Excitonic condensation of strongly correlated electrons

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Akademie věd ČR, v. v. i.

Magnetism of 'non-magnetic ions'

single atom

energy spectrum:

magnetic multiplet

ground state (S=0)

Examples: Co^{3+}, Fe^{2+}, Ir^{5+}, Os^{4+}, Ru^{4+} in cubic crystal field

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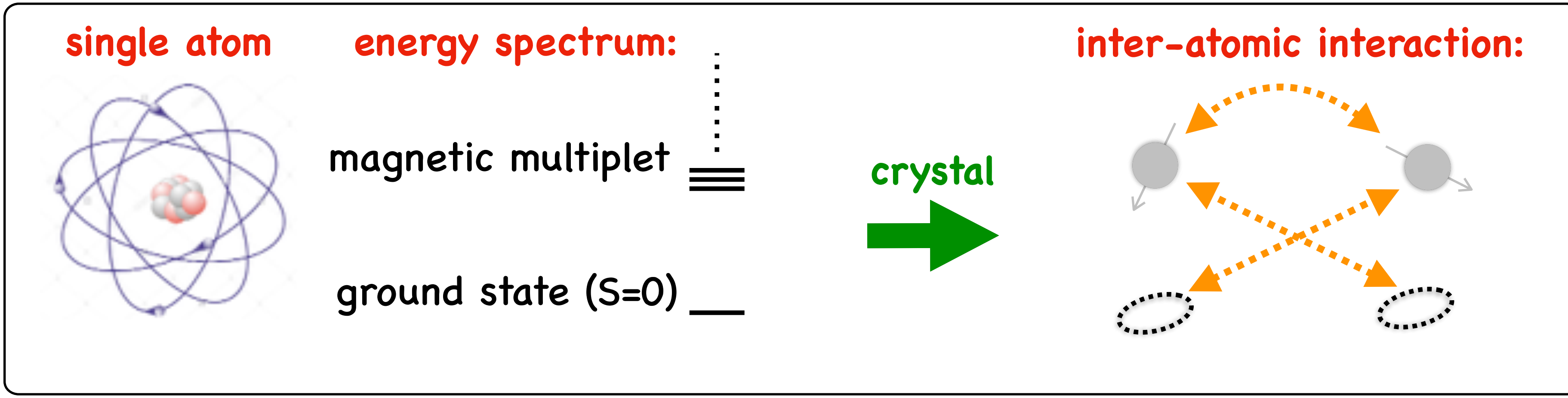
combing a bold head

Magnetism of ‘non-magnetic ions’

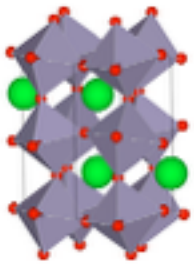


Examples: Co^{3+}, Fe^{2+}, Ir^{5+}, Os^{4+}, Ru^{4+} in cubic crystal field

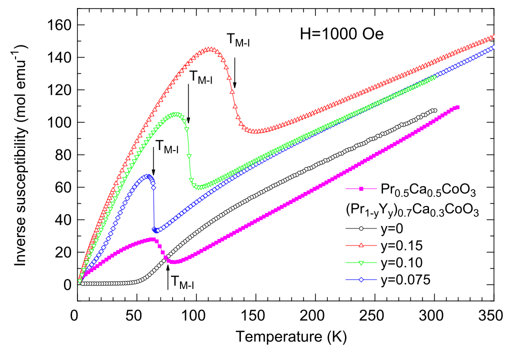
*L. Balents, PRB **62**, 2346 (2000)*
*G. Khaliullin, PRL **111**, 197201 (2013)*
*JK and P. Augustinský, PRB **90**, 235112 (2014)*
T. Kaneko, Y. Ohta and S. Yunoki, PRB 97, 155131 (2018)
*J. Nasu et al., PRB **93**, 205136 (2016)*
T. Tatsuno et al., JPSJ 85, 083706 (2016)
*T. Yamaguchi, K. Sugimoto and Y. Ohta, JPSJ **86**, 043701 (2017)*

⋮

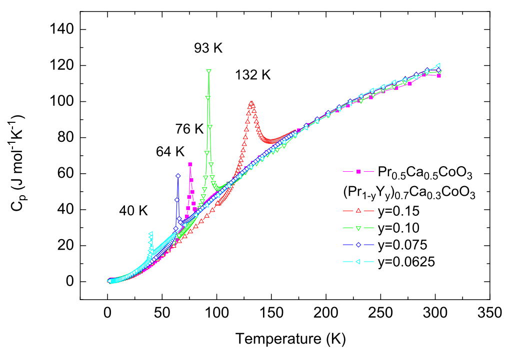
$(Pr_{1-y}Y_y)_{1-x}Ca_xCoO_3$



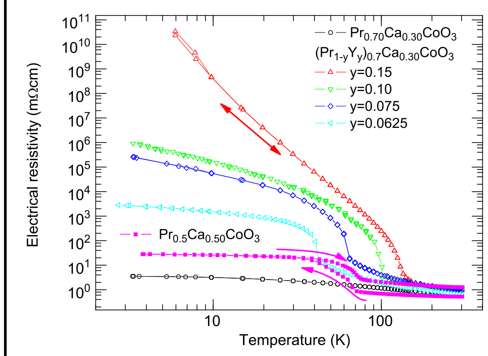
spin susceptibility



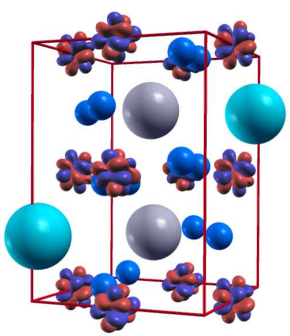
specific heat



transport

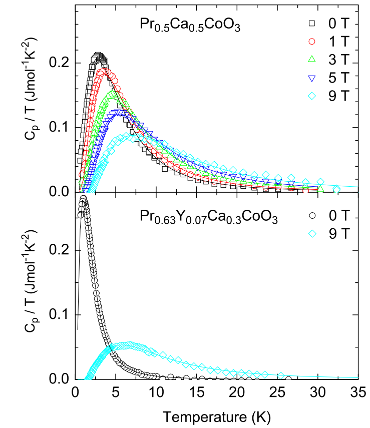


theory

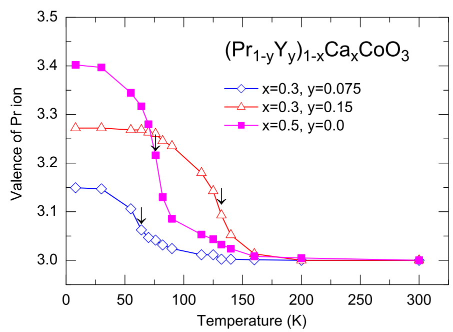


Crystal field

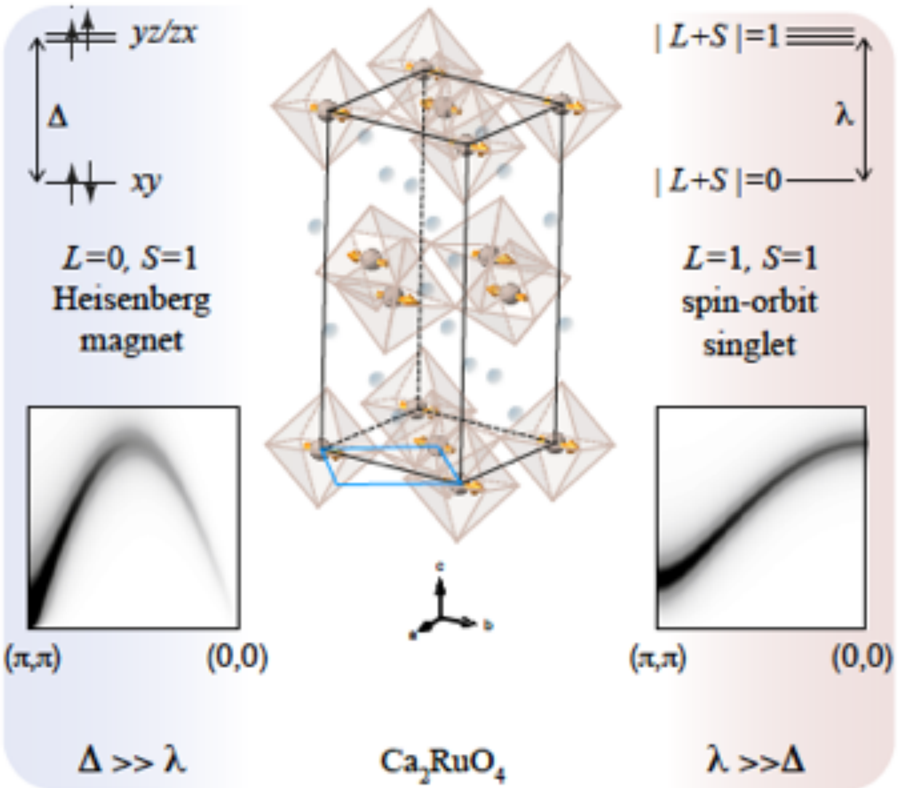
low-T specific heat

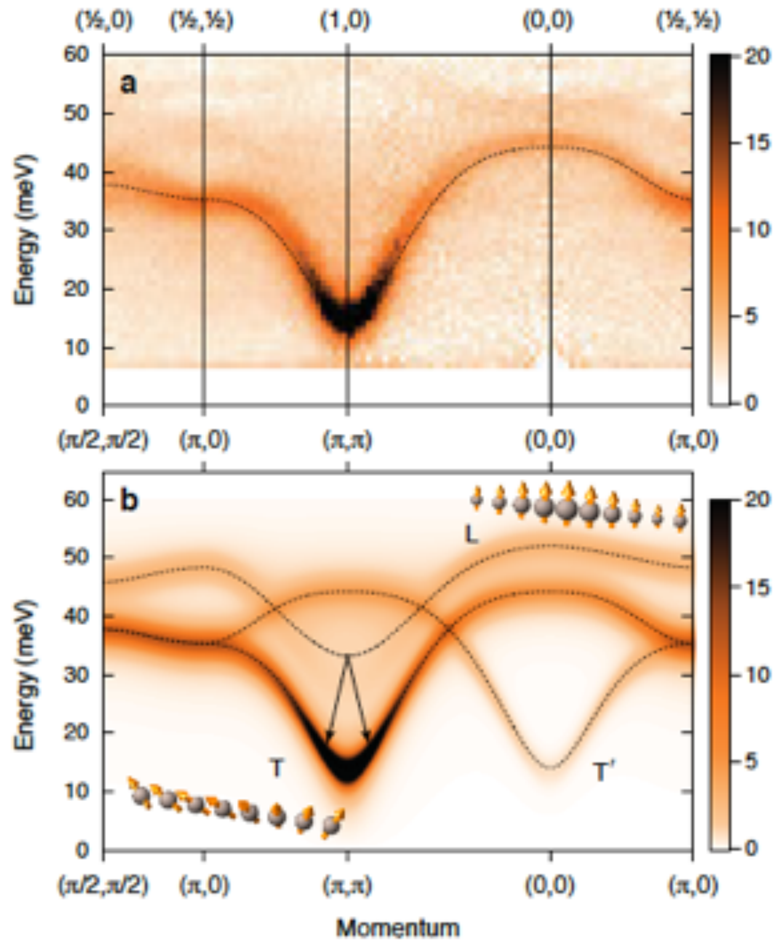


XAS->valence transition



Ca_2RuO_4

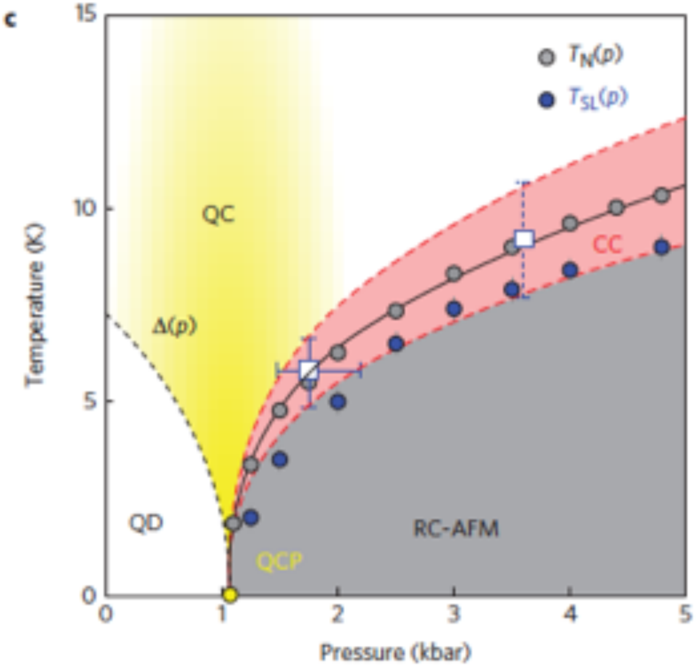


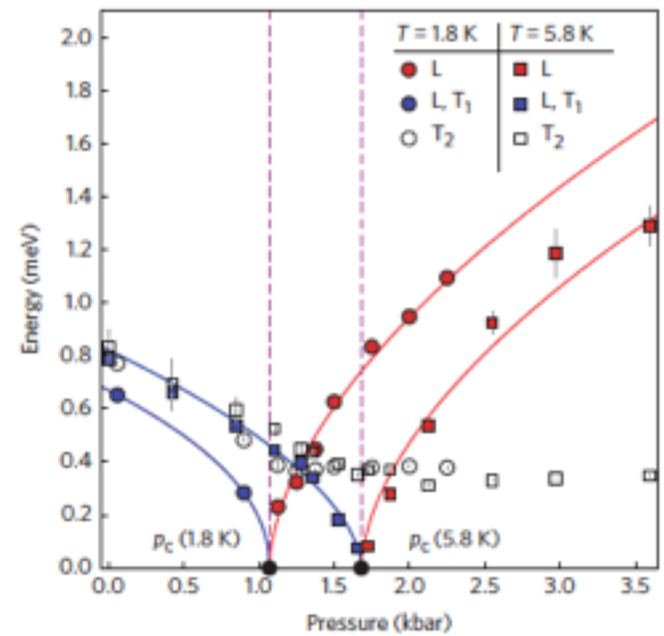


Spin-orbit coupling

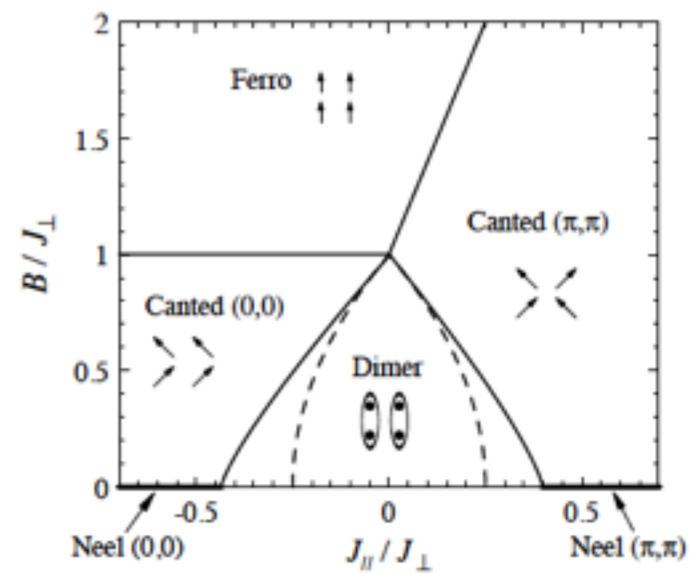
A. Jain et al., Nat. Phys. **13**, 633 (2017)

$TlCuCl_3$





bi-layer Heisenberg model

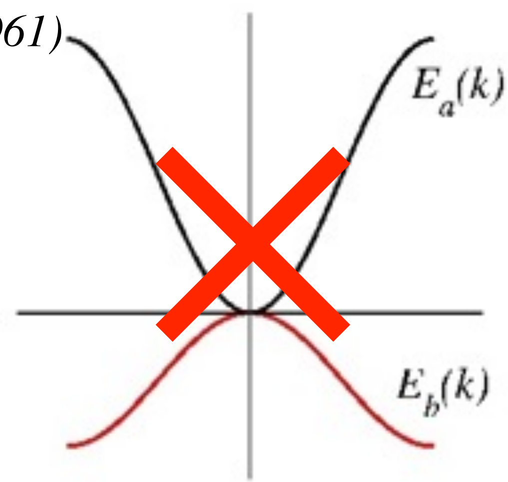


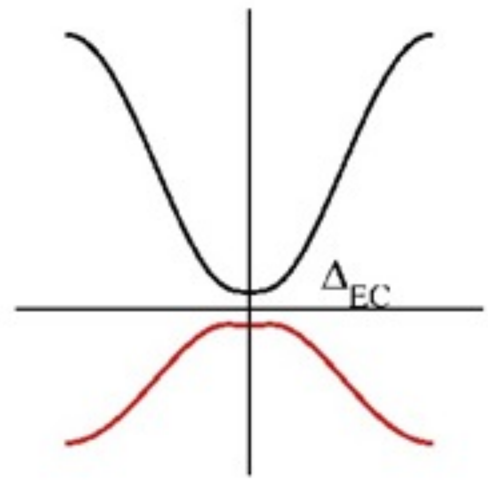
P. Merchant et al., Nat. Phys. ***10****, 373 (2014)*

Excitonic insulator

"The first point that we must make about this model is that the predicted continuous increase in the number of free electrons and holes from the value zero is not possible. An electron and a positive hole will attract each other … the electron and hole will always, when in the state of lowest energy, form pairs (excitons) …"

N. F. Mott, Philos. Mag. ***6****, 287 (1961)*



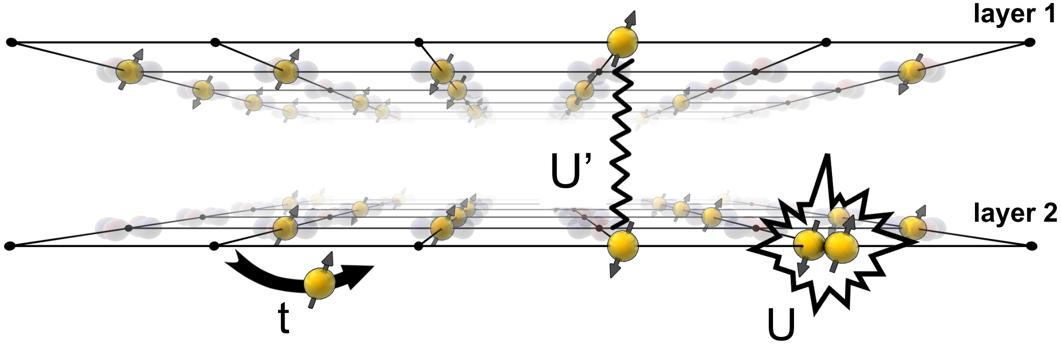


R. S. Knox, Solid State Phys. Sup
L. V. Keldysh and Y. V. Kopaev, Sov. Phys. Solid State ***6****, 2219 (1965)*
J. des Cloizeaux, J. Phys. Chem. Solids ***26****, 259 (1965)*
B. I. Halperin and T. M. Rice, Solid state physics, (1968)

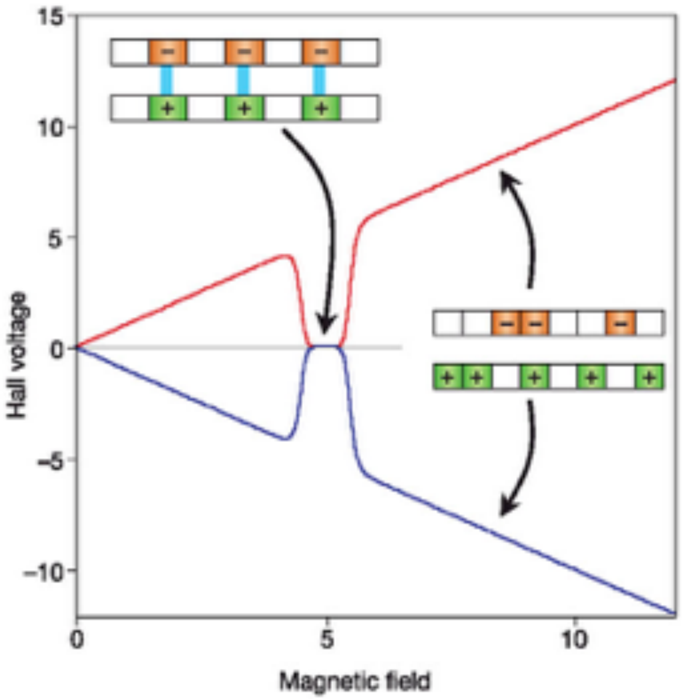
} BCS theories

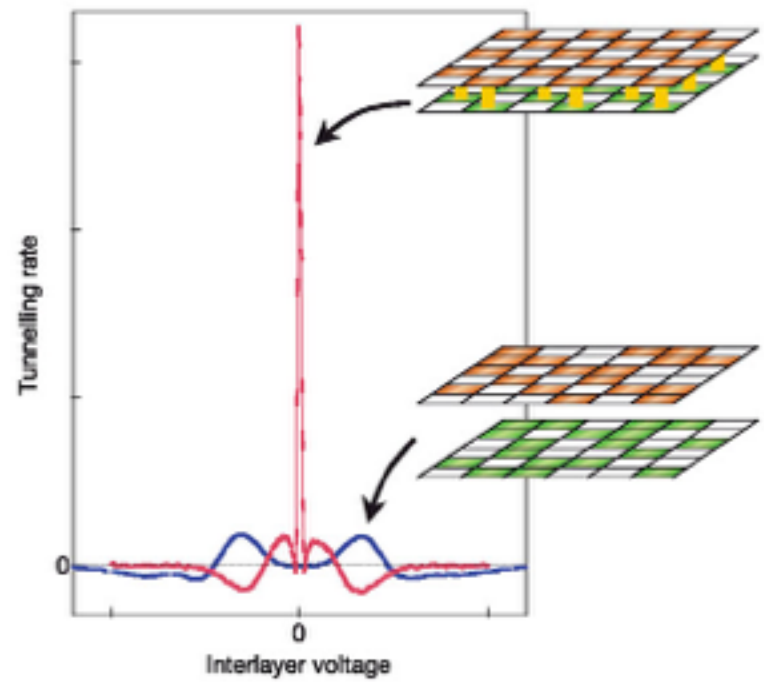
H. Cercellier et al., PRL ***99****, 146403 (2007); C. Monney et al. PRL* ***106****, 106404 (2011)*

Experimental proof of exciton condensation: bilayers



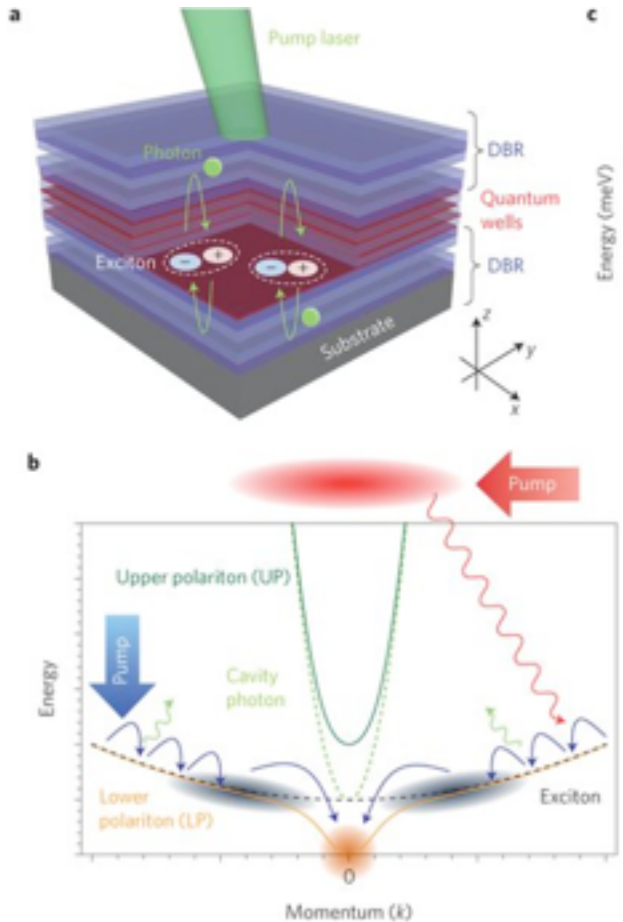
from Rademaker, 2014





*J. P. Eisenstein and A. H. MacDonald, Nature **432**, 691 (2004)*

Exciton-polariton condensates



PHYSICAL REVIEW LETTERS **122,** 017401 (2019)

Superradiant Quantum Materials

Giacomo Mazza[1,2,*] and Antoine Georges[2,3,1,4]

T. Byrnes, N. Young Kim and Y. Yamamoto, Nat. Phys. ***10****, 803 (2014)*

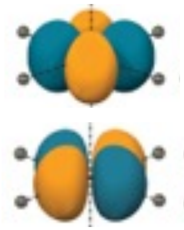
Outline

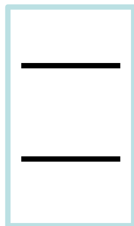
- Two-orbital Hubbard model: strong coupling
  DMFT results
- Real materials $(Pr_xR_{1-x})_yCa_{1-y}CoO_3$
- How to detect exciton condensate?

