Numerical renormalization group approach for simulating quantum nano-devices

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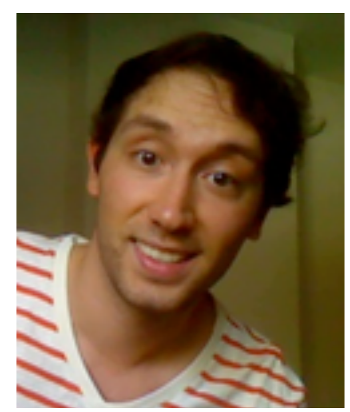

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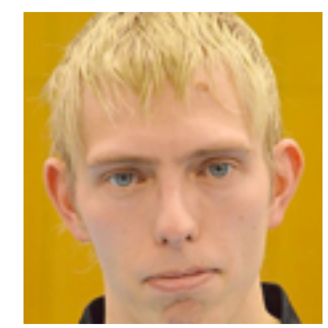

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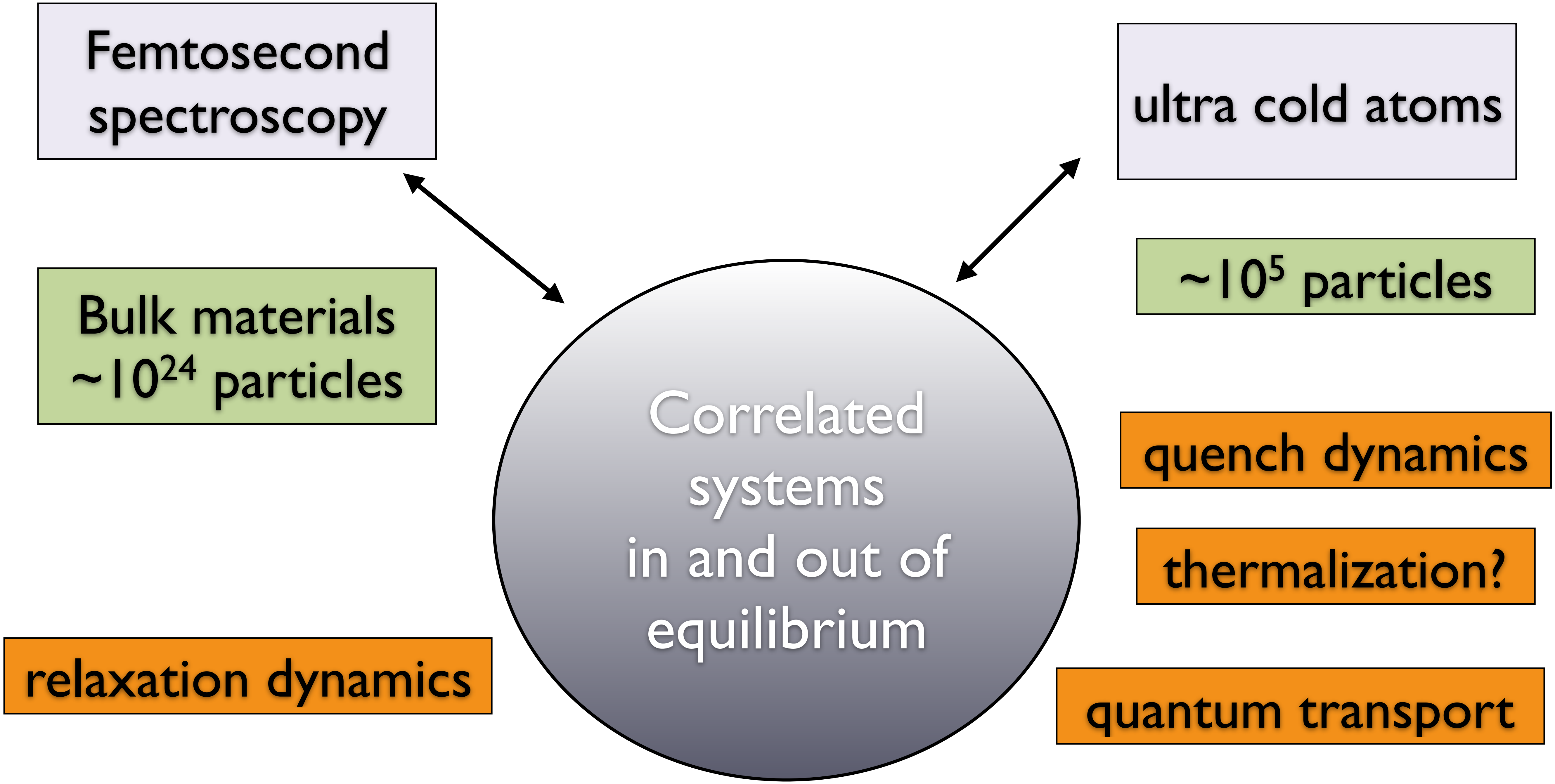
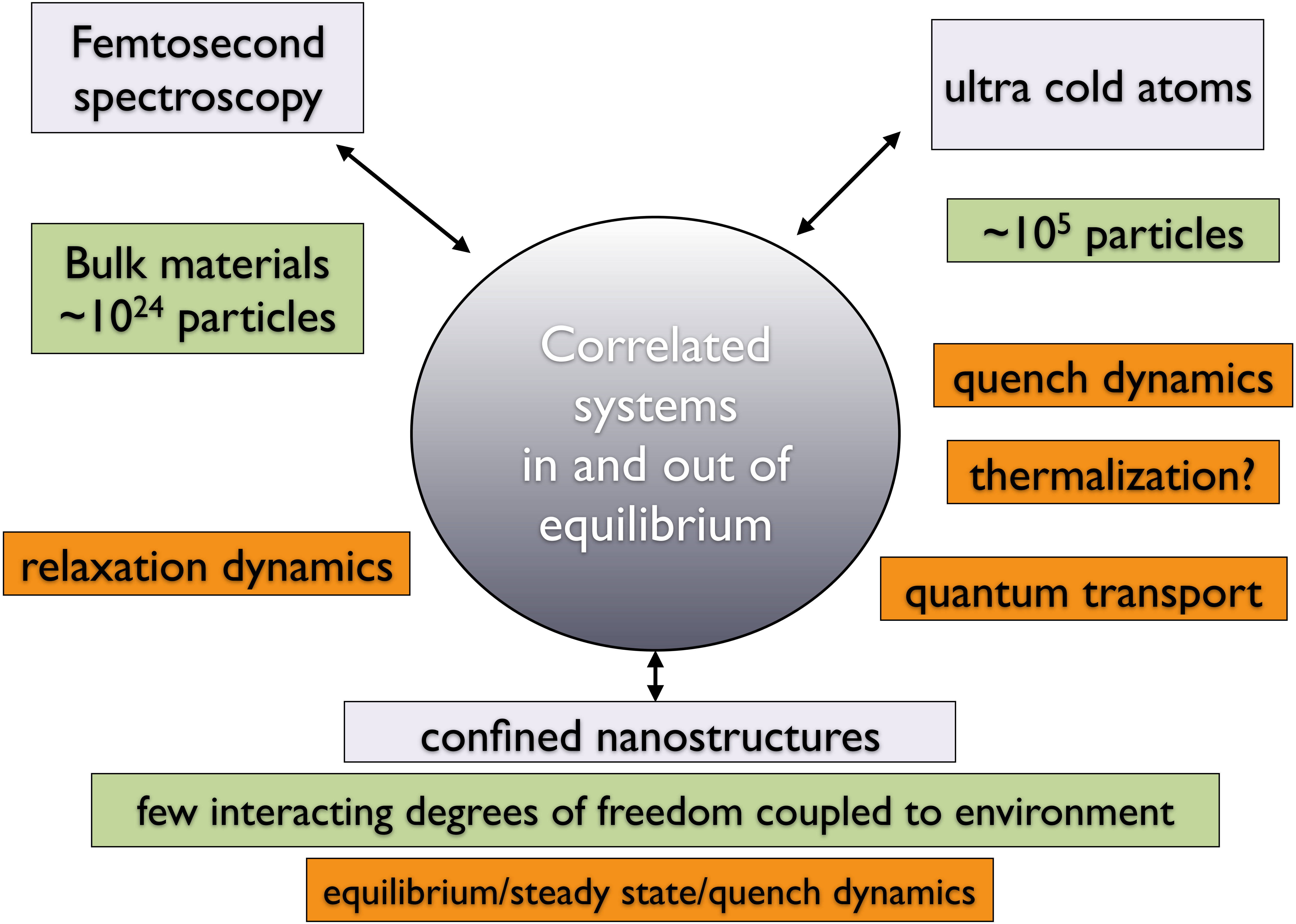
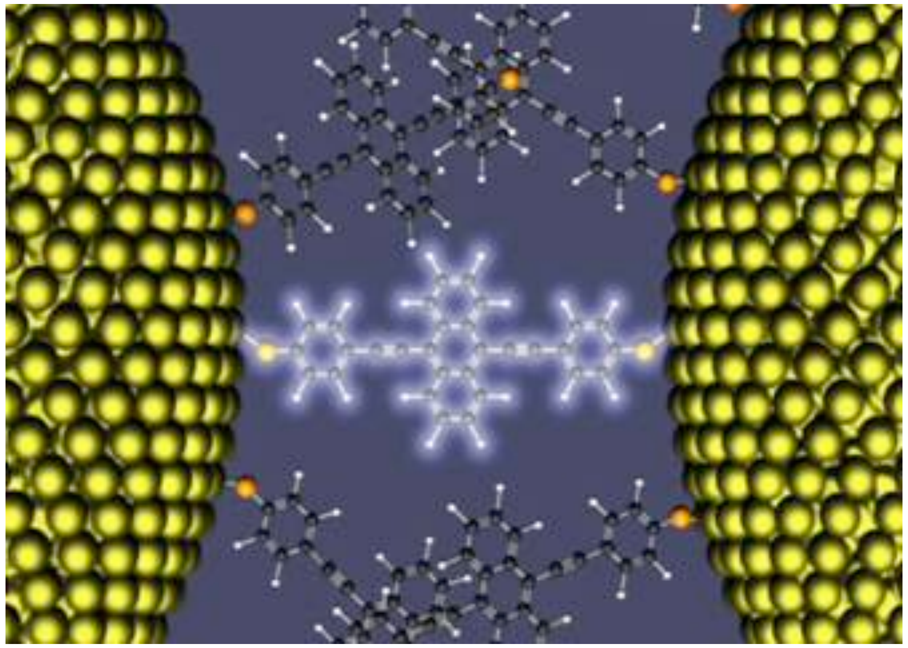
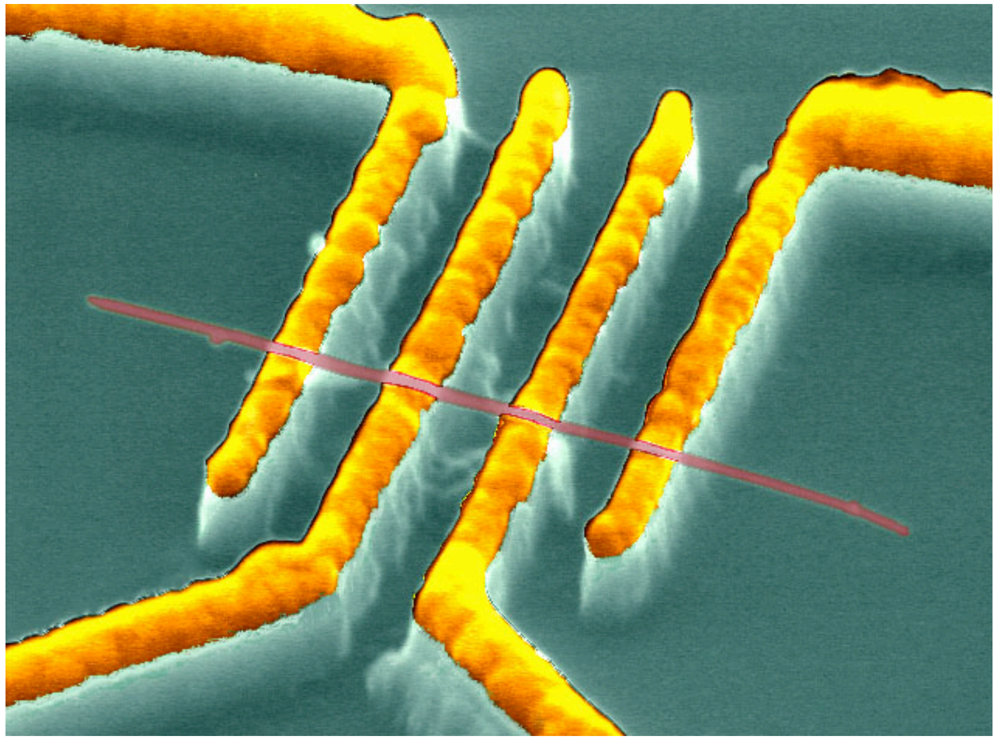

Femtosecond spectroscopy
ultra cold atoms
Bulk materials
~10²⁴ particles
~10⁵ particles
Correlated systems in and out of equilibrium
quench dynamics
thermalization?
relaxation dynamics
quantum transport


Femtosecond spectroscopy
ultra cold atoms
Bulk materials
~10^{24} particles
~10^{5} particles
Correlated systems in and out of equilibrium
quench dynamics
thermalization?
relaxation dynamics
quantum transport
confined nanostructures
few interacting degrees of freedom coupled to environment
equilibrium/steady state/quench dynamics



source: Forschungszentrum Karlsruhe



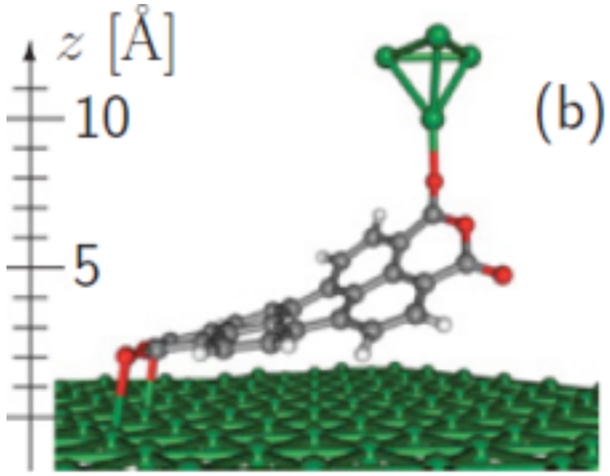
NCCR Nanoscale Science
Institute of Physics, University of Basel

Electron confinement:
strong Coulomb interaction

challenges:

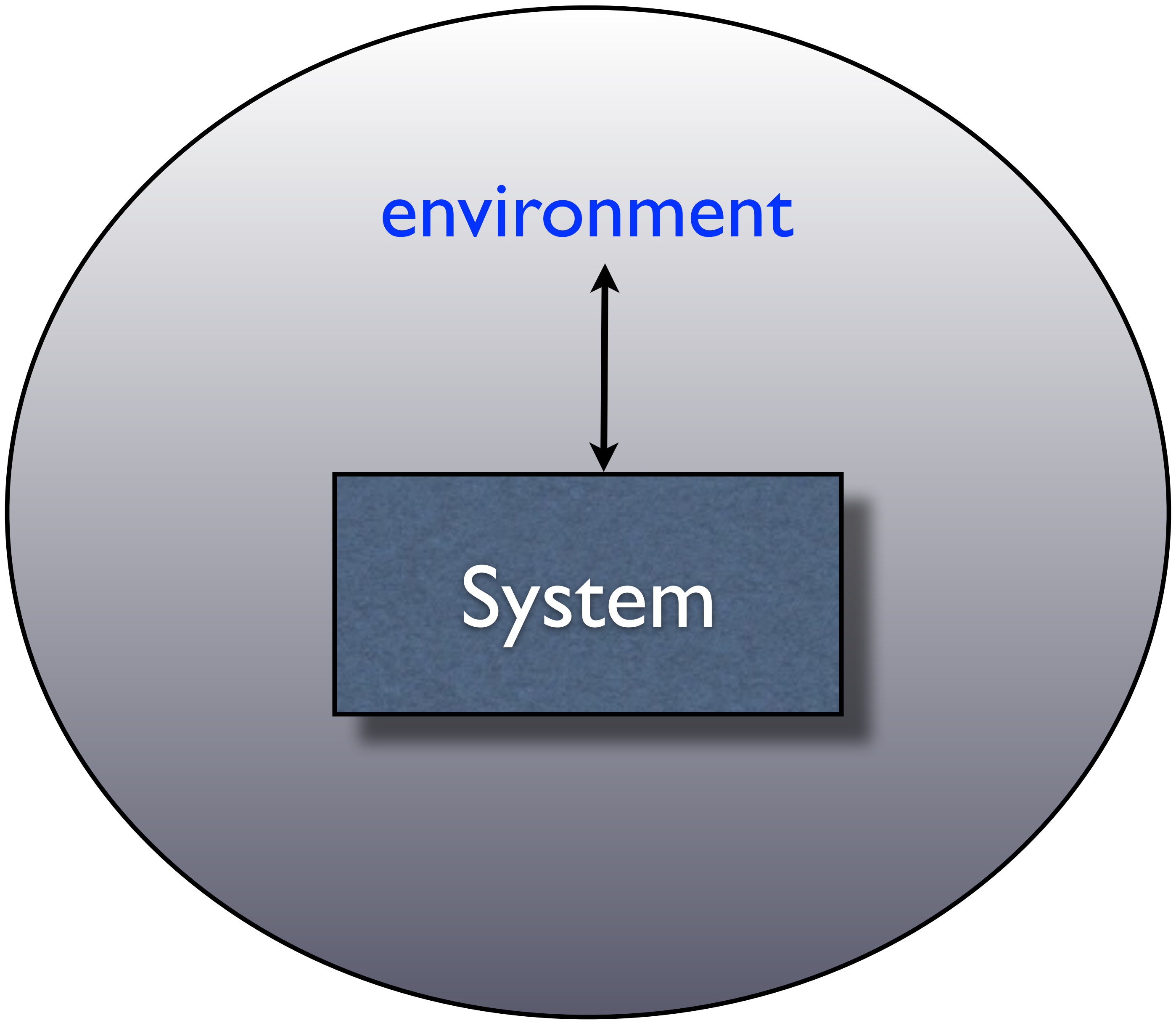
- controllable, reproducible molecular connections
- induced local moments for spin manipulations
- **modeling correlated systems in and out of equilibrium**

