Kourovka-35

Correlation Effects in Nanosystems

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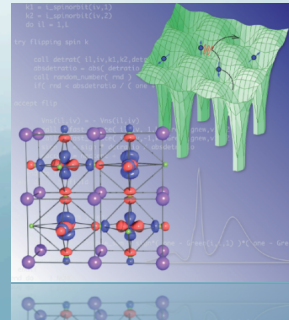
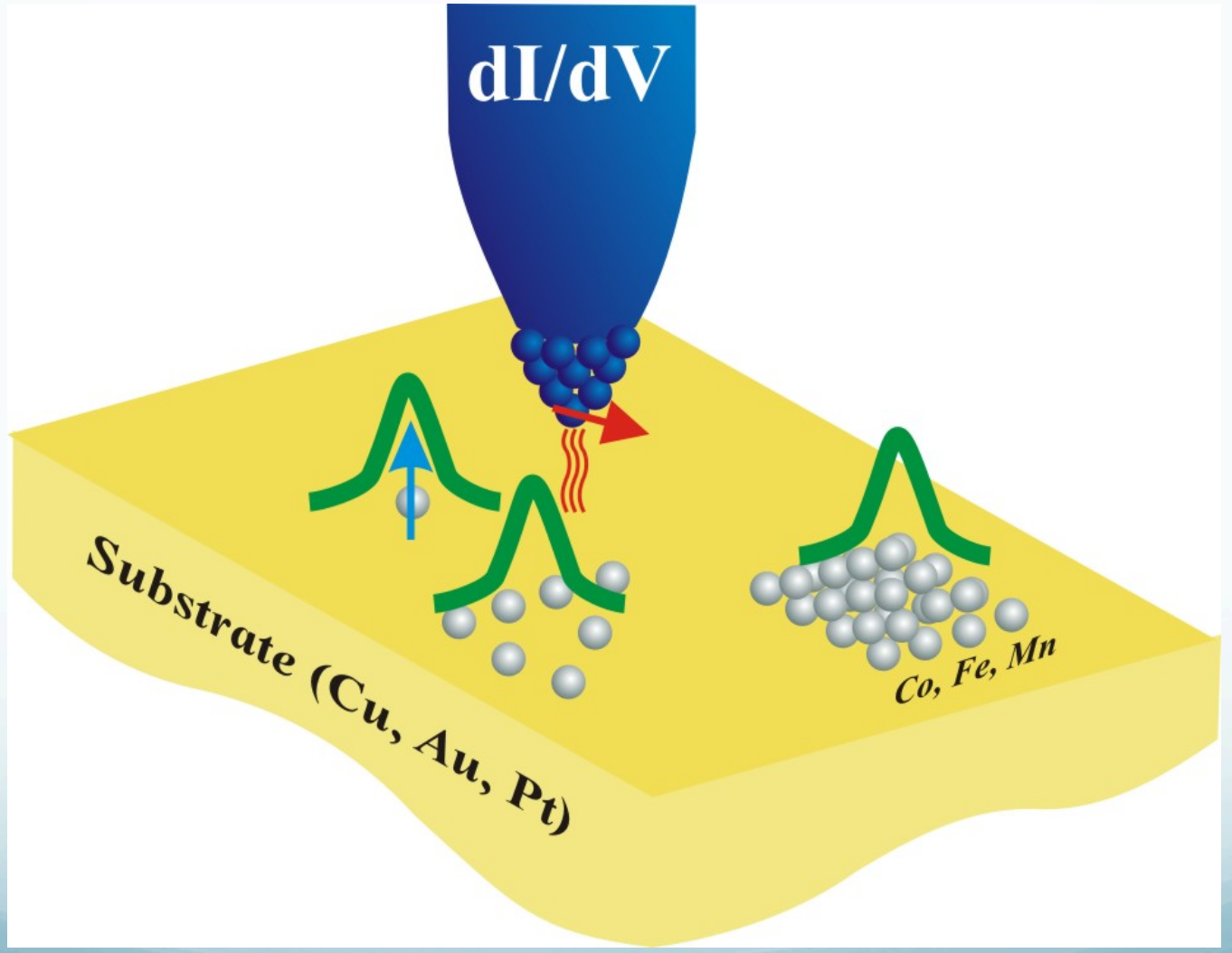


Outline

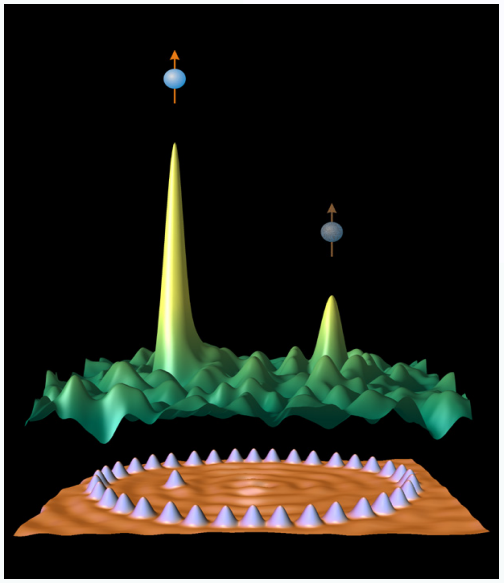
- Magnetic nanosystems
- Path Integral for Impurity Model
- Numericallly Exact Impurity Solver: CT-QMC
- Transition Metal adatom on Surface
- Summary

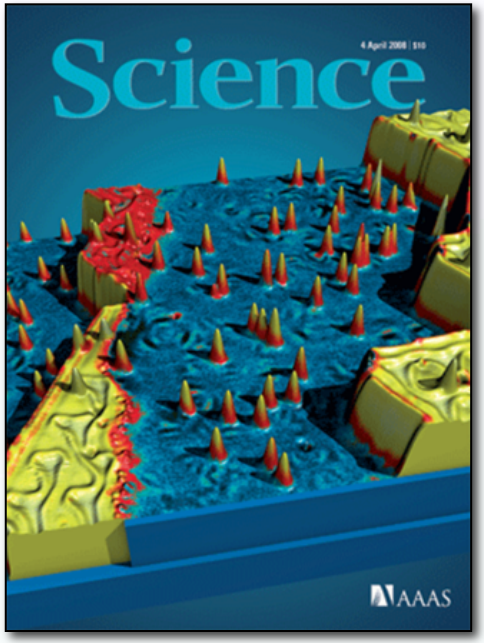
STM: Quantum Tools for Nanosystems



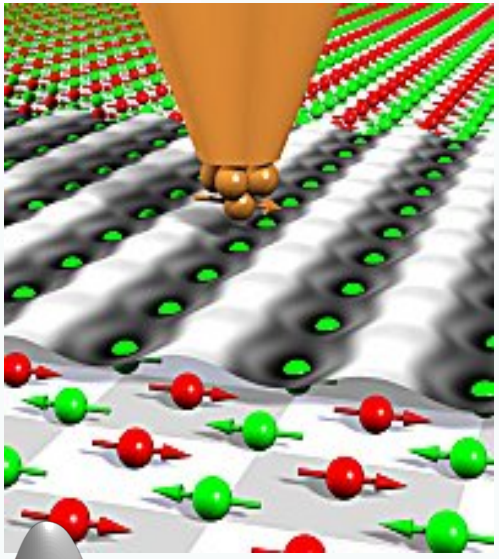
STM and magnetic nanostructures

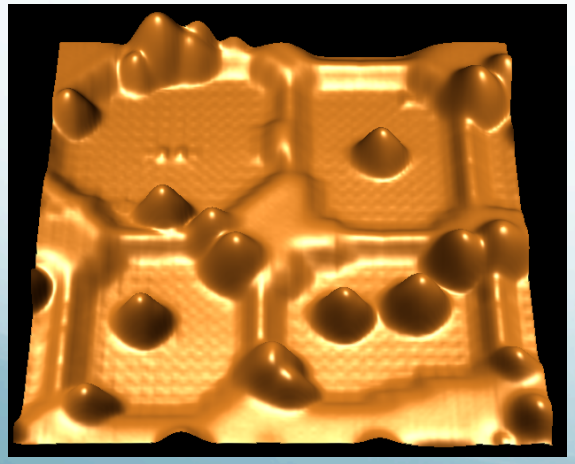
IBM-Almaden

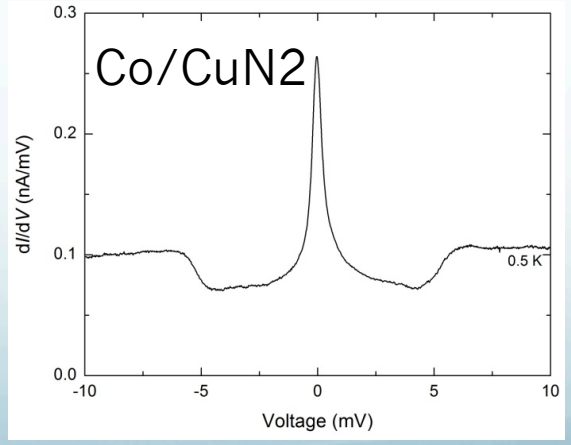


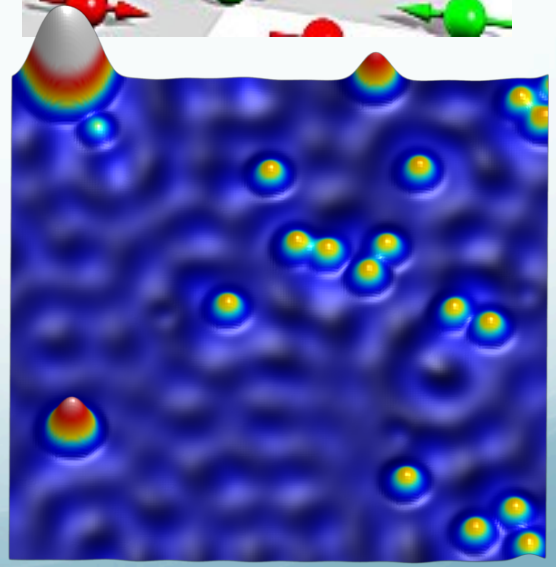


UH-Hamburg

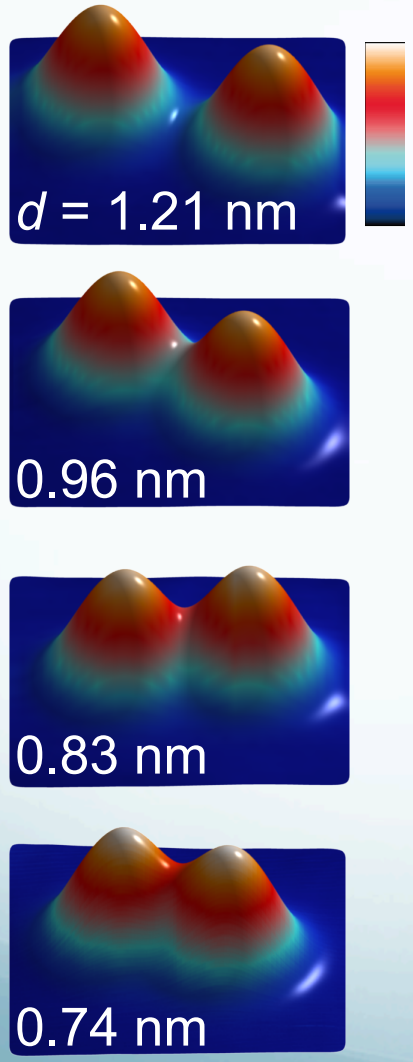


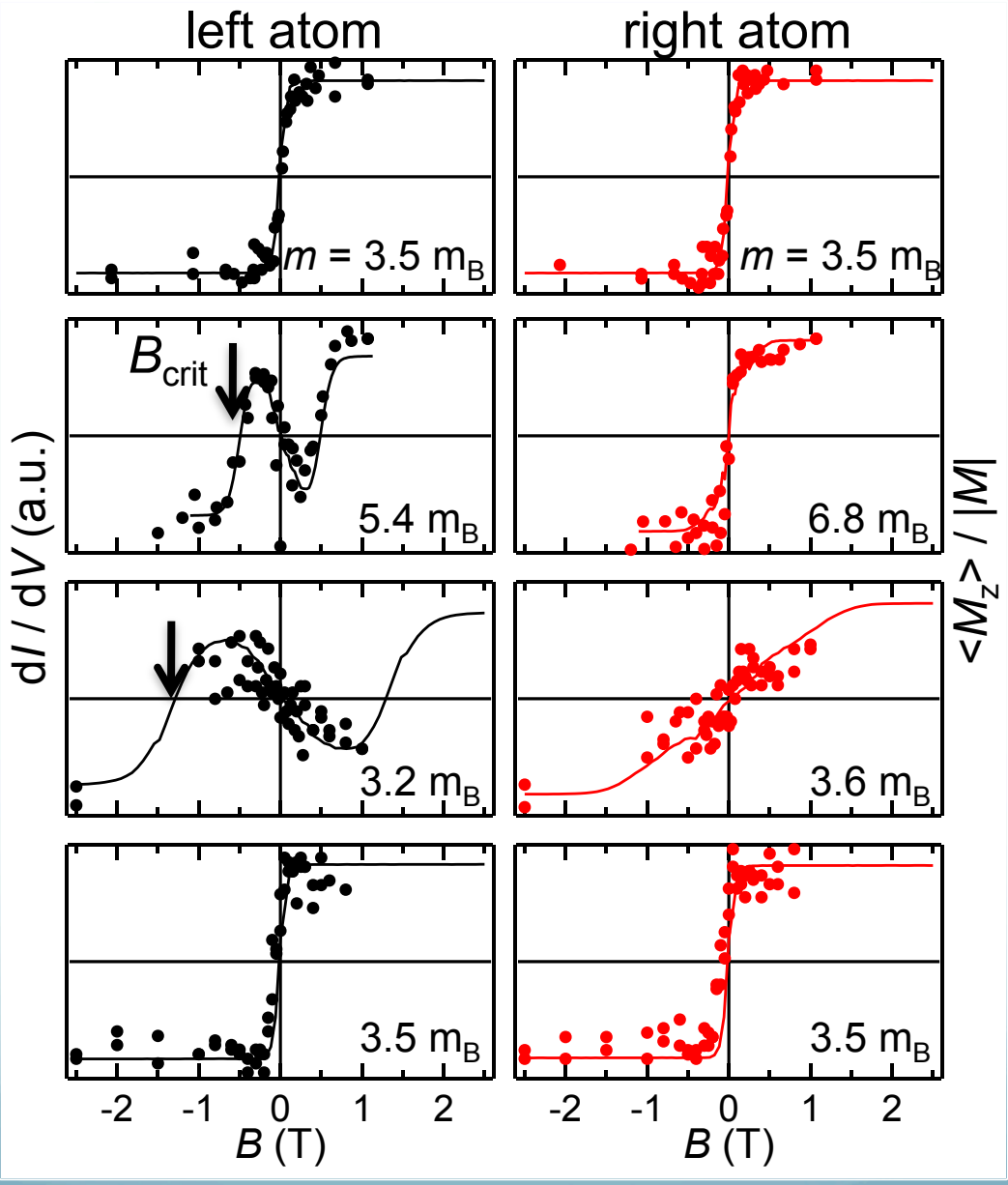


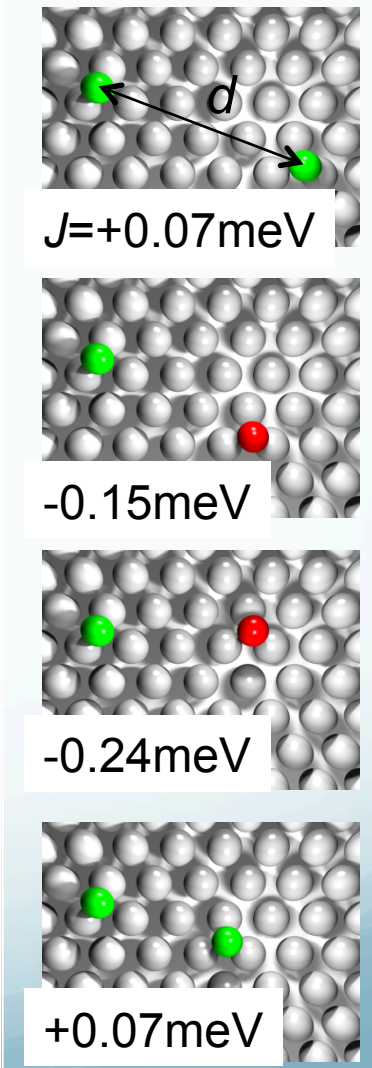




Single Atom Magnetometry
R. Wiesendanger (Uni Hamburg)

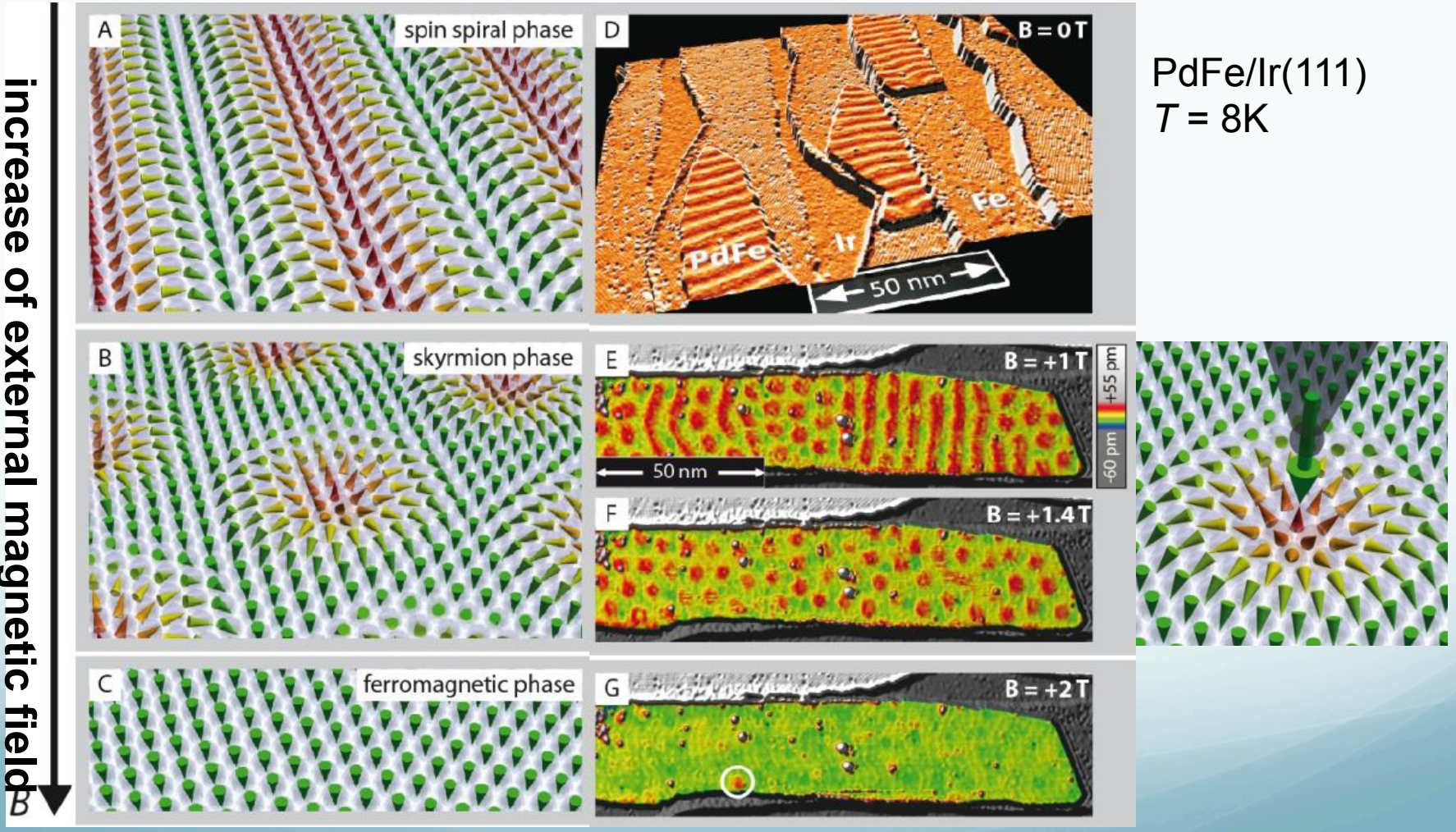






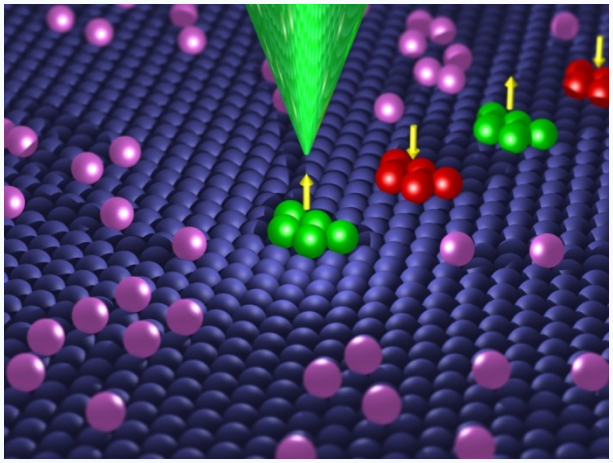
Control of nano-magnetism by STM

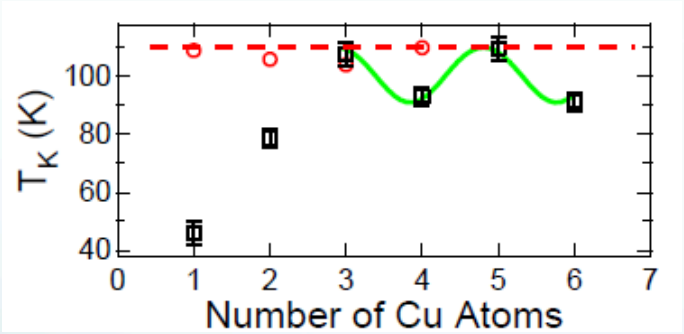
UH-Hamburg (R. Wiesendanger)

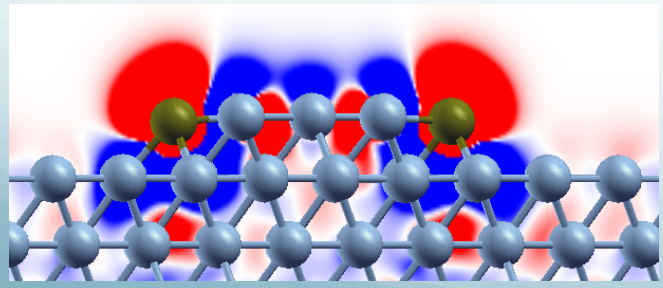


PdFe/Ir(111)
T = 8K

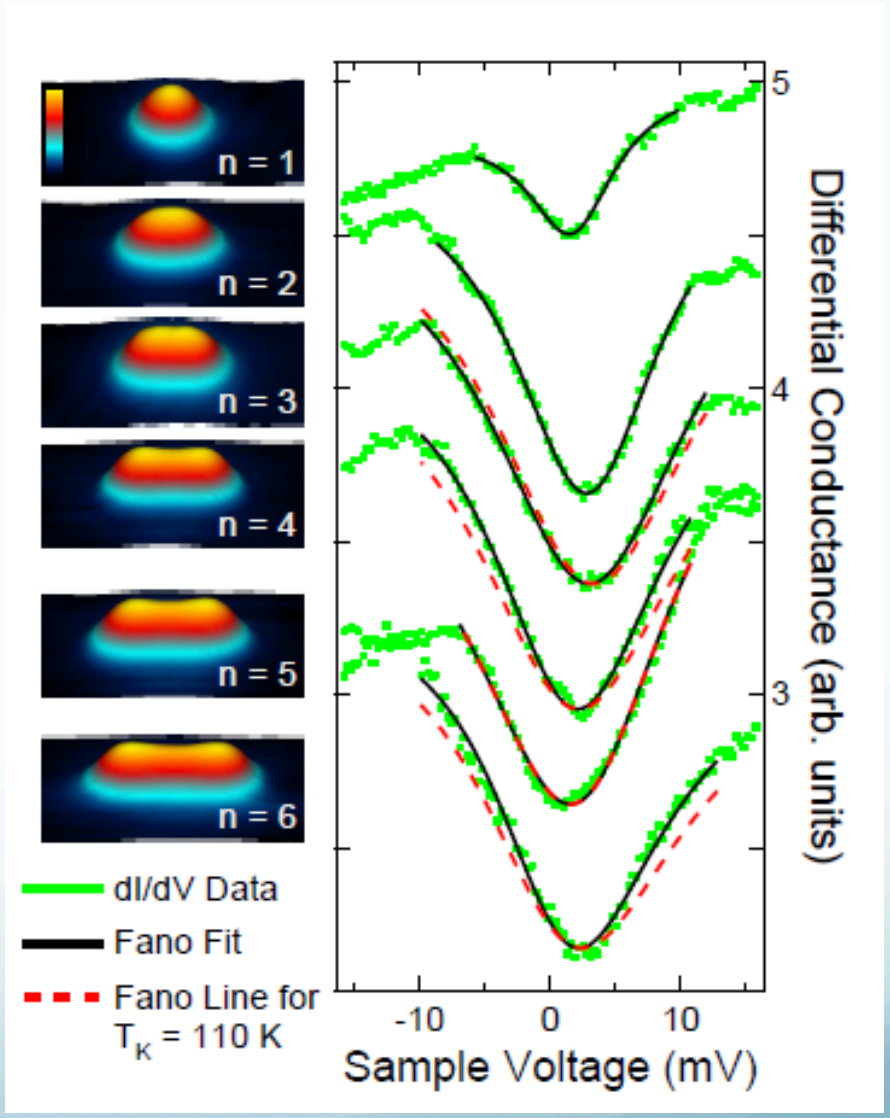
Non-Local correlations: STM







$CoCu_nCo$ on Cu(111)



N. Neél, et al, PRL. 107, 106804 (2011)

Path Integrals for Fermions

from A. Kamenev "Field Theory of Non-Equilibrium Systems" (Cambridge, 2011)

Single Fermion second-quantization operators

$$\begin{aligned} \hat{c}_i|0\rangle &= 0 \\ \hat{c}_i|1\rangle &= |0\rangle \\ \hat{c}_i^+|0\rangle &= |1\rangle \\ \hat{c}_i^+|1\rangle &= 0 \end{aligned}$$

$$\begin{aligned} \hat{c}^+\hat{c}|n\rangle &= n|n\rangle \\ \hat{c}^2 &= 0 \\ (\hat{c}^+)^2 &= 0 \\ \{\hat{c},\hat{c}^+\} &= \hat{1} \end{aligned}$$

