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Mechanism for orbital ordering in transition-metal oxides

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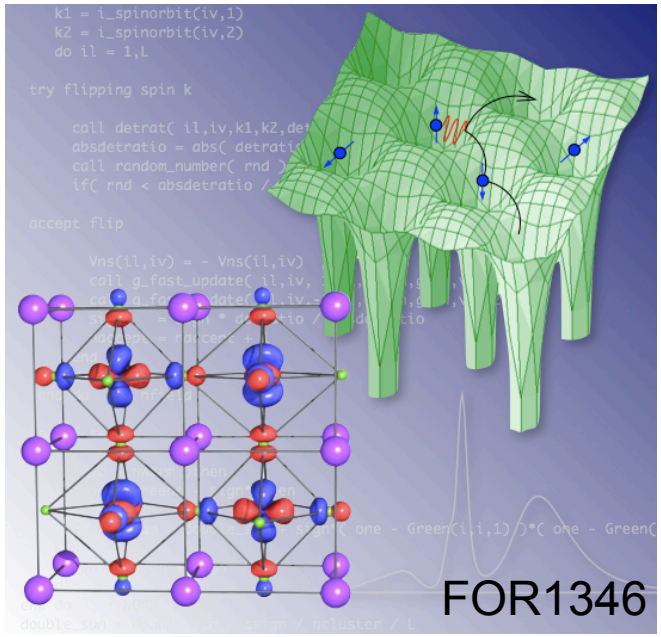

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orbital order

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 specific atomic site, has three attributes: charge, spin, and orbital. The orbital represents the shape of the electron cloud in solid. In transition-metal oxides with anisotropic-shaped d-orbital electrons, the Coulomb interaction between the electrons (strong electron correlation effect) is of importance for understanding their metal-insulator transitions and properties such as high-temperature superconductivity and colossal magnetoresistance. The orbital degree of freedom occasionally plays an important role in these phenomena, and its correlation and/or order-disorder transition causes a variety of phenomena through strong coupling with charge, spin, and lattice dynamics. An overview is given here on this "orbital physics," which will be a key concept for the science and technology of correlated electrons.

When more than two orbitals are involved, a variety of situations can be realized, and this quantum mechanical process depends on the orbitals (*4, 5*). In this way, the spin $\vec{S}$ and the orbital pseudospin $\vec{T}$ are coupled. In more general cases, the transfer integral t_{ij} depends on the direction of the bond ij and also on the pair of the two orbitals a, $b = (x^2 - y^2)$ or $(3z^2 - r^2)$. This gives rise to the anisotropy of the Hamiltonian in the pseudospin space as well as in the real space. For example, the transfer integral between the two neighboring Mn atoms in the crystal lattice is determined

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.

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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,

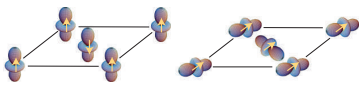

Figure 1 Possible arrangements of Mn^{3+} *d*-orbitals on a square lattice. The patterns are two-dimensional versions of orbitally ordered states actually observed in manganese oxides. The corresponding magnetic states are indicated by yellow arrows.

Ionic relaxation contribution to the electronic reconstruction at the *n*-type $LaAlO_3/SrTiO_3$ interface

Rossitza Pentcheva[1] and Warren E. Pickett[2]
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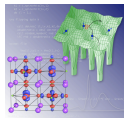
[2]*Department of Physics, University of California, Davis, California 95616, USA*
(Received 30 June 2008; revised manuscript received 19 September 2008; published 7 November 2008)

Density-functional theory calculations reveal that the compensation mechanism at the isolated *n*-type interface in $LaAlO_3/SrTiO_3$ superlattices involves both ionic and electronic degrees of freedom. Strong polar distortions screen the local electric field and reduce the band discontinuity across the interface. We find that the electronic reconstruction depends sensitively on whether structural optimization is performed within GGA (conventional exchange and correlation effects) or GGA+*U* (which includes strong intra-atomic interactions). For a structural optimization within GGA+*U* the excess charge is confined to the interface TiO_2 layer with a charge-ordered, orbitally polarized arrangement of Ti^{3+} and Ti^{4+}. While the charge-ordered phase represents the ground state, optimization within GGA leads to more pronounced lattice polarization, suppression of charge order (with remaining d_{xy}-orbital occupation in the interface layer), and a delocalization of the excess charge

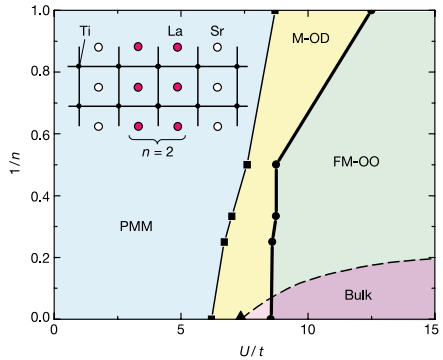


Electronic reconstruction at an interface between a Mott insulator and a band insulator

Satoshi Okamoto & Andrew J. Millis

Department of Physics, Columbia University 538 West 120th Street, New York, New York 10027, USA

Surface science is an important and well-established branch of materials science involving the study of changes in material



RAPID COMMUNICATIONS

PHYSICAL REVIEW B **78**, 201102(R) (2008)

Magnetism, conductivity, and orbital order in $(LaMnO_3)_{2n}/(SrMnO_3)_n$ superlattices

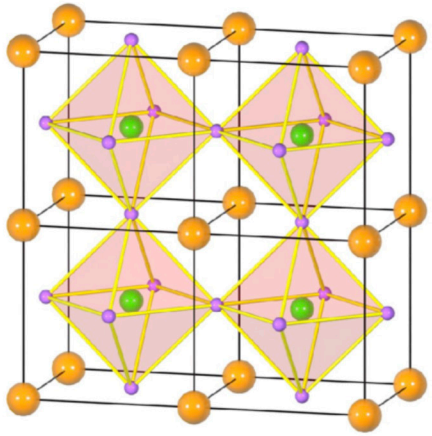
Shuai Dong,[1,2,3] Rong Yu,[1,2] Seiji Yunoki,[4,5] Gonzalo Alvarez,[6] J.-M. Liu,[3] and Elbio Dagotto[1,2]
[1]*Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA*
[2]*Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 32831, USA*
[3]*Nanjing National Laboratory of Microstructures, Nanjing University, Nanjing 210093, China*
[4]*Computational Condensed Matter Physics Laboratory, RIKEN, Wako, Saitama 351-0198, Japan*
[5]*CREST, Japan Science and Technology Agency (JST), Kawaguchi, Saitama 332-0012, Japan*
[6]*Computer Science and Mathematics Division and Center for Nanophase Materials Science, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*
(Received 8 October 2008; published 21 November 2008)

The modulation of charge density and spin order in $(LaMnO_3)_{2n}/(SrMnO_3)_n$ ($n=1-4$) superlattices is studied via Monte Carlo simulations of the double-exchange model. *G*-type antiferromagnetic barriers in the

hallmark of orbital order

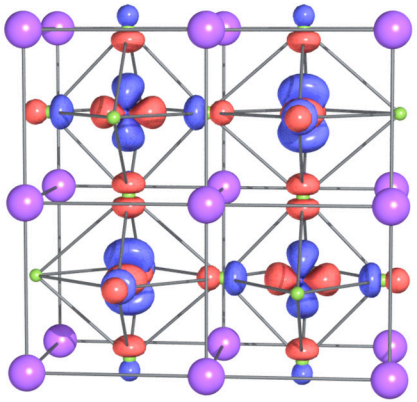
cooperative Jahn-Teller distortions

orbitally degenerate partially filled d states



paradigmatic systems

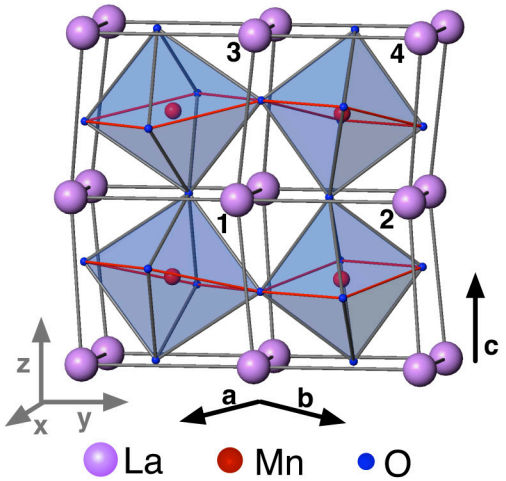
$KCuF_3$



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

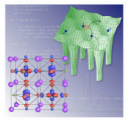
2-fold orbitally degenerate

$LaMnO_3$



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

2-fold orbitally degenerate



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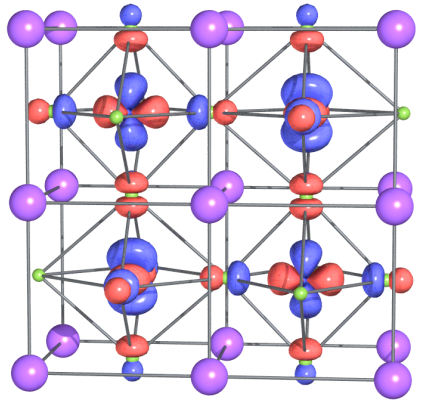
orbital order melting

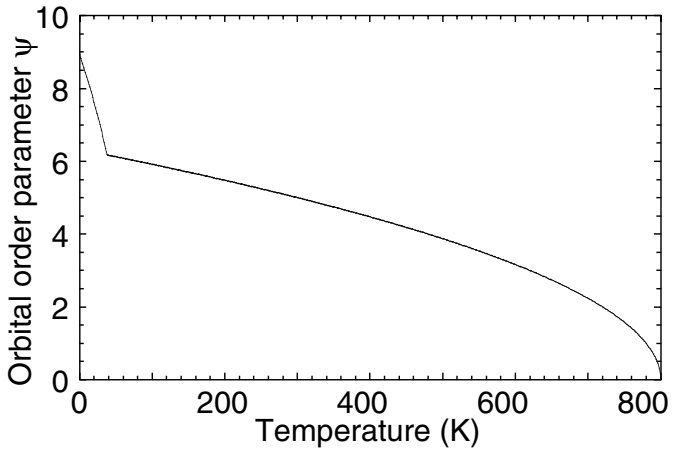
T_N~40 K T_{OO}~800-1400 K

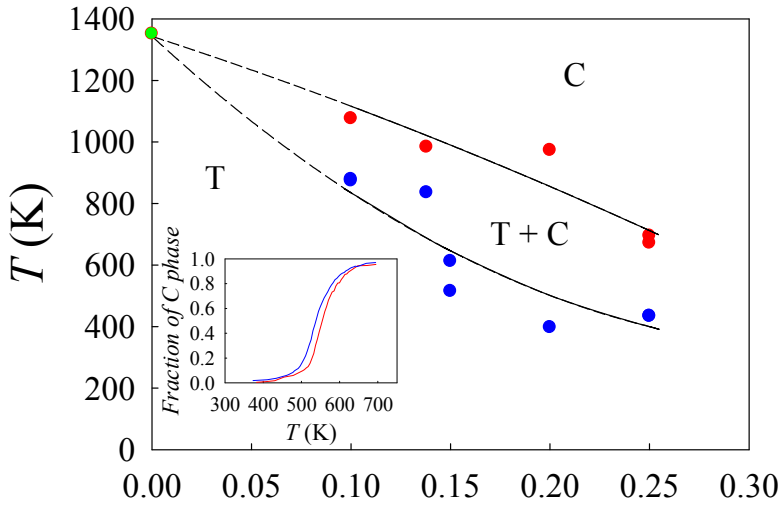


VOLUME 88, NUMBER 10 PHYSICAL REVIEW LETTERS 11 MARCH 2002

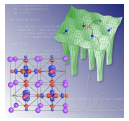
Coupling between Spin and Orbital Degrees of Freedom in $KCuF_3$

L. Paolasini,[1] R. Caciuffo,[2] A. Sollier,[1] P. Ghigna,[3] and M. Altarelli[4]
[1]*European Synchrotron Radiation Facility, B.P. 220, F-38043 Grenoble, France*
[2]*Istituto Nazionale per la Fisica della Materia, Dipartimento di Fisica ed Ingegneria dei Materiali e del Territorio, Università di Ancona, Via Brecce Bianche, I-60131 Ancona, Italy*
[3]*Dipartimento di Chimica Fisica, Università di Pavia, I-27100 Pavia, Italy*
[4]*Sincrotrone Trieste and Abdus Salam International Centre for Theoretical Physics, Trieste, Italy*
(Received 14 August 2001; published 25 February 2002)





