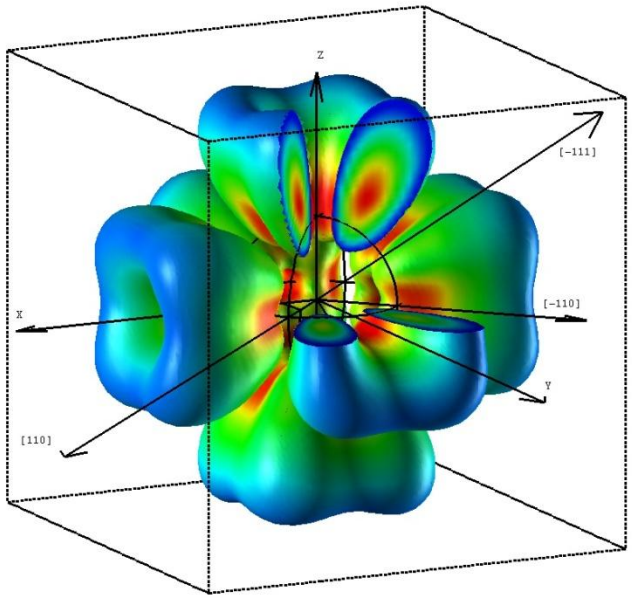
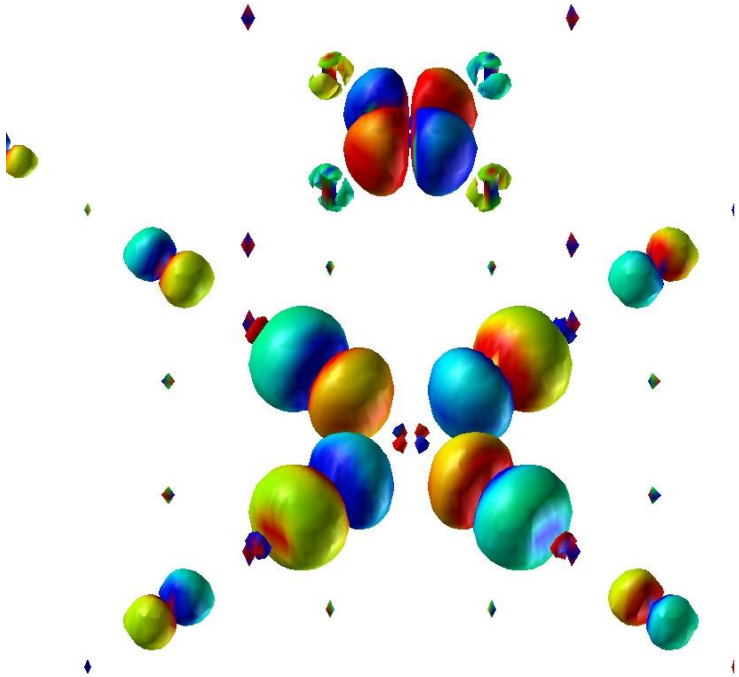
Local Excitations in Strongly Correlated Systems and Their Propagation





Wei Ku

Brookhaven National Lab & SUNY Stony Brook

Acknowledgement

Funding sources

- Basic Energy Science, Department of Energy
- DOE-CMSN
- Taiwanese Student Fellowship

Theoretical works

- Weiguo Yin (BNL)
- Chi-Cheng Lee (BNL & Tamkang Univ.) ← about to move on
- Chen-Lin Yeh (BNL & Tamkang Univ.)
- Hung-Chung Hsueh (Tamkang Univ.)

Experiments

- Ben C. Larson (ORNL)
- Igor Zaliznyak (BNL)
- Peter Abbamonte (UIUC)

Discussions

- George Sawatzky (UBC)
- Adolfo G. Eguiluz (UT-Knoxville / ORNL)

Outline

- Strong correlation in crystals: intra-atomic vs. inter-atomic
  - defining representative local unit
- Energy reduction: many-body down-folding
  - RG: energy-dependent perspective of physics
  - a proper derivation of low-energy physics
- QP excitations: ARPES and Fermi surface
- Length reduction in simulation: flavored twisted boundary condition
  - circumventing the finite size effects
- P-H excitations of large momentum: NIXS
  - Momentum transfer: anisotropy & local orbitals
  - Spectrum: perturbation, TD-LDA+*U* & beyond
  - Propagation in space time: effective hopping kernel

Strong correlation in crystals

- Large intra-atomic interaction
  - → atomic multiplets beyond single determinant
- + large short-range kinetic effects
  - → beyond atomic multiplets
  - → strong short-range correlation
- + short-range inter-atomic interaction
  - → stronger short-range correlation
- No well-defined systematic framework
  - → many different approximate approaches
  - → qualitative description already difficult
- Main interest: phase transitions at low-*T* and/or high P
- Opportunity for 1st-principles methods: material- and *P*-dependence

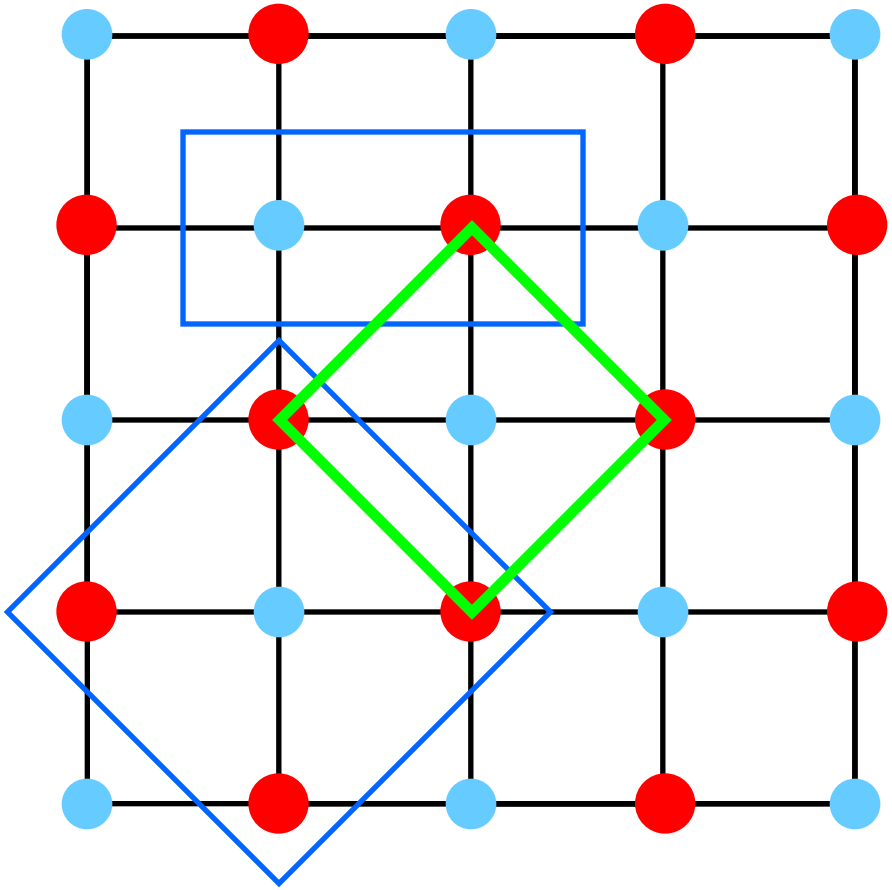
Local Picture for Strongly Correlated Systems

$$H = H_0 + V = H_{local} + H_{nonlocal}$$

- V too big for perturbation
- Maximize the terms in the "local" part

  → symmetric Wannier Representation → defines "local"
- Treat local part "accurately" and leave non-local part as modification

How to define “local” in CT-insulators?

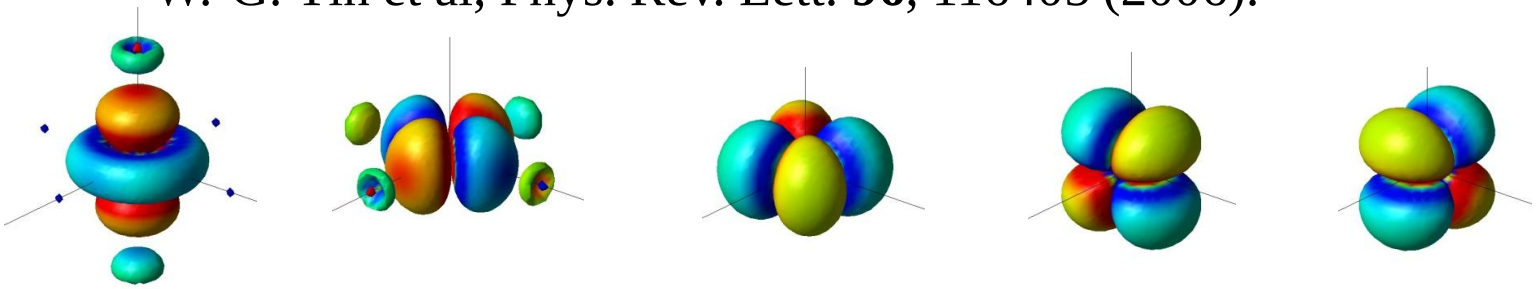


- Periodic symmetry
- Point group symmetry
- Simultaneously keep both? How to split the Hilbert space?

Symmetric Wannier Functions for CT-Insulators

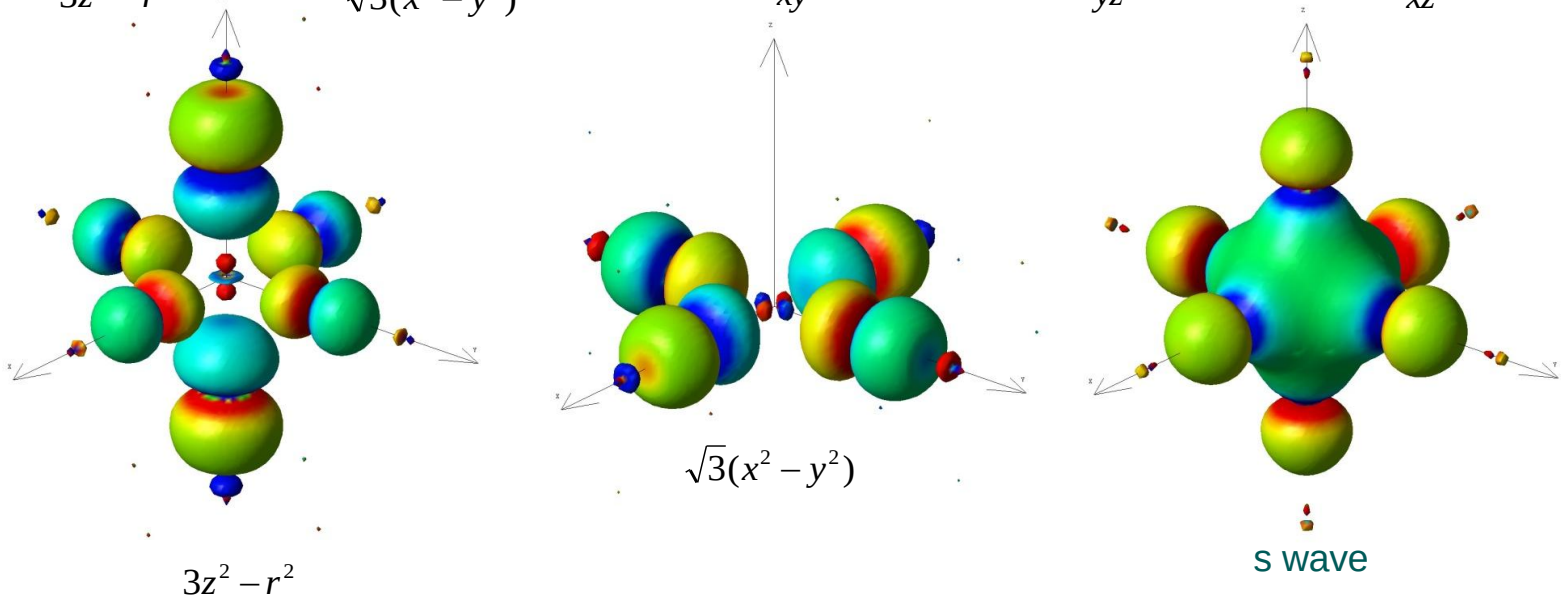
W. Ku et al., Phys. Rev. Lett. **89**, 167204 (2002).
R. L. Barnett et al., Phys. Rev. Lett. **96**, 026406 (2006).
W.-G. Yin et al, Phys. Rev. Lett. **96**, 116405 (2006).

