Excitonic Condensation of Strongly Correlated Electrons

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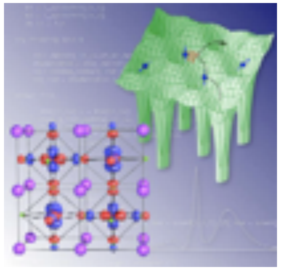
DFG FOR1346



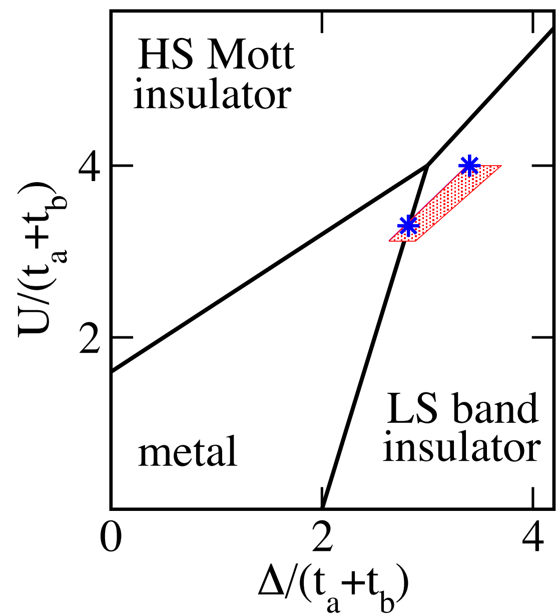
Minimal lattice model with spin-state transition

Two-band Hubbard model at n=2 (half filling)

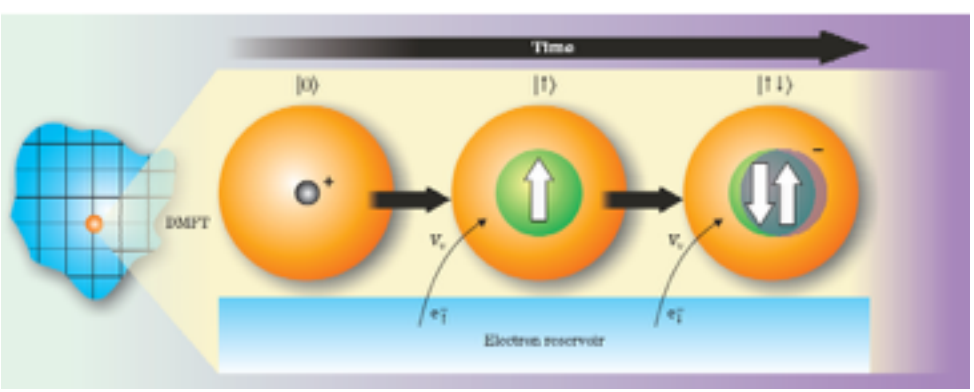
$$H_{\rm t} = \frac{\Delta}{2}\sum_{i,\sigma}(n^a_{i\sigma} - n^b_{i\sigma}) + \sum_{i,j,\sigma}(t_a a^\dagger_{i\sigma}a_{j\sigma} + t_b b^\dagger_{i\sigma}b_{j\sigma})$$
$$+ \sum_{\langle ij\rangle,\sigma}(V_1 a^\dagger_{i\sigma}b_{j\sigma} + V_2 b^\dagger_{i\sigma}a_{j\sigma} + c.c.)$$

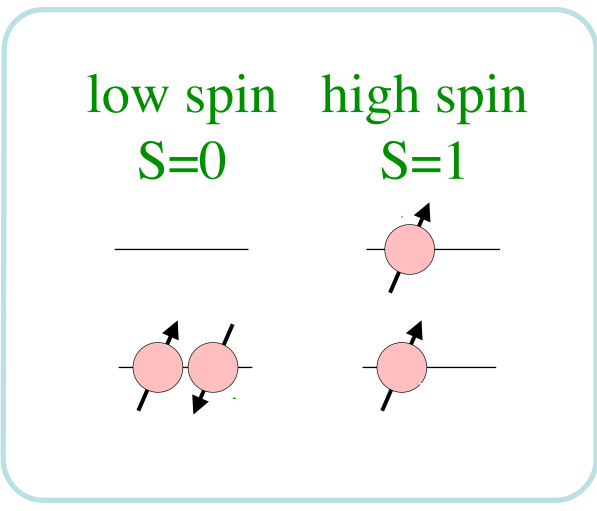
$$H^{\rm dd}_{\rm int} = U\sum_i(n^a_{i\uparrow}n^a_{i\downarrow} + n^b_{i\uparrow}n^b_{i\downarrow}) + (U-2J)\sum_{i,\sigma}n^a_{i\sigma}n^b_{i-\sigma}$$
$$+ (U-3J)\sum_{i\sigma}n^a_{i\sigma}n^b_{i\sigma}$$

$$H'_{\rm int} = J\sum_{i\sigma}a^\dagger_{i\sigma}b^\dagger_{i-\sigma}a_{i-\sigma}b_{i\sigma} + J'\sum_i(a^\dagger_{i\uparrow}a^\dagger_{i\downarrow}b_{i\downarrow}b_{i\uparrow} + c.c.).$$



