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Swiss Federal Institute of Technology Zurich

Ultrafast Dynamics of Strongly Correlated Systems at SwissFEL

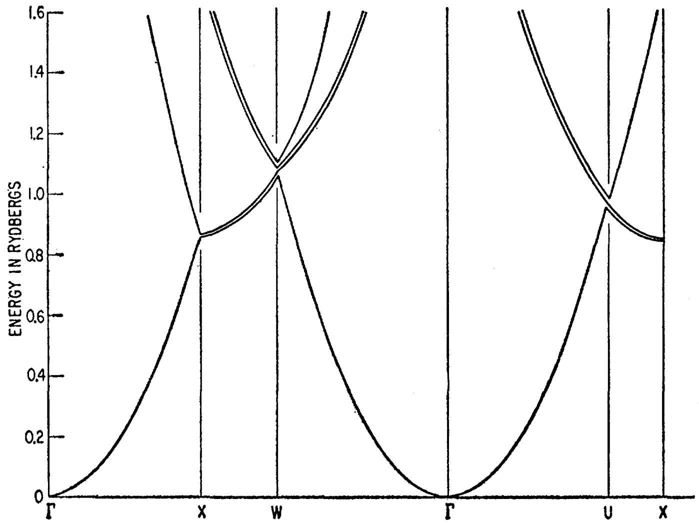
Steve Johnson, ETHZ

Outline

- Introduction: problems in strongly correlated systems
- Examples
  - CDW melting in $TiSe_2$
  - Diffuse scattering as a probe of nonequilibrium phonons
  - Lattice, charge & orbital order dynamics in manganites
  - Nonlinear phonon-phonon interactions
- Enabling technologies for ESB

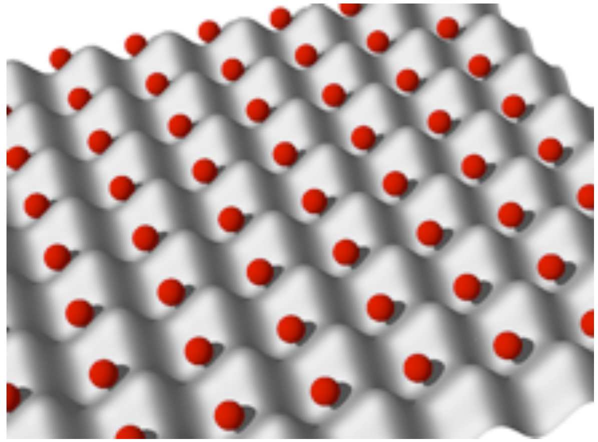
Strongly correlated systems

Al: nearly free electron model



[Harrison, Phys. Rev. 118 (1960)]

Mott insulator

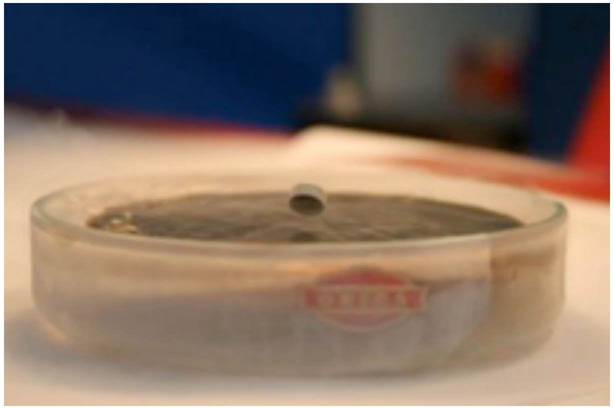


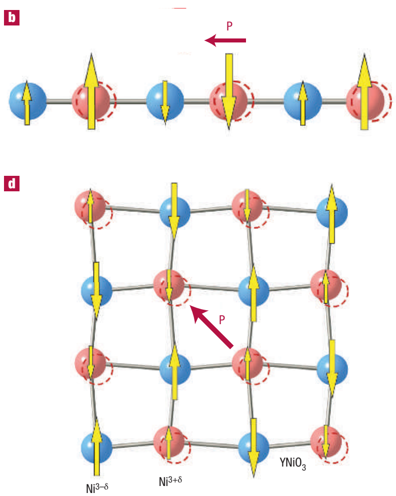
Increasing e-e correlation

- Strong correlations between electronic states
- Breakdown of independent electron picture

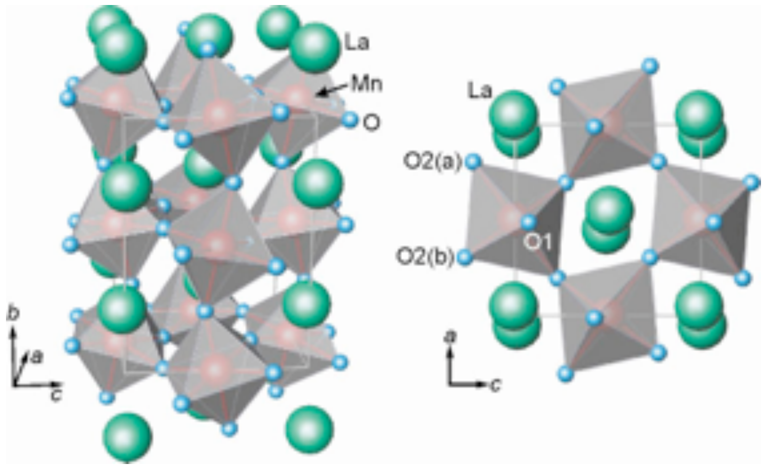
Strongly correlated systems

- High-T_c superconductors



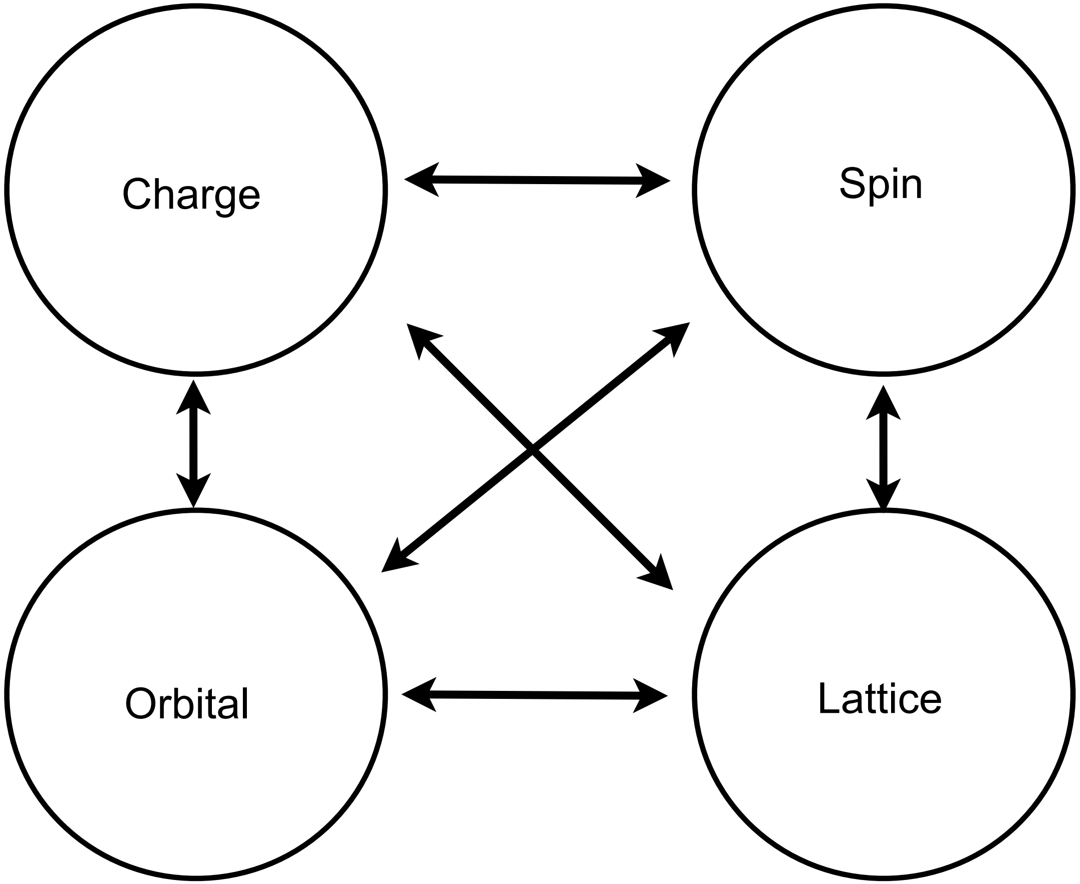


- Manganites (CMR, CO/OO)



