



COLLÈGE
DE FRANCE
— 1530 —

Chaire de Physique de la Matière Condensée

***Des oxydes supraconducteurs
aux atomes froids
- la matière à fortes corrélations quantiques -***

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**Cycle 2009-2010
Cours 8 – 23 juin 2010**

Cours 8:

Corrélations électroniques dans les oxydes : effets physiques liés à la dégénérescence orbitale et au couplage de Hund.



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Séminaire:

Leon BALENTS

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Strong Spin-Orbit Physics and the Mott Regime

OUTLINE

- Interactions in a multi-orbital context
- Key energy scales: summary
- Effect of orbital degeneracy on the Mott transition – model considerations
- Effect of crystal-field splitting: orbital polarization - the titanates
- Effect of Hund's coupling (I): orbital compensation – BaVS_3
- Effect of Hund's coupling (II): suppressed quasiparticle coherence – Fe-pnictides, Sr_2RuO_4

WARNING:

- A lot of the material presented here touches on ongoing research – not everything is understood !
- → This last lecture is ~ more seminar-style
- **Special thanks to:**

M.Aichhorn, S.Biermann, M.Ferrero, F.Lechermann,
O.Parcollet, L.Pourovskii

*for discussions and collaborations on the topic(s) of
this lecture*

1. Interactions in a multi-orbital context

Recall lecture 6: must take matrix element of screened interaction within symmetry-adapted Wannier-like (or atomic-like) wave-functions in the solid-state environment

→ Local (on-site) interaction hamiltonian:

$$H_{\text{int}} = \frac{1}{2} \sum_{\sigma\sigma'} \sum_{m_1 \dots m_4} U_{m_1 m_2 m_3 m_4} d_{m_1 \sigma}^\dagger d_{m_2 \sigma'}^\dagger d_{m_4 \sigma'} d_{m_3 \sigma}$$

$$\begin{aligned} U_{m_1 m_2 m_3 m_4} &\equiv \langle \chi_{m_1} \chi_{m_2} | V_{\text{scr}} | \chi_{m_3} \chi_{m_4} \rangle \\ &= \int d\mathbf{r} d\mathbf{r}' \chi_{m_1}^*(\mathbf{r}) \chi_{m_2}^*(\mathbf{r}') V_{\text{scr}}(\mathbf{r}, \mathbf{r}') \chi_{m_3}(\mathbf{r}) \chi_{m_4}(\mathbf{r}') \end{aligned}$$

- $V(\mathbf{r}, \mathbf{r}')$ depends on both \mathbf{r}, \mathbf{r}' – not on distance $|\mathbf{r} - \mathbf{r}'|$ only
- The wave-functions are not the ones of the isolated atom

1.1 The case of an isolated atom - spherical symmetry -

Pioneering works:

- Friedrich Hund, ~ 1926
- John Slater, Phys Rev 34 (1929) 1293
- Giulio Racah, an amazing series of 4 Phys Rev articles from 1941 [Phys Rev 61 (1942) 186] till 1949 [Phys Rev 76 (1949) 1352]
→ pioneered powerful group-theory methods for spectra of multi-electron atoms.

Books: Condon&Shortley, Slater, Fano, Judd, Sobelman, ...

Unfortunately, references on extension to the solid-state are scattered and rather still incomplete...

Mostly in spectroscopy (a pioneer: G.Sawatzky) and 'LDA+U' literature

Giulio (Yoel) Racah (Hebrew: רַקַּח (יואל) ג'וליו, 1909-1965)

Giulio Racah's pioneering work: - a rather moving story -

Theory of Complex Spectra. I

GIULIO RACAH

The Hebrew University, Jerusalem, Palestine

(Received November 14, 1941)

This paper gives a closed formula which entirely replaces for the two-electron spectra the previous lengthy calculations with the diagonal-sum method. Applications are also made to some configurations with three or more electrons and to the p'' configurations of the nuclei.

Theory of Complex Spectra. IV

GIULIO RACAH

The Hebrew University, Jerusalem, Israel

(Received February 7, 1949)

§2. TWO-ELECTRON CONFIGURATIONS

If ω is the angle between the radii vectors of the two electrons, the coefficients f_k of F^k are⁴ the eigenvalues of the matrix

$$(l_1 l_2 m_1 m_2 | P_k(\cos \omega) | l_1 l_2 m_1' m_2') ; \quad (1)$$

here P_k is the Legendre polynomial of the order k . The transformation which diagonalizes this matrix is $(l_1 l_2 LM | l_1 l_2 m_1 m_2)$, and therefore

$$f_k(l_1 l_2 L) = \sum_{m_1 m_2 m_1' m_2'} (l_1 l_2 LM | l_1 l_2 m_1 m_2) (l_1 l_2 m_1 m_2 | P_k(\cos \omega) | l_1 l_2 m_1' m_2') (l_1 l_2 m_1' m_2' | l_1 l_2 LM), \quad (2)$$

or

$$f_k(l_1 l_2 L) = (l_1 l_2 LM | P_k(\cos \omega) | l_1 l_2 LM). \quad (3)$$

In the same way, if $\pm g_k$ are the coefficients of G^k for the singlet and for the triplet terms, we have

$$g_k(l_1 l_2 L) = \sum_{m_1 m_2 m_1' m_2'} (l_1 l_2 LM | l_1 l_2 m_1 m_2) (l_1 l_2 m_1 m_2 | P_k(\cos \omega) | l_1 l_2 m_1' m_2') (l_1 l_2 m_1' m_2' | l_1 l_2 LM), \quad (4)$$

and in view of⁵

$$(l_1 l_2 m_1' m_2' | l_1 l_2 LM) = (-1)^{l_1 + l_2 - L} (l_1 l_2 m_1 m_2 | l_1 l_2 LM), \quad (5)$$

this becomes

$$g_k(l_1 l_2 L) = (-1)^{l_1 + l_2 - L} (l_1 l_2 LM | P_k(\cos \omega) | l_1 l_2 LM). \quad (6)$$

Slater calculated the matrix elements of $P_k(\cos \omega)$ in the $l_1 l_2 m_1 m_2$ scheme, and then obtained the eigenvalues of this operator by means of the diagonal-sum procedure; we will calculate the matrix elements of $\cos \omega$ directly in the $l_1 l_2 LM$ scheme by the method of Güttinger and Pauli,⁶ and then calculate f_k and g_k with the ordinary methods of matrix calculations.

If \mathbf{u}_i is the unit vector in the direction from the origin to the electron i , by comparing TAS 4³21 with TAS 9³11, we have

$$(l_i \hat{z} | \mathbf{u}_i | l_i) = 0, \quad (l_i \hat{z} | \mathbf{u}_i | l_i - 1) = (l_i - 1 | \hat{z} | l_i) = \frac{1}{[(2l_i - 1)(2l_i + 1)]^{1/2}}; \quad (7)$$

and since

$$\cos \omega = (\mathbf{u}_1 \cdot \mathbf{u}_2), \quad (8)$$

introducing (7) in TAS 12³2 we find that the only non-vanishing elements of $(l_1 l_2 LM | \cos \omega | l_1' l_2' LM)$ are

$$\begin{aligned} (l_1 l_2 LM | \cos \omega | l_1 - 1 l_2 - 1 LM) &= -\frac{[(l_1 + l_2 + L + 1)(l_1 + l_2 + L)(l_1 + l_2 - L)(l_1 + l_2 - L - 1)]^{1/2}}{2[(2l_1 - 1)(2l_1 + 1)(2l_2 - 1)(2l_2 + 1)]^{1/2}}, \\ (l_1 l_2 LM | \cos \omega | l_1 + 1 l_2 - 1 LM) &= \frac{[(L + l_1 - l_2 + 2)(L + l_1 - l_2 + 1)(L + l_2 - l_1)(L + l_2 - l_1 - 1)]^{1/2}}{2[(2l_1 + 1)(2l_1 + 3)(2l_2 - 1)(2l_2 + 1)]^{1/2}}, \\ (l_1 l_2 LM | \cos \omega | l_1 - 1 l_2 + 1 LM) &= \frac{[(L + l_1 - l_2)(L + l_1 - l_2 - 1)(L + l_2 - l_1 + 2)(L + l_2 - l_1 + 1)]^{1/2}}{2[(2l_1 - 1)(2l_1 + 1)(2l_2 + 1)(2l_2 + 3)]^{1/2}}, \\ (l_1 l_2 LM | \cos \omega | l_1 + 1 l_2 + 1 LM) &= -\frac{[(l_1 + l_2 + L + 3)(l_1 + l_2 + L + 2)(l_1 + l_2 - L + 2)(l_1 + l_2 - L + 1)]^{1/2}}{2[(2l_1 + 1)(2l_1 + 3)(2l_2 + 1)(2l_2 + 3)]^{1/2}}. \end{aligned} \quad (9)$$

