The Gutzwiller Density Functional Theory

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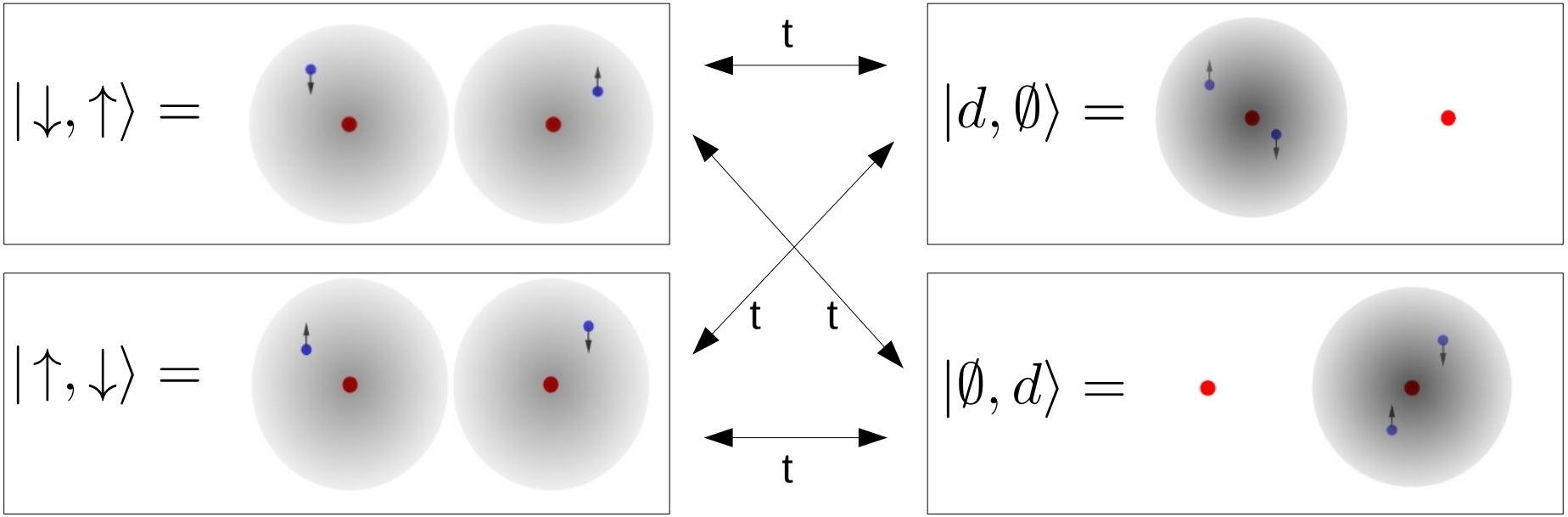
I) Introduction

1. Model for an H_2-molecule:

basis:

(spin: $S = 0$)



energies: $E_{\uparrow,\downarrow} = E_{\downarrow,\uparrow} = 0$

$E_{\emptyset,d} = E_{d,\emptyset} = U$

$t =$ matrix element for transitions:

$$|\uparrow,\downarrow\rangle \quad |d,\emptyset\rangle$$
$$|\downarrow,\uparrow\rangle \overset{t}{\leftrightarrow} |\emptyset,d\rangle$$

U : Coulomb interaction (intra-atomic)

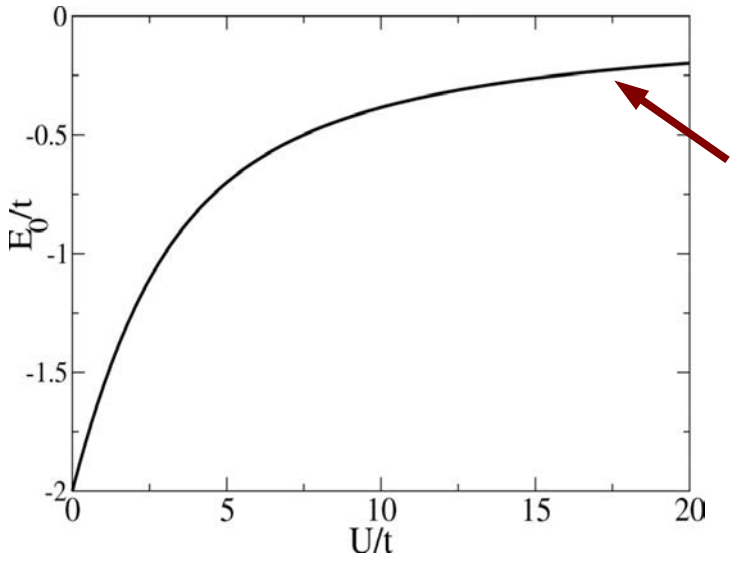
1.1. Perspective of elementary quantum mechanics:

$$\hat{H}_H = t\Big[\Big(|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle\Big)\Big(\langle d,\emptyset| + \langle\emptyset,d|\Big) + \text{h.c.}\Big] + U\Big[|d,\emptyset\rangle\langle d,\emptyset| + |\emptyset,d\rangle\langle\emptyset,d|\Big]$$

$\hat{=}\ 4\times 4-$ matrix

$$\longrightarrow\ E_0 = \frac{1}{2}(U - \sqrt{U+16t^2})$$

(ground-state energy)



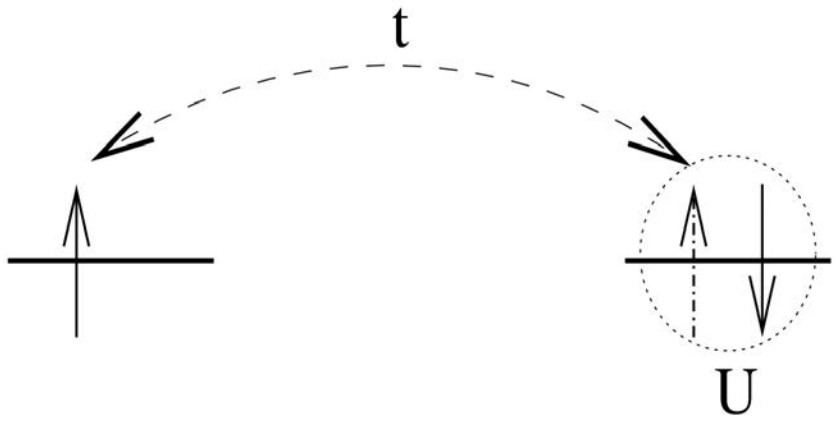
$$\sim -\frac{t^2}{U}$$

ground state: $|\Psi_0^U\rangle = \frac{1}{\sqrt{2}}\Big[\alpha_1(|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle) - \alpha_2(|d,\emptyset\rangle + |\emptyset,d\rangle)\Big]$

$$\xrightarrow[(U=0)]{} |\Psi_0^0\rangle = \frac{1}{2}\Big[(|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle) - (|d,\emptyset\rangle + |\emptyset,d\rangle)\Big]$$

1.2. Perspective of solid-state theory

$$\hat{H}_H = t \sum_{s=\uparrow,\downarrow} \left(\hat{c}^\dagger_{1s} \hat{c}_{2s} + \hat{c}^\dagger_{2s} \hat{c}_{1s} \right)$$
$$+U \sum_{i=1}^{2} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$



first: solve the 'single-particle problem' ($U = 0$) :

$\hat{h}^\dagger_{\pm,s} \equiv \frac{1}{\sqrt{2}} (\hat{c}^\dagger_{1s} \pm \hat{c}^\dagger_{2s})$ ('molecular orbitals': 'bonding' & 'anti-bonding')

$\longrightarrow \quad \hat{H}_H = -t \sum_{s=\uparrow,\downarrow} \left(\hat{h}^\dagger_{-,s} \hat{h}_{-,s} - \hat{h}^\dagger_{+,s} \hat{h}_{+,s} \right)$

ground state: $|\Psi^0_0\rangle = \hat{h}^\dagger_{-,\uparrow} \hat{h}^\dagger_{-,\downarrow} |0\rangle$

$U = 0 \longrightarrow$ ground state is 'single-particle product state'

Effective single-particle ('Hartree-Fock') theory:

Idea: find the single-particle product state $|\Psi_0^{\mathrm{HF}}\rangle$ with the lowest energy $E_0^{\mathrm{HF}} = \langle\Psi_0^{\mathrm{HF}}|\hat{H}_H|\Psi_0^{\mathrm{HF}}\rangle$

Problems: i) for $U \neq 0$, $|\Psi_0^U\rangle$ is never a single-particle product state

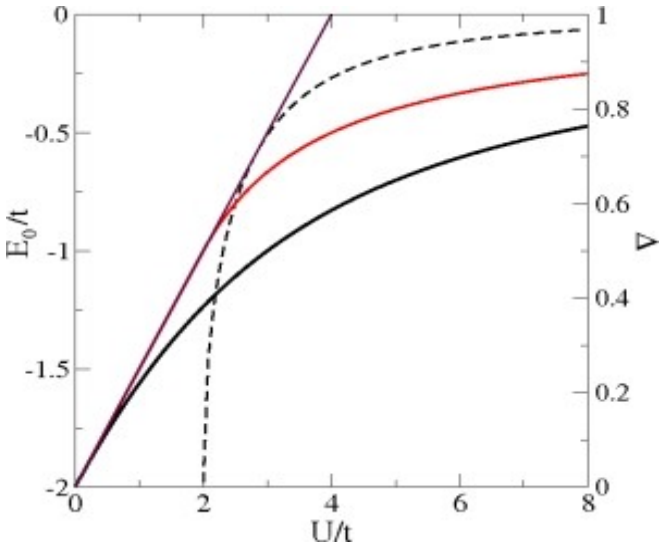
ii) the HF ground state with the 'correct' spin symmetry is $|\Psi_0^0\rangle$

('correct' spin symmetry: eigenstate of $\hat{\vec{S}}^2$ with eigenvalue $S = 0$)

→ energy $E_0^0 = \langle\Psi_0^0|\hat{H}_H|\Psi_0^0\rangle = -2t + U/2 \overset{(U\to\infty)}{\to} \infty$

iii) Hartree-Fock ground state breaks spin-symmetry for $U > 2t$:

$$\Delta \equiv \langle n_{1\uparrow}\rangle - \langle n_{1\downarrow}\rangle = \langle n_{2\downarrow}\rangle - \langle n_{2\uparrow}\rangle \geq 0$$



1.3. Perspective of many-particle theory

$$\hat{H}_H = t \sum_{s=\uparrow,\downarrow} \left(\hat{c}^\dagger_{1s} \hat{c}_{2s} + \hat{c}^\dagger_{2s} \hat{c}_{1s} \right) + U \sum_{i=1}^{2} \underbrace{\hat{n}_{i\uparrow} \hat{n}_{i\downarrow}}_{\equiv \hat{d}_i} \longrightarrow \hat{D} = \sum_i \hat{d}_i$$

(operator for double occupancies)

Gutzwiller's idea: with increasing U, double occupancies are more and more suppressed in the ground state

Gutzwiller wave function: (M. C. Gutzwiller, PRL **10**, 159 (1963))

$$|\Psi_G\rangle \equiv g^{\hat{D}} |\Psi_0^0\rangle = g^{\hat{d}_1} g^{\hat{d}_2} |\Psi_0^0\rangle = \frac{1}{2} \Big[(|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle) - g(|d,\emptyset\rangle + |\emptyset,d\rangle) \Big]$$

compare: $$|\Psi_0^U\rangle = \frac{1}{\sqrt{2}} \Big[\alpha_1 (|\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle) - \alpha_2 (|d,\emptyset\rangle + |\emptyset,d\rangle) \Big]$$

proper choice of the variational parameter g

→ reproduces the exact ground state

2. Transition metals and their compounds

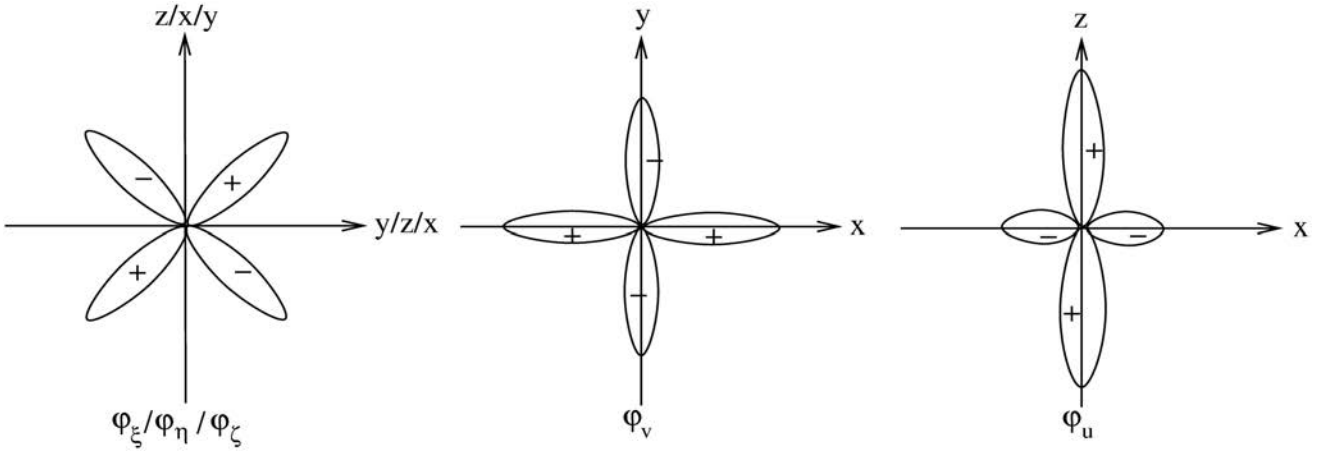
2.1. Transition-metal atoms

partially filled d-shell: $Y_{l,m}$ with $l = 2$ and $m = -2, -1, 0, 1, 2$

in cubic environment:

$$\left.\begin{aligned} \varphi_\xi &\sim \mathrm{i}(Y_{2,1} + Y_{2,-1}) \sim yz \\ \varphi_\eta &\sim (Y_{2,1} - Y_{2,-1}) \sim xz \\ \varphi_\zeta &\sim \mathrm{i}(Y_{2,2} - Y_{2,-2}) \sim xy \end{aligned}\right\} \; t_{2\mathrm{g}}- \text{ orbitals}$$

$$\left.\begin{aligned} \varphi_u &= Y_{2,0} \sim (3z^2 - r^2) \\ \varphi_v &\sim (Y_{2,2} + Y_{2,-2}) \sim (x^2 - y^2) \end{aligned}\right\} \; e_{\mathrm{g}}- \text{ orbitals}$$



2.2. Lattice models

$$\hat{H}_{\mathrm{H}} = \sum_{i,j,s=\uparrow,\downarrow} t_{i,j}\hat{c}^{\dagger}_{is}\hat{c}_{js} + U\sum_{i}\hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$ ('single-band Hubbard model')

Hamiltonian for transition metals with partially filled d-shells:

$$\hat{H}_{\mathrm{H}} = \sum_{i\neq j;\sigma,\sigma'} t^{\sigma,\sigma'}_{i,j}\hat{c}^{\dagger}_{i,\sigma}\hat{c}_{j,\sigma'} + \sum_{i}\hat{H}_{\mathrm{loc},i}$$ ('multi-band Hubbard models")

with the local 'atomic' Hamiltonian:

$$\hat{H}_{\mathrm{loc},i} = \sum_{\sigma_1,\sigma_2,\sigma_3,\sigma_4} U_{\sigma_1,\sigma_2,\sigma_3,\sigma_4}\hat{c}^{\dagger}_{\sigma_1}\hat{c}^{\dagger}_{\sigma_2}\hat{c}_{\sigma_3}\hat{c}_{\sigma_4} + \sum_{\sigma}\varepsilon_{\sigma}\hat{c}^{\dagger}_{\sigma}\hat{c}_{\sigma}$$

σ : combined spin-orbital index

$U_{\sigma_1,\sigma_2,\sigma_3,\sigma_4}$: local Coulomb interaction

ε_{σ} : orbital energies

Local Hamiltonian for d-orbitals in cubic environment:

$$\hat{H}_{\rm loc} = \frac{1}{2}\sum_{b,s} U(b,b)\hat{n}_{b,s}\hat{n}_{b,\bar{s}} + \frac{1}{2}\sum_{\substack{b(\neq)b' \\ s,s'}} (U(b,b') - \delta_{s,s'}J(b,b'))\hat{n}_{b,s}\hat{n}_{b',s'}$$
$$+\frac{1}{2}\sum_{b(\neq)b'} J(b,b')\left[\left(\hat{c}^\dagger_{b,\uparrow}\hat{c}^\dagger_{b,\downarrow}\hat{c}_{b',\downarrow}\hat{c}_{b',\uparrow} + \text{h.c.}\right) + \sum_s \hat{c}^\dagger_{b,s}\hat{c}^\dagger_{b',\bar{s}}\hat{c}_{b,\bar{s}}\hat{c}_{b',s}\right]$$
$$+ \sum_{\substack{t,e(\neq)e' \\ s,s'}} (T(t;e,e') - \delta_{s,s'}A(t;e,e'))\hat{n}_{t,s}\hat{c}^\dagger_{e,s'}\hat{c}_{e',s'}$$
$$+ \sum_{t,e(\neq)e'} A(t;e,e')\left[\left(\hat{c}^\dagger_{t,\uparrow}\hat{c}^\dagger_{t,\downarrow}\hat{c}_{e,\downarrow}\hat{c}_{e',\uparrow} + \text{h.c.}\right) + \sum_s \hat{c}^\dagger_{t,s}\hat{c}^\dagger_{e,\bar{s}}\hat{c}_{t,\bar{s}}\hat{c}_{e',s}\right]$$
$$+ \sum_{\substack{t(\neq)t'(\neq)t'',e \\ s,s'}} S(t,t';t'',e)\left(\hat{c}^\dagger_{t,s}\hat{c}^\dagger_{t',s'}\hat{c}_{t'',s'}\hat{c}_{e,s} + \text{h.c.}\right)$$

with 10 independent parameters $U(b,b'),\ldots,S(t,t';t'',e)$

in spherical approximation: 3 Racah parameters A, B, C

alternatively: use mean values $U' \sim \sum_{b\neq b'} U(b,b'), J \sim \sum_{b\neq b'} J(b,b')$ and $C/B = 4$