STM Au/PTCDA



see als Fabian Pauly talk

Goal



simulation of

- real-time dynamics in
- quantum transport

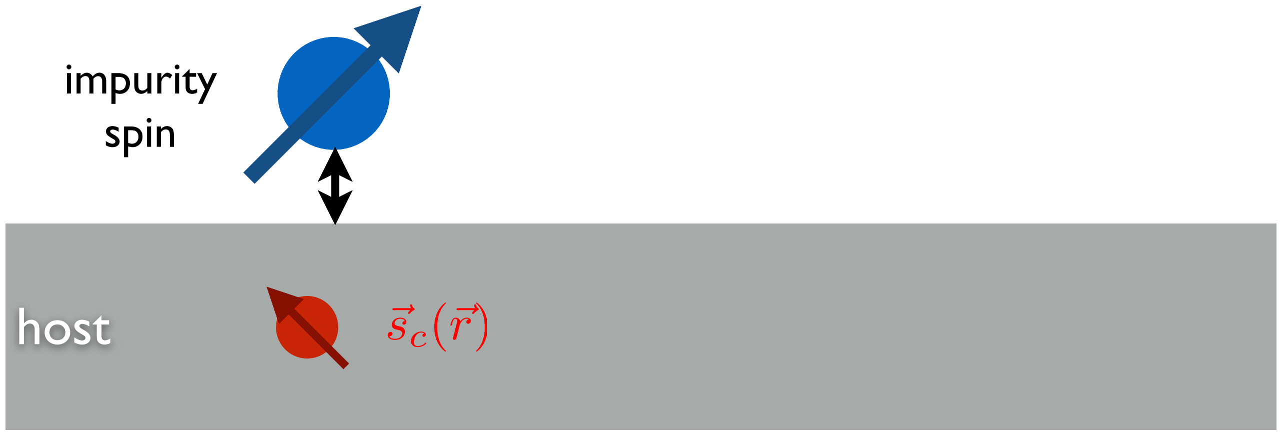
though

small quantum systems

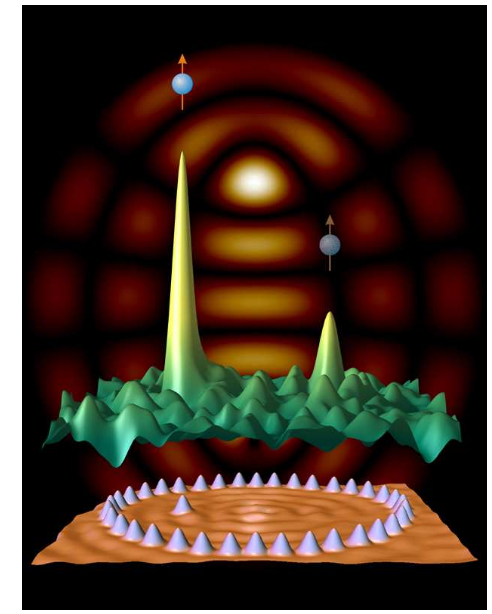
problem:

- entanglement with the environment
- change of ground state

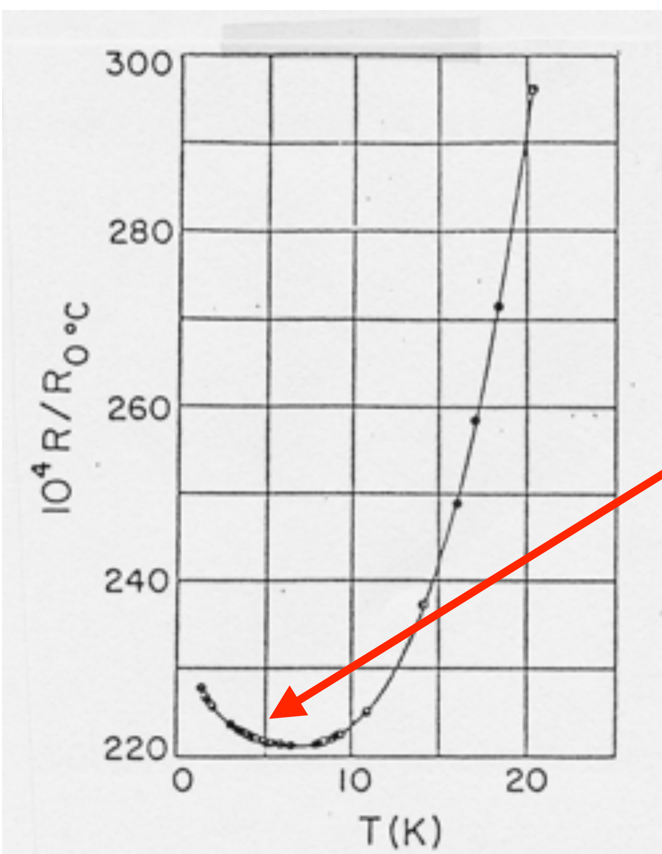
Kondo model: drosophila of solid state theory



Quantum-mirage: Co on Cu



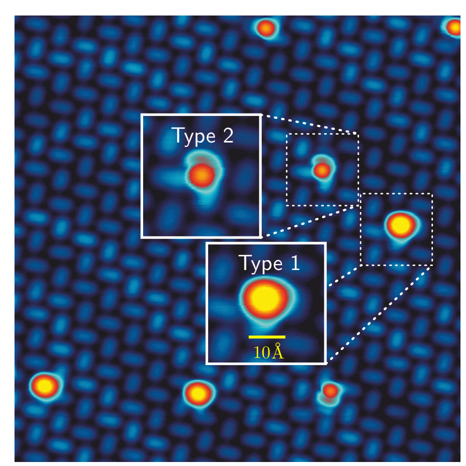
Manoharan et al,
Nature 403, 512 (2000)



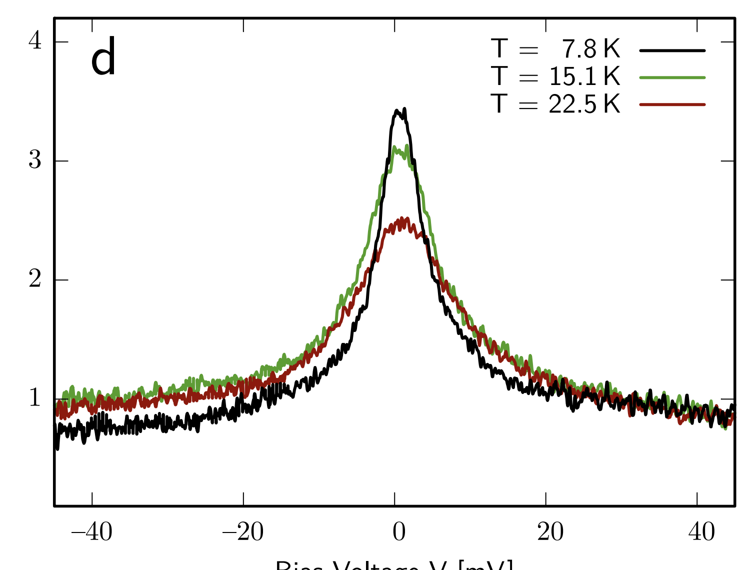
Kondo scale T_K

$$T_K = De^{-\frac{1}{\rho J}}$$

de Haas, de Boer, van den Berg
Physica 1,1115 (1934)

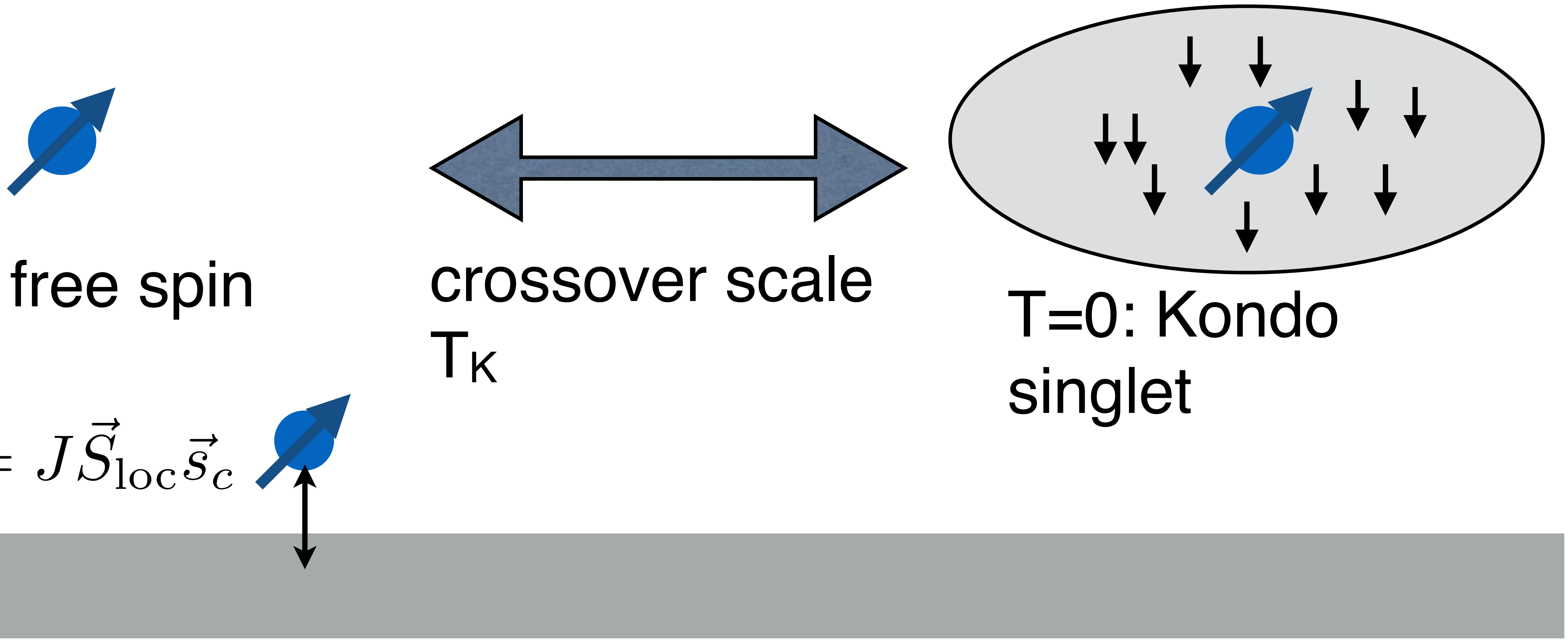


Au/PTCDA on Au(111)



Esat et al, PRB 2015

Kondo model: drosophila of solid state theory



T=∞: free spin

crossover scale T_K

T=0: Kondo singlet

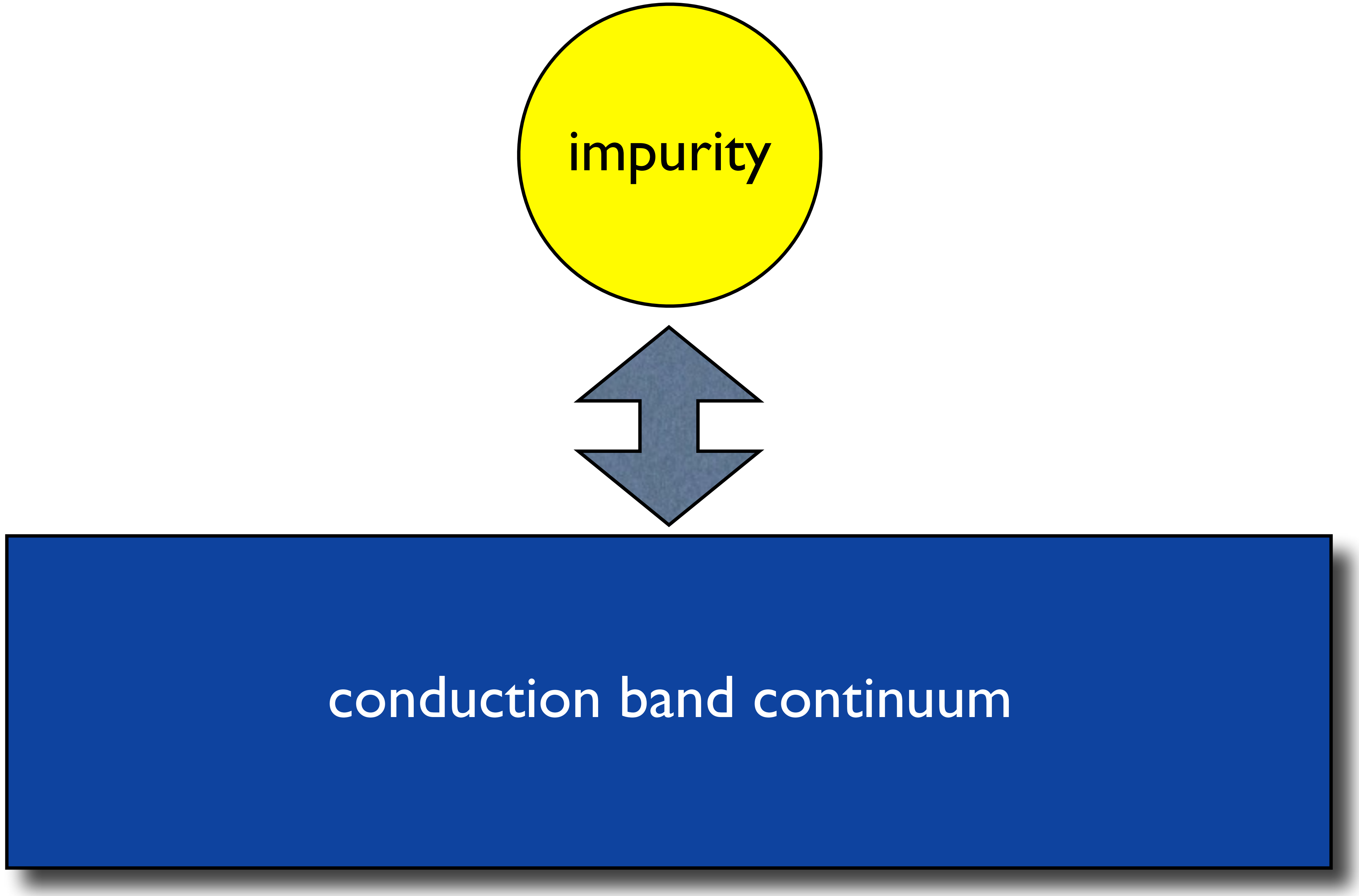
$$H_K = J\vec{S}_{\rm loc}\vec{s}_c$$

s=1/2: Kondo singlet formation

non perturbative since orthogonal ground state

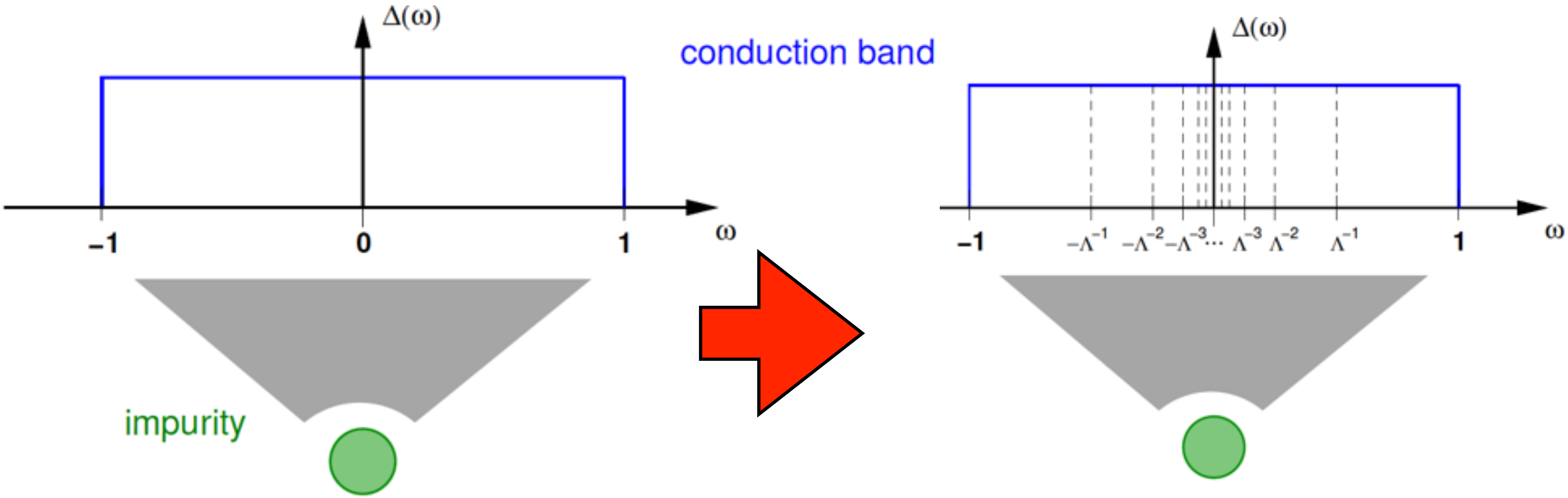
K. Wilson, RMP 1975

Numerical renormalization group



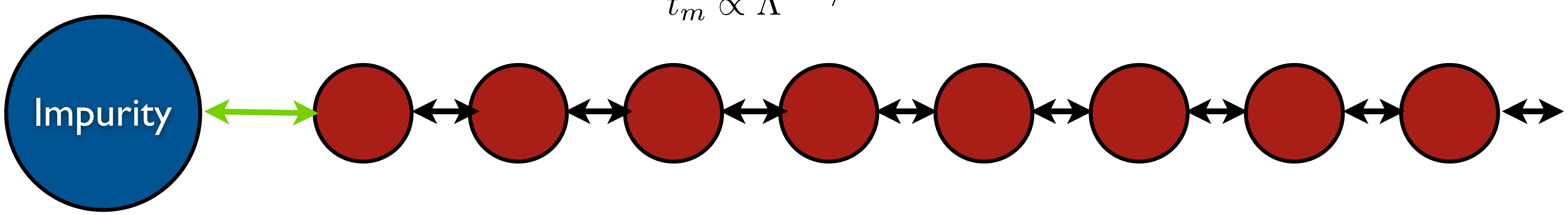
Numerical renormalization group

Ken Wilson 1975, Nobel price 1982



Mapping onto a semi-infinite chain

$t_m \propto \Lambda^{-m/2}$



NRG review: Bulla, Costi, Pruschke, RMP 80, 395 (2008)

