Kondo effect in mesoscopic and nanoscopic systems

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Recap Lectures 1 & 2

• Standard spin ½ Kondo model
  - Complicated quantum impurity problem
  - Needed the developments of new numeric tools to solve it
  - The low temperature strong coupling fixed point is a Fermi liquid
  - There a single energy scale in the Kondo regime: universal functions of $k_B T_K$
Outline of lecture 3

- Mesoscopic and nanoscopic devices
- Transport through quantum dots and molecular transistors
  - Meir and Wingreen conductance formula
- Molecular transistors.
  - Kondo effect in molecular transistors
  - The role of molecular vibrations
Moore’s law (1965)

The number of transistors in an processor doubles every two years while the price remains constant.
Moore’s law in aeronautics?

- A flight between New York and Paris was worth in 1975 $900 and lasted 9 hours.
- It should cost now 1 penny and last less than a second.
Why molecular transistors?

- Moore’s law is expected to breakdown by 2020. (gate size ~6nm, tunneling, heating)
- An alternative to semiconductor based transistors may be needed.
- **Molecular based devices** offer the possibility of creating transistors with an area $\sim 10^5$ times smaller than current technology
Quantum dots

- The starting point is a two dimensional electron gas generated in a GaAs/Al$_x$Ga$_{1-x}$As heterostructure.
- The gas is protected by an insulated layer and metallic gates can be deposited on the surface and negatively charged to repel the electrons below.
- The electrons are confined in small regions forming a quantum dot
- Charging energies $U \sim 1K - 10K$
- Kondo temperatures $T_K < 1K$
Molecular transistors

- Metal-metal junction:
  - Electromigration
  - Break junctions
- Add molecules at the junction to get one bridging the gap
  - Lack of reproducibility
- Charging energy \( U \sim 1000K - 10000K \)
- Kondo temperatures \( T_K < 200K \)
Atoms or molecules on surfaces

- Studied using Scanning tunneling microscopy (STM)
- Spectroscopy: access to spectral properties of the atom
- STM tip couples to bulk electrons: Fano lineshapes
- Lack of tunability
- Surface states that can be modified using quantum corrals
- Kondo effect given mostly by bulk states.
Transport through an interacting region

\[ H = \sum_{\ell=L,R} \sum_{k\alpha} \epsilon_{\ell k} c_{\ell k\alpha}^\dagger c_{\ell k\alpha} + H_{\text{int}}(\{d_n\}; \{d_n^\dagger\}) \]

\[ + \sum_{\ell=L,R} \sum_{k\alpha} \sum_n (V_{n\ell k\alpha} c_{\ell k\alpha}^\dagger d_n + V_{n\ell k\alpha}^* d_n^\dagger c_{\ell k\alpha}) \]

Transport through an interacting region

Meir and Wingreen showed that

\[ J = \frac{ie}{2\hbar} \int dE \left( \text{Tr}[ (f_L(E)\Gamma^L - f_R(E)\Gamma^R)(G^r - G^a) + (\Gamma^L - \Gamma^R)G^< ] \right) \]

where:

\[ \Gamma^L_{n,m} = 2\pi \sum_\alpha \rho_L(E)V_{nL\alpha}(E)V^*_{mL\alpha}(E) \]

\[ G^<_{n,\ell k\alpha}(t) = i\langle\langle c_{\ell k\alpha}, d_n(t) \rangle\rangle \]


\[ eV = \mu_R - \mu_L \]
Transport through an interacting region

- **Simplifications:**
  - In the high temperature regime \( k_B T \gg \Gamma_L, \Gamma_R \)
    - No Kondo physics
  - **Proportionate couplings:** \( \Gamma_L = \lambda \Gamma_R \)
    - No need to calculate: \( G_{n,\ell k \alpha}^< \)
    - But \( G^r(E, V) \)
  - **Zero bias:** \( V \rightarrow 0 \)
    - \( G^r(E, 0) \) but \( G_{n,\ell k \alpha}^<(E, V \rightarrow 0) \)
  - **Asymmetric couplings:** \( \Gamma_L \ll \Gamma_R \)
    - Tunneling situation: \( G^r(E, 0) \)
Single level quantum dot