Model set-up

2-orbital atom with 2 electrons

orbitals:

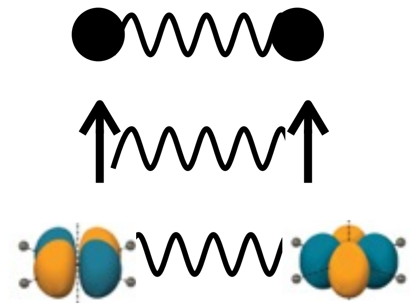




$E_0+\Delta$

E_0

e-e interaction:



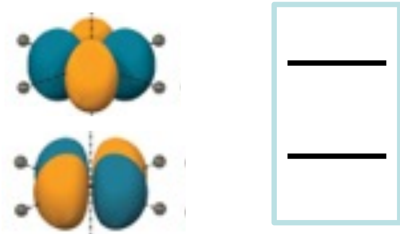
U

$-J$

$-2J$

2-orbital atom with 2 electrons

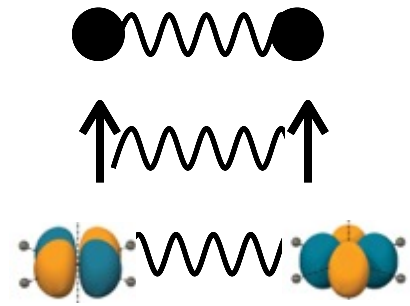
orbitals:



$E_0+\Delta$

E_0

e-e interaction:



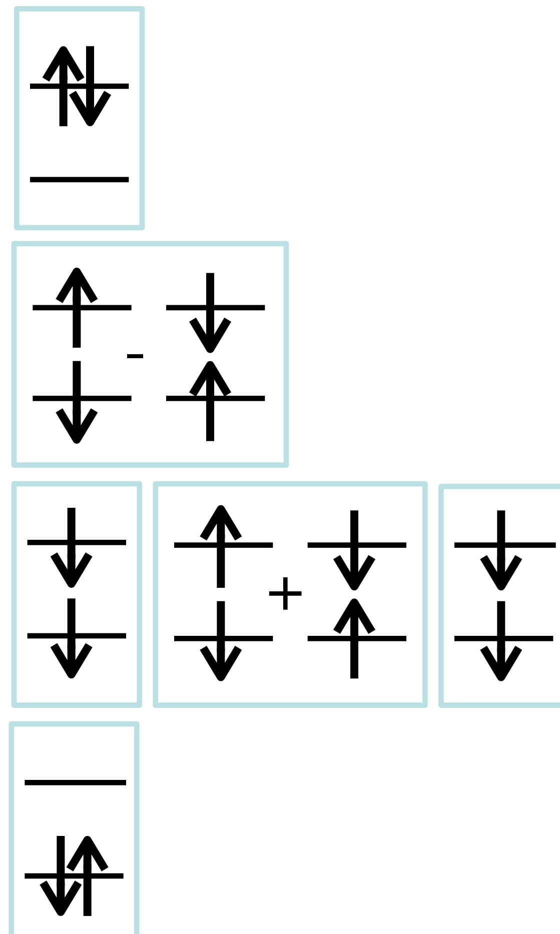
U

$-J$

$-2J$

2-electron configurations:

$\Delta \gtrsim 3J$



$2E_0+U+2\Delta$

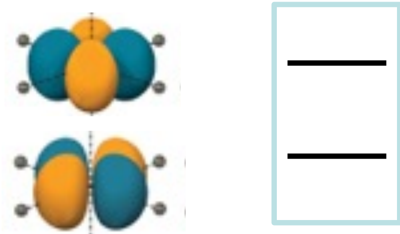
$2E_0+U+\Delta-2J$

$2E_0+U+\Delta-3J$

$2E_0+U$

2-orbital atom with 2 electrons

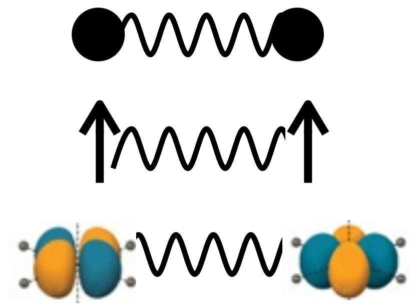
orbitals:



$E_0+\Delta$

E_0

e-e interaction:



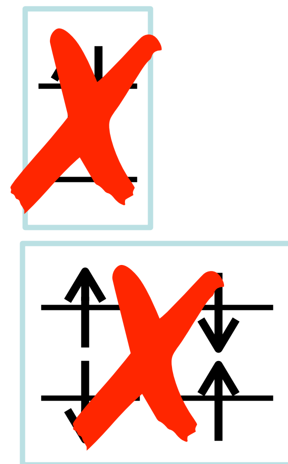
U

-J

-2J

2-electron configurations:

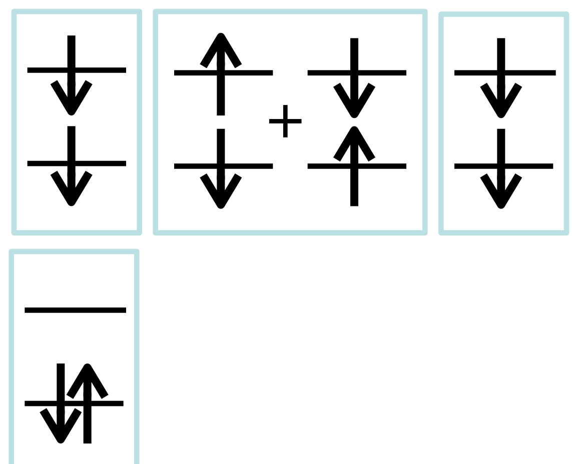
$\Delta \gtrsim 3J$



$2E_0+U+2\Delta$

$2E_0+U+\Delta-2J$

High-spin state (S=1)

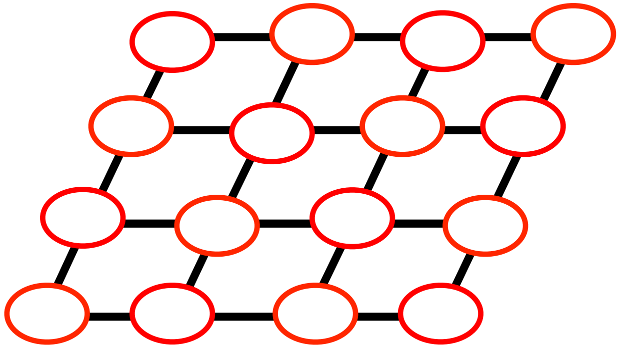


$2E_0+U+\Delta-3J$

Low-spin state (S=0)

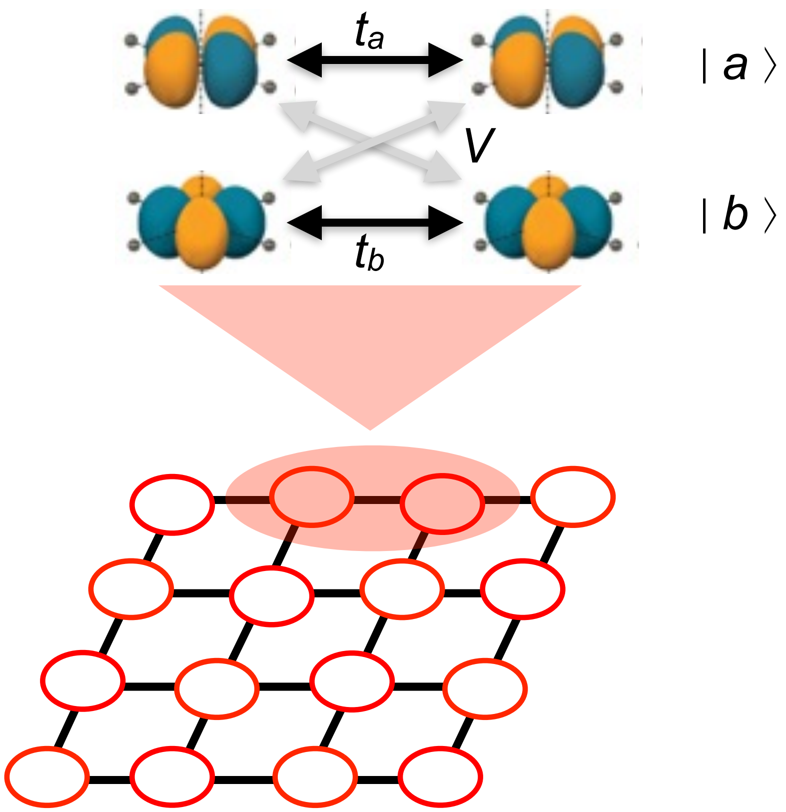
$2E_0+U$

Crystal of 2-orbital atoms



Crystal of 2-orbital atoms

Hopping between neighbors

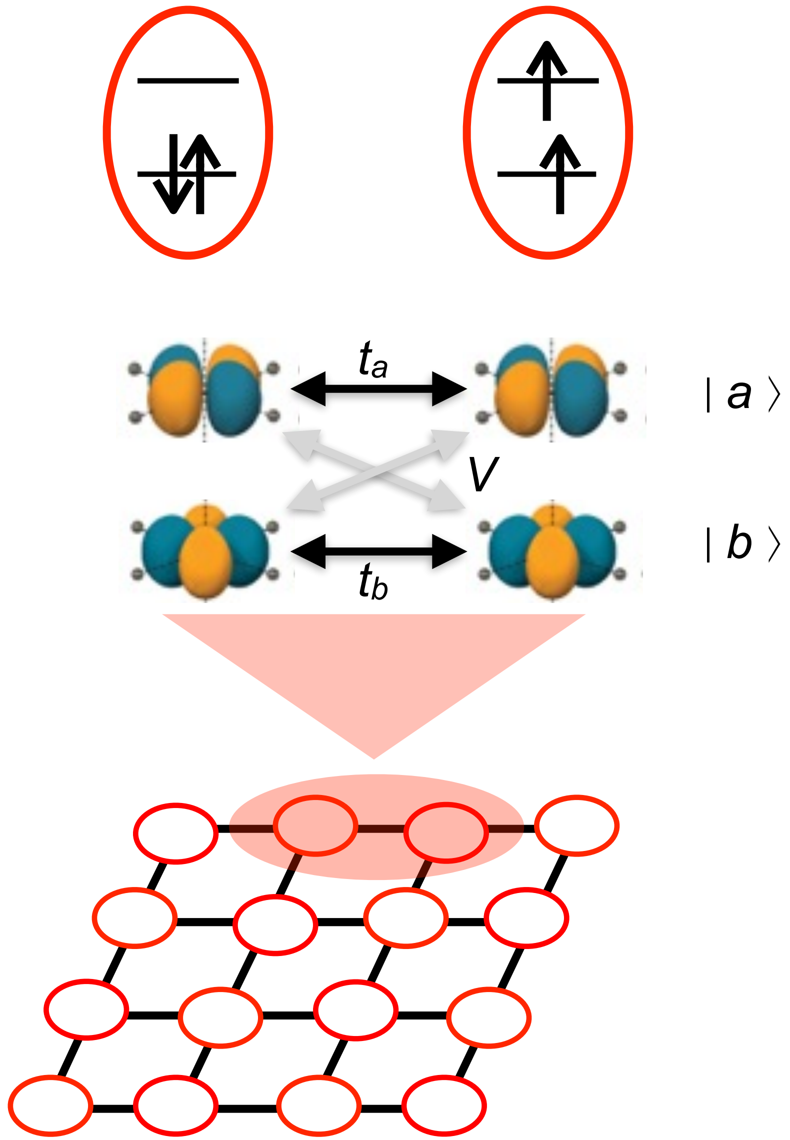


No Coulomb interaction between neighbors => Hubbard model

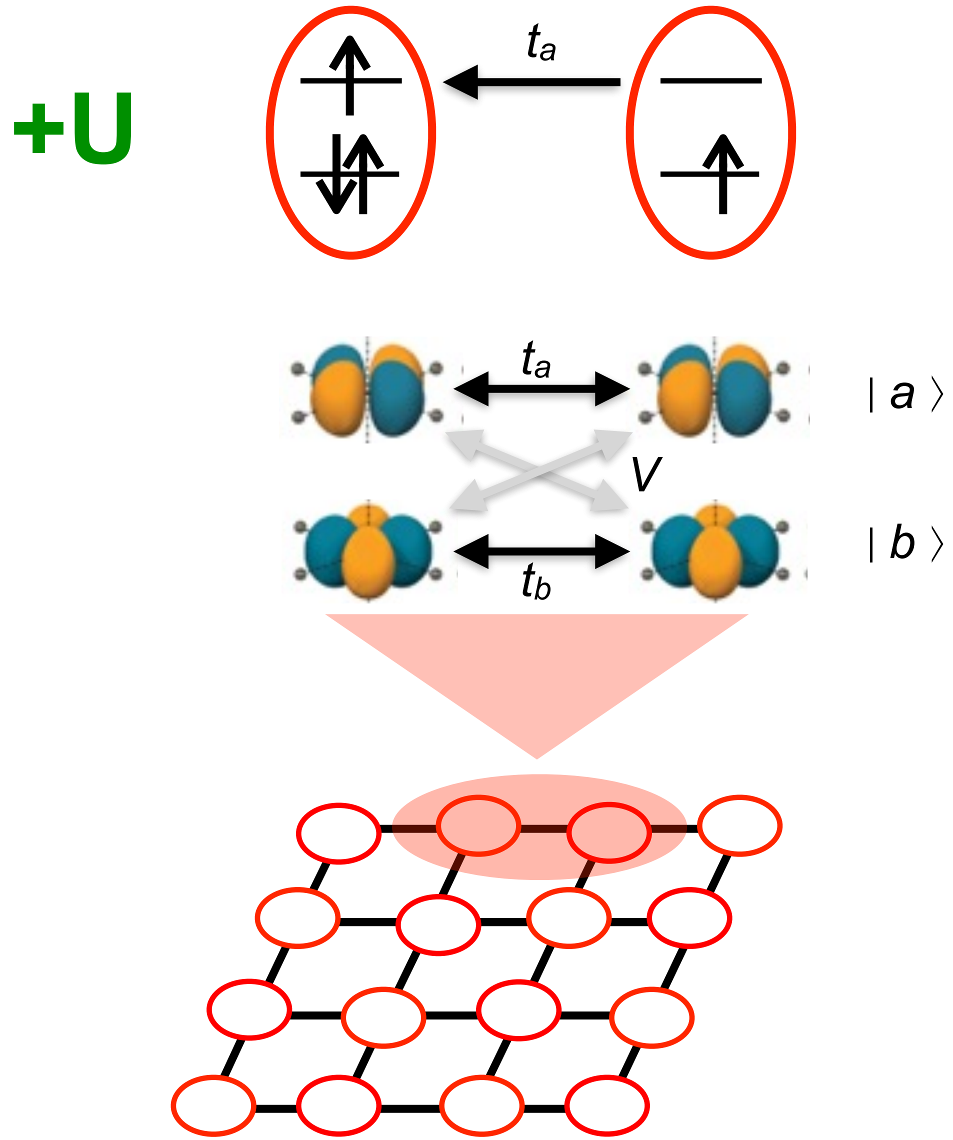
Strong-coupling limit (hard-core bosons)