- Multiferroics

Strongly correlated systems

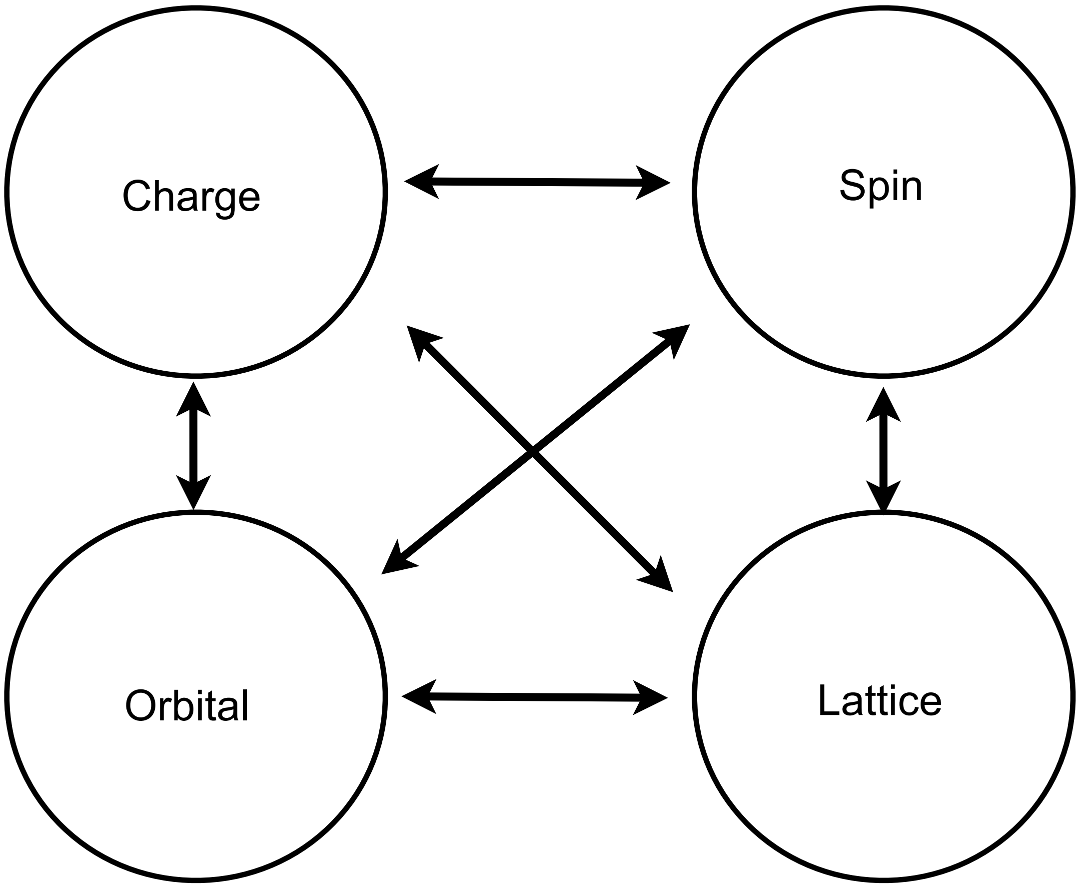


- Correlations from strong, competing interactions

Ultrafast: non-thermodynamic states in correlated systems

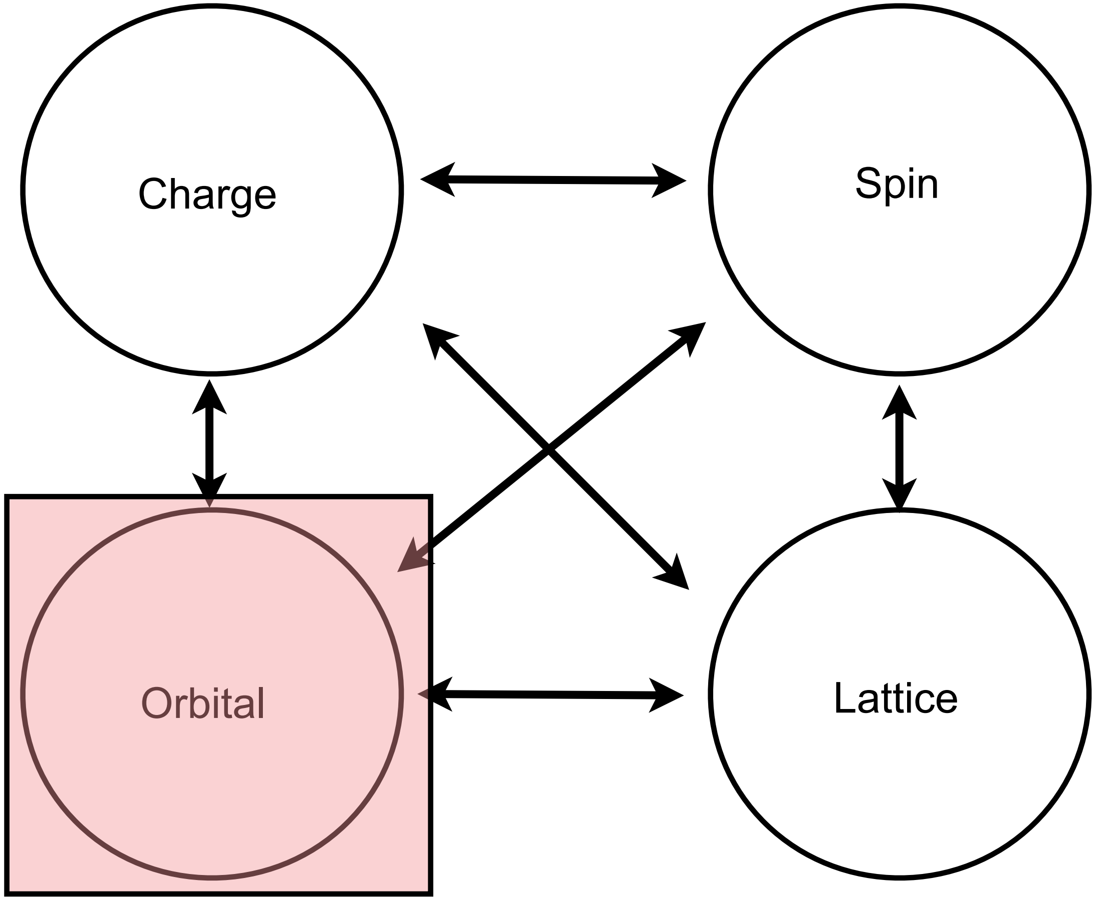
- New ways to control the state of correlated systems
  - More efficient?
  - New states?
  - Faster?
- Important test of theoretical models

Strongly correlated systems



- Ideal experiment: selective, fast pump & selective, fast probe

Strongly correlated systems



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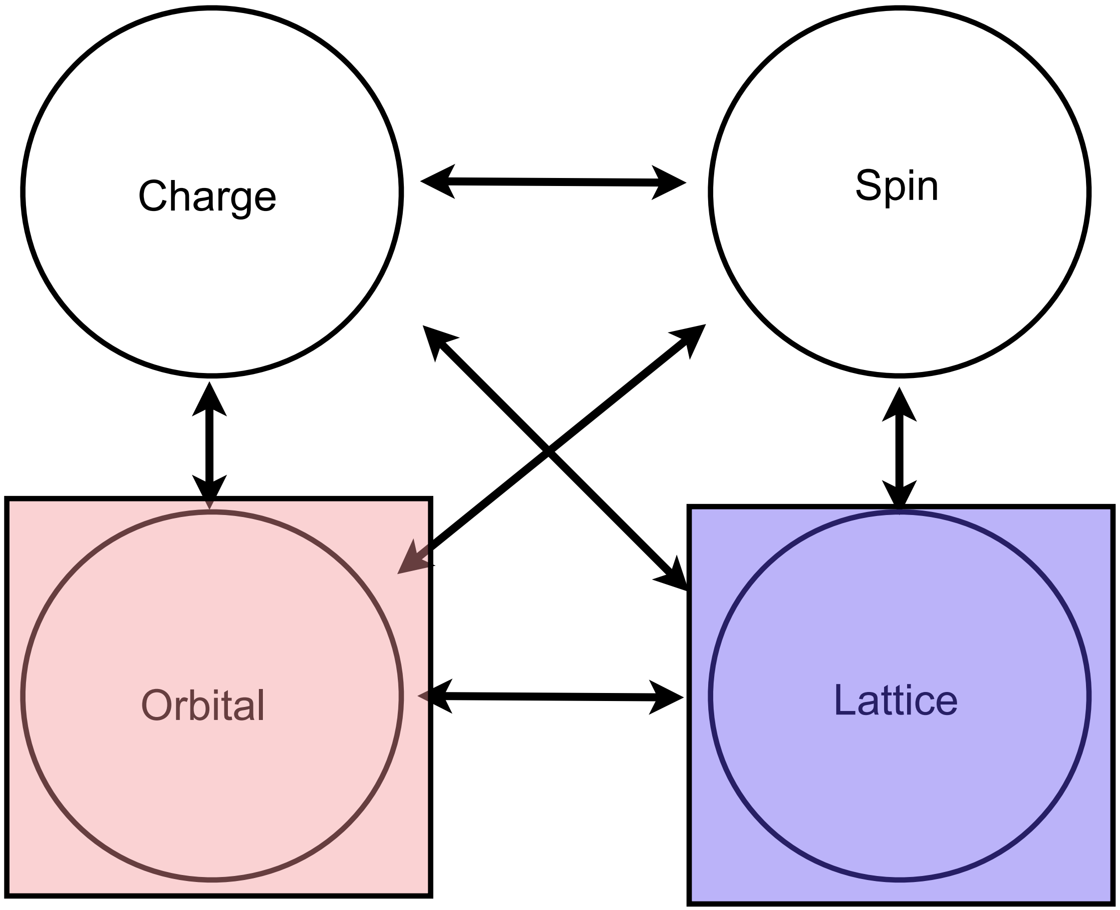

