Beyond DFT: DMFT and its extensions

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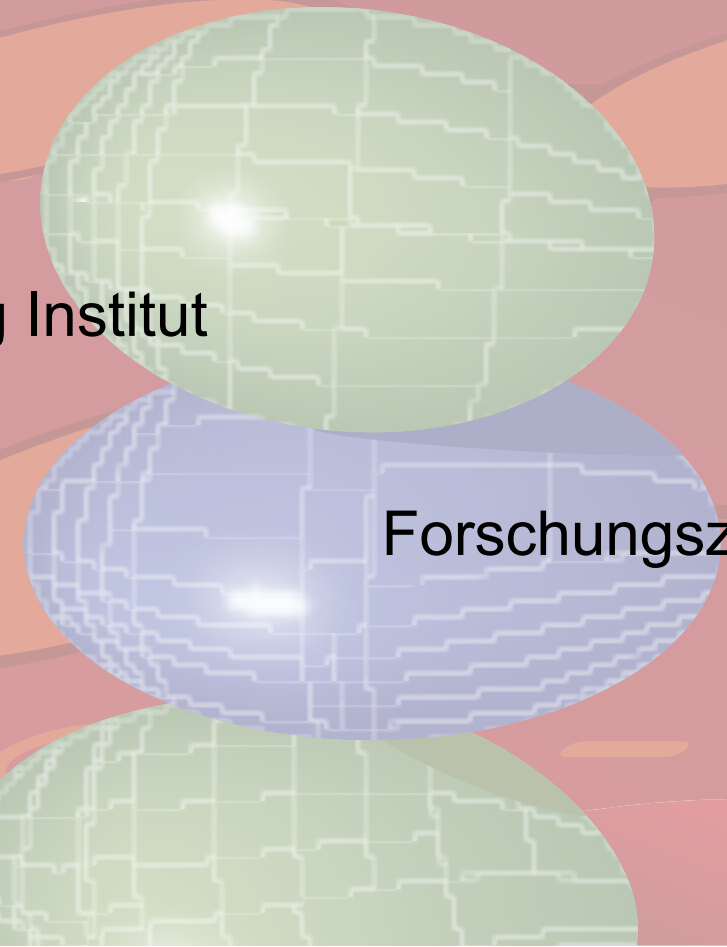

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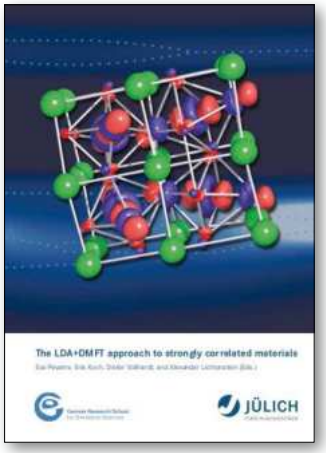
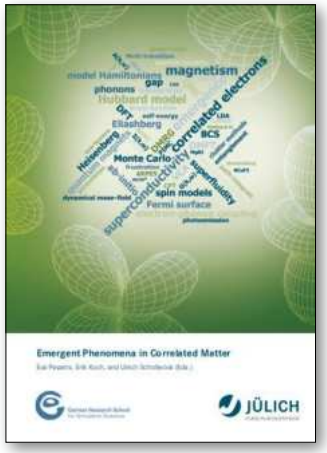
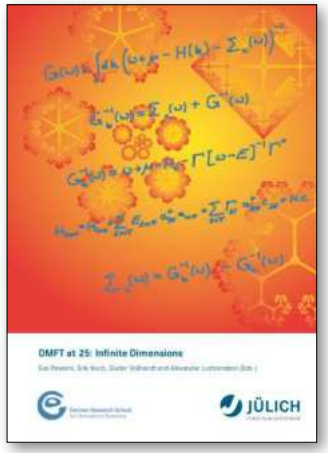
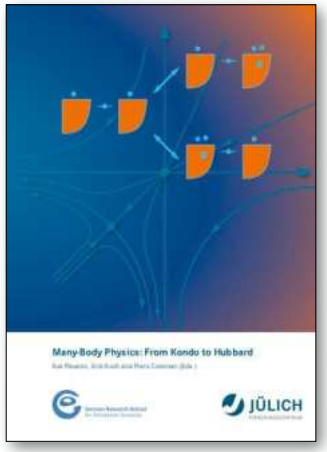
Autumn School on Correlated Electrons

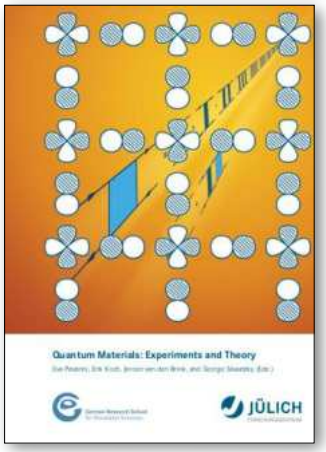
www.cond-mat.de/events/correl.html

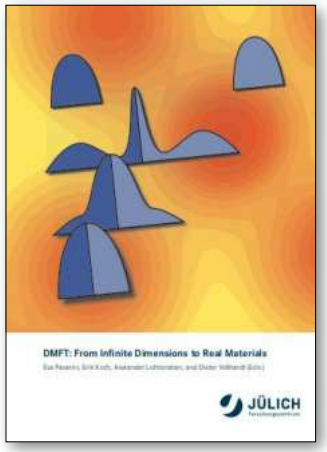


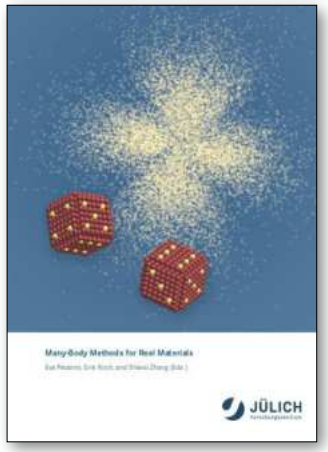


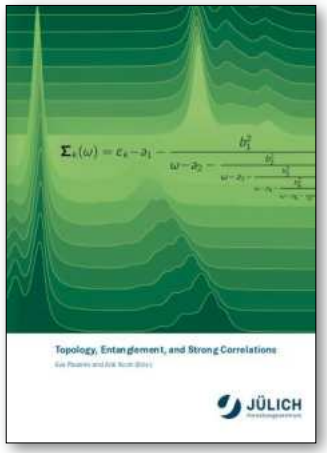


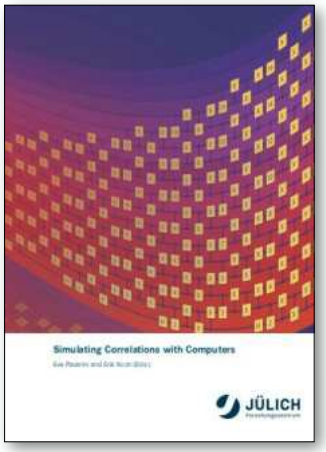


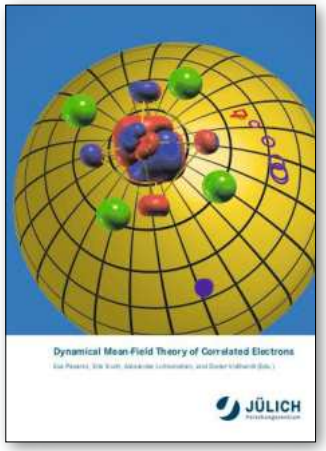


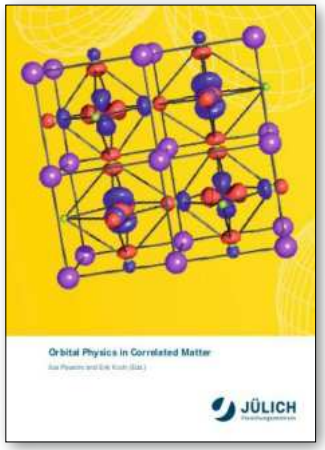


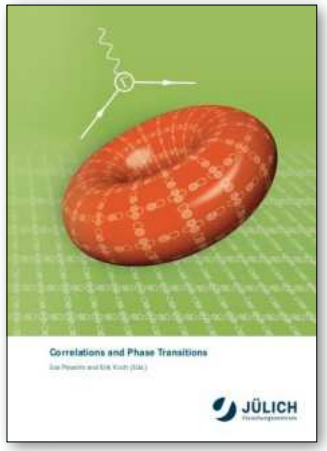


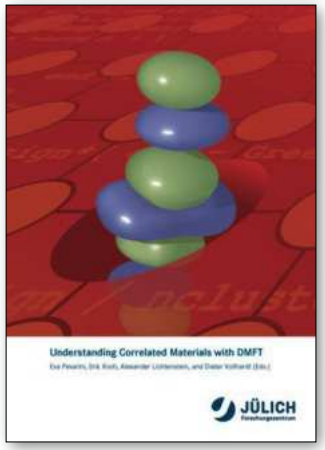






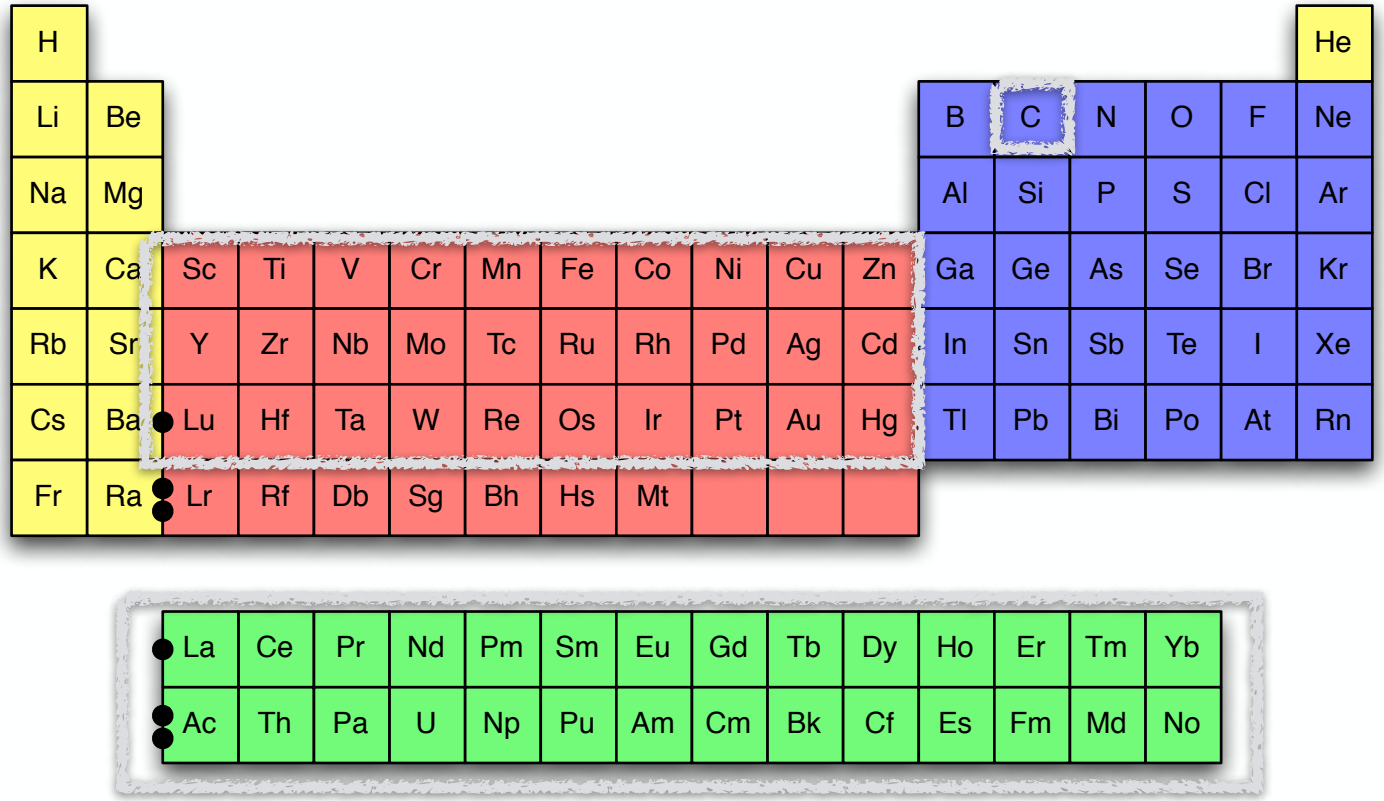






what are correlated materials?

systems for which *single-electron picture* fails *qualitatively*
because atomic-like physics survives in the solid



Coulomb-induced metal-insulator transition

heavy-Fermions

unconventional superconductivity

spin-charge separation

.....

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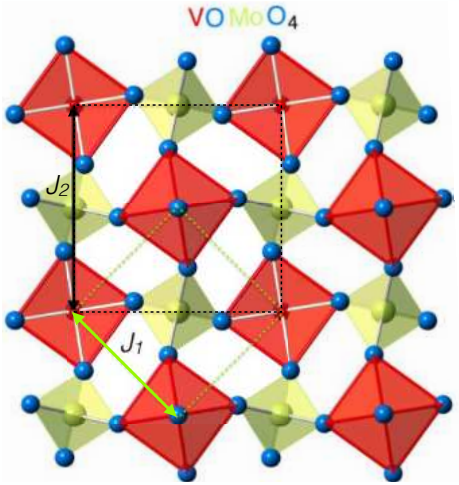
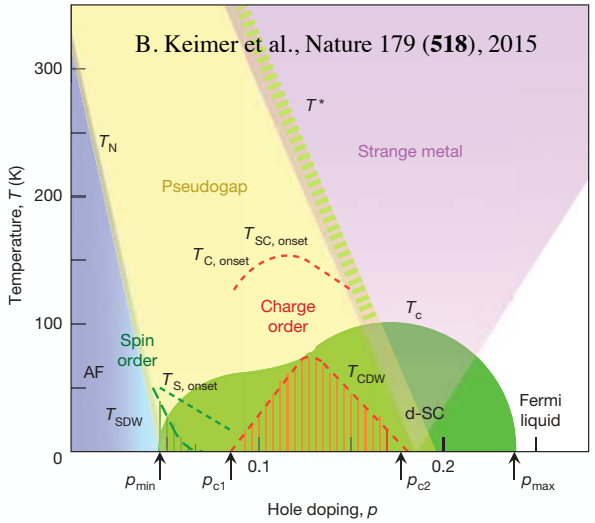
representative Mott systems

single-band systems

magnetic phases & frustration



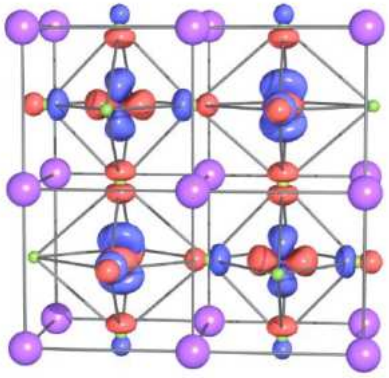
HTSC

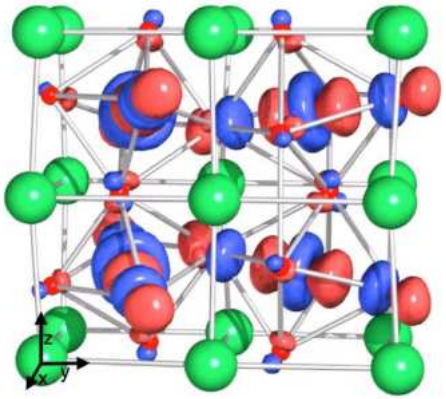


two-band systems (e_g)

orbital physics and orbital ordering

$KCuF_3$





$LaMnO_3$

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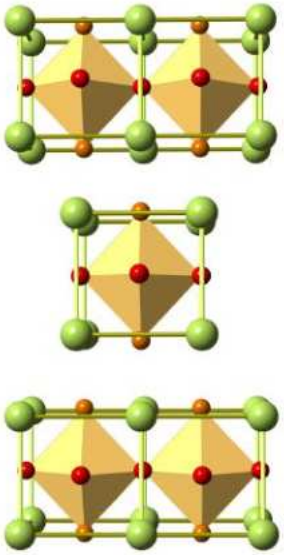
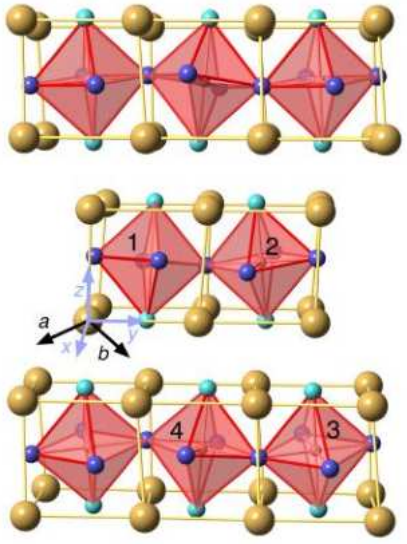
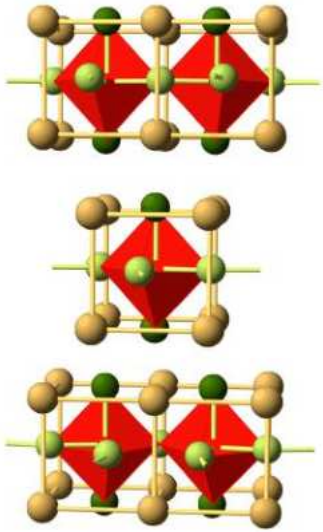
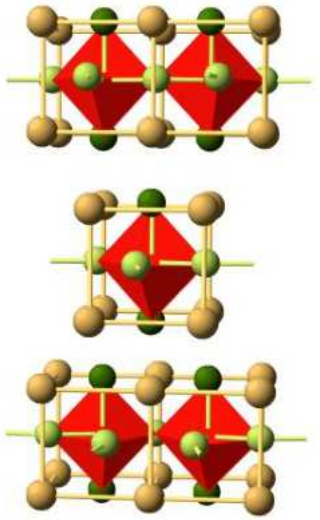
three-band t_{2g} materials

Sr_2RuO_4	Ca_2RuO_4	Sr_2RhO_4	Sr_2IrO_4
			
metal	*insulator*	*metal*	*insulator*
Hund's metal	*orbitally ordered Higgs modes*	*small masses*	*spin-orbit Mott*

organization of the lecture

I. recap: the many-body problem

II. Mott systems, Hubbard model and DMFT

III. DMFT for materials: DFT+DMFT

IV. two paradigmatic examples

PART 1

recap: the many-body problem

the theory of nearly everything

Born-Oppenheimer approximation, non-relativistic

$$\hat{H}_e = -\frac{1}{2}\sum_i \nabla_i^2 + \frac{1}{2}\sum_{i\neq i'} \frac{1}{|\mathbf{r}_i - \mathbf{r}_{i'}|} - \sum_{i,n} \frac{Z_n}{|\mathbf{r}_i - \mathbf{R}_n|} + \frac{1}{2}\sum_{n\neq n'} \frac{Z_n Z_{n'}}{|\mathbf{R}_n - \mathbf{R}_{n'}|}$$

$\mathbf{r}_i$ electrons

$\mathbf{R}_n$ nuclei

$$\hat{H}_e \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = E_\alpha \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$$

solution in four steps

the many-body problem

1 $\hat{H}_e = \sum_i h(\mathbf{r}_i) + \frac{1}{2}\sum_{i,i'} u(\mathbf{r}_i, \mathbf{r}_{i'})$

single-electron two-electron

split Hamiltonian

2 $h(\mathbf{r})\psi_\lambda(\mathbf{r}) = \varepsilon_\lambda \psi_\lambda(\mathbf{r})$

build single-electron complete basis $\{\psi_\lambda(\mathbf{r})\}$

3 $\Phi_{\Lambda=\{\lambda_i\}} = \prod_{i=1,\ldots,N} c^\dagger_{\lambda_i}|0\rangle$

build complete basis of Slater determinants $\{\Phi_\Lambda(\mathbf{r})\}$

4 $\hat{H}_e \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = E_\alpha \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$

$\Psi_\alpha = \sum_\Lambda C^\alpha_\Lambda \, \Phi_\Lambda$ .. and diagonalize...

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bad news: the exact solution is not an option

Hilbert space of Slater determinants
grows exponentially (or faster)

eigenstates are **typically** combinations of
a **very** large number of Slater determinants!

example: 4 determinants per site, 4^{N_s}

$4^2=16$ $4^{10}\sim10^6$ $4^{20}\sim10^{12}$

the non-interacting many-body problem

1 $$\hat{H}_e = \sum_i h(\mathbf{r}_i)$$

single-electron

2 $$h(\mathbf{r})\psi_\lambda(\mathbf{r}) = \varepsilon_\lambda \psi_\lambda(\mathbf{r})$$

build single-electron complete basis $\{\psi_\lambda(\mathbf{r})\}$

3 $$\Phi_{\Lambda=\{\lambda_i\}} = \prod_{i=1,\ldots,N} c^\dagger_{\lambda_i}|0\rangle$$

$\{\Phi_\Lambda(\mathbf{r})\}$

all eigenstates are **single** Slater determinants!
size of Hilbert space irrelevant

$$E = \sum_i \varepsilon_{\lambda_i}$$

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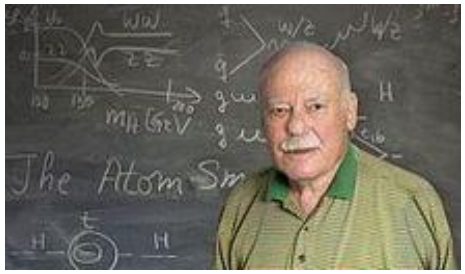
good news: it would be anyway useless



H.J. Lipkin

On the other hand, the exact solution of a many-body problem is really irrelevant since it includes a large mass of information about the system which although measurable in principle is never measured in practice.

[..] An incomplete description of the system is considered to be sufficient if these measurable quantities and their behavior are described correctly.

$$\hat{H}_e \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = E_\alpha \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$$

E. Pavarini and E. Koch, Autumn School on Correlated Electron 2013, Introduction

the many-body problem: ***more is different***

Born-Oppenheimer approximation, non-relativistic

$$\hat{H}_e = -\frac{1}{2}\sum_i \nabla_i^2 + \frac{1}{2}\sum_{i\neq i'} \frac{1}{|\mathbf{r}_i - \mathbf{r}_{i'}|} - \sum_{i,\alpha} \frac{Z_\alpha}{|\mathbf{r}_i - \mathbf{R}_\alpha|} + \frac{1}{2}\sum_{\alpha\neq\alpha'} \frac{Z_\alpha Z_{\alpha'}}{|\mathbf{R}_\alpha - \mathbf{R}_{\alpha'}|}$$

i=electrons

α = nuclei

simple interactions among many particles
can lead to unexpected *emergent* co-operative behavior

Coulomb-induced metal-insulator transition
heavy-Fermions
unconventional superconductivity
spin-charge separation
.....

Philip Warren Anderson

4 August 1972, Volume 177, Number 4047

SCIENCE

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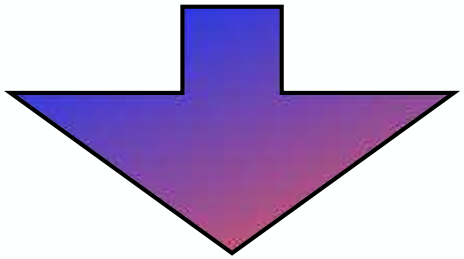
emergent energy scales

general theory & many parameters *{P}*

$$\hat{H}_e = -\frac{1}{2}\sum_i \nabla_i^2 + \frac{1}{2}\sum_{i\neq i'} \frac{1}{|\mathbf{r}_i - \mathbf{r}_{i'}|} - \sum_{i,n} \frac{Z_n}{|\mathbf{r}_i - \mathbf{R}_n|} + \frac{1}{2}\sum_{n\neq n'} \frac{Z_n Z_{n'}}{|\mathbf{R}_n - \mathbf{R}_{n'}|}$$



low energy: **simpler model,** effective elementary entities and fewer effective parameters *p*

but *p*=*p*(*{P}*)

e.g., Hubbard model, Heisenberg model

$$\hat{H}_e \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = E_\alpha \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$$

what can be done then ?

a way out: density-functional theory

1964

PHYSICAL REVIEW VOLUME 136, NUMBER 3B 9 NOVEMEBR 1964

Inhomogeneous Electron Gas*

P. HOHENBERG†
École Normale Superieure, Paris, France
AND
W. KOHN‡
École Normale Superieure, Paris, France and Faculté des Sciences, Orsay, France
and
University of California at San Diego, La Jolla, California
(Received 18 June 1964)

This paper deals with the ground state of an inter...
proved that there exists a universal functional of the ...
pression $E \equiv \int v(\mathbf{r})n(\mathbf{r})d\mathbf{r}+F[n(\mathbf{r})]$ has as its minimum ...
$v(\mathbf{r})$. The functional $F[n(\mathbf{r})]$ is then discussed for ...
(2) $n(\mathbf{r})=\varphi(\mathbf{r}/r_0)$ with φ arbitrary and $r_0\rightarrow\infty$. In bot...
relation energy and linear and higher order electronic ...
also sheds some light on generalized Thomas-Fermi ...
these methods are presented.

INTRODUCTION

DURING the last decade there has been considerable progress in understanding the properties of a homogeneous interacting electron gas.[1] The point of view has been, in general, to regard the electrons as similar to a collection of noninteracting particles with the important additional concept of collective excitations.

On the other hand, there has been in existence since the 1920's a different approach, represented by the Thomas-Fermi method[2] and its refinements, in which the electronic density $n(\mathbf{r})$ plays a central role and in which the system of electrons is pictured more like a classical liquid. This approach has been useful, up to now, for simple though crude descriptions of inhomogeneous systems like atoms and impurities in metals.

Lately there have been also some important advances along this second line of approach, such as the work of Kompaneets and Pavlovskii,[3] Kirzhnits,[4] Lewis,[5] Baraff and Borowitz,[6] Baraff,[7] and DuBois and Kivelson.[8] The present paper represents a contribution in the same area.

1965

PHYSICAL REVIEW VOLUME 140, NUMBER 4A 15 NOVEMBER 1965

Self-Consistent Equations Including Exchange and Correlation Effects*

W. KOHN AND L. J. SHAM
University of California, San Diego, La Jolla, California
(Received 21 June 1965)

From a theory of Hohenberg and Kohn, approximation methods for treating an inhomogeneous system of interacting electrons are developed. These methods are exact for systems of slowly varying or high density. For the ground state, they lead to self-consistent equations analogous to the Hartree and Hartree-Fock equations, respectively. In these equations the exchange and correlation portions of the chemical potential of a uniform electron gas appear as additional effective potentials. (The exchange portion of our effective potential differs from that due to Slater by a factor of $\frac{2}{3}$.) Electronic systems at finite temperatures and in magnetic fields are also treated by similar methods. An appendix deals with a further correction for systems with short-wavelength density oscillations.

I. INTRODUCTION

IN recent years a great deal of attention has been given to the problem of a homogeneous gas of interacting electrons and its properties have been established with a considerable degree of confidence over a wide range of densities. Of course, such a homogeneous gas represents only a mathematical model, since in all real systems (atoms, molecules, solids, etc.) the electronic density is nonuniform.

It is then a matter of interest to see how properties of the homogeneous gas can be utilized in theoretical

In Secs. III and IV, we describe the necessary modifications to deal with the finite-temperature properties and with the spin paramagnetism of an inhomogeneous electron gas.

Of course, the simple methods which are here proposed in general involve errors. These are of two general origins[4]: a too rapid variation of density and, for finite systems, boundary effects. Refinements aimed at reducing the first type of error are briefly discussed in Appendix II.

II. THE GROUND STATE

the *standard model*: density-functional theory

$$\hat{H}_e \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = E_\alpha \Psi_\alpha(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N)$$

$$n_G(\mathbf{r}), \qquad E_G[n(\mathbf{r})], \qquad \ldots$$

(in practice: LDA,GGA,…)

1998: Nobel Prize in Chemistry to Walter Kohn

In my view DFT makes two kinds of contribution to the science of multi-particle quantum systems, including problems of electronic structure of molecules and of condensed matter:

The first is in the area of fundamental *understanding*. Theoretical chemists and physicists, following the path of the Schroedinger equation, have become accustomed to think in a truncated *Hilbert space of single particle orbitals*. The spectacular advances achieved in this way attest to the fruitfulness of this perspective. However, when high accuracy is required, so many Slater determinants are required (in some calculations up to ~ 10^9!) that *comprehension* becomes difficult. DFT provides a complementary perspective. It focuses on quantities in the real, 3-dimensional coordinate space, principally on the electron density $n(r)$ of the groundstate. Other quantities of great interest

(from the Nobel lecture)

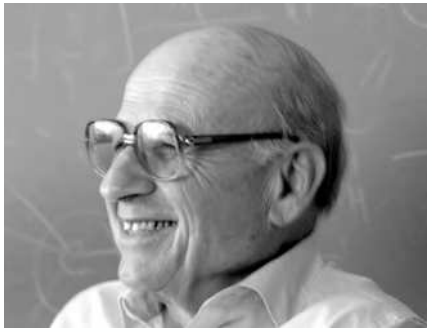
but, how do we calculate the density?