Ni



$3z^2 - r^2$ $\sqrt{3}(x^2 - y^2)$ xy yz xz

O

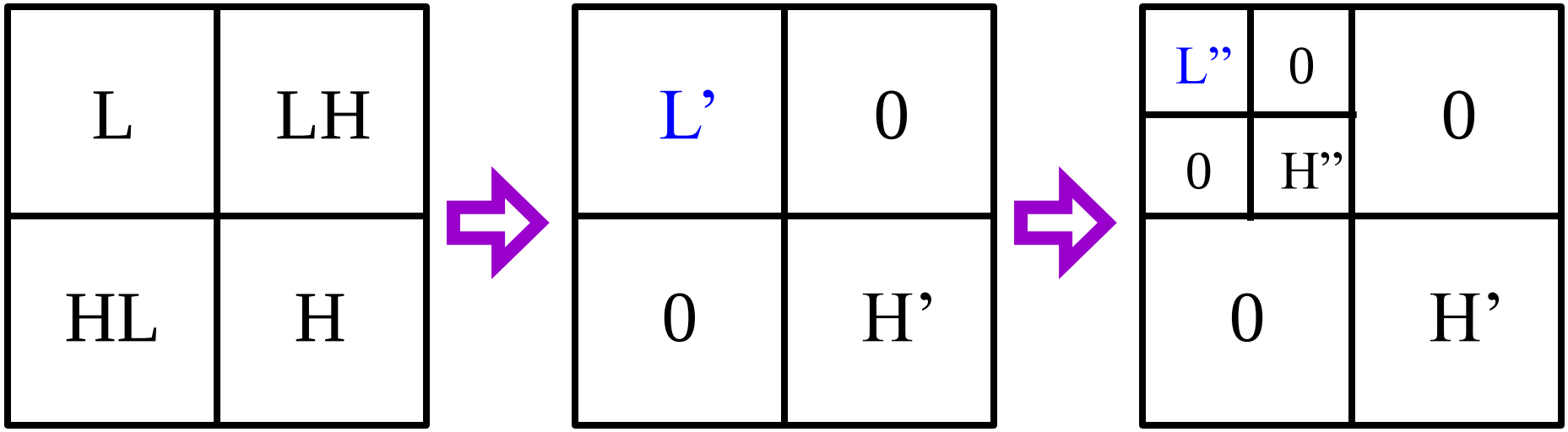


$3z^2 - r^2$ $\sqrt{3}(x^2 - y^2)$ s wave

- O-*p* orbitals → additional Ni-*d* orbitals (no double counting of O orbitals)
- "local" is now defined by this "super-atom"
- Maximize the H^{local}

Energy reduction: RG perspective

- A simple "matrix view" of RG: block diagonalization of H_L and H_H



- Many-body Hamiltonian changes at different energy scale
  - → dominant physics varies at different energy scale
  - → needs a multi-resolution procedure to lower the energy scale

Many-Body Down-Folding

- A three-step procedure

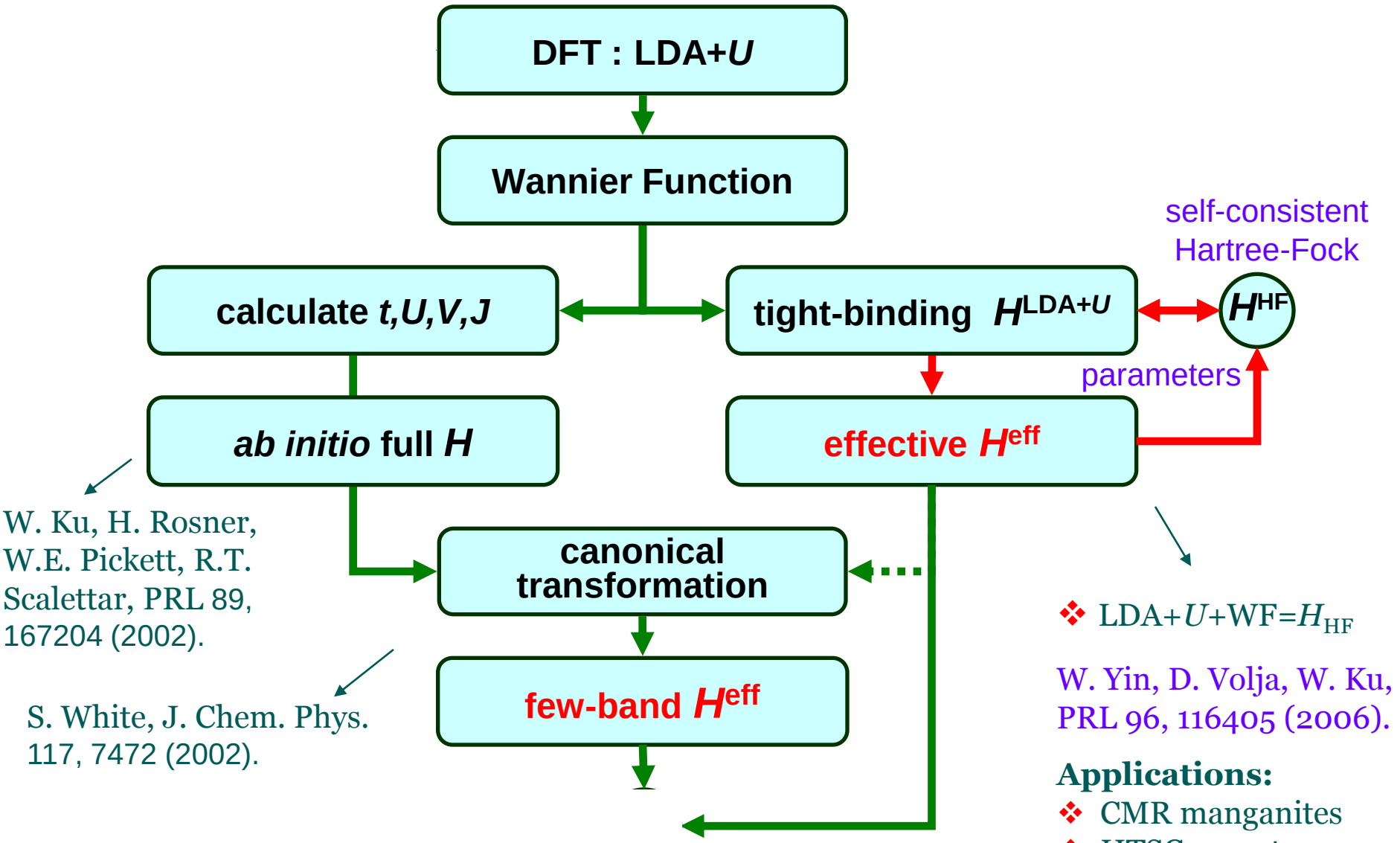
  Full energy → ~10 eV → ~1 eV
- Full energy: LDA+U or hybrid functional of insulating phase

  W. Ku et al., Phys. Rev. Lett. **89**, 167204 (2002).
- ~10 eV: matching interacting Hamiltonian

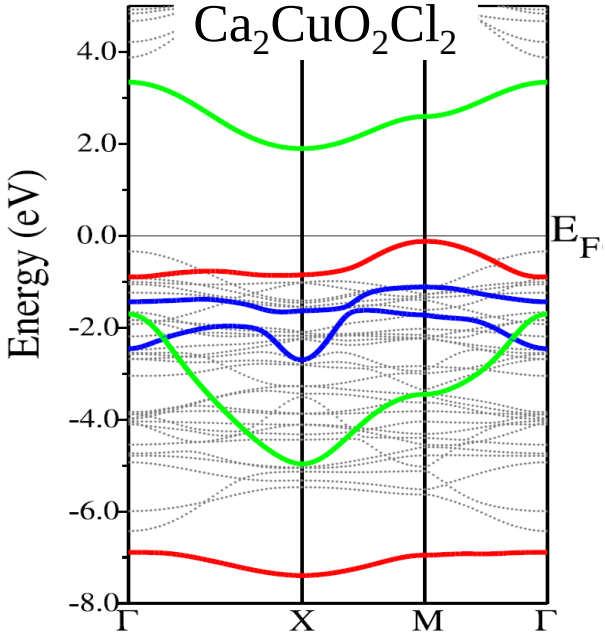
  W.-G. Yin et al, Phys. Rev. Lett. **96**, 116405 (2006).
- ~1 eV: exact diagonalization + canonical transformation

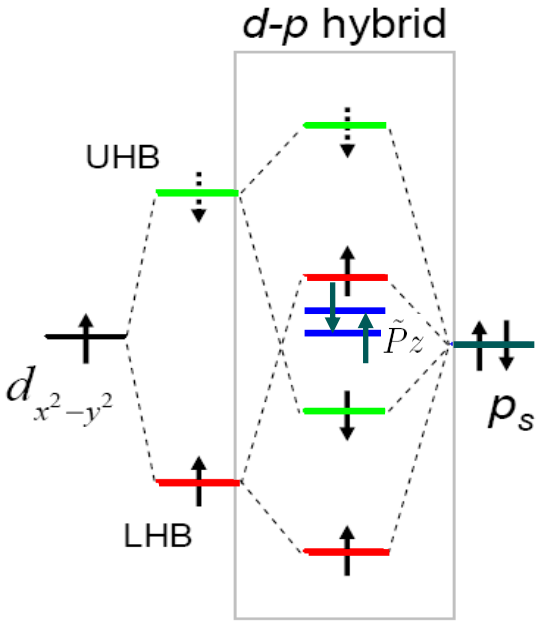
  W.G. Yin and W. Ku, Phys. Rev. B **79**, 214512 (2009).
  - applicable for large doping range
  - visualization of new interactions

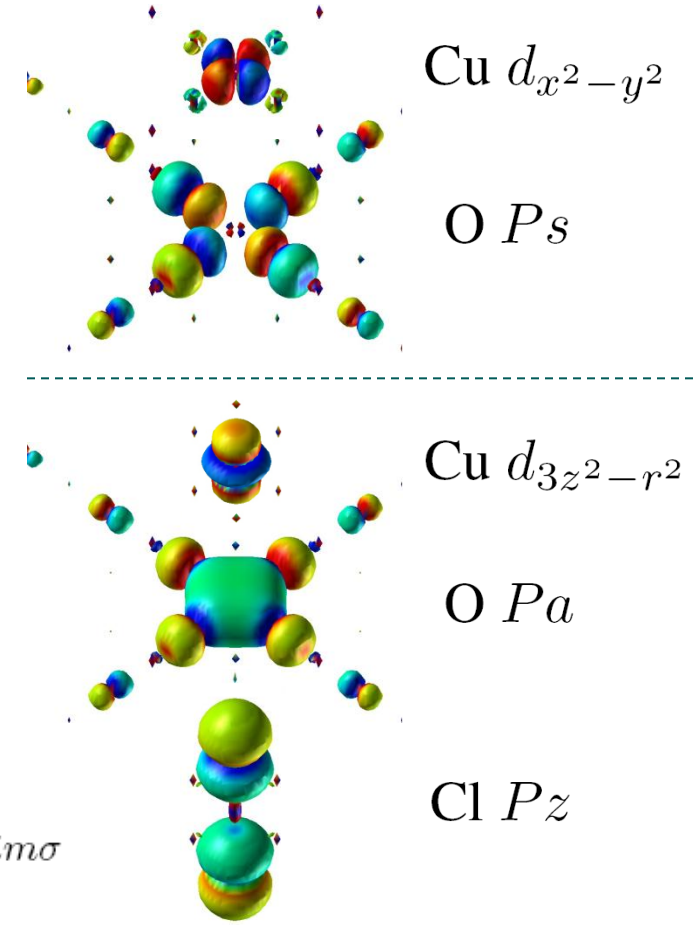
Many-body down-folding



~10 eV: LDA+U → 5-band Hamiltonian







$$H^{5b} = \sum_{ijmm'\sigma} (T_{ij}^{mm'} C_{im\sigma}^{\dagger} C_{jm'\sigma} + H.c.) + \sum_{im\sigma} \varepsilon_m n_{im\sigma}$$
$$+ U_{\text{eff}} \sum_{i,m=1,2} n_{im\uparrow} n_{im\downarrow} + U'_{\text{eff}} \sum_{i\sigma\sigma'} n_{i1\sigma} n_{i2\sigma'} - J_{\text{eff}} \sum_{i\sigma\sigma'} C_{i1\sigma}^{\dagger} C_{i1\sigma'} C_{i2\sigma'}^{\dagger} C_{i2\sigma}$$

- ε_{P_Z} is the most significant material-dependent

~1 eV: effective one-band Hamiltonian

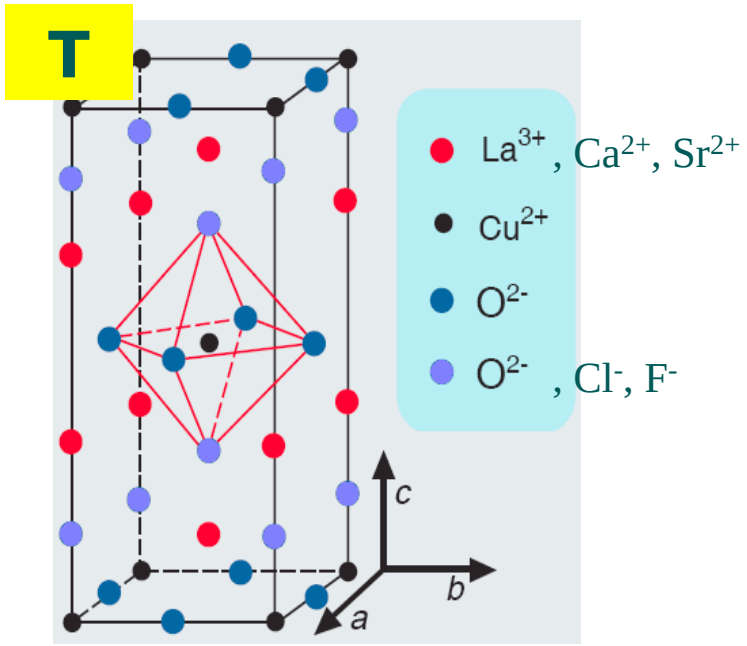
- Numerical canonical transformation:
- Solve local problem exactly
- Include non-local terms in the basis of local multiplets:

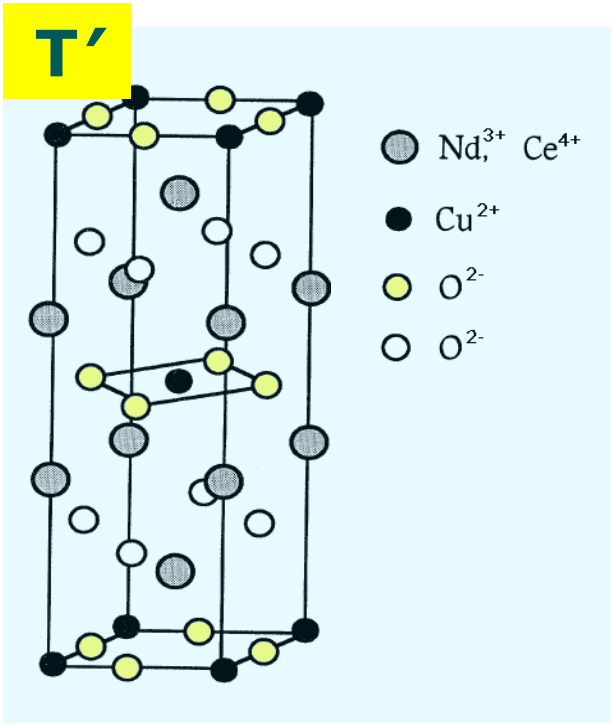
  Hubbard X-operator

$$H^{1b} = -\sum_{ij\sigma} t_{ij}(\tilde{c}^{\dagger}_{i\sigma}\tilde{c}_{j\sigma} + H.c.) + J\sum_{\langle ij\rangle}(\vec{S}_i\cdot\vec{S}_j - \frac{n_i n_j}{4})$$
$$+ \frac{J}{4}\sum_{\langle ijk\rangle\sigma}(\tilde{c}^{\dagger}_{i\sigma}\tilde{c}^{\dagger}_{j\bar{\sigma}}\tilde{c}_{j\sigma}\tilde{c}_{k\bar{\sigma}} - \tilde{c}^{\dagger}_{i\sigma}n_{j\bar{\sigma}}\tilde{c}_{k\sigma} + H.c.)$$
$$+ \sum_{\langle ij\rangle} V_{ij}n_i n_j + \sum_i \mu_i n_i + \sum_{\langle ij\rangle}\varepsilon_{ij},$$

