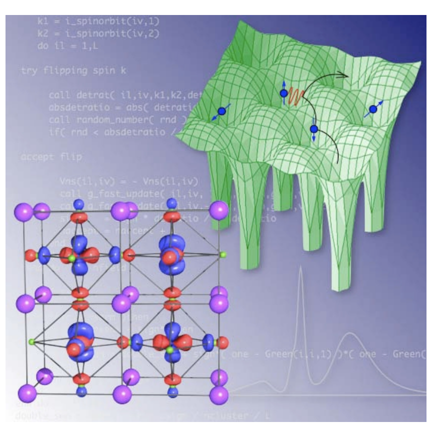
Surprising effect of electronic correlations in band insulators

Jan Kuneš and Vlastimil Křápek

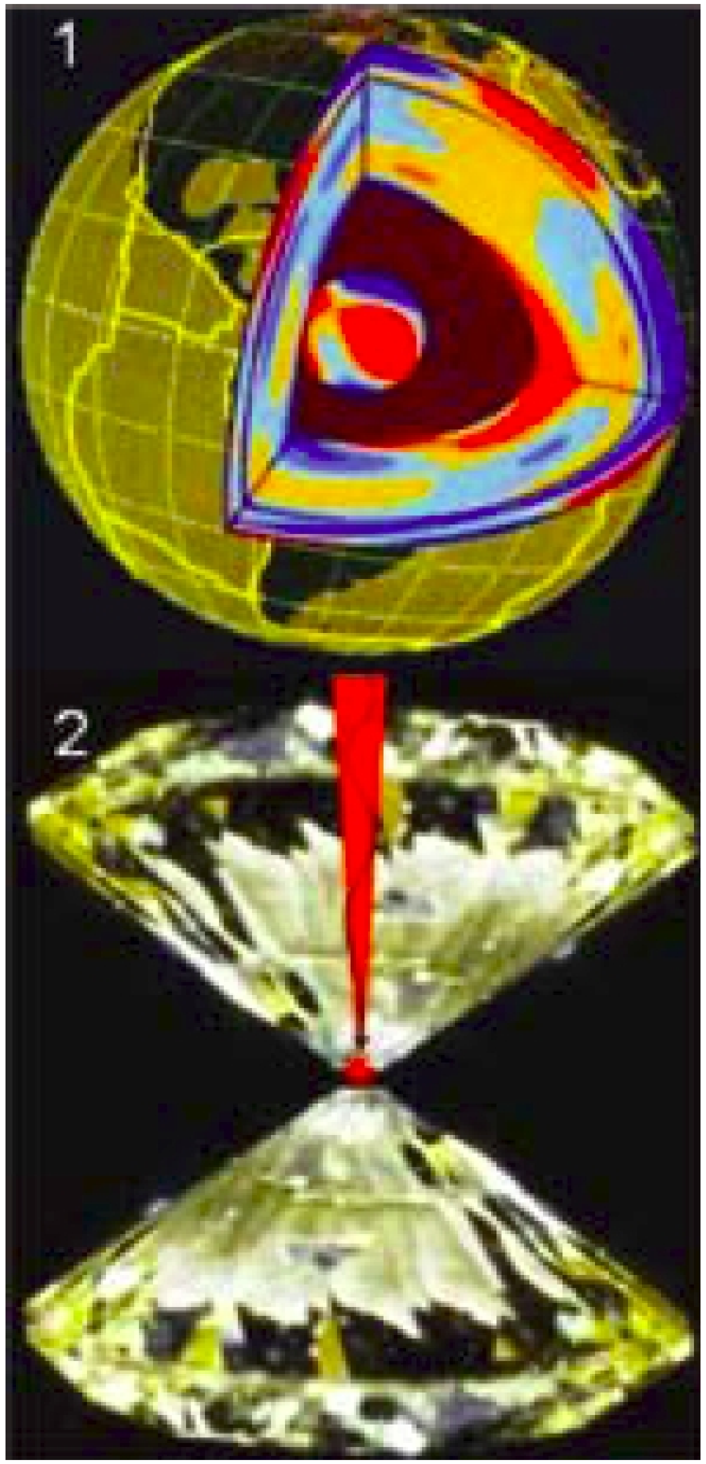
Institute of Physics AS CR



DFG FOR 1346
Dynamical Mean-Field Approach with Predictive Power for Strongly Correlated Materials
&
GA CR P204/10/0284

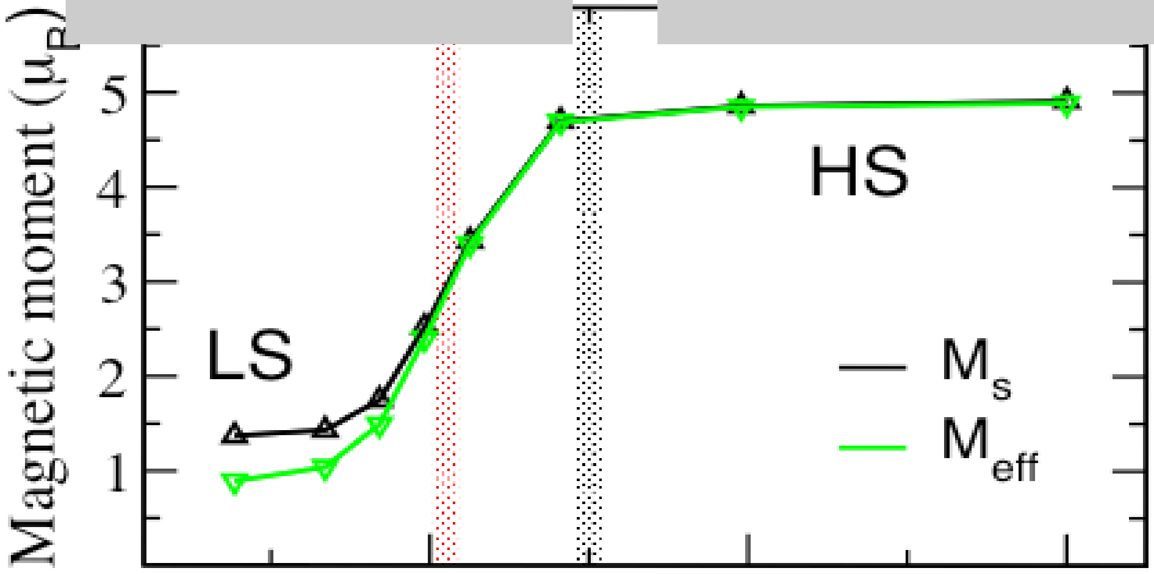
TM oxides under high pressures

Simultaneous moment collapse metal-insulator transition



LOW SPIN

HIGH SPIN

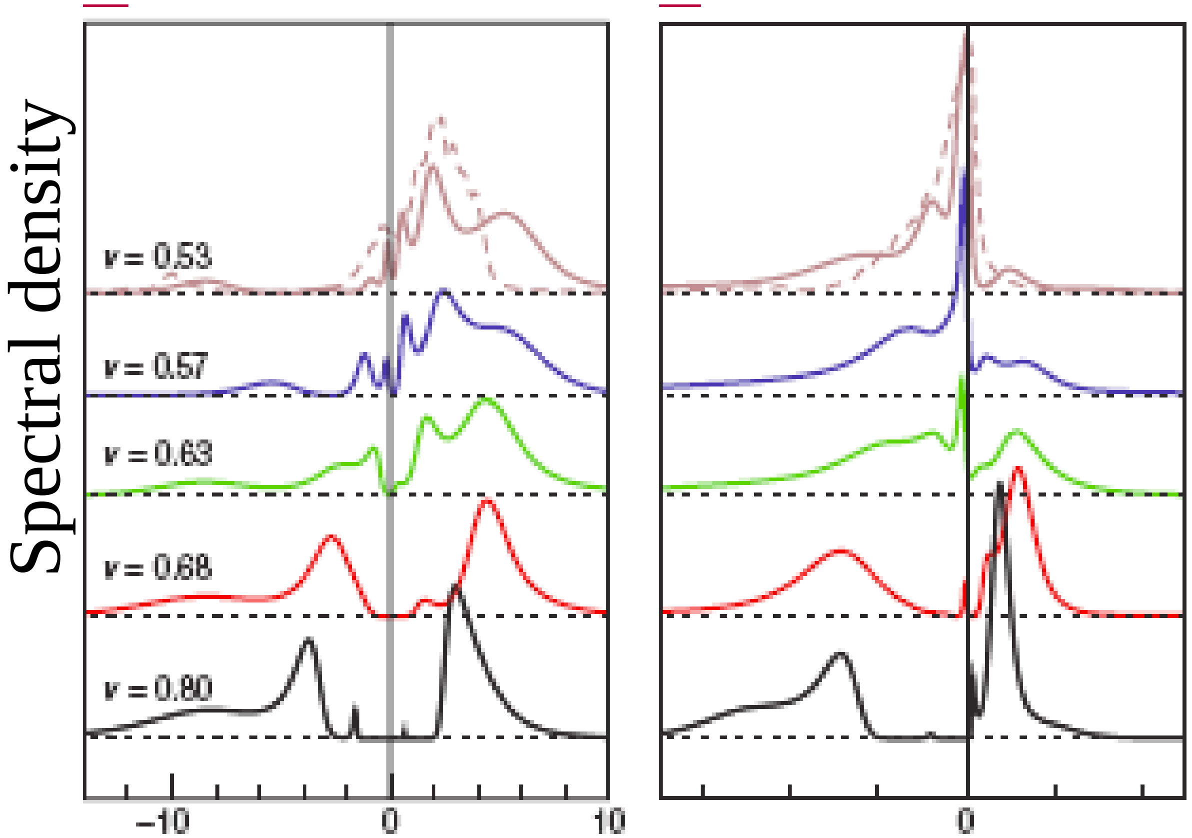


MnO

α-Fe_2O_3

PRESSURE

METAL

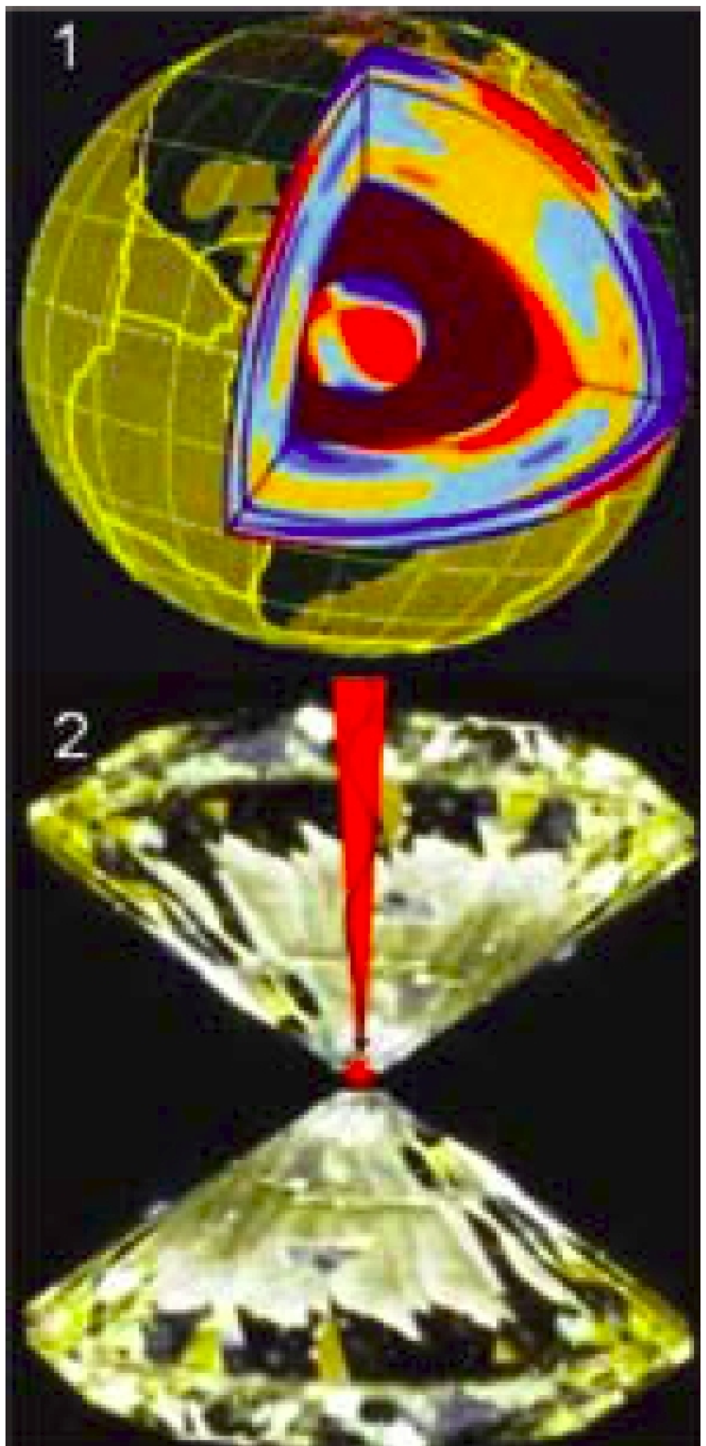


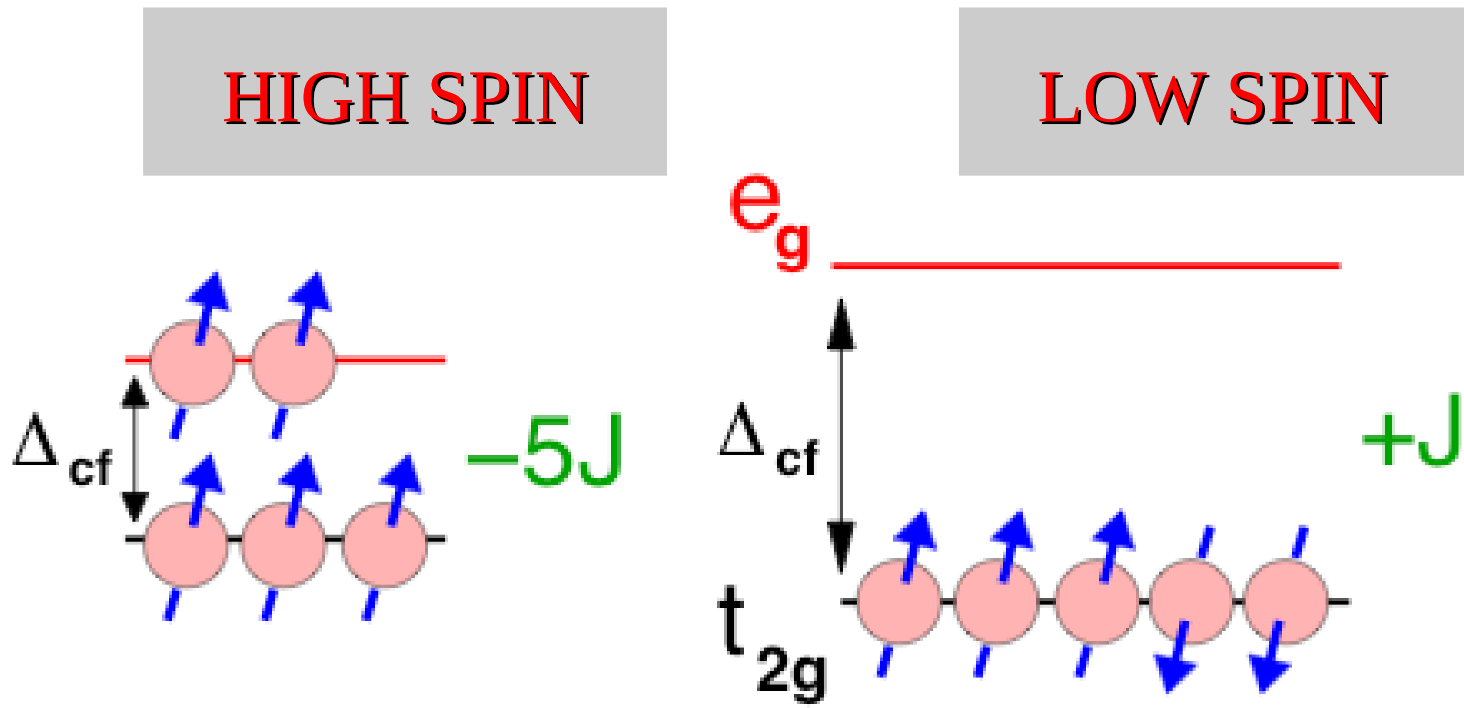
PRESSURE

INSULATOR

TM oxides under high pressures

Simultaneous moment collapse metal-insulator transition



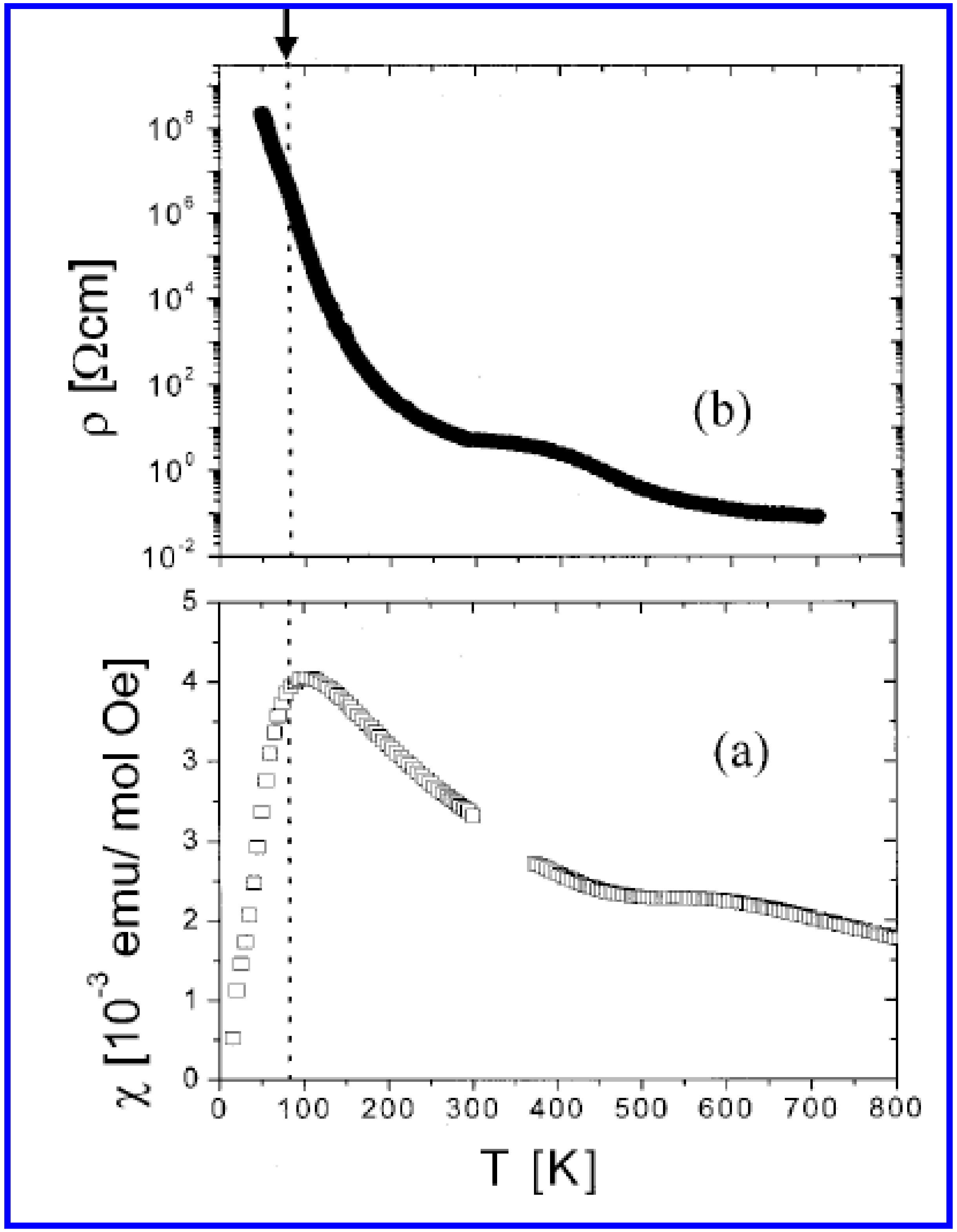


What happens right at the transition?

Outline

- $LaCoO_3$
- Dynamical mean-field theory
- HS-LS transitions in 2-band model
- HS-LS order on bipartite lattice
- Blume-Emery-Griffiths model for fermionic systems
- From cobaltites to manganites
- Conclusions

$LaCoO_3$

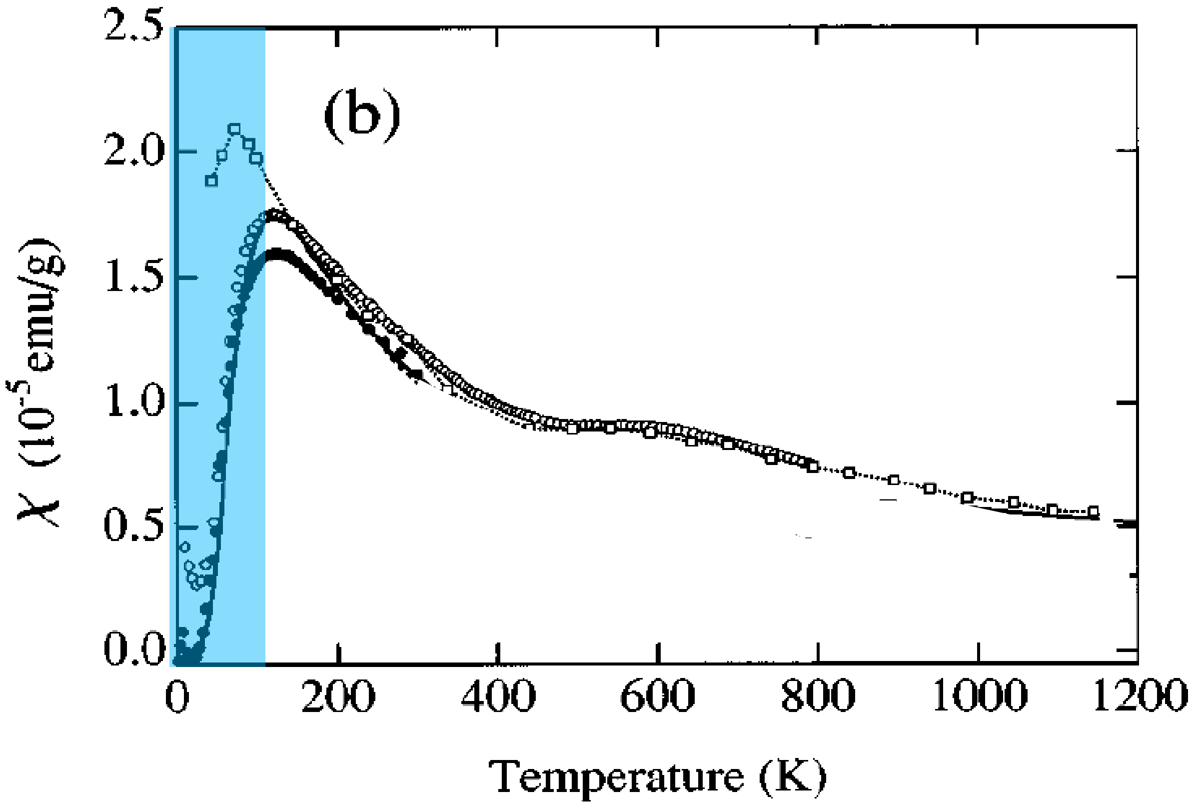


Resistivity

Magnetic susceptibility

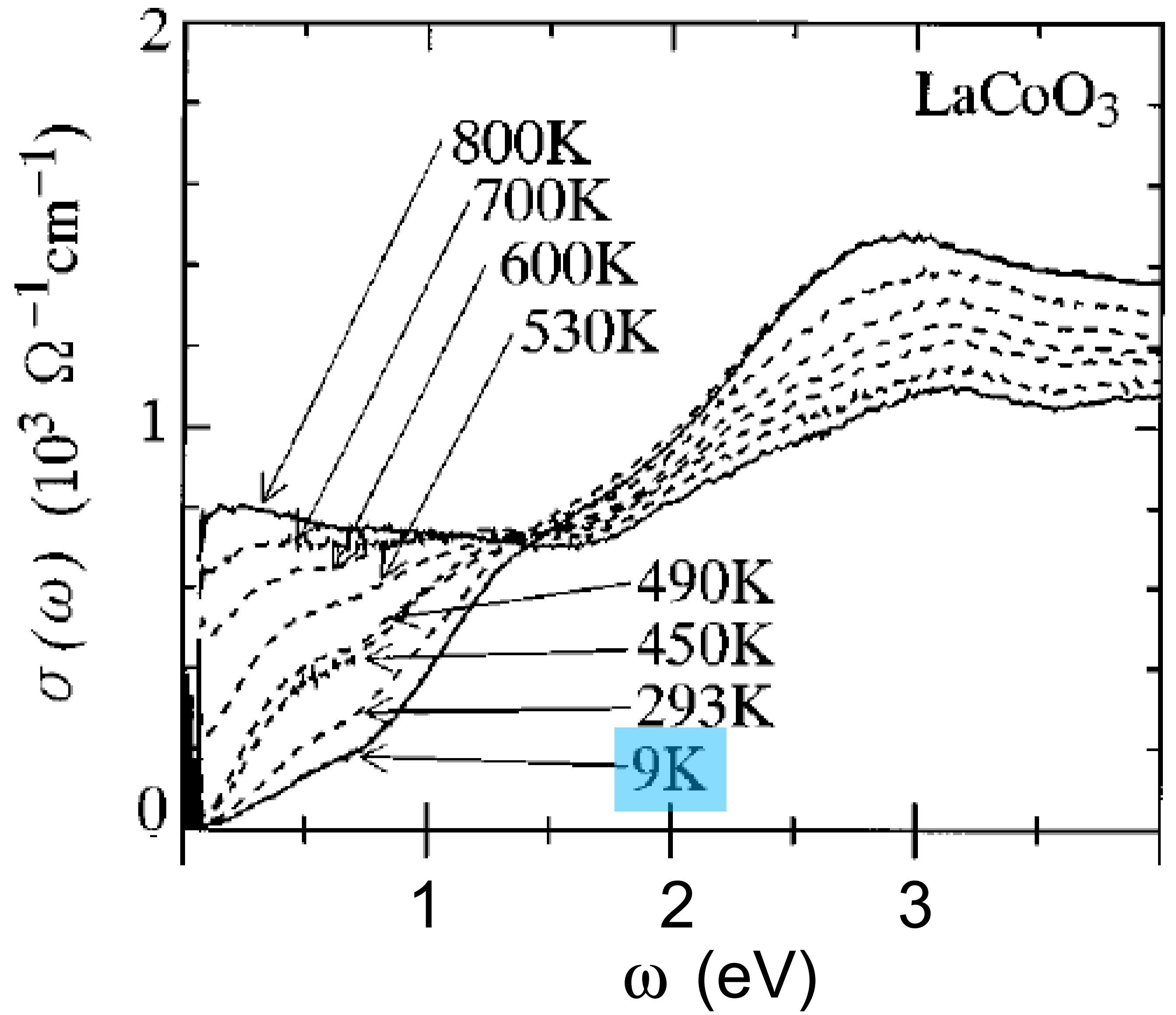
*English et al. Phys. Rev. B **65**, 220407 (2002)*