Kohn-Sham **auxiliary** Hamiltonian

$$\hat{h}_e = \sum_i \left[-\frac{1}{2}\nabla_i^2 + v_R(\boldsymbol{r}_i) \right] = \sum_i \hat{h}_e(\boldsymbol{r}_i)$$

$$v_R(\boldsymbol{r}) = -\sum_\alpha \frac{Z_\alpha}{|\boldsymbol{r} - \boldsymbol{R}_\alpha|} + \int d\boldsymbol{r}' \frac{n(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|} + \frac{\delta E_{\rm xc}[n]}{\delta n} = v_{en}(\boldsymbol{r}) + v_H(\boldsymbol{r}) + v_{xc}(\boldsymbol{r})$$

Hartree **exchange-correlation**



Walter Kohn

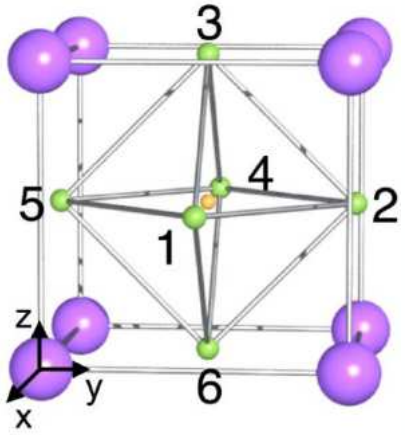
a non-interacting problem
with the **same electron density** of
the original many-electron problem

Nobel Prize in Chemistry (1998)

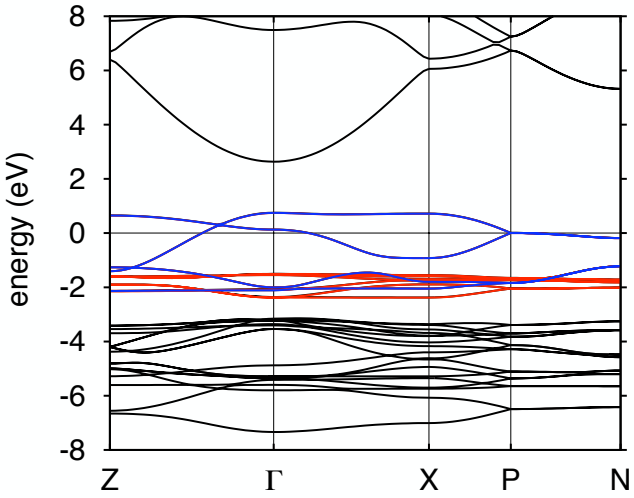
unexpected successes of DFT

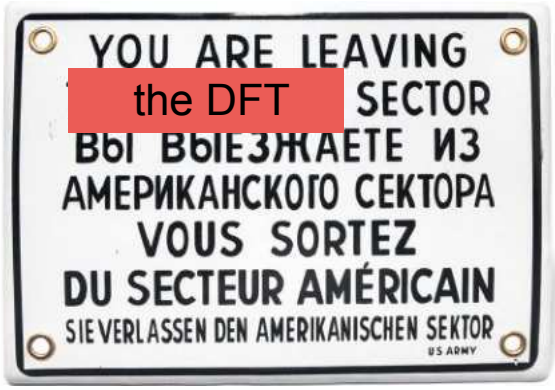
Kohn-Sham band-structures work well as

approximated elementary excitations



$KCuF_3$





material trends, prediction

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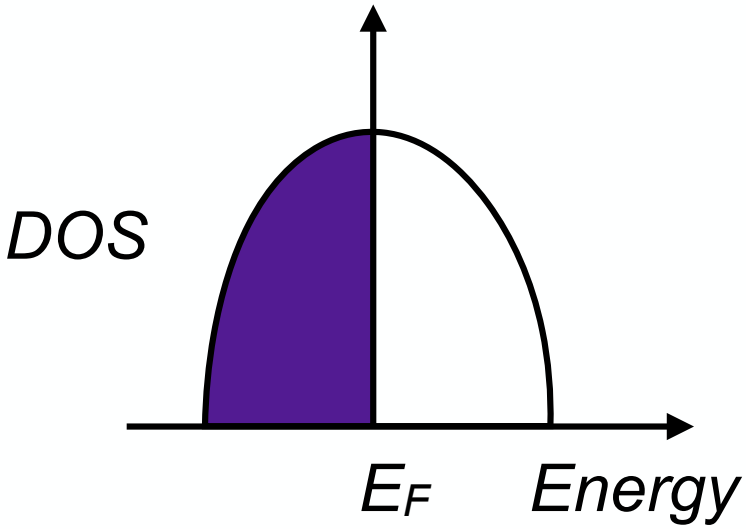
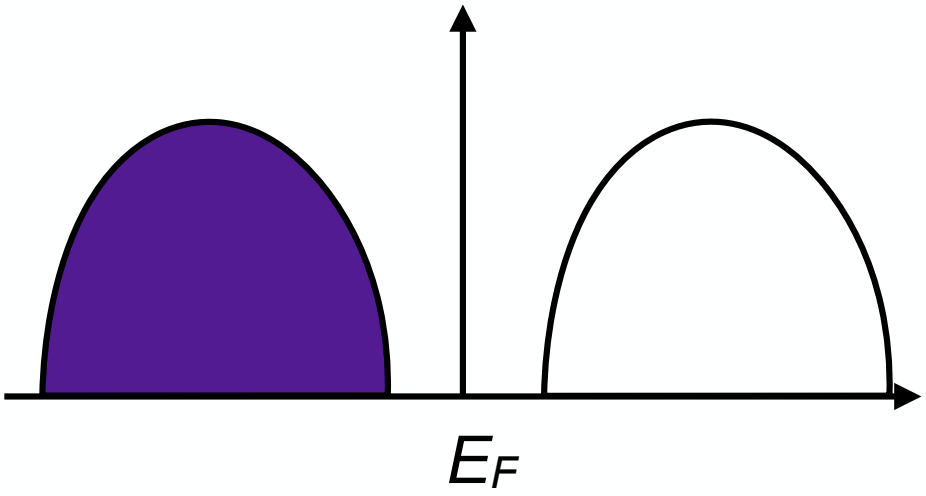
remarkably, it works well in many cases!





metal
(e.g., Cu, Na)

band partially filled

insulator
(e.g. diamond)

band full

why? mean-field-**like** model or Fermi-liquid theory

$$\hat{H}_e = \sum_i h(\mathbf{r}_i) + \frac{1}{2}\sum_{i,i'} u(\mathbf{r}_i, \mathbf{r}_{i'})$$

$$\hat{H}_e \sim \sum_i \tilde{h}(\mathbf{r}_i)$$

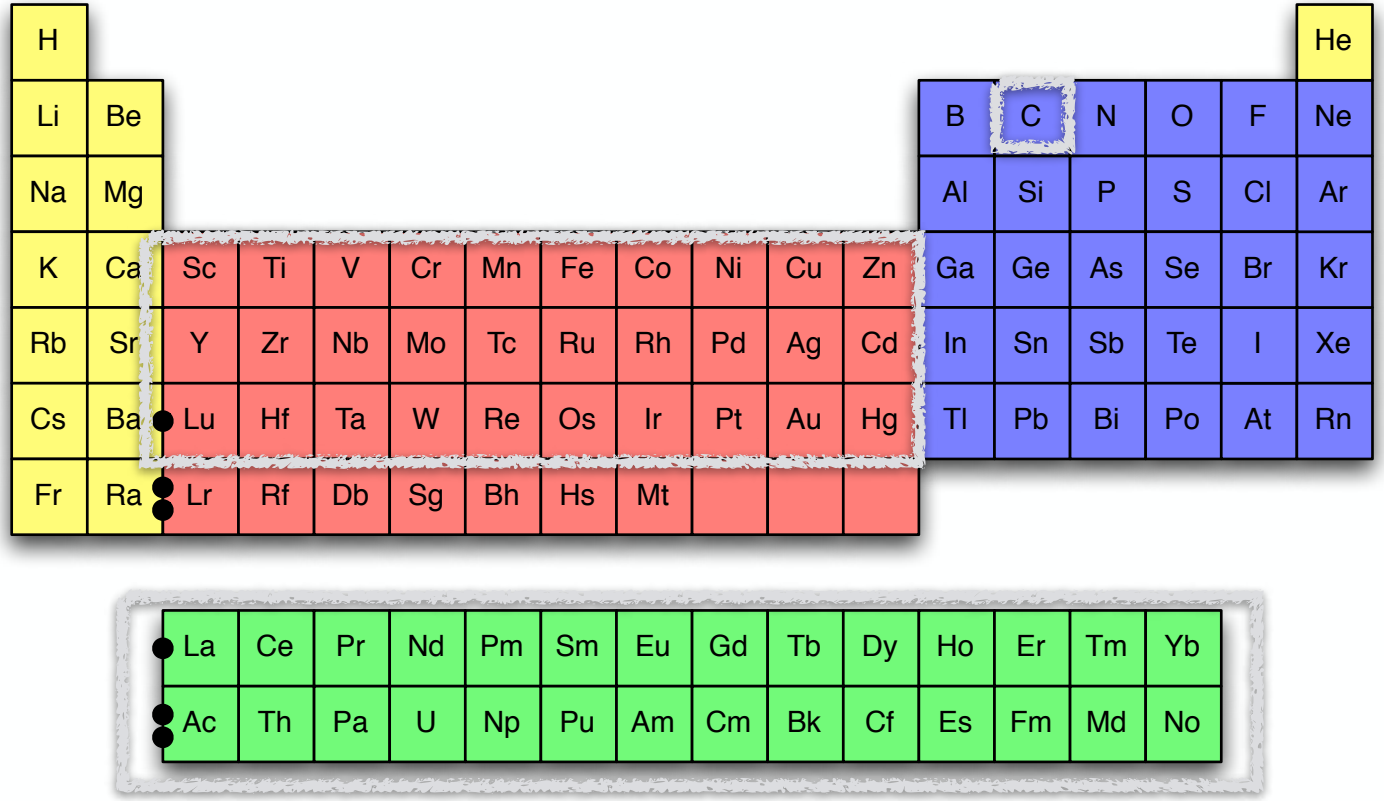
Kohn Sham

- correct symmetry
- average long-range Coulomb effect
- very good ground-state density
- sum rules

what are correlated materials?

systems for which *the single-electron picture* fails *qualitatively*

atomic-like physics survives in the solid



atomic-like = local Coulomb repulsion

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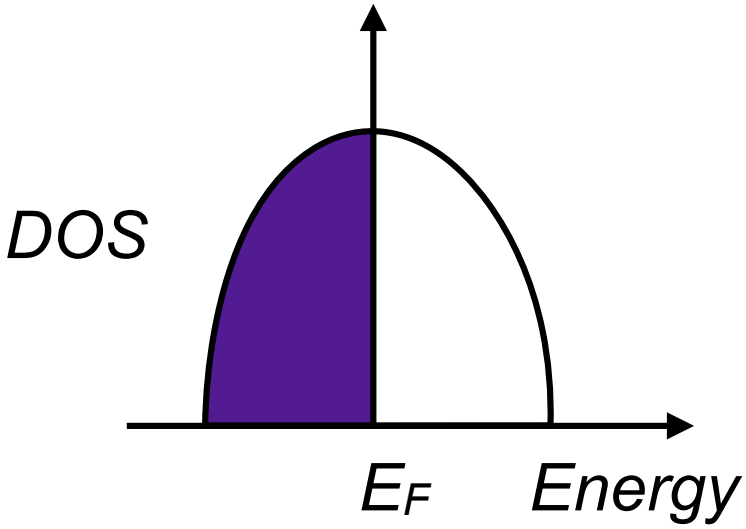
deep problems: Mott systems

single-electron picture:

band partially filled, it should be a metal

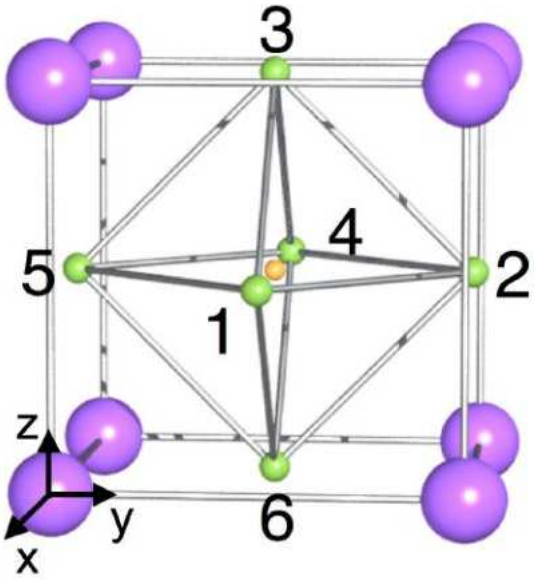


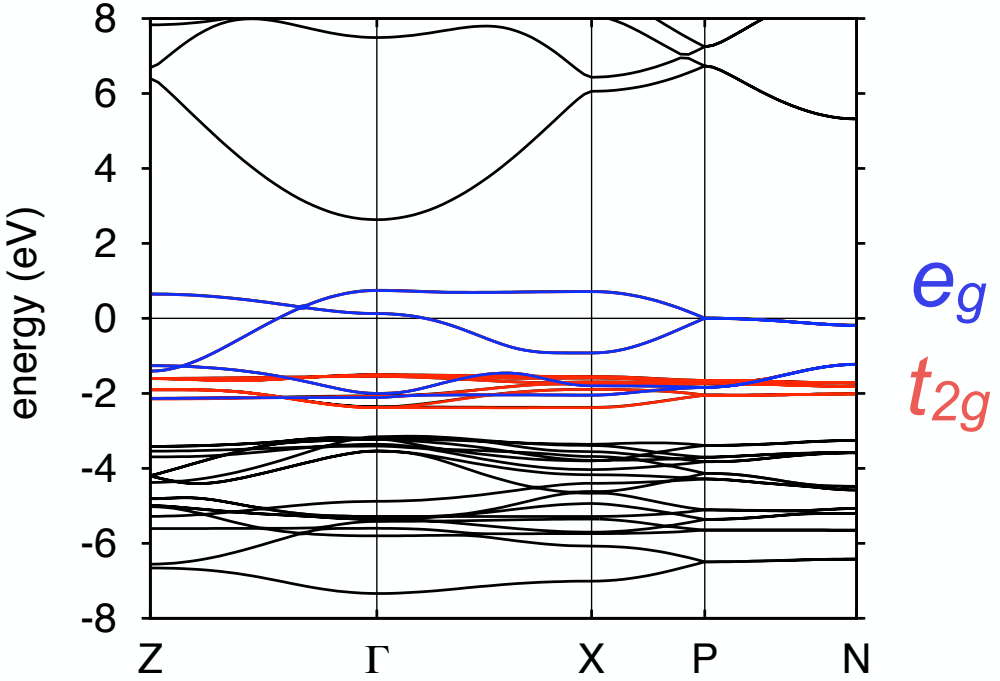
*but experimentally there is an **excitation** gap*

*that can be explained by **local** electron-electron repulsion*

deep problems: Mott systems

$KCuF_3$





Experiments: insulator. Above 40 K a **paramagnetic** insulator

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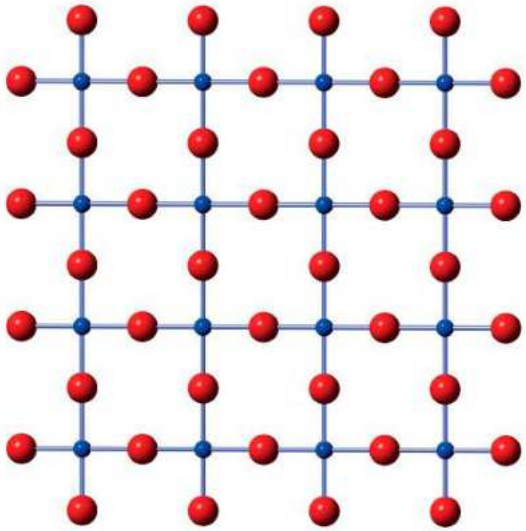
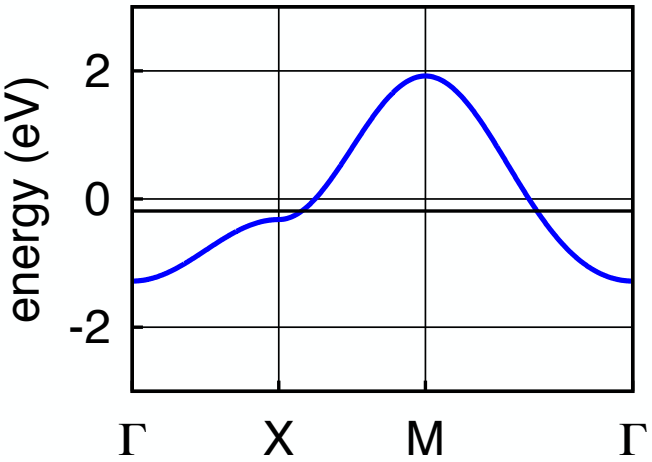
high-T_c superconducting cuprates

CuO_2 planes

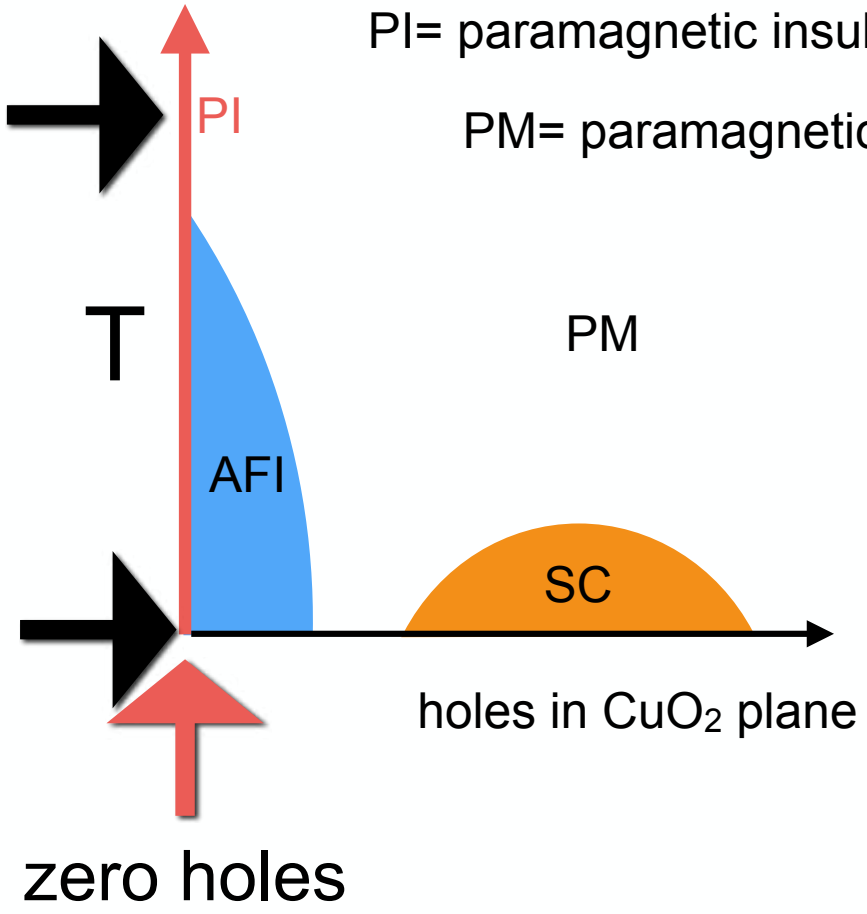




AFI= antiferromagnetic insulator

PI= paramagnetic insulator

PM= paramagnetic metal



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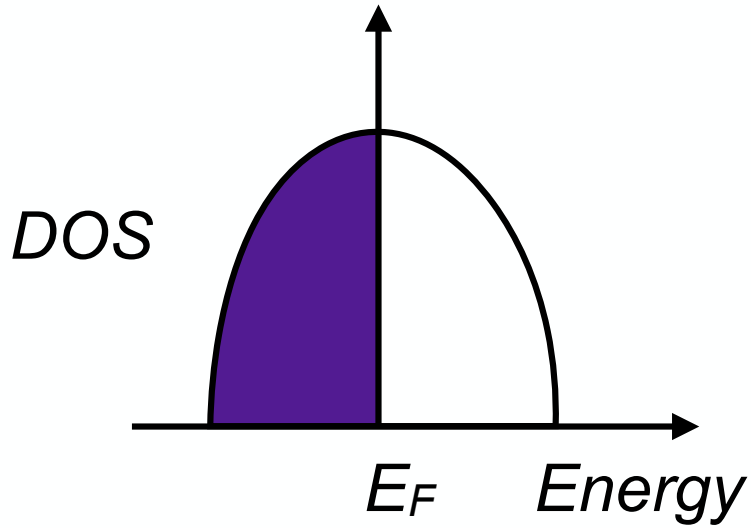
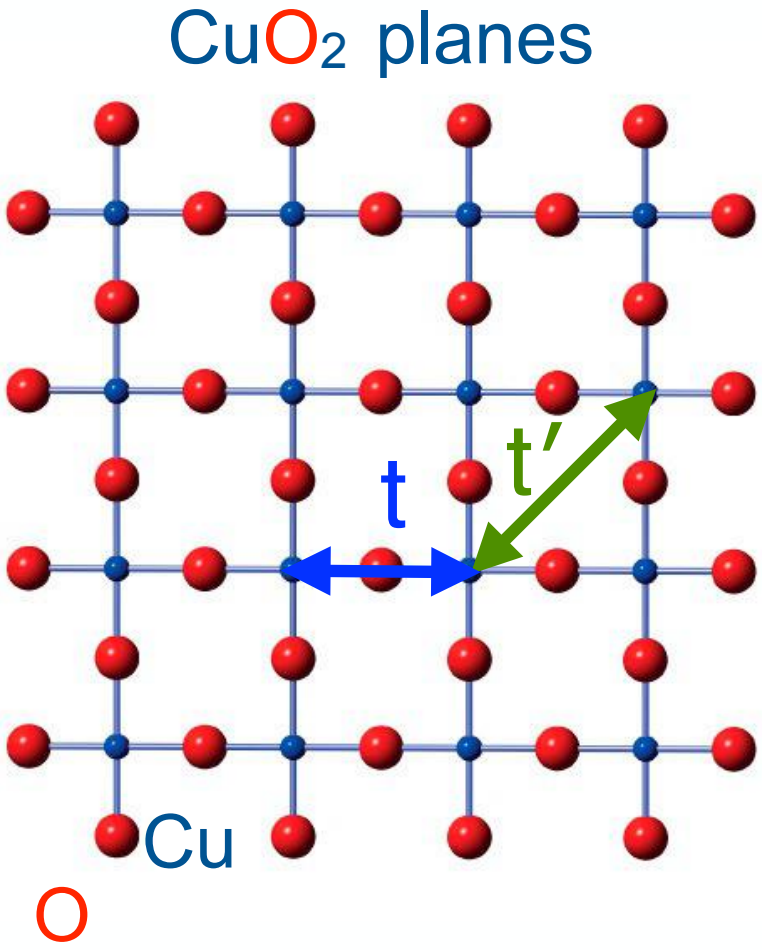
why *does the single-electron picture fails ?*

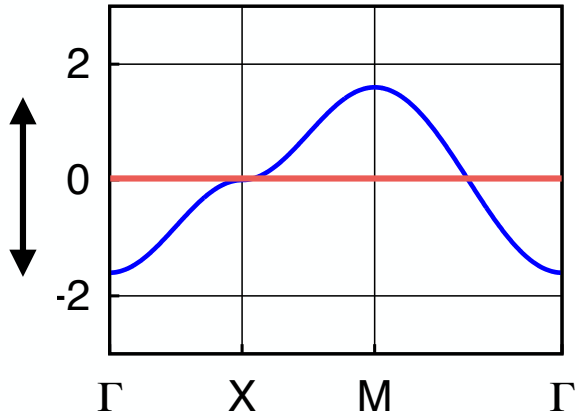


cuprates: tight-biding model



Kramers degeneracy:
Fermi level in the center of band

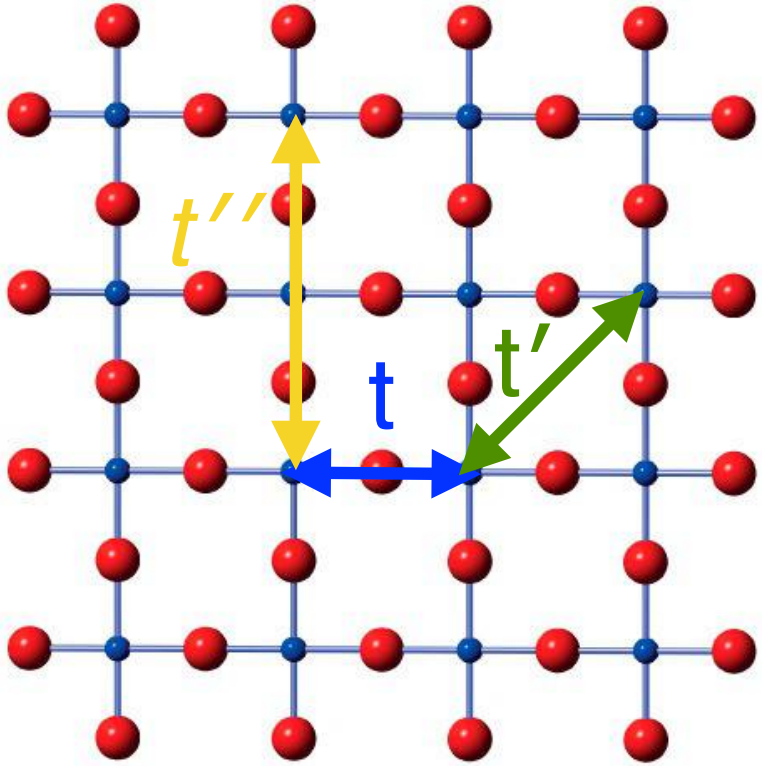
W= 8t

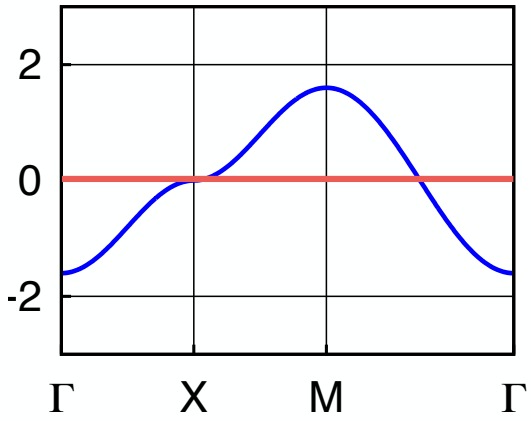


$$\varepsilon_{\boldsymbol{k}} = -2t[\cos k_x + \cos k_y]$$

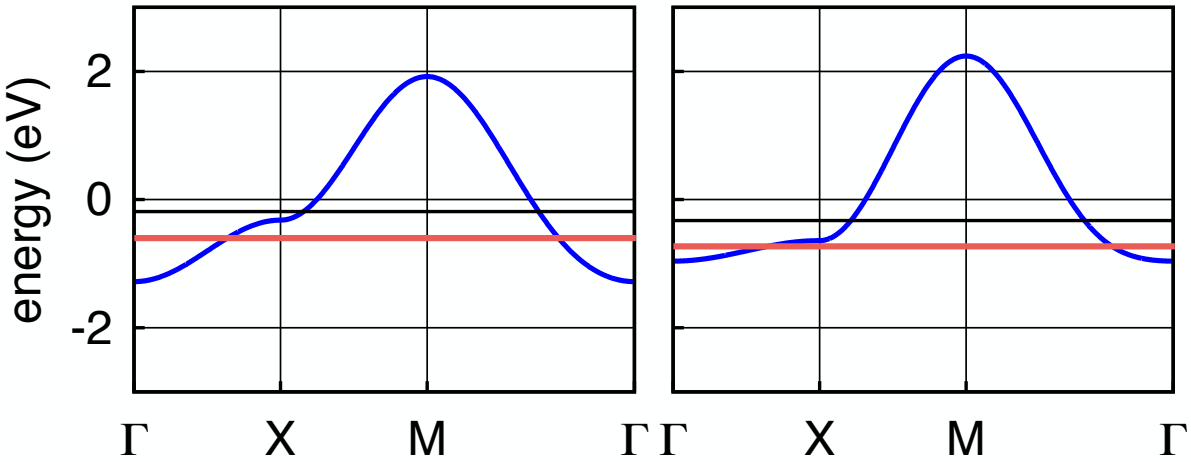
$$\hat{H} = \varepsilon_d \sum_i \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma} - t \sum_{\langle ii' \rangle} \sum_\sigma c^\dagger_{i\sigma} c_{i'\sigma} = \varepsilon_d \sum_k \sum_\sigma c^\dagger_{k\sigma} c_{k\sigma} + \sum_k \sum_\sigma \varepsilon_{\mathbf{k}} c^\dagger_{k\sigma} c_{k'\sigma}$$

modify potential but keep symmetry





t′/t= =0.2 t′/t= =0.4



but since Kramers degeneracy stays we always have a metal..

"symmetry protected"

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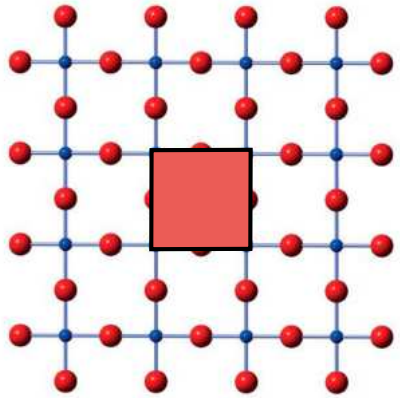
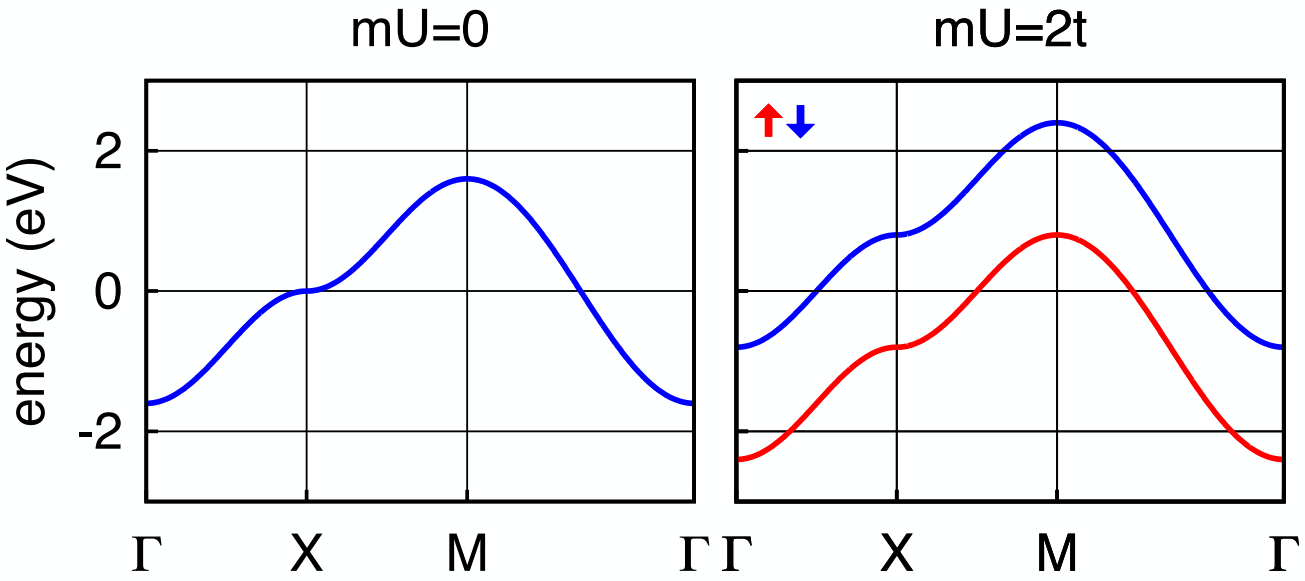
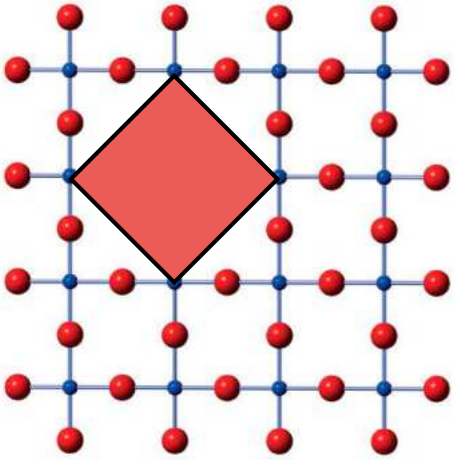
to open a gap we must lower the symmetry