- t', t'', V_{ij}, μ_i, ε_{ij} strongly depend on ε_{Pz}
- “super-exchange” J & “super-replusion” V_{ij}

Material dependence of prototypical cuprates

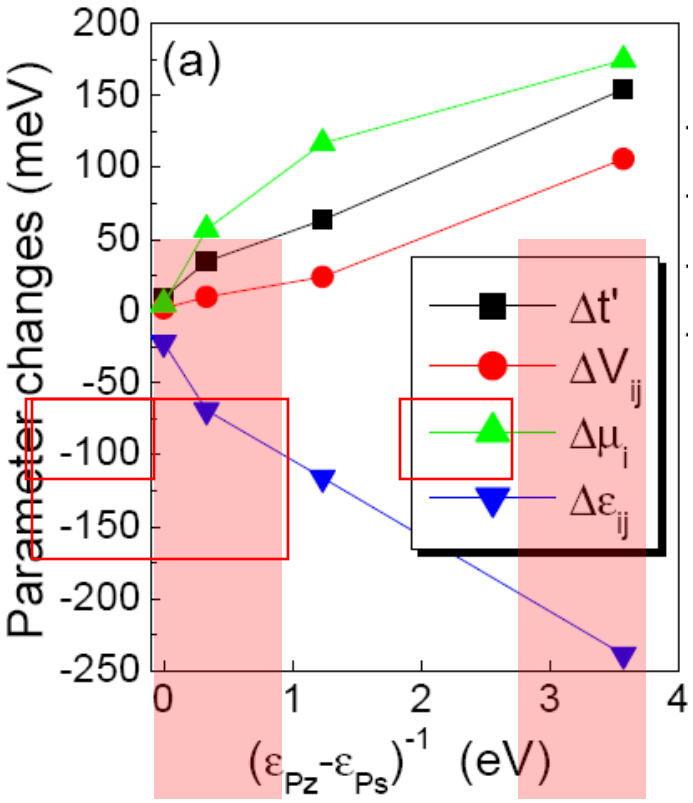




- La_2CuO_4 apical O
- $Ca_2CuO_2Cl_2$ apical Cl
- $Sr_2CuO_2F_2$ apical F
- Nd_2CuO_4 no apical atoms

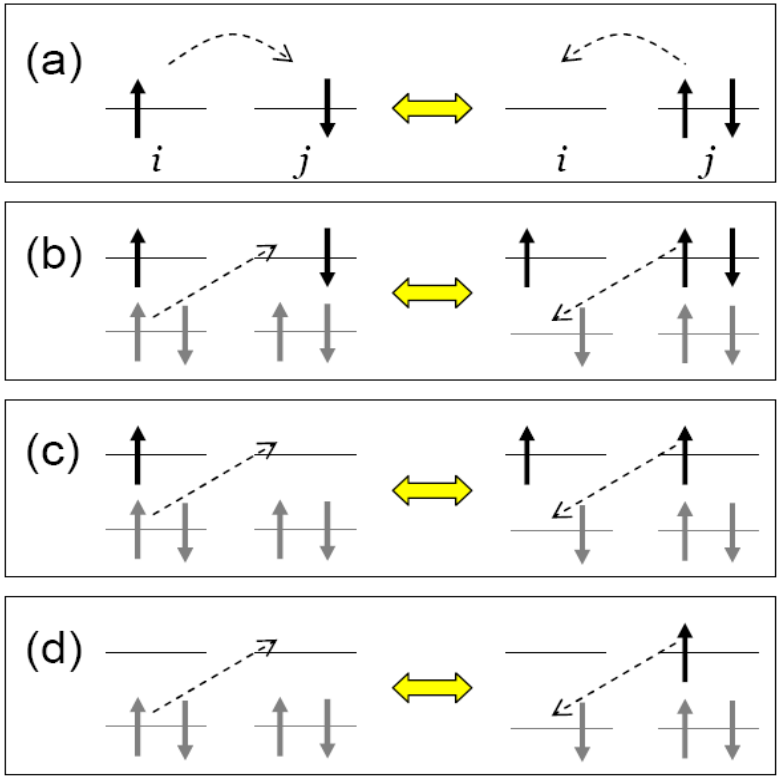
One-Band Parameters

	Nd_2CuO_4	$Sr_2CuO_2F_2$	$Ca_2CuO_2Cl_2$	La_2CuO_4
$T_c^{\max}$ (K)	N/A	46		
$\varepsilon_{Pz} - \varepsilon_{Ps}$	∞	3010		
$J_{\mathrm{LDA}+U}{}^b$	131	155		
$J/2$	138 (138)	138 (138)		
t	431 (431)	467 (467)		
$t'/\lvert t\rvert$	-0.33 (-0.35)	-0.25 (-0.32)		
$t''/\lvert t\rvert$	0.23 (0.24)	0.19 (0.23)		
V_{ij}	29 (27)	39 (29)		
μ_i	-795 (-800)	-337 (-394)		
ε_{ij}	-36 (-14)	-86 (-17)		



- Universality in contributions from the b_1 orbitals (Cu x^2-y^2, O Ps) → *t* and *J*.
- Contributions from the a_1 orbitals (Cu $3z^2$-r^2, O Pa, apical Pz) : Significant
  - Small from Cu z^2 and O *Pa*
  - Material-dependence mainly from the apical *Pz* orbital.
  - LDA's problem of Fermi surface of $Ca_2CO_2Cl_2$ fixed ! Tanaka et al., PRB (2004).

Super-exchange & super-repulsion



$-\frac{J}{2} n_{i\uparrow} n_{j\downarrow}$ super-exchange

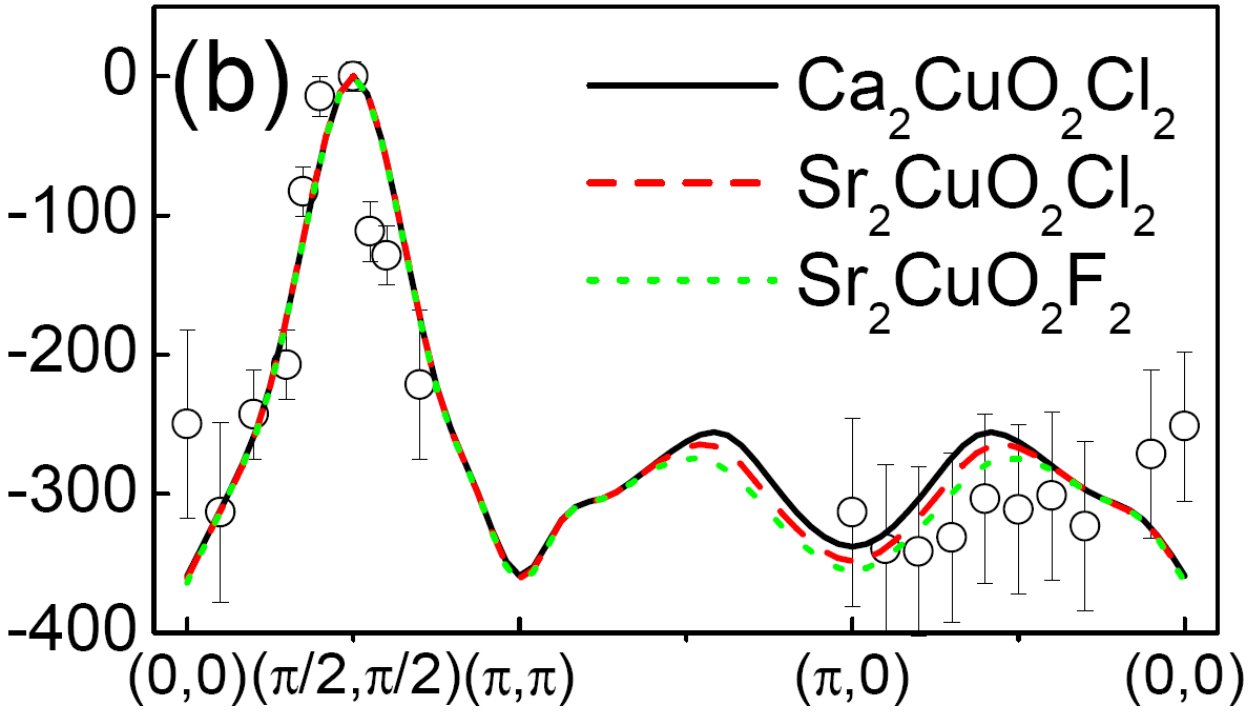
$-v_{11}\ n_i n_j$ e-e attraction

$-v_{10}\ n_i(1-n_j)$ e-h attraction

$-v_{00}\ (1-n_i)(1-n_j)$ h-h attraction

- "Super-repulsion" $V_{ij} = 2v_{10} - v_{00} - v_{11}$
- Site potential $\mu_i = 4(v_{00} - v_{10})$
- "Super-repulsion" != direct Coulomb:
  - controlled by p_z & harder to screen

Quasi-particle dispersion



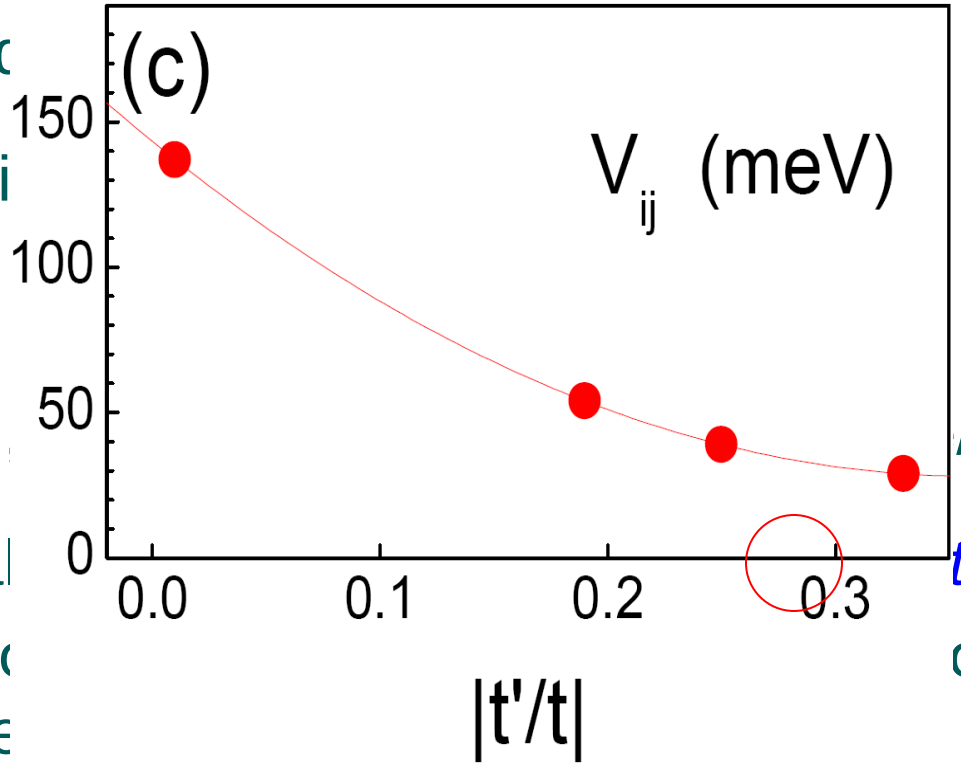
- low hole concentration: self-consistent Born approximation.
  Yin *et al.*, PRL (1998).
- exp.: Wells *et al.*, PRL (1995) on $Sr_2CuO_2Cl_2$.
- Significant mass enhancement obtained!

Implications

- Super-repulsion V_{ij} weakens local *d*-wave pairing.
  - Anticorrelation between V_{ij} and $|t'/t|$ → $T_c^{max} \sim |t'/t|$
- Charge c
  - μ_i → i
  - V_{ij} →

  (SI-STM /strain, …)
- New rea t'' , μ_i
  - Phon ormed pairs → phase
- Examining high-T_c SC in multilayer systems:

  Layer-dependent influence of the apical *Pz* orbital.