Dynamical Mean-Field Theory

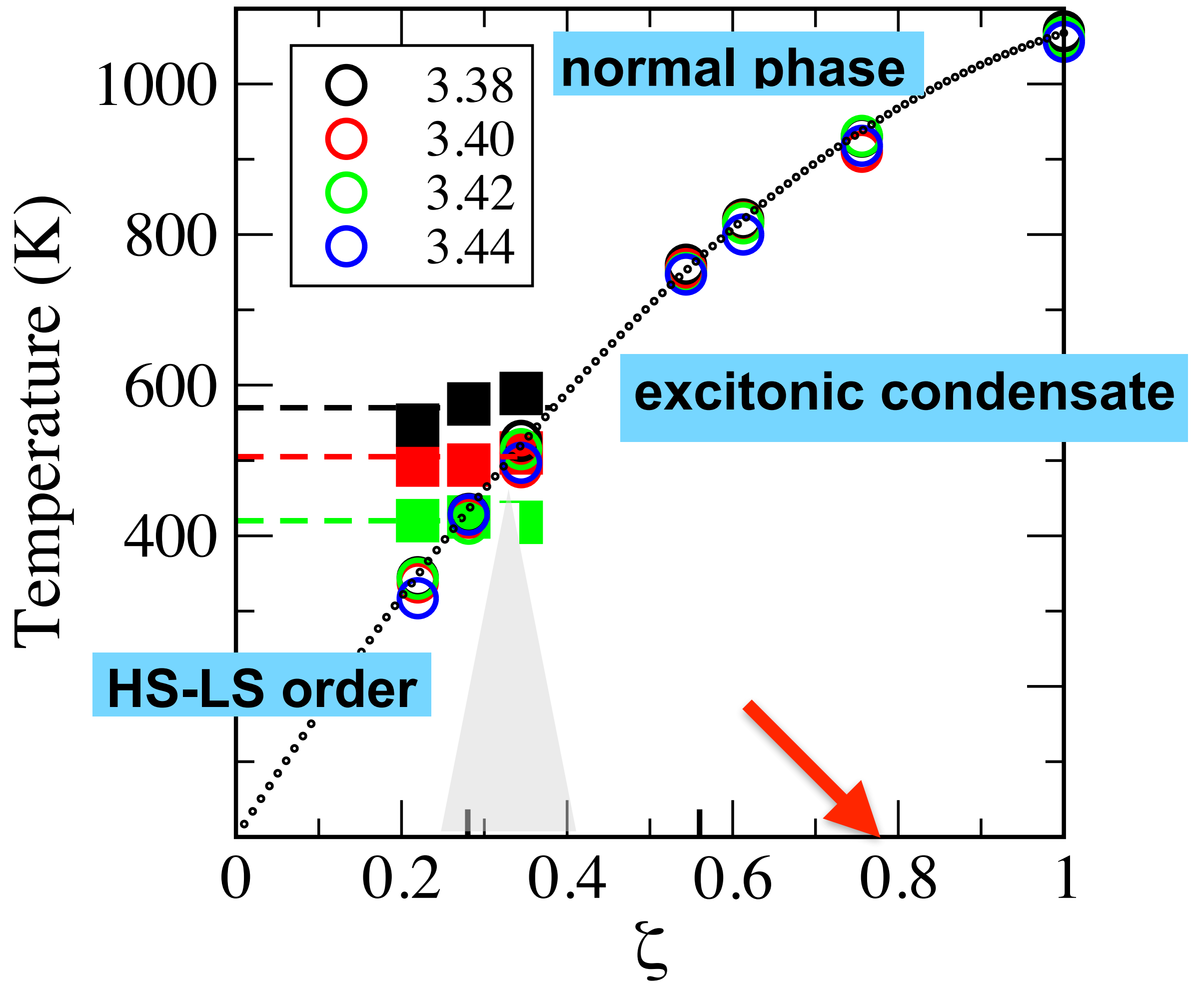




Excitonic instability - DMFT linear response

U=4, J=1
$t_a^2+t_b^2=const$
$\Delta=3.40+\delta$
V=0

$$\zeta = \frac{2t_a t_b}{t_a^2+t_b^2}$$



JK and Augustinsky, 2014

Excitonic condensate - oder parameter

Order parameter is a complex vector: $\phi^\gamma = \sum_{\alpha\beta} \sigma^\gamma_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$

z-axis parallel to: $i(\bar{\phi} \wedge \phi)$

No cross hopping (V=0) -> overall phase of ϕ does not matter
-> phases are distinguished by $|\phi^+|$ and $|\phi^-|$

3 possible phases:

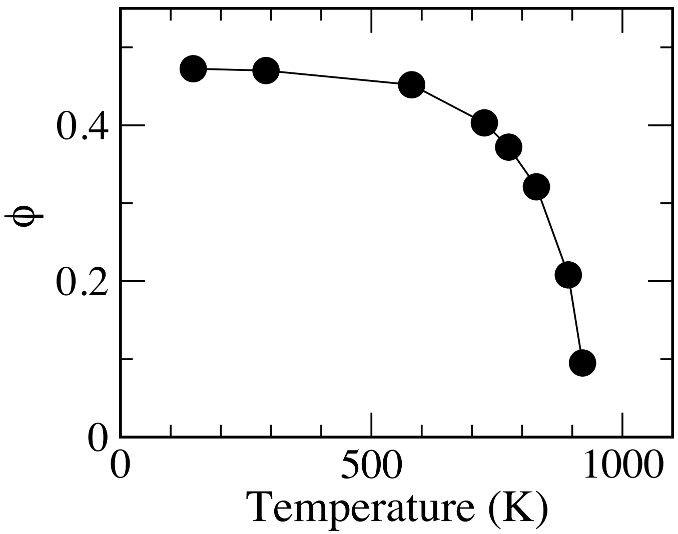
linear (L) $|\phi^+| = |\phi^-| \neq 0$

circular (C) $|\phi^+| = 0, |\phi^-| \neq 0$

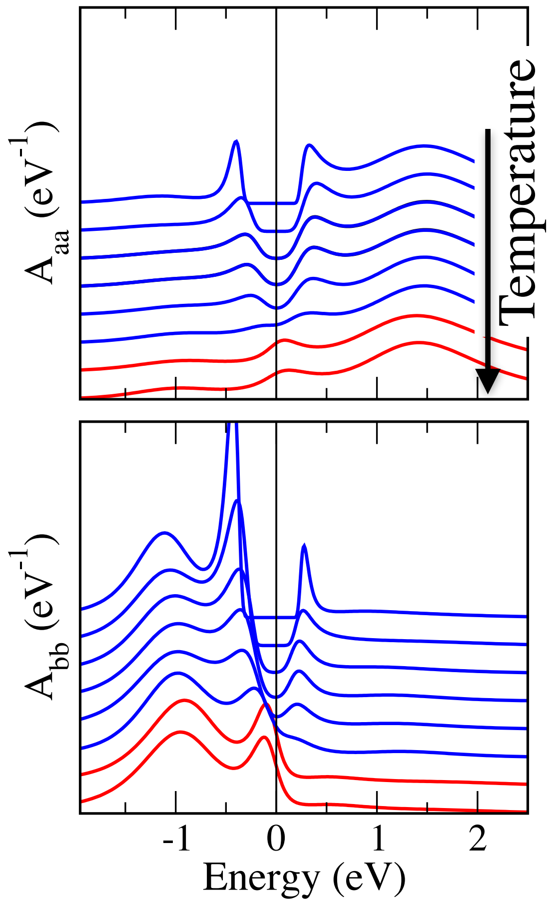
elliptic (E) $0 \neq |\phi^+| \neq |\phi^-| \neq 0$

Excitonic condensation (undoped) - L phase

order parameter $|\phi^+| = |\phi^-|$



Spectral density (diagonal elements)