Ghigna et al. PRB (2010)



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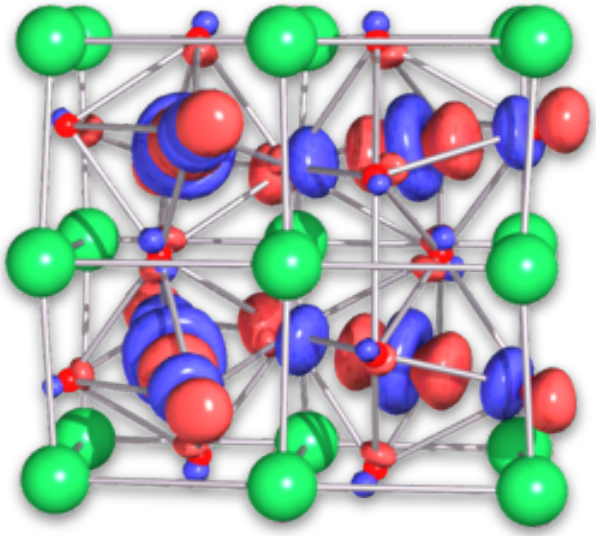
orbital order melting



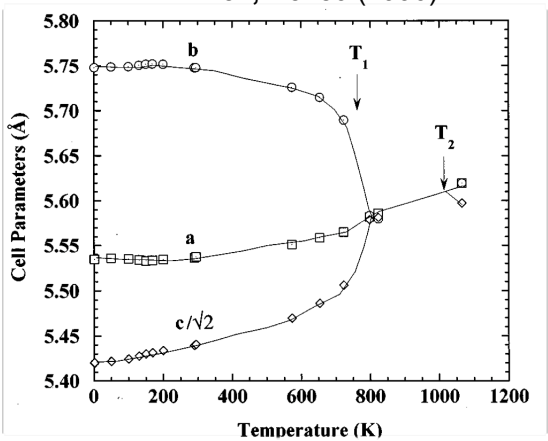
$3l^2-r^2$

OO hallmark: JT distortion

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

$LaMnO_3$

Rodrıguez-Carvajal et al.
PRB **57,** R3189 (1996)



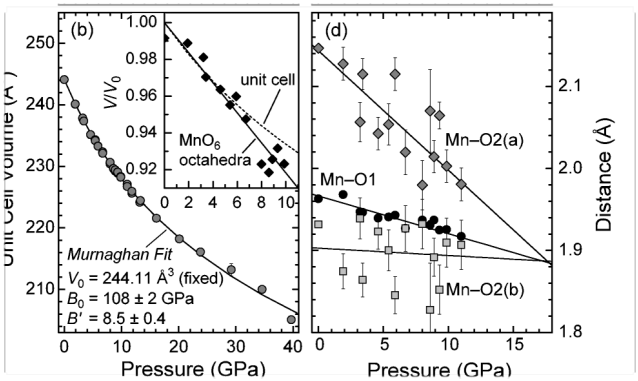
temperature

T_N~140 K

T_{KK}~750 K

$LaMnO_3$

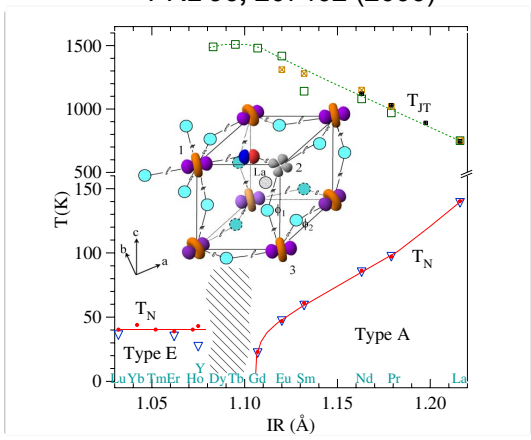
Loa et al.
PRL **87,** 125501 (2001)



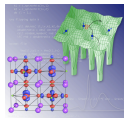
pressure

$RMnO_3$

Zou and Goodenough
PRL **96,** 207402 (2006)



IR radius



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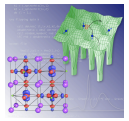
mechanism of orbital-order

what is the origin?

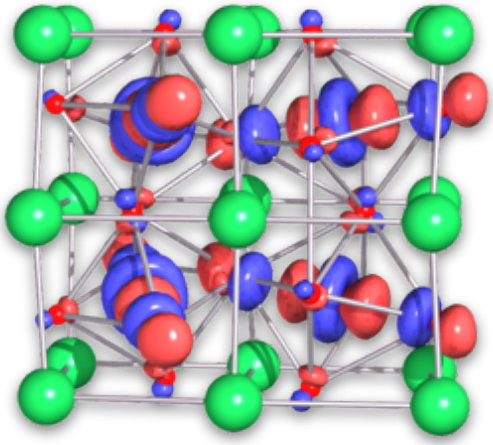
electron-phonon coupling (+*U*)
vs
electronic super-exchange

$KCuF_3$ and $RMnO_3$

super-exchange effects remarkably large
but electron-phonon coupling essential



understanding orbital order



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.

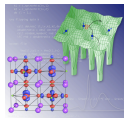
electron-phonon

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.

super-exchange



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electron-phonon coupling

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Institute for the Study of Metals, University of Chicago, Chicago 37, Illinois

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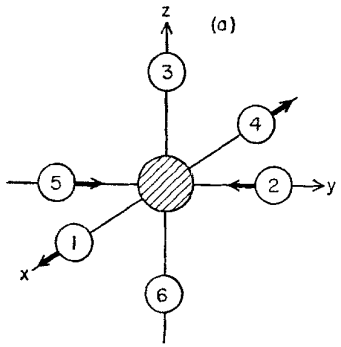
Jahn-Teller theorem



The Normal Mode
Q_2 (Q_2 >0)

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

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

partially filled 2-fold degeenerate e_g shell

$$H = -g\sqrt{C}(\tau_z Q_3 + \tau_x Q_2)$$

τ : pseudospins in orbital space

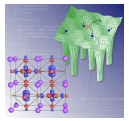
$E_0 + \Delta/2$ ——

Δ

$E_0 - \Delta/2$ —— θ

JT splitting

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



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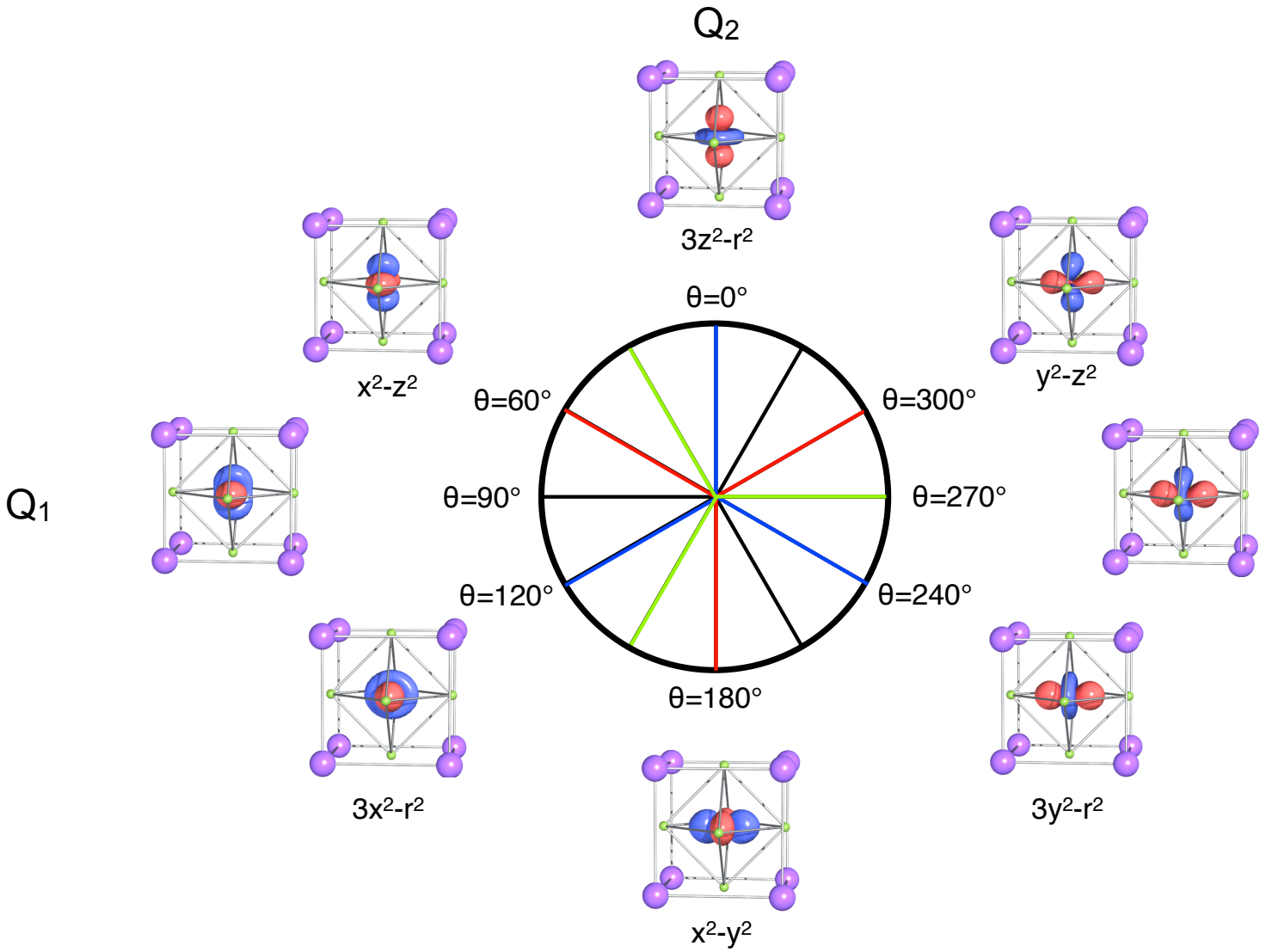
e_g^1 orbitals



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

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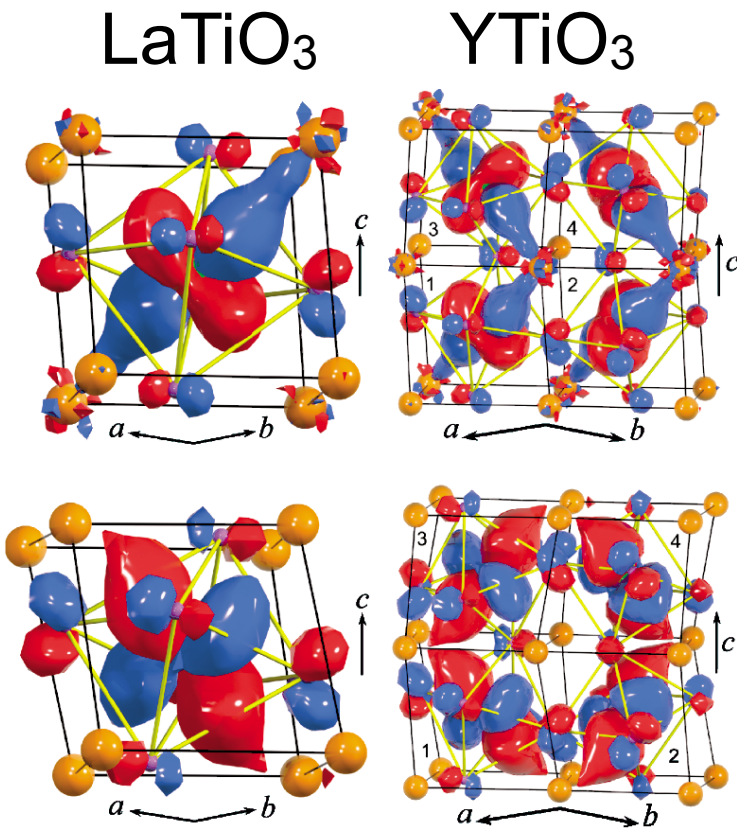
do we need a large crystal-field?