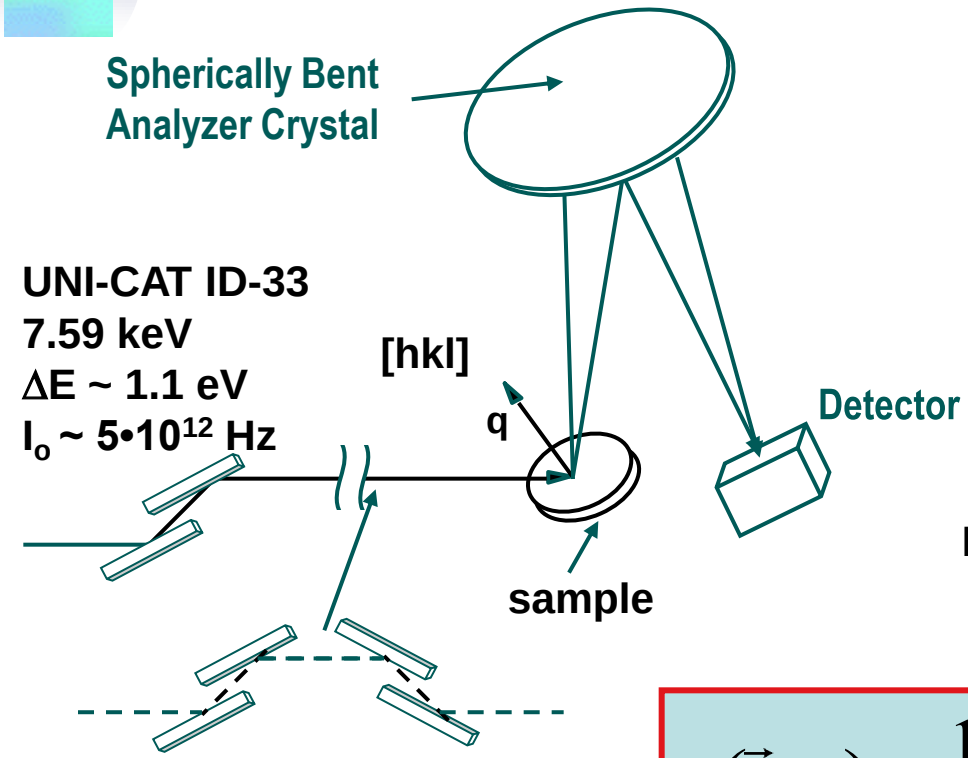


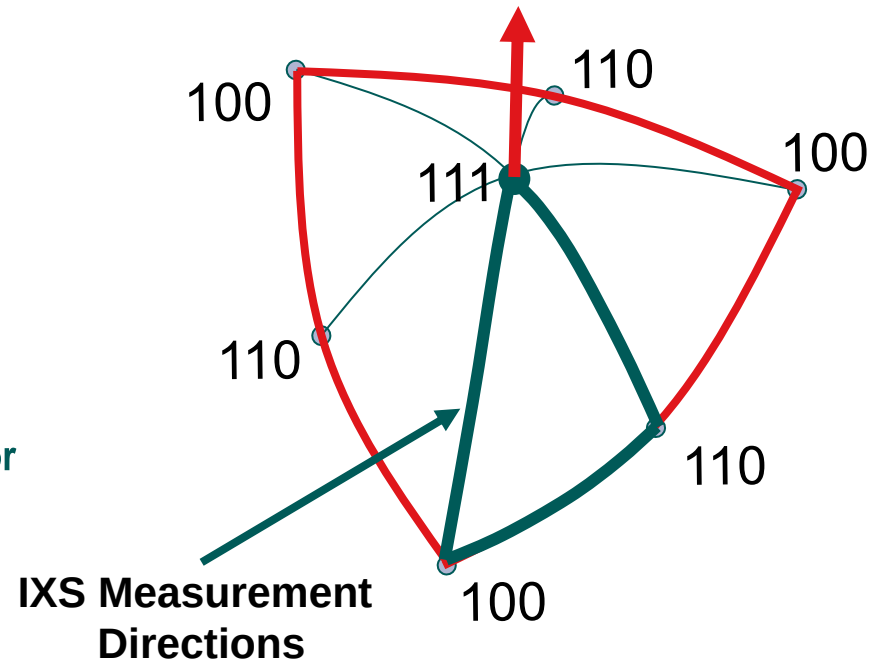
Flavored twisted boundary condition

- Fluctuation at length scale longer than the size of computation
  → Finite-size effects
- Treating a large MB system as a collection of smaller systems that couple to each other: rigorous derivation
- approximate the coupling between the small systems
  → flavored twisted B.C.

  W.-G. Yin and W. Ku, Phys. Rev. B **80**, 180402 (2009).
  → twisted B.C.
  → periodic B.C.
- ex: bond energy of Heisenberg model → only needs two sites!

Non-Resonant Inelastic X-Ray Scattering (NIXS)





$$s(\vec{q},\omega) = \frac{1}{V} S(\vec{q},\omega) = -2\hbar \,\mathrm{Im}\,\chi_{\vec{G}\vec{G}'}(\vec{q}-\vec{G},\omega)$$

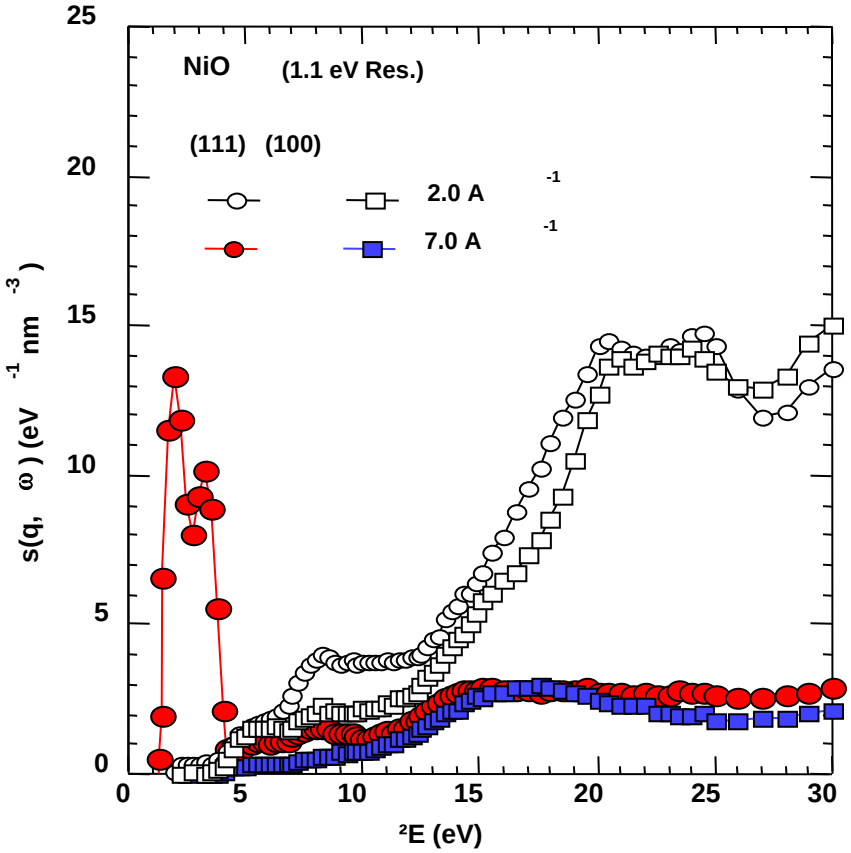
Absolute IXS Measurements

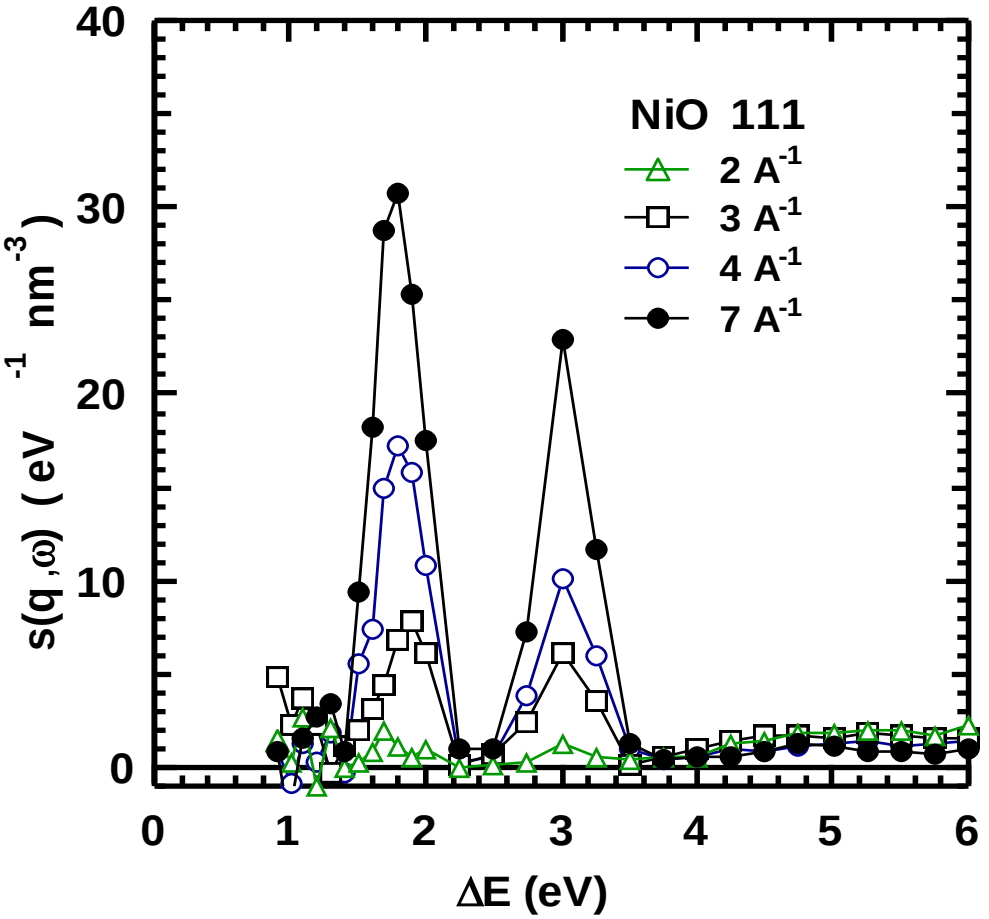
$$\frac{d^2\sigma}{d\Omega d\omega} =: r_0^2 \left(\vec{e}_i \cdot \vec{e}_f\right)^2 \left(\frac{\omega_f}{\omega_i}\right) S(\vec{q},\omega)$$

Absolute Response Calculations

$$S(\vec{q},\omega) = -2\hbar V \,\mathrm{Im}\,\chi_{\vec{G},\vec{G}}(\vec{q}-\vec{G},\omega)$$

Local Excitations in the Mott Gap in NiO



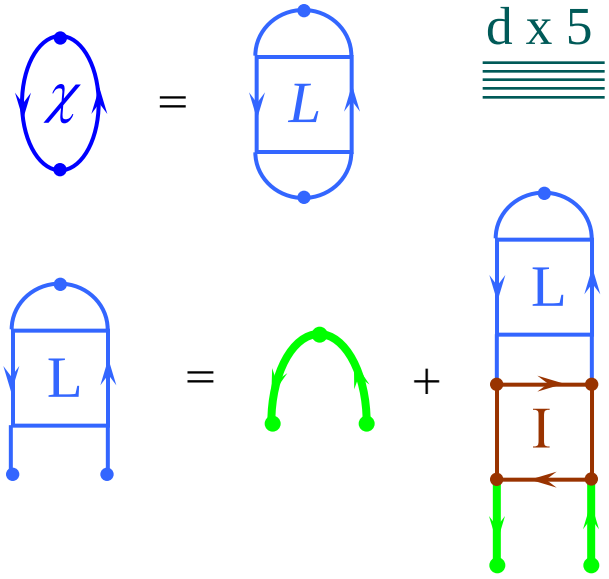


- New features at large *q* in the Mott gap!
- Short wave length → local excitations
- Strong angular dependence: (100) != (111)

