Variational wave functions for multiband Hubbard models

Federico Becca

CNR IOM-DEMOCRITOS and International School for Advanced Studies (SISSA)

ICTP Trieste, November, 2017

C. de Franco (SISSA), L.F. Tocchio (Torino)

R. Kaneko (Tokyo), R. Valenti (Frankfurt)

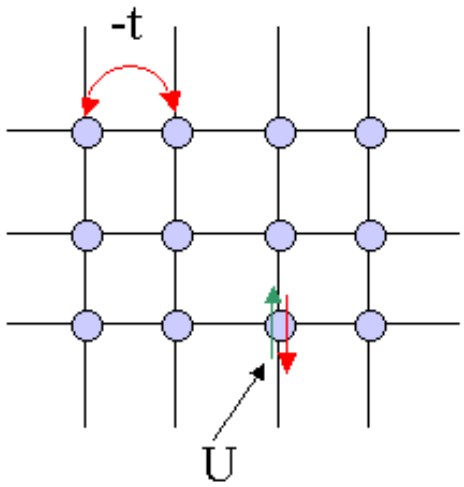
1. Variational wave functions for the Hubbard model
   - The Jastrow-Slater wave functions
   - How to distinguish between metals and insulators

2. Results for the two-band Hubbard model
   - The orbital-selective Mott transition on the square lattice
   - Charge orders in organic charge-transfer salts

3. Conclusions

The one-band Hubbard Model

$$H = -t \sum_{i,j,\sigma} c^{\dagger}_{i,\sigma} c_{j,\sigma} + h.c. + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$



The Hubbard model is the prototype for correlated electrons on the lattice
NO exact solution in $D > 1$

- Does it give rise to (high-temperature) superconductivity?
- Benchmark for several numerical methods (mostly in 2D):
  - Several quantum Monte Carlo techniques (variational, diffusion, path integral)
  - Density-matrix renormalization group and tensor networks (iPEPS)
  - Dynamical mean-field therory and cluster extensions
  - Embedding schemes (density-matrix embedding theory)

Le Blanc *et al.* (Simons collaboration), PRX (2015)

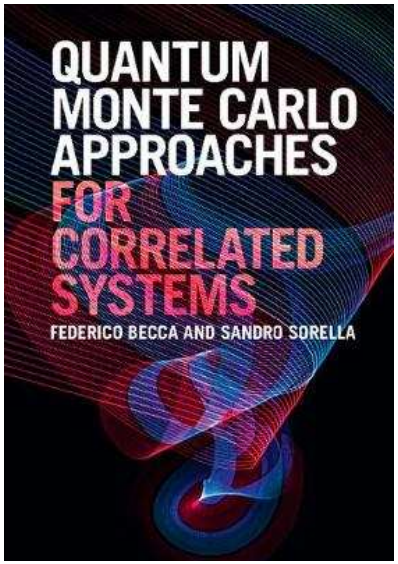
Motivations and strategy

GOAL: capture the ground state by variational wave functions

We want to construct flexible variational states that may describe:

- Metals and superconductors
- Phases with charge and/or spin order, both metallic and insulating
- Mott insulators without any local order
- (Topologocal phases, including chiral spin liquids are also possible)

We employ Jastrow-Slater wave functions and Monte Carlo sampling



Cambridge University Press (November 2007)

Non-interacting (Slater or BCS) determinant

Long-range Jastrow factor

Capello, Becca, Fabrizio, Sorella, and Tosatti, PRL (2005)

Kaneko, Tocchio, Valenti, Becca, and Gros, PRB (2016)

(Backflow correlations and Lanczos steps)

Tocchio, Becca, Parola, and Sorella, PRB (2008)

Tocchio, Becca, and Gros, PRB (2011)

Becca and Sorella, PRL (2001)

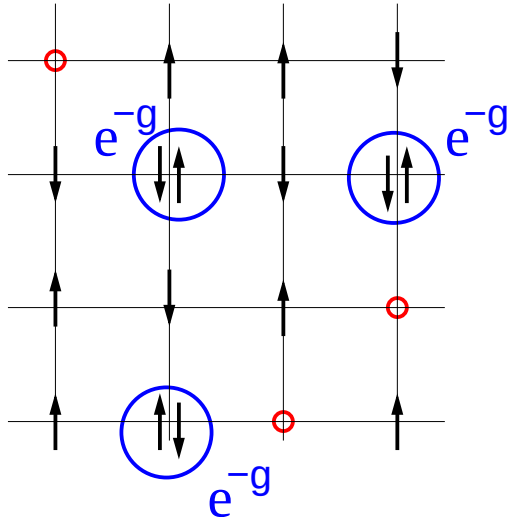
Prehistory of correlated wave functions for Mott insulators

Gutzwiller wave function

$$|\Phi_g\rangle = e^{-g \sum_i n_{i,\uparrow} n_{i,\downarrow}} |\Psi_0\rangle$$

Gutzwiller, PRL (1963)

Yokoyama and Shiba, JPSJ (1987)



It does not correlate empty and doubly occupied sites

Metallic for $g \neq \infty$ (any finite U/t)

Empty and doubly occupied sites play a crucial role for the conduction

They must be correlated otherwise an electric field would induce a current

The Jastrow-Slater wave functions

The low-energy properties reflect the long-distance behavior
We must change the density-density correlations of $|\Psi_0\rangle$ at large distance

$$\boxed{|\Psi\rangle = \mathcal{J}|\Psi_0\rangle}$$

$$\mathcal{J} = \exp\left(-\frac{1}{2}\sum_{i,j} v_{i,j} n_i n_j\right) = \exp\left(-\frac{1}{2}\sum_{q} v_q n_{-q} n_q\right)$$

$|\Psi_0\rangle$ is an uncorrelated determinant obtained from a non-interacting Hamiltonian:

$$\mathcal{H}_0 = \sum_{i,j,\sigma} t_{i,j} c^\dagger_{i,\sigma} c_{j,\sigma} + \sum_{i,j} \Delta_{i,j} c^\dagger_{i,\uparrow} c^\dagger_{j,\downarrow} + h.c.$$

$$\boxed{|\Psi_0\rangle = \exp\left\{\sum_{i,j} f_{i,j} c^\dagger_{i,\uparrow} c^\dagger_{j,\downarrow}\right\} |0\rangle}$$

For $v_{i,i} \to \infty$
The RVB physics is recovered

Anderson, Science (1987)

Find the optimal set of parameters $v_{i,j}$, $t_{i,j}$ and $\Delta_{i,j}$ which minimizes the energy

Sorella, PRB (2005)

Metal or insulator?

Ansatz for the low-energy excitations Feynman, Phys. Rev. (1954)

$$\boxed{|\Psi_q\rangle = n_q|\Psi\rangle} \qquad N_q = \langle\Psi|n_{-q}n_q|\Psi\rangle/\langle\Psi|\Psi\rangle$$

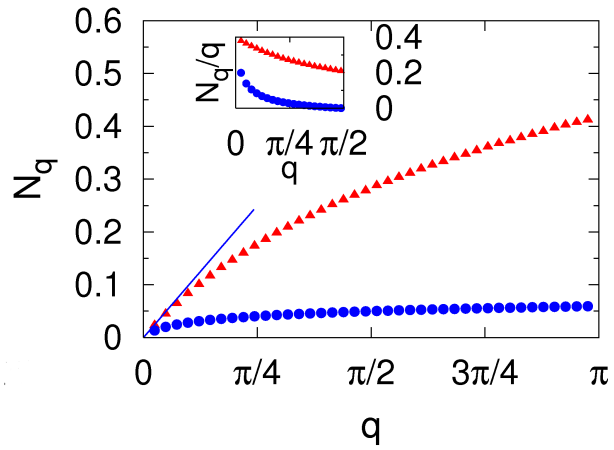
f-sum rule

$$\boxed{\Delta E_q = \frac{\langle\Psi_q|(H-E_0)|\Psi_q\rangle}{\langle\Psi_q|\Psi_q\rangle} = \frac{\langle\Psi|[n_{-q},[H,n_q]]|\Psi\rangle}{2N_q} \approx \frac{q^2}{N_q}}$$

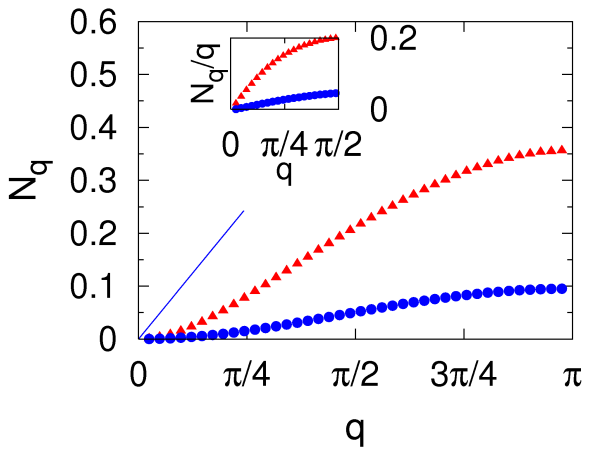
$N_q \sim |q| \Rightarrow \Delta E_q \to 0 \quad \Rightarrow$ metal
$N_q \sim q^2 \Rightarrow \Delta E_q$ is finite $\Rightarrow$ insulator