From these formulas it is possible to calculate the matrix elements of $P_k(\cos \omega)$ with the ordinary methods of matrix calculations; in order that these elements may have a value different from zero, k must satisfy the conditions

$$k + l_1 + l_2 = 2g_1, \quad k + l_2 + l_1 = 2g_2 \quad (10)$$

⁴ TAS §8.

⁵ TAS 14³ 7.

⁶ Güttinger and Pauli, Zeits. f. Physik 67, 743 (1931); TAS §10³ et seq.

Excerpt
from paper 1
(sub. 1941, pub. 1942 !)
A beautiful illustration
of the spiritual strength
of a man in barbarian
times...

Isolated atom:

$$V_c(\mathbf{r} - \mathbf{r}') = \frac{e^2}{|\mathbf{r} - \mathbf{r}'|}$$
$$\chi_m(\mathbf{r}) = R_{nl}(\mathbf{r}) Y_{lm}(\theta, \phi)$$

Separate radial and angular variables:

$$V_c(\mathbf{r} - \mathbf{r}') = \sum_{k=0}^{\infty} \frac{4\pi}{2k+1} \frac{r_{<}^k}{r_{>}^{k+1}} \sum_{q=-k}^{+k} Y_{kq}(\theta, \phi) Y_{kq}^*(\theta', \phi')$$

$r_{>}, r_{<}$:
largest/smallest
of r, r'

This yields:

$$U_{m_1 m_2 m_3 m_4}^{\text{at}} = \sum_k \omega_k(m_1, m_2, m_3, m_4) F^k$$

In which:

- The F 's are Slater integrals involving radial wave-functions
- The ω 's are entirely known numbers (Racah-Wigner)
- The sum involves only F_0, F_2, F_4 for d-shell, F_0, F_2, F_4, F_6 for f-shell

$$F^k = e^2 \int_0^\infty r^2 dr \int_0^\infty r'^2 dr' \frac{r^k}{r^{k+1}} R_{nl}^2(r) R_{nl}^2(r')$$

$$\omega_k(m_1, m_2, m_3, m_4) = \frac{4\pi}{2k+1} \sum_{q=-k}^{+k} \langle Y_{lm_1} | Y_{kq} | Y_{lm_3} \rangle \langle Y_{lm_2} | Y_{kq}^* | Y_{lm_4} \rangle$$

$$\langle Y_{lm} | Y_{kq} | Y_{lm'} \rangle \equiv \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi Y_{lm}^*(\theta, \phi) Y_{kq}(\theta, \phi) Y_{lm'}(\theta, \phi)$$

$$= (2l+1) \sqrt{\frac{2k+1}{4\pi}} \begin{pmatrix} l & k & l \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & k & l \\ -m & q & m' \end{pmatrix}$$

Wigner-
-Racah
3j symbols

If hydrogen atom wave functions are used:

$$F^4/F^2 \simeq 0.625 \quad (3d - \text{shell})$$

$$F^4/F^2 \simeq 451/675 \simeq 0.67 \quad , \quad F^6/F^2 \simeq 1001/2025 \simeq 0.49 \quad (4f\text{-shell})$$

1.2 Atom in the solid-state

- Two key differences: i) Wave-functions are atomic-like Wannier functions, transforming according to irreducible representation of the crystal point group and ii) The interaction potential is screened: $V_c(r-r') \rightarrow V_{scr}(r,r')$
- Such calculations start to become possible (from first principle), e.g. `constrained RPA' (GW) approach discussed in lecture 6
- No complete understanding or consensus yet...

For 1st understanding, let's select
 specific terms:
 density-density interactions

(Beware: keeping those terms only violates orbital symmetries)

Hamiltonian then simplifies to: (often used in practical calculations !)

$$H_{int} = \frac{1}{2} \sum_{mm',\sigma} U_{mm'}^{\sigma\sigma} n_{m\sigma} n_{m'\sigma} + \frac{1}{2} \sum_{mm'} U_{mm'}^{\sigma\bar{\sigma}} (n_{m\uparrow} n_{m'\downarrow} + n_{m\downarrow} n_{m'\uparrow})$$

with:

$$U_{mm'}^{\sigma\bar{\sigma}} = U_{mm'mm'} \equiv U_{mm'} \quad \text{Antiparallel spins}$$

$$U_{mm'}^{\sigma\sigma} = U_{mm'mm'} - U_{mm'm'm} \equiv U_{mm'} - J_{mm'}$$

Parallel spins → Interaction reduced by exchange J

→ `2nd Hund rule' (largest possible L)

Empirical knowledge/beliefs...

- When considering localized-enough Wannier (~ atomic orbitals), interaction matrix can be constructed in the spherical-symmetry approximation, with:
 - Slater F_0 strongly reduced by screening (overall electrostatic interaction)
 - F_2, F_4, F_6 not so different from isolated atom values.
 - Hence, Hund's exchange, which depends only on F_2, F_4 , is much less severely screened
 - Rationale for this: these Slater integrals have a lot more weight close to the nucleus.

Let's look at some recent first-principle results (cRPA/GW)

Iron pnictide '1111' LaOFeAs, retaining full Fe-d, O-p and As-p bands → quite localized Wannier functions:

[M.Aichhorn,...T.Miyake... et al. PRB 80 (2009) 085101]

$$U_{mm'}^{\sigma\bar{\sigma}}|_{\text{cRPA}} = \begin{pmatrix} 3.77 & 2.35 & 2.21 & 2.71 & 2.71 \\ 2.35 & 3.94 & 2.87 & 2.44 & 2.44 \\ 2.21 & 2.87 & 3.31 & 2.29 & 2.29 \\ 2.71 & 2.44 & 2.29 & 3.48 & 2.29 \\ 2.71 & 2.44 & 2.29 & 2.29 & 3.48 \end{pmatrix}$$

$$U_{mm'}^{\sigma\sigma}|_{\text{cRPA}} = \begin{pmatrix} 0.00 & 1.61 & 1.55 & 2.26 & 2.26 \\ 1.61 & 0.00 & 2.50 & 1.82 & 1.82 \\ 1.55 & 2.50 & 0.00 & 1.70 & 1.70 \\ 2.26 & 1.82 & 1.70 & 0.00 & 1.74 \\ 2.26 & 1.82 & 1.70 & 1.74 & 0.00 \end{pmatrix}$$

The ordering of orbitals in those matrices is d_{z^2} , $d_{x^2-y^2}$, d_{xy} , d_{xz} , and d_{yz} .