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_{2g}^1

LDA+DMFT 770 K

Δ=200-300 meV

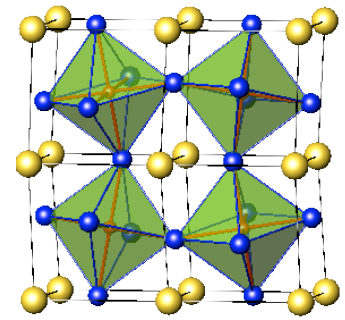
$\Delta + U$ correlation effects open d shell

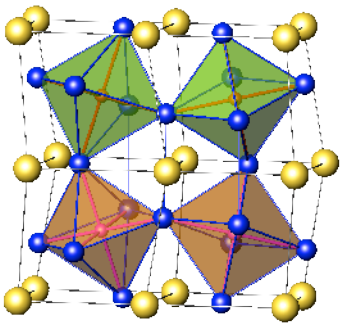
do we need a large crystal-field?

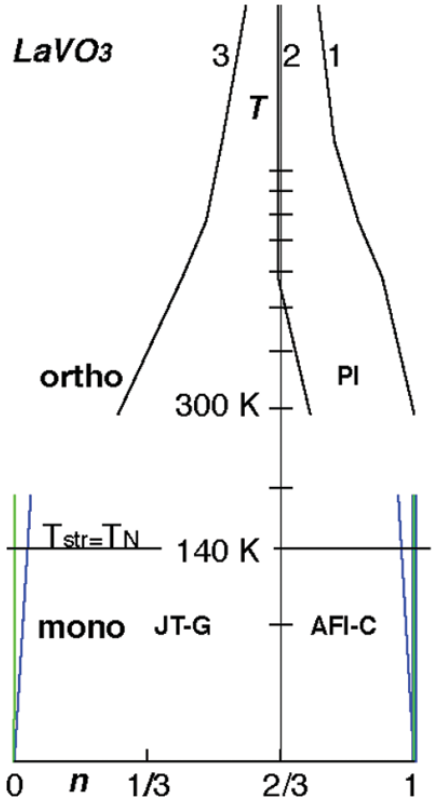
PRL **99,** 126402 (2007) PHYSICAL REVIEW LETTERS week ending 21 SEPTEMBER 2007

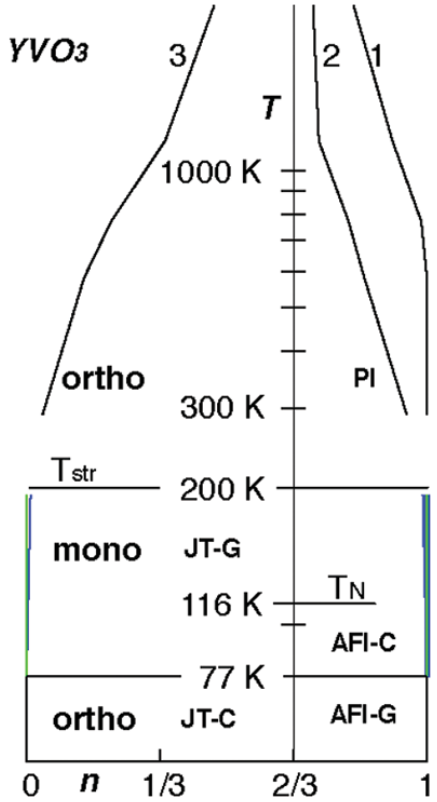
Orbital Fluctuations in the Different Phases of $LaVO_3$ and YVO_3

M. De Raychaudhury,[1,2] E. Pavarini,[3,4] and O. K. Andersen[1]









t_{2g}^2

Δ=100-200 meV

do we need a large crystal-field?

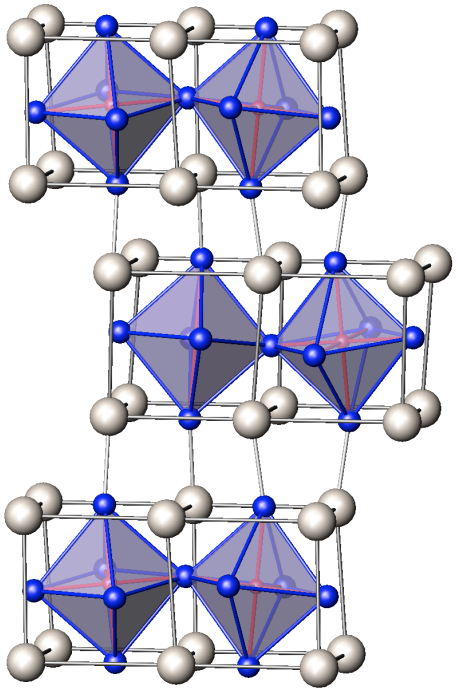
PRL **104,** 226401 (2010) PHYSICAL REVIEW LETTERS week ending 4 JUNE 2010

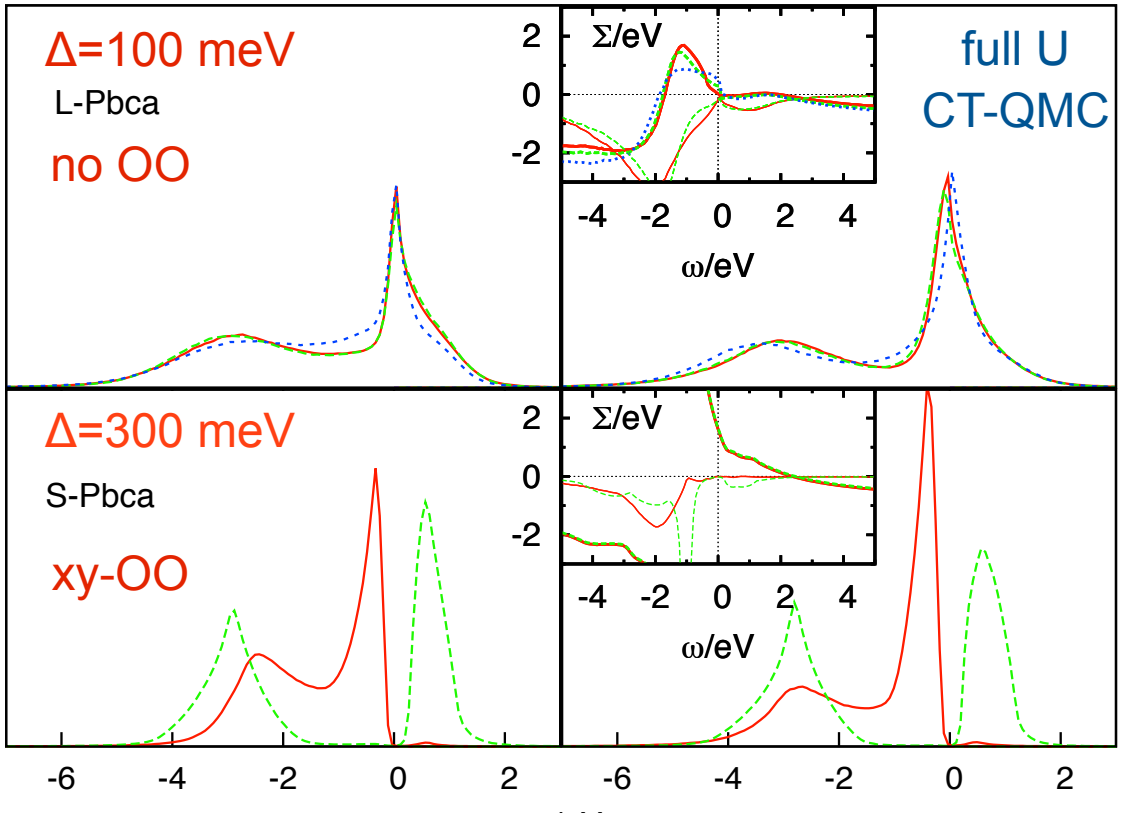
Nature of the Mott Transition in Ca_2RuO_4

E. Gorelov,[1] M. Karolak,[2] T. O. Wehling,[2] F. Lechermann,[2] A. I. Lichtenstein,[2] and E. Pavarini[1]

layered ruthenates

t_{2g}^4





do we need a large crystal-field?

el-ph coupling

$$H = -g\sqrt{C}(\tau_z Q_3 + \tau_x Q_2)$$

$E_0 + \Delta/2$ ——

Δ

$E_0 - \Delta/2$ —— θ

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

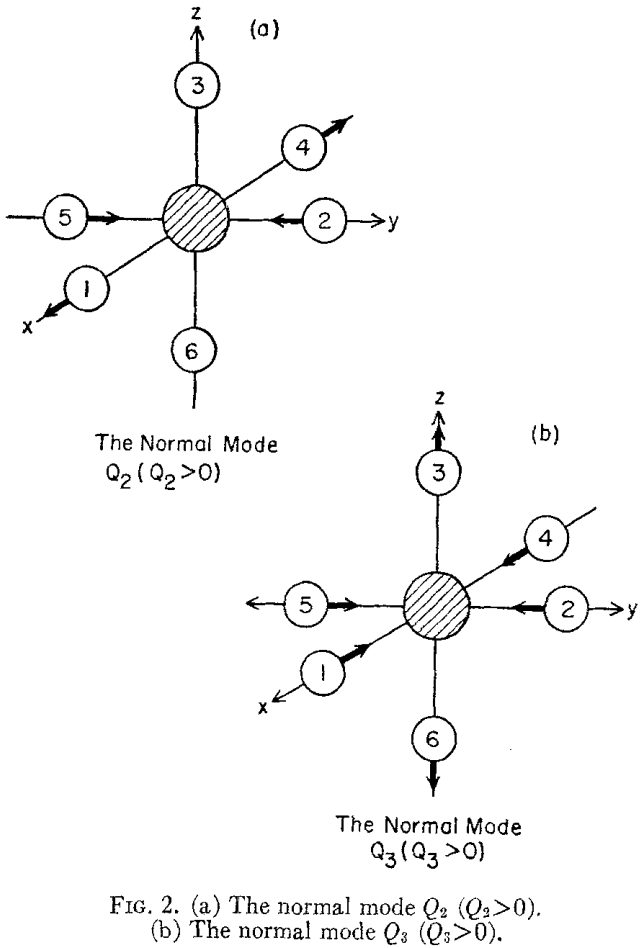


FIG. 2. (a) The normal mode Q_2 (Q_2>0). (b) The normal mode Q_3 (Q_3>0).

no, a 100 meV crystal-field could be enough

super-exchange

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.

no static JT

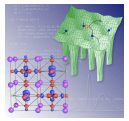
$$H = -\sum_{imjm'\sigma} t_{imjm'} c^{\dagger}_{im\sigma} c_{jm'\sigma} + U \sum_{im\sigma jm'\sigma'} n_{im\sigma} n_{jm'\sigma'}$$

perturbation t/U

↓

super-exchange Hamiltonian

$$H = -J_{SS} S_1 \cdot S_2 - J_{OO} \tau_1 \tau_2 - J_{SO}[\tau_1 S_2 + \tau_2 S_1]$$



τ : pseudospins in orbital space

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what state is occupied?

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

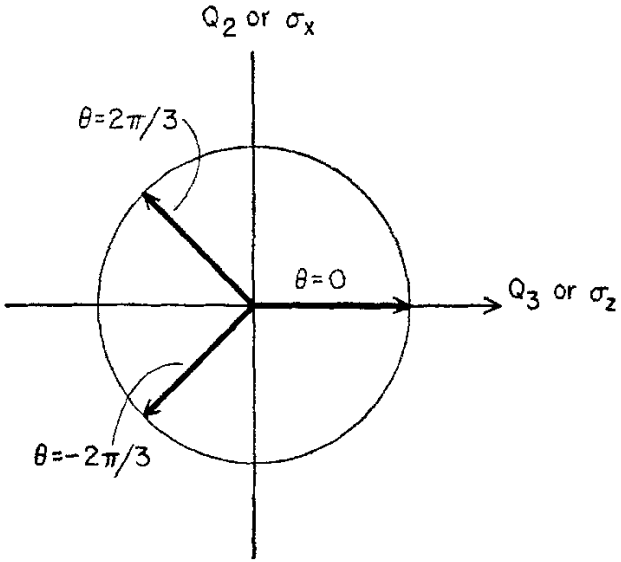

FIG. 3. The Q_3Q_2 and $\sigma_z\sigma_x$ plane.