Example: 1D Hubbard model at half filling with $U/t = 4$ and 10

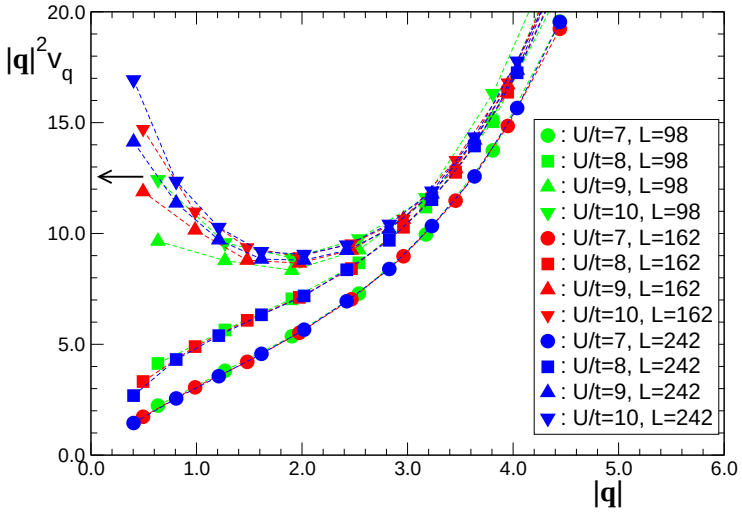
Gutzwiller WF

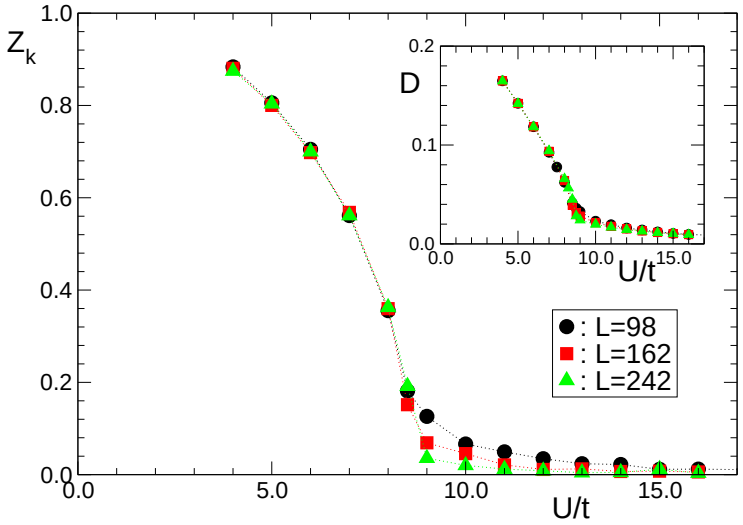


Long-range Jastrow WF



Two-dimensional (paramagnetic) Hubbard model





$$N_q \approx \frac{N_q^0}{1+2v_q N_q^0} \approx \frac{1}{v_q}$$

$$Z_k = \frac{\left|\langle \Psi_{N-1} | c_{k,\sigma} | \Psi_N \rangle\right|^2}{\langle \Psi_N | \Psi_N \rangle \langle \Psi_{N-1} | \Psi_{N-1} \rangle}$$

N_q^0 is the uncorrelated structure factor $\qquad |\Psi_{N-1}\rangle = \mathcal{J}\, c_{k,\sigma}\, |\Psi_0\rangle$

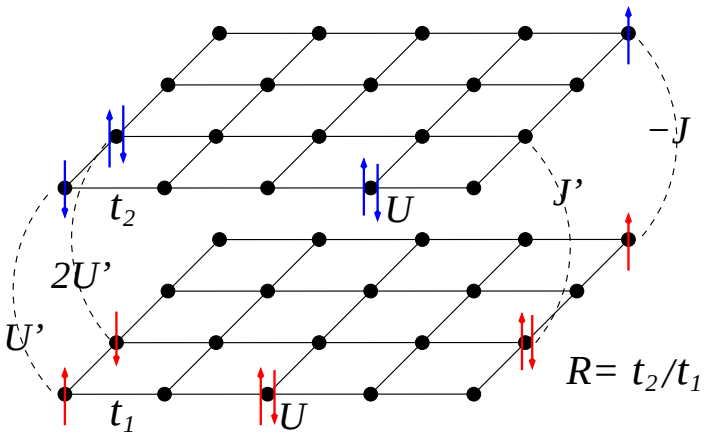
- $U/t \lesssim 8.5$: $v_q \sim \frac{1}{|q|}$ with Z_k finite: **FERMI LIQUID**
- $U/t \gtrsim 8.5$: $v_q \sim \frac{1}{q^2}$ with vanishing Z_k: **MOTT INSULATOR**
- AF parameter in the Slater determinant: AF order for $U > 0$ (BAND INSULATOR)

The two-band Hubbard model on the square lattice

$$H_{\text{kin}} = -\sum_{\langle i,j\rangle,\alpha,\sigma} t_\alpha c^\dagger_{i,\alpha,\sigma} c_{j,\alpha,\sigma} + \text{h.c.}$$

$$H_{\text{int}} = U \sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow} + (U - 2J) \sum_{i} n_{i,1} n_{i,2}$$

$$H_{\text{Hund}} = -J \sum_{i,\sigma,\sigma'} c^\dagger_{i,1,\sigma} c_{i,1,\sigma'} c^\dagger_{i,2,\sigma'} c_{i,2,\sigma} - J \sum_{i} c^\dagger_{i,1,\uparrow} c^\dagger_{i,1,\downarrow} c_{i,2,\uparrow} c_{i,2,\downarrow} + \text{h.c.}$$

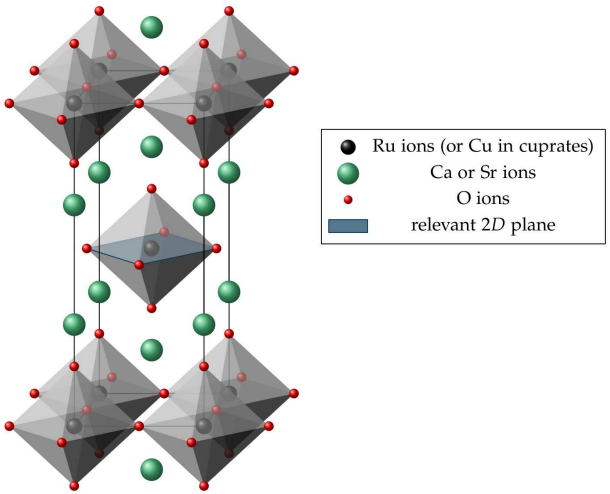


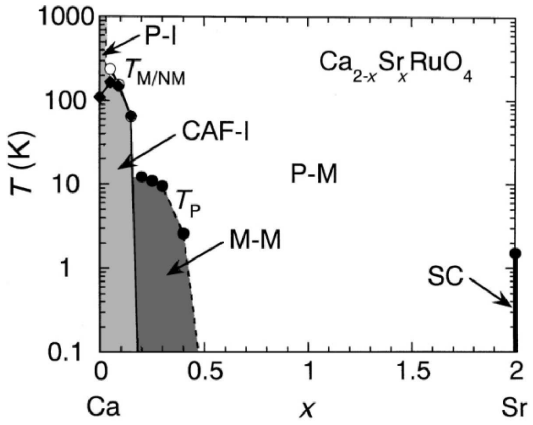
- Half-filling (2 electrons/site)
- Rotational symmetry of degenerate orbitals
  $U' = U - 2J$
  $J' = J$

  Kanamori, Prog.Theor.Phys. (1963)
- Small enough $R = t_2/t_1 \Rightarrow$ **OSMI**
  one orbital undergoes the MIT
  while the other remains metallic

Tocchio, Arrigoni, Sorella, and Becca, J. of Phys.: Cond. Matter (2016)

$Ca_{2-x}Sr_xRuO_4$ ruthenate: an orbital selective state?





Maeno *et al.*, Nature (1994)

- Coexistence of spin-1/2 moments and metallicity (M-M phase)
- Possible explanation: presence of both localized and delocalized bands

Anisimov, Nekrasov, Kondakov, Rice, and Sigrist, EPJB (2002)

Several works that used dynamical mean-field theory and slave-particle approaches

Liebsch, PRL (2003)

Koga, Kawakami, Rice, and Sigrist, PRL (2004)

Ferrero, Becca, Fabrizio, and Capone, PRB (2005)

de Medici, Georges, and Biermann, PRB (2005)

Arita and Held, PRB (2005)

Rüegg, Indergand, Pilgram, and Sigrist, EPJB (2005)

Inaba and Koga, PRB (2006)

The non-magnetic variational wave function

$$|\Psi\rangle = \mathcal{J}|\Psi_0\rangle$$

$$\mathcal{J} = \exp\left(-\frac{1}{2}\sum_{i,j}\sum_{\alpha\beta} v_{i,j}^{\alpha\beta} n_{i,\alpha} n_{j,\beta}\right)$$

$|\Psi_0\rangle$ is the ground state of a non-interacting Hamiltonian with

- Intra-orbital hopping

  $\sum_{k,\alpha,\sigma} \{-2\tilde{t}_\alpha(\cos k_x + \cos k_y) - \mu_\alpha\} c^\dagger_{k,\alpha,\sigma} c_{k,\alpha,\sigma}$
- Intra-orbital singlet pairing with *d*-wave symmetry

