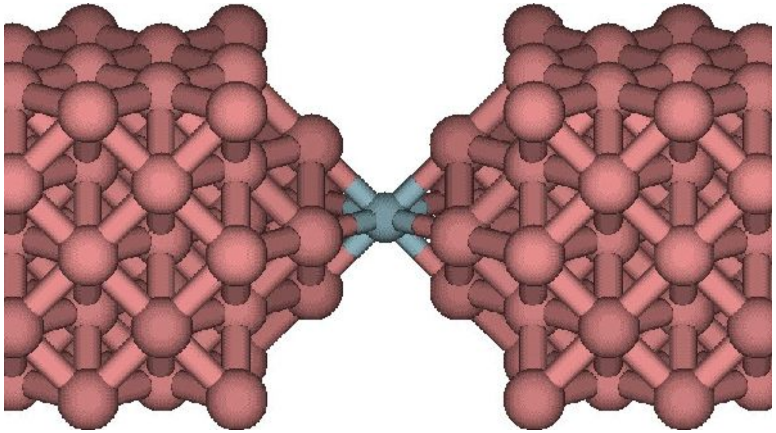
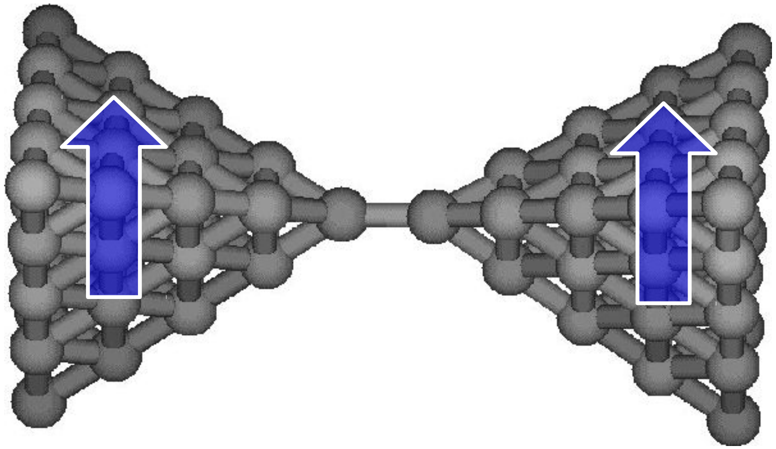
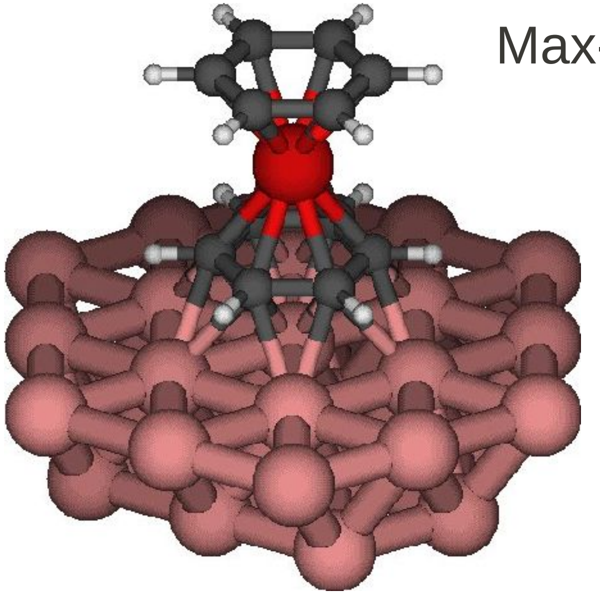
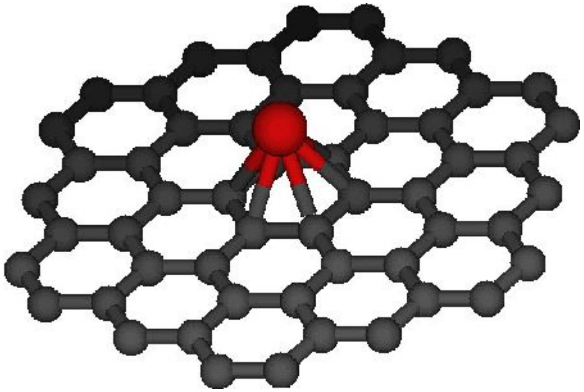



Dynamical Mean-Field Theory For Molecular Electronics

David Jacob



Max-Planck-Institut für Mikrostrukturphysik
Halle, Germany

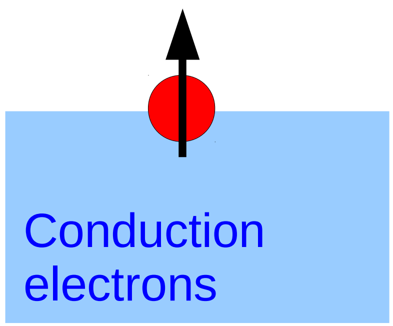


TNT2010

Braga, 7.9.2010

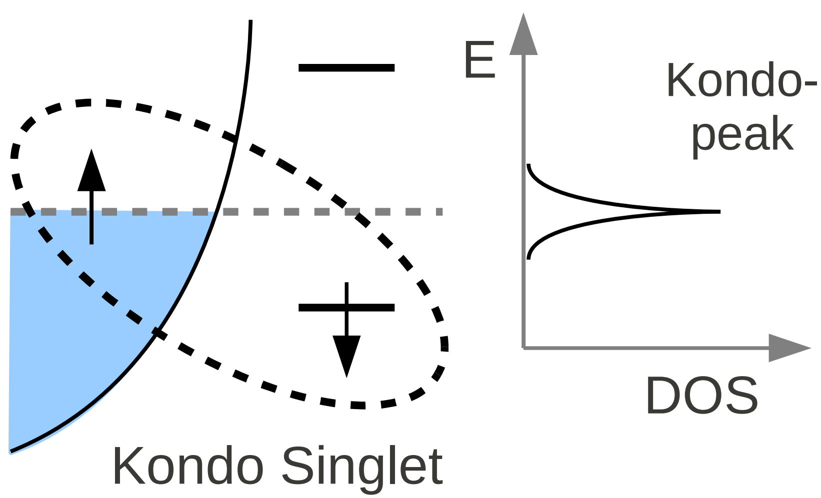
The Kondo effect in a nutshell

Anderson Impurity Model



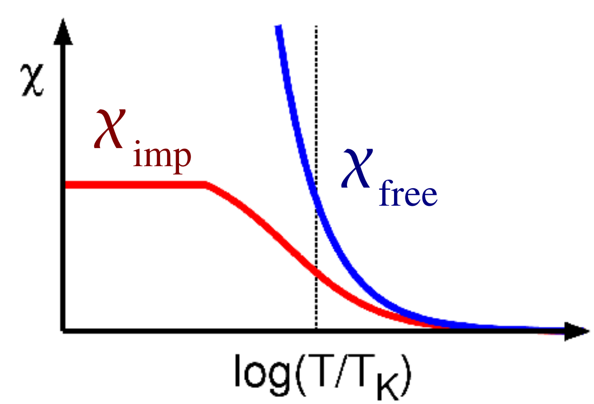
$$\hat{H}_{\text{AIM}} = \sum_{\sigma} \epsilon_d \, d^\dagger_\sigma d_\sigma + U \, d^\dagger_\uparrow d_\uparrow \, d^\dagger_\downarrow d_\downarrow + \sum_{k\sigma} \epsilon_k \, c^\dagger_{k\sigma} c_{k\sigma} + \sum_{k\sigma} (V_k \, c^\dagger_{k\sigma} d_\sigma + h.c.)$$

At low temperatures

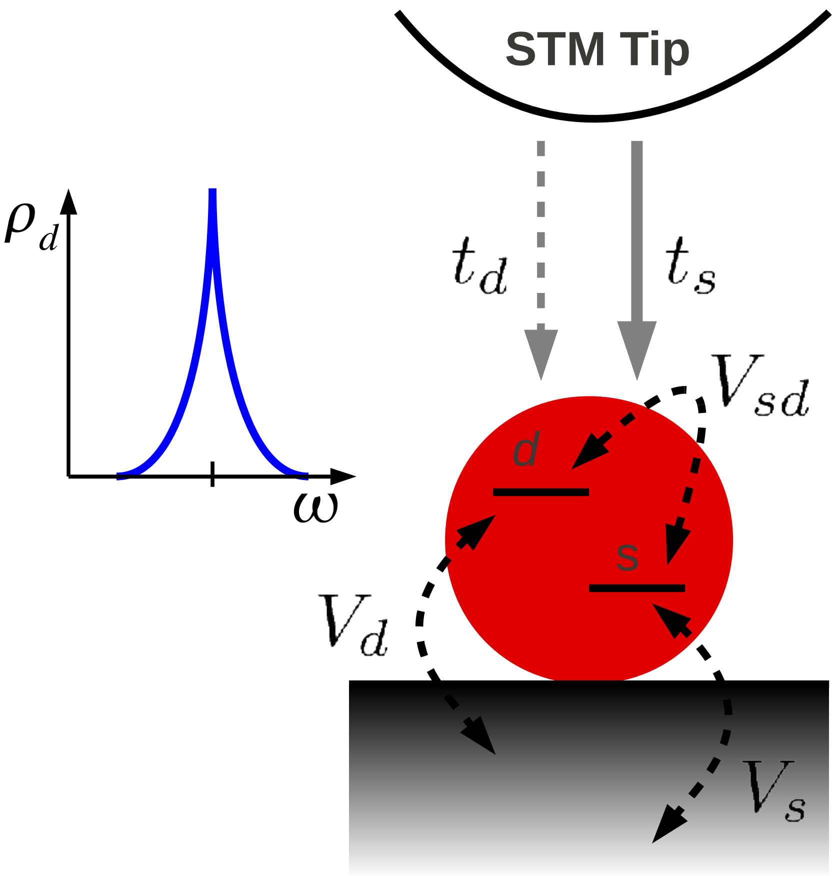


Magnetic Moment
is screened!