Numerical renormalization group

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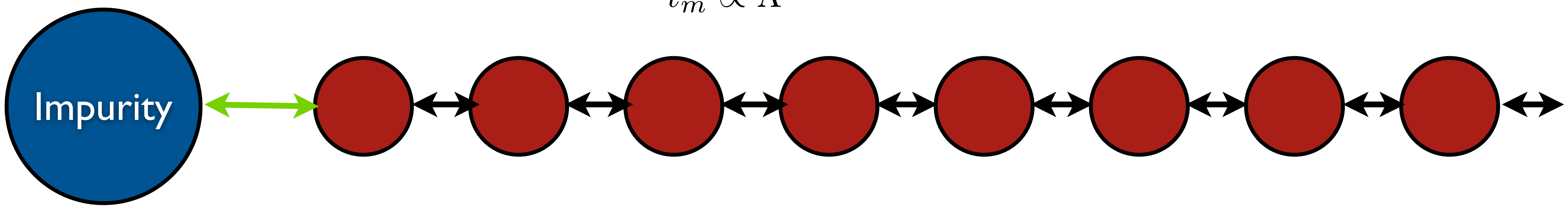


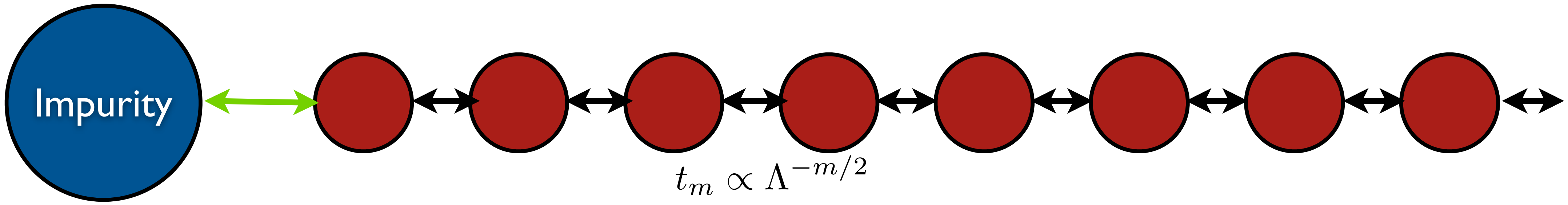
- separation of energy scales:
  discretisation parameter Λ>1
- iterative diagonalisation:
  - ➡ approx. eigenbasis
  - ➡ complete basis set: used for real time dynamics and spectral functions

$$H|l,e;m\rangle \approx E_l|l,e;m\rangle$$

$$\hat{1} = \sum_m^N \sum_l \sum_e |l,e;m\rangle\langle l,e;m|$$

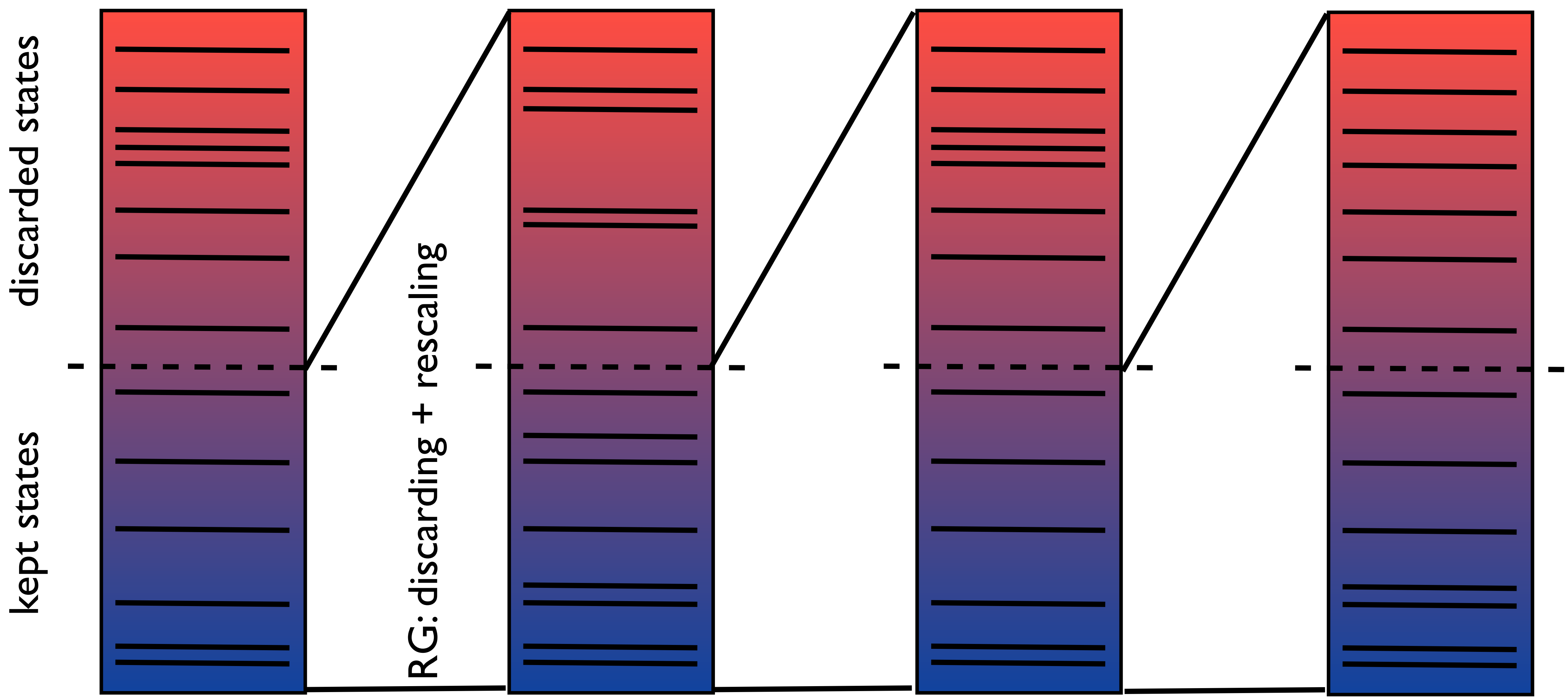
Anders,Schiller, PRL 2005,PRB 2006

Numerical renormalization group



Anders,Schiller, PRL 2005,PRB 2006

iterative diagonalisation



all discarded states: complete basis set

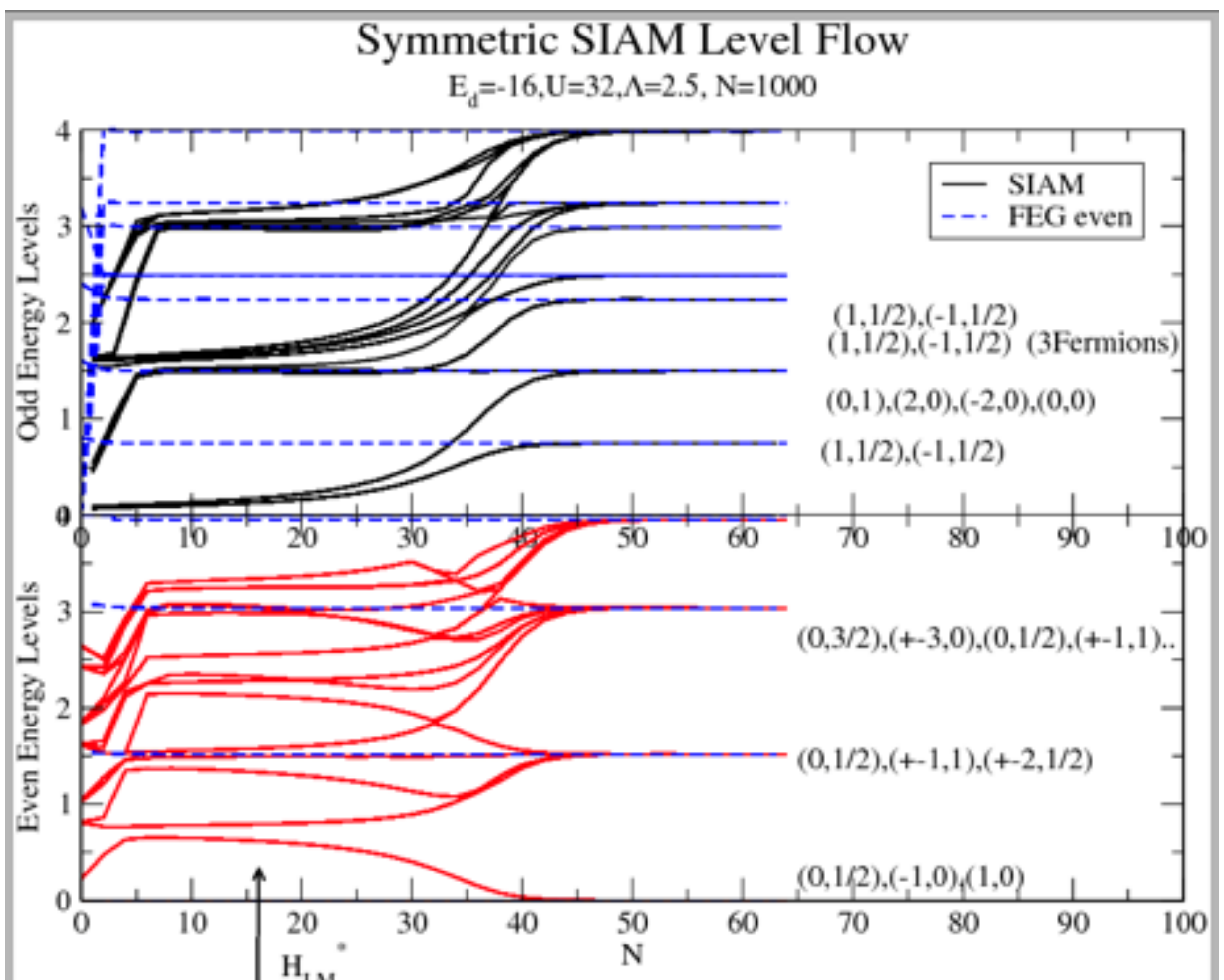
$$\hat{1} = \sum_{m}^{N} \sum_{l} \sum_{e} |l, e; m\rangle\langle l, e; m|$$

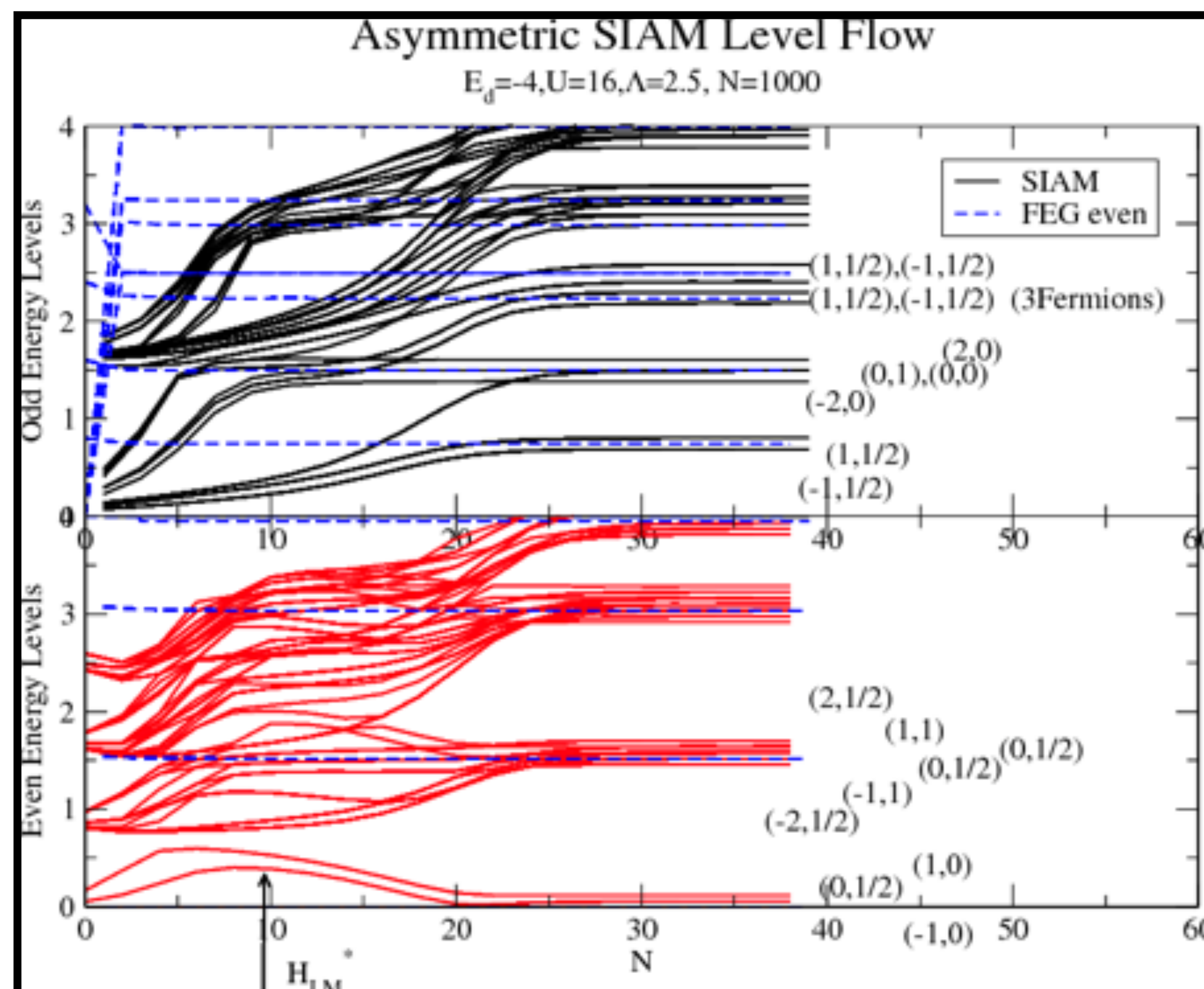
Numerical renormalization group

What does the NRG tells us?

1. thermodynamical expectation values $$\langle \hat{O} \rangle = \sum_l \frac{e^{-\beta E_l}}{Z} \langle l | \hat{O} | l \rangle$$

2. Level flow of low lying excitations: fixed points





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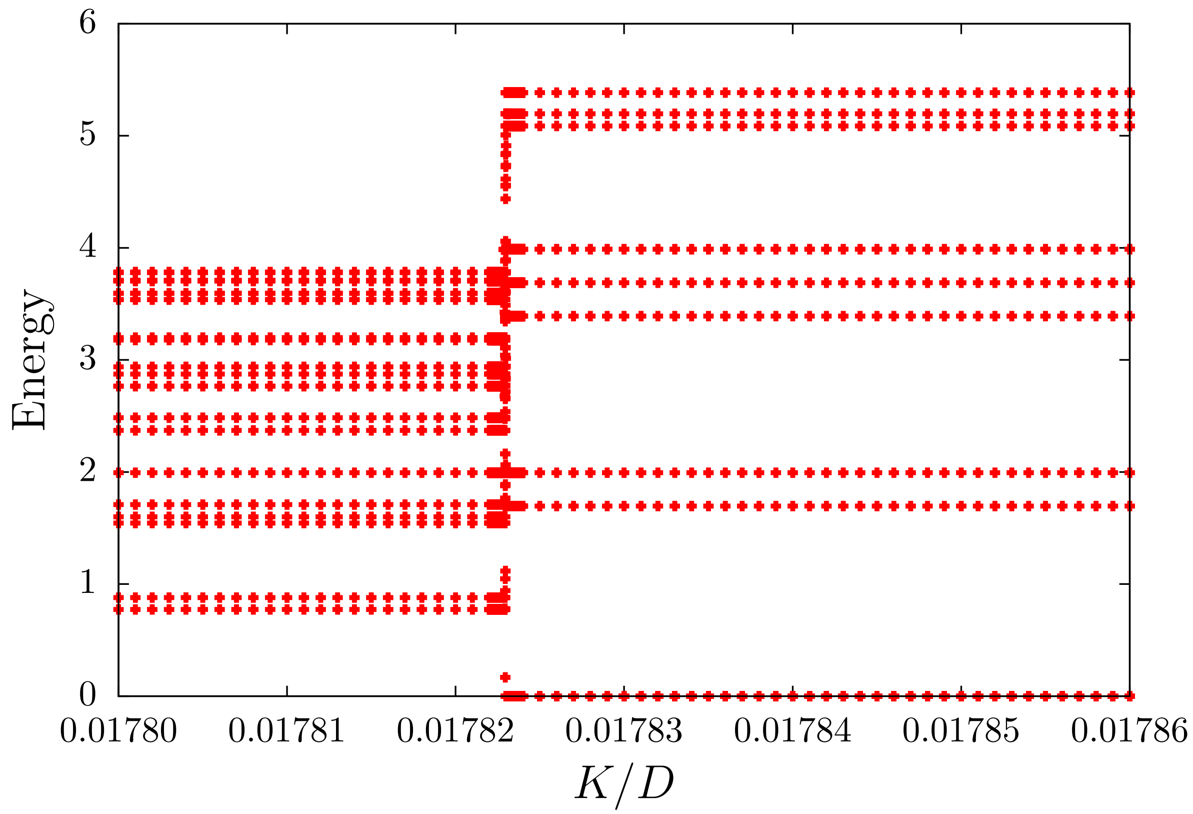
3. Quantum critical points and quantum phase transitions



external control parameter K:
change of fixed points

What does the NRG tells us?