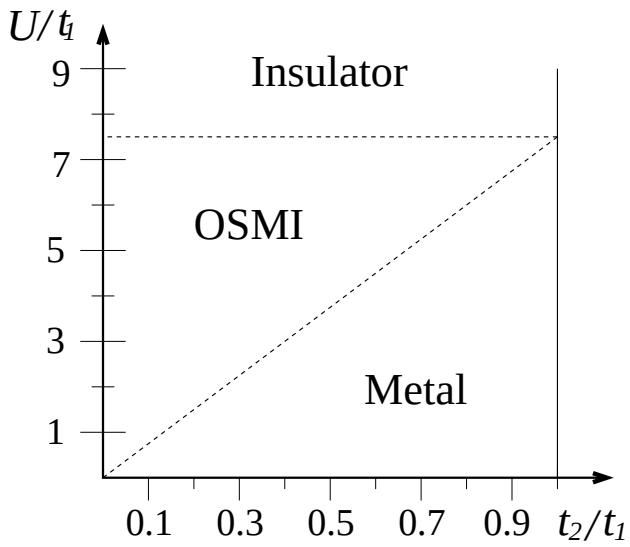
  $\sum_{k,\alpha} 2\Delta_\alpha(\cos k_x - \cos k_y)\left(c^\dagger_{k,\alpha,\uparrow} c^\dagger_{-k,\alpha,\downarrow} + c_{-k,\alpha,\downarrow} c_{k,\alpha,\uparrow}\right)$
- Inter-orbital triplet pairing (finite Hund's coupling)

  $\Delta^t_\perp \sum_i \left(c^\dagger_{i,1,\uparrow} c^\dagger_{i,2,\downarrow} - c^\dagger_{i,2,\uparrow} c^\dagger_{i,1,\downarrow} + c_{i,2,\downarrow} c_{i,1,\uparrow} - c_{i,1,\downarrow} c_{i,2,\uparrow}\right)$

- $\tilde{t}_2$, Δ_α, $\Delta^t_\perp$ and μ_α are variational parameters to be optimized ($\tilde{t}_1 = 1$)
- no further inter-orbital hopping $t_\perp$ can be stabilized in the wave function
  $t_\perp \sum_{i,\sigma}\left(c^\dagger_{i,1,\sigma} c_{1,2,\sigma} + c^\dagger_{i,2,\sigma} c_{i,1,\sigma}\right)$

The phase diagram for decoupled bands

$$H = -\sum_{\langle i,j\rangle,\alpha,\sigma} t_\alpha c^\dagger_{i,\alpha,\sigma} c_{j,\alpha,\sigma} + \text{h.c.} + U \sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow}$$



$$0 < R = t_2/t_1 < 1$$

$$\frac{U_c^1}{t_1} = 7.5 \pm 0.5$$

$$\frac{U_c^2}{t_1} = R\frac{U_c^1}{t_1}$$

The two orbitals are decoupled and each one undergoes a MIT independently

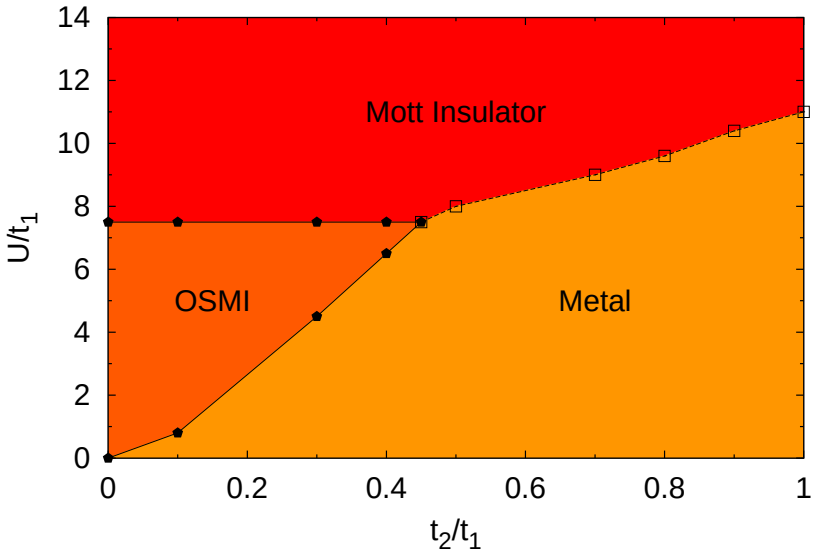
trivial OSMI

Do they still have separated MIT when they are no longer decoupled?

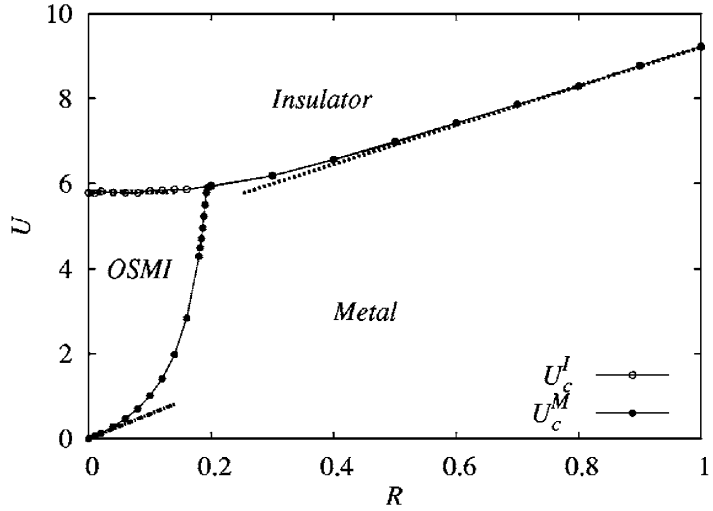
The phase diagram for $J = 0$

$$H = -\sum_{\langle i,j\rangle,\alpha,\sigma} t_\alpha c^\dagger_{i,\alpha,\sigma} c_{j,\alpha,\sigma} + \text{h.c.} + U\sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow} + U\sum_i n_{i,1} n_{i,2}$$

Variational Monte Carlo



DMFT



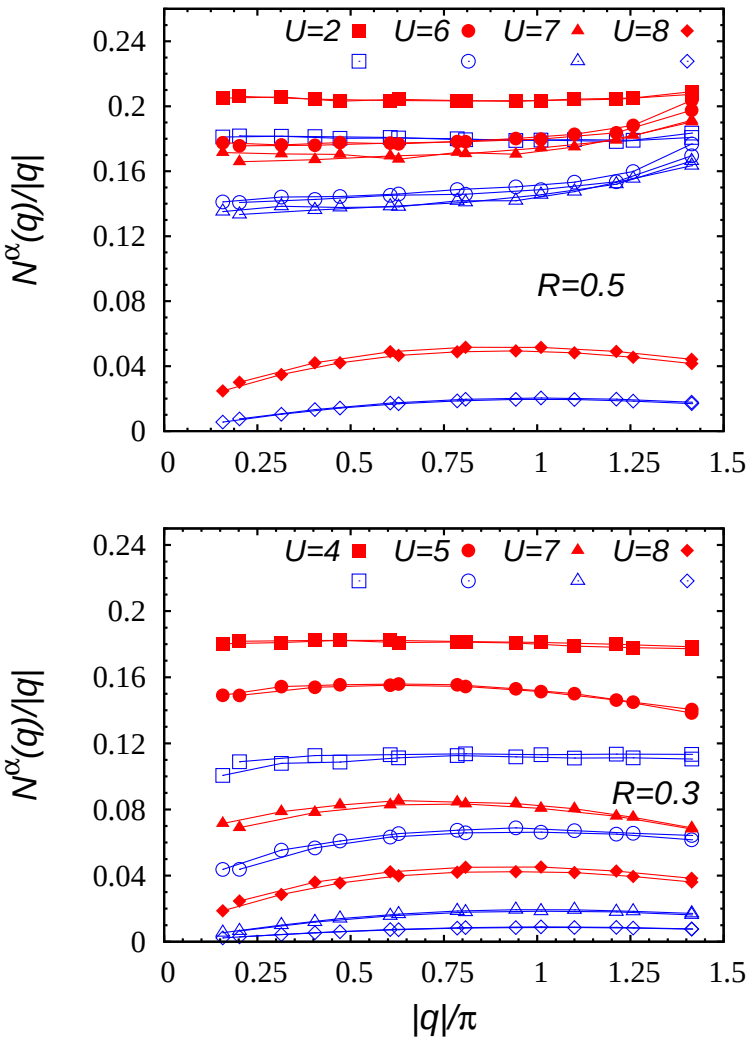
Inaba and Koga, PRB (2006)

see also: de Medici, Georges, and Biermann, PRB (2005)

and Ferrero, Becca, Fabrizio, and Capone PRB (2005)

The presence of the inter-band U favors a metallic phase

Metal-insulator transitions



- $N^\alpha(q) = \langle n^\alpha_q n^\alpha_{-q} \rangle \sim q^2$ for $|q| \to 0$: band α is insulating (gapped)
- $N^\alpha(q) = \langle n^\alpha_q n^\alpha_{-q} \rangle \sim q$ for $|q| \to 0$: band α is metallic (gapless)

Three phases can be found:

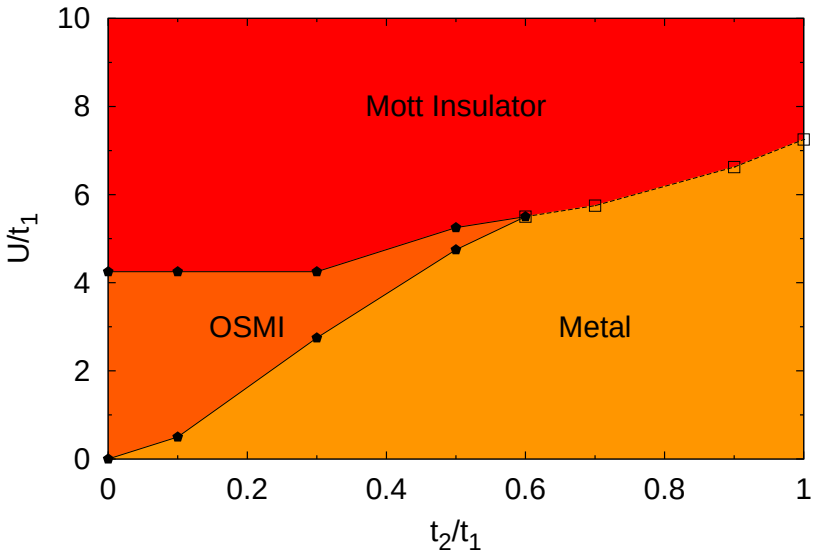
- Metal (e.g., $U/t_1 = 6, R = 0.5$)
- Mott (e.g., $U/t_1 = 8, R = 0.5$)
- OSMI (e.g., $U/t_1 = 7, R = 0.3$)

- Small R: smooth metal-OSMI-Mott transitions
- Large R: first-order metal-Mott transition

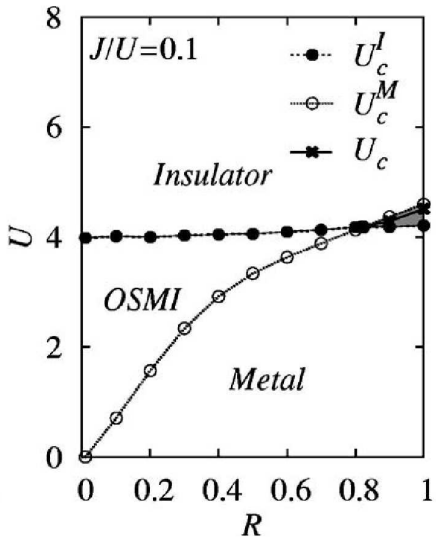
The phase diagram at $J/U = 0.1$

$$H = -\sum_{\langle i,j\rangle,\alpha,\sigma} t_\alpha c^\dagger_{i,\alpha,\sigma} c_{j,\alpha,\sigma} + \text{h.c.} + U\sum_{i,\alpha} n_{i,\alpha,\uparrow} n_{i,\alpha,\downarrow} + U\sum_i n_{i,1} n_{i,2}$$
$$-J\sum_{i,\sigma,\sigma'} c^\dagger_{i,1,\sigma} c_{i,1,\sigma} c^\dagger_{i,2,\sigma'} c_{i,2,\sigma} - J\sum_i c^\dagger_{i,1,\uparrow} c^\dagger_{i,1,\downarrow} c_{i,2,\uparrow} c_{i,2,\downarrow} + \text{h.c.}$$

Variational Monte Carlo



DMFT

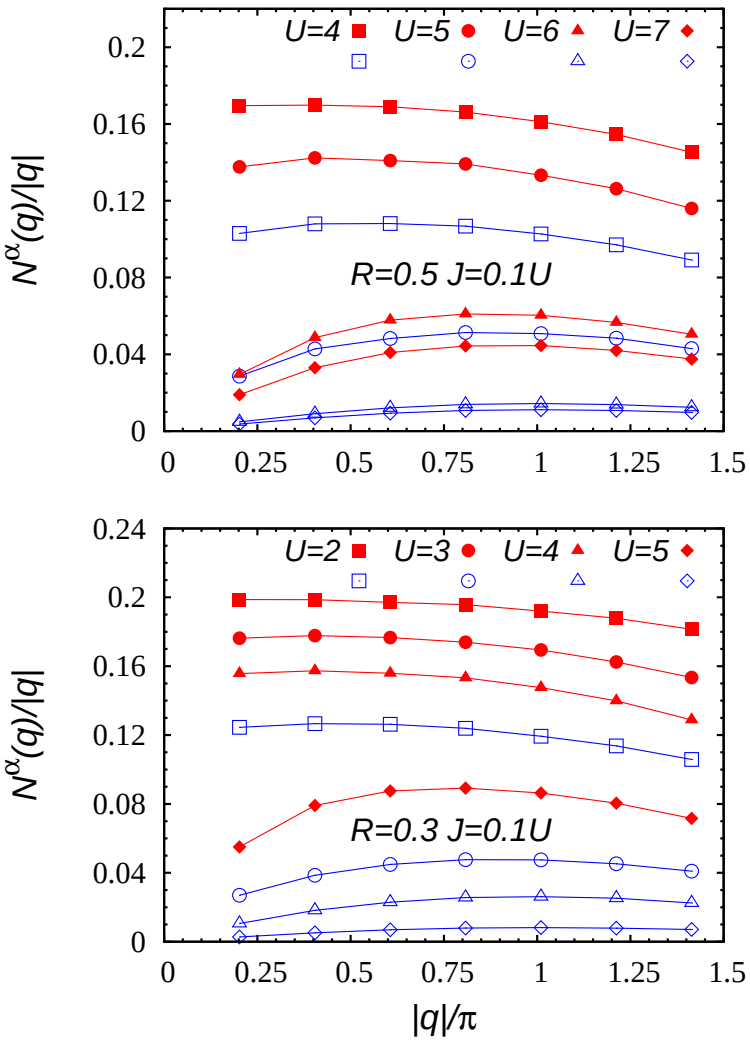


Inaba and Koga, PRB (2006)

The Hund's coupling J favors the Mott phase at half filling

de Medici PRB (2011)

Metal-insulator transitions

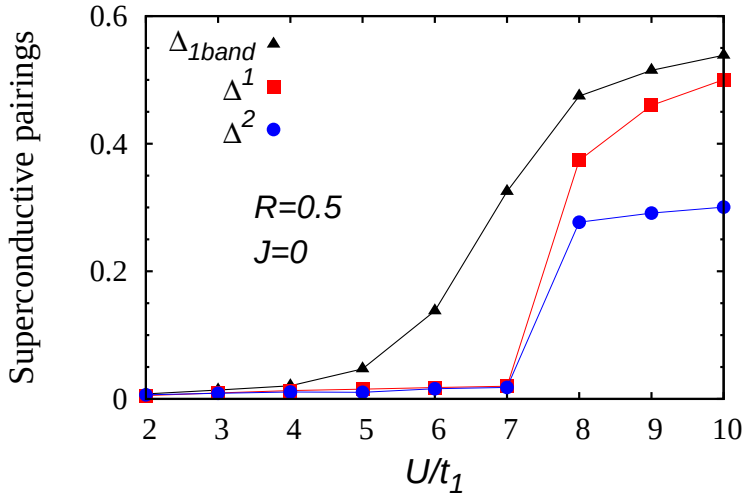


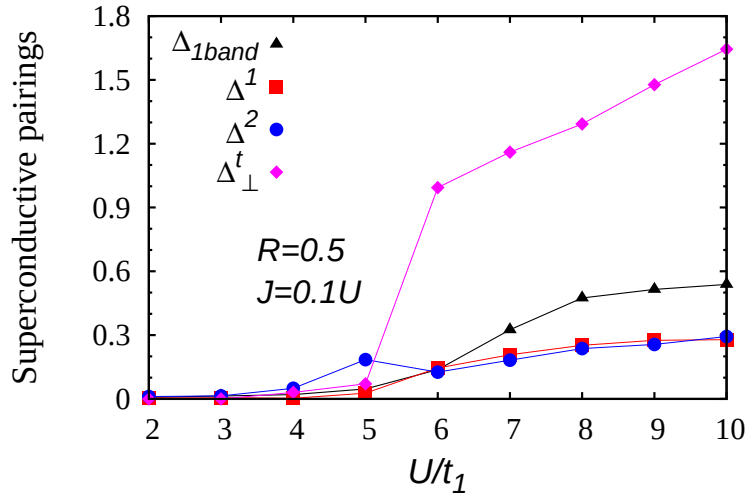
- $N^\alpha(q) = \langle n^\alpha_q n^\alpha_{-q} \rangle \sim q^2$ for $|q| \to 0$: band α is insulating (gapped)
- $N^\alpha(q) = \langle n^\alpha_q n^\alpha_{-q} \rangle \sim q$ for $|q| \to 0$: band α is metallic (gapless)

Three phases can be found:

- Metal (e.g., $U/t_1 = 4, R = 0.5$)
- Mott (e.g., $U/t_1 = 7, R = 0.5$)
- OSMI (e.g., $U/t_1 = 4, R = 0.3$)

The importance of BCS paring





- For $J = 0$, intra-orbital singlet pairing with *d*-wave symmetry
  (similarly to the one-band Hubbard model: RVB picture)

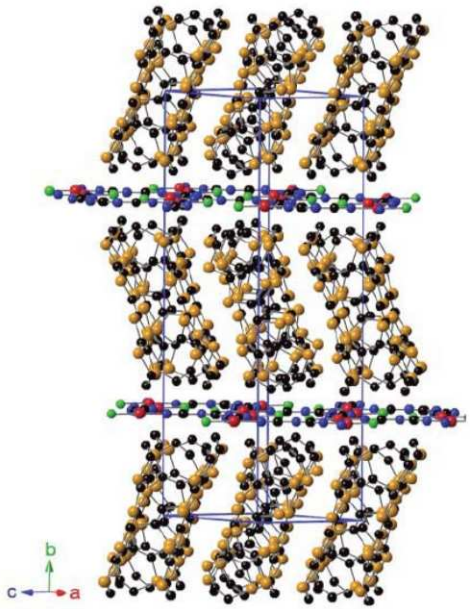
  Anderson, Science (1987)