Algebra of Grassmann anti-commuting numbers: $(\hat{c}_i^+, \hat{c}_i) \to (c_i^*, c_i)$

$$\begin{aligned} c_i c_j &= -c_j c_i \\ c_i^2 &= 0 \\ f(c) &= f_0 + f_1 c \\ f(c^*, c) &= f_{00} + f_{10}c^* + f_{01}c + f_{11}c^* c \end{aligned}$$

Grassmann numbers anticommute with fermionic operators

$$\{c, \hat{c}\} = \{c, \hat{c}^+\} = 0$$

Grassmann calculus

Differentiation: $$\frac{\partial c_i}{\partial c_j} = \delta_{ij}$$

N.B. order: $$\frac{\partial}{\partial c_2} c_1 c_2 = -c_1$$

Example: $$f(c^*, c) = f_{00} + f_{10}c^* + f_{01}c + f_{11}c^*c$$

$$\frac{\partial}{\partial c^*}\frac{\partial}{\partial c} f(c^*, c) = \frac{\partial}{\partial c^*}(f_{01} - f_{11}c^*) = -f_{11} = -\frac{\partial}{\partial c}\frac{\partial}{\partial c^*} f(c^*, c)$$

Integration: $$\int 1 dc = 0$$
$$\int c dc = 1$$

equivalent to differetition

$$\int \ldots dc \rightarrow \frac{\partial}{\partial c} \ldots$$

Coherent State

Eigenstate of annihilation operator

$$\hat{c}\,|c\rangle \;=\; c\,|c\rangle$$

Diefinition of coherent states

$$|c\rangle \;=\; e^{-c\hat{c}^{+}}\,|0\rangle = (1 - c\hat{c}^{+})\,|0\rangle = |0\rangle - c\,|1\rangle$$

Proof

$$\hat{c}\,|c\rangle \;=\; \hat{c}\,(|0\rangle - c\,|1\rangle) = -\hat{c}c\,|1\rangle = c\,|0\rangle = c\,|c\rangle$$

Left Coherent State: c^{*} is just another Grassman number

$$\begin{aligned}
\langle c|\,\hat{c}^{+} &= \langle c|\,c^{*} \\
\langle c| &= \langle 0|\,e^{-\hat{c}c^{*}} = \langle 0|\,(1 - \hat{c}c^{*}) = \langle 0| - \langle 1|\,c^{*} \\
\langle c|\,\hat{c}^{+} &= (\langle 0| - \langle 1|\,c^{*})\,\hat{c}^{+} = -\langle 1|\,c^{*}\hat{c}^{+} = \langle 0|\,c^{*} = \langle c|\,c^{*}
\end{aligned}$$

Unity operator in coherent states

Overlap of Coherent States (non-orthogonal)

$$\langle c^* | c \rangle = (\langle 0| - \langle 1| c^*)(|0\rangle - c|1\rangle) = 1 + c^* c = e^{c^* c}$$

Resolution of Unity

$$\hat{1} = \int\int dc^* dc e^{-c^* c} |c\rangle \langle c|$$

Proof

$$\begin{aligned} \int\int dc^* dc e^{-c^* c} |c\rangle \langle c| &= \int\int dc^* dc (1 - c^* c)(|0\rangle - c|1\rangle)(\langle 0| - \langle 1| c^*) \\ &= -\int\int dc^* dc c^* c (|0\rangle \langle 0| + |1\rangle \langle 1|) = \hat{1} \end{aligned}$$

Trace of Fermionic Operators

Matrix elements of normally ordered operators

$$\langle c^* | \widehat{H}(\hat{c}^+, \hat{c}) | c \rangle = H(c^*, c) \langle c^* | c \rangle = H(c^*, c) e^{c^* c}$$

Trace-formula

$$\begin{aligned} Tr\left(\hat{O}\right) &= \sum_{n=0,1} \langle n | \hat{O} | n \rangle = \sum_{n=0,1} \int\int dc^* dc e^{-c^* c} \langle n | c \rangle \langle c | \hat{O} | n \rangle = \\ &= \int\int dc^* dc e^{-c^* c} \sum_{n=0,1} \langle -c | \hat{O} | n \rangle \langle n | c \rangle = \int\int dc^* dc e^{-c^* c} \langle -c | \hat{O} | c \rangle \end{aligned}$$

"Minus" due to commutation Left and Right coherent state

$$c^* c = -c c^*$$

$$|-c\rangle = |0\rangle + c\,|1\rangle$$

Path Integral for Fermions

Partition function

$$Z = \lim_{N\to\infty} \int \Pi_{i=1}^{2N} dc_i^* dc_i e^{-\sum_i c_i^* c_i} \left\langle -c_1 \left| \rho_0 \right| c_{2N} \right\rangle \cdots \left\langle c_i \left| \widehat{U}_{-\Delta t} \right| c_{i-1} \right\rangle \cdots$$
$$\cdots \left\langle c_{N+2} \left| \widehat{U}_{-\Delta t} \right| c_{N+1} \right\rangle \left\langle c_{N+1} \left| \widehat{1} \right| c_N \right\rangle \left\langle c_N \left| \widehat{U}_{\Delta t} \right| c_{N+1} \right\rangle \cdots \left\langle c_2 \left| \widehat{U}_{\Delta t} \right| c_1 \right\rangle$$

Using $$\left\langle c_i \left| \widehat{U}_{\pm\Delta t} \right| c_{i-1} \right\rangle \approx e^{c_i^* c_{i-1}} e^{\mp i H(c_i^* c_{i-1})\Delta t}$$

We get for real-time

$$Z = \lim_{N\to\infty} \int \Pi_{i=1}^{2N} dc_i^* dc_i e^{iS(c^* c)}$$

$$S(c^* c) = \Delta t \sum_{i=2}^{2N} \left[i c_i^* \frac{c_i - c_{i-1}}{\Delta t} - H(c_i^* c_{i-1}) \right] + i c_1^* \left(\rho_0 c_{2N} + c_1 - c_{N+1} \right)$$

$$S\left[c^* c\right] = \int_C dt \left[c^*(t) i \partial_t c(t) - H\left(c^*(t), c(t)\right) \right]$$

Gaussian Path Integrals

Only one analytical path integral:

$$Z\left[J^*, J\right] = \int\int \prod_{i=1}^{N} \left[dc_i^* dc_i\right] e^{-\sum_{i,j=1}^{N} c_i^* M_{ij} c_j + \sum_{i=1}^{N}\left[c_i^* J_i + J_i^* c_i\right]} = \det\left[M\right] e^{-\sum_{i,j=1}^{N} J_i^* M_{ij}^{-1} J_j}$$

Short notation

$$\int D\left[c^* c\right] e^{-c^* M c} = \det M$$

Proof - "det": expand the exponent only N-th oder is non-zero

$$e^{-\sum_{i,j=1}^{N} c_i^* M_{ij} c_j} = \frac{\left(-\sum_{i,j=1}^{N} c_i^* M_{ij} c_j\right)^N}{N!}$$

Permutations of c_i^* and c_j gives $\det M$

Examples:

N=1

$$\int D\left[c^* c\right] e^{-c_1^* M_{11} c_1} = \int D\left[c^* c\right] \left(-c_1^* M_{11} c_1\right) = M_{11} = \det M$$

N=2

$$\int D\left[c^* c\right] e^{-c_1^* M_{11} c_1 - c_1^* M_{12} c_1 - c_2^* M_{21} c_1 - c_2^* M_{22} c_2} =$$

$$\frac{1}{2!} \int D\left[c^* c\right] \left(-c_1^* M_{11} c_1 - c_1^* M_{12} c_1 - c_2^* M_{21} c_1 - c_2^* M_{22} c_2\right)^2 =$$

$$M_{11} M_{22} - M_{12} M_{21} = \det M$$

Correlation Function: U=0

Change of variables

$$c \to c - M^{-1} j$$

Using: $$c^* M c - c^* j - j^* c = \left(c^* - j^* M^{-1}\right) M \left(c - M^{-1} j\right) - j^* M^{-1} j$$

Single-particle correlation function:

$$\left\langle c_i c_j^* \right\rangle = \frac{1}{Z\left[0,0\right]} \frac{\delta Z\left[J^*, J\right]}{\delta J_j \delta J_i^*}|_{J=0} = M_{ij}^{-1}$$

Two-particle correlation function:

$$\left\langle c_i c_j c_k^* c_l^* \right\rangle = \frac{1}{Z\left[0,0\right]} \frac{\delta Z\left[J^*, J\right]}{\delta J_l \delta J_k \delta J_j^* \delta J_i^*}|_{J=0} = M_{il}^{-1} M_{jk}^{-1} - M_{ik}^{-1} M_{jl}^{-1}$$

Path Integral for Everything

Euclidean action

$$
\begin{aligned}
Z &= \int \mathcal{D}[c^*, c] e^{-S} \\
S &= \sum_{12} c_1^* \left(\partial_\tau + t_{12}\right) c_2 + \frac{1}{4} \sum_{1234} c_1^* c_2^* U_{1234} c_4 c_3
\end{aligned}
$$

One- and two-electron matrix elements:

$$
\begin{aligned}
t_{12} &= \int d\mathbf{r}\, \phi_1^*(\mathbf{r}) \left(-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \mu \right) \phi_2(\mathbf{r}) \\
U_{1234} &= \int d\mathbf{r} \int d\mathbf{r}'\, \phi_1^*(\mathbf{r}) \phi_2^*(\mathbf{r}')\, U(\mathbf{r} - \mathbf{r}')\, \phi_3(\mathbf{r}) \phi_4(\mathbf{r}')
\end{aligned}
$$

Shot notation:

$$
\sum_1 \ldots \equiv \sum_{im} \int d\tau \ldots
$$

One- and Two-particle Green Functions

One-particle Green function

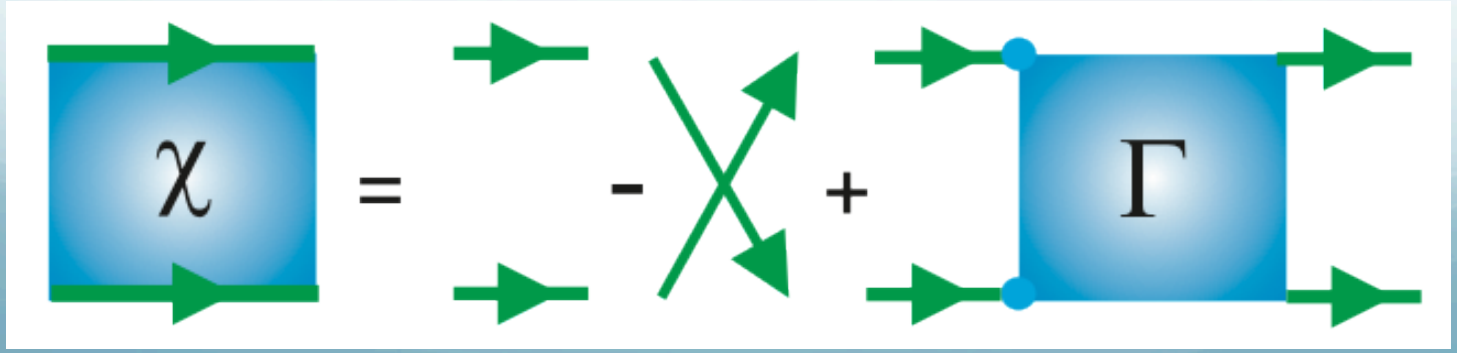
$$G_{12} = -\langle c_1 c_2^* \rangle_S = -\frac{1}{Z} \int \mathcal{D}[c^*, c]\, c_1 c_2^*\, e^{-S}$$

Two-particle Green function (generalized susceptibilities)

$$\chi_{1234} = \langle c_1 c_2 c_3^* c_4^* \rangle_S = \frac{1}{Z} \int \mathcal{D}[c^*, c]\, c_1 c_2 c_3^* c_4^*\, e^{-S}$$

Vertex function:

$$X_{1234} = G_{14}G_{23} - G_{13}G_{24} + \sum_{1'2'3'4'} G_{11'}G_{22'}\Gamma_{1'2'3'4'}G_{3'3}G_{4'4}$$



Baym-Kadanoff functional

Source term $S[J] = S + \sum_{ij} c_i^* J_{ij} c_j$

Partition function and Free-energy:

$$Z[J] = e^{-F[J]} = \int \mathcal{D}[c^*, c]\, e^{-S[J]}$$

Legendre transforming from J to G:

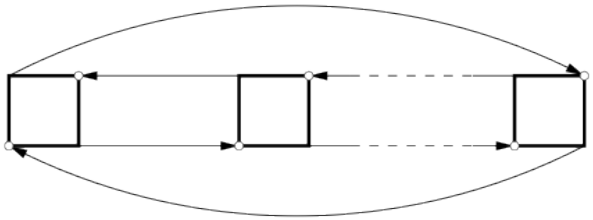
$$F[G] = F[J] - \mathrm{Tr}(JG)$$

$$G_{12} = \frac{1}{Z[J]} \frac{\delta Z[J]}{\delta J_{12}}\bigg|_{J=0} = \frac{\delta F[J]}{\delta J_{12}}\bigg|_{J=0}$$

Decomposition into the single particle part and correlated part

$$F[G] = \mathrm{Tr}\ln G - \mathrm{Tr}\,(\Sigma G) + \Phi[G]$$

$\Phi[G] = \sum_i$



Functional Family

$$F[G] = -Tr\ln[-(G_0^{-1} - \Sigma[G])] - Tr(\Sigma[G]G) + \Phi[G]$$

Exact representation of Φ: $V^{\alpha}_{ee} = \alpha\, V_{ee}$

$$\Phi = \frac{1}{2}\int_0^1 d\alpha Tr[V_{ee}^{\alpha} < \psi^{+}\psi^{+}\psi\psi >]$$

Different Functionals and constrained field J:

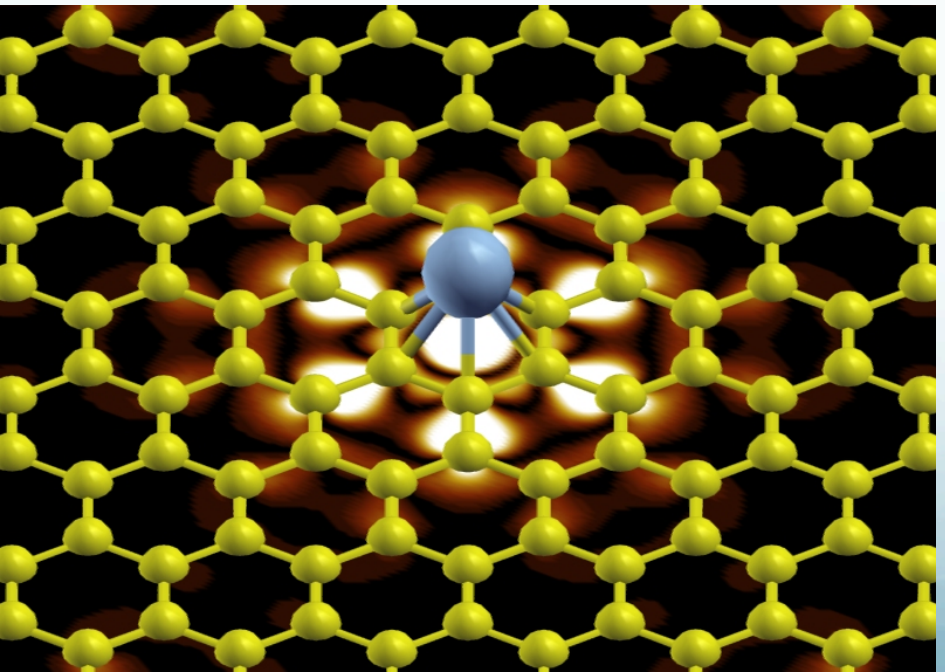
$G=\rho$	$J=V=V_h+V_{xc}$	DFT
$G=G(i\omega)$	$J=\Sigma_{loc}(i\omega)$	LDA+DMFT
$G=G(k,i\omega)$	$J=\Sigma(k,i\omega)$	GW++

G. Kotliar et. al. RMP (2006)

Path Integral for impurity problem

Partition function: $$Z = \int \mathcal{D}[d^\dagger, d] e^{-S}$$

$$S = \sum_{ab} \iint_0^\beta d\tau d\tau' d_a^\dagger(\tau)[(\partial_\tau + E^{ab})\delta(\tau - \tau') + \Delta^{ab}(\tau - \tau')]d_b(\tau') + \int_0^\beta d\tau H^I_{\text{loc}}(\tau)$$



Bath Green-function

$$\mathcal{G}^0 = -(\partial_\tau + \mathbf{E} + \Delta)^{-1}$$

Hybridization

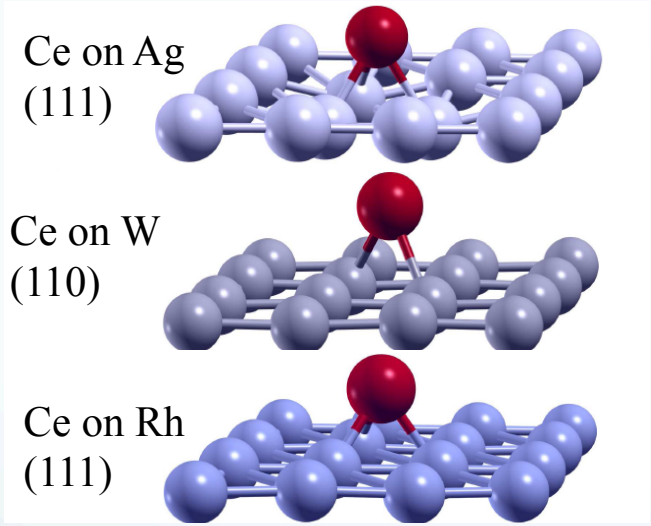
$$\Delta^{ab}(i\omega_n) = \sum_{k\alpha} V_k^{*a\alpha}(i\omega_n - \varepsilon_{k\alpha})^{-1} V_k^{\alpha b}$$

Local Interactions

$$H^I_{\text{loc}} = \sum_{pqrs} I^{pqrs} d_p^\dagger d_q^\dagger d_r d_s + \cdots$$

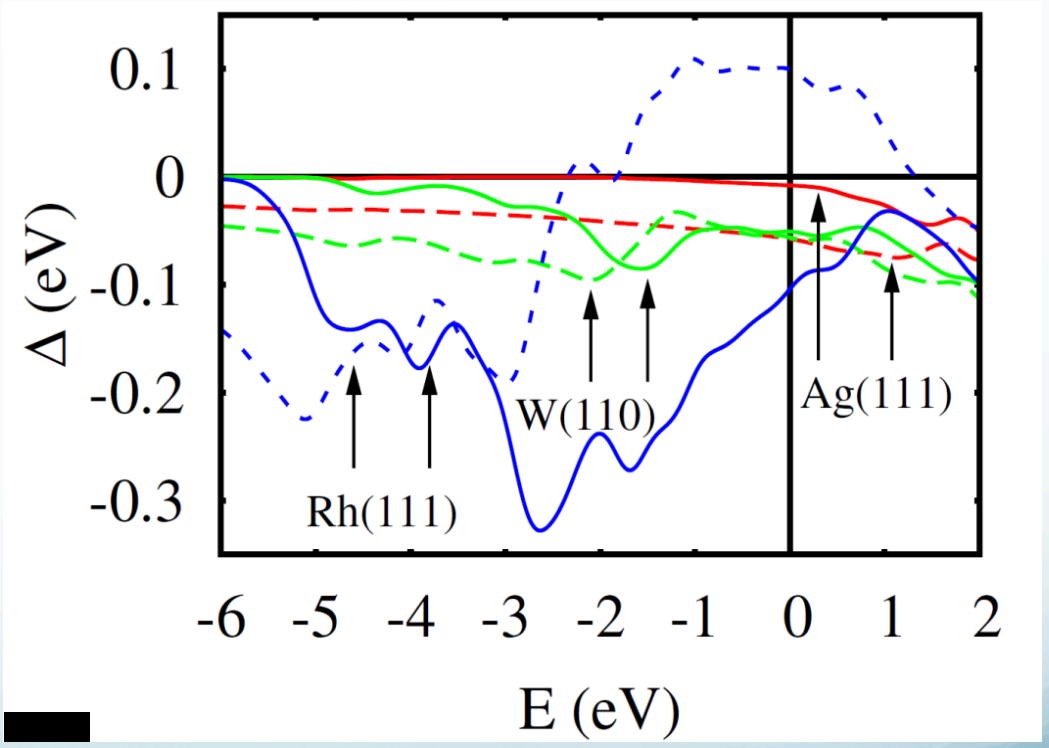
DFT calculations: hybridization

Relaxed structures



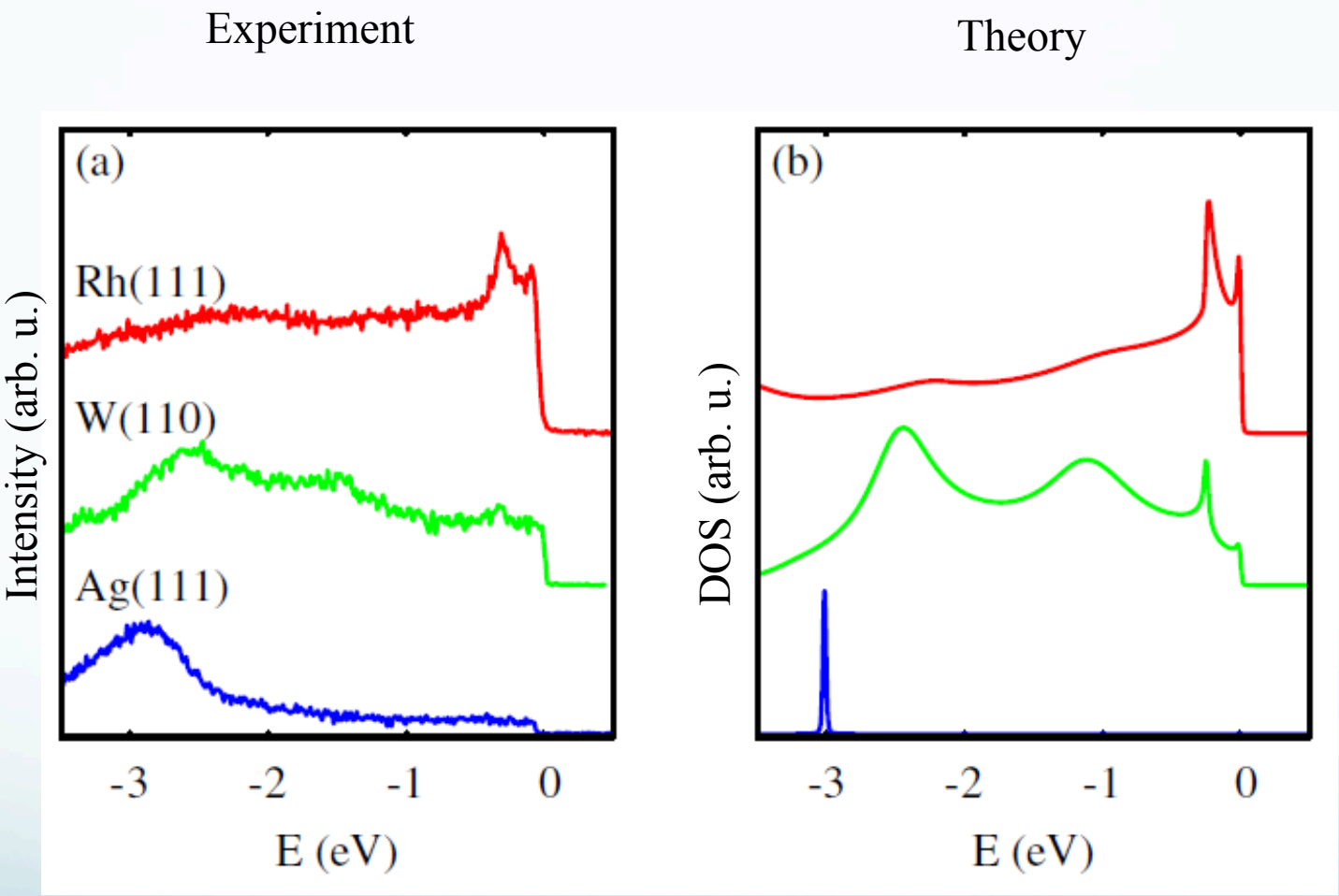
$$\Delta(\omega) = \frac{1}{7}\sum_m \Delta^{\mathrm{LDA}}_{mm}(\omega)$$

Hybridization functions



Solid: Im Δ. Dashed: Re Δ

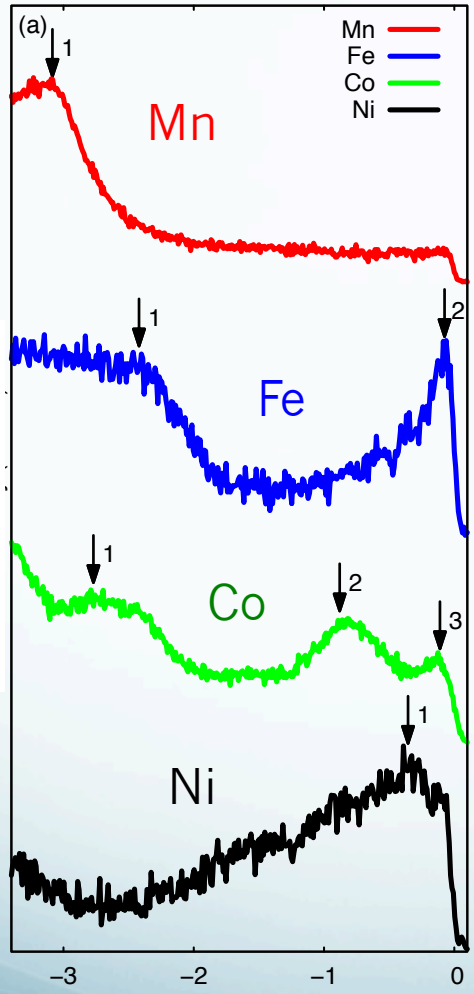
Photoemisson spectra: Ce on TM



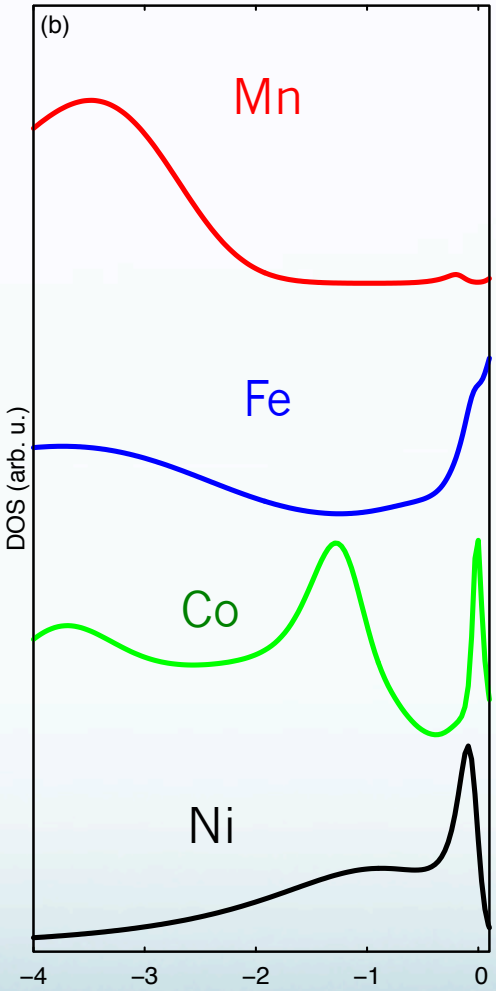
S. Gardonio, T. Wehling et al., PRL (2011)