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4. Stability of fixed points

$$R[H^* + \delta H] = H^* + \delta H' = H^* + \sum_i \lambda_i c_i \Delta H_i$$

- λ_i <1: irrelevant operator
- λ_i =1: marginal operator (marginal relevant: Kondo coupling)
- λ_i >1: relevant operator: flow away from the fixed point

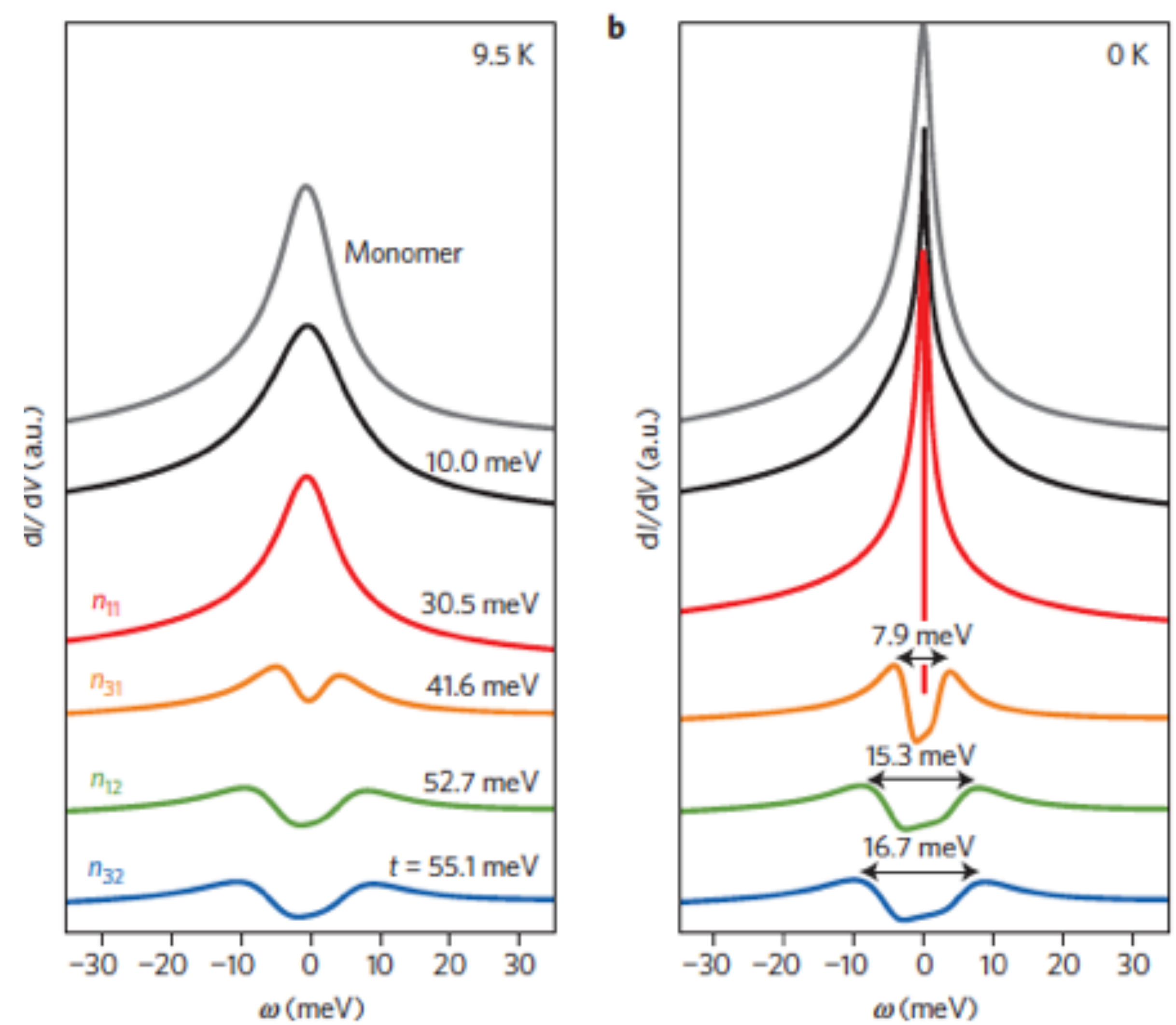
Numerical renormalization group

What does the NRG tells us?

1. thermodynamical expectation values $$\langle \hat{O} \rangle = \sum_l \frac{e^{-\beta E_l}}{Z} \langle l | \hat{O} | l \rangle$$
2. Level flow of low lying excitations: fixed points
3. Quantum critical points and quantum phase transitions
4. Stability of fixed points
5. **Spectral functions**

Hofstetter, PRL 2001
Peters et al,PRB 2006
Weichselbaum et al, PRL 2007

example: spectra for Au/PTCDA dimers
Esat et al, Nature Physics 2016



Numerical renormalization group

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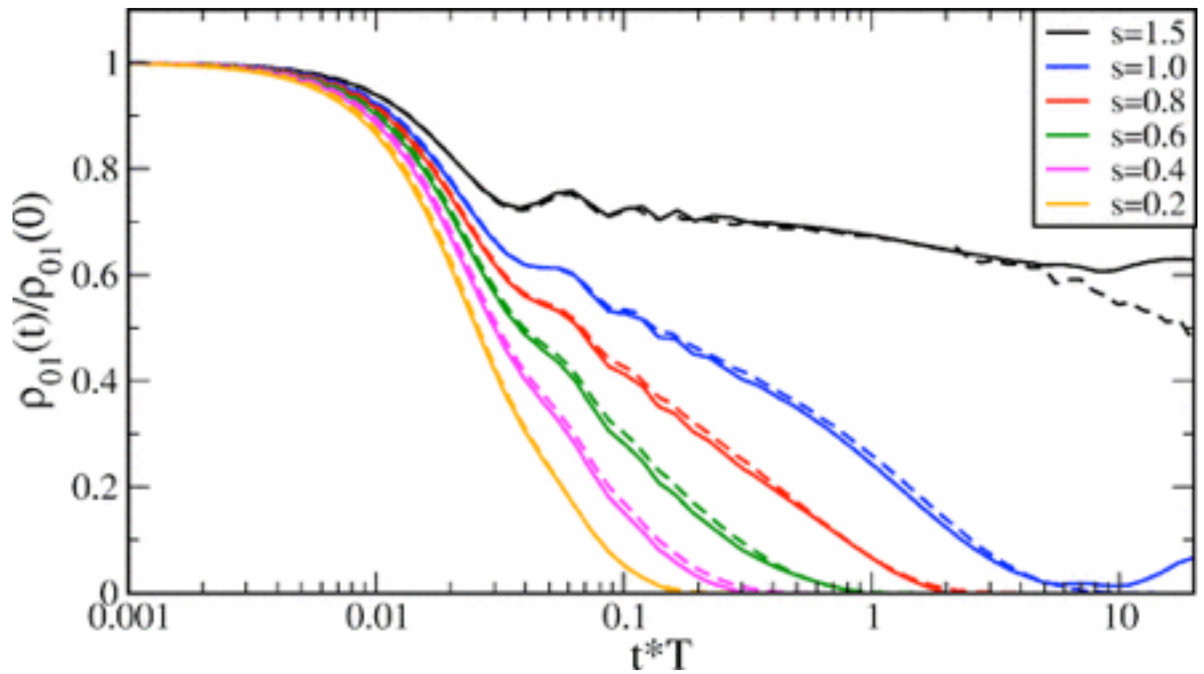
4. Stability of fixed points
5. Spectral functions
6. **Real time dynamics after quenches: TD-NRG**

$$\langle \hat{O}(t) \rangle = \sum_m \sum_{rl}{}' \rho^{\mathrm{red}}_{rl} \langle l | \hat{O} | r \rangle e^{i(E_l - E_r)t}$$

Anders, Schiller, PRL 2005, PRB 2006



dephasing im spin-boson model

Numerical renormalization group

What does the NRG tells us?

1. thermodynamical expectation values $$\langle \hat{O} \rangle = \sum_l \frac{e^{-\beta E_l}}{Z} \langle l | \hat{O} | l \rangle$$

2. Level flow of low lying excitations: fixed points

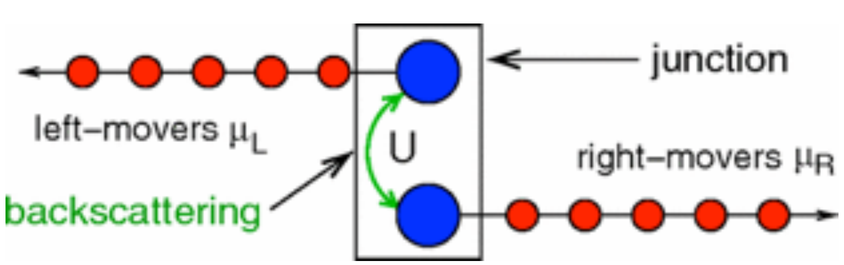
3. Quantum critical points and quantum phase transitions

4. Stability of fixed points

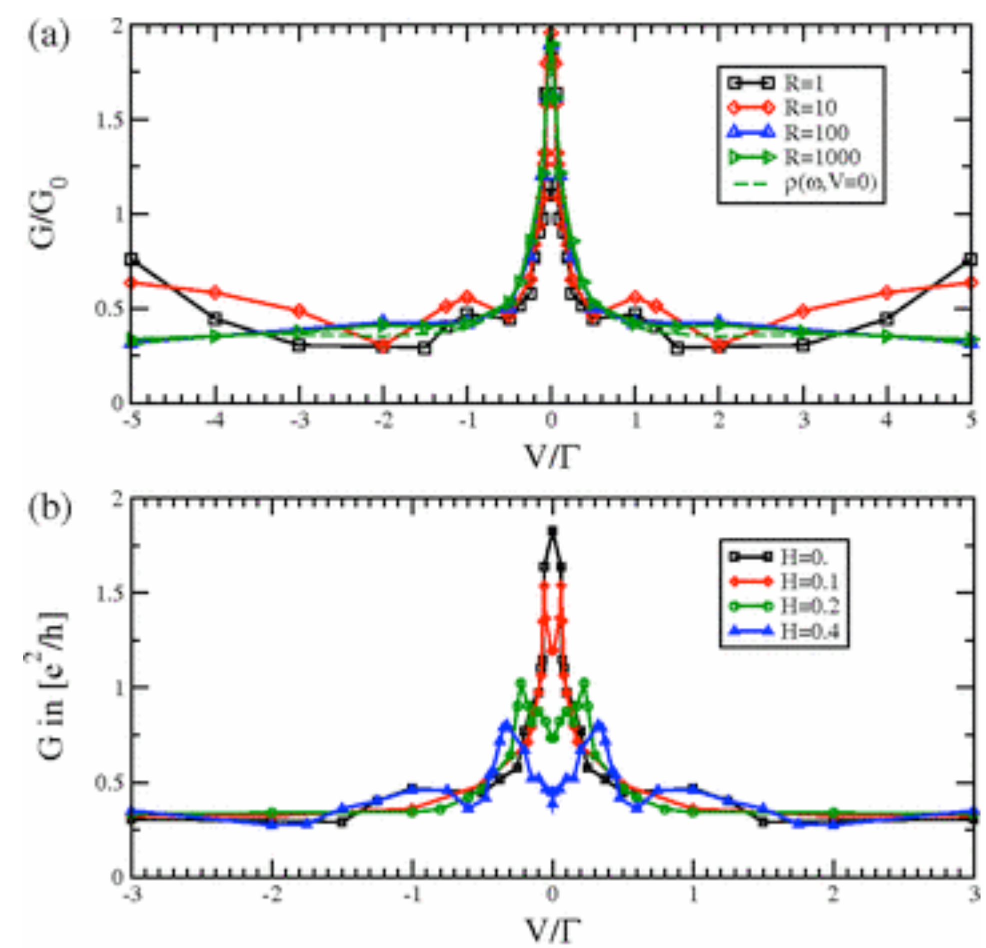
5. Spectral functions

6. Real time dynamics after quenches (TD-NRG)

7. Steady-State currents: (SNRG)



Anders, PRL 2008



NEQ numerical renormalization group

$$\langle \hat{O} \rangle(t) = \mathrm{Tr}\left[\hat{O}\hat{\rho}(t)\right]$$

- equilibrium: one condition $\hat{\rho}(t) = \hat{\rho}_0 = \exp(-\beta H)/Z$
- non-equilibrium: two conditions: $\hat{\rho}_0$ and H^f

$$\hat{\rho}(t) = e^{-iH^f t}\hat{\rho}_0 e^{iH^f t}$$

- calculation of the trace using energy eigenstate

$$\langle \hat{O} \rangle(t) = \sum_{n,m} \langle E_n|\hat{O}|E_m\rangle\langle E_m|\rho_0|E_n\rangle e^{-i(E_m-E_n)t}$$

TD-NRG complete basis set:

$$\langle \hat{O} \rangle(t) \quad = \quad \sum_m \quad \sum_{l,l}^{l \text{ or } l' \text{ discarded}} \quad \langle l|\hat{O}|l'\rangle e^{i(E_l-E_{l'})t}\rho^{red}_{l'l}(m)$$

Applications

1. TD-NRG: propagation of Kondo correlations
2. Steady-state currents
3. chemically driven quantum phase transition in Au/PTCDA dimers on a gold surface

Applications

1. **TD-NRG: propagation of Kondo correlations**
2. Steady-state currents
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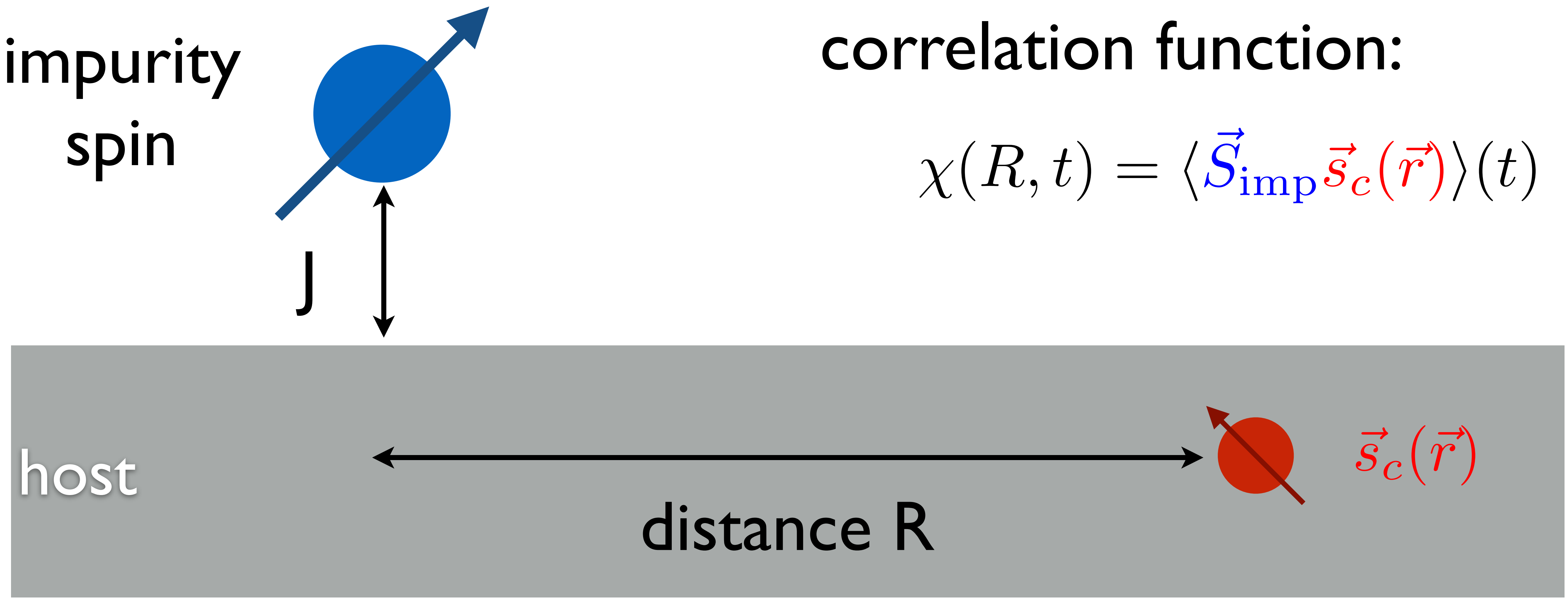
Kondo model: finite R spin-spin correlations