B. C. Larson et al.,PRL 2007

q-dependence of Localized Excitons

Energy-resolved Wannier states

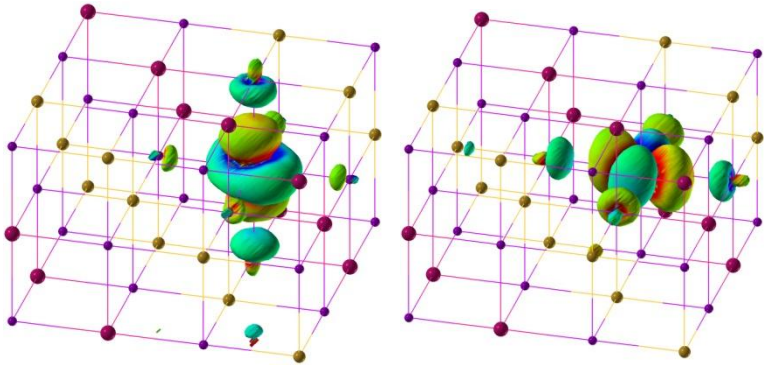


d x 5

e_g x 2

$e_g\ 1 \propto 3z^2 - r^2$

$e_g\ 2 \propto \sqrt{3}\left(x^2 - y^2\right)$

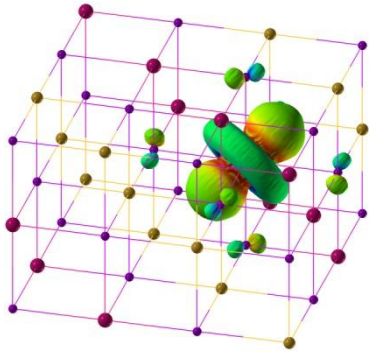


E_F of NiO

t_{2g} x 3

a_{1g} x 1

$a_{1g} : 2\left(yz + zx + xy\right)$

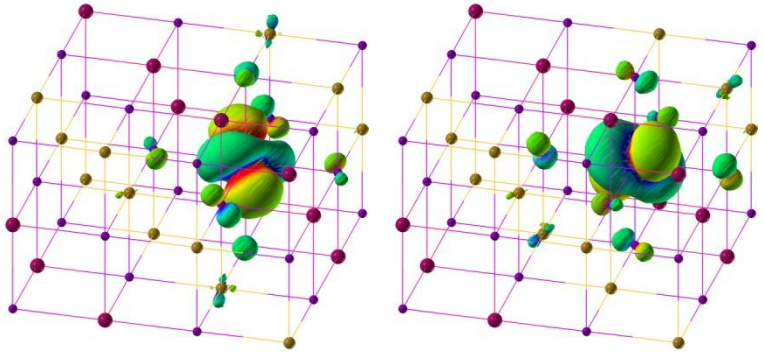


E_F of CoO

e'_g x 2

$e'_g\ 1 \propto \left(1+\sqrt{3}\right)yz + \left(1-\sqrt{3}\right)zx - 2xy$

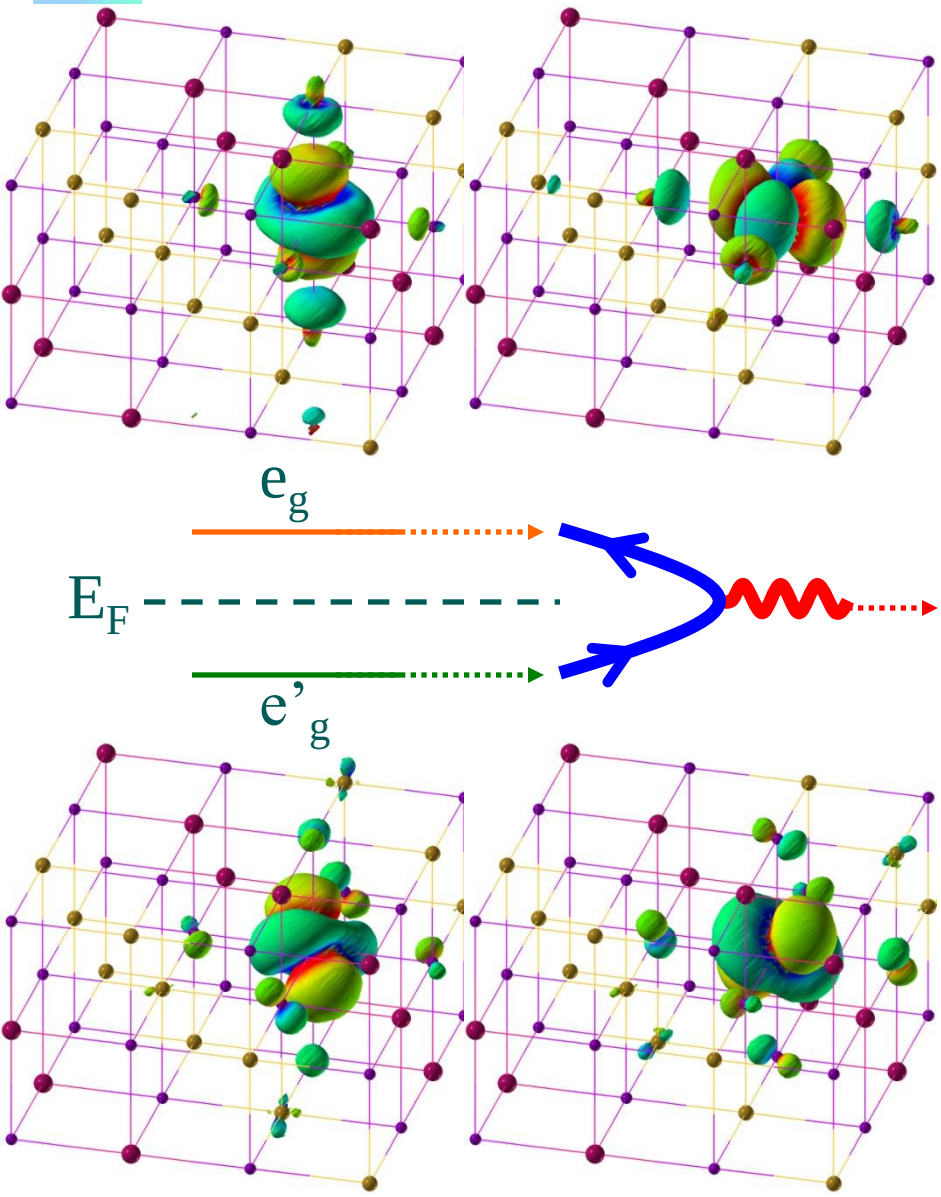
$e'_g\ 2 \propto \left(1-\sqrt{3}\right)yz + \left(1+\sqrt{3}\right)zx - 2xy$



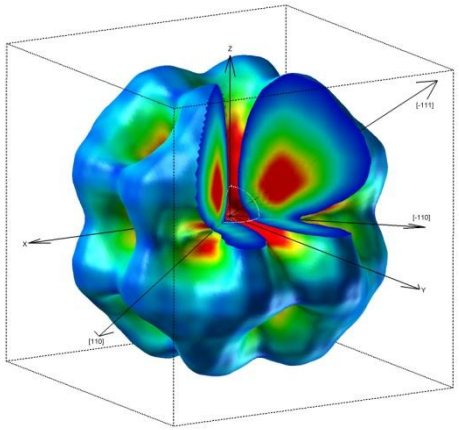
$$\chi\left(\mathbf{q},\mathbf{q}';w\right) = \sum_{11'} \left\langle 1\left| e^{-i\mathbf{q}\cdot\hat{\mathbf{x}}} \right| 1' \right\rangle \left\langle 1'\left| e^{i\mathbf{q}'\cdot\hat{\mathbf{x}}} \right| 1 \right\rangle L\left(1,1';1',1;w\right)$$

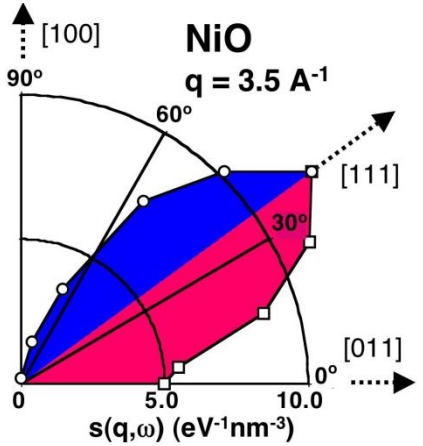
Local Excitations in NoO and CoO

Point group symmetry and new selection rules

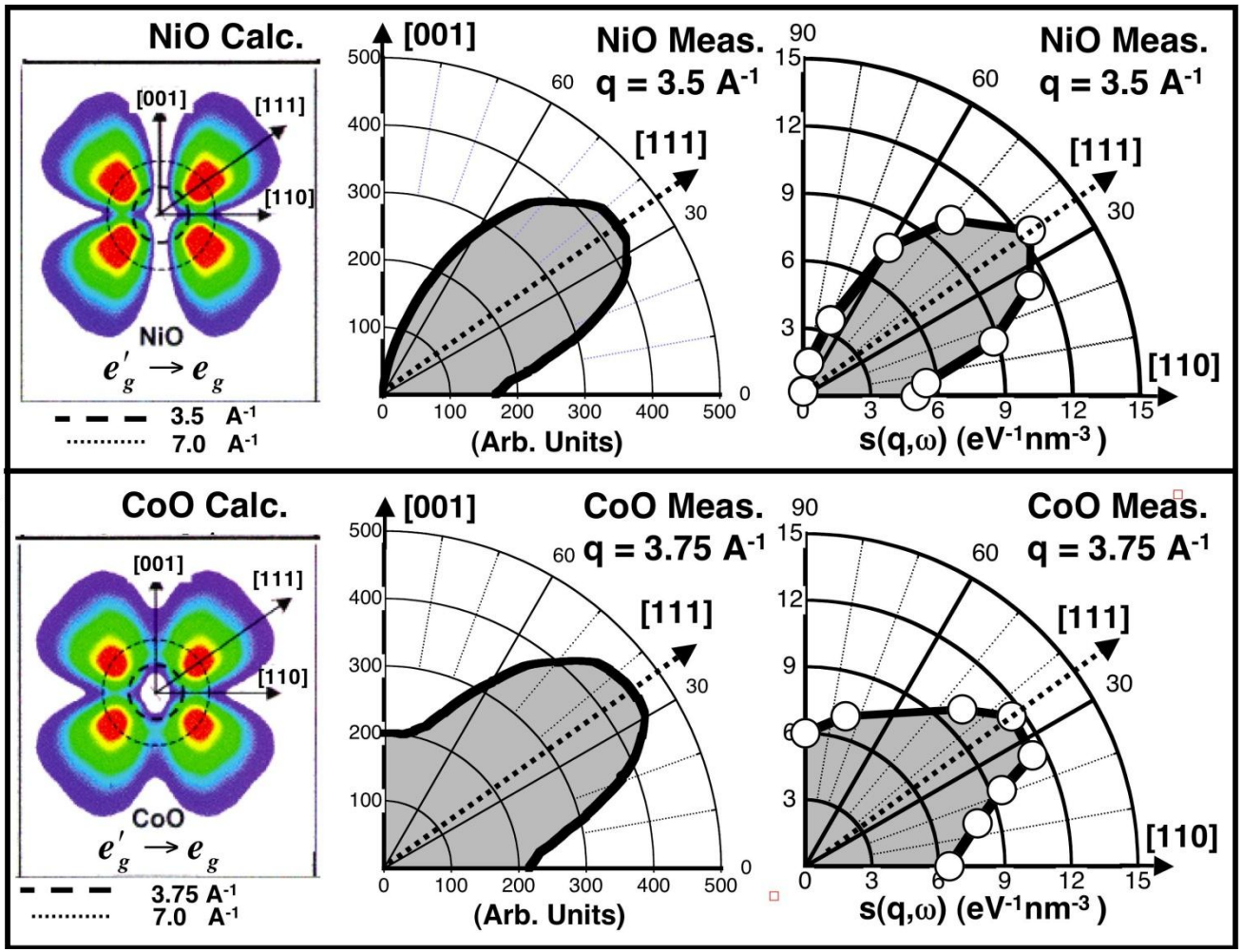


- Local cubic point group symmetry
  - → nodal directions
  - → new selection rules





Angular Dependence of Local Excitations

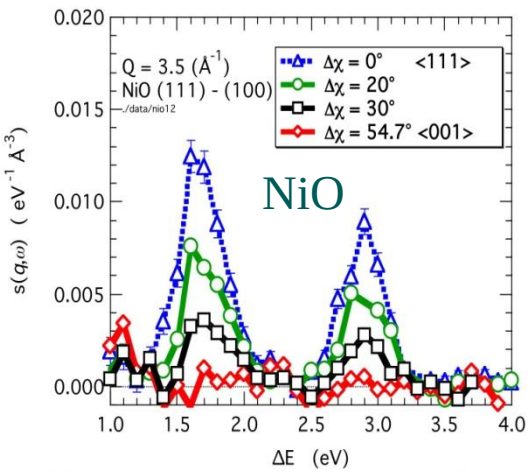


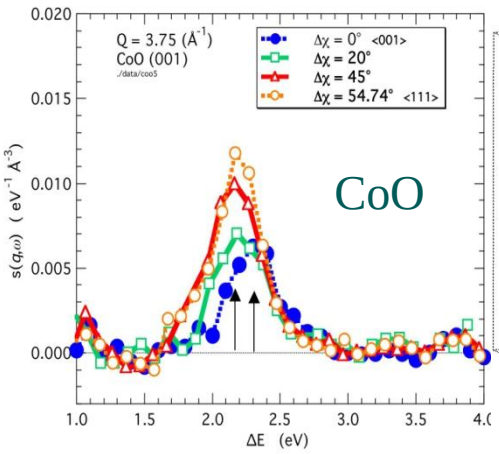
- Nodal direction ← point group symmetry
- Lack of [100] node in CoO → weak symmetry breaking

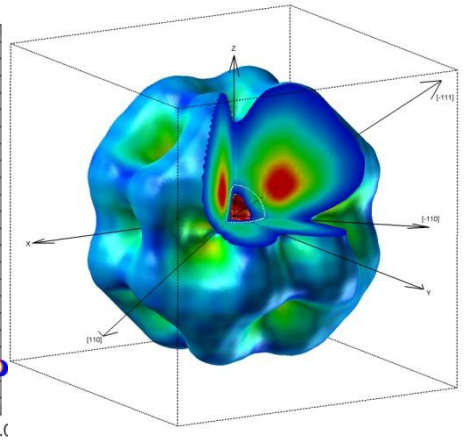
B. C. Larson, Wei Ku, Tischler, Chi-Cheng Lee, *et al.*, PRL **99**, 026401 (2007)

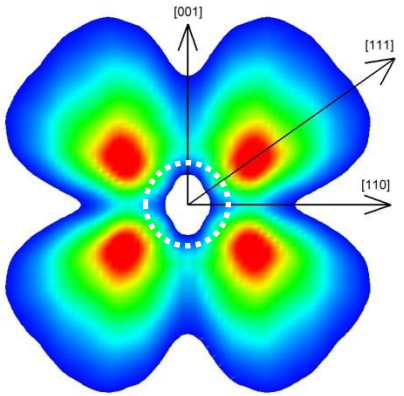
Local Excitations in NoO and CoO

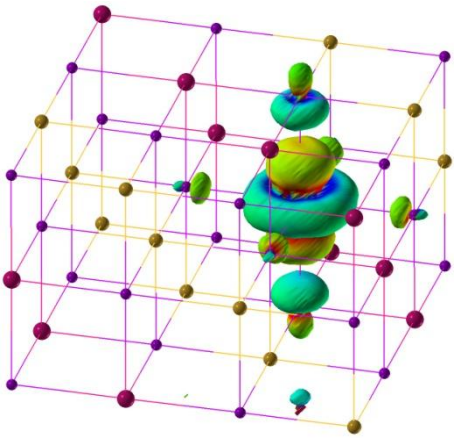
Sensitive probe of weak symmetry breaking