Magnetic Susceptibility

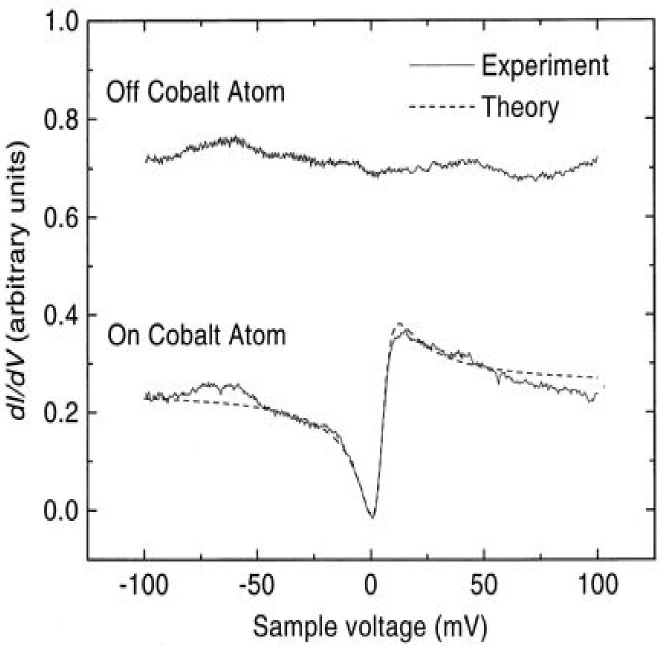


Kondo effect of Magnetic Adatoms on Metal Surfaces

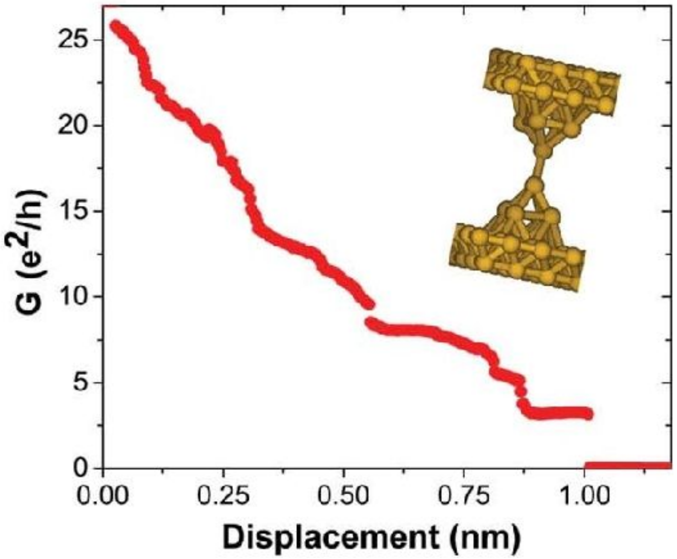


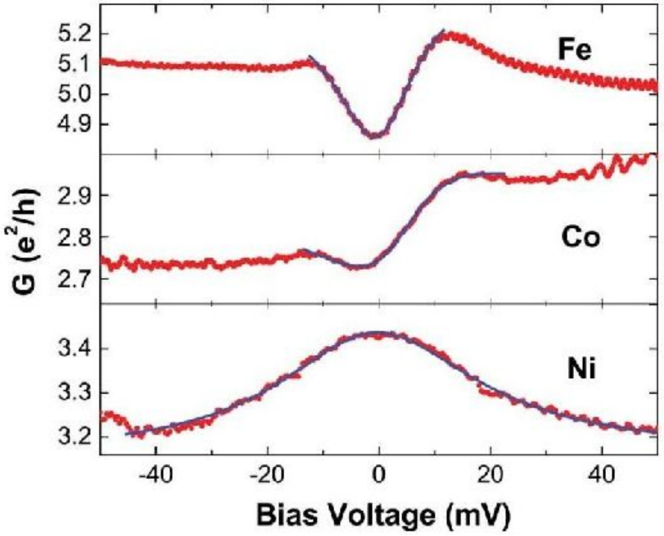
Fano formula for conductance:

$$G(V) \propto \frac{q+\epsilon}{1+\epsilon^2} \qquad \epsilon = (eV - \tilde{\epsilon}_d)/k_B T_K$$



Madhavan *et al.*, Science **280**, 567 (1998)
Schiller and Hershfield, PRB **61**, 9036 (2000)

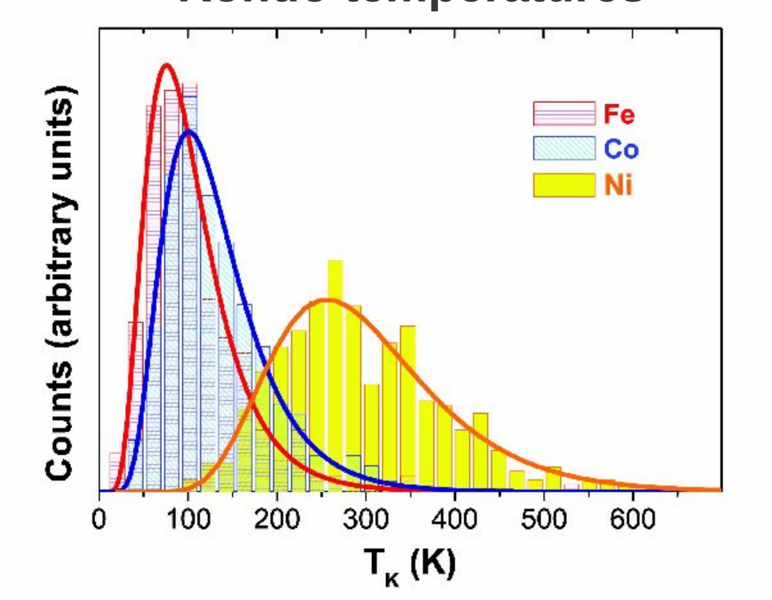




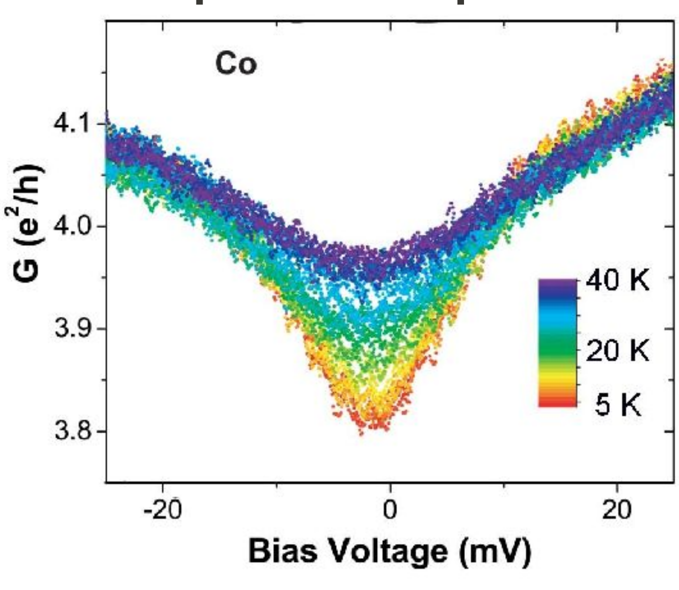
Kondo effect in *ferromagnetic* nanocontacts?