$LaCoO_3$



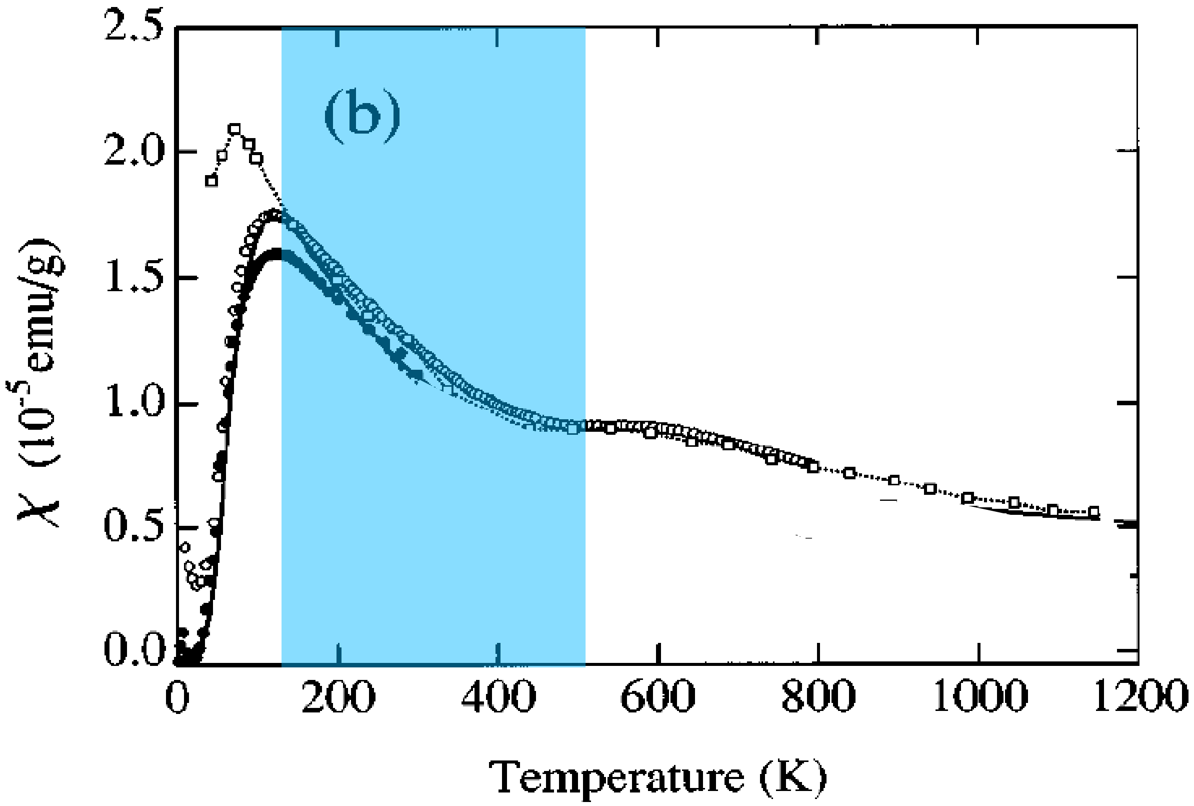
Saitoh et al. Phys. Rev. B ***55,*** *4257 (1997)*

Optical conductivity



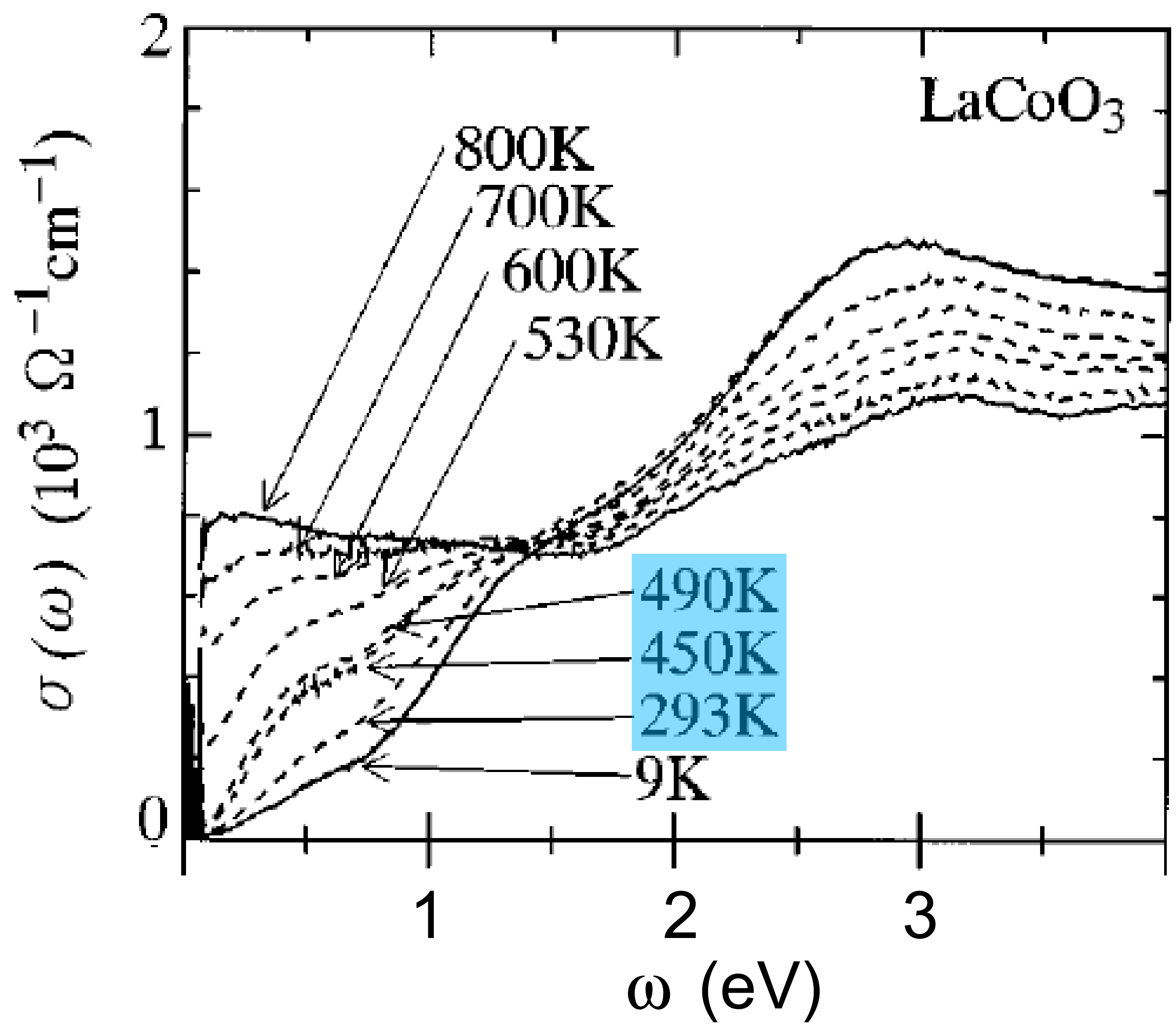
Tokura et al. Phys. Rev. B ***58****, R1699 (1998)*

$LaCoO_3$



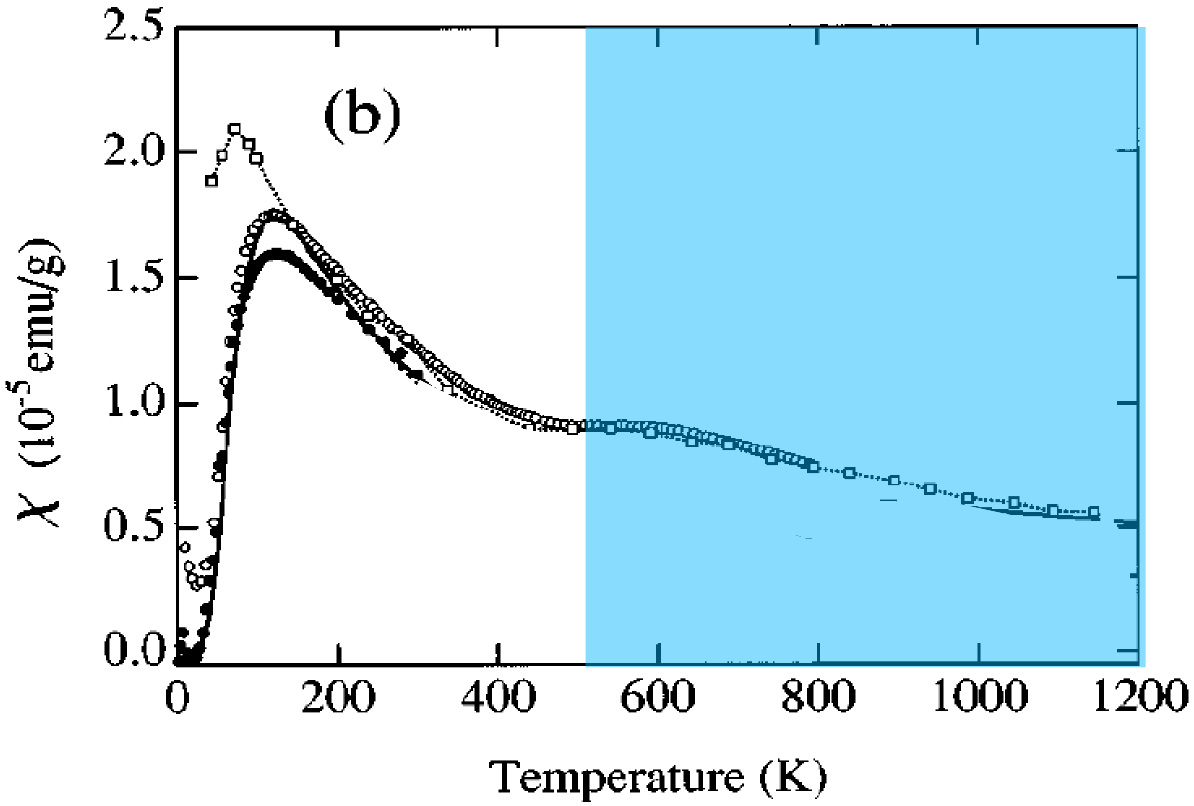
Saitoh et al. Phys. Rev. B ***55,*** *4257 (1997)*

Optical conductivity



Tokura et al. Phys. Rev. B ***58****, R1699 (1998)*

$LaCoO_3$



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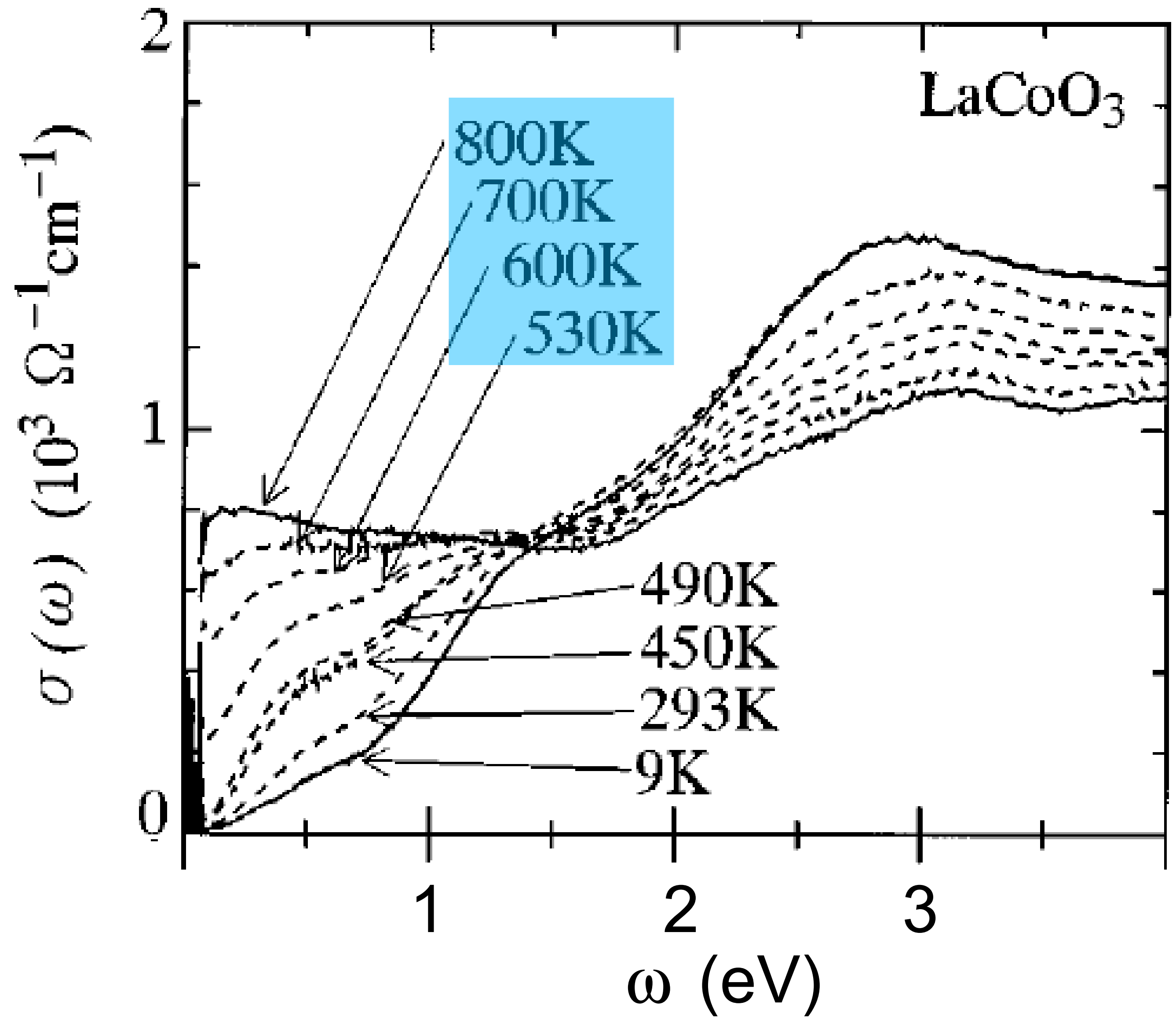
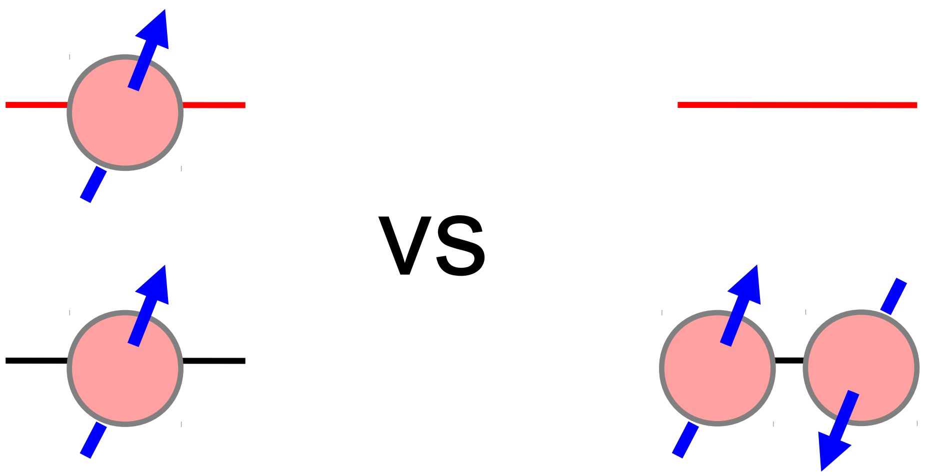
Optical conductivity



Tokura et al. Phys. Rev. B ***58****, R1699 (1998)*

Two-band Hubbard model

$$H = \sum_{i,\sigma} \left((\Delta - \mu) n^a_{i,\sigma} - \mu n^b_{i,\sigma} \right) + \sum_{\langle ij \rangle,\sigma} \left(t_{aa} a^\dagger_{i,\sigma} a_{j,\sigma} + t_{bb} b^\dagger_{i,\sigma} b_{i,\sigma} \right)$$
$$+ U \sum_i \left(n^a_{i,\uparrow} n^a_{i,\downarrow} + n^b_{i,\uparrow} n^b_{i,\downarrow} \right) + (U - 2J) \sum_{i,\sigma} n^a_{i,\sigma} n^b_{i,-\sigma}$$
$$+ (U - 3J) \sum_{i,\sigma} n^a_{i,\sigma} n^b_{i,\sigma}$$

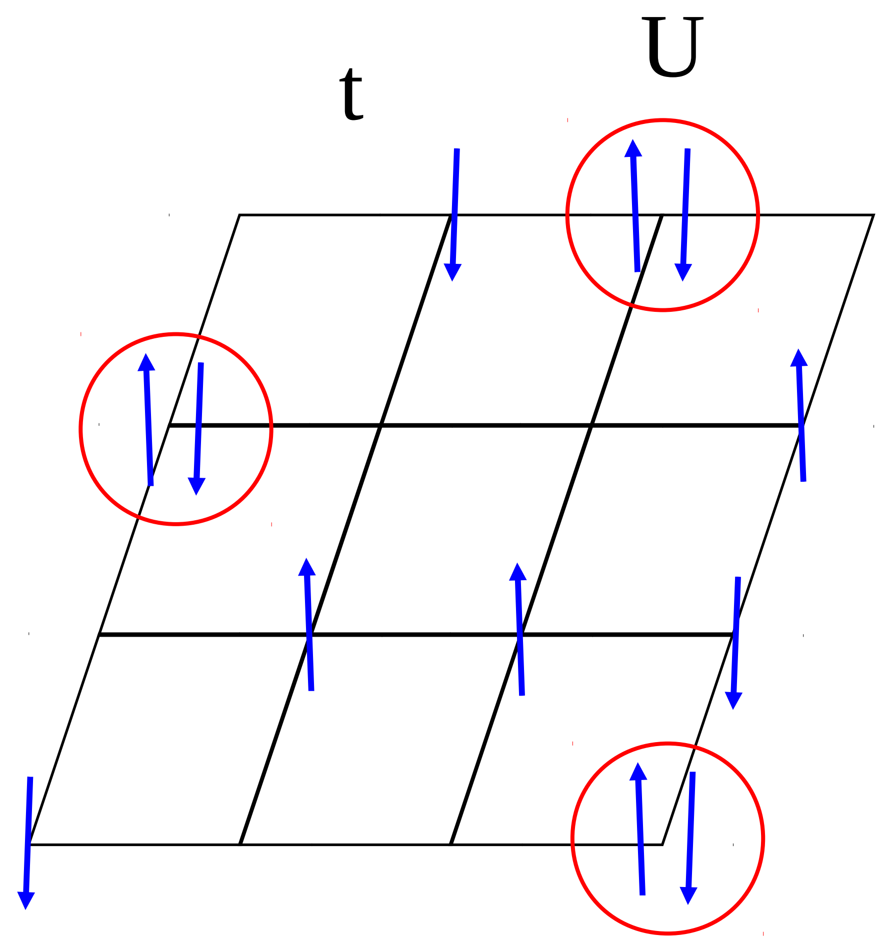


Dynamical mean-field theory

Hubbard model in d=∞

Hamiltonian = Hopping + Local interaction

$$H = \sum_{i,j,\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



Large dimension limit - classical Heisenberg model

$$Z = \sum_{\{s_i\}} \exp(-S)$$

Cavity construction: $S = S_0 + \Delta S + S_{(0)}$ $\qquad \Delta S = \sum_i J_{0i} s_0 s_i$

Expansion in 'hybridization':

$$Z = Z_{(0)} \sum_{s_0} \exp(-S_0) \left(1 + \sum_i J_{i0} \langle s_i \rangle_{(0)} s_0 + \frac{1}{2} \sum_{i,j} J_{i0} J_{j0} \langle s_i s_j \rangle_{(0)} s_0^2 + \ldots \right)$$

Cumulant expansion:

$$Z = Z_{(0)} \sum_{s_0} \exp(-S_{\text{eff}})$$

$$S_{\text{eff}} = h s_0 + \sum_i J_{i0} \langle s_i \rangle_{(0)} s_0 + \frac{1}{2} \sum_{i,j} J_{i0} J_{j0} \Big(\langle s_i s_j \rangle_{(0)} - \langle s_i \rangle_{(0)} \langle s_j \rangle_{(0)} \Big) s_0^2 + \ldots$$

Scaling:

$$J = \frac{J^*}{d}$$

Dynamical mean-field theory

- Weak coupling expansion

In d=∞ limit only local contributions to the self-energy survive!

$$\Sigma(\mathbf{k},\omega) \equiv \Sigma(\omega)$$
$$\Sigma = \Phi[G_{\mathrm{loc}}]$$

- Cumulant expansion (cavity construction)

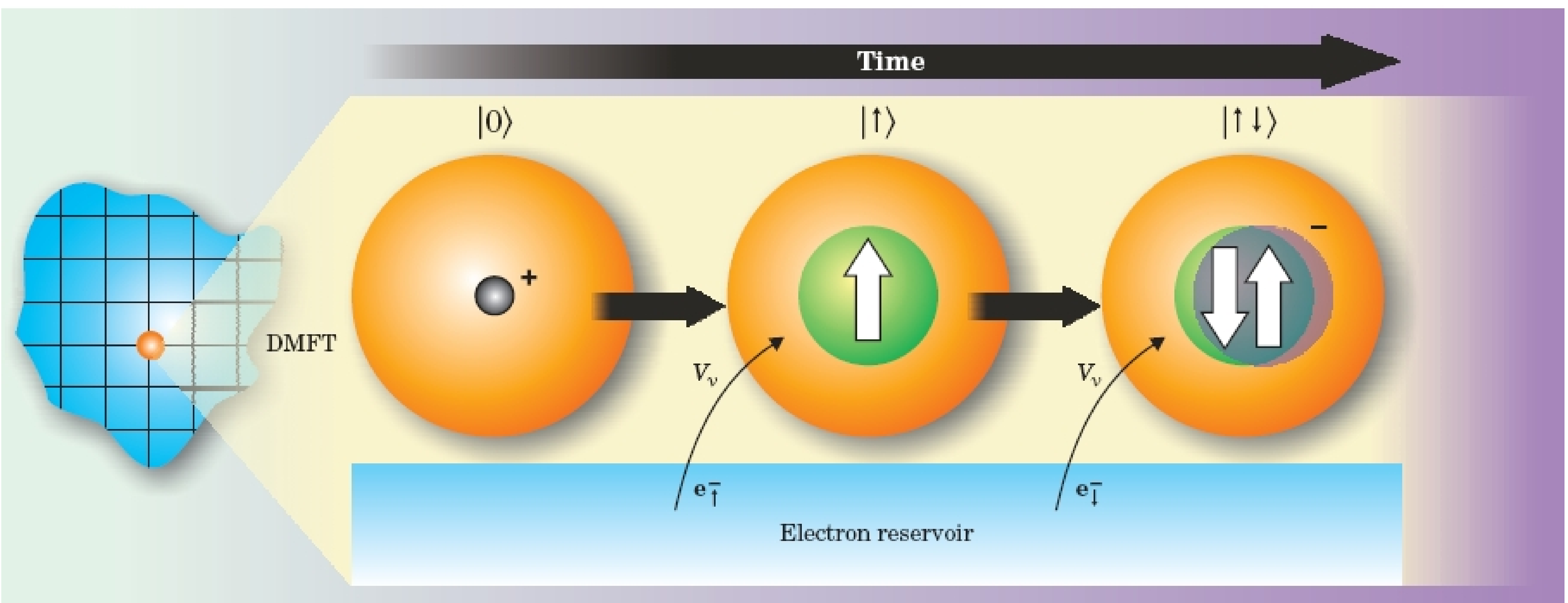
In d=∞ limit tha action reduces to interacting site subject to "time dependent" external field equivalent to fermionic bath

Georges and Kotliar,
*PRB **45**, 6479 (1992)*

DMFT	Weiss molecular field
$$H = \sum_{i,j,\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$	$$H = \sum_{i,j} J_{ij} S_i S_j + h \sum_i S_i$$
$$G_{ii}(\tau) = -\langle T c_i(\tau) c_i^\dagger(0) \rangle$$	$$s_i = \langle S_i \rangle$$
$$H_{\mathrm{loc}} = (\varepsilon - \mu) \sum_\sigma c^\dagger_\sigma c_\sigma + U n_\uparrow n_\downarrow + \sum_{r\sigma} \eta_r b^\dagger_{r\sigma} b_{r\sigma} + \sum_{r\sigma} V_r c^\dagger_\sigma b_{r\sigma} + c.c.$$	$$H_{\mathrm{loc}} = \tilde{h} S$$
$$G_{ii}(\omega) = \sum_k \frac{1}{\omega + \mu - \varepsilon_k - \Sigma(\omega)}$$	$$\tilde{h} = \sum_i J_{0i} s_i + h$$

Dynamical Mean-Field Theory (DMFT)

- Single out a site from the lattice
- Replace the rest of the lattice by an effective medium
- Solve the impurity many-body problem
- Reconstruct lattice quantities



A. Georges et al. RMP ***68****, 13 (1996*) ***Physics Today*** (March 2004) Kotliar, Vollhardt

Impurity problem

Hamiltonian formulation

$$H_{\rm loc} = (\varepsilon - \mu)\sum_\sigma c_\sigma^\dagger c_\sigma + U n_\uparrow n_\downarrow + \sum_{r\sigma} \eta_r b_{r\sigma}^\dagger b_{r\sigma} + \sum_{r\sigma} V_r c_\sigma^\dagger b_{r\sigma} + c.c.$$

$$\Delta(\omega) = \sum_r \frac{|V_r|^2}{\omega - \eta_r}$$

Action formulation

$$S_{\rm imp} = -\int_0^\beta d\tau \sum_\sigma \bar{c}_\sigma(\tau)(\frac{\partial}{\partial\tau} + \mu - \varepsilon)c_\sigma(\tau) + U n_\uparrow(\tau) n_\downarrow(\tau) -$$

$$\int_0^\beta d\tau \int_0^\beta d\tau' \sum_\sigma \bar{c}_\sigma(\tau)\Delta(\tau - \tau')c_\sigma(\tau')$$

$$\boxed{G_{ii}(\tau) = -\langle T c_i(\tau) c_i^\dagger(0)\rangle}$$

Two-band Hubbard model

$$H = \sum_{i,\sigma} \left((\Delta - \mu) n^a_{i,\sigma} - \mu n^b_{i,\sigma} \right) + \sum_{\langle ij \rangle, \sigma} \left(t_{aa} a^\dagger_{i,\sigma} a_{j,\sigma} + t_{bb} b^\dagger_{i,\sigma} b_{i,\sigma} \right)$$