2.3. Interaction effects in transition-metal compounds

3d wave functions are rather localised

→ in solids, the local Coulomb interaction and the band-width are of the same order of magnitude

Experiment: - magnetism

- orbital order

- metal-insulator transitions

- magneto-resistance ('giant', 'colossal')

- high-temperature superconductivity

Theory: effective single-particle theories often fail

(Hartree-Fock or local density approximation in density-functional theory)

→ simplest example: fcc nickel

II) Gutzwiller variational theory

1. Gutzwiller wave functions

1.1 Definitions

for the single-band Hubbard model $\hat{H}_{\mathrm{H}} = \sum_{i,j,s} t_{i,j} \hat{c}^{\dagger}_{is} \hat{c}_{js} + U \sum_{i} \hat{d}_i$

one defines: $|\Psi_{\mathrm{G}}\rangle \equiv \hat{P}_{\mathrm{G}} |\Psi_0\rangle$ (Gutzwiller wave function)

mit i) $|\Psi_0\rangle$ arbitrary single-particle product wave function

ii) $\hat{P}_{\mathrm{G}} \equiv \prod_{i} \hat{P}_i$ und $\hat{P}_i = g^{\hat{d}_i} = 1 - (1-g)\hat{d}_i$

alternative formulation: $\hat{P}_i = \sum_{\Gamma} \lambda_{\Gamma} |\Gamma\rangle_{i\,i} \langle\Gamma|$

with 'atomic' eigenstates $|\Gamma\rangle_i$

and variational parameters λ_{Γ} :

$$|\Gamma\rangle_i = \begin{cases} |d\rangle_i = \hat{c}^{\dagger}_{i,\uparrow} \hat{c}^{\dagger}_{i,\downarrow} |\mathrm{vac}\rangle_{\mathrm{i}} \\ |s\rangle_i = \hat{c}^{\dagger}_{i,s} |\mathrm{vac}\rangle_{\mathrm{i}} \\ |\emptyset\rangle_i = |\mathrm{vac}\rangle_{\mathrm{i}} \end{cases}$$

multi-band Hubbard models

$$\hat{H}_{\mathrm{H}} = \sum_{i \neq j;\sigma,\sigma'} t_{i,j}^{\sigma,\sigma'} \hat{c}_{i,\sigma}^{\dagger} \hat{c}_{j,\sigma'} + \sum_{i} \hat{H}_{\mathrm{loc},i} \quad \text{mit} \quad H_{\mathrm{loc},i} = \sum_{\Gamma} E_{\Gamma} \underbrace{|\Gamma\rangle_{i\,i}\langle\Gamma|}_{= \hat{m}_{i;\Gamma}}$$

$|\Gamma\rangle$: atomic eigenstates with energies E_{Γ}

e.g.: 3d-shell: $2^{10} = 1024-$ dimensional local Hilbert space

(2^{10}: 10 spin-orbitals σ)

generalised Gutzwiller wave function:

$$|\Psi_{\mathrm{G}}\rangle \equiv \prod_{i} \hat{P}_i |\Psi_0\rangle \quad \text{with } \hat{P}_i = \sum_{\Gamma,\Gamma'} \lambda_{\Gamma,\Gamma'} |\Gamma\rangle_{i\,i}\langle\Gamma'|$$

$\lambda_{\Gamma,\Gamma'}$: matrix of variational parameters (in this lecture $\lambda_{\Gamma,\Gamma'} = \delta_{\Gamma,\Gamma'}\lambda_{\Gamma}$)

problem: $|\Psi_G\rangle$ is still a complicated many-particle wave function

$$\longrightarrow E^{\mathrm{GA}} = \frac{\langle\Psi_{\mathrm{G}}|\hat{H}|\Psi_{\mathrm{G}}\rangle}{\langle\Psi_{\mathrm{G}}|\Psi_{\mathrm{G}}\rangle}$$ cannot be evaluated in general

1.2. Evaluation: Diagrammatic expansion

We need to calculate

$$\langle \Psi_{\mathrm{G}} | \hat{c}^{\dagger}_{i,\sigma} \hat{c}_{j,\sigma'} | \Psi_{\mathrm{G}} \rangle = \left\langle (\hat{P}_i \hat{c}^{\dagger}_{i,\sigma} \hat{P}_i)(\hat{P}_j \hat{c}_{j,\sigma'} \hat{P}_j) \prod_{l \neq (i,j))} \hat{P}_l^2 \right\rangle_{\Psi_0}$$

$$\langle \Psi_{\mathrm{G}} | \hat{m}_{i;\Gamma} | \Psi_{\mathrm{G}} \rangle = \left\langle (\hat{P}_i \hat{m}_{i;\Gamma} \hat{P}_i) \prod_{l \neq i} \hat{P}_l^2 \right\rangle_{\Psi_0}$$

$$\langle \Psi_{\mathrm{G}} | \Psi_{\mathrm{G}} \rangle = \left\langle \prod_{l} \hat{P}_l^2 \right\rangle_{\Psi_0}$$

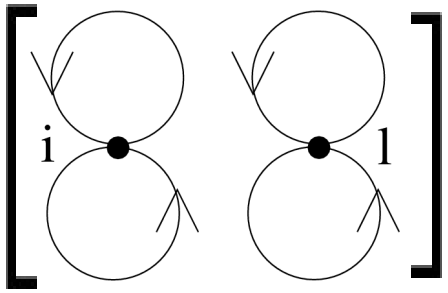
$|\Psi_0\rangle$ is a single-particle product wave function

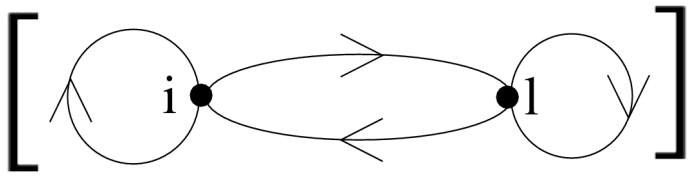
→ Wick theorem applies, e.g.,

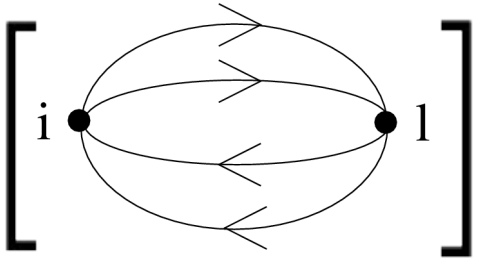
$$\langle \hat{c}^{\dagger}_1 \hat{c}_1 \hat{c}^{\dagger}_2 \hat{c}_2 \rangle_{\Psi_0} = \langle \hat{c}^{\dagger}_1 \hat{c}_1 \rangle_{\Psi_0} \langle \hat{c}^{\dagger}_2 \hat{c}_2 \rangle_{\Psi_0} - \langle \hat{c}^{\dagger}_1 \hat{c}_2 \rangle_{\Psi_0} \langle \hat{c}^{\dagger}_2 \hat{c}_1 \rangle_{\Psi_0}$$

→ Diagrammatic representation of all terms, e.g.,

$$\langle \hat{d}_i \hat{d}_l \rangle_{\Psi_0} = \left\langle (\hat{c}^\dagger_{i,\uparrow}\hat{c}_{i,\uparrow}\hat{c}^\dagger_{i,\downarrow}\hat{c}_{i,\downarrow})(\hat{c}^\dagger_{l,\uparrow}\hat{c}_{l,\uparrow}\hat{c}^\dagger_{l,\downarrow}\hat{c}_{l,\downarrow}) \right\rangle_{\Psi_0}$$

$=$ 

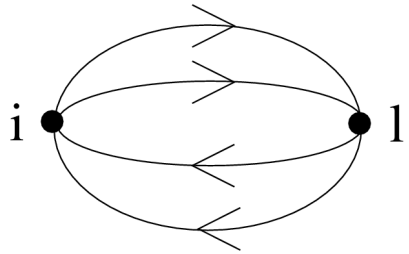
$-2\times$ 

$+$ 

with lines  $= \langle \hat{c}^\dagger_{l,\sigma}\hat{c}_{l',\sigma'} \rangle_{\Psi_0} = P^{\sigma,\sigma'}_{l,l'}$

1.3. Simplifications in infinite dimensions

A) Diagrams with three or more lines



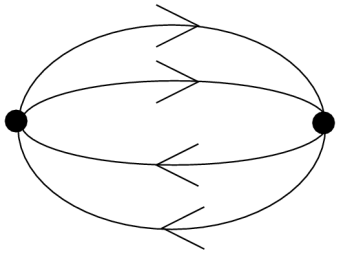
Kinetic energy per site on a D-dimensional lattice:

$$T_i = \sum_j t_{i,j} P_{i,j} \sim D t_{i,j} P_{i,j}$$

← (only n.n. hopping)

$$T_i \overset{!}{<} \infty \quad \text{for} \quad D \to \infty \quad \longrightarrow \quad t_{i,j}, P_{i,j} \sim 1/\sqrt{D}$$

$$\longrightarrow \quad \sum_l$$



$$= 0$$

B) Diagrams with two lines

$\langle \hat{d}_i \hat{P}_l^2 \rangle_{\Psi_0} =$ 

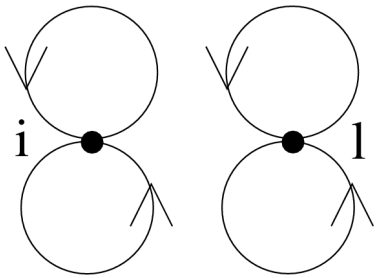
Idea: make sure that all these diagrams cancel each other

This is achieved by the (local) constraints

$$\langle \hat{c}^\dagger_{l,\sigma} \hat{c}_{l,\sigma'} \rangle_{\Psi_0} = \langle \hat{c}^\dagger_{l,\sigma} \hat{c}_{l,\sigma'} \hat{P}_l^2 \rangle_{\Psi_0}$$

$$1 = \langle \hat{P}_l^2 \rangle_{\Psi_0}$$

C) Disconnected diagrams are cancelled by the norm



Conclusion: All diagrams with internal vertices vanish

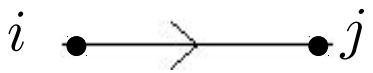
$$\langle \hat{m}_{i;\Gamma} \rangle_{\Psi_G} = \left\langle (\hat{P}_i \hat{m}_{i;\Gamma} \hat{P}_i) \right\rangle_{\Psi_0}$$

$$\langle \hat{c}^\dagger_{i,\sigma} \hat{c}_{j,\sigma'} \rangle_{\Psi_G} = \left\langle (\hat{P}_i \hat{c}^\dagger_{i,\sigma} \hat{P}_i)(\hat{P}_j \hat{c}_{j,\sigma'} \hat{P}_j) \right\rangle_{\Psi_0}$$

The remaining evaluation is rather straightforward:

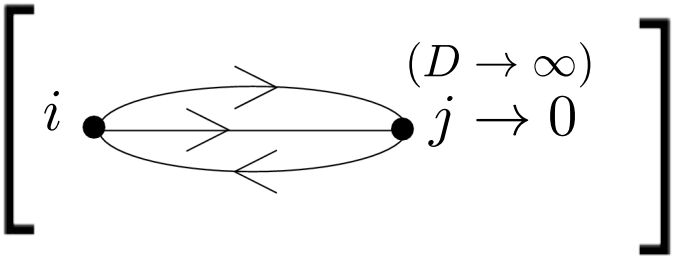
$$\langle \hat{m}_{i;\Gamma} \rangle_{\Psi_G} = \lambda^2_\Gamma \langle \hat{m}_{i;\Gamma} \rangle_{\Psi_0}$$

$$\langle \hat{c}^\dagger_{i,\sigma} \hat{c}_{j,\sigma'} \rangle_{\Psi_G} = \sum_{\gamma,\gamma'} q^\gamma_\sigma q^{\gamma'}_{\sigma'} \langle \hat{c}^\dagger_{i,\gamma} \hat{c}_{j,\gamma'} \rangle_{\Psi_0} =$$



with $q^\gamma_\sigma = q^\gamma_\sigma(\lambda_\Gamma)$

='renormalisation matrix'



1.4. Summary: energy functional in infinite spatial dimensions

Evaluation of $E^{\mathrm{GA}} = \langle \hat{H} \rangle_{\Psi_{\mathrm{G}}} = E_0 + E_{\mathrm{loc}}$

in the limit of infinite dimensions leads to

$$E_{\mathrm{loc}} = L \sum_{\Gamma} E_\Gamma m_\Gamma \quad (m_\Gamma \equiv \langle \hat{m}_\Gamma \rangle_{\Psi_{\mathrm{G}}})$$

$$E_0 = \sum_{i \neq j} \sum_{\gamma,\gamma'} \underbrace{\sum_{\sigma,\sigma'} q_\sigma^\gamma q_{\sigma'}^{\gamma'} t_{i,j}^{\sigma,\sigma'} \langle \hat{c}_{i,\gamma}^\dagger \hat{c}_{i,\gamma'} \rangle_{\Psi_0}}_{\equiv \tilde{t}_{i,j}^{\gamma,\gamma'}}$$

(effective hopping parameters)

m_Γ and q_σ^γ are analytic functions of

$$\lambda_\Gamma \text{ and } C^0_{\sigma,\sigma'} = \langle \hat{c}_{i,\sigma}^\dagger \hat{c}_{i,\sigma'} \rangle_{\Psi_0}$$

Remaining numerical problem:

Minimisation of E^{GA} with respect to λ_Γ and $|\Psi_0\rangle$

1.5. Minimisation of the energy functional

We consider E^{var} as a function of λ_Γ and of the non-interacting density matrix $\tilde{\rho}$ with the elements

$$\rho_{(i\sigma),(j\sigma')} \equiv \langle \hat{c}^\dagger_{j,\sigma'} \hat{c}_{i,\sigma} \rangle_{\Psi_0}$$

'non-interacting' $\longrightarrow$ $\tilde{\rho}^2 = \tilde{\rho}$

Minimisation with respect to $\tilde{\rho}$ leads to the effective single particle equation $\hat{H}_0^{\mathrm{eff}} |\Psi_0\rangle = E_0 |\Psi_0\rangle$

with $$\hat{H}_0^{\mathrm{eff}} = \sum_{i \neq j} \sum_{\sigma,\sigma'} \tilde{t}^{\sigma,\sigma'}_{i,j} \hat{c}^\dagger_{i,\sigma} \hat{c}_{j,\sigma'} + \sum_i \sum_{\sigma,\sigma'} \eta_{\sigma,\sigma'} \hat{c}^\dagger_{i,\sigma} \hat{c}_{i,\sigma'}$$

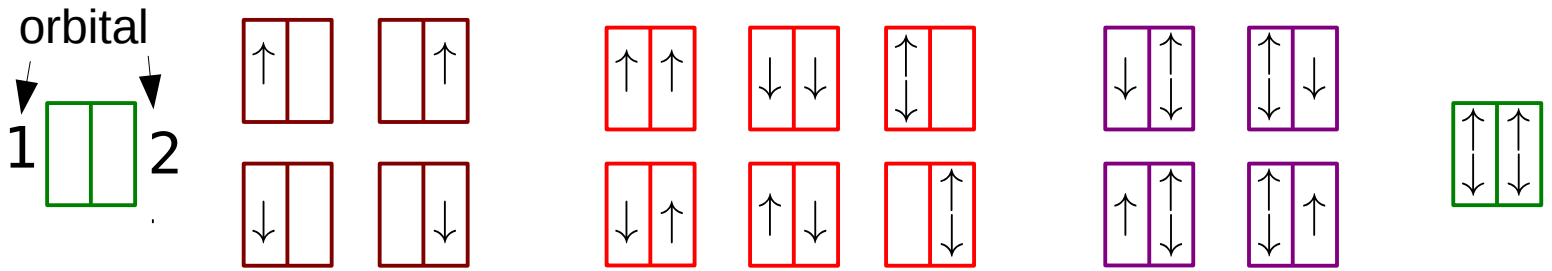
$$= \sum_{\vec{k},\gamma} E_{\vec{k},\gamma} \hat{h}^\dagger_{\vec{k},\gamma} \hat{h}_{\vec{k},\gamma}$$

$\longrightarrow$ bands are i) mixed (q^γ_σ) and shifted ($\eta_{\sigma,\sigma'}$)

ii) renormalised (q^γ_σ)

2. Ferromagnetism in two-band models

16 local states:



local Hamiltonian:

$$\hat{H}_{\rm loc} = U \sum_{b} \hat{n}_{b,\uparrow}\hat{n}_{b,\downarrow} + U' \sum_{s,s'} \hat{n}_{1,s}\hat{n}_{2,s'}$$

$$-J \sum_{s} \hat{n}_{1,s}\hat{n}_{2,s}$$

$$+J \sum_{s} \hat{c}^{\dagger}_{1,\sigma}\hat{c}^{\dagger}_{2,-s}\hat{c}_{1,-s}\hat{c}_{2,s}$$

$$+J_C \left(\hat{c}^{\dagger}_{1,\uparrow}\hat{c}^{\dagger}_{1,\downarrow}\hat{c}_{2,\downarrow}\hat{c}_{2,\uparrow} + \text{h.c.} \right)$$

$e_{\rm g}$ -orbitals:

$$J = J_C \quad \text{and} \quad U - U' = 2J$$

2-particle states	energy	Sym.
$\lvert\uparrow,\uparrow\rangle$ $(\lvert\uparrow,\downarrow\rangle + \lvert\downarrow,\uparrow\rangle)/\sqrt{2}$ $\lvert\downarrow,\downarrow\rangle$	$U - 3J$	${}^3A_2$
$(\lvert\uparrow,\downarrow\rangle - \lvert\downarrow,\uparrow\rangle)/\sqrt{2}$ $(\lvert\uparrow\downarrow,0\rangle - \lvert 0,\downarrow\uparrow\rangle)/\sqrt{2}$	$U - J$	${}^1E_1$
$(\lvert\uparrow\downarrow,0\rangle + \lvert 0,\downarrow\uparrow\rangle)/\sqrt{2}$	$U + J$	${}^1A_1$