\[ H = \sum_{\sigma} \epsilon_d n_{\sigma} + U n_{d\downarrow} n_{d\uparrow} + \sum_{\ell k} \epsilon_{\ell k} c_{\ell k\sigma}^\dagger c_{\ell k\sigma} + \sum_{\ell k} (V_{\ell k} c_{\ell k\sigma}^\dagger d_{\sigma} + V^* d_{\sigma}^\dagger c_{\ell k\sigma}) \]

\[ \Gamma_L = \lambda \Gamma_R, \quad V \to 0 \]

\[ g_\sigma = \frac{4\pi e^2}{h} \int dE \frac{\Gamma_L \Gamma_R}{\Gamma_L + \Gamma_R} A_{d\sigma}(E, T, V = 0) \left( -\frac{\partial f}{\partial E} \right) \]

The Anderson impurity is coupled to a single effective electron bath with an hybridization \( \Gamma = \Gamma_L + \Gamma_R \)

The d-level couples to the symmetric combination \( V_L f_{L0\sigma} + V_R f_{R0\sigma} \)
Zero temperature conductance

\[ g_\sigma(T = 0) = \frac{4\pi e^2}{h} \frac{\Gamma^L \Gamma^R}{\Gamma^L + \Gamma^R} A_{d\sigma}(E = 0, T = 0, V = 0) \]

Non-interacting system:

\[ A_{d\sigma}(E = 0, T = 0, V = 0) = \frac{\Gamma/\pi}{\epsilon_d^2 + \Gamma^2} \]

\[ \Gamma = \Gamma^L + \Gamma^R \]

\[ g_\sigma(T = 0) = \frac{4e^2}{h} \frac{\Gamma^L \Gamma^R}{\epsilon_d^2 + (\Gamma^L + \Gamma^R)^2} \]

\[ g_\sigma(T = 0) = \frac{4e^2}{h} \frac{\Gamma^L \Gamma^R}{(\Gamma^L + \Gamma^R)^2} \frac{1}{1 + \frac{\epsilon_d^2}{(\Gamma^L + \Gamma^R)^2}} \]

\[ g_\sigma(T = 0) = \frac{4e^2}{h} \frac{\Gamma^L \Gamma^R}{(\Gamma^L + \Gamma^R)^2} \sin^2(\pi n_{d\sigma}) \]
Zero temperature conductance

For $\Gamma^L = \Gamma^R$ and $\epsilon_d = 0$

$$g_{\sigma}(T = 0) = \frac{e^2}{h} \quad \text{(unitary limit)}$$

The total conductance as a function of the gate voltage has a Lorentzian shape.

For an interacting level, the occupation of the level has a two step behavior as predicted by the Hartree-Fock solution and we expect the conductance to have a plateau of height $2 \, e^2/h$ in the magnetic moment regime.

$$g_{\sigma}(T = 0) = \frac{4e^2}{h} \frac{\Gamma^L \Gamma^R}{(\Gamma^L + \Gamma^R)^2} \sin^2(\pi n_{d\sigma})$$
High temperature regime

$k_B T \gg \Gamma$

We can treat the hybridization as a perturbation. To lowest order
We use the spectral density of the isolated quantum dot (atomic limit)

$$A_{d\sigma}(E) = \frac{1}{Z} \sum_{i,j} (e^{-\beta E_i} + e^{-\beta E_j}) \langle \Psi_j | d_{\sigma}^\dagger | \Psi_i \rangle \langle \Psi_i | d_{\sigma} | \Psi_j \rangle \delta[\epsilon - (E_j - E_i)]$$

$$g = \frac{e^2}{\hbar} \frac{\Gamma}{k_B T} \sum_{i,j,\sigma} (P_i + P_j) f(E_i - E_j) f(E_j - E_i) |\langle \Psi_j | d_{\sigma}^\dagger | \Psi_i \rangle|^2$$

$$P_i = e^{-\beta E_i} / Z$$

We expect two peaks of width $k_B T$ as a function of the gate voltage at $\epsilon_d = 0, -U$