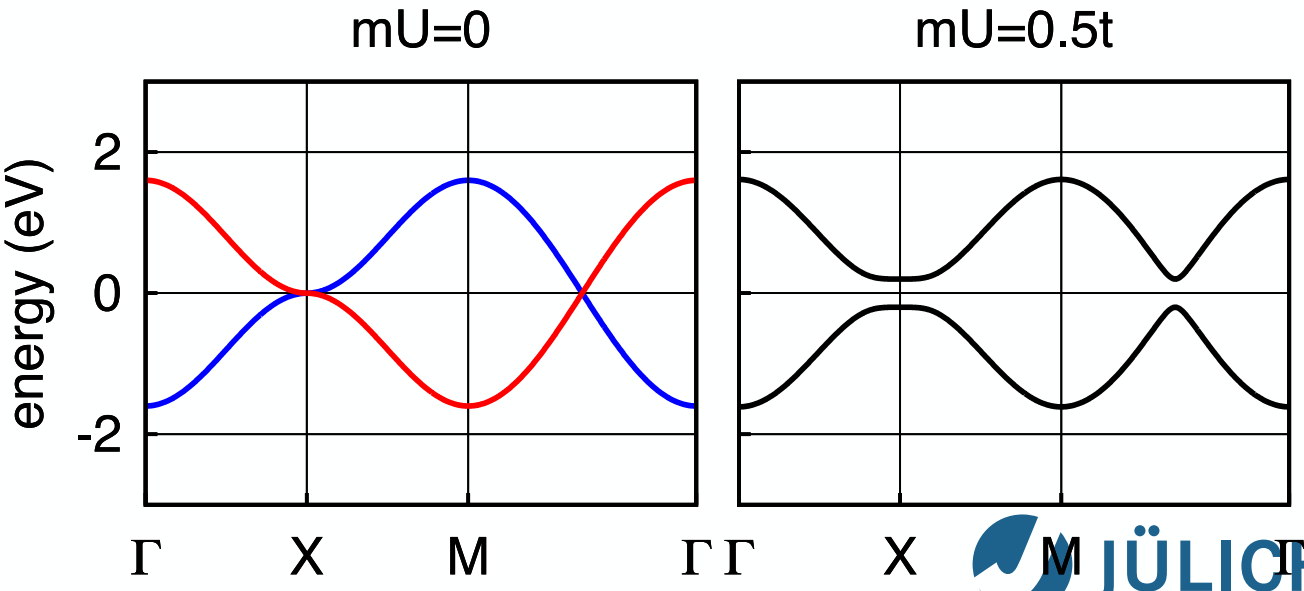
ferro





antiferro





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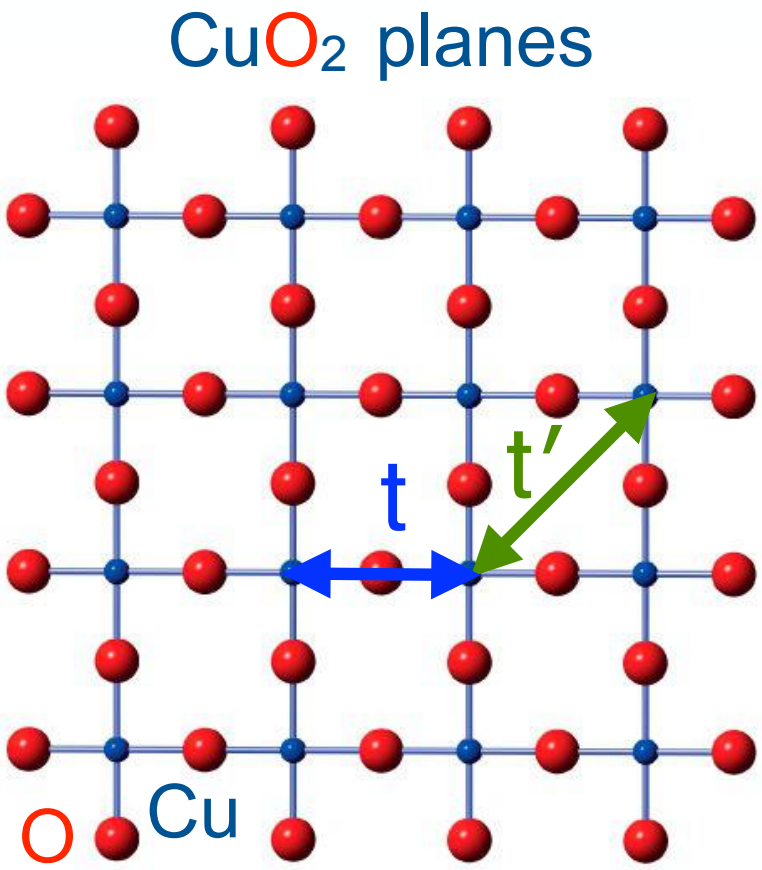
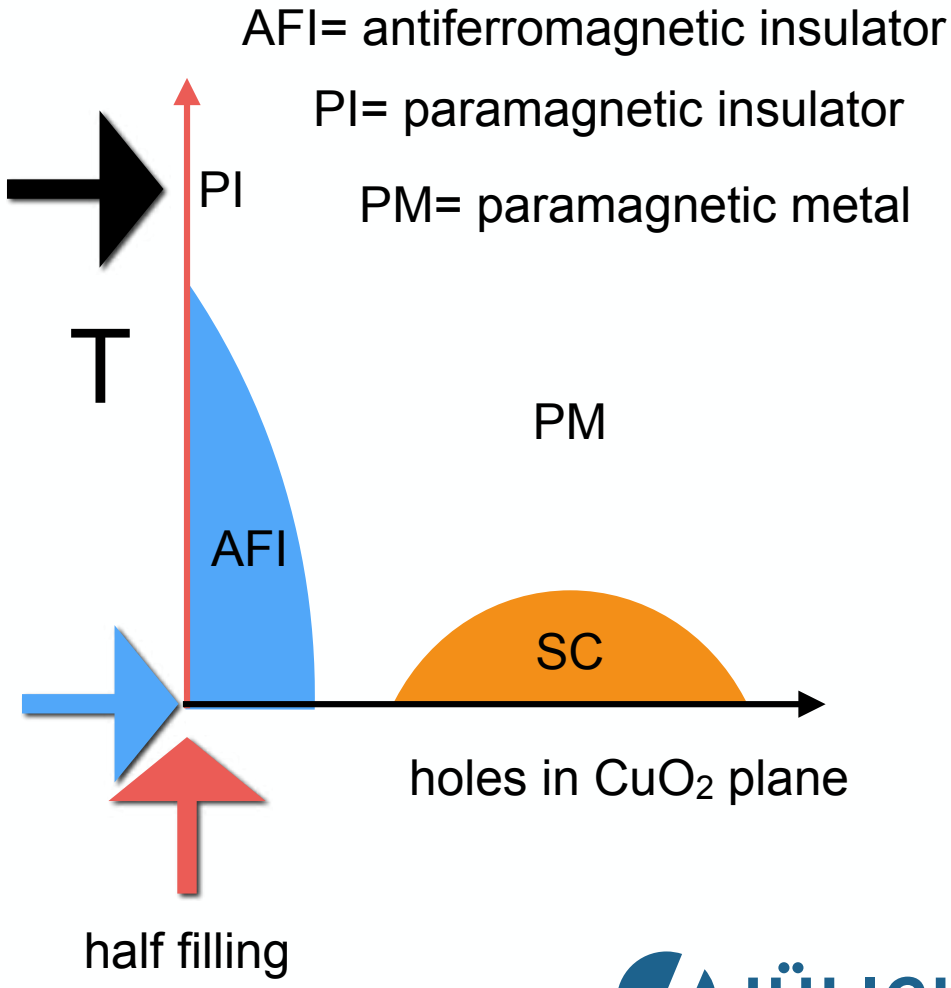
high-T_c superconducting cuprates

phase diagram





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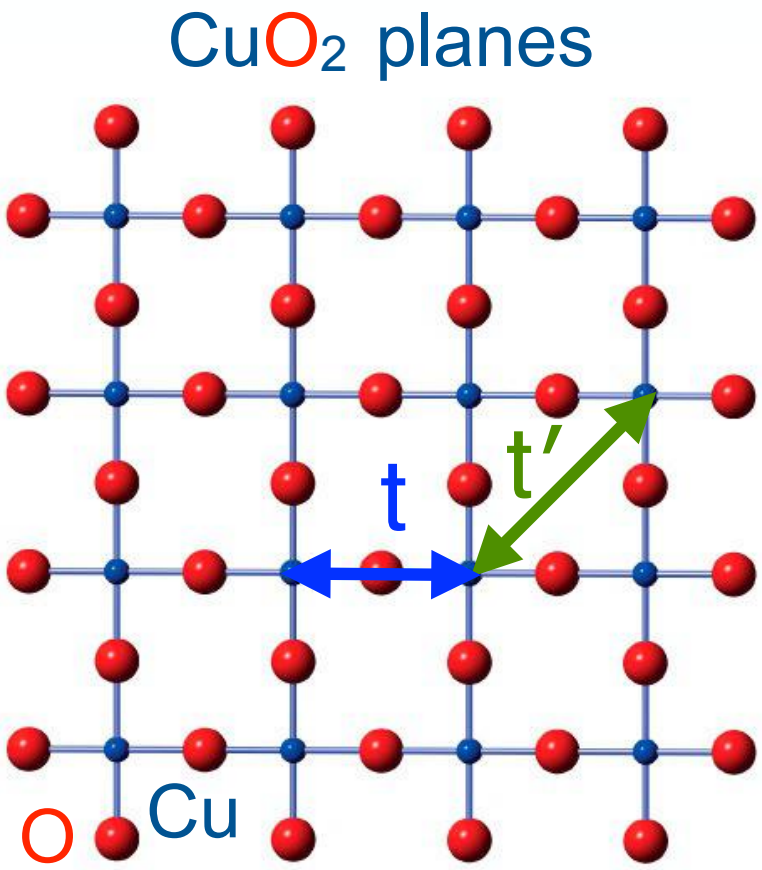
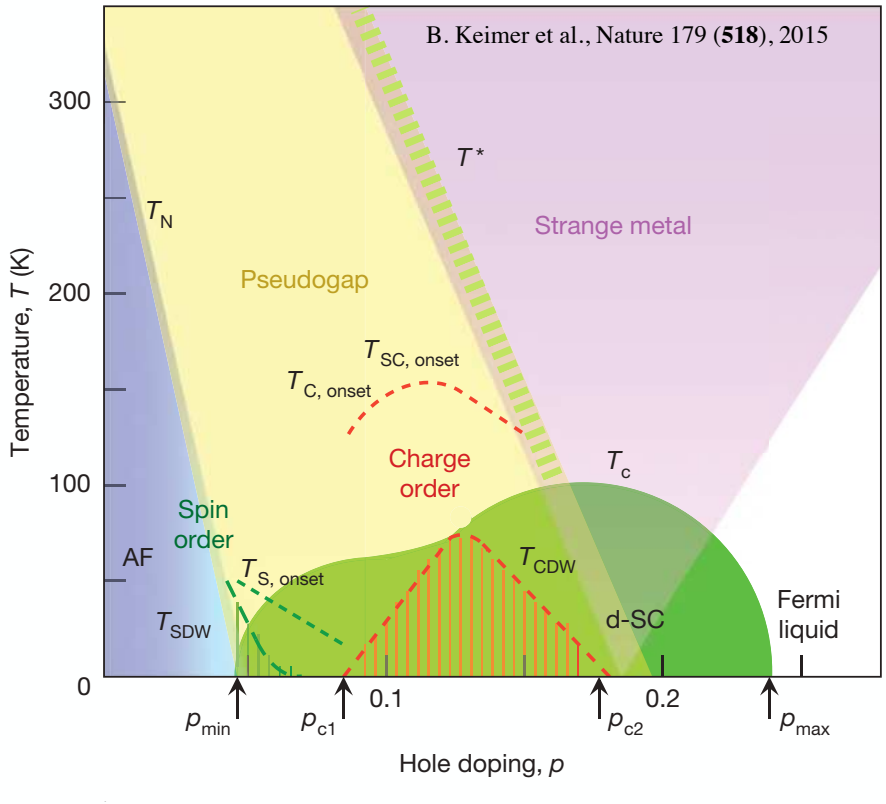
high-T_c superconducting cuprates

phase diagram





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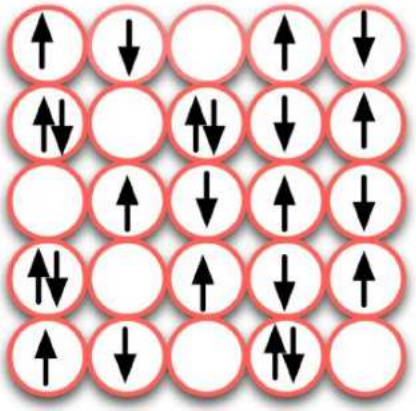
PART 2

Mott transition, Hubbard model and DMFT

the paradigmatic model



Hubbard model

half filling

$$\hat{H} = \varepsilon_d \sum_i \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma} - t \sum_{\langle ii' \rangle} \sum_\sigma c^\dagger_{i\sigma} c_{i'\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

U/W=0:
metal

$m^* \to \infty$

$\Delta E \to U - W$

W/U=0:
insulator

metal with heavy quasiparticles (large masses)

insulator with increasing gap

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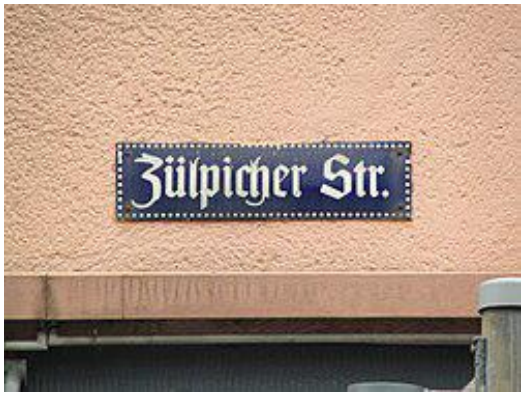
effective masses

2 October, speed: 50 m ~ 30 seconds



(photo from wikipedia)

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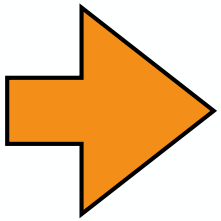
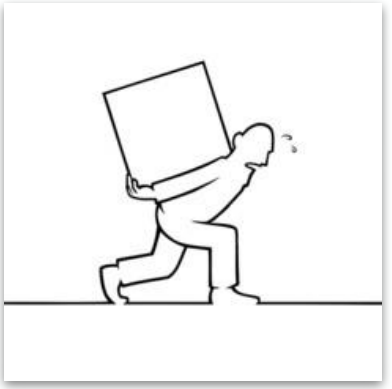
effective masses

Rosenmontag, speed: 50 m ~ 30 minutes

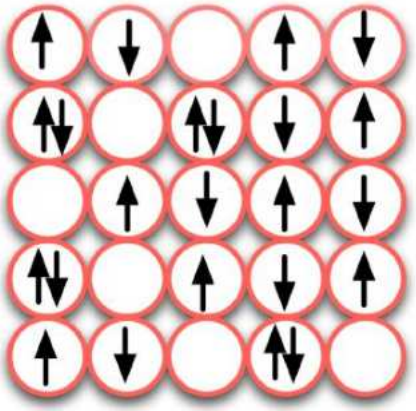
.. but also on a normal day if carrying a very big weight





(photo from wikipedia)

the paradigmatic model



Hubbard model

half filling

$$\hat{H} = \varepsilon_d \sum_i \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma} - t \sum_{\langle ii' \rangle} \sum_\sigma c^\dagger_{i\sigma} c_{i'\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

U/W=0:
metal

$m^* \to \infty$

$\Delta E \to U - W$

W/U=0:
insulator

metal with heavy quasiparticles (large masses)

insulator with increasing gap

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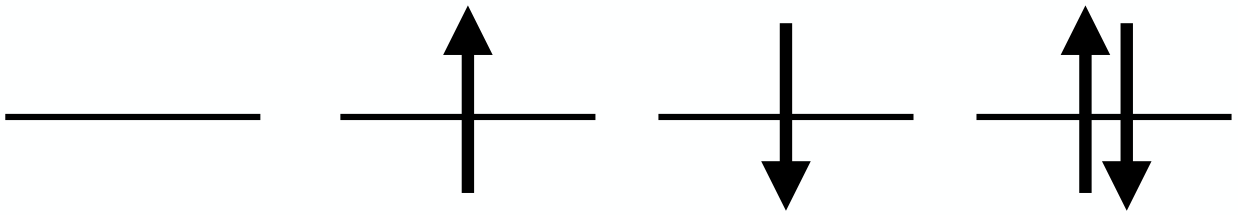
bad news: the exact solution is not an option

Hilbert space of Slater determinants
grows exponentially (or faster)

example: 4 determinants per site, 4^{Ns}

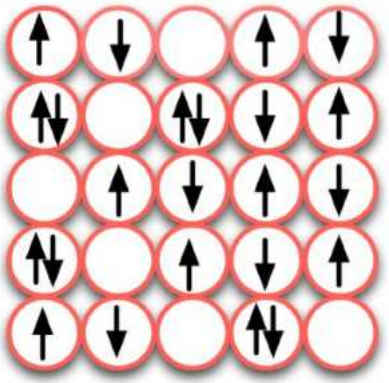
$4^2=16$ $4^{10}\sim10^6$ $4^{20}\sim10^{12}$



1989-1992: dynamical mean-field theory

Hubbard model

$$\hat{H} = \varepsilon_d \sum_i \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma} - t \sum_{\langle ii' \rangle} \sum_\sigma c^\dagger_{i\sigma} c_{i'\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

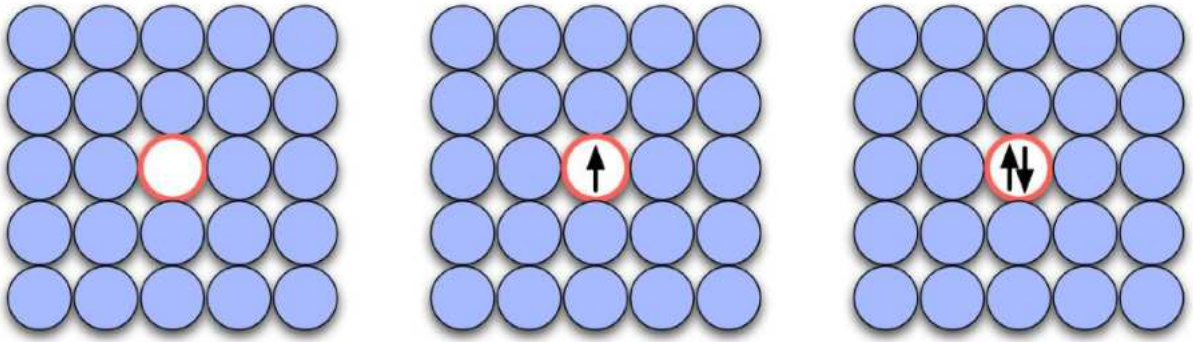


$G^{i,j}$

H^{LDA}

$U^{i,i}$

self-consistent
quantum-impurity model



$$\mathcal{G}^{-1} = G^{-1} + \Sigma$$

$$G = G^{i,i}$$

k-independent self-energy

exact in the infinite coordination number limit

Metzner and Vollhardt, PRL **62**, 324 (1989); Georges and Kotliar, PRB **45**, 6479 (1992).

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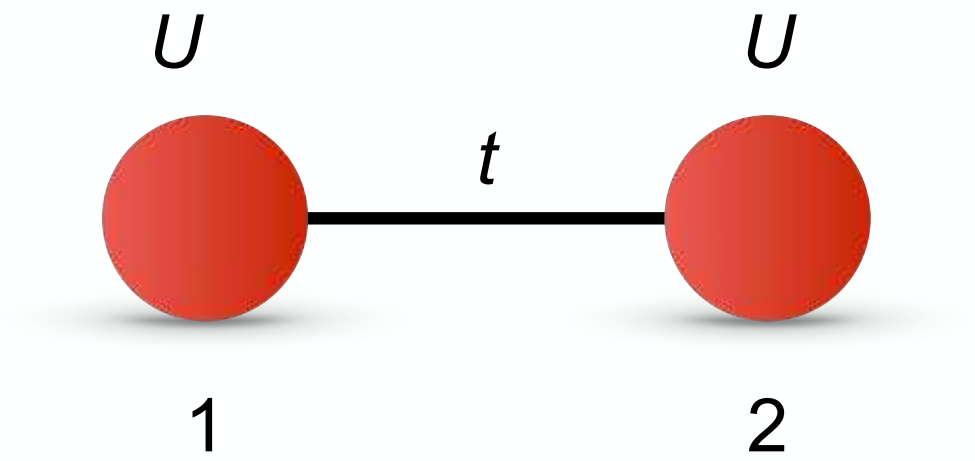
how does it work?

example: Hubbard dimer

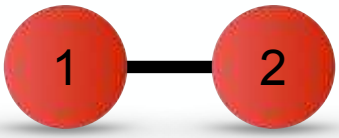
$$\hat{H} = \varepsilon_d \sum_{i\sigma} \hat{n}_{i\sigma} - t \sum_{\sigma} \left(c^{\dagger}_{1\sigma} c_{2\sigma} + c^{\dagger}_{2\sigma} c_{1\sigma} \right) + U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$



half filling : N=2

this is a **toy** model: coordination number is **one**

all analytic results are in lecture notes of Autumn School

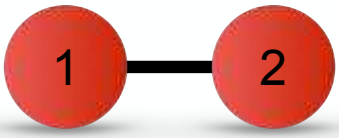


finite *t*: exact diagonalization

half filling (N=2)

$\lvert 2,S,S_z\rangle_\alpha$	$E_\alpha(2,S)$	$d_\alpha(2,S)$
$\lvert 2,0,0\rangle_+ = b_1\lvert 2,0,0\rangle_0 - \frac{b_2}{\sqrt{2}}\left(\lvert 2,0,0\rangle_1 + \lvert 2,0,0\rangle_2\right)$	$2\varepsilon_d + \frac{U}{2} + \frac{1}{4}\left(U + 2\Delta(t,\frac{U}{2})\right)$	1
$\lvert 2,0,0\rangle_o = \frac{1}{\sqrt{2}}\left(\lvert 2,0,0\rangle_1 - \lvert 2,0,0\rangle_2\right)$	$2\varepsilon_d + U$	1
$\lvert 2,1,m\rangle_o = \lvert 2,1,m\rangle$	$2\varepsilon_d + \frac{U}{2}$	3
$\lvert 2,0,0\rangle_- = b_2\lvert 2,0,0\rangle_0 + \frac{b_1}{\sqrt{2}}\left(\lvert 2,0,0\rangle_1 + \lvert 2,0,0\rangle_2\right)$	$2\varepsilon_d + \frac{U}{2} + \frac{1}{4}\left(U - 2\Delta(t,\frac{U}{2})\right)$	1

$$\Delta(t,U) = \sqrt{U^2 + 16t^2}$$

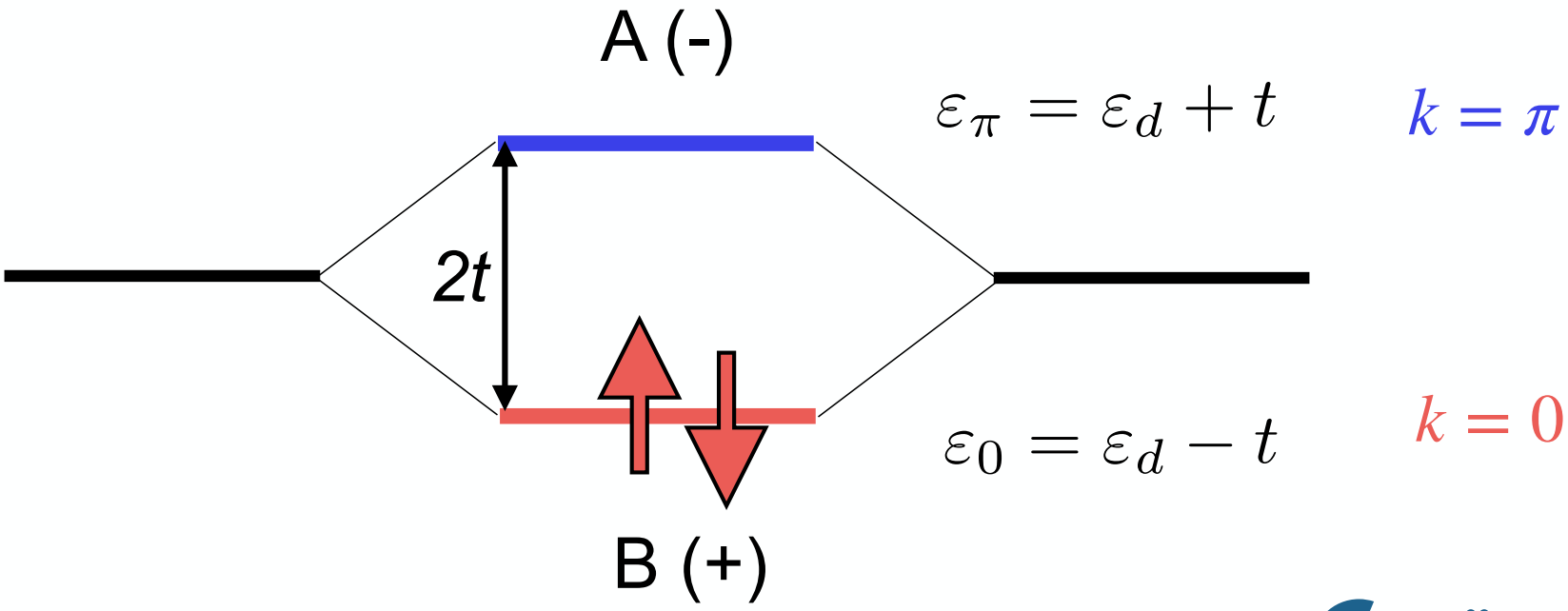


U=0: bonding and antibonding

change from **site** to ***k*** representation

$$c_{k\sigma} = \frac{1}{\sqrt{2}}\left(c_{1\uparrow} \mp c_{2\uparrow}\right)$$

$$\varepsilon_k = \varepsilon_d - t\cos(k)$$



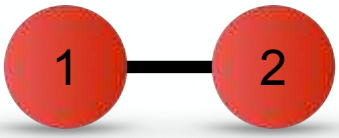
local Green function

$i\nu_n \longrightarrow \omega + i\delta$ Lehmann representation

$$G_{ii,\sigma}(i\nu_n) = \frac{1}{Z}\sum_{nn'N} e^{-\beta(E_n(N)-\mu N)}\left[\frac{|\langle n'N-1|c_{i\sigma}|nN\rangle|^2}{i\nu_n - [E_n(N) - E_{n'}(N-1) - \mu]}\right.$$
$$\left. + \frac{|\langle n'N+1|c^\dagger_{i\sigma}|nN\rangle|^2}{i\nu_n - [E_{n'}(N+1) - E_n(N) - \mu]}\right],$$

we need *all N, N+1* and *N-1* states

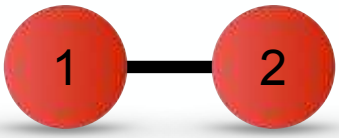
N= number of electrons



finite *t*: exact diagonalization N=1,3

$\lvert 1, S, S_z\rangle_\alpha$	$E_\alpha(1,S)$	$d_\alpha(1,S)$
$\lvert 1, 1/2, \sigma\rangle_+ = \frac{1}{\sqrt{2}}\left(\lvert 1, 1/2, \sigma\rangle_1 - \lvert 1, 1/2, \sigma\rangle_2\right)$	$\varepsilon_d + t$	2
$\lvert 1, 1/2, \sigma\rangle_- = \frac{1}{\sqrt{2}}\left(\lvert 1, 1/2, \sigma\rangle_1 + \lvert 1, 1/2, \sigma\rangle_2\right)$	$\varepsilon_d - t$	2

$\lvert 3, S, S_z\rangle_\alpha$	$E_\alpha(3)$	$d_\alpha(3,S)$
$\lvert 3, 1/2, \sigma\rangle_+ = \frac{1}{\sqrt{2}}\left(\lvert 1, 1/2, \sigma\rangle_1 + \lvert 1, 1/2, \sigma\rangle_2\right)$	$3\varepsilon_d + U + t$	2
$\lvert 3, 1/2, \sigma\rangle_- = \frac{1}{\sqrt{2}}\left(\lvert 1, 1/2, \sigma\rangle_1 - \lvert 1, 1/2, \sigma\rangle_2\right)$	$3\varepsilon_d + U - t$	2



the local Green function

$i\nu_n \longrightarrow \omega + i\delta$ Lehmann representation

$d^1 \rightarrow d^0$

$$|\langle 1|c_\sigma|2\rangle|^2$$

$$\mu = \varepsilon_d + \frac{U}{2}$$

$$G^{\sigma}_{i,i}(i\nu_n) = \frac{1}{4}\left(\frac{1 + w(t,U)}{i\nu_n - (E_0(2) - \varepsilon_d + t - \mu)} + \frac{1 - w(t,U)}{i\nu_n - (E_0(2) - \varepsilon_d - t - \mu)} \right.$$

E(2)-E(1)

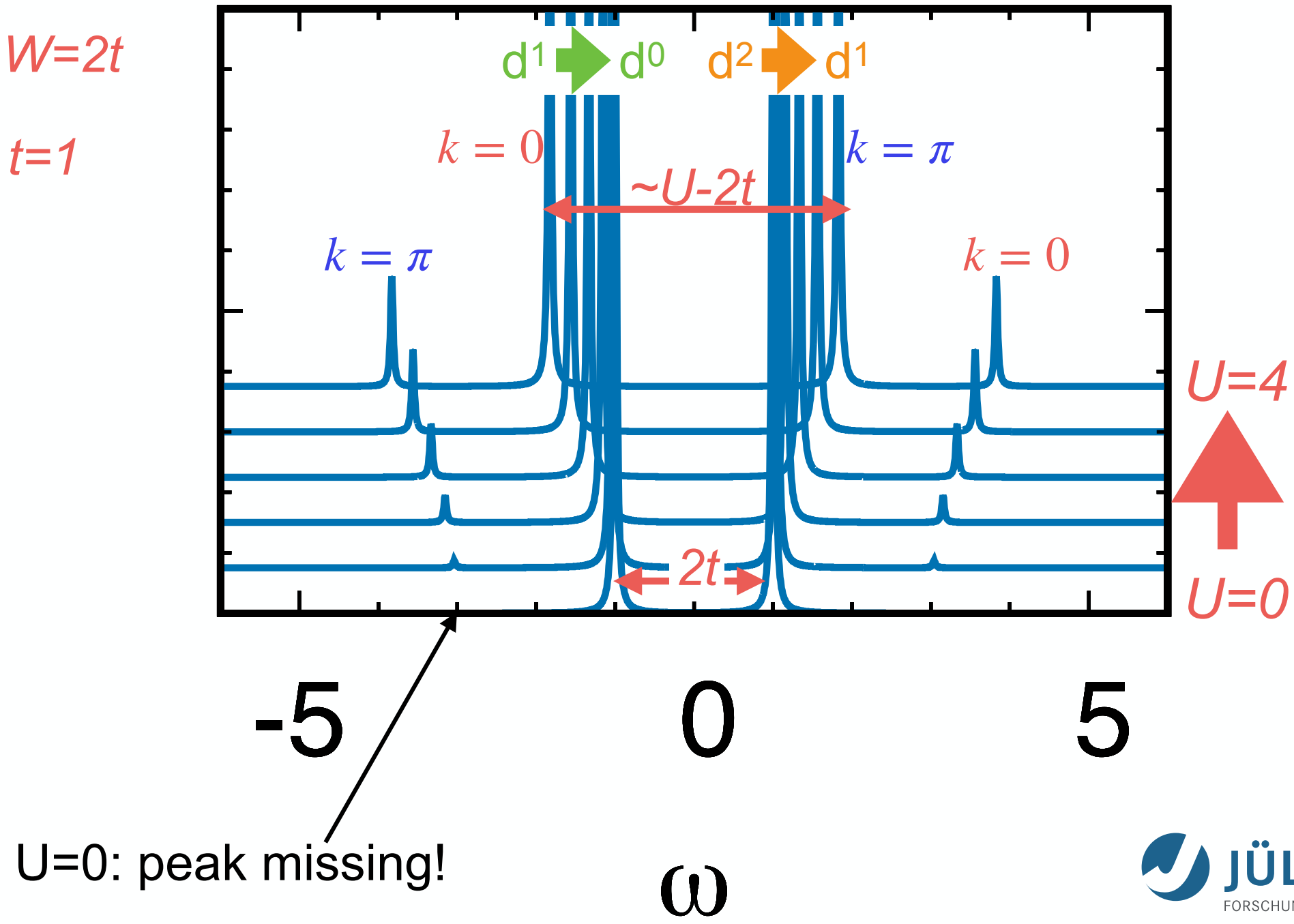
$$|\langle 3|c^\dagger_\sigma|2\rangle|^2$$

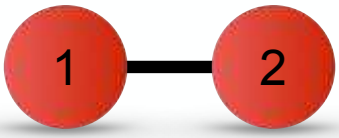
$$\left. + \frac{1 - w(t,U)}{i\nu_n - (-E_0(2) + U + 3\varepsilon_d + t - \mu)} + \frac{1 + w(t,U)}{i\nu_n - (-E_0(2) + U + 3\varepsilon_d - t - \mu)} \right)$$

E(3)-E(2)

$d^2 \rightarrow d^1$

the exact local spectral function





the local Green function

$i\nu_n \longrightarrow \omega + i\delta$ Lehmann representation

$$\mu = \varepsilon_d + \frac{U}{2}$$

d^1 ➡ d^0

$$G^{\sigma}_{i,i}(i\nu_n) = \frac{1}{4}\Bigg(\underbrace{\frac{1 + w(t,U)}{i\nu_n - (E_0(2) - \varepsilon_d + t - \mu)}}_{k=0} + \underbrace{\frac{1 - w(t,U)}{i\nu_n - (E_0(2) - \varepsilon_d - t - \mu)}}_{k=\pi}$$

$$+ \underbrace{\frac{1 - w(t,U)}{i\nu_n - (-E_0(2) + U + 3\varepsilon_d + t - \mu)}}_{k=0} + \underbrace{\frac{1 + w(t,U)}{i\nu_n - (-E_0(2) + U + 3\varepsilon_d - t - \mu)}}_{k=\pi} \Bigg),$$

d^2 ➡ d^1

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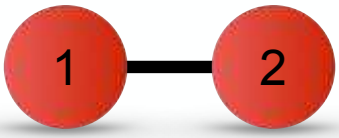


self-energy: local and non-local

$$G^{\sigma}_{11}(i\nu_n) = \frac{1}{2}\sum_k \frac{1}{i\nu_n - (\varepsilon_k + \Sigma^{\sigma}(k, i\nu_n) - \mu)}$$

$$\varepsilon_k = \varepsilon_d - t\cos(k)$$

Σ_l

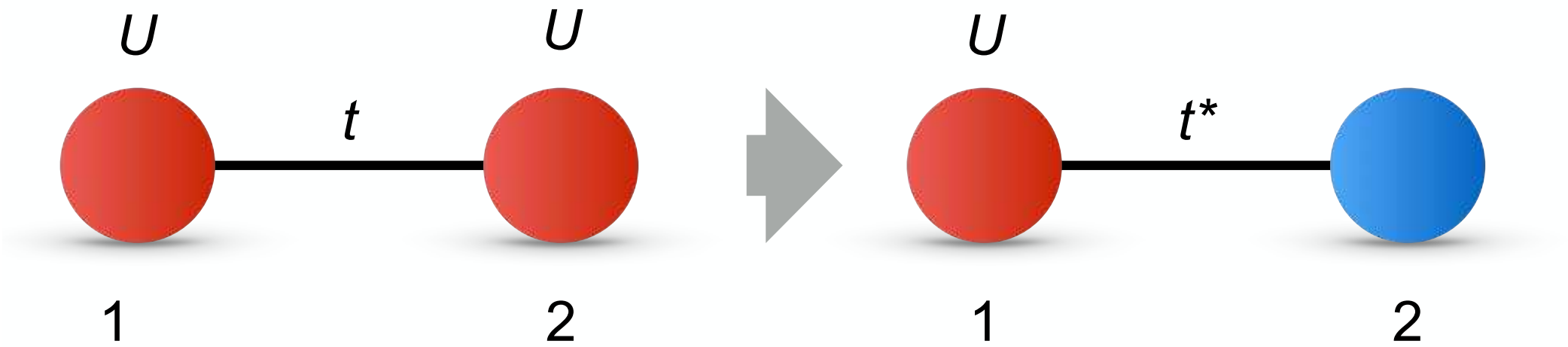
$$\Sigma^{\sigma}_l(i\nu_n) = \frac{1}{2}\Big(\Sigma^{\sigma}(\pi, i\nu_n) + \Sigma^{\sigma}(0, i\nu_n)\Big) = \frac{U}{2} + \frac{U^2}{4}\,\frac{i\nu_n + \mu - \varepsilon_d - \frac{U}{2}}{(i\nu_n + \mu - \varepsilon_d - \frac{U}{2})^2 - (3t)^2}$$

$\Delta\Sigma_l$

$$\Delta\Sigma^{\sigma}_l(i\nu_n) = \frac{1}{2}\Big(\Sigma^{\sigma}(\pi, i\nu_n) - \Sigma^{\sigma}(0, i\nu_n)\Big) = \frac{U^2}{4}\,\frac{3t}{(i\nu_n + \mu - \varepsilon_d - \frac{U}{2})^2 - (3t)^2}$$

map to a quantum impurity model

the Anderson molecule



$$\hat{H}^{\mathrm{A}} = \varepsilon_s \sum_{\sigma} \hat{n}_{s\sigma} - t^* \sum_{\sigma} \left(c^{\dagger}_{d\sigma} c_{s\sigma} + c^{\dagger}_{s\sigma} c_{d\sigma} \right) + \varepsilon_d \sum_{\sigma} \hat{n}_{d\sigma} + U \hat{n}_{d\uparrow} \hat{n}_{d}$$

~ same local self-energy?

find value of ε_s *solution: $\varepsilon_s = \varepsilon_d + U/2 = \mu$, $t^* = t$*