$t \ll U$

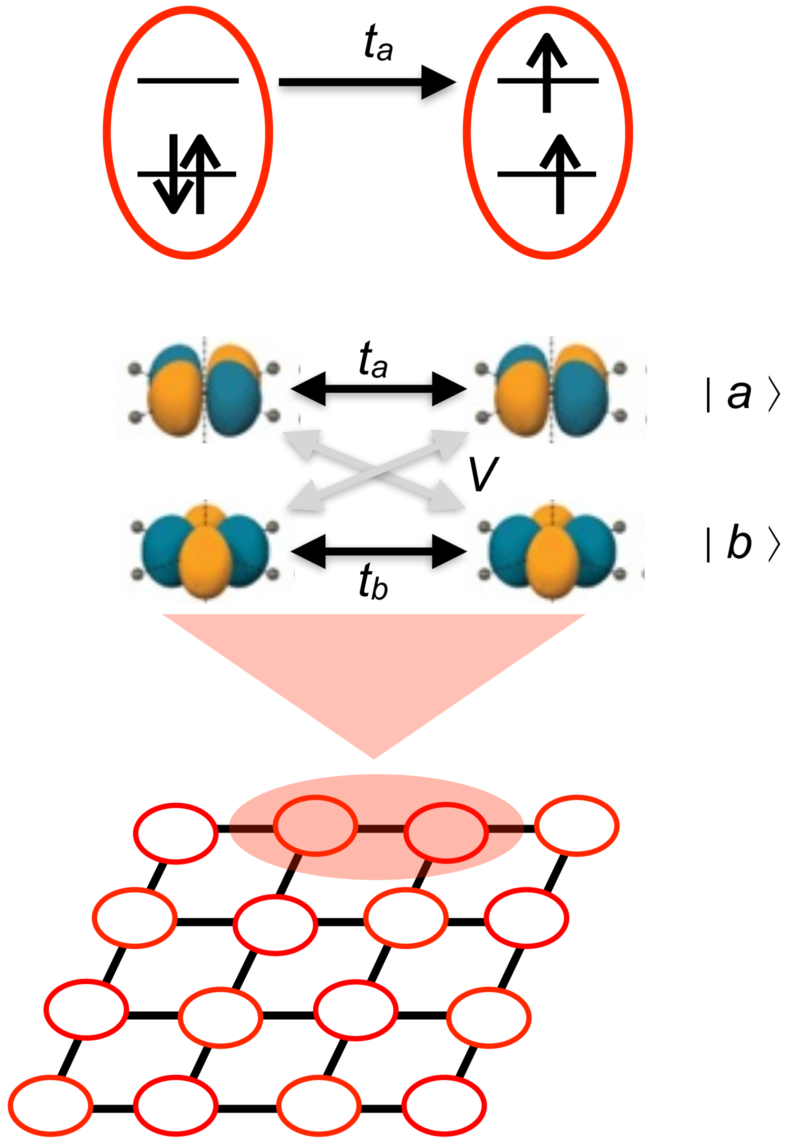
Crystal of 2-orbital atoms



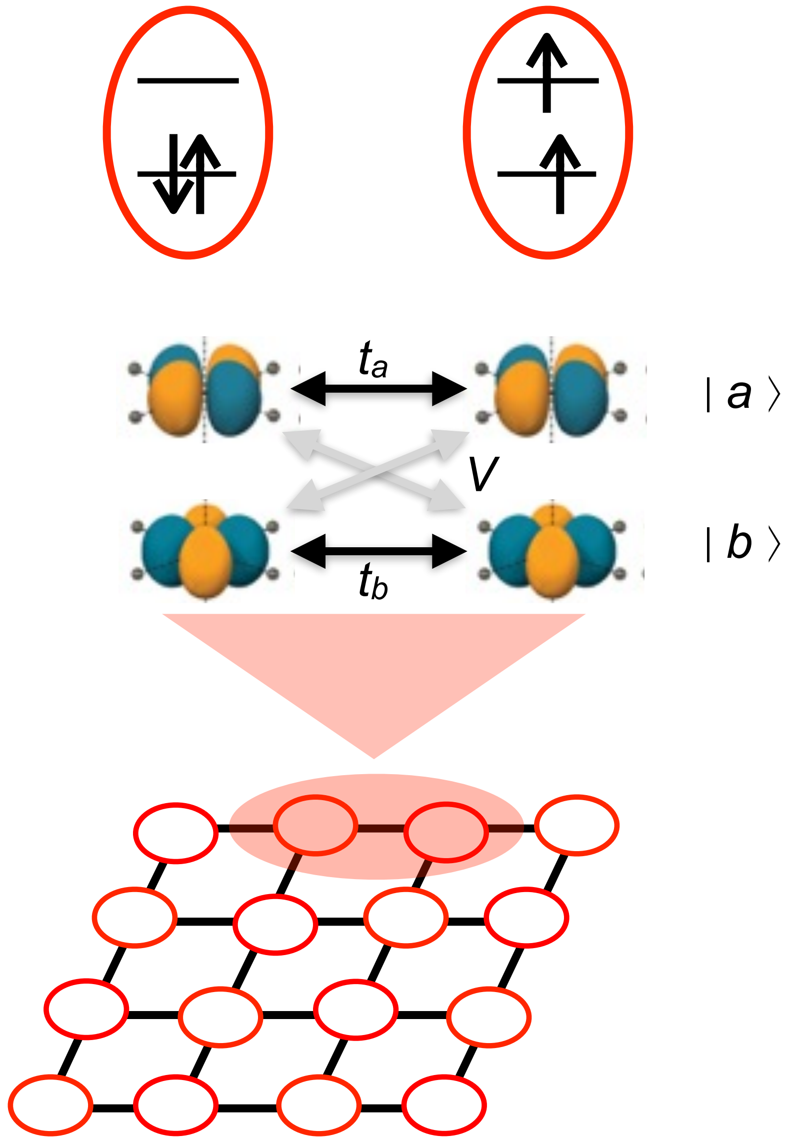
Crystal of 2-orbital atoms



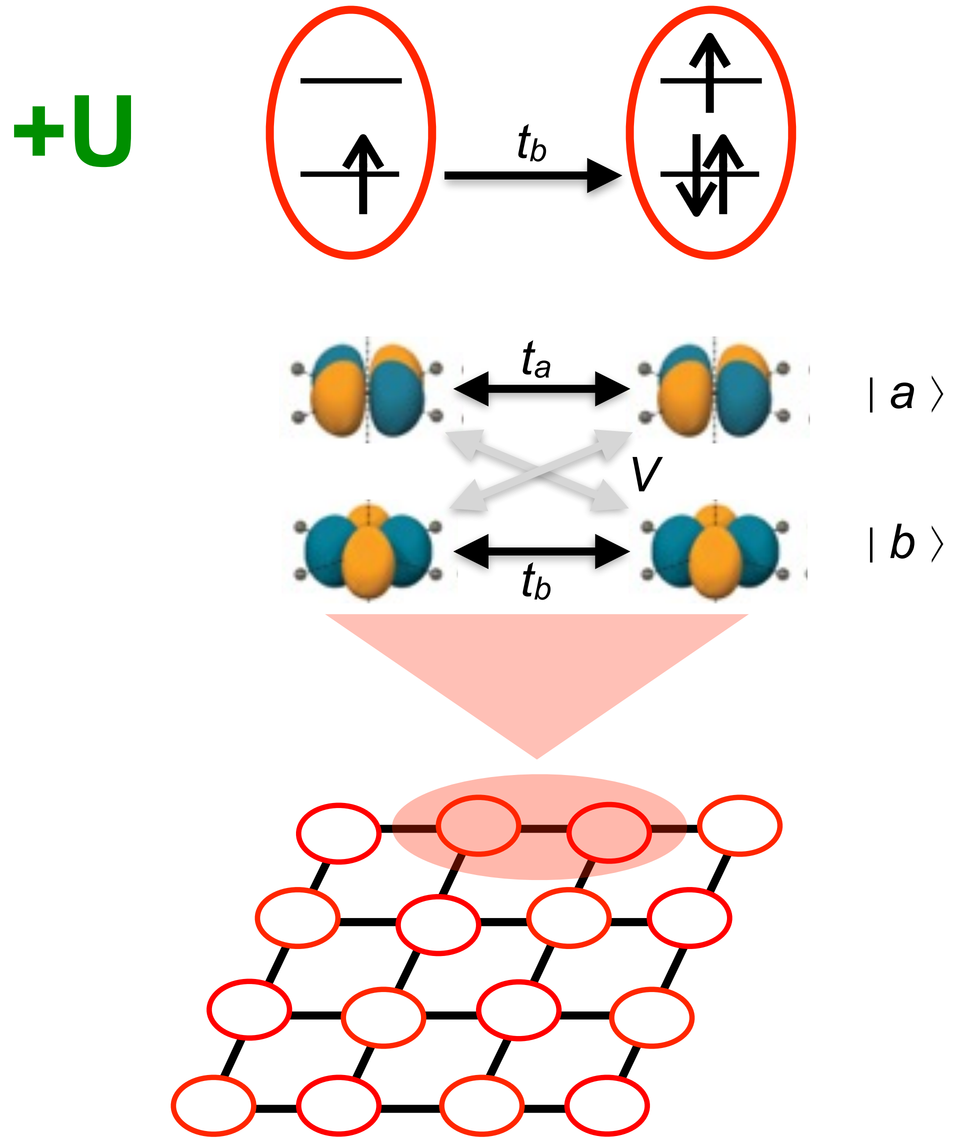
Crystal of 2-orbital atoms



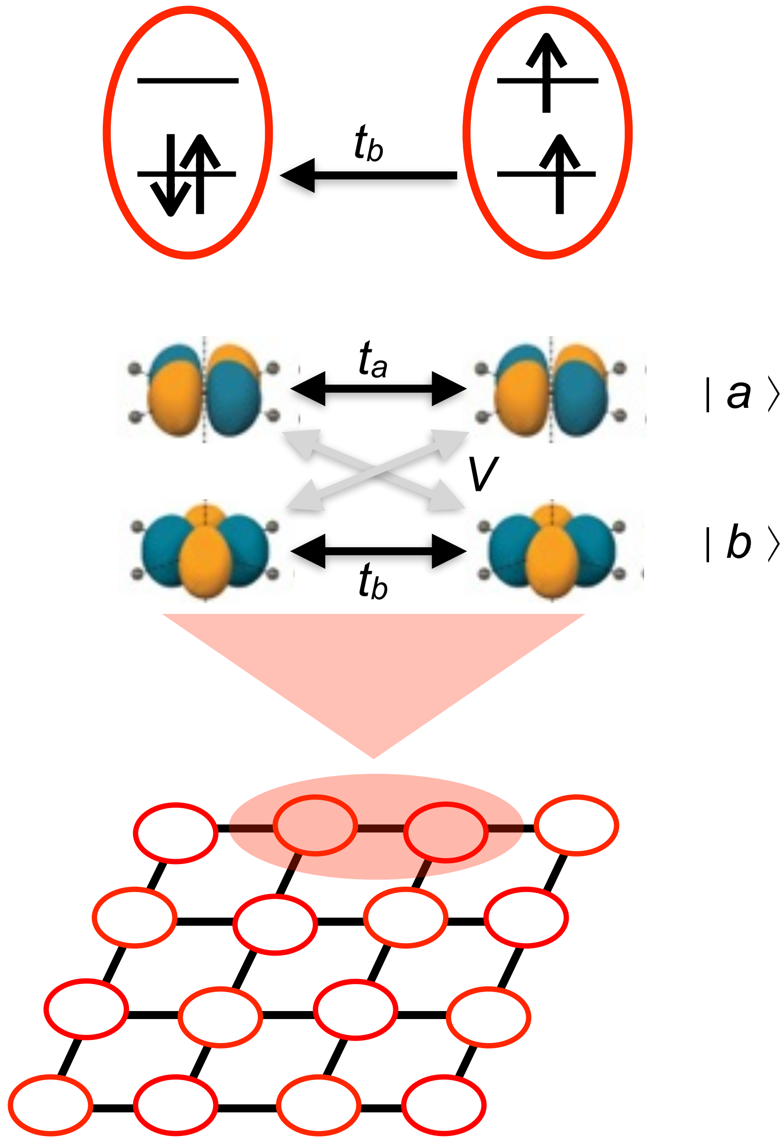
Crystal of 2-orbital atoms



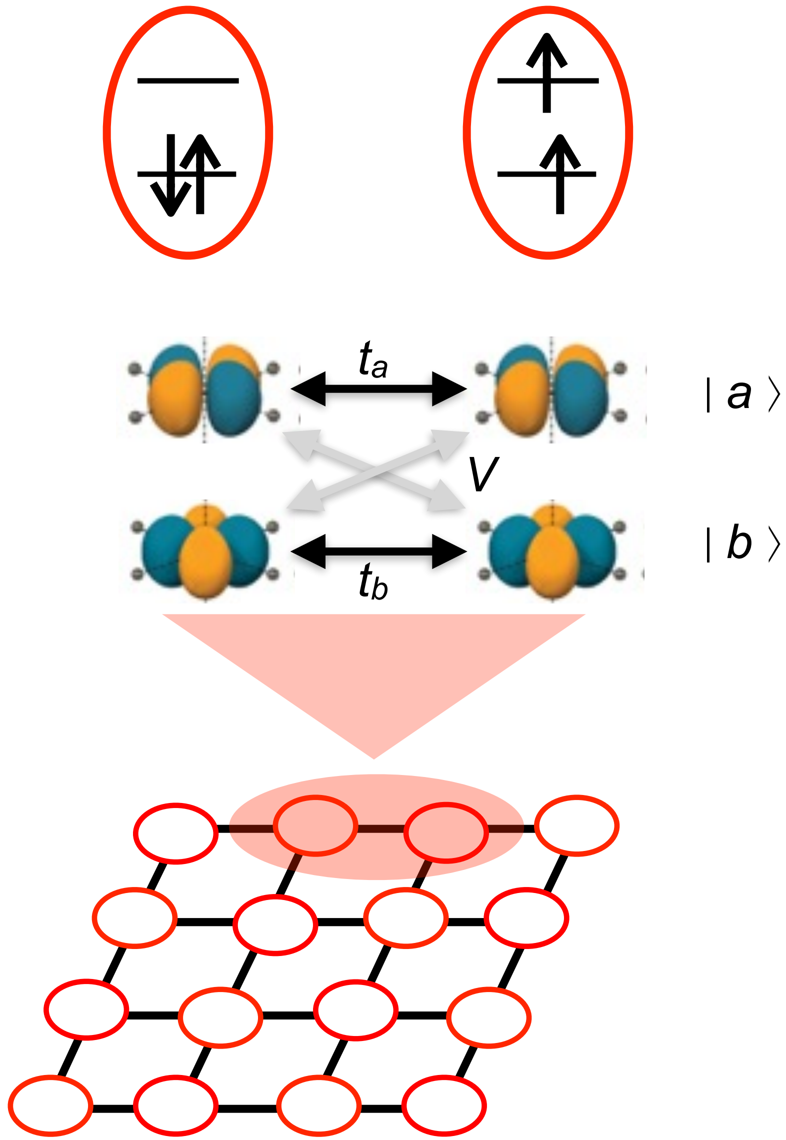
Crystal of 2-orbital atoms



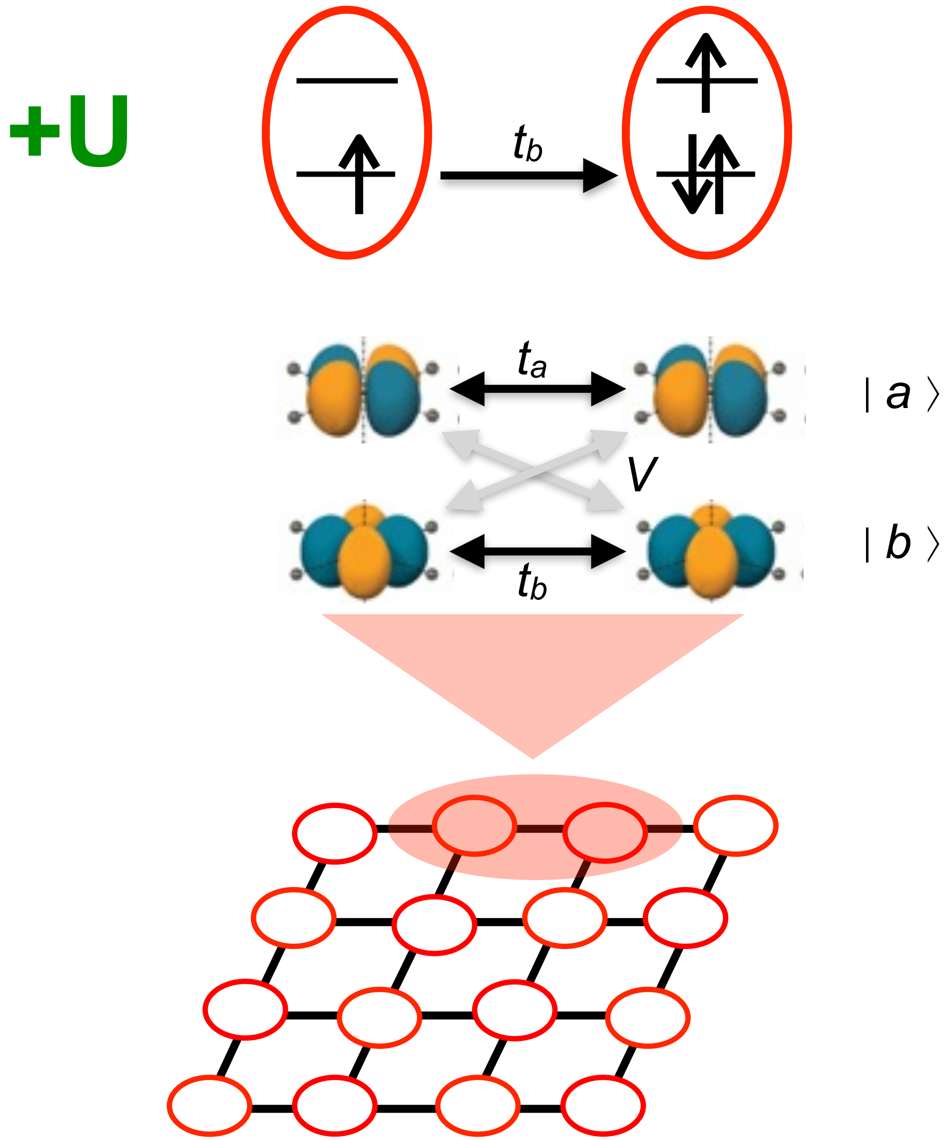
Crystal of 2-orbital atoms



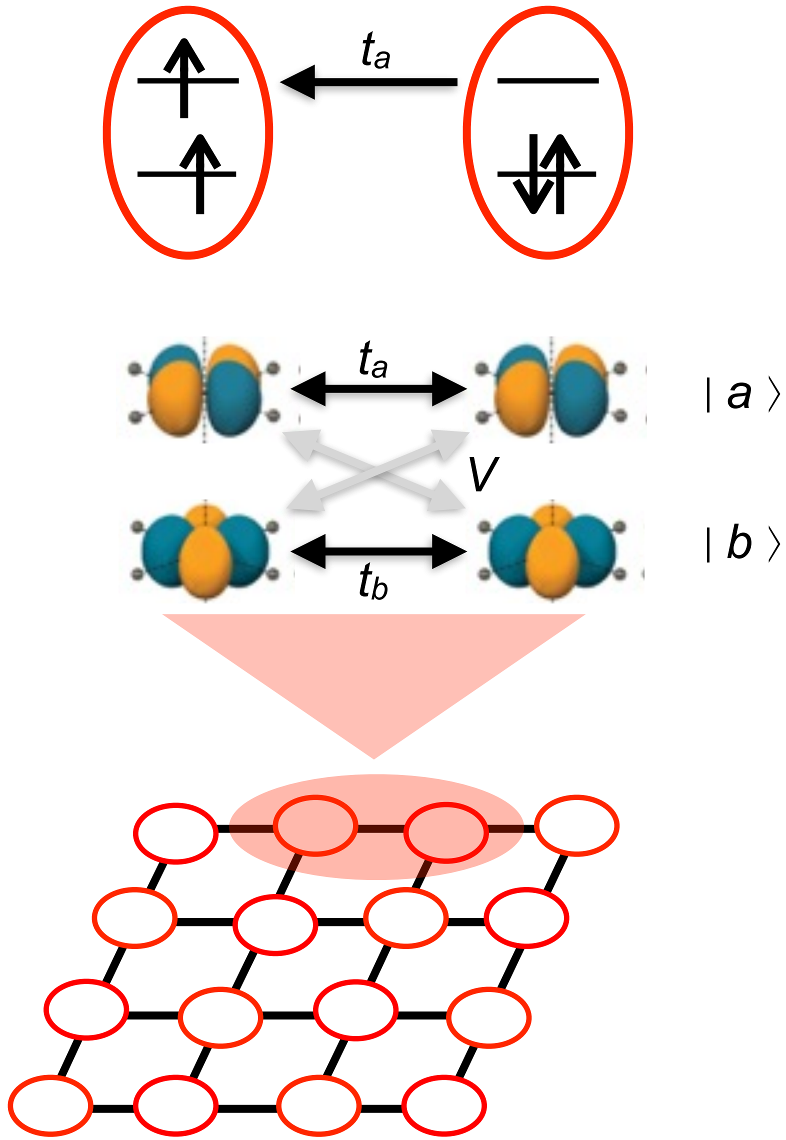
Crystal of 2-orbital atoms



Crystal of 2-orbital atoms

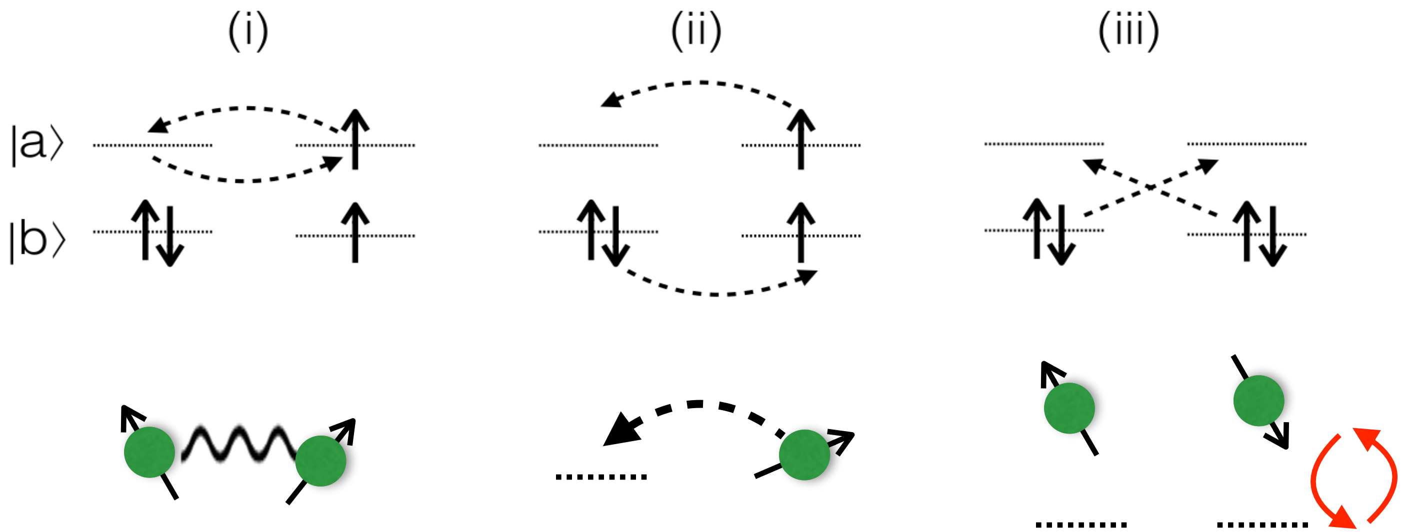


Crystal of 2-orbital atoms

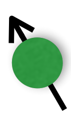


Strong coupling theory

Typical 2nd order hopping processes:

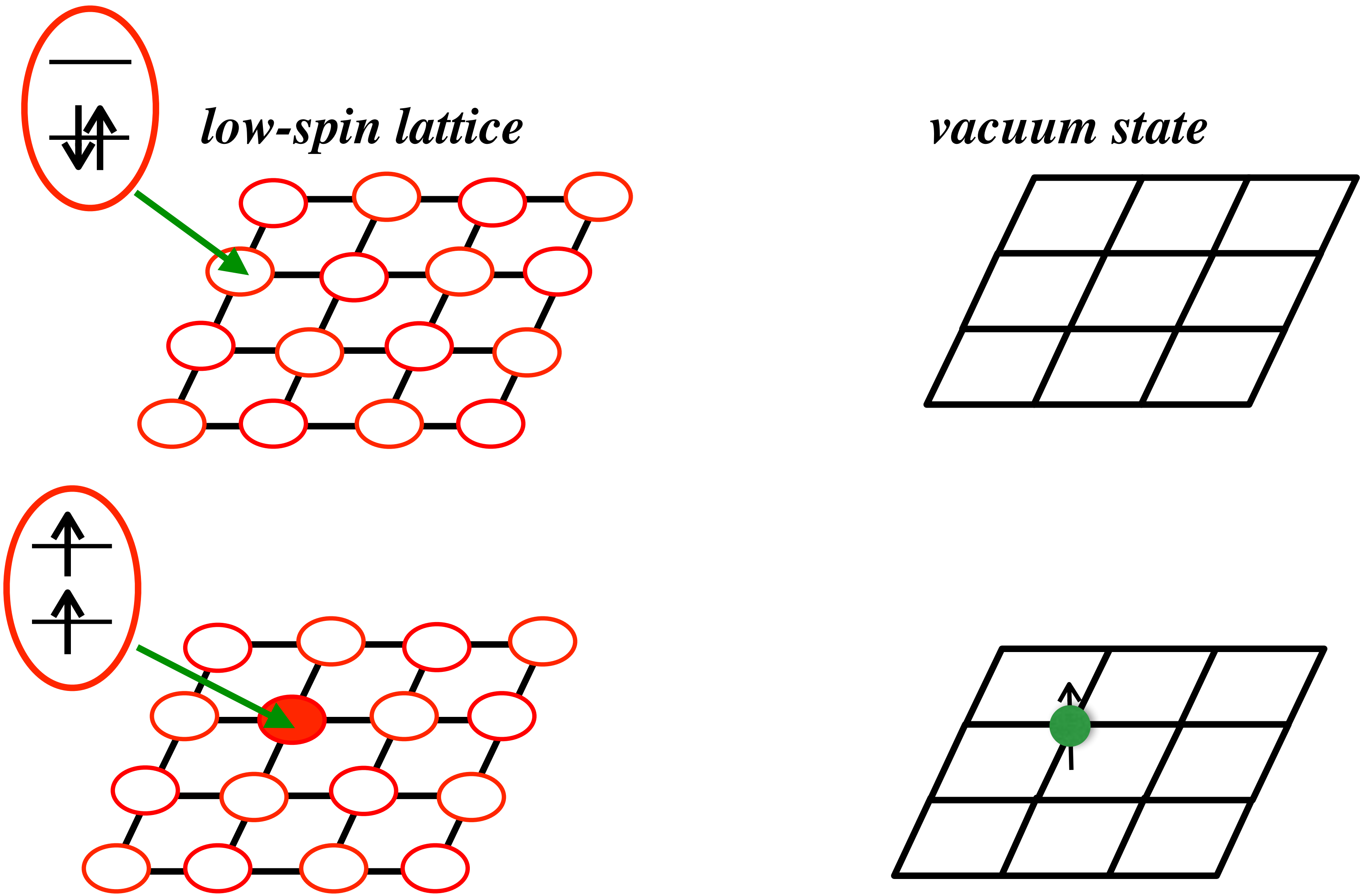


Effective Hamiltonian:

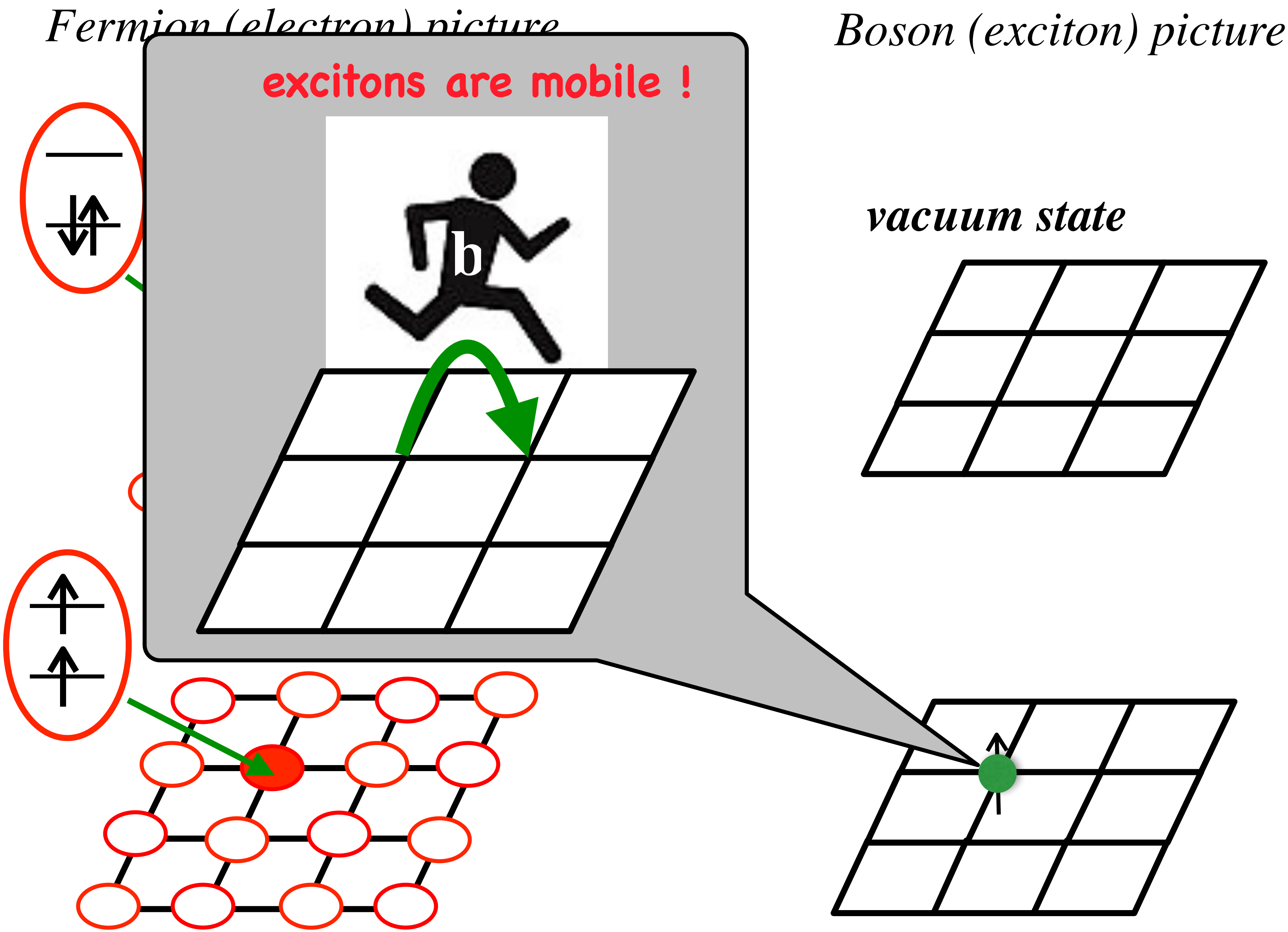
The new  particles are **bosons** and carry **spin S=1**.

Strong coupling theory

Fermion (electron) picture | *Boson (exciton) picture*

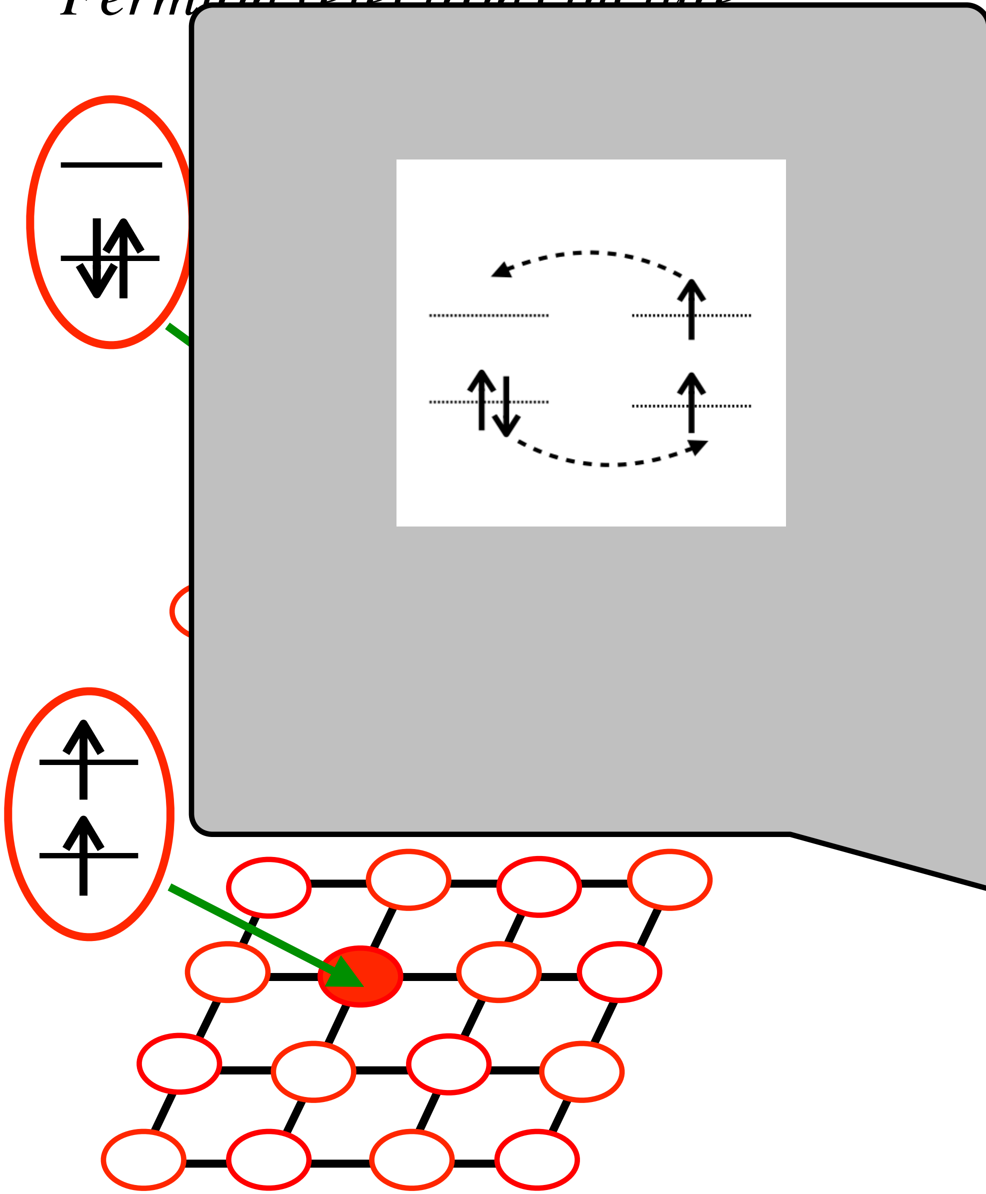


Strong coupling theory



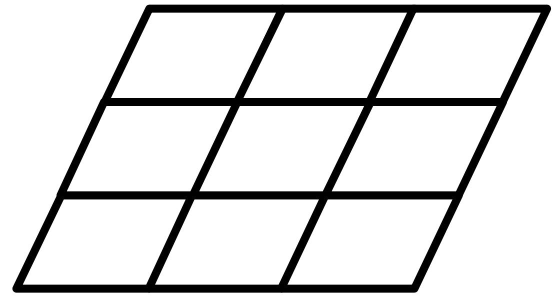
Strong coupling theory

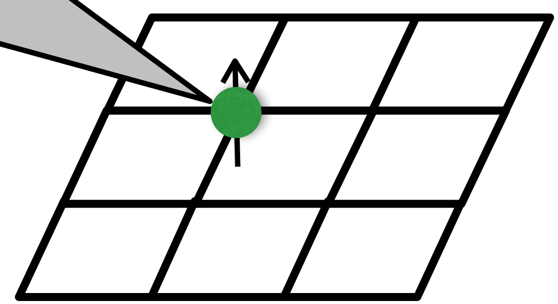
Fermion (electron) picture



Boson (exciton) picture

vacuum state

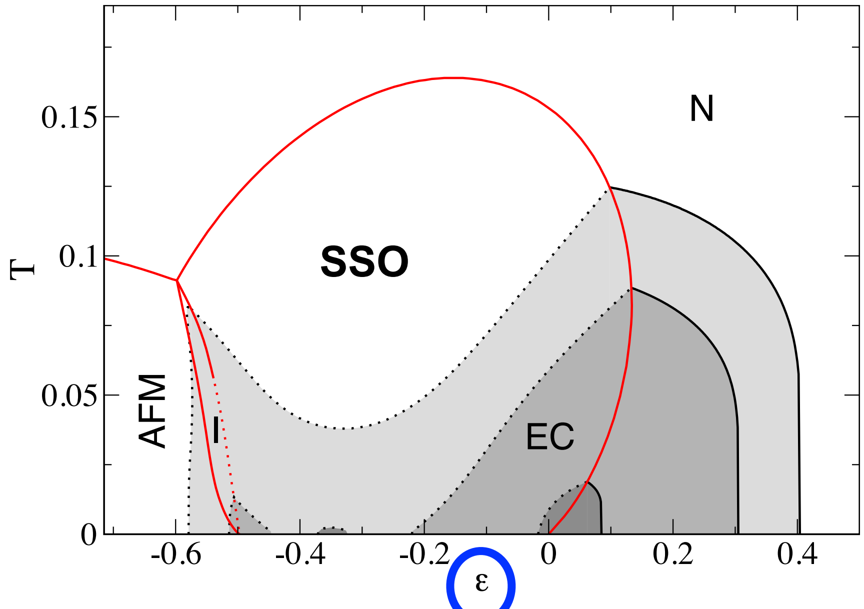




Mean-field theory for excitons

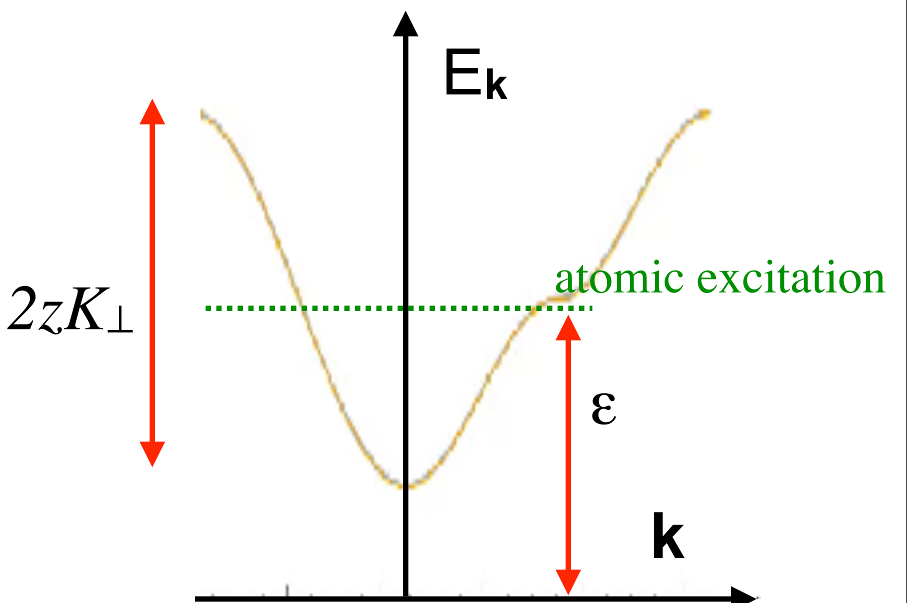
$$H_{\text{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

