Magnetic Excitations in Cuprates and Titanates

neutron scattering:
C. Ulrich, H. He, B. Keimer, MPI Stuttgart
P. Bourges, Y. Sidis, CEA Saclay
L.P. Regnault, CEA Grenoble
M. Reehuis, Hahn-Meitner Institut
A. Ivanov, M. Ohl, ILL
M. Reehuis, Hahn-Meitner Institut

crystals:
B. Liang, C.T. Lin, MPI Stuttgart
N. Kolesnikov, Russian Acad. of Sciences
S. Kondo, H. Takagi, Univ. Tokyo
Y. Taguchi, Y. Tokura, Univ. Tokyo

theory:
G. Khaliullin, MPI Stuttgart
S. Okamoto, RIKEN

Outline

1. Copper Oxides

Interaction between spin and charge excitations

1. Summary of neutron scattering work on
$La_{2-x}Sr_xCuO_4$ (single-layer, low T_C)
$YBa_2Cu_3O_{6+x}$, $Bi_2Sr_2CaCuO_{8+x}$ (bilayer, high T_C)

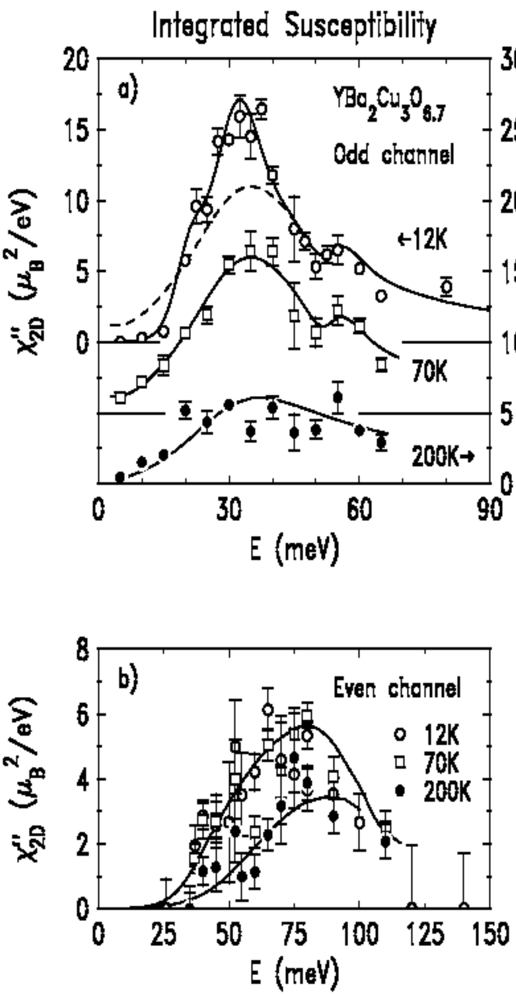
2. Neutron scattering from
$Tl_2Ba_2CuO_6$ (single-layer, high T_C) and
$La_{2-x}Sr_xCaCu_2O_6$ (bilayer, low T_C)

2. Titanium Oxides

Interaction between spin and orbital excitations

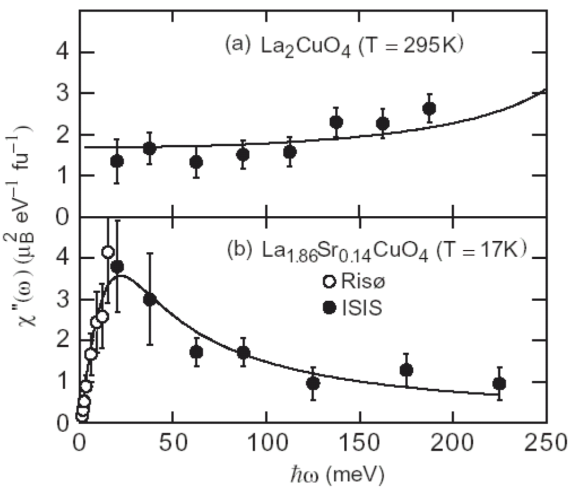
Q-Integrated Spin Susceptibility

underdoped YBCO



Fong et al., PRB 61, 14773 (2000)