$\epsilon_d \propto -V_g$

At the charge degeneracy points.
Goldhaber-Gordon et al
Nature 2000

QD

$V_g$

$n_d$

$V_g$

$g$

$V_g$

Temperature dependence of linear conductance

(a) 90 mK

(b) 400 mK

(c) 800 mK

$G (e^2/h)$

$V_g$ (mV)

$V_g$ (mV)
Kondo effect in quantum dots

![Image of Kondo effect in quantum dots](image-url)
Scanning tunneling microscopy

\[ \frac{dI}{dV} \propto A_{STM}(V) \]

\[ t_c \Psi_\sigma^\dagger + t_d d_\sigma^\dagger \]

Access to the spectral density

Fano lineshapes

\[ q = \frac{t_d}{t_c} \]
A molecule as a building block

- Large level quantization.
- Large charging energies.

\[ \begin{align*}
U + \epsilon_d & \quad \downarrow \quad \epsilon_d \\
H_d & = \epsilon_d ( n_\uparrow + n_\downarrow ) + U n_\uparrow n_\downarrow
\end{align*} \]

Anderson model!
Kondo effect in molecules

Molecular vibrations

Outline

- Molecular vibrations
- Anderson-Holstein
- Negative U Kondo effect
- Franck Condon effect
- Franck Condon blockade and Kondo effect

Kondo effect in C60 molecular transistors:

Phononic effects in Suspended Quantum dots:
A molecule as a building block

- Large level quantization.
- Large charging energies.
- Electron vibron interaction: coupling to a mode with coordinate $x$ and frequency $\omega_0$

$$H_d = \epsilon_d(x)(n_{\uparrow} + n_{\downarrow}) + U n_{\uparrow} n_{\downarrow} + H_{phon}$$

$$\epsilon_d(x) \sim \epsilon_d + c_1(x - x_0) + \cdots$$
Quantum Harmonic Oscillator

\[ H_{vib} = \frac{\hat{p}^2}{2m} + \frac{1}{2}m\omega_0^2\hat{x}^2 \]

\[ a = \sqrt{\frac{m\omega_0}{2}} \left( \hat{x} + \frac{i}{m\omega_0}\hat{p} \right), \quad a^\dagger = \sqrt{\frac{m\omega_0}{2}} \left( \hat{x} - \frac{i}{m\omega_0}\hat{p} \right) \]

\[ \hat{x} = \sqrt{\frac{1}{2m\omega_0}}(a + a^\dagger), \quad \hat{p} = i\sqrt{\frac{m\omega_0}{2}}(a^\dagger - a) \]

\[ H_{vib} = \omega_0 \left( a^\dagger a + \frac{1}{2} \right) \]
Isolated molecule

\[ H_d = \epsilon_d n_d + U n_\uparrow n_\downarrow - \lambda (a^\dagger + a)(n_d - 1) + \omega_0 \left( \frac{1}{2} + a^\dagger a \right) \]

\[ n_d = n_\uparrow + n_\downarrow \]

The Hamiltonian can be diagonalized on each charge sector using a phonon displacement operator

\[ \tilde{U} = e \frac{\lambda}{\omega_0} (n_d - 1)(a^\dagger - a) \]

\[ \tilde{U}^\dagger a \tilde{U} = a + \frac{\lambda}{\omega_0} (n_d - 1) \]

\[ \tilde{H}_d = \tilde{U}^\dagger H_d \tilde{U} \]

\[ \tilde{H}_d = \left( \epsilon_d + \frac{\lambda^2}{\omega_0} \right) n_d + \left( U - \frac{2\lambda^2}{\omega_0} \right) n_\uparrow n_\downarrow + \omega_0 \left( \frac{1}{2} + a^\dagger a \right) - \frac{\lambda^2}{\omega_0} \]
Isolated molecule