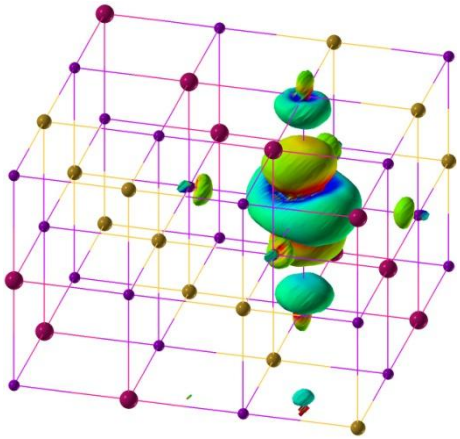




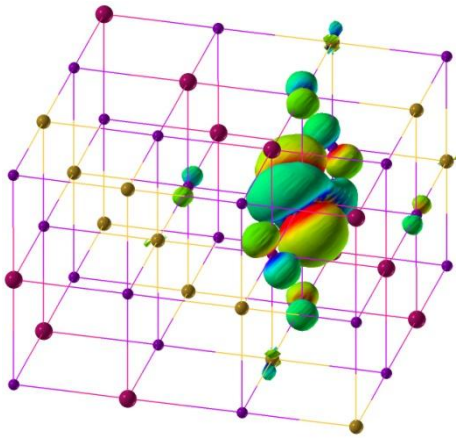




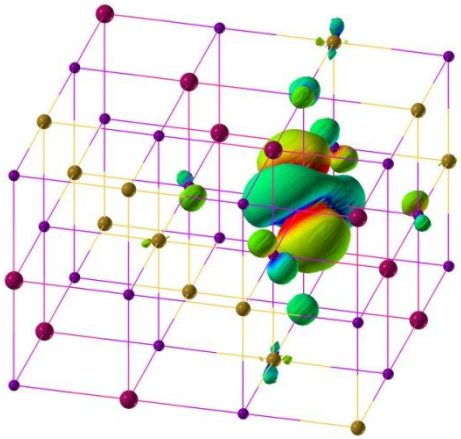
NiO



CoO



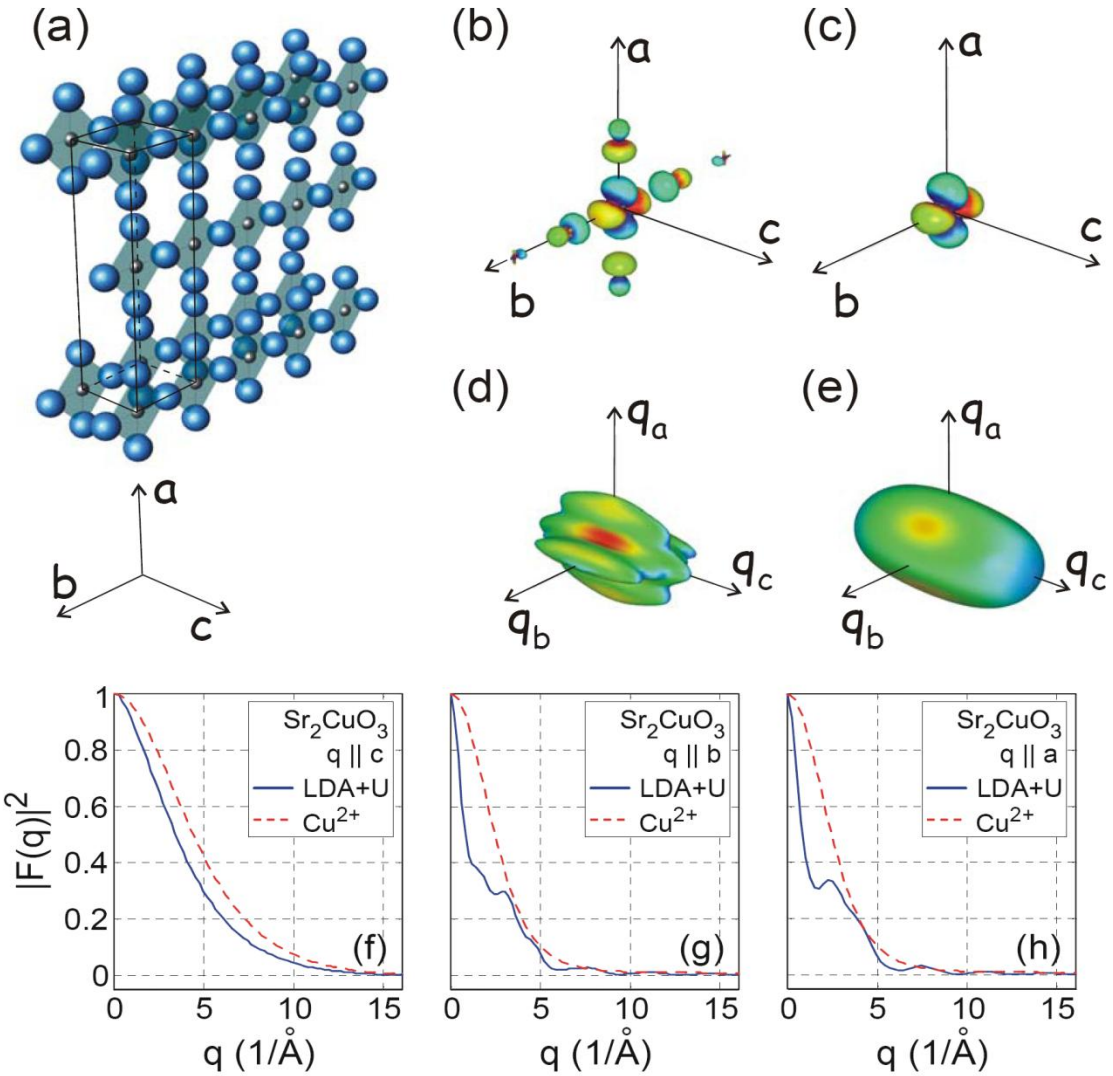
NiO



CoO

- Lost of nodal directions : extremely sensitive to weak symmetry breaking
- Visualization of symmetry breaking via Wannier functions

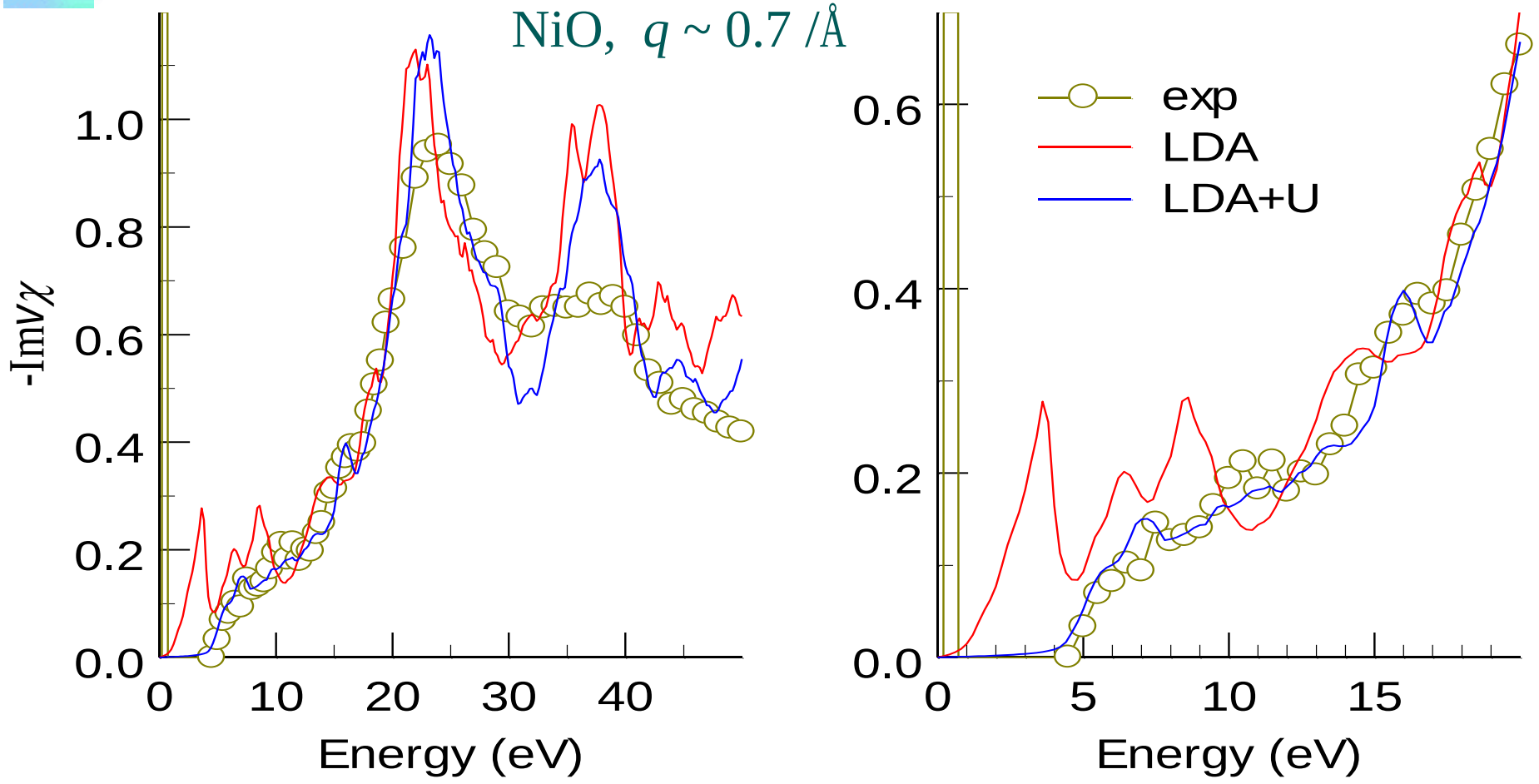
70% Missing Spectral Weight of INX in Cuprates



Igor Zaliznyak *et al*., Nature Physics 2009

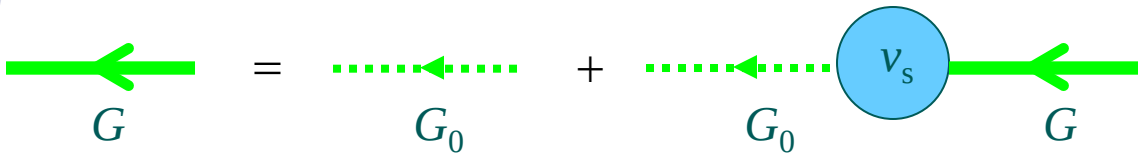
Charge Excitations in NoO

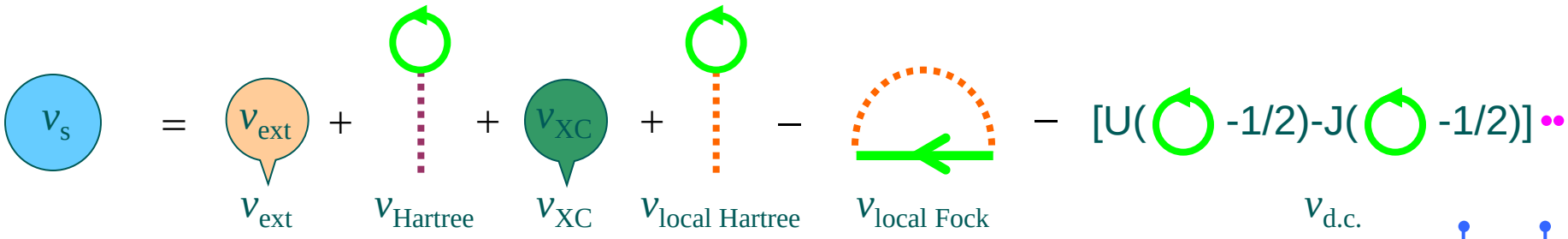
Small momentum transfer



- Small q → inter-site excitations
- LDA+U approximation greatly improves the gap and line shape
- Good agreement at small q in absolute unit
- Can't produce peaks in the Mott gap at large q ☹

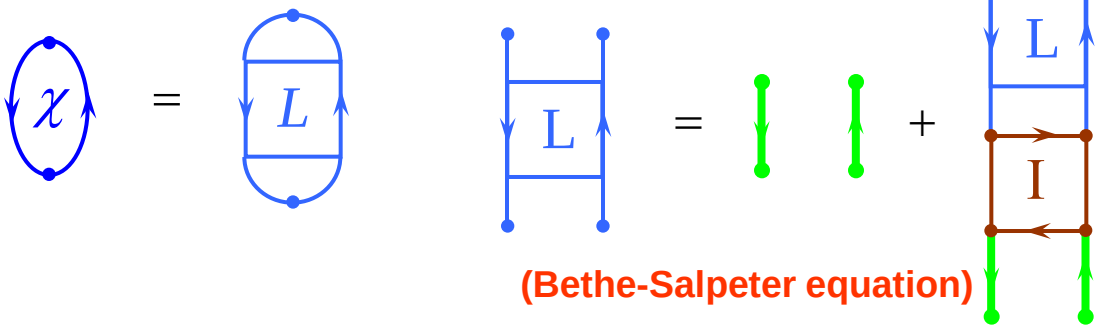
TDDFT via LDA+U Functional (TD-LDA+U)

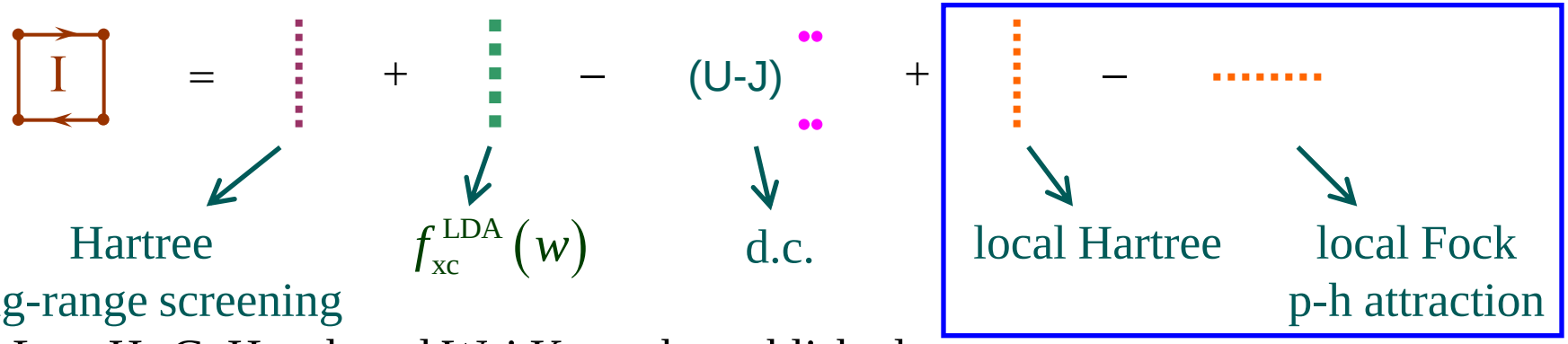




response function

$$L = \frac{\delta G}{\delta v_{ext}} = -G\frac{\delta G^{-1}}{\delta v_{ext}}G$$