M. R. Calvo *et al*.,Nature **458**, 1150 (2009)

Kondo temperatures

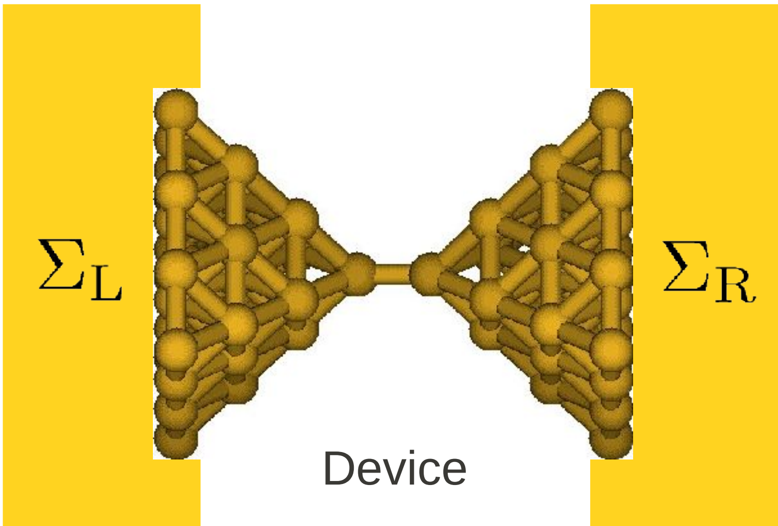


Temperature dependence



- Is it really Kondo effect?
- If yes how is it possible?
- Very interesting and rich physics
- Important questions for possible applications
- Ab initio theory needed that is able to predict strong correlation effects such as Kondo

DFT based quantum transport calculations



(1) Ab initio Density Functional Theory calculations of device and leads

(2) Lead Self-energies:

$$\mathbf{\Sigma}_{\rm L}(\omega) = \mathbf{V}_{\rm L}(\omega + \mu - \mathbf{H}_{\rm L})^{-1}\mathbf{V}_{\rm L}^{\dagger}$$

$$\mathbf{\Sigma}_{\rm R}(\omega) = \mathbf{V}_{\rm R}(\omega + \mu - \mathbf{H}_{\rm R})^{-1}\mathbf{V}_{\rm R}^{\dagger}$$

(3) Device Green's function:

$$\mathbf{G}_{\rm D}^{\rm KS}(\omega) = \frac{1}{\omega + \mu - \mathbf{H}_{\rm D}^{\rm KS} - \mathbf{\Sigma}_{\rm L}(\omega) - \mathbf{\Sigma}_{\rm R}(\omega)}$$

Landauer Transport formalism:

$$T(\omega) = {\rm Tr}\left[\Gamma_{\rm L}(\omega)(G_{\rm D}^{\rm KS})^{\dagger}(\omega)\ \Gamma_{\rm R}(\omega)G_{\rm D}^{\rm KS}(\omega)\right]$$

$$I(V) = \frac{2e}{h}\int d\omega (f_{\rm L}(\omega) - f_{\rm R}(\omega))T(\omega)$$

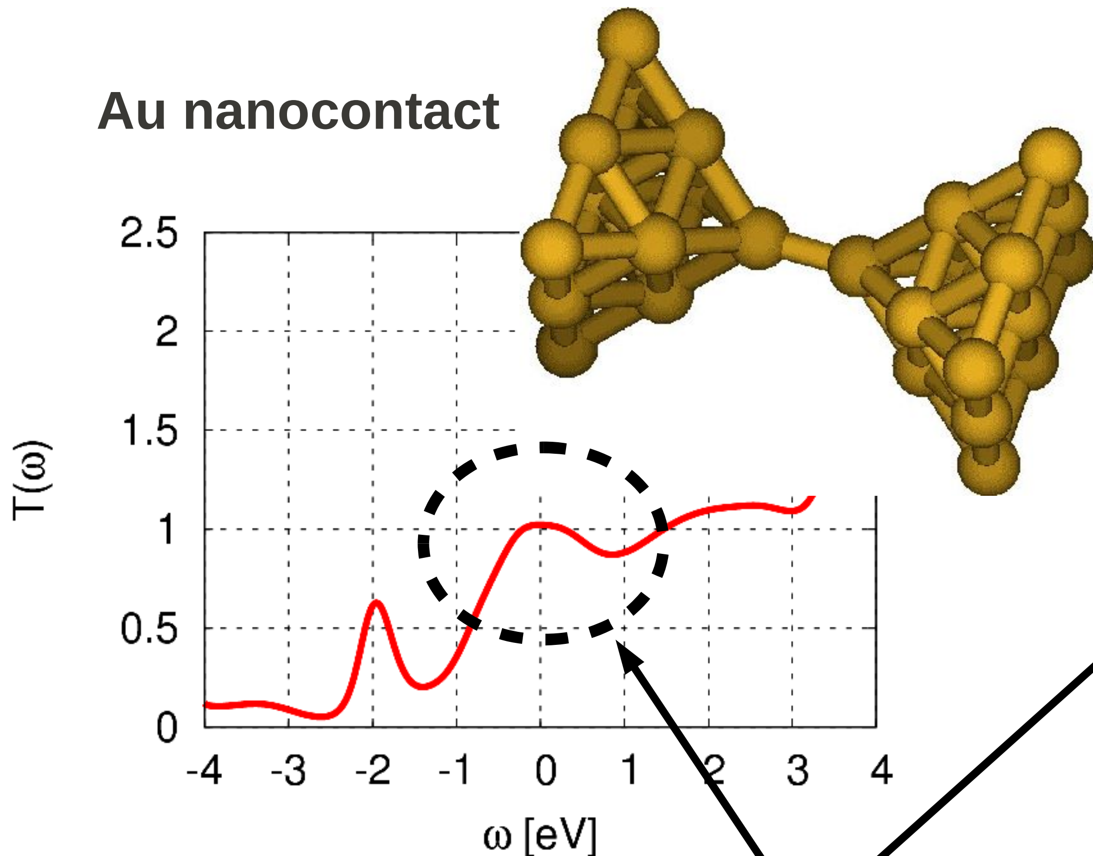
$$\mathcal{G}(V) = \frac{2e^2}{h} \times T(eV)$$

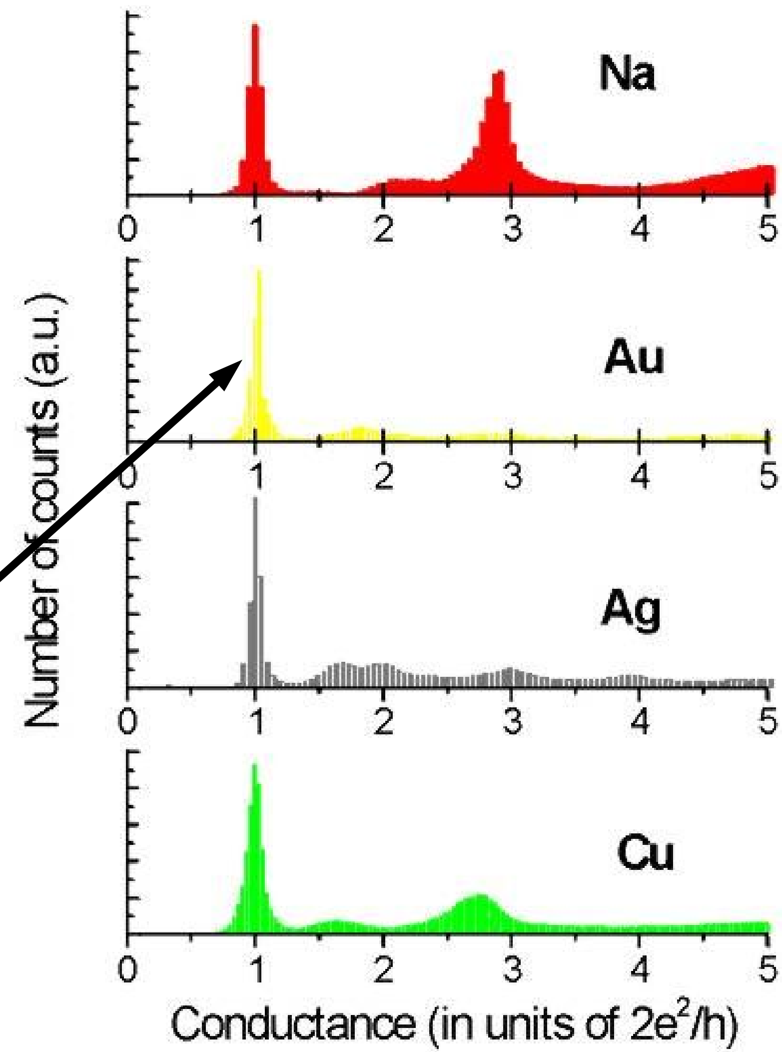
Implemented in **ALACANT** software package based on GAUSSIAN and CRYSTAL (J.J. Palacios and D. Jacob)

