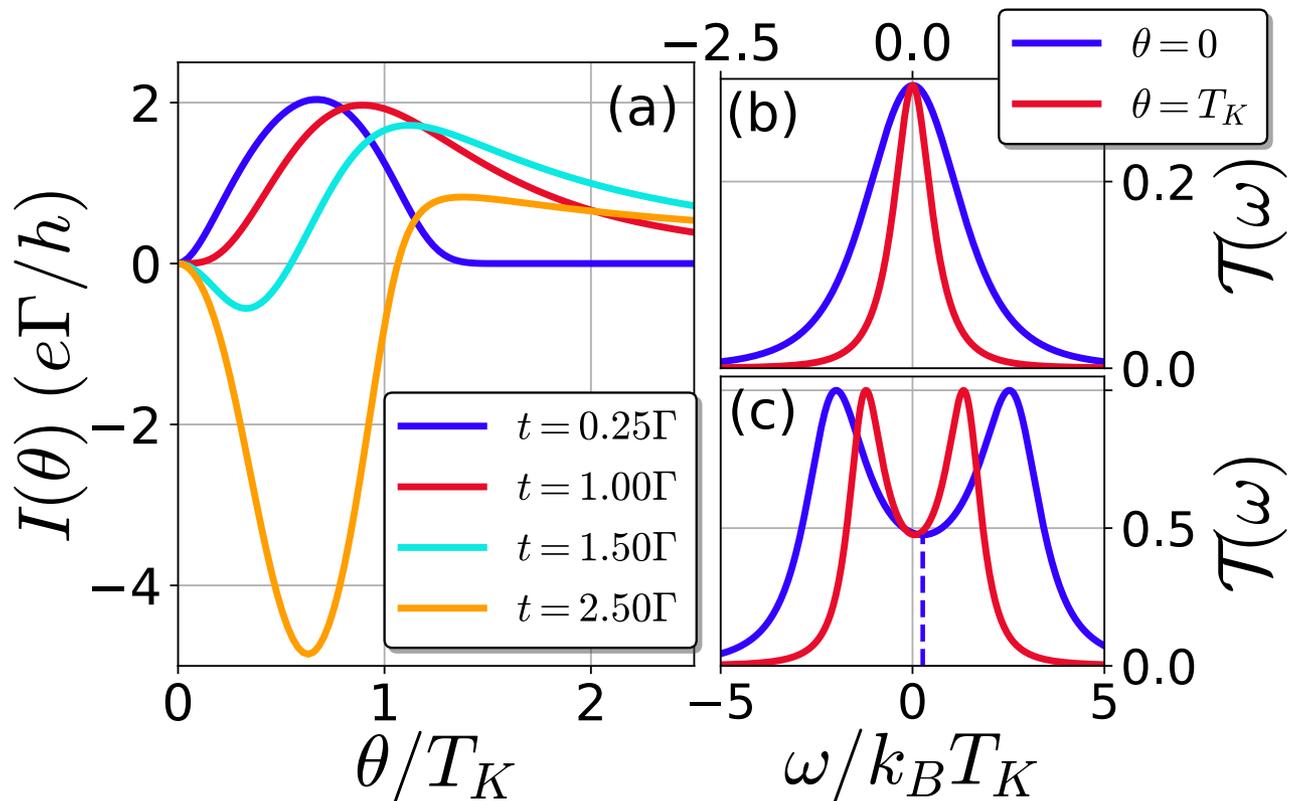
$$I = \frac{e}{h} \int d\omega [f_L(\omega) - f_R(\omega)] \mathcal{T}(\omega)$$

Transmission function

$$\mathcal{T}(\omega) = 4 \sum_{\sigma} \tilde{\Gamma}_L \tilde{\Gamma}_R |G_{fL\sigma, fR\sigma}^r|^2$$