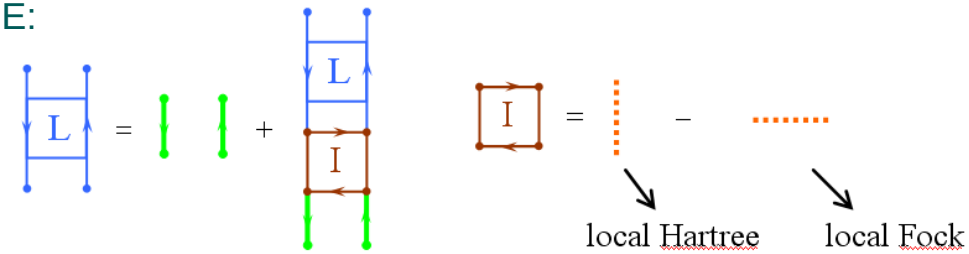




C.-C. Lee, H.-C. Hsueh and Wei Ku, to be published

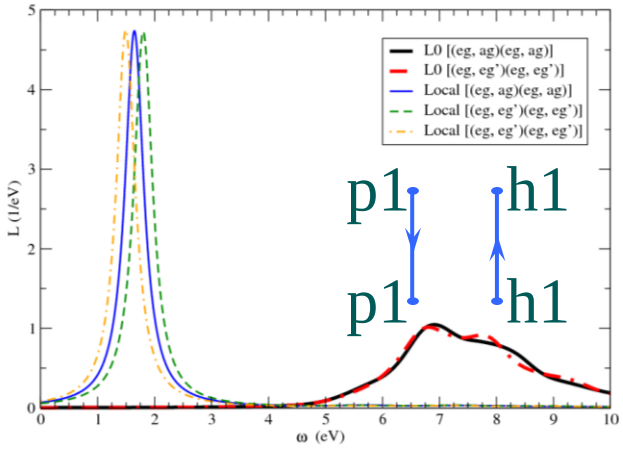
Formation of Frenkel Excitons in Local Picture

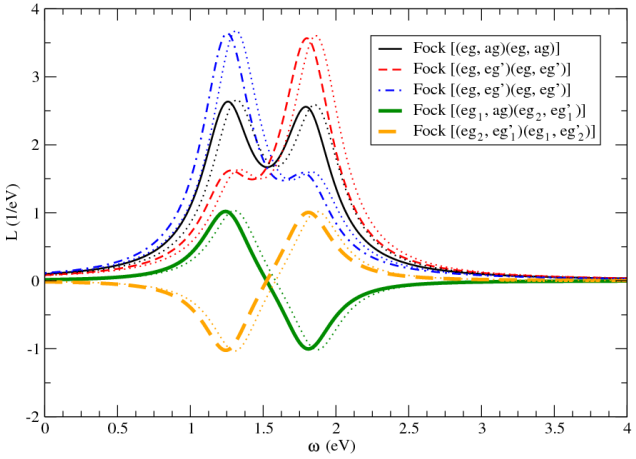
BSE:

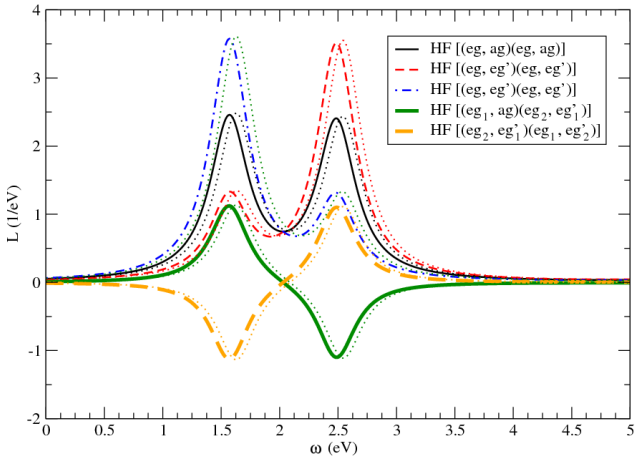


(Wannier basis, laptop done all the job)

bind energy ~ 8 eV







\+ p1 h1 / p1 h1 **binding**

same pair

p-h attraction

\+ p2 h2 / p1 h1 **scattering**

local Fock

\+ p1 h1 / p1 h1 **screening**

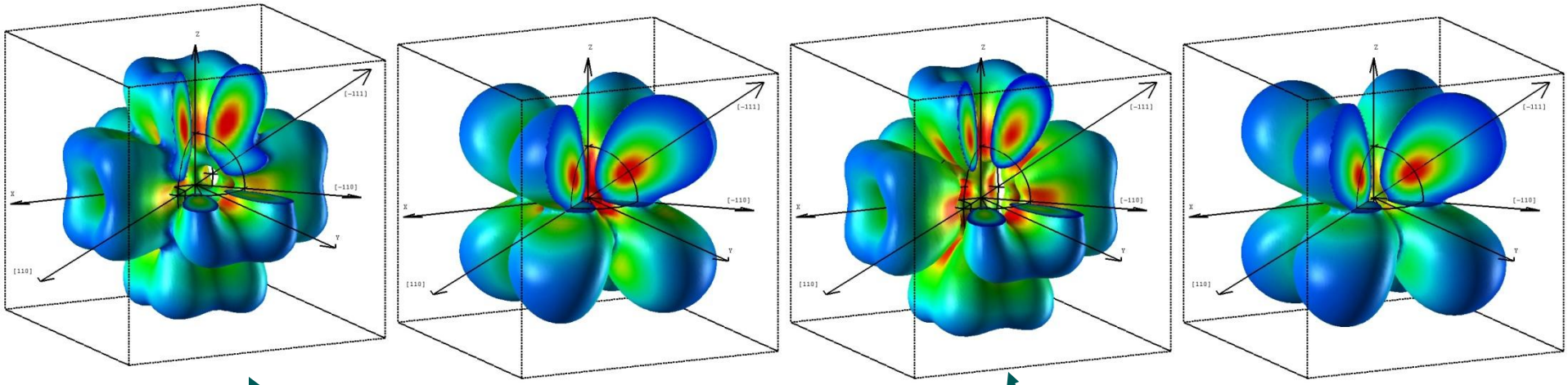
local Hartree

➔ Exciton energy in reasonable agreement with experiment

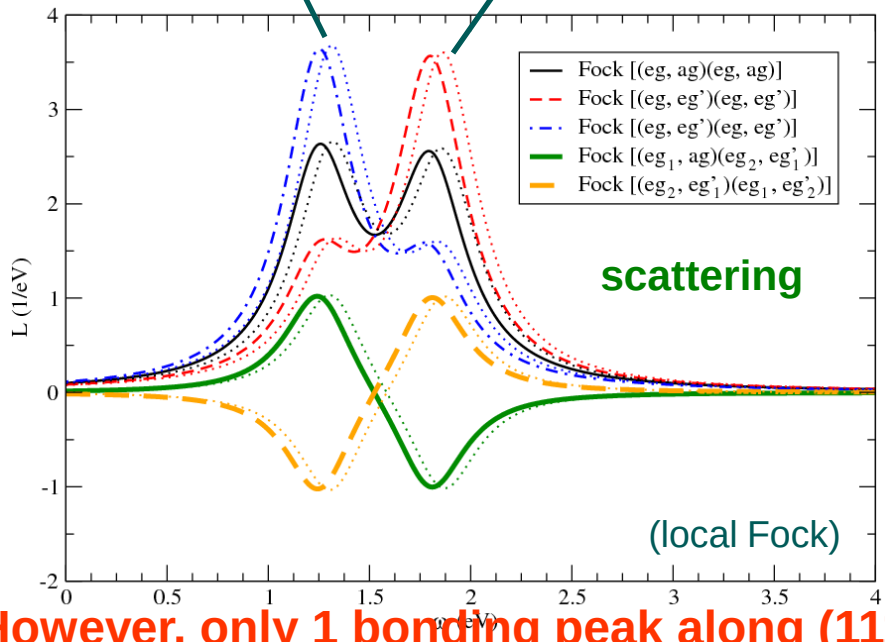
➔ Strongly hybridized Frenkel excitons

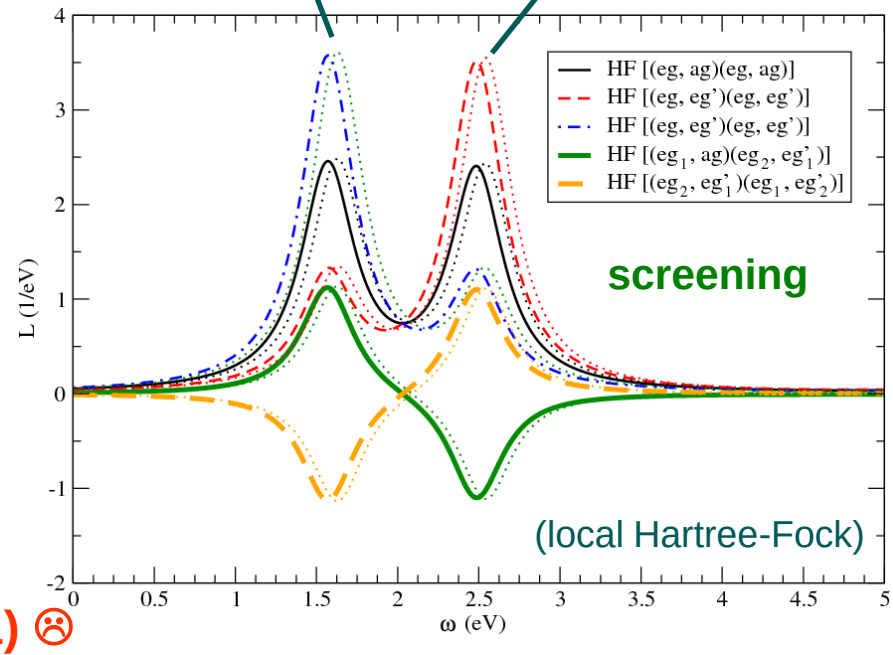
C.-C. Lee, H.-C. Hsueh and Wei Ku, to be published

Capability and Limitation of the Approximate Functional



(anti-bonding) (bonding) (anti-bonding) (bonding)



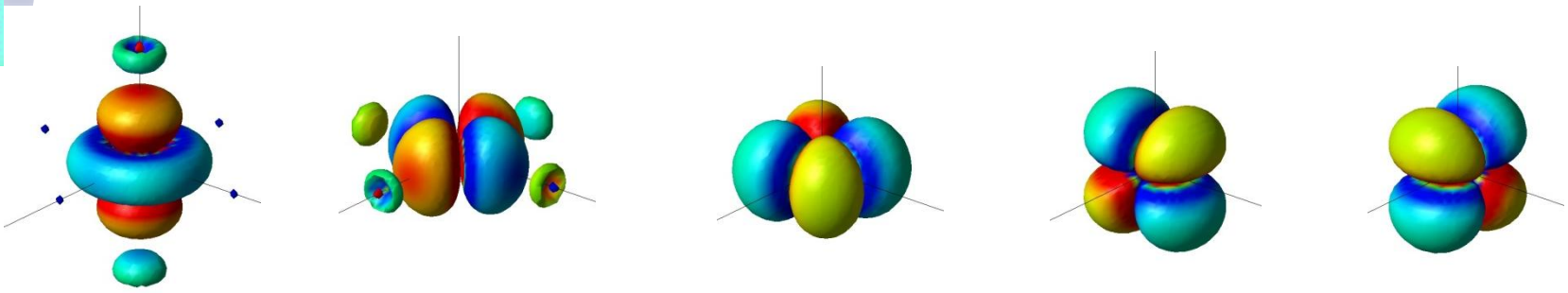


However, only 1 bonding peak along (111) ☹

← Generic problem in solving BSE with simple kernel or with QP approximation

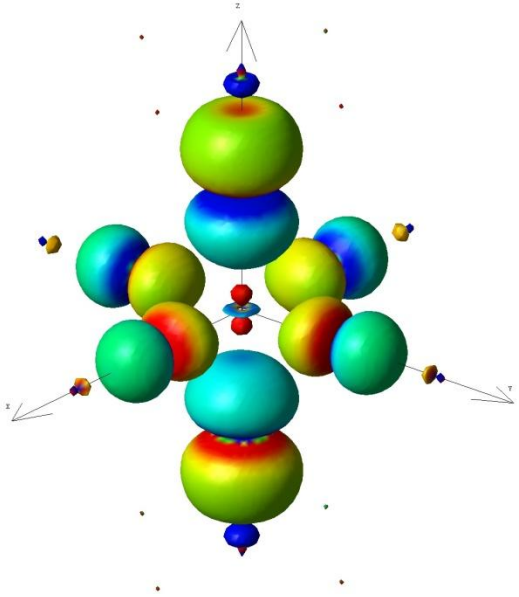
Super Atom for Charge Transfer Insulator