opimally doped LSCO

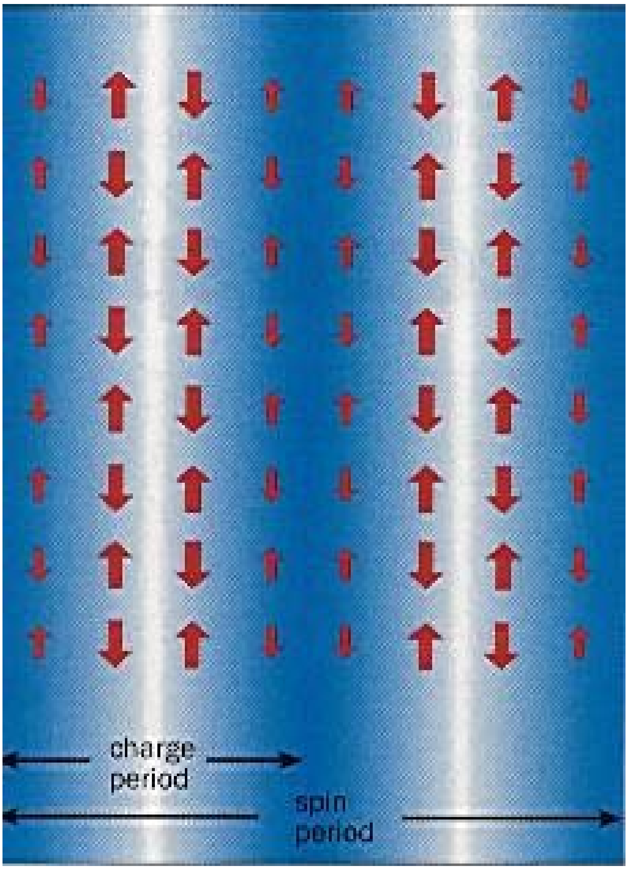


Hayden et al, Physica B 241, 765 (1998)

$\int d^2q\, \chi''(q, \omega)$

- comparable to spin waves in undoped Heisenberg antiferromagnet
- broadly similar in YBCO and LSCO
- qualitatively described as “overdamped spin waves”