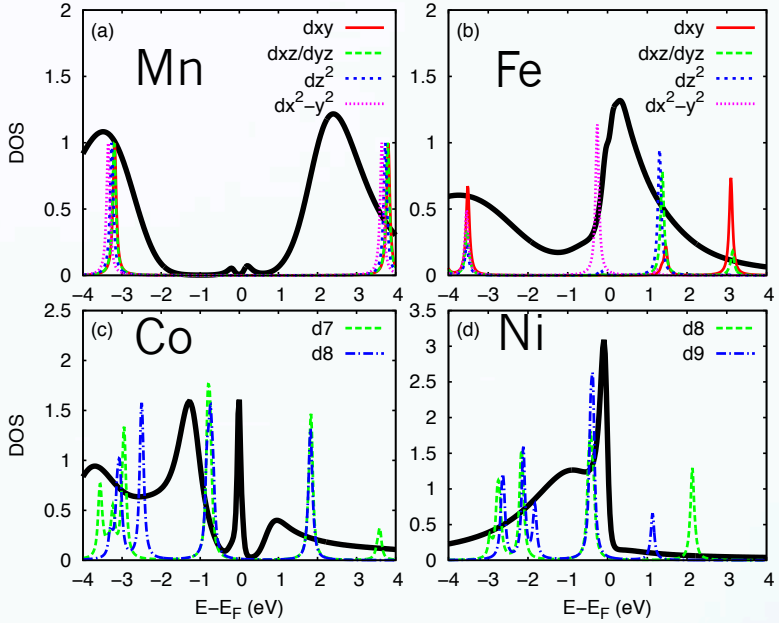
TM on Ag: PES

Experiment



Theory



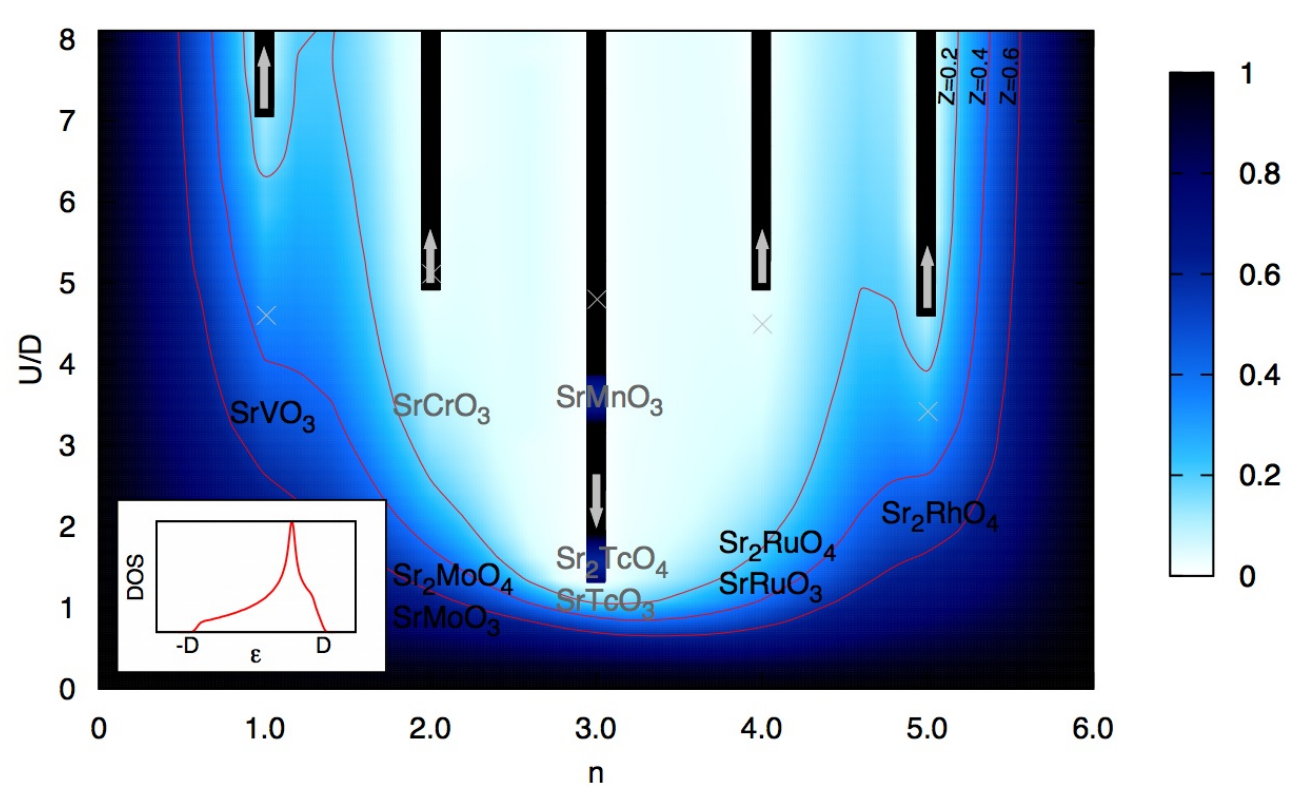


Hund's rule physics:

$$U_{\text{eff}}(n) \approx \begin{cases} U + 4J & \text{for } n = 5 \\ U - (3/2)J & \text{for } n = 6 \text{ and } n = 9 \\ U - (1/2)J & \text{for } n = 7 \text{ and } n = 8 \end{cases}$$

S. Gardonio, M. Karolak et al., PRL (2013)

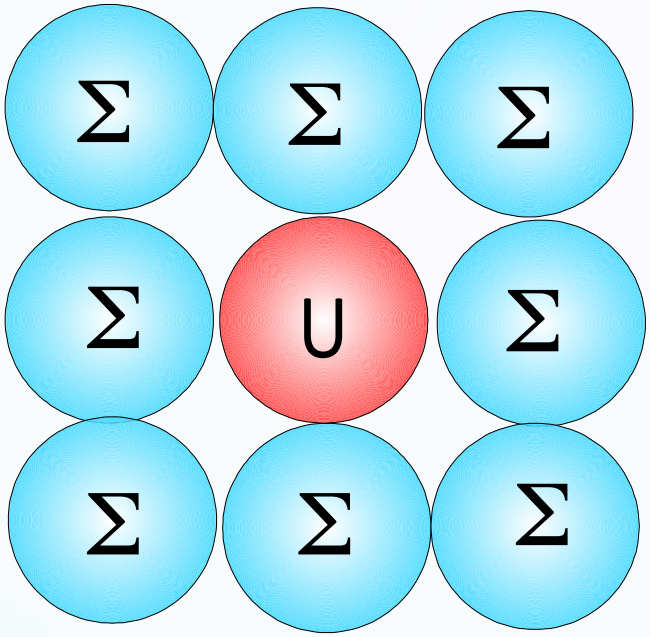
J - Hund's Materials



$$H_{t_{2g}} = (U - 3J)\,\frac{\hat{N}(\hat{N}-1)}{2} - 2J\,\vec{S}^2 - \frac{J}{2}\vec{L}^2 + \frac{5}{2}J\,\hat{N}$$

A. Georges et.al, PRL (2012)

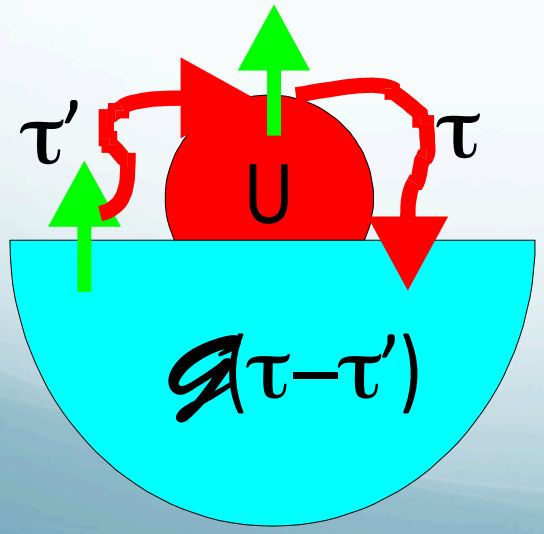
Quantum Impurity Solver



$$Z = \int \mathcal{D}[c^*, c] e^{-S_{simp}},$$

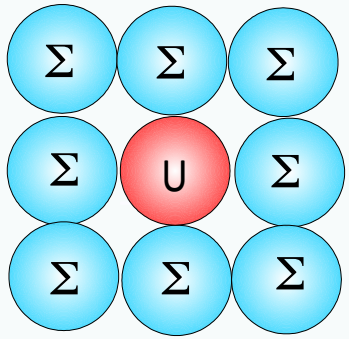
$$S_{simp} = -\sum_{I,J=0}^{N} \int_0^\beta d\tau \int_0^\beta d\tau' \, c^*_{I\sigma}(\tau) \left[\mathcal{G}^{-1}_\sigma(\tau - \tau')\right]_{IJ} c_{J\sigma}(\tau')$$

$$+ \sum_{I=1}^{N} \int_0^\beta d\tau U n_{I,\uparrow}(\tau) n_{I,\downarrow}(\tau),$$



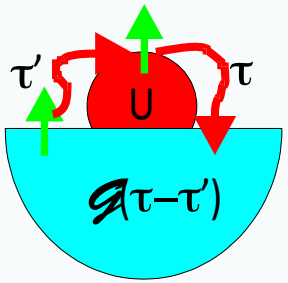
Numerically Exact Solver:
Quantum Monte Carlo

Continuous Time Quantum Monte Carlo



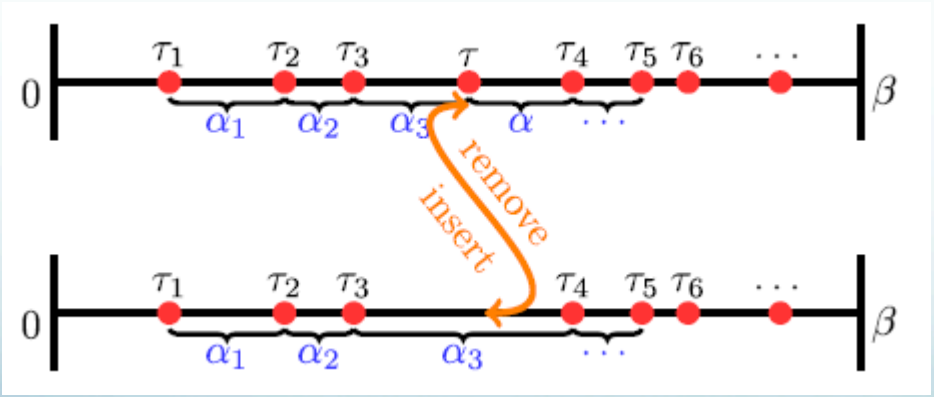
Partition function: $H = H_0 + V$

$$Z = \mathbf{Tr}\left[e^{-\beta H_0}\mathbf{T}_\tau e^{-\int_0^\beta d\tau V(\tau)}\right]$$



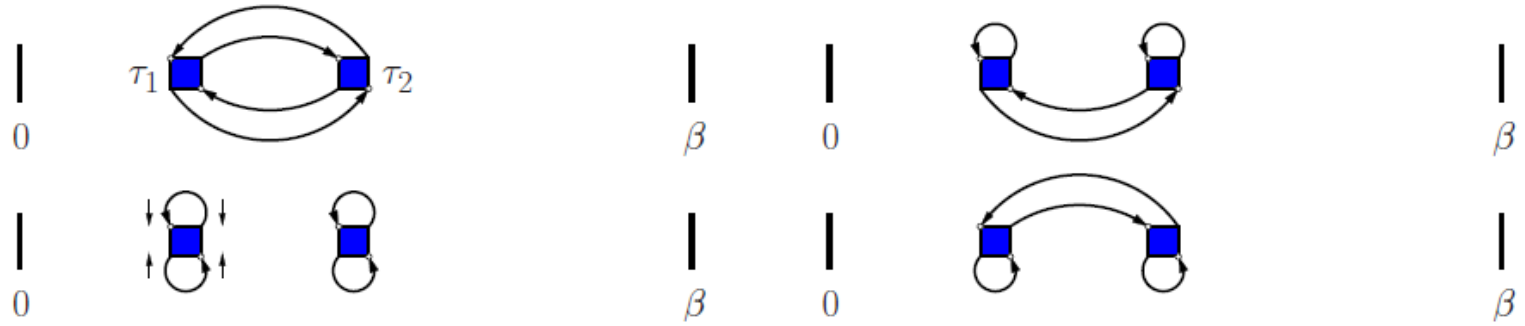
Continuous Time Quantum Monte Carlo (CT-QMC)

$$Z = \sum_{k=0}^{\infty}\int_0^\beta d\tau_1 \int_{\tau_1}^\beta d\tau_2 \ldots \int_{\tau_{k-1}}^\beta d\tau_k \mathbf{Tr}\left[e^{-\beta H_0}e^{-\tau_k H_0}(-V)\ldots e^{-(\tau_2-\tau_1)H_0}(-V)e^{-\tau_1 H_0}\right]$$



E. Gull, A. Millis, A.L., A. Rubtsov, M. Troyer, Ph. Werner, Rev. Mod. Phys. **83**, 349 (2011)

Weak coupling QMC: CT-INT



$$\mathcal{Z}_{\text{imp}} = \int e^{-S_0[c^*,c]+U\int_0^\beta n_\uparrow(\tau)n_\downarrow(\tau)}\mathcal{D}[c^*,c]$$

$$= \int e^{-S_0[c^*,c]} \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} U^k \int_0^\beta d\tau_1 \ldots \int_0^\beta d\tau_k \, c_\uparrow^*(\tau_1)c_\uparrow(\tau_1)$$

$$\times c_\downarrow^*(\tau_1)c_\downarrow(\tau_1)\ldots c_\uparrow^*(\tau_k)c_\uparrow(\tau_k)c_\downarrow^*(\tau_k)c_\downarrow(\tau_k)\,\mathcal{D}[c^*,c]$$

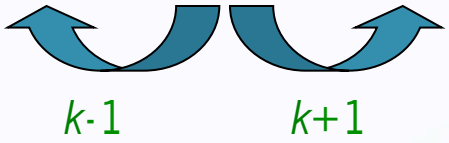
$$= \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} U^k \int d\tau_1 \ldots \int d\tau_k \text{sgn}(\det \hat{G}) \underbrace{|\det \hat{G}|}_{P(k)}$$

$$\hat{G}_{ij} = g_0(\tau_i - \tau_j)$$

A. Rubtsov, 2004

Random walks in the *k*-space

$Z = \ldots Z_{k-1} + Z_k + Z_{k+1} + \ldots$

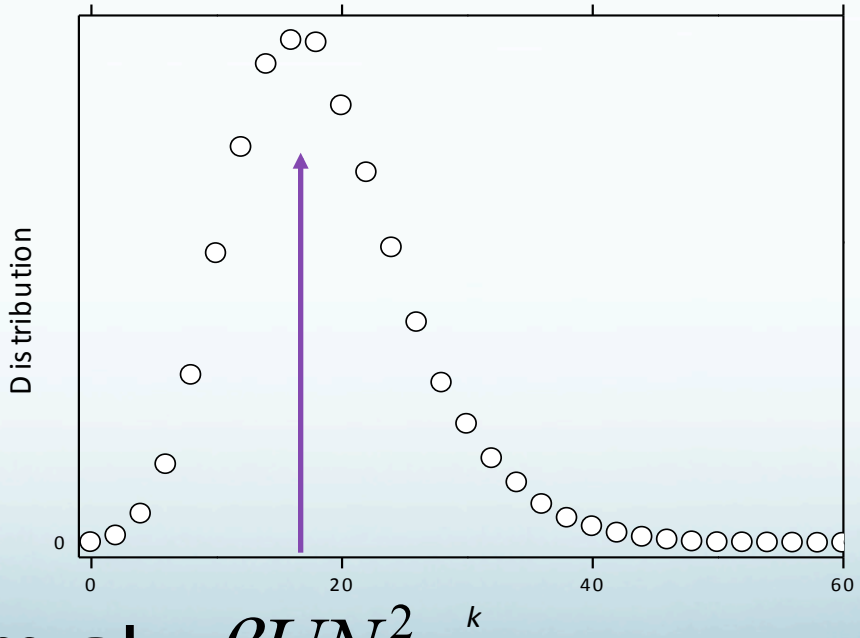


Acceptance ratio

decrease

Step *k*-1

$$\frac{k}{|w|}\frac{D^{k-1}}{D^k}$$



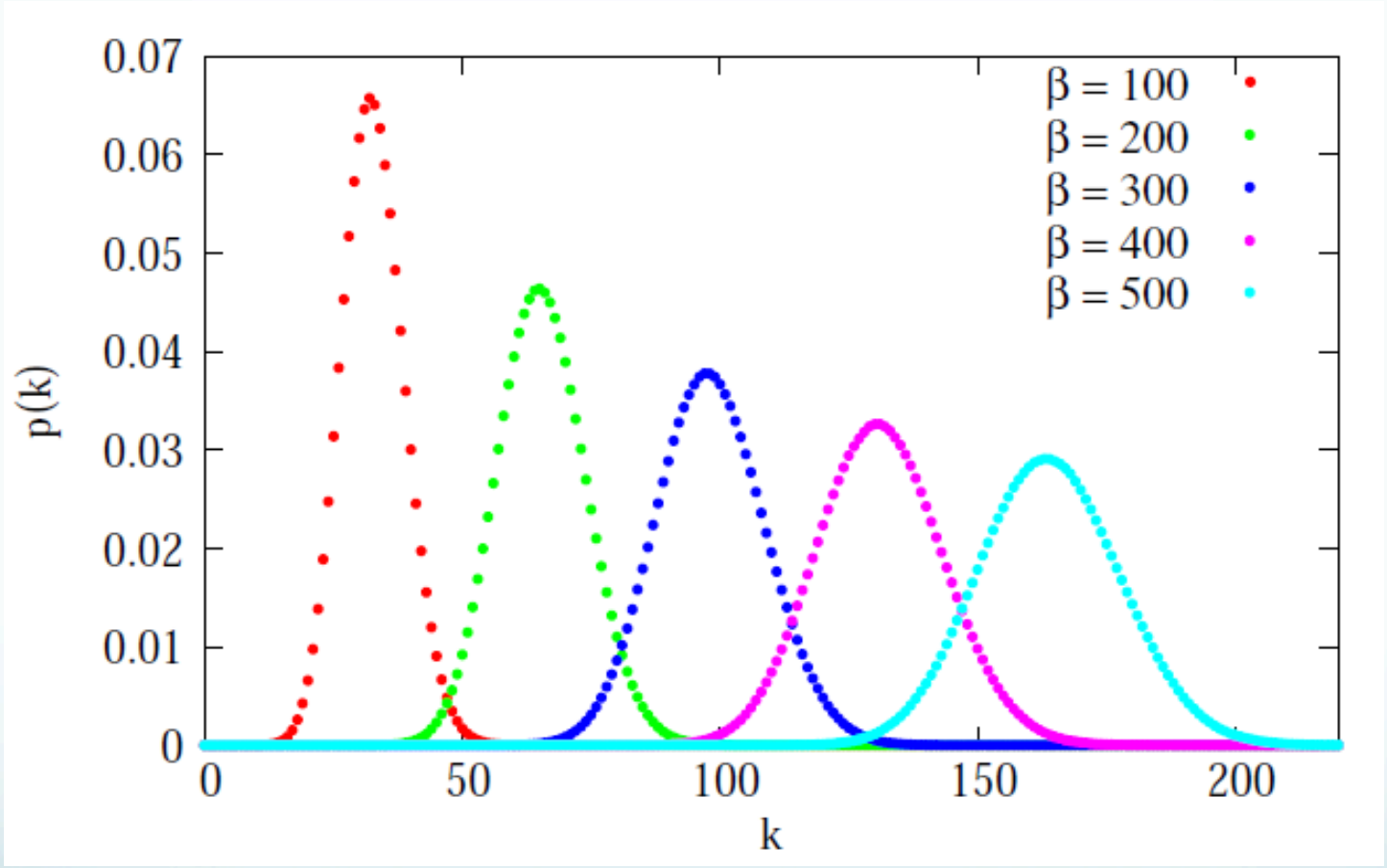
increase

Step *k*+1

$$\frac{|w|}{k+1}\frac{D^{k+1}}{D^k}$$

Maximum at βUN^2

Convergence with Temperature: CT-INT



Maximum: $\beta U N^2$

CT-QMC: Hybridization expansion (CT-HYB)

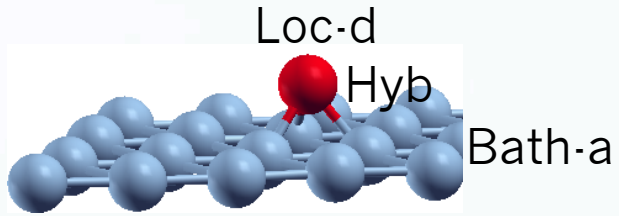
Hamiltonian:

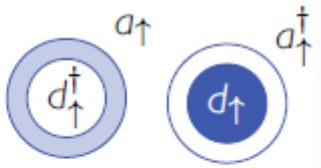
$$H = H_{\text{bath}} + H_{\text{hyb}} + H_{\text{loc}}$$

$$H_{\text{bath}} = \sum_p \epsilon_p a_p^\dagger a_p$$

$$H_{\text{hyb}} = \sum_{p,j} V_p^j d_j^\dagger a_p + V_p^{j*} a_p^\dagger d_j$$

$$H_{\text{loc}} = U \sum_i n_{i\uparrow} n_{i\downarrow}$$



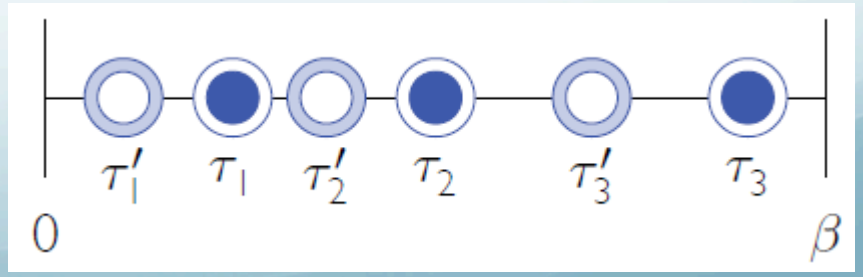


$$H_0 = H_{\text{bath}} + H_{\text{loc}}$$

$$V = H_{\text{hyb}}$$

$$V = H_{\text{hyb}} = \sum_{p,j} V_p^j d_j^\dagger a_p + V_p^{j*} a_p^\dagger d_j = \tilde{H}_{\text{hyb}}^\dagger + \tilde{H}_{\text{hyb}}$$

$$Z = \sum_{k=0}^{\infty} \int_0^\beta d\tau_1 \ldots \int_{\tau_{k-1}}^\beta d\tau_k \int_0^\beta d\tau_1' \ldots \int_{\tau_{k-1}'}^\beta d\tau_k' \mathbf{Tr}\left[e^{-\beta(H_{\text{loc}}+H_{\text{bath}})} \mathbf{T}_\tau \tilde{H}_{hyb}^\dagger(\tau_k) \tilde{H}_{hyb}(\tau_k') \ldots \tilde{H}_{hyb}^\dagger(\tau_1) \tilde{H}_{hyb}(\tau_1') \right]$$

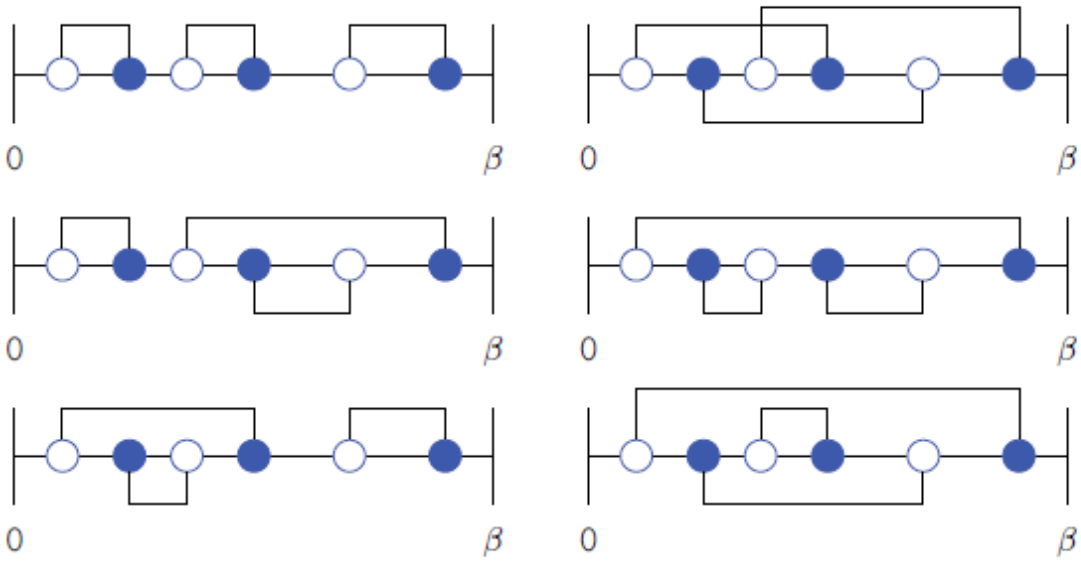


Ph. Werner, et al PRL **97**, 076405 (2006)