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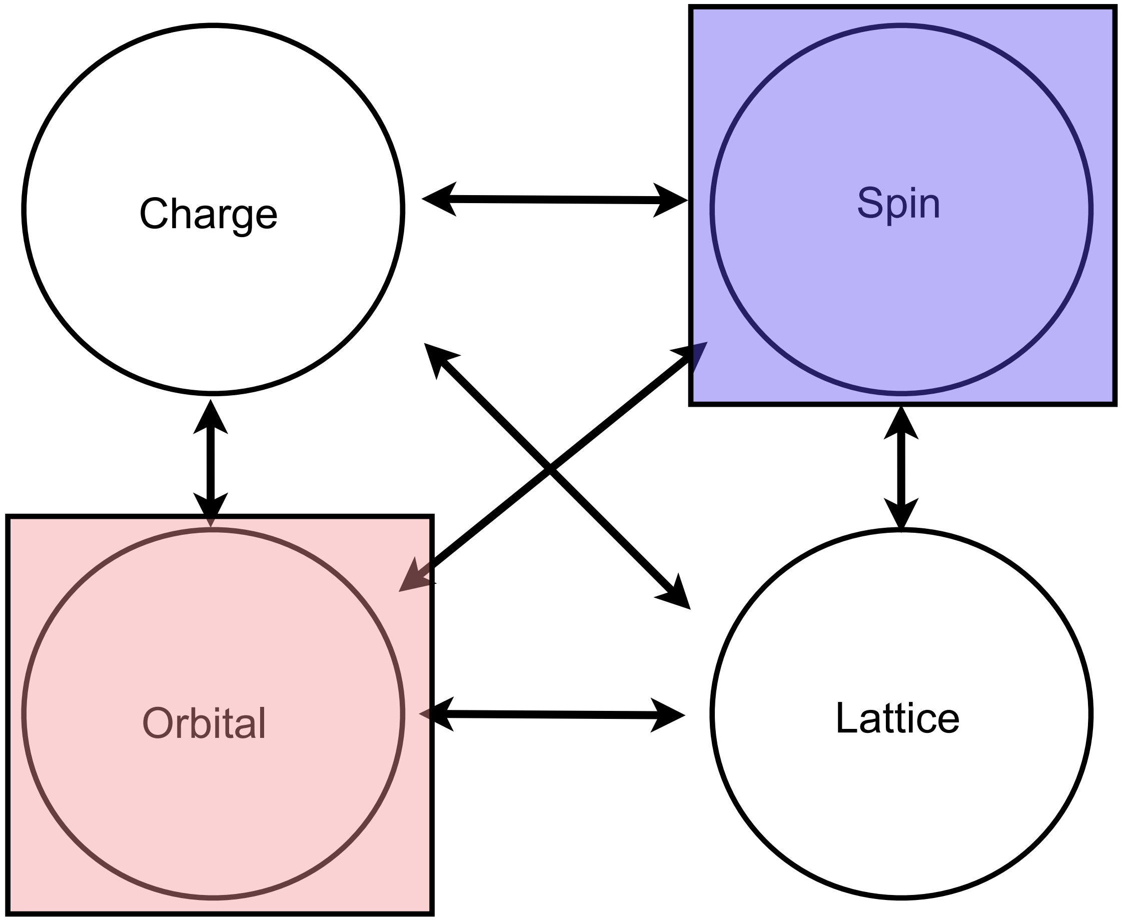
Strongly correlated systems



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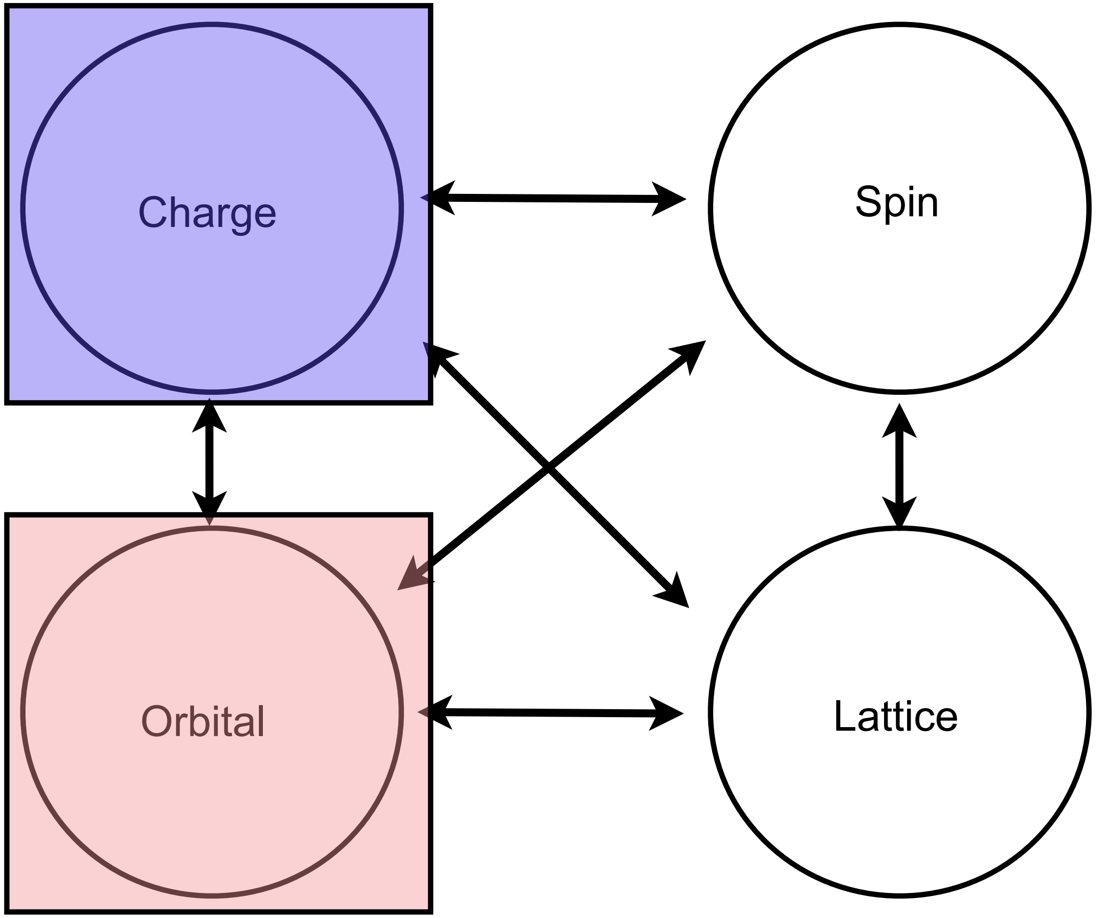

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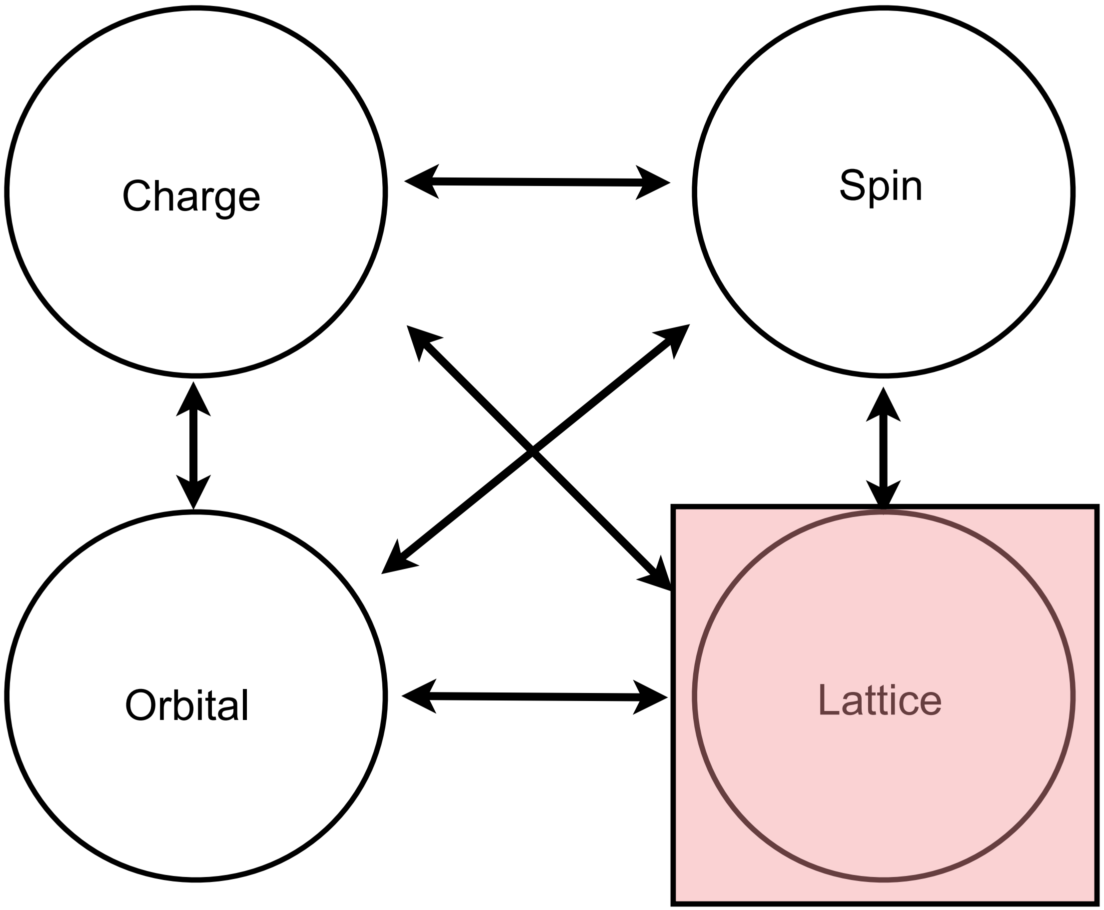