Striped Phase in $La_{2-x}Sr_xCuO_4$

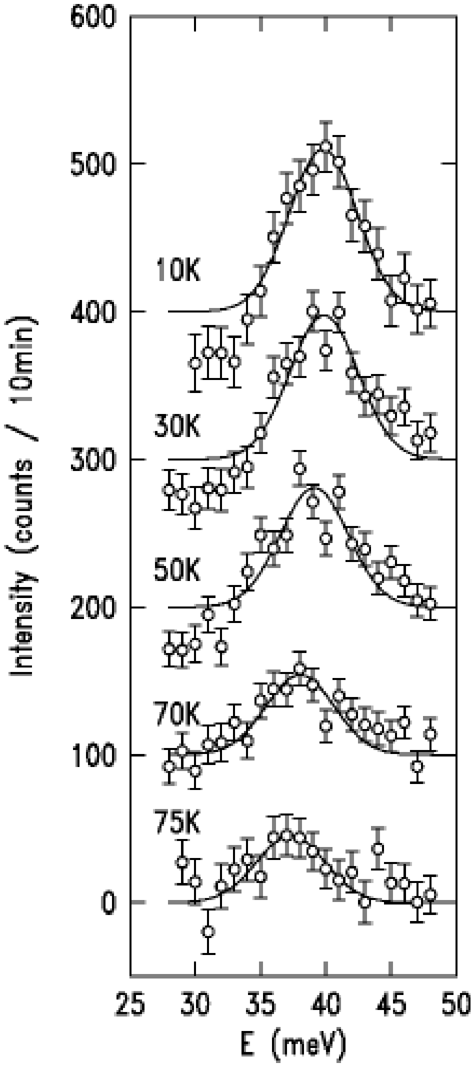


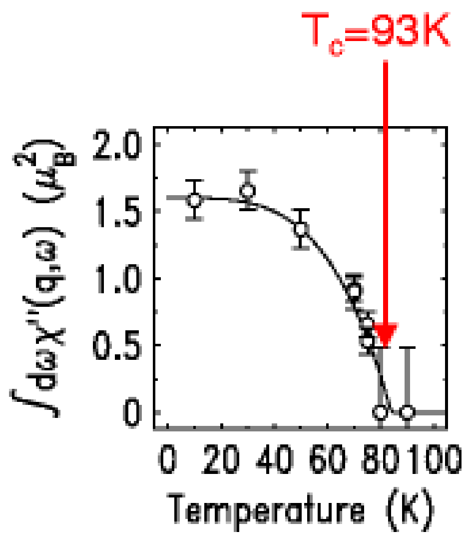
- *incommensurate* magnetic & lattice superstructure at x~1/8 in $La_{2-x}Sr_xCuO_4$:Nd

- low energy excitations with same incommensurate geometry in $La_{2-x}Sr_xCuO_4$: fluctuating stripes?

- modification of $\chi''(q, \omega)$ below T_C only at low energies, no resonant mode