Satisfies many of the above expectations

'Best fit' to spherically symmetric form → ~ 9% accurate:

$$U_{mm'}^{\sigma\bar{\sigma}} = \begin{pmatrix} 3.59 & 2.19 & 2.19 & 2.73 & 2.73 \\ 2.19 & 3.59 & 2.91 & 2.37 & 2.37 \\ 2.19 & 2.91 & 3.59 & 2.37 & 2.37 \\ 2.73 & 2.37 & 2.37 & 3.59 & 2.37 \\ 2.73 & 2.37 & 2.37 & 2.37 & 3.59 \end{pmatrix}$$

$$U_{mm'}^{\sigma\sigma} = \begin{pmatrix} 0.00 & 1.49 & 1.49 & 2.30 & 2.30 \\ 1.49 & 0.00 & 2.57 & 1.76 & 1.76 \\ 1.49 & 2.57 & 0.00 & 1.76 & 1.76 \\ 2.30 & 1.76 & 1.76 & 0.00 & 1.76 \\ 2.30 & 1.76 & 1.76 & 1.76 & 0.00 \end{pmatrix}$$

$U \sim 2.7 \text{ eV}$
 $J \sim 0.8 \text{ eV}$

Construct (much) more extended Wannier's
from 5 d-bands only:

$$U_{mm'}^{\sigma\bar{\sigma}}|_{\text{cRPA}} = \begin{pmatrix} 3.17 & 2.02 & 1.72 & 2.22 & 2.22 \\ 2.02 & 3.36 & 2.16 & 2.04 & 2.04 \\ 1.72 & 2.16 & 2.17 & 1.73 & 1.73 \\ 2.22 & 2.04 & 1.73 & 2.73 & 1.84 \\ 2.22 & 2.04 & 1.73 & 1.84 & 2.73 \end{pmatrix}$$

$$U_{mm'}^{\sigma\sigma}|_{\text{cRPA}} = \begin{pmatrix} 0.00 & 1.41 & 1.26 & 1.87 & 1.87 \\ 1.41 & 0.00 & 1.91 & 1.54 & 1.54 \\ 1.26 & 1.91 & 0.00 & 1.33 & 1.33 \\ 1.87 & 1.54 & 1.33 & 0.00 & 1.44 \\ 1.87 & 1.54 & 1.33 & 1.44 & 0.00 \end{pmatrix}$$

- Spherical approximation becomes very bad (~ 25% error)
- Smaller (average) U and J, as expected
- Strongly orbital-dependent

Take-home message:

- For systems with strong hybridization to ligand orbitals (e.g late TMOs), beware of models involving d-only orbitals AND local interactions only in the d-shell → keeping the ligand is safest, possibly treating U_{pd} in Hartree approximation
- For early TMOs, much less of a problem → d-only hamiltonian ~ OK

1.3 Full hamiltonian for t_{2g} in cubic symmetry (consistent with all symmetries)

From symmetry considerations, only 2 independent parameters entirely specify the full interaction tensor

Non-zero matrix elements are :

$$\begin{aligned} u &= U_{aaaa} \\ u' &= U_{abab} = u - 2j \\ j &= U_{aabb} = U_{abba} \end{aligned}$$

Can be expressed from 'screened' Slater integrals:

$$\begin{aligned} u &= F^0 + \frac{4}{49}F^2 + \frac{4}{49}F^4 \\ u' &= F^0 - \frac{2}{49}F^2 - \frac{4}{441}F^4 \\ j &= \frac{3}{49}F^2 + \frac{20}{441}F^4 \end{aligned}$$

$$\begin{aligned} H_{t_{2g}} &= U \sum_a n_{a\uparrow} n_{a\downarrow} + \sum_{a < b, \sigma} [U' n_{a\sigma} n_{b\bar{\sigma}} + (U' - J) n_{a\sigma} n_{b\sigma}] \\ &- J c_{a\sigma}^\dagger c_{a\bar{\sigma}} c_{b\bar{\sigma}}^\dagger c_{b\sigma} - J c_{a\sigma}^\dagger c_{a\bar{\sigma}} c_{b\sigma} c_{b\bar{\sigma}} \end{aligned}$$

← density-density terms

← spin-flip and pair-hopping terms (non density-density)

t_{2g} : Manifestly invariant form

$$\hat{n} = \sum_{a\sigma} \hat{n}_{a\sigma}$$

Charge

$$\vec{S} = \frac{1}{2} \sum_a d_{a\sigma}^\dagger \vec{\sigma}_{\sigma\sigma'} d_{a\sigma'}$$

Spin

$$\vec{T} = \sum_{\sigma} d_{a\sigma}^\dagger \vec{\tau}_{ab} d_{b\sigma}$$

$$T_c = i \sum_{\sigma} \sum_{ab} \epsilon_{abc} d_{a\sigma}^\dagger d_{b\sigma}$$

Orbital momentum
(flavor isospin)

$$H_{t_{2g}} = (U - 3J) \frac{1}{2} \hat{n}(\hat{n} - 1) - 2J \vec{S}^2 - \frac{J}{2} \vec{T}^2$$

Question: how important are the non density-density terms ?

2. TMOs: Summary of key energy scales (qualitative)

- Bandwidth, or rather kinetic energy in occupied part of the d-band (related to t_{pd} and Δ)
- Distance to ligand (O states): charge-transfer energy Δ
- U_{dd} on d-states
- J Hund's (exchange) coupling: density-density and spin-flip/pair hopping
- U_{pd} between metal and ligand (is Hartree approx enough for this one ?)

3. Mott transition with orbital degeneracy: results of model studies

3.1) N degenerate orbitals, U-only, J=0

'Brinkman-Rice' critical value:

i) increases with N

ii) is largest for half-filled band

e.g from Gutzwiller, slave-bosons or DMFT calculation (U_{c2})

$$U_c(N, n) = |\bar{\epsilon}(n)| \frac{\left[\sqrt{n(2N - n + 1)} + \sqrt{(n + 1)(2N - n)} \right]^2}{2N - n}$$

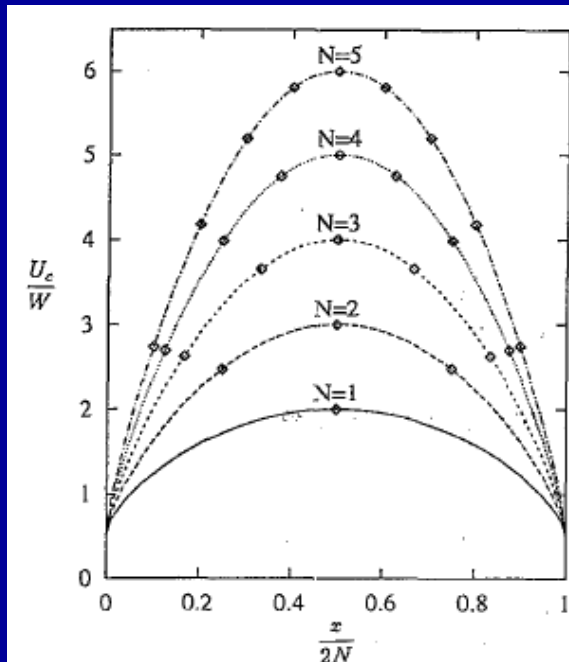
For n electrons per site,

$\bar{\epsilon}(n)$ = kinetic energy per electron

Half-filling $n=N$ (U_c largest):

$$U_c(n = N) = 4|\bar{\epsilon}(n)| (N + 1) \quad \text{increases } \sim N \text{ with } N$$

Lu, PRB (1994)



Note (subtlety, possibly relevant in practice though):

This is the BR critical coupling at which $Z \rightarrow 0$

However, an insulating solution with sizeable gap may exist much below this, for $U > U_{c1} \sim N^{1/2}$

cf: Gunnarsson et al., PRB (1996), Florens et al. PRB (2002)

3.2) The Hund's exchange strongly affects these results, leading to qualitatively different behavior:

- Critical value is strongly reduced as compared to $J=0$
- For density-density (Ising) coupling, transition is strongly 1st order
- Non density-density terms are not so important far from the transition (i.e do not affect Z very much) but become crucially important in the transition region

Several studies 1995-2010, renewed interest w/ iron pnictides

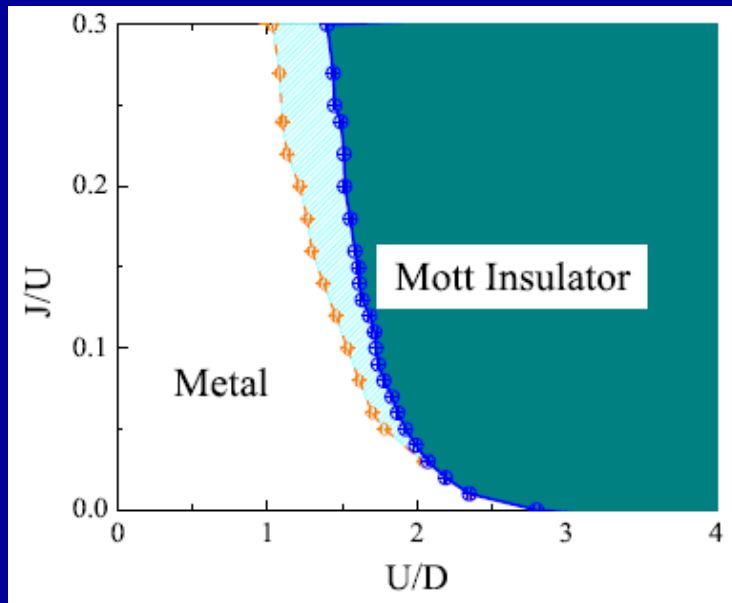


FIG. 3: (Color online) Phase diagram in the $J - U$ plane of the two-orbital model at half-filling, the blue circles (orange diamonds) give the phase boundary of the metal-to-Mott-insulator transition if the full (Ising-type) Hund's coupling is considered.

Yu and Si, arXiv:1006.2337

(June, 15, 2010 !)

'slave-spin' formalism

See also: Ono et al. PRB (2003)

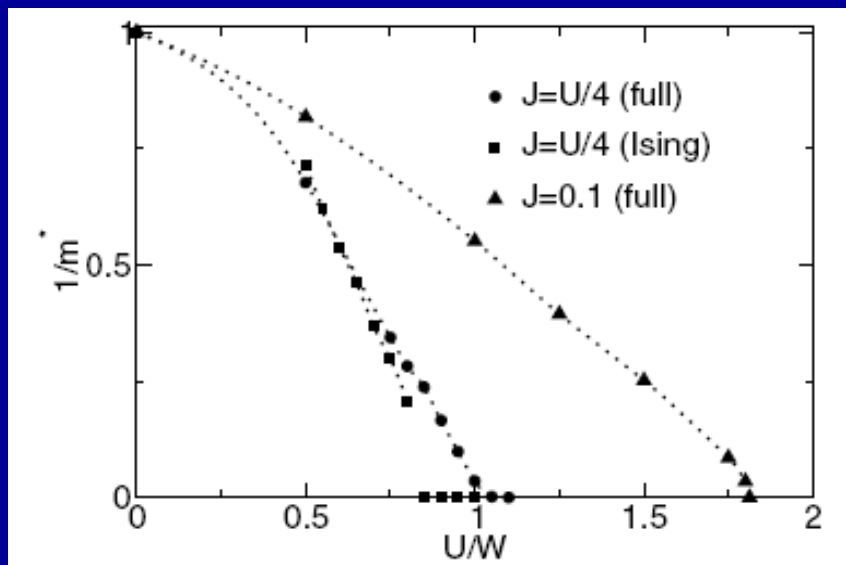


Fig. 8. Inverse effective mass as function of U/W and $J = U/4$ for the full (circles) and Ising-like exchange interaction (squares). The latter shows a strong jump in $1/m^*$ at U_c , leading to a first order transition, while the former vanishes continuously. The triangles represent a calculation with fixed $J = 0.1$. The lines are meant as guide to the eyes.

T.Pruschke and R.Bulla

EPJB 44 (2005) 217

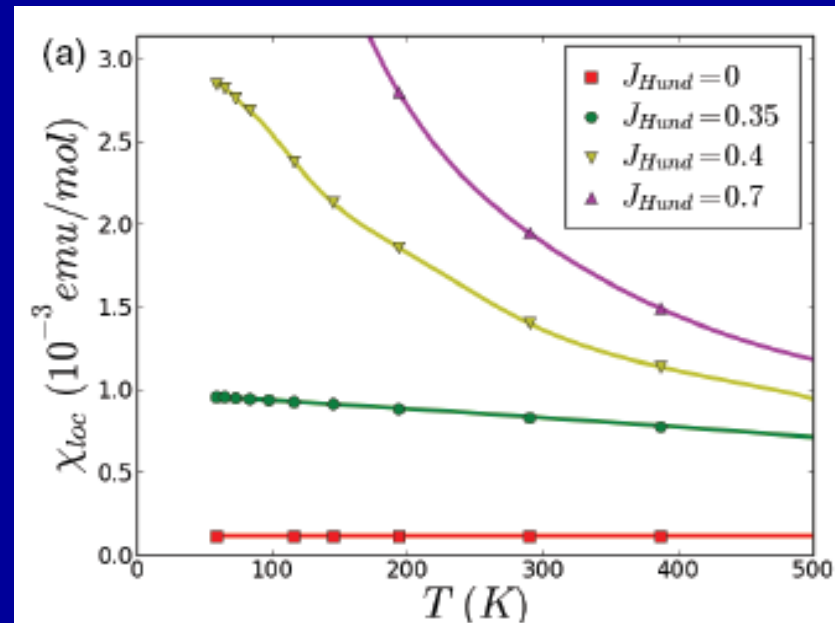
DMFT

First (?) careful study of non density-density terms

Importance of Hund's coupling in practice

- e.g Iron pnictides: would be very weakly correlated if not for finite J
- Indeed: 5 bands, 6 electrons, bandwidth $\sim 4\text{eV}$
- Above formula with $J=0$ would yield $U_c \sim 25 \text{ } \epsilon \sim 10\text{-}20 \text{ eV} !$