correlation function:

$$\chi(R,t) = \langle \vec{S}_{\mathrm{imp}} \vec{s}_c(\vec{r}) \rangle (t)$$

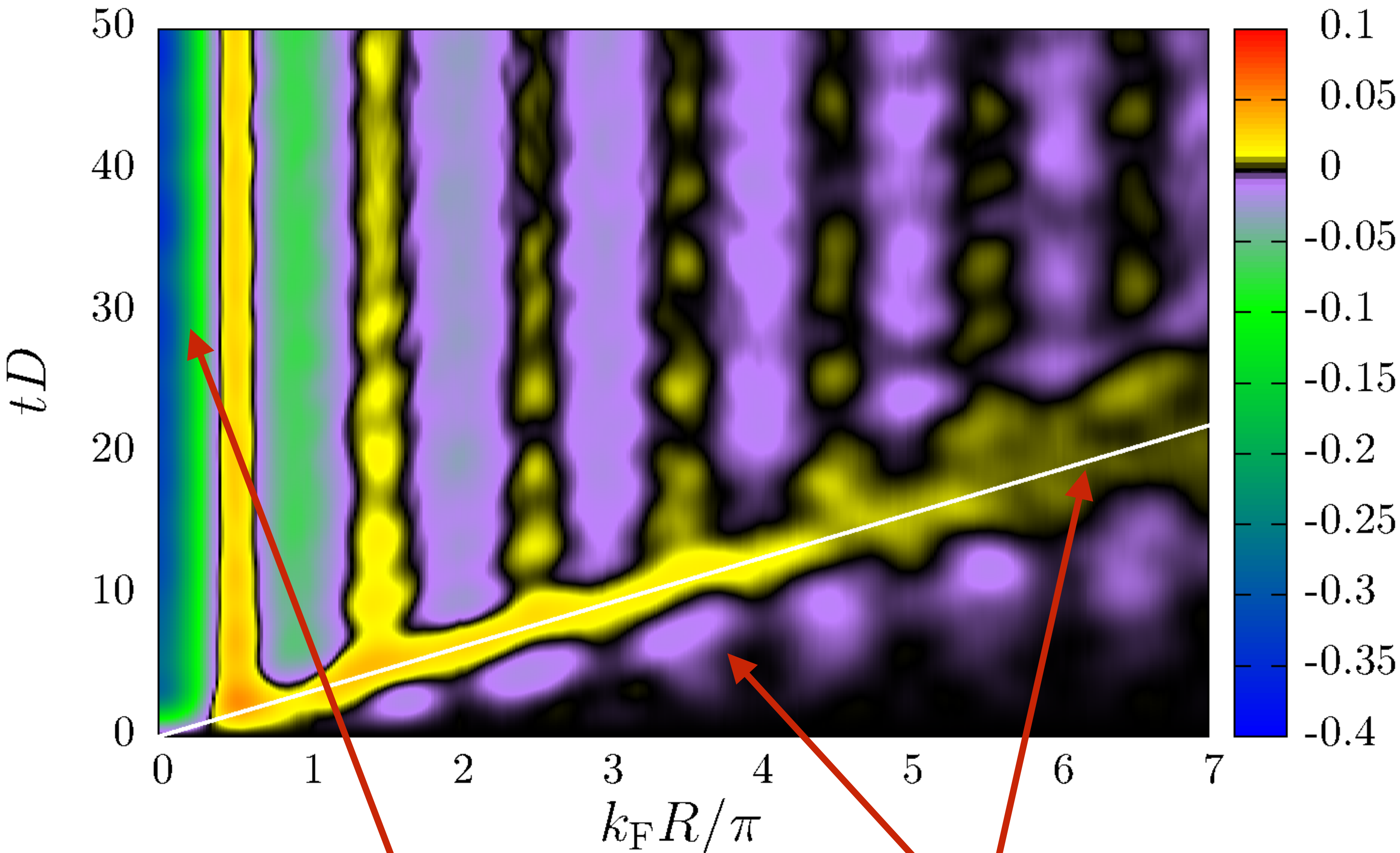


J. Kondo, Prog. Theor. Phys 32, 37 (1964)

$$H = \sum_{\sigma} \int_{-D}^{D} d\varepsilon \, \varepsilon \, c^{\dagger}_{\varepsilon\sigma} c_{\varepsilon\sigma} + J \vec{S}_{\mathrm{imp}} \vec{s}_c(0)$$

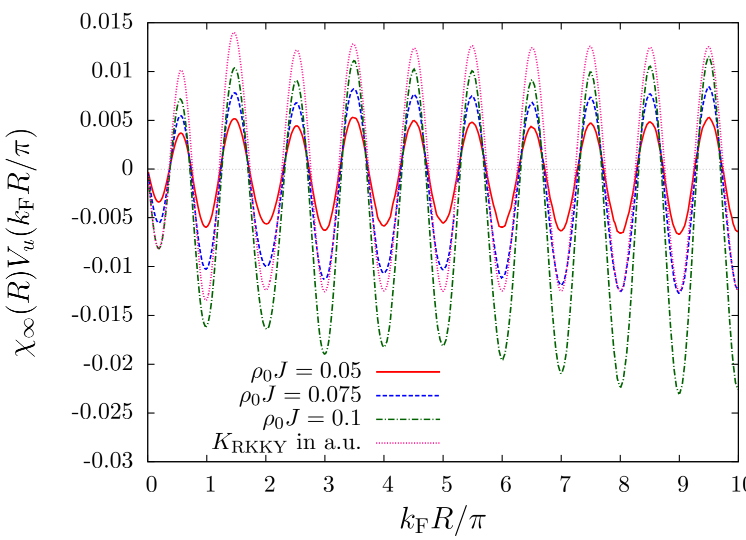
R dependency: mapping on a two band model: even and odd parity

non-equilibrium dynamics



$$\chi(R,t) = \langle \vec{S}_{\rm imp} \vec{s}_{\rm c}(\vec{r}) \rangle (t)$$

$$\rho_0 J = 0.3$$

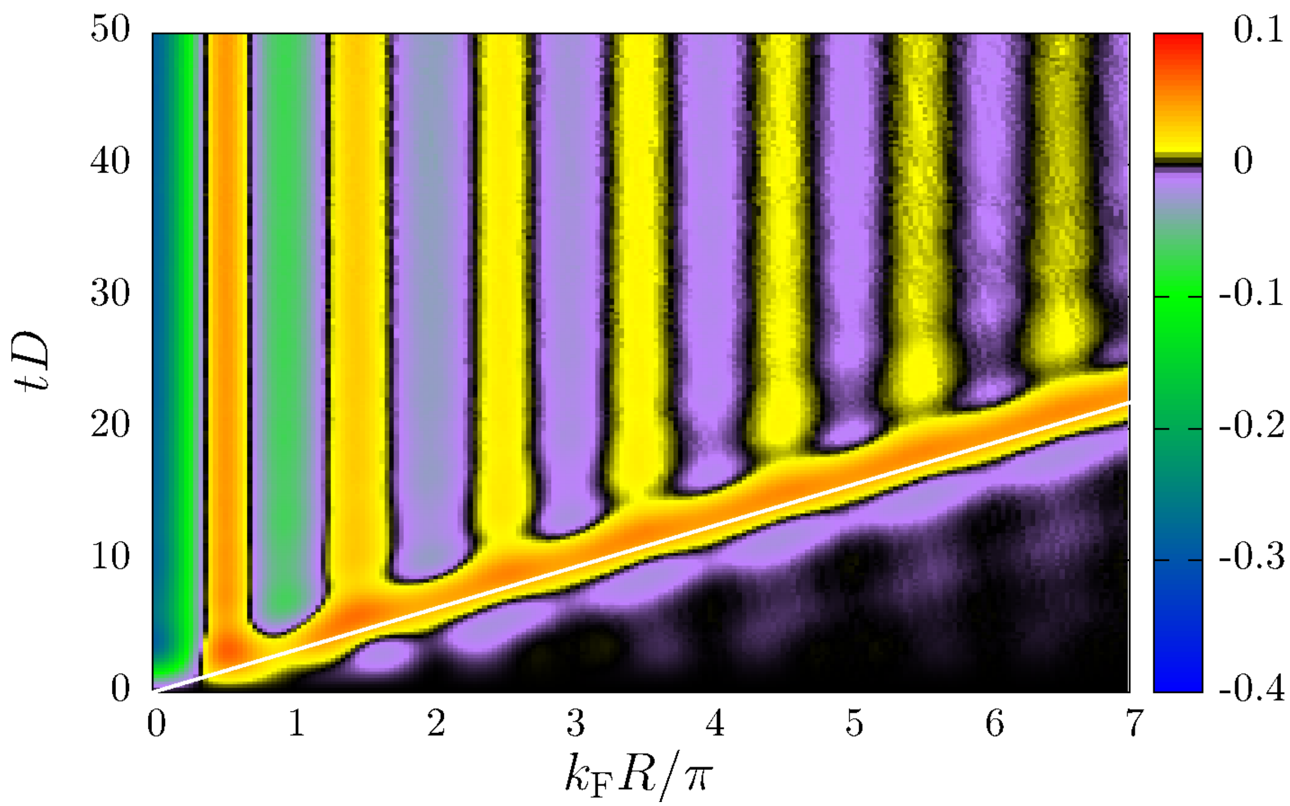


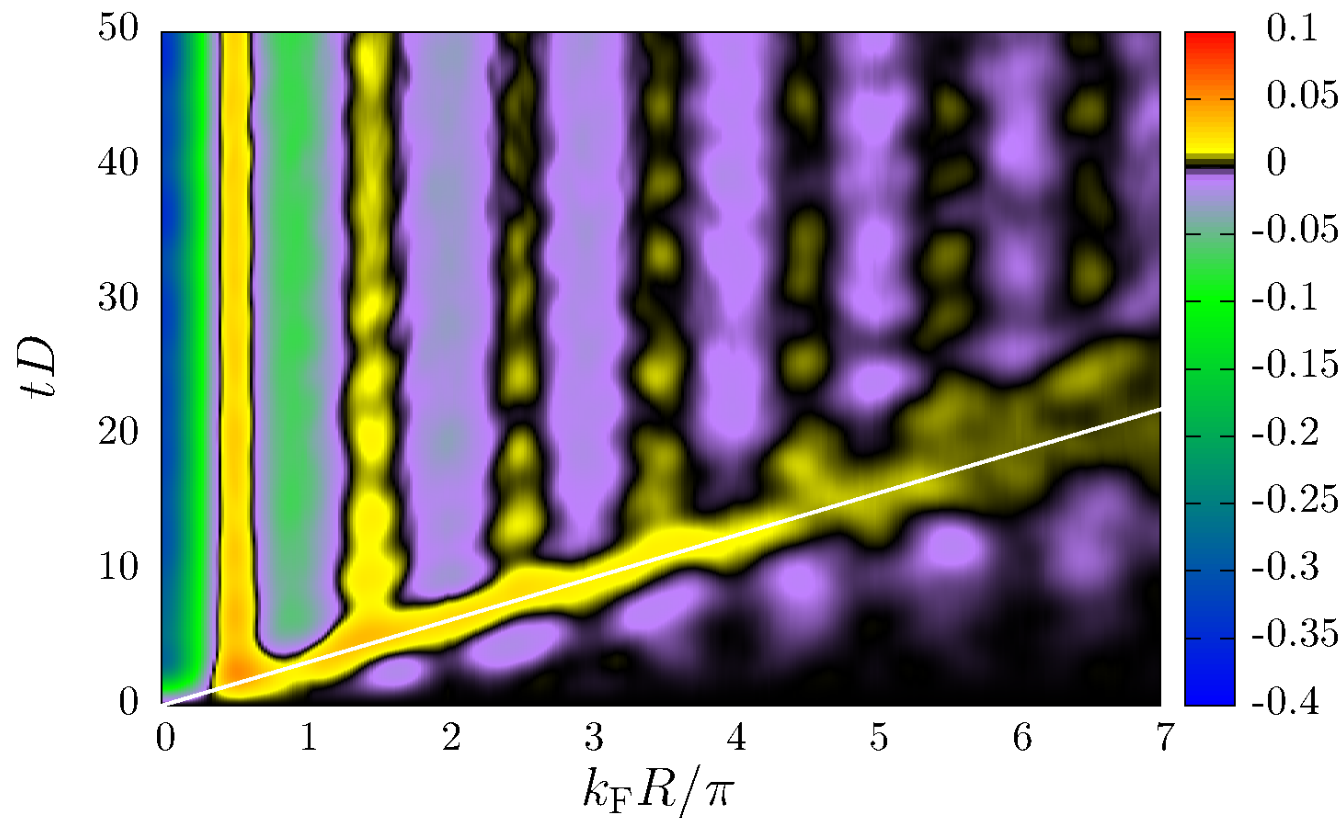
- "light-cone" physics R=v_Ft
- surprice: buildup of correlations outside the light-cone
- fast short time scale and thermalisation inside the light-cone

Lechtenberg, FBA Phys. Rev. B 90, 045117 (2014)

Comparison: perturbation theory

$$\rho^I(t) = \rho_0 + i\int_0^t \left[\rho_0, H_K^I(t_1)\right] dt_1 - \int_0^t \int_0^{t_1} \left[\left[\rho_0, H_K^I(t_2)\right] H_K^I(t_1)\right] dt_2 dt_1 + O(J^2)$$





second order perturbation theory
no Kondo effect

TD-NRG
Kondo effect included

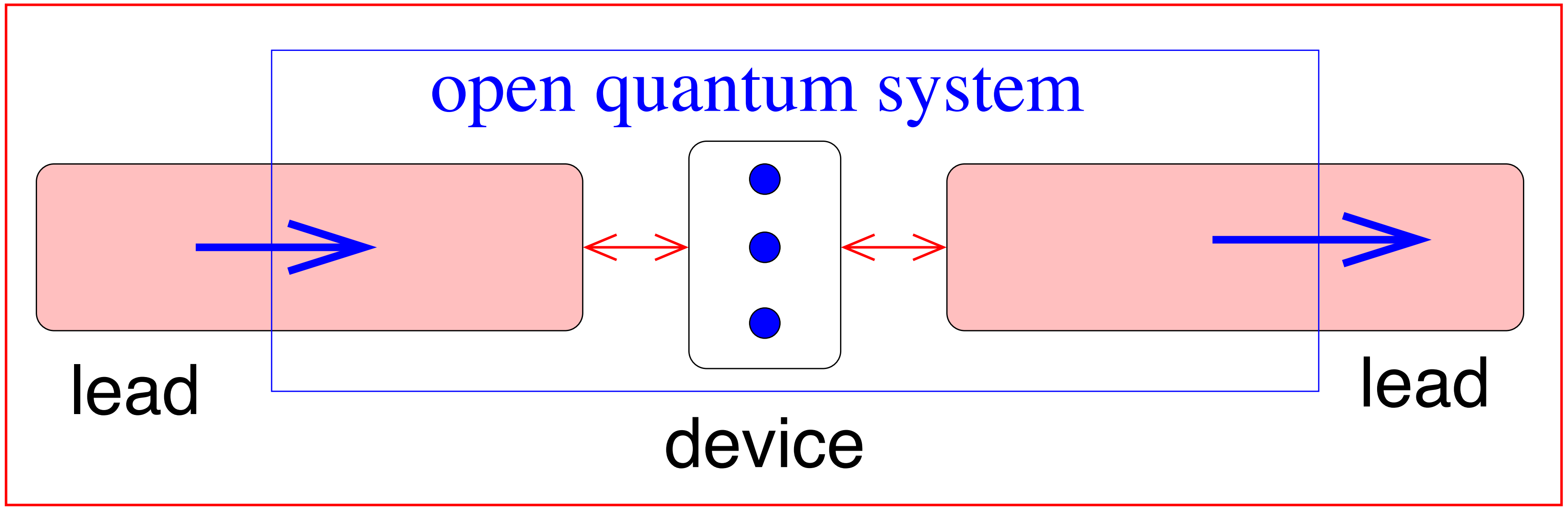
qualitative very good agreement: correlations outside the light cone persist

Lechtenberg, FBA Phys. Rev. B 90, 045117 (2014)

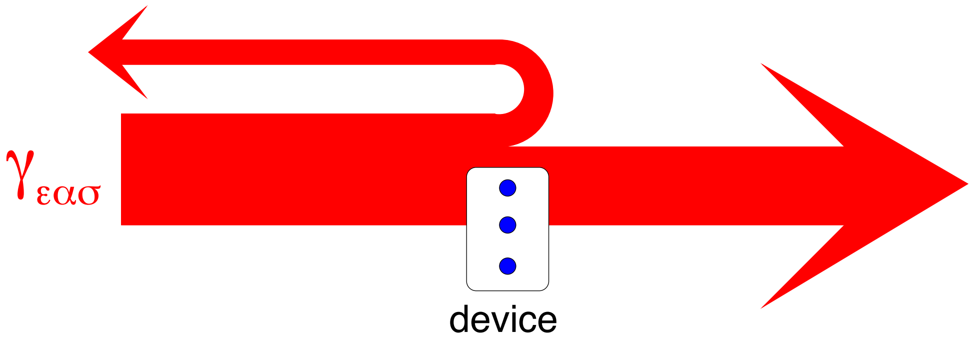
Applications

1. TD-NRG: propagation of Kondo correlations
2. **Steady-state currents**
3. chemically driven quantum phase transition in Au/PTCDA dimers on a gold surface