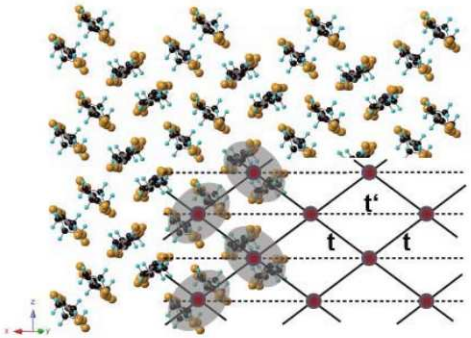
  $$\sum_{k,\alpha} 2\Delta_\alpha (\cos k_x - \cos k_y) \left(c^\dagger_{k,\alpha,\uparrow} c^\dagger_{-k,\alpha,\downarrow} + c_{-k,\alpha,\downarrow} c_{k,\alpha,\uparrow} \right)$$

- For $J > 0$, also inter-orbital triplet pairing
  (to favor spin alignment from the Hund's coupling)

  $$\Delta^t_\perp \sum_i \left(c^\dagger_{i,1,\uparrow} c^\dagger_{i,2,\downarrow} - c^\dagger_{i,2,\uparrow} c^\dagger_{i,1,\downarrow} + c_{i,2,\downarrow} c_{i,1,\uparrow} - c_{i,1,\downarrow} c_{i,2,\uparrow} \right)$$

The organic charge-transfer salts κ-$(ET)_2X$





- Layers of organic ET molecules (bis(ethylenedithio)tetrathiafulvalene)
- Insulating anion sheets X=$Cu[N(CN)_2]Cl$
- κ packing of ET molecules with strong dimerization
- 3/4 filling within the ET layers (3 electrons per dimer)

M. Lang *et al.*, IEEE Trans. Magn. (2014)

Ferroelectricity in κ-$(ET)_2Cu[N(CN)_2]Cl$

- Ferroelectric transition (peak in the dielectric constant)

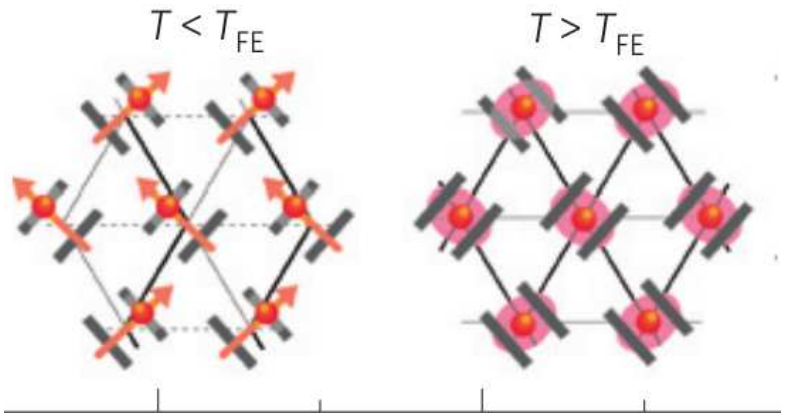
  Lunkenheimer *et al.*, Nature Mat. (2012)

- At the ferroelectric transition the hole per dimer localizes on one molecule

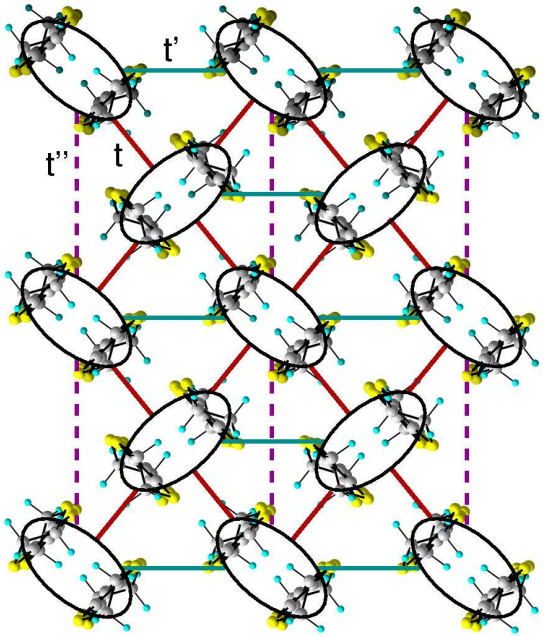
- No spin-driven mechanism for ferroelectricity
  (data do not depend on an external magnetic field)

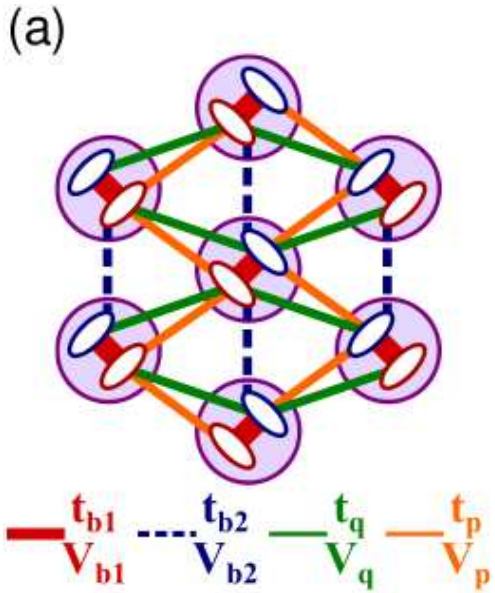
- The critical temperature is similar to the one for magnetic order

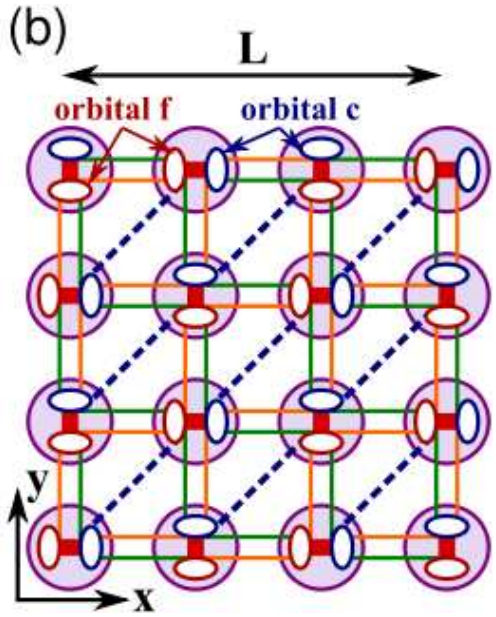
  Shimizu *et al.*, PRL (2003)



An *ad hoc* two-band Hubbard model on the square lattice







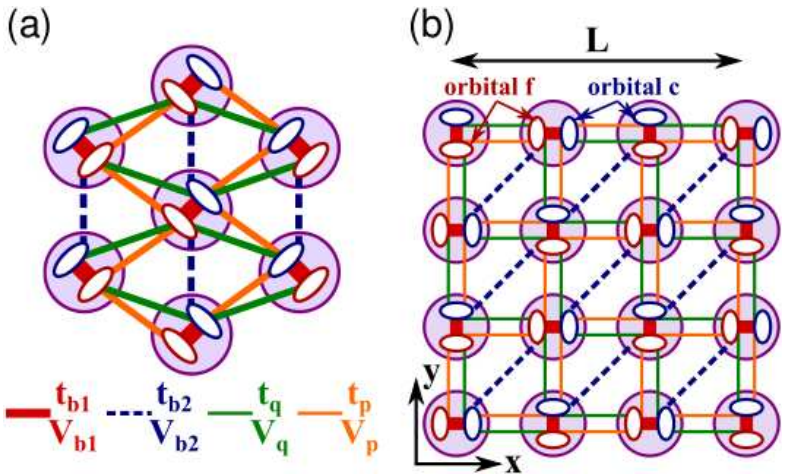
Kino and Fukuyama, JPSJ (1996); Seo, JPSJ (2000)

Hotta, PRB (2010)

Watanabe, Seo, and Yunoki, JPSJ (2017)

Kaneko, Tocchio, Valenti, and Becca, NJP (2017)

An *ad hoc* two-band Hubbard model on the square lattice



- 2 orbitals per site
- 3/4 filling (3 electrons/site)
- No Hund coupling
- $t_{b1} = 1,\ t_{b2} = 0.359,$
  $t_p = 0.539,\ t_q = 0.221$
  For κ-$(ET)_2Cu[N(CN)_2]Cl$

$$\mathcal{H} = \mathcal{H}_t + \mathcal{H}_V + \mathcal{H}_U$$

$$\mathcal{H}_t = t_{b1} \sum_{i,\sigma} c^\dagger_{i,\sigma} f_{i,\sigma} + t_{b2} \sum_{i,\sigma} c^\dagger_{i,\sigma} f_{i+x+y,\sigma} + t_q \sum_{i,\sigma} (c^\dagger_{i,\sigma} f_{i+x,\sigma} + c^\dagger_{i,\sigma} f_{i+y,\sigma})$$