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

From this point of view, we can understand also the properties of substances with Mn^{3+} and Cr^{2+}. The calculations of the local distortions near Jahn-Teller ions usually indicate[1] that both near Cu^{2+} and near Mn^{3+} the anion octahedron becomes elongated (one long axis and two short ones). It was found above that from the point of view of superexchange, the favored orbitals in perovskites are the singly-occupied ones of the type $d_{z^2-x^2}$ ($\theta = \pi/3$). For the Cu^{2+} ion (hole orbital), the local octahedron then becomes elongated; thus, the two factors, superexchange, and the local Jahn-Teller effect act in the same direction in this case. Indeed, in $KCuF_3$ we actually have $\theta \approx \pm 60°$ [12]. On the other hand, for ions with d^4 structure (Mn^{3+}, Cr^{2+}), an orbital $d_{z^2-x^2}$ occupied by an electron would lead to a local oblate octahedron and to a pseudotetragonal deformation of the crystal with $c/a > 1$; in actual fact, however, $c/a < 1$ in $LaMnO_3$ and $KCrF_3$. The local Jahn-Teller effect, however, calls for the electron to occupy an orbital of the type d_{ε}^2 ($\theta = \pm 120°$); the simultaneous action and compe- and superexchange, can indeed explain the intermediate type of occupied orbital with $\theta \approx 90°$.

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)

JT instability in afm LDA+U

Density-functional theory and strong interactions: Orbital ordering in Mott-Hubbard insulators

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(Received 15 May 1995)

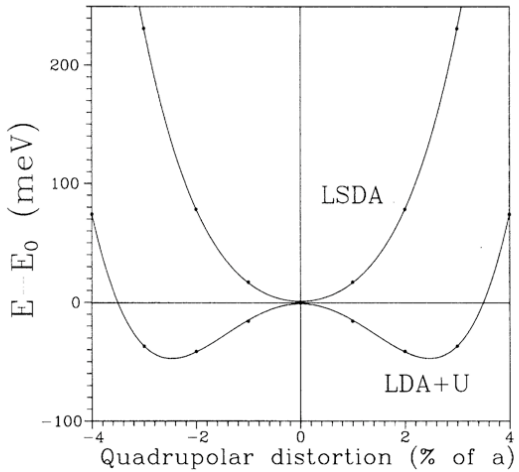
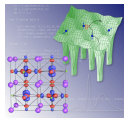


FIG. 1. The dependence of the total energy of $KCuF_3$ on the quadrupolar lattice distortion obtained in calculations with LSDA and LDA+U functionals.

The situation changes drastically if we allow for orbital polarization. Because U exceeds the bandwidth, the orbital sector is already strongly polarized (as are the spins) before the lattice is allowed to react. Overlooking some unimportant details concerning the coherence of the intermediate states, the well-known rule that electronic MFT in strong coupling maps onto the classical "spin" problem holds also in this case. In other words, we find the quadrupolar orbital-ferromagnetic spin phase to be most stable (for the same reasons as Kugel and Khomskii[6]). Obviously the cubic lattice is unstable in the presence of this orbital order parameter. In fact, despite large-scale changes in the electronic system the deformation is modest, indicating a rather weak electron-phonon coupling.

KK-like mechanism !

however this is not the energy gain due to orbital polarization!



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JT instability in pm LDA+DMFT

PRL **101,** 096405 (2008) PHYSICAL REVIEW LETTERS week ending 29 AUGUST 2008

Structural Relaxation due to Electronic Correlations in the Paramagnetic Insulator $KCuF_3$

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[3]*Institute of Metal Physics, South Kovalevskoy Street 18, 620219 Yekaterinburg GSP-170, Russia*
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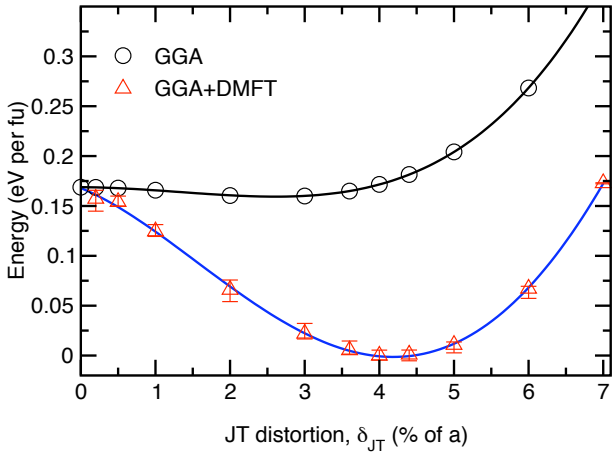
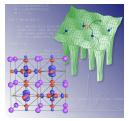
(Received 7 April 2008; published 29 August 2008)



DMFT para and LDA+U AFM give similar results

energy gain ~ 175 meV



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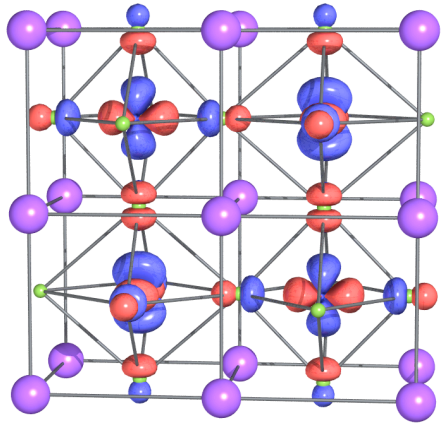
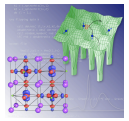
what is the mechanism?



- super-exchange
  - why T_N (40K) much smaller than T_{OO} (800-1400 K) ?
- electron-phonon coupling
  - what about LDA+U,HF,GGA+DMFT results?

Our approach: single out Kugel-Khomskii mechanism using LDA+DMFT



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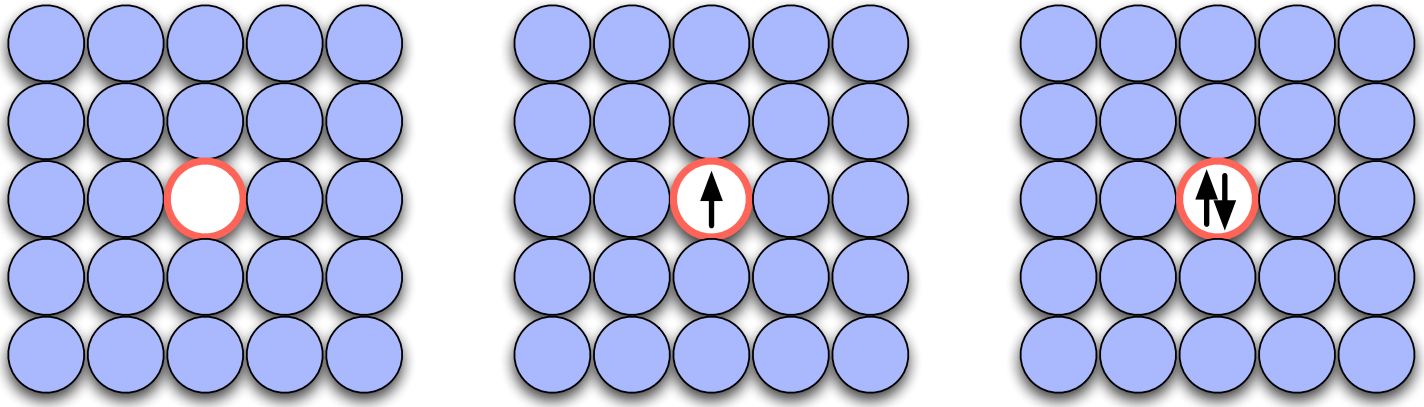
dynamical mean-field theory

Hubbard model:

$$H = -t \sum_{i \neq i'} \sum_{\sigma} c^{\dagger}_{i\sigma} c_{i\sigma} + U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

DMFT approximation: from a lattice problem to a self-consistent one-impurity model

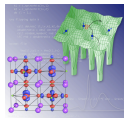


$$G_0^{-1} - G^{-1} = \Sigma(\omega)$$

dynamics captured — self-energy local

exact in infinite dimensions

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



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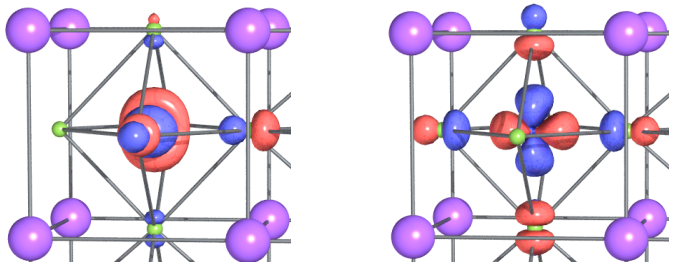
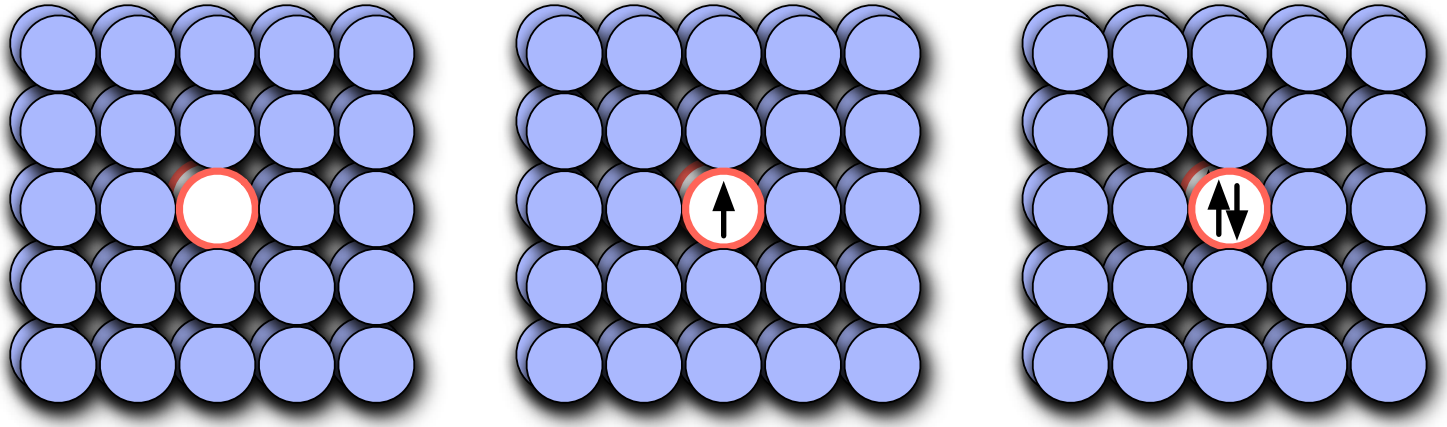
general LDA+DMFT scheme with Wannier functions

multiband Hubbard model

$$H = -\sum_{m,m',i,i',\sigma} t_{mm'}^{i,i'} c_{im\sigma}^{\dagger} c_{im'\sigma} + U \sum_{i\ m} \hat{n}_{im\uparrow}\hat{n}_{im\downarrow}$$
$$+ \frac{1}{2} \sum_{\substack{i\sigma\sigma' \\ m\neq m'}} (U - 2J - J\delta_{\sigma,\sigma'}) \hat{n}_{im\sigma}\hat{n}_{im'\sigma'}$$
$$- J \sum_{i\ m\neq m'} \left[c_{im\uparrow}^{\dagger} c_{im\downarrow}^{\dagger} c_{im'\uparrow} c_{im'\downarrow} + c_{im\uparrow}^{\dagger} c_{im\downarrow} c_{im'\downarrow}^{\dagger} c_{im'\uparrow} \right] - \hat{H}_{\mathrm{DC}}^{e_g}$$



Wannier functions



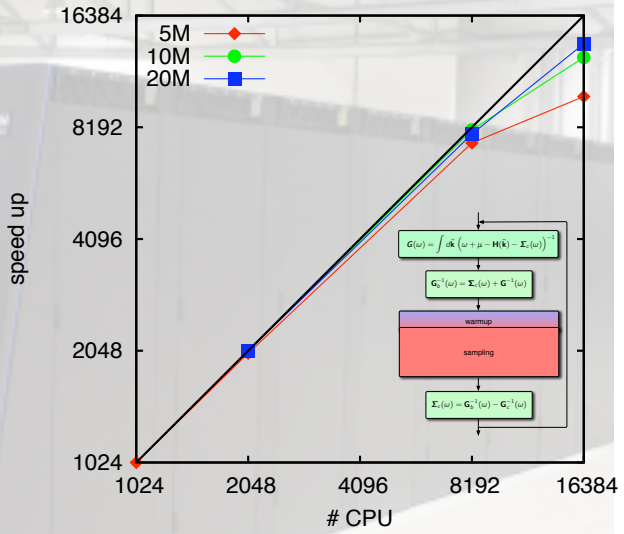
$$G_{m,m'} = \sum_{\mathbf{k},\mathbf{n}} \left[\frac{1}{i\omega_n + \mu - \mathrm{H}_{0\mathbf{k}} - \Sigma(i\omega_n)} \right]_{m,m'}$$