MF phase diagram:



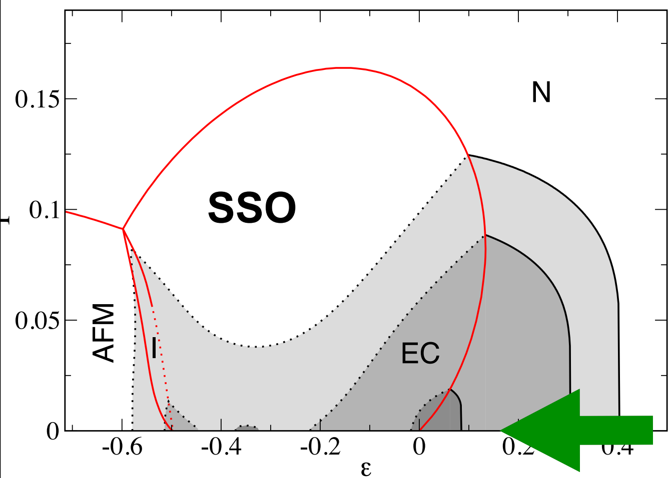
JK, JPCM ***27****, 333201 (2015)*

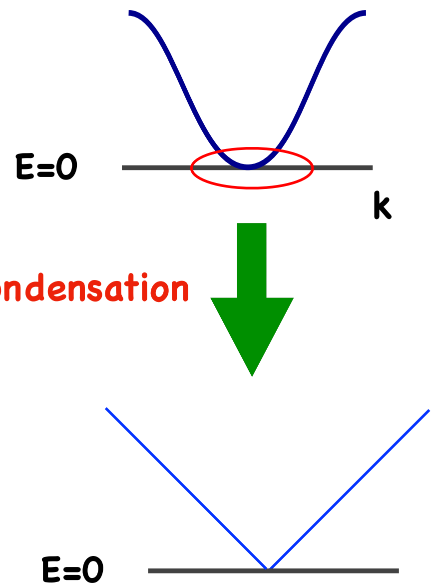
elementary excitations of normal phase



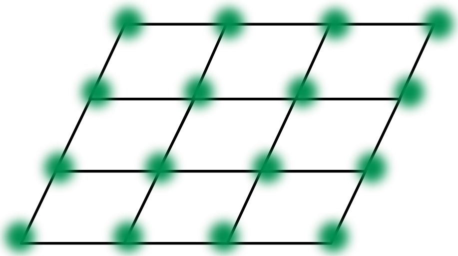
Mean-field theory for excitons

MF phase diagram:

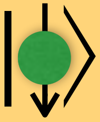




Exciton condensate (superfluid):



Normal state (T>0)

statistical mixture of vacuum $|\cdots\rangle$ and boson  states

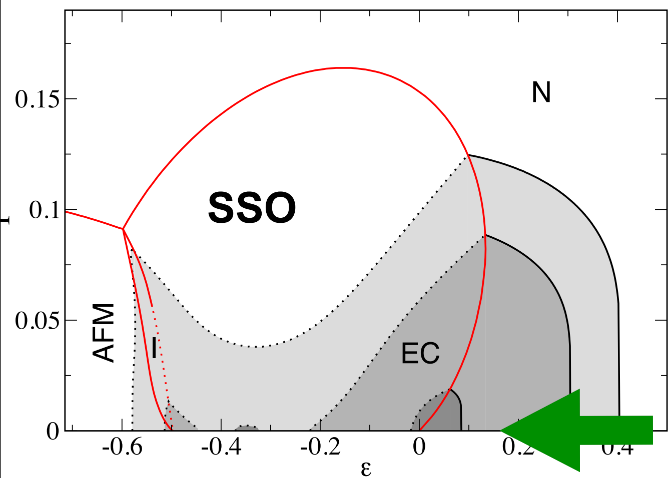
$$\langle n \rangle > 0, \quad \langle d_s \rangle = 0$$

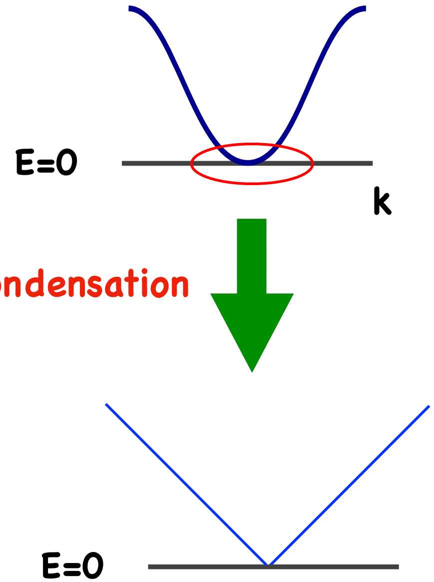
low-spin ground state + thermally populated high-spin states

$$\chi \sim \frac{1}{T}, \quad \langle a^\dagger_\sigma b_{-\sigma} \rangle = 0$$

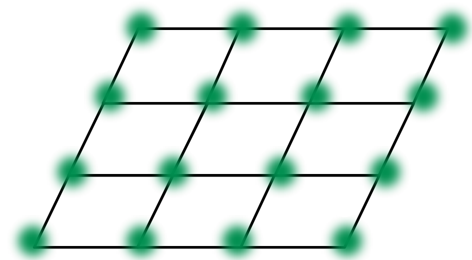
Mean-field theory for excitons

MF phase diagram:

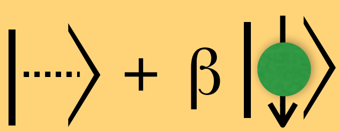




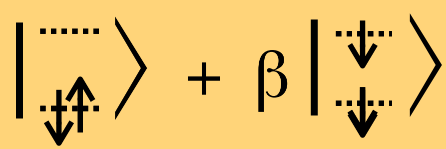
Exciton condensate (superfluid):



Condensate

superposition α  ferromagnetic

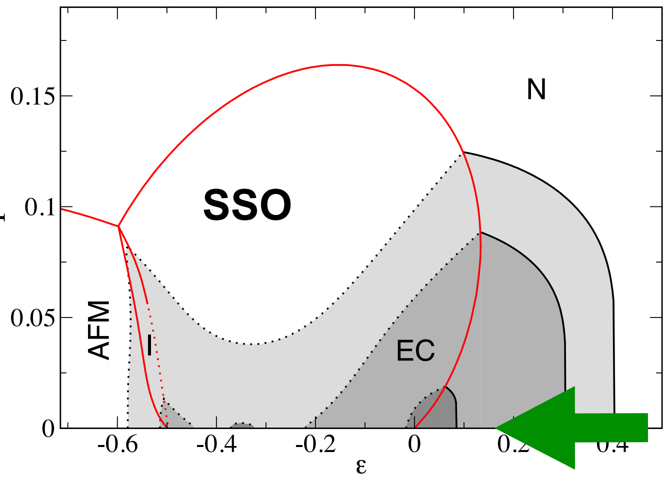
$$\langle n \rangle > 0, \quad \langle d_s \rangle \neq 0$$

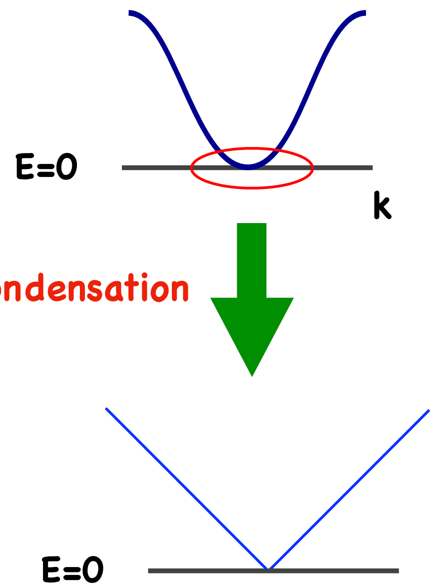
hybridized state low-spin and high-spin states α 

$$\langle a_\sigma^\dagger b_{-\sigma} \rangle \neq 0$$

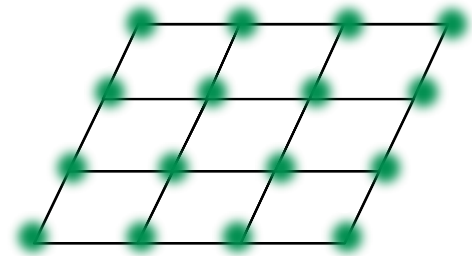
Mean-field theory for excitons

MF phase diagram:





Exciton condensate (superfluid):



Condensate

superposition $\alpha\,|\cdots\rangle + \beta\,|\downarrow\rangle + \beta\,|\uparrow\rangle$ polar

$\langle n\rangle > 0, \quad \langle d_s\rangle \neq 0$

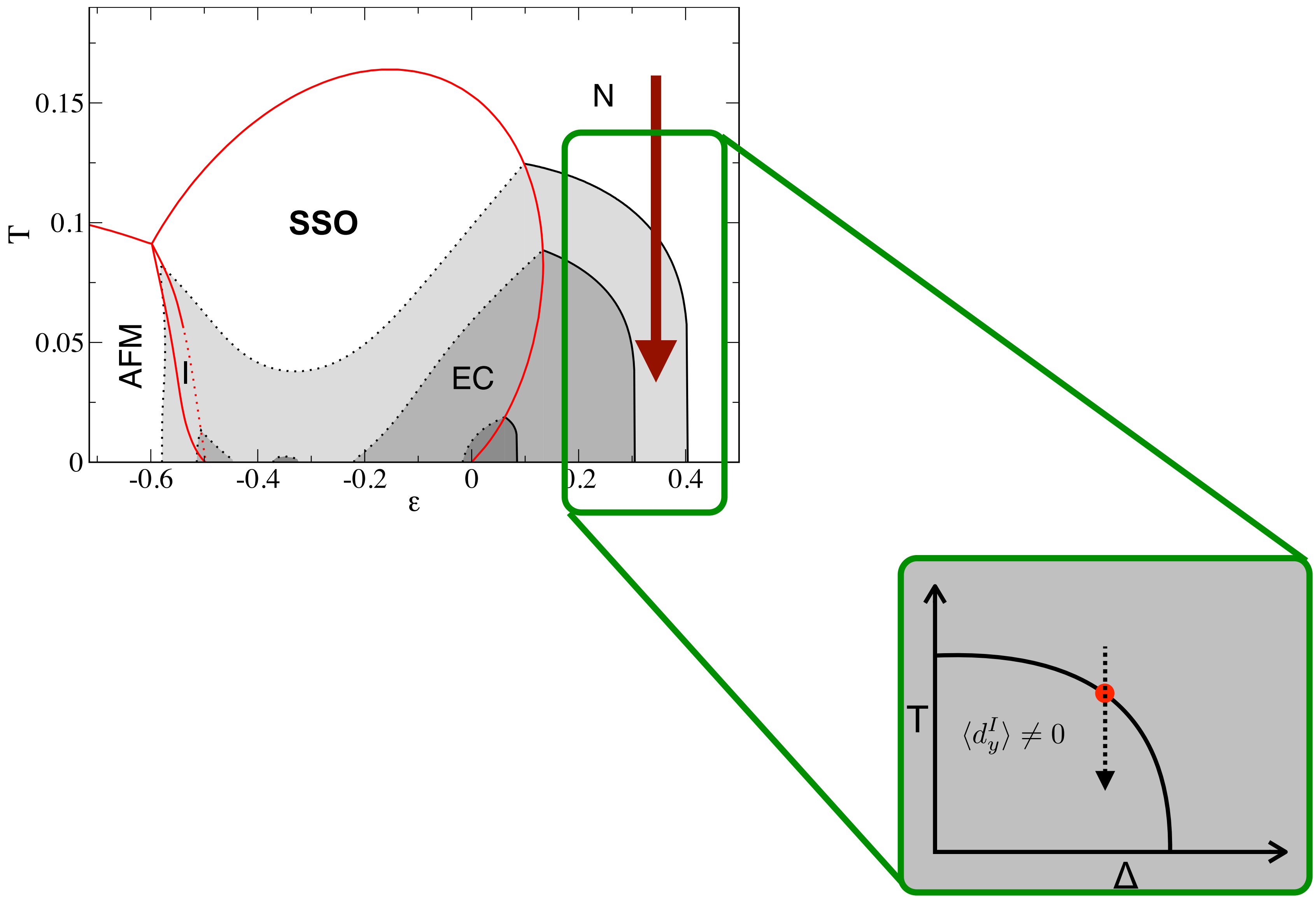
hybridized state low-spin and high-spin states $\alpha\,|\cdots\rangle + \beta\,|\downarrow\downarrow\rangle$

$\langle a^\dagger_\sigma b_{-\sigma}\rangle \neq 0$

Back to fermions
(Dynamical mean-field theory)

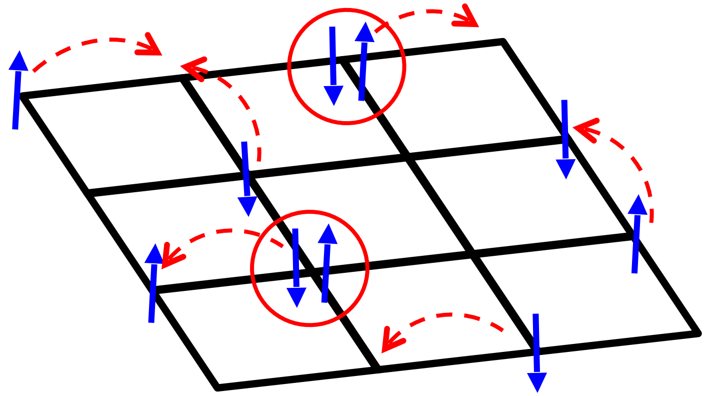
$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$

Numerical results (DMFT) excitonic instability:



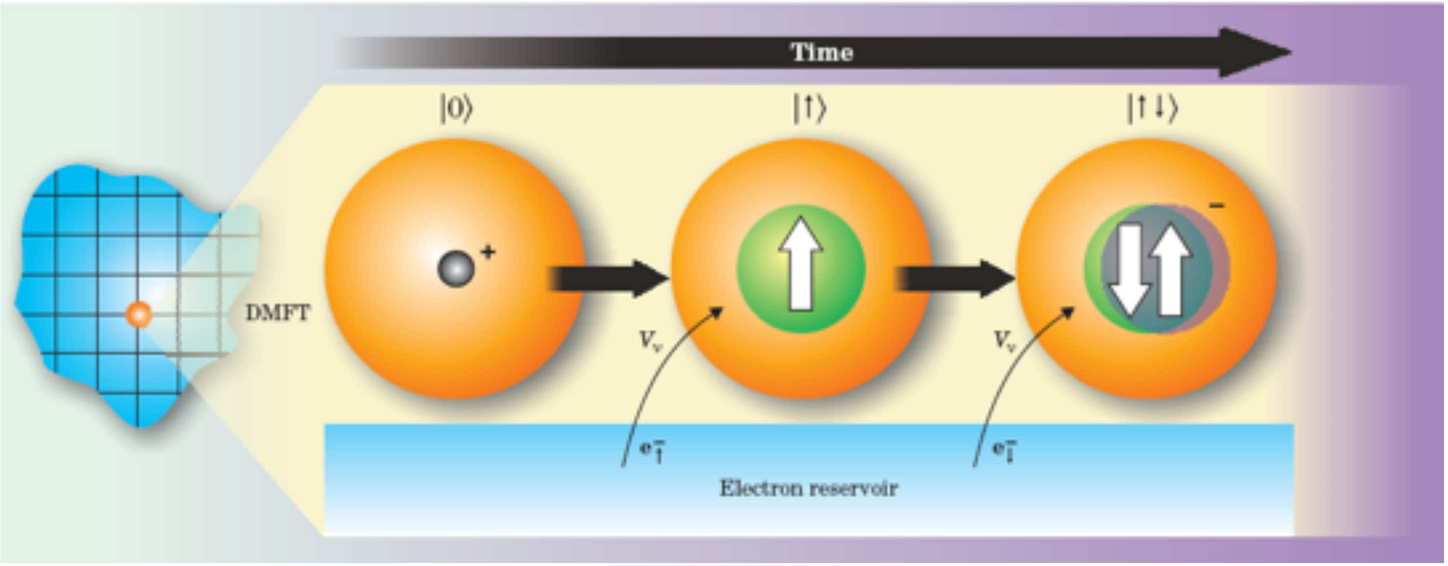
Hubbard model

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



$$
\begin{aligned}
H_{\mathrm{t}} &= \frac{\Delta}{2}\sum_{i,\sigma}(n^a_{i\sigma}-n^b_{i\sigma}) + \sum_{i,j,\sigma}(t_a a^\dagger_{i\sigma}a_{j\sigma} + t_b b^\dagger_{i\sigma}b_{j\sigma}) \\
&\quad + \sum_{\langle ij\rangle,\sigma}(V_1 a^\dagger_{i\sigma}b_{j\sigma} + V_2 b^\dagger_{i\sigma}a_{j\sigma} + c.c.) \\
H^{\mathrm{dd}}_{\mathrm{int}} &= U\sum_i (n^a_{i\uparrow}n^a_{i\downarrow} + n^b_{i\uparrow}n^b_{i\downarrow}) + (U-2J)\sum_{i,\sigma} n^a_{i\sigma}n^b_{i-\sigma} \\
&\quad + (U-3J)\sum_{i\sigma} n^a_{i\sigma}n^b_{i\sigma} \\
H'_{\mathrm{int}} &= J\sum_{i\sigma} a^\dagger_{i\sigma}b^\dagger_{i-\sigma}a_{i-\sigma}b_{i\sigma} + J'\sum_i (a^\dagger_{i\uparrow}a^\dagger_{i\downarrow}b_{i\downarrow}b_{i\uparrow} + c.c.).
\end{aligned}
$$

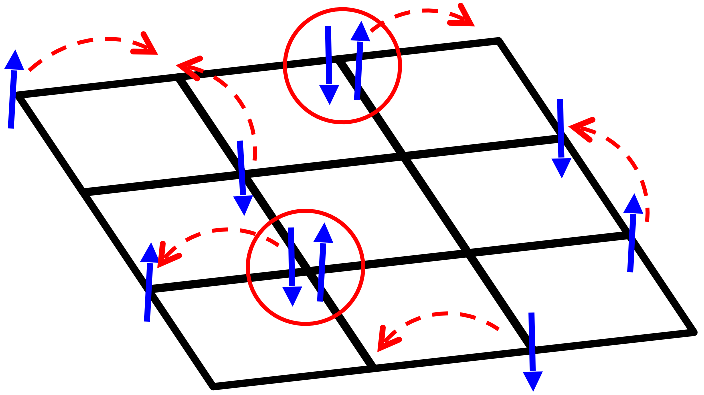
Dynamical mean-field theory



Physics Today (March 2004) Kotliar, Vollhardt

Hubbard model

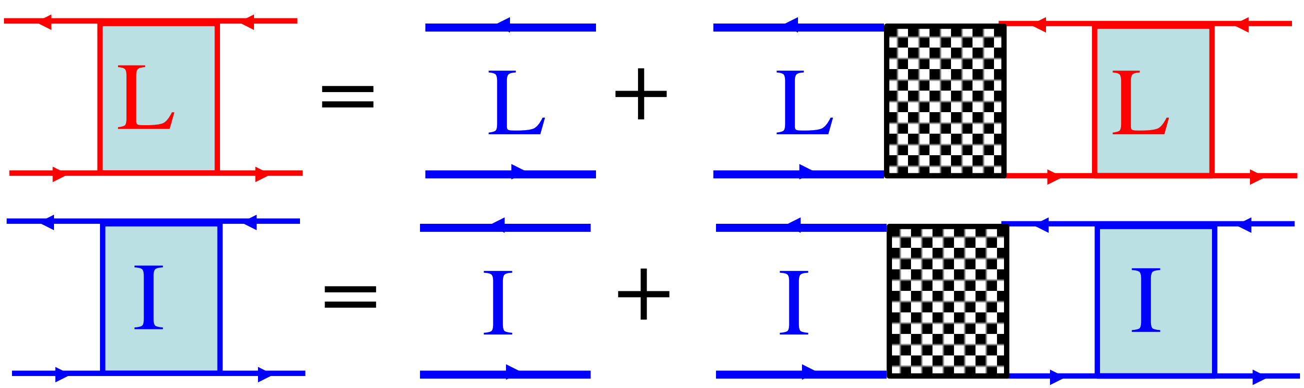
Two-band Hubbard model at n=2 (half filling)



$$
\begin{aligned}
H_{\mathrm{t}} &= \frac{\Delta}{2}\sum_{i,\sigma}(n^a_{i\sigma}-n^b_{i\sigma}) + \sum_{i,j,\sigma}(t_a a^\dagger_{i\sigma}a_{j\sigma}+t_b b^\dagger_{i\sigma}b_{j\sigma}) \\
&\quad + \sum_{\langle ij\rangle,\sigma}(V_1 a^\dagger_{i\sigma}b_{j\sigma}+V_2 b^\dagger_{i\sigma}a_{j\sigma}+c.c.) \\
H^{\mathrm{dd}}_{\mathrm{int}} &= U\sum_i (n^a_{i\uparrow}n^a_{i\downarrow}+n^b_{i\uparrow}n^b_{i\downarrow}) + (U-2J)\sum_{i,\sigma} n^a_{i\sigma}n^b_{i-\sigma} \\
&\quad + (U-3J)\sum_{i\sigma} n^a_{i\sigma}n^b_{i\sigma} \\
H'_{\mathrm{int}} &= J\sum_{i\sigma} a^\dagger_{i\sigma}b^\dagger_{i-\sigma}a_{i-\sigma}b_{i\sigma} + J'\sum_i (a^\dagger_{i\uparrow}a^\dagger_{i\downarrow}b_{i\downarrow}b_{i\uparrow}+c.c.).
\end{aligned}
$$

Bethe-Salpeter equation

- full (ω,ν,ν') frequency structure
- local ph-irreducible vertex
- multi-orbital; no symmetry constraints
- Maxent for analytic continuation