Known: ferromagnetism is hardly found in single-band models

→ requires pathological densities of states and/or very large Coulomb parameters U

→ orbital degeneracy is an essential ingredient

therefore, we consider:

e_g-orbitals on a simple cubic 3-dimensional lattice

with hopping to nearest and next-nearest neighbours

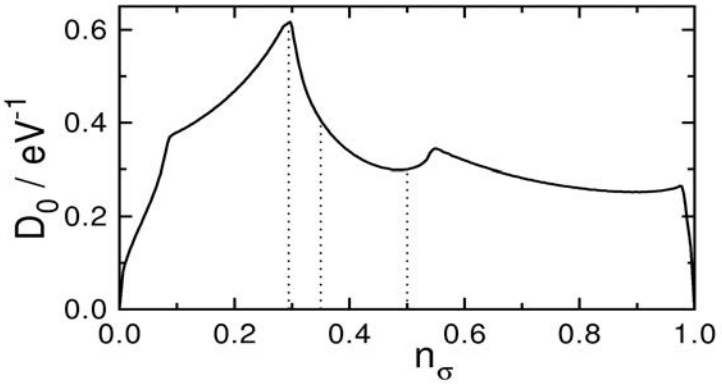
Gutzwiller wave function:

i) no multiplet coupling

$\lambda_{\Gamma,\Gamma'} \sim \delta_{\Gamma,\Gamma'}$

ii) $|\Psi_0\rangle$: spin-polarised Fermi sea

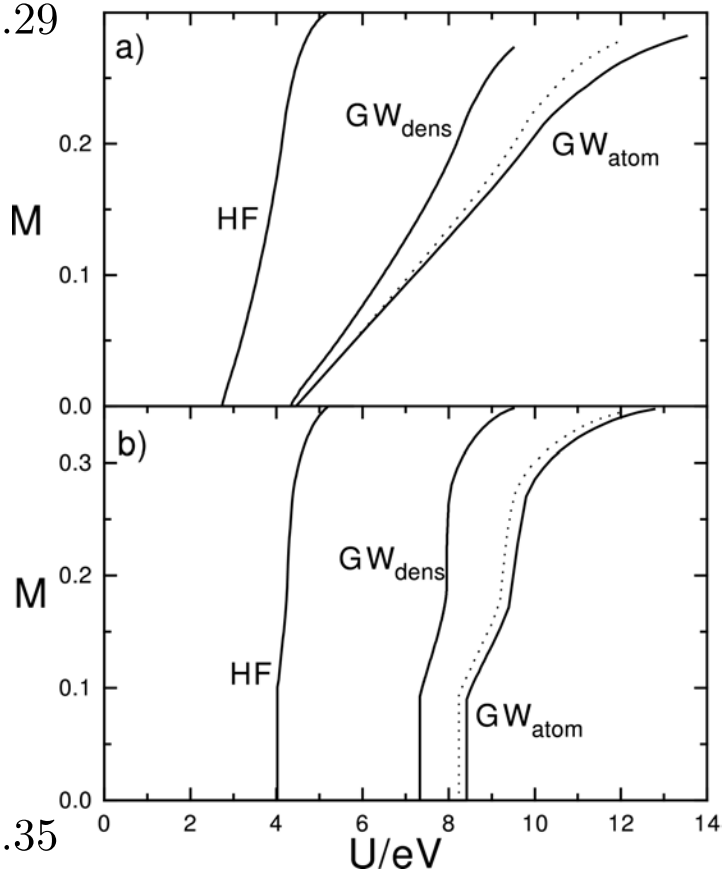
density of states:



Results:

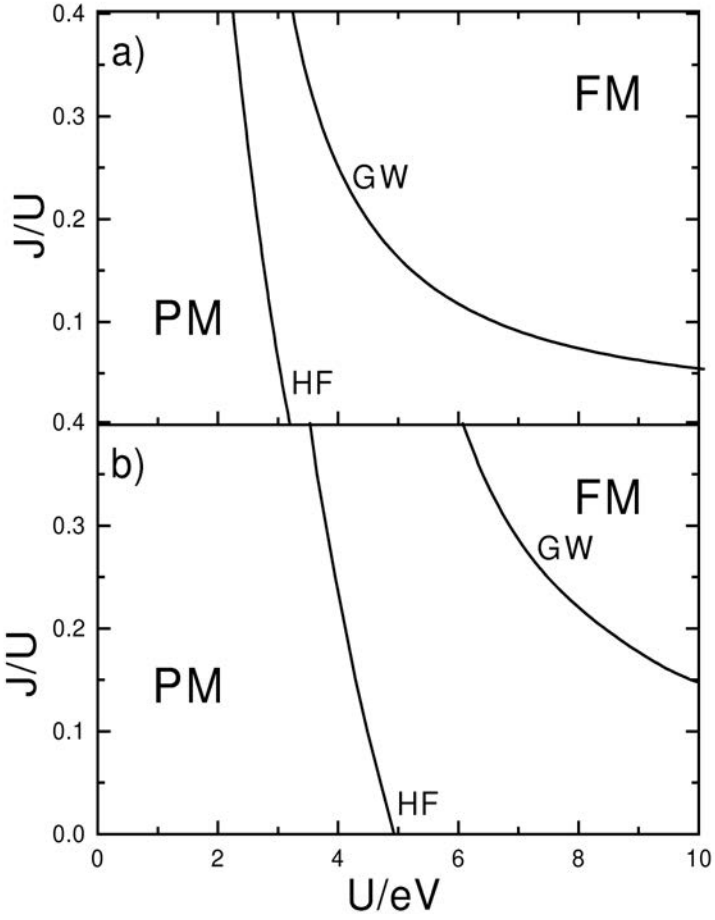
Magnetisation ($J = 0.2U$)

$n_\sigma = 0.29$

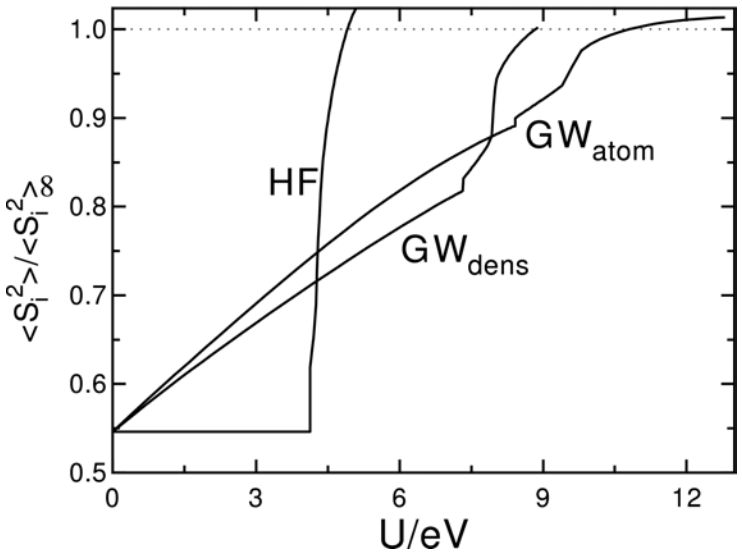


$n_\sigma = 0.35$

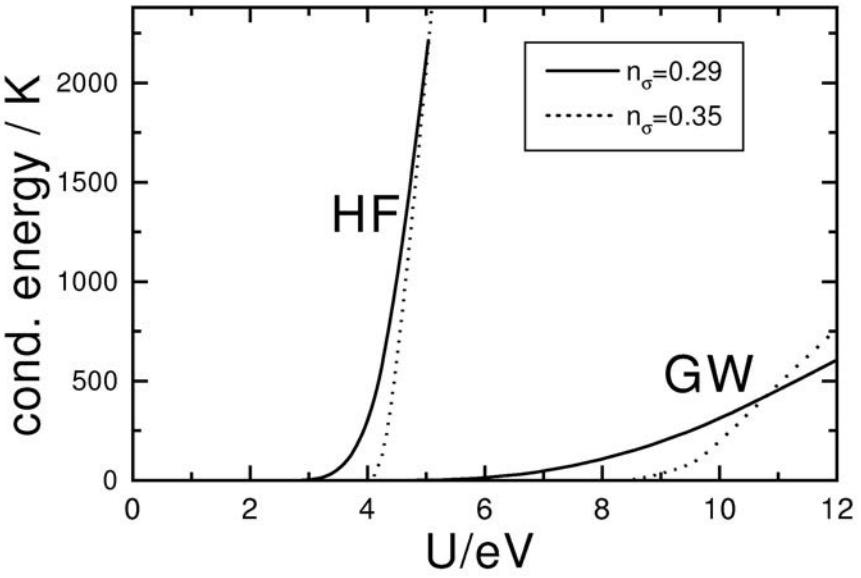
phase diagram



size of the local spin



condensation energy

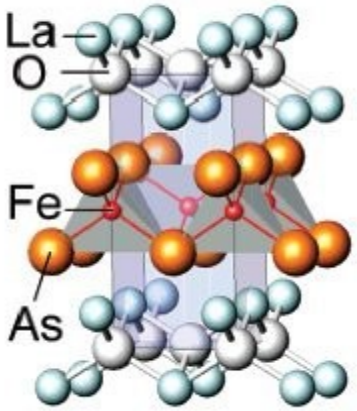


i) Orbital degeneracy and exchange interaction are essential for ferromagnetic order

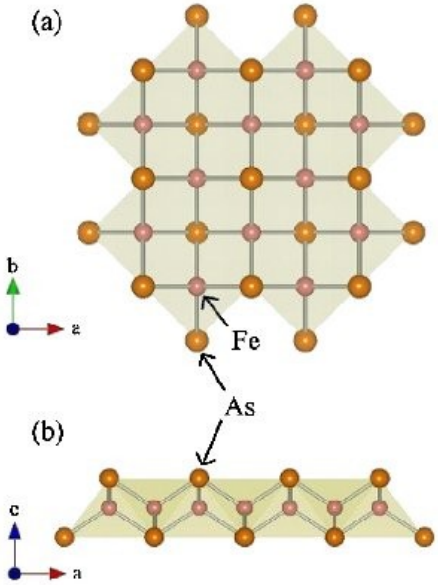
ii) Single-particle approaches are insufficient

3. Magnetic order in LaFeAsO

3.1 Electronic structure of LaFeAsO



(Y. Kamihara et al., J. Am. Chem. Soc 130, 3296 (2008))



(K. Ishida et al. J. Phys. Soc. Jpn. 78, 062001 (2009))

i) Metal with conductivity mainly in FeAs layers

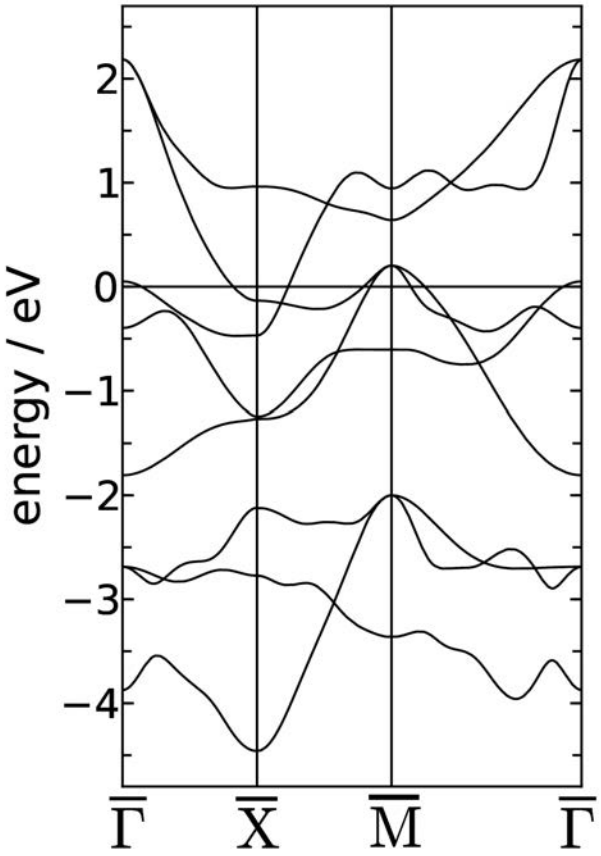
ii) AF ground state with a magnetic moment of $\approx 0.6\mu_{\mathrm{B}}/\mathrm{Fe}$

(DFT: $\approx 2.0\mu_{\mathrm{B}}/\mathrm{Fe}$)

3.2 LDA band-structure and effective tight-binding models

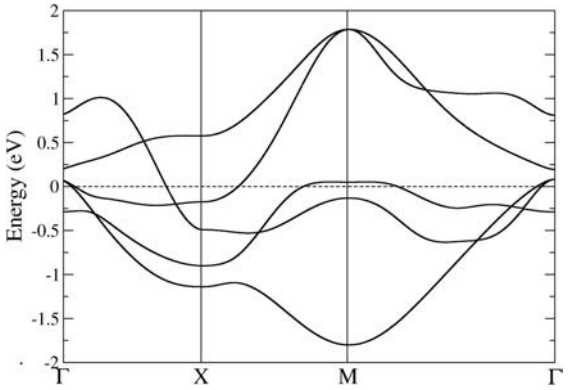
Eight-band model

(O. K Andersen and L. Boeri, Ann. Physik 523, 8 (2011))



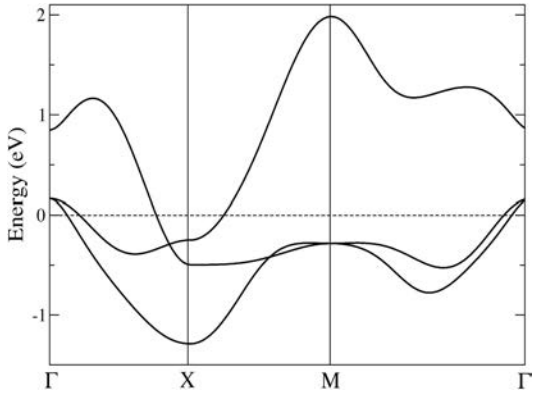
Five-band model:

(S. Graser et al., New J. Phys. 11, 025016 (2009))



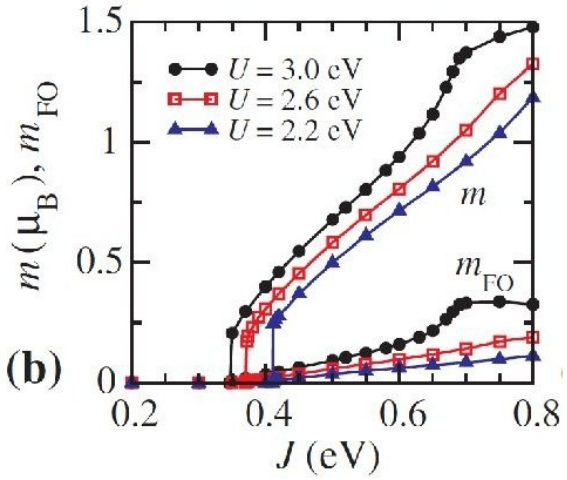
Three-band model:

(S. Zhou et al., PRL 105, 096401 (2010))



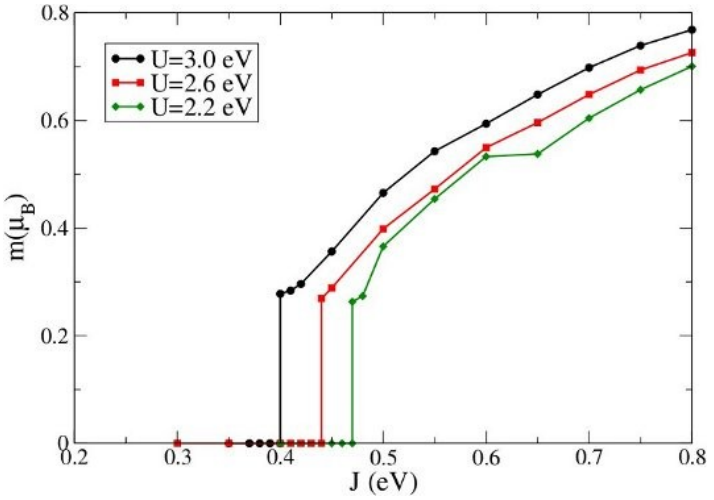
3.3 Magnetic order in three-band models

Without spin-flip terms:

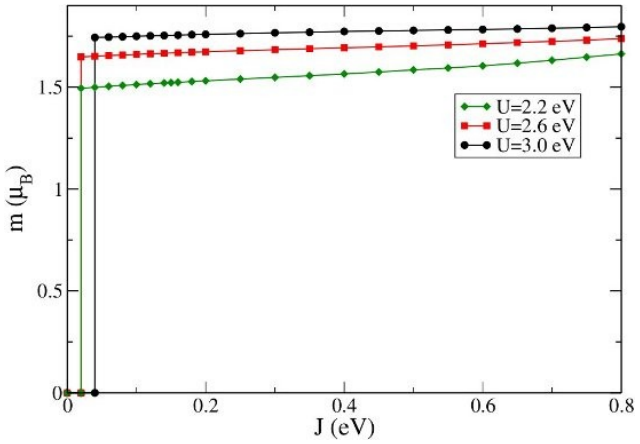


(S. Zhou et al., PRL 105, 096401 (2010))

With spin-flip terms:



Hartree-Fock:

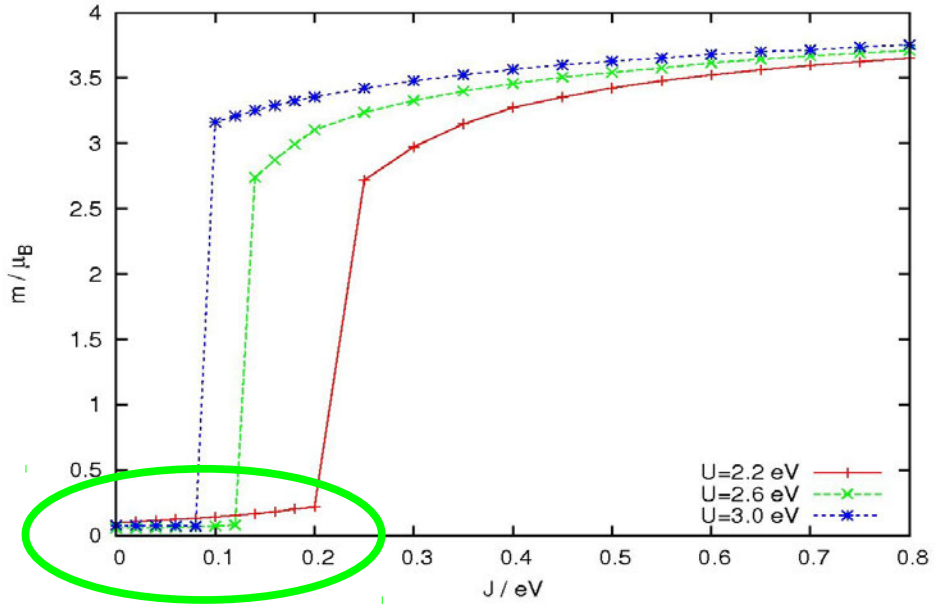


Conclusion (?):

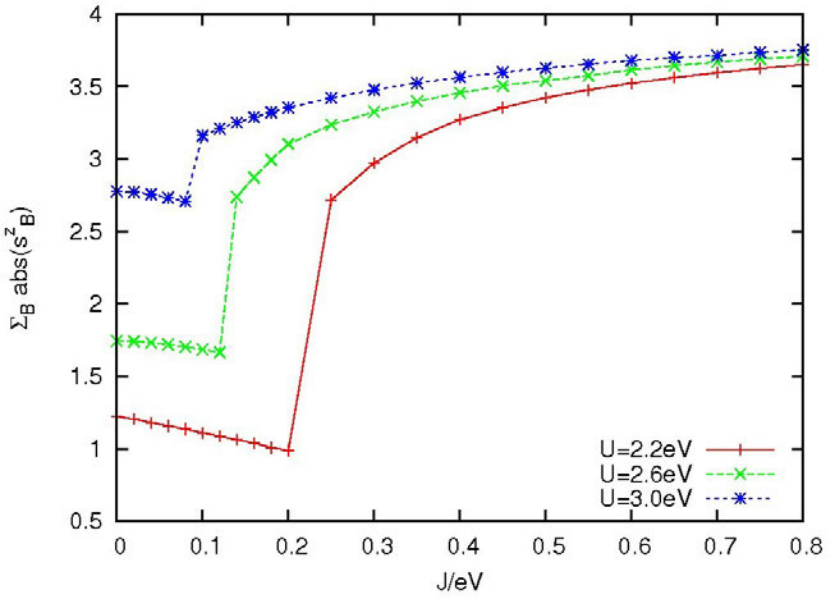
Gutzwiller theory yields a reasonable magnetic moment without fine-tuning of model parameters

3.4 Five-band model

A) Hartree-Fock:

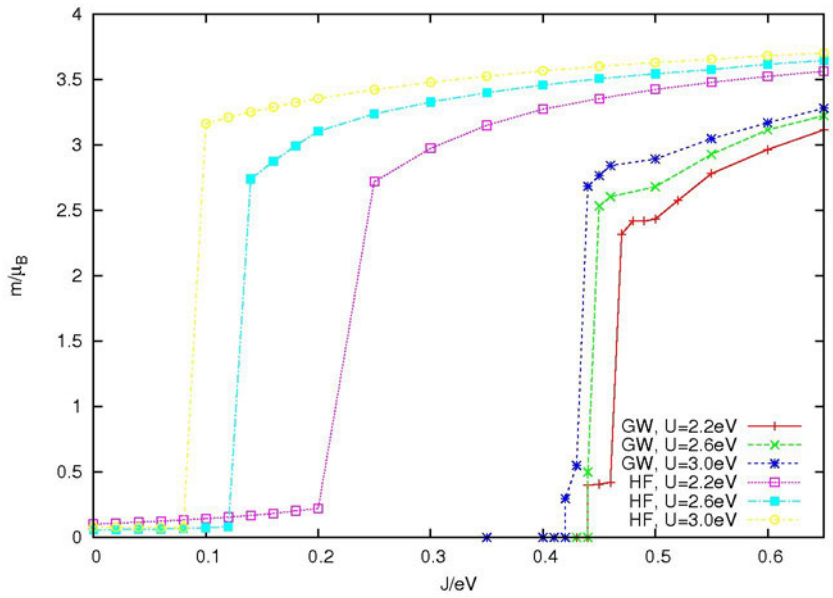


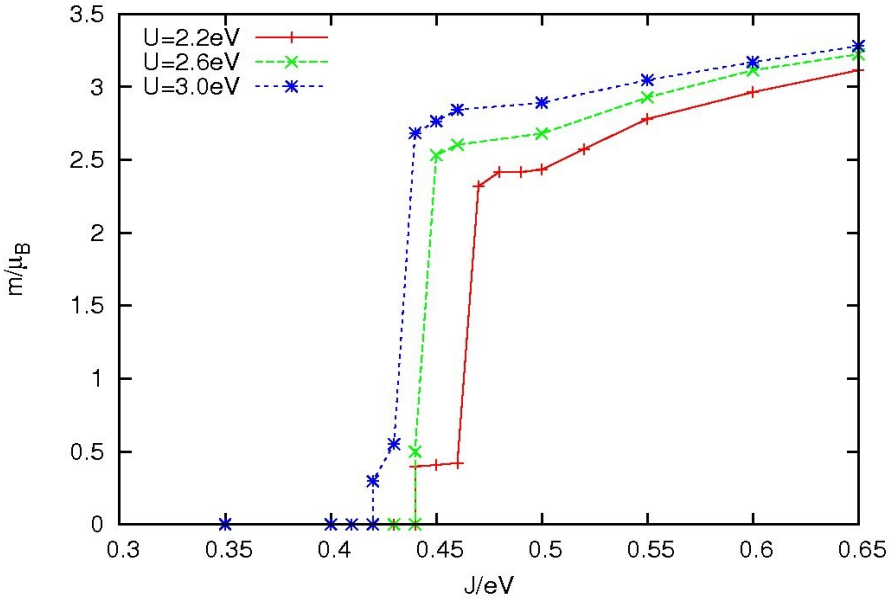
$$\sum_b |S_{z,b}| \neq 0$$



3.4 Five-band model

(T. Schickling et al., PRL 106, 156402 (2011))





Conclusions:

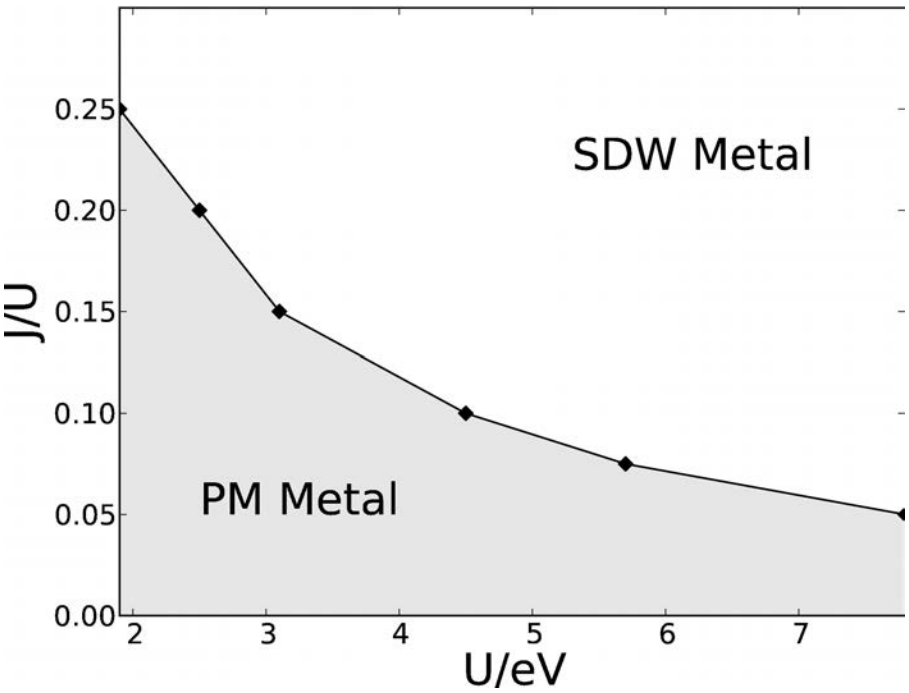
- no orbital order (in contrast to Hartree-Fock)
- small magnetic moments appear only in a small range of correlation parameters

→ still no satisfactory explanation for the magnetic order observed in LaFeAsO

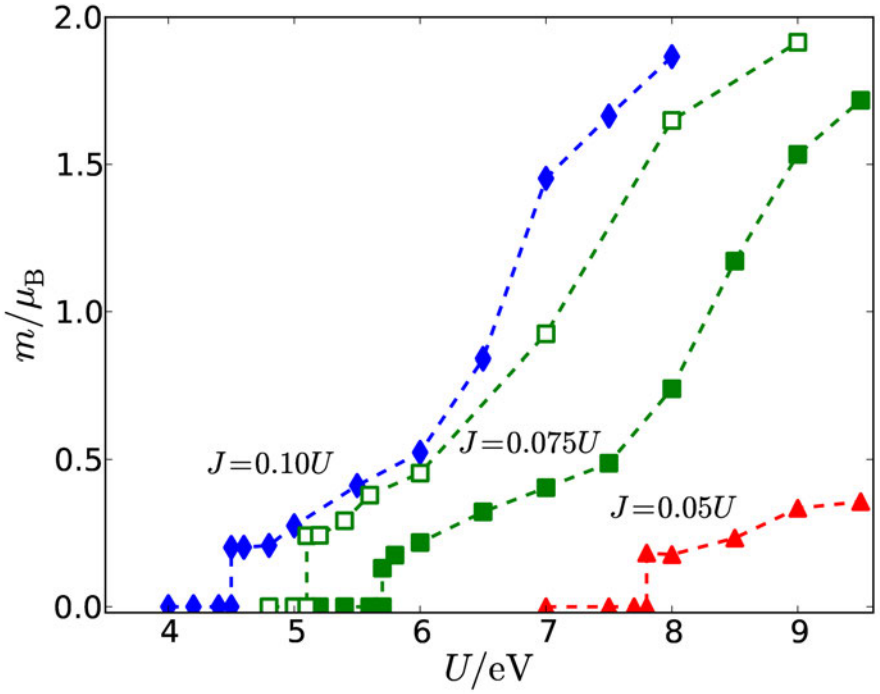
3.5 Electronic properties of LaAsFeO (eight-band model)

(T. Schickling et al., PRL 108, 036406 (2012))

Phase diagram:



Magnetic moment:



→ Reasonable values of the magnetic moment over a large range of Coulomb parameters

III) The Gutzwiller Density Functional Theory

1. Remainder: The Densitiy Functional Theory

Electronic Hamiltonian in solid-state physics

$$\hat{H}_{\mathrm{el}} = \sum_s \int \mathrm{d}^3 r \; \hat{\psi}_s^\dagger(\mathbf{r}) \left(-\frac{\Delta_{\mathbf{r}}}{2m} + V(\mathbf{r}) \right) \hat{\psi}_s(\mathbf{r})$$

$$+ \frac{1}{2} \sum_{s,s'} \int \mathrm{d}^3 r \int \mathrm{d}^3 r' \; \hat{\psi}_s^\dagger(\mathbf{r}) \hat{\psi}_{s'}^\dagger(\mathbf{r}') \frac{e^2}{|\mathbf{r} - \mathbf{r}'|} \hat{\psi}_{s'}(\mathbf{r}') \hat{\psi}_s(\mathbf{r})$$

Hohenberg-Kohn theorem:

Existence of a universal functional $W[n(\mathbf{r})]$ of the density $n(\mathbf{r})$ such that

$$E[n(\mathbf{r})] = \int \mathrm{d}^3 r V(\mathbf{r}) n(\mathbf{r}) + W[n(\mathbf{r})]$$

has its minimum at the exact ground-state density $n_0(\mathbf{r})$ of $\hat{H}_{\mathrm{el}}$

('universal' = independent of $V(\mathbf{r})$)

One usually writes

$$W[n(\mathbf{r})] = T[n(\mathbf{r})] + \frac{e^2}{2}\int \mathrm{d}^3 r \int \mathrm{d}^3 r' \, \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + E_{\mathrm{xc}}[n(\mathbf{r})]$$

with $T[n(\mathbf{r})]$: 'kinetic energy functional'

$E_{\mathrm{xc}}[n(\mathbf{r})]$: 'exchange correlation functional'

common approximations:

$$T[n(\mathbf{r})] = \frac{3}{10m}(3\pi^2)^{2/3}\int \mathrm{d}^3 r \, n(\mathbf{r})^{5/3}.$$

free electron gas

$$E_{\mathrm{xc}}[n(\mathbf{r})] = -\int \mathrm{d}^3 r \frac{3e^2}{4\pi}(3\pi)^{1/3} n(\mathbf{r})^{4/3}$$

HF approximation for free electron gas

Kohn-Sham scheme:

Instead of $\hat{H}_{\rm el}$, consider the effective single-particle Hamiltonian

$$\hat{H}_0^{\rm eff} = \sum_s \int \mathrm{d}^3 r \hat{\psi}_s^\dagger(\mathbf{r}) \left[-\frac{\Delta_{\mathbf{r}}}{2m} + V(\mathbf{r}) \right] \hat{\psi}_s(\mathbf{r})$$

$$+ \sum_s \int \mathrm{d}^3 r \hat{\psi}_s^\dagger(\mathbf{r}) \left[e^2 \int \mathrm{d}^3 r' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + V_{\rm xc}^{\rm KS}\{n(\mathbf{r})\} \right] \hat{\psi}_s(\mathbf{r})$$

with the 'Kohn-Sham potential'

Kinetic energy of non-interacting particles

$$V_{\rm xc}^{\rm KS}\{n(\mathbf{r})\} = \frac{\partial}{\partial \tilde{n}(\mathbf{r})} \Big(T\{\tilde{n}(\mathbf{r})\} - T'\{\tilde{n}(\mathbf{r})\} + E_{\rm xc}\{\tilde{n}(\mathbf{r})\} \Big) \Big|_{\tilde{n}(\mathbf{r})=n(\mathbf{r})}$$

→ $\hat{H}_{\rm el}$ and $\hat{H}_0^{\rm eff}$ have the same ground-state density

We introduce a basis of local orbitals $\phi_{i,\sigma}(\mathbf{r})$

Kohn-Sham equations:

$$\hat{H}_0^{\text{eff}}|\Psi_0\rangle = E_0|\Psi_0\rangle$$

$$\hat{H}_0^{\text{eff}} = \sum_{i,j}\sum_{\sigma,\sigma'} t_{i,j}^{\sigma,\sigma'}[n(\mathbf{r})]\,\hat{c}_{i,\sigma}^{\dagger}\hat{c}_{j,\sigma'}$$

$$t_{i,j}^{\sigma,\sigma'}[n(\mathbf{r})] \equiv \int \mathrm{d}^3 r \phi_{i,\sigma}^*(\mathbf{r})\left(-\frac{\Delta_{\mathbf{r}}}{2m} + V(\mathbf{r}) + e^2\int \mathrm{d}^3 r' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + V_{\text{xc}}^{\text{KS}}[n(\mathbf{r})]\right)\phi_{j,\sigma}(\mathbf{r})$$

$$n(\mathbf{r}) = \sum_{i,j}\sum_{\sigma,\sigma'} \phi_{i,\sigma}^*(\mathbf{r})\phi_{j,\sigma'}(\mathbf{r})\langle \hat{c}_{i,\sigma}^{\dagger}\hat{c}_{j,\sigma'}\rangle_{\Psi_0}$$

2. The Gutzwiller Kohn-Sham equations

We distinguish 'localised' ($\sigma \in \ell$) and 'delocalised' orbitals ($\sigma \in \mathrm{d}$)

$$\hat{H}_{\mathrm{H}} = \hat{H}_0 + \sum_i \hat{H}_{i;\mathrm{c}}$$

$$\hat{H}_0 \equiv \sum_{i \neq j} \sum_{\sigma,\sigma'} t_{i,j}^{\sigma,\sigma'} \hat{c}_{i,\sigma}^{\dagger} \hat{c}_{j,\sigma'} + \sum_i \sum_{\sigma,\sigma' \in \mathrm{d}} \epsilon_i^{\sigma,\sigma'} \hat{c}_{i,\sigma}^{\dagger} \hat{c}_{i,\sigma'}$$

$$\hat{H}_{i;\mathrm{c}} \equiv \sum_{\sigma,\sigma' \in \ell} \epsilon_i^{\sigma,\sigma'} \hat{c}_{i,\sigma}^{\dagger} \hat{c}_{i,\sigma'} + \sum_{\sigma_1,\sigma_2,\sigma_3,\sigma_4 \in \ell} U_i^{\sigma_1,\sigma_2,\sigma_3,\sigma_4} \hat{c}_{i,\sigma_1}^{\dagger} \hat{c}_{i,\sigma_2}^{\dagger} \hat{c}_{i,\sigma_3} \hat{c}_{i,\sigma_4}$$

where $t_{i,j}^{\sigma,\sigma'} = t_{i,j}^{\sigma,\sigma'}[n(\mathbf{r})]$ and $\epsilon_i^{\sigma,\sigma'} = \epsilon_i^{\sigma,\sigma'}[n(\mathbf{r})]$

are now functionals of the density $n(\mathbf{r})$

Problem: Coulomb interaction is counted twice in the localised orbitals → 'double-counting problem'