E. Pavarini, S. Biermann, A. Poteryaev, A.I. Lichtenstein, A.Georges, O.K. Andersen, PRL 2004

LDA+DMFT with Wannier functions

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

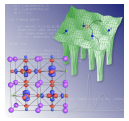
$$
\begin{array}{c|cccc}
\Sigma^l & |\alpha\sigma\rangle_1 & |\beta\sigma\rangle_1 & |\alpha\sigma\rangle_2 & |\beta\sigma\rangle_2 \\
\hline
|\alpha\sigma\rangle_1 & \Sigma^1_{\alpha\alpha} & \Sigma^1_{\alpha\beta} & 0 & 0 \\
|\beta\sigma\rangle_1 & \Sigma^1_{\beta\alpha} & \Sigma^1_{\beta\beta} & 0 & 0 \\
|\alpha\sigma\rangle_2 & 0 & 0 & \Sigma^1_{\alpha\alpha} & -\Sigma^1_{\alpha\beta} \\
|\beta\sigma\rangle_2 & 0 & 0 & -\Sigma^1_{\beta\alpha} & \Sigma^1_{\beta\beta}
\end{array}
$$



DMFT and cDMFT

QMC quantum impurity solvers: HF and CT-HYB

Wannier functions: NMTOs, MLWF or PWF from LAPW (Wien2k)



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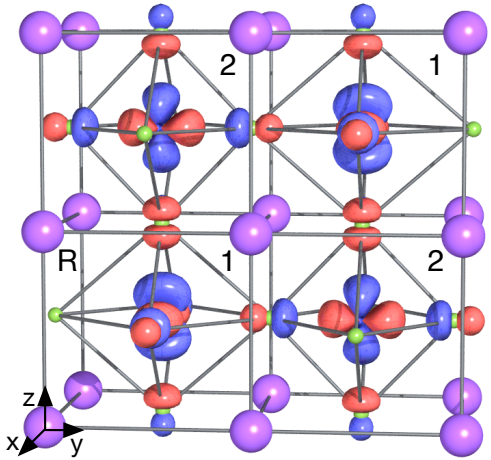
$KCuF_3$



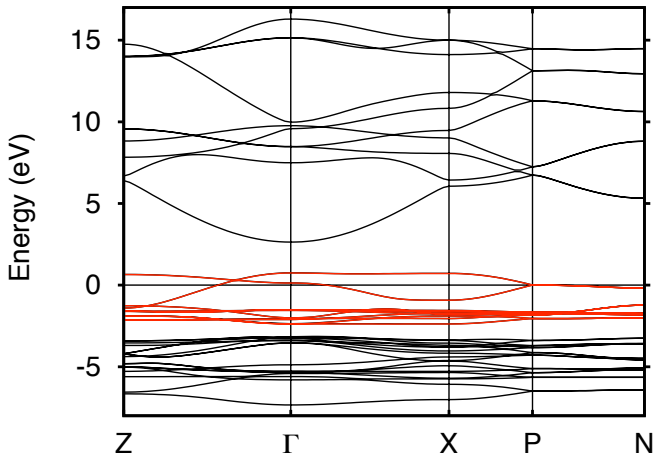
K

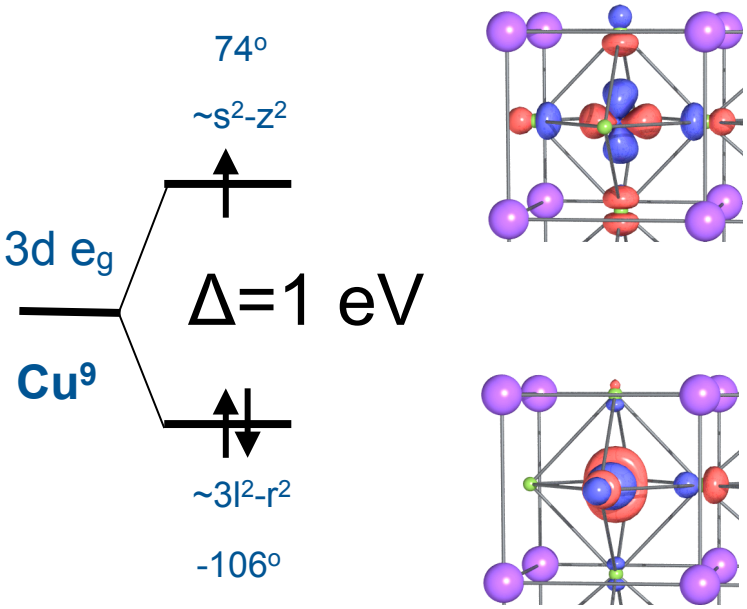
F

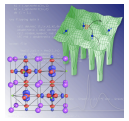
$K^+ Cu^{2+} F^-$

$K\ 4s^0\ Cu\ 3d^9\ F\ 2p^6$

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







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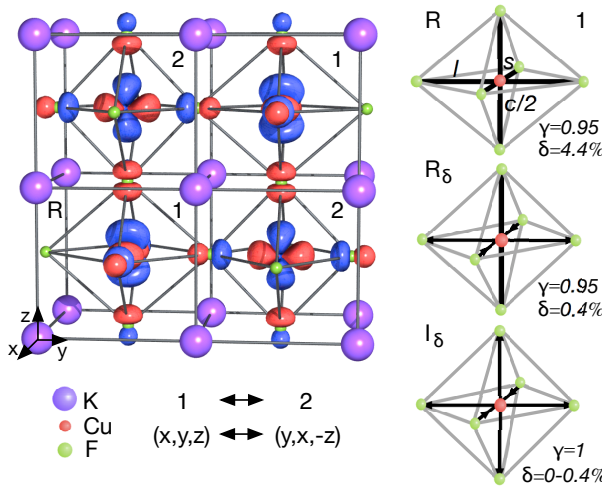
single out super-exchange

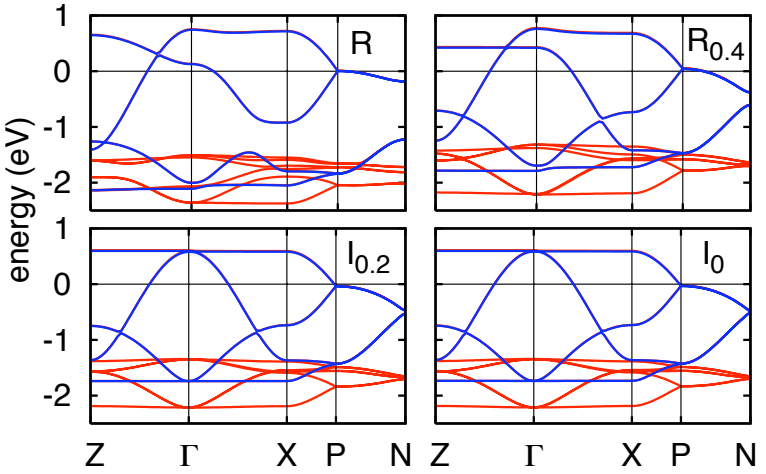


ideal structures

δ=(l-s)/(l+s)/2 Jahn-Teller distortion

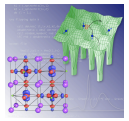
λ=c/a/2 tetragonal distortion

LDA bands



corresponding two-band Hubbard model

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



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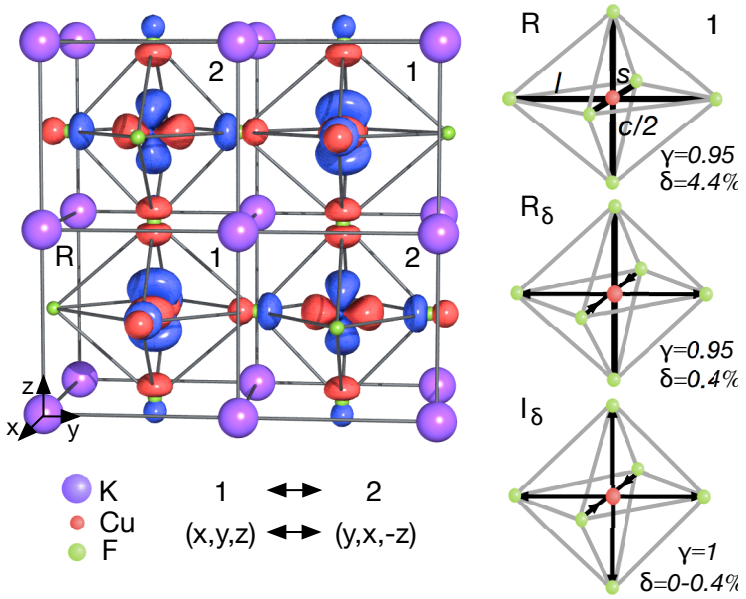
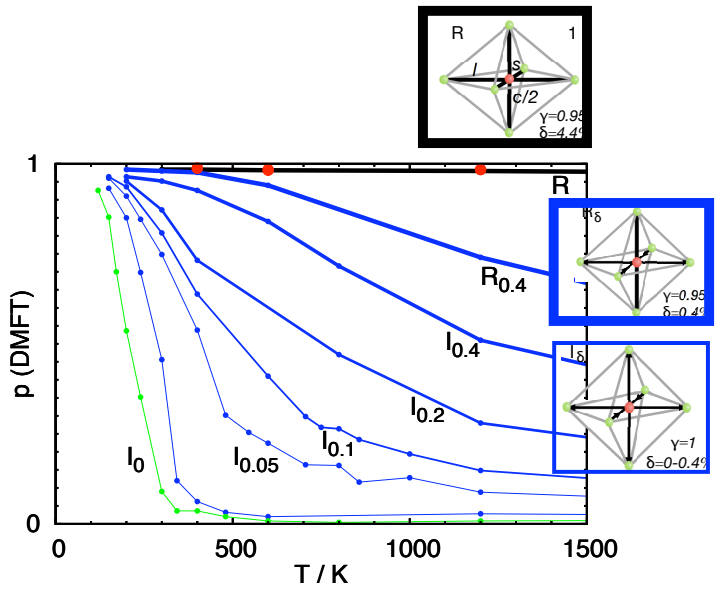
KK mechanism in $KCuF_3$

orbital polarization $p=n_1-n_2$



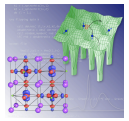


experiments: T_{JT}~800-1400 K

theory: T_{KK}~350 K

T_N vs T_{KK} ratio ok!

reminder: mean-field theory overestimates T_{KK}



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

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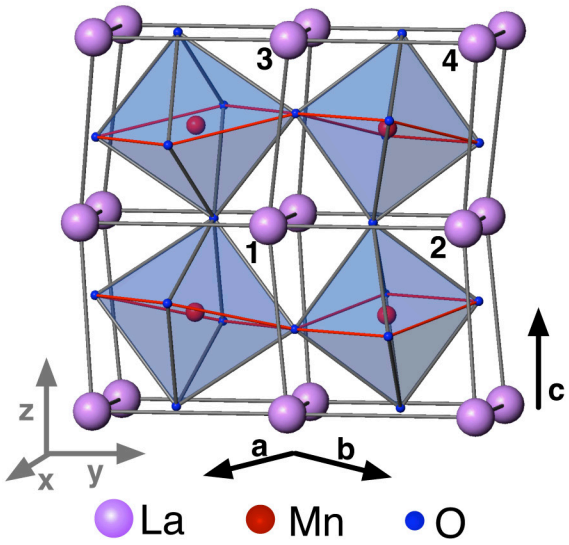
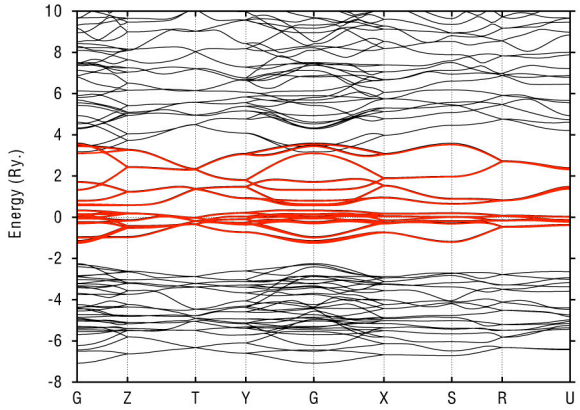
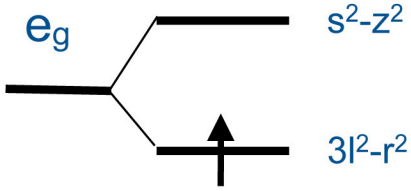
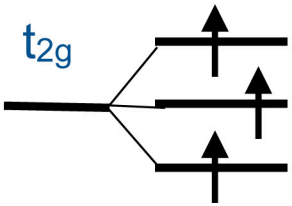
cooperative JT distortion in $LaMnO_3$