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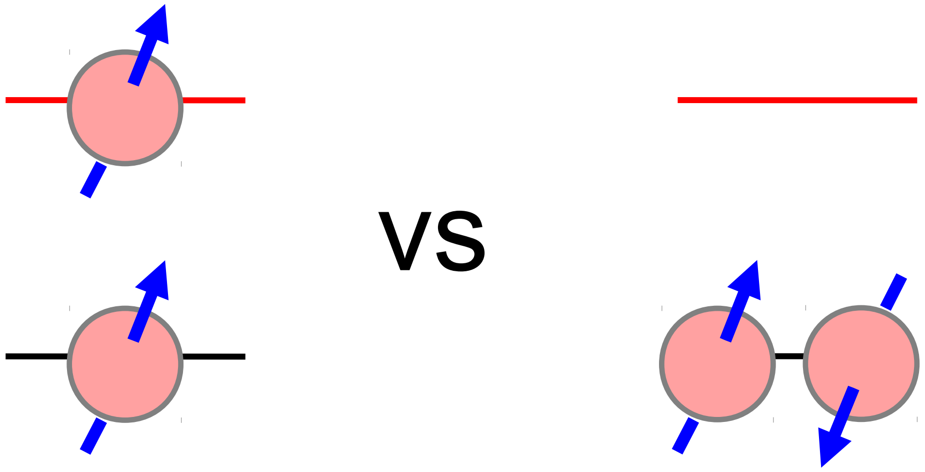
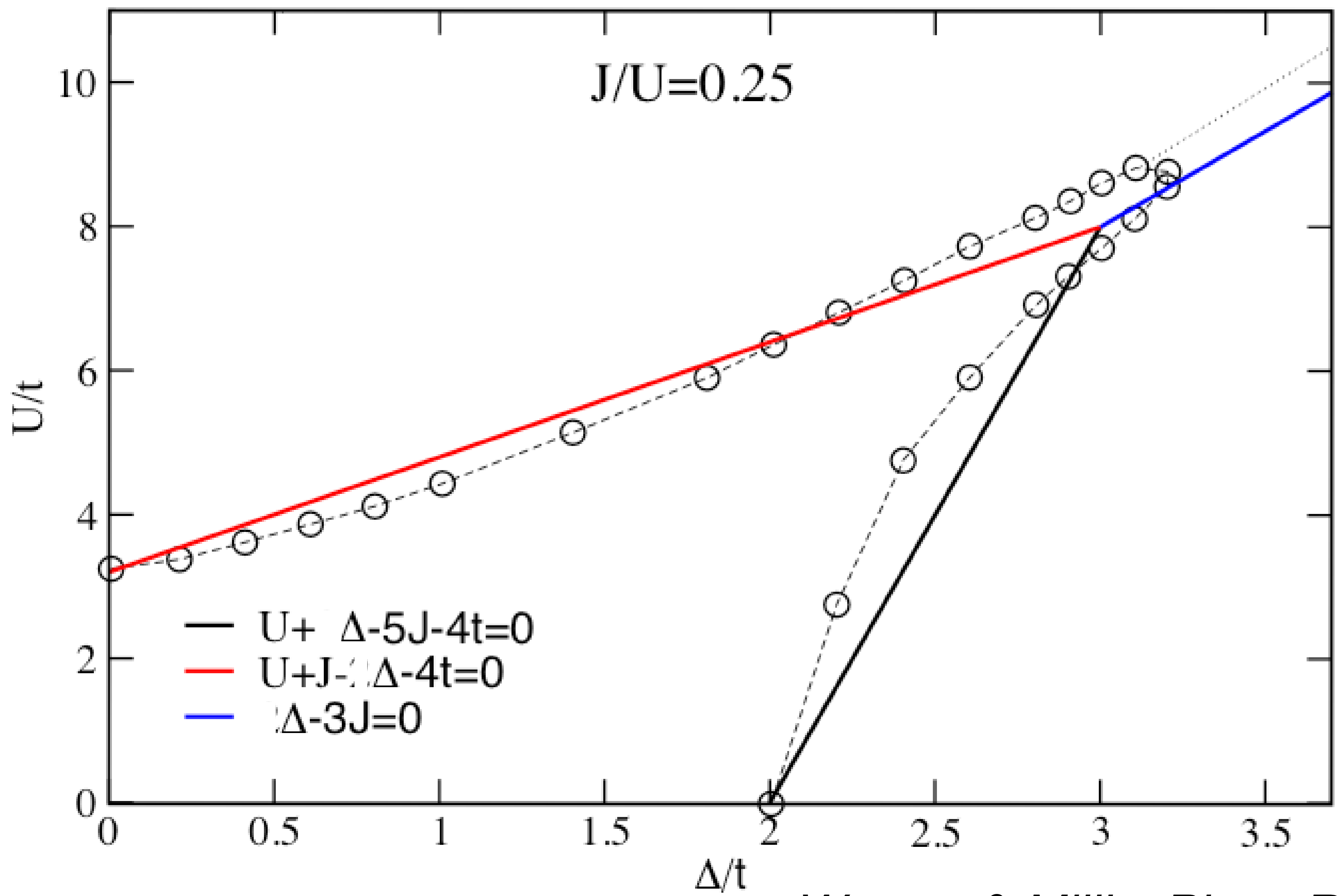


U-Δ phase diagram

Δ - crystal field

J/U - fixed

Uniform phase (arbitrary lattice - DMFT)



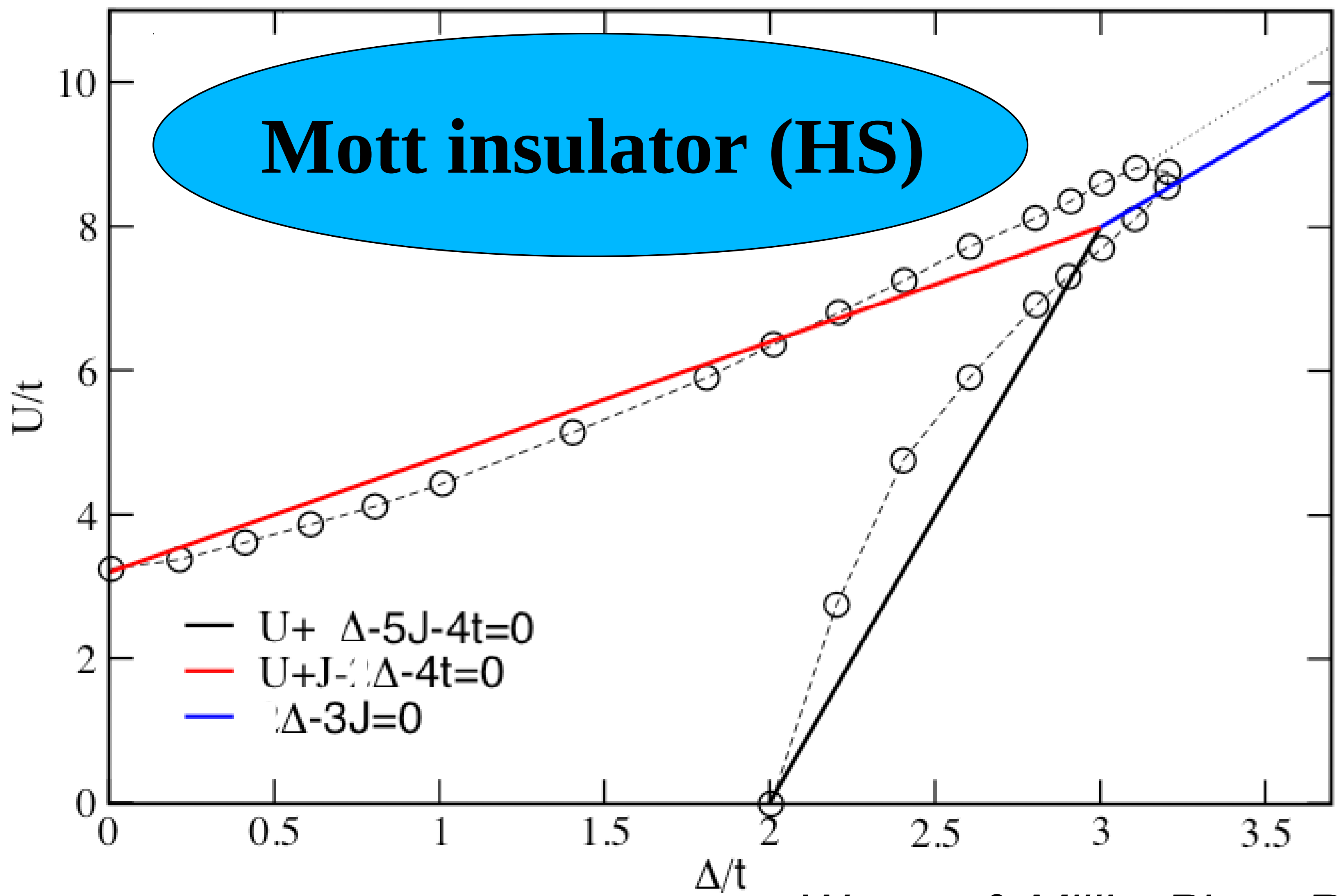
Werner & Millis, Phys. Rev. Lett. ***99,*** *126405 (2007)*
JK et al. Eur. Phys. J. Special Topics ***180****, 5 (2009)*

U-Δ phase diagram

Δ - crystal field

J/U - fixed

2D - bipartite lattice (square lattice)



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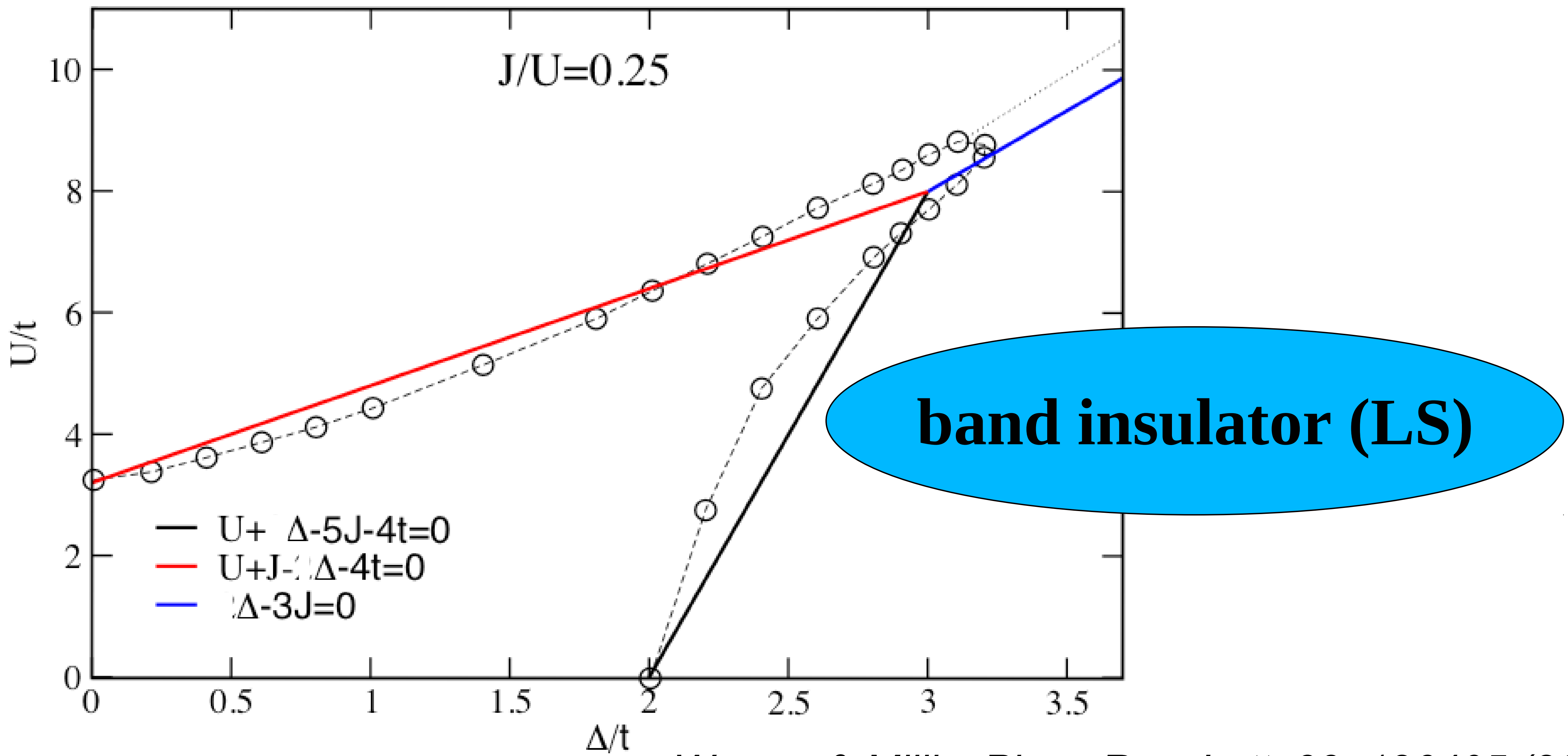

Werner & Millis, Phys. Rev. Lett. ***99,*** *126405 (2007)*
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U-Δ phase diagram

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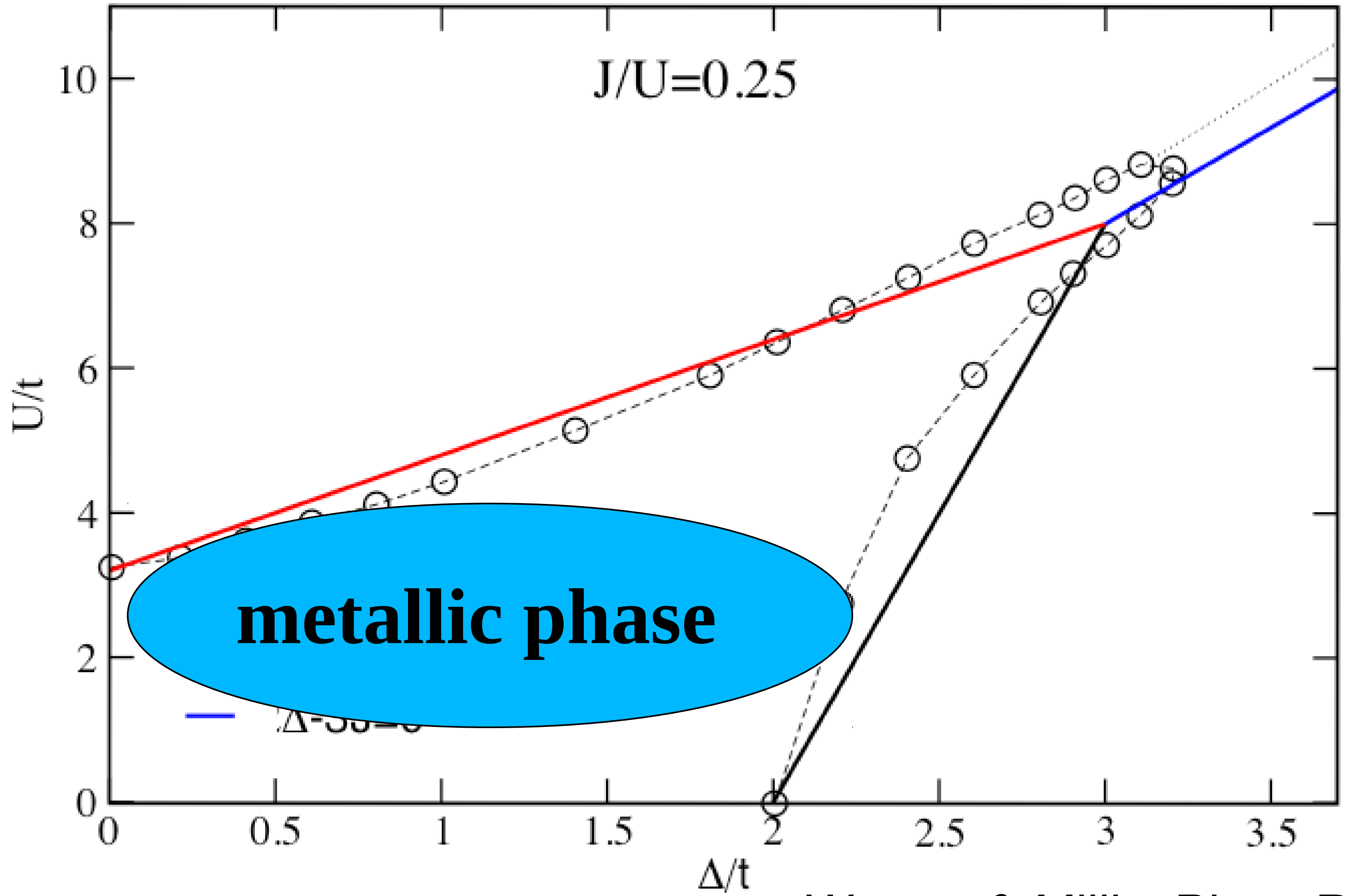
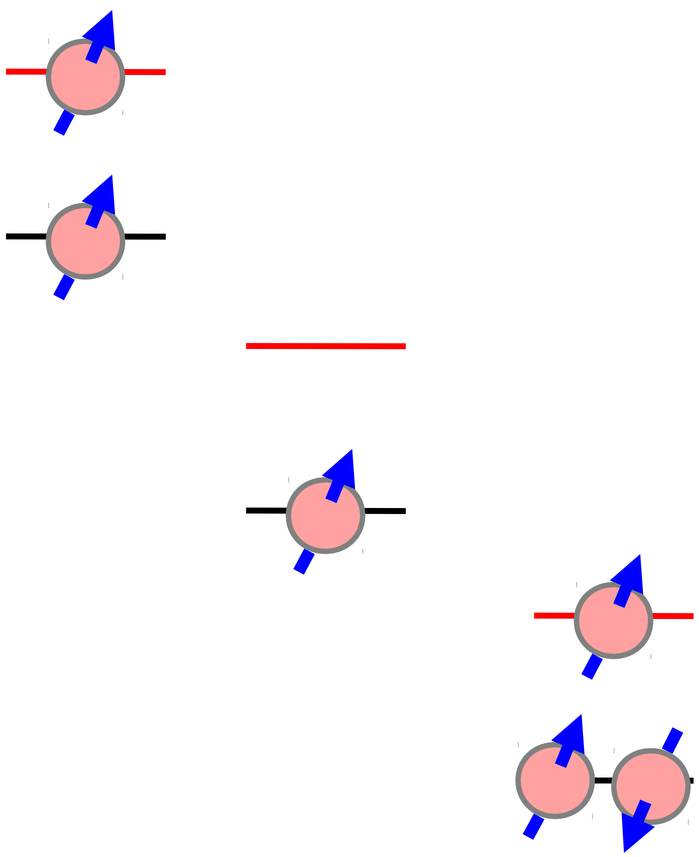


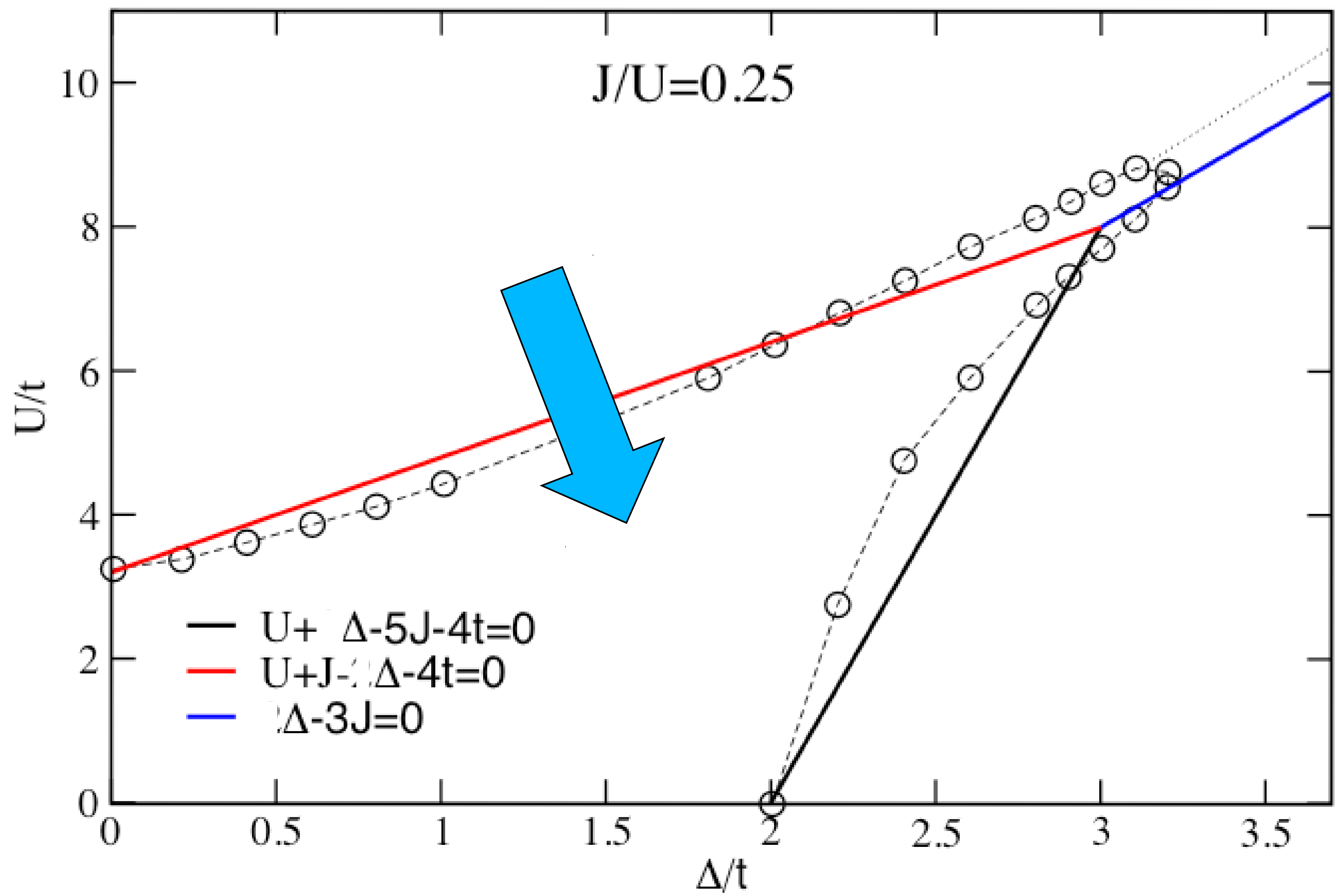
Werner & Millis, Phys. Rev. Lett. ***99,*** *126405 (2007)*
JK et al. Eur. Phys. J. Special Topics ***180****, 5 (2009)*

Gap closing

'Mott gap = 0'

E_g=2E(N)-E(N-1)-E(N+1)

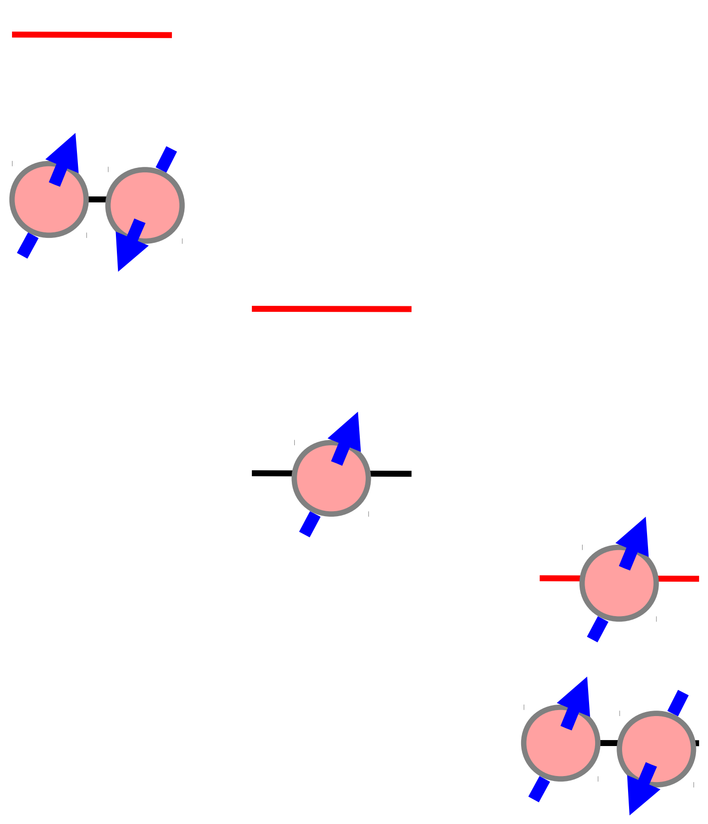


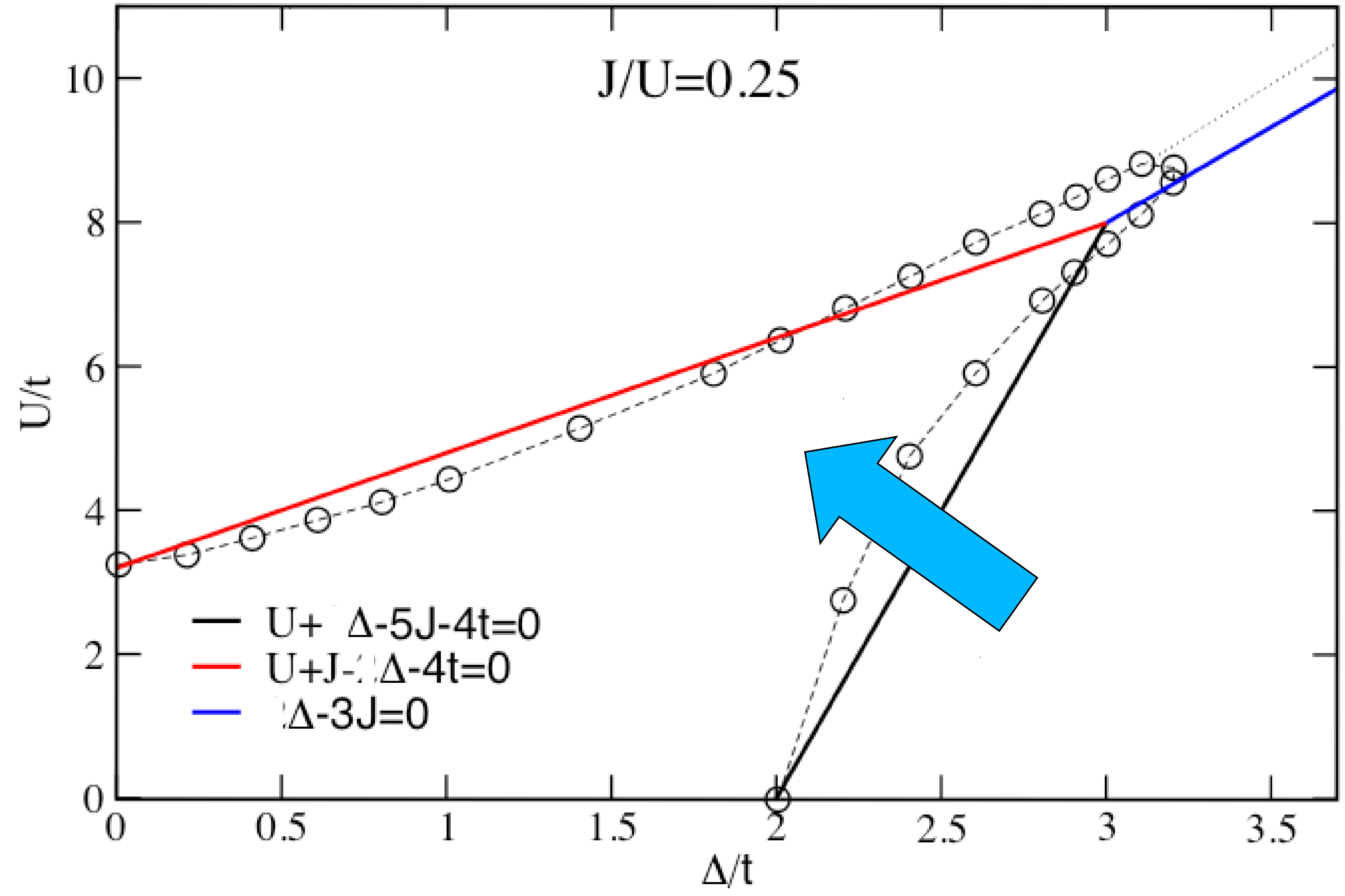


Band gap

'Band gap = 0'