Boundary condition



Lippmann-Schwinger
scattering states



Steady state:

$$\rho(\mu_L, \mu_R) = \frac{1}{Z} e^{-\beta(H - Y(\mu_L, \mu_R))}$$

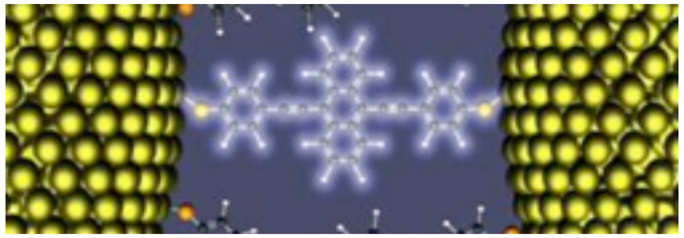
Hershfield, PRL, **70**, 2134 (1993)

SNRG:

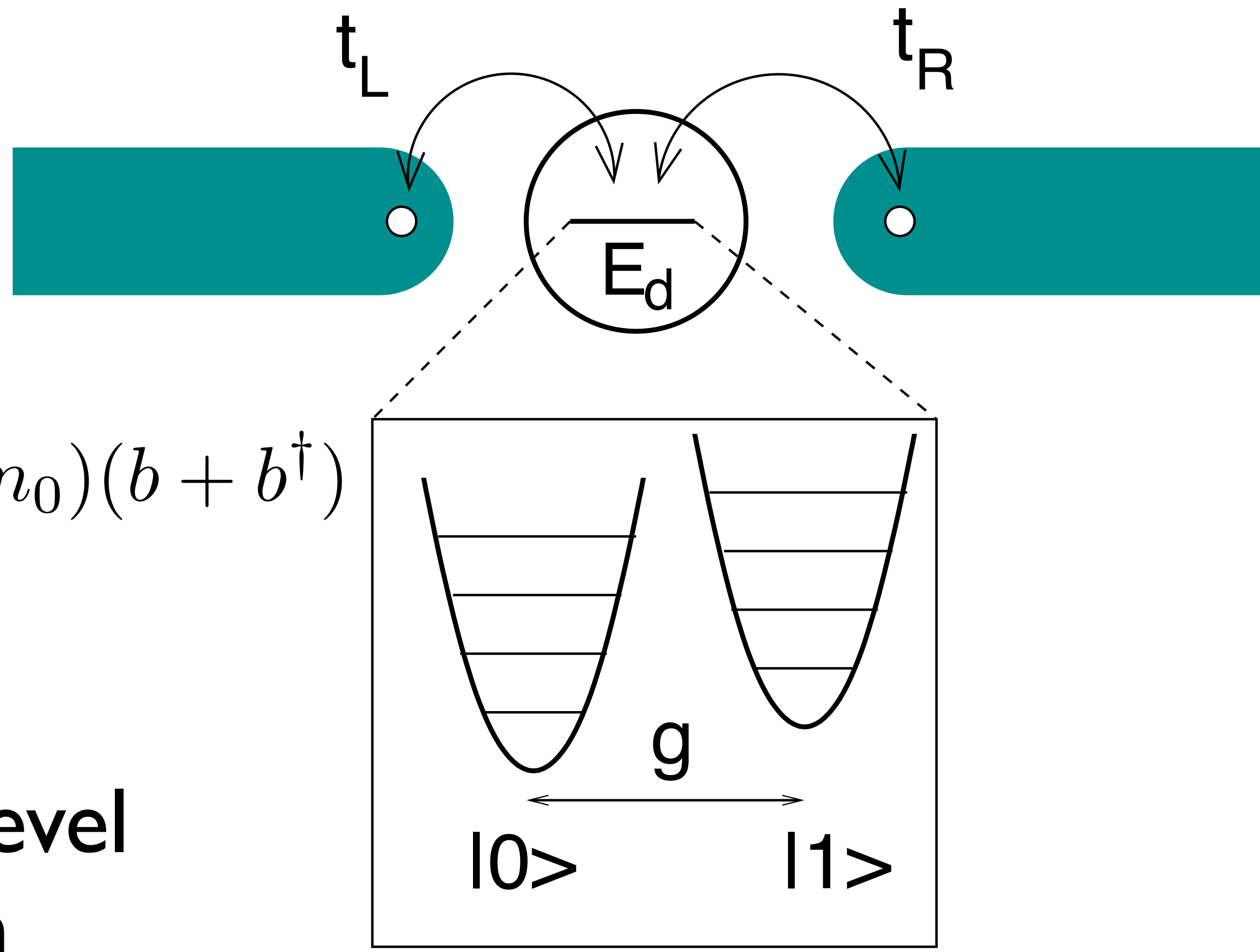
$$\rho(\mu_L, \mu_R) = \lim_{t \to \infty} e^{iHt} \rho_0(\mu_L, \mu_R) e^{-iHt}$$

including charging energy exactly

Anders, PRL, 101, 066804 (2008)



Holstein Anderson model



$$H_{\mathrm{e-ph}} = \lambda_{\mathrm{ph}}(\hat{n} - n_0)(b + b^\dagger)$$

$g = \lambda_{ph}/\omega_0$

rel. el.-phonon coupling

- two leads
- molecular level
- tunnel term
- phonon mode
- electron phonon coupling

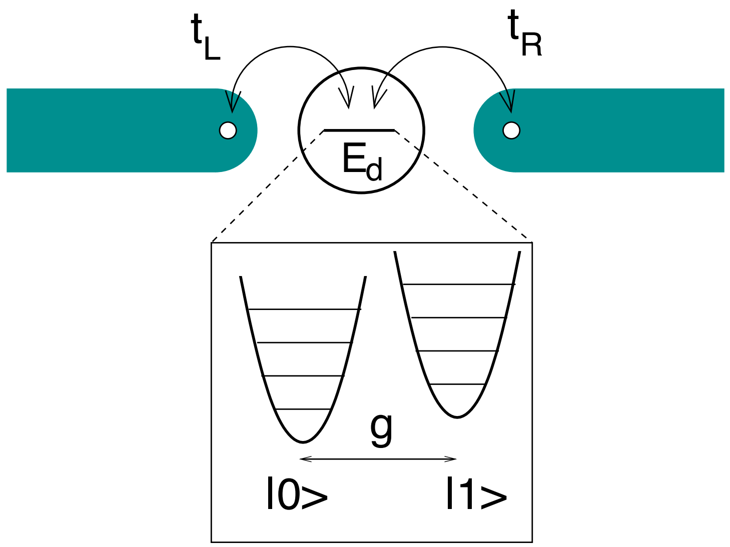
Lang, Firsov, JETP 16, 1301 (1962)

$t_L = t_R = 0$: exactly solvable local problem

polaron formation: entanglement between electron and phonon

M. Galperin et al, J.of Physics: Condensed Matter 19, 103201 (2007)

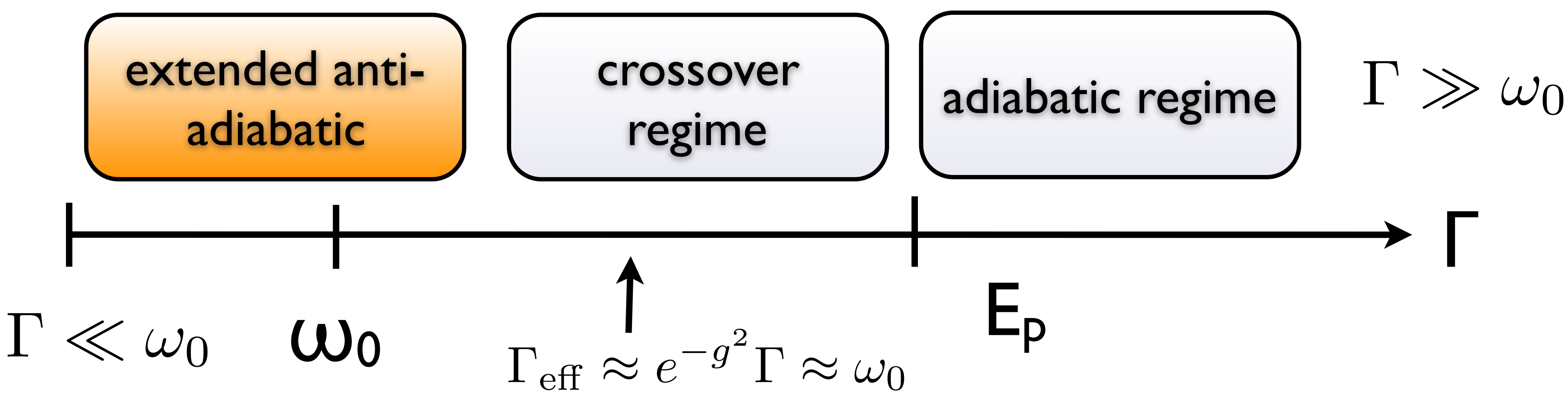
Equilibrium properties



Three energy scales

- charge fluctuation $\Gamma = \pi t^2 \rho_0$
- phonon frequency ω_0
- polaronic shift $E_p = \frac{\lambda_{ph}^2}{\omega_0} = g^2 \omega_0$

ω_0 finite



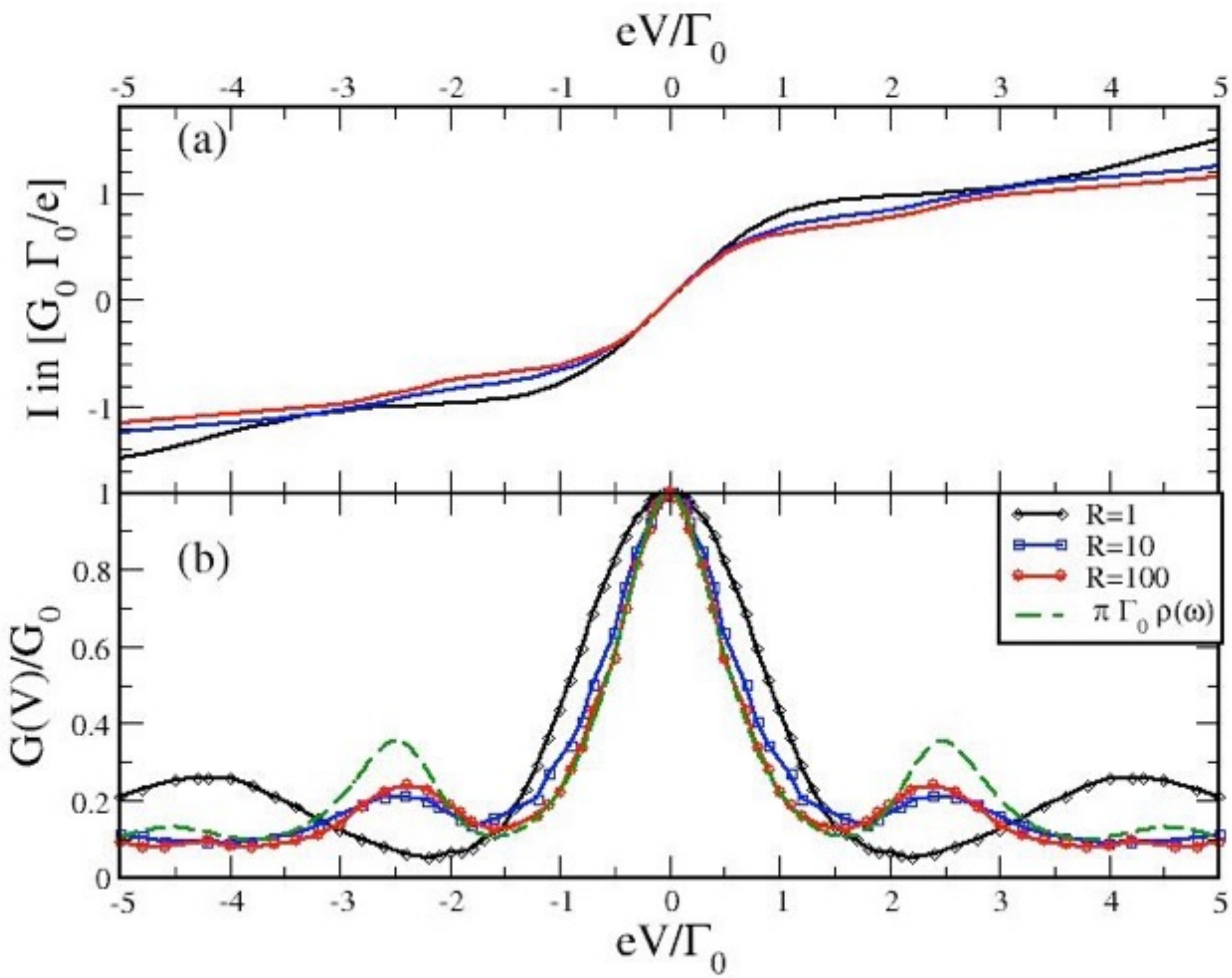
Γ_{eff} : Polaron formation, tunneling suppressed

Lang, Firsov, JETP 16, 1301 (1962)

Eidelstein et al, PRB 87, 075319 (2013)

A. Jovchev, FBA, PRB 87, 195112 (2013)

ph-symmetric case



$$G_0 = \frac{e^2}{h}\frac{4\Gamma_L\Gamma_R}{\Gamma_0^2}$$

$$R = \frac{\Gamma_L}{\Gamma_R}$$

$$\frac{\lambda_{\mathrm{ph}}}{\Gamma_0} = \frac{\omega_0}{\Gamma_0} = 2$$

tunnel regime

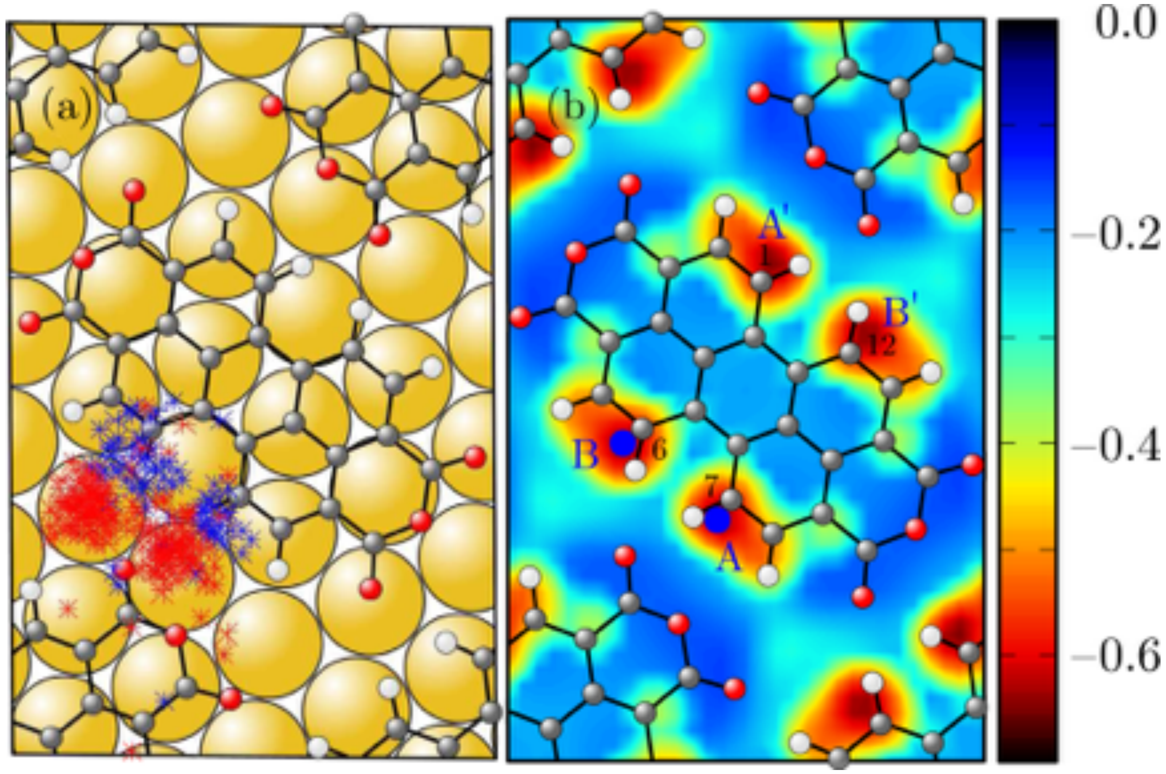
$R \to \infty$

A. Jovchev, FBA, PRB 87, 195112 (2013)

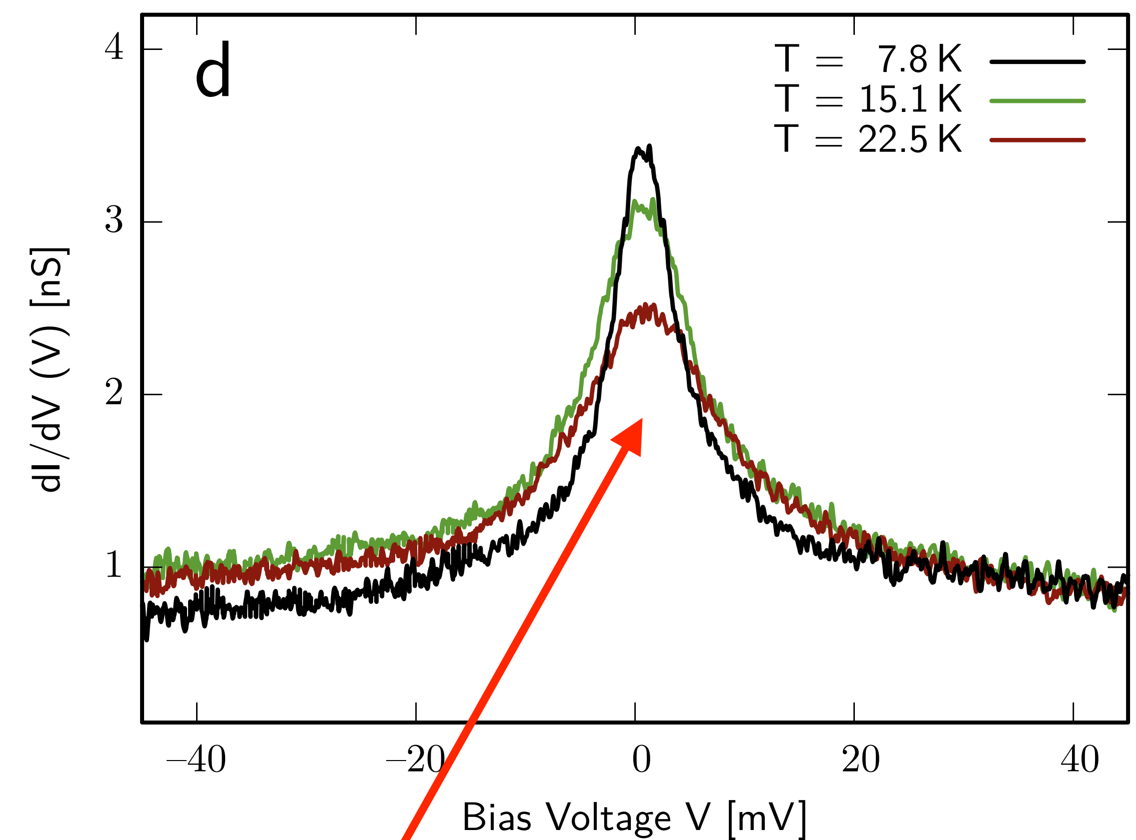
Applications

1. TD-NRG: propagation of Kondo correlations
2. Steady-state currents
3. **chemically driven quantum phase transition in Au/PTCDA dimers on a gold surface**