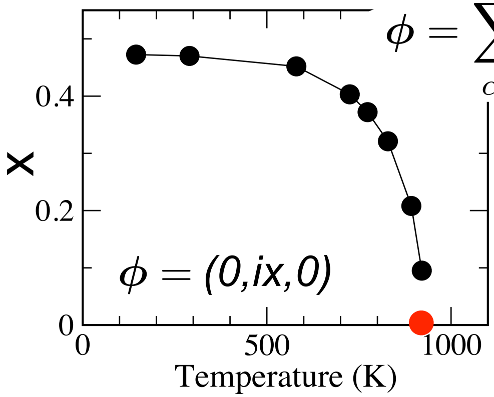
lattice model

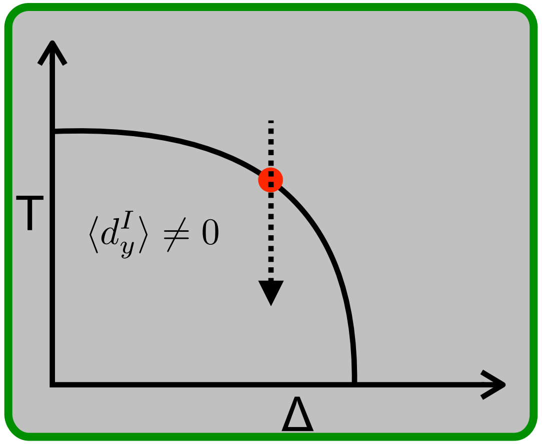
impurity model

Polar excitonic condensate (half filling)

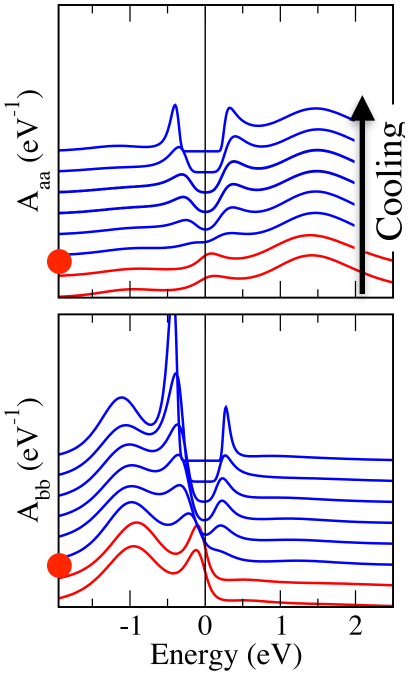
Order parameter

$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^{\dagger}_{\alpha} b_{\beta} \rangle$$

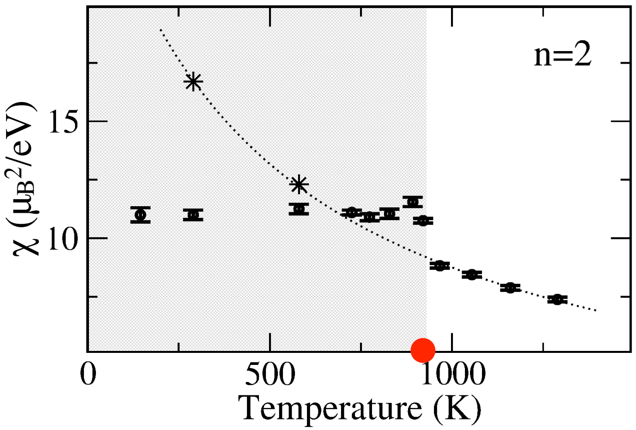




Spectral density



Uniform spin susceptibility



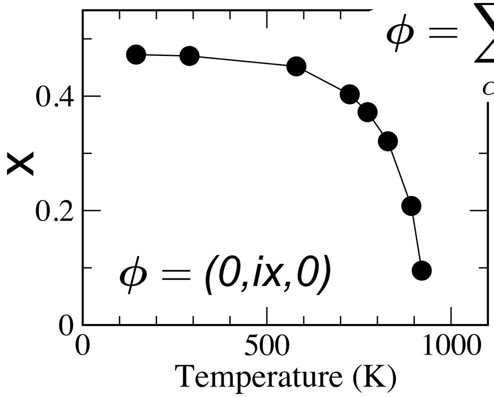
Model parameters (eV):
U=4, J=1, Δ_{CF}=3.40
t_a=0.4118, t_b=-0.1882
square lattice

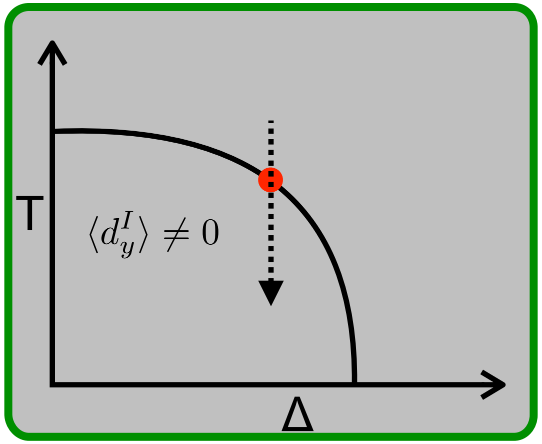
*JK and P. Augustinský, PRB **90**, 235112 (2014)*

Polar excitonic condensate (half filling)

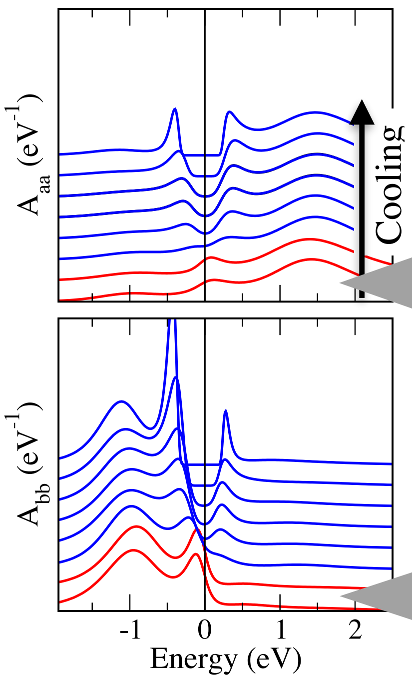
Order parameter

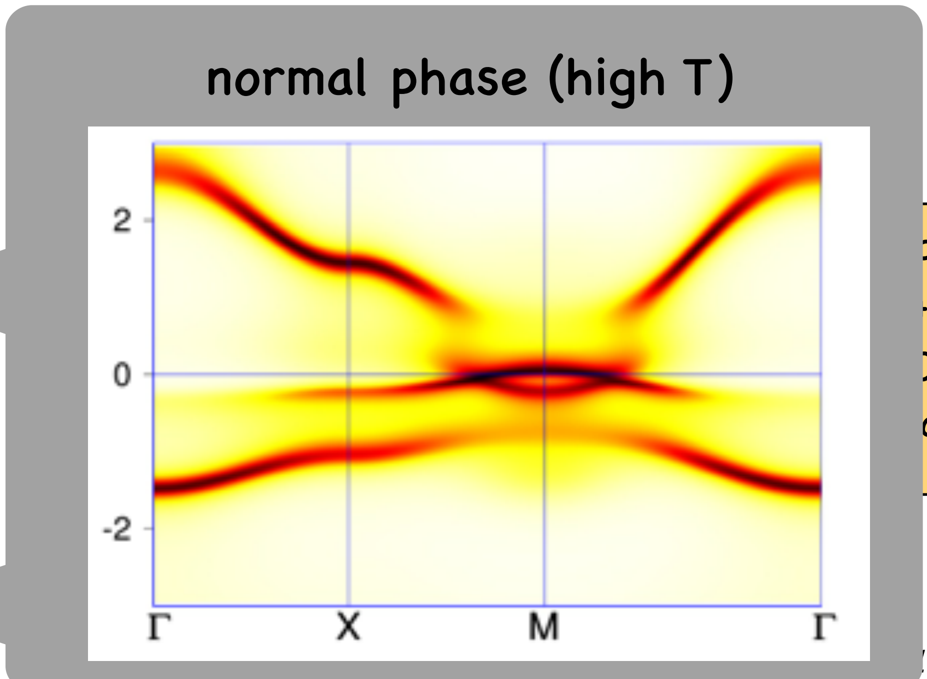
$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$





Spectral density





del parameters (eV):

, J=1, Δ_{CF}=3.40

.4118, t_b=-0.1882

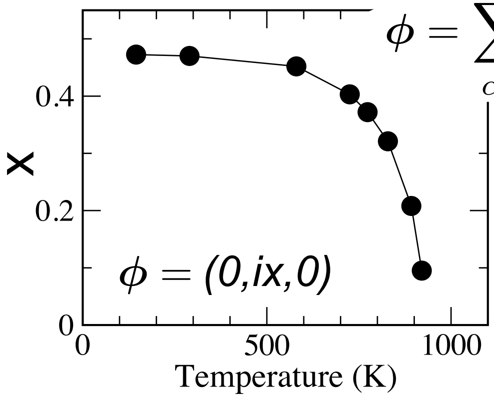
are lattice

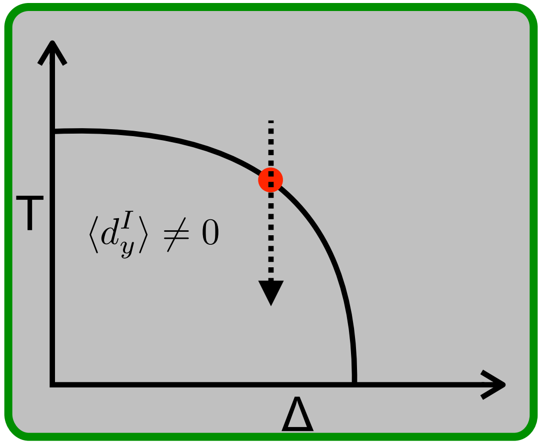
)

Polar excitonic condensate (half filling)

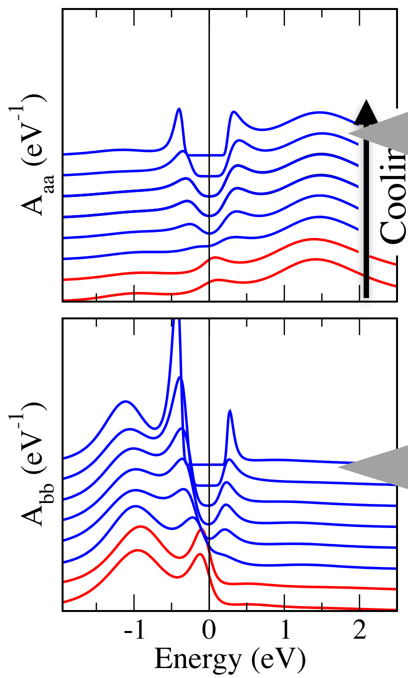
Order parameter

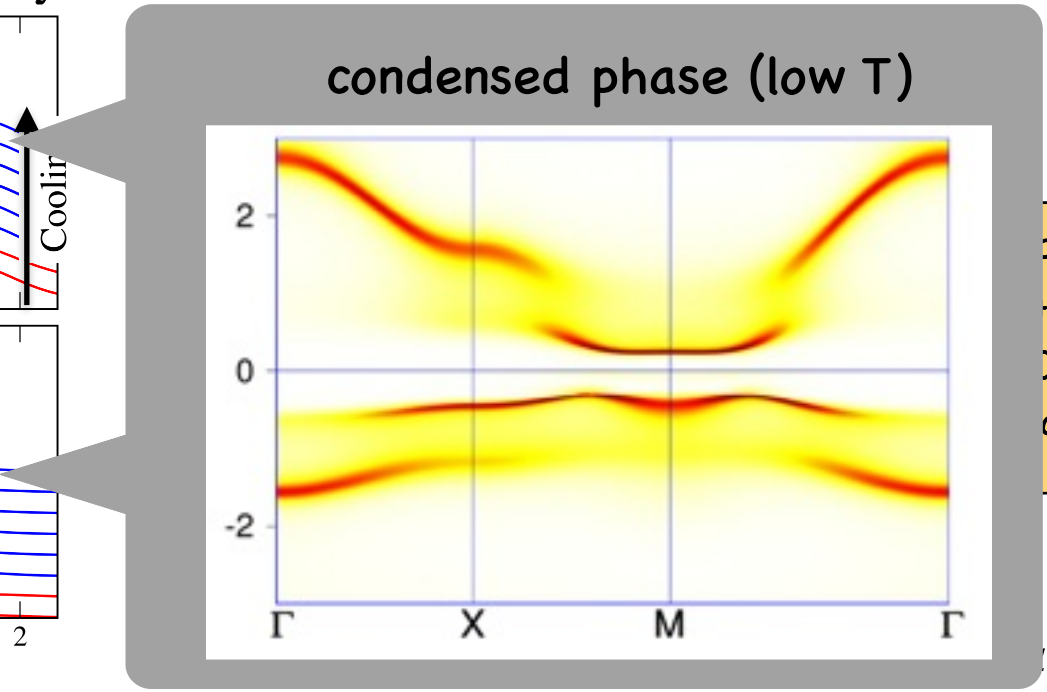
$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$





Spectral density

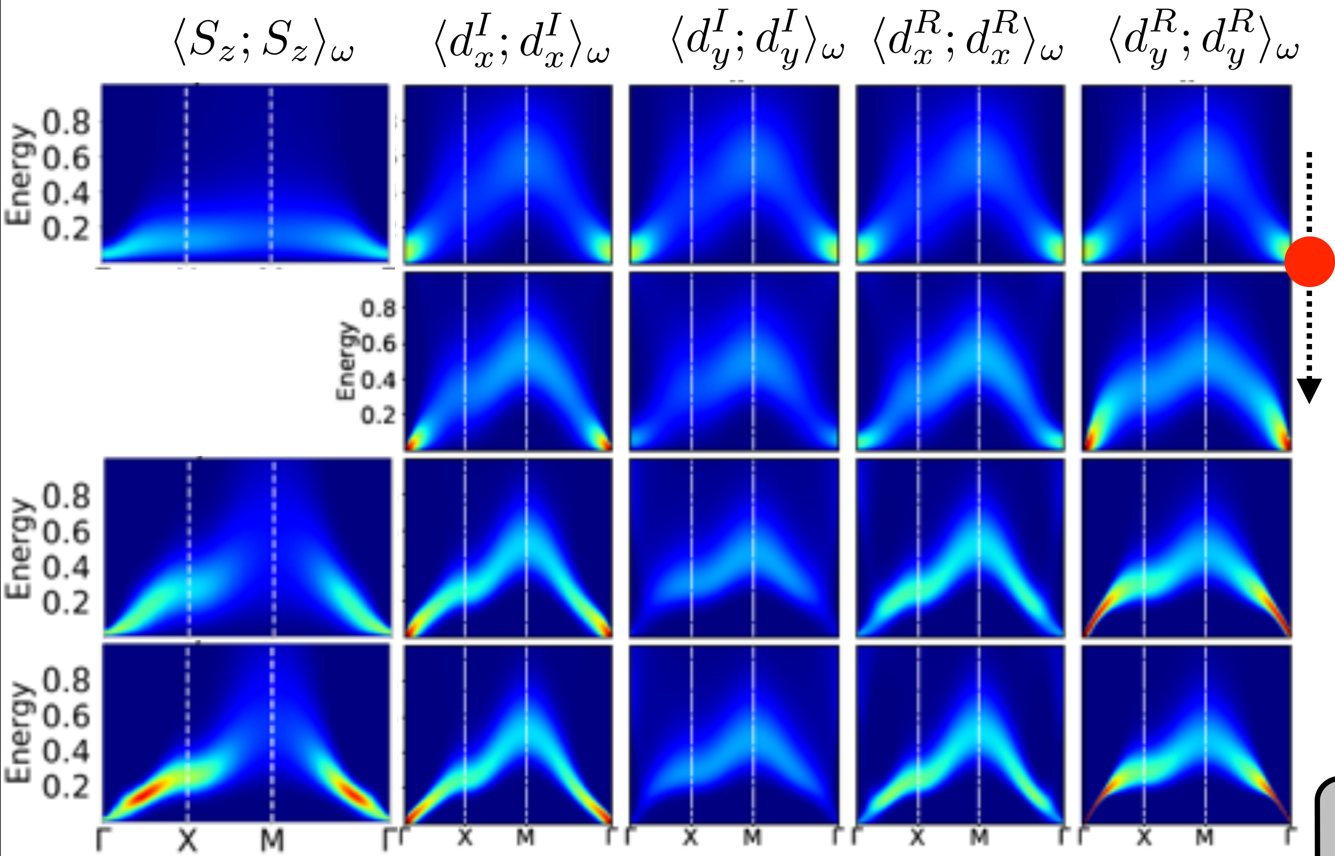


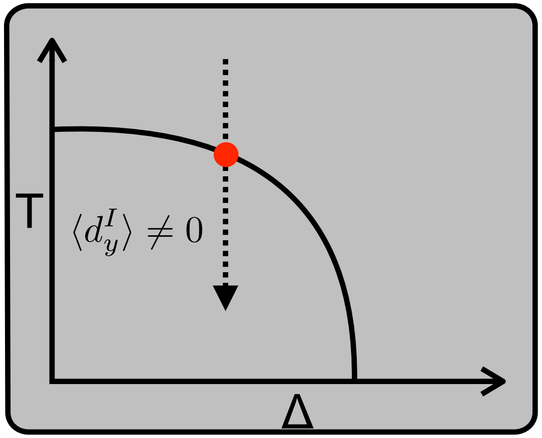


del parameters (eV):
-, J=1, Δ_{CF}=3.40
.4118, t_b=-0.1882
are lattice

)

Dynamical susceptibility





D. Geffroy et al., PRL ***122****, 127601 (2019)*

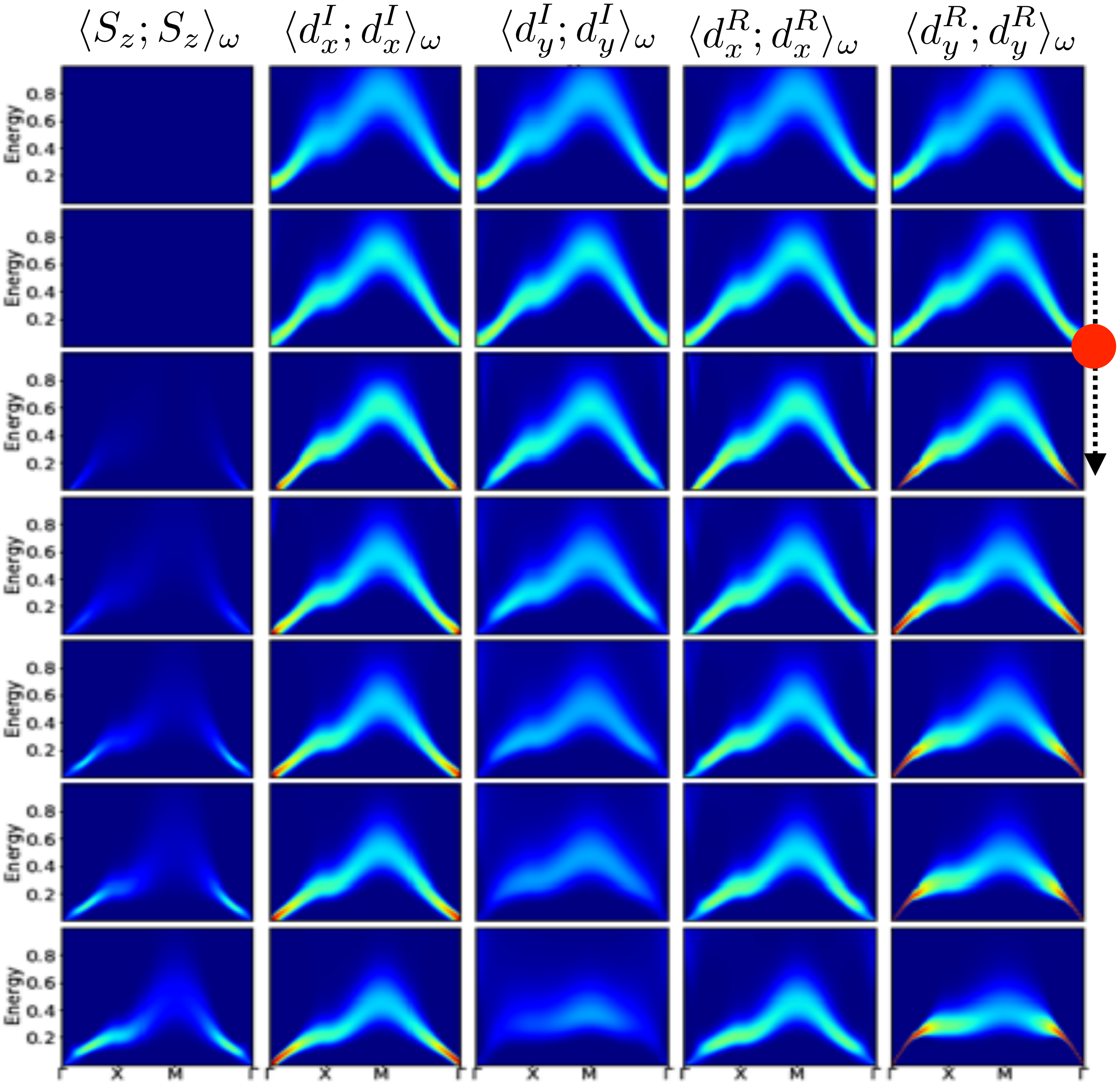
Formal defs:

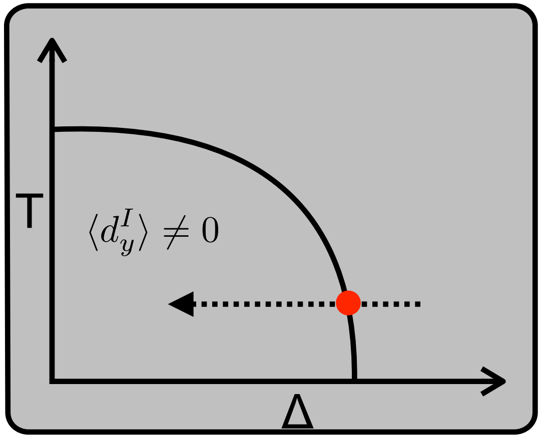
$$d_{i\eta}^R = \sum_{\alpha\beta} \sigma_{\alpha\beta}^\eta (a_{i\alpha}^\dagger b_{i\beta} + b_{i\alpha}^\dagger a_{i\beta})$$

$$d_{i\eta}^I = i \sum_{\alpha\beta} \sigma_{\alpha\beta}^\eta (a_{i\alpha}^\dagger b_{i\beta} - b_{i\alpha}^\dagger a_{i\beta})$$

$$S_i^z = \sum_{\alpha\beta} \sigma_{\alpha\beta}^z (a_{i\alpha}^\dagger a_{i\beta} + b_{i\alpha}^\dagger b_{i\beta})$$

Dynamical susceptibility





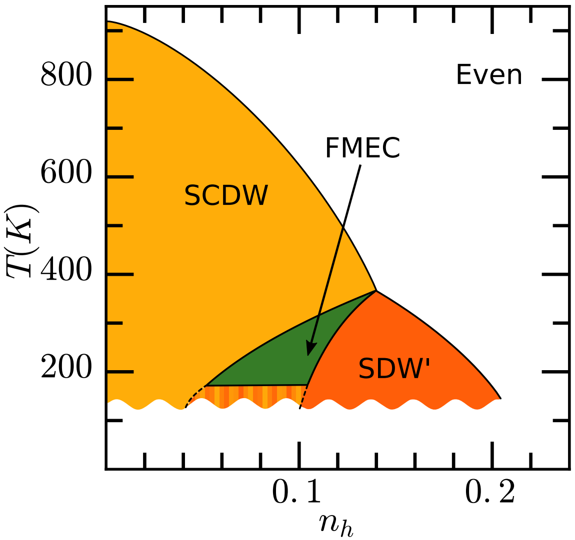
Formal defs:

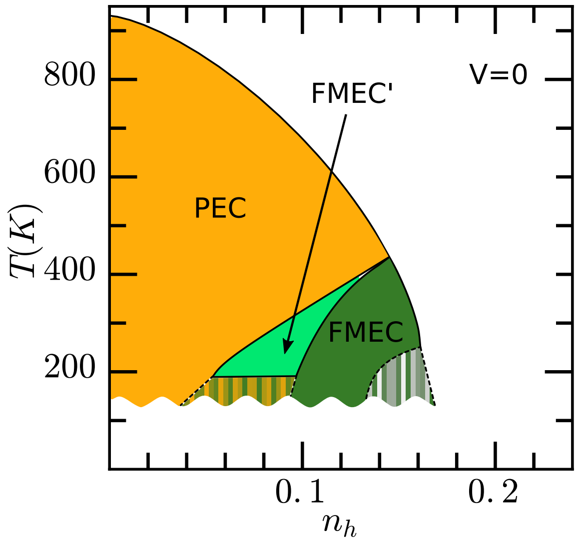
$$d^R_{i\eta} = \sum_{\alpha\beta} \sigma^\eta_{\alpha\beta}(a^\dagger_{i\alpha} b_{i\beta} + b^\dagger_{i\alpha} a_{i\beta})$$

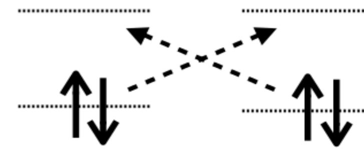
$$d^I_{i\eta} = i\sum_{\alpha\beta} \sigma^\eta_{\alpha\beta}(a^\dagger_{i\alpha} b_{i\beta} - b^\dagger_{i\alpha} a_{i\beta})$$

$$S^z_i = \sum_{\alpha\beta} \sigma^z_{\alpha\beta}(a^\dagger_{i\alpha} a_{i\beta} + b^\dagger_{i\alpha} b_{i\beta})$$