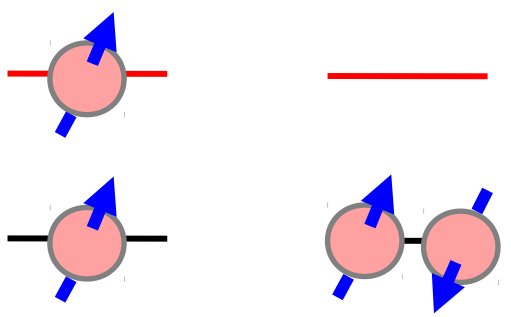
E_g=2E(N)-E(N-1)-E(N+1)

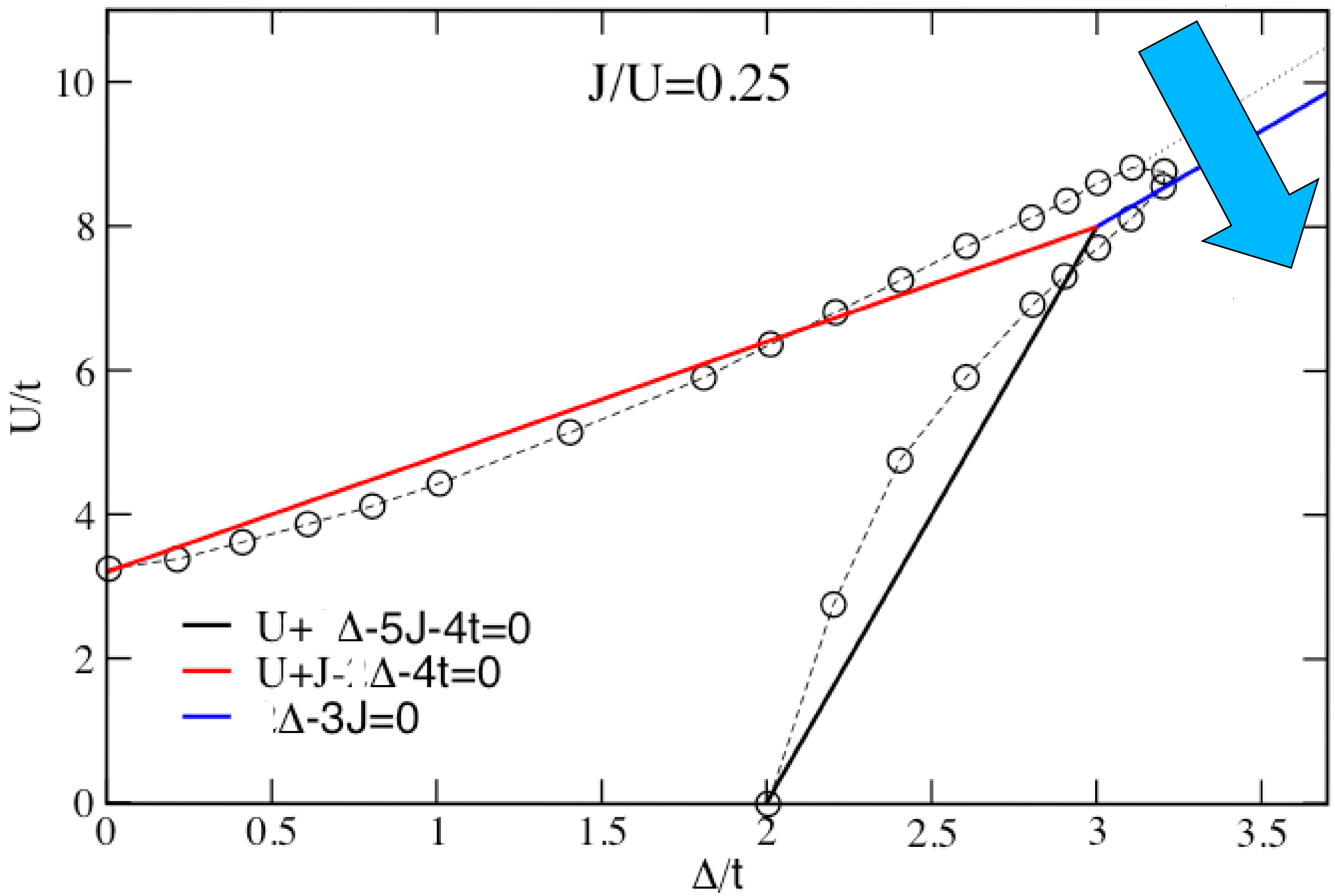




Local state transition

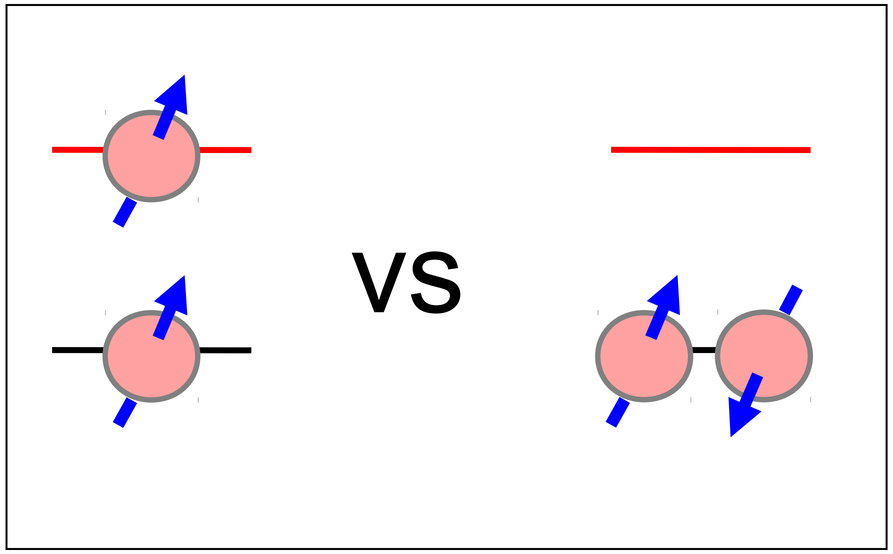
'E(HS)-E(LS) = 0'



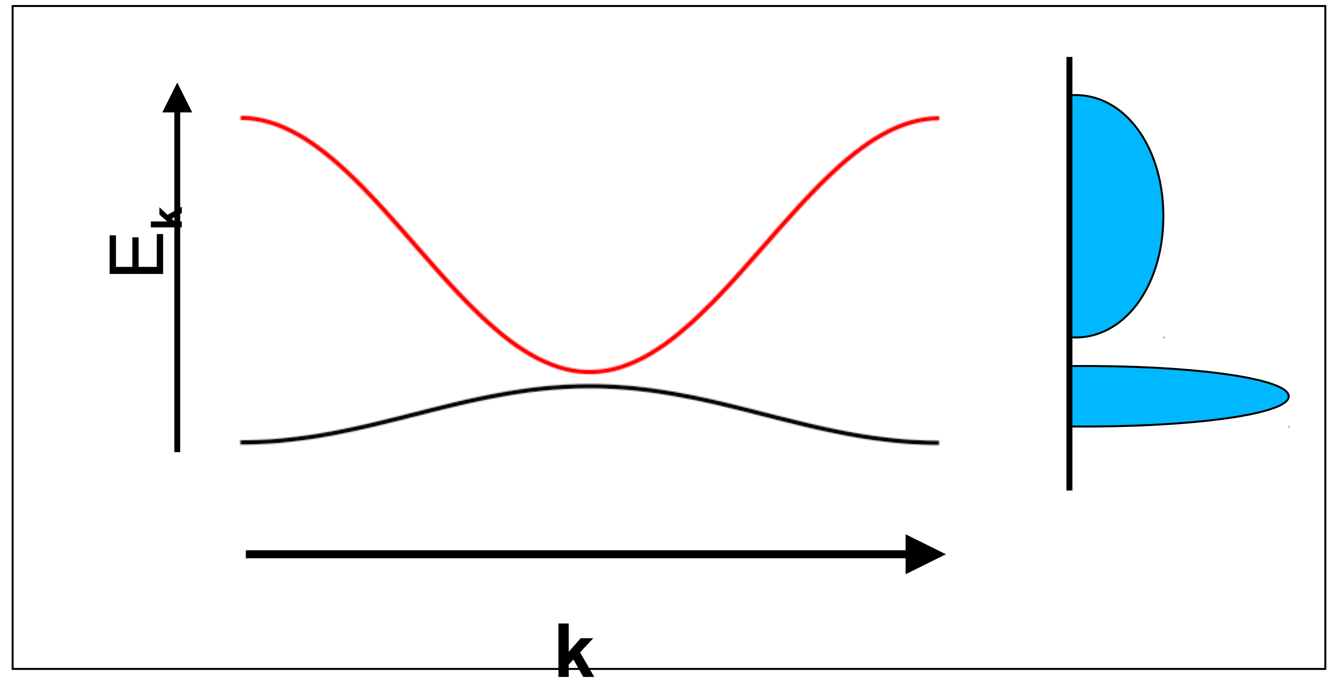


Two-band Hubbard model at half filling

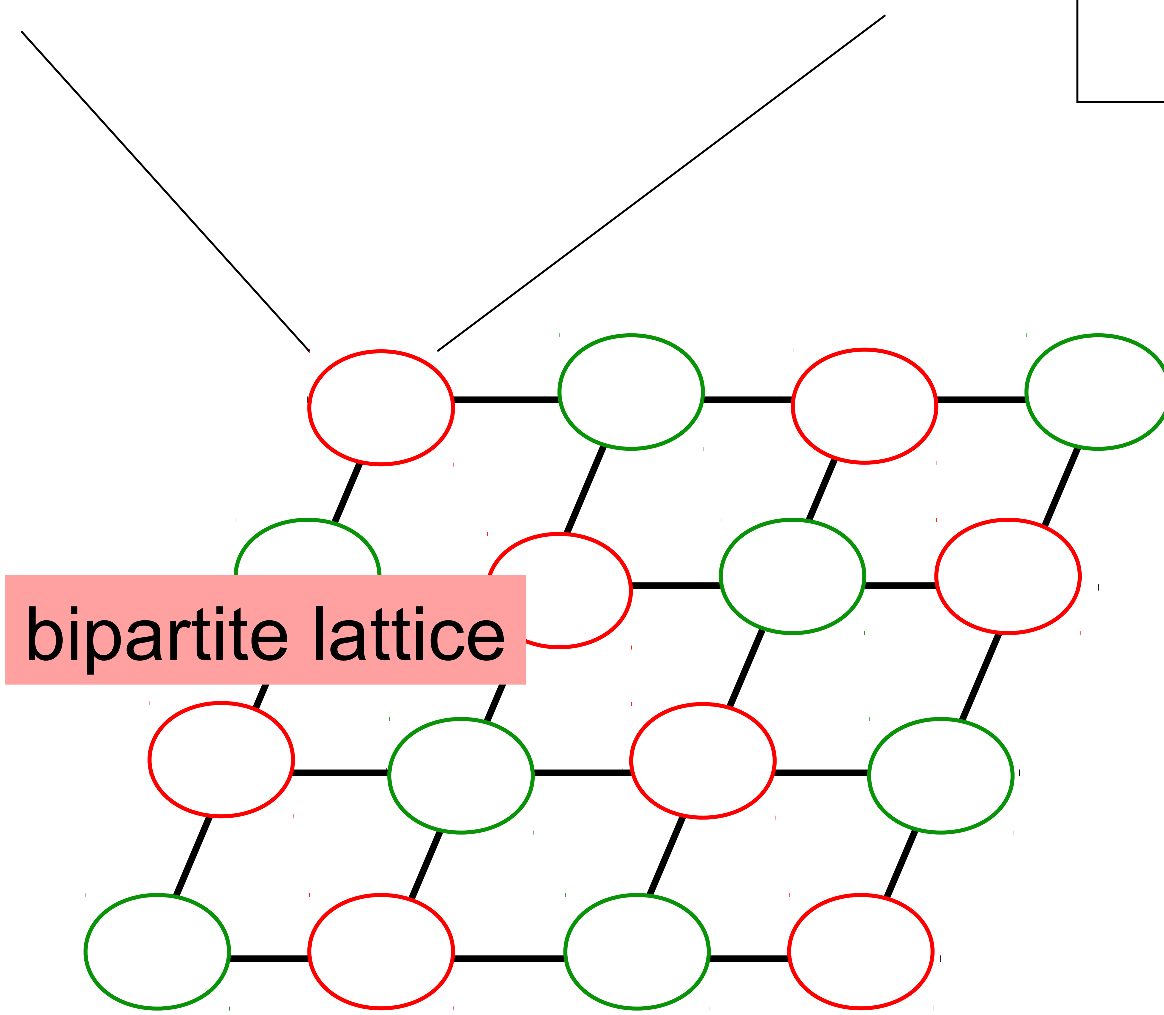
on-site HS-LS competition



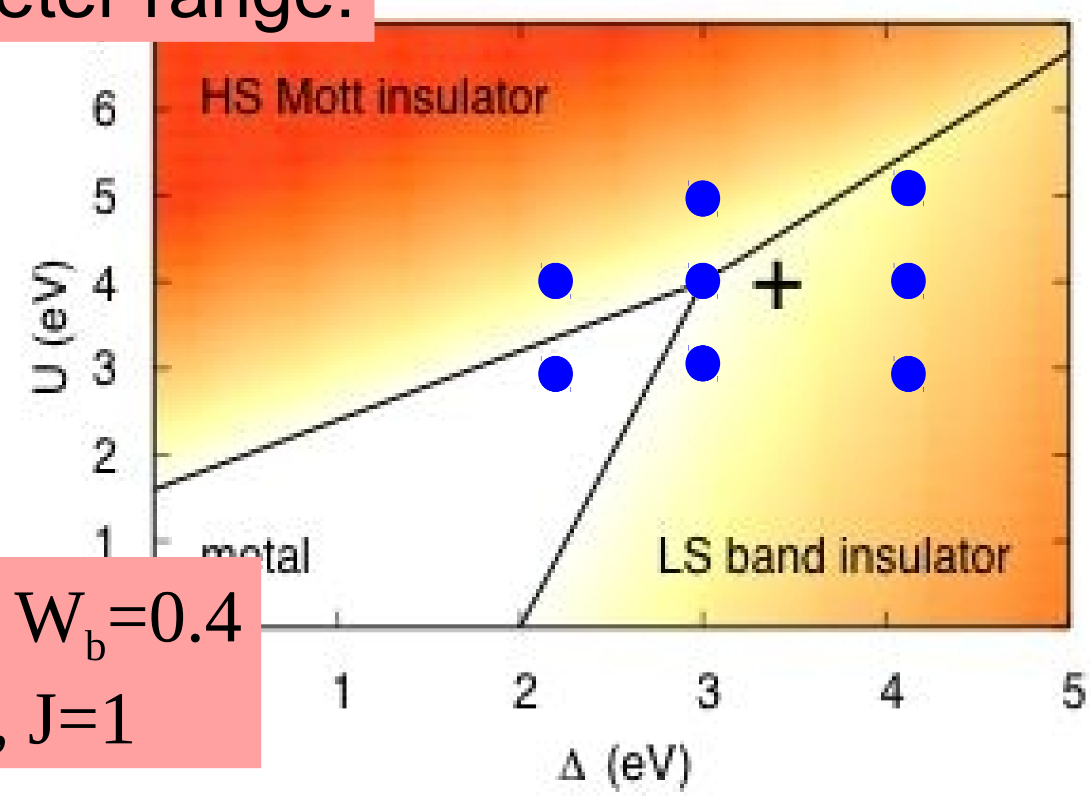
non-interacting band structure



bipartite lattice

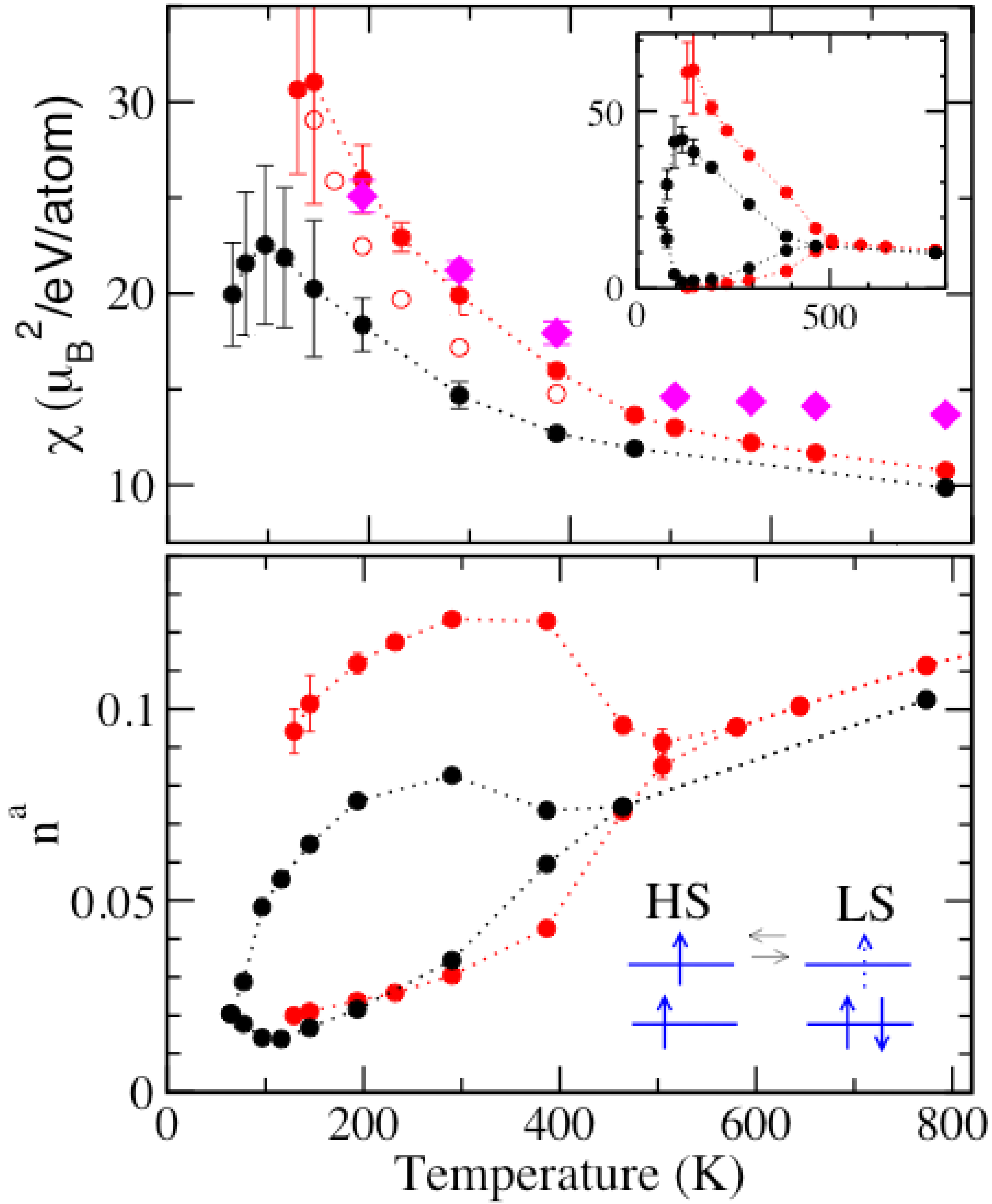


parameter range:



W_a=3.6, W_b=0.4
U=4, J=1

Spin susceptibility and disproportionation

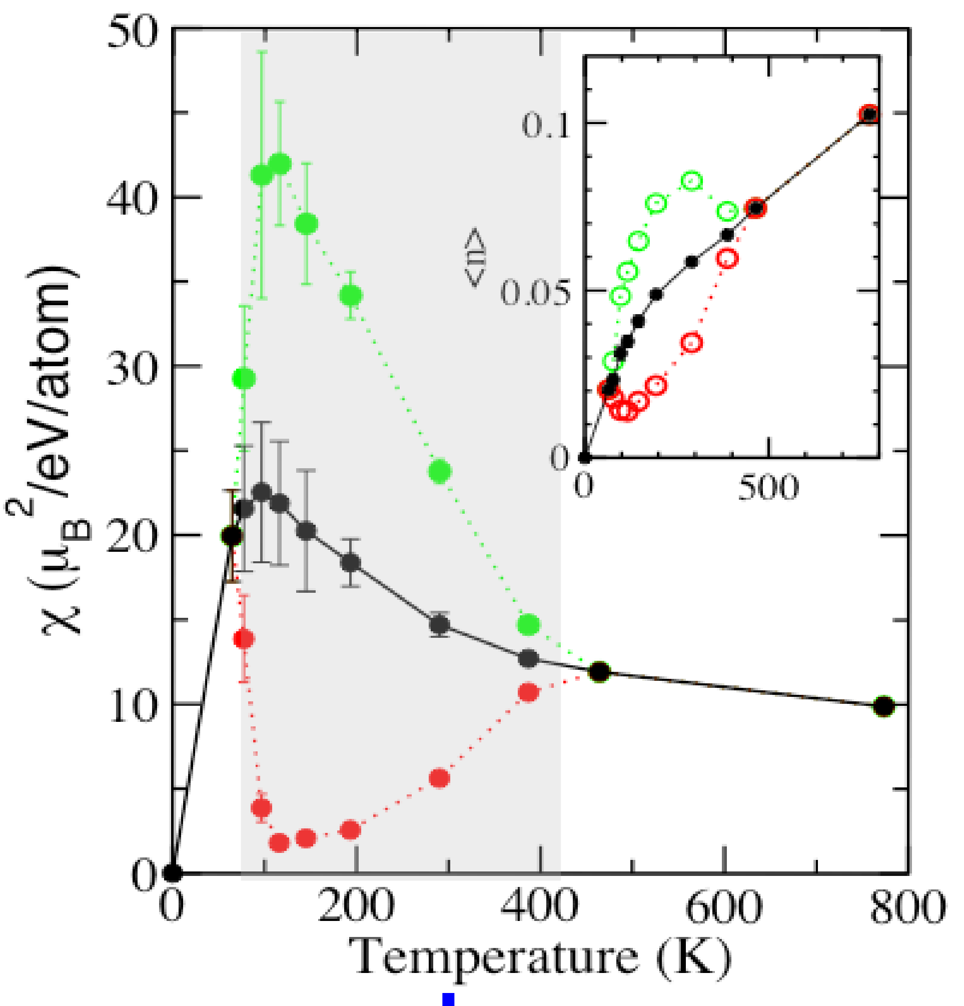


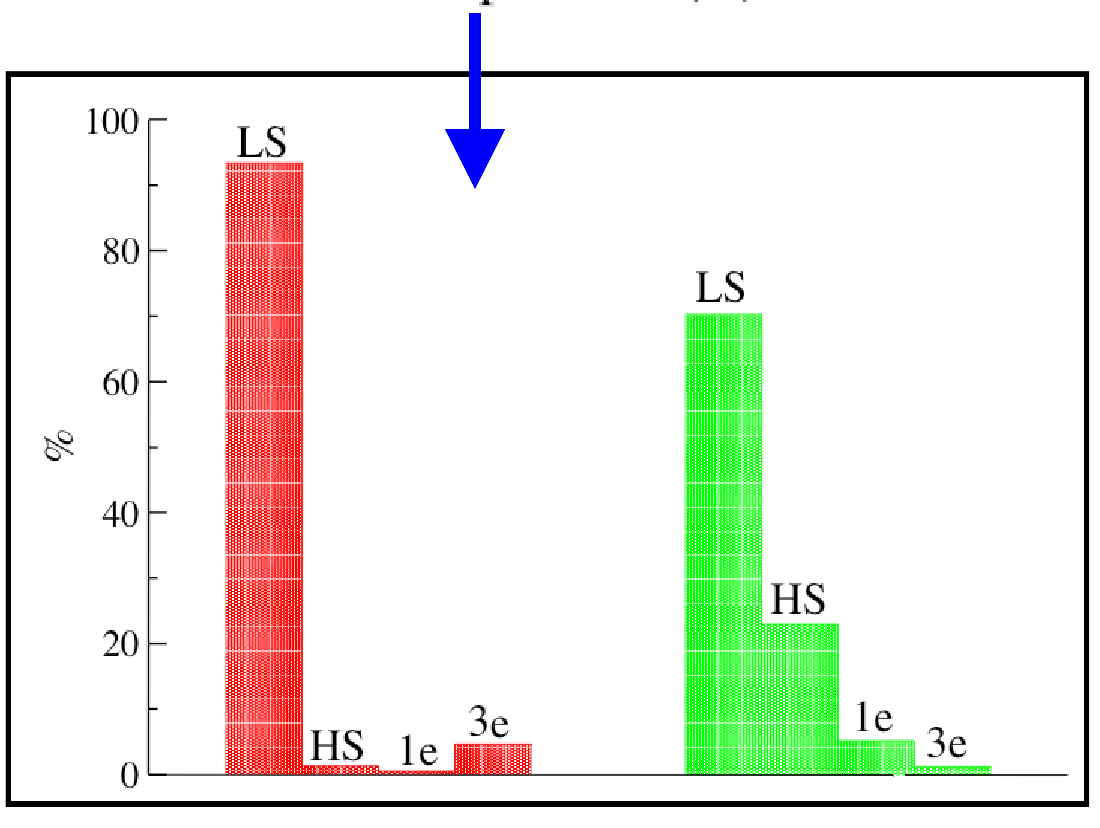
Δ-3J=0.42
local susceptibility
Δ-3J=0.40
local susceptibility
local susceptibility (homog. ph.)
uniform susceptibility