CT-HYB: determinant weght

higher orders:

H_{bath} is non-interacting apply Wick's theorem



bath contributions can be combined into

$$\det(\Delta) = \begin{vmatrix} \Delta_{11'} & \Delta_{12'} & \Delta_{13'} \\ \Delta_{21'} & \Delta_{22'} & \Delta_{23'} \\ \Delta_{31'} & \Delta_{32'} & \Delta_{33'} \end{vmatrix}$$

$$Z = Z_{\text{bath}} \sum_{k=0}^{\infty} \int_0^{\beta} d\tau_1 \ldots \int_{\tau'_{k-1}}^{\beta} d\tau'_k \sum_{\substack{j_1,\ldots j_k \\ j'_1,\ldots j'_k}} \text{Tr}_d \left[e^{-\beta H_{\text{loc}}} \mathsf{T}_\tau d_{j_k}(\tau_k) d^\dagger_{j'_k}(\tau'_k) \ldots d_{j_1}(\tau_1) d^\dagger_{j'_1}(\tau'_1) \right] \det(\Delta)$$

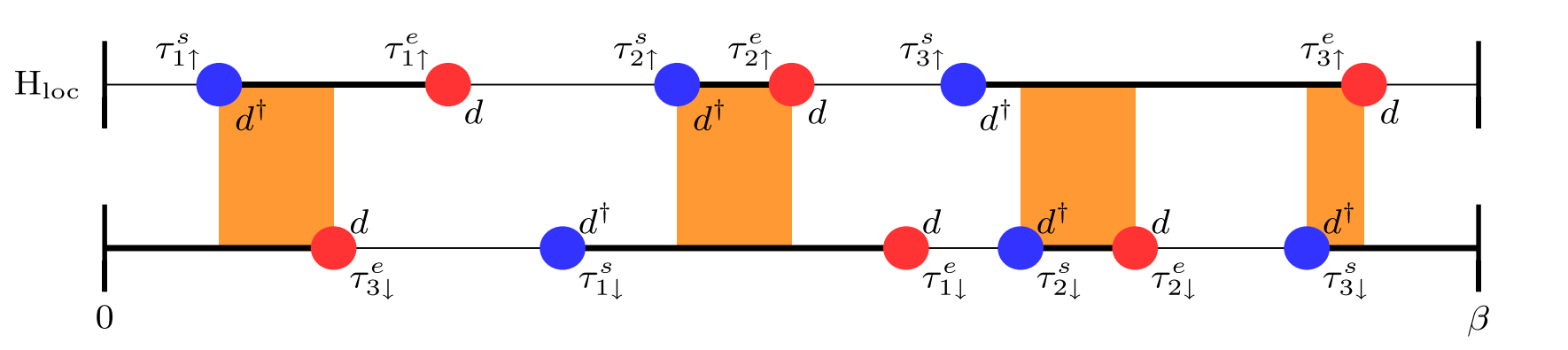
Strong-Coupling Expansion: CT-HYB

$$S_{\mathrm{at}} = \int_0^\beta d\tau \sum_\sigma c_\sigma^*(\tau)[\partial_\tau - \mu]c_\sigma(\tau) + U\int_0^\beta d\tau c_\uparrow^*(\tau)c_\uparrow(\tau)c_\downarrow^*(\tau)c_\downarrow(\tau)$$

Hybridization:

$$S_\Delta = -\int_0^\beta d\tau' \int_0^\beta d\tau \sum_\sigma c_\sigma(\tau)\Delta(\tau-\tau')c_\sigma^*(\tau')$$

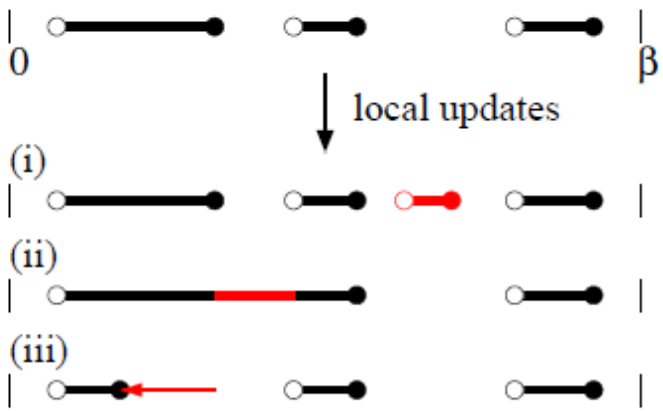
$$\mathcal{Z} = \mathcal{Z}_{\mathrm{at}} \sum_k \int_0^\beta d\tau_1' \int_{\tau_1'}^\beta d\tau_1 \ldots \int_{\tau_{k-1}}^\beta d\tau_k' \int_{\tau_k'}^{\circ\tau_k'} d\tau_k \times$$
$$\times \langle c(\tau_k)c^*(\tau_k') \ldots c(\tau_1)c^*(\tau_1')\rangle_{\mathrm{at}} \det \hat{\Delta}^{(k)}$$



P. Werner, et al, 2006

CT-HYB: Monte Carlo sampling

- Sampling of Z through local updates
  - (i) insertion/removal of segments
  - (ii) insertion/removal of anti-segments
  - (iii) shifts of the segment end points



- Detailed balance

$$s_k \to s_{k+1} = s_k + \tilde{s} \qquad \frac{p_{ins}(\tilde{s})}{p_{rem}(\tilde{s})} = \frac{Z_{k+1}(s_{k+1})}{Z_k(s_k)} \frac{\beta l_{max}}{k+1} e^{\tilde{l}\mu}$$

- Store and update the matrix $M = F^{-1}$

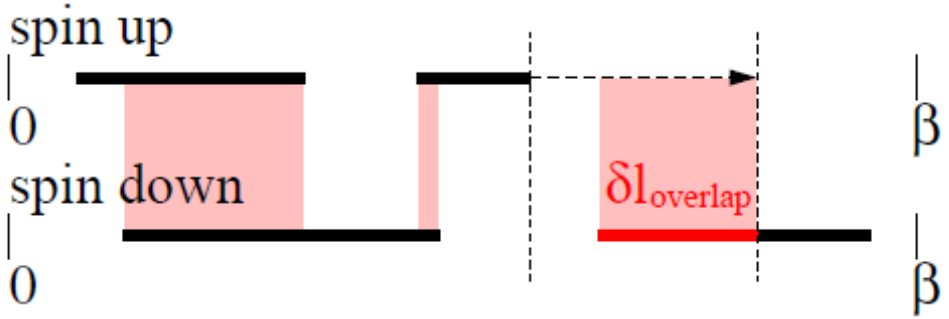
  $\to$ access to determinant ratios

  $\to$ efficient computation of G

$$G(\tau) = \left\langle \frac{1}{\beta} \sum_{i,j} M_{j,i} \Delta(\tau, \tau_i^e - \tau_j^s) \right\rangle$$

CT-HYB: segment scheme

- Hubbard model ($U \neq 0$): Segment configurations for spin up/down



- Acceptance rate for MC moves now also depends on segment overlap
- Detailed balance

$$s \to \tilde{s} \qquad \frac{p(s \to \tilde{s})}{p(\tilde{s} \to s)} = \frac{Z_k(\tilde{s})}{Z_k(s)} e^{(\tilde{l}-l)\mu - U\delta l_{overlap}}$$

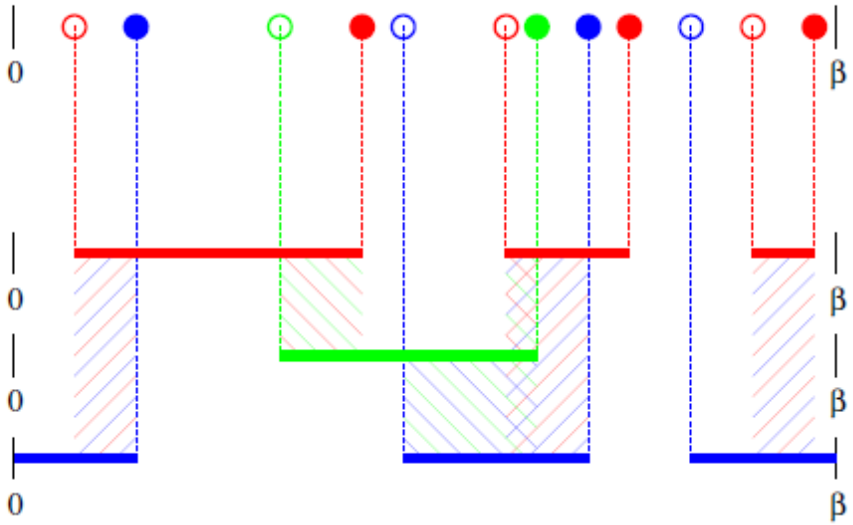
- Obviously: $E_{pot} = U\langle l^{total}_{overlap}\rangle$

$$n_\sigma = G_\sigma(\beta) = \beta^{-1}\langle l^{total}_\sigma\rangle$$

CT-HYB: multi-orbital segment picture

$$H_{\text{int}} = \sum_{ij} U_{ij} n_i n_j$$

- If $\psi^\dagger$, ψ (for each flavor) must occur in alternating order ...

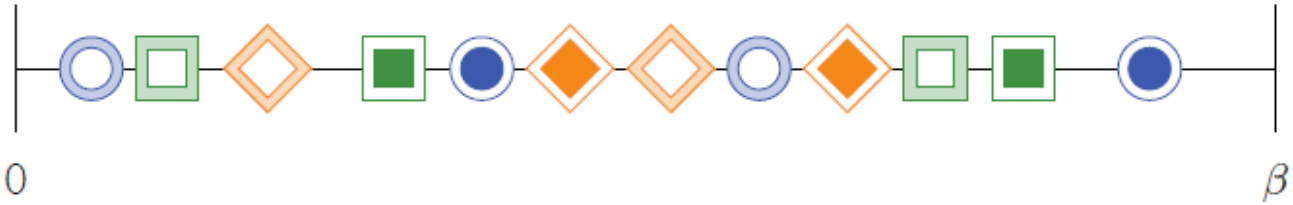


... the segment picture allows an efficient representation of all configurations with non-zero trace

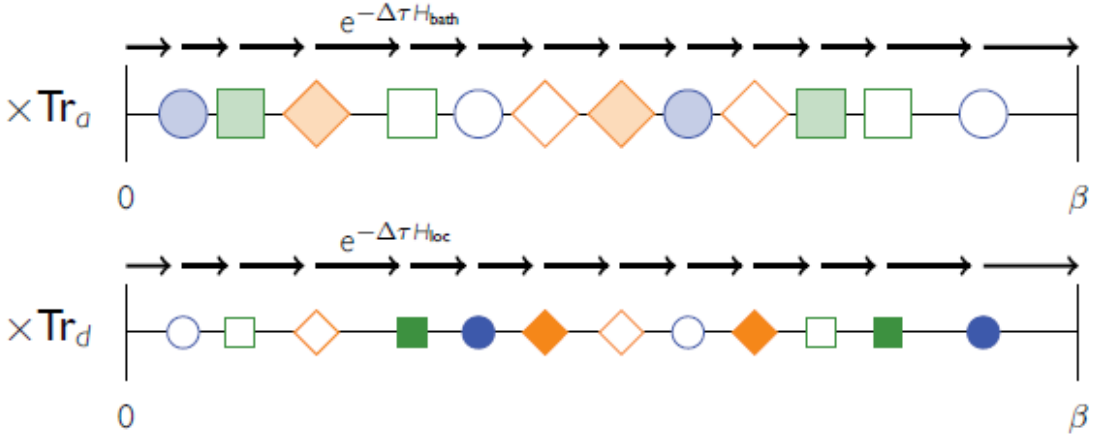
CT-HYB: General Interaction

P. Werner and A.J. Millis, PRB 74, 155107 (2006)

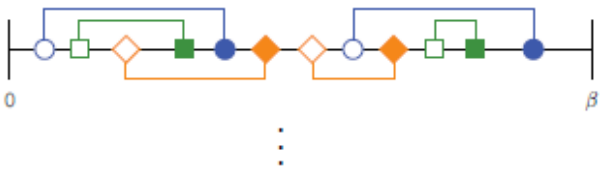
creation and annihilation operators for different orbitals



$$Z = \sum_{k=0}^{\infty} \int_0^\beta d\tau_1 \ldots \int_{\tau_{k-1}}^\beta d\tau_k \int_0^\beta d\tau'_1 \ldots \int_{\tau'_{k-1}}^\beta d\tau'_k \sum_{\substack{j_1,\ldots,j_k \\ j'_1,\ldots,j'_k}} \sum_{p_1,\ldots,p_k \; p'_1,\ldots,p'_k} V^{j_1}_{p_1} V^{j'_1 *}_{p'_1} \ldots V^{j_k}_{p_k} V^{j'_k *}_{p'_k}$$



tracing out bath degrees of freedom gives rise to determinant weight as before

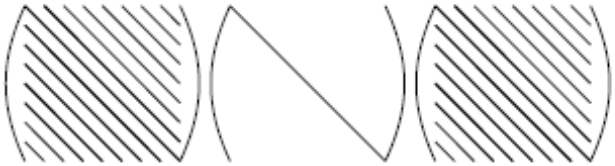


CT-HYB: matrix code

Transform operators into eigenbasis of $H_{\mathbf{loc}}$

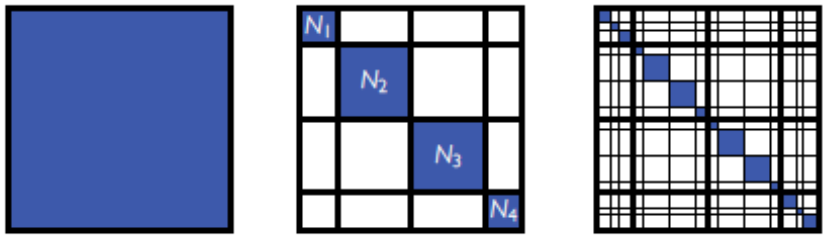
$e^{-\tau H_{\mathrm{loc}}} = \mathbf{diag}(e^{-\tau\alpha_1}, \ldots, e^{-\tau\alpha_N})$ $\quad O_k(\tau_k)$: dense matrices

$$\mathbf{Tr}_d\,[\ldots] = \mathbf{Tr}\left[e^{-(\beta-\tau_k)H_{\mathrm{loc}}} O_k(\tau_k) e^{-(\tau_k-\tau_{k-1})H_{\mathrm{loc}}} \ldots O_1(\tau_1) e^{-\tau_1 H_{\mathrm{loc}}}\right]$$



Matrix-matrix multiplications in the trace calculation

Exploiting symmetries and block-diagonalizing operators



only symmetry sectors contributing to the trace need to be considered

CT-HYB: Krylov code

A. M. Läuchli and P. Werner, PRB 80, 235117 (2009)

$e^{-\Delta\tau H_{loc}}$ $O_k(\tau_k)$ sparse in the occupation number basis

Krylov time evolution

$$\mathrm{Tr}_d\,[\ldots] = \sum_{|\psi\rangle} \langle\psi| e^{-(\beta-\tau_k)H_{loc}} O_k(\tau_k) e^{-(\tau_k-\tau_{k-1})H_{loc}} \ldots O_1(\tau_1) e^{-\tau_1 H_{loc}} |\psi\rangle$$

Idea: compute $e^{-\tau H_{loc}}|\psi\rangle$ using Lanczos recursion Park and Light, J. Chem. Phys (1986)

sparse matrix-vector multiplication

construct Krylov subspace $\mathcal{K} = \left\{ |\psi\rangle, H_{loc}|\psi\rangle, H_{loc}^2|\psi\rangle, \ldots, H_{loc}^p|\psi\rangle \right\}$

efficiently represents $e^{-\tau H_{loc}}|\psi\rangle$ for a small number p τ small $\longrightarrow$ p small

Hochbruck & Lubich, SIAM J. Numer. Anal. (1997)

CT-HYB: Krylov code

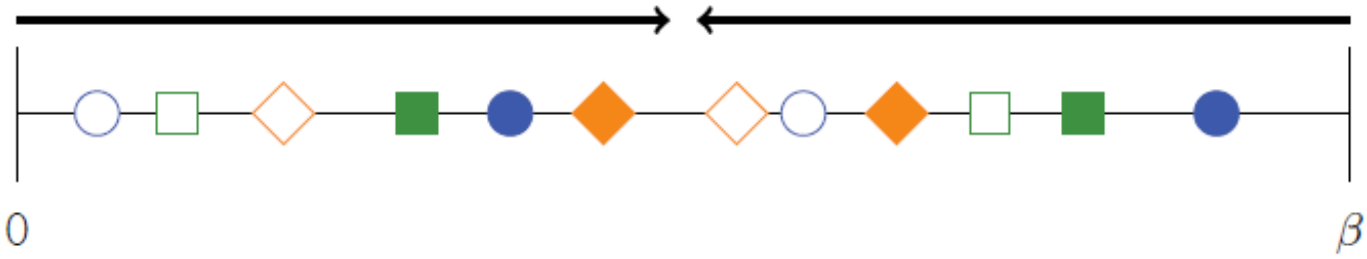
$$\mathrm{Tr}_d\,[\ldots] = \sum_{|\psi\rangle} \langle\psi| e^{-(\beta-\tau_k)H_{\mathrm{loc}}} O_k(\tau_k) e^{-(\tau_k-\tau_{k-1})H_{\mathrm{loc}}} \ldots O_1(\tau_1) e^{-\tau_1 H_{\mathrm{loc}}} |\psi\rangle$$

Trace truncation

sum only over low-energy states $|\psi\rangle$ in $\sum_{|\psi\rangle}$

include at least lowest multiplet of the local problem

different from truncating the Hilbert space

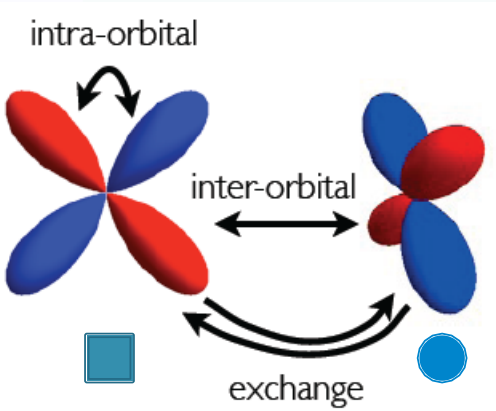


all states remain accessible in the propagation at intermediate times

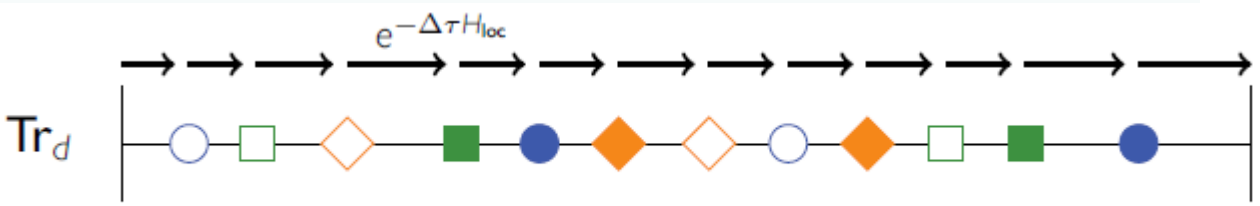
Multiorbital impurity with general U

General Interaction:

$$U = \frac{1}{2}\sum_{\substack{ijkl\\ \sigma\sigma'}} \left\langle ij \left| \frac{1}{r_{12}} \right| kl \right\rangle d^{+}_{i\sigma} d^{+}_{j\sigma'} d_{l\sigma'} d_{k\sigma}$$



Krylov-CT-QMC



$$\mathrm{Tr}_d\,[\ldots] = \mathrm{Tr}\left[e^{-(\beta-\tau_k)H_{\mathrm{loc}}} O_k(\tau_k) e^{-(\tau_k-\tau_{k-1})H_{\mathrm{loc}}} \ldots O_1(\tau_1) e^{-\tau_1 H_{\mathrm{loc}}}\right]$$

$$Z_{\mathrm{AIM}} = Z_{\mathrm{bath}} \sum_k \int d\tau_1 \cdots d\tau'_k \mathrm{Tr}_{\mathrm{d}}\,[\ldots]\,\det \boldsymbol{\Delta}$$

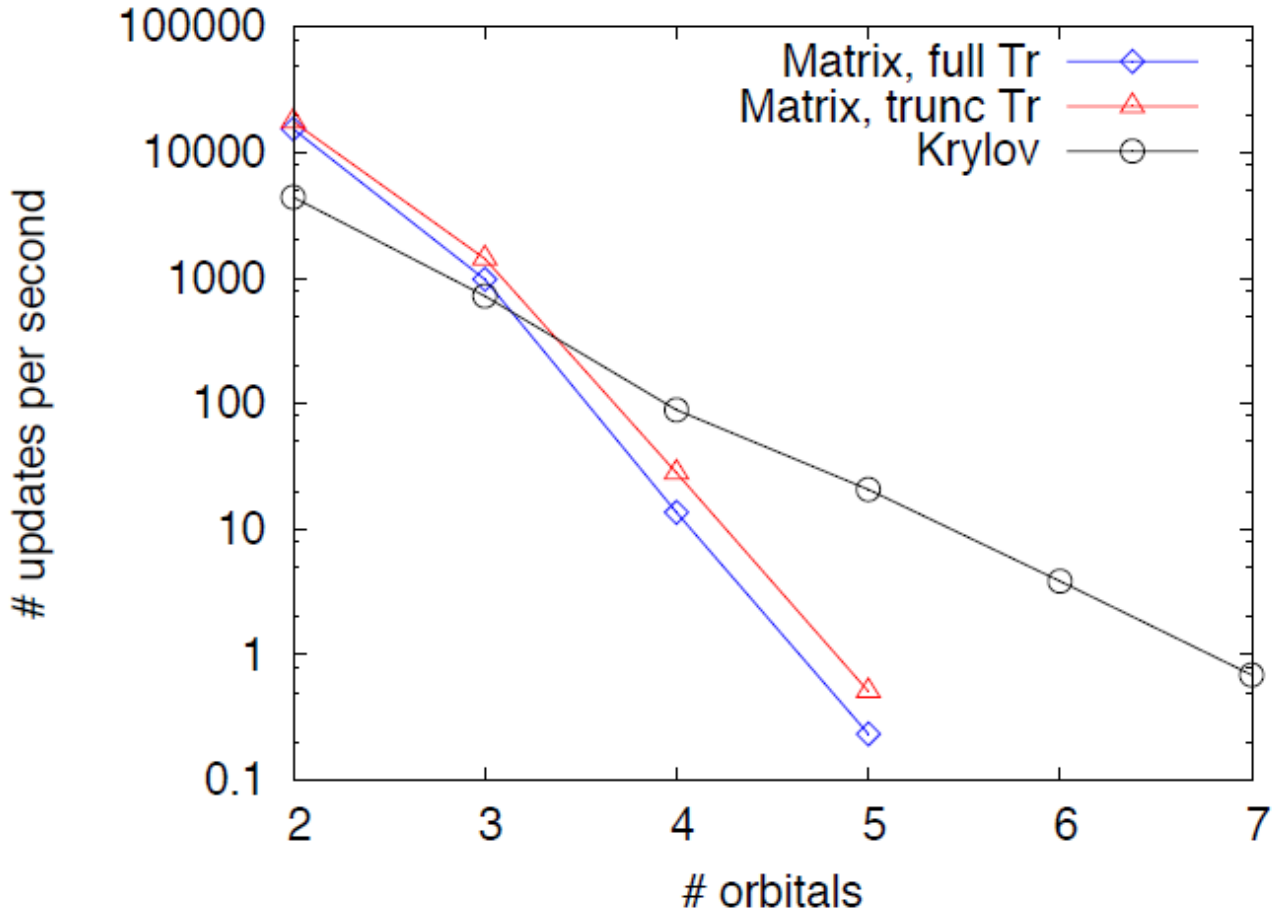
$$\mathcal{K}_p(|v\rangle) = \{|v\rangle, H_{\mathrm{loc}}|v\rangle, H^2_{\mathrm{loc}}|v\rangle, \ldots, H^p_{\mathrm{loc}}|v\rangle\}$$

$$e^{-H_{\mathrm{loc}}\tau_k}|v\rangle \approx V e^{-h_{\mathrm{p}}\tau_k} V^{\dagger}|v\rangle$$

A. Läuchli and Ph. Werner, et al PRB **80**, 235117 (2009)

CT-QMC-Krylov: performance

A. M. Läuchli and P. Werner, PRB 80, 235117 (2009)



Miracle of CT-QMC

$$S = \sum_{\sigma\sigma'} \int_0^\beta d\tau \int_0^\beta d\tau [-G_0^{-1}(\tau-\tau')c_\sigma^+(\tau)c_\sigma(\tau') + \frac{1}{2}U\delta(\tau-\tau')c_\sigma^+(\tau)c_{\sigma'}^+(\tau)c_{\sigma'}(\tau')c_\sigma(\tau')]$$

$$G_0^{-1}(\tau-\tau') = \delta(\tau-\tau')[\frac{\partial}{\partial\tau} + \mu] - \Delta(\tau-\tau')$$