Resonant Mode in $YBa_2Cu_3O_7$

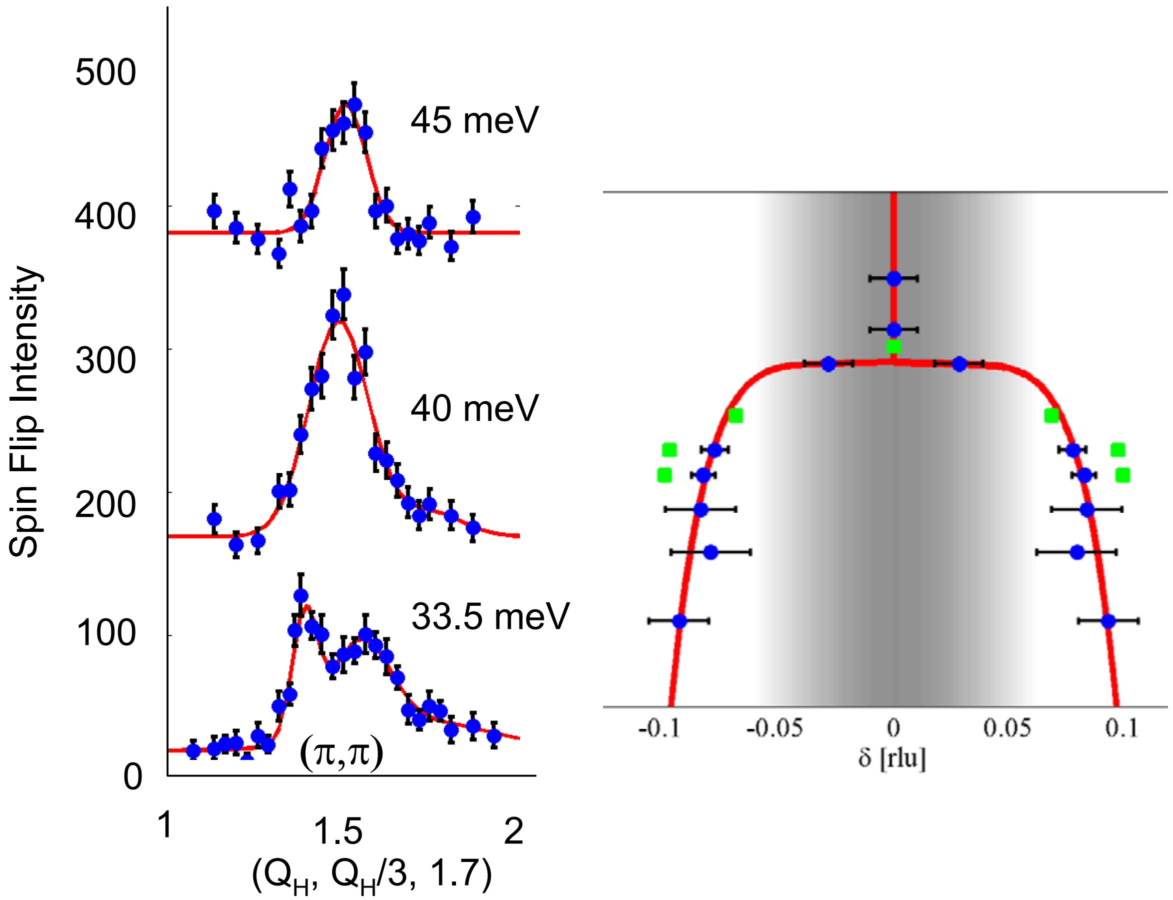




- spectral weight concentrated around commensurate wave vector q=(π,π)
- comparable to spin waves in undoped $YBa_2Cu_3O_{6+x}$
- induced by superconductivity

H.F. Fong et al, Phys. Rev. Lett. 75, 316 (1995);
Phys. Rev. B 54, 6708 (1996)

Resonant Mode Dispersion

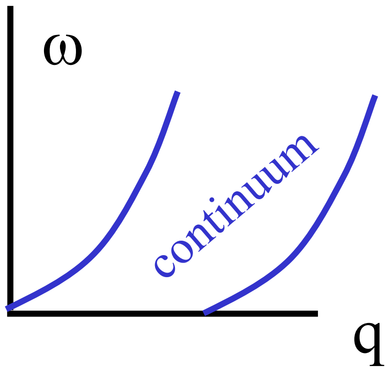


incommensurate excitations below resonant mode:

- low spectral weight
- same T-dependence as resonant mode
- best interpreted as "downward" mode dispersion

P. Bourges et al., Science 288, 1234 (2000)

RPA Model

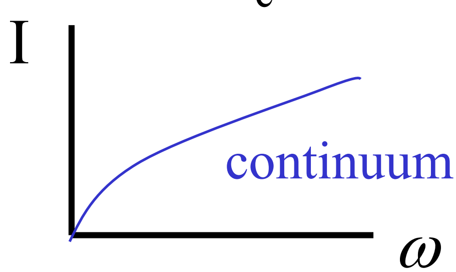


starting point:
band susceptibility $\chi_0(q,\omega)$

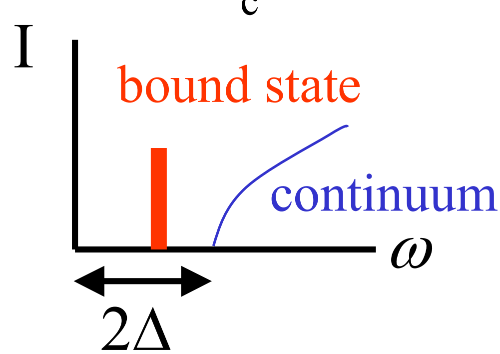
renormalization of $\chi_0(q,\omega)$ by interactions:

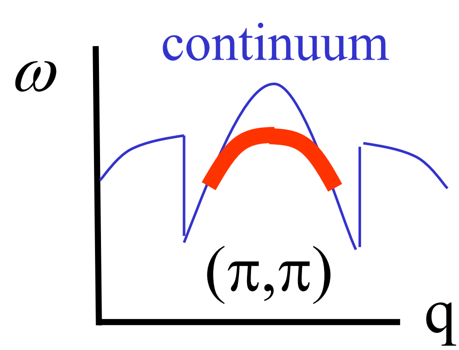
$$\text{Stoner model: } \chi(q,\omega) = \frac{\chi_0(q,\omega)}{1 - J(q)\,\chi_0(q,\omega)}$$