D. Jacob and J.J. Palacios, arXiv:1008.4538

DFT based quantum transport calculations

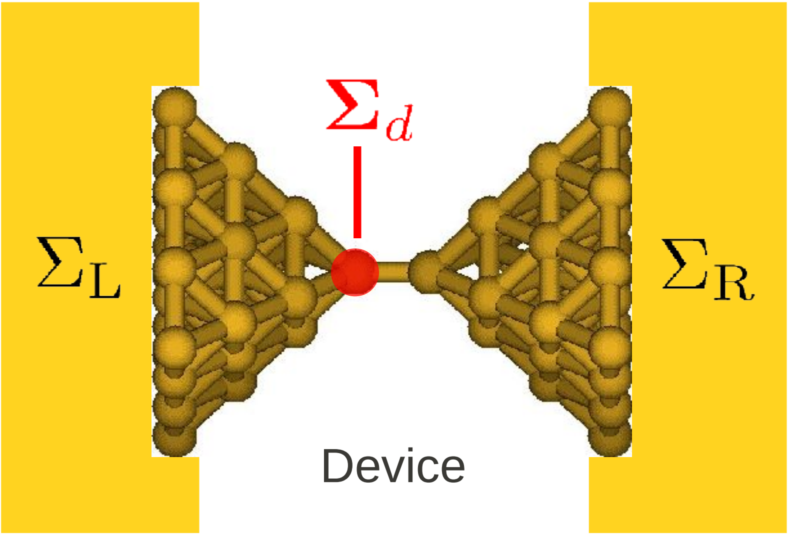
Au nanocontact





Small bias: **G ~ 1 x $2e^2/h$**

How to incorporate dynamic correlations



Starting point:

Ab initio **Density Functional Theory** calculations of **Device** and **Leads**

Correlated **Device Green's function:**

$$\mathbf{G}_{\mathrm{D}}(\omega) = \frac{1}{\omega + \mu - \mathbf{H}_{\mathrm{D}}^{\mathrm{KS}} - \mathbf{\Sigma}_{\mathrm{L}}(\omega) - \mathbf{\Sigma}_{\mathrm{R}}(\omega) - \mathbf{\Sigma}_{\mathbf{d}}(\omega)}$$

Conductance and current in general: Meir-Wingreen

$$I(V) = \frac{1}{2} \int d\omega \, \mathrm{Tr} \left[(\Gamma_{\mathrm{L}} - \Gamma_{\mathrm{R}}) \mathbf{G}_{\mathrm{D}}^{<} + (f_{\mathrm{L}} \Gamma_{\mathrm{L}} - f_{\mathrm{R}} \Gamma_{\mathrm{R}}) \mathbf{A}_{\mathrm{D}} \right]$$

For small bias and low temperature: Landauer

$$\mathcal{G}(V) = \frac{2e^2}{h} \times T(eV)$$

$$T(\omega) = \mathrm{Tr} \left[\Gamma_{\mathrm{L}}(\omega) G_{\mathrm{D}}^{\dagger}(\omega) \; \Gamma_{\mathrm{R}}(\omega) G_{\mathrm{D}}(\omega) \right]$$

$$I(V) = \frac{2e}{h} \int d\omega (f_{\mathrm{L}}(\omega) - f_{\mathrm{R}}(\omega)) T(\omega)$$

How to calculate the Self-Energy: OCA Impurity solver

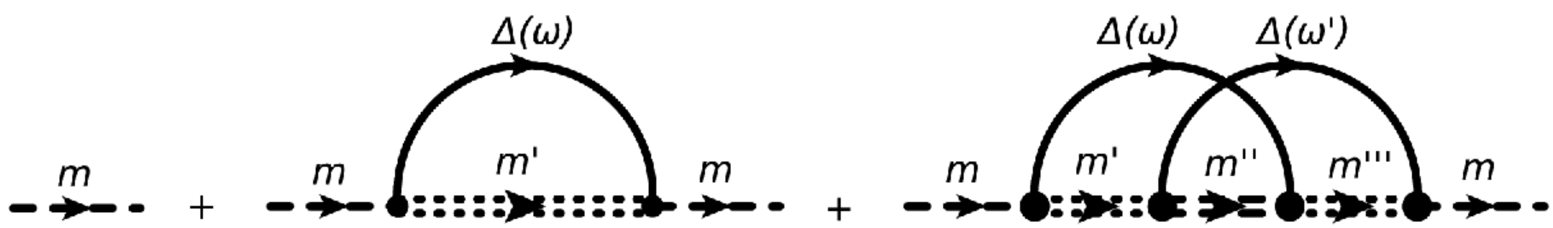
$$\mathcal{H}_{3d} = \sum_i \epsilon_d^{(i)} \hat{n}_d^{(i)} + \sum_{ijkl\sigma\sigma'} U_{ijkl} d^\dagger_{i\sigma} d^\dagger_{j\sigma'} d_{k\sigma'} d_{l\sigma} \xrightarrow[\text{Diagonalization}]{\text{Exact}} \sum_m E_m |m\rangle\langle m|$$

Many-body eigenstates $|m\rangle$ ⟶ **Pseudo Particles** $\hat{a}^\dagger_m, \hat{a}_m$

Hybridization function: $\mathbf{\Delta}_d(\omega) = \omega + \mu - \mathbf{H}_d - [\mathbf{P}_d \mathbf{G}^{\mathrm{KS}}_{\mathrm{D}}(\omega) \mathbf{P}_d]^{-1}$

Perturbation Expansion in *Hybridization Strength*:

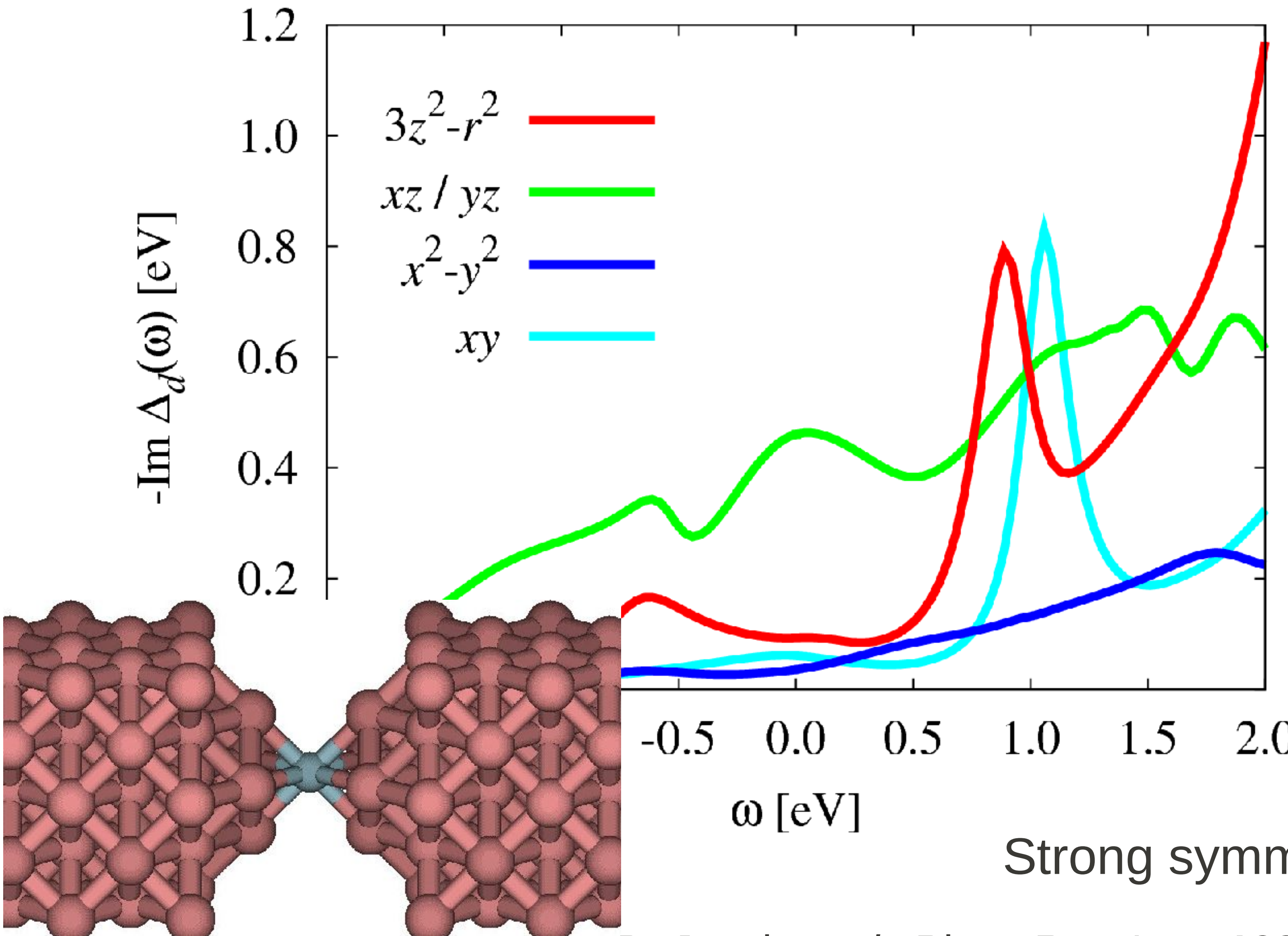
$$G_m(\omega) = (\omega + \mu - E_m - \Sigma_m(\omega))^{-1} =$$