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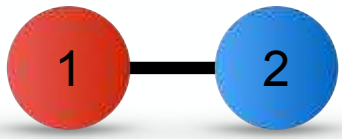
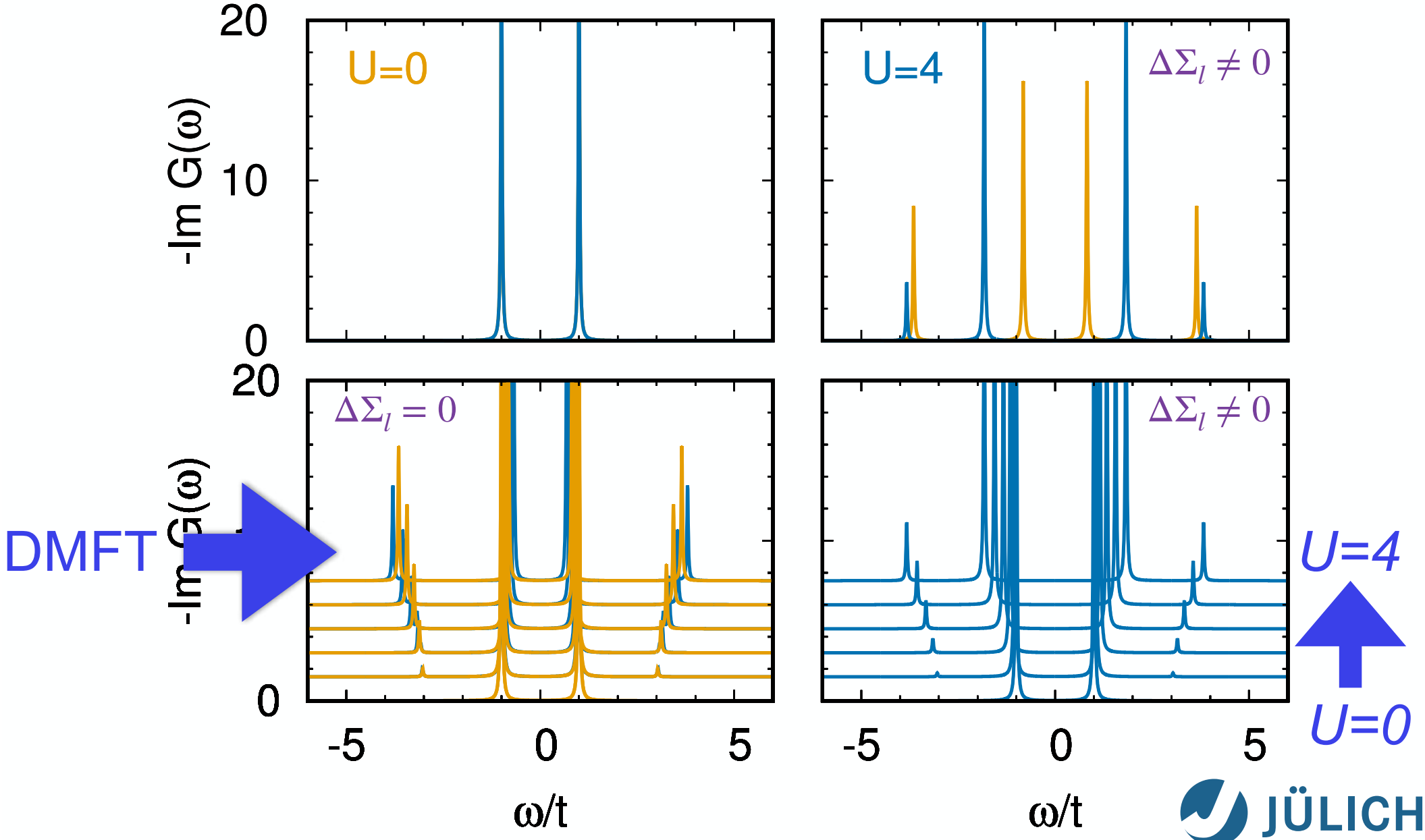


DMFT for the Hubbard dimer



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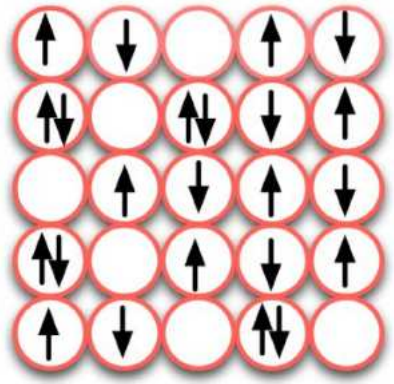
DMFT for the one-band Hubbard model

$$\hat{H} = \varepsilon_d \sum_i \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma} - t \sum_{\langle ii' \rangle} \sum_\sigma c^\dagger_{i\sigma} c_{i'\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

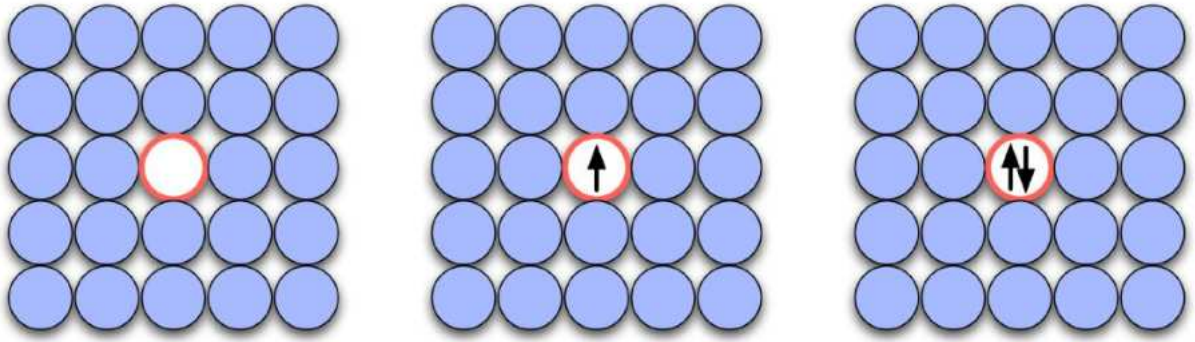


$G^{i,j}$

H^{LDA}

$U^{i,i}$

self-consistent
quantum-impurity model



$$\mathcal{G}^{-1} = G^{-1} + \Sigma$$

$$G = G^{i,i}$$

QIP: QMC, NRG, DMRG, ED, Lanczos…

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dynamical mean-field theory

$$\hat{H} = \varepsilon_d \sum_i \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma} - t \sum_{\langle ii' \rangle} \sum_\sigma c^\dagger_{i\sigma} c_{i'\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

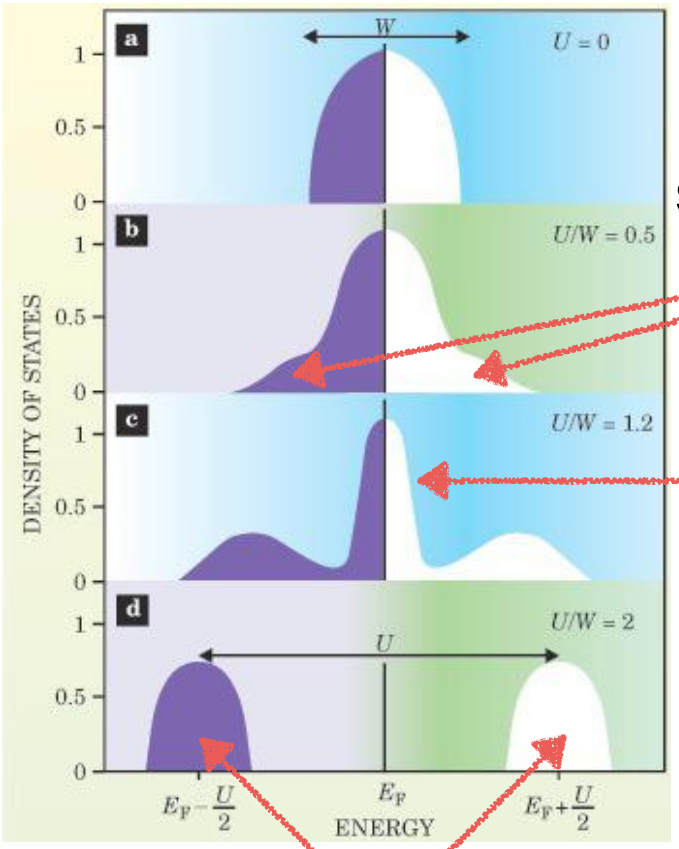
U=0

W: band width

Bethe Lattice

U~W

U>U_c



spectral weight transfer

narrow quasi-particle peak

Hubbard bands

G. Kotliar and D. Vollhardt, Physics Today **57,** 53 (2004)

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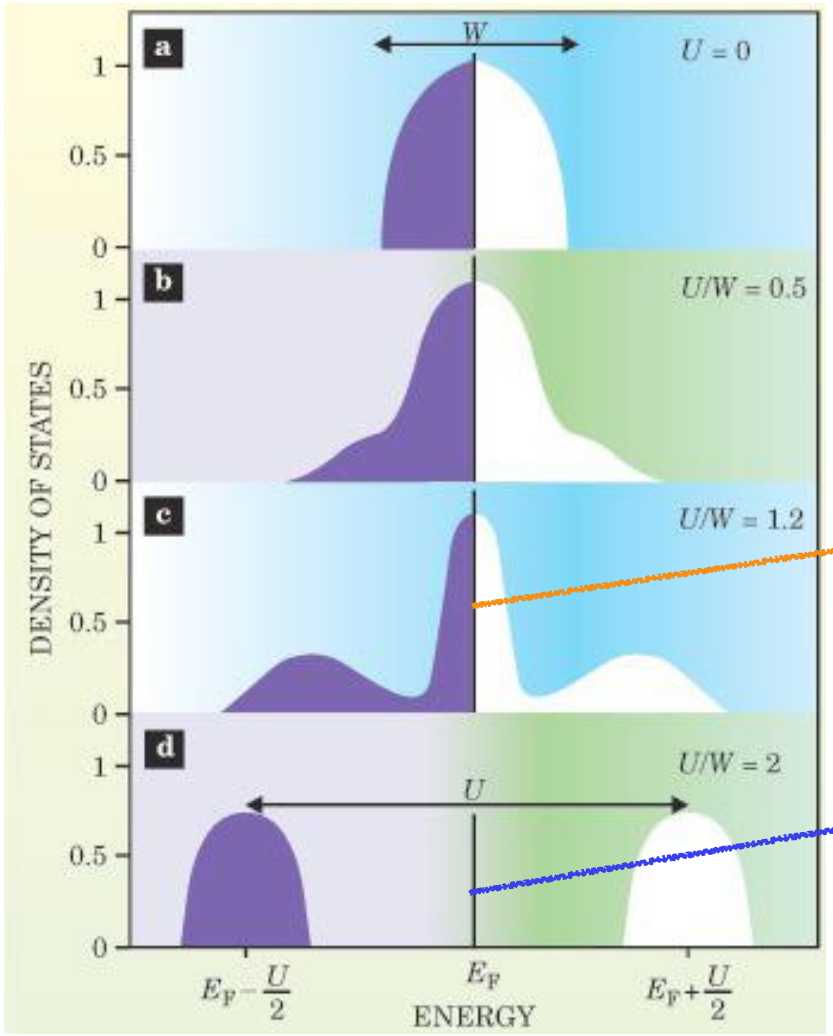
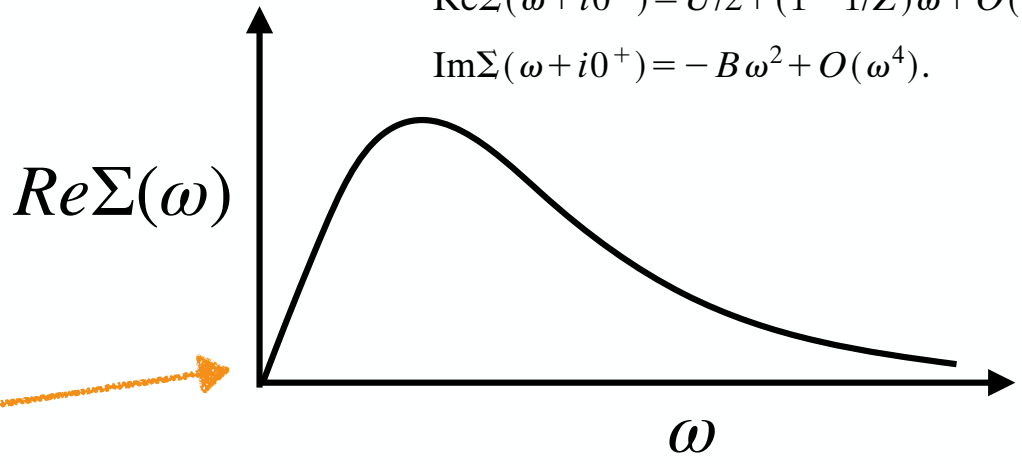
dynamical mean-field theory

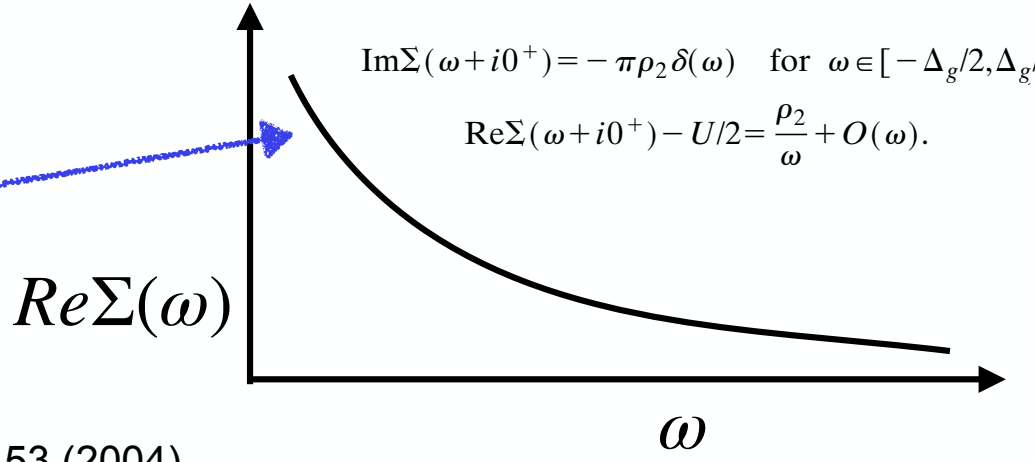
low frequency



$$\mathrm{Re}\Sigma(\omega + i0^+) = U/2 + (1 - 1/Z)\omega + O(\omega^3),$$
$$\mathrm{Im}\Sigma(\omega + i0^+) = -B\omega^2 + O(\omega^4).$$



$$\mathrm{Im}\Sigma(\omega + i0^+) = -\pi\rho_2\delta(\omega) \quad \text{for} \quad \omega \in [-\Delta_g/2, \Delta_g/2]$$
$$\mathrm{Re}\Sigma(\omega + i0^+) - U/2 = \frac{\rho_2}{\omega} + O(\omega).$$



G. Kotliar and D. Vollhardt, Physics Today **57,** 53 (2004)

A. Georges et al. RMP **63**, 13 (1996)

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effective masses

$$G_{\mathbf{k}}(\omega) \sim \frac{1}{\omega - (\varepsilon_{\mathbf{k}} - \mu + \Sigma_R(\omega) + i\Sigma_I(\omega))}$$

U finite

low frequency expansion of self-energy

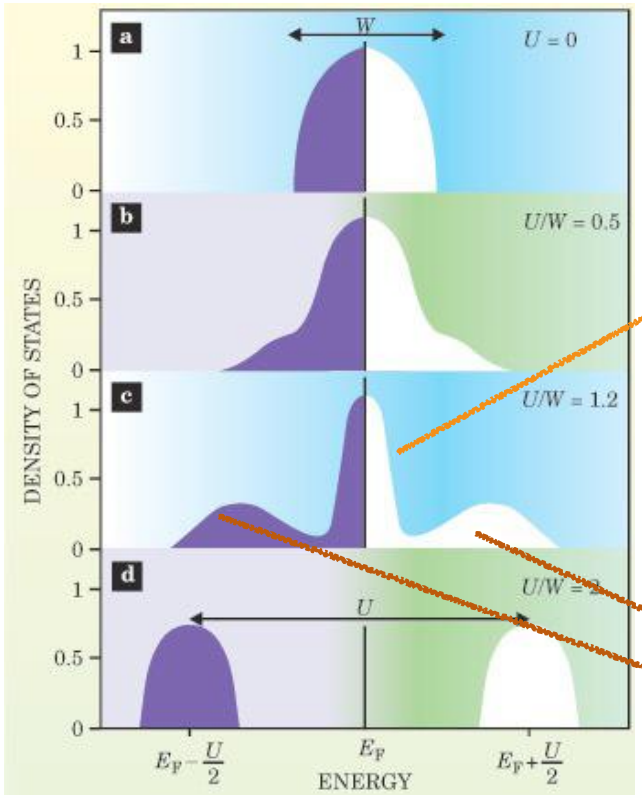
$$G_{\mathbf{k}}(\omega) \sim \frac{1}{\omega - (\varepsilon_{\mathbf{k}} - \mu + \omega(1 - 1/Z) - iB\omega^2)}$$

$$G_{\mathbf{k}}(\omega) \sim \frac{Z}{\omega - Z(\varepsilon_{\mathbf{k}} - \mu) + i\frac{1}{\tau}}$$

effective masses and lifetime

$$G^0_{\mathbf{k}}(\omega) \sim \frac{1}{\omega - (\varepsilon_{\mathbf{k}} - \mu) + i\delta}$$

$$\tau = \infty \qquad m^*/m = 1$$



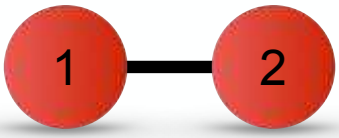
$$G_{\mathbf{k}}(\omega) \sim \frac{Z}{\omega - Z(\varepsilon_{\mathbf{k}} - \mu) + i\frac{1}{\tau}}$$

weight Z<1

$$\frac{m^*}{m} \sim \frac{1}{Z}$$

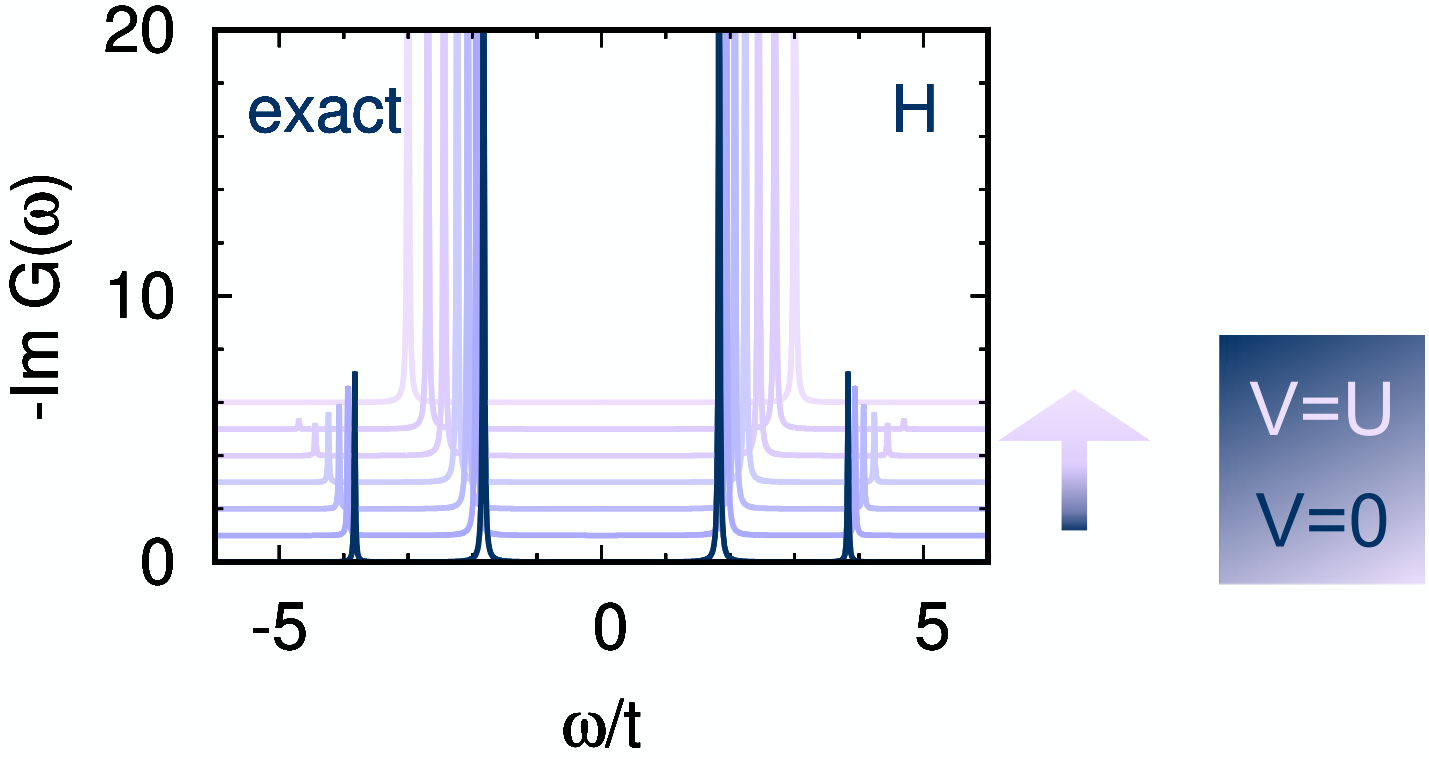
$$\tau = \frac{1}{ZB\omega^2}$$

weight 1-Z



why a **local** Hubbard U?

$$\hat{H} = \varepsilon_d \sum_{i\sigma} \hat{n}_{i\sigma} - t \sum_{\sigma} \left(c^{\dagger}_{1\sigma} c_{2\sigma} + c^{\dagger}_{2\sigma} c_{1\sigma} \right) + U \sum_{i=1,2} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + V \sum_{\sigma\sigma'} \hat{n}_{1\sigma} \hat{n}_{2\sigma'}$$



non-local V makes it **less** correlated

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what about materials?

are there one-band systems?

(systems well described by one-band Hubbard model)

and *how* do we know?

the starting point: DFT band-structures

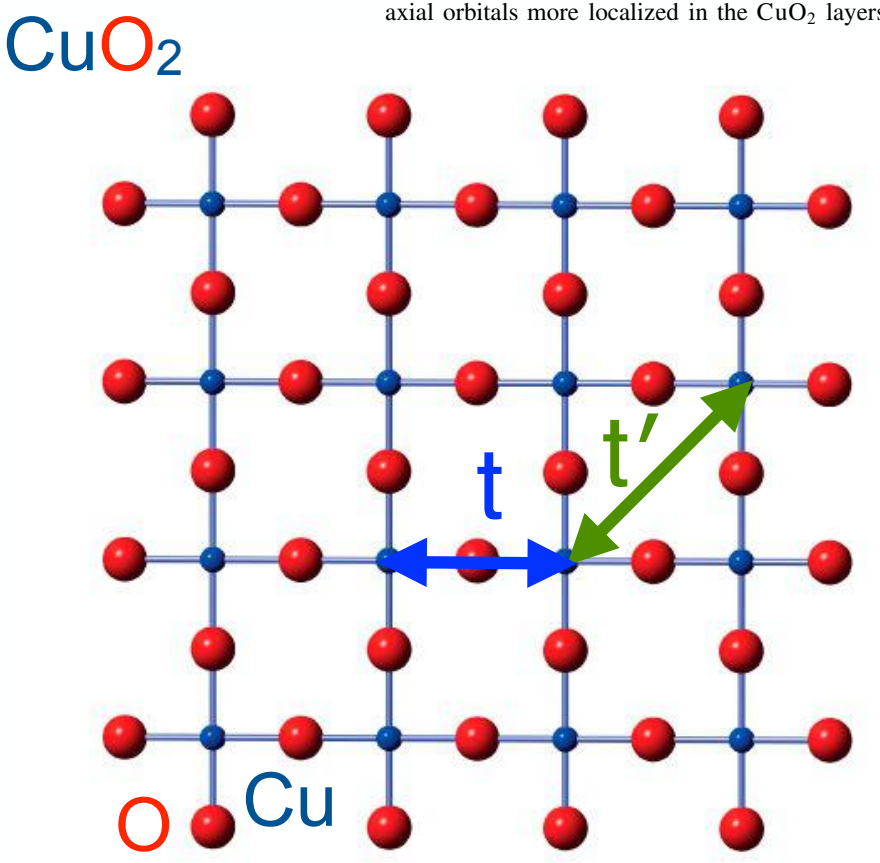
high-T_c superconducting cuprates

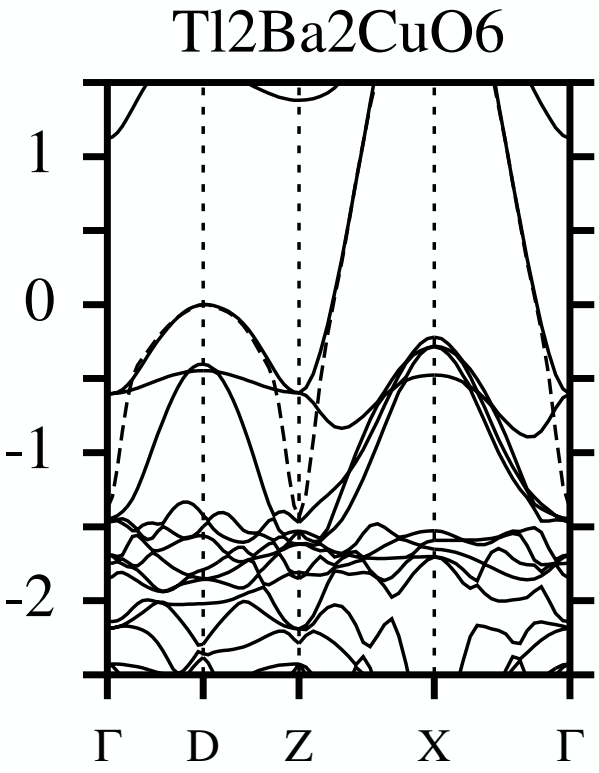
VOLUME 87, NUMBER 4 PHYSICAL REVIEW LETTERS 23 JULY 2001

Band-Structure Trend in Hole-Doped Cuprates and Correlation with $T_{c\,\max}$

E. Pavarini, I. Dasgupta,* T. Saha-Dasgupta,† O. Jepsen, and O. K. Andersen
Max-Planck-Institut für Festkörperforschung, D-70506 Stuttgart, Germany
(Received 4 December 2000; published 10 July 2001)

By calculation and analysis of the bare conduction bands in a large number of hole-doped high-temperature superconductors, we have identified the range of the intralayer hopping as the essential, material-dependent parameter. It is controlled by the energy of the axial orbital, a hybrid between Cu $4s$, apical-oxygen $2p_z$, and farther orbitals. Materials with higher $T_{c\,\max}$ have larger hopping ranges and axial orbitals more localized in the CuO_2 layers.





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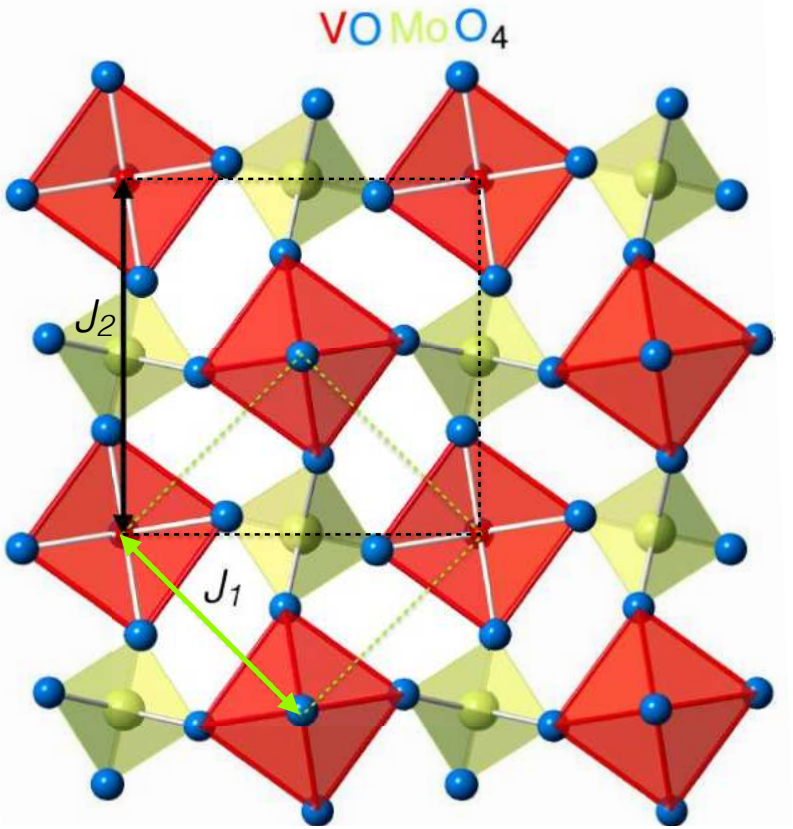
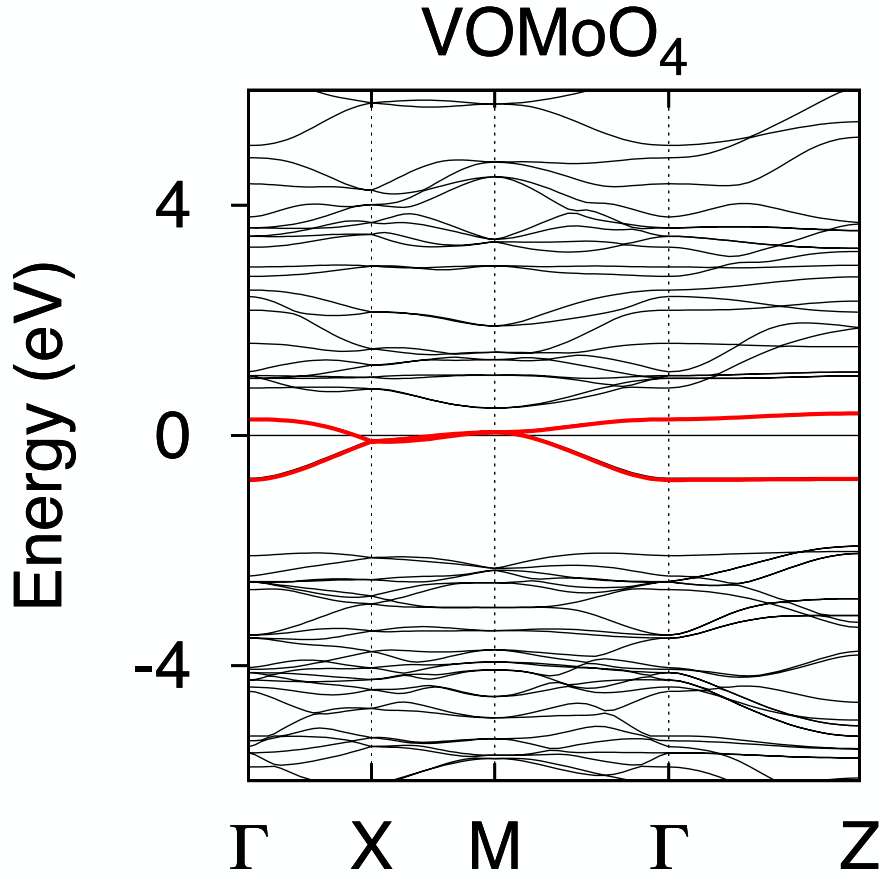
one-band systems: $VOMoO_4$