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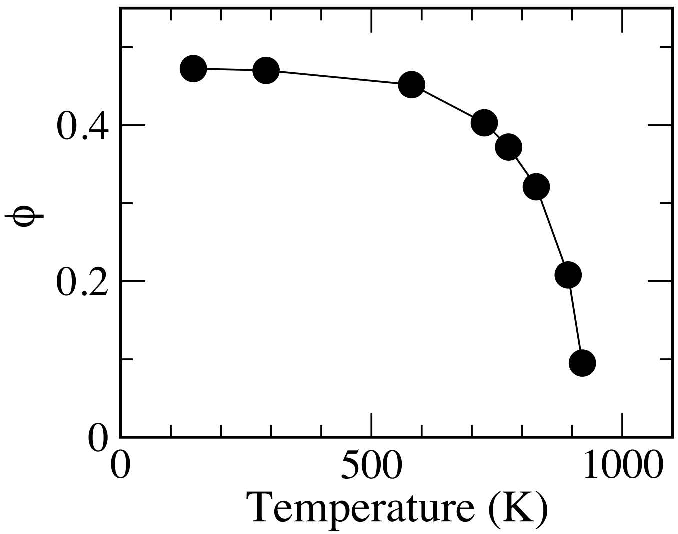
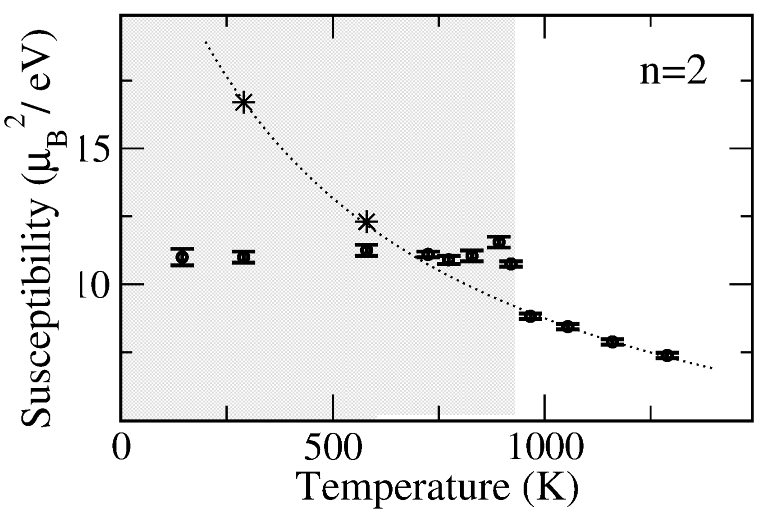
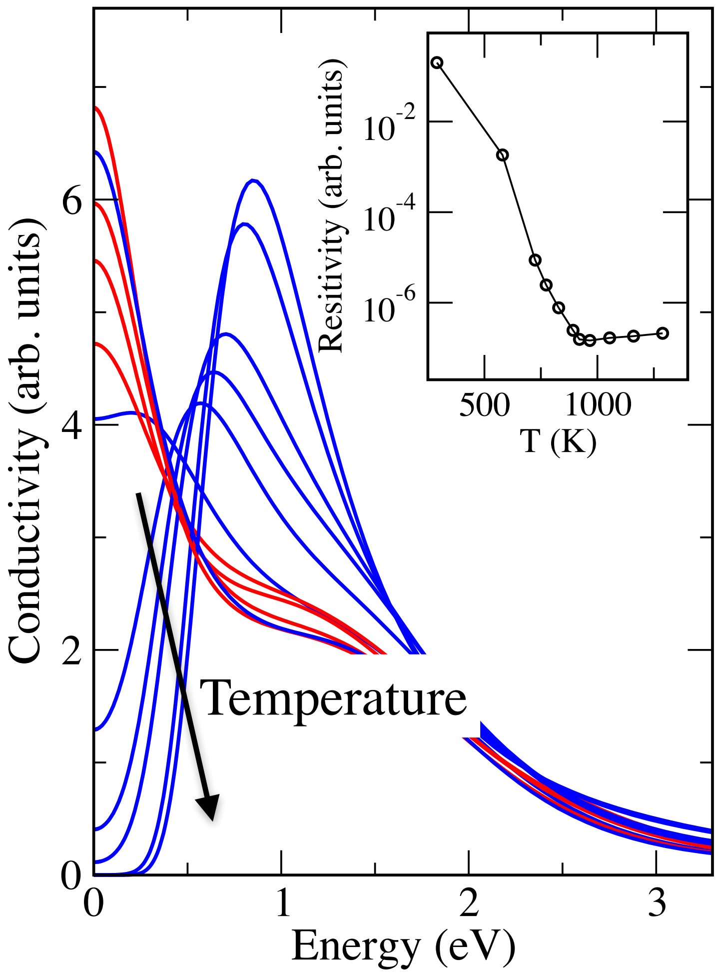


Uniform spin susceptibility

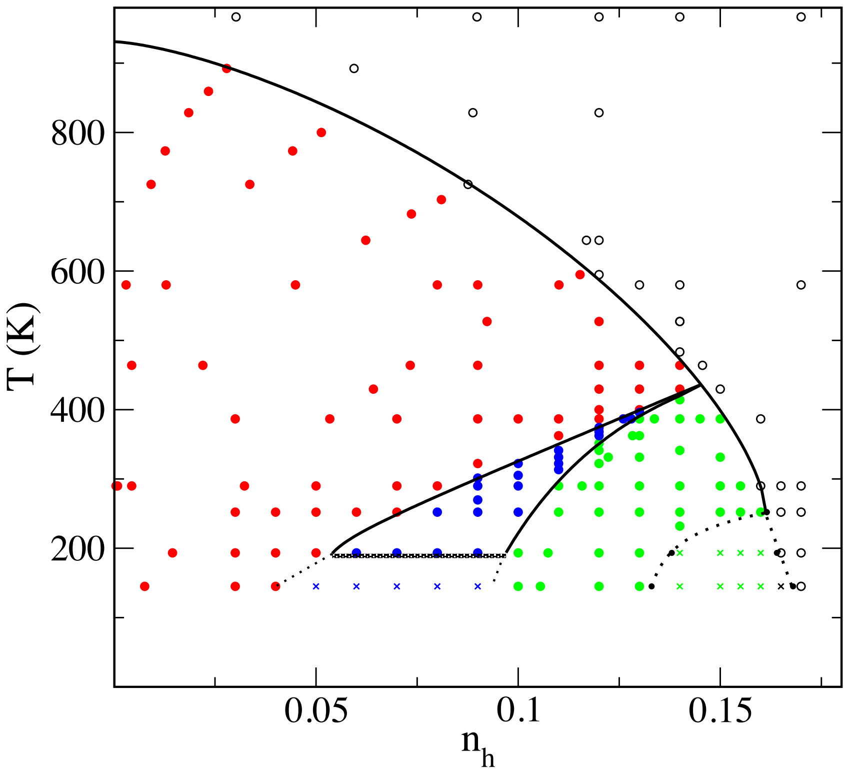


Optical conductivity (dc resistivity)