\[ |0, m\rangle = |0\rangle e^{\frac{\lambda}{\omega_0} (a^\dagger - a)} |m\rangle \]
\[ |\sigma, m\rangle = |\sigma\rangle |m\rangle \]
\[ |2, m\rangle = |\uparrow \downarrow\rangle e^{-\frac{\lambda}{\omega_0} (a^\dagger - a)} |m\rangle \]
\[ E_{0,m} = -\frac{\lambda^2}{\omega_0} + m\omega_0 \]
\[ E_{\sigma,m} = \epsilon_d + m\omega_0 \]
\[ E_{2,m} = 2\epsilon_d + U - \frac{\lambda^2}{\omega_0} + m\omega_0 \]

\[ \tilde{U} = e^{\frac{\lambda}{\omega_0} (n_d - 1)(a^\dagger - a)} \]
\[ \tilde{M}^+ = e^{\frac{\lambda}{\omega_0} (a^\dagger - a)} \]
\[ \tilde{M}^- = e^{-\frac{\lambda}{\omega_0} (a^\dagger - a)} \]

\[ \epsilon_d + U - \frac{\lambda^2}{\omega_0} \]
\[ U_{\text{eff}} = U - \frac{\lambda^2}{\omega_0} \]
Weak coupling to the leads $k_B T > \Gamma$

$$g = \frac{e^2}{\hbar} \frac{\Gamma}{k_B T} \sum_{\mu, \nu, m, n, \sigma} W(E_{\nu n}, E_{\mu m}, T) |\langle \Psi_{\nu n} | d^\dagger_{\sigma} | \Psi_{\mu m} \rangle|^2$$

$$W(E_{\nu n}, E_{\mu m}, T) = (P_{\mu m} + P_{\nu n}) f(E_{\mu m} - E_{\nu n}) f(E_{\nu n} - E_{\mu m})$$

$$P_{\mu m} = e^{-\beta E_{\mu, m}} / Z$$

$$|\langle \Psi_{\nu 0} | d^\dagger_{\sigma} | \Psi_{\mu 0} \rangle|^2 = |\langle \nu | d^\dagger_{\sigma} | \mu \rangle|^2 |\langle 0 | e^{\frac{\lambda}{\omega_0} (a^\dagger - a)} | 0 \rangle| = |\langle \nu | d^\dagger_{\sigma} | \mu \rangle|^2 e^{-\lambda^2 / \omega_0^2}$$

Exponential suppression of the tunneling due to the Franck-Condon effect: “Franck-Condon blockade”

Franck Condon effect

\[ |0, 0\rangle \rightarrow |1, n\rangle \]

\[
\gamma_{n,0}^2 = |\langle 0 | e^{\frac{\lambda}{\omega_0} (a^\dagger - a)} | n \rangle|^2
\]

\[
= e^{-\frac{\lambda^2}{\omega_0^2}} \frac{(\frac{\lambda}{\omega_0})^{2n}}{n!}
\]

J. Franck Trans. Farad. Soc. 21 536 (1926);
E. Condon Phys. Rev. 28 1182(1926)
NRG results

Zero-bias conductance vs. gate voltage at low (black) and high temperatures.

[PRl 2004]
Spectral density \((\epsilon_d = -U/2)\)

\[ A_d(E = 0) = \frac{1}{\pi \Delta} \]
Schrieffer Wolff transformation

$U_{\text{eff}} > 0$

\[ |\downarrow\rangle|0\rangle \xrightarrow{H_V} |\uparrow\downarrow\rangle e^{-\frac{\lambda}{\omega_0}(a^+ - a)} |m\rangle \xrightarrow{H_V} |0\rangle e^{\frac{\lambda}{\omega_0}(a^+ - a)} |m\rangle \xrightarrow{H_V} |\uparrow\rangle|0\rangle \]

Intermediate states

\[ E_{\downarrow} = \epsilon_d \]

\[ E_{2,m} = 2\epsilon_d + U - \frac{\lambda^2}{\omega_0} + m\omega_0 \]

\[ E_{0,m} = -\frac{\lambda^2}{\omega_0} + m\omega_0 \]

\[ E_{\uparrow} = \epsilon_d \]