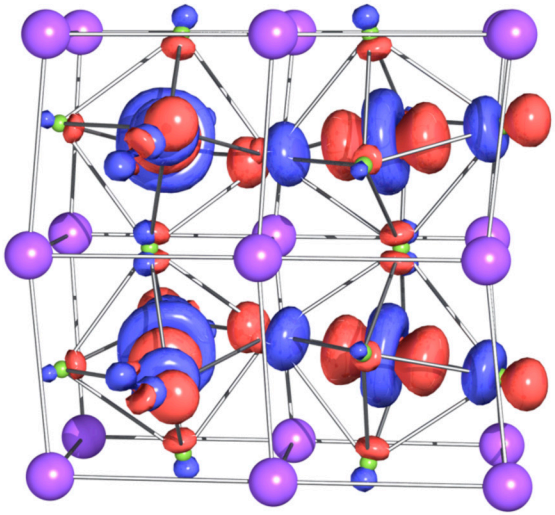




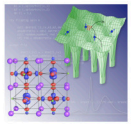




Mn^{3+} $3d^4$



LDA+DMFT



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PHYSICAL REVIEW B VOLUME 61, NUMBER 20 15 MAY 2000-II

Effects of magnetic ordering on the anisotropy and temperature dependence of the optical conductivity in $LaMnO_3$: A tight-binding approach

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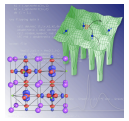
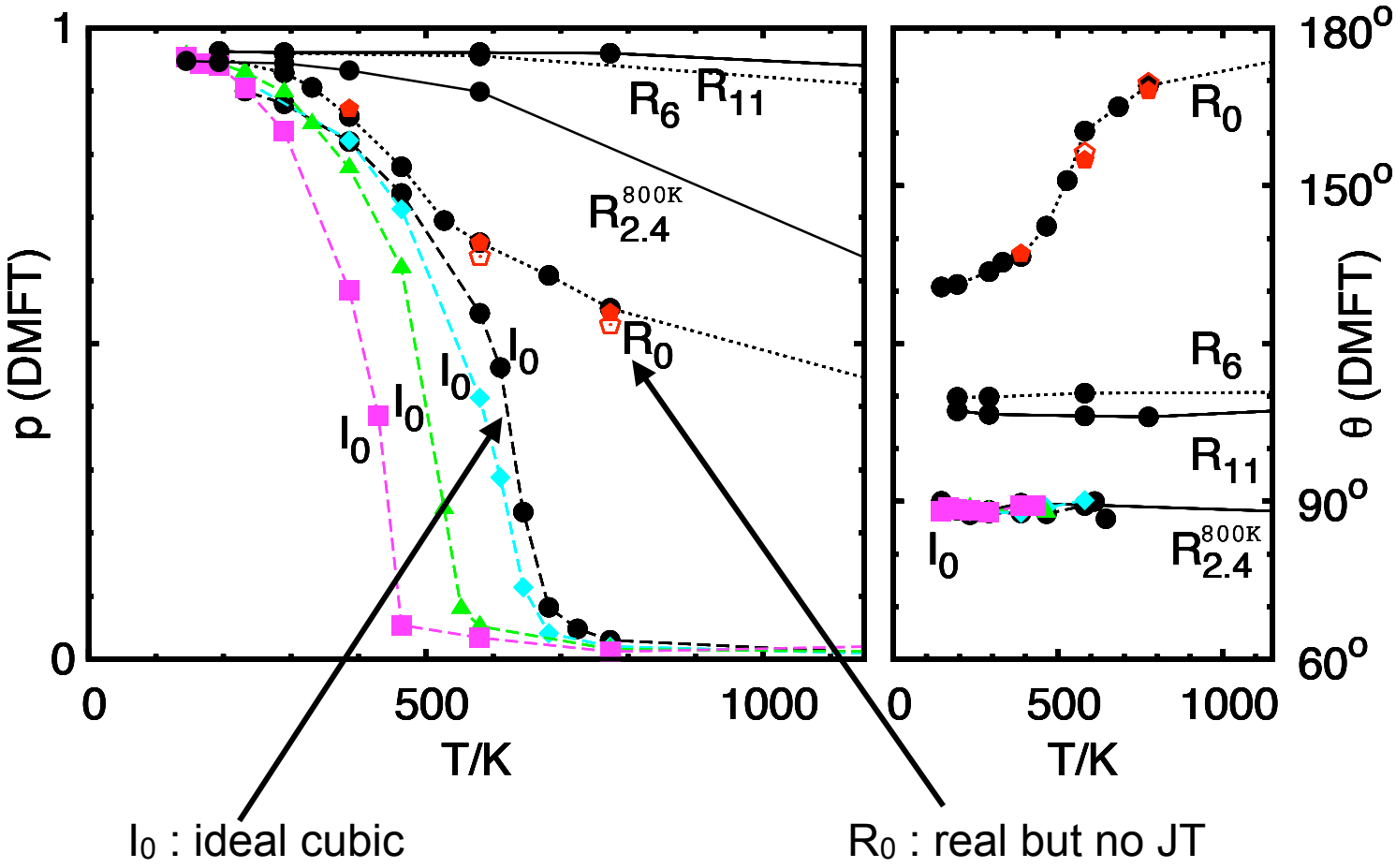
Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey 08854
(Received 14 January 1999; revised manuscript received 29 November 1999)

$$
\begin{aligned}
H = & - \sum_{ii'} \sum_{mm'} \sum_{\sigma\sigma'} t^{ii'}_{mm'} u^{ii'}_{\sigma\sigma'} c^{\dagger}_{im\sigma} c_{i'm'\sigma} \\
& - h \sum_{im} (n_{im\Uparrow} - n_{im\Downarrow}) + U \sum_{im} n_{im\Uparrow} n_{im\Downarrow} \\
& + \frac{1}{2} \sum_{im \neq m'\sigma\sigma'} (U - 2J - J\delta_{\sigma\sigma'}) n_{im\sigma} n_{im'\sigma'} \\
& - J \sum_{m \neq m'} (c^{\dagger}_{m\Uparrow} c^{\dagger}_{m'\Downarrow} c_{m'\Uparrow} c_{m\Downarrow} + c^{\dagger}_{m\Uparrow} c^{\dagger}_{m\Downarrow} c_{m'\Uparrow} c_{m'\Downarrow})
\end{aligned}
$$



T_{KK}~ 600 K !!



non-JT crystal-field

experiments

super-exchange

red: cDMFT 4 sites
black: DMFT

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

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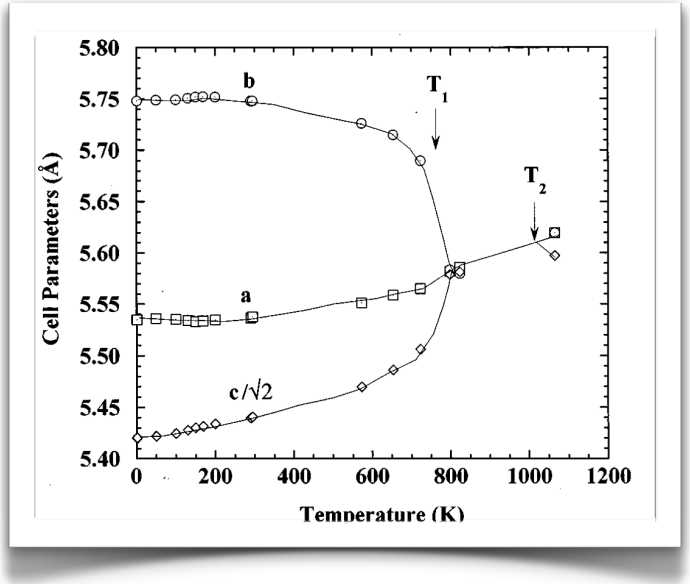
$T_{KK} \sim T_{JT}$



PHYSICAL REVIEW B VOLUME 57, NUMBER 6 1 FEBRUARY 1998-II

Neutron-diffraction study of the Jahn-Teller transition in stoichiometric $LaMnO_3$

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L. Pinsard and A. Revcolevschi
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(Received 2 September 1997)

The parent compound of the giant magnetoresistance Mn-perovskite, $LaMnO_3$, has been studied by thermal analysis and high-resolution neutron-powder diffraction. The orthorhombic *Pbnm* structure at room temperature is characterized by an antiferrodistorsive orbital ordering due to the Jahn-Teller effect. This ordering is evidenced by the spatial distribution of the observed Mn-O bond lengths. $LaMnO_3$ undergoes a structural phase transition at $T_{JT} \approx 750$ K, above which the orbital ordering disappears. There is no change in symmetry although the lattice becomes metrically cubic on the high-temperature side. The MnO_6 octahedra become nearly regular above T_{JT} and the thermal parameter of oxygen atoms increases significantly. The observed average cubic lattice is probably the result of dynamic spatial fluctuations of the underlying orthorhombic distortion. [S0163-1829(98)51706-7]

VOLUME 81, NUMBER 3 PHYSICAL REVIEW LETTERS 20 JULY 1998

Resonant X-Ray Scattering from Orbital Ordering in $LaMnO_3$

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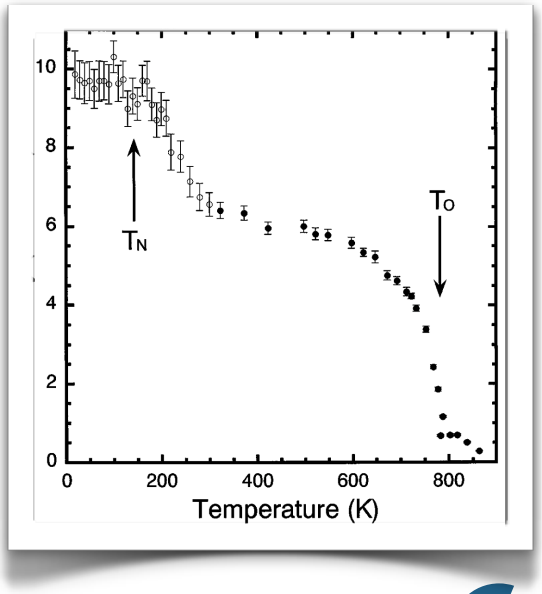
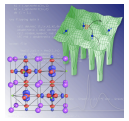
[2]*Core Research for Evolutional Science and Technology (CREST), Tsukuba 305-0047 Japan*
[3]*Department of Physics, Brookhaven National Laboratory, Upton, New York 11973*
[4]*Institute of Materials Science, University of Tsukuba, Tsukuba 305-8573, Japan*
[5]*Department of Applied Physics, University of Tokyo, Tokyo 113-0033, Japan and Joint Research Center for Atom Technology (JRCAT), Tsukuba 305-0046, Japan*
[6]*Department of Physics, Tohoku University, Sendai 980-8578, Japan*
(Received 22 April 1998)





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however...

PRL **94,** 177203 (2005) PHYSICAL REVIEW LETTERS week ending 6 MAY 2005

Orbital Correlations in the Pseudocubic *O* and Rhombohedral *R* Phases of $LaMnO_3$

Xiangyun Qiu,[1] Th. Proffen,[2] J. F. Mitchell,[3] and S. J. L. Billinge[1]

[1]*Department of Physics and Astronomy, Michigan State University, E. Lansing, Michigan 48824, USA*
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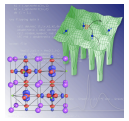
(Received 12 August 2004; revised manuscript received 24 February 2005; published 5 May 2005)

The local and intermediate structure of stoichiometric $LaMnO_3$ has been studied in the pseudocubic and rhombohedral phases at high temperatures (300–1150 K). Neutron powder diffraction data were collected and a combined Rietveld and high real space resolution atomic pair distribution function analysis was carried out. The nature of the Jahn-Teller (JT) transition around 750 K is confirmed to be orbital order to disorder. In the high-temperature orthorhombic (*O*) and rhombohedral (*R*) phases, the MnO_6 octahedra are still fully distorted locally. More importantly, the intermediate structure suggests the presence of local ordered clusters of diameter ~16 Å (~4 MnO_6 octahedra) implying strong nearest-neighbor JT antiferrodistortive coupling. These clusters persist well above the JT transition temperature even into the high-temperature *R* phase.