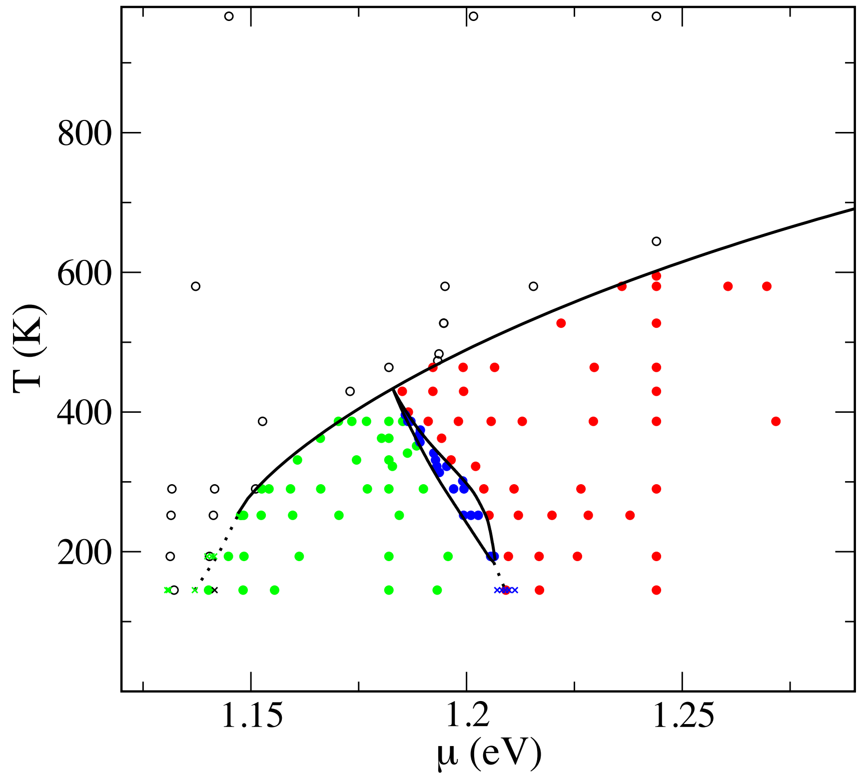


Excitonic condensation (doping) - all phases

n-T phase diagram



μ-T phase diagram

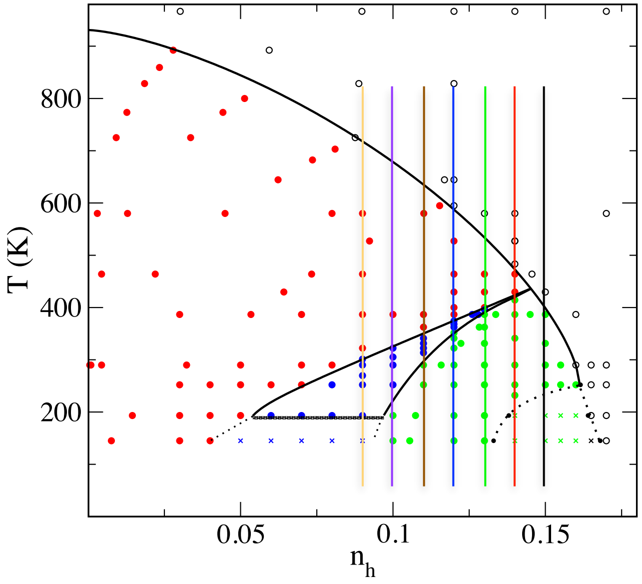


- L phase
- E phase
- C phase

n_h - hole concentration ($N=2-n_h$)

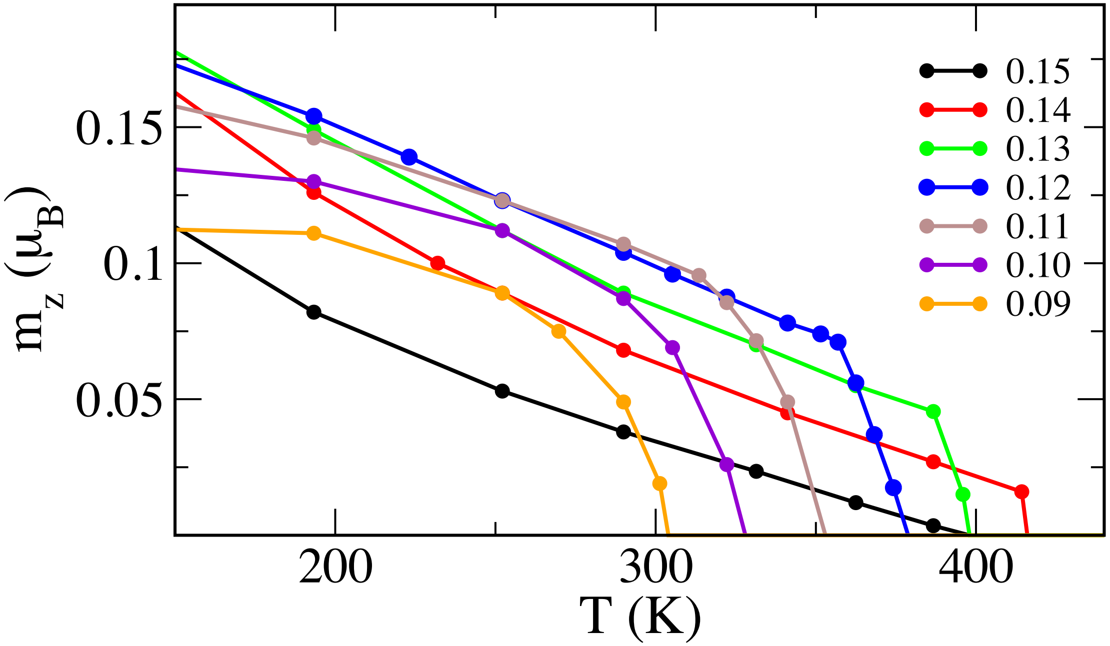
Excitonic condensation (doping) - all phases

n-T phase diagram



Ferromagnetic magnetization

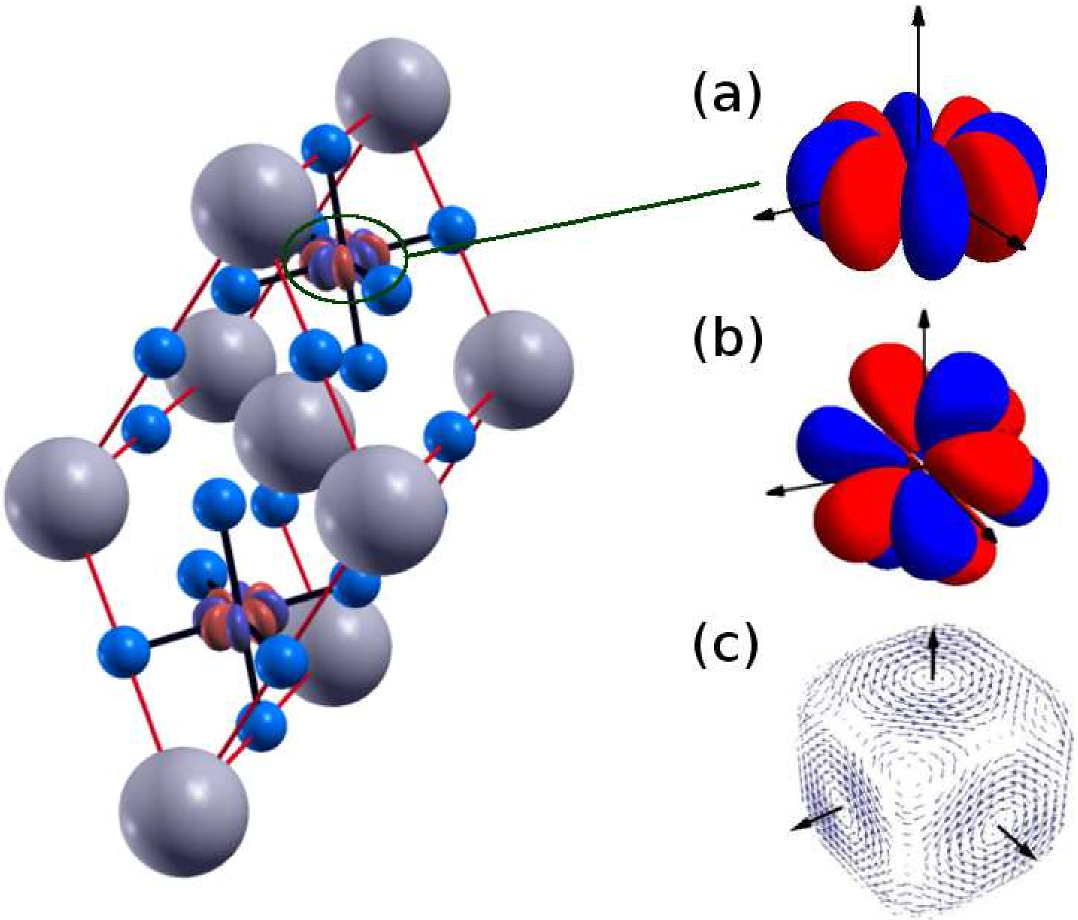
$$\langle m_z \rangle \quad || \quad i(\bar{\phi} \wedge \phi)$$