Ni

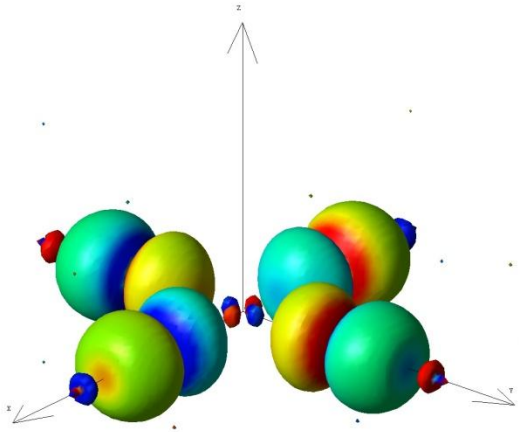


$3z^2 - r^2$ $\sqrt{3}(x^2 - y^2)$ xy yz xz

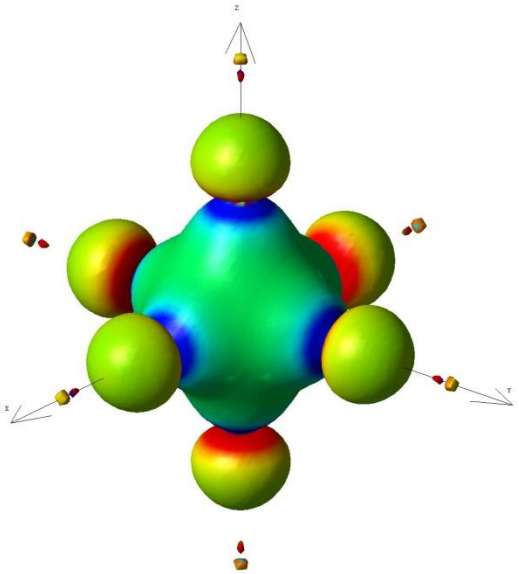
O



$3z^2 - r^2$



$\sqrt{3}(x^2 - y^2)$



s wave

$$H = H_{local} + H_{nonlocal}$$

(exact) **(modification)**

Maximize the contributions of “local atom”

Local density-density response function

$$H_{local} = T_{ij}^{\sigma} C_{i\sigma}^{+} C_{j\sigma} + \frac{1}{2} V_{ijkl} C_{i\sigma}^{+} C_{j\sigma'}^{+} C_{l\sigma'} C_{k\sigma}$$ (in Wannier basis)

one-particle Green's function
two-particle Green's function

$$\chi(\vec{q},\vec{q};\omega) = \sum_m \left(\frac{\left|\langle \Psi_m | \hat{\rho}(-\vec{q}) | \Psi_0 \rangle\right|^2}{\hbar\omega - (E_m - E_0) + i\eta} - \frac{\left|\langle \Psi_m | \hat{\rho}(\vec{q}) | \Psi_0 \rangle\right|^2}{\hbar\omega - (E_0 - E_m) - i\eta} \right)$$

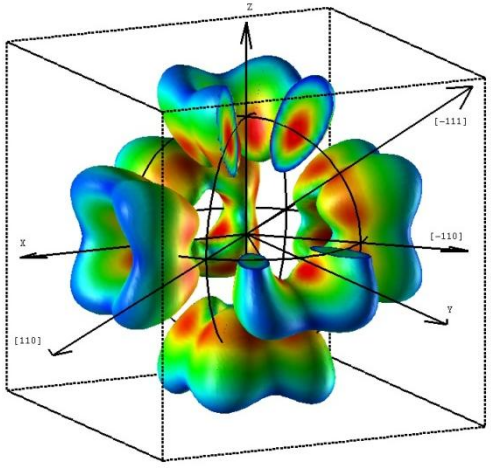
T_{ij}^{local} :

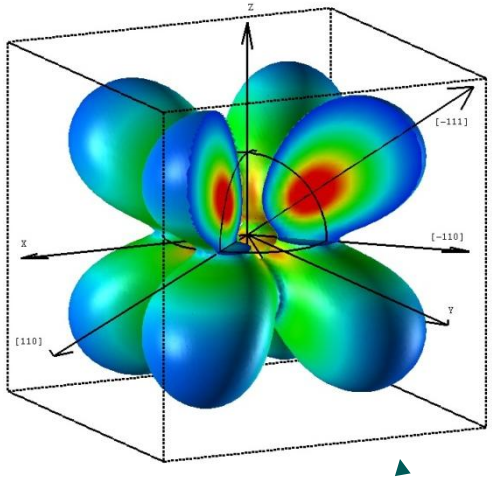
					O		
$3z^2-r^2$	$\sqrt{3}(x^2-y^2)$	yz	xz	xy	s	$3z^2-r^2$	$\sqrt{3}(x^2-y^2)$
-52.21	0.00	0.00	0.00	0.00	0.00	1.79	0.00
0.00	-52.21	0.00	0.00	0.00	0.00	0.00	1.79
0.00	0.00	-52.09	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	-52.09	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	-52.09	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	-4.40	0.00	0.00
1.79	0.00	0.00	0.00	0.00	0.00	-2.35	0.00
0.00	1.79	0.00	0.00	0.00	0.00	0.00	-2.35

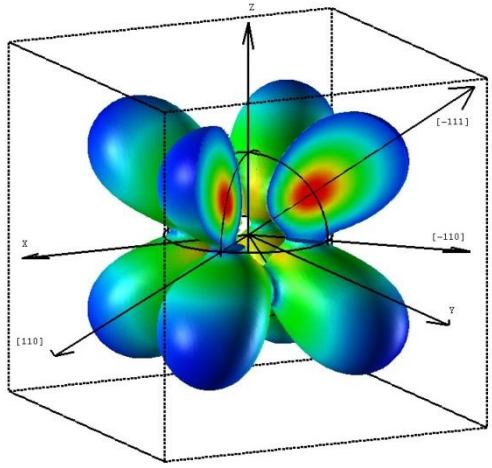
$\varepsilon = 1.097$, U = 8eV, J = 0.95eV

Density response for super atom

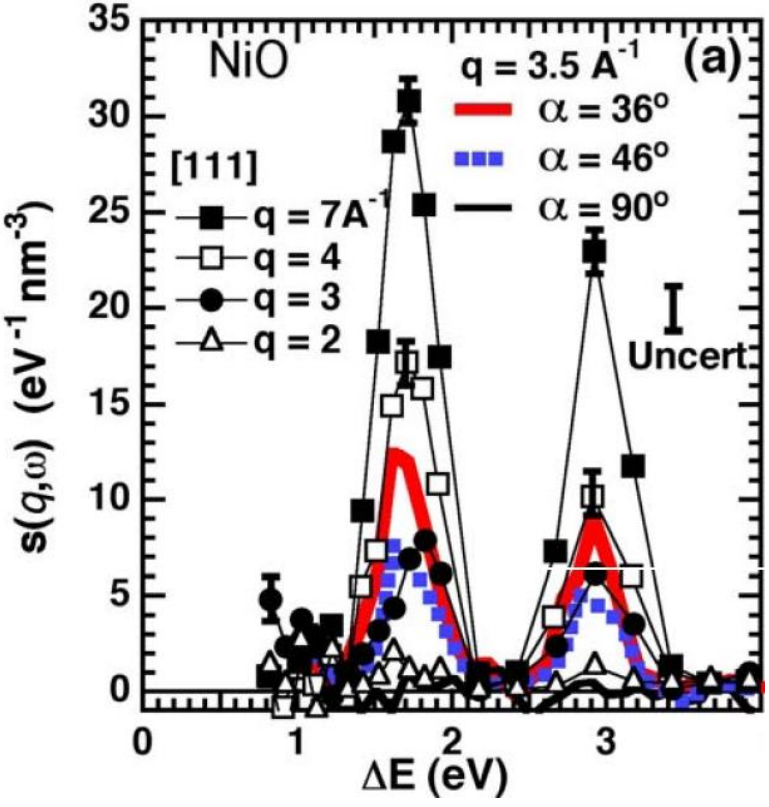
(eg-t2g=0.65eV)

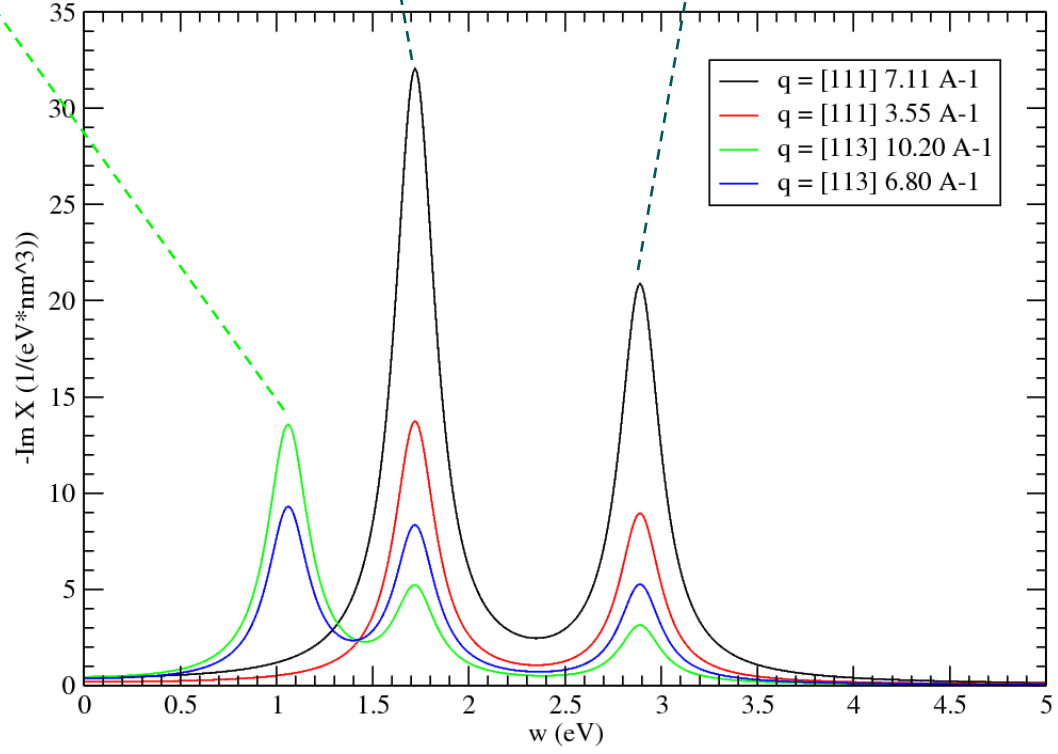






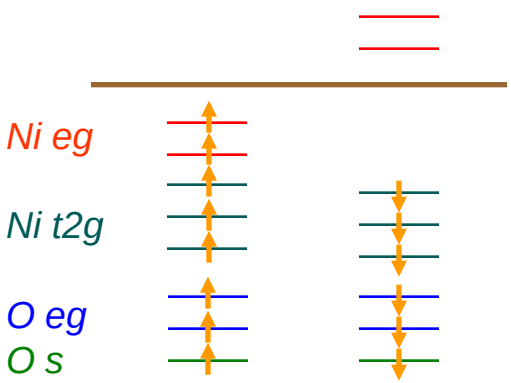
(antibonding-type)



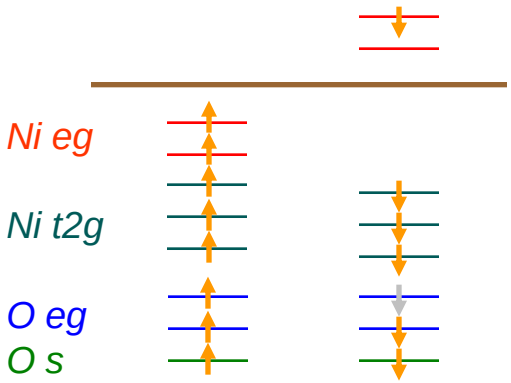


Multiplet Splitting Made Possible with MB Hilbert Space

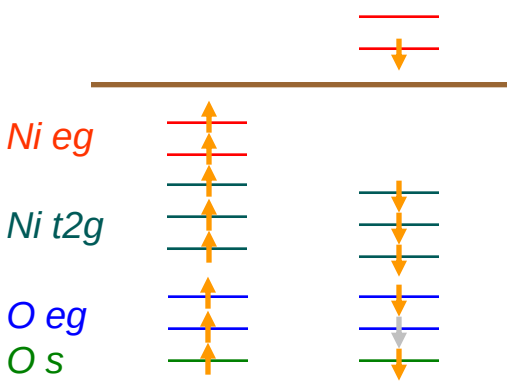
GS



(0.9484)

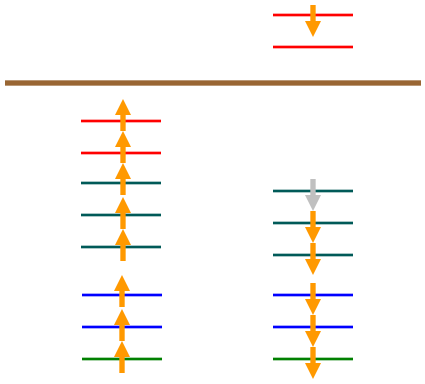


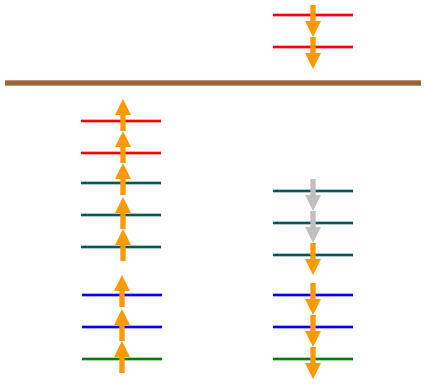
(-0.2226)

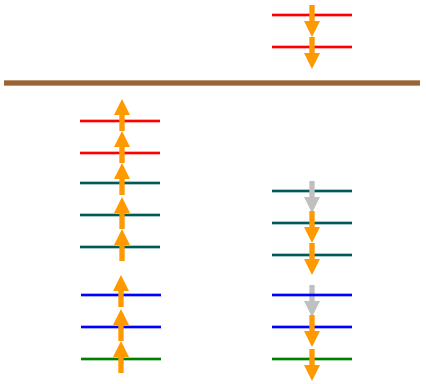


(0.2226)

T_{1g}







Summary

- Strong correlation in crystals: intra-atomic vs. inter-atomic
  - defining representative local unit
- Energy reduction: many-body down-folding
  - RG: energy-dependent perspective of physics
  - a proper derivation of low-energy physics
- QP excitations: ARPES and Fermi surface
- Length reduction in simulation: flavored twisted boundary condition
  - circumventing the finite size effects
- P-H excitations of large momentum: NIXS & INS
  - Momentum transfer: anisotropy & local orbitals
  - Spectrum: perturbation, TD-LDA+*U* & beyond
  - Propagation in space time: effective hopping kernel