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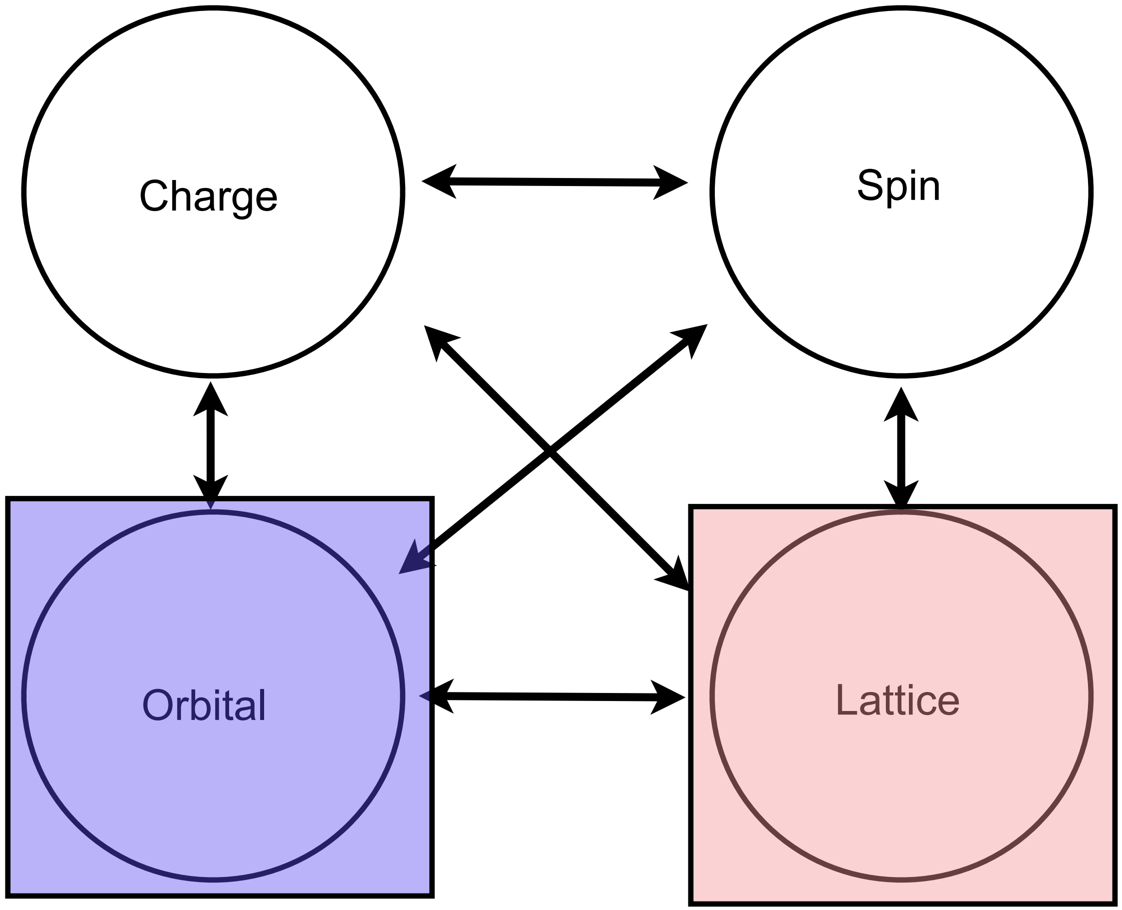
Strongly correlated systems

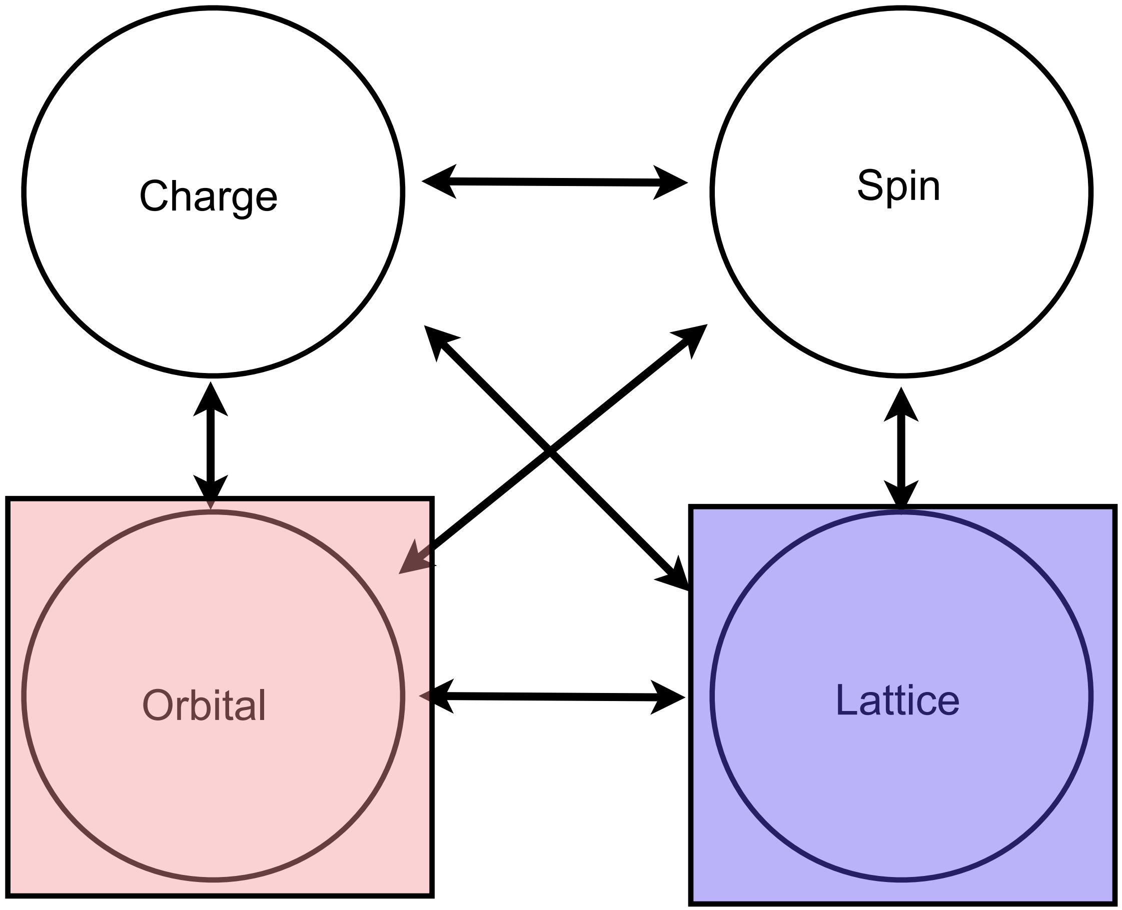


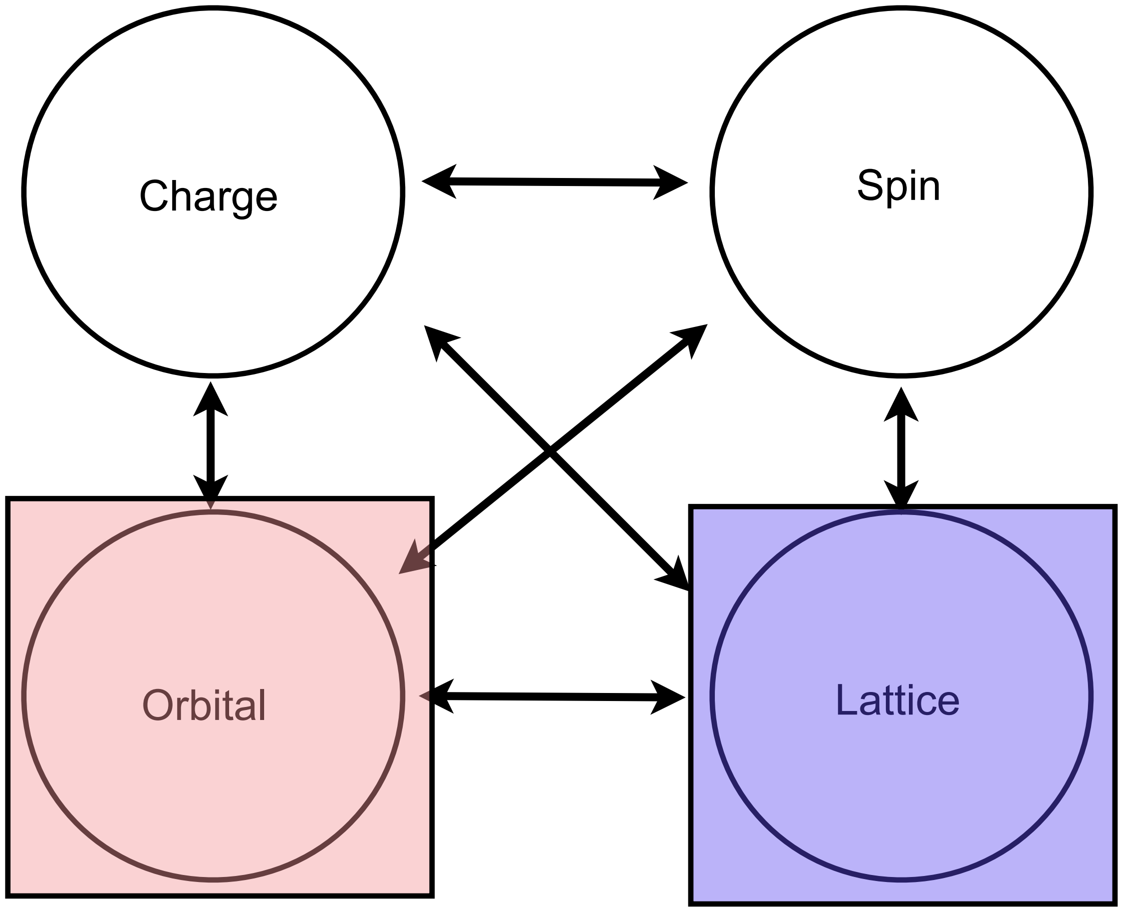
- Ideal experiment: selective, fast pump & selective, fast probe