K. Haule *et al.*, PRB **64**, 155111 (2001)

G. Kotliar *et al.*, RMP **78**, 865 (2006)

Magnetic impurity in Cu nanocontact
Hybridization function

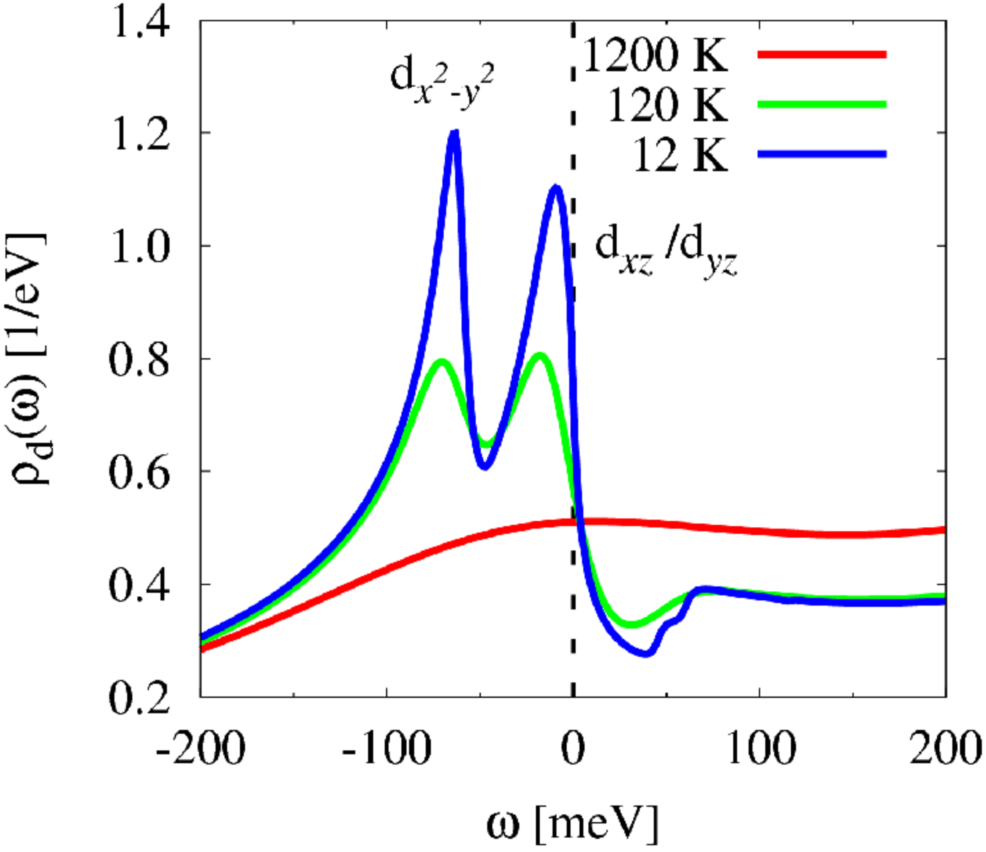


Strong symmetry breaking!

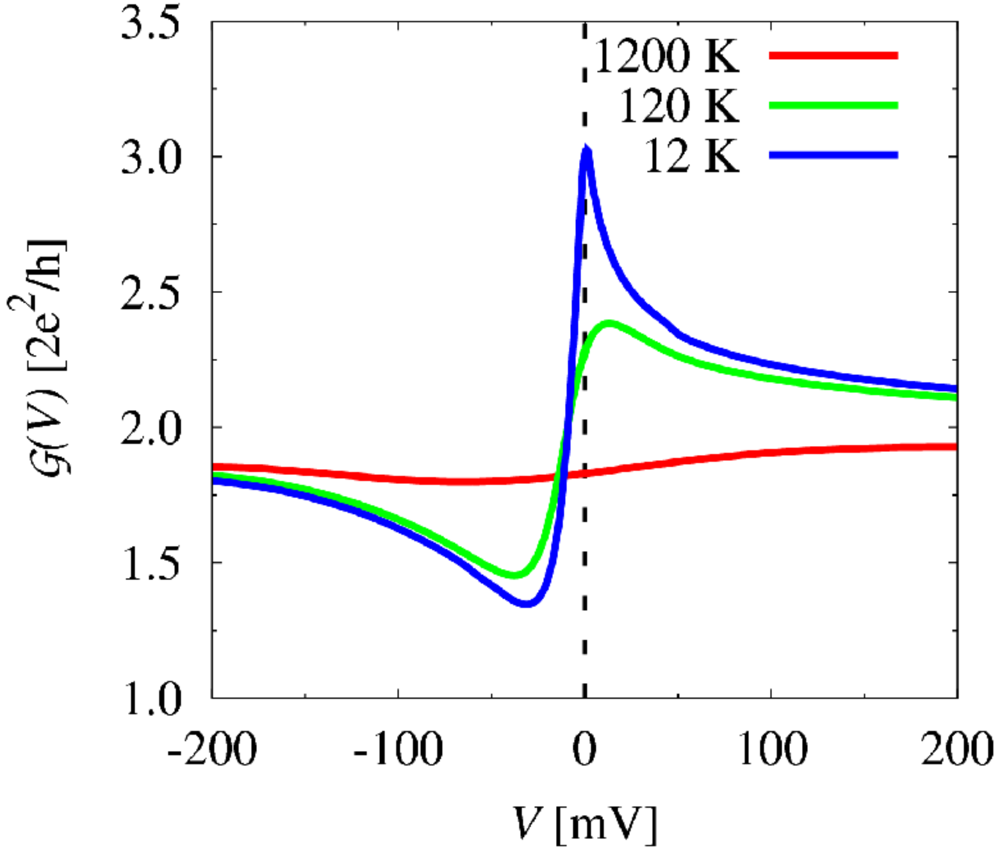
D. Jacob *et al.*, Phys. Rev. Lett. **103**, 016803 (2009)