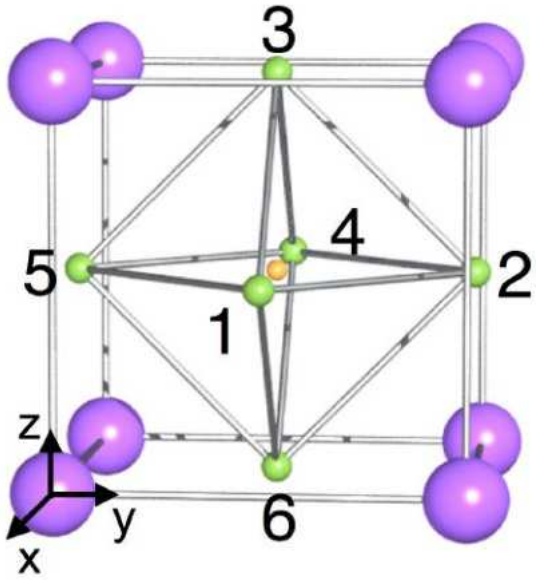


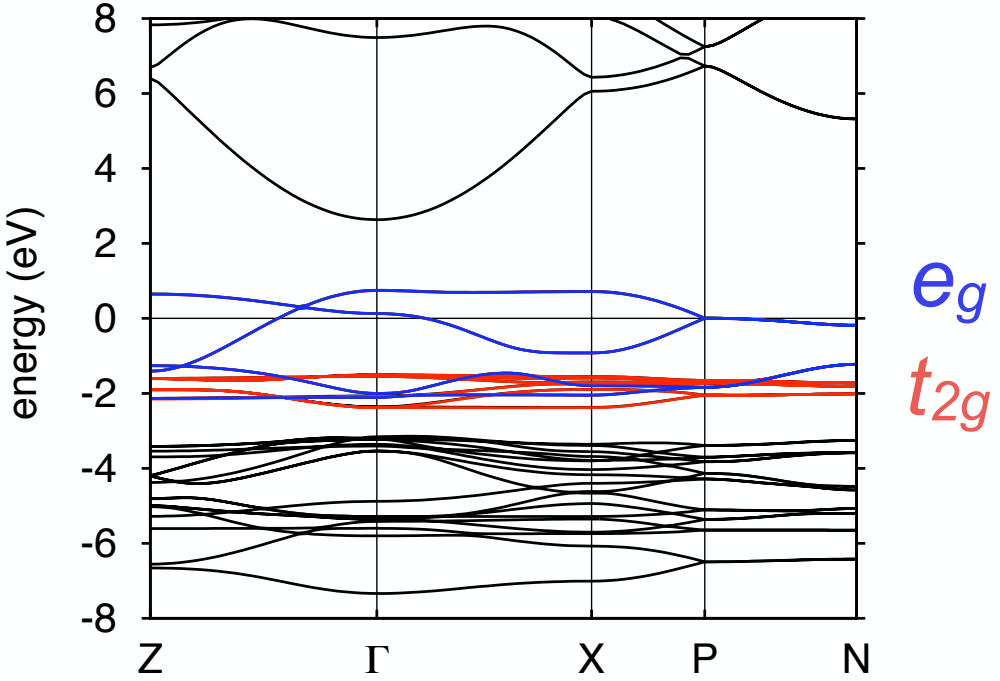
A. Kiani and E. Pavarini, Phys. Rev. B **94**, 075112 (2016)

correlated materials: not only W/U

orbital physics: interplay of spin, orbital, charge, lattice degrees of freedom and correlations

$KCuF_3$





Experiments: insulator. Above 40 K a **paramagnetic** insulator

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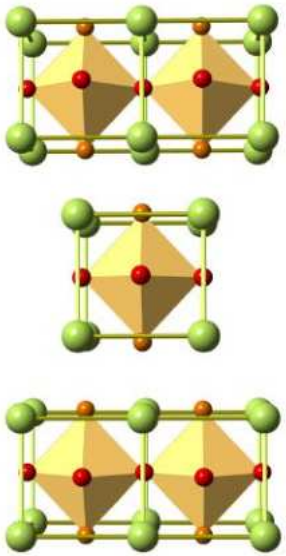
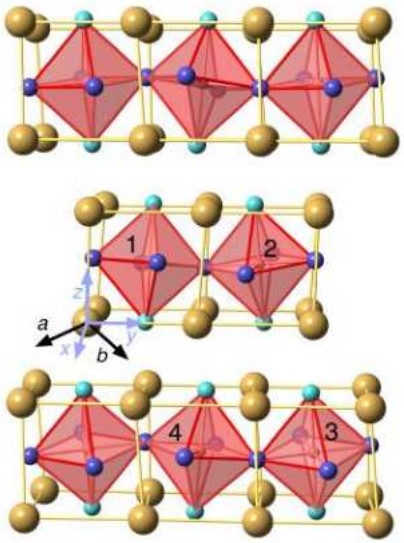
correlated materials: not only W/U

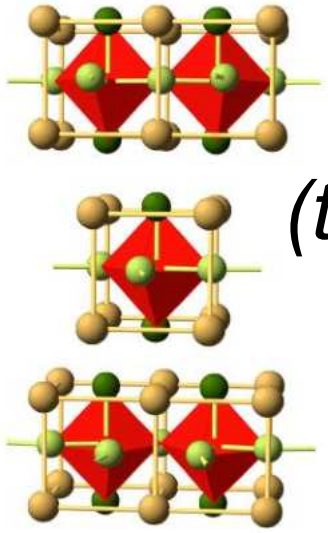
Sr_2RuO_4 Ca_2RuO_4 Sr_2RhO_4 Sr_2IrO_4

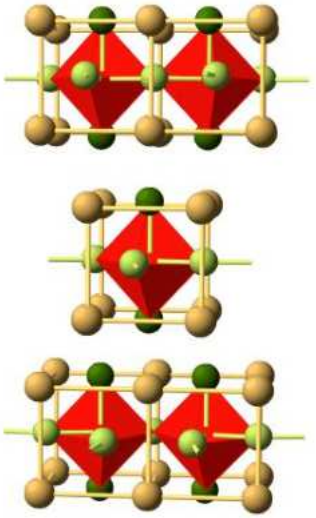


$(t_{2g})^4$



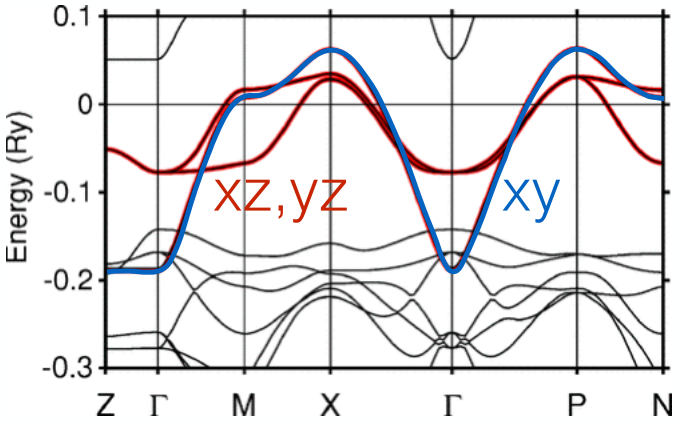
$(t_{2g})^5$





metal *insulator* *metal* *insulator*

orbital physics: interplay of spin, orbital, charge, lattice degrees of freedom and correlations

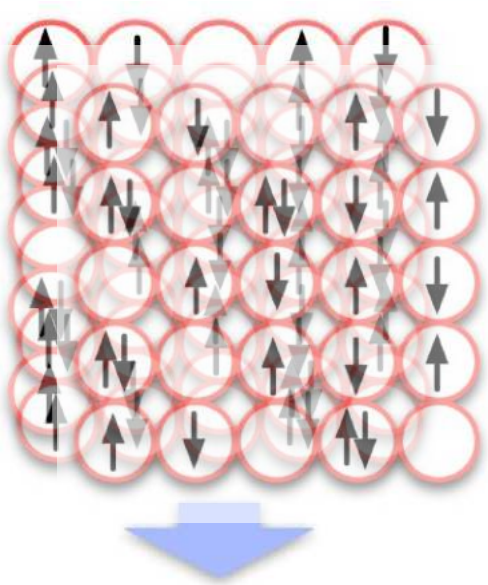


PART 3

DMFT for Materials: DFT+DMFT

DMFT for materials (multi-orbital systems)

$$\hat{H}_e = \underbrace{-\sum_{ab} t_{ab} c_a^\dagger c_b}_{\hat{H}_0} + \underbrace{\frac{1}{2} \sum_{aa'bb'} U^{\text{local}}_{aa'bb'} \; c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\hat{H}_U}$$

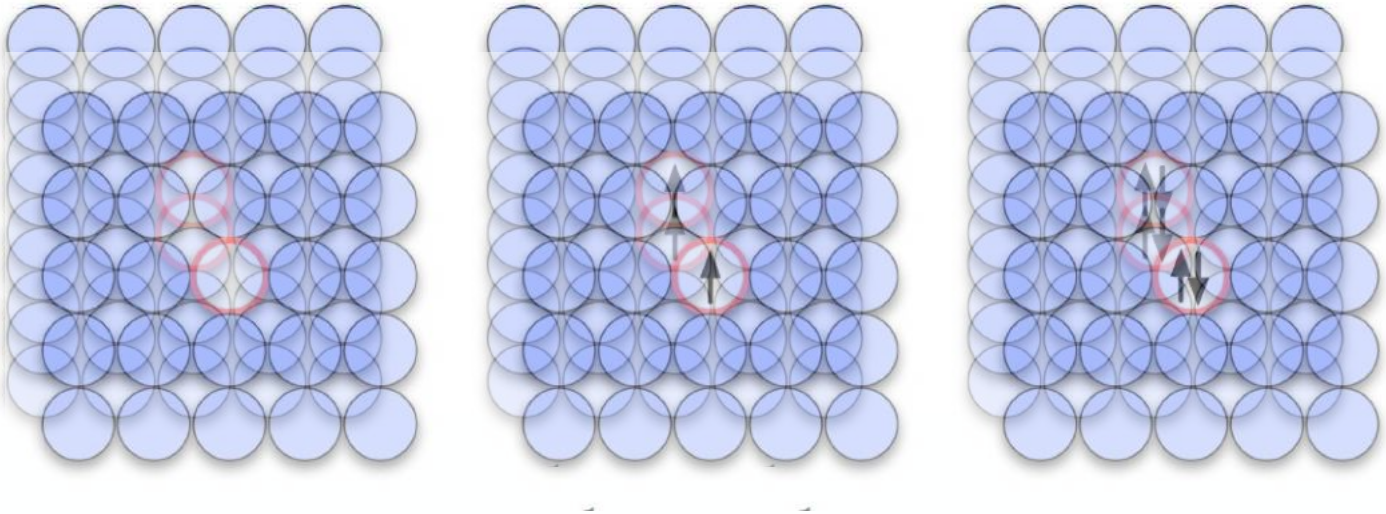


$G^{i,j}$

H^{LDA}

$U^{i,i}$

realistic
self-consistent
quantum-impurity
(QI) model



we need 1. **minimal** material-specific models
and 2. **flexible and efficient** solvers

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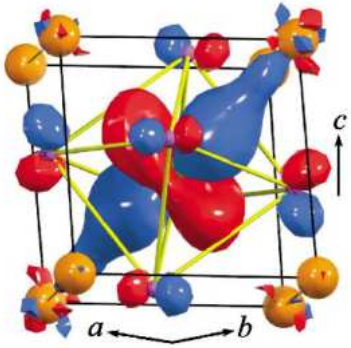
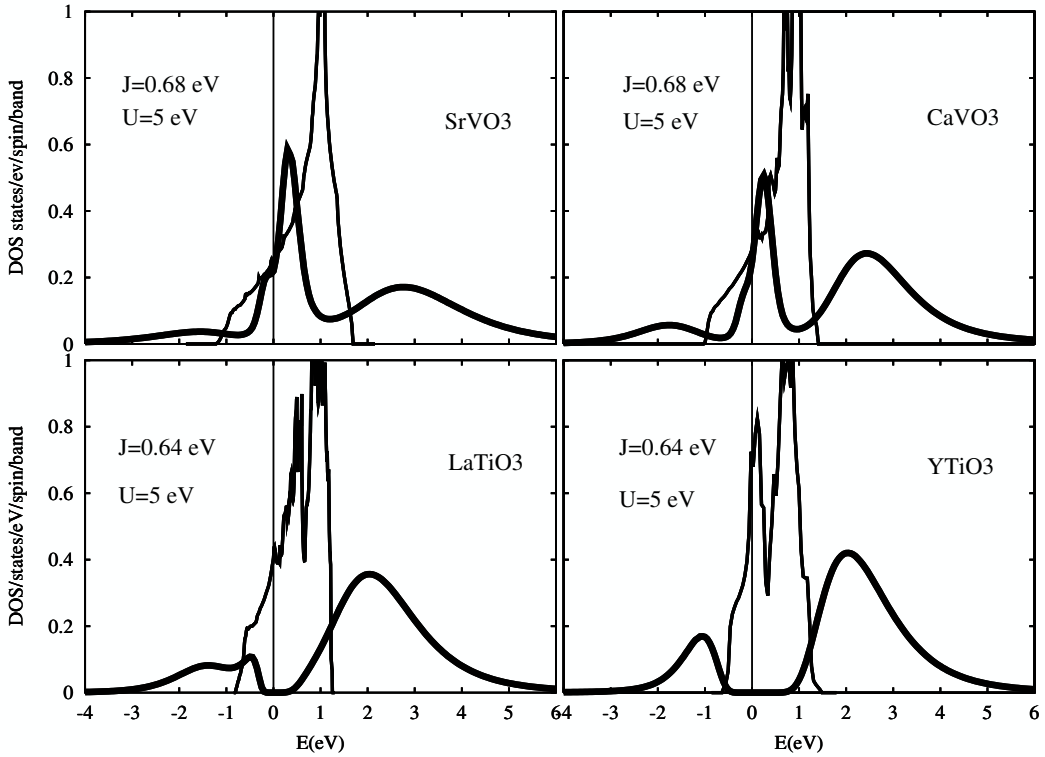
1. minimal models: DFT-based Wannier functions

VOLUME 92, NUMBER 17 PHYSICAL REVIEW LETTERS week ending 30 APRIL 2004

Mott Transition and Suppression of Orbital Fluctuations in Orthorhombic $3d^1$ Perovskites

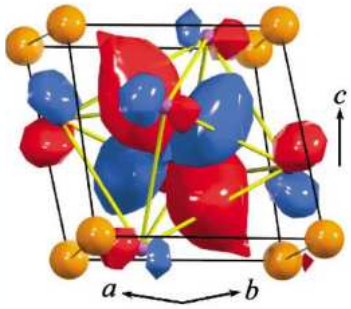
E. Pavarini,[1] S. Biermann,[2] A. Poteryaev,[3] A. I. Lichtenstein,[3] A. Georges,[2] and O. K. Andersen[4]





T~800 K

t_{2g}^1



small crystal-field+hoppings play key role

Δ=200-300 meV

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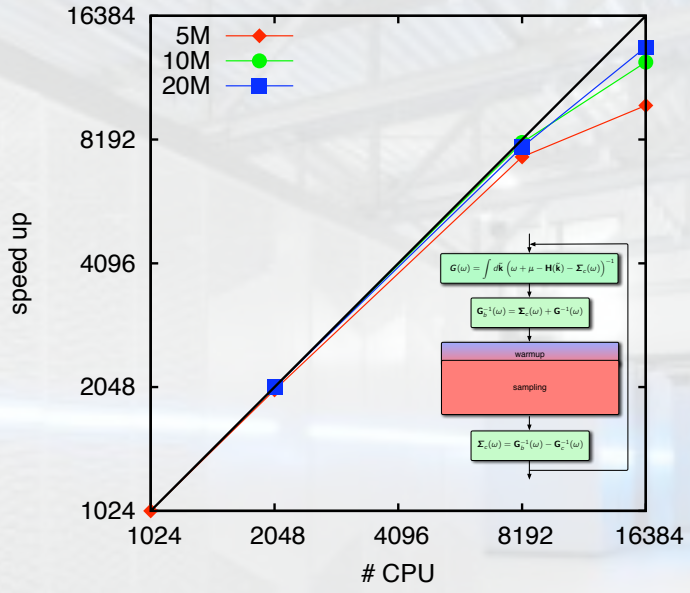
2. flexible and efficient solvers: our package

$$H = -\sum_{ii'}\sum_{mm'}\sum_{\sigma} t^{ii'}_{mm'} c^{\dagger}_{im\sigma} c_{i'm'\sigma}$$
$$+ U \sum_{im} n_{im\uparrow} n_{im\downarrow}$$
$$+ \frac{1}{2} \sum_{im \neq m' \sigma\sigma'} (U - 2J - J\delta_{\sigma\sigma'}) n_{im\sigma} n_{im'\sigma'}$$
$$- J \sum_{m \neq m'} (c^{\dagger}_{m\uparrow} c^{\dagger}_{m'\downarrow} c_{m'\uparrow} c_{m\downarrow} + c^{\dagger}_{m\uparrow} c^{\dagger}_{m\downarrow} c_{m'\uparrow} c_{m'\downarrow})$$

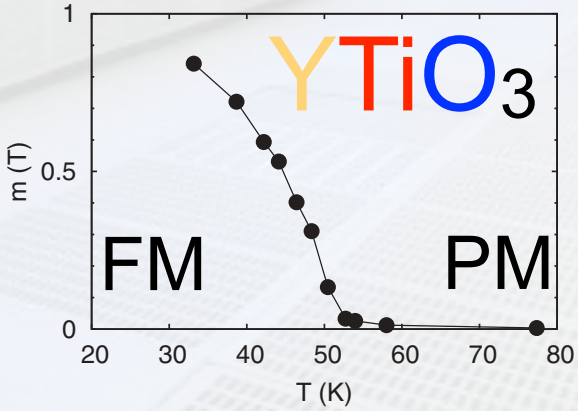
self-energy matrix in spin-orbital space



DMFT and cDMFT

generalized quantum impurity solvers:

general HF QMC
general CT-INT QMC
general CT-HYB QMC



- CT-HYB: A. Flesch, E. Gorelov, E. Koch and E. Pavarini
  Phys. Rev. B **87**, 195141 (2013)
- CT-INT: E. Gorelov et al, PRL 104, 226410 (2010)
- CT-INT+SO: G. Zhang, E. Gorelov, E. Sarvestani, and E. Pavarini,
  Phys. Rev. Lett. 116, 106402 (2016)

sign problem: smart adapted basis choice

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build minimal material-specific models

the many-body problem

$$\hat{H}_e = -\frac{1}{2}\sum_i \nabla_i^2 + \frac{1}{2}\sum_{i\neq i'} \frac{1}{|\mathbf{r}_i - \mathbf{r}_{i'}|} - \sum_{i,\alpha} \frac{Z_\alpha}{|\mathbf{r}_i - \mathbf{R}_\alpha|} + \frac{1}{2}\sum_{\alpha\neq\alpha'} \frac{Z_\alpha Z_{\alpha'}}{|\mathbf{R}_\alpha - \mathbf{R}_{\alpha'}|}$$

electronic Hamiltonian in 2nd quantization

$$\hat{H}_e = \underbrace{-\sum_{ab} t_{ab} c_a^\dagger c_b}_{\hat{H}_0} + \underbrace{\frac{1}{2}\sum_{aa'bb'} U_{aa'bb'}\, c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\hat{H}_U}$$

complete one-electron basis set!

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the quantum many-body problem

$$\hat{H}_e = \underbrace{-\sum_{ab} t_{ab} c_a^\dagger c_b}_{\hat{H}_0} + \underbrace{\frac{1}{2} \sum_{aa'bb'} U_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\hat{H}_U}$$

hopping integrals

$$t_{ab} = -\int d\mathbf{r}\; \overline{\phi_a}(\boldsymbol{r}) \Big(-\frac{1}{2}\nabla^2 \underbrace{- \sum_\alpha \frac{Z_\alpha}{|\boldsymbol{r} - \boldsymbol{R}_\alpha|}}_{v_{\mathrm{en}}(\boldsymbol{r})} \Big) \phi_b(\boldsymbol{r})$$

$$U_{aa'bb'} = \int d\mathbf{r}_2 \int d\mathbf{r}_2\; \overline{\phi_a}(\boldsymbol{r}_1)\, \overline{\phi_{a'}}(\boldsymbol{r}_2)\; \frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|}\; \phi_{b'}(\boldsymbol{r}_2)\, \phi_b(\boldsymbol{r}_1)$$

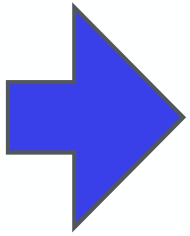
Coulomb integrals

1. basis choice

in theory all basis are identical

$$\hat{H}_e = \underbrace{-\sum_{ab} t_{ab} c_a^\dagger c_b}_{\hat{H}_0} + \underbrace{\frac{1}{2} \sum_{aa'bb'} U_{aa'bb'}\ c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\hat{H}_U}$$

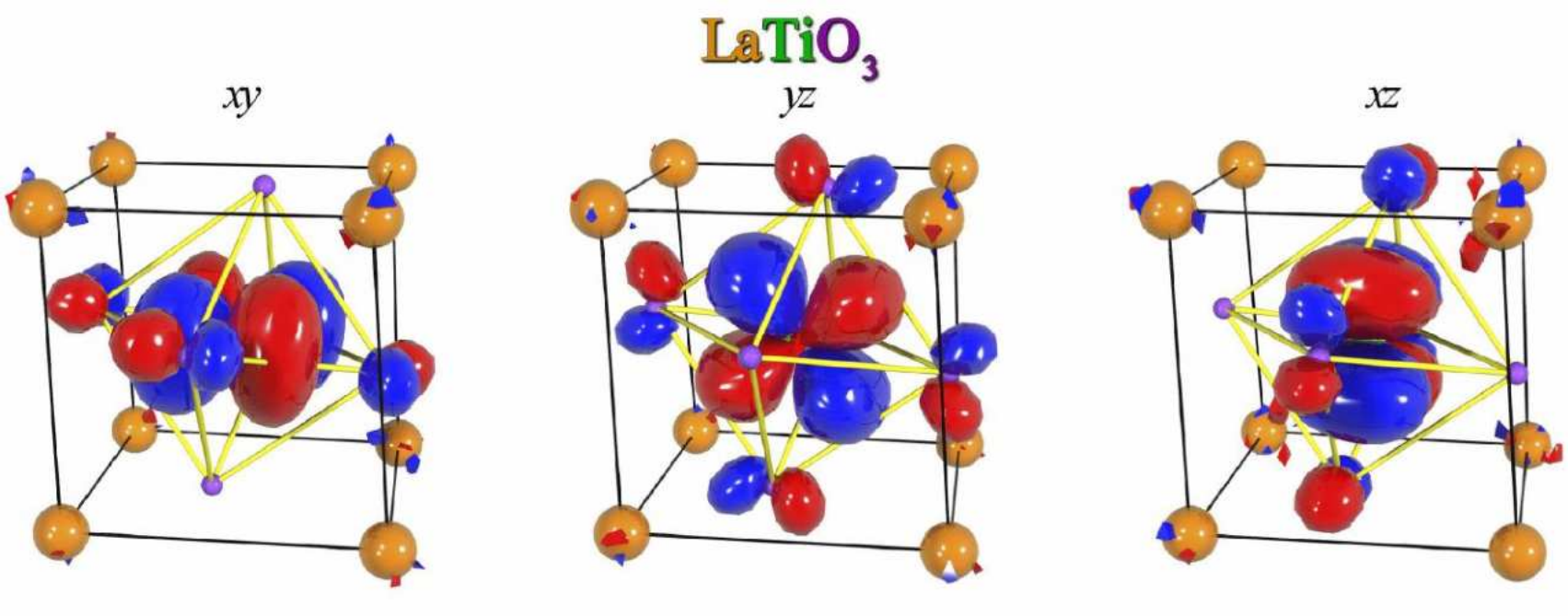
in practice some bases are better than others



DFT-based (Kohn-Sham) **Wannier** functions

Wannier functions: Fourier transform of Bloch states

why Kohn-Sham Wannier functions ?



localized, orthogonal, span bands exactly

carry information on lattice and chemistry

E. Pavarini, A. Yamasaki, J. Nuss, O.K. Andersen, New. J. Phys ***7****, 188 (2005)*

a price to pay: double-counting correction

$$\hat{H}_e = \underbrace{-\sum_{ab} t_{ab} c_a^\dagger c_b}_{\hat{H}_0} + \underbrace{\frac{1}{2}\sum_{aa'bb'} U_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\hat{H}_U}$$

Kohn-Sham Wannier orbitals

$$\hat{H}_e = \underbrace{-\sum_{ab} \tilde{t}_{ab}\, c_a^\dagger c_b}_{\hat{H}_0 = \hat{H}_e^{\mathrm{LDA}}} + \underbrace{\frac{1}{2}\sum_{aba'b'} \tilde{U}_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b - \hat{H}_{\mathrm{DC}}}_{\Delta\hat{H}_U}$$

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a price to pay: double-counting correction

$$\tilde{t}_{ab} = -\int d\mathbf{r}\, \overline{\phi_a^{\mathrm{KS}}}(\boldsymbol{r}) \left(-\frac{1}{2}\nabla^2 + v_{\mathrm{R}}(\boldsymbol{r}) \right) \phi_b^{\mathrm{KS}}(\boldsymbol{r})$$

$$v_R(\boldsymbol{r}) = -\sum_\alpha \frac{Z_\alpha}{|\boldsymbol{r} - \boldsymbol{R}_\alpha|} + \int d\boldsymbol{r}' \frac{n(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|} + \frac{\delta E_{\mathrm{xc}}[n]}{\delta n} =$$

Hartree **exchange-correlation**

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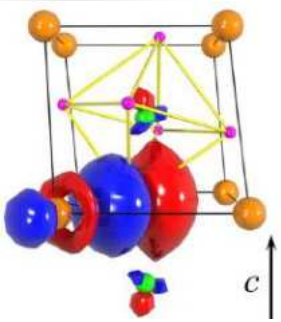
2. divide electrons in *light* and *heavy*

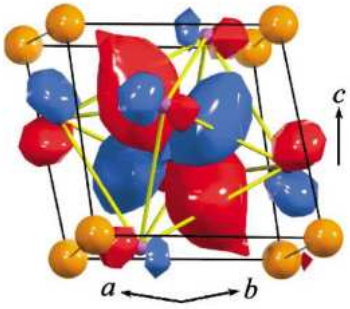
$YTiO_3$

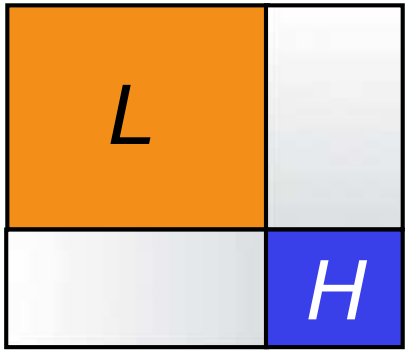
light= weakly correlated

O *p*

Ti 3d t_{2g}







heavy= strongly correlated

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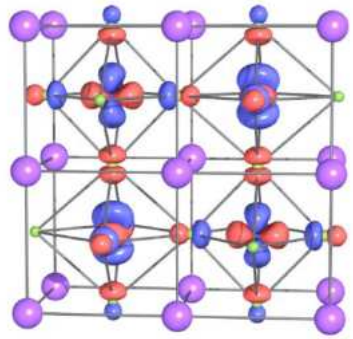
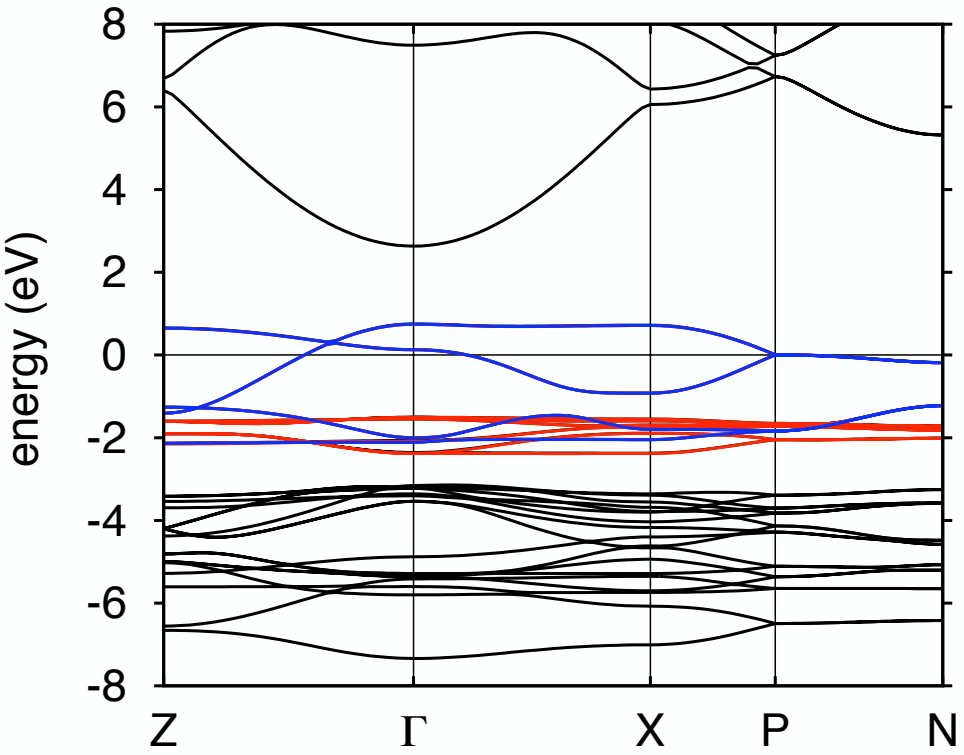
which electrons are *heavy*?

$KCuF_3$



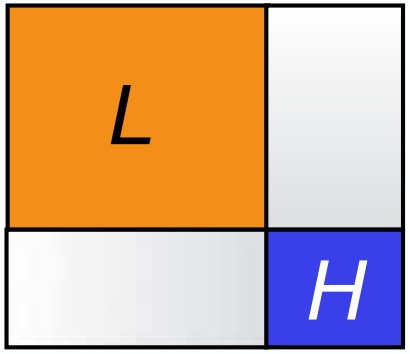
K

F p



Cu e_g

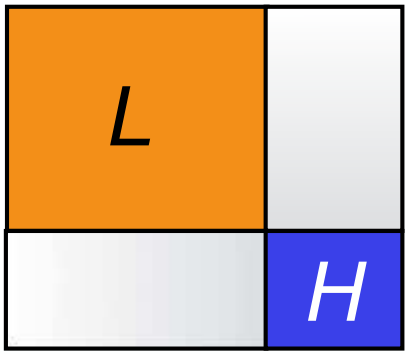
Cu t_{2g}



typically bands from partially filled
d/f shells at the Fermi level

weakly-correlated or light electrons

$$\hat{H}_e = \underbrace{-\sum_{ab} \tilde{t}_{ab}\, c_a^\dagger c_b}_{\hat{H}_0 = \hat{H}_e^{\mathrm{LDA}}} + \underbrace{\frac{1}{2}\sum_{aba'b'} \tilde{U}_{aa'bb'}\, c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\Delta \hat{H}_U} - \hat{H}_{\mathrm{DC}}$$



$$\hat{H}_{\mathrm{eff}} \sim \hat{S}^{-1} \hat{H}_e \hat{S} \sim \hat{H}_e^{\mathrm{LDA}}$$

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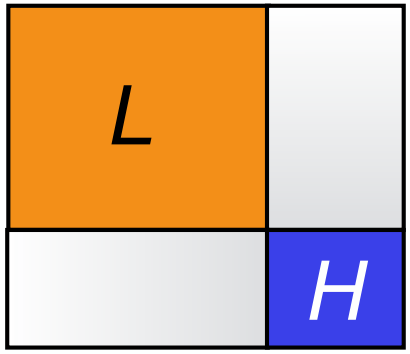
strongly-correlated or heavy electrons

Hubbard-like approximation

$$\hat{H}_e = \underbrace{-\sum_{ab} \tilde{t}_{ab}\, c_a^\dagger c_b}_{\hat{H}_0 = \hat{H}_e^{\mathrm{LDA}}} + \underbrace{\frac{1}{2} \sum_{aba'b'} \tilde{U}_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b - \hat{H}_{\mathrm{DC}}}_{\Delta \hat{H}_U}$$



$$\hat{H}_{\mathrm{eff}} \sim \hat{S}^{-1} \hat{H}_e \hat{S} \sim \hat{H}_{\mathrm{Hubbard-like}}$$

$$\hat{H}_e = \underbrace{-\sum_{ab} \tilde{t}_{ab}\, c_a^\dagger c_b}_{\hat{H}_0 = \hat{H}_e^{\mathrm{LDA}}} + \underbrace{\frac{1}{2} \sum_{aba'b'} \tilde{U}^{\mathrm{local}}_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b - \hat{H}^{\mathrm{local}}_{\mathrm{DC}}}_{\Delta \hat{H}_U}$$

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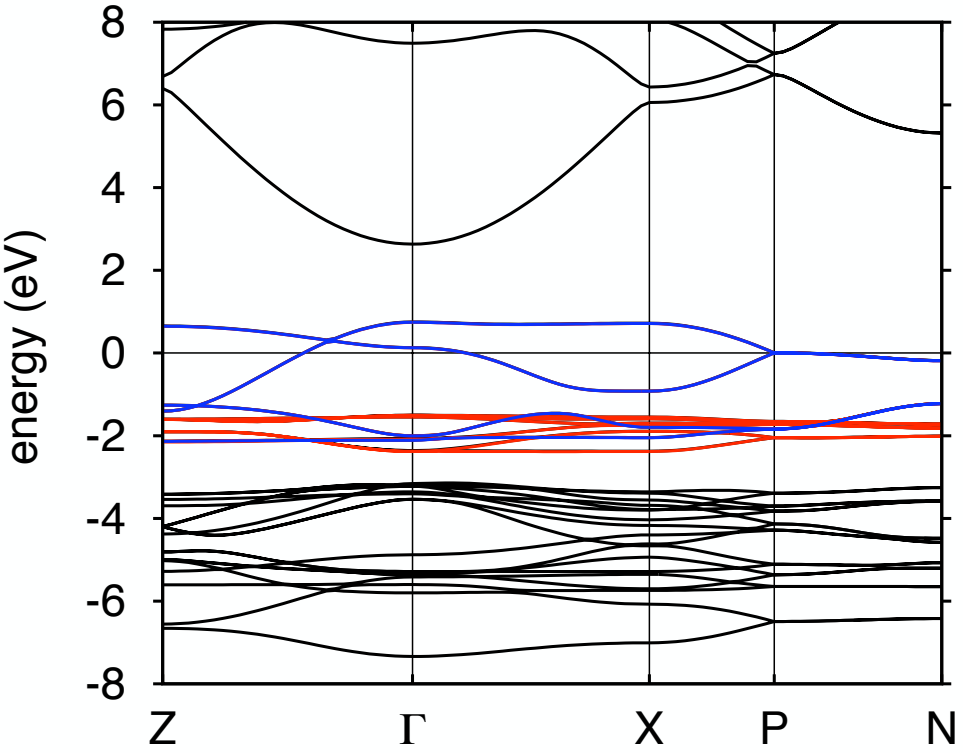
3. how much do we downfold?