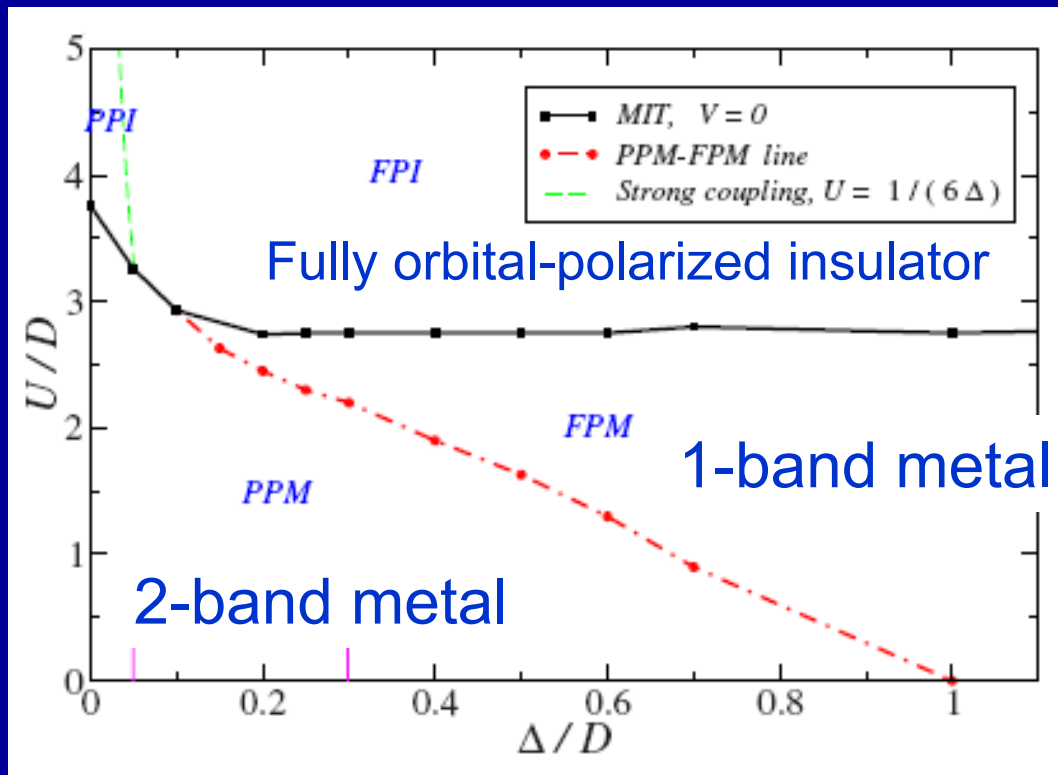
- Actually, physics of iron pnictides is quite sensitive to J :
cf. Haule-Kotliar NJP 2009
Aichhorn et al. (FeSe
→ Hund's correlated)
arXiv:1003.1286



4. Splitting of orbital degeneracy

- orbital polarization and the Mott transition-

Model example: 1 electron in 2 orbitals, split by Δ , $J=0$
(Manini et al., PRB 2002; Poteryaev et al. PRB 2008)



Lifting of orbital
Degeneracy:

- Favors insulating state
- Triggers orbital polarization

Strong-coupling (Kugel-Khomskii) analysis of the insulator :

$$\hat{H}_{\text{eff}} = -\Delta \sum_i \hat{T}_i^z + \sum_{\langle ij \rangle} \{J_s(\vec{S}_i \vec{S}_j) + J_o(\vec{T}_i \vec{T}_j) + J_m(\vec{S}_i \vec{S}_j)(\vec{T}_i \vec{T}_j)\}.$$

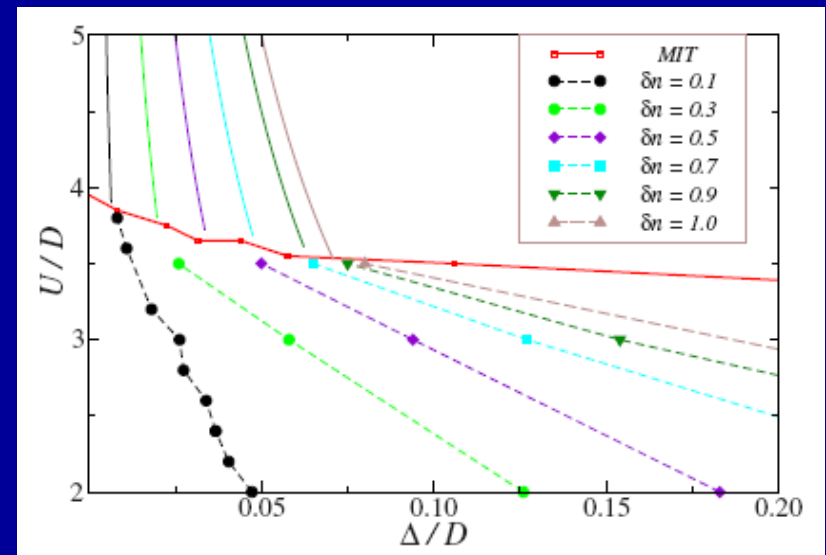
For U-only model:

$$J_s = J_o = \frac{J_m}{4} = \frac{2t^2}{U}$$

$$\delta n = \tanh\left[\frac{\beta}{2}(\Delta - \Delta_c \delta n)\right] \quad \Delta_c = \frac{zJ}{2} = z \frac{t^2}{U},$$

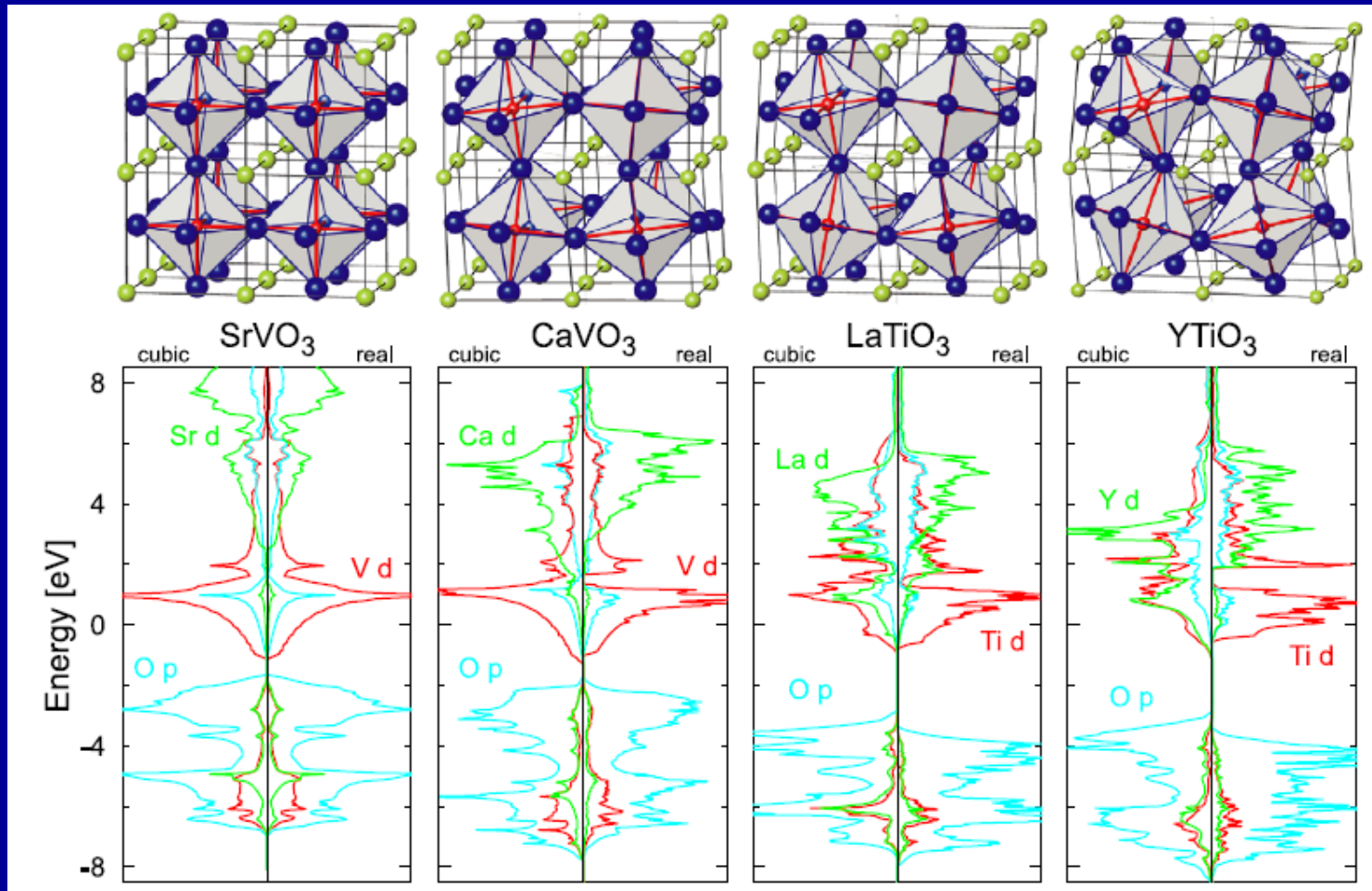
Hence, orbital polarisability of PM insulator is LARGE $\sim U/t^2$

As soon as inter-orbital splitting (can be fraction of an eV) becomes larger than J_0 (a few ~ 1000 K) insulator becomes orbitally polarized



Iso-orbital polarization contours

We believe that this is important in explaining why d1 titanates RTiO_3 are Mott insulators, while d1 vanadates RVO_3 are not.