Orbital degeneracy - d^6 perovskites

LDA+U (static mean-field) solutions with excitonic order for hypothetical cubic structure of $LaCoO_3$

$LaCoO_3$ AF-EC order



	local spin density	ϕ^{α}_{β}	residual group
(a)		$\begin{pmatrix} 0 & 0 & X \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	$\tilde{D}_{4h} \times U(1)$
(b)		$\begin{pmatrix} 0 & 0 & X \\ 0 & 0 & X \\ 0 & 0 & X \end{pmatrix}$	$\tilde{D}_{3d} \times U(1)$
(c)		$\begin{pmatrix} X & 0 & 0 \\ 0 & X & 0 \\ 0 & 0 & X \end{pmatrix}$	O_h

Conclusions

• Solids close to spin-state transition may be unstable towards condensation of spinful excitons.

• The excitonic order parameter has complex structure and allows multiple phases.

• The excitonic order may lead to a long-range order of magnetic multipoles or local spin currents, but can also induce ferromagnetic polarisation.

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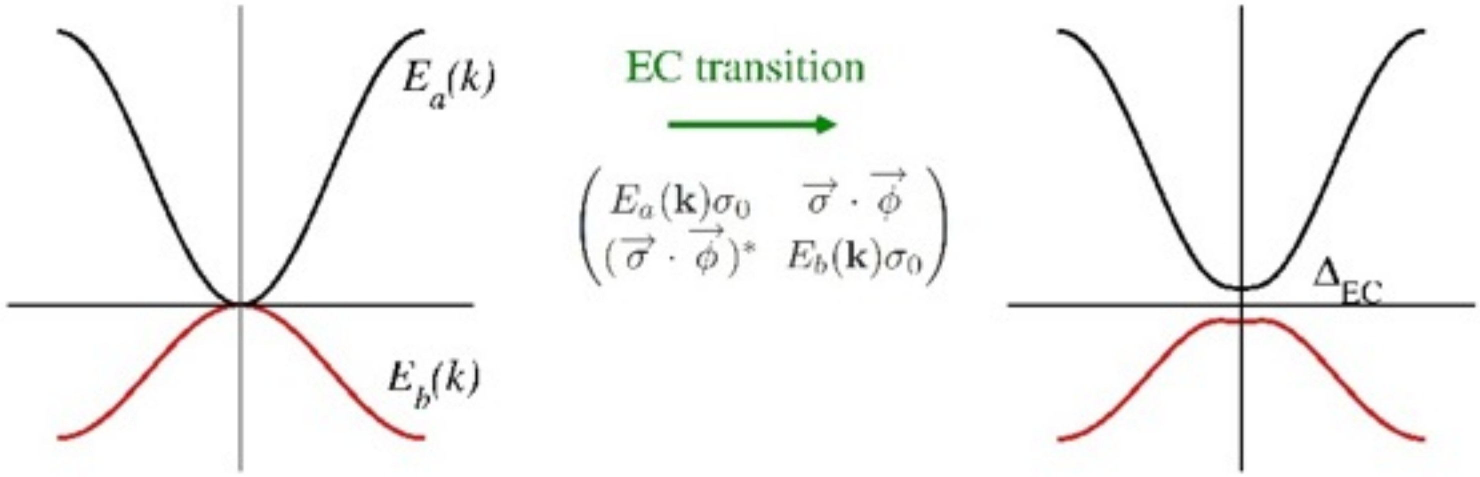
arXiv:1410.5198

Excitonic insulator



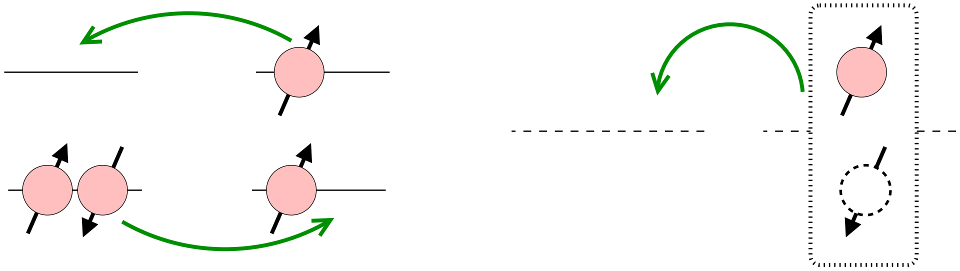
A band insulator with a very narrow gap (positive or negative) is unstable towards opening of a gap due to electron-hole attraction - condensation of excitons.

The gap can have spin-singlet or spin-triplet symmetry and be real or imaginary. Which of these options is realised depends on the interaction term and details of the band structure.

Mott, 1961
Halperin and Rice, 1968

Strong coupling picture of excitonic condensation

Strong coupling: HS states behave as hard-core bosons with the vacuum state $|\mathrm{vac}\rangle \equiv |\mathrm{LS}\rangle$



$$H_{\rm eff} = \sum_{i,s} \mu n_{i,s} + J_{\perp} \sum_{i,j,s} d^{\dagger}_{i,s} d_{j,s} + J_{\parallel} \sum_{\langle ij \rangle, s, s'} n_{i,s} n_{j,s'} + J_0 \sum_{\langle ij \rangle} S^z_i S^z_j$$

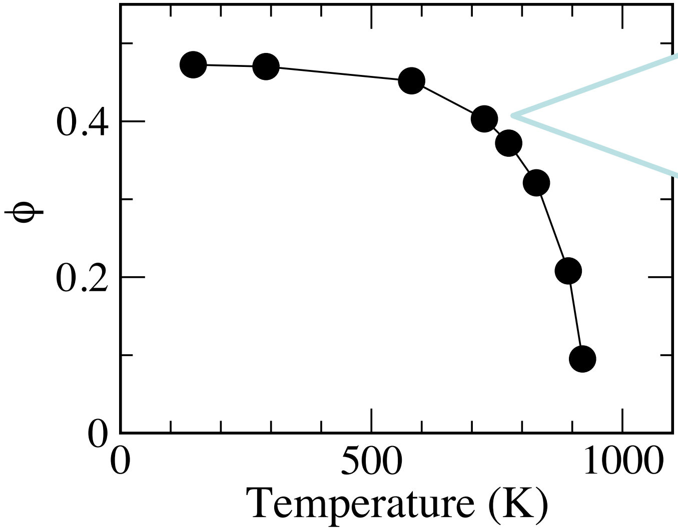
$$d^{\dagger}_{1} = a^{\dagger}_{\uparrow} b_{\downarrow}$$
$$d^{\dagger}_{-1} = a^{\dagger}_{\downarrow} b_{\uparrow}$$