Ultrafast: non-thermodynamic states in correlated systems

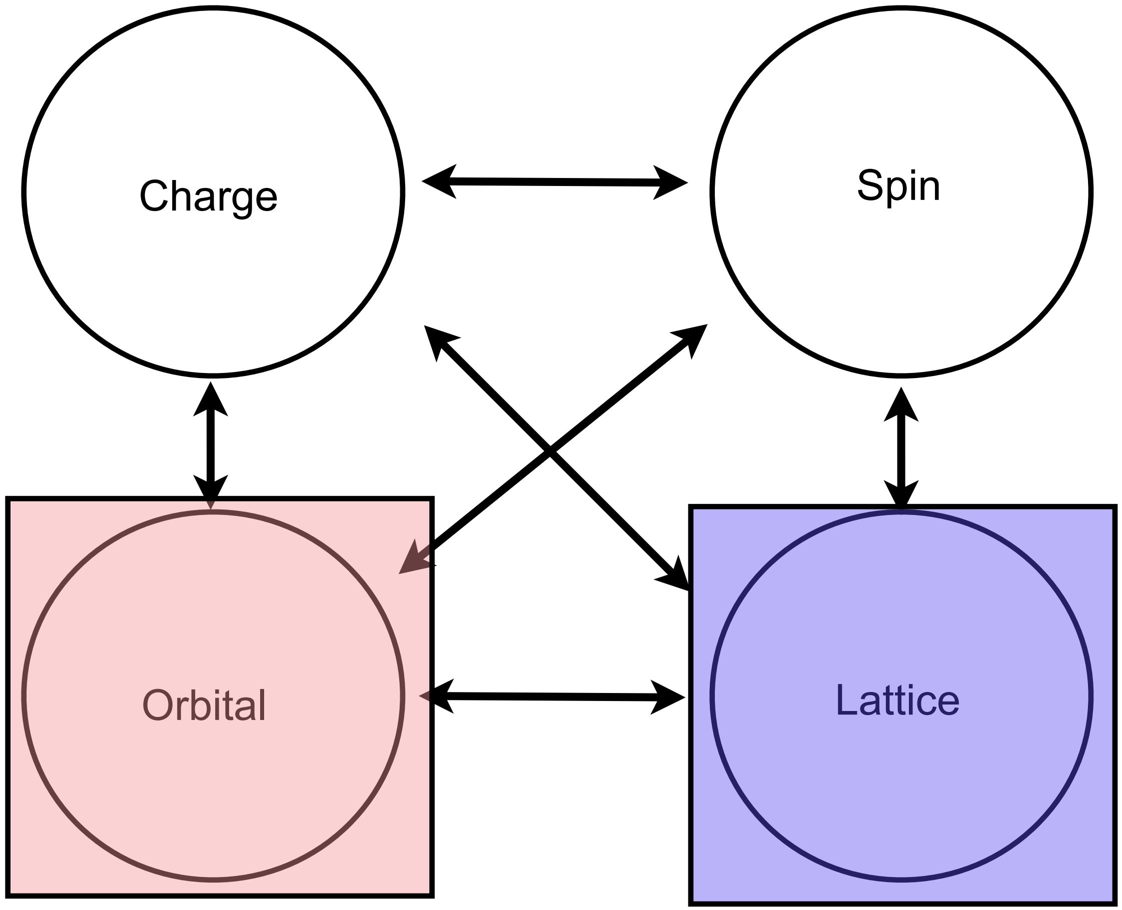
- New ways to control the state of correlated systems
  - More efficient?
  - New states?
  - Faster?
- Important test of theoretical models

- Requires:
  - Pump, probe faster than coupling time
  - Selectivity in pump and probe


Charge
Spin
Orbital
Lattice

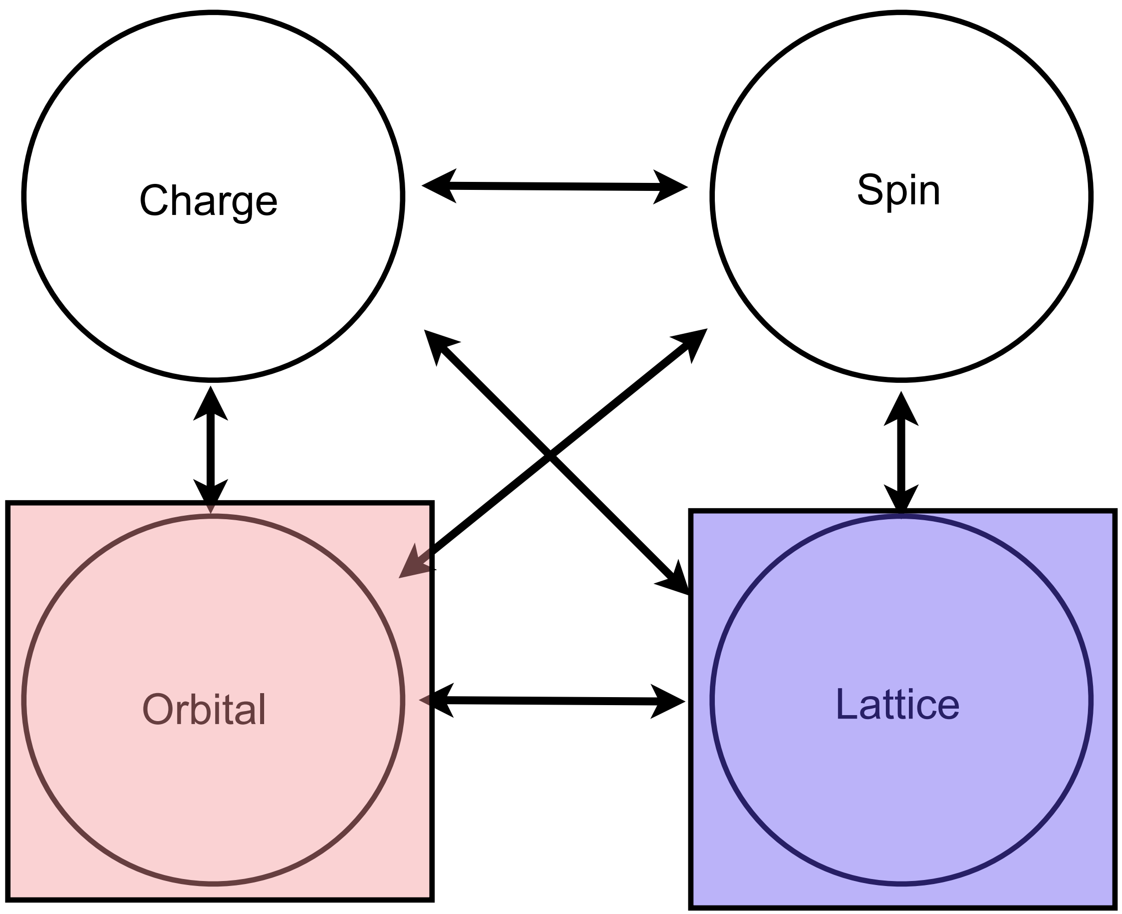


Interband absorption (optical)



Interband absorption (optical)

X-ray diffraction



Interband absorption (optical)

X-ray diffraction
Diffuse scattering

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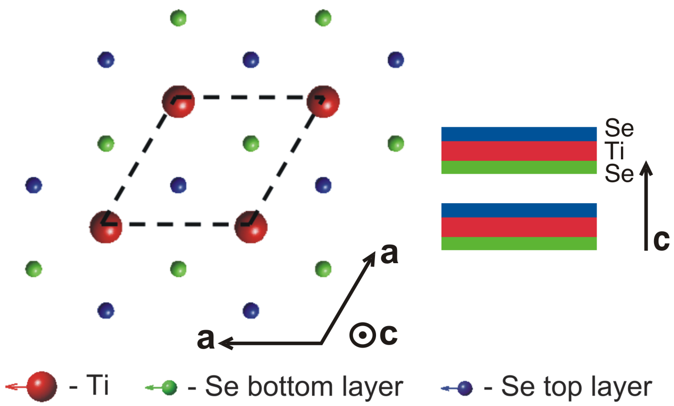
$TiSe_2$: charge density wave

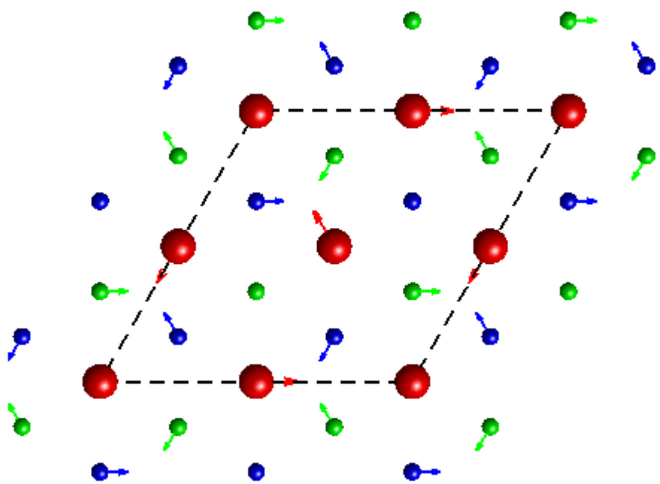
T > 200 K

- 1*T*- structure
- P-3m1
- Semimetal

T < 200 K

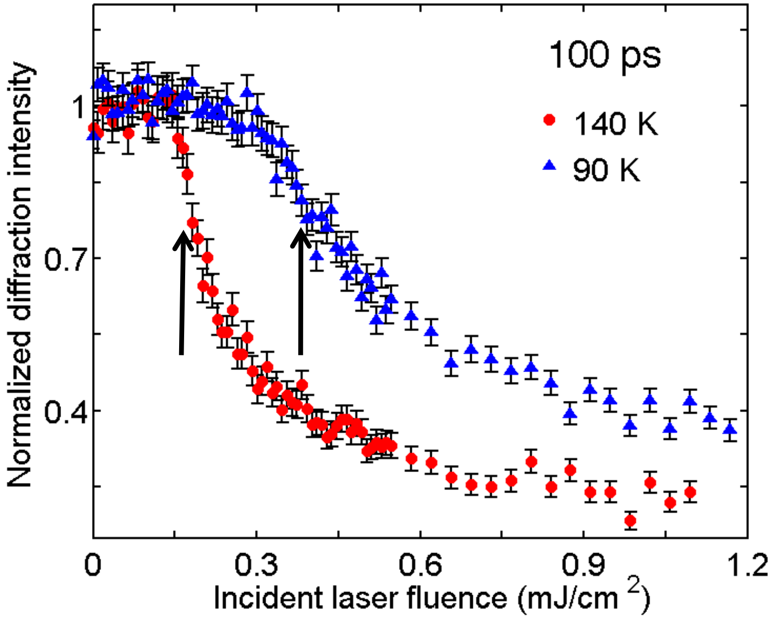
- CDW commensurate phase
- (2a×2a×2c) Superlattice
- Distorts towards 2*H*-structure
- Semimetal





XRD: satellite peaks

Time-resolved XRD: Laser-induced transition nonthermal



	E_{con}	Laser-induced	Thermal
140 K	5.7 meV/(u.c.)	7.9 meV/(u.c.)	36.7 meV/(u.c.)
90 K	9.0 meV/(u.c.)	16.7 meV/(u.c.)	60.0 meV/(u.c.)