The two effects of distortion:

- 1) Reduction of total t_{2g} bandwidth:

Table 8. t_{2g} edge-to-edge ($W_{t_{2g}}$) and rms (W) bandwidths in eV.

	SrVO ₃ [42]	CaVO ₃ [43]	LaTiO ₃ [44]	LaTiO ₃ [12]	YTiO ₃ [20]
$W_{t_{2g}}$	2.85	2.45	2.09	1.92	2.05
W	2.85	2.39	2.18	2.08	1.87

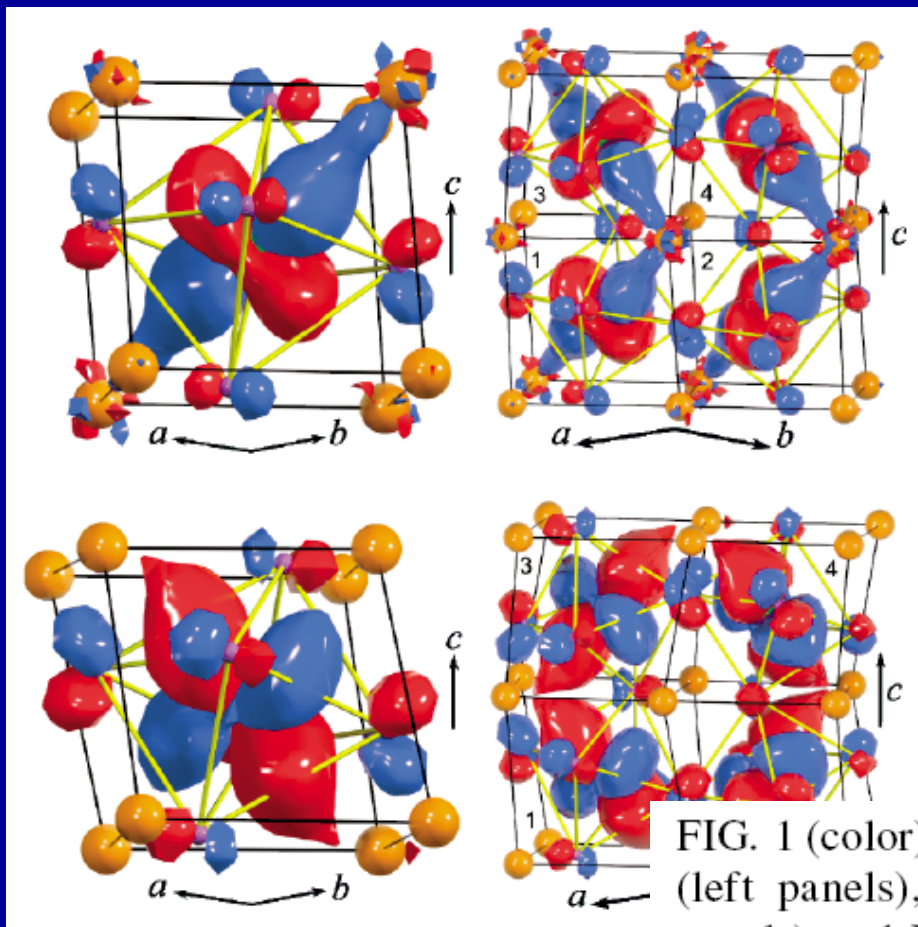
This is because the O-M-O bond is no longer straight
→ pi-bonding less efficient

-2) Splitting between t_{2g} orbitals (lifting of orbital degeneracy)

(140,200) meV for LaTiO₃ ; (200,330) meV for YTiO₃

→ **Both effects** are responsible for the Mott insulating state of LaTiO₃ and YTiO₃

One specific linear combination of orbitals dominantly populated:



$$0.586|xy\rangle + 0.275|xz\rangle + 0.762|yz\rangle \text{ for LaTiO}_3$$

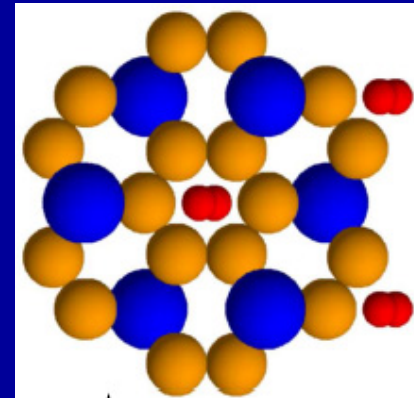
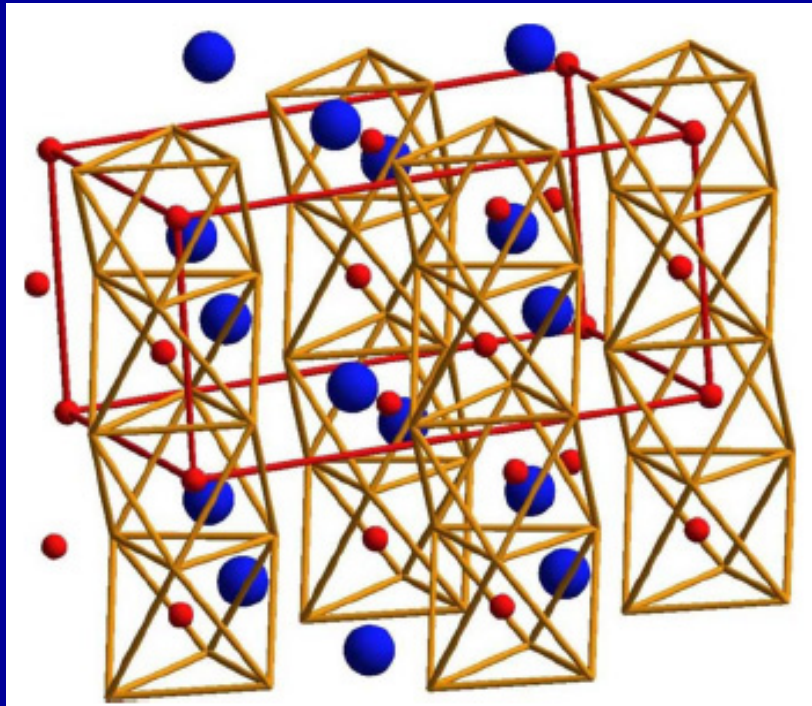
(88% of d1 electron population)

$$0.622|xy\rangle - 0.029|xz\rangle + 0.782|yz\rangle \text{ for YTiO}_3$$

(96% of d1 electron population)

FIG. 1 (color). *Pbnm* primitive cells (right panels), subcells 1 (left panels), and the occupied t_{2g} orbitals for LaTiO_3 (top panels) and YTiO_3 (bottom panels) according to the LDA + DMFT calculation. The oxygens are violet, the octahedra yellow, and the cations orange. In the global, cubic xyz system

5. Hund's coupling can fight crystal-field
→ compensation of orbital populations



BaVS3: A zoo of phase transitions

(all 2nd order)

-Structural transition T=240K

hexagonal > orthorombic, zig-zag distortion of chains

-Metal-Insulator at T=70K to **NON-MAGNETIC** insulator

Now known to be CDW ordering

(Inami et al, 2002; Fagot et al, 2003)