Density in the Gutzwiller ground state

$$n(\mathbf{r}) = \sum_{i \neq j} \sum_{\sigma,\sigma',\gamma,\gamma'} \phi^*_{i,\gamma}(\mathbf{r}) \phi_{j,\gamma'}(\mathbf{r}) q^{\sigma}_{\gamma} \left(q^{\sigma'}_{\gamma'} \right)^* \rho_{(j\sigma'),(i\sigma)}$$

$$+ \sum_{i} \sum_{\sigma} |\phi_{i,\sigma}(\mathbf{r})|^2 \rho_{(i\sigma),(i\sigma)}$$

depends on λ_Γ and $\rho_{(i\sigma),(j\sigma')} \equiv \langle \hat{c}^\dagger_{j,\sigma'} \hat{c}_{i,\sigma} \rangle_{\Psi_0}$

$$\longrightarrow \quad t^{\sigma,\sigma'}_{i,j} = t^{\sigma,\sigma'}_{i,j}(\tilde{\rho}, \lambda_\Gamma)$$

Gutzwiller-DFT energy functional:

$$E^{\mathrm{GDFT}}(\tilde{\rho}, \lambda_\Gamma) = \sum_{\sigma,\sigma',\gamma,\gamma'} q^{\sigma}_{\gamma} \left(q^{\sigma'}_{\gamma'} \right)^* \sum_{i \neq j} t^{\gamma,\gamma'}_{i,j}(\tilde{\rho}, \lambda_\Gamma) \rho_{(j\sigma'),(i\sigma)}$$

$$+ \sum_{i,\sigma \in \mathrm{d}} \epsilon^{\sigma,\sigma}_i \rho_{(i\sigma),(i\sigma)} + L \sum_{\Gamma} E_\Gamma \lambda^2_\Gamma m^0_\Gamma$$

Minimisation

$$\frac{\partial}{\partial \rho_{(i\sigma),(j\sigma')}} E^{\mathrm{GDFT}}\left(\tilde{\rho}, \lambda_\Gamma\right) = 0 \quad , \quad \frac{\partial}{\partial \lambda_\Gamma} E^{\mathrm{GDFT}}\left(\tilde{\rho}, \lambda_\Gamma\right) = 0$$

leads to 'Gutzwiller Kohn-Sham equations' with

$$\hat{H}_0^{\mathrm{eff}} = \sum_{i \neq j} \sum_{\sigma,\sigma',\gamma,\gamma'} q_\gamma^\sigma \left(q_{\gamma'}^{\sigma'}\right)^* t_{i,j}^{\gamma,\gamma'}(\tilde{\rho}, \lambda_\Gamma) \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'} + \sum_{i,\sigma \in \ell} \eta_\sigma \hat{c}_{i,\sigma}^\dagger \hat{c}_{i,\sigma}$$

($q_\gamma^\sigma = \delta_{\sigma,\gamma}$ for $\sigma, \gamma \in \mathrm{d}$)

and

$$\eta_{\tilde{\sigma}} \equiv \frac{1}{L} \sum_{\sigma,\sigma',\gamma,\gamma'} \left[\frac{\partial}{\partial n_{\tilde{\sigma}}} q_\gamma^\sigma \left(q_{\gamma'}^{\sigma'}\right)^* \right] \sum_{i \neq j} t_{i,j}^{\gamma,\gamma'}(\tilde{\rho}, \lambda_\Gamma) \rho_{(j\sigma'),(i\sigma)}$$

$$+ \frac{\partial}{\partial n_{\tilde{\sigma}}} \sum_\Gamma E_\Gamma \lambda_\Gamma^2 m_\Gamma^0$$

Correlation-induced changes of $n(\mathbf{r})$:

i) $$n(\mathbf{r}) = \sum_{i \neq j} \sum_{\sigma,\sigma',\gamma,\gamma'} \phi^*_{i,\gamma}(\mathbf{r}) \phi_{j,\gamma'}(\mathbf{r}) q^{\sigma}_{\gamma} \left(q^{\sigma'}_{\gamma'} \right)^* \rho_{(j\sigma'),(i\sigma)}$$

$$+ \sum_{i} \sum_{\sigma} |\phi_{i,\sigma}(\mathbf{r})|^2 \rho_{(i\sigma),(i\sigma)}$$

differs from the DFT expression

ii) Correlated bands are shifted (via η_σ)

⟶ change of $\tilde{\rho}$ ⟶ change of $n(\mathbf{r})$

iii) Correlated bands are renormalised in $\hat{H}_0^{\text{eff}}$

⟶ change of $\tilde{\rho}$ and λ_Γ in $t_{i,j}^{\sigma,\sigma'}(\tilde{\rho}, \lambda_\Gamma)$

⟶ change of $n(\mathbf{r})$

Problems:

1. The local Coulomb interactions $U_i^{\sigma_1,\sigma_2,\sigma_3,\sigma_4}$ are usually considered as adjustable parameters

→ 'ab-initio' character is partially lost

2. Double-counting problem:

Coulomb interaction appears in $U_i^{\sigma_1,\sigma_2,\sigma_3,\sigma_4}$ and in $\epsilon_i^{\sigma,\sigma'}$

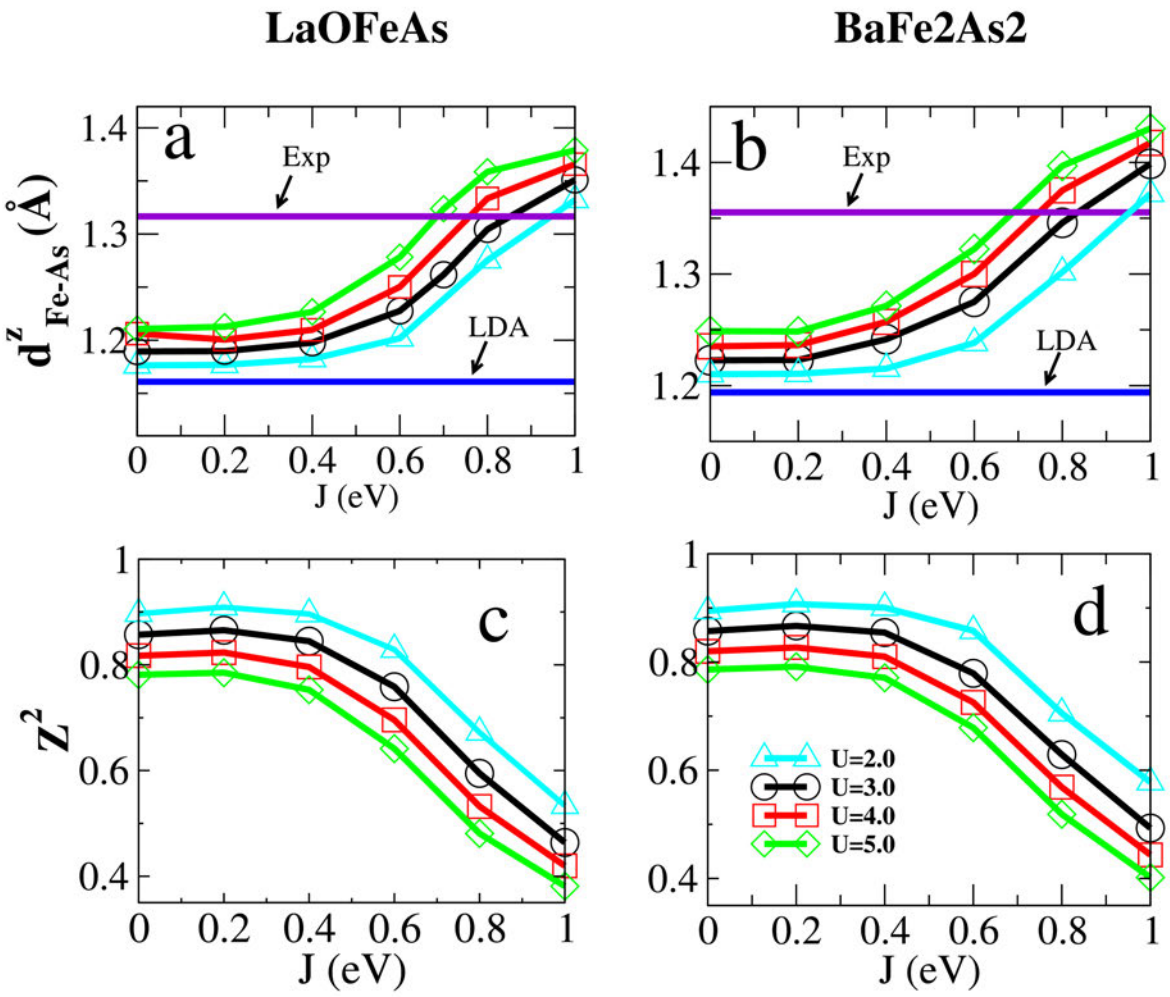
One possible solution: subtract

$$\hat{H}_{\rm dc} = 2 \sum_{\sigma,\sigma',\gamma\in\ell} (U_i^{\sigma,\gamma,\gamma,\sigma'} - U_i^{\gamma,\sigma,\gamma,\sigma'}) n_\gamma^0 \hat{c}_{i,\sigma}^\dagger \hat{c}_{i,\sigma'}$$

from $\hat{H}_{i;\rm c}$

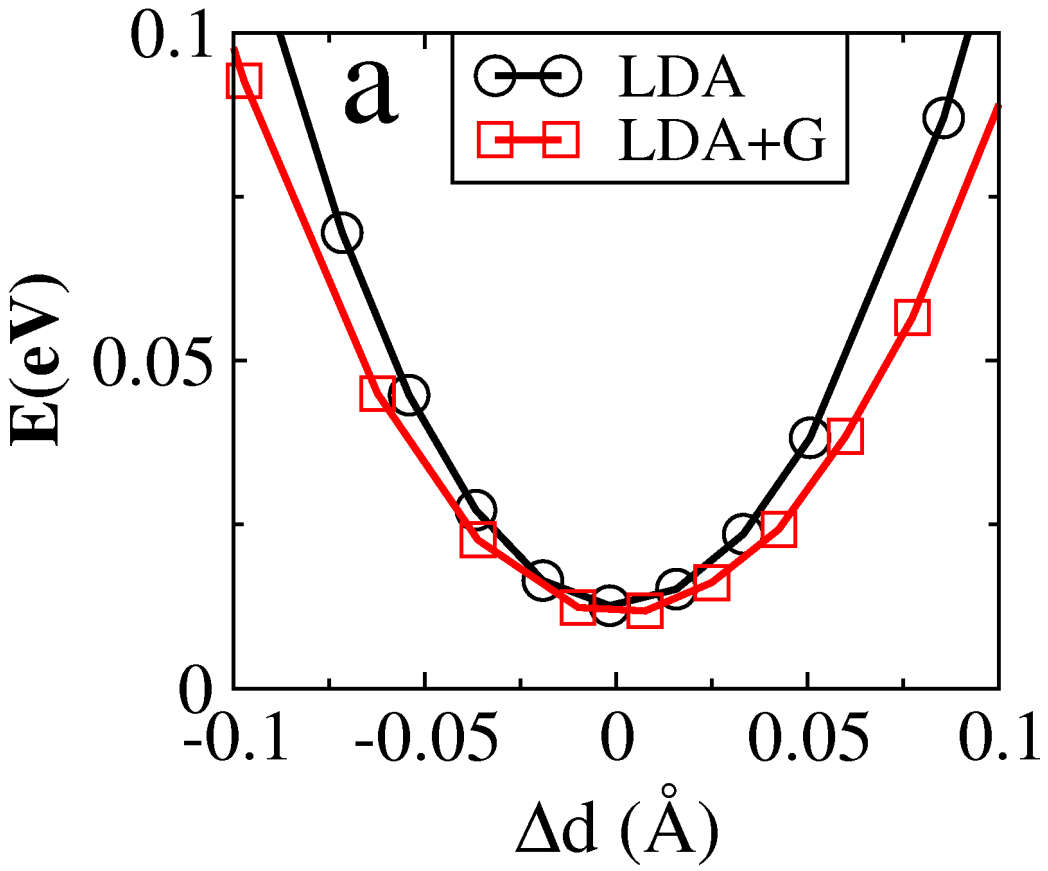
3. Example: Lattice parameters of iron pnictides

Interlayer distance $d^z_{\mathrm{Fe-As}}$ and (average) band renormalisation



from G. Wang et. al, Phys. Rev. Lett. 104, 047002 (2010)

Elastic constants:



$$E''_{\mathrm{LDA+G}} \approx 0.7 E''_{\mathrm{LDA}}$$

→ softening of the corresponding phonon mode

in agreement with experiment

IV) Further developments

1. Superconductivity: beyond the Gutzwiller approximation

1.1 Diagrammatic expansion

(J. Bünemann et al., EPL 98, 27006 (2012))

In the single-band case we need to calculate

i) $$\langle \Psi_{\mathrm{G}} | \Psi_{\mathrm{G}} \rangle = \left\langle \prod_l \hat{P}_l^2 \right\rangle_0 \qquad \left[\langle \ldots \rangle_0 \equiv \langle \Psi_0 | \ldots | \Psi_0 \rangle \right]$$

ii) $$\langle \Psi_{\mathrm{G}} | \hat{d}_i | \Psi_{\mathrm{G}} \rangle = \left\langle \hat{P}_i \hat{d}_i \hat{P}_i \prod_{l(\neq i)} \hat{P}_l^2 \right\rangle_0$$

iii) $$\langle \Psi_{\mathrm{G}} | \hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} | \Psi_{\mathrm{G}} \rangle = \left\langle (\hat{P}_i \hat{c}_{i\sigma}^{\dagger} \hat{P}_i)(\hat{P}_j \hat{c}_{j\sigma} \hat{P}_j) \prod_{l(\neq i,j)} \hat{P}_l^2 \right\rangle_0$$

Procedure:

A) Proper choice of the expansion parameter

B) Use of Wick's theorem and the linked-cluster theorem

→ Diagrammatic representation

C) Numerical evaluation in real space

A) Proper choice of the expansion parameter

Main idea: Choose parameters λ_Γ such that

$$\hat{P}_l^2 \stackrel{!}{=} 1 + x \cdot \hat{d}_l^{\mathrm{HF}} \quad \text{with}$$

$$\hat{d}_l^{\mathrm{HF}} \equiv (\hat{n}_{l\uparrow} - n_\uparrow^0)(\hat{n}_{l\downarrow} - n_\downarrow^0) = \hat{n}_{l\uparrow}^{\mathrm{HF}} \hat{n}_{l\downarrow}^{\mathrm{HF}}$$

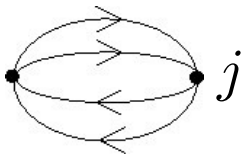
$$n_\sigma^0 \equiv \langle \hat{n}_{l\sigma}^0 \rangle_{\Psi_0} \quad , \quad \hat{n}_{l\sigma}^{\mathrm{HF}} \equiv \hat{n}_{l\sigma} - n_\sigma^0$$

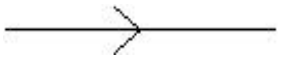
This fixes three parameters λ_Γ and it remains only one (λ_d or x)

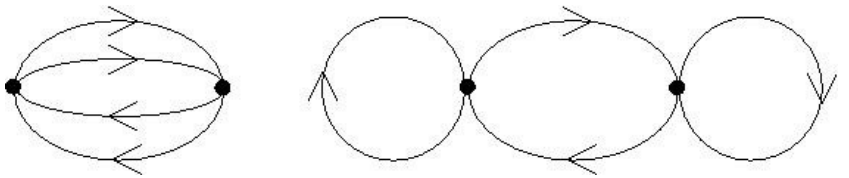
$$\lambda_d^2 = 1 + x(1 - n_\uparrow^0)(1 - n_\downarrow^0) \quad \longleftrightarrow \quad x = \frac{\lambda_d^2 - 1}{(1 - n_\uparrow^0)(1 - n_\downarrow^0)}$$

Main advantage of the HF-operators: no 'Hartree bubbles'

(Wick's theorem)

e.g.: $\langle \hat{d}_i^{\mathrm{HF}} \hat{d}_j^{\mathrm{HF}} \rangle_0 = |P_{i,j,\uparrow}^0|^2 |P_{i,j,\downarrow}^0|^2 =$  j

with $P_{i,j,\sigma} \equiv \langle \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} \rangle_{\Psi_0} =$ i  j

In contrast: $\langle \hat{d}_i \hat{d}_j \rangle_0 =$  $+ \cdots$

The missing of Hartree bubbles has two consequences:

i) Number of diagrams is significantly reduced, e.g.

ii) Each line is $\sim 1/D^{|i-j|/2}$ ($D =$ number of spatial dimensions)

→ Diagrams are fairly localised and the power series in x converges rapidly (can be tested for $D = 1$)

This suggests the following strategy:

i) Calculate

$$P_{i,j,\sigma} \equiv \langle \hat{c}^{\dagger}_{i\sigma} \hat{c}_{j\sigma} \rangle_{\Psi_0}$$

in momentum space (i.e., with negligible numerical error)

ii) Calculate all diagrams (power series in x) in real space up to a certain order in x

iii) Minimise the energy with respect to x

1.2 The one-dimensional Hubbard model

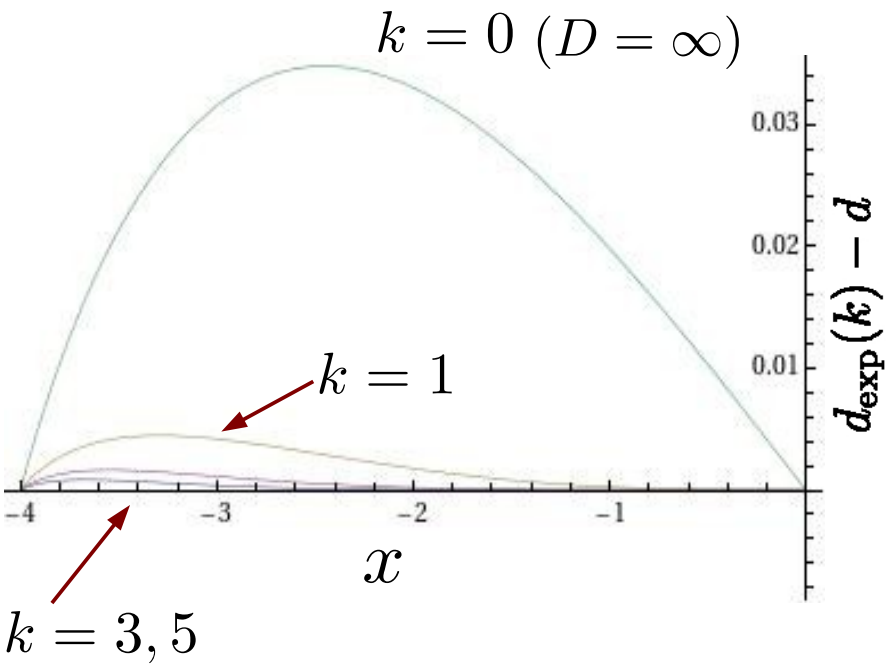
In one dimension one can evaluate Gutzwiller wave functions exactly [W. Metzner and D. Vollhardt, PRL 59, 121 (1987)]