Doping + cross-hopping





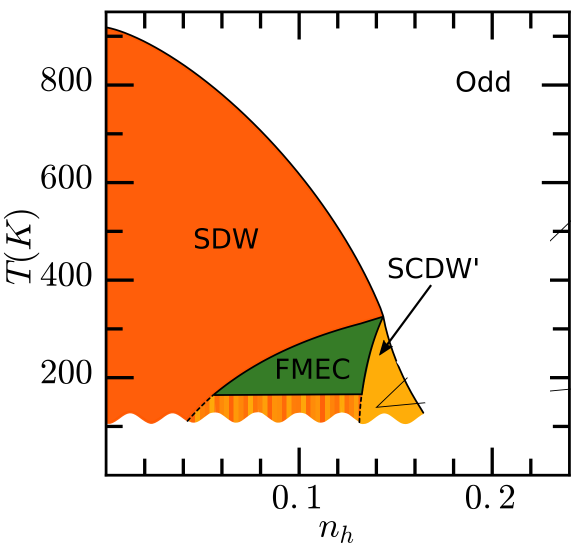


$V_1=V_2$ even cross-hopping

$V_1=-V_2$ odd cross-hopping

$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^{\dagger}_{\alpha} b_{\beta} \rangle$$

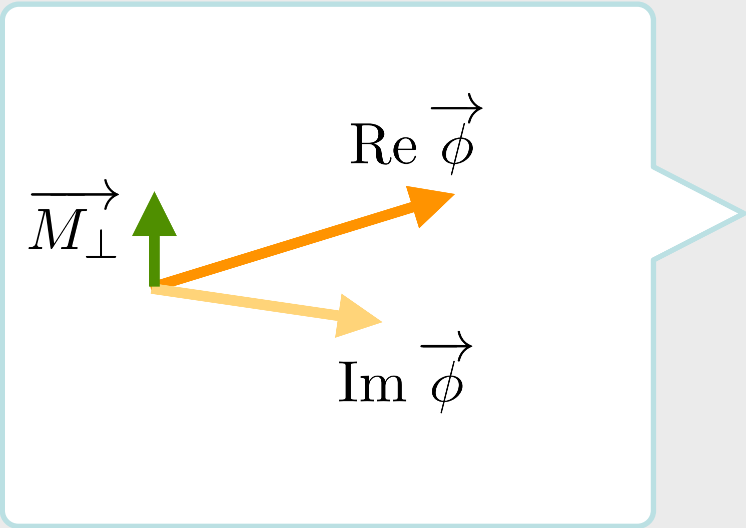
$$\text{PEC} \begin{cases} \text{SDW} & \phi = \mathbf{x} \\ \text{SCDW} & \phi = i\mathbf{x} \end{cases}$$



Condensate state	$M_\perp$	$M_\parallel$	$\mathbf{m}(\mathbf{r})$	$\mathbf{m}_\mathbf{k}$	Re ϕ	Im ϕ
FMEC	✓	✓, 0	✓	✓	✓	✓
SDW	0	0	✓	0	✓	0
SCDW	0	0	0	0	0	✓
SDW’	0	✓, 0	✓	✓	✓	0
SCDW’	0	0	0	✓	0	✓

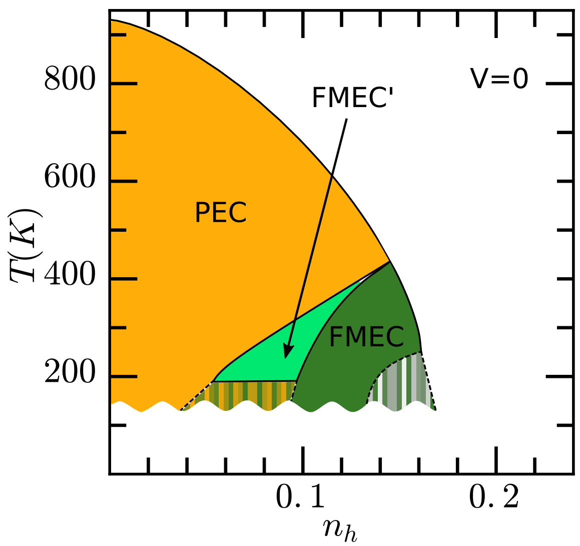
$$\mathbf{m}_\mathbf{k} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^{\dagger}_{\mathbf{k}\alpha} a_{\mathbf{k}\beta} + b^{\dagger}_{\mathbf{k}\alpha} b_{\mathbf{k}\beta} \rangle$$

*JK and D. Geffroy, PRL **116**, 256403 (2016)*



$$\boldsymbol{\phi} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$

Doping

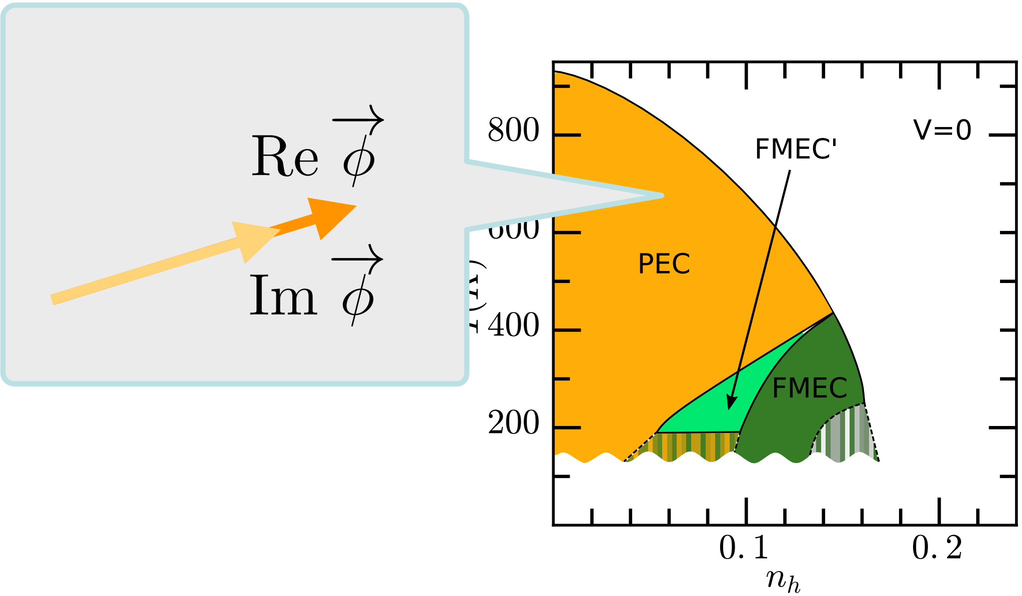


$$H_{\text{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$K_0 > 0$ PEC: $\psi = \prod_i (\alpha + \beta d_{ix}^\dagger)|0\rangle$

$K_0 < 0$ FMEC: $\psi = \prod_i (\alpha + \beta d_{i1}^\dagger)|0\rangle$

Doping



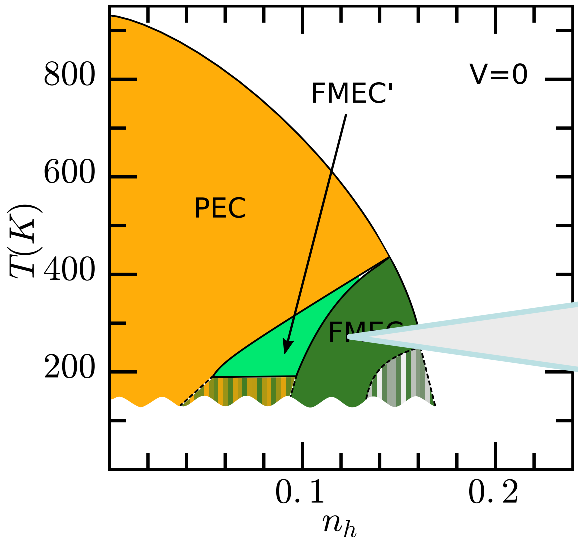
$$H_{\mathrm{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

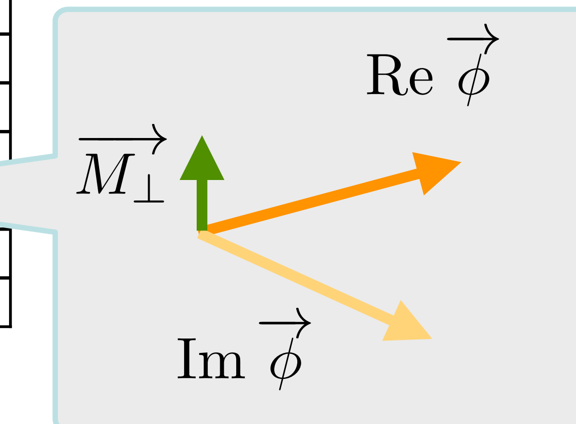
$K_0 > 0$ PEC: $\psi = \prod_i (\alpha + \beta d_{ix}^\dagger)|0\rangle$

$K_0 < 0$ FMEC: $\psi = \prod_i (\alpha + \beta d_{i1}^\dagger)|0\rangle$

Doping

$$\mathbf{S}_i = i(\mathbf{d}_i^\dagger \times \mathbf{d}_i)$$





$$H_{\text{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$K_0 > 0$ PEC: $\psi = \prod_i (\alpha + \beta d_{ix}^\dagger)|0\rangle$

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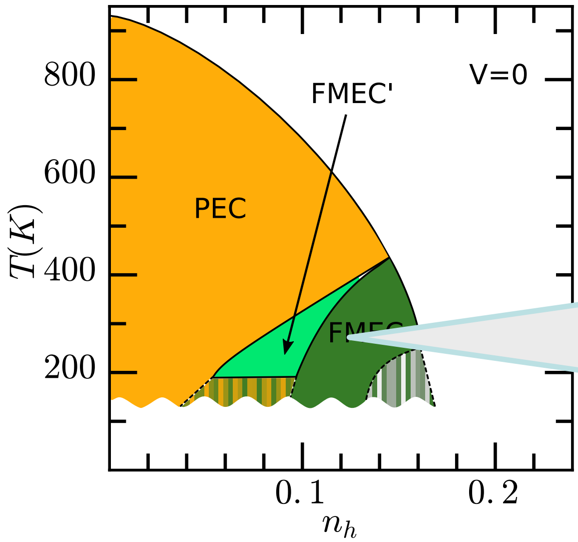
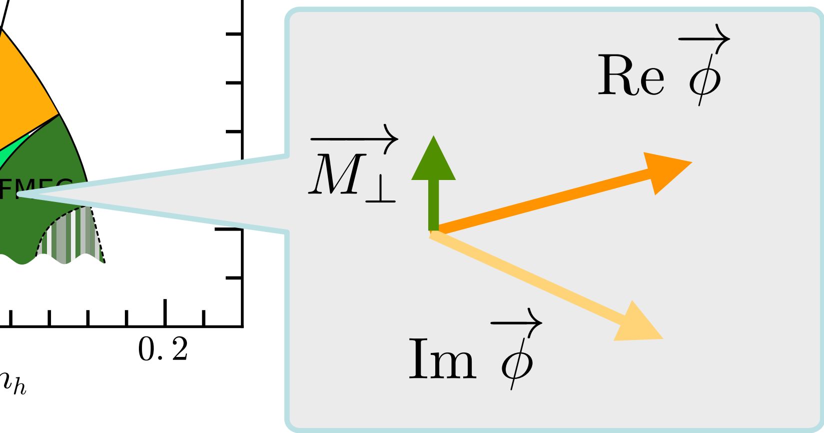
Doping



$$S_z = d_1^\dagger d_1 - d_{-1}^\dagger d_{-1}$$
$$= i(d_x^\dagger d_y - d_y^\dagger d_x)$$

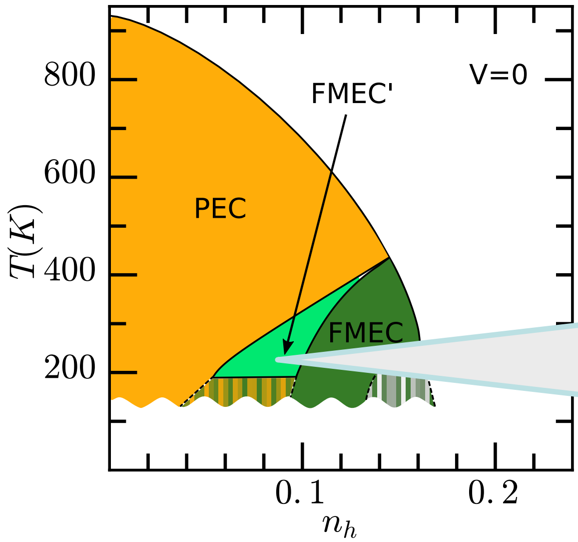


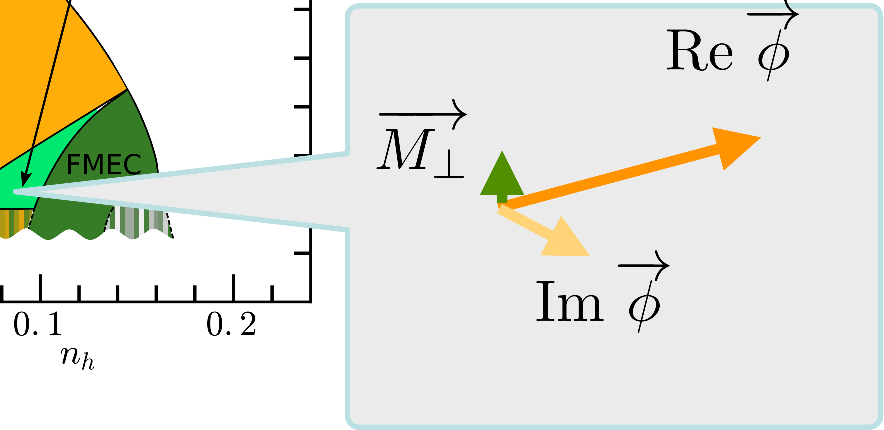
$$H_{\text{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$K_0 > 0$ PEC: $\psi = \prod_i (\alpha + \beta d_{ix}^\dagger)|0\rangle$

$K_0 < 0$ FMEC: $\psi = \prod_i (\alpha + \beta d_{i1}^\dagger)|0\rangle$

Doping



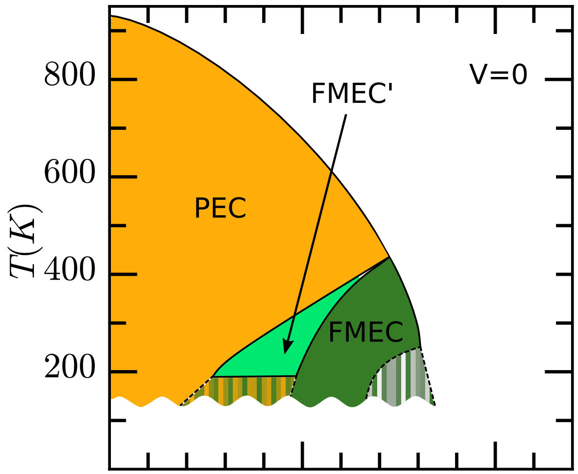


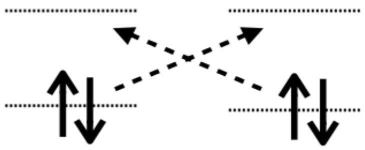
$$H_{\text{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

$K_0 > 0$ PEC: $\psi = \prod_i (\alpha + \beta d_{ix}^\dagger)|0\rangle$

$K_0 < 0$ FMEC: $\psi = \prod_i (\alpha + \beta d_{i1}^\dagger)|0\rangle$

Cross-hopping

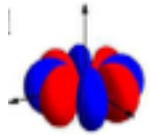




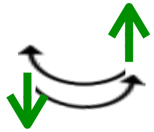
V_1=V_2 even cross-hopping
V_1=-V_2 odd cross-hopping

$$H_{\text{eff}} = \varepsilon \sum_i n_i + K_\perp \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j + H.c. \right) + K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$
$$+ K_1 \sum_{\langle ij \rangle} \left(\mathbf{d}_i^\dagger \cdot \mathbf{d}_j^\dagger + \mathbf{d}_i \cdot \mathbf{d}_j \right)$$

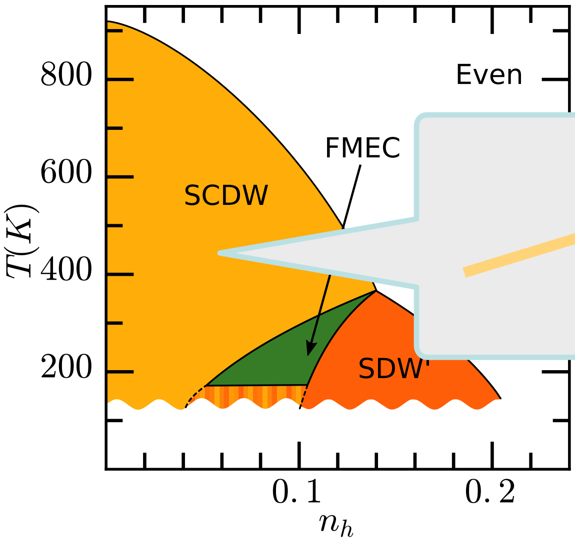
$K_\perp K_1 > 0$ SDW: $\psi = \prod_i (\alpha + \beta d_{ix}^\dagger)|0\rangle$

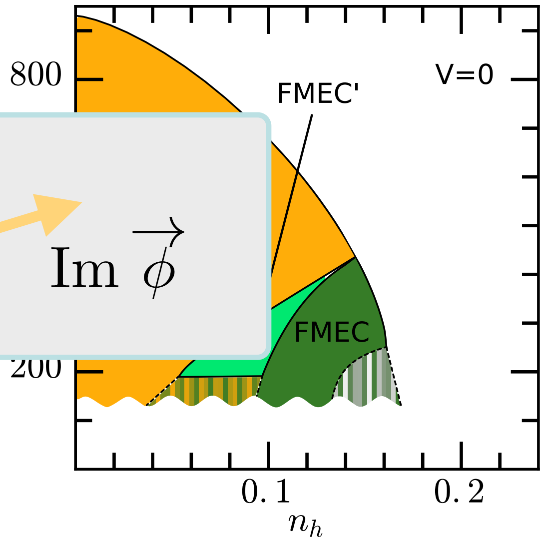


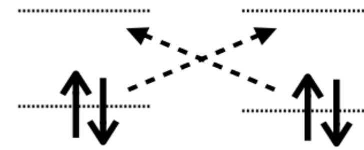
$K_\perp K_1 < 0$ SCDW: $\psi = \prod_i (\alpha + i\beta d_{ix}^\dagger)|0\rangle$



Doping + cross-hopping



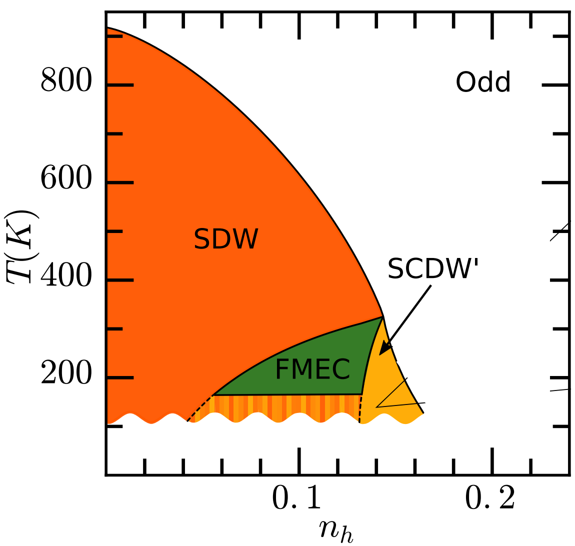




V$_1$=V$_2$ even cross-hopping
V$_1$=-V$_2$ odd cross-hopping

$$\boldsymbol{\phi} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$

PEC $\begin{cases} \text{SDW} & \boldsymbol{\phi} = \mathbf{x} \\ \text{SCDW} & \boldsymbol{\phi} = i\mathbf{x} \end{cases}$

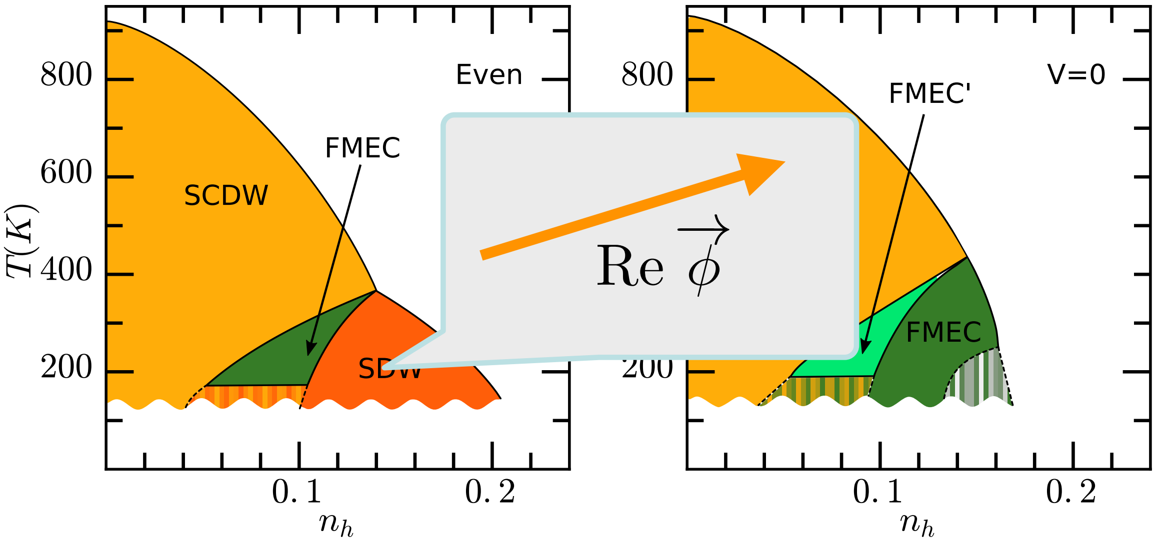


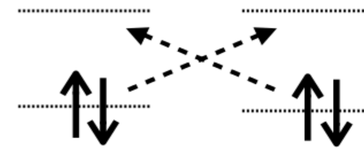
Condensate state	$M_\perp$	$M_\parallel$	$\mathbf{m}(\mathbf{r})$	$\mathbf{m}_\mathbf{k}$	Re $\boldsymbol{\phi}$	Im $\boldsymbol{\phi}$
FMEC	✓	✓, 0	✓	✓	✓	✓
SDW	0	0	✓	0	✓	0
SCDW	0	0	0	0	0	✓
SDW’	0	✓, 0	✓	✓	✓	0
SCDW’	0	0	0	✓	0	✓

$$\mathbf{m}_\mathbf{k} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_{\mathbf{k}\alpha} a_{\mathbf{k}\beta} + b^\dagger_{\mathbf{k}\alpha} b_{\mathbf{k}\beta} \rangle$$

*JK and D. Geffroy, PRL **116**, 256403 (2016)*

Doping + cross-hopping



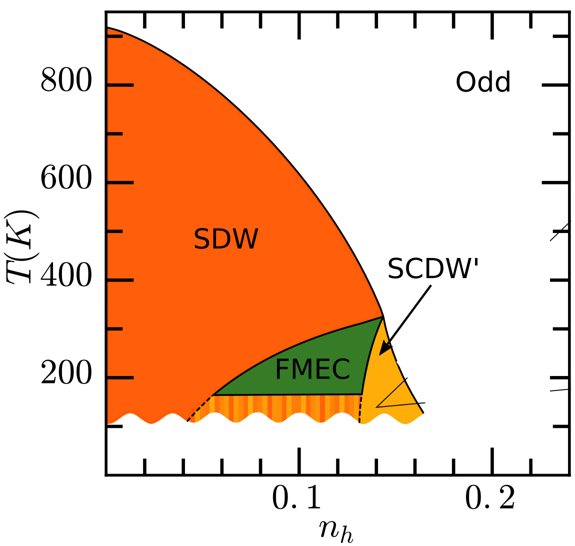


V₁=V₂ even cross-hopping

V₁=-V₂ odd cross-hopping

$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$

$$\text{PEC} \begin{cases} \text{SDW} & \phi = \mathbf{x} \\ \text{SCDW} & \phi = i\mathbf{x} \end{cases}$$