Kondo Hamiltonian with modified couplings

\[ J_K = V^2 \sum_m \gamma_{0,m}^2 \left( \frac{1}{-\epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} + \frac{1}{U + \epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} \right) \]
Schrieffer Wolff transformation

$U_{\text{eff}} < 0$

$H_V$

$|\sigma\rangle |m\rangle$

$E_\sigma = \epsilon_d + m\omega_0$

$H_V$

$|\uparrow\downarrow\rangle e^{-\frac{\lambda}{\omega_0}(a^\dagger - a)} |0\rangle$

$|0\rangle e^{\frac{\lambda}{\omega_0}(a^\dagger - a)} |0\rangle$

intermediate states

$J_{K}^{\perp} = V^2 \sum_m \gamma_{0,m}^2 \left( \frac{(-1)^m}{-\epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} + \frac{(-1)^m}{U + \epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} \right)$

$J_{K}^{\parallel} = V^2 \sum_m \gamma_{0,m}^2 \left( \frac{1}{-\epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} + \frac{1}{U + \epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} \right)$

Anisotropic Kondo Hamiltonian for the pseudospin:

$|\uparrow\rangle = |0\rangle$, $|\downarrow\rangle = |\uparrow\downarrow\rangle$
Charge Kondo effect (anisotropic Kondo)

\[ \frac{dj_\perp}{d \ln D} = -2j_\parallel j_\perp \quad \frac{dj_\parallel}{d \ln D} = -2j_\perp^2 \]

\[ T_K \sim D \left( \frac{J_\perp}{J_\parallel} / J_K \right) e^{1/\rho_0 J_K} \]

\[ (J_\perp / J_K) \sim e^{-2(\lambda/\omega_0)^2} \]
Kondo temperature

\[ T_K = 12.2 e^{-\frac{1}{\rho_0 J_K}} \]

\[ T_K = 3.0 (J_\perp / J_\parallel)^{1/\rho_0 J_\parallel} \]
A small gate voltage destroys the charge Kondo effect but there is no peak splitting as in the spin-Kondo with a magnetic field.
Spin-Kondo effect and vibrations

\[ \Gamma \propto \Gamma_0 e^{-\lambda^2/\omega_0^2} \]

peaked at \[ m^* = (\lambda/\omega_0)^2 \]

\[ J_K = V^2 \sum_m \gamma_{0,m}^2 \left( \frac{1}{-\epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} + \frac{1}{U + \epsilon_d + m\omega_0 - \frac{\lambda^2}{\omega_0}} \right) \]

\[ \sim V^2 \left( \frac{1}{-\epsilon_d} + \frac{1}{U + \epsilon_d} \right) \equiv J_K(\lambda = 0) \]
Spin-Kondo effect and vibrations

\[ g = \left( \frac{\lambda}{\omega_0} \right)^2 \]

PSC, G. Usaj, and C.A. Balseiro, PRB R (2007)
Other electron phonon couplings

- Breathing modes
  - Effective hybridization
- Shuttle modes
  - New channel opening: no longer possible to map the left and right leads to a single electron bath.
- Stretching modes
  - Coupling to magnetic anisotropy can change the nature of ground state in magnetic molecules [PRB (2012)]
Coulomb blockade diamond edges

\[ g(V) \sim \frac{2e^2}{h} \pi \Gamma \left[ A_d(V/2) + A_d(-V/2) \right] \]
Coulomb blockade diamond edges
Conclusions

- The electron vibron interaction in molecular transistors leads to a rich variety of behavior:
  - New anisotropic charge Kondo effect
  - Anomalous gate-voltage dependence of the Kondo temperature.
  - Blurring of Coulomb-blockade diamond edges due to Franck-Condon effects.
  - Conduction channel opening (asymmetric coupling)

- Next lecture:
  - Exotic Kondo effects in double quantum dots and magnetic molecules.
Breathing mode
Breathing mode