Note: - **2nd order transition !**

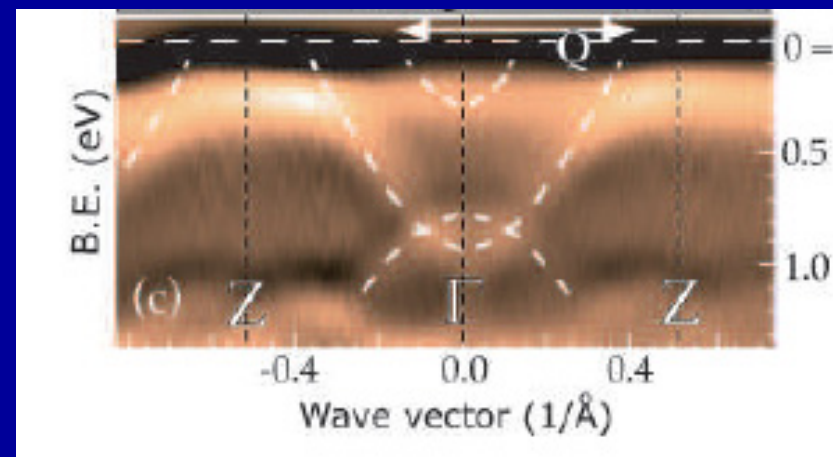
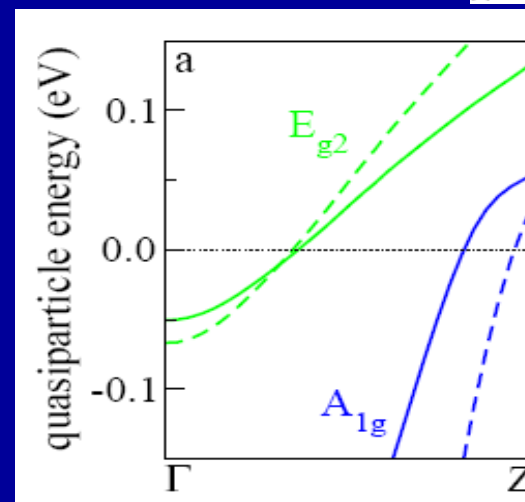
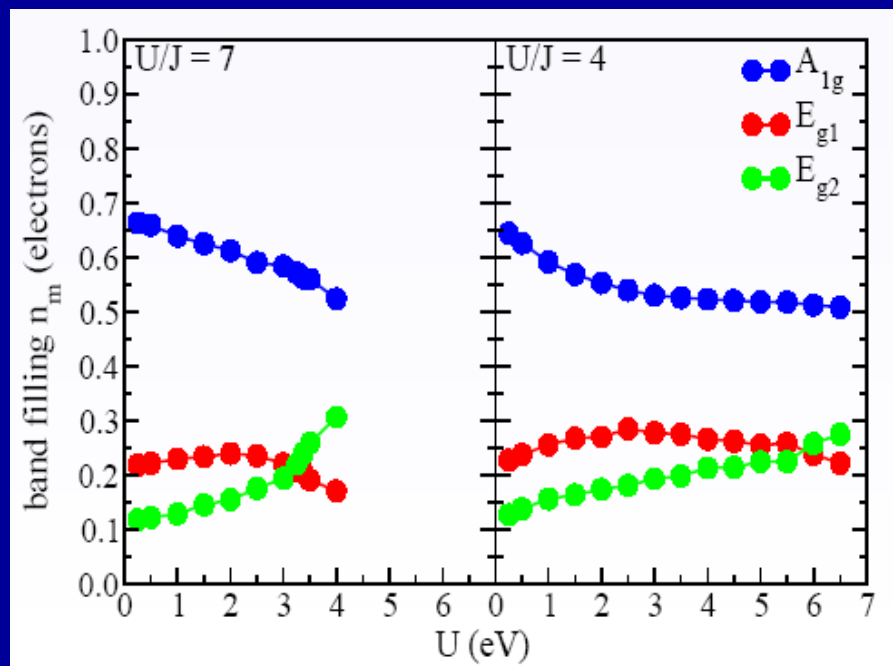
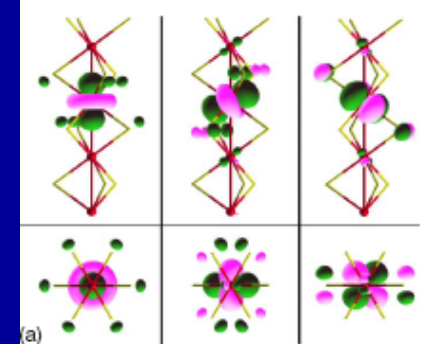
- MIT can be driven to T=0 under pressure (20kbar)

>> QCP (Forro et al., 2000)

-Magnetic ordering at T=30K (incommensurate SDW ?)

(Nakamura, 2000)

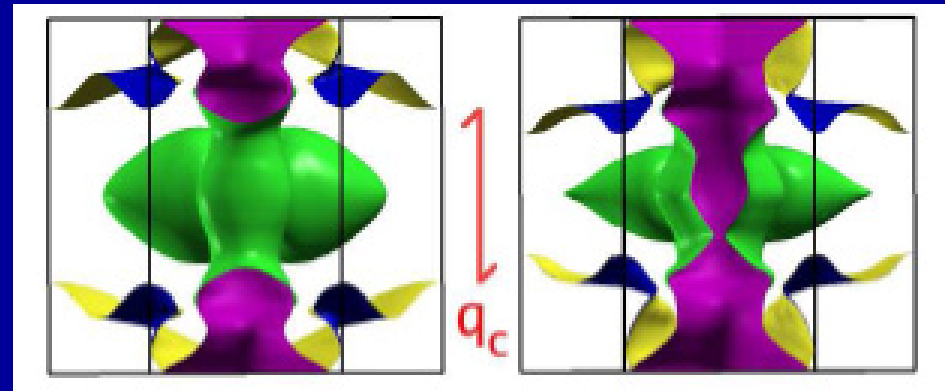
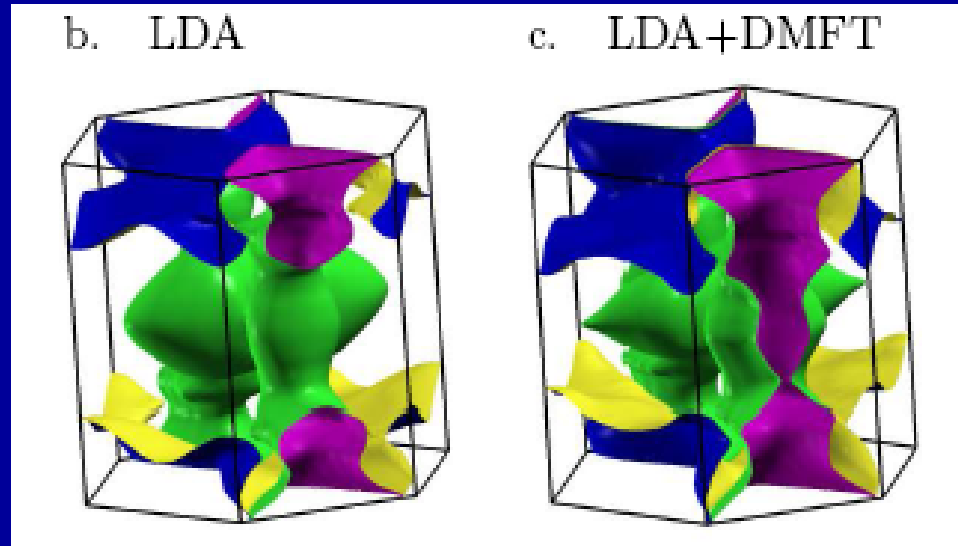
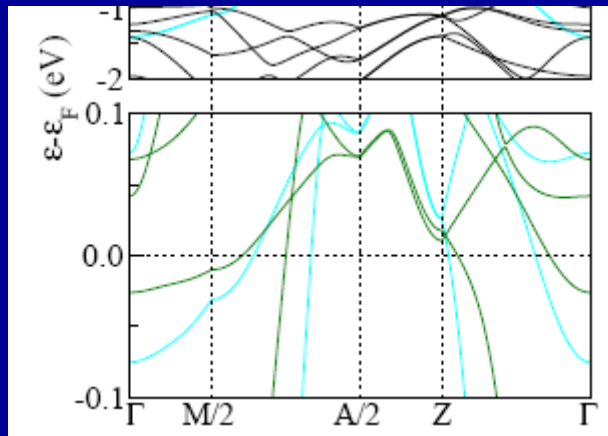
Lechermann, Biermann and A.G: BaVS3
 PRL 2005; PRB 2007