DOI: 10.1103/PhysRevLett.94.177203 PACS numbers: 75.47.Lx, 61.12.−q, 75.47.Gk

still $T_{KK} \sim T_{JT}$...



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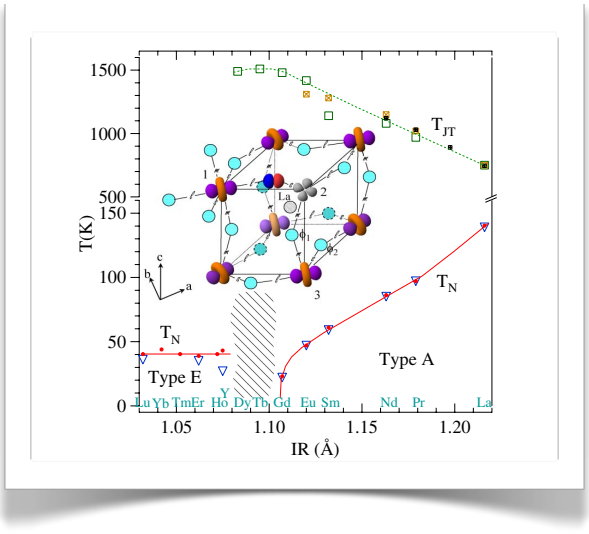
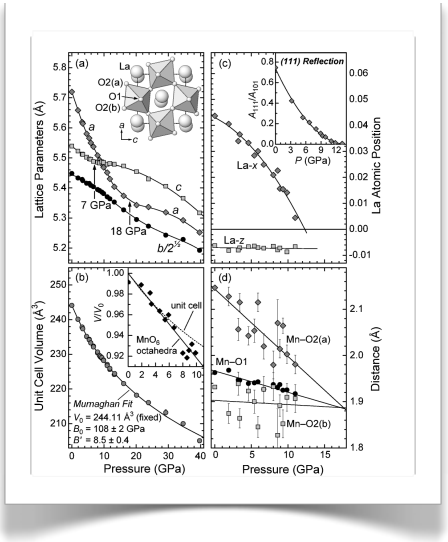
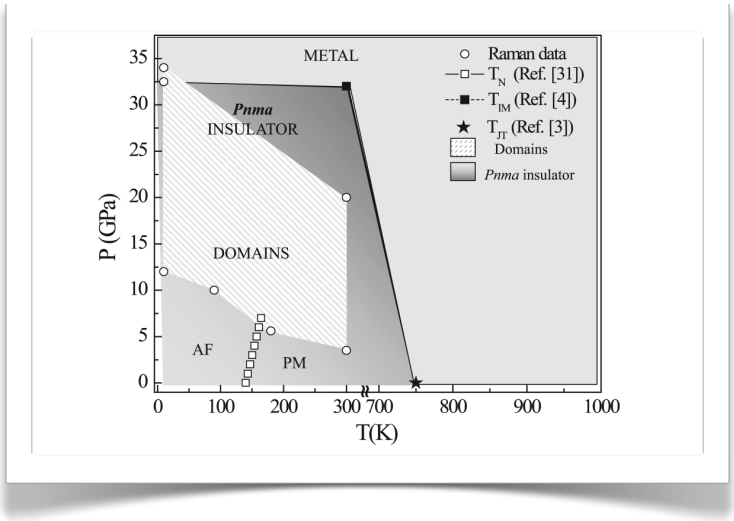
is super-exchange driving oo melting?

approach: RMnO$_3$ series + high pressure data

T_{JT} .....

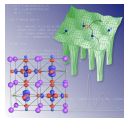






.. increases with decreasing IR

decreases with increasing P



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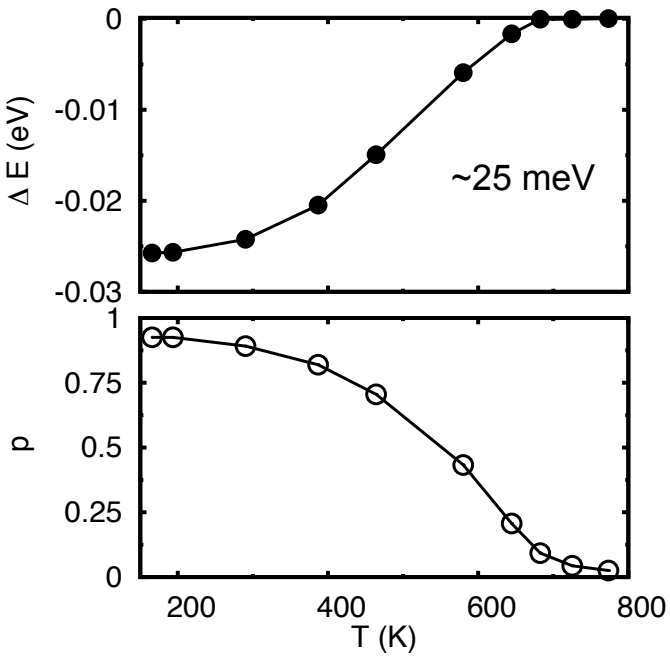
two methods



we need small error bars!

orbital polarization

$$p = n_1 - n_2$$

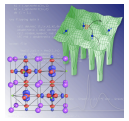
total energy

$$E = E_{\mathrm{LDA}} + \langle \hat{H}^{\mathrm{LDA}} \rangle - E^{e_g}_{\mathrm{LDA}} + \langle \hat{H}_U \rangle - E_{\mathrm{DC}}.$$

$$H = -J_{SS} S_1 \cdot S_2 - J_{OO} \tau_1 \tau_2 - J_{SO}[\tau_1 S_2 + \tau_2 S_1]$$

mean-field for pseudo-spin operators

$T_{KK} \sim 2\,\Delta E$



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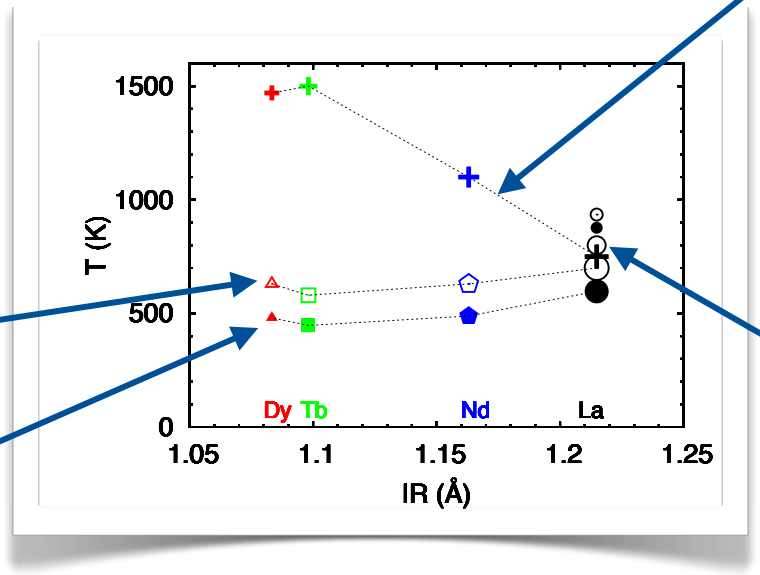
without distortions

T_{KK} decreases with IR

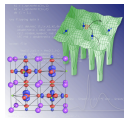
T_{JT} in experiments



orbital polarization

total energy

$LaMnO_3$ under pressure



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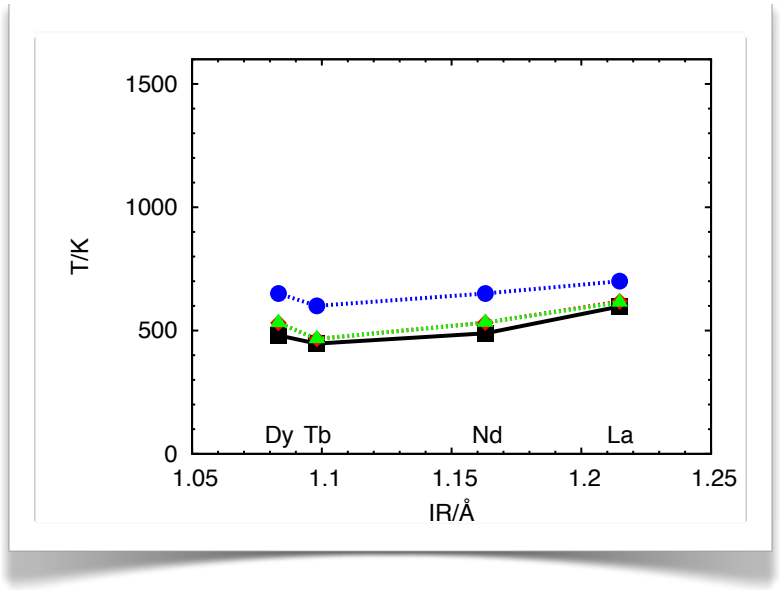
without distortions

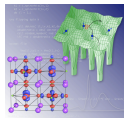
T_{KK} decreases with IR



CT-HYB segment (green)

CT-HYB Krylov (red)

spin flip and pair hopping do not change trends



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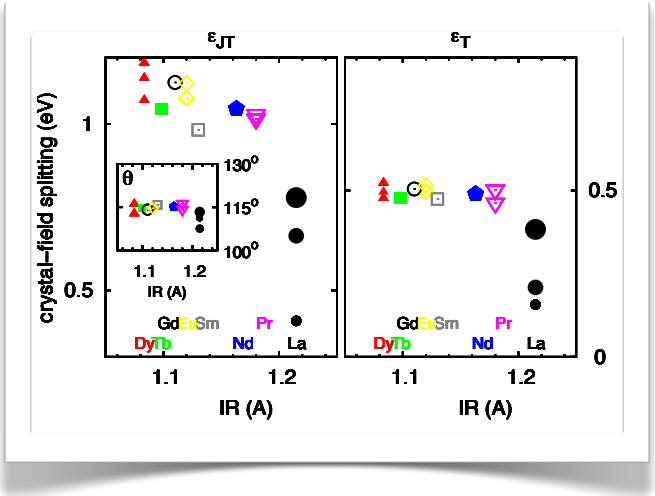
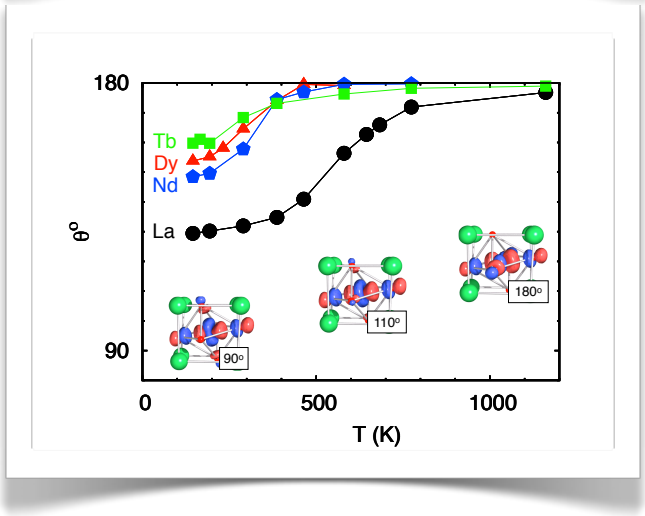
tetragonal splitting



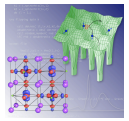


transition for all systems but $LaMnO_3$. These results can be understood qualitatively in static mean-field theory. In the simplest case, super-exchange yields just an effective Jahn-Teller splitting $\varepsilon_{KK} = \langle \tau_x \rangle \lambda_{KK}$, where λ_{KK} is the molecular field parameter; the self-consistency condition for orbital order is