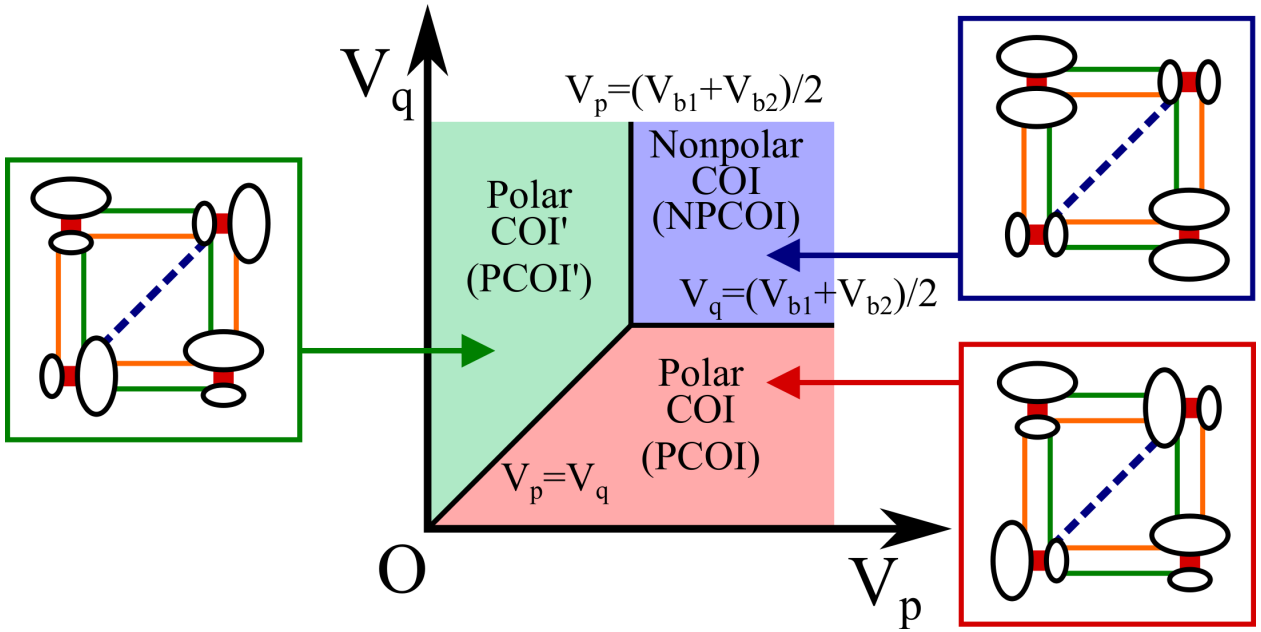
$$+ t_p \sum_{i\in\mathcal{A},\sigma} (c^\dagger_{i,\sigma} c_{i+x,\sigma} + c^\dagger_{i,\sigma} c_{i-y,\sigma} + f^\dagger_{i,\sigma} f_{i-x,\sigma} + f^\dagger_{i,\sigma} f_{i+y,\sigma}) + \text{h.c.}$$

$$\mathcal{H}_V = V_{b1} \sum_i n^c_i n^f_i + V_{b2} \sum_i n^c_i n^f_{i+x+y} + V_q \sum_i (n^c_i n^f_{i+x} + n^c_i n^f_{i+y}) + V_p \sum_{i\in\mathcal{A}} (n^c_i n^c_{i+x} + n^c_i n^c_{i-y} + n^f_i n^f_{i-x} + n^f_i n^f_{i+y})$$

$$\mathcal{H}_U = U \sum_i (n^c_{i,\uparrow} n^c_{i,\downarrow} + n^f_{i,\uparrow} n^f_{i,\downarrow})$$

The atomic limit $t_{b1} = t_{b2} = t_p = t_q = 0$

Look at simple and relevant cases that show regular patterns of charge order



$$
\begin{aligned}
E_{\text{polar}} &= E + V_q \\
E_{\text{polar}'} &= E + V_p \\
E_{\text{nonpolar}} &= E + \frac{1}{2}(V_{b1} + V_{b2})
\end{aligned}
$$

where $E = U + 2V_{b1} + 4V_p + 4V_q + 2V_{b2}$

The variational wave functions

$$|\Psi\rangle = \mathcal{J}|\Psi_0\rangle$$

$$\mathcal{J} = \exp\left(-\frac{1}{2}\sum_{i,j}\sum_{\alpha\beta} v_{i,j}^{\alpha\beta} n_{i,\alpha} n_{j,\beta}\right)$$

$|\Psi_0\rangle$ is the ground state of a non-interacting Hamiltonian with

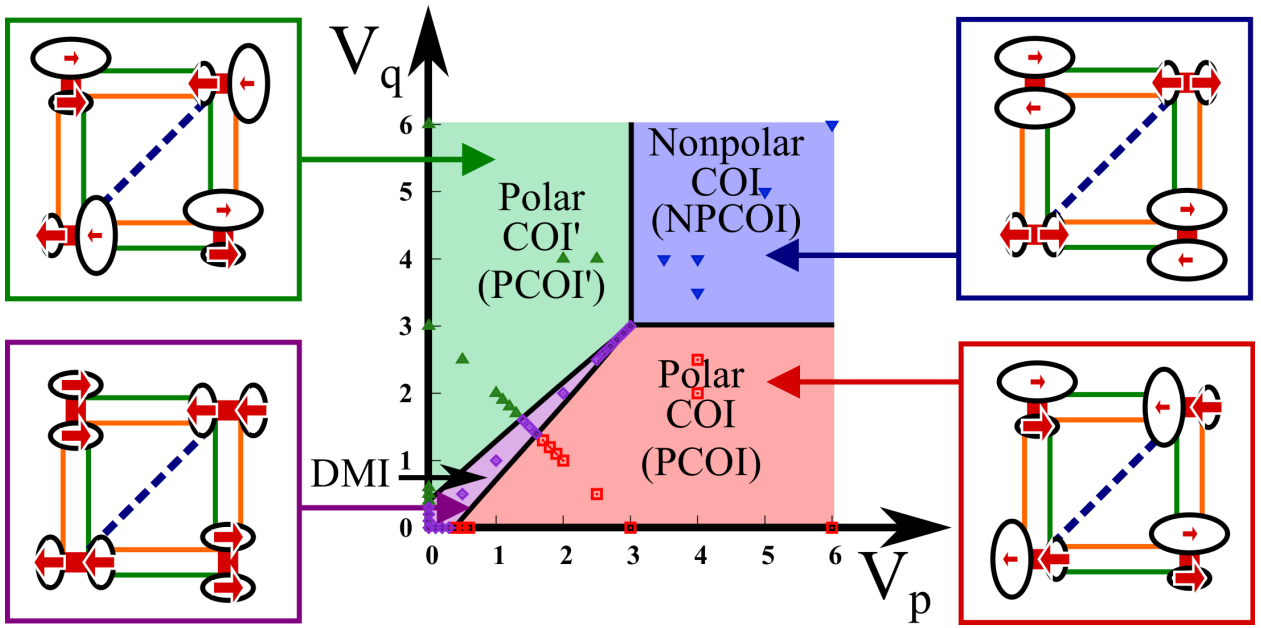
- The kinetic part described by t_{b1}, t_{b2}, t_p, and t_q
- A staggered charge-order pattern
  $\sum_i e^{i\mathbf{Q}\cdot\mathbf{R_i}}(\mu^c n_i^c + \mu^f n_i^f)$
- An antiferromagnetic pattern
  $\sum_i [m_i^c (c_{i,\uparrow}^\dagger c_{i,\downarrow} + c_{i,\downarrow}^\dagger c_{i,\uparrow}) + m_i^f (f_{i,\uparrow}^\dagger f_{i,\downarrow} + f_{i,\downarrow}^\dagger f_{i,\uparrow})]$

$\mathbf{Q} = (\pi, \pi)$ with $\mu^c = \mu^f \Longrightarrow$ the NPCOI
$\mathbf{Q} = (\pi, \pi)$ with $\mu^c = -\mu^f \Longrightarrow$ the PCOI
$\mathbf{Q} = (\mathbf{0}, \mathbf{0})$ with $\mu^c = -\mu^f \Longrightarrow$ the PCOI$'$

$$m_i^\alpha = \begin{cases} m_1^\alpha & \text{if } e^{i\mathbf{Q}\cdot\mathbf{R_i}}\mu^\alpha < 0 \\ m_2^\alpha & \text{if } e^{i\mathbf{Q}\cdot\mathbf{R_i}}\mu^\alpha > 0 \end{cases}$$