Interaction expansion CT-INT: A. Rubtsov et al, JTP Lett (2004)

$$Z = Z_0 \sum_{k=0}^{\infty} \frac{(-U)^k}{k!} Tr \det[G_0(\tau-\tau')]$$

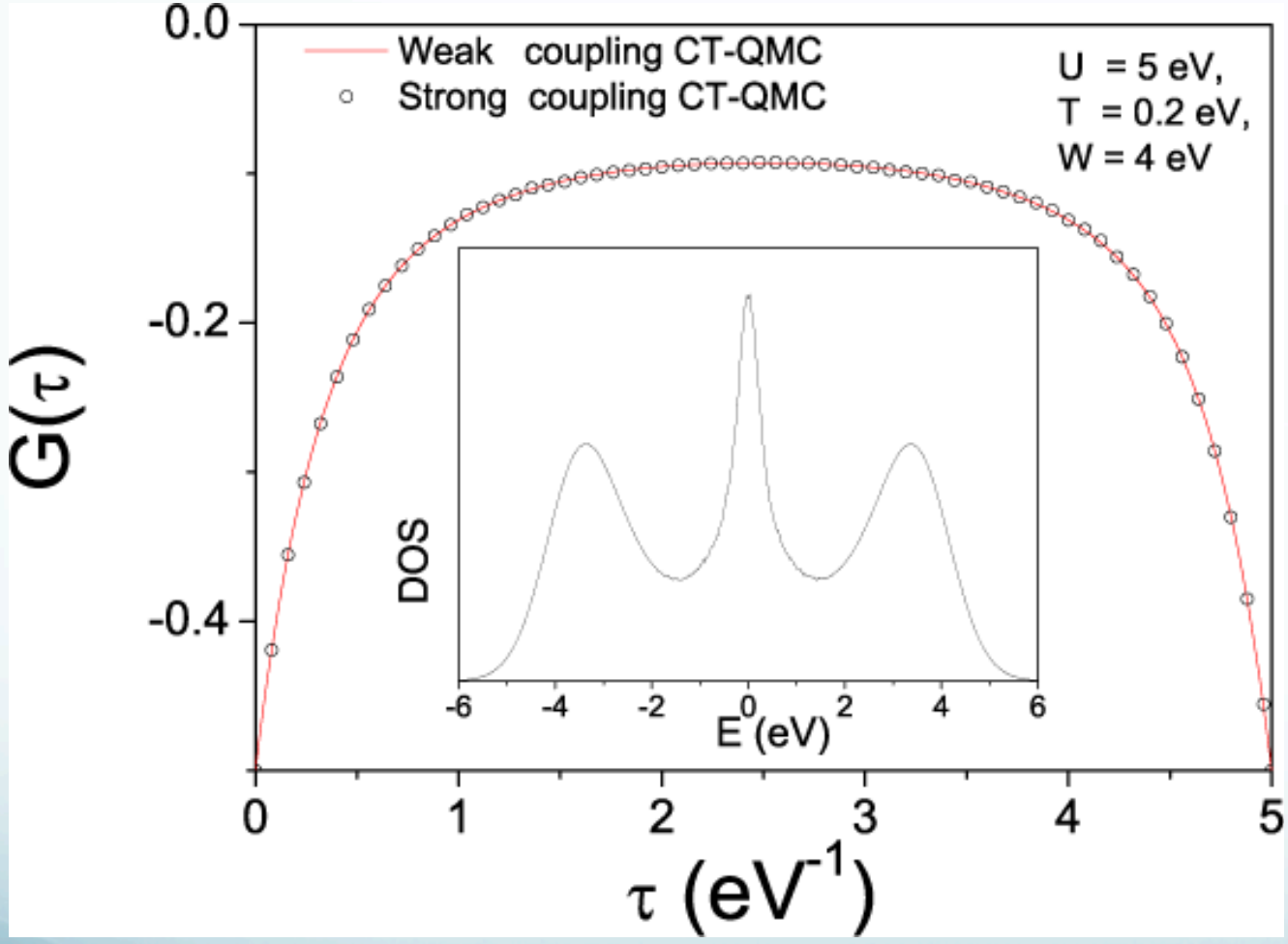
Hybridization expansion CT-HYB: P. Werner et al, PRL (2006)

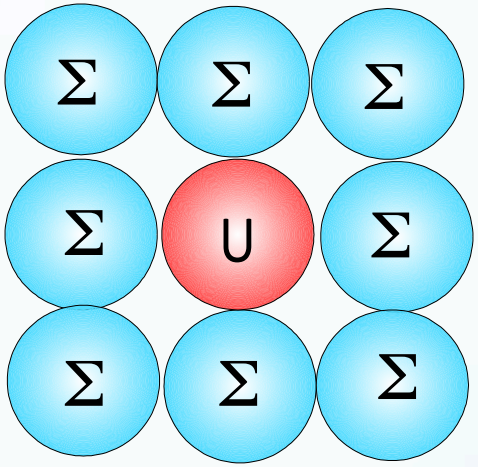
$$Z = Z_0 \sum_{k=0}^{\infty} \frac{1}{k!} Tr \left\langle c_\sigma^+(\tau)c_\sigma(\tau')...c_{\sigma'}^+(\tau)c_{\sigma'}(\tau') \right\rangle_0 \det[\Delta(\tau-\tau')]$$

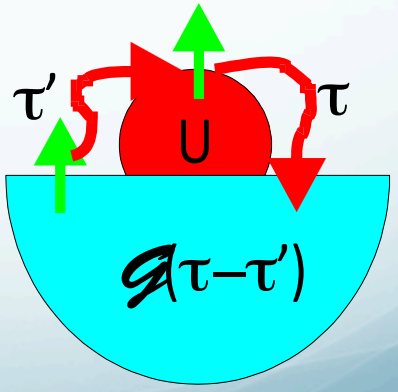
Efficient Krylov scheme: A. Läuchli and P. Werner, PRB (2009)

E. Gull, et al, RMP (2011) + ALPS & TRIQS implementations

Comparison of different CT-QMC



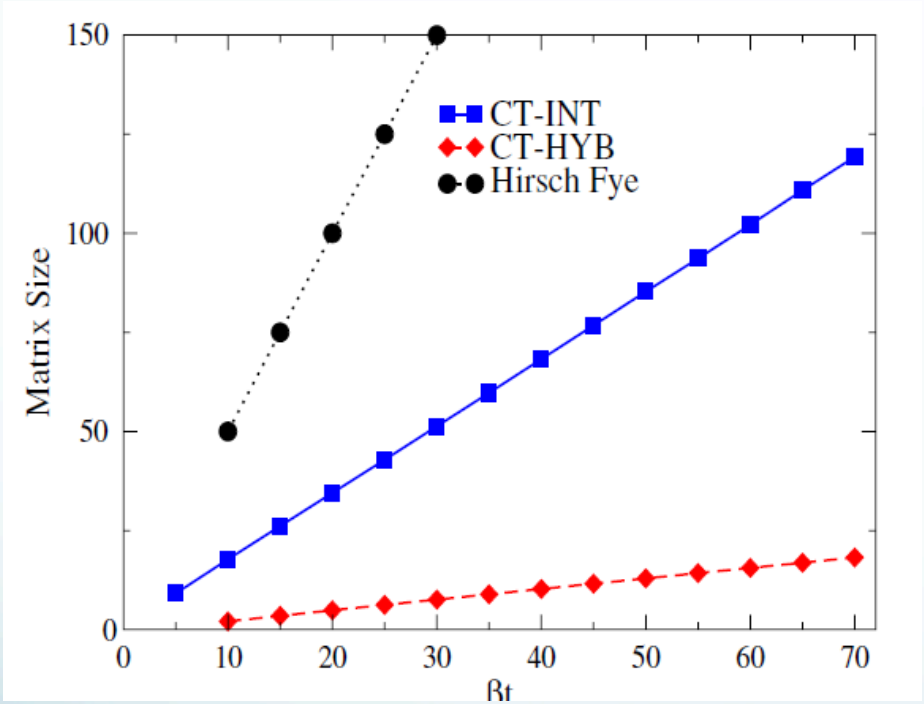




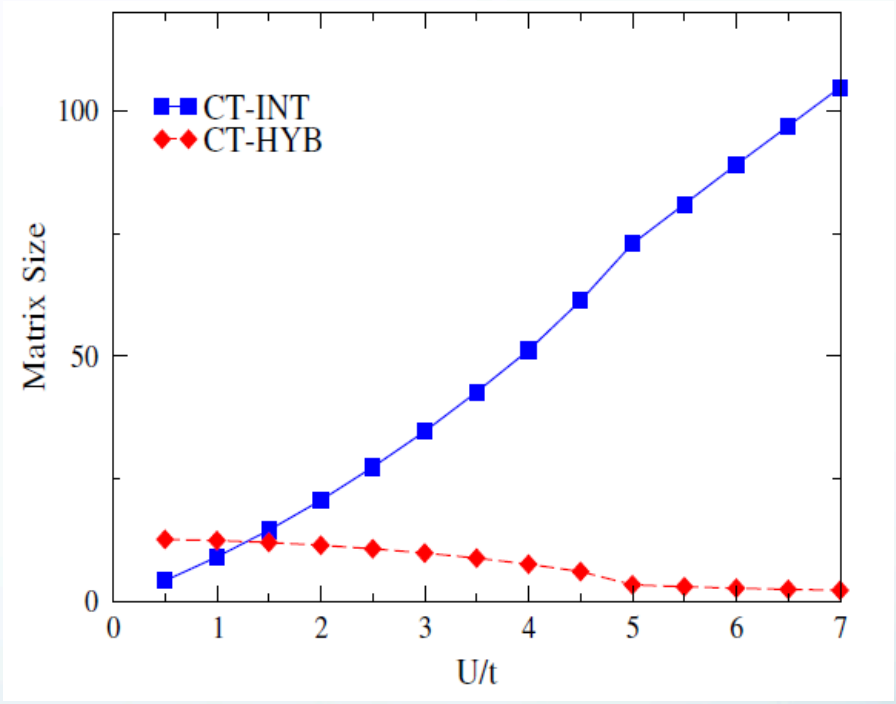
Ch. Jung, unpublished

CT-QMC review:E. Gull et al. RMP (2011)

Scaling of CT-QMC



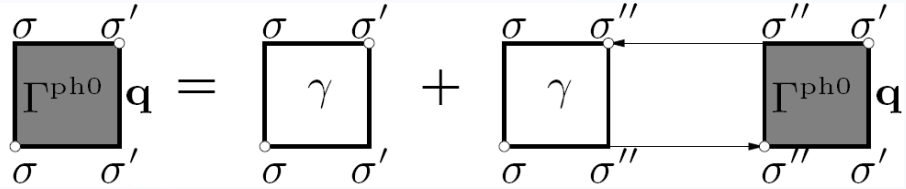
Temperature



Interactions

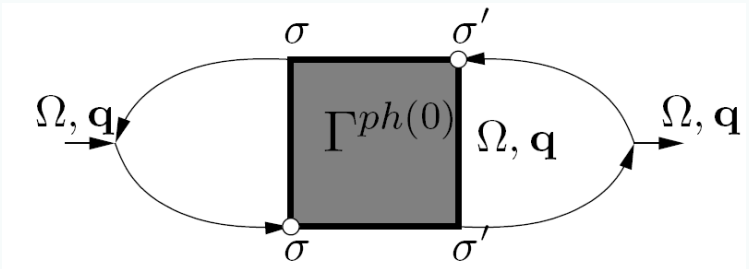
Magnetic susceptibility: nanosystems

Bethe-Salpeter Equation:

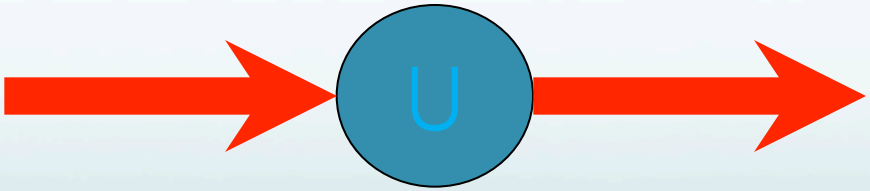


Susceptibility:

$$\tilde{\chi}^{\sigma\sigma'}(\Omega, \mathbf{q})$$

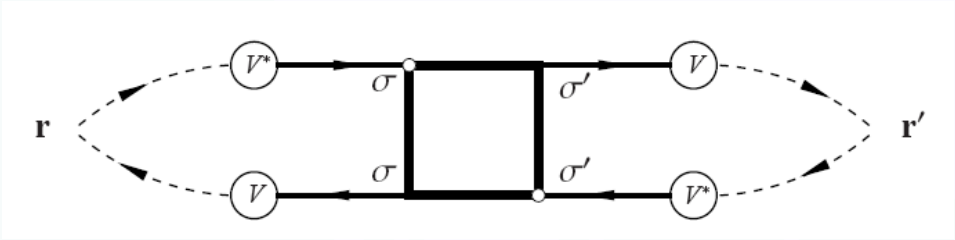


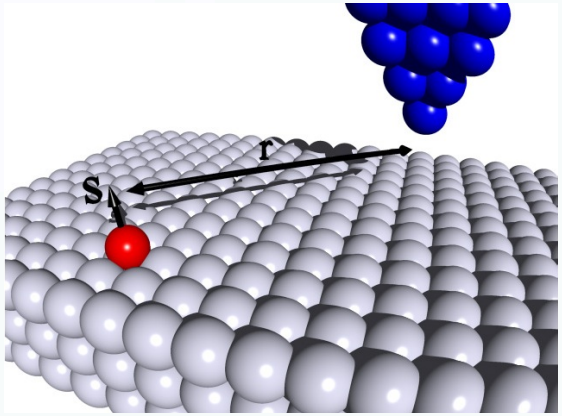
Local correlated nano-system:

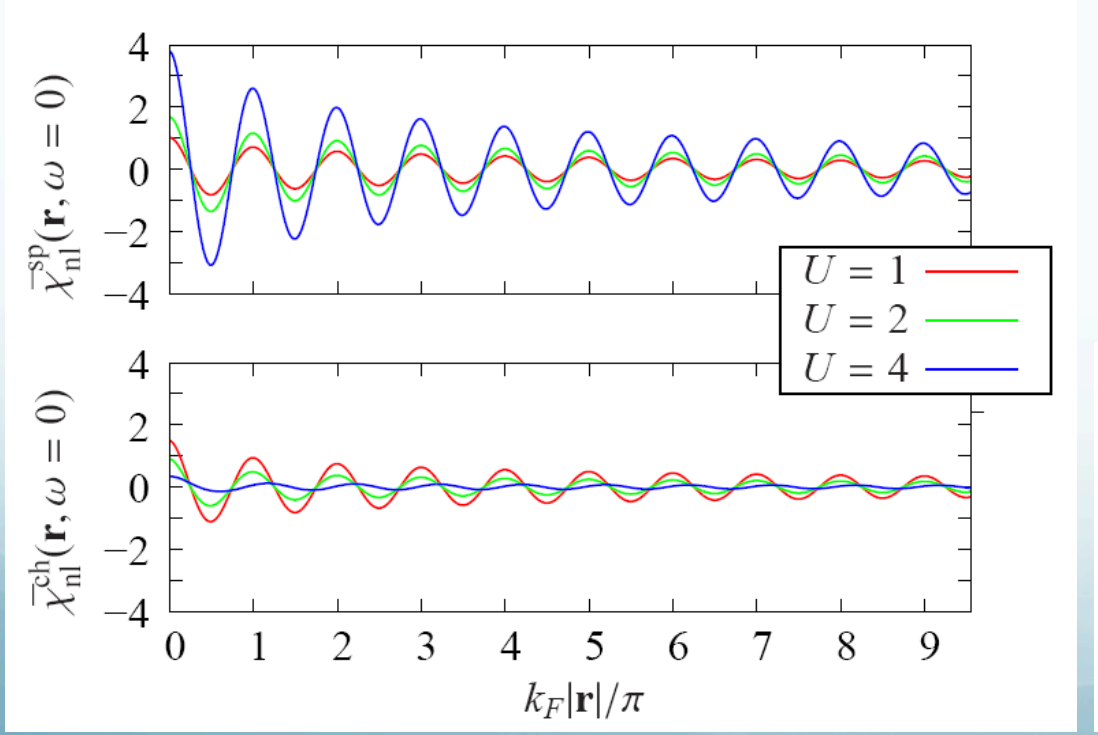


$$\chi^{-1}_{\nu,\nu'}(\vec{q},\omega) = \chi^{-1}_{0,\nu,\nu'}(\vec{q},\omega) - \gamma_{\nu,\nu'}(\omega)$$

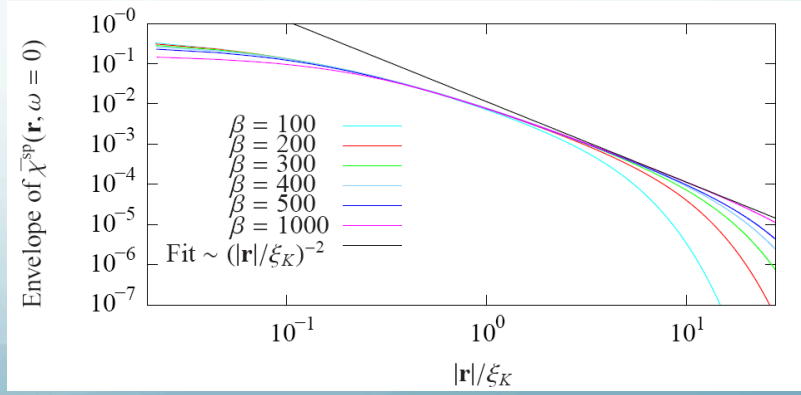
Spin and Charge susceptibility near impurity