80 K (Optics) =>16.5 meV/(u.c.)

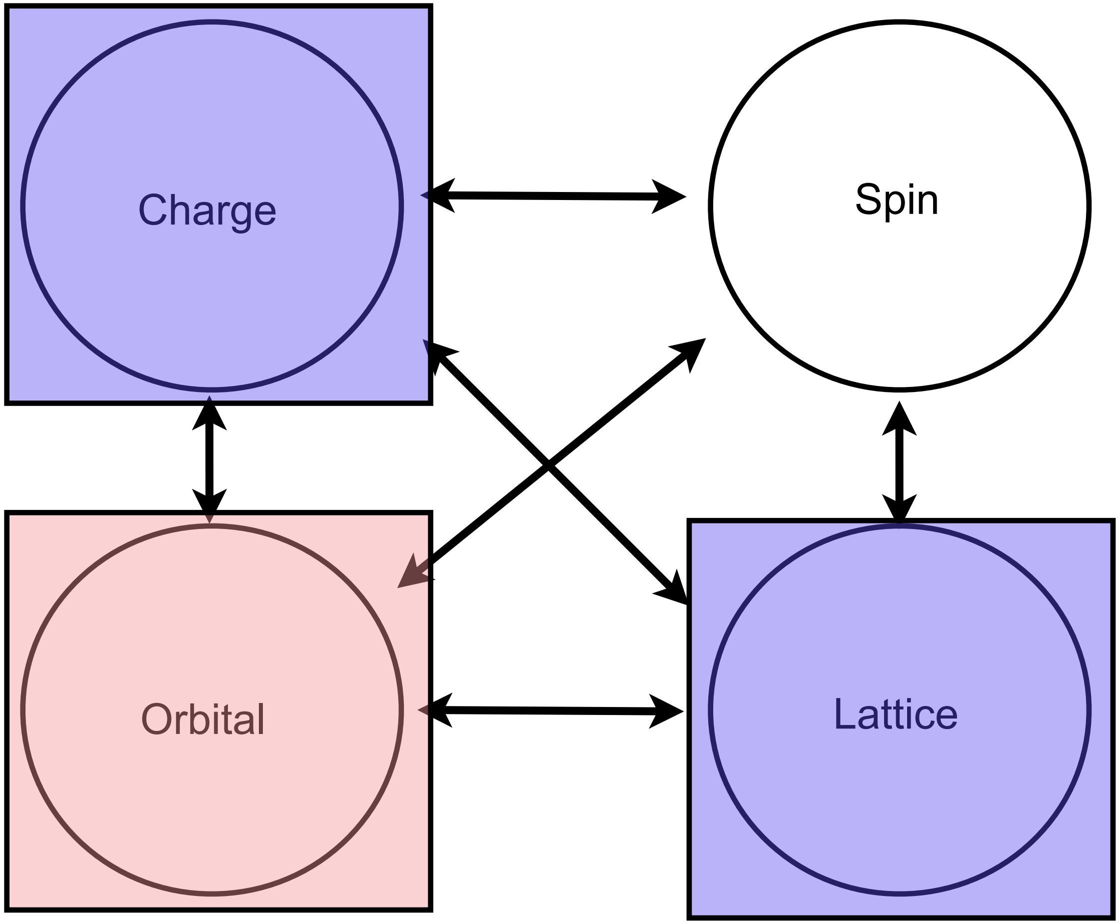
Contrasts with "conventional" CDW from FS nesting

More "efficient" way to drive transition

Supports excitonic model for mechanism of CDW

[E. Möhr-Vorobeva et al. PRL 107, 036403 (2011)]

Resonant x-ray diffraction



Interband absorption (optical)

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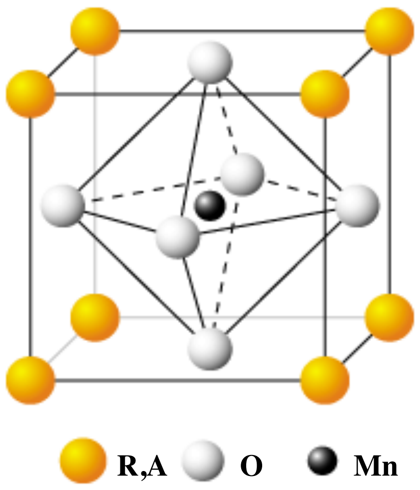
Manganese oxides: $R_{1-x}A_xMnO_3$



Transition metal oxides with perovskite structure

- R: 3^+ cation as rare earths (La, Pr,…)
- A: 2^+ cation as Ca, Na, Sr
- Mn: 3^+,4^+

Many types of long range order ...

- Structural modulation arising from Jahn-Teller distortion on Mn^{3+} sites
- Charge order: modulation of Mn valence
- Orbital order: modulation of orientation of occupied e_g orbitals in Mn^{3+}

Collaboration: PSI RESOX (U. Staub), FEMTO (P. Beaud, G. Ingold), ETHZ

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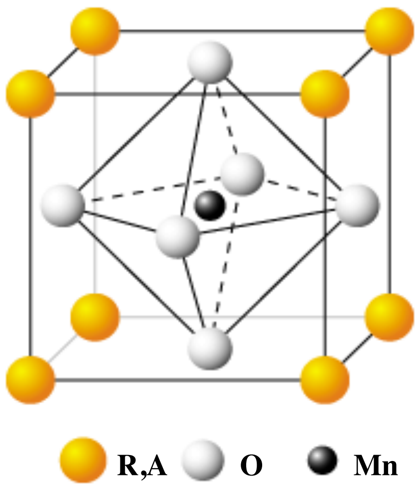

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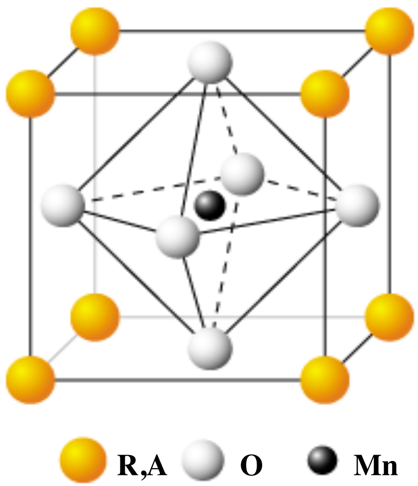

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Resonant x-ray diffraction

Non-resonant x-ray diffraction

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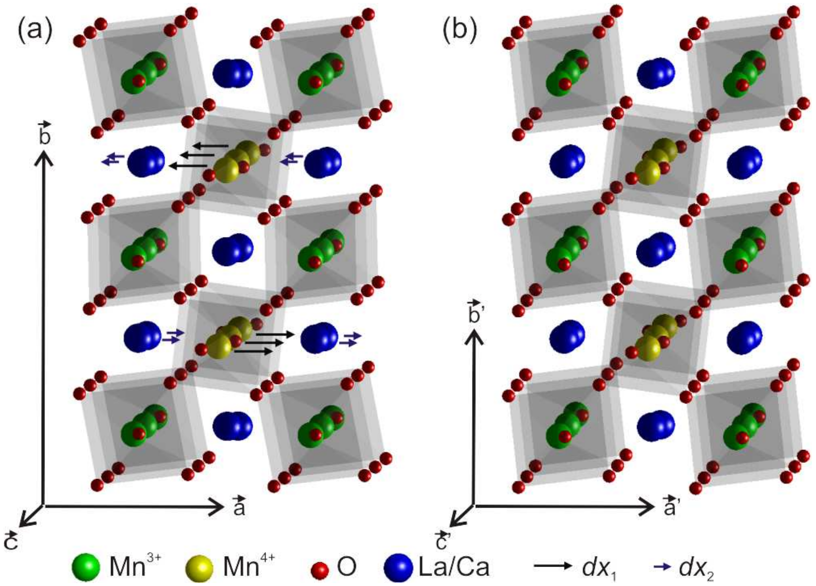
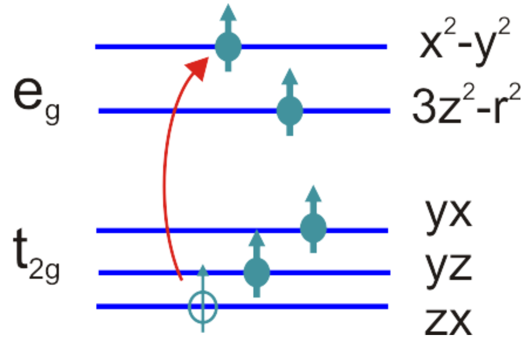
$La_{0.42}Ca_{0.58}MnO_3$

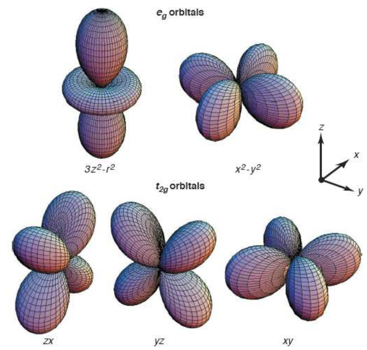
20 K
Monoclinic ($P2_1/m$)

300 K
Orthorhombic ($Pbnm$)

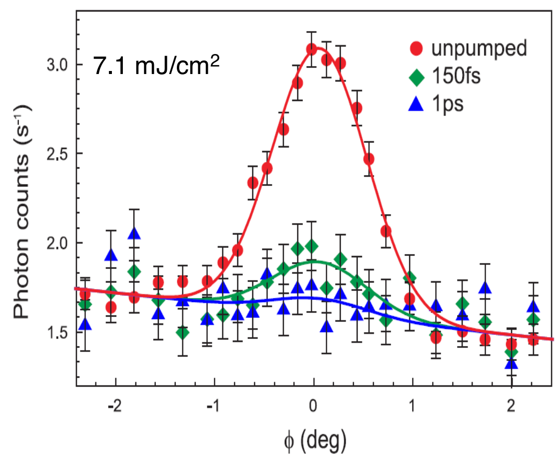


Rodriguez *et al.*, PRB 71, 104430 (2005)