Strong-coupling phases

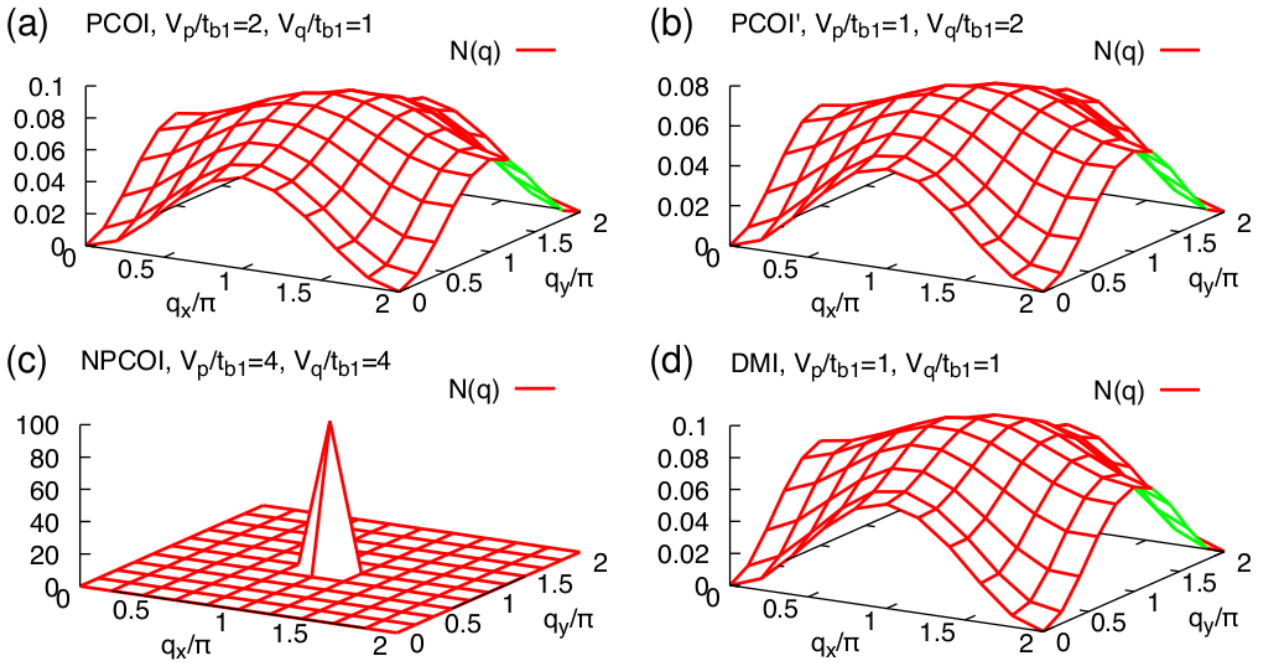
We fix $U/t_{b1} = 10$, $V_{b1}/t_{b1} = 4$, and $V_{b2}/t_{b1} = 2$, and vary V_p and V_q



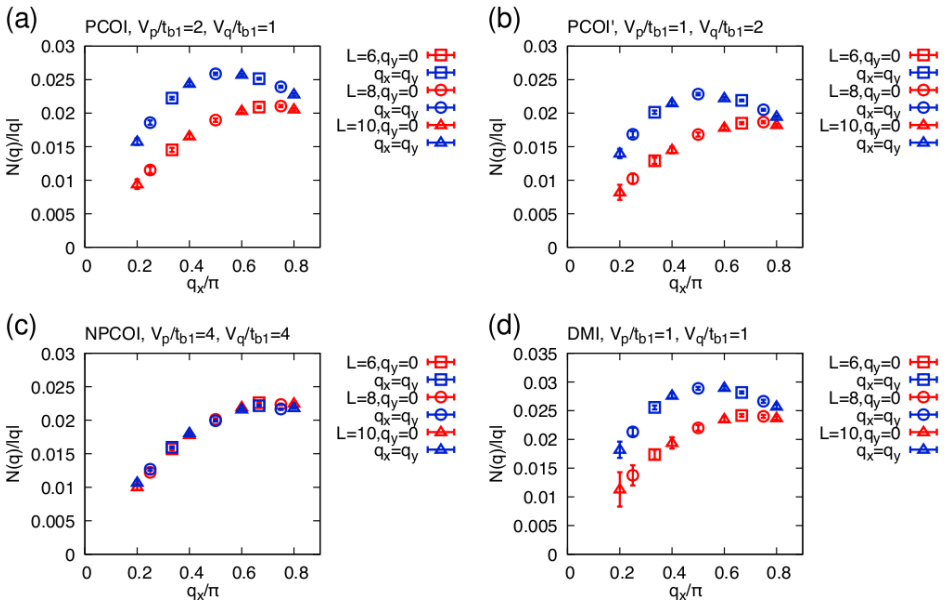
- The dimer-Mott insulator (DMI) intrudes between polar phases
- Polar states acquire ferromagnetic correlations between molecules
- Non-polar state shows antiferromagnetic correlations between molecules

Charge correlations

$$N(q) = \frac{1}{N_s} \sum_{i,j} \langle (n_i^c + n_i^f)(n_j^c + n_j^f) \rangle e^{i\mathbf{q}\cdot(\mathbf{R_i}-\mathbf{R_j})}$$

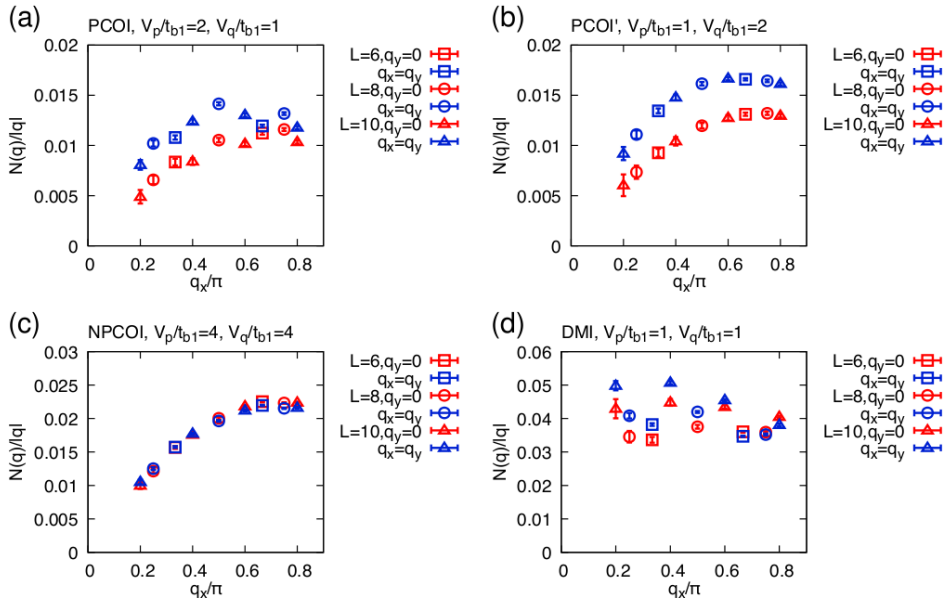


Charge order without antiferromagnetism



Full variational wave function with AF order

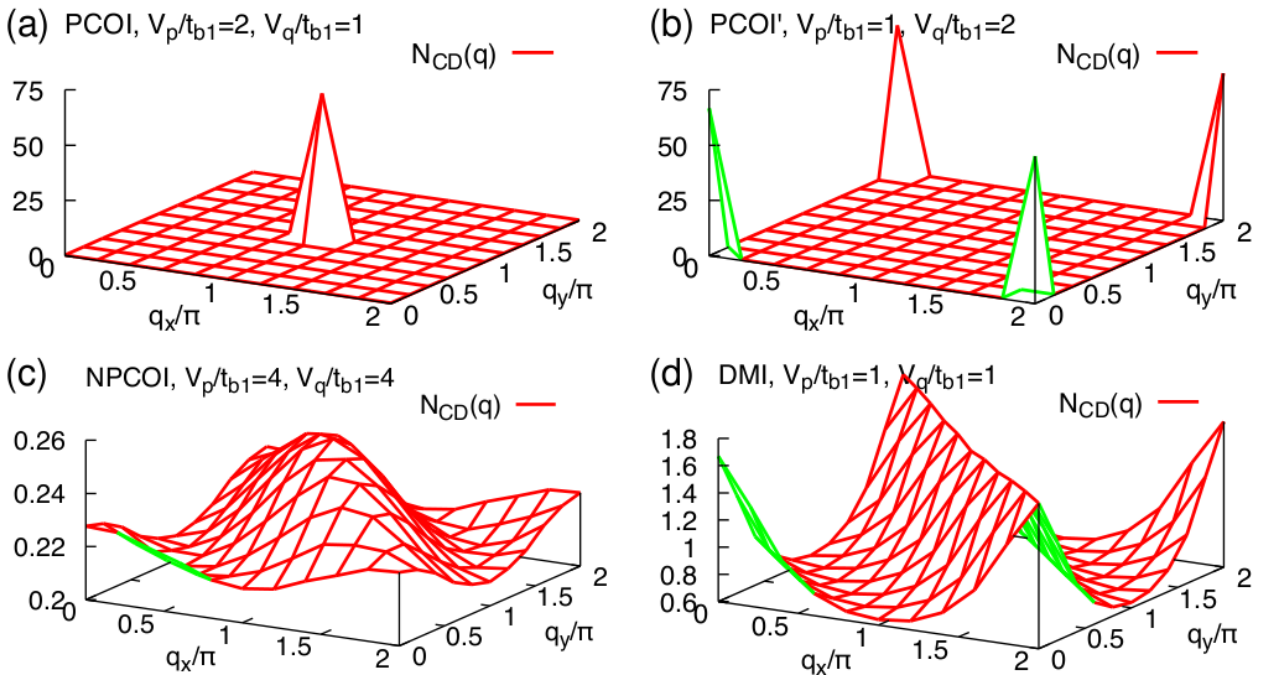
Our results suggest that charge order is not driven by magnetism