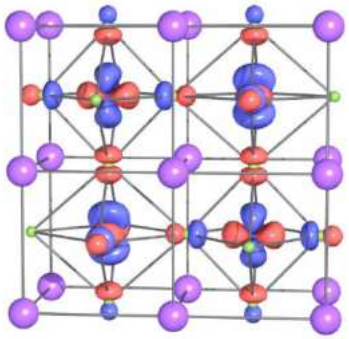
$KCuF_3$

K

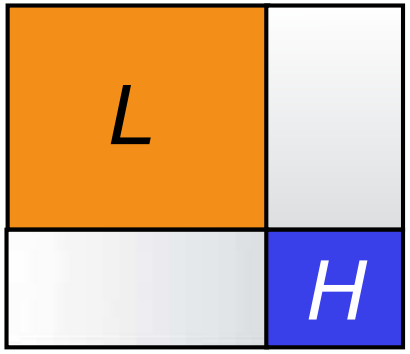
Cu t_{2g}

F p





Cu e_g



e_g

e_g

H

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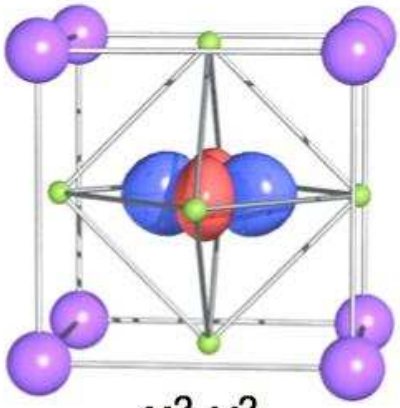
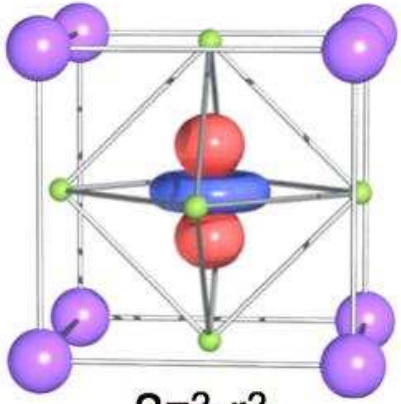
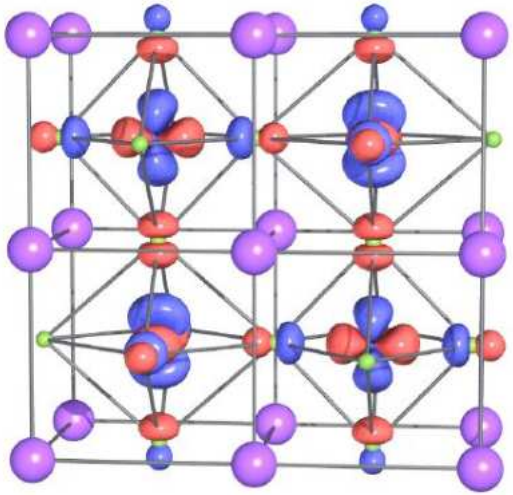
dowfolding level and Wannier orbitals

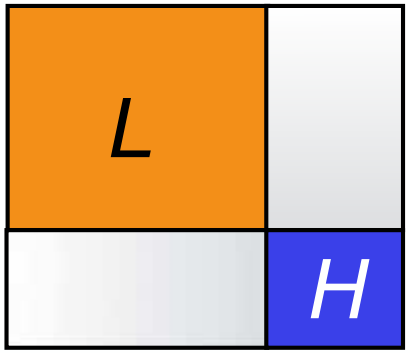




no downfolding

massive downfolding





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4. double-counting correction

$$\hat{H}_e = \underbrace{-\sum_{ab} \tilde{t}_{ab}\, c_a^\dagger c_b}_{\hat{H}_0 = \hat{H}_e^{\mathrm{LDA}}} + \underbrace{\frac{1}{2} \sum_{aba'b'} \tilde{U}^{\mathrm{local}}_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b - \hat{H}^{\mathrm{local}}_{\mathrm{DC}}}_{\Delta \hat{H}_U}$$

for massive downfolding (e.g., one band)
= shift of chemical potential

massive downfolding: DC correction

around mean-field approximation

$$\hat{H}_U = U \sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

$$\hat{H}_{\mathrm{DC}} = U \sum_i \left(\hat{n}_{i\uparrow}\bar{n}_{i\downarrow} + \bar{n}_{i\uparrow}\hat{n}_{i\downarrow} - \bar{n}_{i\uparrow}\bar{n}_{i\downarrow} \right)$$

$$\bar{n}_{i\sigma} = n/2$$

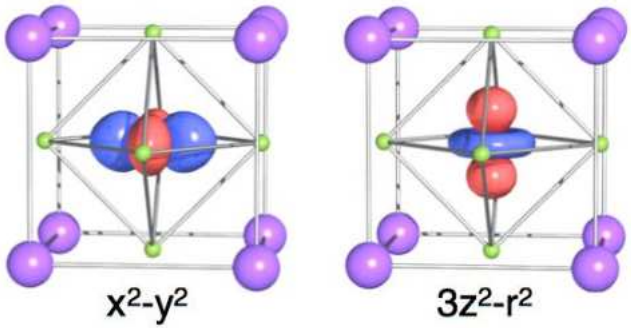
$$\hat{H}_{\mathrm{DC}} = \frac{n}{2} U \sum_i \left(\hat{n}_{i\uparrow} + \hat{n}_{i\downarrow} - \frac{n}{2} \right) = \delta\mu\hat{N} - \mathrm{const}$$

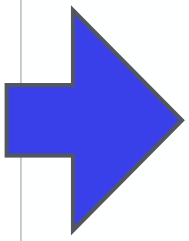
other often used approximate scheme: FLL

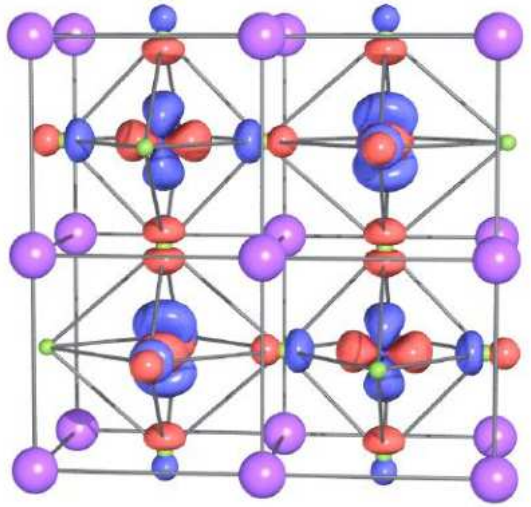
5. the Coulomb integrals: screening

$$U_{aa'bb'} = \int d\mathbf{r}_2 \int d\mathbf{r}_2 \, \overline{\phi_a}(\boldsymbol{r}_1)\, \overline{\phi_{a'}}(\boldsymbol{r}_2)\, \frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|}\, \phi_{b'}(\boldsymbol{r}_2)\, \phi_b(\boldsymbol{r}_1)$$

bare Coulomb integrals







5. the Coulomb integrals: screening

$$U_{aa'bb'} = \int d\mathbf{r}_2 \int d\mathbf{r}_2 \; \overline{\phi_a}(\boldsymbol{r}_1)\, \overline{\phi_{a'}}(\boldsymbol{r}_2)\; \frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|}\; \phi_{b'}(\boldsymbol{r}_2)\, \phi_b(\boldsymbol{r}_1)$$

bare Coulomb integrals

however, screening effects

$$U_{bare} \rightarrow U_{screened}$$

(approximate schemes: cLDA, **cRPA**)

cRPA

$$U_{aa'bb'} = \int d\mathbf{r}_2 \int d\mathbf{r}_2 \, \overline{\phi_a}(\mathbf{r}_1)\, \overline{\phi_{a'}}(\mathbf{r}_2)\, W(\mathbf{r}_1 - \mathbf{r}_2)\, \phi_{b'}(\mathbf{r}_2)\, \phi_b(\mathbf{r}_1)$$

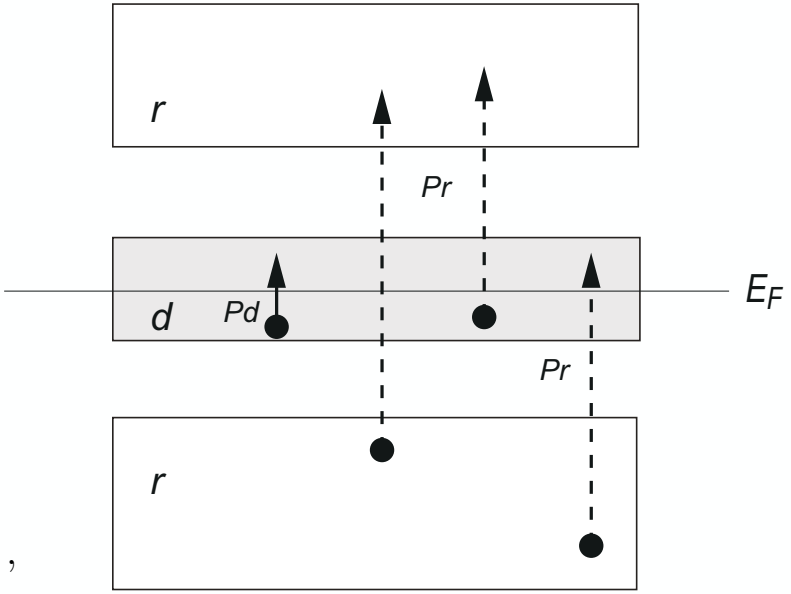
screened Coulomb integrals

$$W = \frac{U}{1 - U P_r}$$

$$P = P_d + P_r$$ polarization function

$$P = P^0$$

$$P^0(\mathbf{r}, \mathbf{r}'; \omega) = -2 \sum_i^{\text{occ}} \sum_j^{\text{unocc}} \left(\frac{f_{ij}(\mathbf{r}, \mathbf{r}')}{\omega + (\varepsilon_j - \varepsilon_i - i\delta)} - \frac{f_{ij}^*(\mathbf{r}, \mathbf{r}')}{\omega - (\varepsilon_j - \varepsilon_i - i\delta)} \right),$$

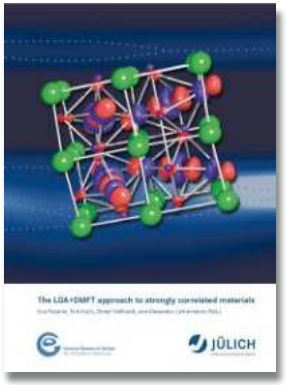


(figure from F. Aryasetiawan, correl18)

t_{2g} or e_g only models, spherical Coulomb

$$\hat{H}_e = \underbrace{-\sum_{ab} \tilde{t}_{ab}\, c_a^\dagger c_b}_{\hat{H}_0 = \hat{H}_e^{\mathrm{LDA}}} + \underbrace{\frac{1}{2} \sum_{aba'b'} \tilde{U}_{aa'bb'}\; c_a^\dagger c_{a'}^\dagger c_{b'} c_b}_{\Delta \hat{H}_U} - \hat{H}_{\mathrm{DC}}$$

Hund's rule J



$$\hat{H} = -\sum_{\sigma} \sum_{i \neq i'} \sum_{mm'} t_{m,m'}^{i,i'}\; c_{im\sigma}^\dagger c_{i'm'\sigma}$$

$$+ U \sum_{i\; m} \hat{n}_{im\uparrow} \hat{n}_{im\downarrow} + \frac{1}{2} \sum_{\substack{i\sigma\sigma' \\ m \neq m'}} \left(U - 2J - J\delta_{\sigma,\sigma'}\right) \hat{n}_{im\sigma} \hat{n}_{im'\sigma'}$$

$$- J \sum_{i\; m \neq m'} \left(c_{im\uparrow}^\dagger c_{im\downarrow}^\dagger c_{im'\uparrow} c_{im'\downarrow} + c_{im\uparrow}^\dagger c_{im\downarrow} c_{im'\downarrow}^\dagger c_{im'\uparrow} \right)$$

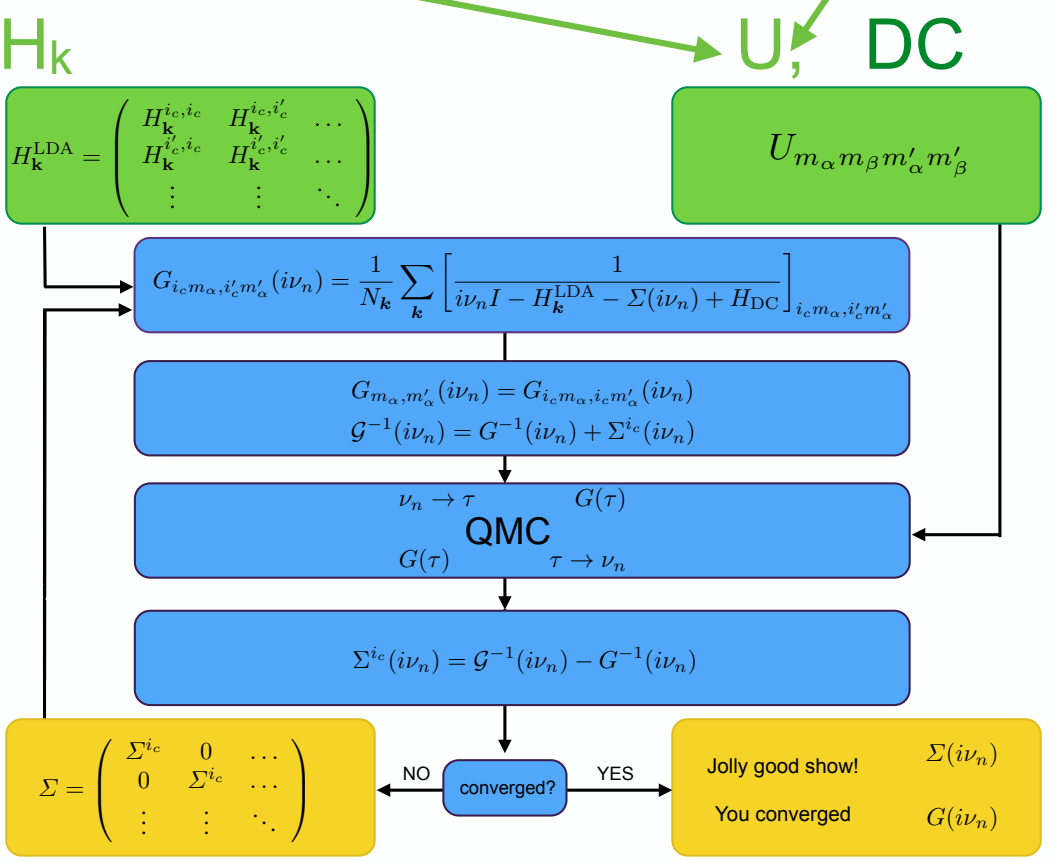
derivation: www.cond-mat.de/events/correl11/manuscripts/pavarini.pdf

collecting everything together

heavy electrons:
hoppings and U_{bare} for DMFT

light electrons:
Kohn-Sham bands and screening

Hubbard model



DMFT

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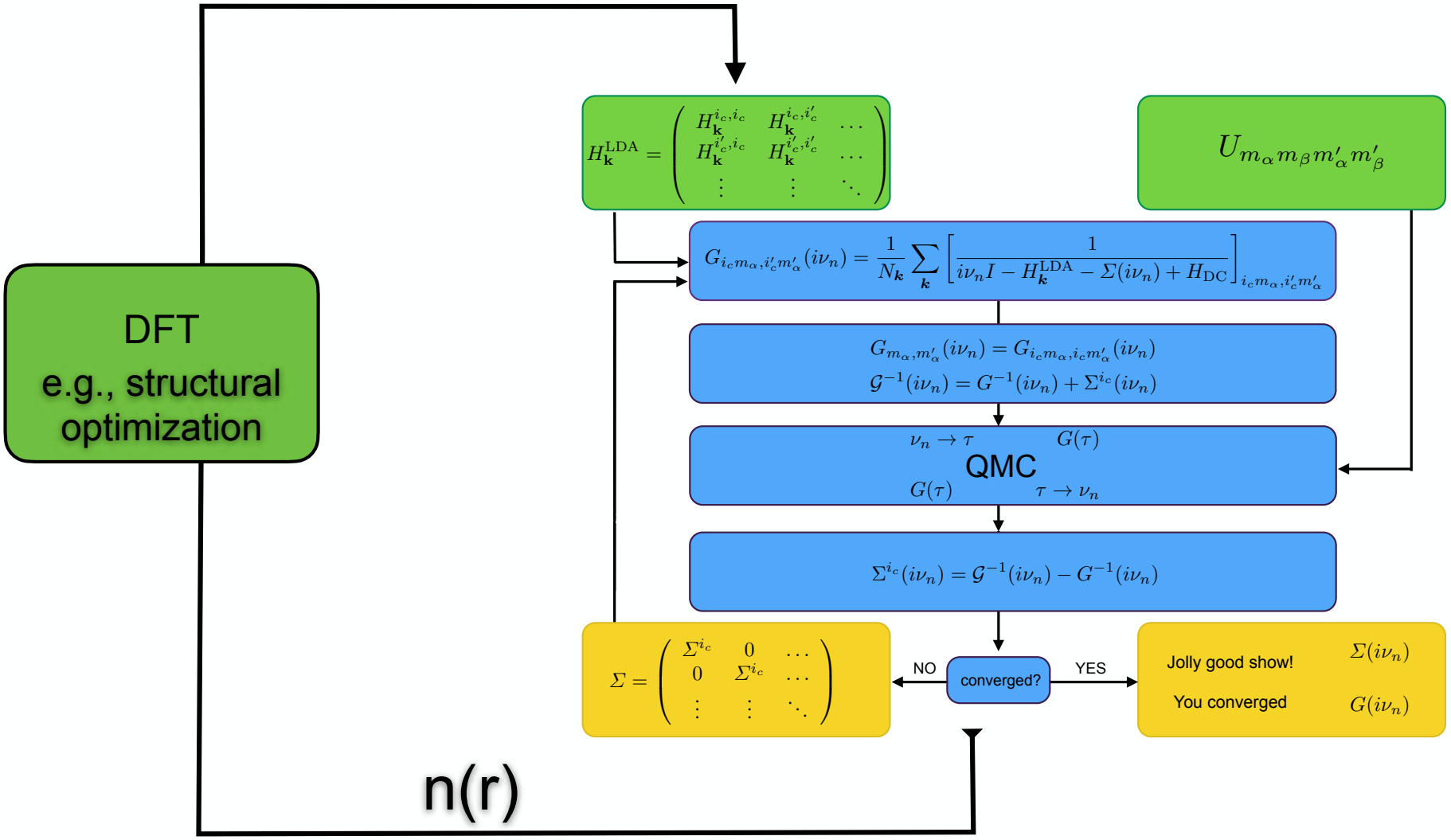
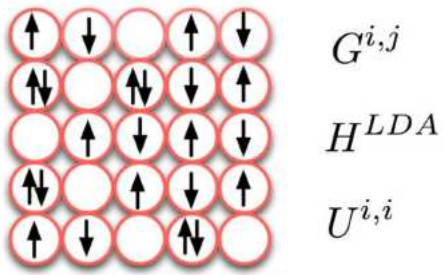
charge self-consistency



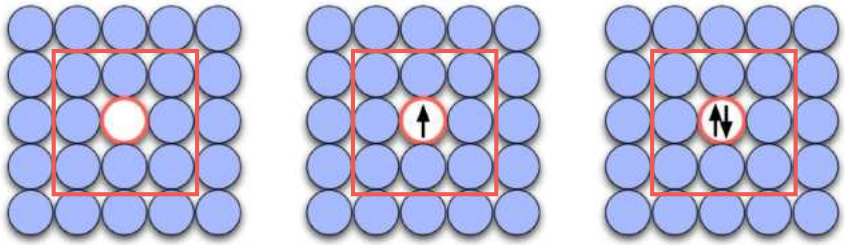
non-local effects: cluster extensions

in DMFT we can account for spatial
long-range correlations

but not for short-range correlations



cluster DMFT



cluster in **k** space: DCA

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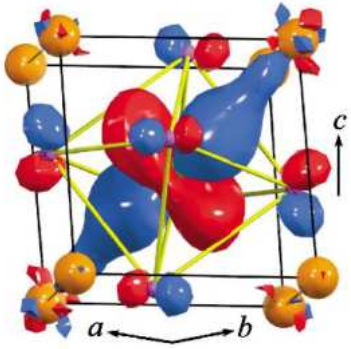
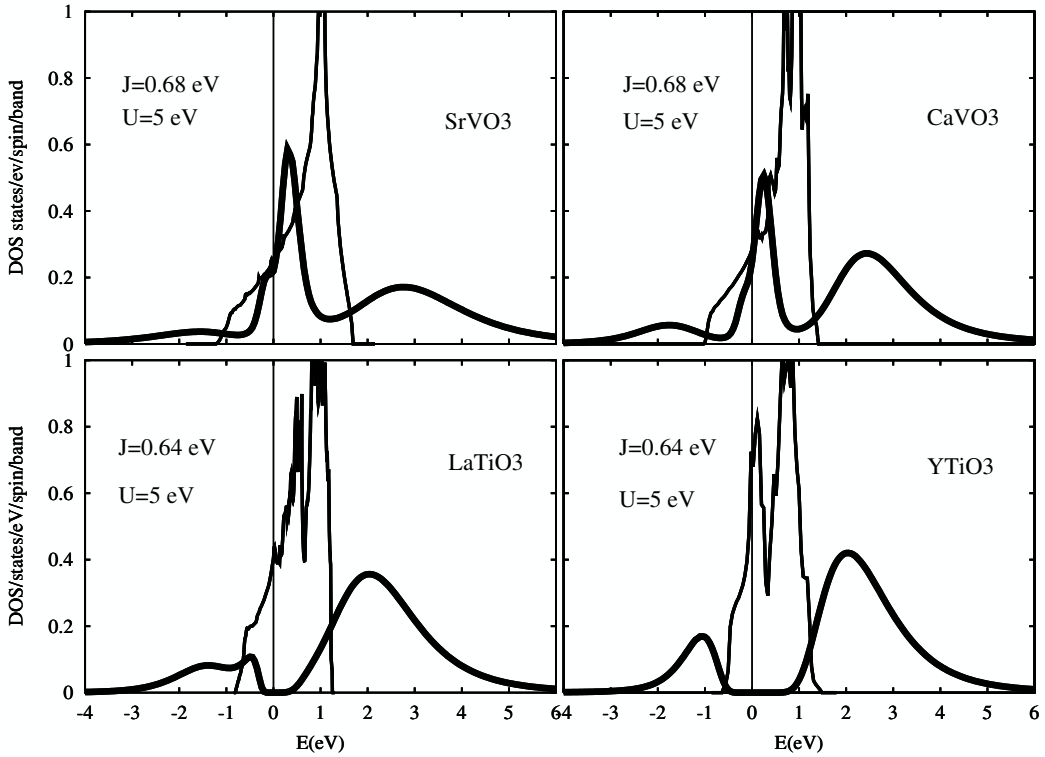
does it work? yes! details matter

VOLUME 92, NUMBER 17 PHYSICAL REVIEW LETTERS week ending 30 APRIL 2004

Mott Transition and Suppression of Orbital Fluctuations in Orthorhombic $3d^1$ Perovskites

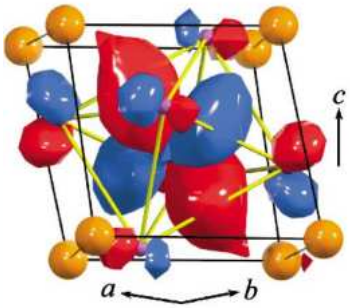
E. Pavarini,[1] S. Biermann,[2] A. Poteryaev,[3] A. I. Lichtenstein,[3] A. Georges,[2] and O. K. Andersen[4]





T~800 K

t_{2g}^1



small crystal-field+hoppings play key role

Δ=200-300 meV

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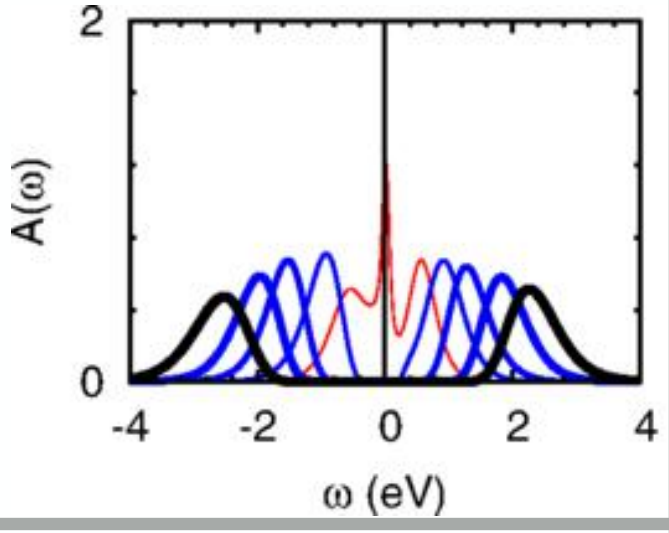
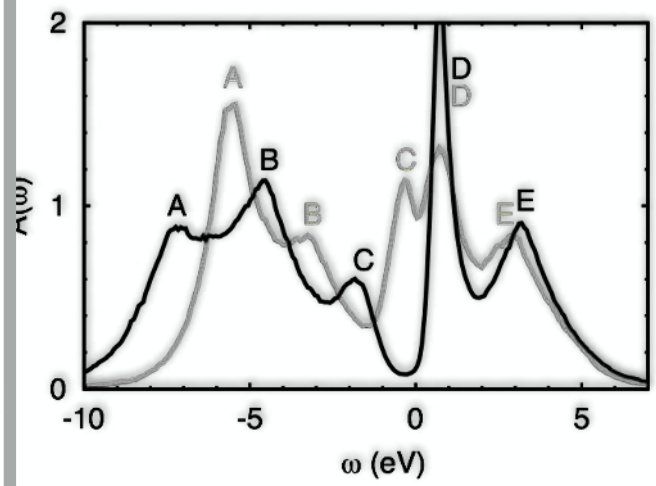
what can we do so far?

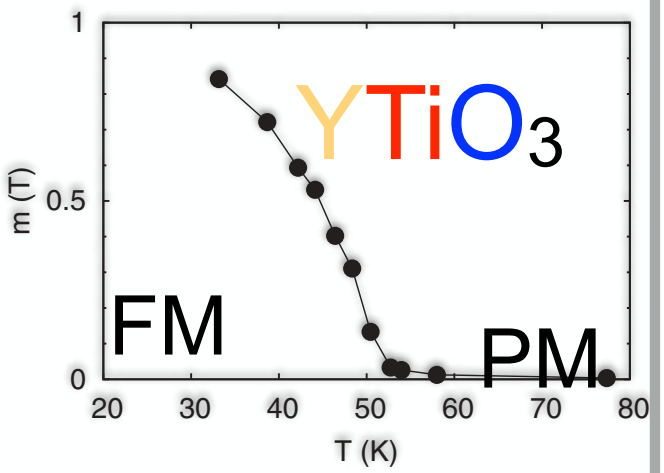
spectral functions



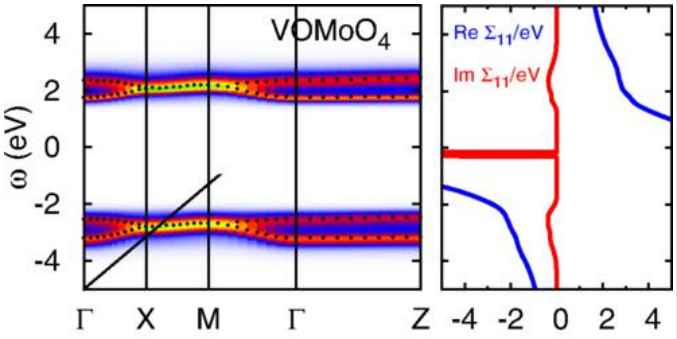
many orbitals



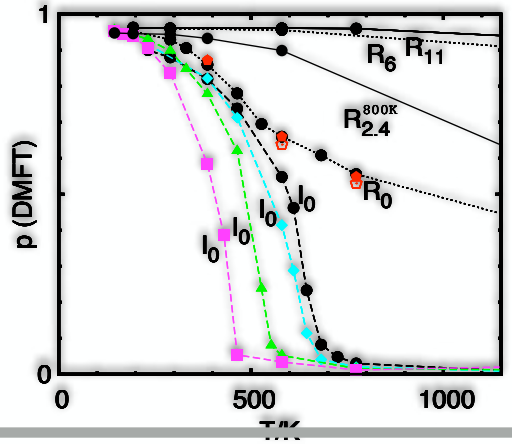
low T



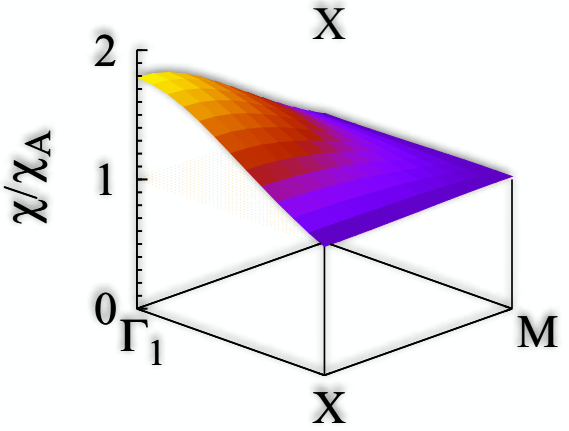
correlated bands



phase transitions



susceptibilities



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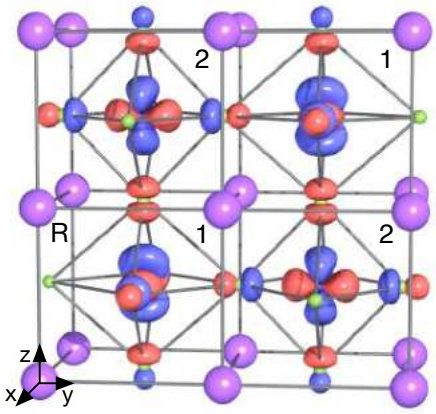
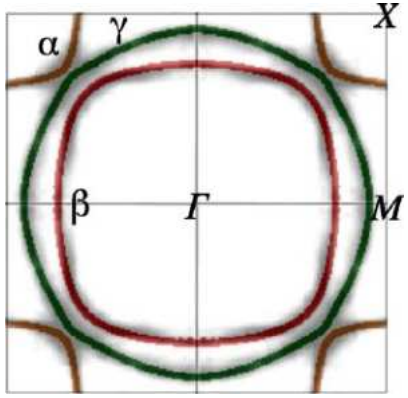
what can we do so far?