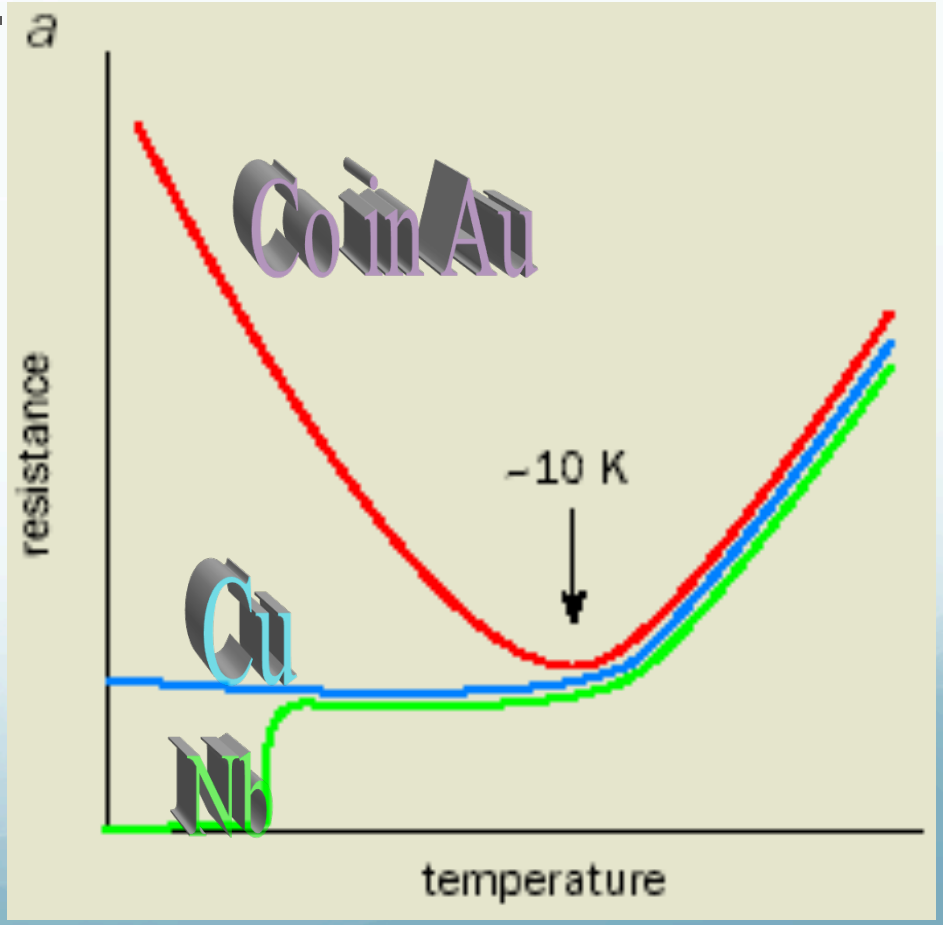


K. Patton, H. Hafermann,et.al
PRB (2009)

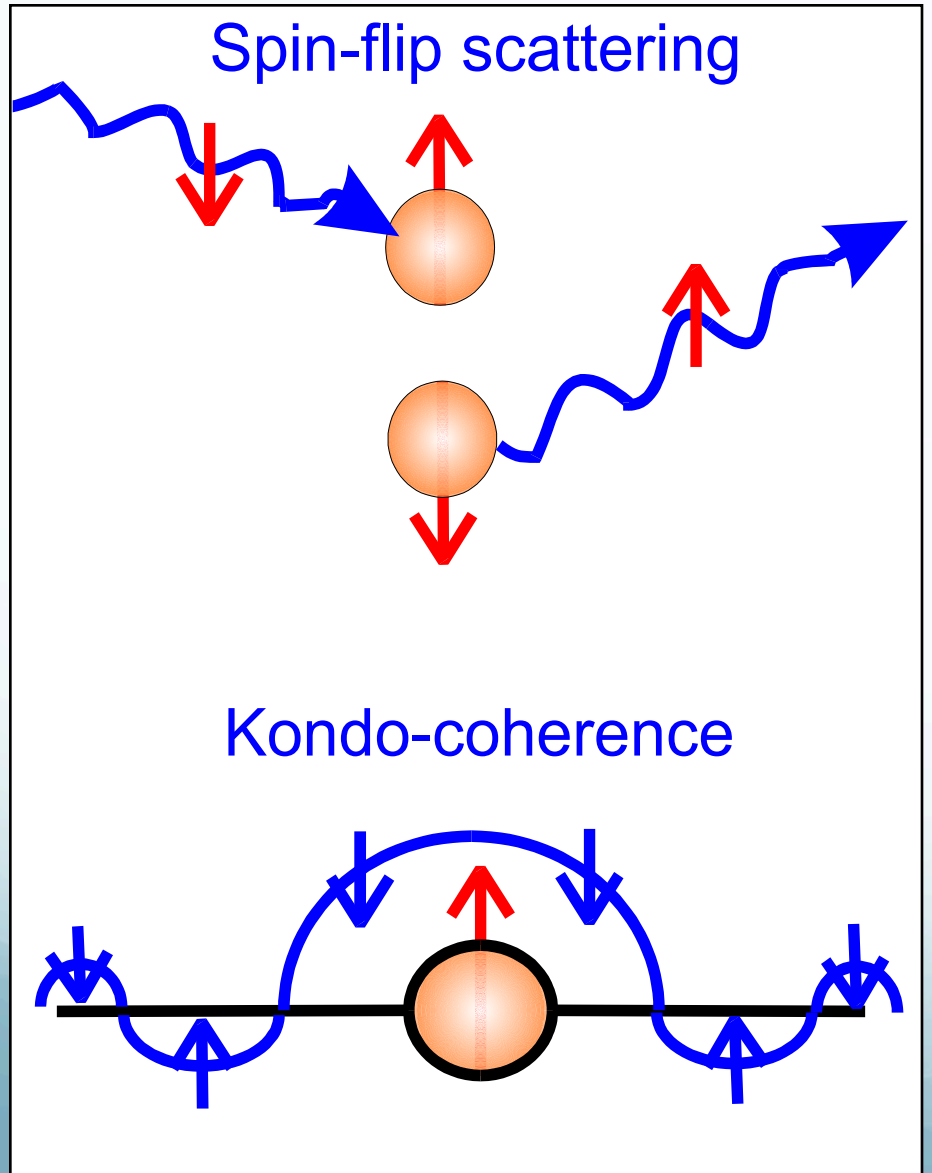


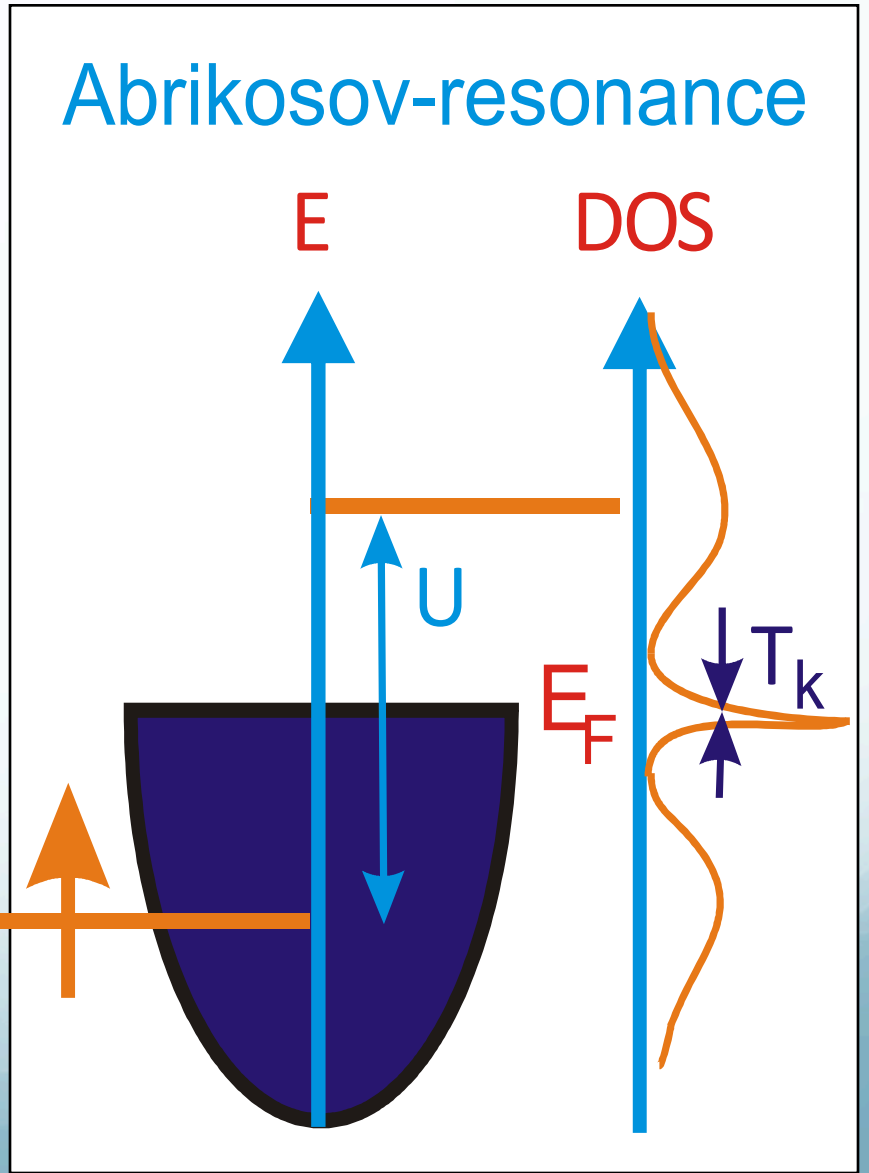
Kondo effect

- 1933 van den Berg: exp.
- 1964 Jun Kondo: theory



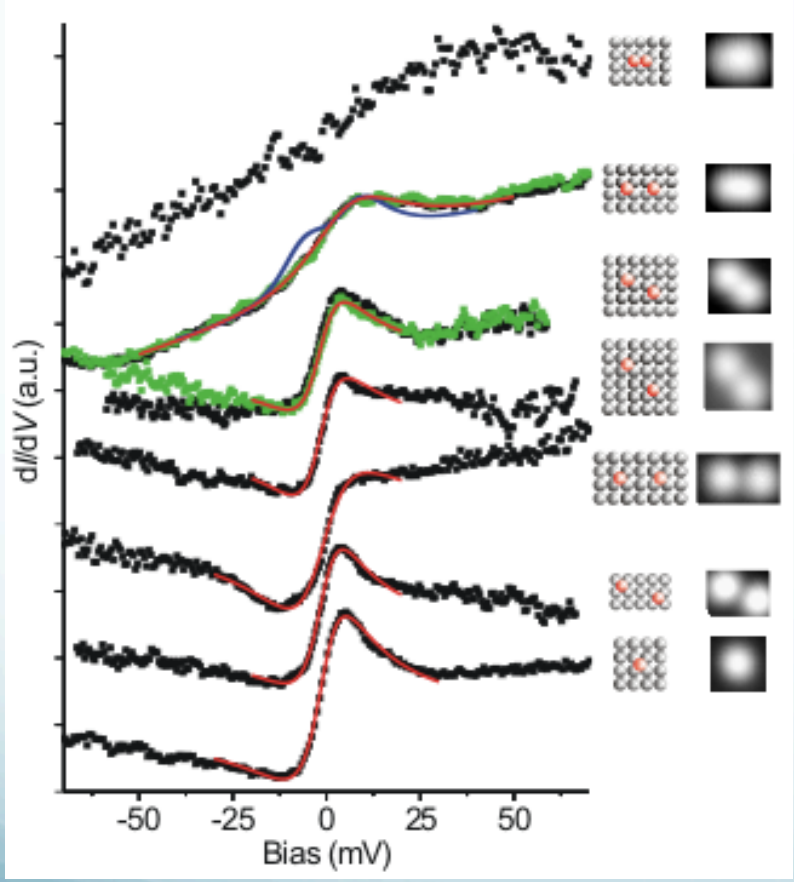
Kondo effect: theory



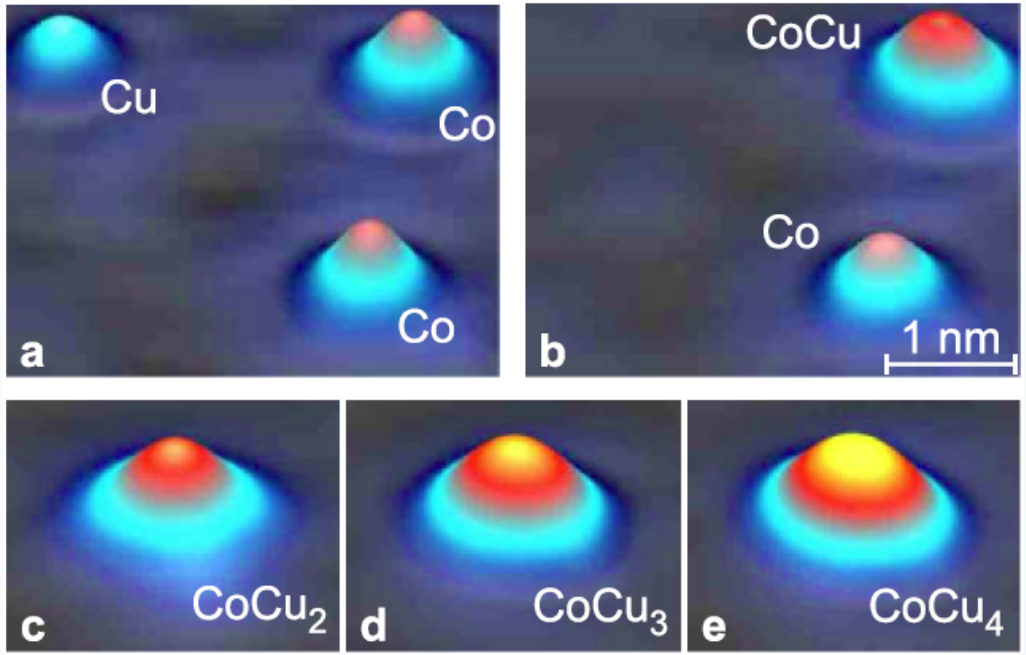


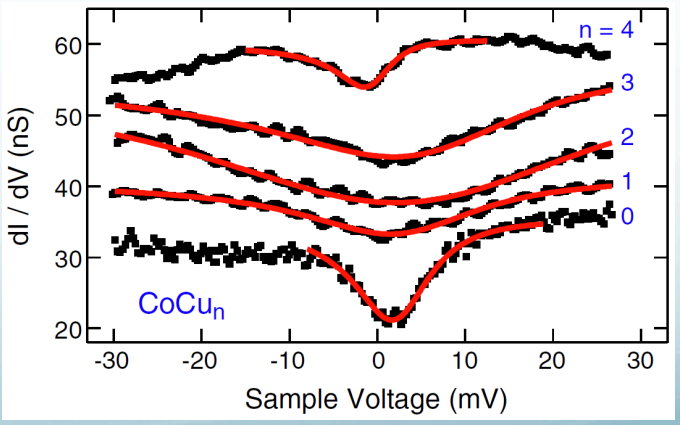
Nanostructures of correlated atoms

Atom manipulation



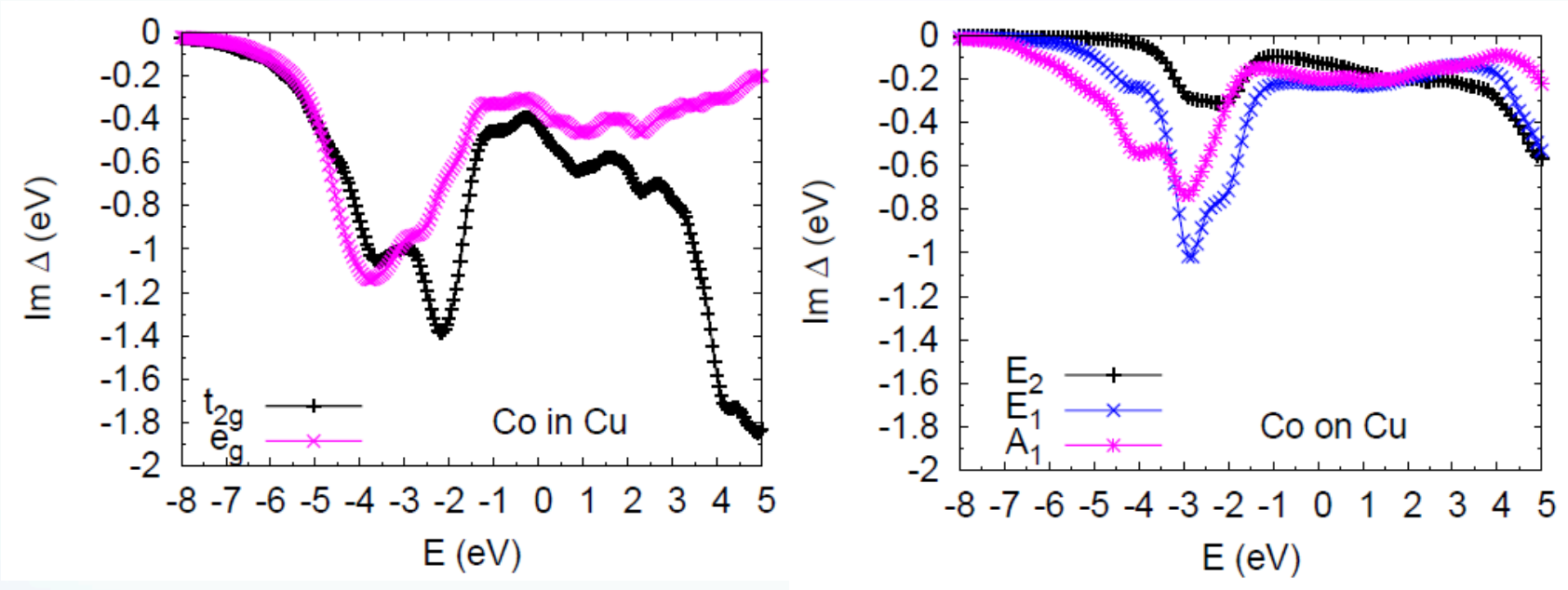
P. Wahl et al., PRL **98**, 056601 (2007)





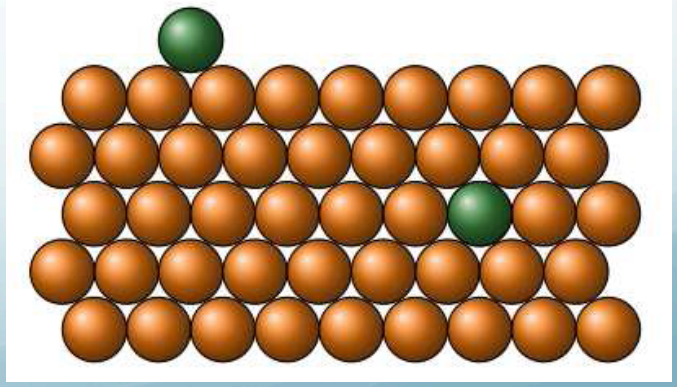
N. Néel , et al., PRL **101**, 266803 (2008)

Hybridization function Co on/in Cu(111)

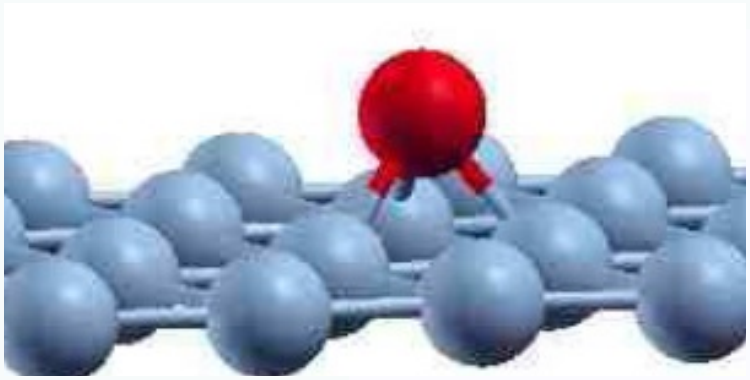


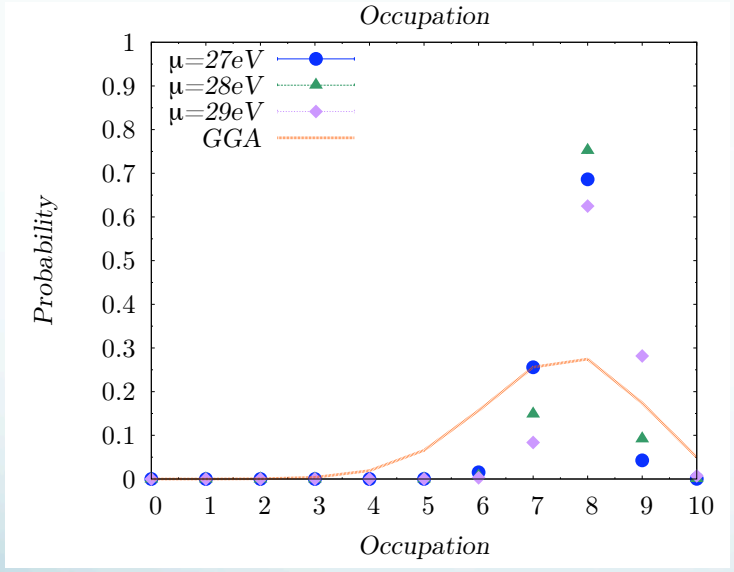
- Hybridization of Co in bulk twice stronger than on surface
- Hybridization in energy range of Cu-d orbitals more anisotropic on surface
- Co-d occupancy: *n*= 7-8

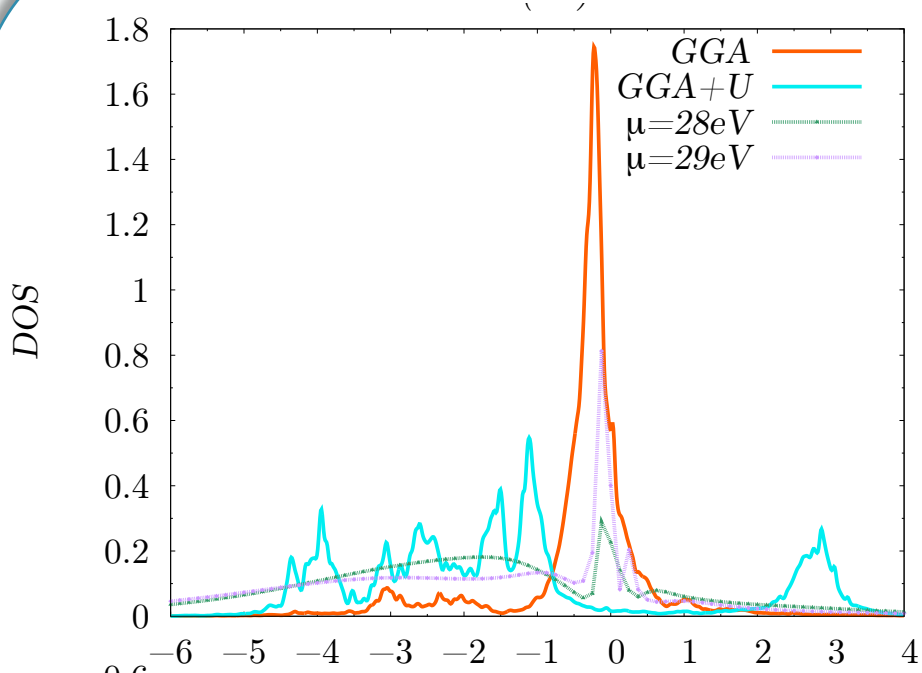
B. Surer, et al PRB (2012)

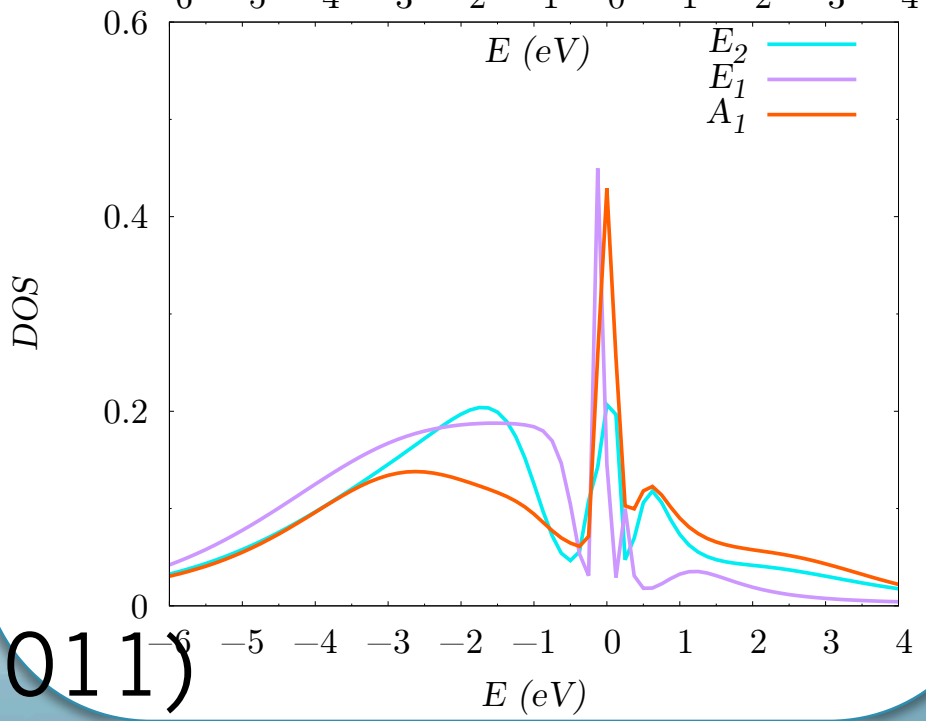


Co on Cu(111)





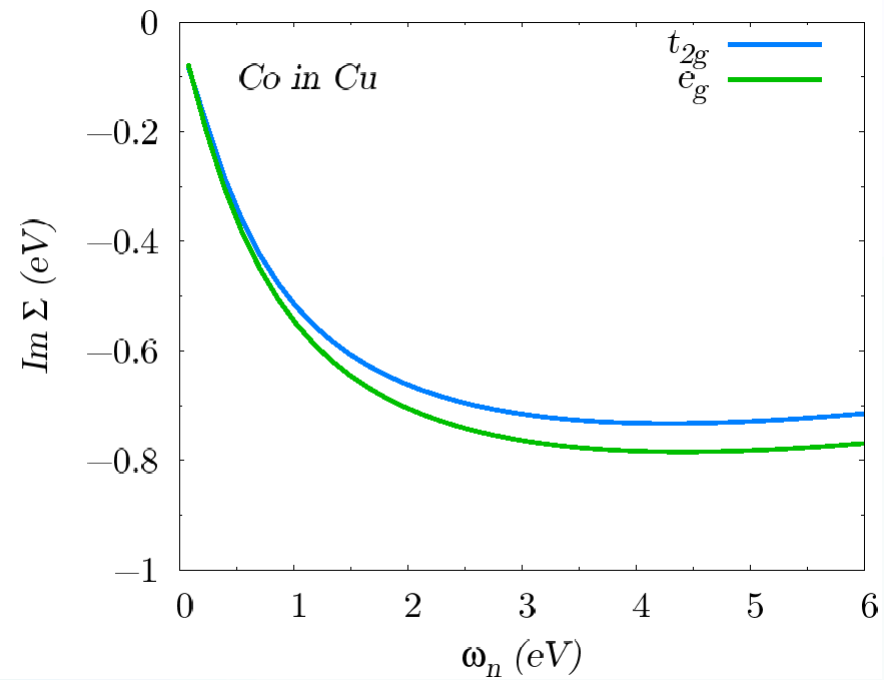


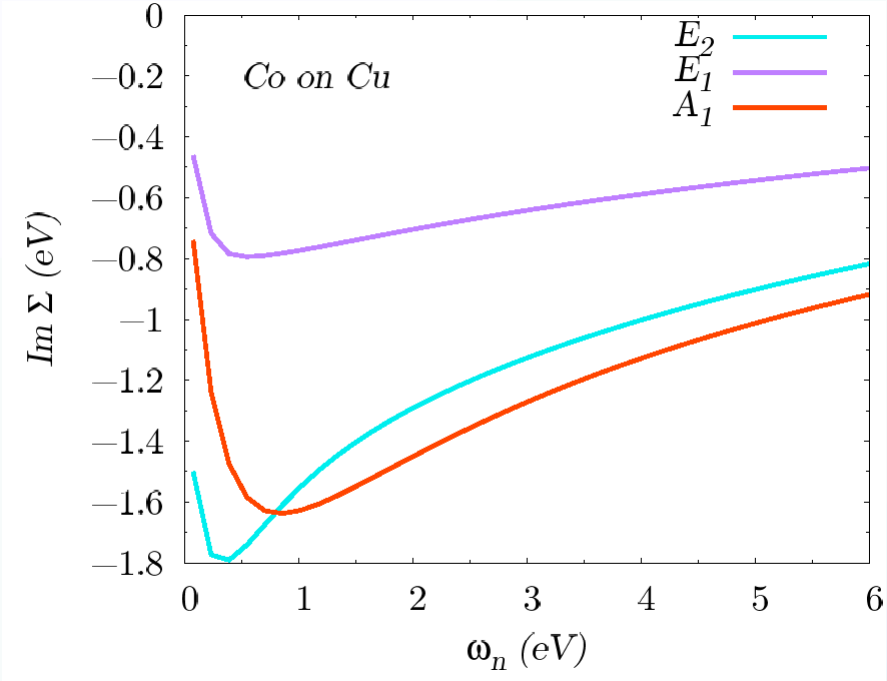


CT-QMC-Krylov

B. Surer, et al. BRB (2011)

Self-energies: Local Fermi liquid





- Fermi liquid: $\Sigma(T,\omega) = \Sigma(T,0) + \Sigma'(T,0)\omega + O(\omega^2)$

$$\mathrm{Im}\,\Sigma(T, i\omega_n) \approx \mathrm{Im}\,\Sigma(T,0) - \mathrm{Im}\,\Sigma'(T,0)\omega_n$$

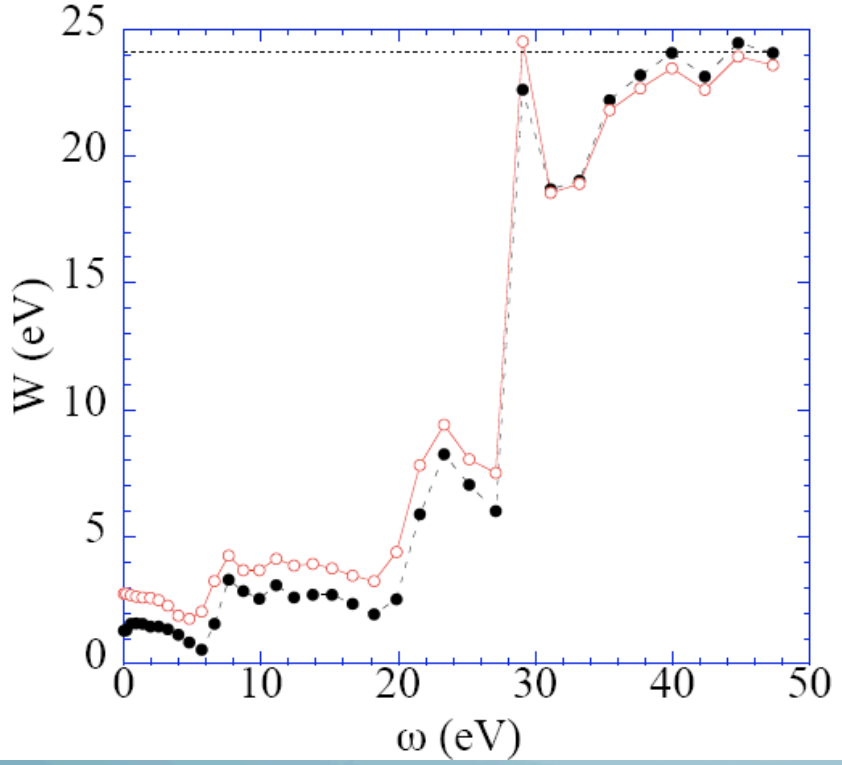
Signatures of low energy Fermi liquids in all orbitals !