STM image Au/PTCDA on Ag(111)



four positions for Au: A,A',B,B'



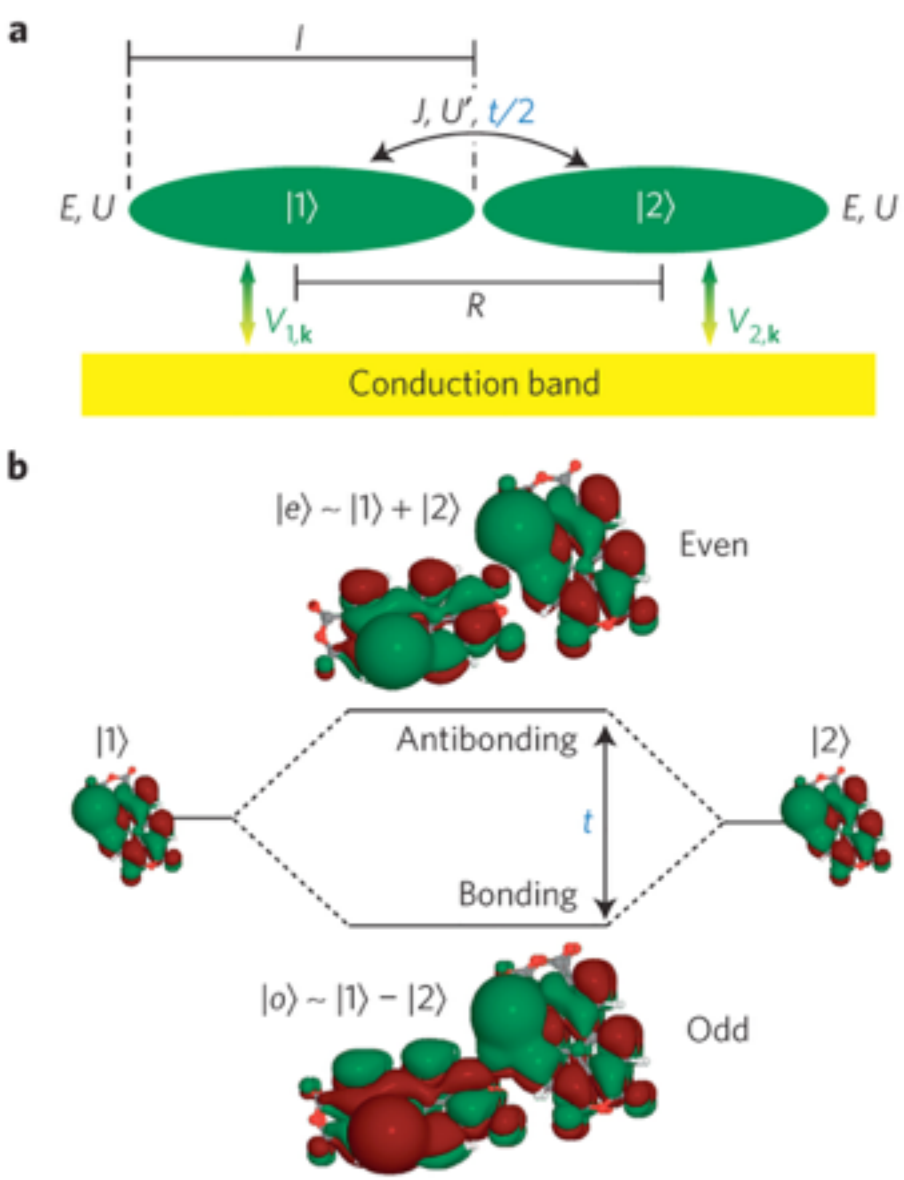
- an extend π orbital is induced in the Au/PTCDA complex

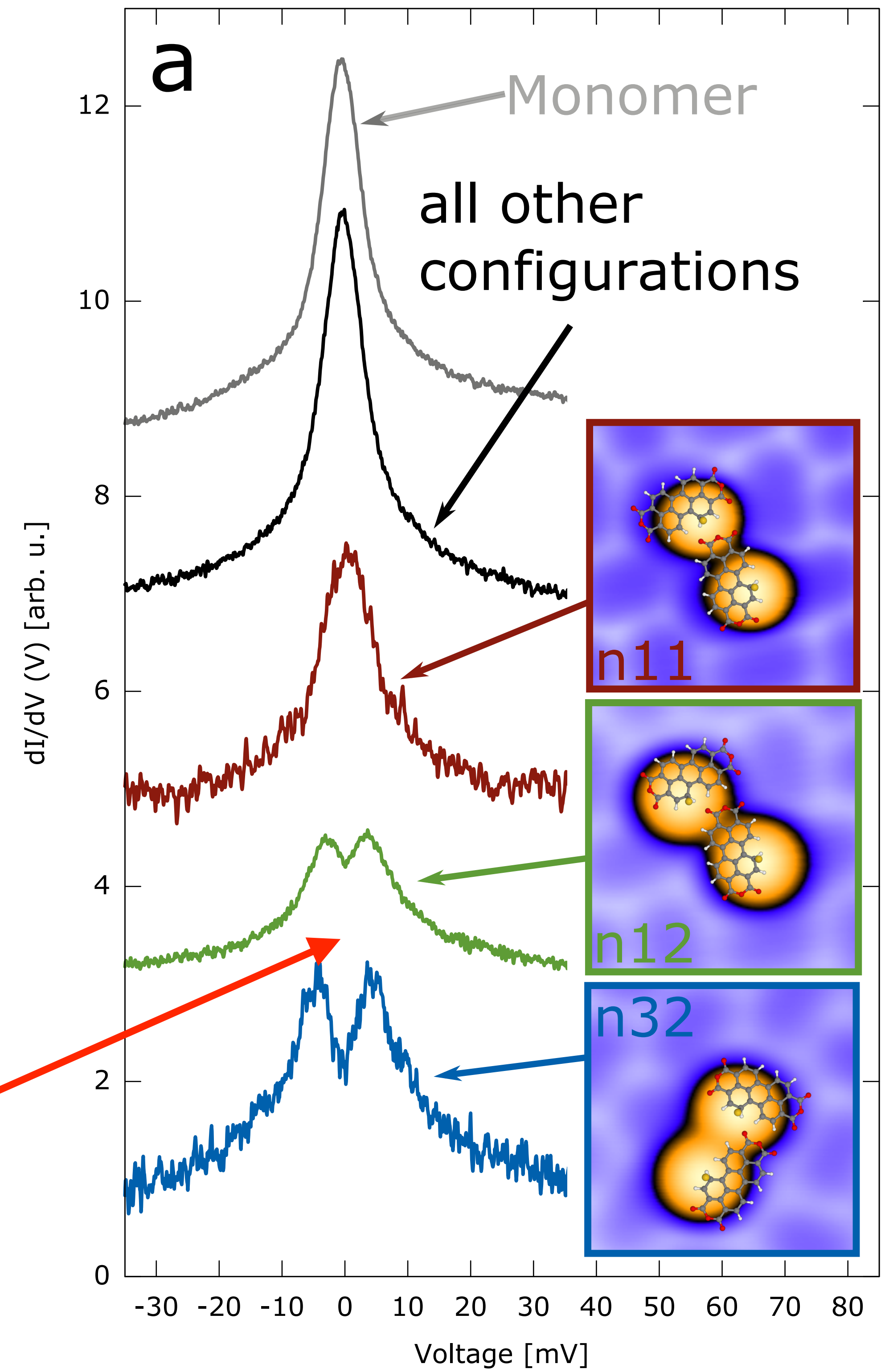
➡ spin moment induced

➡ experimental evidence: Kondo effect measured via STM

Esat et al, PRB 91,144415 (2015)

Dimer Au/PTCDA on Au(111)

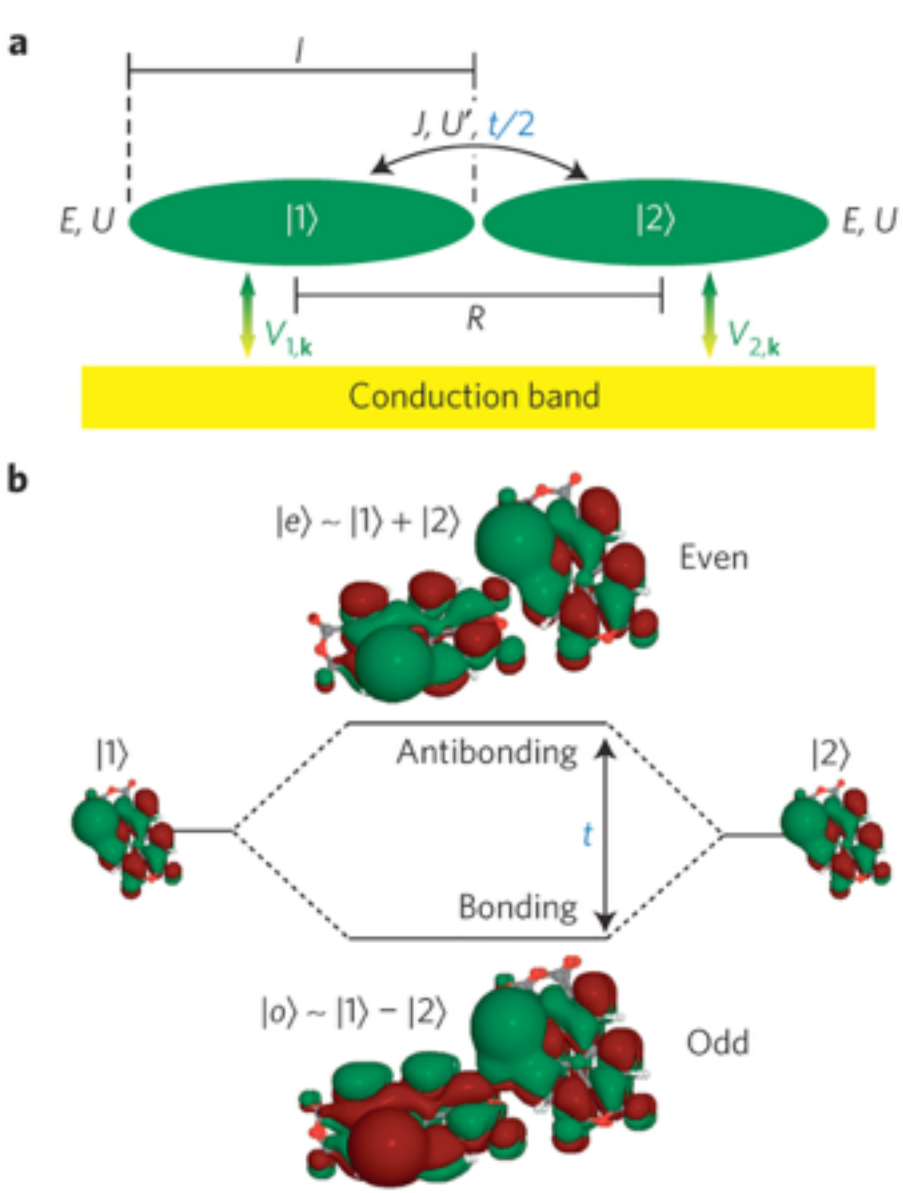


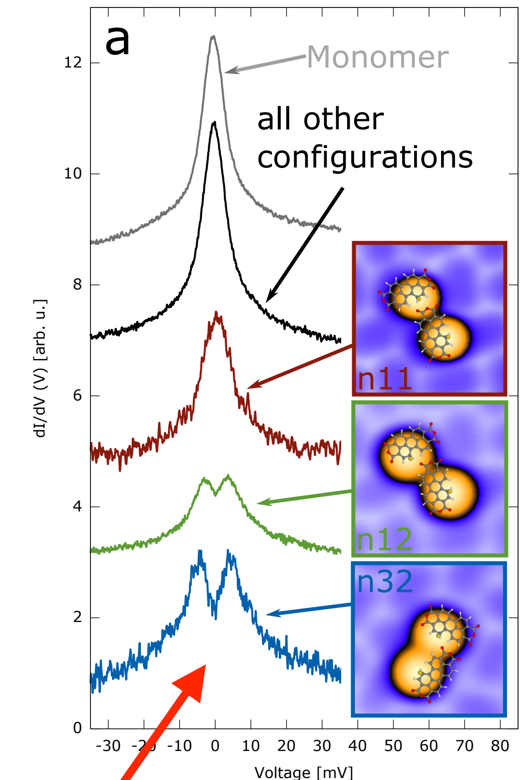


origin of the gap?

Esat et al, Nature Physics 2016

Dimer Au/PTCDA on Au(111)





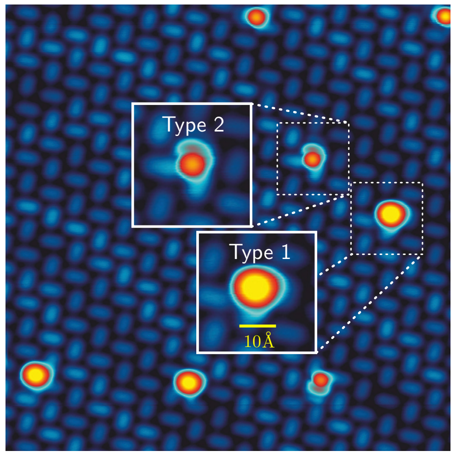
- chemical bonding between the π orbitals
- Hund's J negligible, included via $J = t^2/U$

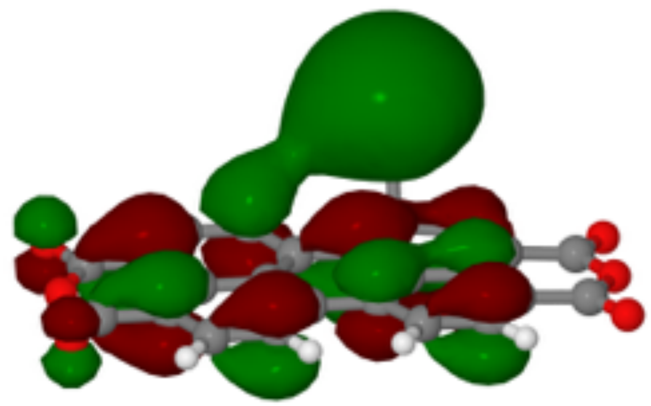
➡ chemically driven quantum phase transition

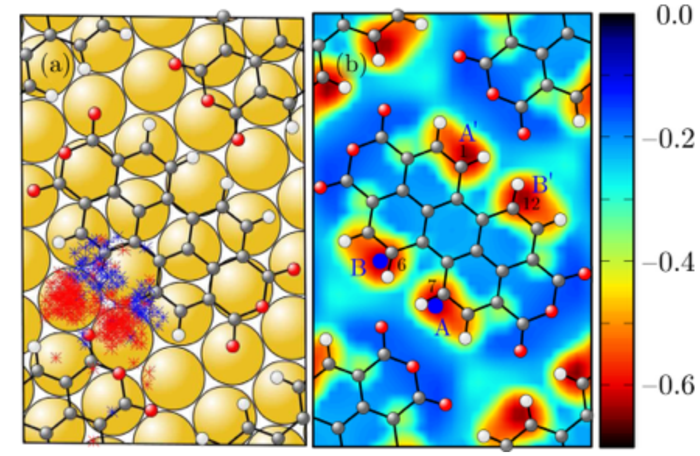
- role of parity breaking?