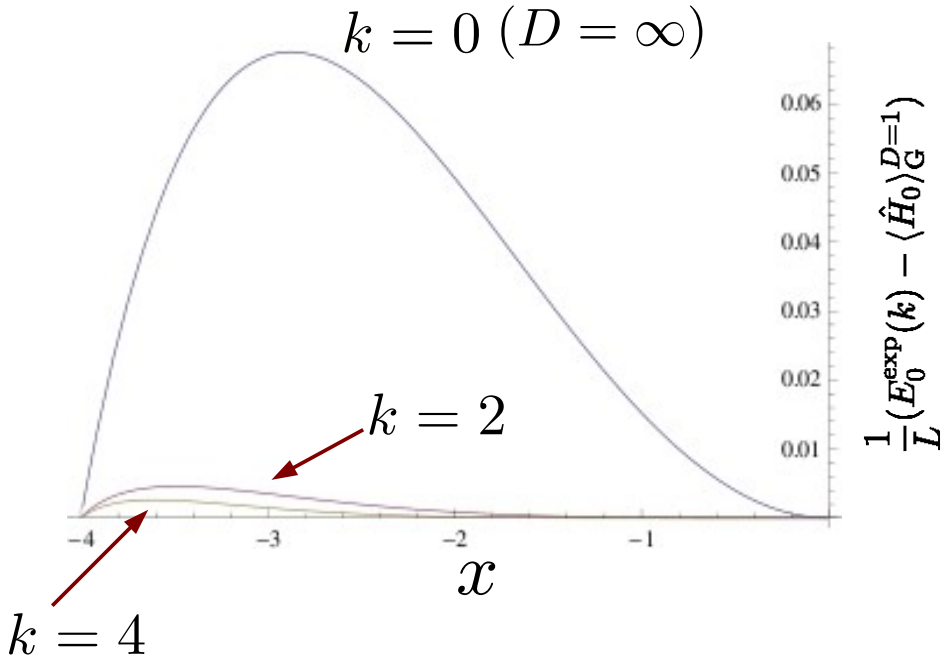
→ convergence of our approach can be tested

With the exact results we may calculate analytically each order of double occupancy $d_{\mathrm{exp}}(k)$ and the kinetic energy $E_0^{\mathrm{exp}}(k)$:

i) Double occupancy

ii) Kinetic energy





1.3 Fermi-surface deformations in two dimensions

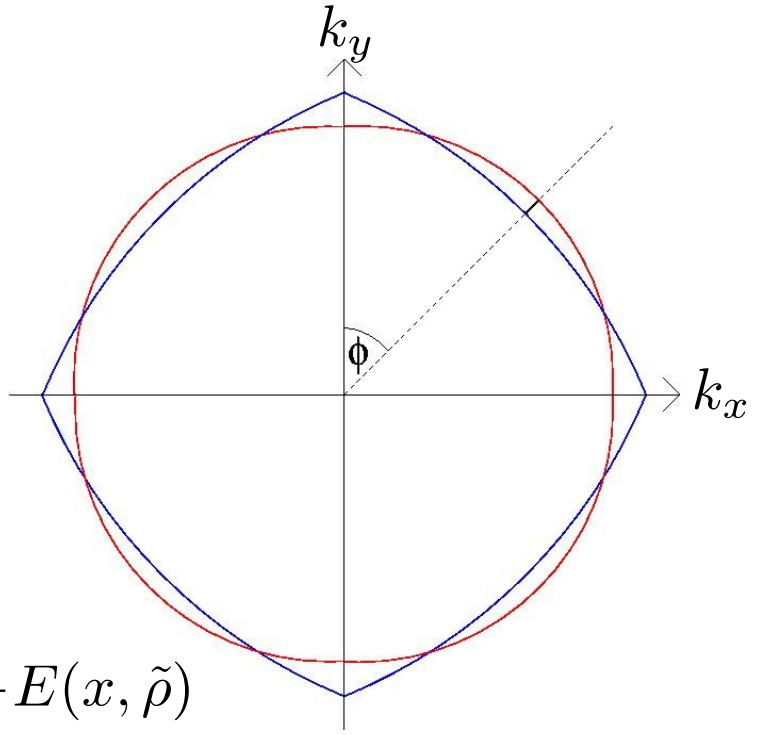
The Gutzwiller wave function

$$|\Psi_G\rangle = \hat{P}|\Psi_0\rangle \quad , \quad \hat{P}_G = \prod_i \hat{P}_i$$

contains as variational objects the parameters λ_Γ (i.e., x) and the wave function $|\Psi_0\rangle$. Without breaking translational or spin-Symmetry the only remaining degree of freedom in a one-band model is the shape of the Fermi surface:

Only constraint: $\frac{N}{L} = \sum_{\vec{k}} \langle \hat{n}_{\vec{k}} \rangle_0 = \text{const.}$

(particle number conservation)



Minimisation with respect to $\tilde{\rho}$:

$\hat{H}_0^{\text{eff}}|\Psi_0\rangle = E_{\text{SP}}|\Psi_0\rangle$ with

$$\hat{H}_0^{\text{eff}} = \sum_{i,j,\sigma} t_{i\sigma,j\sigma}^{\text{eff}} \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \quad , \quad t_{i\sigma,j\sigma}^{\text{eff}} \equiv \frac{\partial}{\partial \rho_{j\sigma,i\sigma}} E(x, \tilde{\rho})$$

Pomeranchuk instabilities

According to fRG calculations it may happen that the Fermi surface spontaneously breaks the rotational symmetry of the system at finite U ('Pomeranchuk instability').

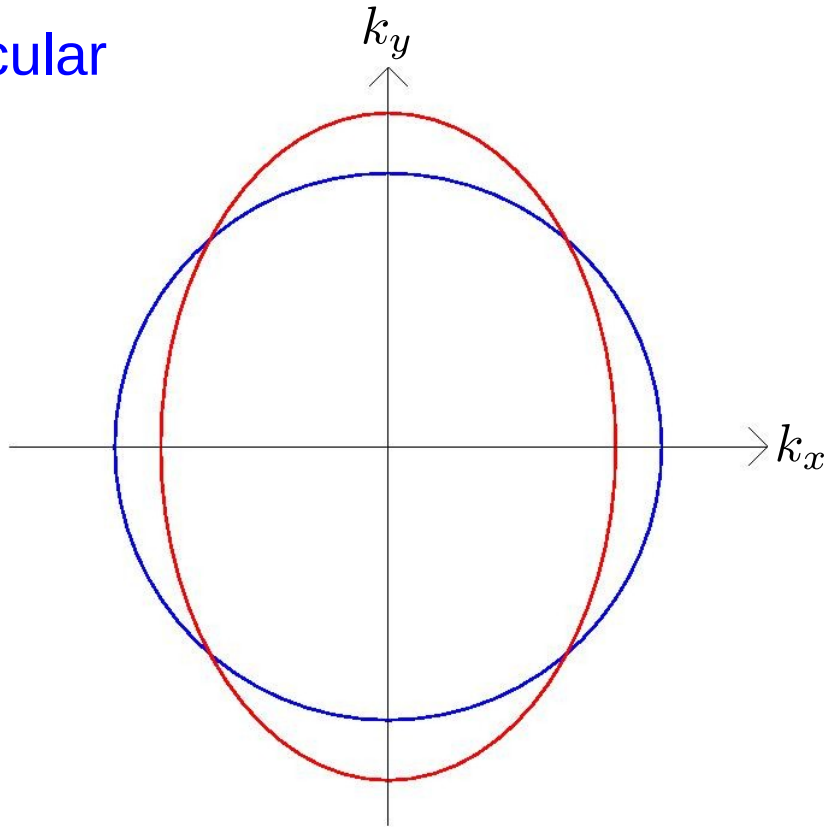
But:

i) fRG is a perturbative approach

ii) Little is know quantitatively, in particular for larger values of U.

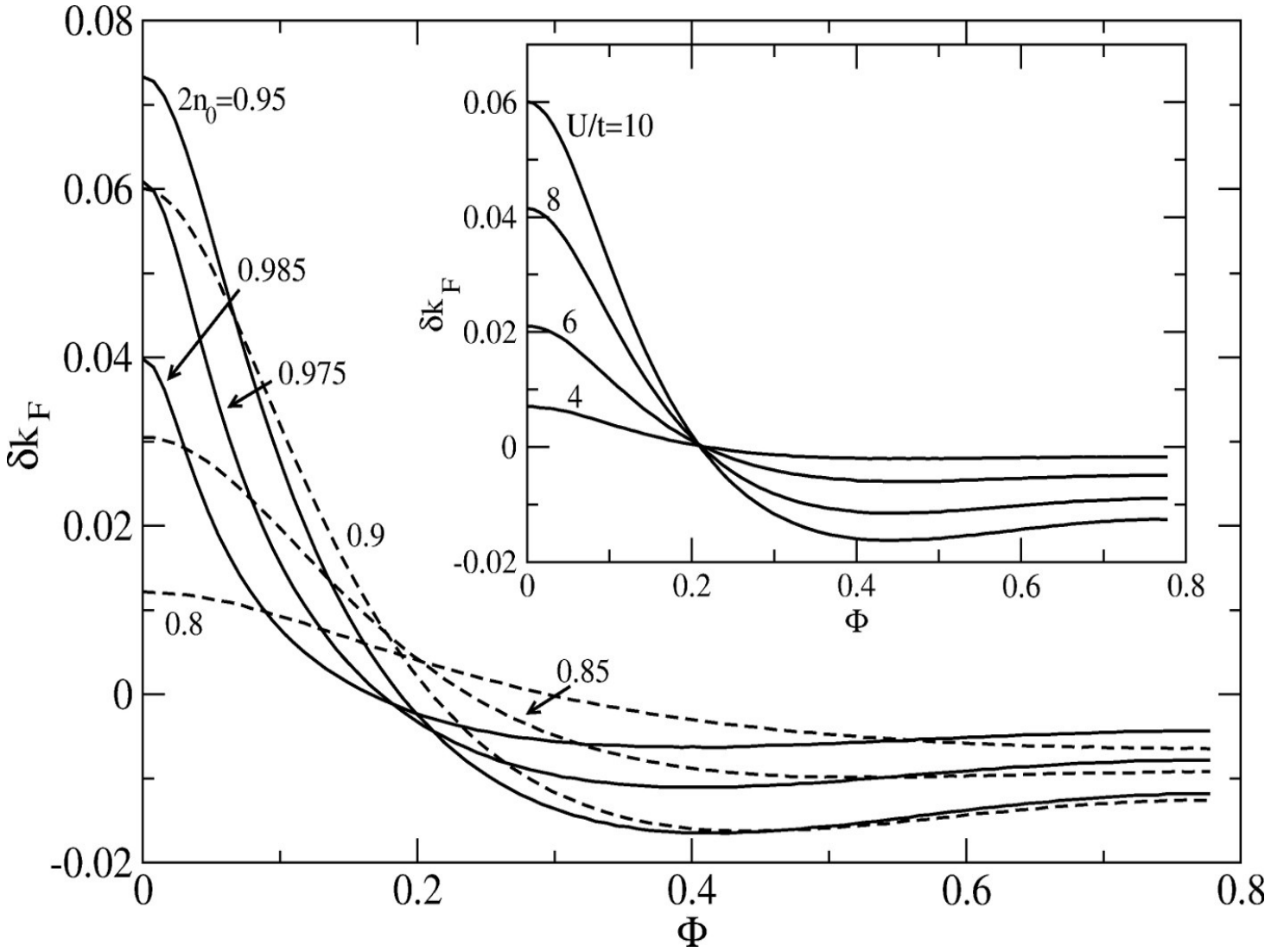
Obvious question:

Do we find a Pomeranchuk instability in our approach?



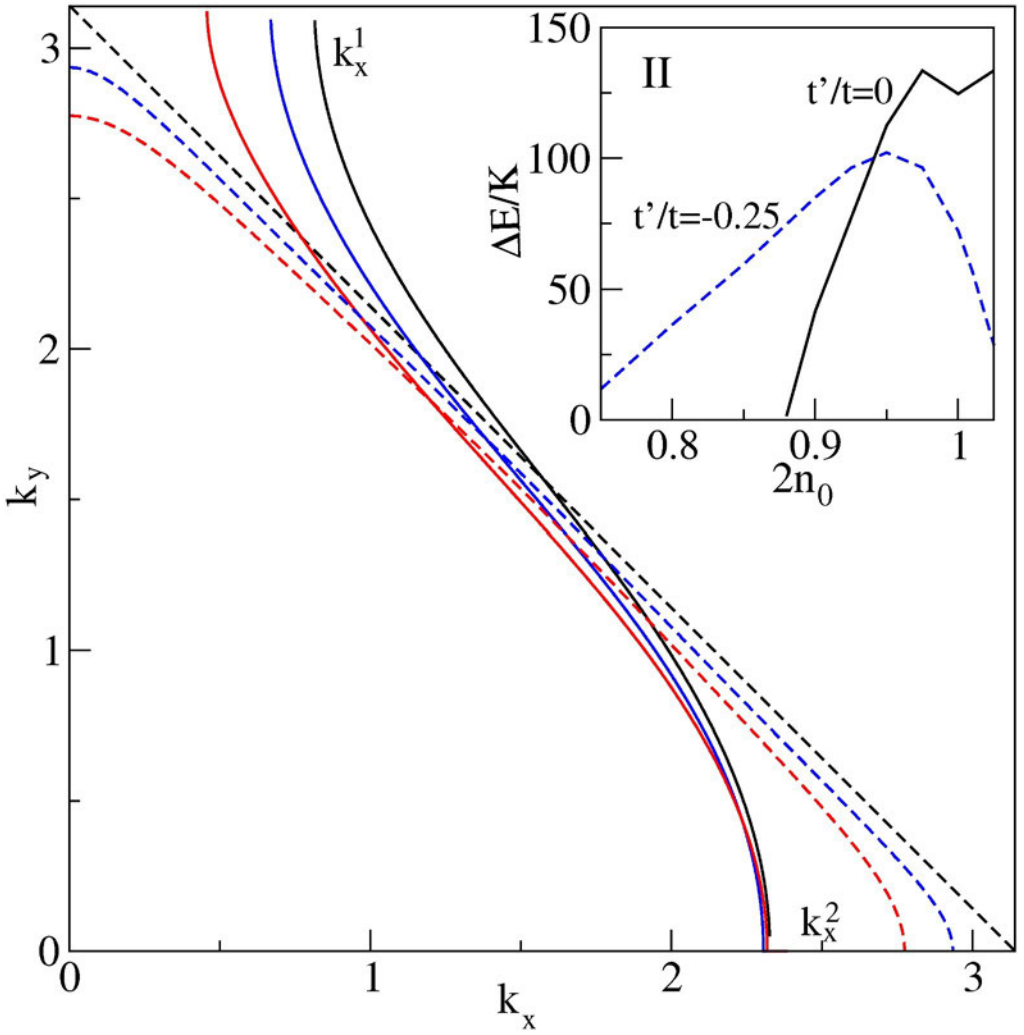
1.4 Hamiltonian with only nearest-neighbor hopping

'Normal' Fermi surface: $U/t = 10$



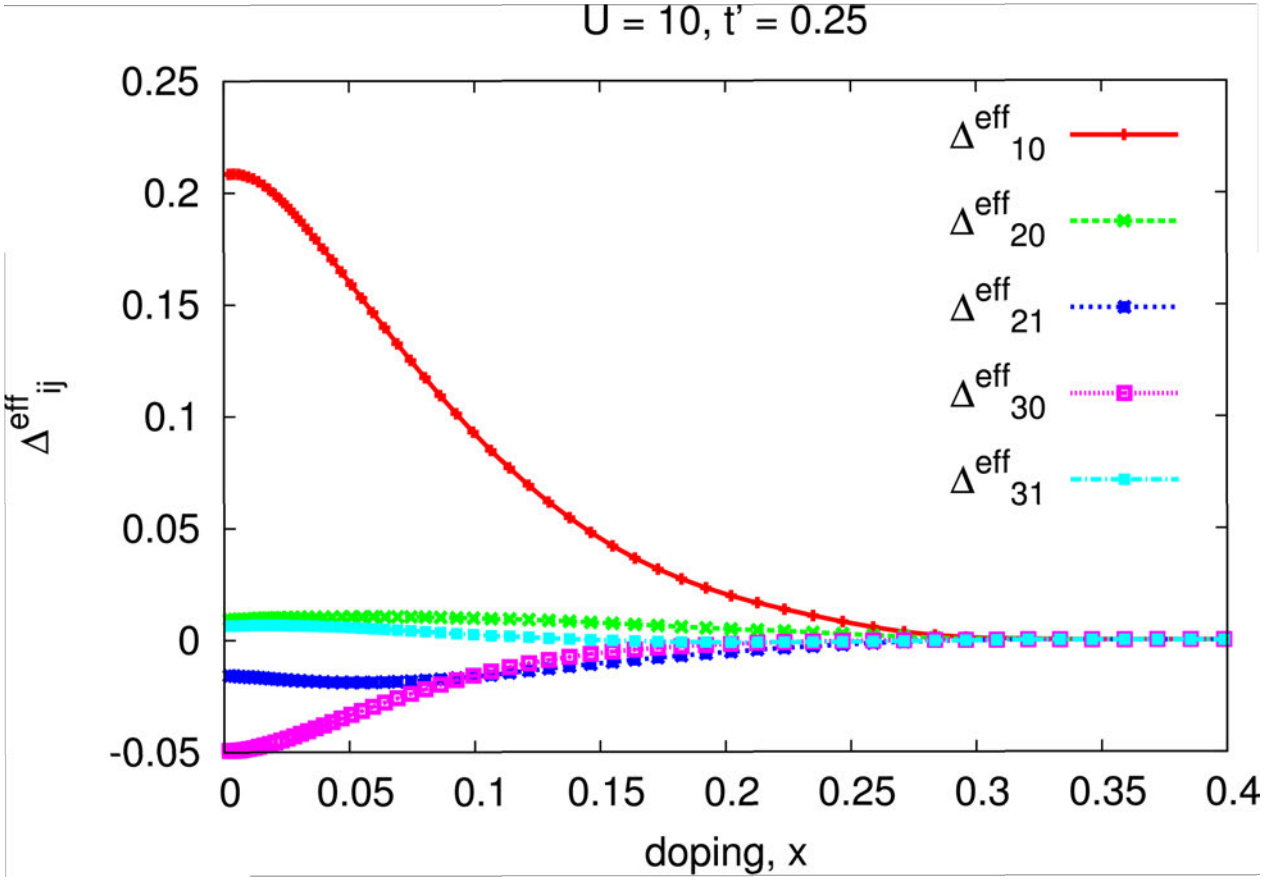
Pomeranchuk phase:

$U/t = 10$ $\qquad$ $2n_0 = 1.0, 0.95, 0.9$



1.5 Superconductivity in two-dimensional Hubbard models

$$\hat{H}_0^{\mathrm{eff}} = \sum_{i,j,\sigma} t_{i\sigma,j\sigma}^{\mathrm{eff}} \hat{c}_{i\sigma}^{\dagger} \hat{c}_{i\sigma} + \sum_{i,j} \left(\Delta_{i,j}^{\mathrm{eff}} \hat{c}_{i,\uparrow}^{\dagger} \hat{c}_{i,\downarrow}^{\dagger} + \mathrm{h.c.} \right)$$



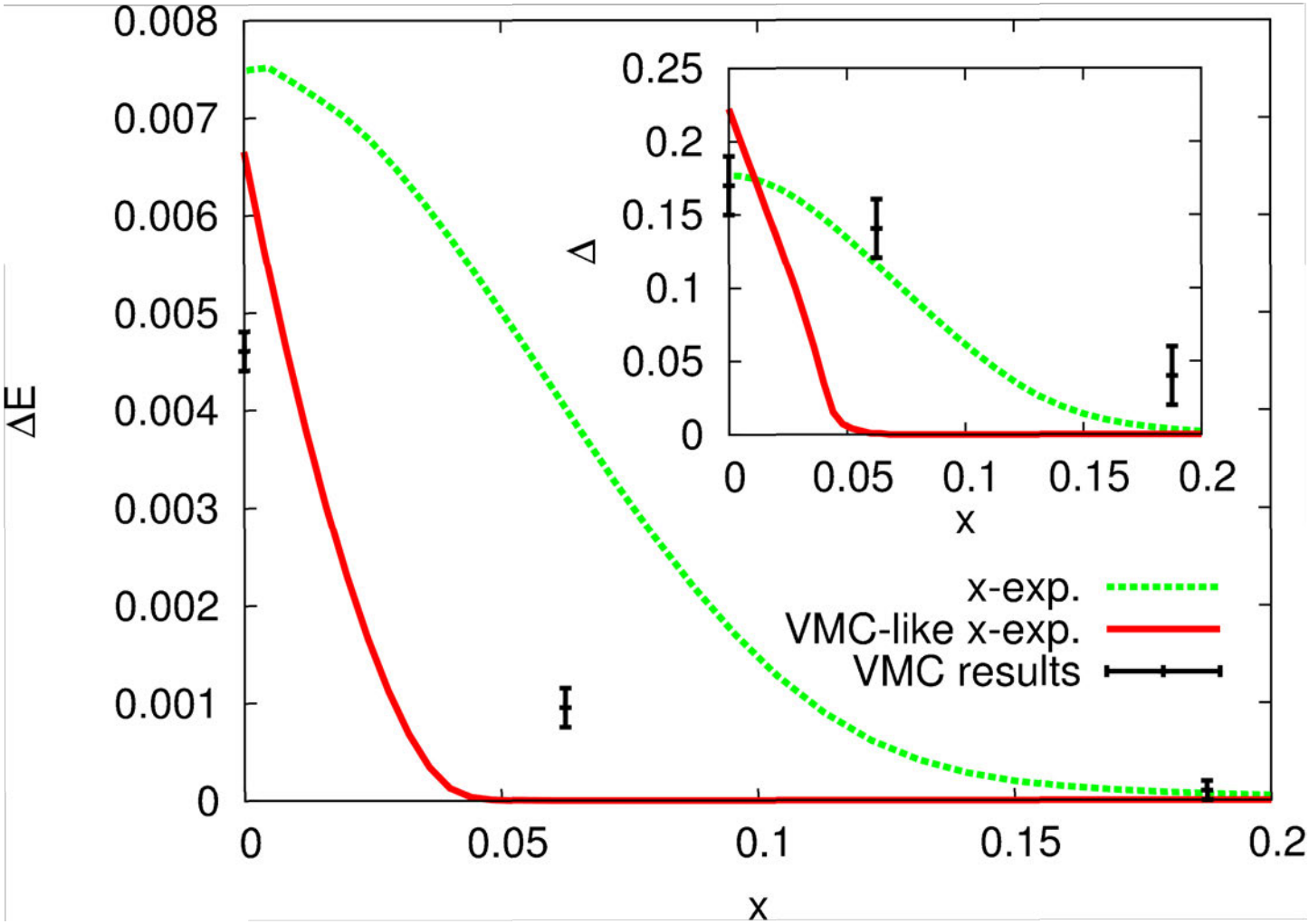
$(t = 1)$

→ Pairing beyond $\Delta_{1,0}^{\mathrm{eff}}$ is relevant

(collaboration with J. Kaczmarczyk, Krakow)

Consequence 1: enhancement of stability region

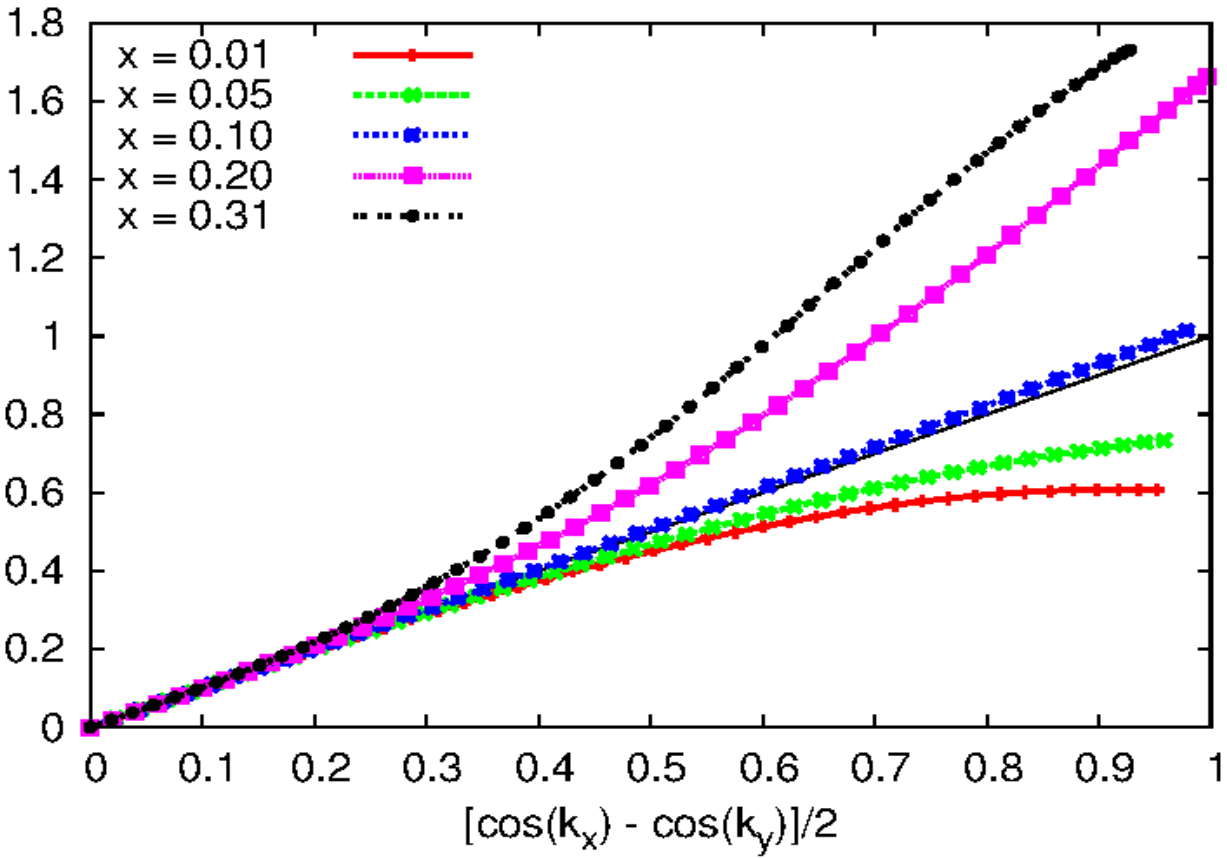
$$U = 10t, t' = 0$$



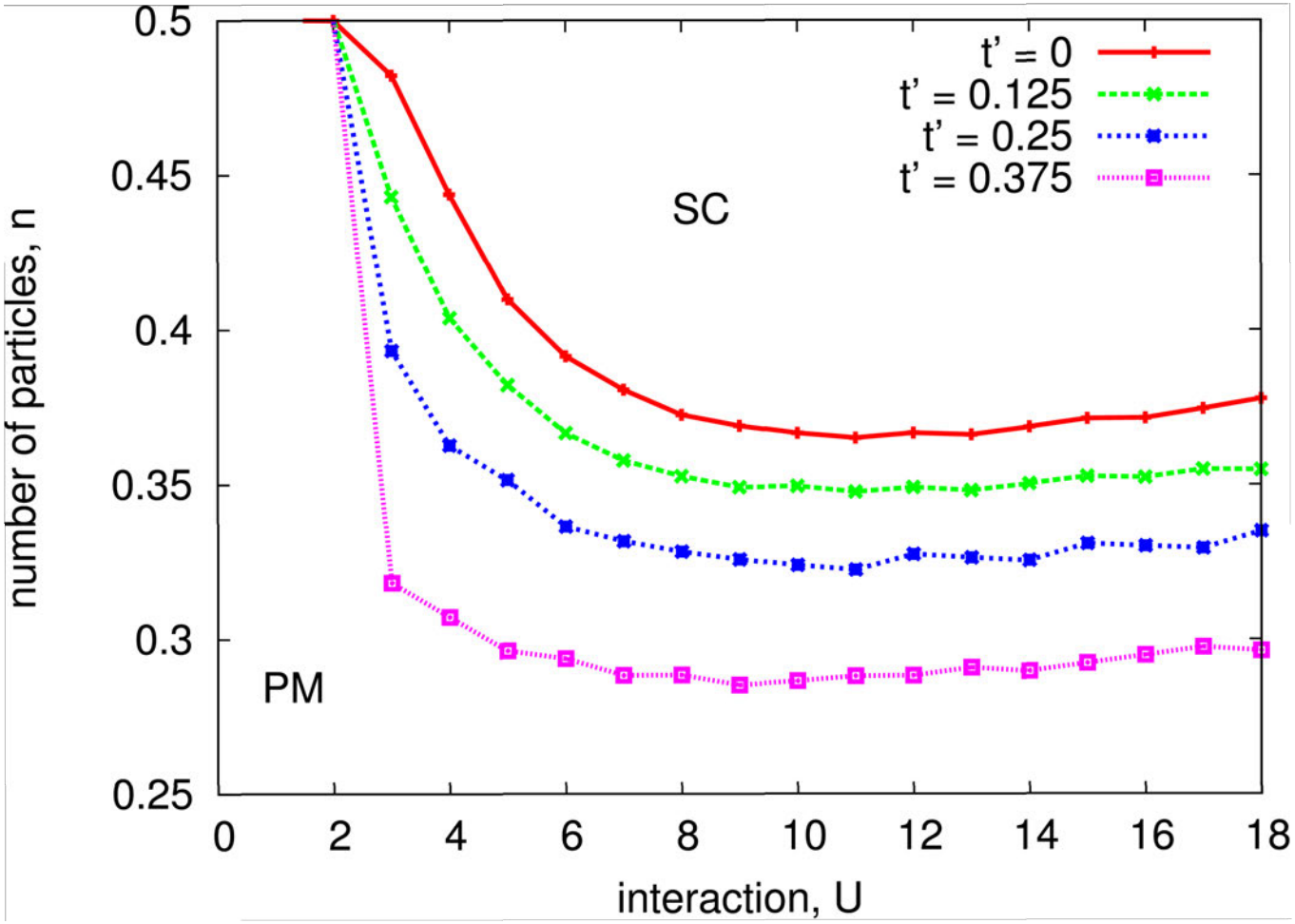
Consequence 2: gap-structure

$$U = 10t, t' = -0.25t$$

$$\frac{\Delta^{\mathrm{eff}}(k_x, k_y)}{\Delta^{\mathrm{eff}}_{1,0}(0, \pi)}$$



Phase diagram ($t = 1$)



2. The time-dependent Gutzwiller theory

2.1 Two-particle excitations: linear response theory

$$\hat{H} \to \hat{H}(t) = \hat{H} + \hat{V}(t) \text{ with } \hat{V}(t) = \sum_{\gamma,\gamma'} f_{\gamma,\gamma'}(t)\hat{c}^\dagger_\gamma \hat{c}_{\gamma'} \quad (f_{\gamma,\gamma'}(-\infty) = 0)$$

"Kubo formula":

$$\delta\underbrace{\langle \hat{c}^\dagger_{\gamma_1}\hat{c}_{\gamma_2}\rangle_t}_{\equiv \rho_{\gamma_1,\gamma_2}} \equiv \langle \hat{c}^\dagger_{\gamma_1}\hat{c}_{\gamma_2}\rangle_t - \langle \hat{c}^\dagger_{\gamma_1}\hat{c}_{\gamma_2}\rangle_{-\infty} = \sum_{\gamma_3,\gamma_4}\int dt' G_{(\gamma_1\gamma_2),(\gamma_3\gamma_4)}(t-t')f_{\gamma_3,\gamma_4}(t')$$

(Fourier transformation)

$$\delta\rho_{\gamma_1,\gamma_2}(\omega) = \sum_{\gamma_3,\gamma_4} G_{(\gamma_2,\gamma_1),(\gamma_3,\gamma_4)}(\omega) f_{\gamma_3,\gamma_4}(\omega)$$

Aim: calculate the density matrix $\tilde{\rho}$ with the elements

$$\rho_{\gamma,\gamma'}(t) = \langle\Psi(t)|\hat{c}^\dagger_\gamma \hat{c}_{\gamma'}|\Psi(t)\rangle \text{ with } \mathrm{i}\frac{\partial}{\partial t}|\Psi(t)\rangle = \hat{H}(t)\Psi(t)\rangle$$

→ equation of motion

$$-\mathrm{i}\dot{\rho}_{\gamma',\gamma}(t) = \langle\Psi(t)|[\hat{H}, \hat{c}^\dagger_\gamma \hat{c}_{\gamma'}]|\Psi(t)\rangle$$

2.2 Time-dependent Hartree-Fock Theory (RPA):

Equation of motion

$$-\mathrm{i}\dot{\rho}_{\gamma',\gamma}(t) = \langle\Psi(t)|[\hat{H}, \hat{c}^\dagger_\gamma \hat{c}_{\gamma'}]|\Psi(t)\rangle$$

Approximation: $|\Psi(t)\rangle$ is assumed to be a single-particle wave function

→ $\langle\Psi(t)|[\hat{H}, \hat{c}^\dagger_\gamma \hat{c}_{\gamma'}]|\Psi(t)\rangle = -[\tilde{h}^{\mathrm{HF}}, \tilde{\rho}(t)]_{\gamma,\gamma'}$ decouples (Wick's theorem)

with $h^{\mathrm{HF}}_{\gamma,\gamma'} = \dfrac{\partial}{\partial\rho_{\gamma',\gamma}} E^{\mathrm{HF}}[\tilde{\rho}(t)]$ and $E^{\mathrm{HF}}[\tilde{\rho}] = \langle\Psi_{\mathrm{HF}}|\hat{H}|\Psi_{\mathrm{HF}}\rangle$

→ $\mathrm{i}\dot{\tilde{\rho}}(t) = [\tilde{h}^{\mathrm{HF}}, \tilde{\rho}(t)]$ is a closed set of differential equations for $\tilde{\rho}(t)$

In linear order with respect to $f_{\sigma,\sigma'}$ this leads to the RPA result

$$\tilde{G}(\omega) = \tilde{G}^{\mathrm{HF}}(\omega)[1 + \tilde{U}\tilde{G}^{\mathrm{HF}}(\omega)]^{-1}$$

with

$$\hat{H}_{\mathrm{int}} = \sum_{\gamma_1,\gamma_2,\gamma_3,\gamma_4} U_{(\gamma_1,\gamma_2),(\gamma_3,\gamma_4)} \hat{c}^\dagger_{\gamma_1} \hat{c}_{\gamma_2} \hat{c}^\dagger_{\gamma_3} \hat{c}_{\gamma_4}$$

2.3 The time-dependent Gutzwiller theory

(G. Seibold und J. Lorenzana
PRL **86**, 2605 (2001))

Comparison:

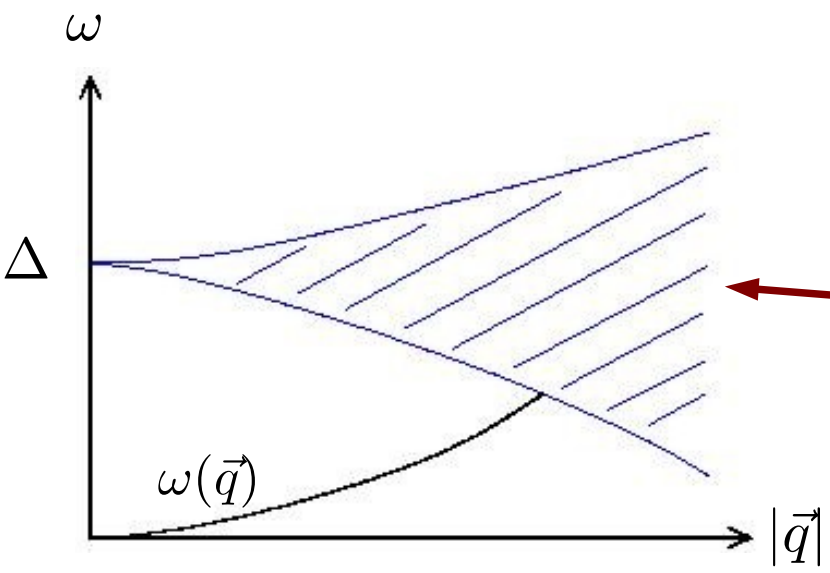
RPA	Time-dep. Gutzwiller theory
$\mathrm{i}\dot{\tilde{\rho}}(t) = [\tilde{h}^{\mathrm{HF}}, \tilde{\rho}(t)]$	$\mathrm{i}\dot{\tilde{\rho}}(t) = [\tilde{h}^{\mathrm{GW}}, \tilde{\rho}(t)]$
$h^{\mathrm{HF}}_{\gamma,\gamma'} = \frac{\partial}{\partial \rho_{\gamma',\gamma}} E^{\mathrm{HF}}[\tilde{\rho}(t)]$	$h^{\mathrm{GW}}_{\gamma,\gamma'} = \frac{\partial}{\partial \rho_{\gamma',\gamma}} E^{\mathrm{GW}}[\tilde{\rho}(t)]$
$E^{\mathrm{HF}}[\tilde{\rho}] = \langle \Psi_{\mathrm{HF}} \vert \hat{H} \vert \Psi_{\mathrm{HF}} \rangle$	$E^{\mathrm{GW}}[\tilde{\rho}] \equiv \min_{\lambda_{\Gamma,\Gamma'}} E(\lambda_{\Gamma,\Gamma'}, \tilde{\rho})$

2.4 RPA for the spin susceptibility of a single-band model

Stoner criterion: $UD(E_\mathrm{F}) > 1$ → ferromagnetism

→ finite exchange interaction $\Delta \equiv U(n_\uparrow - n_\downarrow)$

$$G_\mathrm{T}(\vec{q},\omega) = \frac{G_\mathrm{T}^\mathrm{HF}(\vec{q},\omega)}{1 + UG_\mathrm{T}^\mathrm{HF}(\vec{q},\omega)} \quad \text{with} \quad G_\mathrm{T}^\mathrm{HF}(\vec{q},\omega) = \frac{1}{L}\sum_{\vec{k}} \frac{\langle \hat{n}_{\vec{k}+\vec{q}\uparrow} - \hat{n}_{\vec{k}\downarrow}\rangle}{\omega - \Delta - (\varepsilon_{\vec{k}} - \varepsilon_{\vec{k}+\vec{q}}) + \mathrm{i}\delta}$$



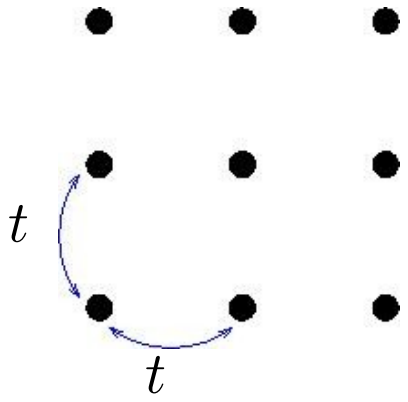
← "Stoner continuum"

$\widehat{=}$ Spin-flip-excitation in the "exchange field" Δ

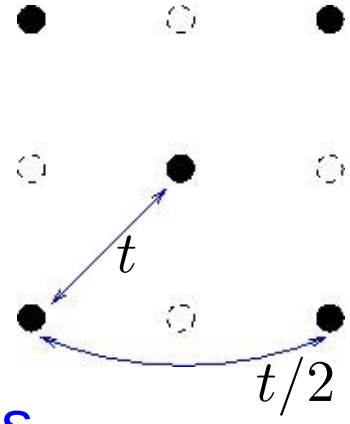
2.5 Spin excitations in single-band models

A) Hypercubic lattices in infinite dimensions $D \to \infty$

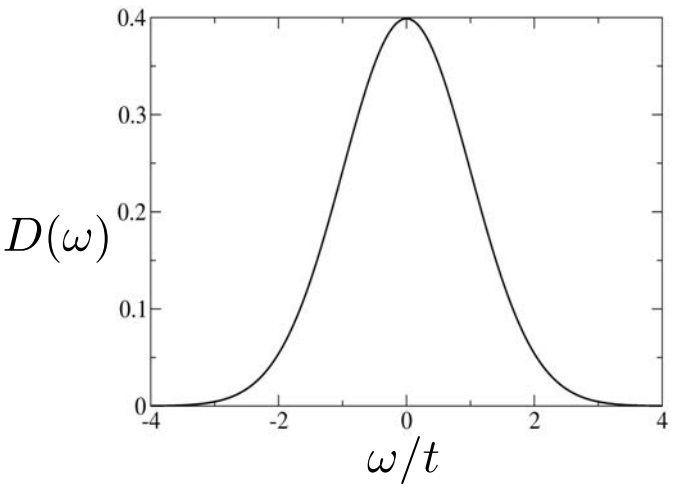
i) simple cubic (sc)

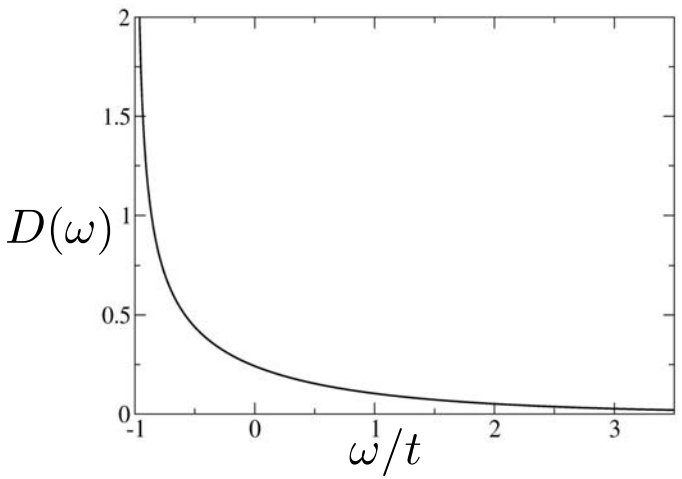


ii) half cubic (hc)



densities of states





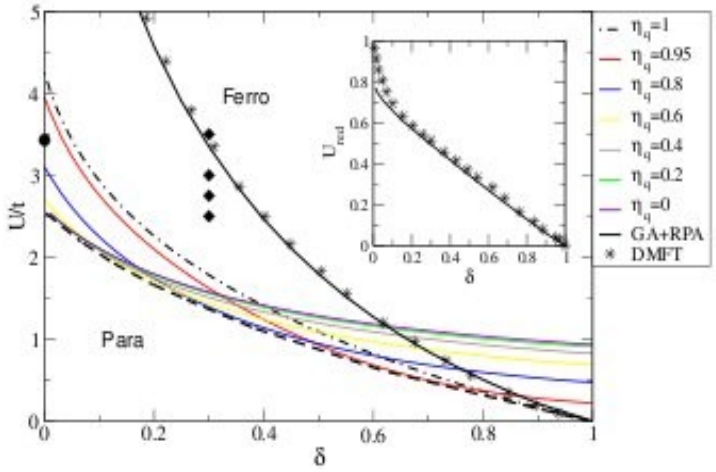
Two-particle response functions in large dimensions only depend on the real parameter

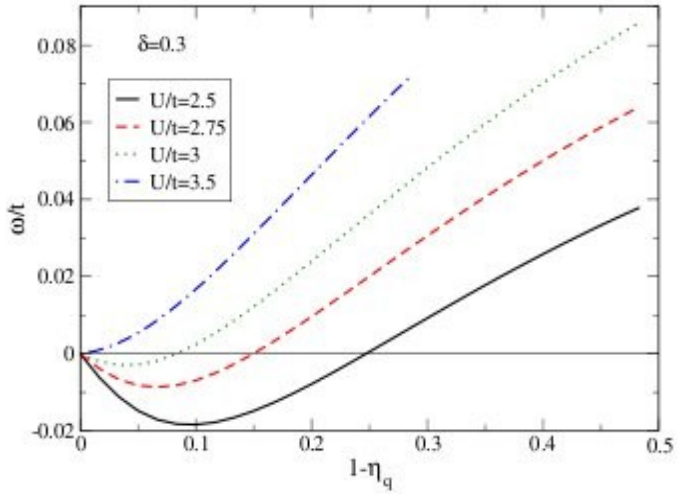
$$\eta_{\vec{q}} \equiv \frac{1}{D}\sum_{i}^{D}\cos\left(q_i\right) \longrightarrow 1 \geq \eta_{\vec{q}} \geq -1$$

$(\vec{q}=\vec{0})$ $(\vec{q}=(\pi,\ldots))$

i) Phase diagram of the hc-lattice: (F. Günther et al., PRL **98**, 176404 (2007))

instability of the paramagnet: spin-wave stability

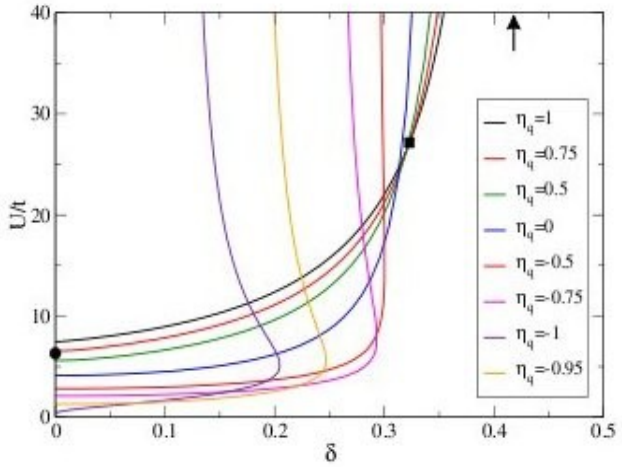




(DMFT: G. Uhrig, PRL **77**, 3629 (1996))

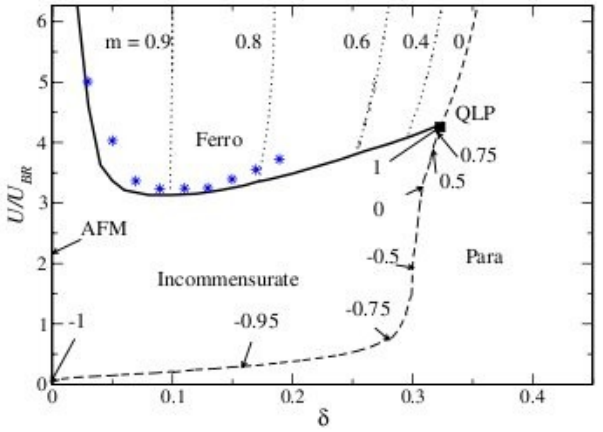
ii) Phase diagram of an sc lattice:

instability of the paramagnet:



stable spin waves

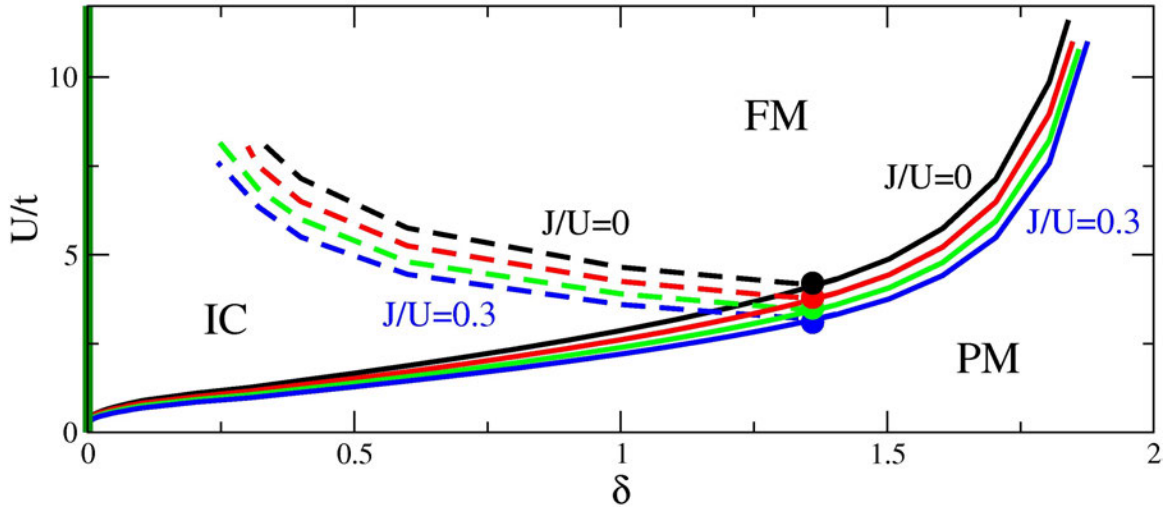
stability of the ferromagnet:



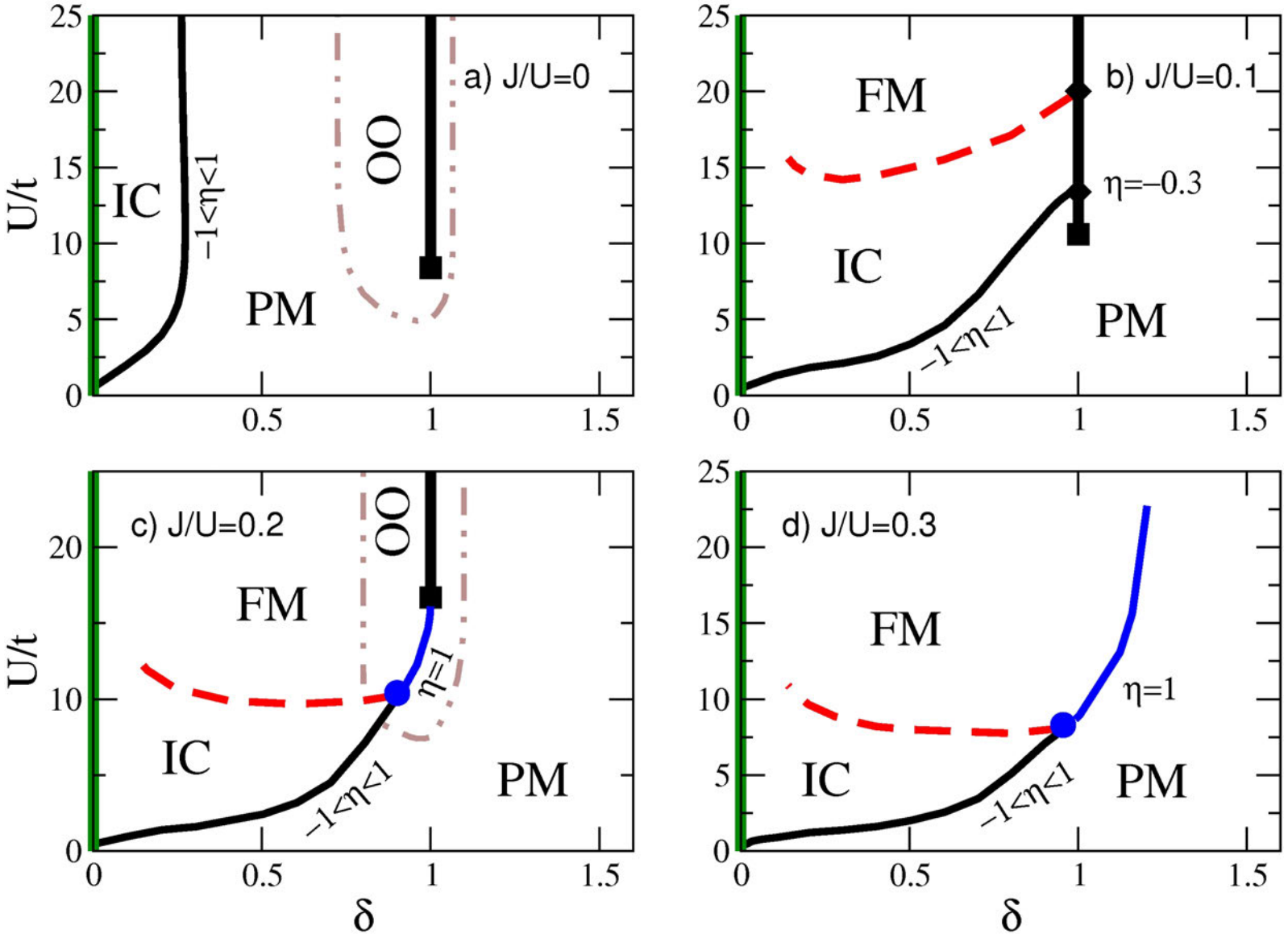
(DMFT: Obermeier et al., PRB **56**, 8479 (1997))

2.6 Spin excitations in a two-band model

Hartree-Fock phase diagrams:



Gutzwiller phase diagrams:



V) Conclusions

1. The Gutzwiller variational approach provides with us a numerically 'Cheap' way to study multi-band Hubbard models for transition metals and their compounds.
2. The modest numerical efforts of this method make it particularly suitable for a self-consistent merger with the DFT.
3. Apart from ground-state properties the method allows us to calculate quasi-particle excitations and two-particle response functions.
4. Systematic improvements of the infinite D limit are possible and sometimes necessary to study, e.g., correlation-induced forms of superconductivity.