Site occupancy n^a (upper band)
Δ-3J=0.42
Δ-3J=0.40

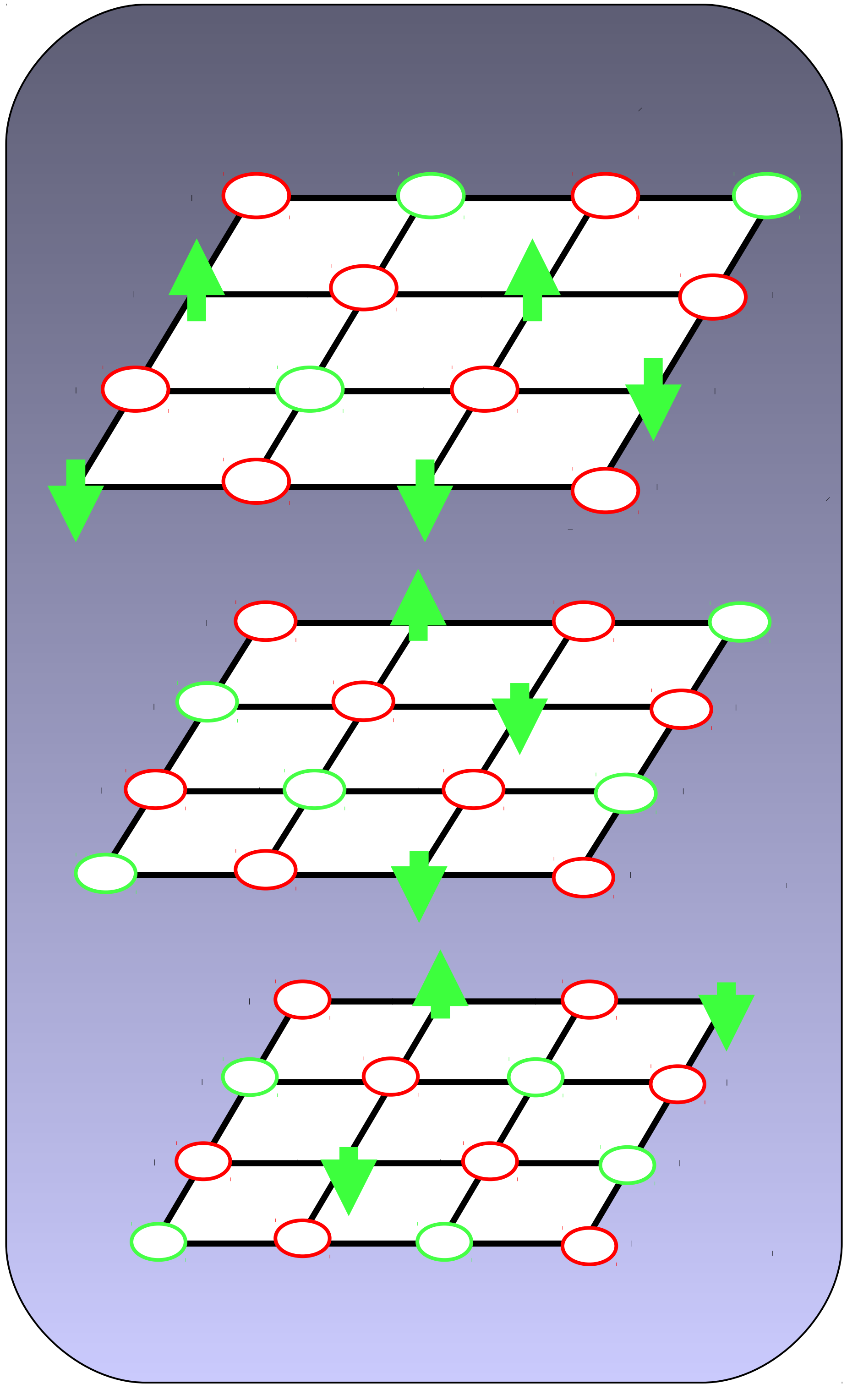
JK & Krapek, Phys. Rev. Lett. ***106****, 256401 (2011)*

Reentrant HS-LS disproportionation

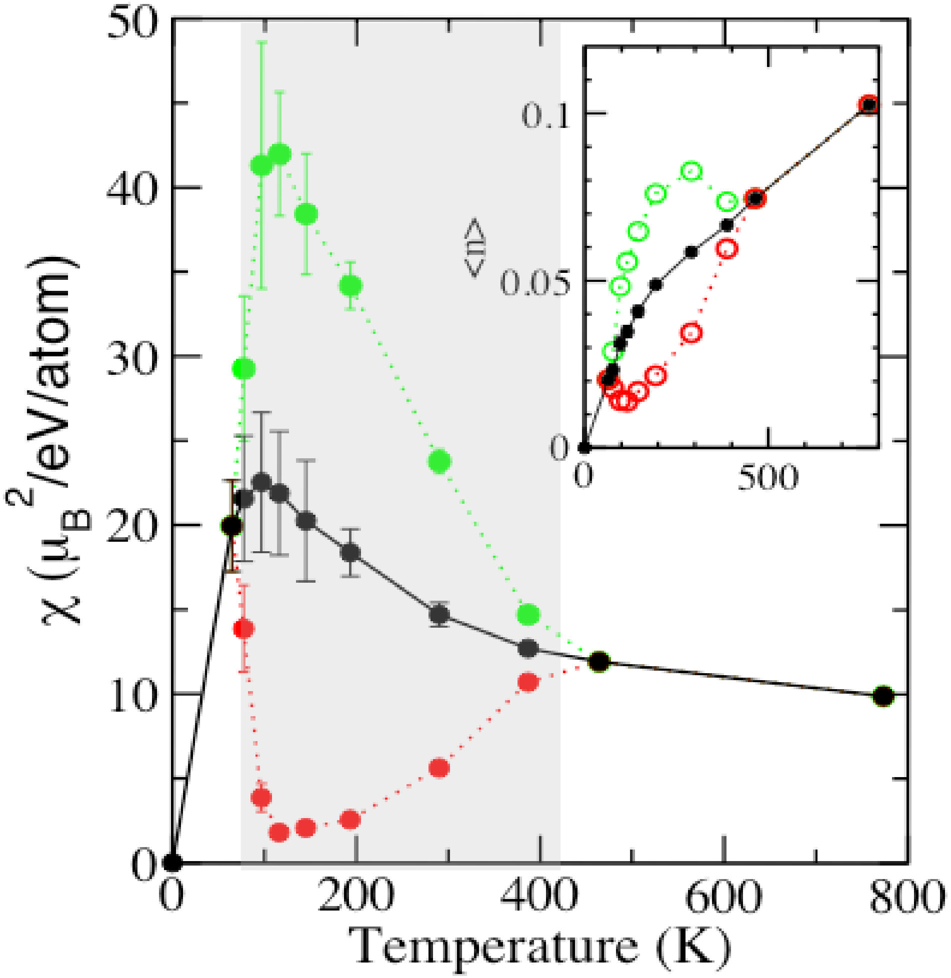


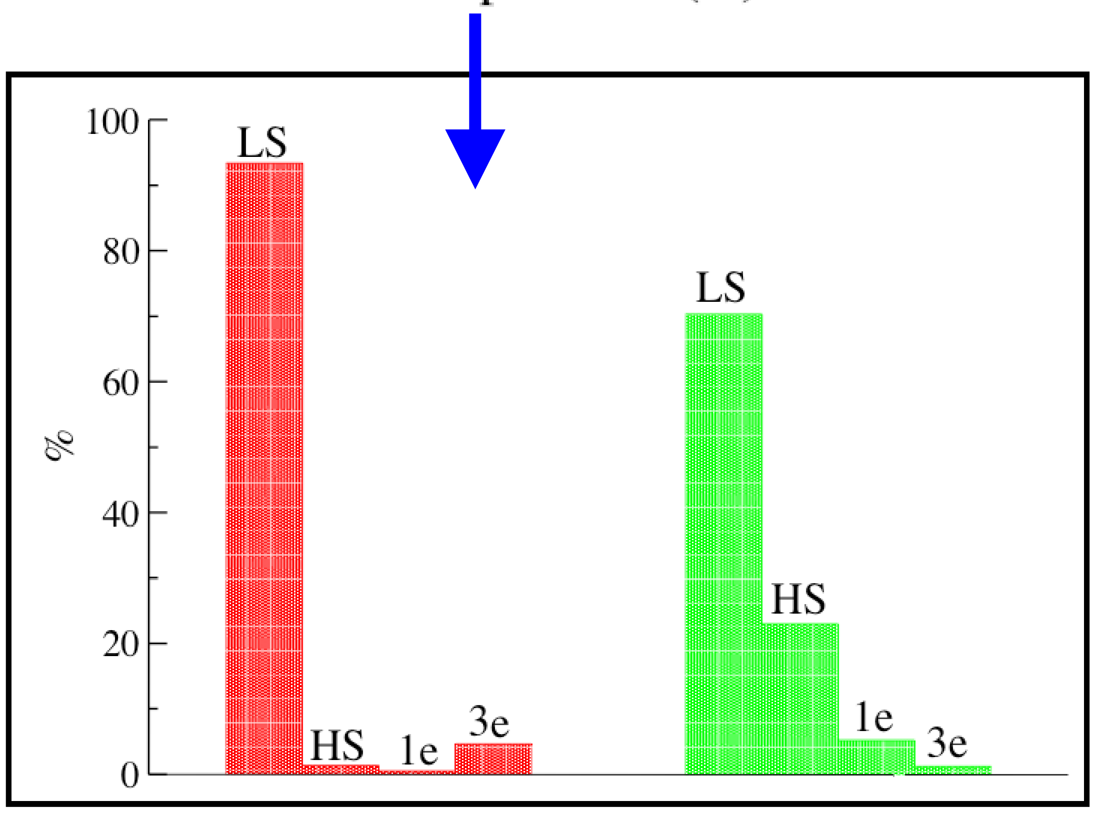


Ordered phase



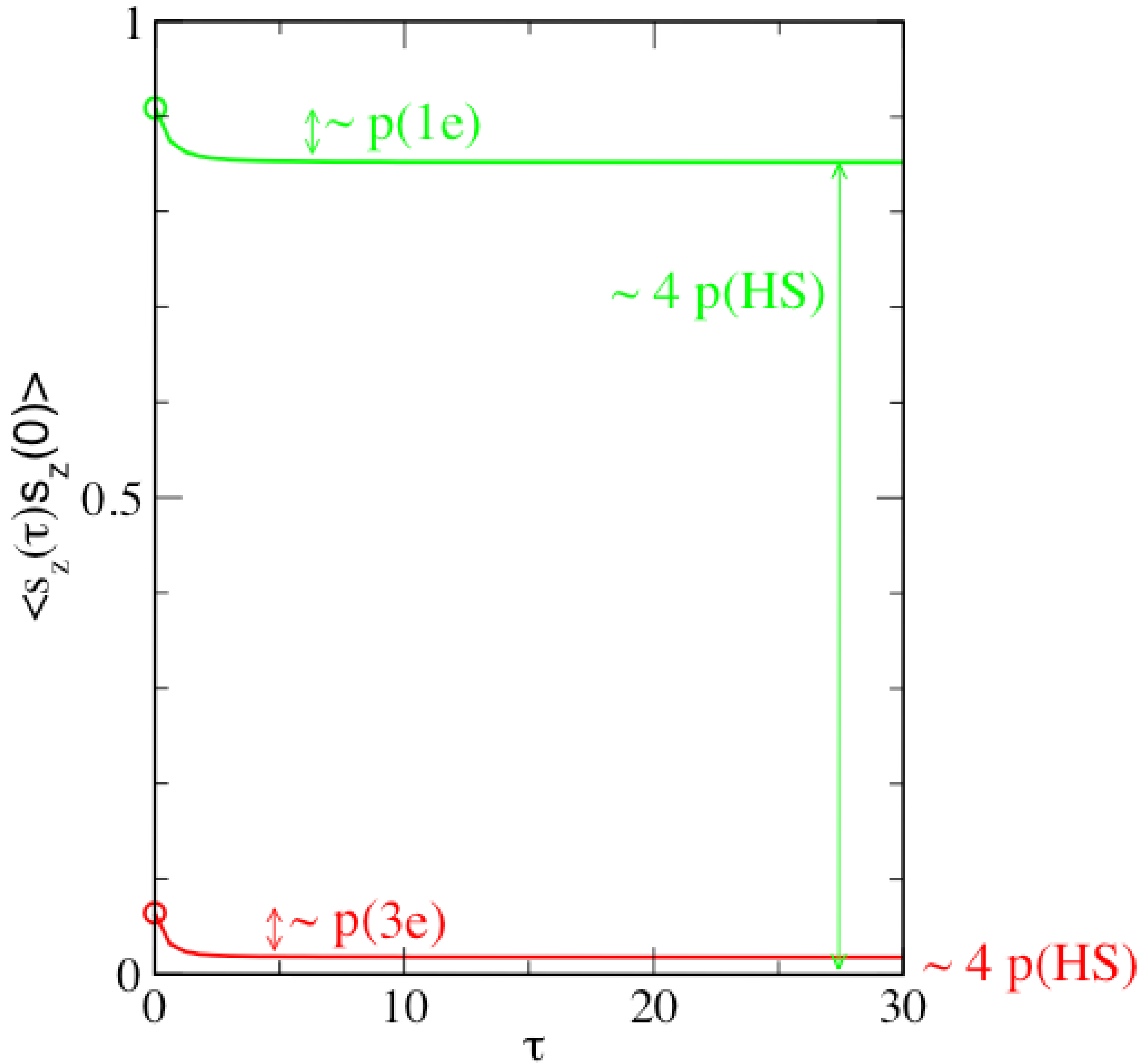
Reentrant HS-LS disproportionation





short excursions
vs
statistical mixture

Spin-spin correlations

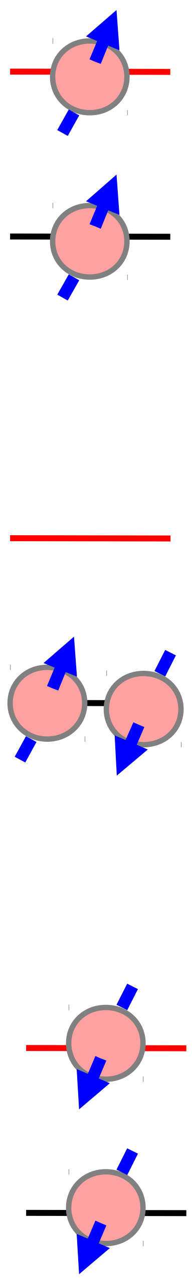


Blume-Emery-Griffiths model

$$\tilde{H} = D\sum_i s_i^2 + K\sum_{\langle ij\rangle} s_i^2 s_j^2 + I\sum_{\langle ij\rangle} s_i s_j$$