Bose-Einstein condensation = spontanous hybridization between HS and LS states on the same site (breaks spin rotational symmetry)

Batista, 2001
Balents, 2000

Excitonic condensation (undoped) - L phase

order parameter $|\phi^+| = |\phi^-|$



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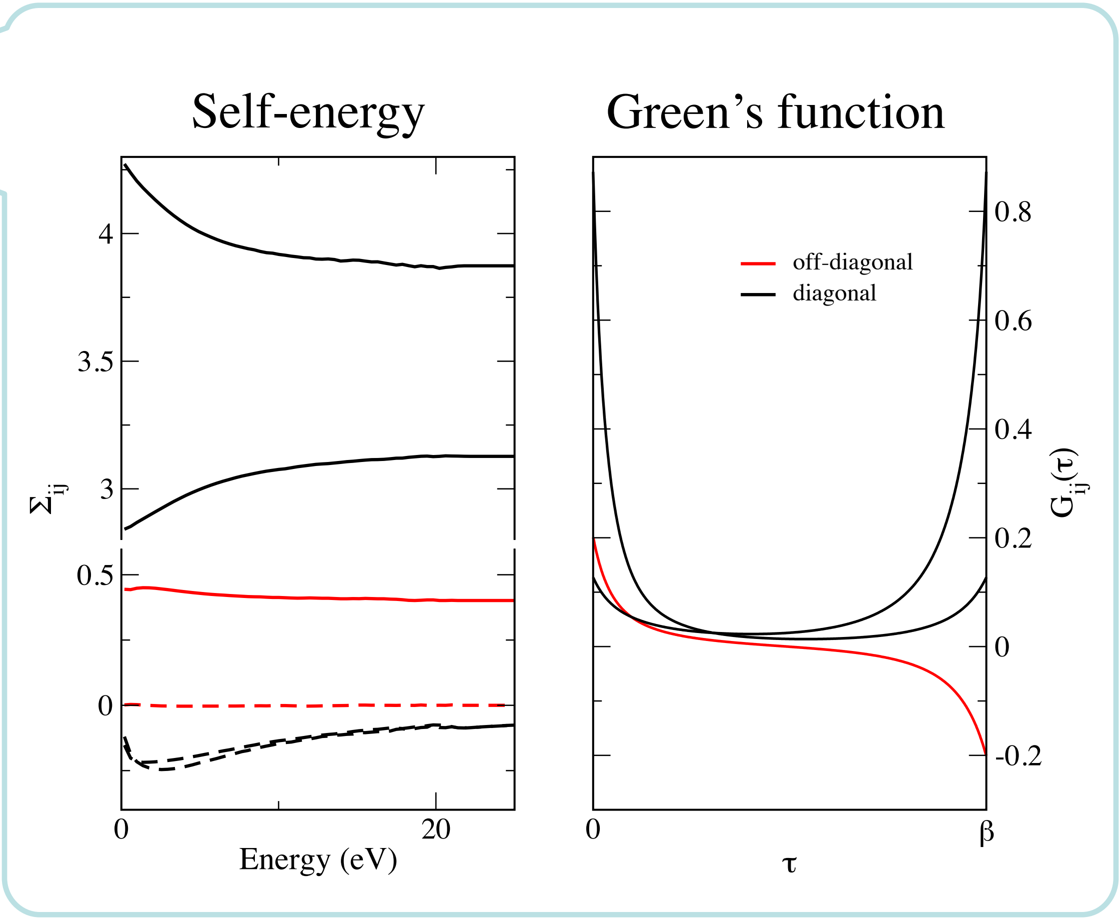
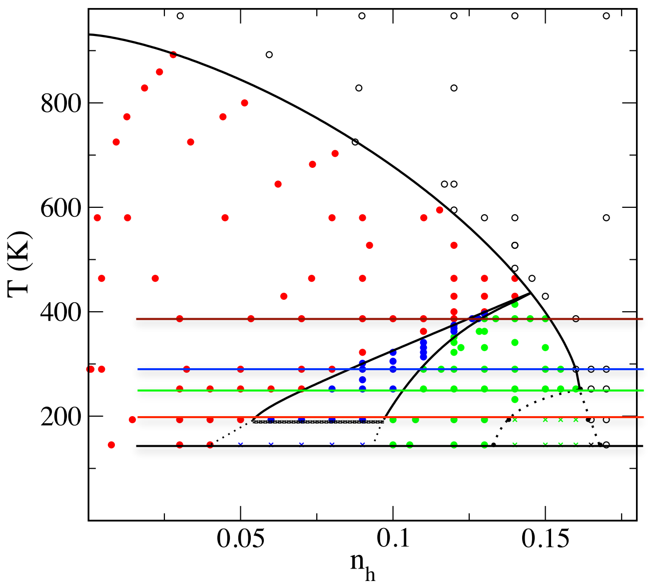


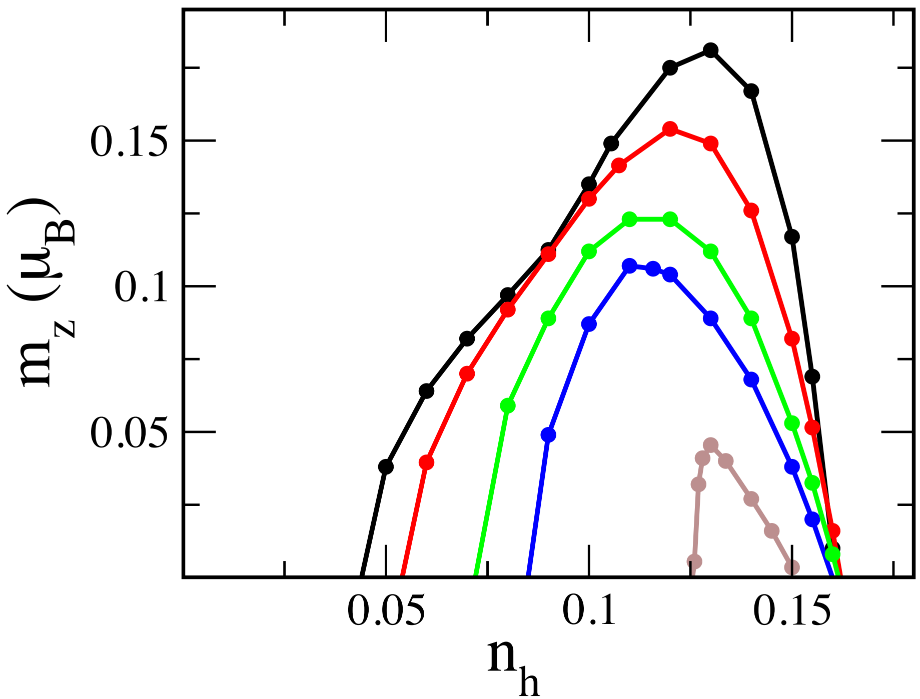
Excitonic condensation (doping) - all phases

n-T phase diagram



Ferromagnetic magnetization

$$\langle m_z \rangle \quad \| \quad i(\bar{\phi} \wedge \phi)$$



EC in cubic d^6 perovskite

Exciton = bound pair of e_g electron and t_{2g} hole

How do we detect the EC order?