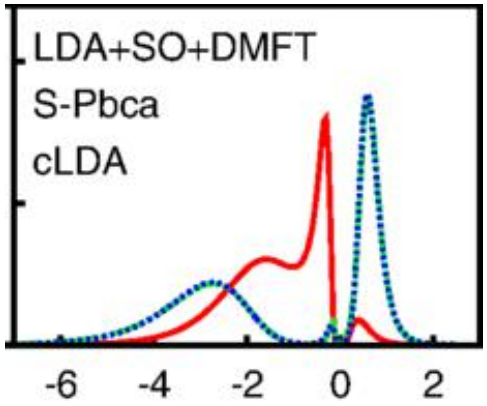
orbital order



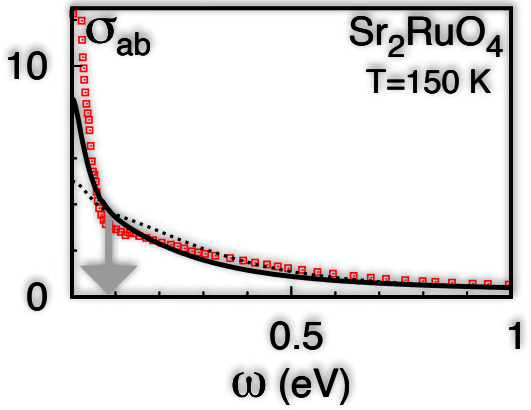
Fermi surface



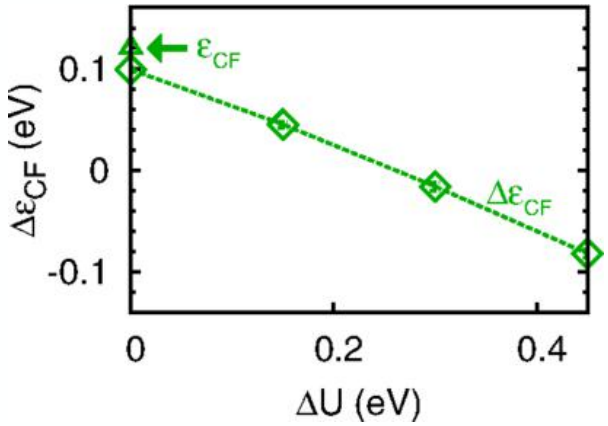
spin-orbit



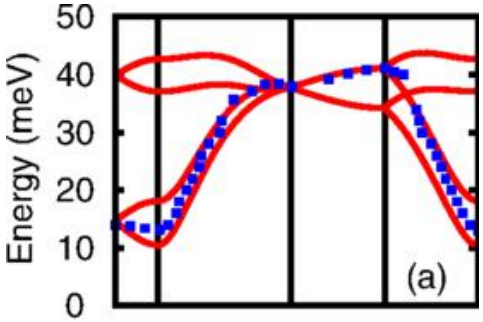
conductivity



realistic Coulomb



spin waves



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PART 4

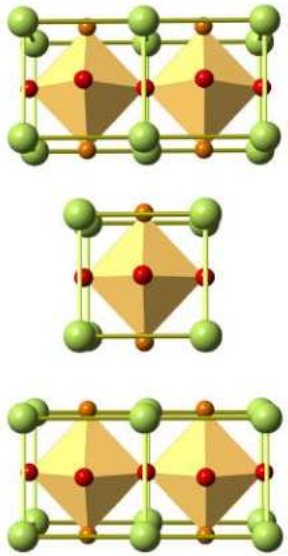
two paradigmatic examples

(a) Sr_2RuO_4 and its family: modeling evolution and unifying picture
(b) origin of orbital ordering: two theories, same prediction

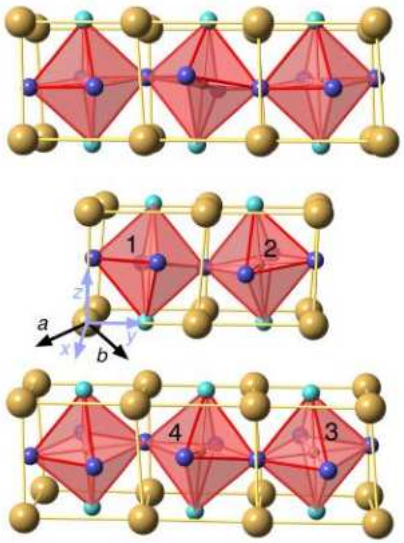
Sr_2RuO_4 and its family: what is the minimal model ?

correlated materials: not only W/U

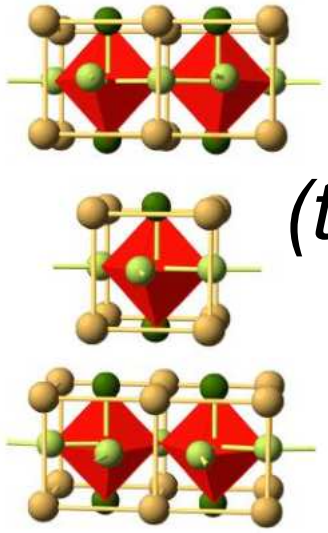
Sr_2RuO_4 Ca_2RuO_4 Sr_2RhO_4 Sr_2IrO_4

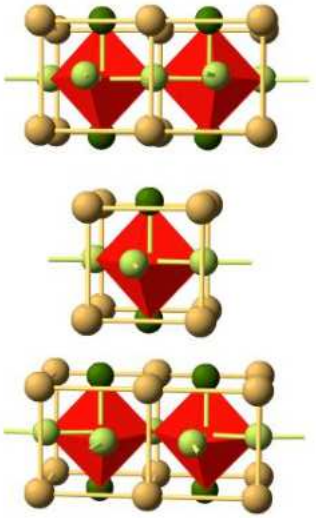


$(t_{2g})^4$



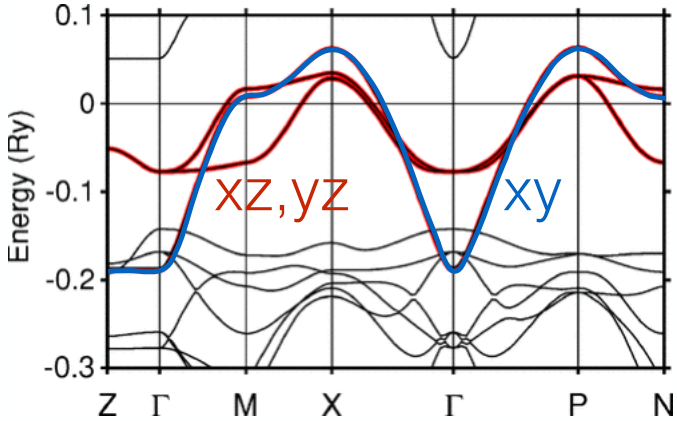
$(t_{2g})^5$





metal *insulator* *metal* *insulator*

orbital physics: interplay of spin, orbital, charge, lattice degrees of freedom in the presence of correlations



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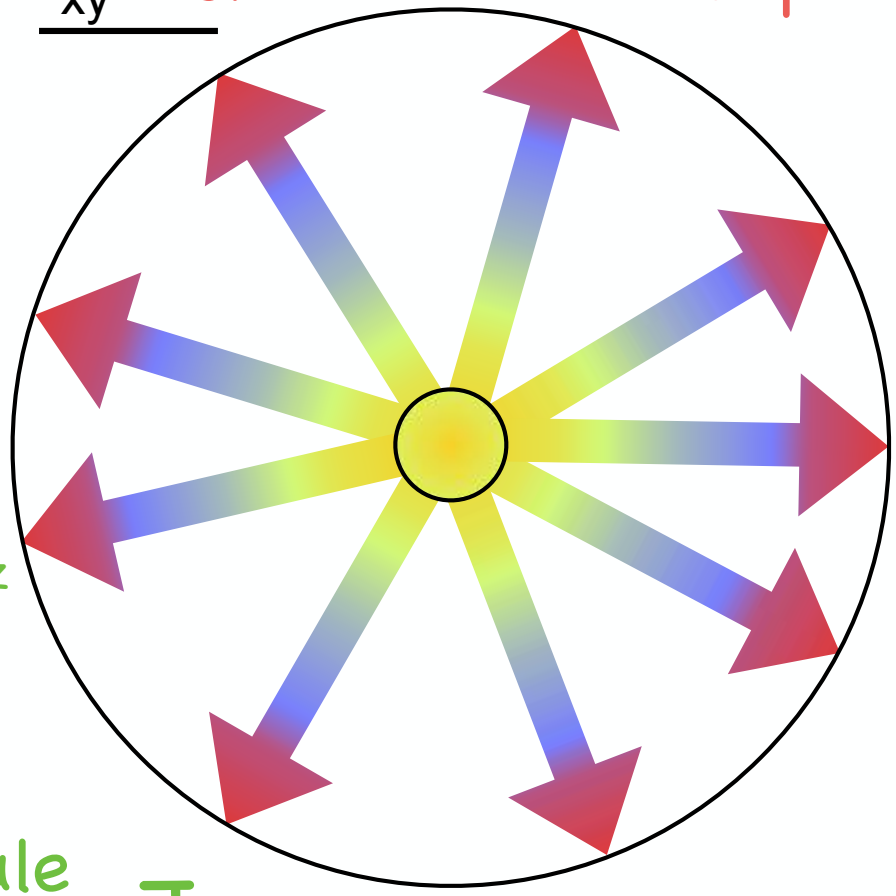
many energy scales are similar



crystal-field splitting

xz/yz

xy

ε_{CF}

spin-orbit coupling

λ

screened Coulomb repulsion

U

dU=U_{xy}-$U_{xz/yz}$

n filling

t_{2g}^n

$W_{xz/yz}$

W_{xy}~2$W_{xz/yz}$

Hund's rule coupling

J

xy vH singularity ?

minimal model evolved with time

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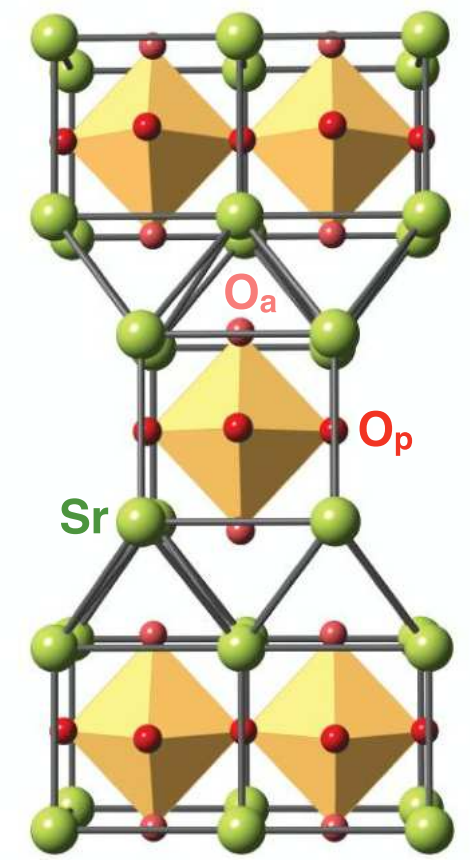
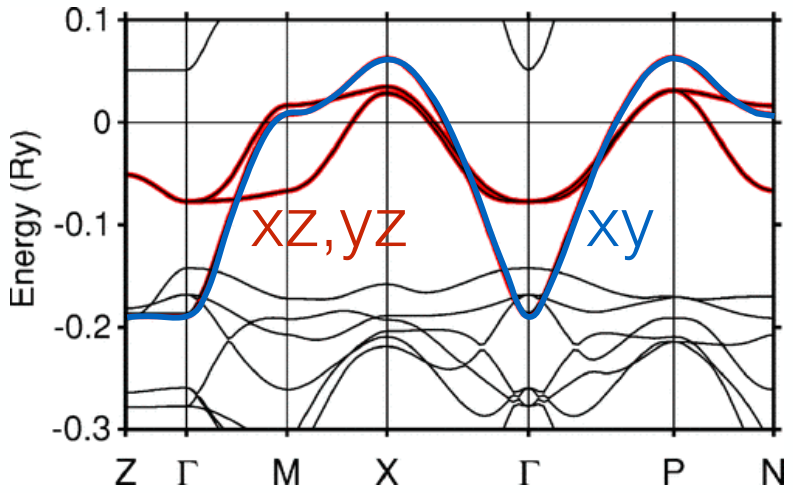
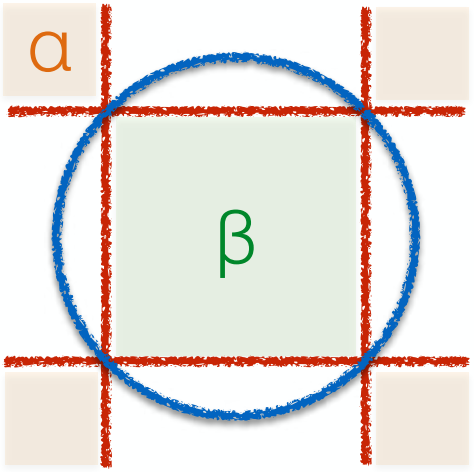
Sr_2RuO_4: Fermi surface riddle





Ru t_{2g}^4

$W_{xy} \sim 2W_{xz/yz}$



Fermi surface

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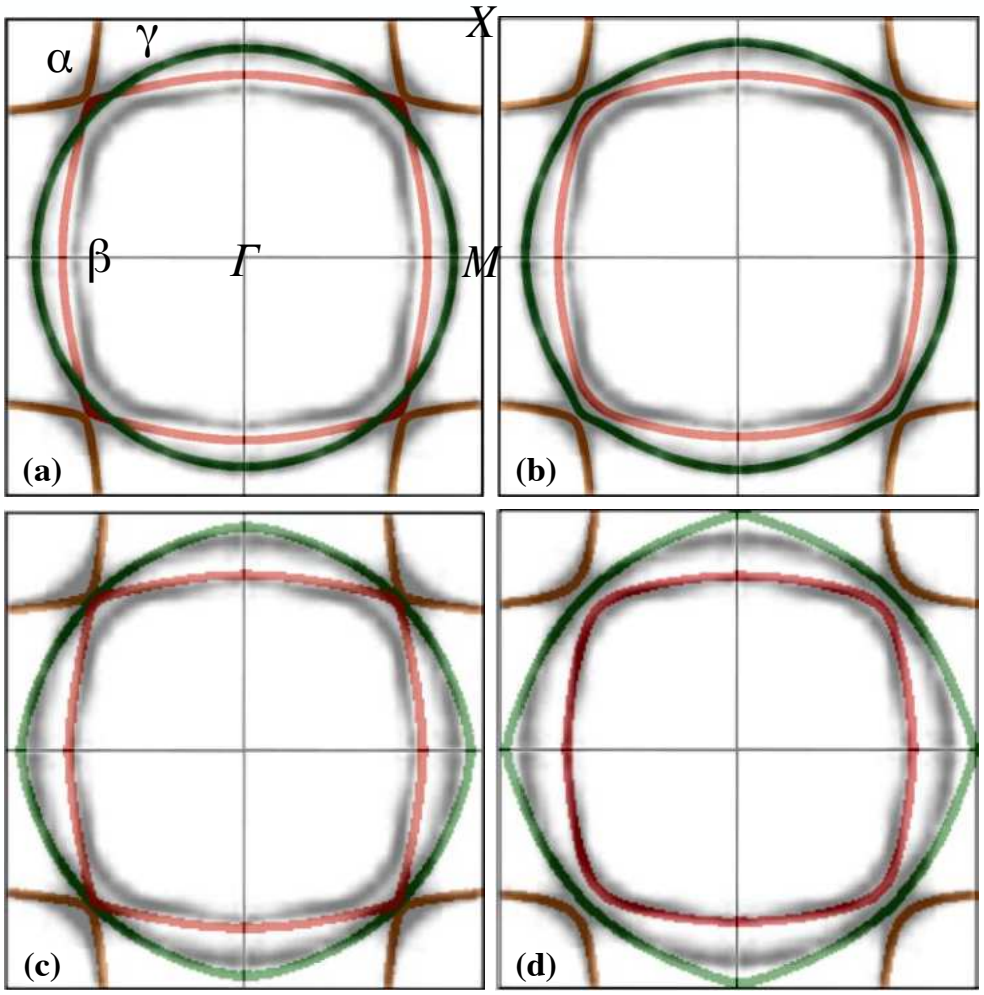
correlations do not improve agreement

experiments (grey map): A. Damascelli et al., Phys. Rev. Lett. **85,** 5194 (2000)

LDA

ε_{CF}



LDA+SO

λ

LDA+DMFT

ε_{CF}

$\varepsilon_{CF} + \Delta\varepsilon_{CF}$

$\sim 2\varepsilon_{CF}$

LDA+SO+DMFT

λ

$\lambda + \Delta\lambda \sim 2\lambda$

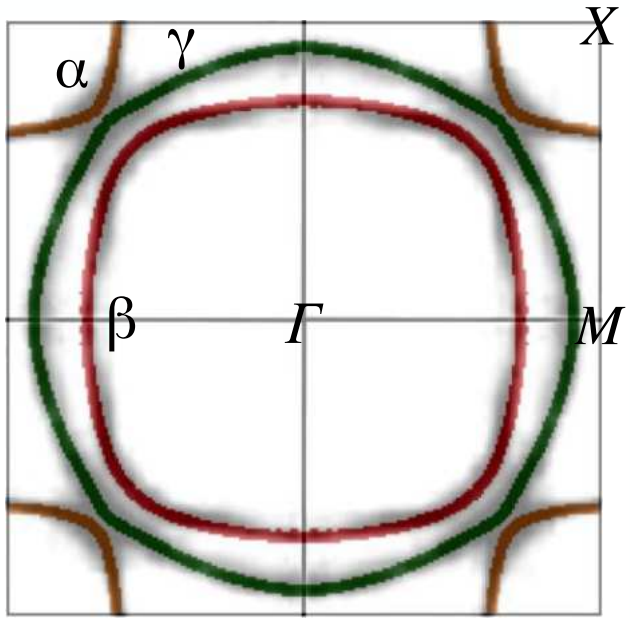
theory: G. Zhang, E. Gorelov, E. Sarvestani, and E. Pavarini, Phys. Rev. Lett. **116**, 106402 (2016)

? a crucial mechanism is still missing ?

the bare Coulomb interaction is spherical dU

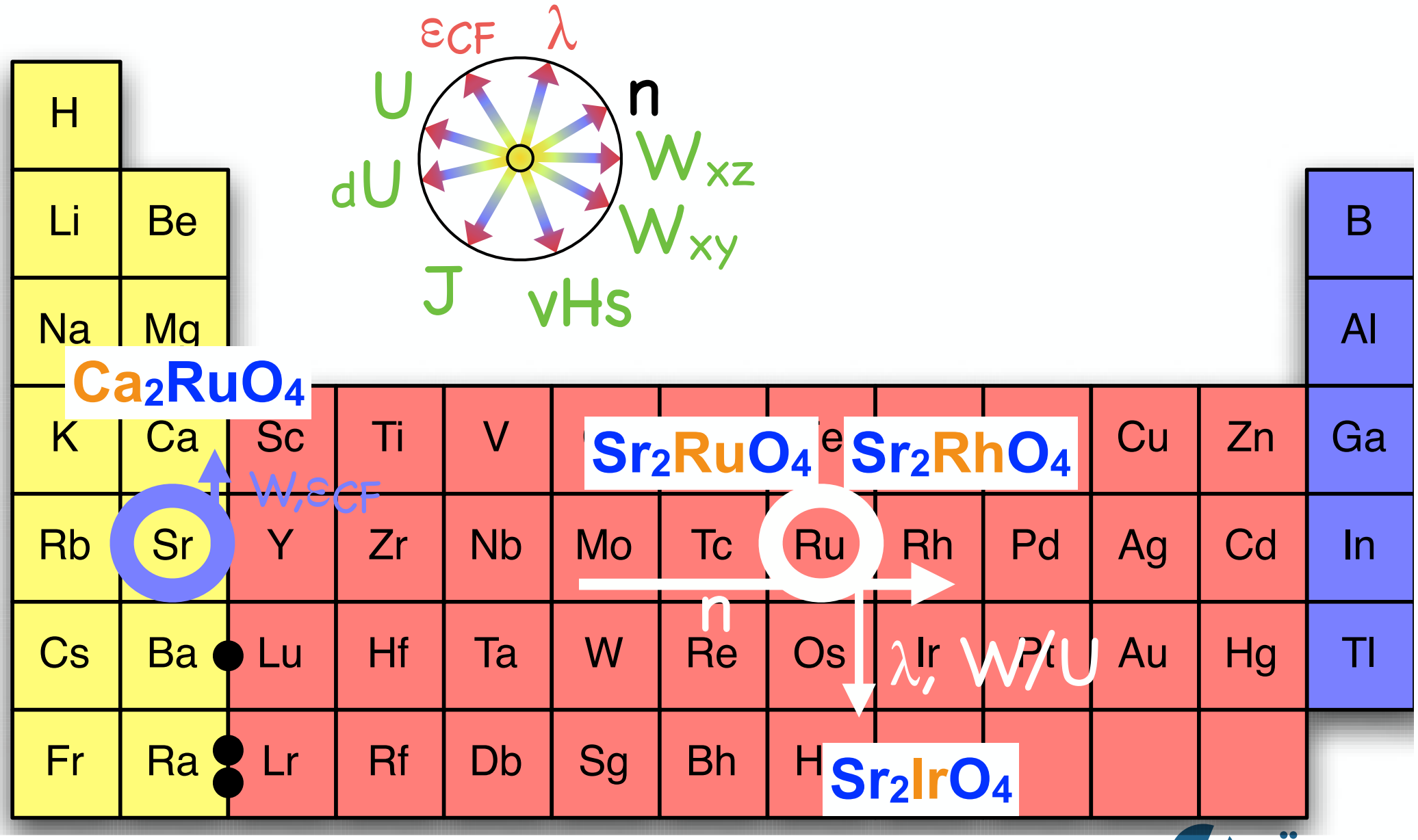
but the **screened** interaction has the symmetry of the site

$\varepsilon_{CF} + \Delta' \varepsilon_{CF} \sim \varepsilon_{CF}$



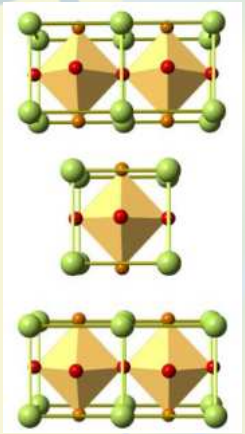
G. Zhang, E. Gorelov, E. Sarvestani, and E. Pavarini, Phys. Rev. Lett. **116**, 106402 (2016)

the Sr_2RuO_4 family: exploit complexity



$(t_{2g})^n$

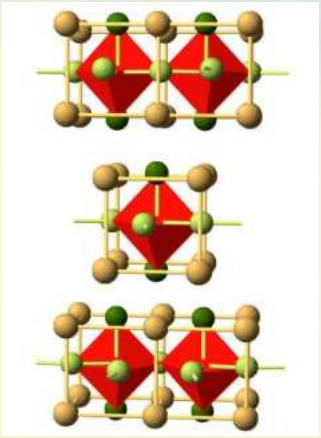
Sr_2RuO_4



correlated metal

$m_{xy} > m_{xz/yz}$

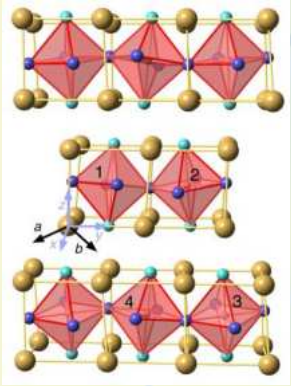
Sr_2RhO_4



weakly correlated metal

only 2-bands
xz/yz at Fermi surface

Ca_2RuO_4



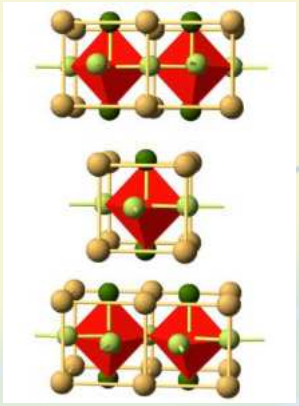
small gap insulator

xy-orbital order

spin-orbit important
only for magnetism

$(t_{2g})^4$

Sr_2IrO_4



spin-orbit Mott insulator
small gap

j=1/2 orbital order
only 1-band half-fillled

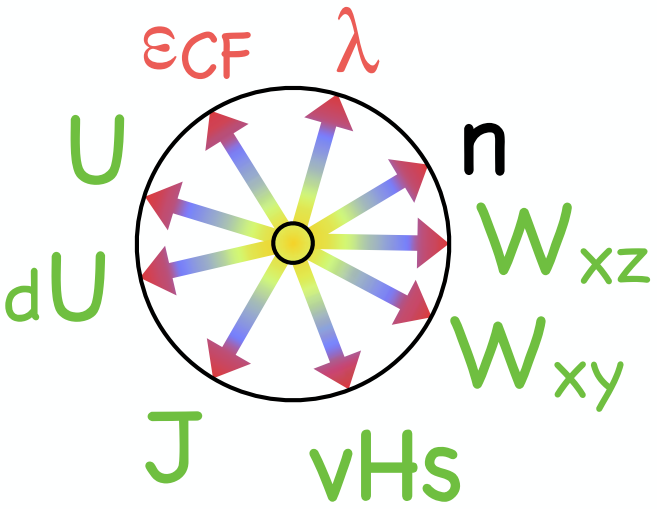
$(t_{2g})^5$

can we build a unifying picture?

yes!

ε_{CF} changes natural occupations

build map of CF (ε_{CF}) effects!



PHYSICAL REVIEW LETTERS **132,** 236505 (2024)

Map of Crystal-Field Effects in Correlated Layered t_{2g}^n Perovskites

Neda Samani,[1] Guoren Zhang,[1,2,3] and Eva Pavarini[1,4]
[1]*Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany*
[2]*School of Physics and Technology, Nantong University, Nantong 226019, People's Republic of China*
[3]*Key Laboratory of Materials Physics, Institute of Solid State Physics, HFIPS, Chinese Academy of Sciences, Hefei 230031, People's Republic of China*
[4]*JARA High-Performance Computing, Forschungszentrum Jülich, Germany*

(natural orbitals=eigenvalues of occupation matrix)

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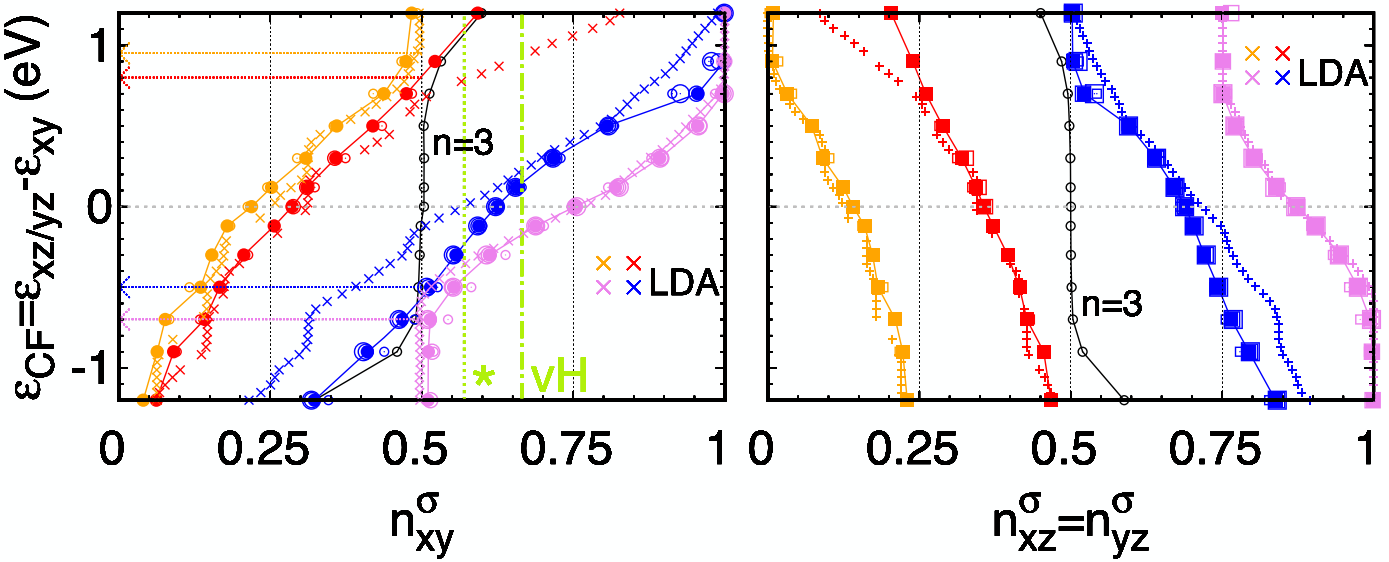
ε_{CF} changes natural occupations

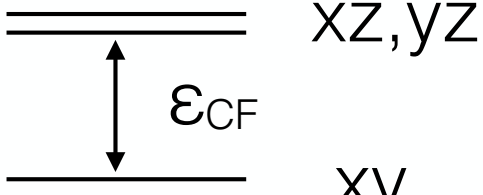
$\lambda = 0$ natural orbitals=xy,xz/yz

t_{2g}^{n}



n=1
n=2
n=4
n=5

CF splitting



$n=n_{xy}+2n_{xz/yz}$

crosses: U=0

closed symbols: finite U

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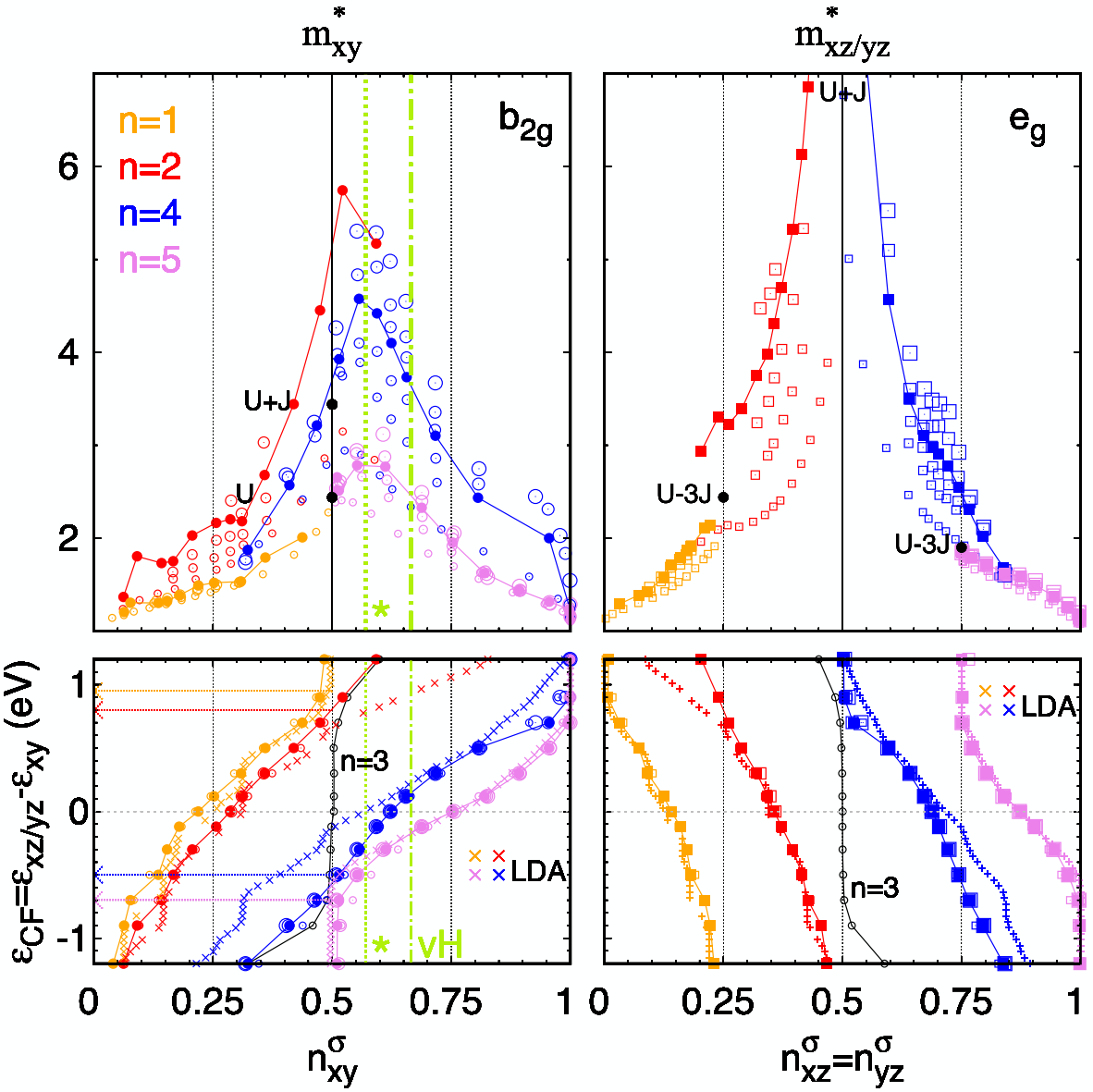
map of crystal-field splitting: effective masses

t_{2g}^n

$\lambda = 0$

n=1
n=2
n=4
n=5



m diverges: insulator*

m=1: not correlated*

moderate (realistic) U,J, fixed U/W

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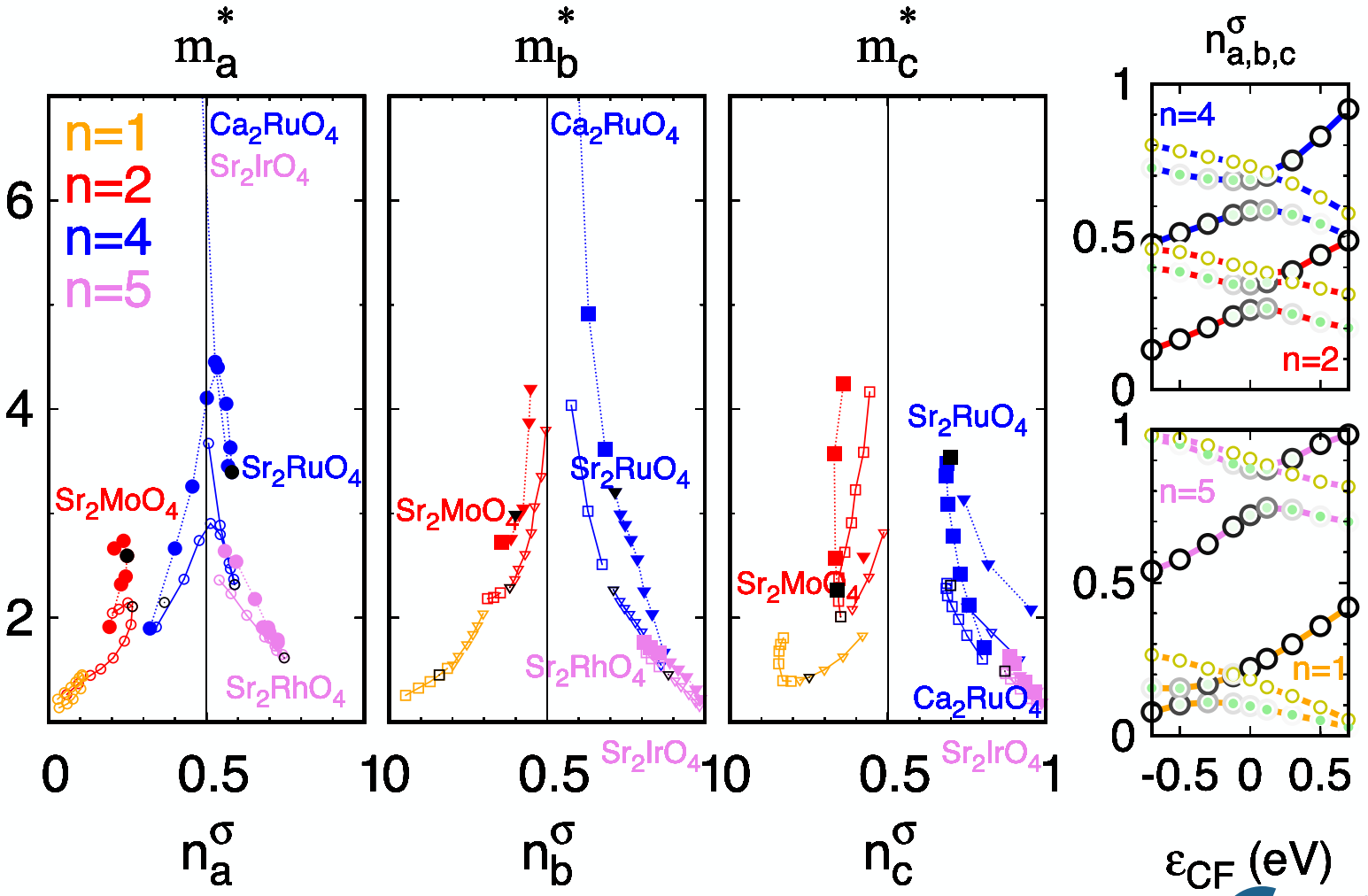
switch on moderate spin-orbit coupling

λ as in Sr_2RuO_4 (4d)



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what is the origin of orbital ordering in materials ?

two theories, same experimental results

spontaneous ordering of orbitals

Crystal structure and magnetic properties of substances with orbital degeneracy

K. I. Kugel' and D. I. Khomskiĭ
P. N. Lebedev Physics Institute
(Submitted November 13, 1972)
Zh. Eksp. Teor. Fiz. **64**, 1429-1439 (April 1973)

Exchange interaction in magnetic substances containing ions with orbital degeneracy is considered. It is shown that, among with spin ordering, superexchange also results in cooperative ordering of Jahn-Teller ion orbitals, which, generally speaking, occurs at a higher temperature and is accompanied by distortion of the lattice (which is a secondary effect here). Concrete studies are performed for substances with a perovskite structure ($KCuF_3$, $LaMnO_3$, MnF_3). The effective spin Hamiltonian is obtained for these substances and the properties of the ground state are investigated. The orbital and magnetic structures obtained in this way without taking into account interaction with the lattice are in accord with the structures observed experimentally. The approach employed also permits one to explain the strong anisotropy of the magnetic properties of these compounds and to obtain a reasonable estimate for the critical temperatures.