Blume et al., Phys. Rev. A ***4****, 1071 (1971)*

$$s_i = \begin{cases} 1 \\ 0 \\ -1 \end{cases}$$

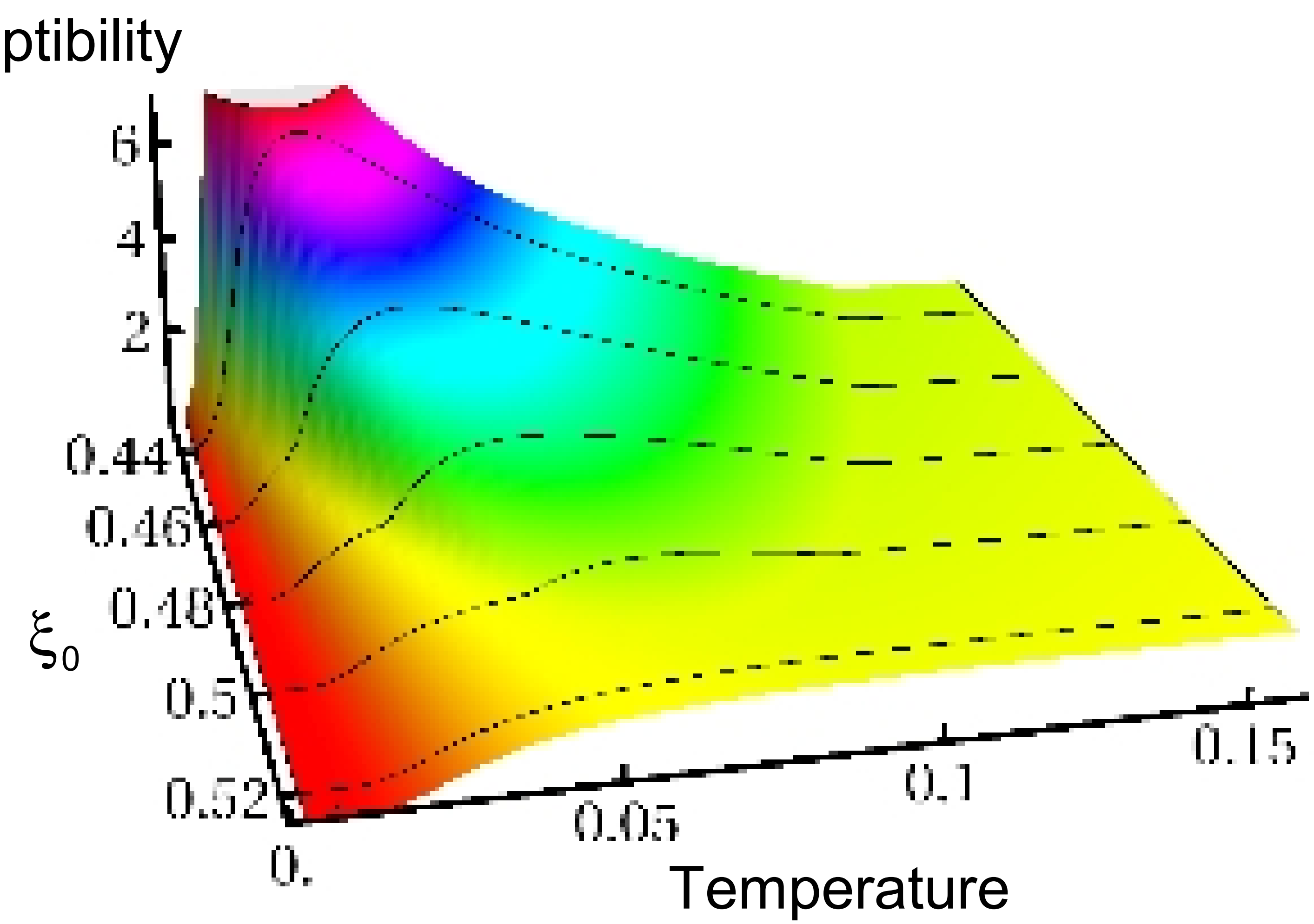


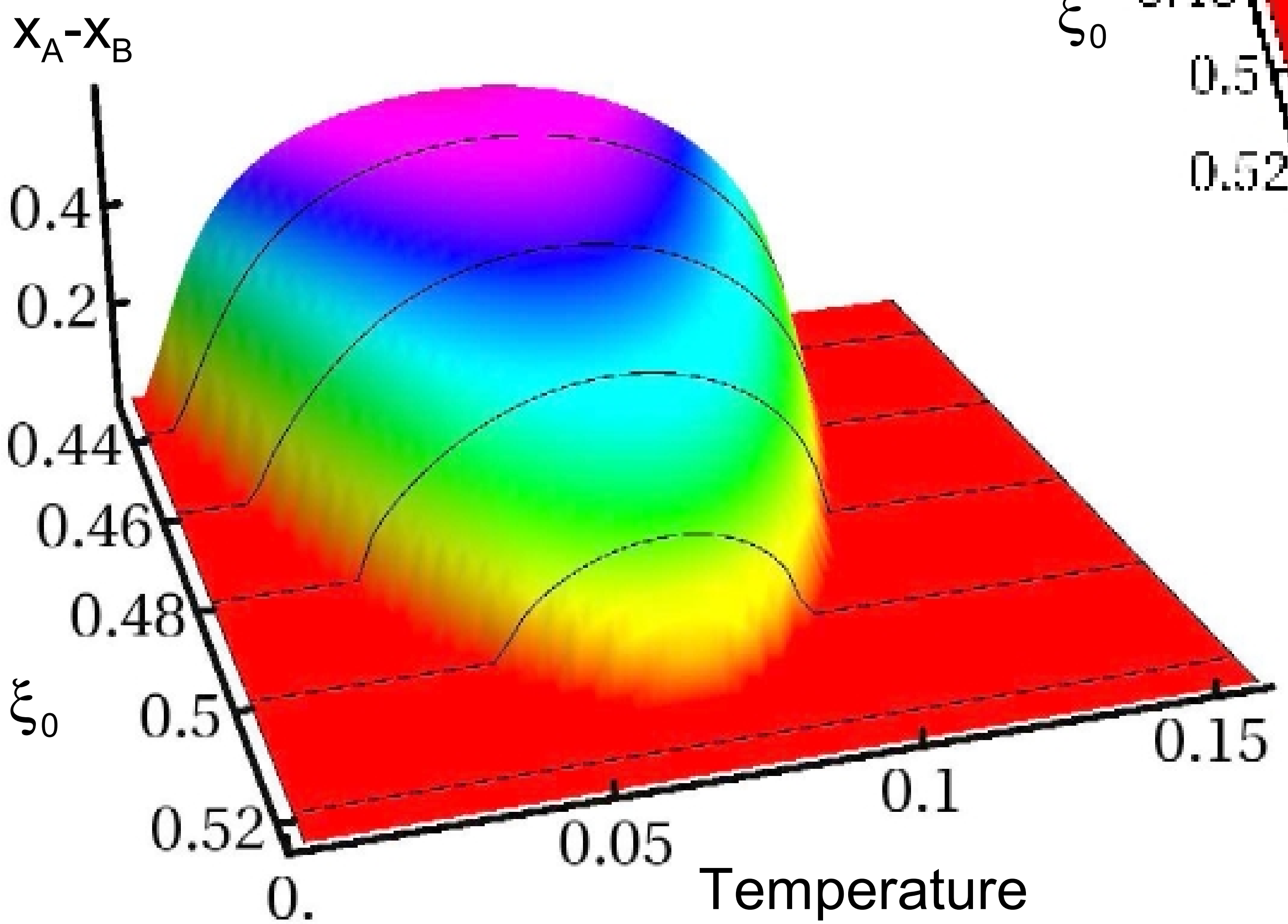
$$D = \Delta - 3J - \frac{Zt^2}{U-2J}$$

$$K = t^2\left(\frac{1}{U-2J} - \frac{1}{U+J}\right)$$

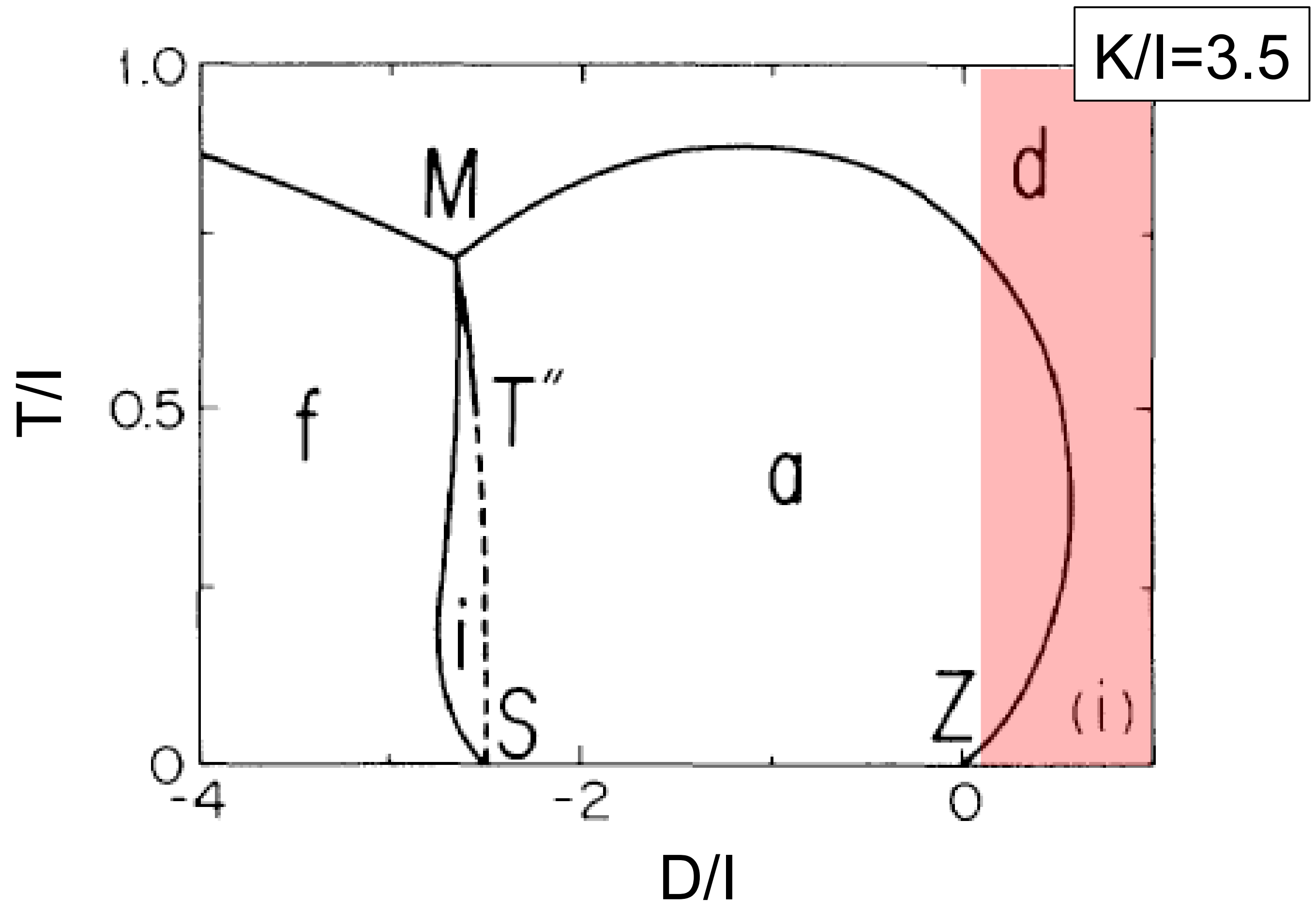
$$I = \frac{t^2}{U+J}$$

Mean-field for the BEG model

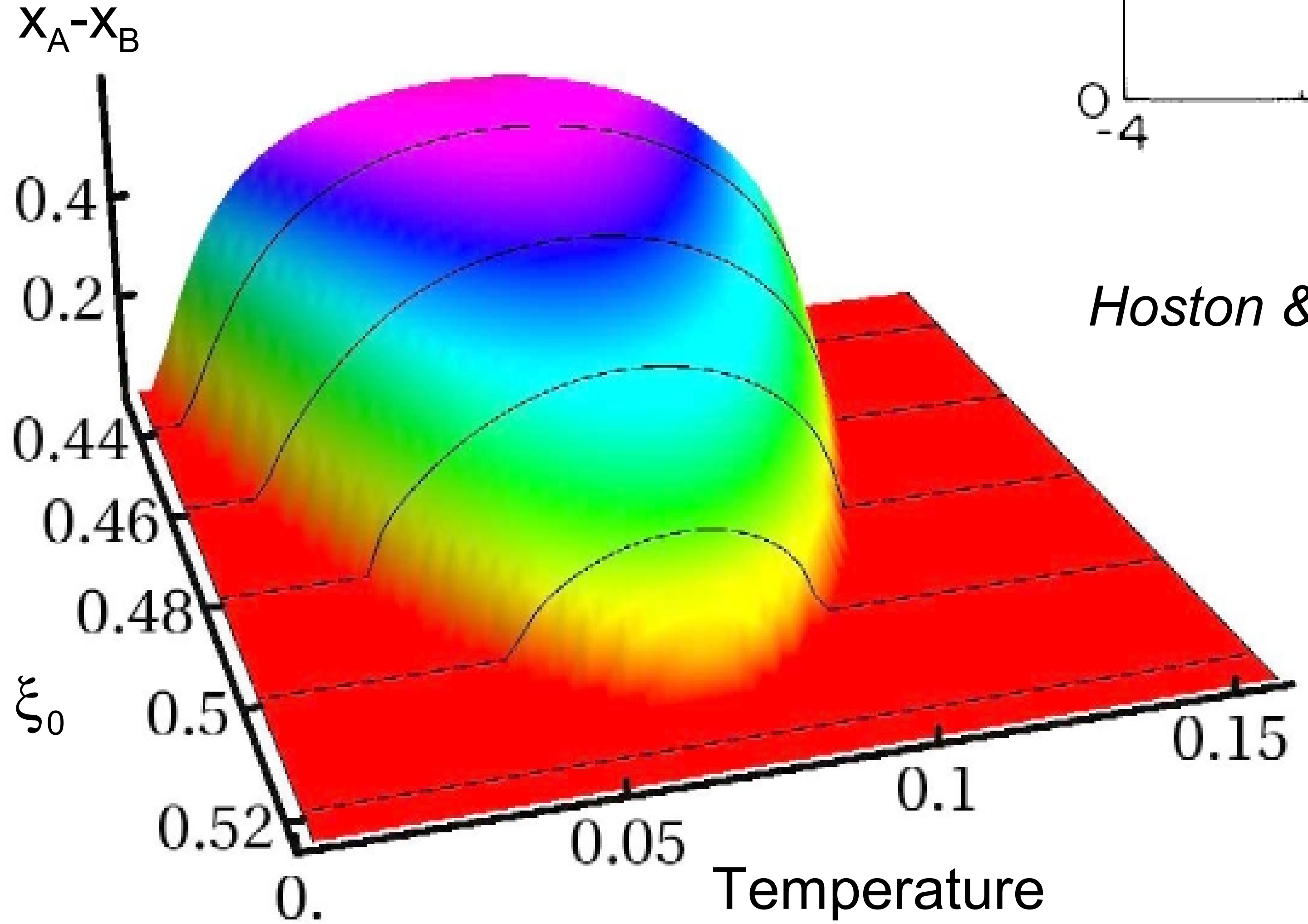




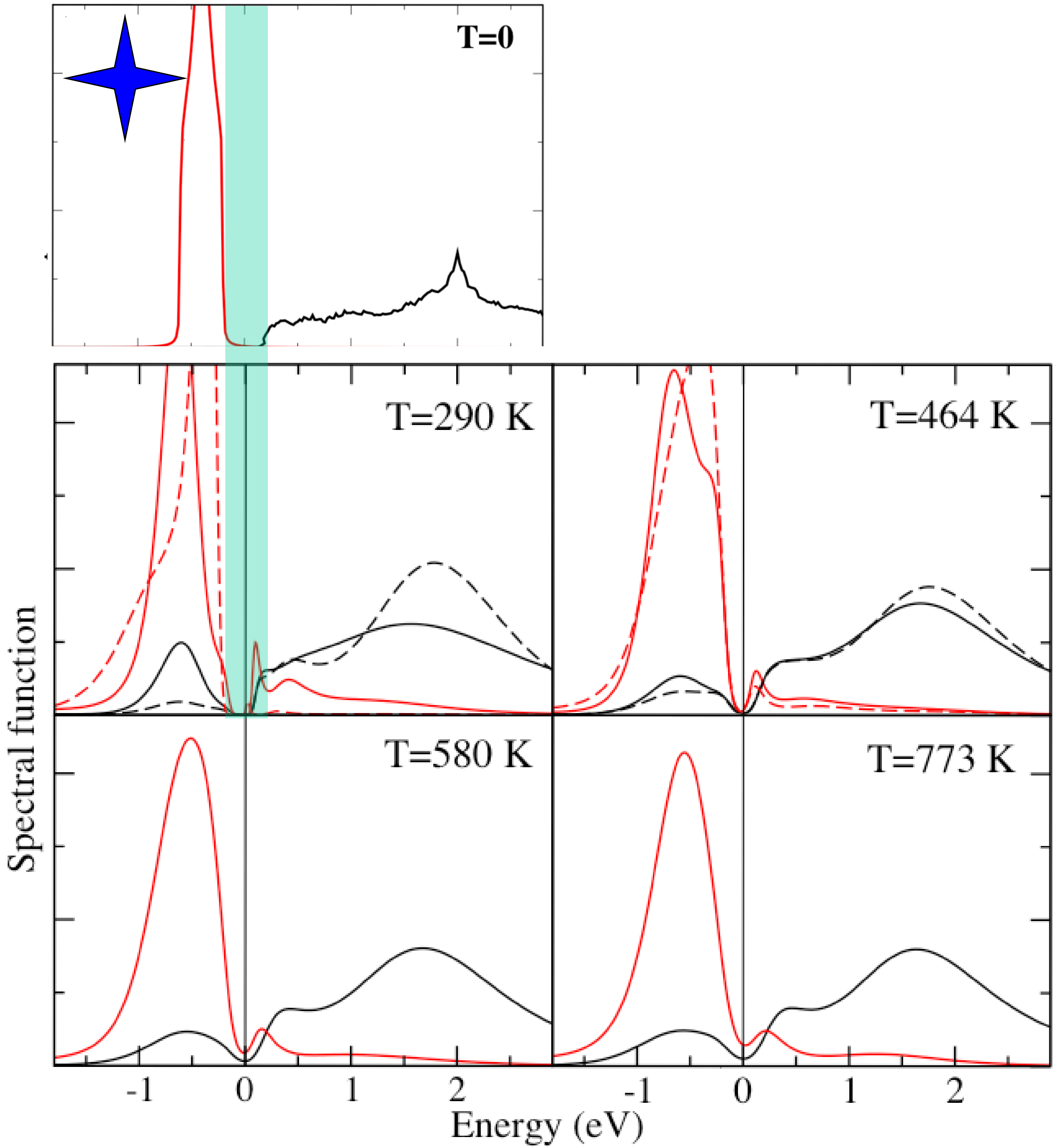
Mean-field for the BEG model

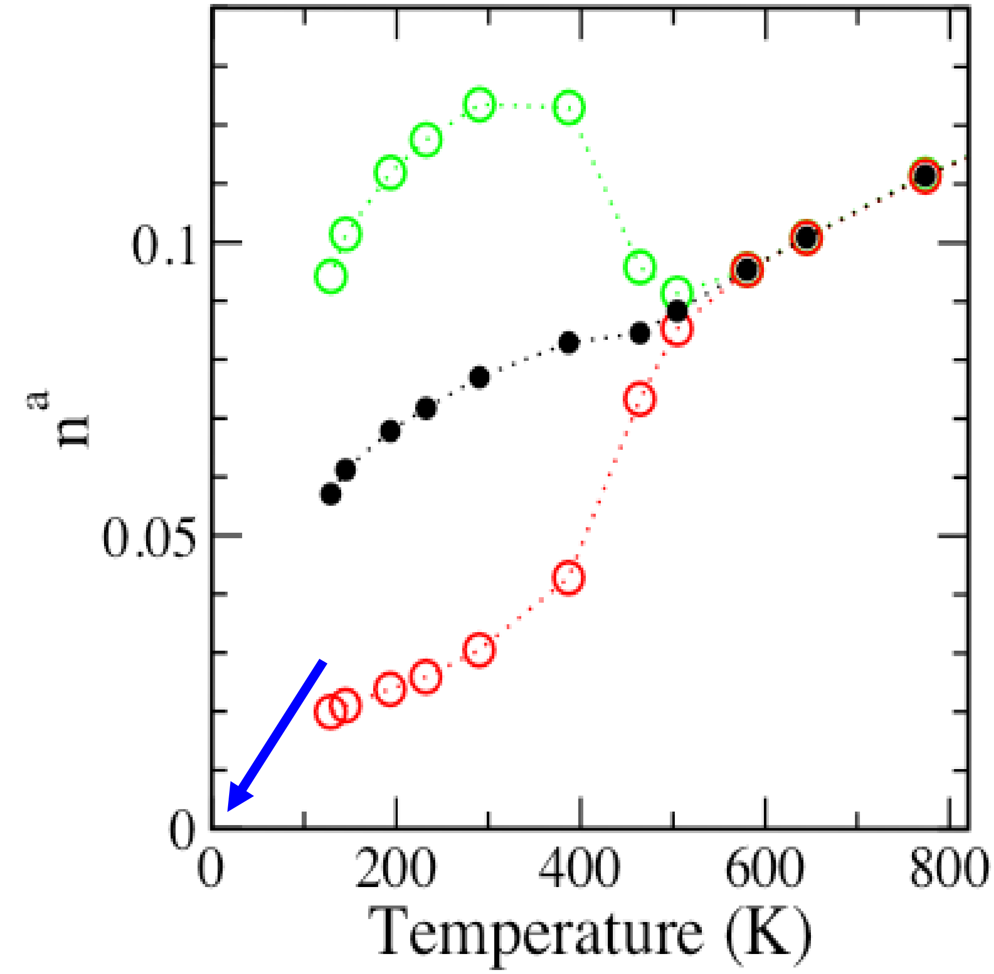


Hoston & Berker, Phys. Rev. Lett. ***67****, 1027 (1991)*



One-particle spectral densities

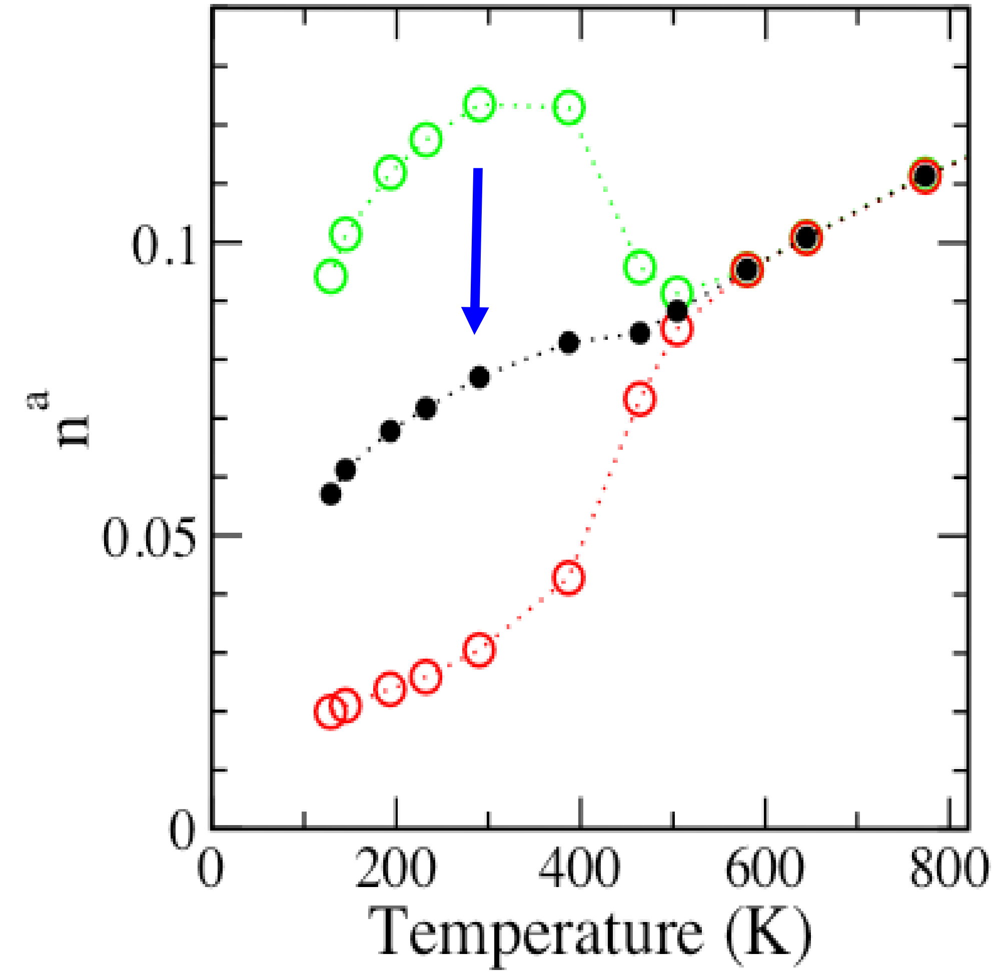


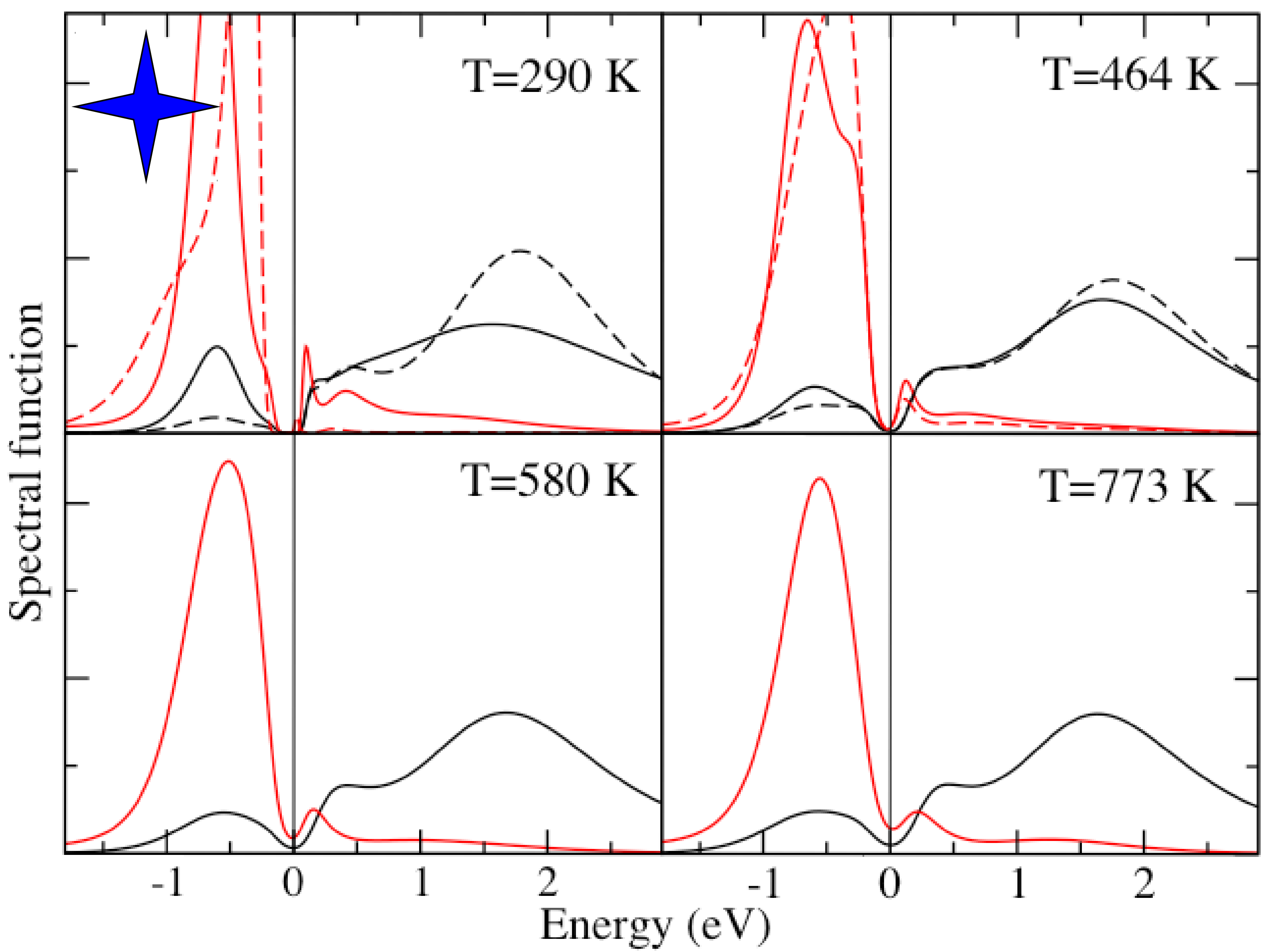


One-particle spectral densities

$A_{aa}(\omega)$ ——

$A_{bb}(\omega)$ ——

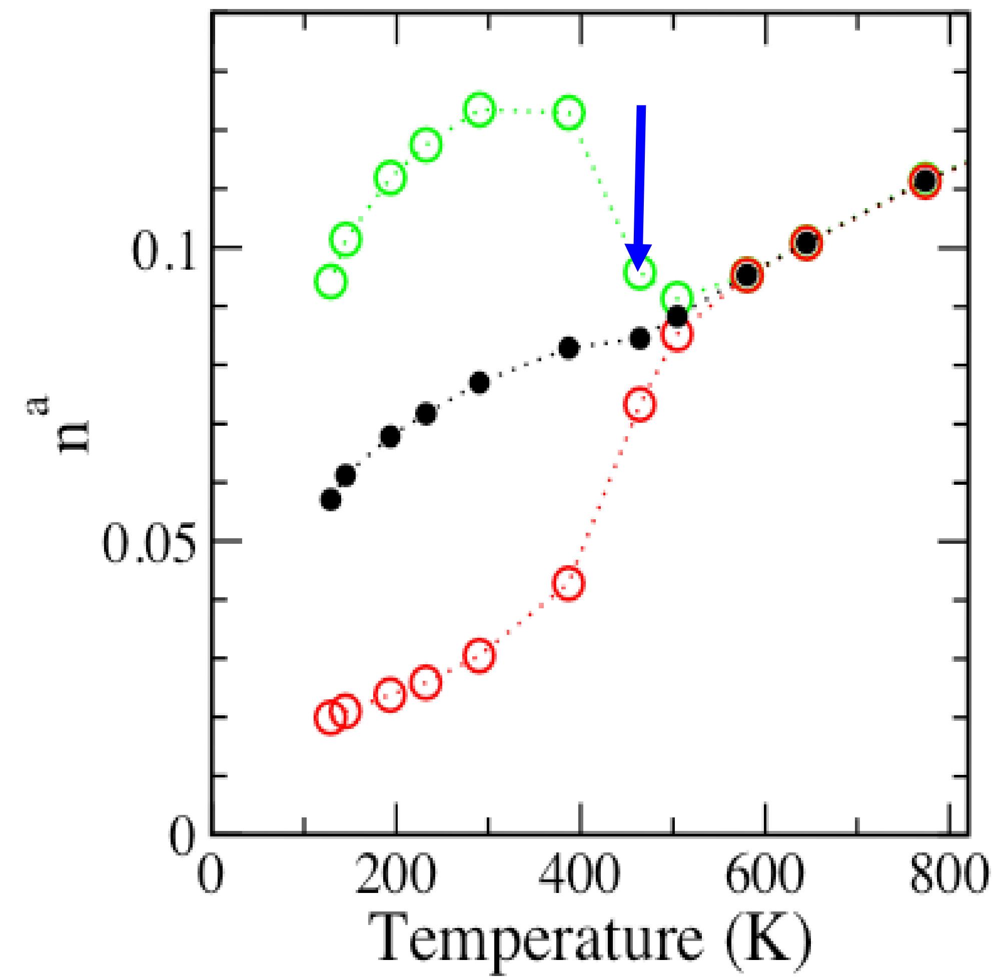


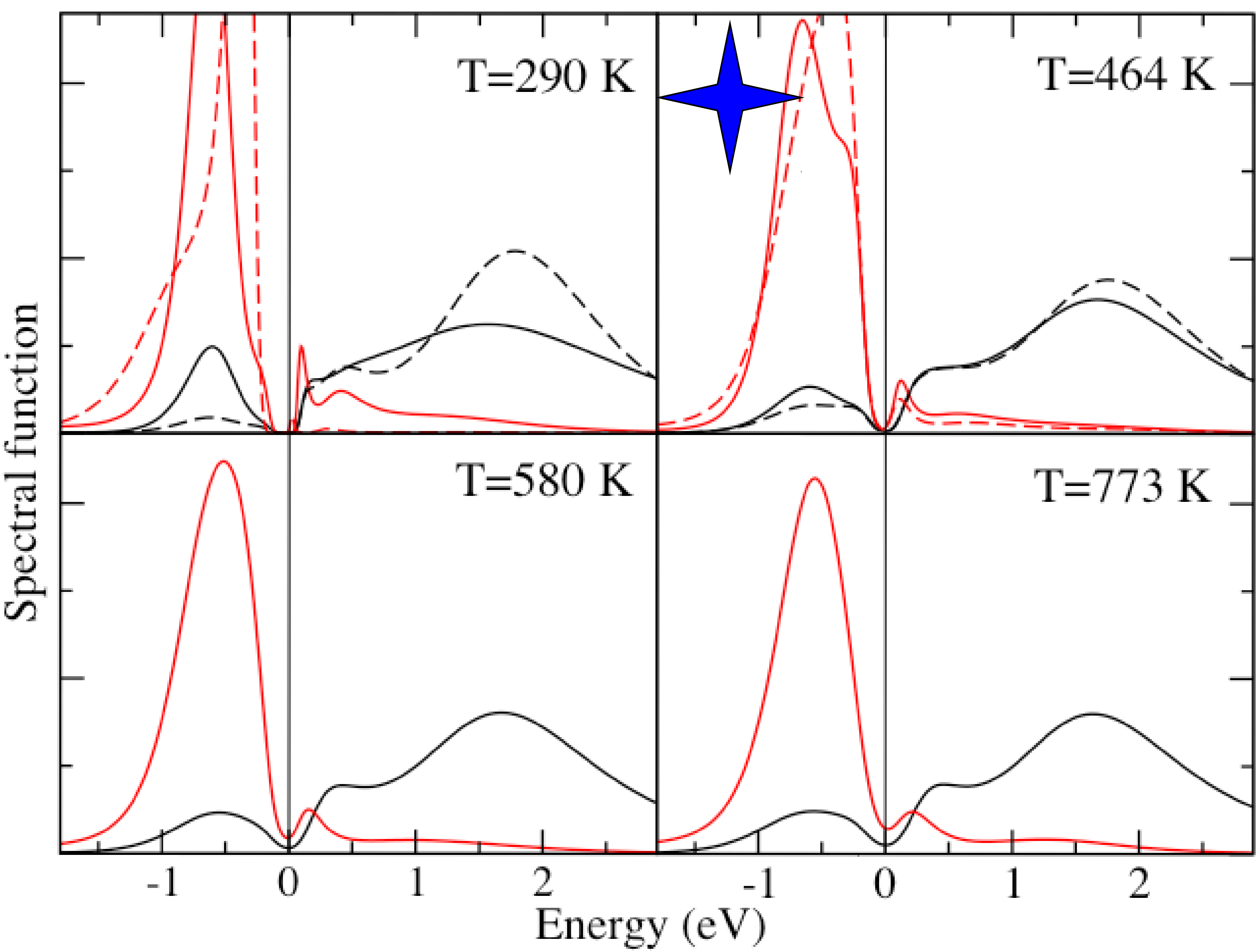


One-particle spectral densities

$A_{aa}(\omega)$ —— (black)

$A_{bb}(\omega)$ —— (red)