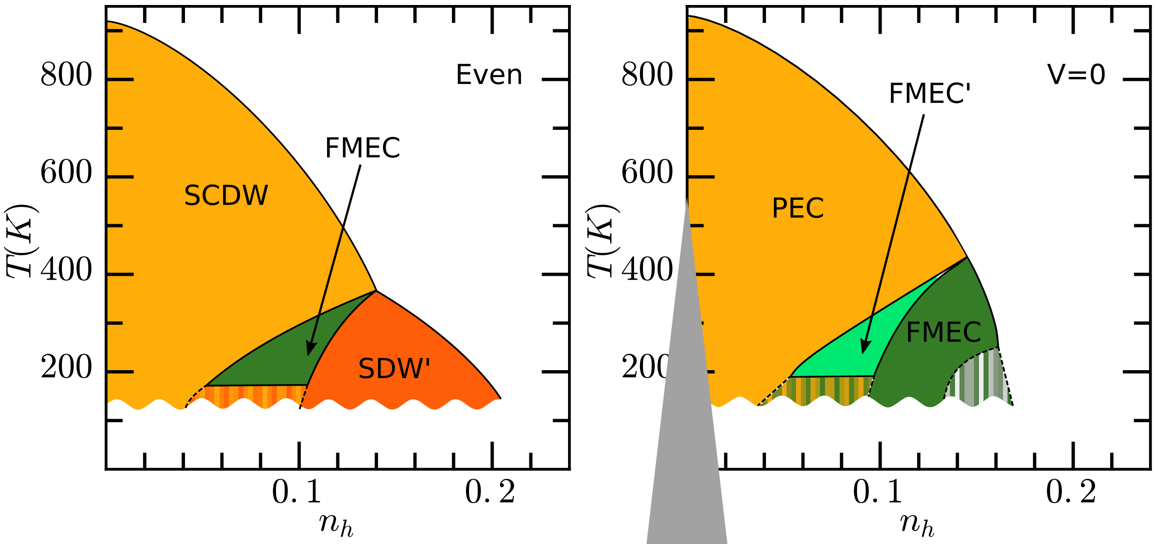


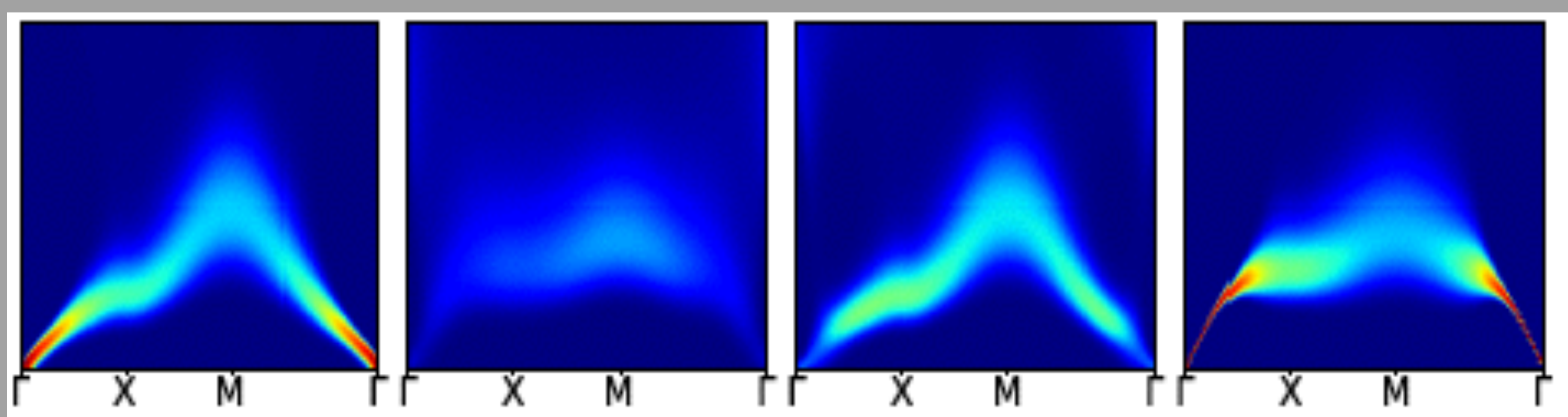
Condensate state	$M_\perp$	$M_\parallel$	$\mathbf{m}(\mathbf{r})$	$\mathbf{m}_\mathbf{k}$	Re ϕ	Im ϕ
FMEC	✓	✓, 0	✓	✓	✓	✓
SDW	0	0	✓	0	✓	0
SCDW	0	0	0	0	0	✓
SDW'	0	|✓, 0|	✓	✓	✓	0
SCDW'	0	0	0	✓	0	✓

$$\mathbf{m}_\mathbf{k} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_{\mathbf{k}\alpha} a_{\mathbf{k}\beta} + b^\dagger_{\mathbf{k}\alpha} b_{\mathbf{k}\beta} \rangle$$

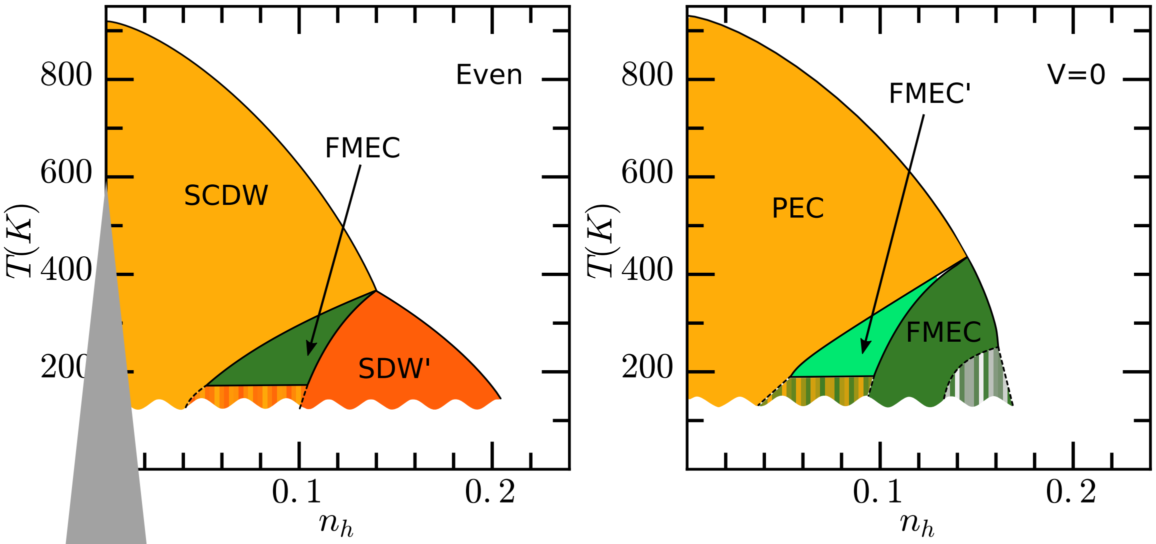
JK and D. Geffroy, PRL ***116****, 256403 (2016)*

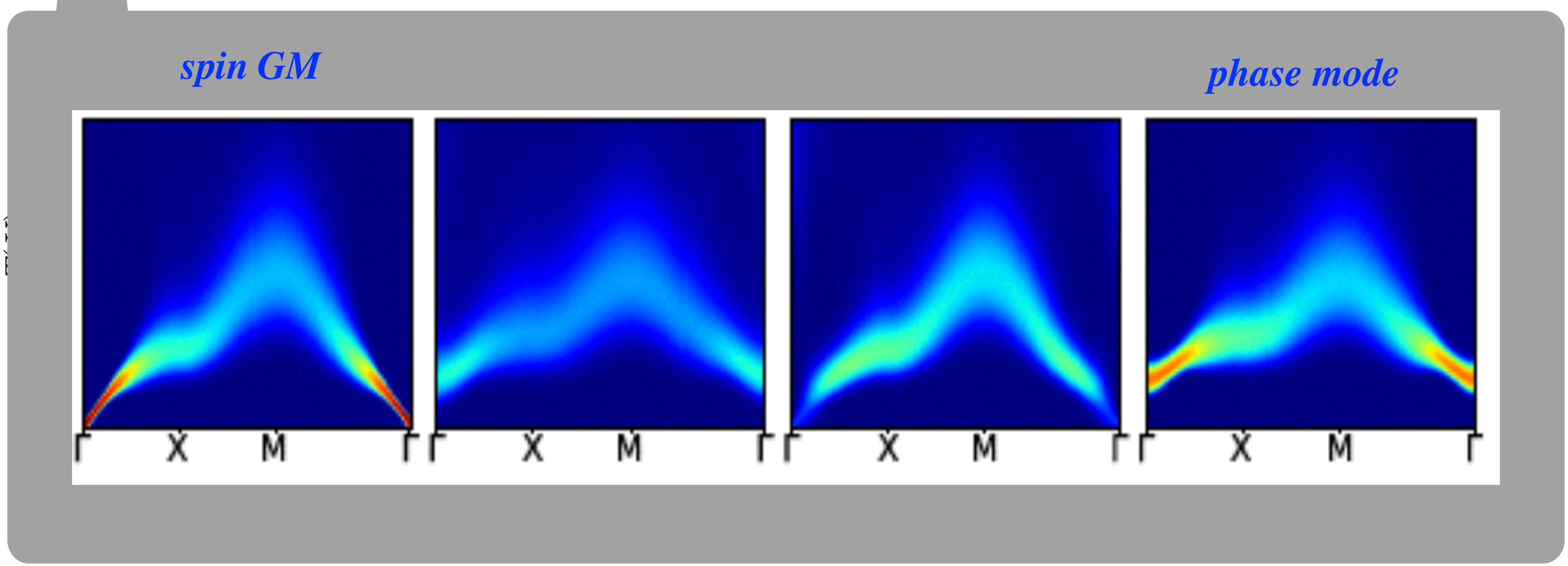
Doping + cross-hopping



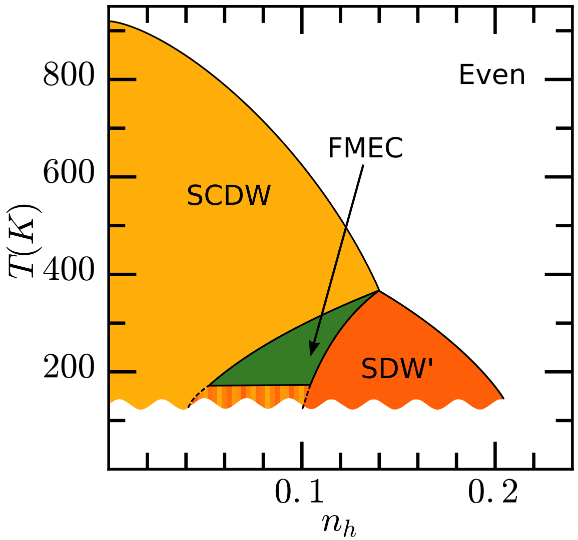


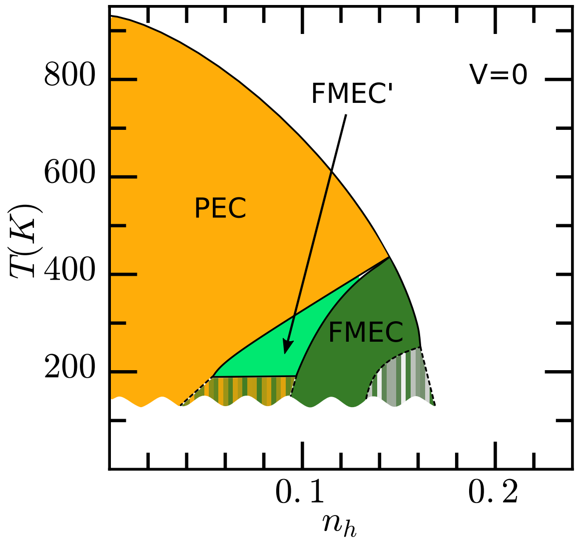
Doping + cross-hopping

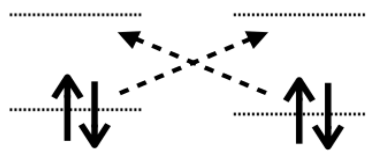




Doping + cross-hopping







$V_1=V_2$ even cross-hopping

$V_1=-V_2$ odd cross-hopping

$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a_\alpha^\dagger b_\beta \rangle$$

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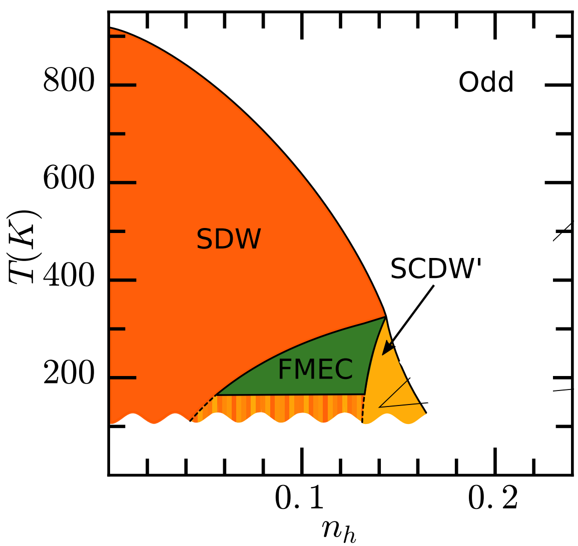
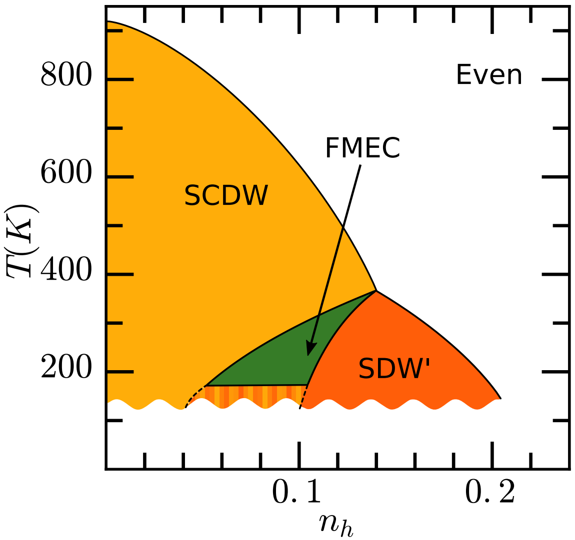


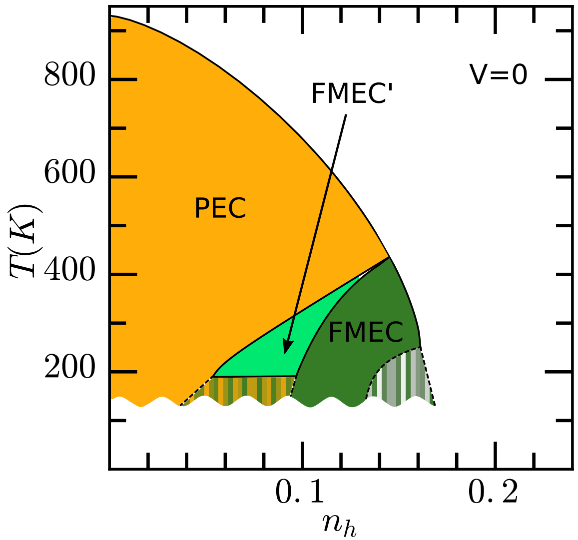
Condensate state	$M_\perp$	$M_\parallel$	$\mathbf{m}(\mathbf{r})$	$\mathbf{m}_\mathbf{k}$	Re ϕ	Im ϕ
FMEC	✓	✓, 0	✓	✓	✓	✓
SDW	0	0	✓	0	✓	0
SCDW	0	0	0	0	0	✓
SDW’	0	✓, 0	✓	✓	✓	0
SCDW’	0	0	0	✓	0	✓

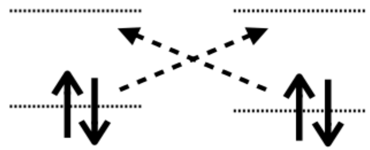
$$\mathbf{m}_\mathbf{k} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a_{\mathbf{k}\alpha}^\dagger a_{\mathbf{k}\beta} + b_{\mathbf{k}\alpha}^\dagger b_{\mathbf{k}\beta} \rangle$$

JK and D. Geffroy, PRL ***116****, 256403 (2016)*

Doping + cross-hopping







V$_1$=V$_2$ even cross-hopping
V$_1$=-V$_2$ odd cross-hopping

$$\phi = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_\alpha b_\beta \rangle$$

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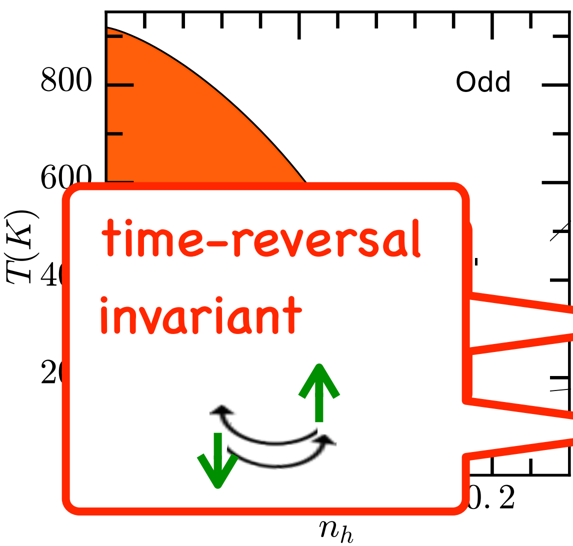
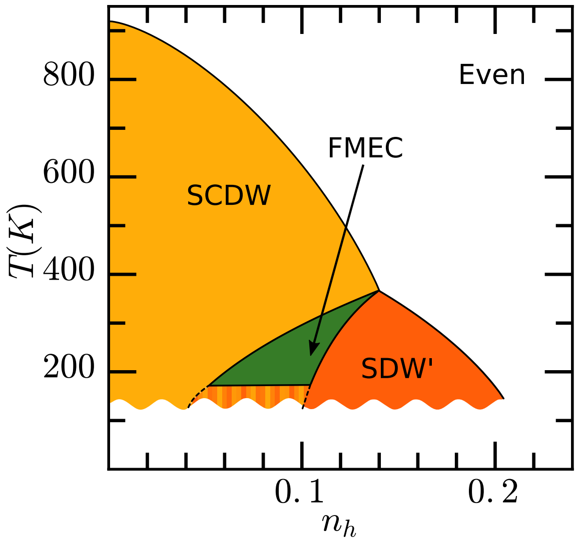


Condensate state	$M_\perp$	$M_\parallel$	$\mathbf{m}(\mathbf{r})$	$\mathbf{m}_\mathbf{k}$	Re ϕ	Im ϕ
FMEC	✓	✓, 0	✓	✓	✓	✓
SDW	0	0	✓	0	✓	0
SCDW	0	0	0	0	0	✓
SDW'	0	✓, 0	✓	✓	✓	0
SCDW'	0	0	0	✓	0	✓

$$\mathbf{m}_\mathbf{k} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_{\mathbf{k}\alpha} a_{\mathbf{k}\beta} + b^\dagger_{\mathbf{k}\alpha} b_{\mathbf{k}\beta} \rangle$$

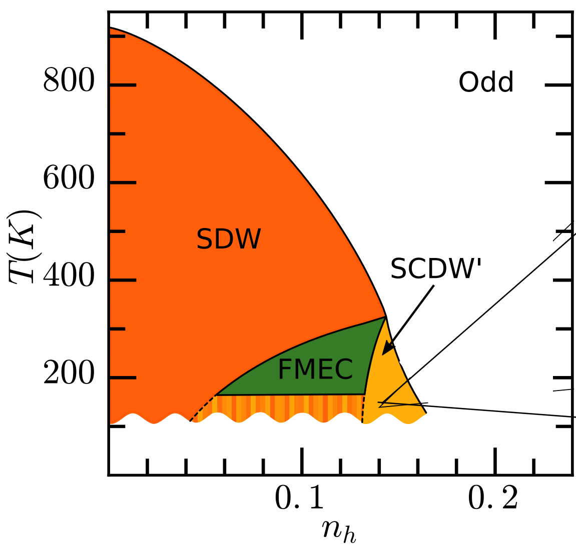
JK and D. Geffroy, PRL ***116****, 256403 (2016)*

Doping + cross-hopping

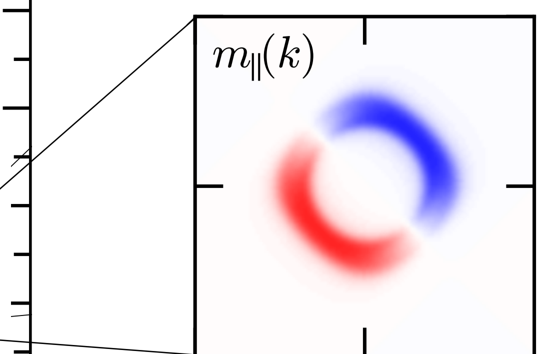


Condensate state	$M_\perp$	$M_\parallel$	$\mathbf{m}(\mathbf{r})$	$\mathbf{m}_\mathbf{k}$	Re ϕ	Im ϕ
FMEC	✓	✓, 0	✓	✓	✓	✓
SDW	0	0	✓	0	✓	0
SCDW	0	0	0	0	0	✓
SDW'	0	✓, 0	✓	✓	✓	0
SCDW'	0	0	0	✓	0	✓

$$\mathbf{m}_\mathbf{k} = \sum_{\alpha\beta} \boldsymbol{\sigma}_{\alpha\beta} \langle a^\dagger_{\mathbf{k}\alpha} a_{\mathbf{k}\beta} + b^\dagger_{\mathbf{k}\alpha} b_{\mathbf{k}\beta} \rangle$$



Spin density in k-space (Brillouin zone)



- k-space spin texture
- bulk spin current ?
- spontaneous spin-orbit coupling

*JK and D. Geffroy, PRL **116**, 256403 (2016)*

Science REPORTS

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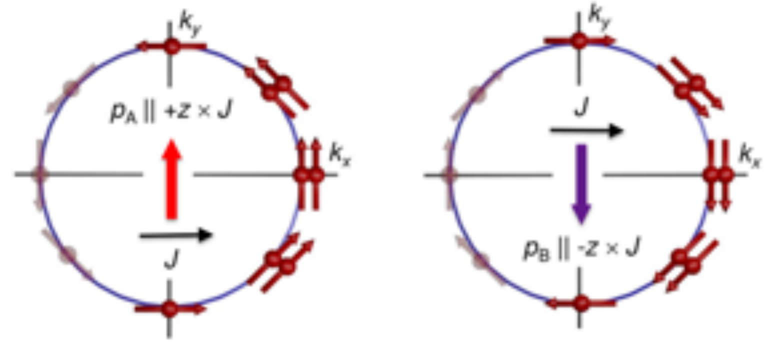
Electrical switching of an antiferromagnet

P. Wadley,[1*†] B. Howells,[1*] J. Železný,[2,3] C. Andrews,[1] V. Hills,[1] R. P. Campion,[1] V. Novák,[2] K. Olejník,[2] F. Maccherozzi,[4] S. S. Dhesi,[4] S. Y. Martin,[5] T. Wagner,[5,6] J. Wunderlich,[2,5] F. Freimuth,[7] Y. Mokrousov,[7] J. Kuneš,[8] J. S. Chauhan,[1] M. J. Grzybowski,[1,9] A. W. Rushforth,[1] K. W. Edmonds,[1] B. L. Gallagher,[1] T. Jungwirth,[2,1]

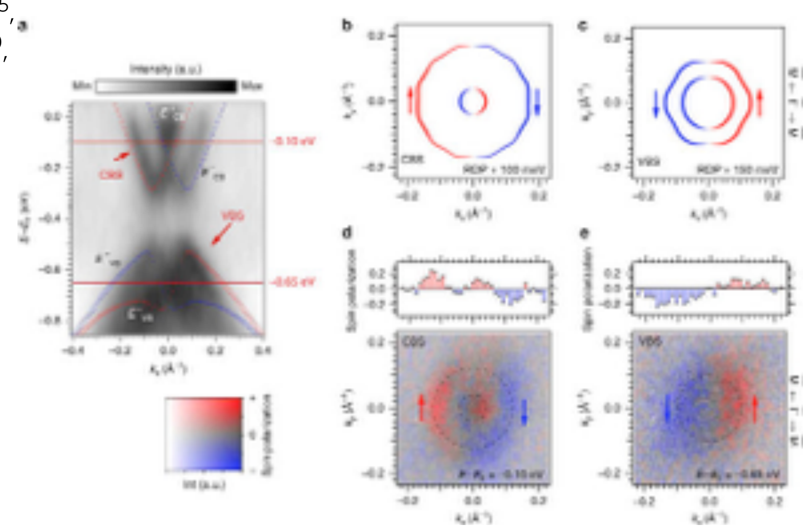


ARTICLE

Received 30 Nov 2015 | Accepted 14 Apr 2016 | Published 18 May 2016 DOI: 10.1038/ncomms11621 OPEN

Spin-texture inversion in the giant Rashba semiconductor BiTeI

Henriette Maaß[1], Hendrik Bentmann[1], Christoph Seibel[1], Christian Tusche[2], Sergey V. Eremeev[3,4,5], Thiago R.F. Peixoto[1], Oleg E. Tereshchenko[5,6,7], Konstantin A. Kokh[5,7,8], Evgueni V. Chulkov[4,5,9,10], Jürgen Kirschner[2] & Friedrich Reinert[1]

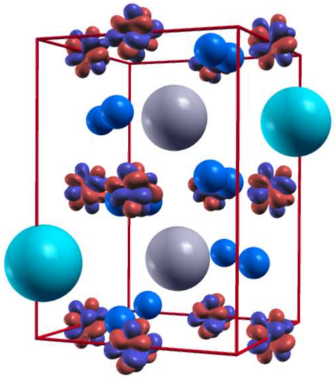


Excitonic condensation in real materials

Immanuel Kant

"Experience without theory is blind, but theory without experience is mere intellectual play."