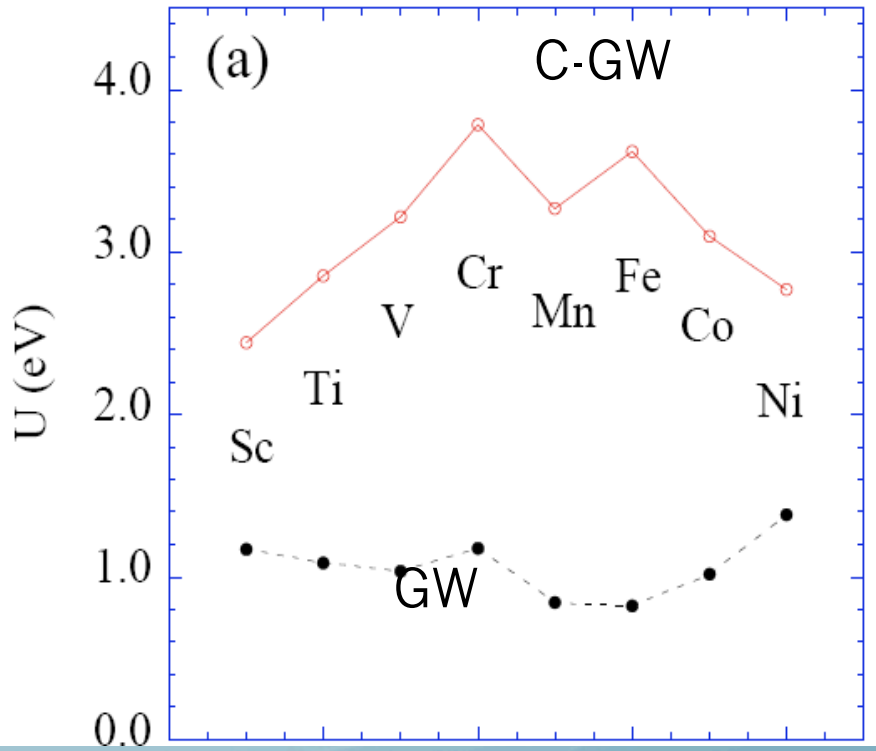
Wannier - GW and effective U(ω)

$$|\varphi_{n\mathbf{R}}\rangle = \frac{V}{(2\pi)^3}\int e^{-i\mathbf{k}\cdot\mathbf{R}}|\psi^{(\mathrm{w})}_{n\mathbf{k}}\rangle d^3k$$

$$|\psi^{(\mathrm{w})}_{n\mathbf{k}}\rangle = \sum_m \mathcal{U}_{mn}(\mathbf{k})|\psi_{m\mathbf{k}}\rangle$$

T. Miyake and F. Aryasetiawan
Phys. Rev. B 77, 085122 (2008)

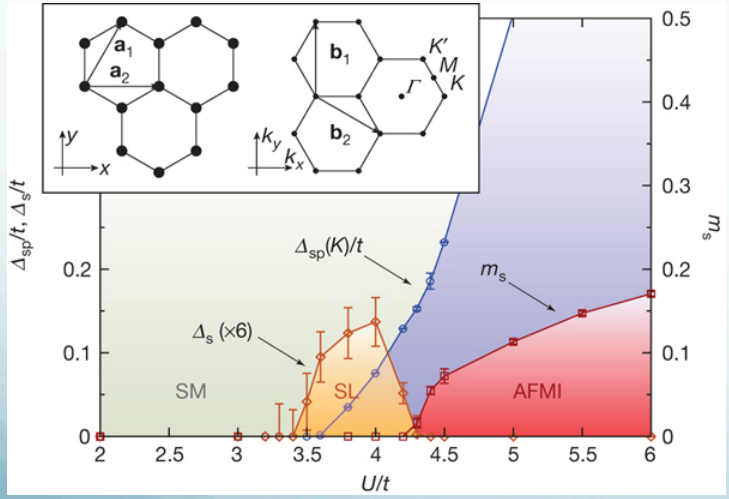




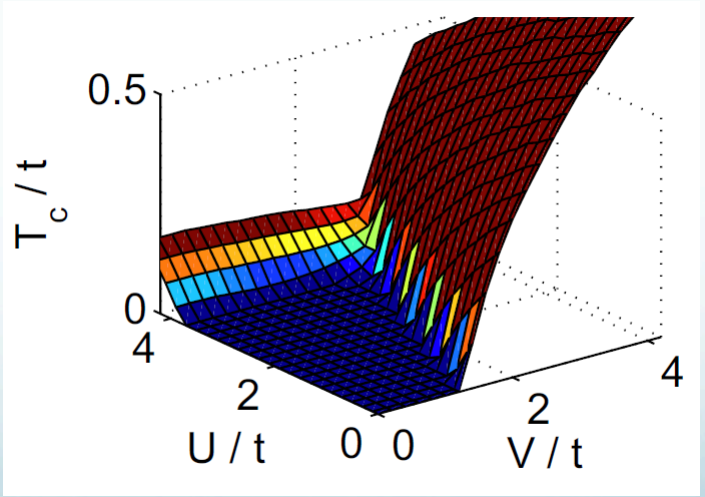
Coulomb interactions in graphene

Generalized Hubbard model

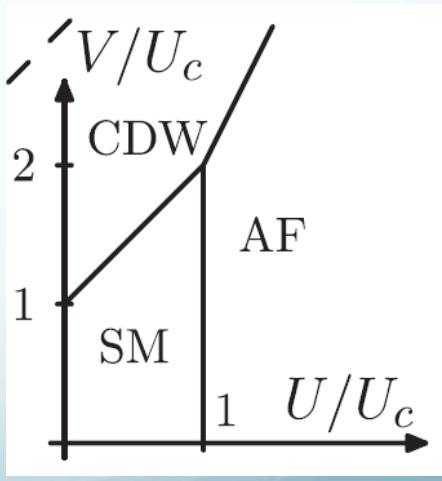
$$\hat{H}_0 = -t \sum_{<\mathbf{i},\mathbf{j}>,\sigma} c^{\dagger}_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma} - t' \sum_{\ll\mathbf{i},\mathbf{j}\gg,\sigma} c^{\dagger}_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma}$$
$$+U_{00} \sum_{\mathbf{i}} n_{\mathbf{i},\uparrow} n_{\mathbf{i},\downarrow} + \frac{1}{2} \sum_{\mathbf{i}\neq\mathbf{j},\sigma,\sigma'} U_{\mathbf{ij}} n_{\mathbf{i},\sigma} n_{\mathbf{j},\sigma'}$$



Meng et al., Nature **464**, 847-851 (2010)



Honerkamp, PRL 100, 146404 (2008)



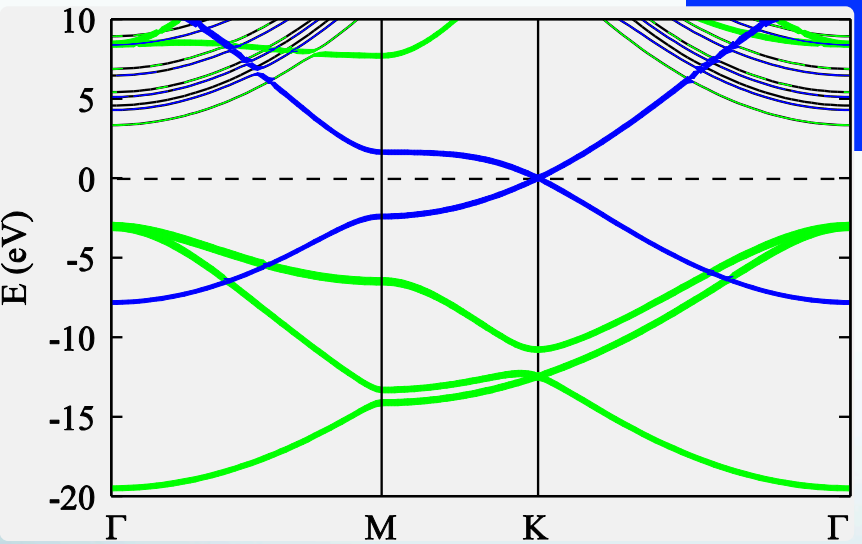
Herbut,
PRL 97, 146401 (2006)

Strength of Coulomb interactions in graphene

Generalized Hubbard model for π-bands only

$$\hat{H}_0 = -t \sum_{<\mathbf{i},\mathbf{j}>,\sigma} c^{\dagger}_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma} - t' \sum_{\ll\mathbf{i},\mathbf{j}\gg,\sigma} c^{\dagger}_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma}$$

$$+U_{00} \sum_{\mathbf{i}} n_{\mathbf{i},\uparrow} n_{\mathbf{i},\downarrow} + \frac{1}{2} \sum_{\mathbf{i}\neq\mathbf{j},\sigma,\sigma'} U_{\mathbf{ij}} n_{\mathbf{i},\sigma} n_{\mathbf{j},\sigma'}$$



π bands (blue) crossing Fermi level

σ bands (green) at higher energies

but all other electrons contribute to screening of the Coulomb in

$$P(\mathbf{r},\mathbf{r}';\omega) = \sum_{i}^{\text{occ}} \sum_{j}^{\text{unocc}} \psi_i(\mathbf{r})\psi_i^*(\mathbf{r}')\psi_j^*(\mathbf{r})\psi_j(\mathbf{r}') \times \left\{ \frac{1}{\omega - \varepsilon_j + \varepsilon_i + i0^+} - \frac{1}{\omega + \varepsilon_j - \varepsilon_i - i0^+} \right\}$$

Polarization function

Partially screened Coulomb interaction *U*

Constrained random phase approximation (cRPA)

Splitting of polarization function

$$P(\mathbf{r},\mathbf{r}';\omega) = \sum_{i}^{\text{occ}} \sum_{j}^{\text{unocc}} \psi_i(\mathbf{r})\psi_i^*(\mathbf{r}')\psi_j^*(\mathbf{r})\psi_j(\mathbf{r}') \times \left\{ \frac{1}{\omega - \varepsilon_j + \varepsilon_i + i0^+} - \frac{1}{\omega + \varepsilon_j - \varepsilon_i - i0^+} \right\}$$

$$P = P_{pz} + P_r$$

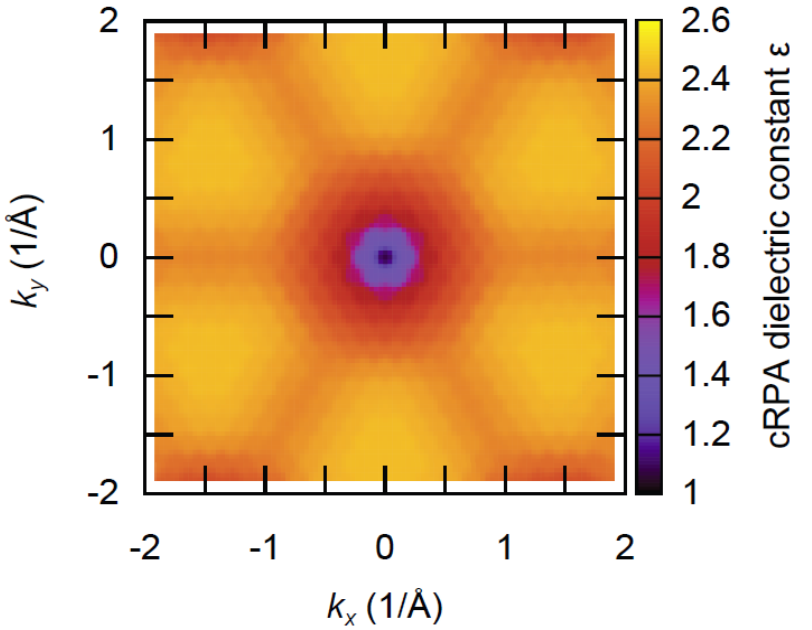
P_{pz}: RPA polarization including only C_{pz}-C_{pz} transitions

P_r: RPA polarization without transitions between C_{pz}-orbitals

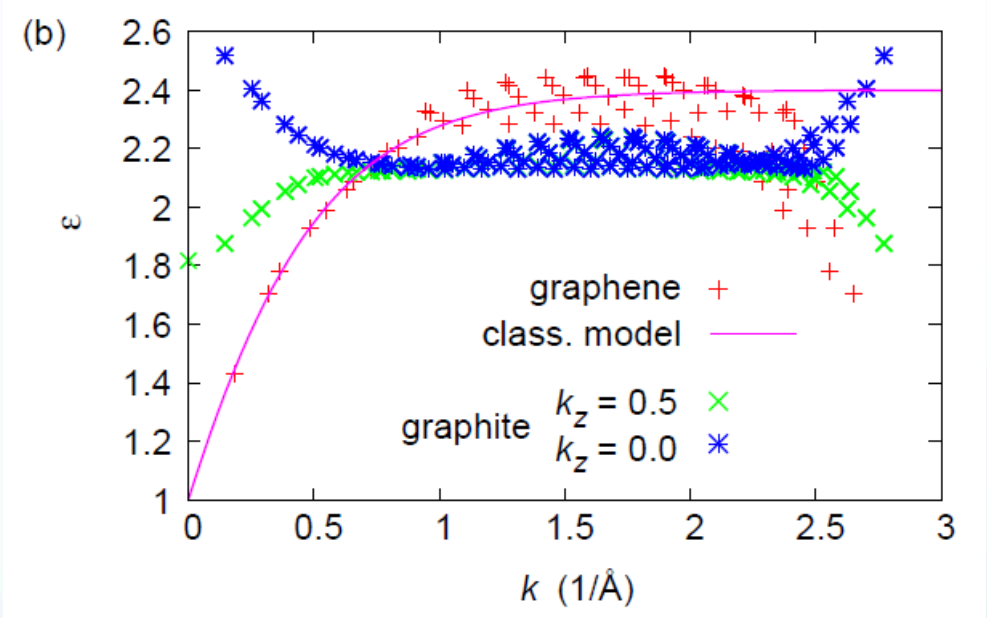
$$U = [1 - vP_r]^{-1} v$$

F. Aryasetiawan et al., PRB 74, 125106 (2006)

Long range tails of Coulomb interactions for effective Hubbard models of graphene and graphite



Static cRPA dielectric ε(**k**) function of graphene in momentum space. (Color coded)



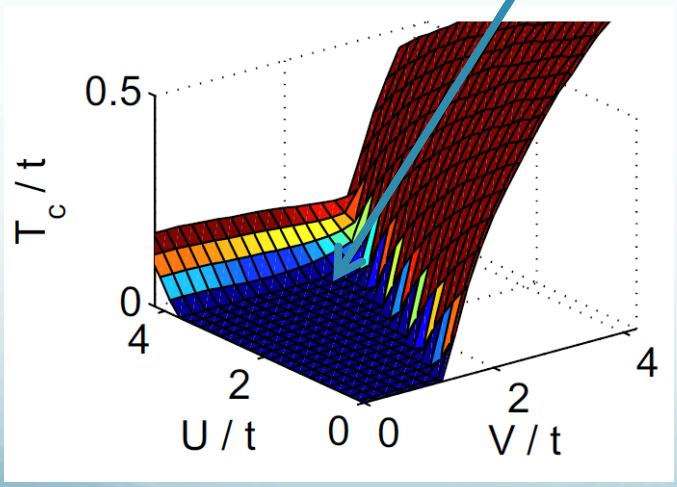
Static cRPA dielectric function ε(|**k**|) f graphene and graphite.

T. Wehling et al., PRL **106**, 236805 (2011)

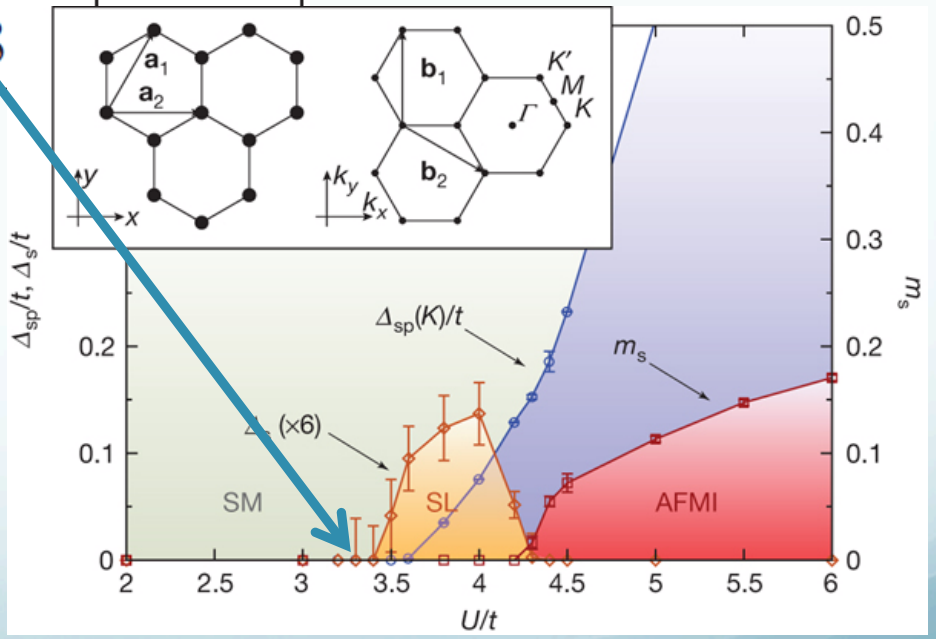
Strength of Coulomb interactions in graphene

	graphene		graphite	
	bare	cRPA	bare	cRPA
U_{00}^{A} (eV)	17.0	9.3	17.5	8.0
U_{00}^{B} (eV)	17.0	9.3	17.7	8.1
U_{01} (eV)	8.5	5.5	8	

TW et al., PRL **106**, 236805 (2011)

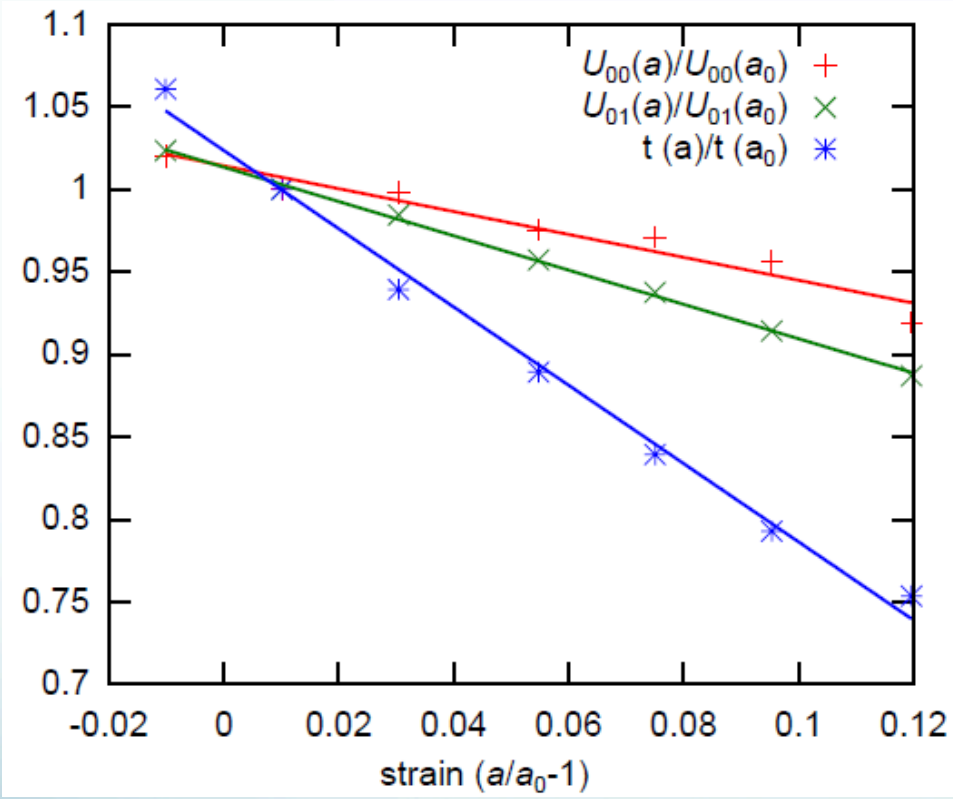


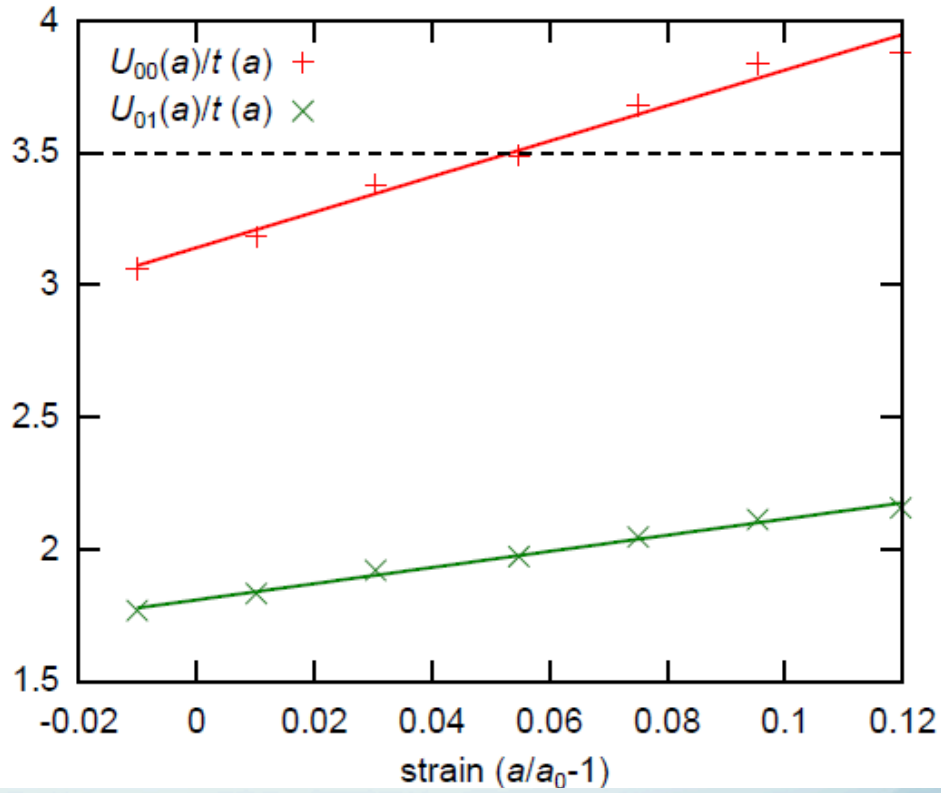
C. Honerkamp, PRL 100, 146404 (2008)



7. Y. Meng et al., Nature **464**, 847-851 (2010)

Effect of strain on Coulomb interactions in graphene



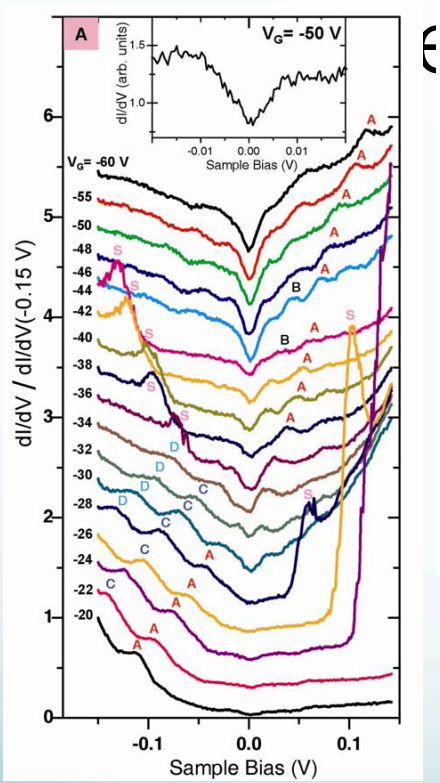


Coulomb interaction U_{ij} and hopping parameters t as function of lattice constant a.
The equilibrium lattice constant is a_0 = 2.47Å.

T. Wehling et al., PRL **106**, 236805 (2011)

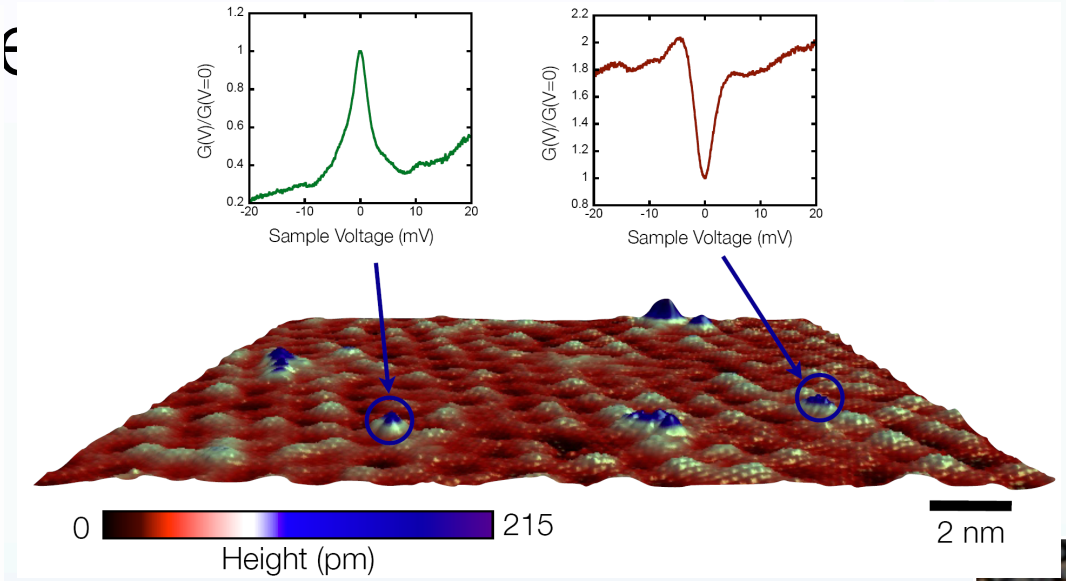
How can graphene's electrons interact with magnetic impurities?

Magnetic ad-atoms on



V. Brar, Nature Phys. 7, 43 (2010)

M. Crommie (U. Berkeley)



H. Manoharan

(U. Stanford)

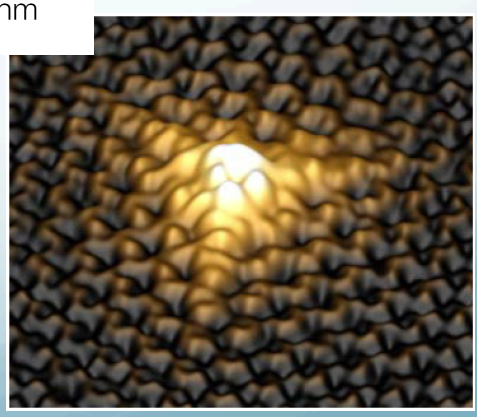
Nature under review

Magnetic lattice defects

STM: M. Ugeda et al., PRL 104, 096804 (2010)

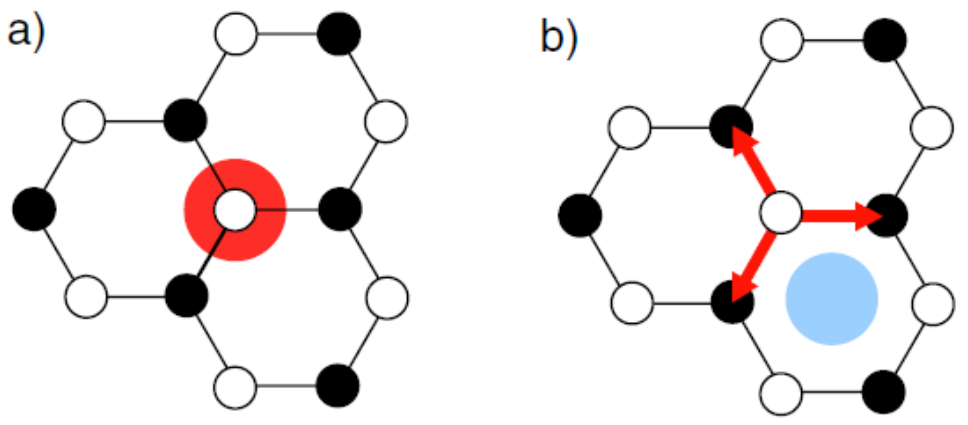
Transport: J. Chen et al., arXiv:1004.3373 (2010)

Theory: T. Wehling et al., PRB 75, 125425 (2007)



Models of spins interacting with graphene

Structure less spin S=½ coupled to different lattice sites



From B. Uchoa et al, PRL **103**, 206804 (2009)

$$\hat{H}_{\mathrm{s-d}} = \sum_{k,\sigma} \epsilon_k c^{\dagger}_{k,\sigma} c_{k,\sigma} + J\mathbf{S}\cdot\mathbf{s}$$

On top:

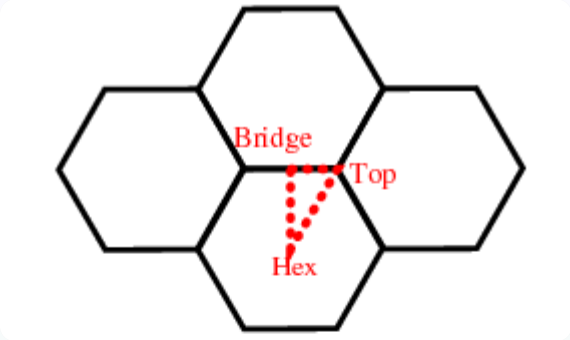
$$\mathbf{s} = \sum_{\alpha,\beta} c^{\dagger}_{0,\alpha} \boldsymbol{\sigma}_{0,\beta} c_{0,\beta}$$

On hex:

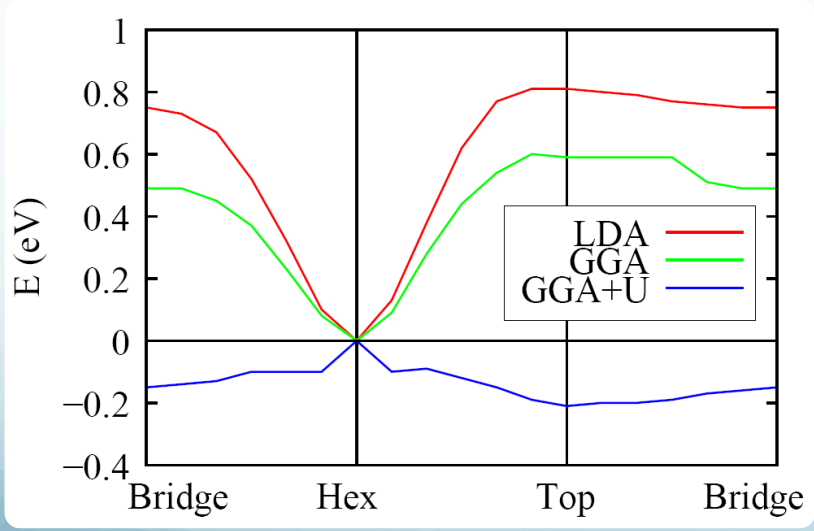
$$\mathbf{s} = \sum_{i\in \mathrm{hex}} \sum_{\alpha,\beta} c^{\dagger}_{i,\alpha} \boldsymbol{\sigma}_{0,\beta} c_{i,\beta}$$

M. Hentschel and F. Guinea, PRB **76,** 115407 (2007).
K. Sengupta and G. Baskaran, PRB **77,** 045417 (2008).
P. S. Cornaglia, G. Usaj, and C. A. Balseiro, PRL **102**, 046801 (2009).
H.-B. Zhuang, Q. feng Sun, and X. C. Xie, EPL **86,** 58004 (2009).
B. Uchoa et al, Phys. Rev. Lett. **103**, 206804 (2009).
L. Dell'Anna, J. Stat. Mech.: Theory Exp. 2010, P01007.