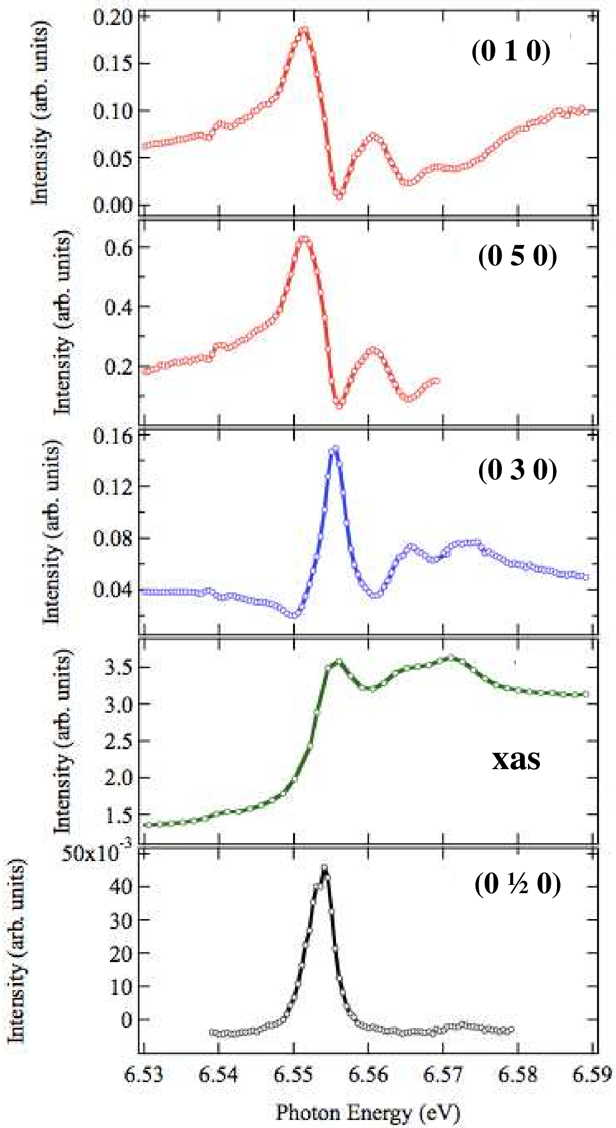
Pump: Interband absorption



PRL 103, 155702 (2009)

- Doubled unit cell due to Jahn-Teller distortion at Mn^{3+} sites
- (5 -5 2) superlattice reflection, sensitive mostly to atomic motion along *x*-axis: 80% (Mn^{4+}), 20% (La/Ca)
- Dissappears heating above $T_{CO} \approx 240$ K or with sufficient laser excitation

$Pr_{0.5}Ca_{0.5}MnO_3$: static resonant XRD



Experiment performed at Material Science beamline at SLS (Phil Wilmott)

sensitive to the charge difference of the Mn ions **Mn^{3+} / Mn^{4+}**

X-ray absorption (fluorescence)

Reflection sensitive to the orbital order (Mn^{3+})
(**Jahn-Teller distortion**)

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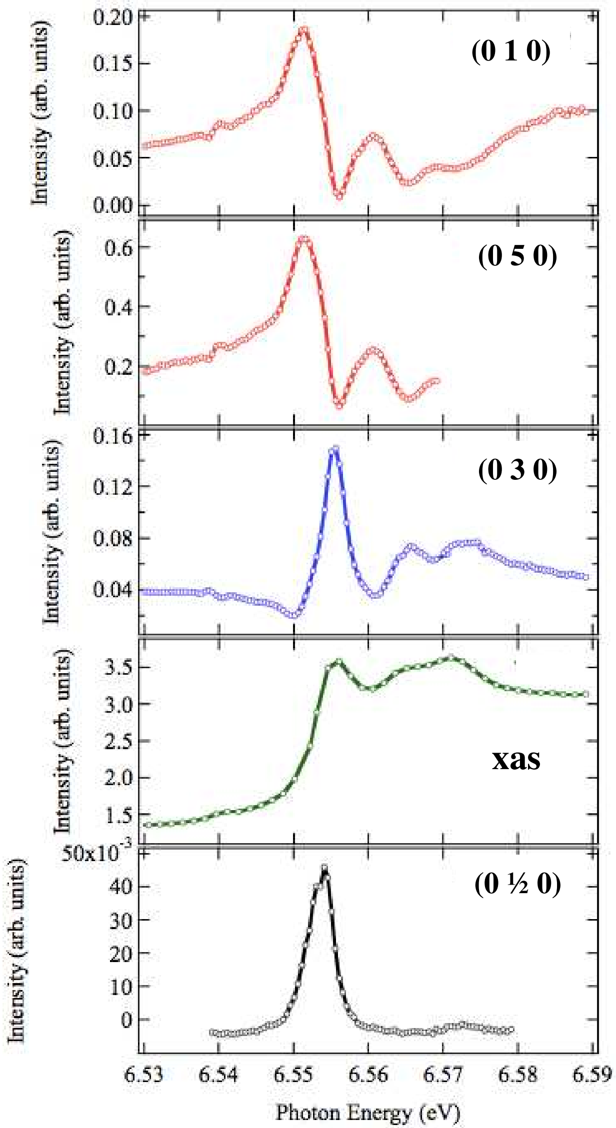