Results: Co impurity

PDOS



Conductance



Occupation of $d_{xz}+d_{yz}$ = 3 => Spin 1/2

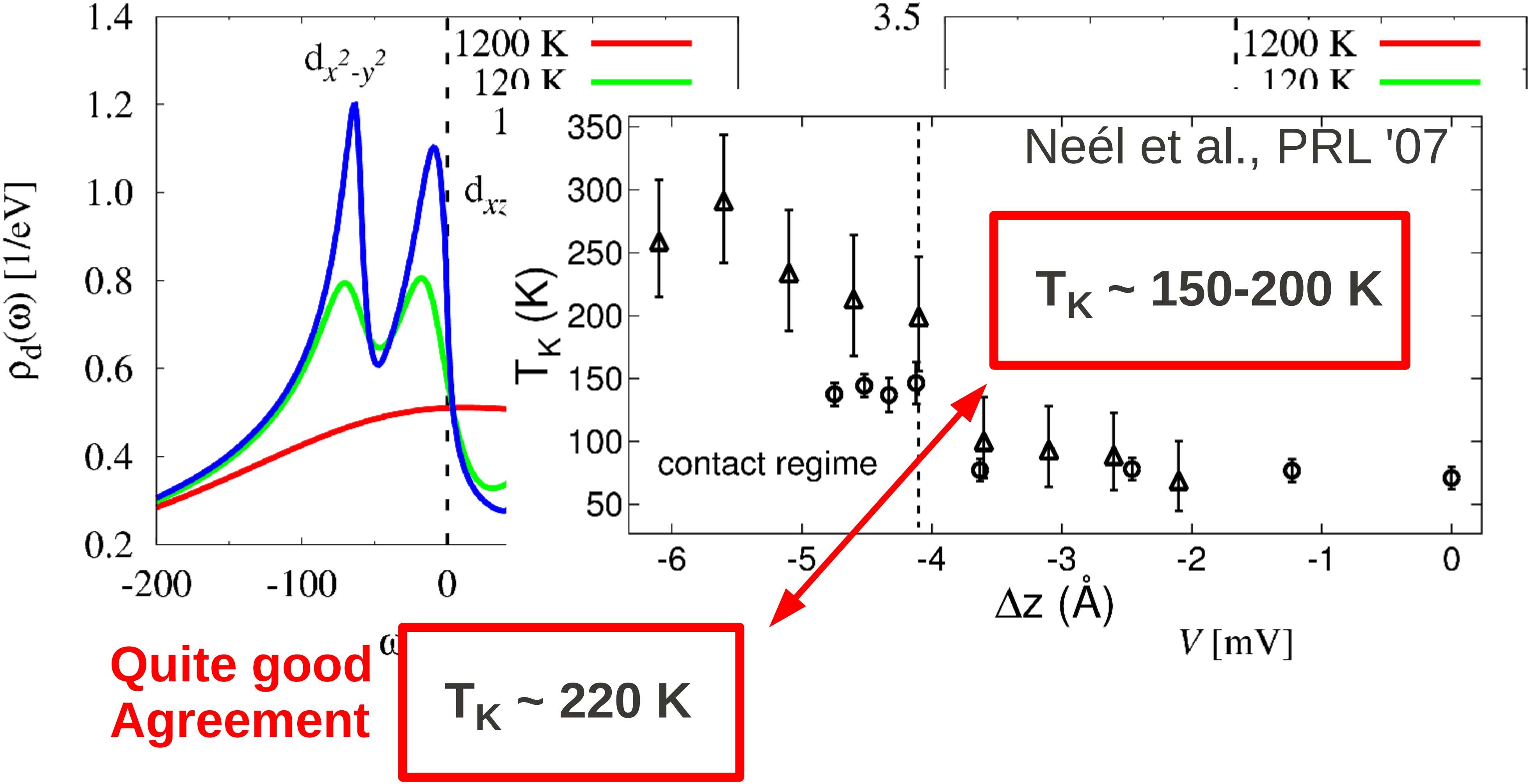
U = 5 eV and J = 1eV

D. Jacob *et al.*, Phys. Rev. Lett. **103**, 016803 (2009)

Results: Co impurity

PDOS

Conductance



Quite good Agreement

D. Jacob *et al.*, Phys. Rev. Lett. **103**, 016803 (2009)

Molecular DMFT:

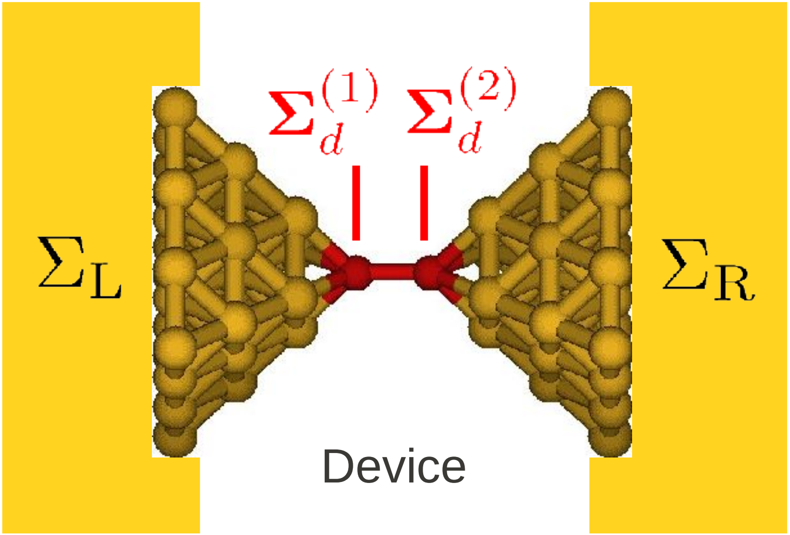
Dynamical Mean-Field Theory for nanoscopic conductors

D. Jacob *et al.*, arXiv:1009.0523

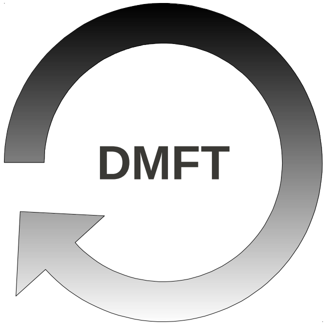
LDA Calc.

$\mathbf{H}_{\mathrm{D}}^{\mathrm{KS}}$

DMFT SCC:
$\mathbf{g}_d^{(i)}(\omega) = \mathbf{P}_d^{(i)} \mathbf{G}_{\mathrm{D}}(\omega) \mathbf{P}_d^{(i)}$



$\mathbf{\Sigma}_d^{(i)}$

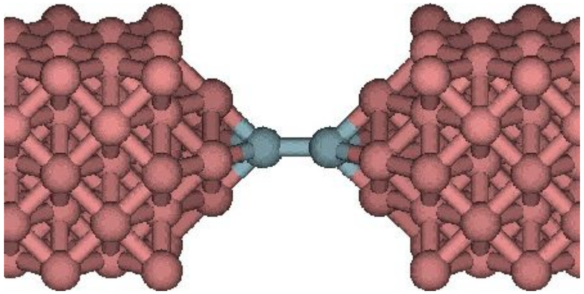


$$\mathbf{\Delta}_d^{(i)}(\omega) = \omega + \mu - \mathbf{H}_d^{(i)} - [\mathbf{g}_d^{(i)}(\omega)]^{-1} - \mathbf{\Sigma}_d^{(i)}(\omega)$$

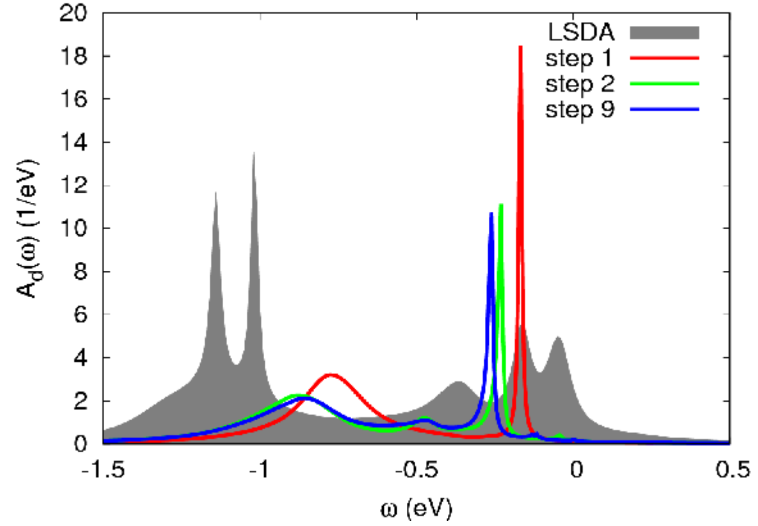
Imp. Solver
(OCA)

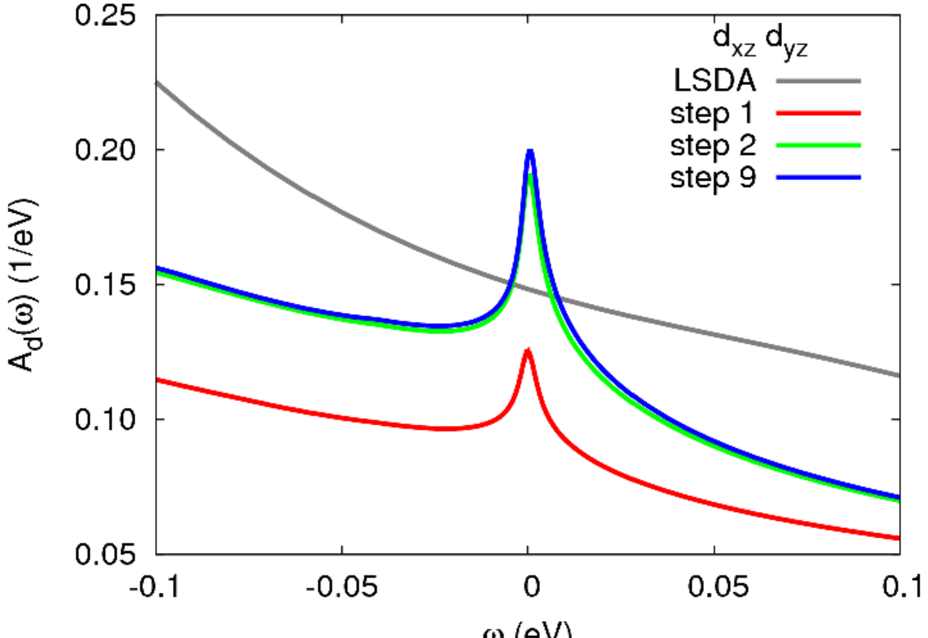
$$\mathbf{G}_{\mathrm{D}}(\omega) = \frac{1}{\omega + \mu - \mathbf{H}_{\mathrm{D}}^{\mathrm{KS}} - \mathbf{\Sigma}_{\mathrm{L}}(\omega) - \mathbf{\Sigma}_{\mathrm{R}}(\omega) - \sum_i \mathbf{\Sigma}_d^{(i)}(\omega)}$$