Local d-occupation matrix (10 x 10):

spin structure:

$$\mathbf{D} = \begin{pmatrix} \mathbf{D}_0 + \boldsymbol{\phi}^z & \boldsymbol{\phi}^x + i\boldsymbol{\phi}^y \\ (\boldsymbol{\phi}^x + i\boldsymbol{\phi}^y)^* & \mathbf{D}_0 - \boldsymbol{\phi}^z \end{pmatrix}$$

orbital structure:

$$\phi^{\alpha}_{xy} d_{x^2-y^2} \otimes d_{xy} + \phi^{\alpha}_{zx} d_{z^2-x^2} \otimes d_{zx} + \phi^{\alpha}_{yz} d_{y^2-z^2} \otimes d_{yz}$$

The order parameter has 9 components (or 18 real components)

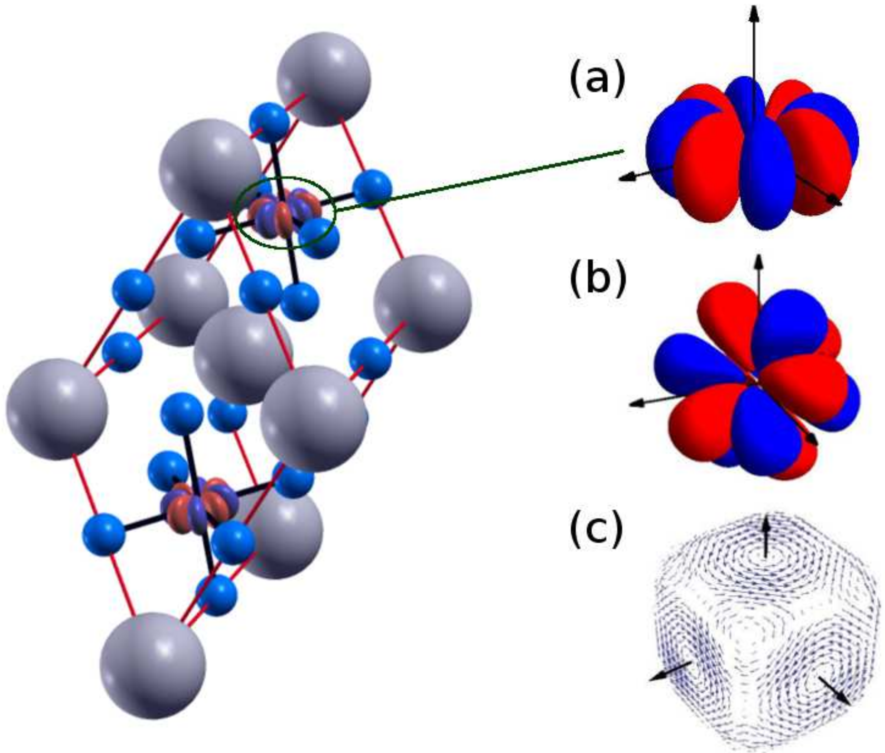
ϕ^{α}_{β}

α = x, y, z transforms like a vector under spin rotations
β = x, ŷ, ẑ transforms like a pseudovector under O_h operations

The spin and orbital symmetry does not specify the ordered phase uniquely, possible solutions can be classified by their residual symmetry.

Examples of LDA+U EC solutions

$LaCoO_3$ AF-EC order



$$\phi_\beta^\alpha$$

$$\begin{pmatrix} 0 & 0 & X \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

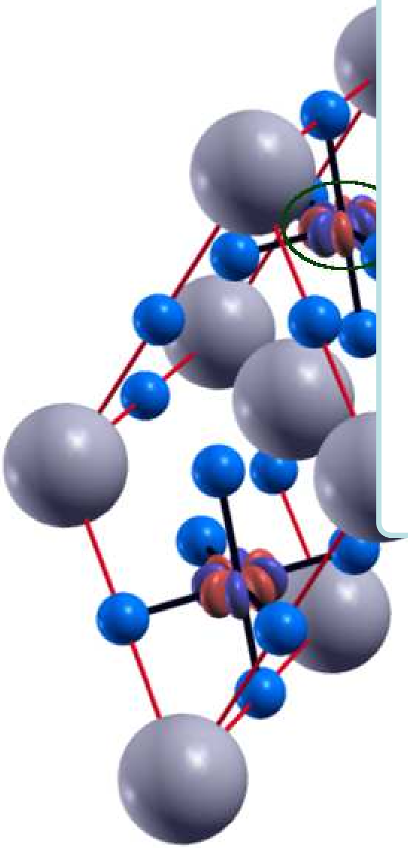
$$\begin{pmatrix} 0 & 0 & X \\ 0 & 0 & X \\ 0 & 0 & X \end{pmatrix}$$

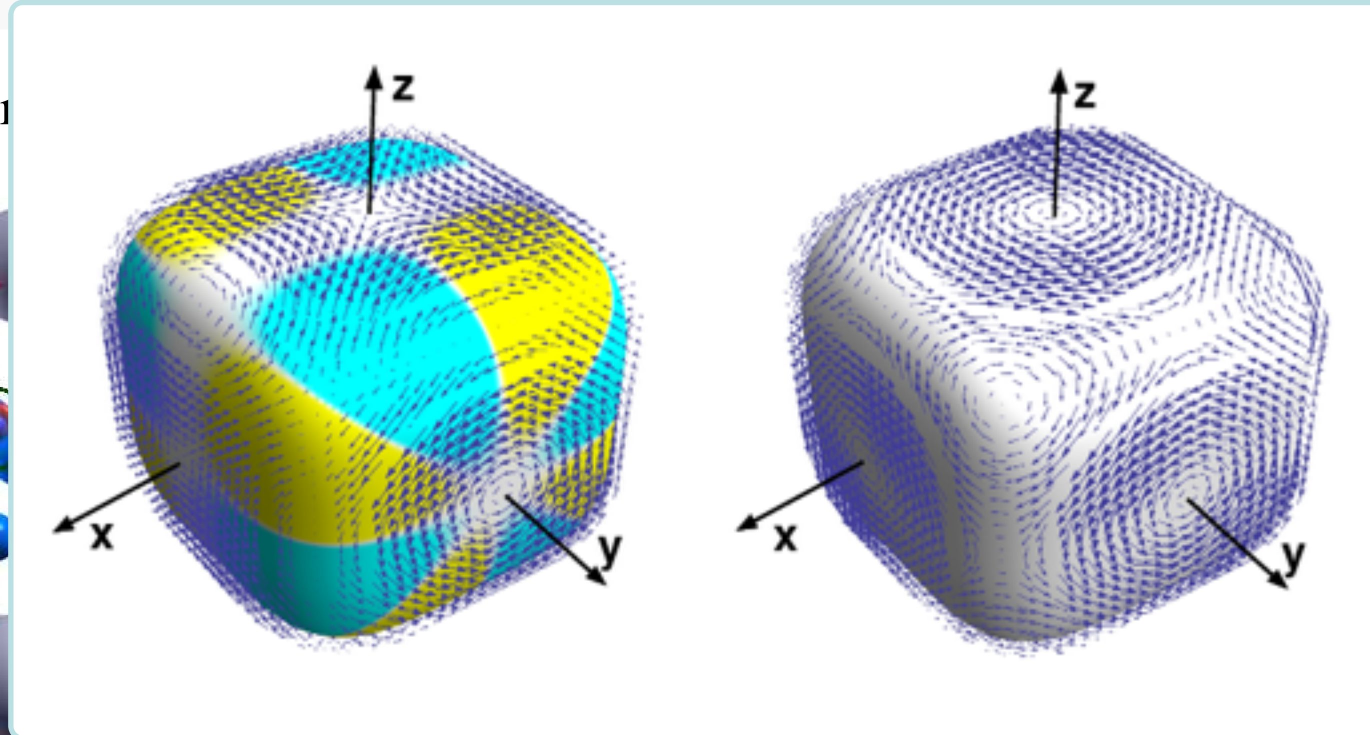
$$\begin{pmatrix} X & 0 & 0 \\ 0 & X & 0 \\ 0 & 0 & X \end{pmatrix}$$