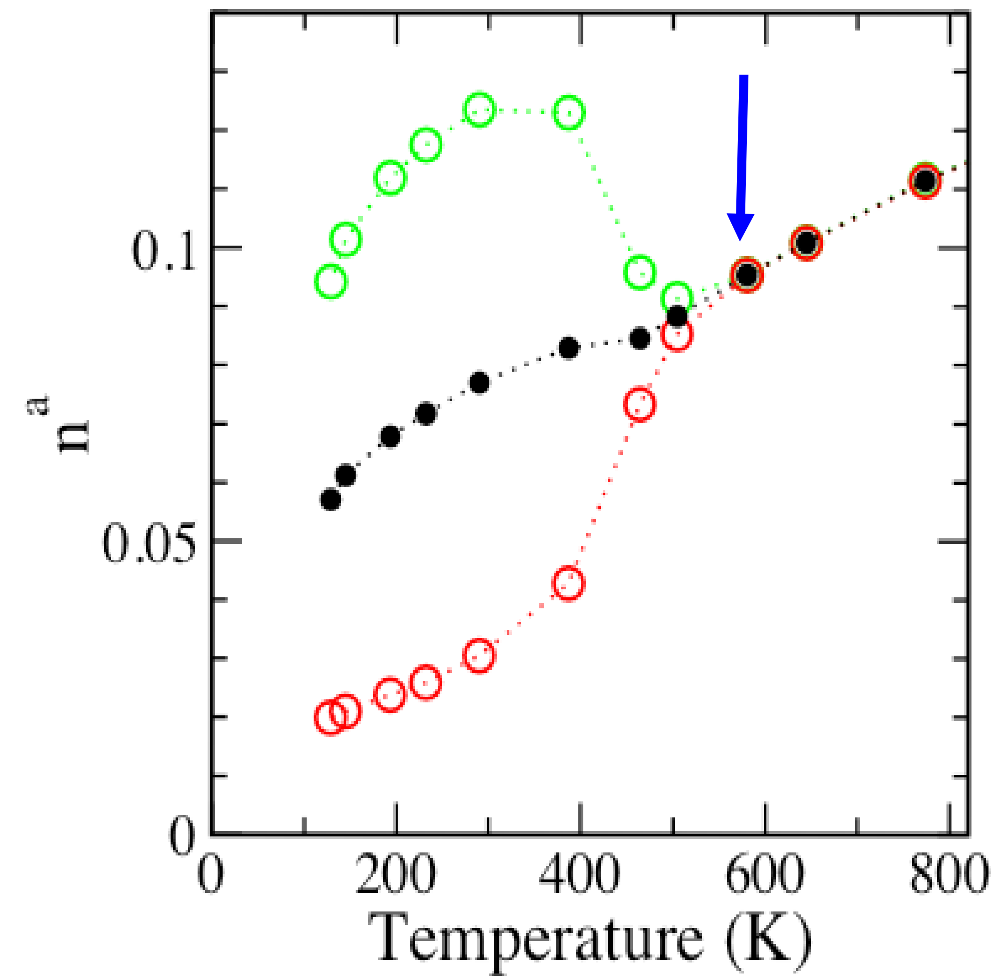


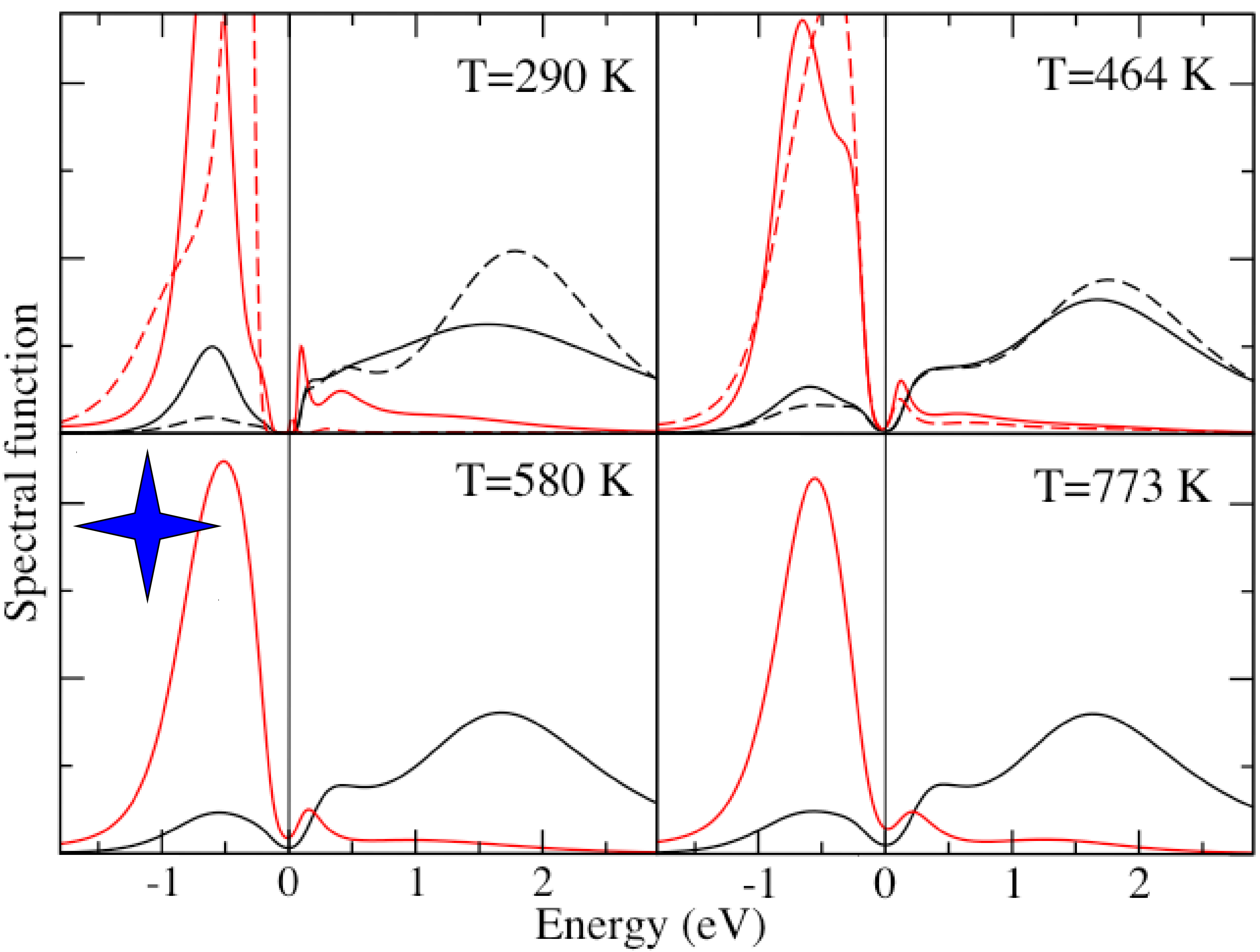


One-particle spectral densities

$A_{aa}(\omega)$

$A_{bb}(\omega)$

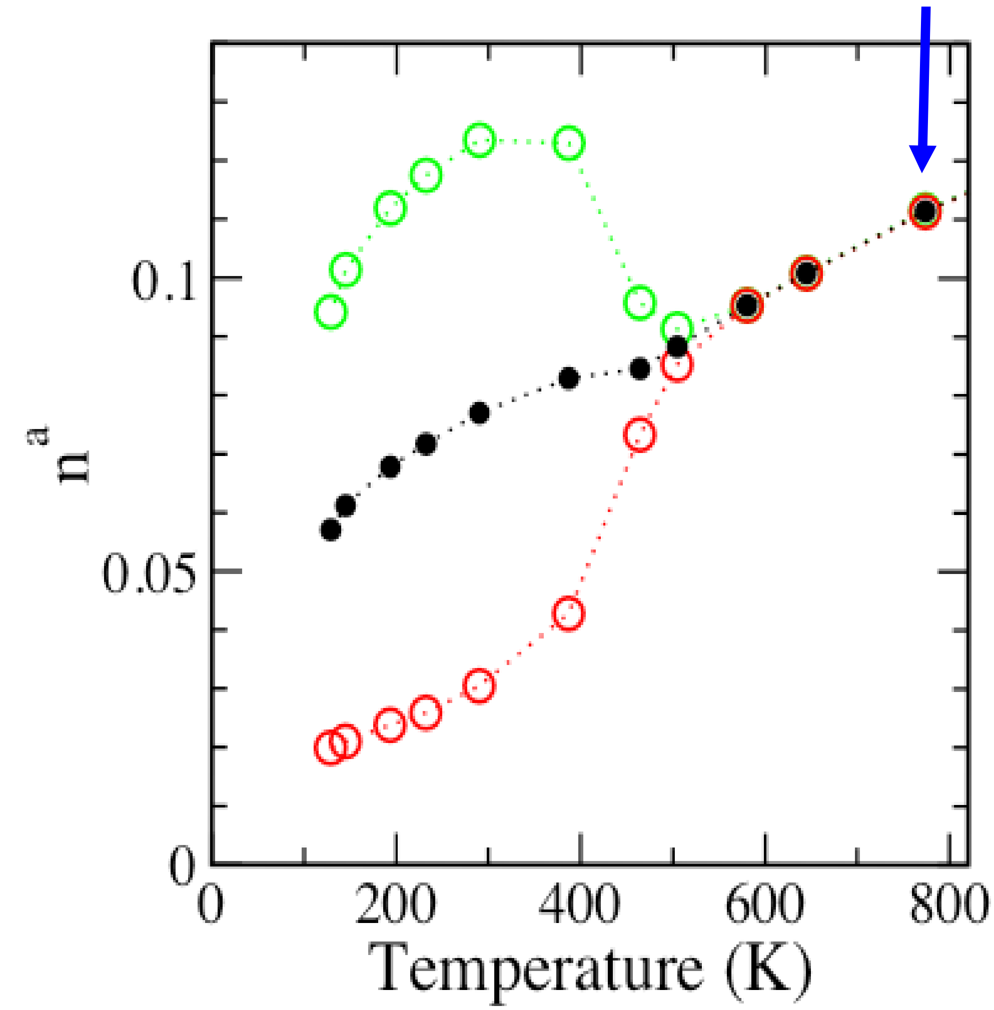


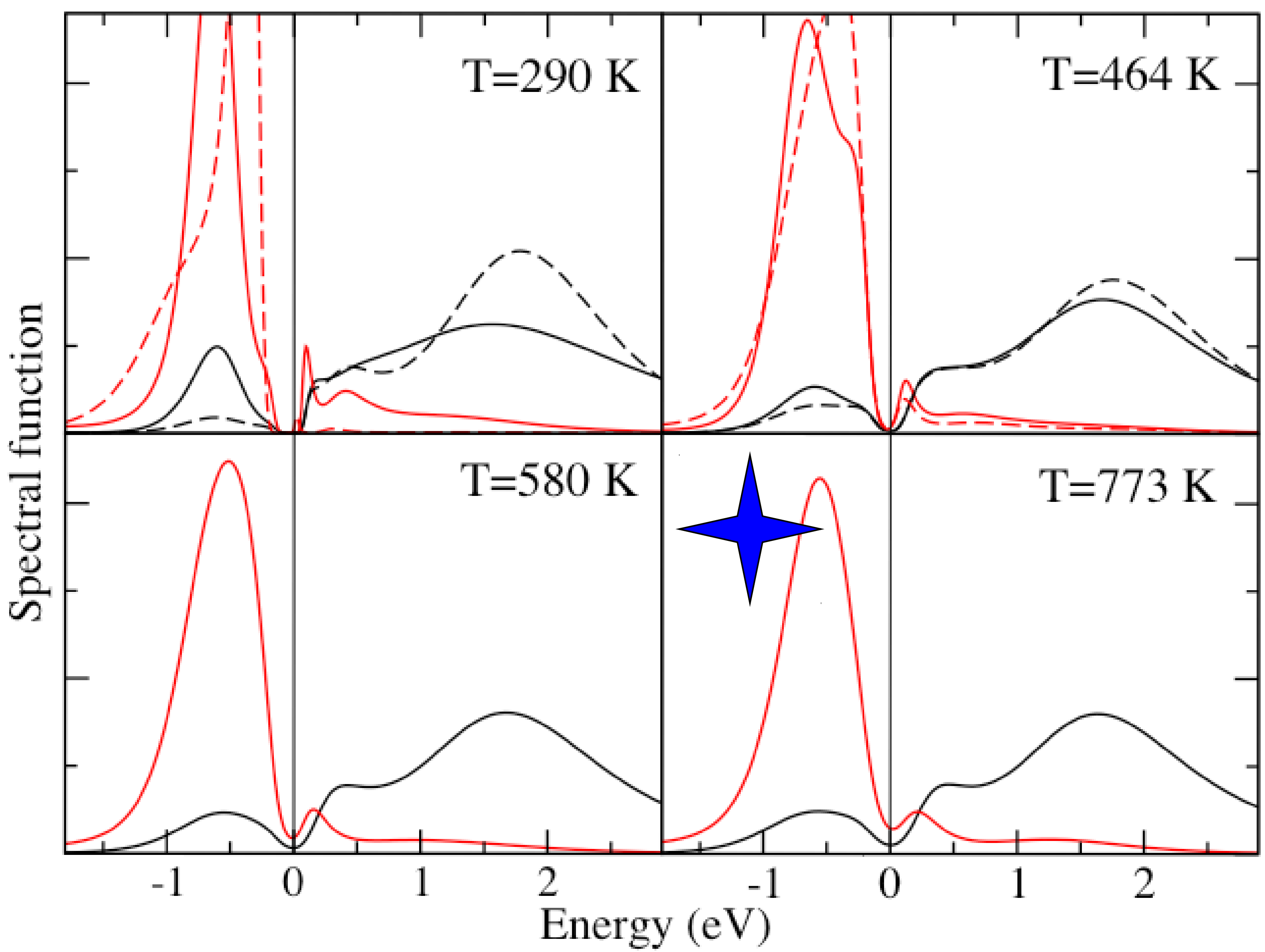


One-particle spectral densities

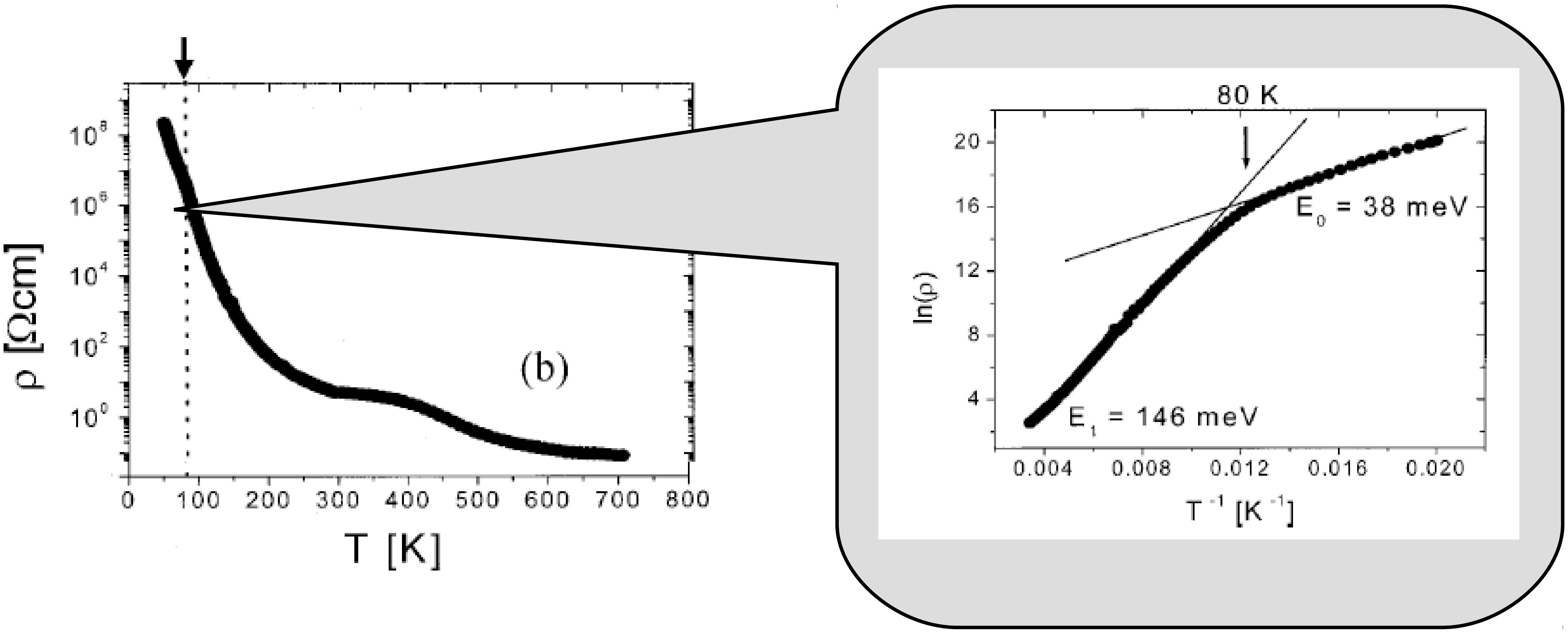
$A_{aa}(\omega)$

$A_{bb}(\omega)$





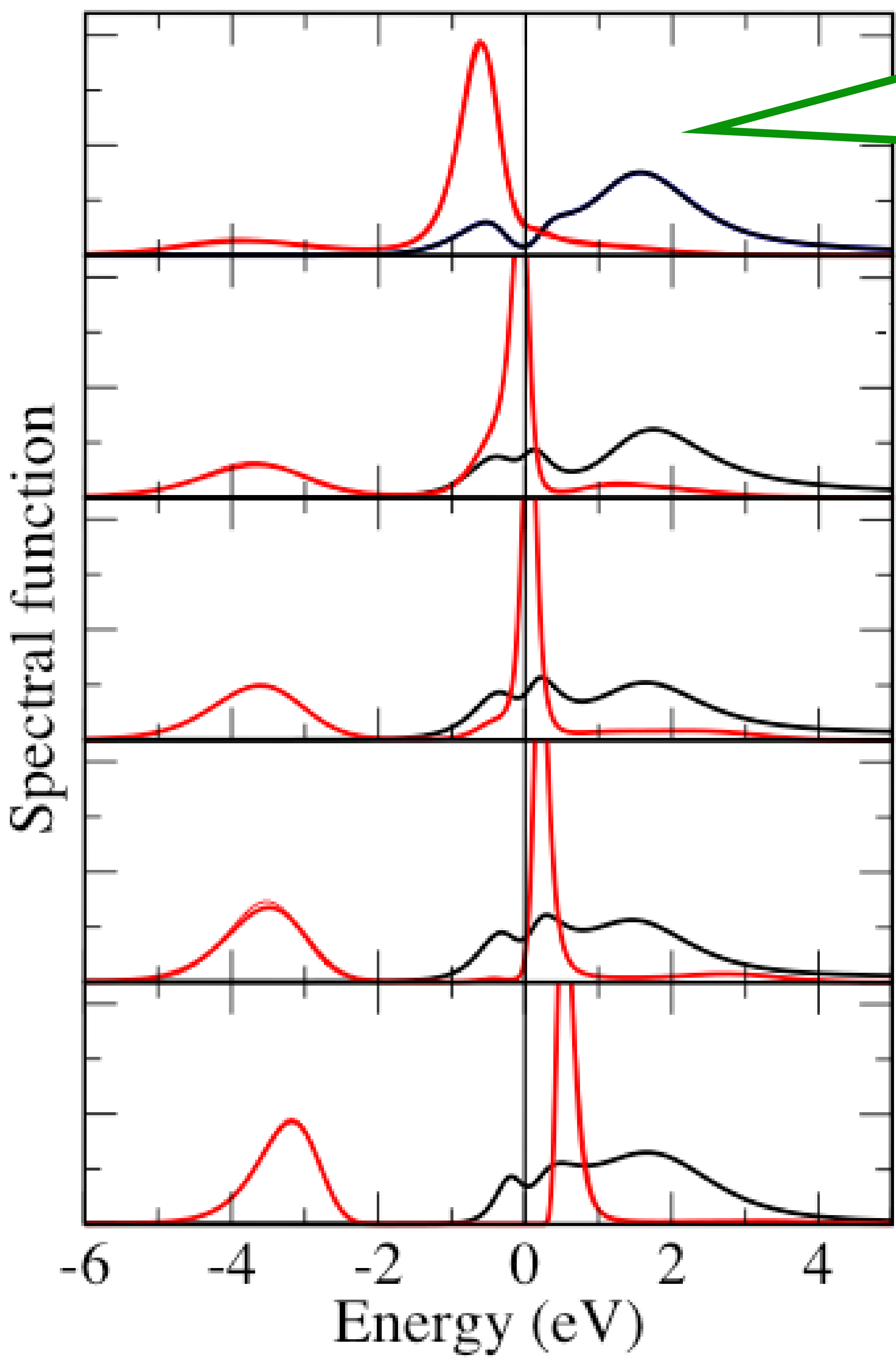
$LaCoO_3$



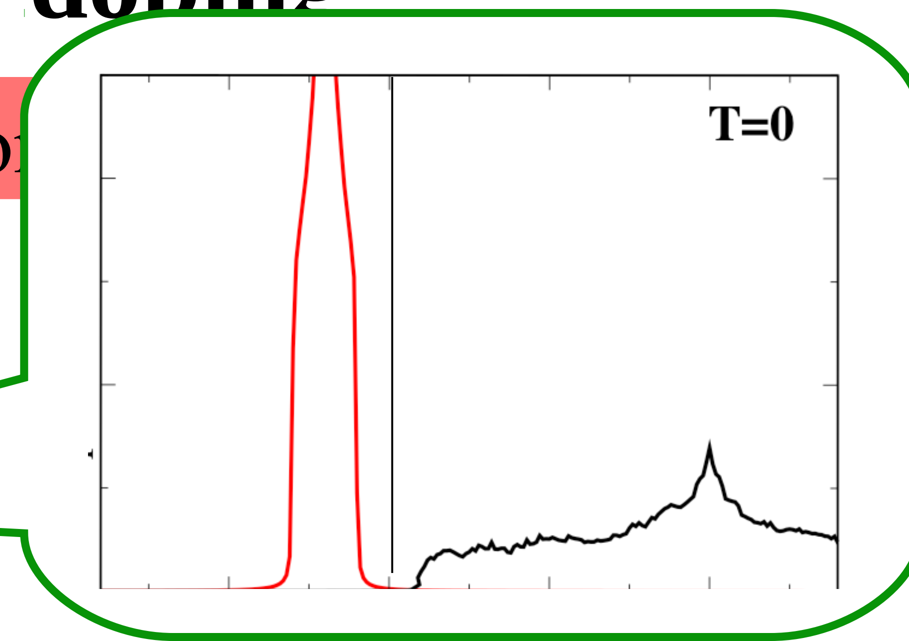
*English et al. Phys. Rev. B **65**, 220407 (2002)*

Hole doping

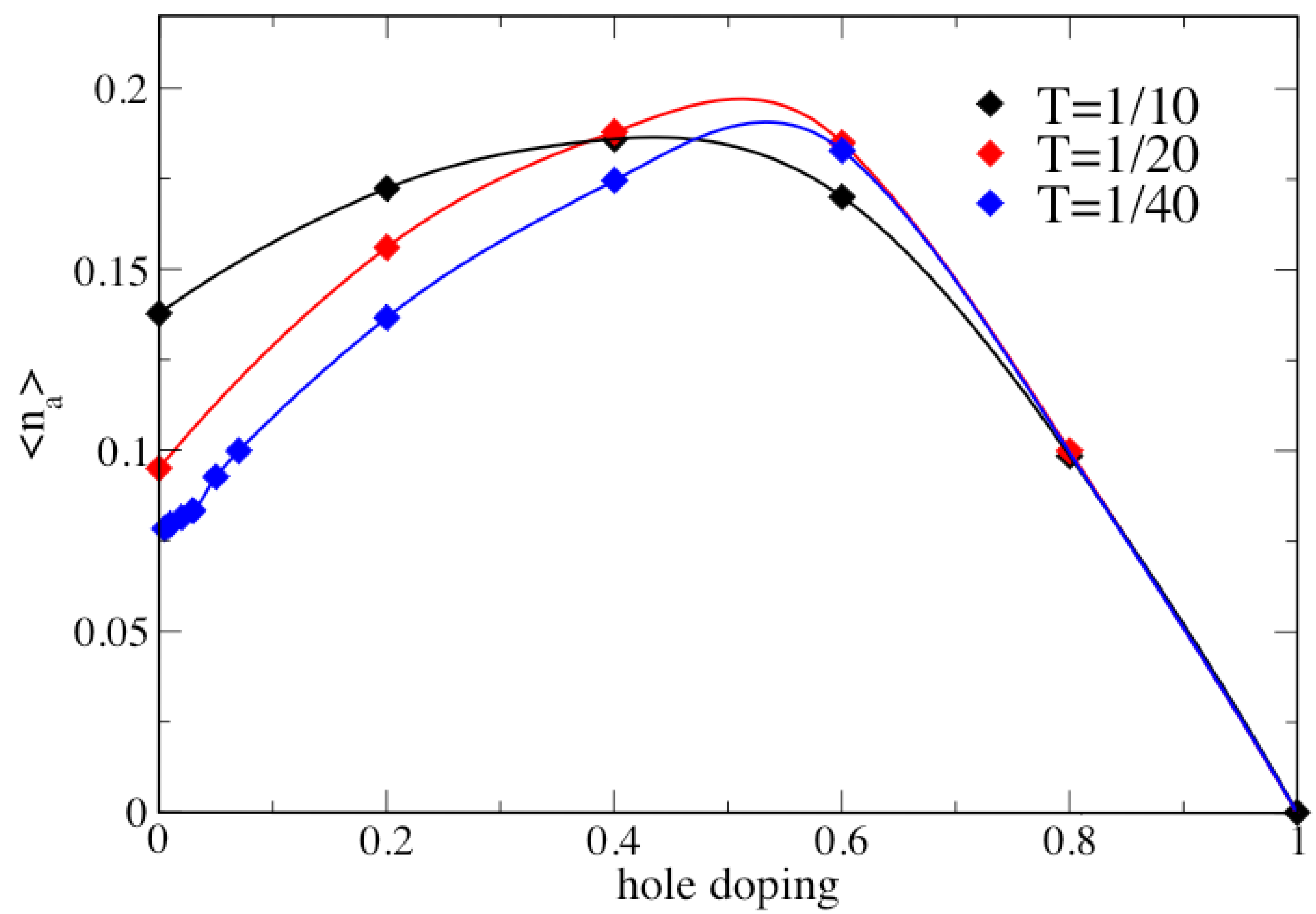
How do we from localized mo … picture?



hole doping

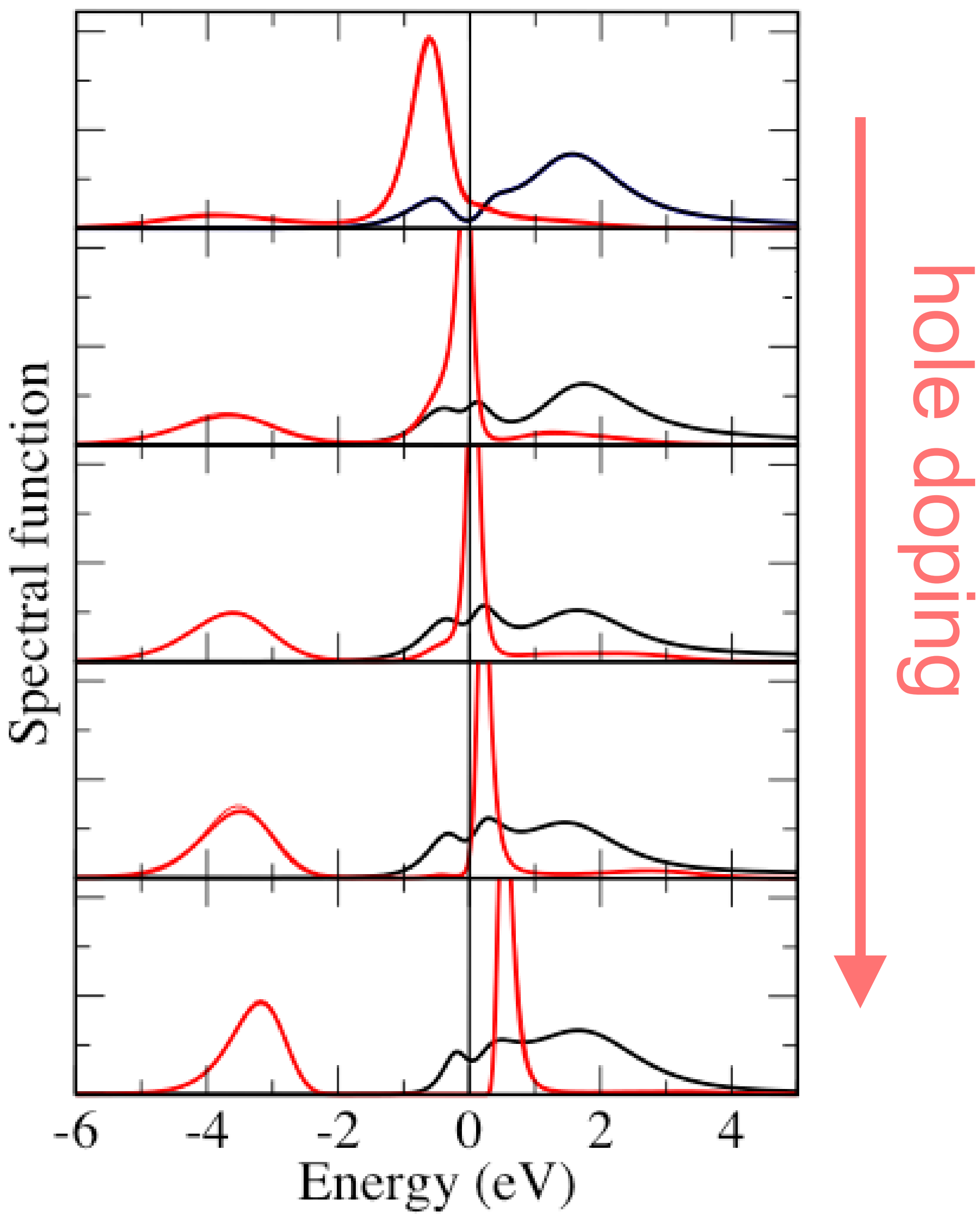


increases !