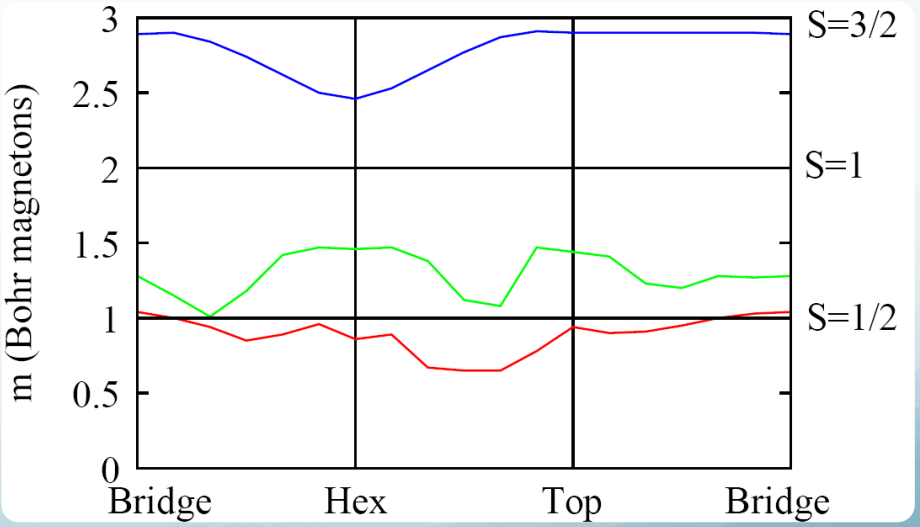
Co ad-atoms: DFT calculations



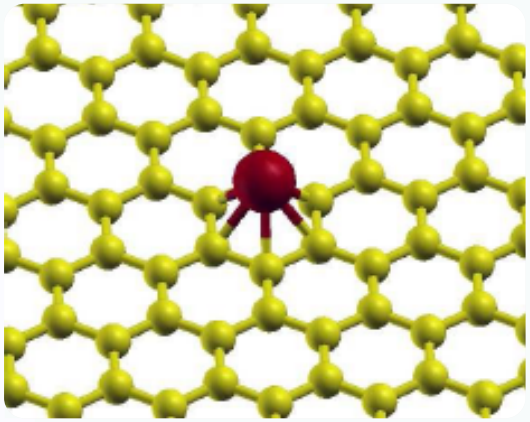
Energy relative to hex-site



Magnetic moment of supercell

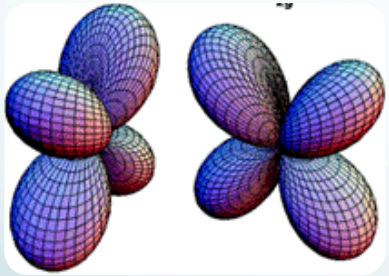


Symmetry analysis

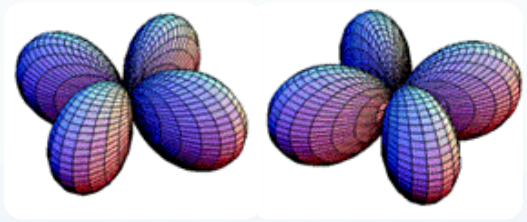


Adatoms at h-site:

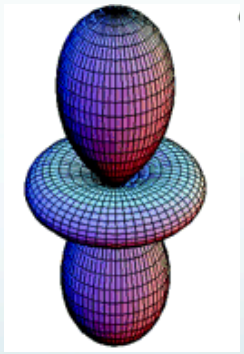
- irreducible representations of C_{6v} point symmetry
  - Atomic *d*-orbitals
    $= E_1 + E_2 + A_1$
  - Graphene Dirac electrons – $E_1 + E_2$



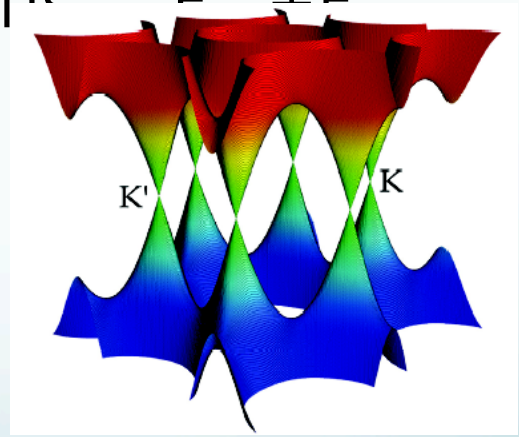
E_1 (d_{xz}, d_{yz}); $|l_z|=1$



E_2 (d_{xy}, $d_{x^2-y^2}$); $|l_z|=2$



A_1 (d_{z^2}); $l_z=0$



T. Wehling et al., PRB **81**, 115427 (2010)

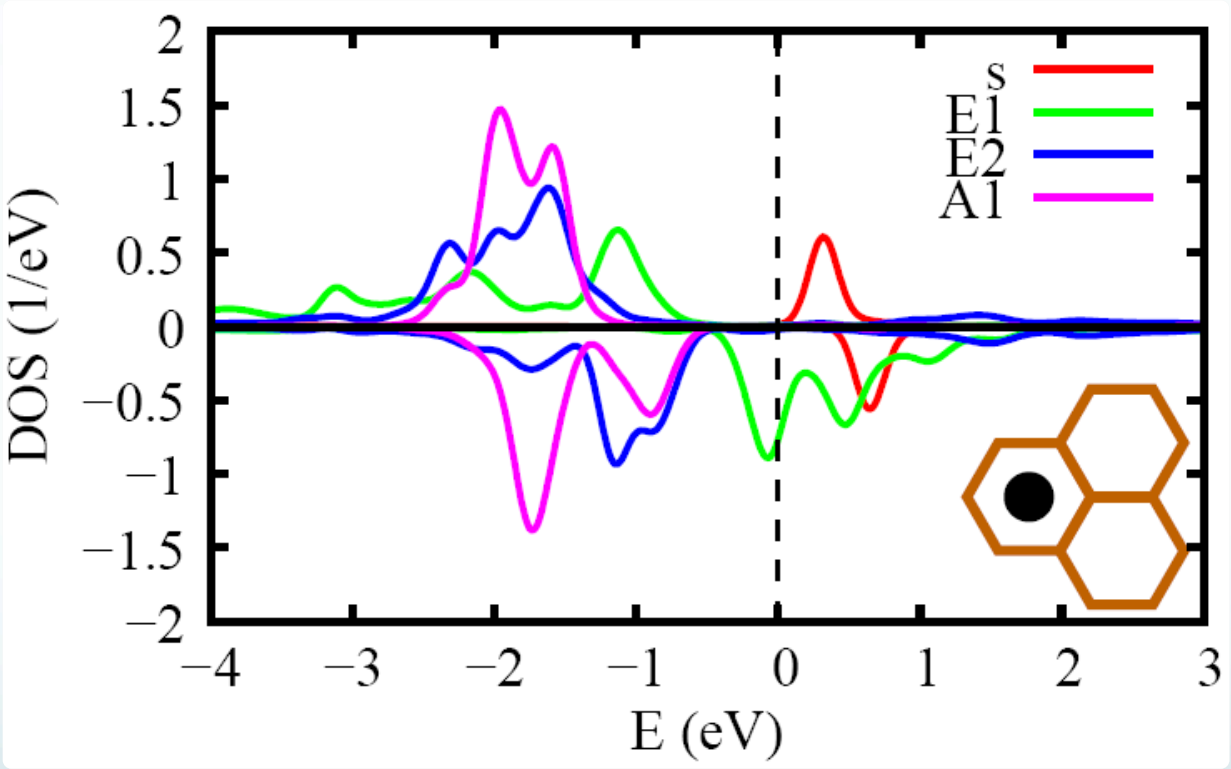
Co on graphene at h-site: electronic structure

S=1/2

$E_1=d_{xz}, d_{yz}$

$E_2=d_{xy}, d_{x^2-y^2}$

$A_1=d_{3r^2-z^2}$



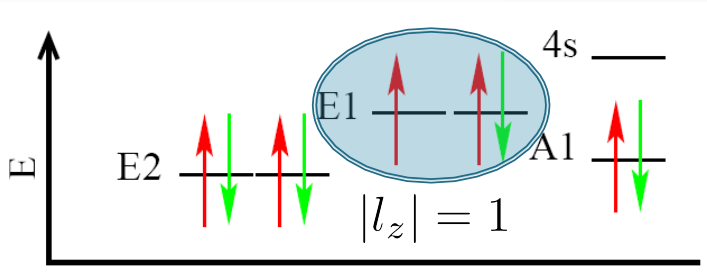
TW et al., PRB 81, 115427 (2010)

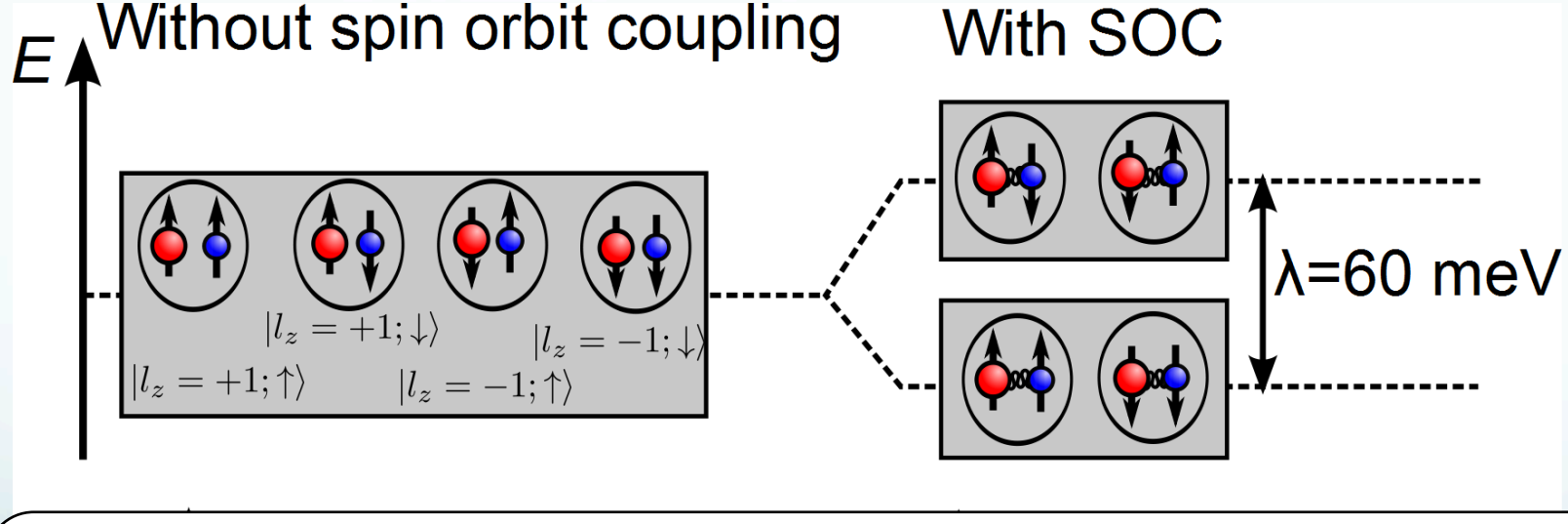
Spin and orbital resolved density of states at Co site from GGA+U (U=2eV, J=0.9eV)

Kondo effect: Co at h-site

Energy level diagram

$E_2=d_{xy}$, $d_{x^2-y^2}$; $E_1=d_{xz}$, d_{yz};
$A_1=d_{3r^2-z^2}$





SU(4)-Kondo above scale of λ, SU(2) below

Realistic models of correlated impurity

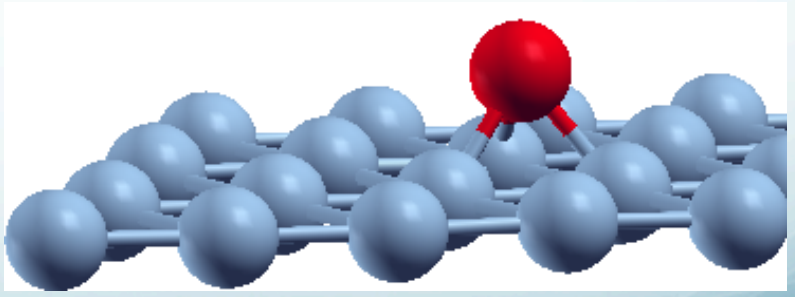
Anderson multiorbital impurity model: U_{1234}

$$\hat{H}_{\mathrm{AIM}} = \sum_{k,\sigma} \epsilon_k c^{\dagger}_{k,\sigma} c_{k,\sigma} + \sum_{k,m,\sigma} \left(V_{km} c^{\dagger}_{k,\sigma} d_{m,\sigma} + \mathrm{H.c.} \right) + \hat{H}_{\mathrm{at}}$$

$$\hat{H}_{\mathrm{at}} = \sum_{m,\sigma} \epsilon_{d,m} d^{\dagger}_{m,\sigma} d_{m,\sigma} + \frac{1}{2} \sum_{\substack{m,m,'m'',m''' \\ \sigma,\sigma'}} U_{mm'm''m'''} d^{\dagger}_{m,\sigma} d^{\dagger}_{m',\sigma'} d_{m'',\sigma'} d_{m''',\sigma}$$

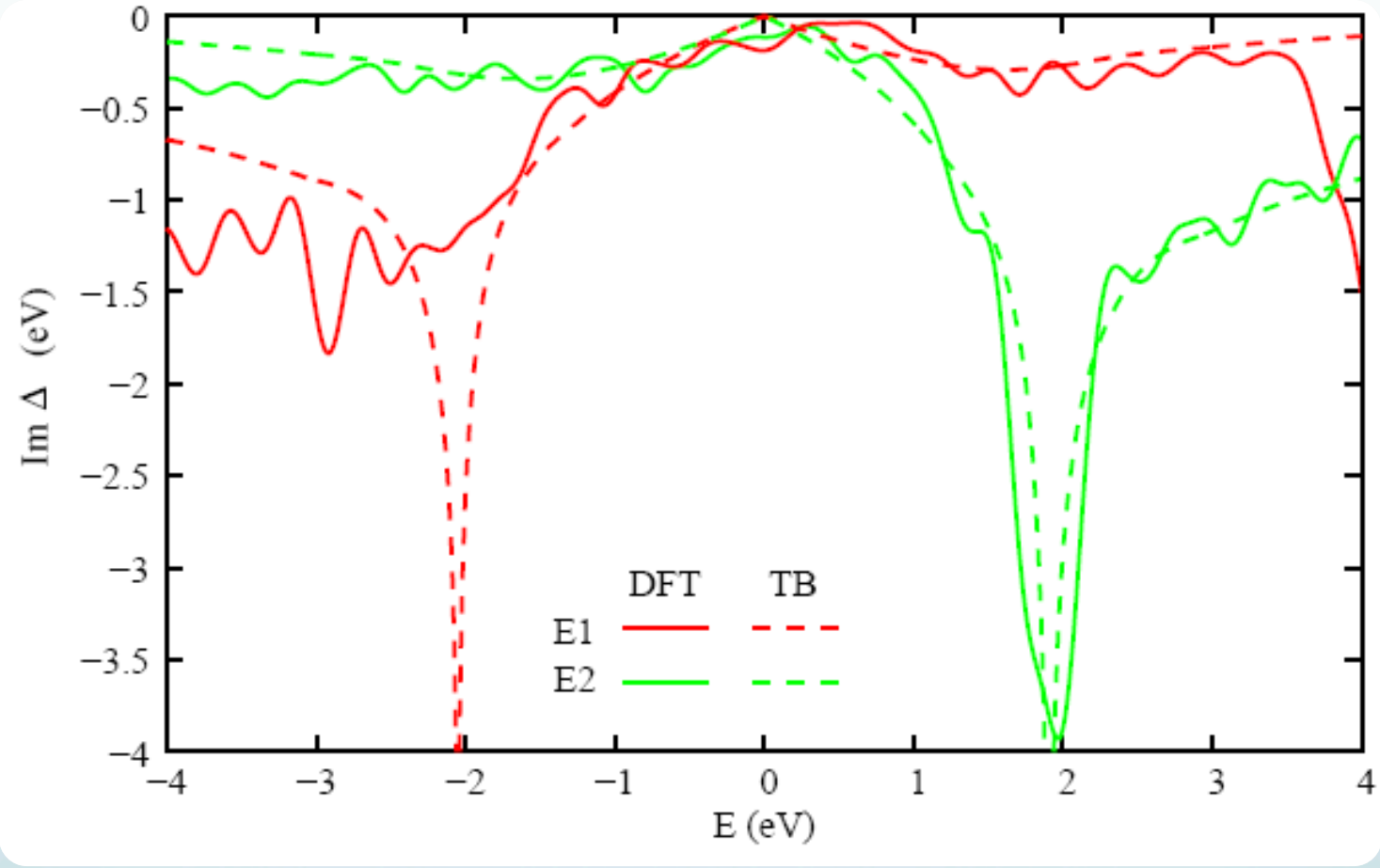
Hybridization function

$$\Delta_{mm'}(\omega) = \sum_{k} \frac{V^{*}_{km} V_{km'}}{\omega + i0^{+} - \epsilon_k}$$



Interface of DFT to many body methods

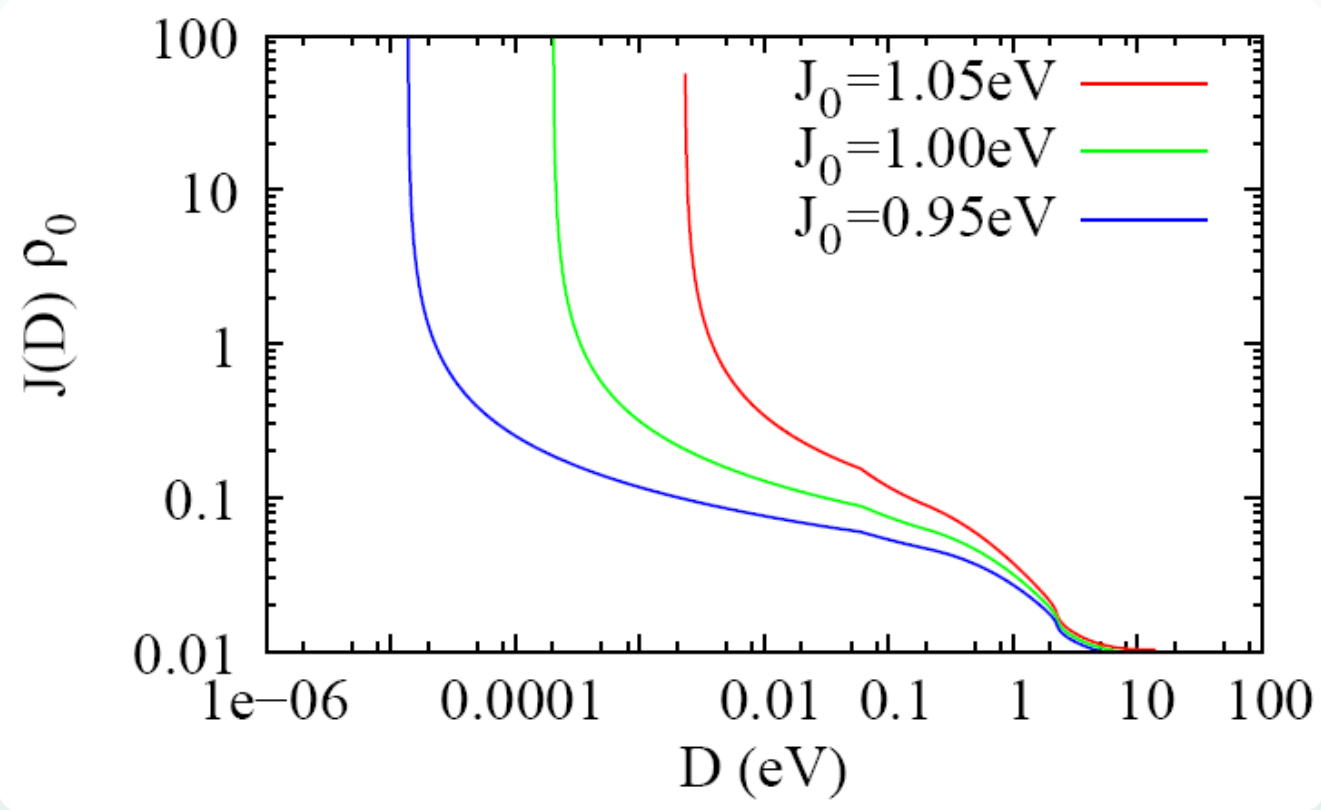
Co at h-site: hybridization functions



Particle hole asymmetry of hybridization functions due to coupling to vHS

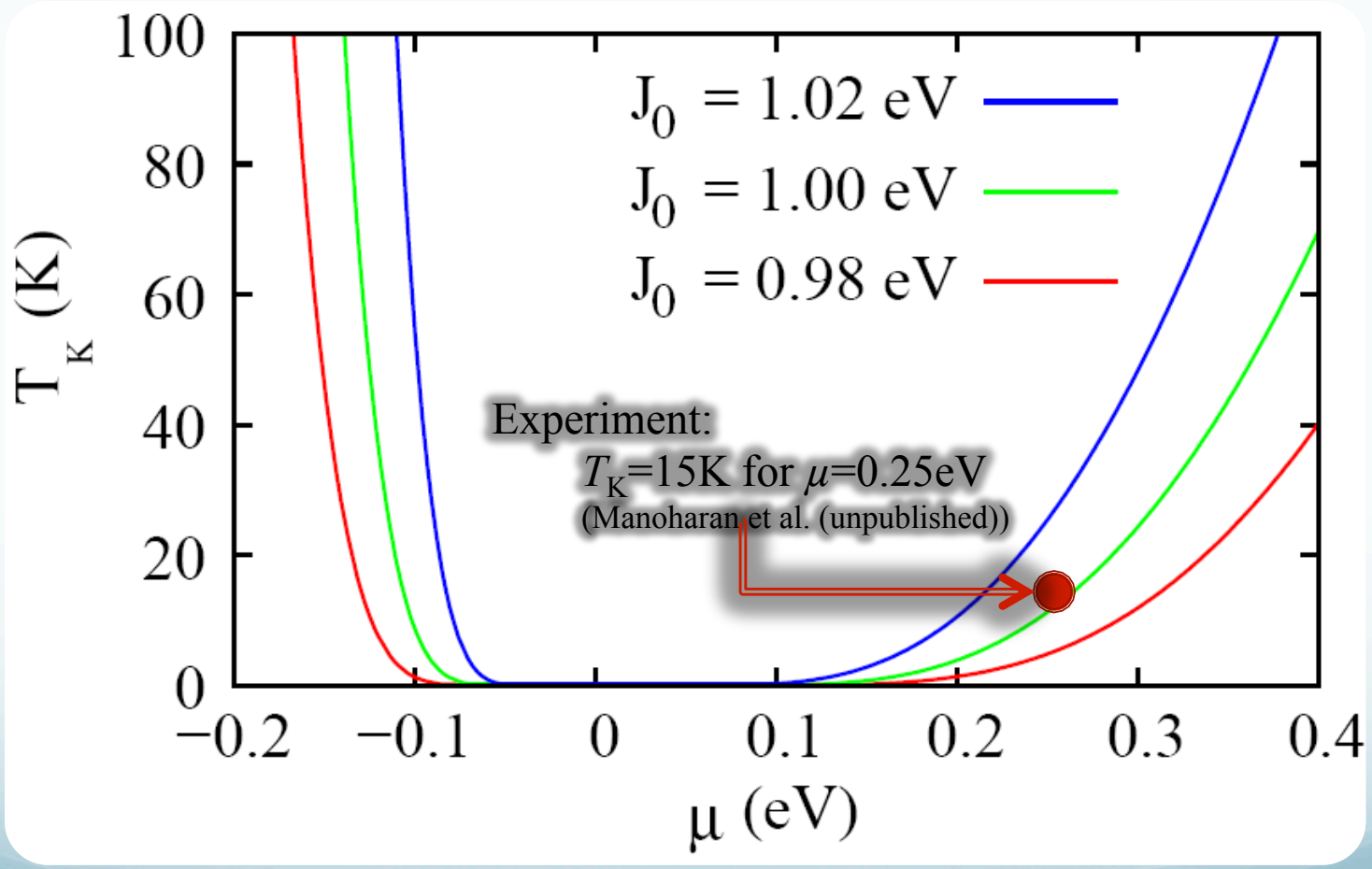
T. Wehling et al., PRB **81**, 115427 (2010)

RG flow of effective coupling



- 10% change in bare coupling J_0 changes T_K by factor > 10
- Proximity to quantum critical point: $J_c \approx 1.1$eV

Kondo temperatures for Co at h-site



T. Wehling et al., Phys. Rev. B 81, 115427 (2010)

J_c=1.1 eV quantum critical point

Summary

- CT-QMC describe local correlations effect
- DMFT is the simplest approach for real system
- DF: non-local correlation effects in nanosystem