i	X	$E_{(i)}$ [meV/f.u.]
1	0.182	-43
2	0.134	-73
3	0.144	-82

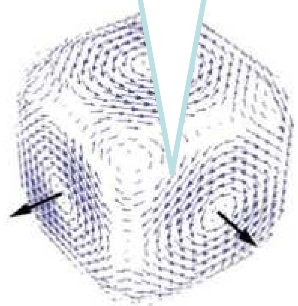
Examples of EC solutions

$LaCoO_3$ AF-EC o… …up





(c)

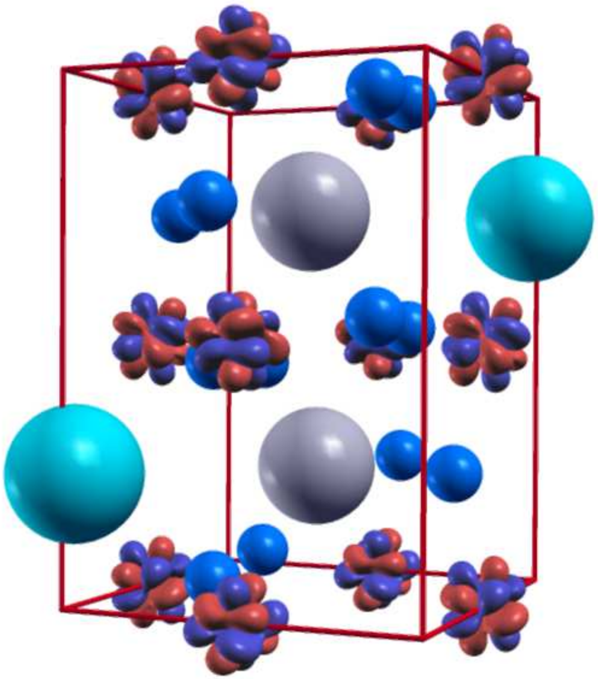


$$\begin{pmatrix} X & 0 & 0 \\ 0 & X & 0 \\ 0 & 0 & X \end{pmatrix} \qquad O_h$$

Examples of LDA+U for PCCO

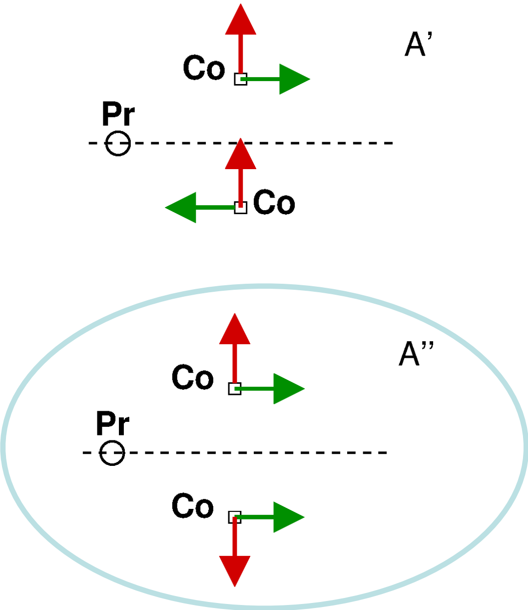
orthorhombic structure: 4 Co atoms per f.u. two inequivalent Co positions

Product solution:



	1	2	3	4
ϕ_{yz}	0.182	0.182	0.216	0.216
ϕ_{zx}	0.228	0.228	-0.212	-0.212
ϕ_{xy}	-0.071	0.071	-0.093	0.093

Orbital pseudovectors on sym. related Co atoms:

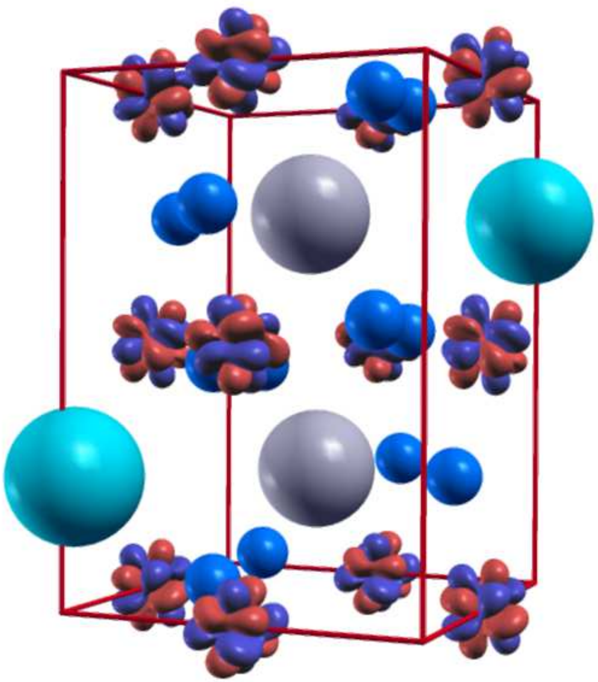


Origin of exchange splitting on Pr

Coupling of Pr 4f[1] spin to p-d orbitals: effective multi-channel Kondo Hamiltonian

$$H^{(n)} = \sum_{\alpha\alpha'} \sum_{mm'} \sum_{i} \mathbf{S} \cdot \boldsymbol{\sigma}_{\alpha\alpha'} J^{(n)}_{i,mm'} c^{\dagger}_{im\alpha} c_{im'\alpha'} + \text{c.c.}$$

Below T_c effective exchange field appears: $h^{(n)}_{\gamma} = \sum_{imm'} J^{(n)}_{i,mm'} \sum_{\alpha\alpha'} 2\,\mathrm{Re}\langle c^{\dagger}_{im\alpha} \sigma^{\gamma}_{\alpha\alpha'} c_{im'\alpha'} \rangle$



The site symmetry of the EC order parameter with respect to the Pr site decides whether contributions of from different Co site interfere constructively or destructively.

For the present EC solution h=0 in the absence of spin-orbit coupling in Pr 4f shell. With SOC splitting on 10 meV scale is obtained.