$KCuF_3$

$LaMnO_3$

$$H_{SE}^{ii'} = J_{SS} S_i \cdot S_{i'} + J_{OO} O_i O_{i'} + J_{SO} (O_i O_{i'})(S_i \cdot S_{i'})$$

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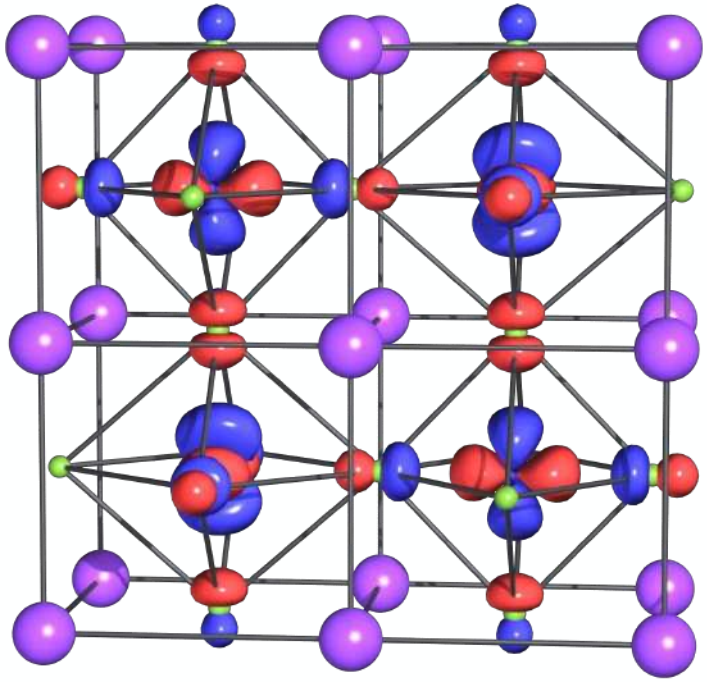
the secondary effect

co-operative Jahn-Teller-like distortion

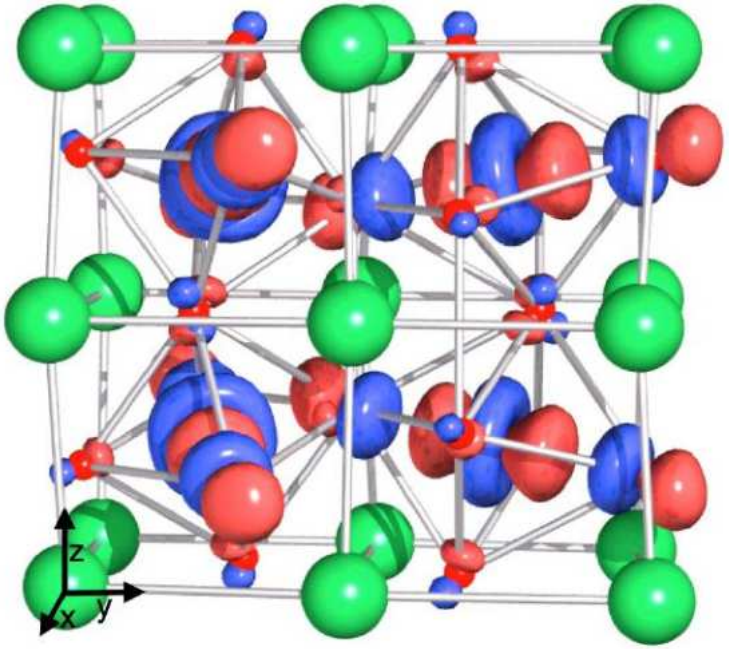
$KCuF_3$



$t_{2g}^6 e_g^3$

G-type OO

$LaMnO_3$



$t_{2g}^3 e_g^1$

C-type OO

orbital ordering and orbital physics

CORRELATED ELECTRON SYSTEMS

REVIEW

Orbital Physics in Transition-Metal Oxides

Y. Tokura[1,2] and N. Nagaosa[1]

An electron in a solid, that is, bound to or nearly localized on the
atomic site, has three attributes: charge, spin, and orbital. Th
represents the shape of the electron cloud in solid. In transiti
oxides with anisotropic-shaped d-orbital electrons, the Coulomb
tion between the electrons (strong electron correlation effe
importance for understanding their metal-insulator transitions a
erties such as high-temperature superconductivity and colossal
toresistance. The orbital degree of freedom occasionally plays a
tant role in these phenomena, and its correlation and/or order
transition causes a variety of phenomena through strong coupl
charge, spin, and lattice dynamics. An overview is given here
"orbital physics," which will be a key concept for the science a
nology of correlated electrons.

[PDF] 1 Metal-Insulator Transition and Orbital Order in ... - arXiv...

arxiv.org/pdf/0811.3378

VOLUME 85, NUMBER 18 PHYSICAL REVIEW LETTERS 30 OCTOBER 2000

Orbital Liquid in Three-Dimensional Mott Insulator: $LaTiO_3$

G. Khaliullin[1,2] and S. Maekawa[2]

[1]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*

[2]*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

(Received 5 June 2000)

of spin and orbital states in Mott insulator $LaTiO_3$. The spin-orbital superexchange
(t_{2g}) ions in cubic crystal suffers from a pathological degeneracy of orbital states
Quantum effects remove this degeneracy and result in the formation of the coherent
the orbital moment of t_{2g} level is fully quenched. We find a finite gap for orbital
isordered state of local degrees of freedom on unfrustrated, simple cubic lattice is
al liquid state naturally explains observed anomalies of $LaTiO_3$.

Electronic reconstruction at an interface between a Mott insulator and a band in

Satoshi Okamoto & Andrew J. Millis

*Department of Physics, Columbia University 538
New York 10027, USA*

**Surface science is an important and we
materials science involving the study**

PRL **99,** 156401 (2007) PHYSICAL REVIEW LETTERS week ending 12 OCTOBER 2007

Superexchange Interaction in Orbitally Fluctuating RVO_3

J.-S. Zhou and J. B. Goodenough

Texas Materials Institute, University of Texas at Austin, Austin, Texas 78712, USA

J.-Q. Yan

Ames Laboratory, Ames, Iowa 50011, USA

Y. Ren

Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA

(Received 18 May 2007; published 8 October 2007)

103, 067205 (2009) PHYSICAL RE

Magnetically Hidden Order of Kra

George Jackeli* and Giniyat Khaliullin

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

(Received 10 June 2009; published 7 August 2009)

We formulate and study an effective Hamiltonian for low-energy Kramers doublets of d^1 ions on a square lattice. We find that the system exhibits a magnetically hidden order in which the expectation values of the local spin and orbital moments both vanish. The order parameter responsible for a time-reversal symmetry breaking has a composite nature and is a spin-orbital analog of a magnetic octupole. We argue that such a hidden order is realized in the layered perovskite Sr_2VO_4.

DOI: 10.1103/PhysRevLett.103.067205 PACS numbers: 75.30.Et, 71.70.Ej, 75.10.Jm

TRANSITION METAL OXIDES

Ferroelectricity driven by orbital order

The discovery that the rotation of the orbital arrangement in manganites induces ferroelectricity exposes an intriguing phase transition that could serve as a blueprint for novel applications.

BERNHARD KEIMER
is at the Max Planck Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany
e-mail: B.Keimer@fkf.mpg.de

Transition metal oxides have fascinated scientists since the 1950s, when the newly developed technique of neutron diffraction was used to show that the compound $La_{1-x}Ca_xMnO_3$ exhibits a rich variety of structural and magnetic phases as the Ca concentration is tuned[1]. The fascination has increased in the wake of the discovery of high-temperature superconductivity in a chemically similar compound,

Figure 1 Possible arrangements of Mn^{3+} *d*-orbitals on a square lattice. The patterns are two-dimensional versions of orbitally ordered states actually observed in manganese oxides. The corresponding magnetic states are indicated by yellow arrows.

ECS Journal of Solid State Science and Technology, 2022 **11** 054004
2162-8777/2022/11(5)/054004/16/$40.00 © 2022 The Electrochemical Society ("ECS"). Published on behalf of ECS by IOP Publishing Limited

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Review—Orbital Physics: Glorious Past, Bright Future

D. I. Khomskii[z]

II. Physikalisches Institut, Universität zu Köln, 50937 Köln, Germany

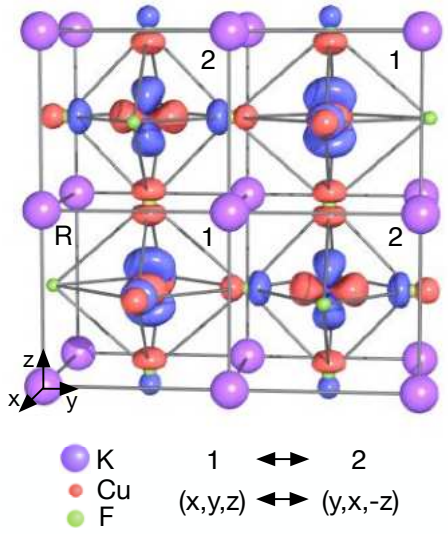
Transition metal (TM) compounds present a very big class of materials with quite diverse properties. Ther
and systems with insulator–metal transitions among them; most magnetic systems are TM compounds; t
superconductors among them. Their very rich properties are largely determined by the strong interplay
freedom: charge; spin; orbital; lattice. Orbital effects play a very important role in these systems—and not o
of this field, initiated by Goodenough almost 70 years ago, turned out to be very fruitful and produced a lot
this short review I discuss the basics of orbital physics and summarize the main achievements in th
Goodenough played a pivotal role, and which are nowadays widely used to explain many properties of TM c
part of the text I discuss novel developments and perspectives in orbital physics, which is still a very a
constantly producing new surprises.

but there is the Jahn-Teller theorem

Crystal Distortion in Magnetic Compounds

JUNJIRO KANAMORI*
Institute for the Study of Metals, University of Chicago, Chicago 37, Illinois

The crystal distortion which arises from the Jahn-Teller effect is discussed in several examples. In the case of compounds containing Cu^{2+} or Mn^{3+} at octahedral sites, the lowest orbital level of these ions is doubly degenerate in the undistorted structure, and there is no spin-orbit coupling in this level. It is shown that, introducing a fictitious spin to specify the degenerate orbital states, we can discuss the problem by analogy with the magnetic problems. The "ferromagnetic" and "antiferromagnetic" distortions are discussed in detail. The transition from the distorted to the undistorted structure is of the first kind for the former and of the second kind for the latter. Higher approximations are discussed briefly. In compounds like FeO, CoO, and $CuCr_2O_4$, the lowest orbital level is triply degenerate, and the spin-orbit coupling is present in this level. In this case the distortion is dependent on the magnitude of the spin-orbit coupling relative to the strength of the Jahn-Teller effect term. The distortion at absolute zero temperature and its temperature dependence are discussed.

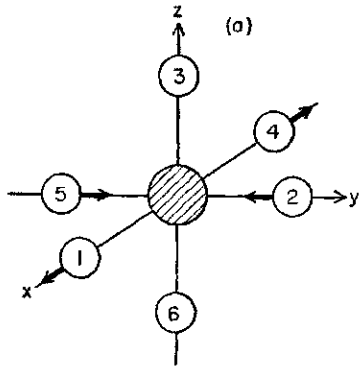


$E_0 + \Delta/2$

$E_0 - \Delta/2$

Δ_{CF}

e_g states



$$|\theta\rangle = \sin\frac{\theta}{2}|3z^2 - 1\rangle + \cos\frac{\theta}{2}|x^2 - y^2\rangle$$

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are there true Kugel-Khomskii materials ?

a chicken-and-egg problem

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solution of chicken and egg problem

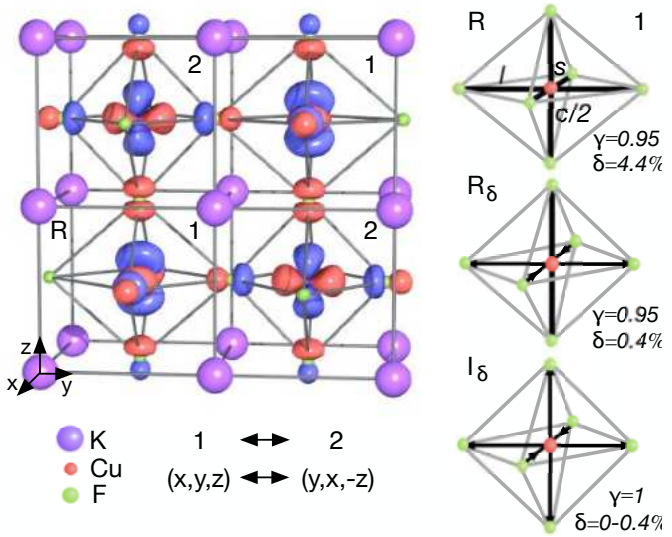
PRL **101,** 266405 (2008) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2008

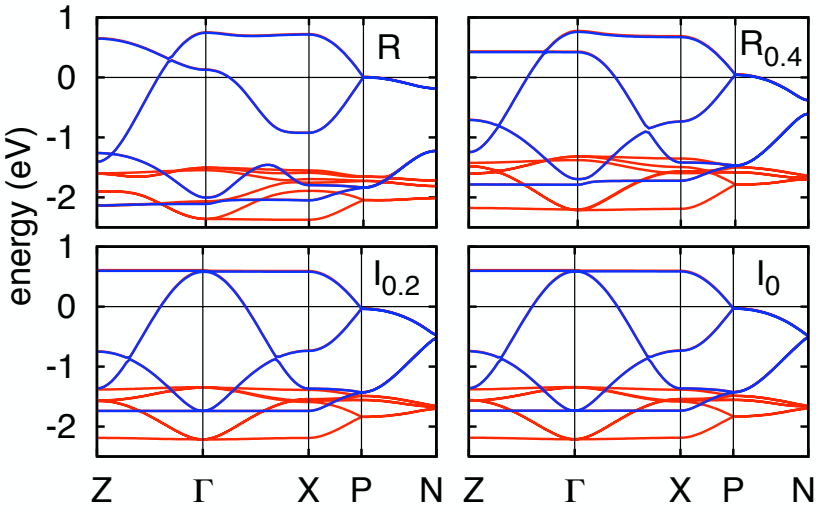
Mechanism for Orbital Ordering in $KCuF_3$

E. Pavarini,[1] E. Koch,[1] and A. I. Lichtenstein[2]

[1]*Institut für Festkörperforschung and Institute for Advanced Simulation, Forschungzentrum Jülich, 52425 Jülich, Germany*
[2]*Institute of Theoretical Physics, University of Hamburg, Jungiusstrasse 9, 20355 Hamburg, Germany*

(Received 18 August 2008; published 31 December 2008)





to single out KK effects:

(i) progressively reduce distortion (hence CF splitting)

(ii) calculate order parameter vs temperature

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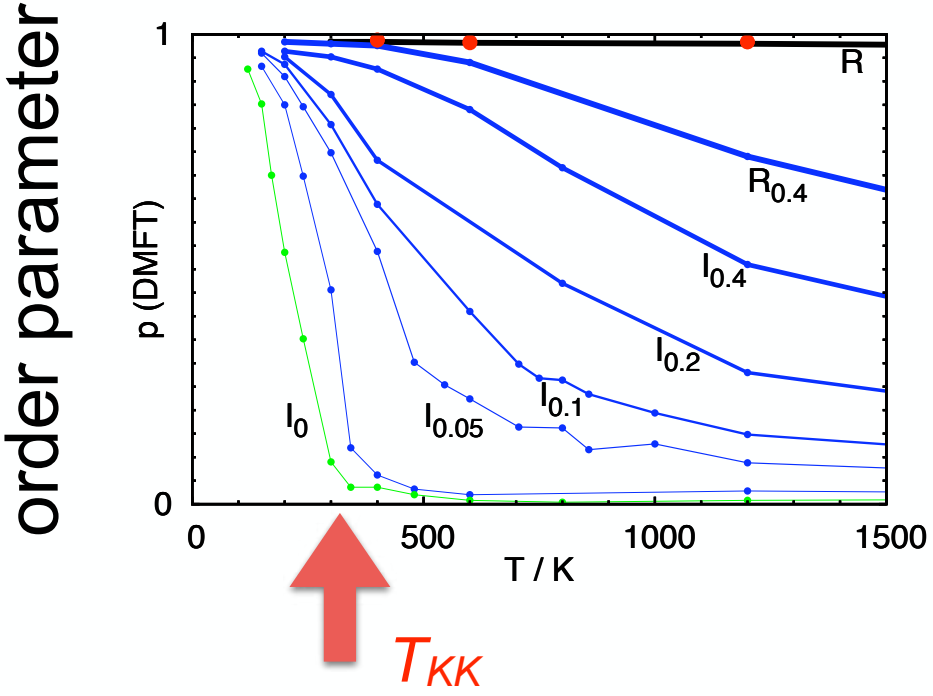
KK SE large but not sufficient alone

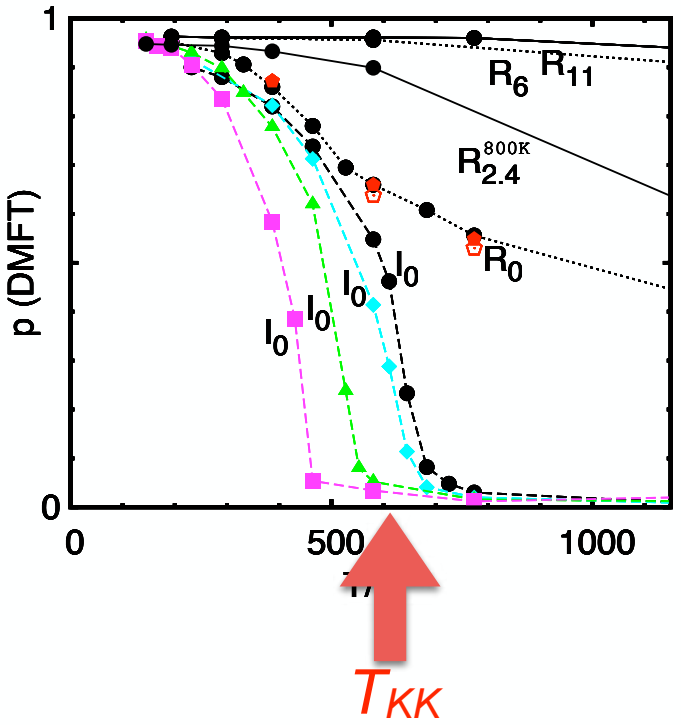
$KCuF_3$



Phys. Rev. Lett. **101**, 266405 (2008)

$T_{KK} \sim 350\ K \ll T_{OO} \sim 1400\ K$

$LaMnO_3$



Phys. Rev. Lett. **104**, 086402 (2010)

$T_{KK} \sim 600\ K \ll T_{OO} \sim 1200\ K$

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RVO_3, t_{2g}^2 : a unique series

PRL **99,** 156401 (2007) PHYSICAL REVIEW LETTERS week ending 12 OCTOBER 2007

Superexchange Interaction in Orbitally Fluctuating *R*VO_3

J.-S. Zhou and J. B. Goodenough
Texas Materials Institute, University of Texas at Austin, Austin, Texas 78712, USA

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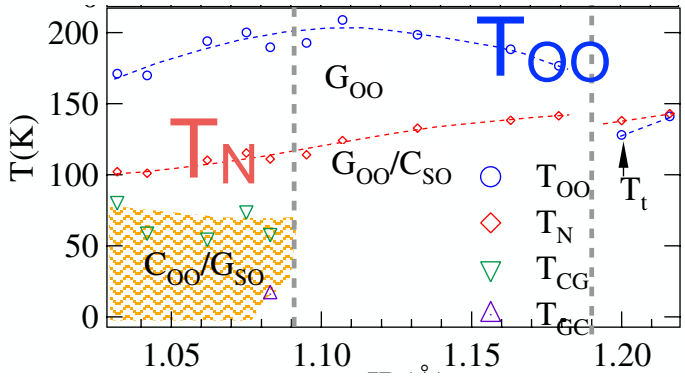

$LaVO_3$

R_I =ionic radius

inversion of T_N and T_{OO}

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multipolar super-exchange expansion

$$\hat{H}_{\mathrm{SE}}^{i,j} = \sum_{qq'} \sum_{\nu\nu'} \sum_{rr'} \sum_{\mu\mu'} \hat{\tau}_i^{r\mu;q\nu} D_{r\mu,r'\mu'}^{ij;q\nu} \hat{\tau}_j^{r'\mu';q\nu}$$

r: orbital rank q:spin rank

orbital monopolar (r=r'=0)

orbital multipolar (r+r'>1)

$$\hat{H}_{\mathrm{SE}}^{i,j} = \hat{H}_{C_{ij}} + \hat{H}_{O_i O_j} + \hat{H}_{S_i S_j} + \hat{H}_{S_i S_j O_i O_j}$$

spin monopolar (q=q'=0)

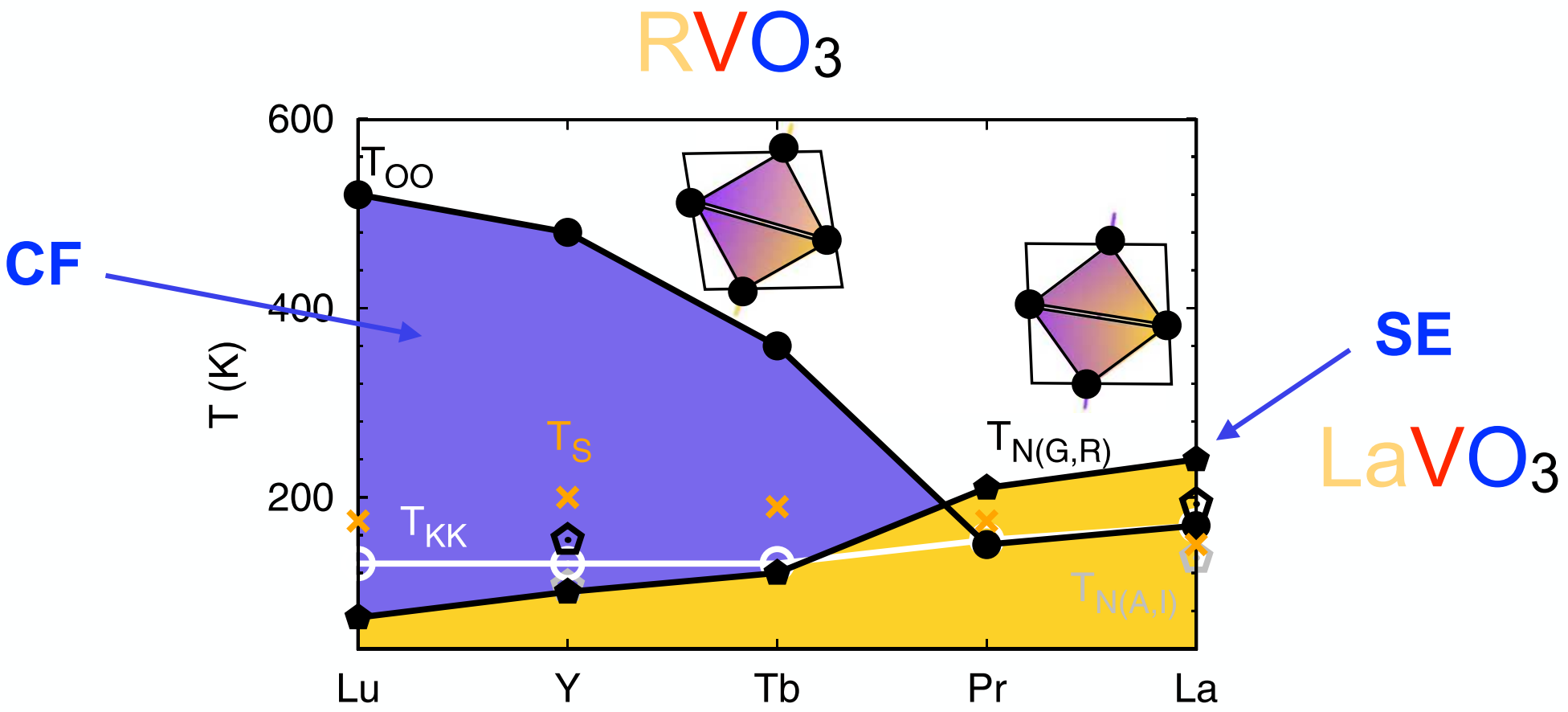
spin dipolar (q=q'=1)

S_iS_j term main contribution to T_N!

PRB **102**, 035113 (2020); PRB **106**, 115110 (2022); PRL **135**, 026508 (2025)

$LaVO_3$: a true Kugel-Khomskii system

PRL **135**, 026508 (2025)

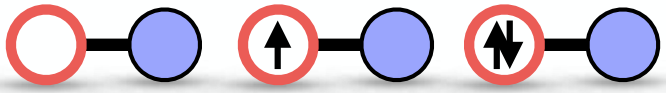


$T_{OO} >> T_{KK}$ R_I =ionic radius $T_{OO} \sim T_{KK}$

V: conclusions

DFT+DMFT

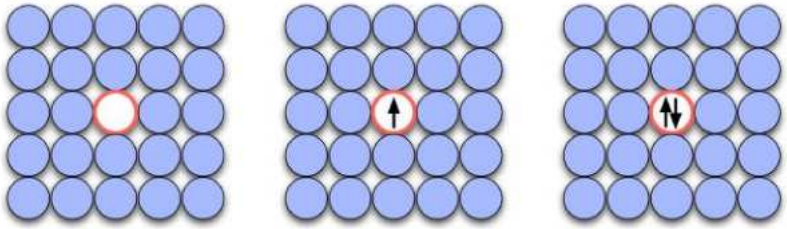
dimer



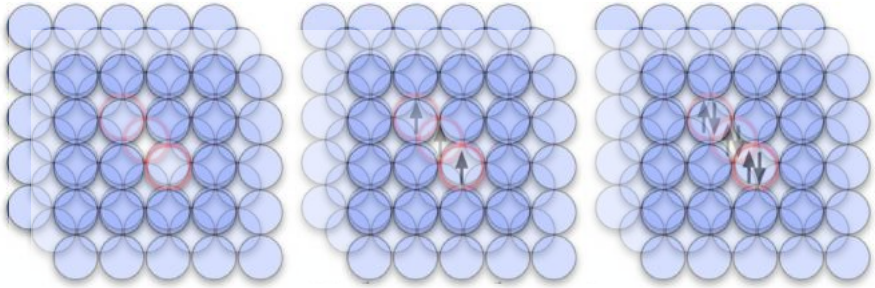
strong-correlations are local

U=V 

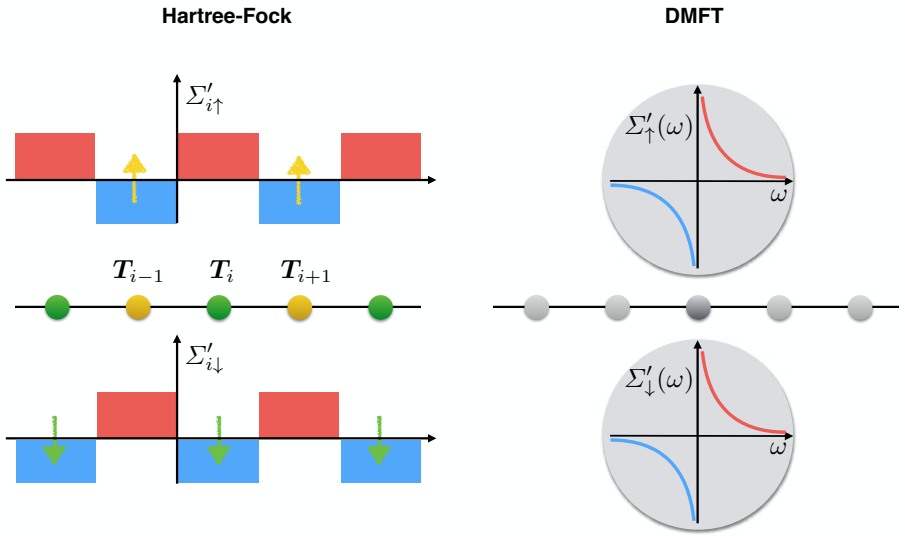
one band



multiband



DFT+U vs DFT+DMFT



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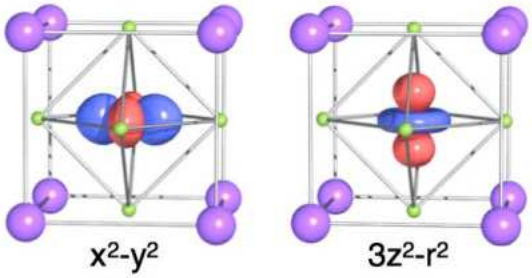
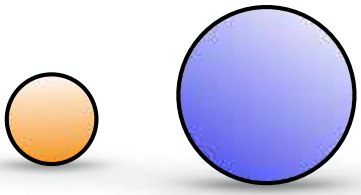
DFT+DMFT

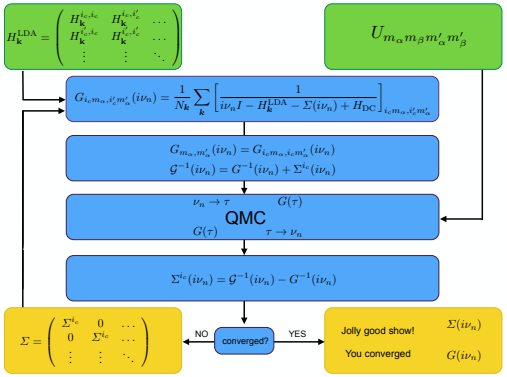
basis choice



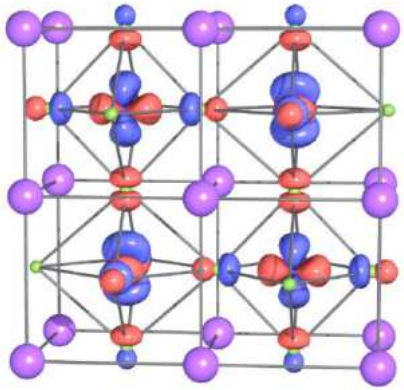
light & heavy electrons



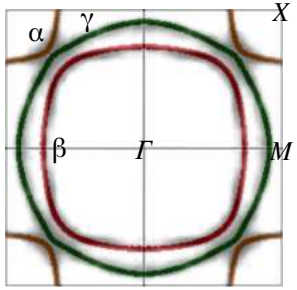
DMFT

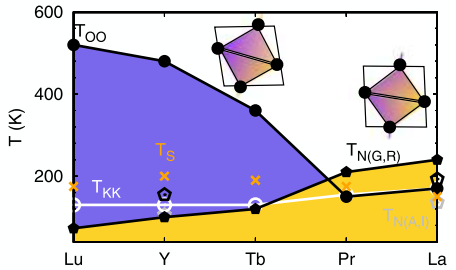


downfolding, localization,
double counting & screening



understanding materials





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