$$\mu_{\alpha} = \varepsilon_F$$

$$\theta = T_L - T_R$$

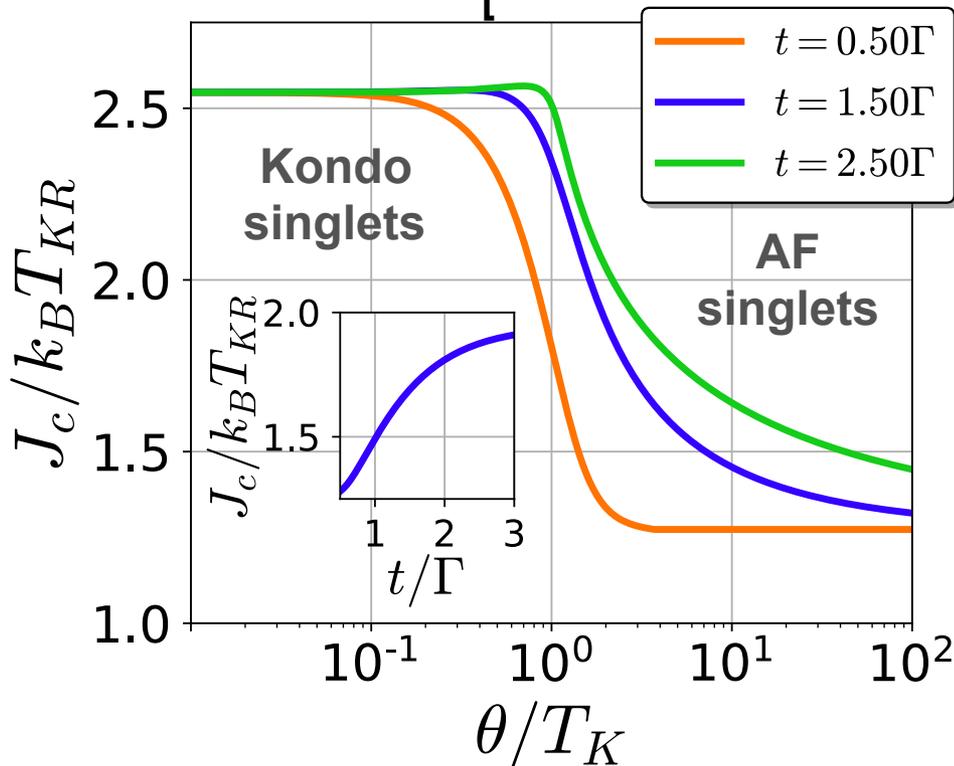


- Nonlinear behavior of the current
- Thermoelectric response depends highly on the ratio  $t/\Gamma$
- Nontrivial zeros due to asymmetric transmissions
- Vanishing current at large thermal biases

Case of relevant antiferromagnetic interactions.  $\mathcal{H}_J = J\hat{S}_L \cdot \hat{S}_R$

A crossover can occur between the Kondo singlet and the dot-dot AF singlet.

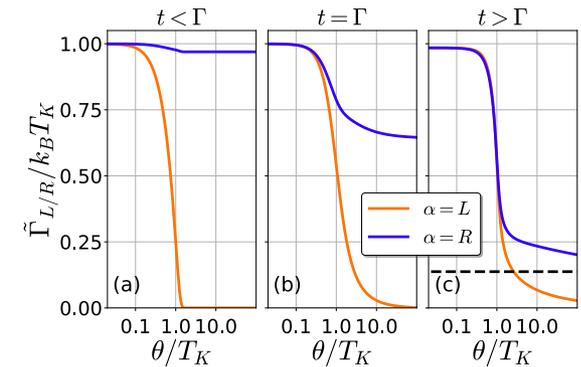
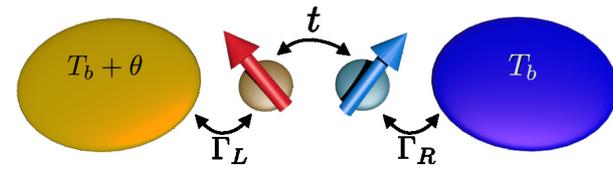
Critical dependent on Kondo Temperatures  $\rightarrow \frac{J_c}{k_B T_{KR}} = \frac{4}{\pi} \left( 1 + \frac{T_{KL}}{T_{KR}} \right)$



- A transition of the critical value occurs:  
 $J_c = \frac{8}{\pi} k_B T_{KR} \rightarrow J_c = \frac{4}{\pi} k_B T_{KR}$
- Kondo correlations are reinforced at large tunneling amplitudes.

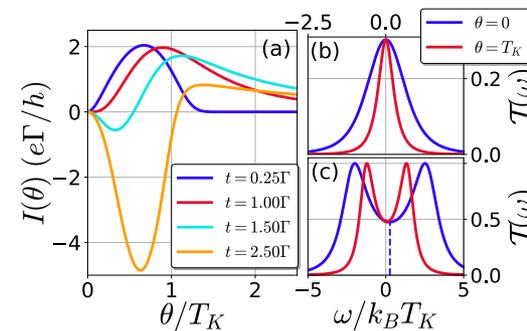
## System

- ➔ A serially-coupled double quantum dot is considered in the Kondo regime.
- ➔ Depending on  $t/\Gamma$ , one can identify different regimes:
  - ▶ Weakly coupled regime
  - ▶ Intermediate coupled regime



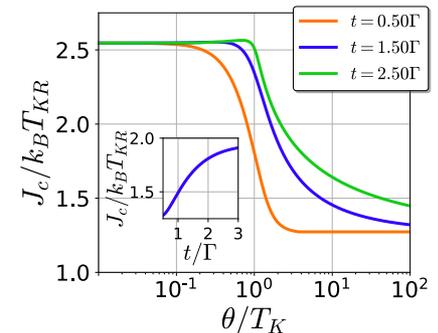
## Transport

- ➔ Heat current
  - ▶ Vanishing at large  $\theta$ .
  - ▶ Thermal diode behavior.
- ➔ Thermocurrent
  - ▶ Nonlinear behavior.



## Critical value

- ➔ Tunneling modifies the critical value of the crossover.





# THANK YOU

for your attention