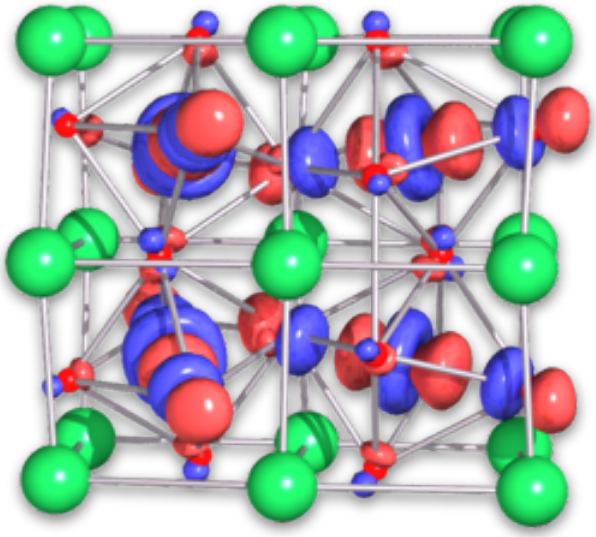
$$\langle \tau_x \rangle = \frac{1}{2} \sin\theta \tanh\left(\beta \sqrt{\varepsilon_T^2 + \varepsilon_{KK}^2}/2\right),$$

with $\sin\theta = \varepsilon_{KK}/\sqrt{\varepsilon_T^2 + \varepsilon_{KK}^2}$. This equation has a non-trivial solution ($\theta \neq 180^o$) only if $\lambda_{KK}/2 > \varepsilon_T$. The critical temperature is

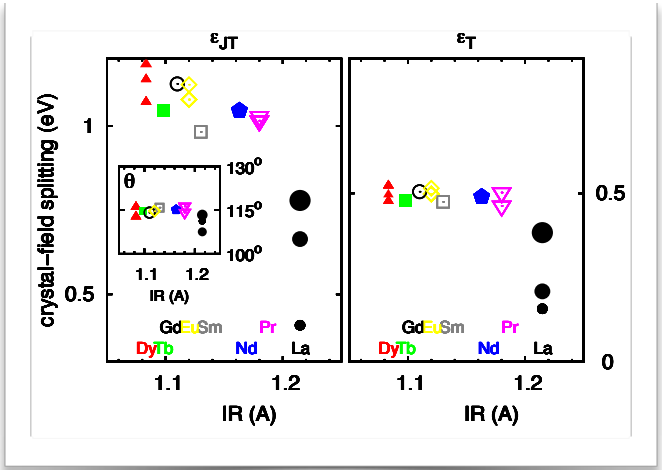
$$T_{KK}^{\varepsilon_T}/T_{KK}^0 = (\varepsilon_T/2k_BT_{KK}^0)/\tanh^{-1}(\varepsilon_T/2k_BT_{KK}^0),$$

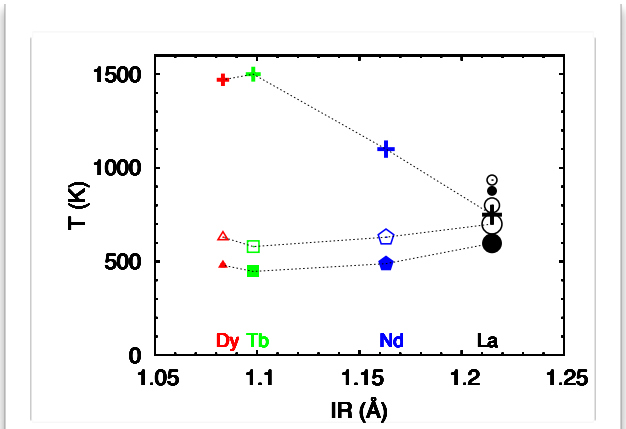


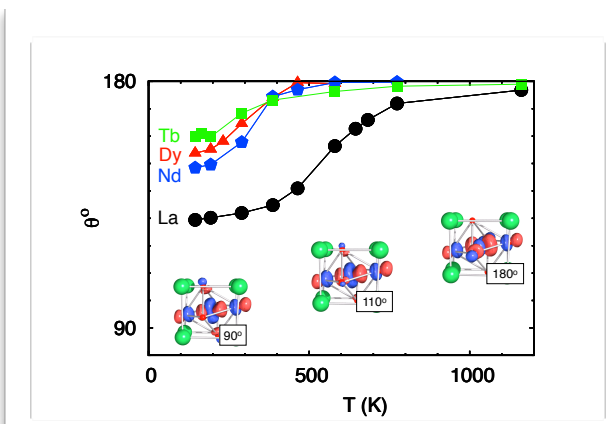
oo and oo melting

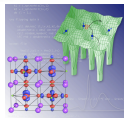


given present experimental data, super-exchange not crucial









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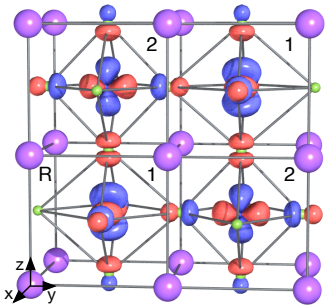
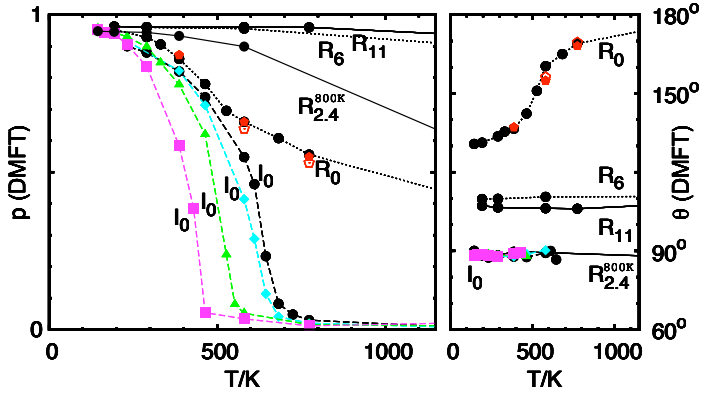
conclusions

$KCuF_3$ and $LaMnO_3$

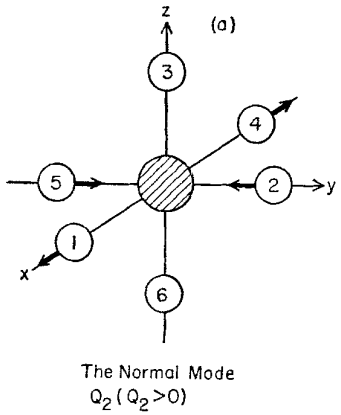
what is the mechanism of orbital-order?

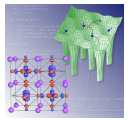


T_{KK} remarkably large



but el-ph coupling essential





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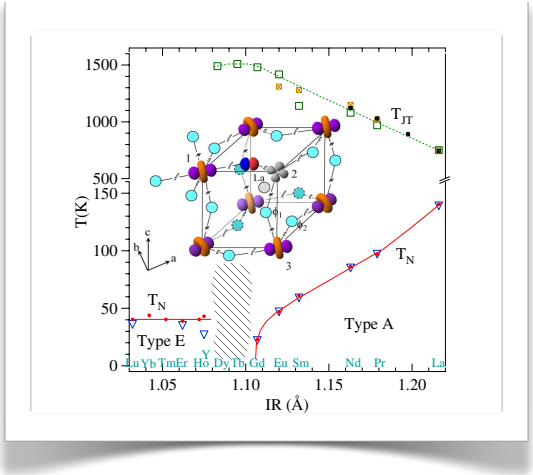
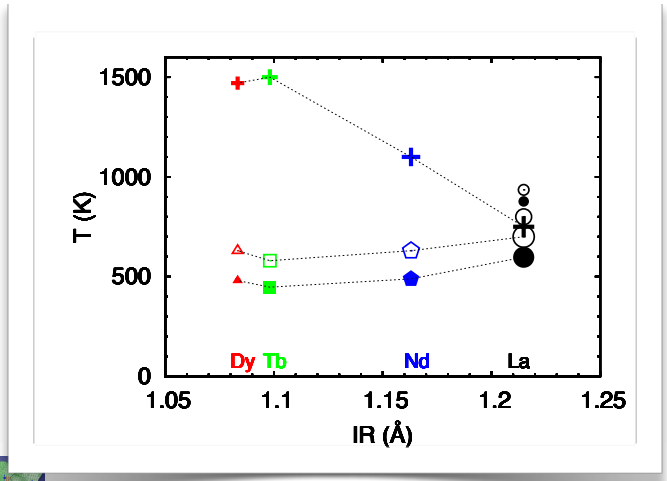
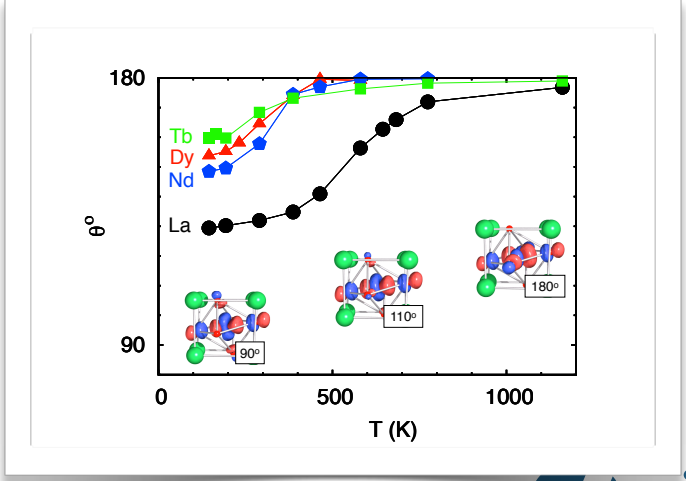
conclusions

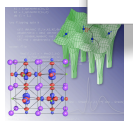
what about order to disorder transitions?



based on present experimental data
super-exchange cannot explain trends







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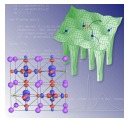
+ A.I. Lichtenstein, Uni Hamburg



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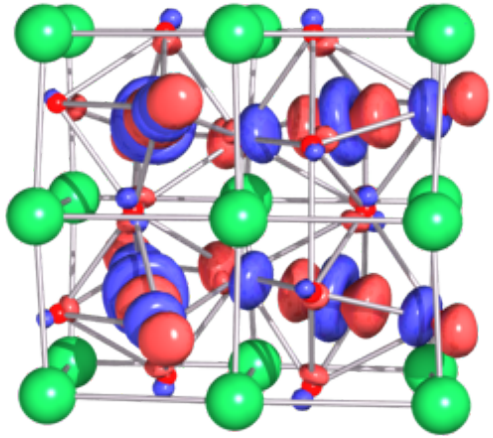
thank you!



Phys. Rev. B **85**, 0354 (2012)

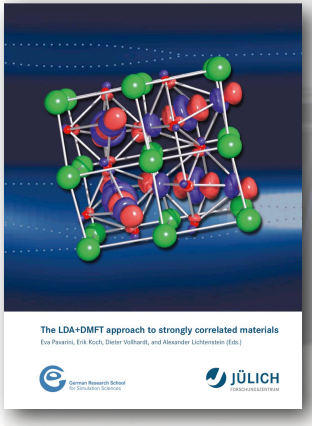
Phys. Rev. Lett. **104**, 086402 (2010)

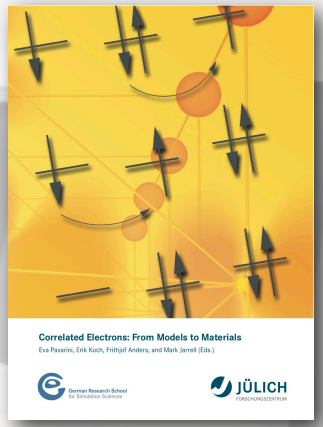
Phys. Rev. Lett. **101**, 266405 (2008)

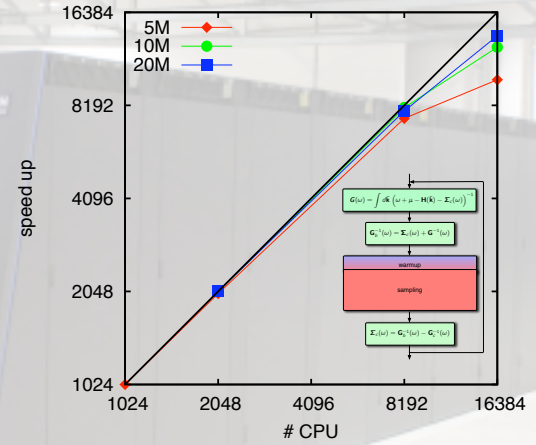
and to be published

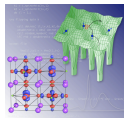
details on the approach

Autumn Schools on correlated electrons









http://www.cond-mat.de/events/correl11/

http://www.cond-mat.de/events/correl12/

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