Intensive studies of this
 material @EPFL (Forro et al)

Mitrovic et al. (2005)

Fermi surface is modified by correlations:



>> Favorable conditions for a CDW transition (\sim nesting) are created by strong correlations

6. Hund's coupling suppresses the quasiparticle coherence scale

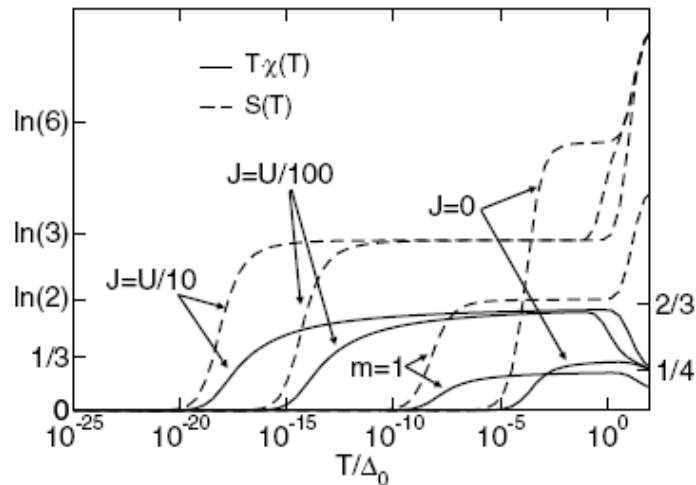


Fig. 2. Thermodynamic properties for a particle-hole symmetric two-orbital impurity model with varying Hund's rule coupling $J = 0, U/100,$ and $U/10$; dashed lines: entropy, solid lines: effective squared moments. The Coulomb parameter U and the hybridization Δ_0 were chosen such that the system is in the Kondo limit. The inter-orbital Coulomb parameter is fixed to $U' = U - 2J$ by rotational invariance. For comparison the results of a single-orbital SIAM are included.

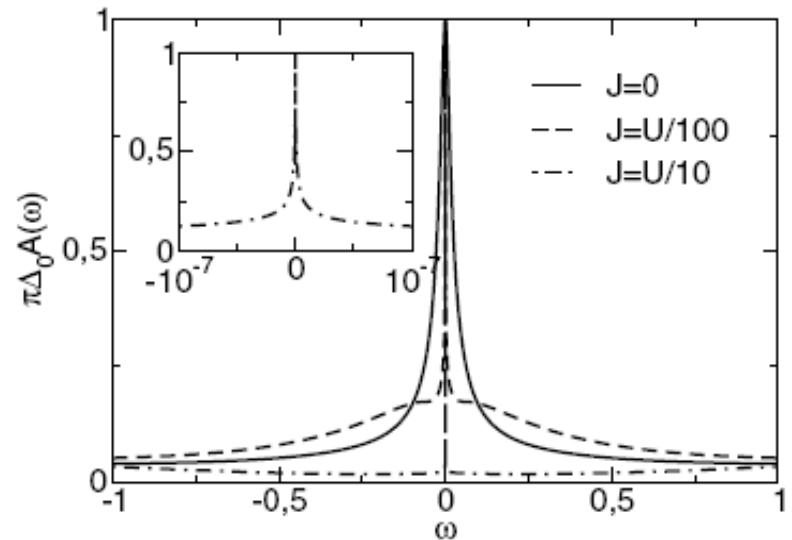


Fig. 3. Local DOS at $T = 0$ for a particle-hole symmetric two-orbital impurity model with Hund's rule coupling $J = 0, U/100$ and $U/10$. Other parameters are the same as in Figure 2.

'Poor man's scaling' analysis

(Nevidomskiy and Coleman, PRL 2010)

$$H = \sum_{\mathbf{k}, \sigma, \mu} \varepsilon_{\mathbf{k}} c_{\mathbf{k}\sigma\mu}^\dagger c_{\mathbf{k}\sigma\mu} - J_H \left(\sum_{\mu=1}^K \mathbf{S}_\mu \right)^2 + J \sum_{\mu=1}^K \mathbf{S}_\mu \cdot \boldsymbol{\sigma}_\mu$$

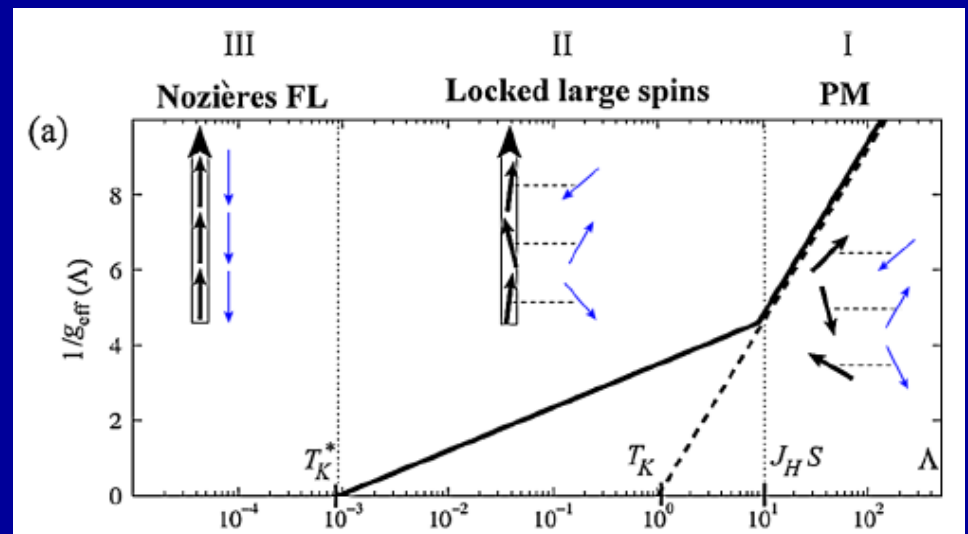
Impurity spin is made of K spin-1/2

K-channels, $J > 0 \rightarrow S = 2K$

$$\frac{d(J\rho)}{d \ln \Lambda} = -2(J\rho)^2 + 2(J\rho)^3,$$

$$\frac{d(J_H \rho)}{d \ln \Lambda} = 4(J\rho)^2 J_H \rho,$$

$$T_K^* \sim J_H S \left(\frac{T_K}{J_H S} \right)^K \equiv T_K \left(\frac{T_K}{J_H S} \right)^{K-1}$$



→ Important to explain low coherence scale/large mass of some 4d oxides, such as Sr₂RuO₄ cf. J.Mravlje

Transition-metal oxides:

- 3d \rightarrow MOTT (U)
- 4d \rightarrow Hund (J)
- 5d \rightarrow Spin-orbit (λ)

?????



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``Cuprates supraconducteurs: où en est-on ?

6 séances = 6 cours + 9 séminaires

Les MARDI de 14h00 a 17h45

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