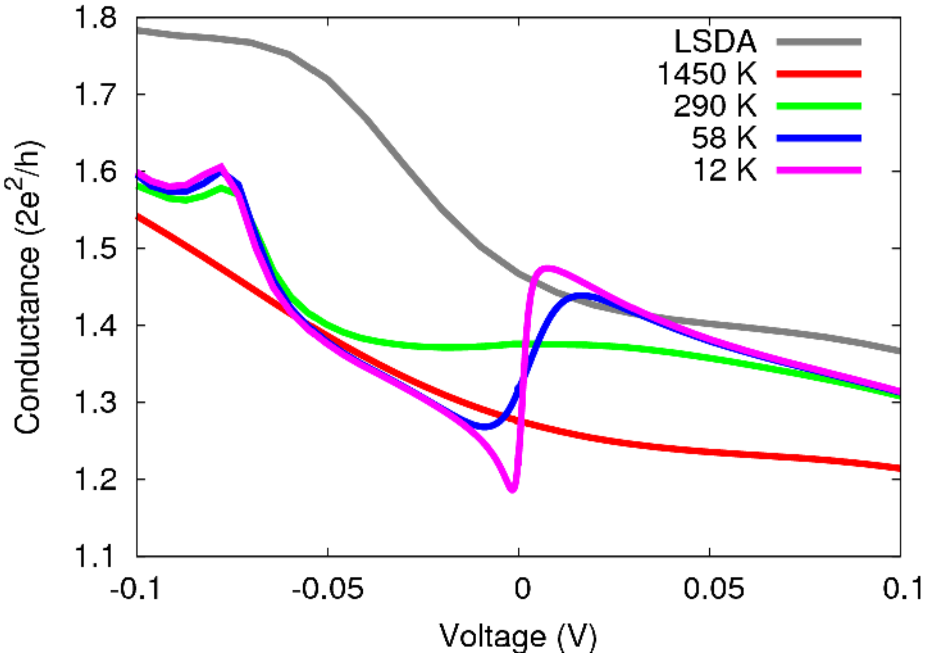
Two Ni atoms in Cu nanocontact



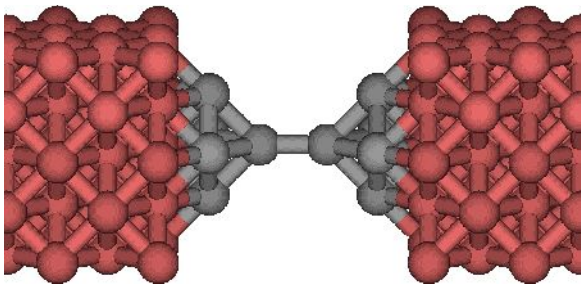
D. Jacob *et al*., arXiv:1009.0523



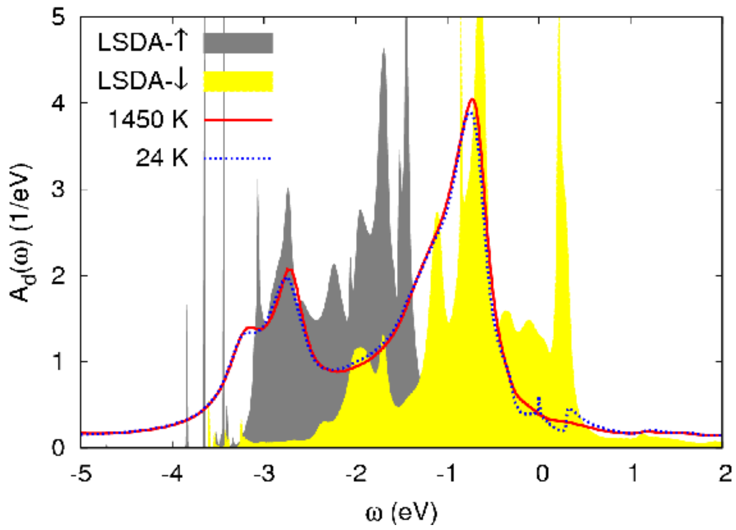


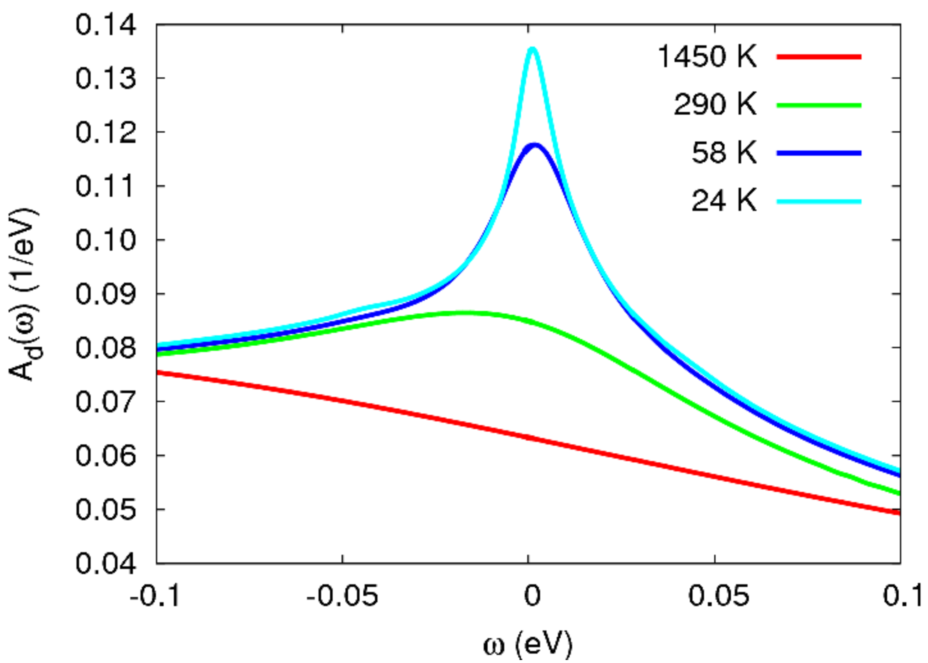


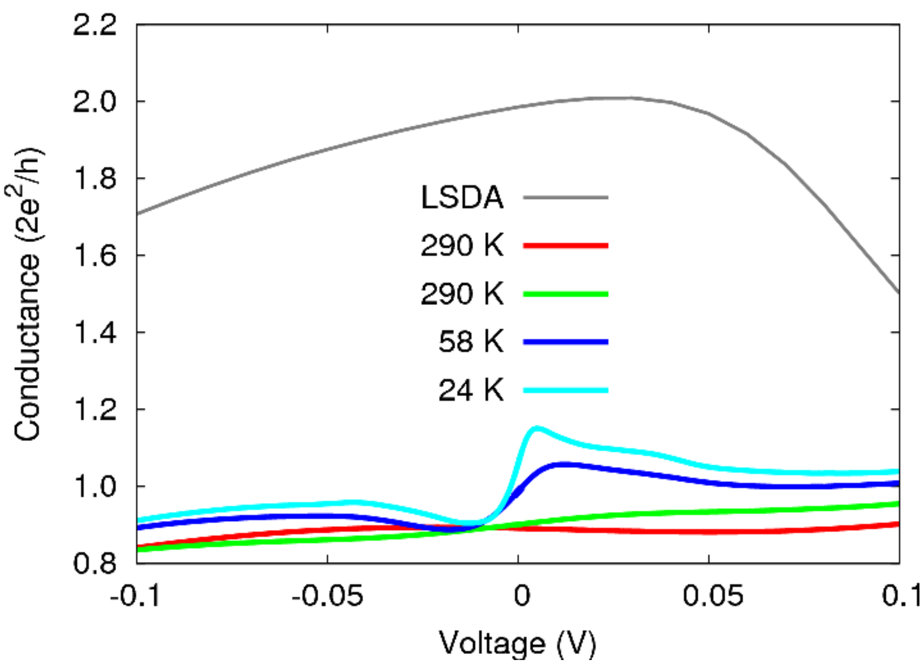
Ni nanocontact between Cu wires



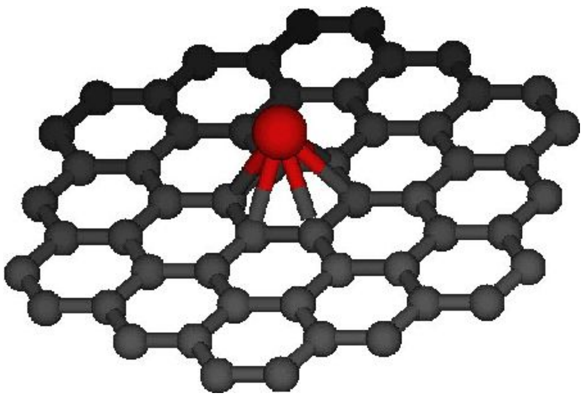
D. Jacob *et al.*, arXiv:1009.0523

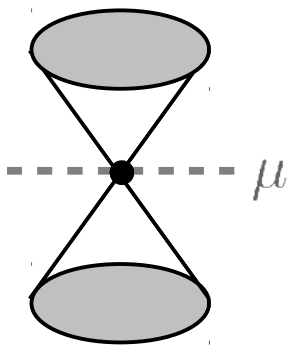




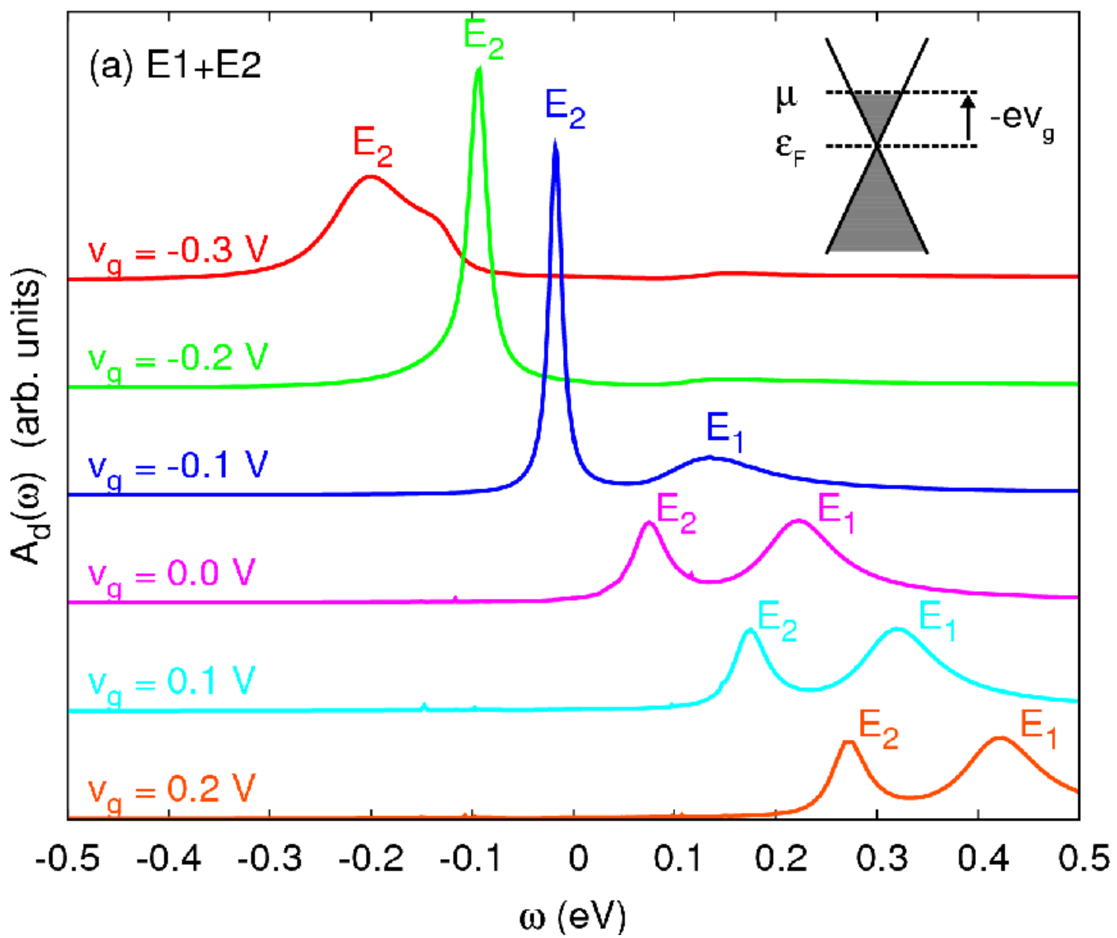


Graphene + Co

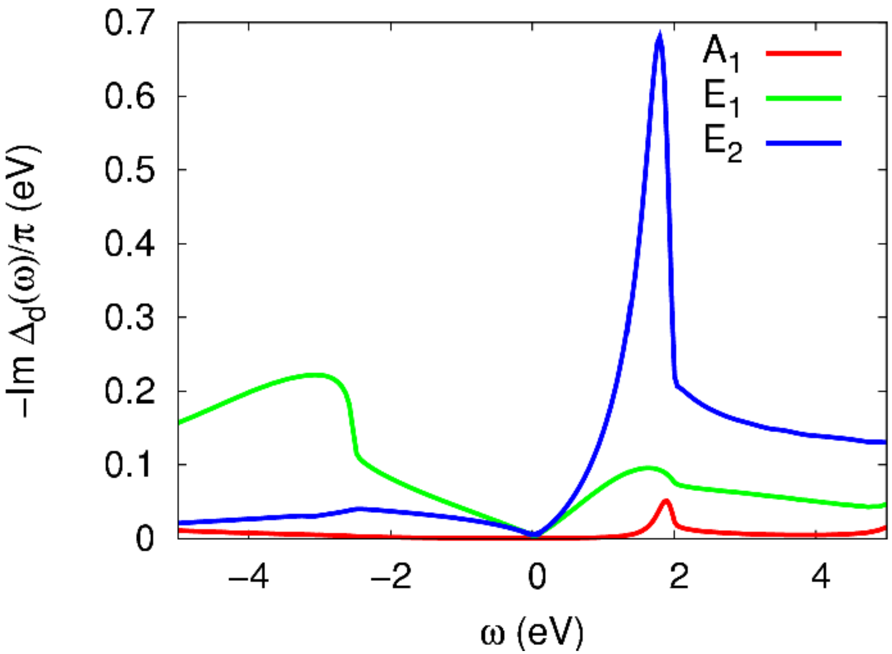




Gate-dependence of 3d-spectra



Hybridization functions



D. Jacob and G. Kotliar, PRB **82**, 085423 (2010)

Conclusions

- Molecular DMFT
- Dynamic Correlations incorporated into ab initio quantum transport