$Pr_{0.5}Ca_{0.5}CoO_3$:

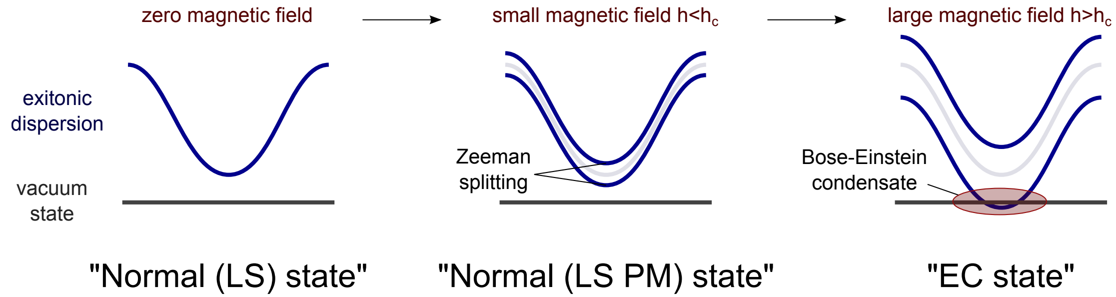


- metal-insulator transition
- disappearance of Co moments
- Pr^{3+} -> Pr^{4+} valence transition
- TR breaking without ordered moments

Tsubouchi et al. 2002, 2004
Hejtmanek et al. 2010, 2013
Knizek et al., 2010, 2013
JK & Augustinsky, 2014

$LaCoO_3$ in high field:

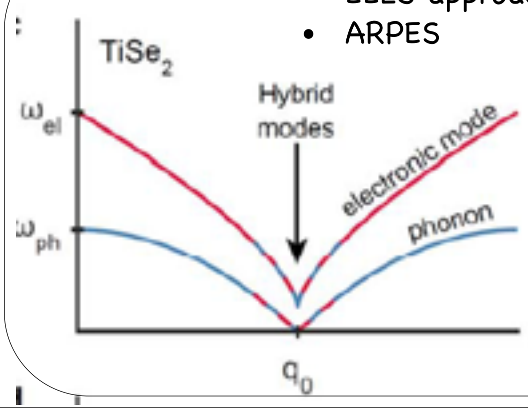
- positive $dB_c/dT>0$



Ikeda et al., 2016; Sotnikov & JK, 2016

1T-$TiSe_2$:

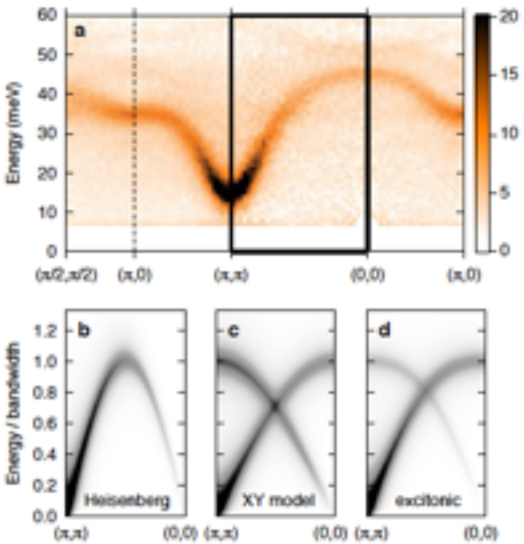
- EELS approaching instability
- ARPES



Kogar et al., 2013

Ca_2RuO_4:

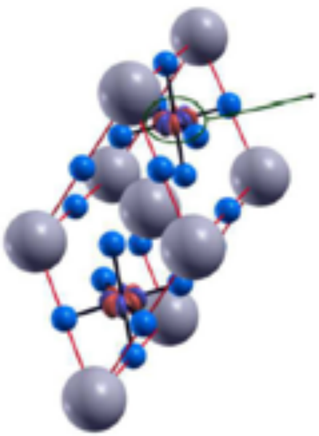
- INS in the ordered state



Jain et al., 2015

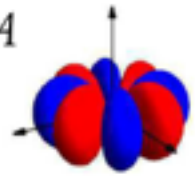
cubic d^6 cobaltite ($LaCoO_3$): LDA+U results

EC phase	$\lvert\phi'\rvert$	$\lvert\phi''\rvert$	$E - E_0[meV/f.u.]$
A-phase	0.151	0.000	-13.32
B-phase	0.191	0.000	-20.39
C-phase	0.202	0.009	-22.81



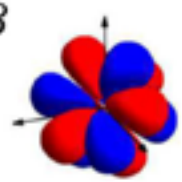
Phase Spin density Order parameter

A



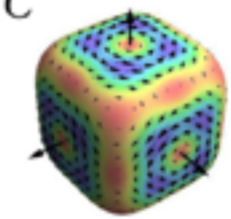
$$\phi_k^{(A)} = (-1)^k \begin{pmatrix} 0 & 0 & \lambda' \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

B



$$\phi_k^{(B)} = (-1)^k \begin{pmatrix} 0 & 0 & \lambda' \\ 0 & 0 & \lambda' \\ 0 & 0 & \lambda' \end{pmatrix}$$

C



$$\phi_k^{(C)} = \underbrace{(-1)^k \begin{pmatrix} \lambda' & 0 & 0 \\ 0 & \lambda' & 0 \\ 0 & 0 & \lambda' \end{pmatrix}}_{\text{staggered}} + i \underbrace{\begin{pmatrix} \lambda'' & 0 & 0 \\ 0 & \lambda'' & 0 \\ 0 & 0 & \lambda'' \end{pmatrix}}_{\text{uniform}}$$

- Numerous excitonic phases are possible
- Rhombohedral distortion (real structure) suppresses excitonic order
- Spin-orbit coupling favors the excitonic order

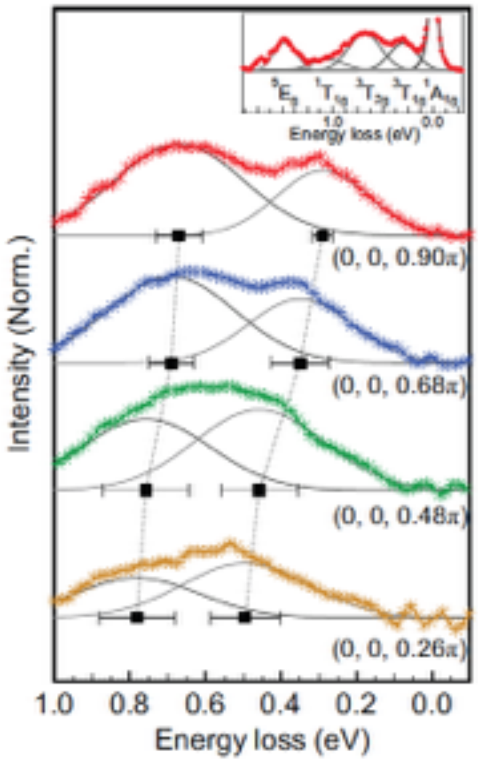
*J. Fernandez Afonso and JK, PRB **95**, 115131 (2017)*

Real $LaCoO_3$

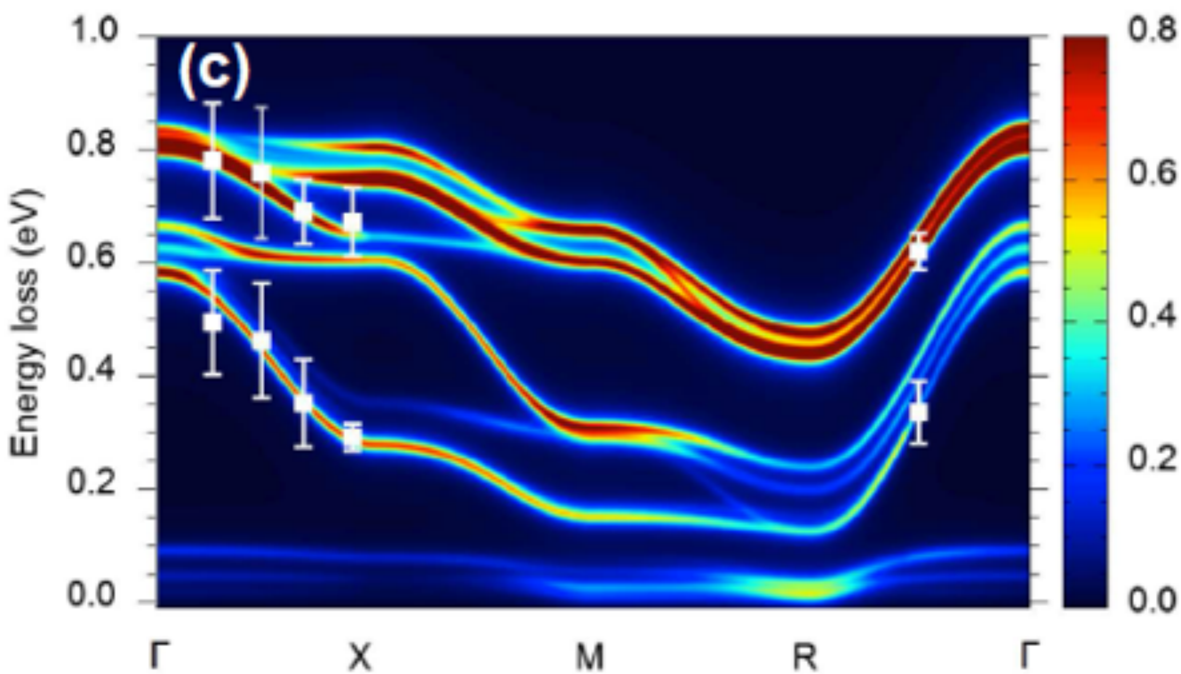
Are IS excitation mobile? **YES**

Do they condense? **NO**

Co L-edge RIXS



Exp + Theory



Wang et al. *PRB* ***90****, 035149 (2018)*

Real $LaCoO_3$

Are IS excitation mobile? **YES**

Do they condense? **NO**

Why not? formation of immobile bi-excitons IS+IS->HS

Why not spin state order HS-LS-HS-LS?

important question

HS $\rightleftarrows$ IS + IS ???

Real $LaCoO_3$

Are IS excitation mobile? **YES**

Do they condense? **NO**

Why not? formation of immobile bi-excitons IS+IS->HS

Why not spin state order HS-LS-HS-LS?

important question

HS $\rightleftarrows$ IS + IS ???

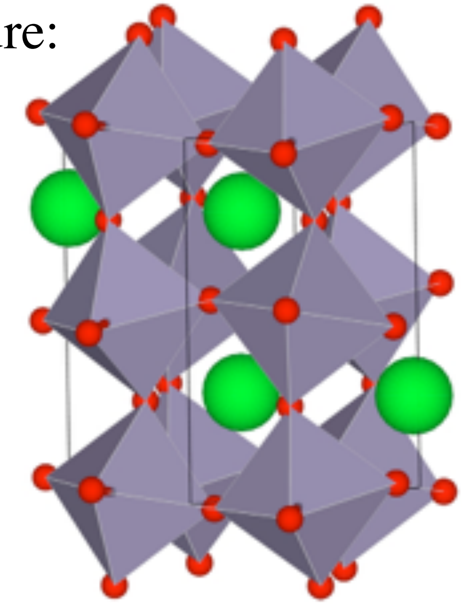
Possible route to exciton magnet in cobaltites:

2D structure, e.g., $(SrLa)_2CoO_4$

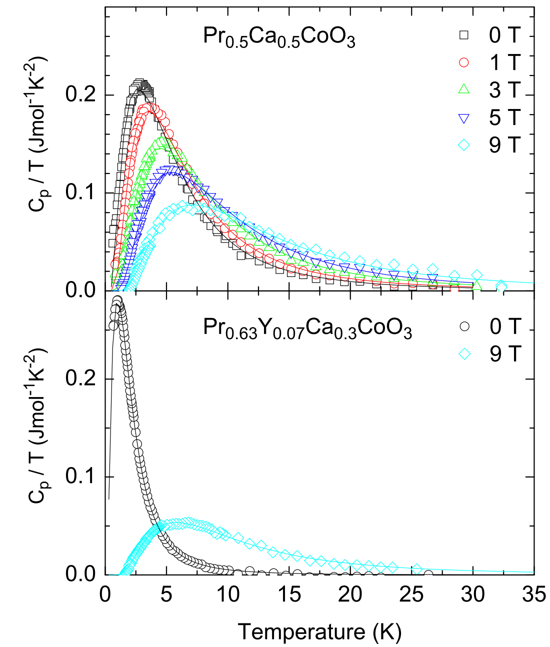
=> smaller E_{IS}-E_{HS}

Exchange splitting in $Pr_{0.5}Ca_{0.5}CoO_3$

crystal structure:

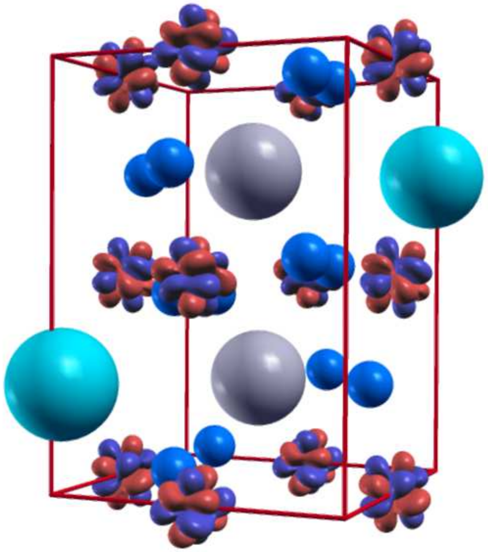


Pr^{4+} Schottky peak:



J. Hejtmánek et al., EPJB 86, 305 (2013)

spin density in condensed phase (LDA+U)



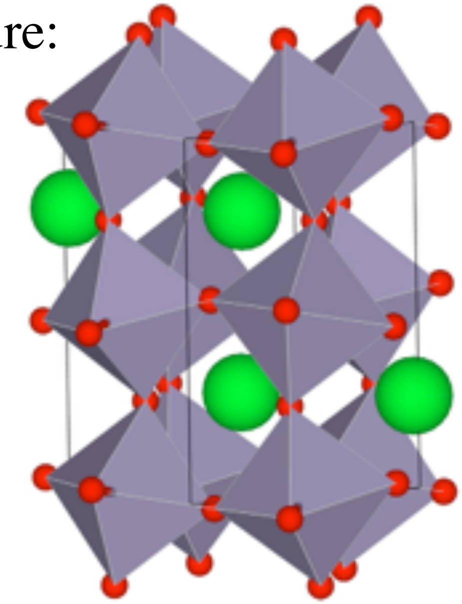
Kondo interaction of Pr moment with Co d-electrons:

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

JK and P. Augustinský, PRB ***90****, 235112 (2014)*

Exchange splitting in $Pr_{0.5}Ca_{0.5}CoO_3$

crystal structure:



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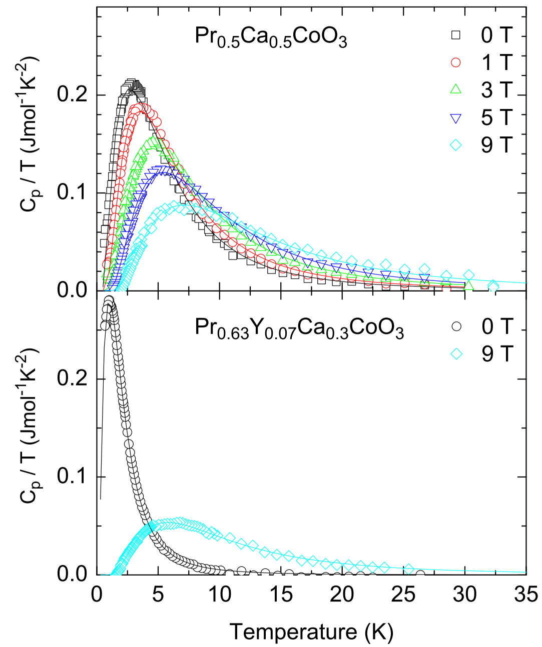

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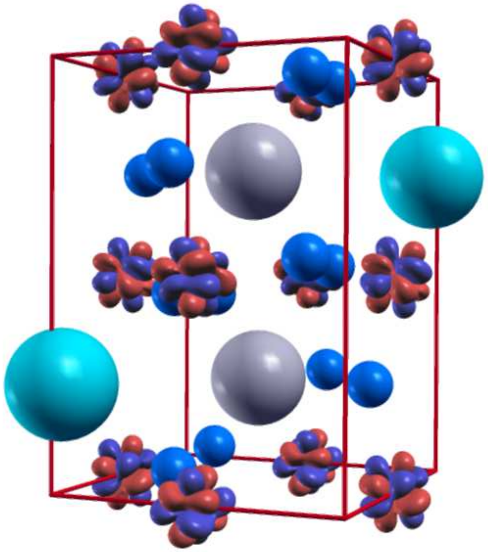
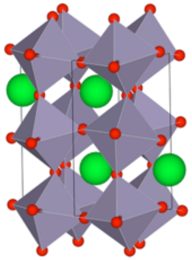


Kondo interaction of Pr moment with Co d-electrons:
in the condensate

$$H^{(n)} = \sum_{\alpha\alpha'} \sum_{mm'} \sum_{i} \mathbf{S} \cdot h_{\gamma}^{(n)}$$

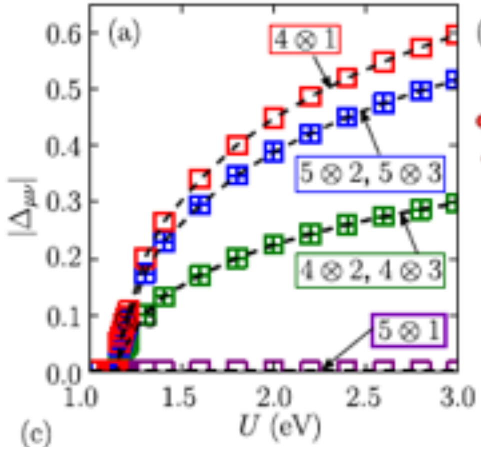
$$h_{\gamma}^{(n)} = \sum_{imm'} J_{i,mm'}^{(n)} \sum_{\alpha\alpha'} 2\,\mathrm{Re}\langle c_{im\alpha}^{\dagger} \sigma_{\alpha\alpha'}^{\gamma} c_{im'\alpha'} \rangle$$

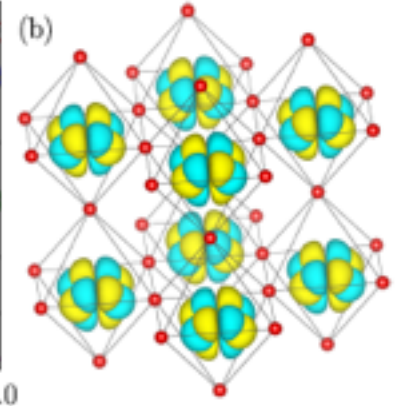
*JK and P. Augustinský, PRB **90**, 235112 (2014)*



$Pr_{0.5}Ca_{0.5}CoO_3$

Order parameter:

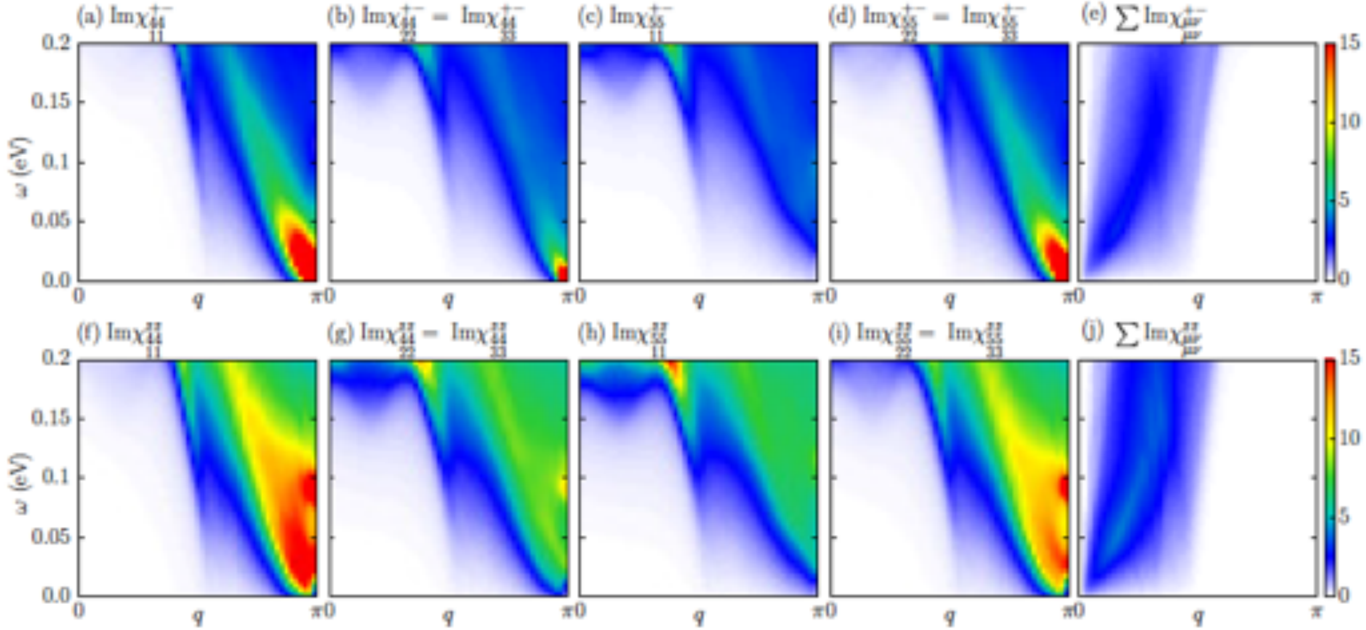




RPA calculation in ideal cubic structure

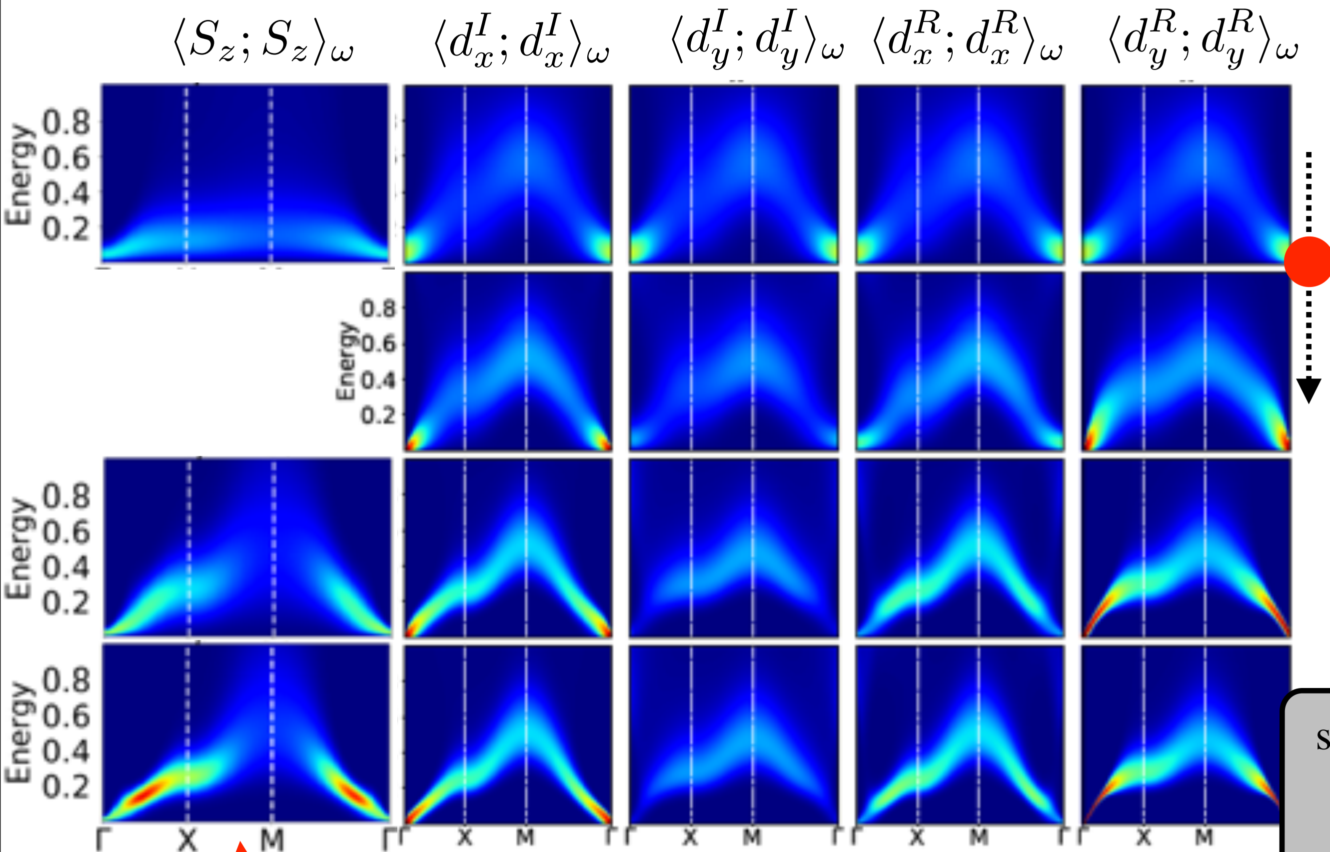
Excitonic and spin susceptibility:

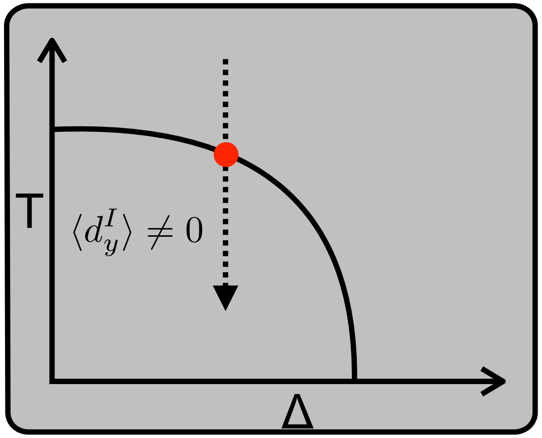
(c)



T. Yamaguchi, K. Sugimoto and Y. Ohta, JPSJ ***86****, 043701 (2017)*

How to detect excitonic condensate (PEC)?





Similar behavior recently observed in INS experiment on $(Pr_{1-y}Y_y)_{1-x}Ca_xCoO_3$

T. Mojoshi et al. PRB ***98****, 205105 (2018)*

strong-coupling picture:

$$S_z = d_1^\dagger d_1 - d_{-1}^\dagger d_{-1}$$
$$= i(d_x^\dagger d_y - d_y^\dagger d_x)$$

in the PEC phase:

$$\langle d_y\rangle = i\phi \quad \Rightarrow$$
$$S_z \approx -\phi(d_x^\dagger + d_x) = -\phi d_x^R$$

Collaborators:

Atsushi Hariki

Andrii Sotnikov

Juan Fernandez Afonso

Dominique Geffroy

Pavel Augustinský

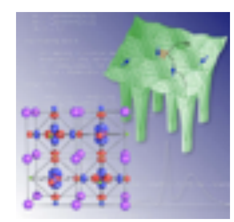
Ru-Pan Wang

Frank de Groot

Zdenek Jirak

Sponsors:

ERC CoG EXMAG



DFG Research Unit FOR1346

Conclusions

• Excitonic magnetism provides a rich field of new physics with potentially interesting application

• Excitonic magnets have yet to be found (promising candidates exist)

• Experimental techniques for unambiguous identification of excitonic condensate have to be established

Higgs mode