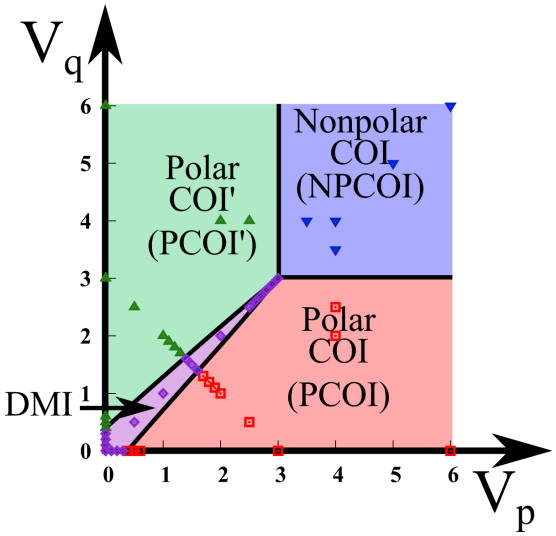
Variational wave function without AF order
(imposing $m_i^\alpha = 0$)

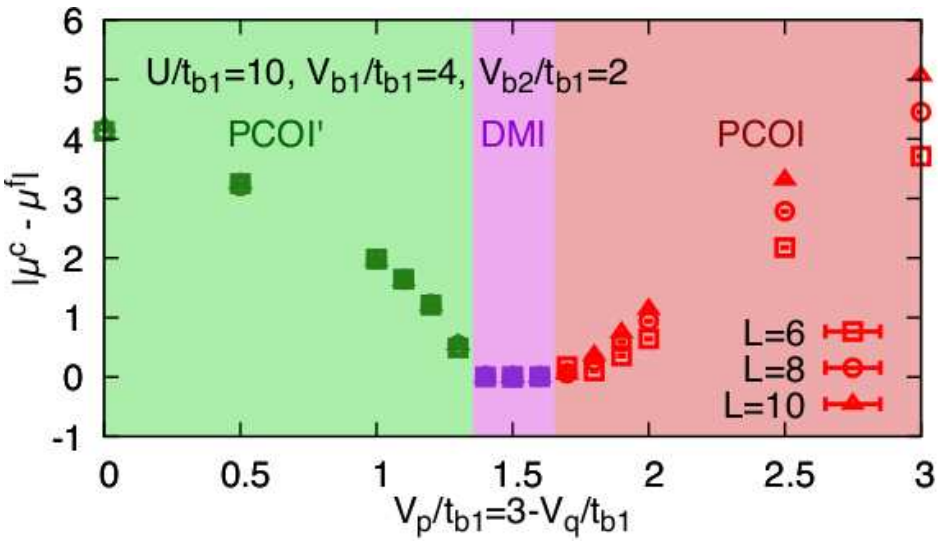
Charge correlations

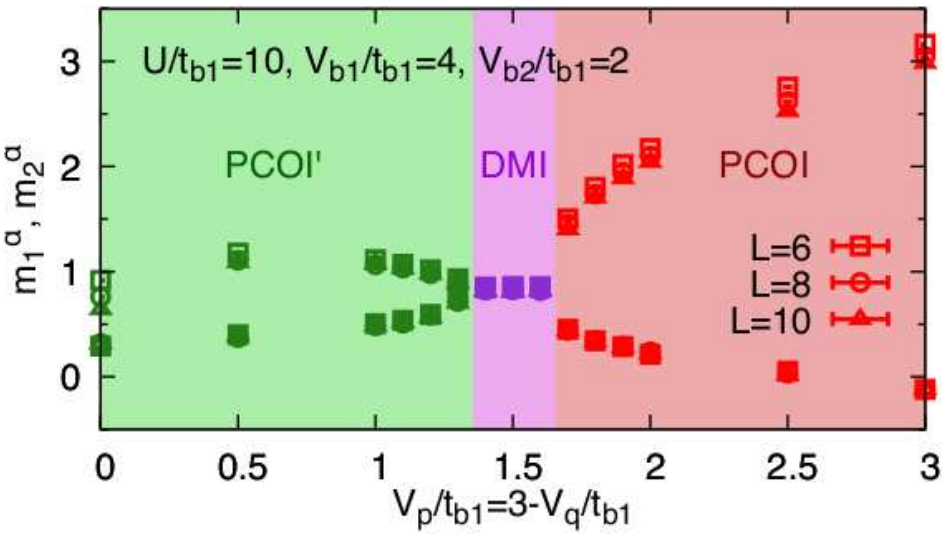
$$N_{\mathrm{CD}}(q) = \frac{1}{N_s} \sum_{i,j} \langle (n_i^c - n_i^f)(n_j^c - n_j^f) \rangle e^{i\mathbf{q}\cdot(\mathbf{R_i}-\mathbf{R_j})}$$



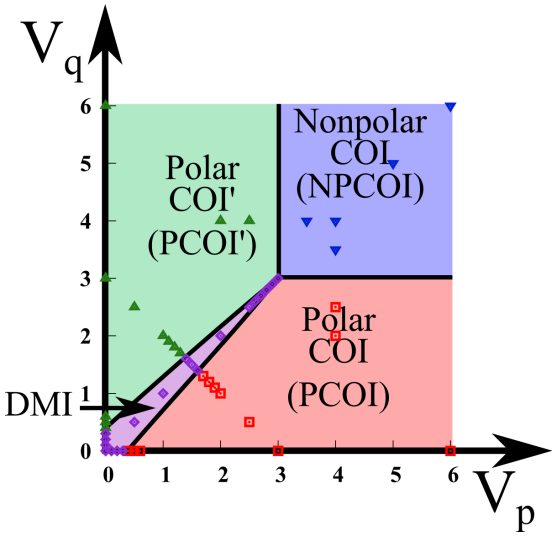
Phase transitions: variational parameters

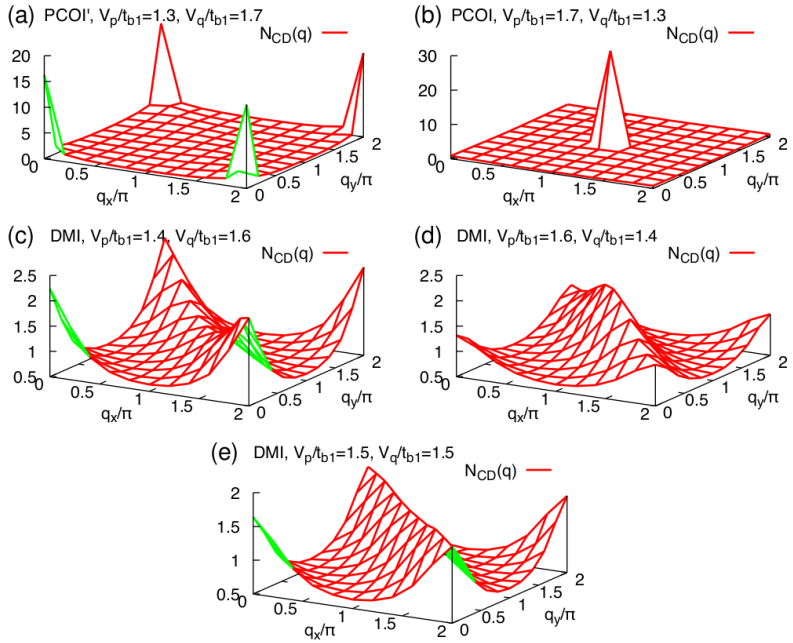


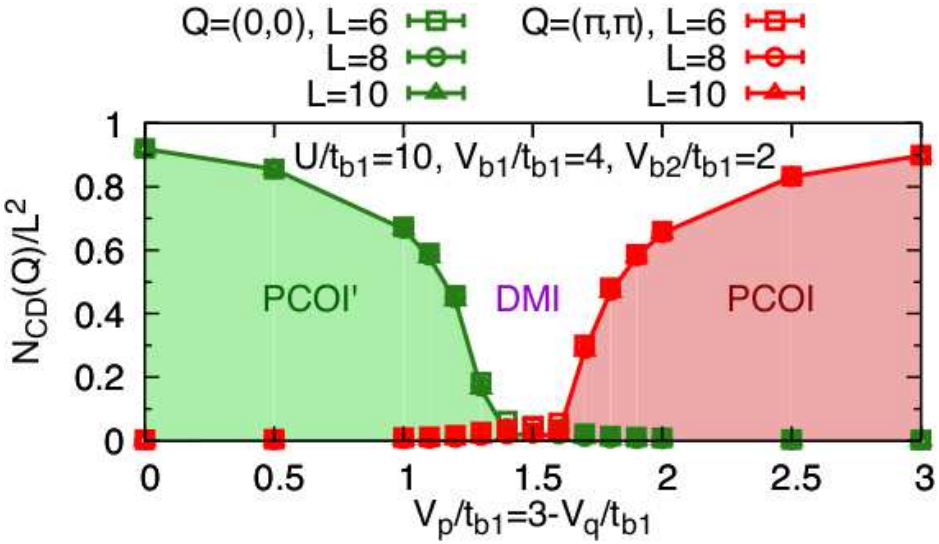




Phase transitions: correlations







Conclusions

Jastrow-Slater wave functions can be easily defined in multiband models

- They can be easily treated within Monte Carlo sampling
- They can be flexibly parametrized in order to reproduce different phases:
  Metals and superconductors
  charge/spin ordered states
  Pure Mott insulators
  ... more exotic states (with orbital order, currents)

Initial benchmarks are promising

- The OSMT is observed within a two-band model
- The Mott transitions in isotropic models are obtained
  (and triplet superconductivity for $J > 0$)
- Polar and nonpolar insulators are found in a two-band model