- Kondo effect in nanoscopic conductors from first principles

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- G. Kotliar (Rutgers U)
- K. Haule (Rutgers U)
- J. J. Palacios (UAM)
- J. Fernández-Rossier (UA)
- C.Untiedt (UA)
- M.R. Calvo (Imperial College)
- E. K. U. Gross (MPI Halle)

References:

D. Jacob *et al*., arXiv:1009.0523

D. Jacob *et al.*, Phys. Rev. Lett. **103**, 016803 (2009)

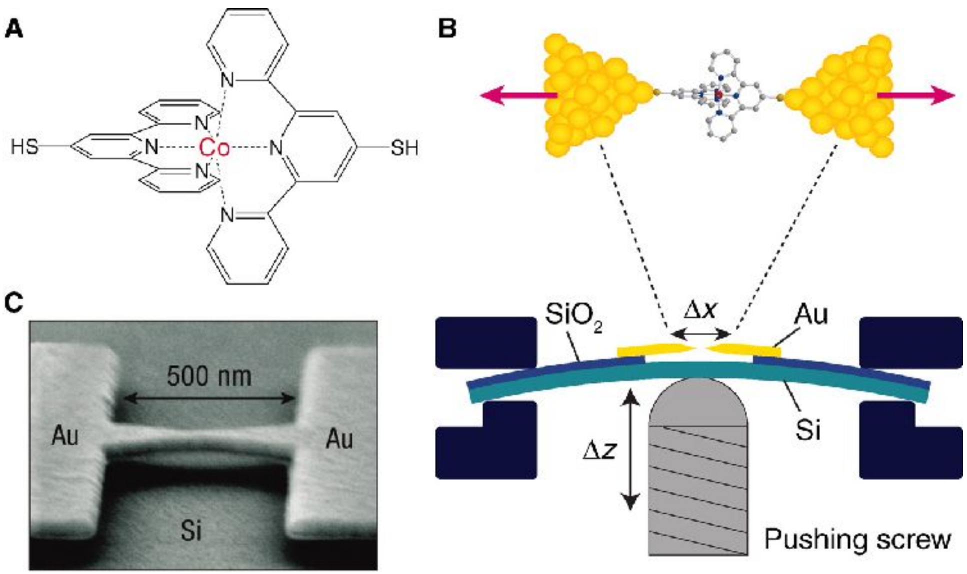
M. R. Calvo *et al.*, Nature **458**, 1150 (2009)

ALACANT Software: www.alacant.dfa.ua.es

D. Jacob and G. Kotliar, Phys. Rev. B **82**, 085423 (2010)

THANK YOU!!!!

Kondo effect in Spin-1 Molecules



Parks et al., Science **328**, 1370 (2010)