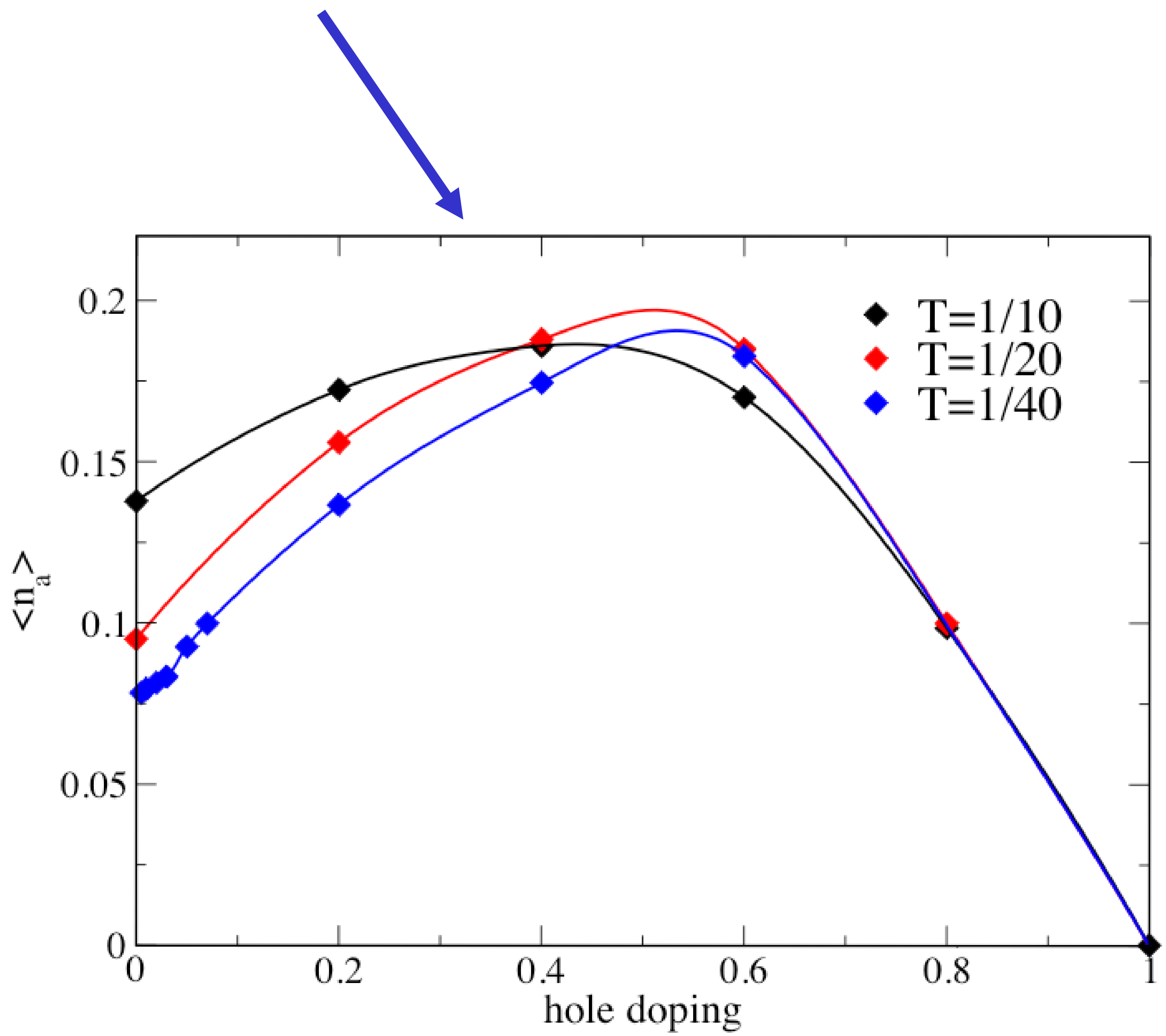


Hole doping

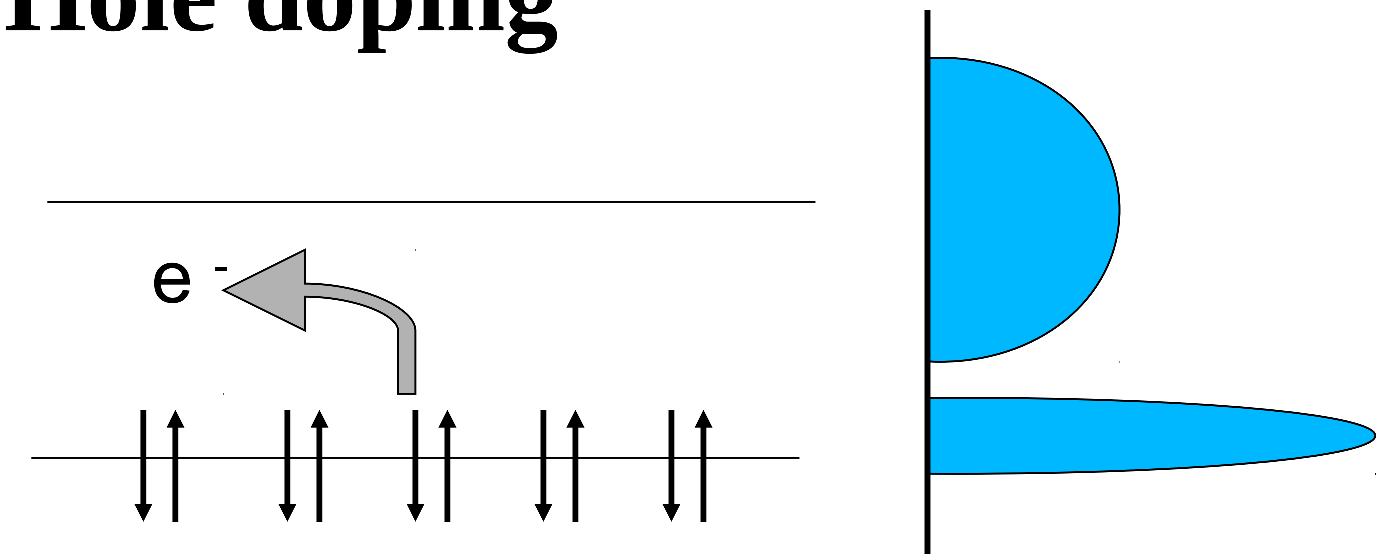
How do we from localized moments to double exchange picture?

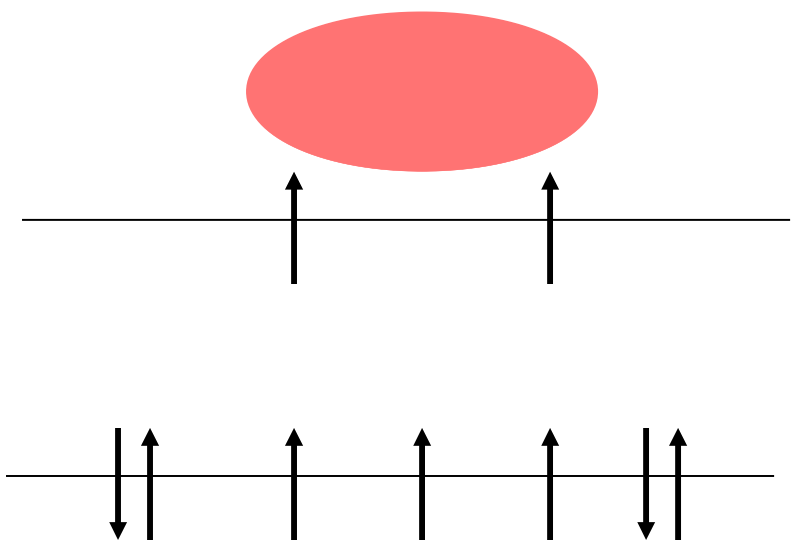


Upper band occupancy increases !

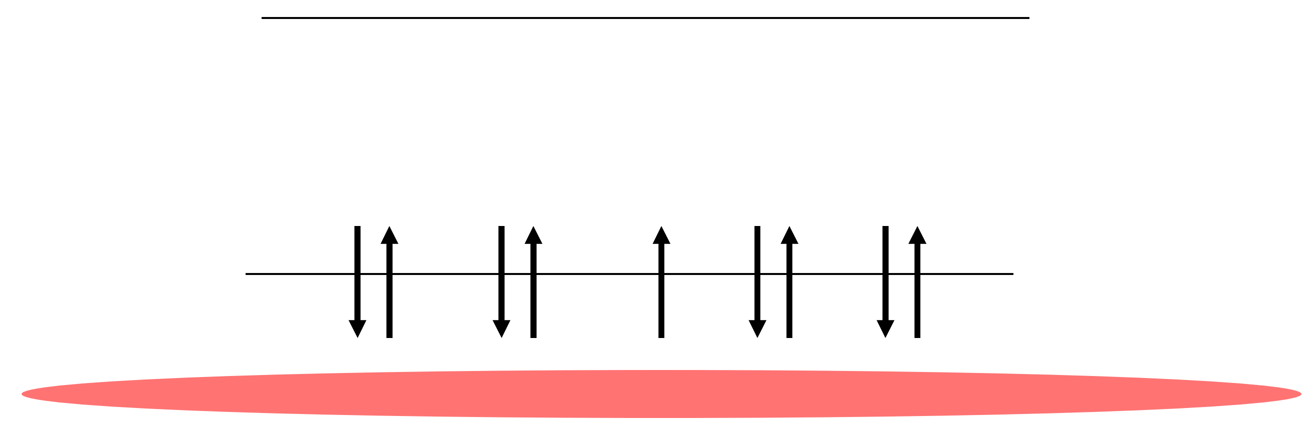


Hole doping





OR



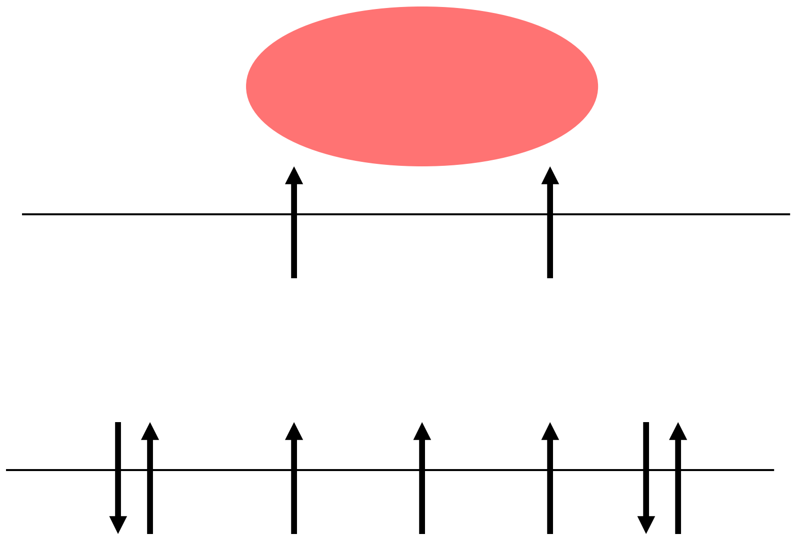
$$\sim -\sqrt{2}t_a + 2\xi_0$$

$$\sim -2t_b$$

localized magnetic polaron

itinerant hole in lower band

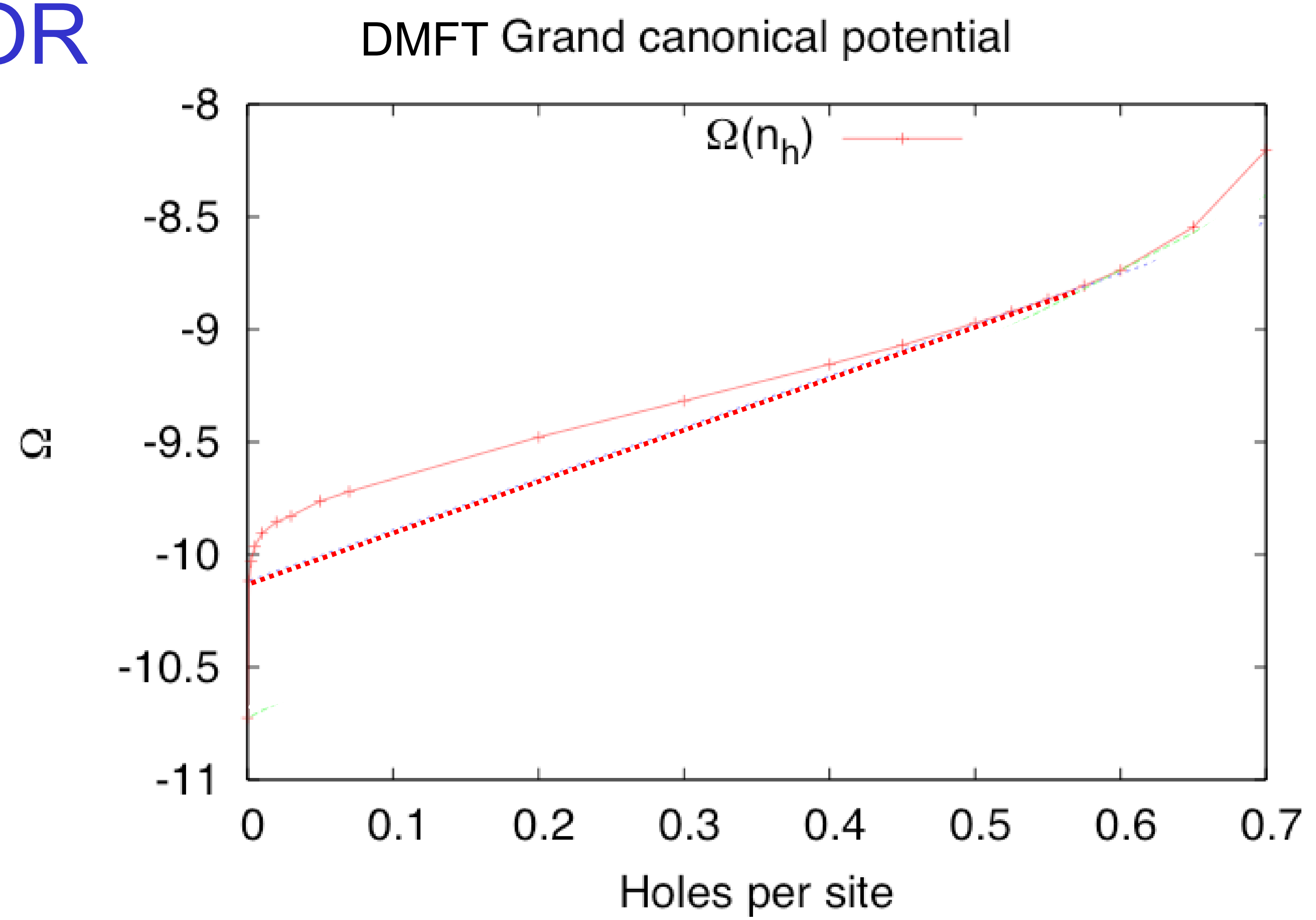
Hole doping



isolated magnetic polarons

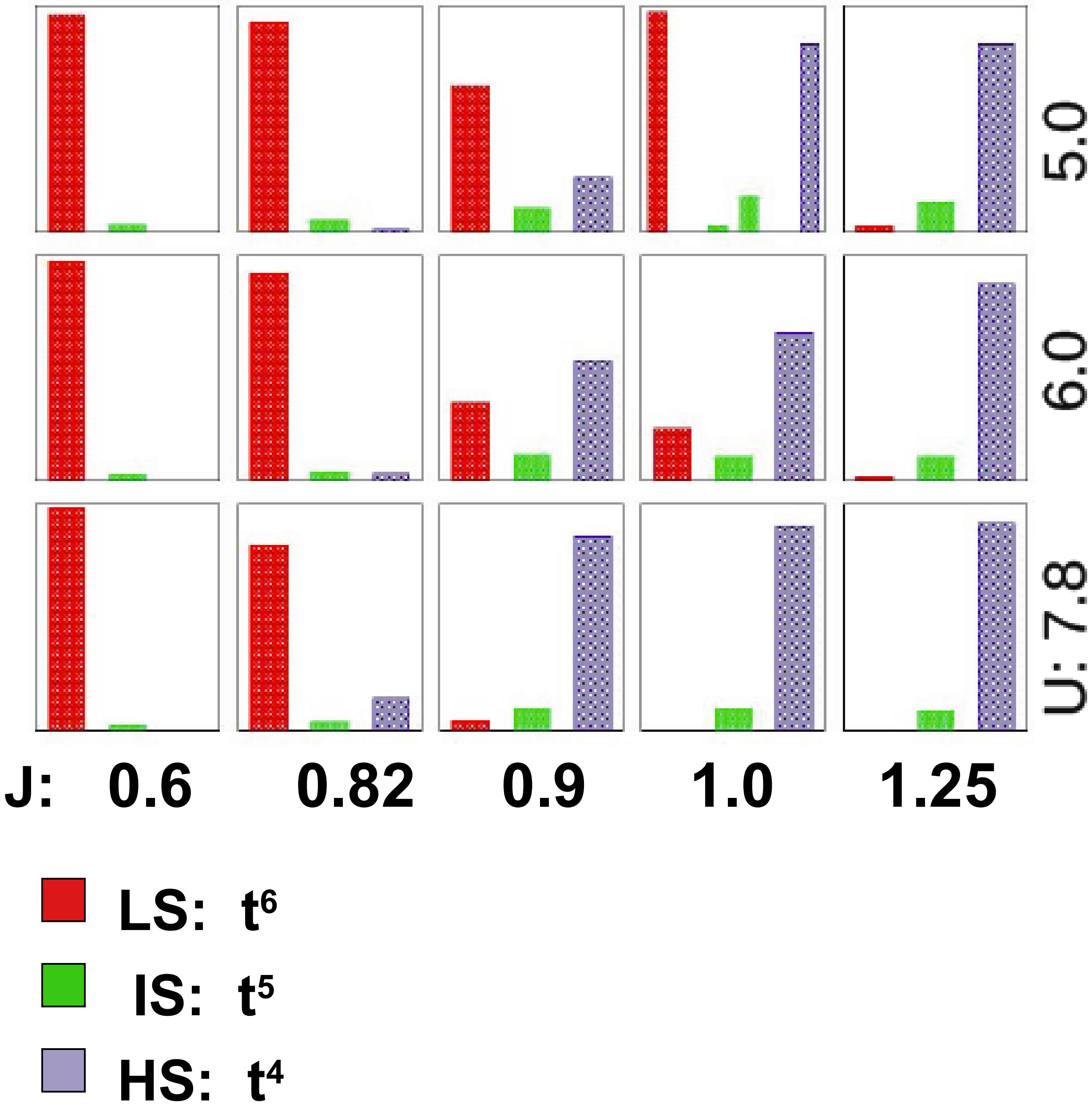
OR

phase separation

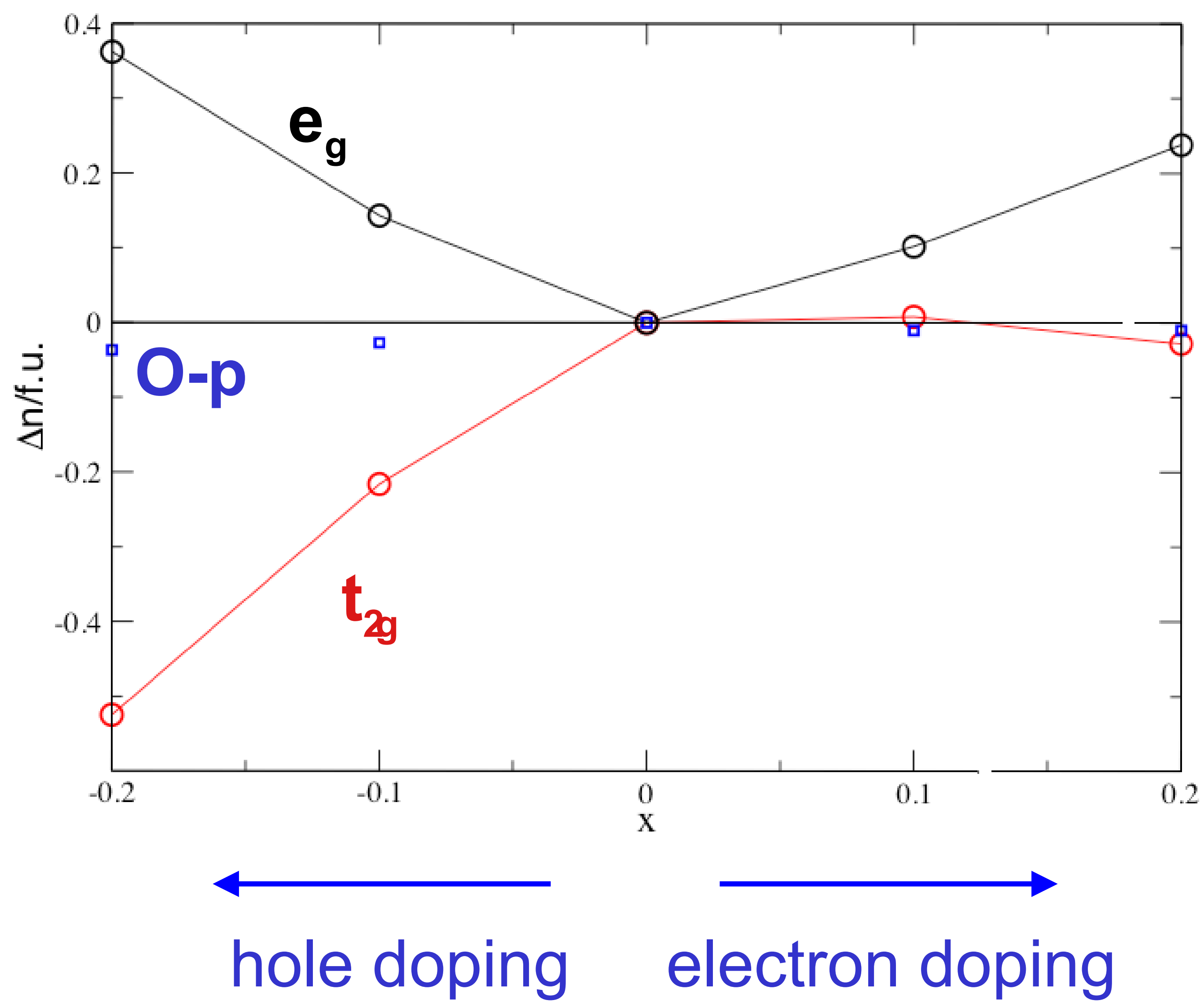


$LaCoO_3$ - LDA+DMFT results

Magnetic excitations = HS



Hole doping ($Sr_xLa_{1-x} CoO_3$)



Conclusions

- (Quasi)degeneracy of ionic multiplets leads to rich phase diagrams in strongly correlated systems.

- Effective HS-LS attraction at the HS/LS transitions leads to a ordered state with reduced translational symmetry.

- 2-band Hubbard model with crystal field provides fermionic realization of BEG model and introduces new parameter - doping.

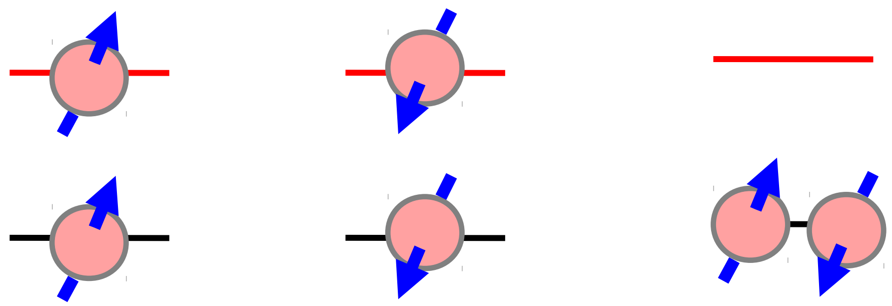
- Under certain circumstances ($W_a >> W_b$) doping leads to formation of inhomogeneities - magnetic polarons

- We observe similar physics in LDA+DMFT calculations for $LaCoO_3$ (LS vs HS competition in stoichiometric system, disproportionation, generation of IS by hole doping)

Low-energy model

Integrate out the charge fluctuations:

- keep 3 local states



- treat hopping as perturbation

Hamiltonian

$$\tilde{H} = \xi_0 \sum_{i,\sigma} n_{i,\sigma}^{\mathrm{HS}} + \sum_{\langle ij \rangle,\sigma} \left(\xi_1 n_i^{\mathrm{LS}} n_{j,\sigma}^{\mathrm{HS}} + \xi_2 n_{i,\sigma}^{\mathrm{HS}} n_{j,-\sigma}^{\mathrm{HS}} \right)$$

$$\xi_0 = \Delta - 3J,\ \xi_1 = -\frac{t_{aa}^2}{U-2J},\ \xi_2 = -\frac{2t_{aa}^2}{U+J}$$

Mean-field free energy

$$F(T) = \frac{\xi_0}{2}(x_A + x_B) + 2\xi_1 \left(x_A + x_B - 2x_A x_B\right) - \xi_2 x_A x_B$$
$$+ \frac{T}{2}(1 - x_A)\ln(1 - x_A) + \frac{T}{2}(1 - x_B)\ln(1 - x_B)$$
$$+ \frac{T}{2} x_A \ln(\frac{x_A}{2}) + \frac{T}{2} x_B \ln(\frac{x_B}{2}),$$