Esat et al, Nature Physics 2016

Modelling of the Au/PTCDA: combine LDA+NRG

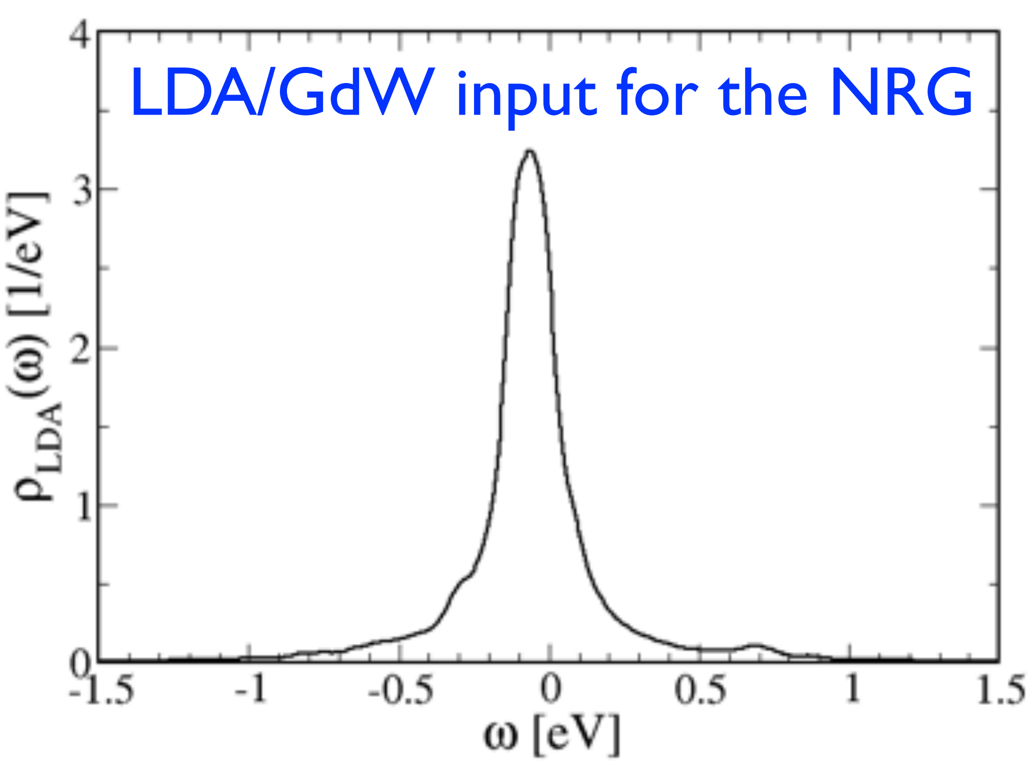






Esat et al, PRB 91,144415 (2015)

Input from LDA+GdW: $\rho_\pi(\omega)$ interpreted as MF DOS



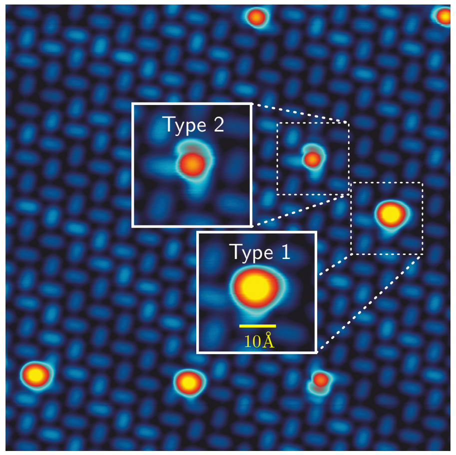
$$G_\sigma^{LDA}(z) = \frac{1}{z - E^\pi - U\langle n_{-\sigma}\rangle - \Delta(z)}$$

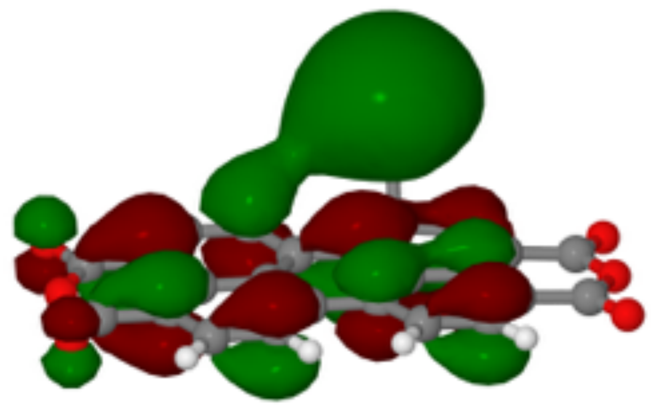
$$\Delta(z) = \sum_k \frac{V_k^2}{z - \varepsilon_k} = \frac{1}{\pi}\int_{-\infty}^{\infty} d\omega \frac{\Gamma(\omega)}{z-\omega}$$

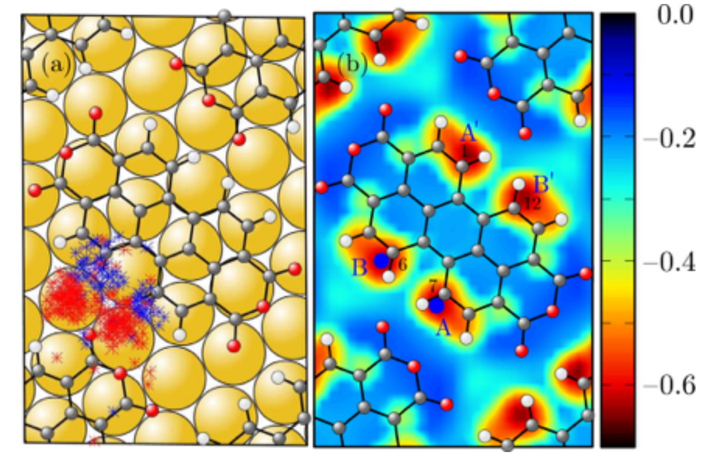
$$U = \int d^3r d^3r' |\psi_\pi(\mathbf{r})|^2 W(\mathbf{r},\mathbf{r}') |\psi_\pi(\mathbf{r}')|^2$$

U known from LDA:
calculate E^π and $\Gamma(\omega)$

Modelling of the Au/PTCDA

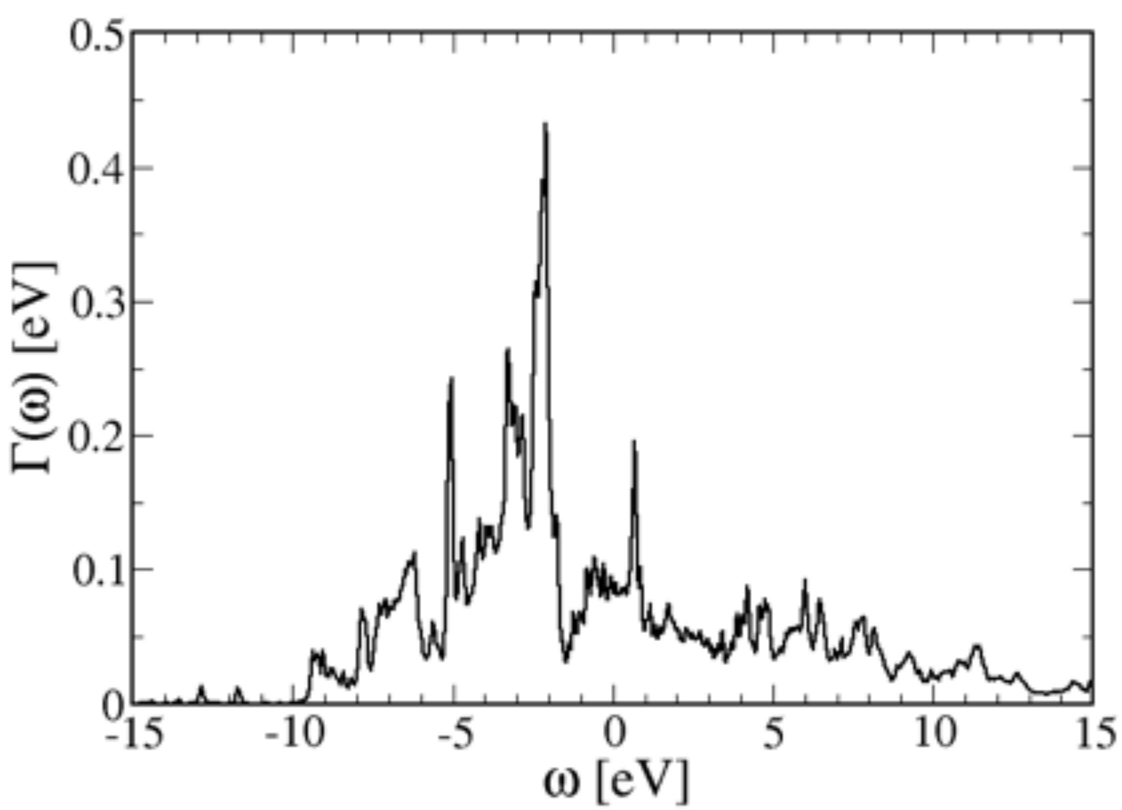






Esat et al, PRB 91,144415 (2015)

Input from LDA: $\rho_\pi(\omega)$ interpreted as MF DOS



$$G_\sigma^{LDA}(z) = \frac{1}{z - E^\pi - U\langle n_{-\sigma}\rangle - \Delta(z)}$$

$$\Delta(z) = \sum_k \frac{V_k^2}{z - \varepsilon_k} = \frac{1}{\pi}\int_{-\infty}^{\infty} d\omega \frac{\Gamma(\omega)}{z-\omega}$$

U known: calculate E^π and $\Gamma(\omega)$

U_{dimer} = 1.4 eV

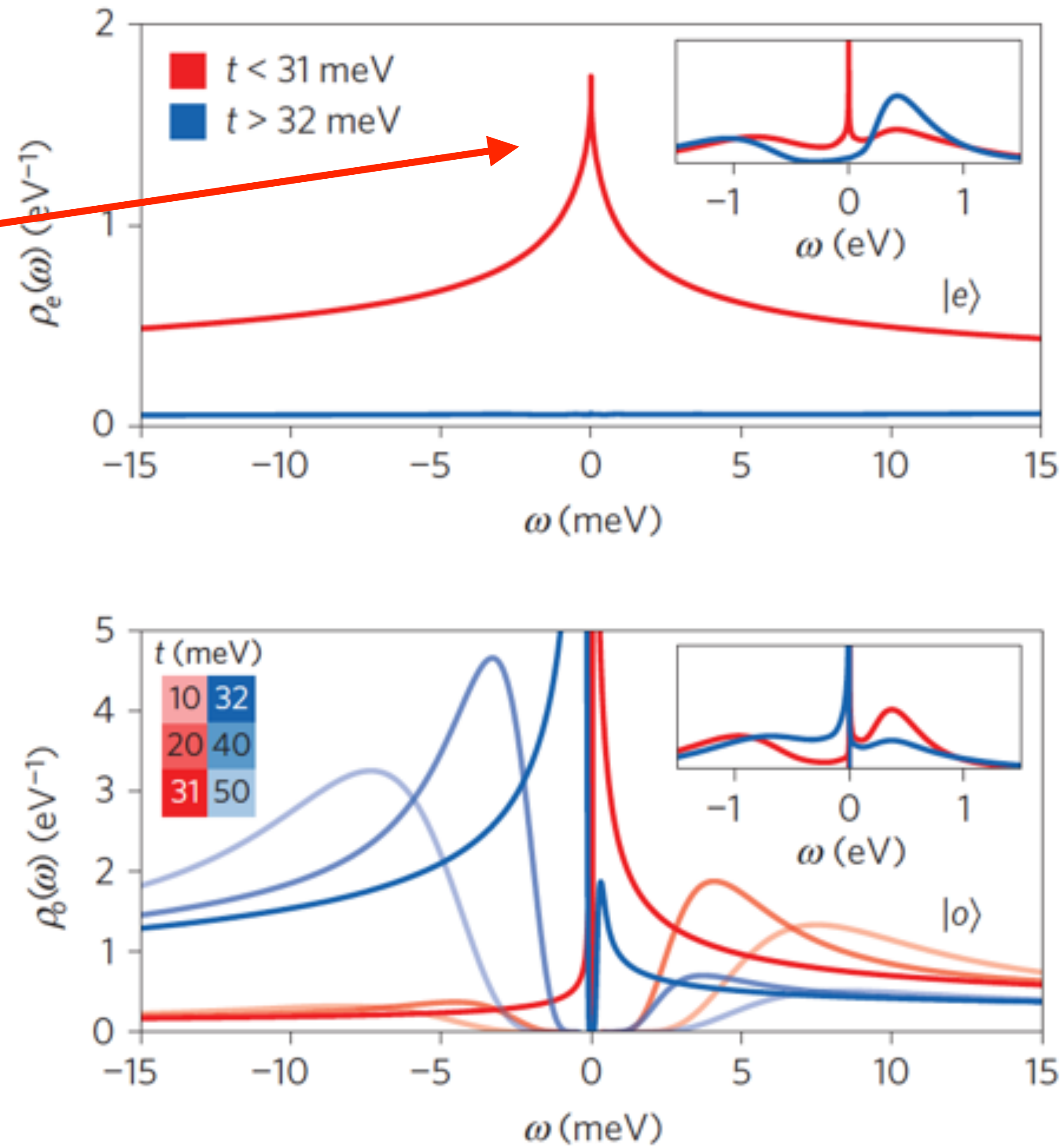
$E^\pi \sim -0.95$ eV

$\Gamma(\omega)$ input for the NRG

Dimer: $(Au/PTCDA)_2$

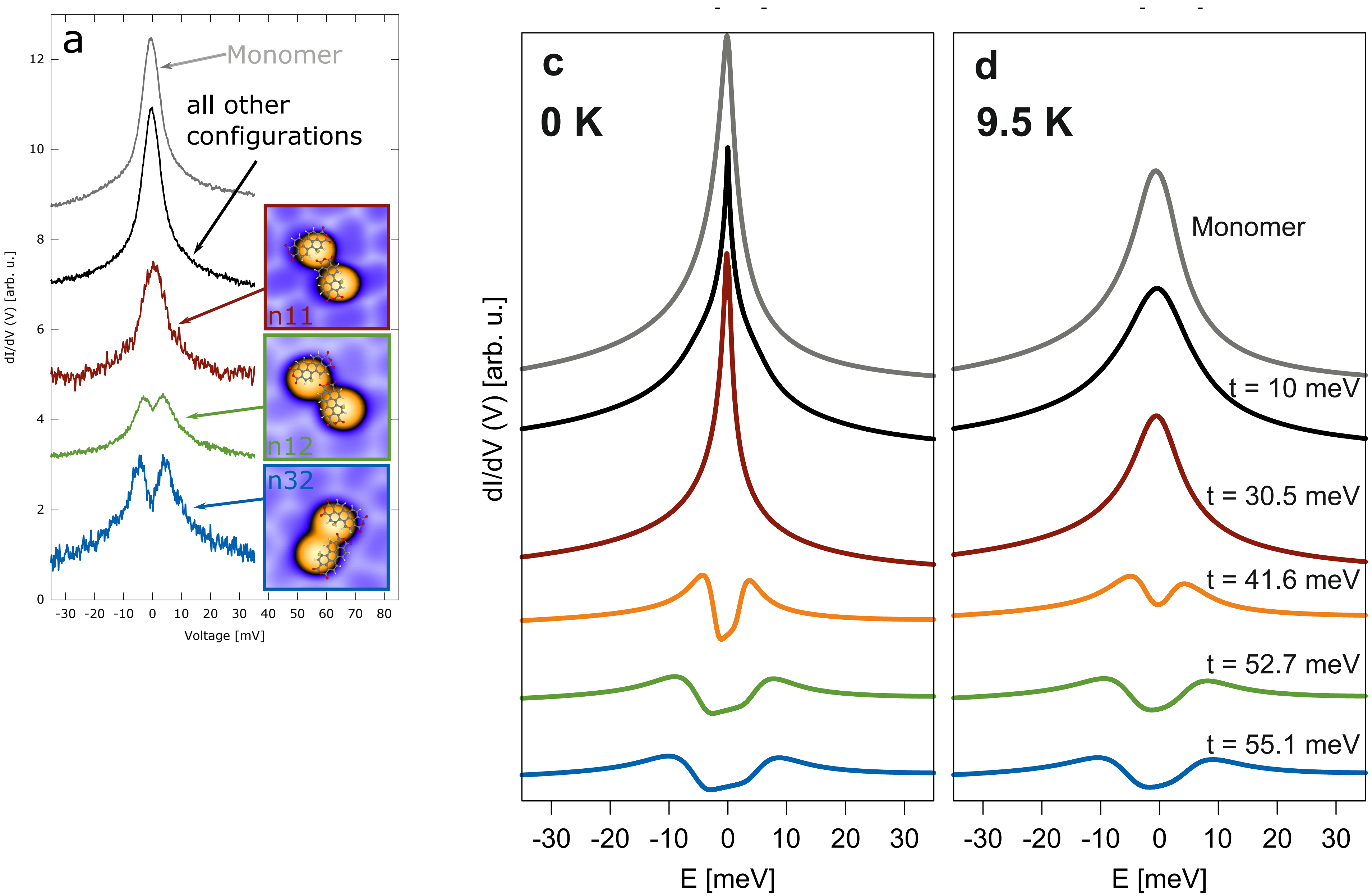
Parity conserving:

- Kondo effect: even orbital
- beyond the QCP: total collapse of Kondo resonance
- x-ray edge physics: odd orbital
- no gap!



Esat et al, Nature Physics 2016

Dimer Au/PTCDA on Au(111)



- different n_{ij}: different tunneling t
- parity breaking: gap formation

LDA+NRG calculations: excellent agreement with experiment with no adjustable parameter

Esat et al, Nature Physics 2016

Summary

- The numerical renormalization group is a powerful tool to address nano-devices in the strong coupling regime
- Combining the NRG with ab-initio methods leads to an approach with predictive power.
- The NRG can address
    - ✦ non-equilibrium dynamics
    - ✦ steady-state currents in quantum impurity systems