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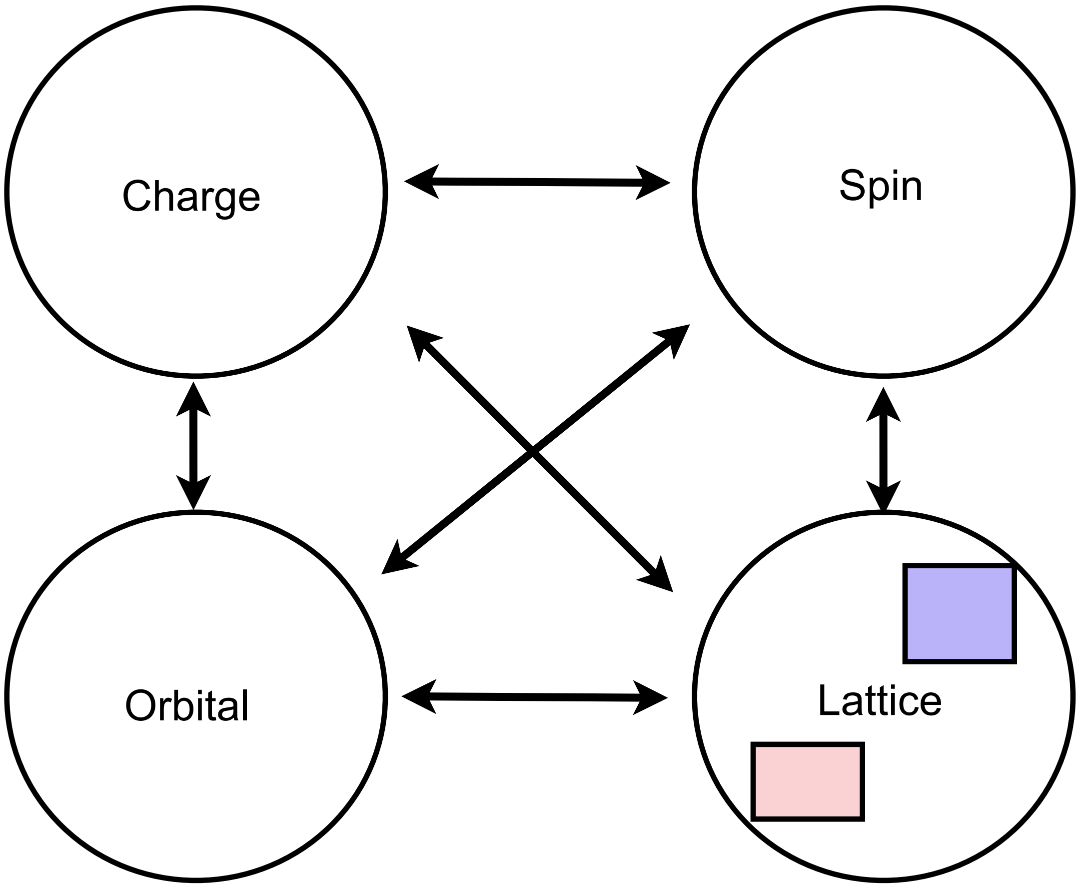
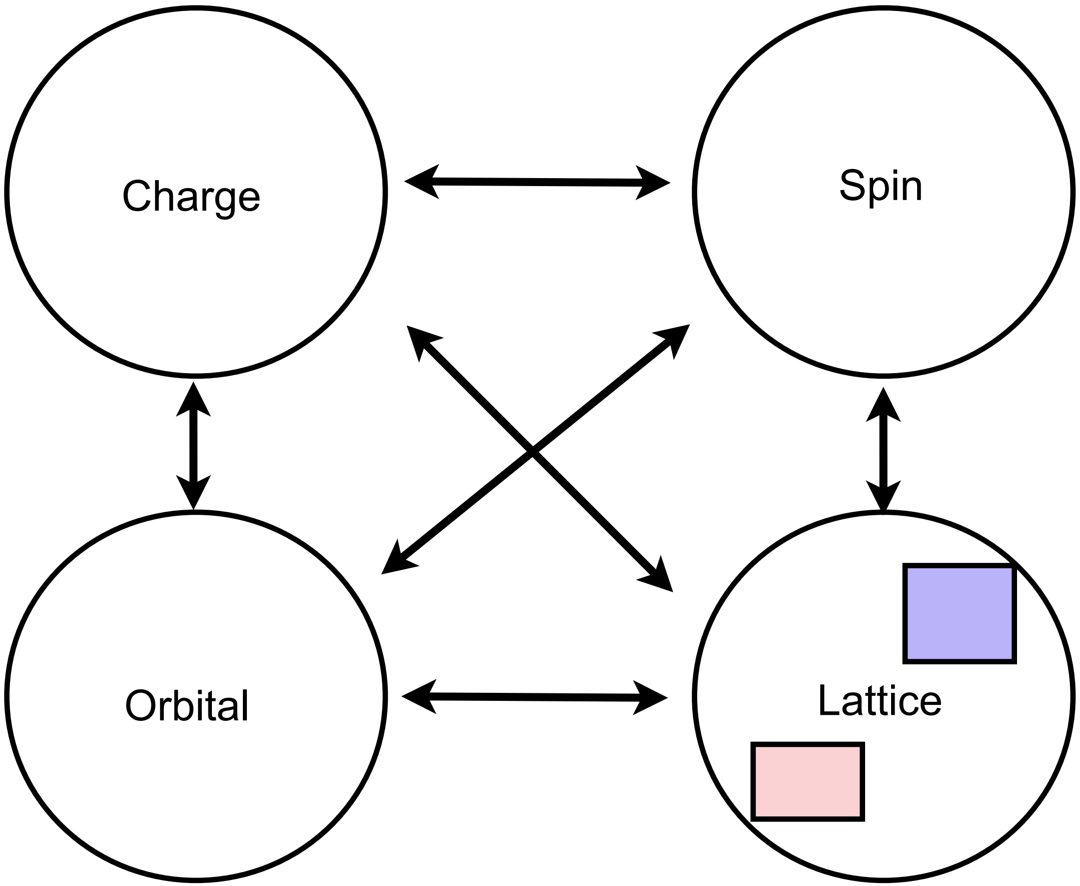
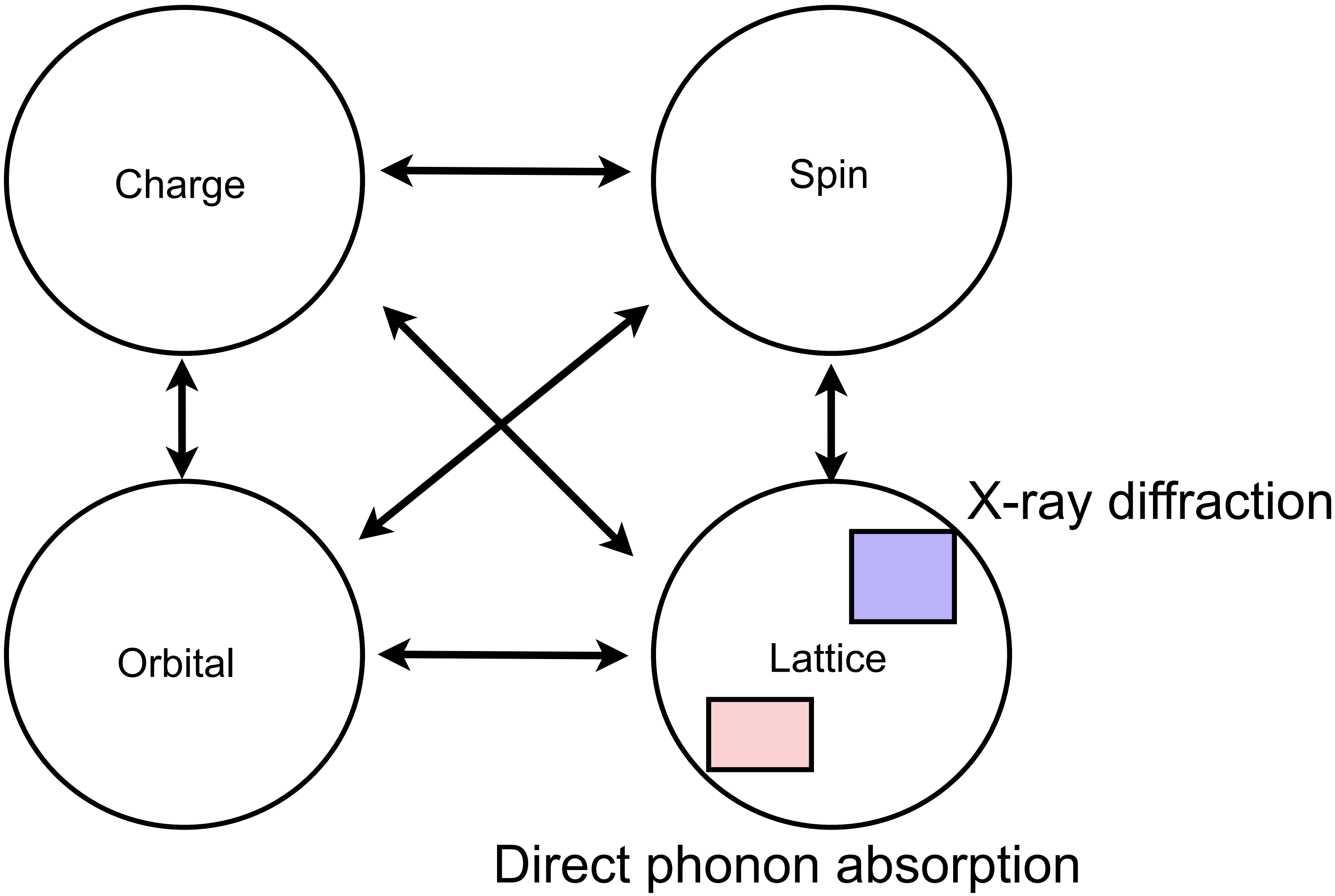
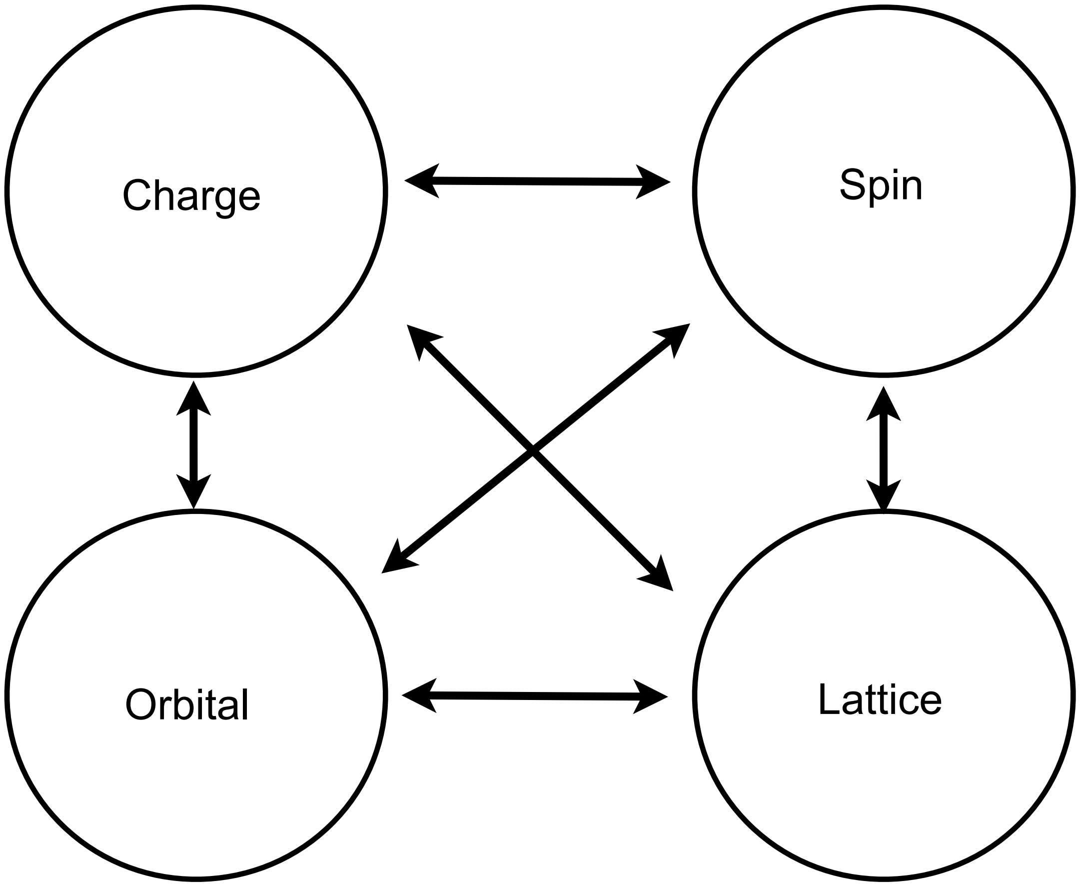

Charge
Spin
Orbital
Lattice



Direct phonon absorption


Charge
Spin
Orbital
Lattice
X-ray diffraction
Direct phonon absorption

Just the beginning!



- Effect of specific lattice modes on charge, spin & orbitals?
- Spin excitations?
- Plasmon (charge) excitations?

Pump characteristics

Direct lattice mode excitations (resonance)

- 2 - 50 THz (6-150 μm)
- 1-30% bandwidth (depends on mode)
- CEP stable: resolve dynamics within the cycle

Direct spin wave excitations similar

- 2-10 THz

Impulsive “kicking”

- Wide bandwidth, single cycle pulses

Driving plasmon resonances, phase modes

- < 1 THz, single-cycle

Orbital/charge excitations

- Visible/UV range, < 10 fs

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Sample environment

- Vacuum
- Temperature 5-500 K
- Electrical contacts
- Strong magnetic fields (> 1 T)
- Flexible sample & detector angles
    - Grazing incidence for bulk samples

Probe characteristics

- 4-12 keV
- Polarization control via phase plates
- Need effective time resolution of ~10 fs
  - Time arrival monitor is *essential*
- Monochromatic beam (0.01% BW)
- High stability of beam on sample
  - Presently the limiting factor in real experiments
  - I_0 often does not “see” critical instabilities in spectrum or pointing
  - May be best to control pointing with apertures
- I_0 with precision of better than 0.1% ???
- Variable focus down to < 5 microns in either direction

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A. Titov (RAS)

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M. Trigo (SLAC)

LCMO sample:
Q. Jia (LANL)

must

PCMO:
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M. Kawasaki,
Y. Tokura (U Tokyo)