$T > T_c$

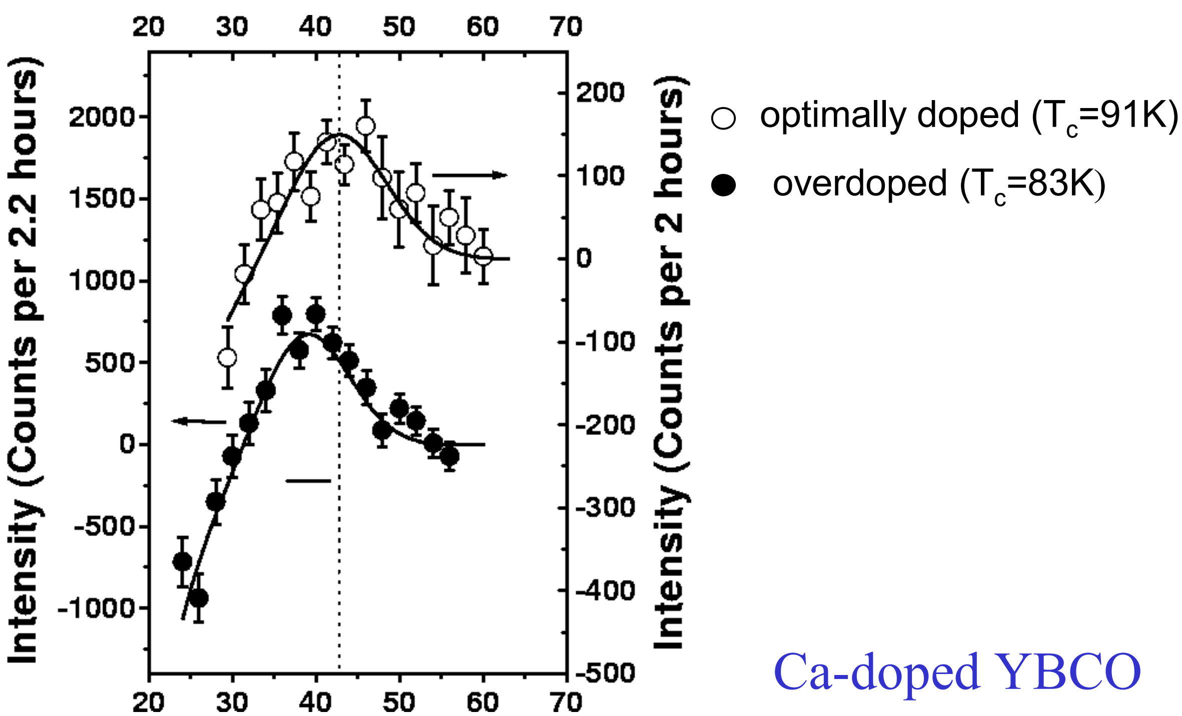


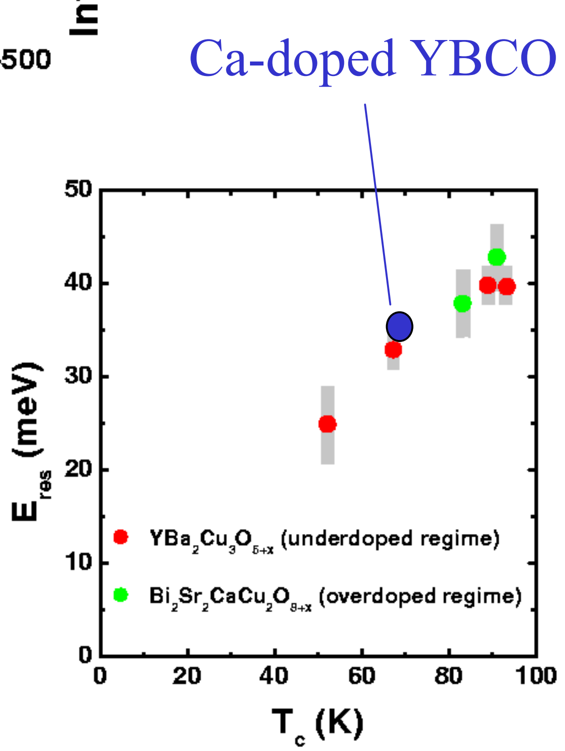
$T < T_c$





Resonant Mode in $Bi_2Sr_2CaCu_2O_{8+x}$

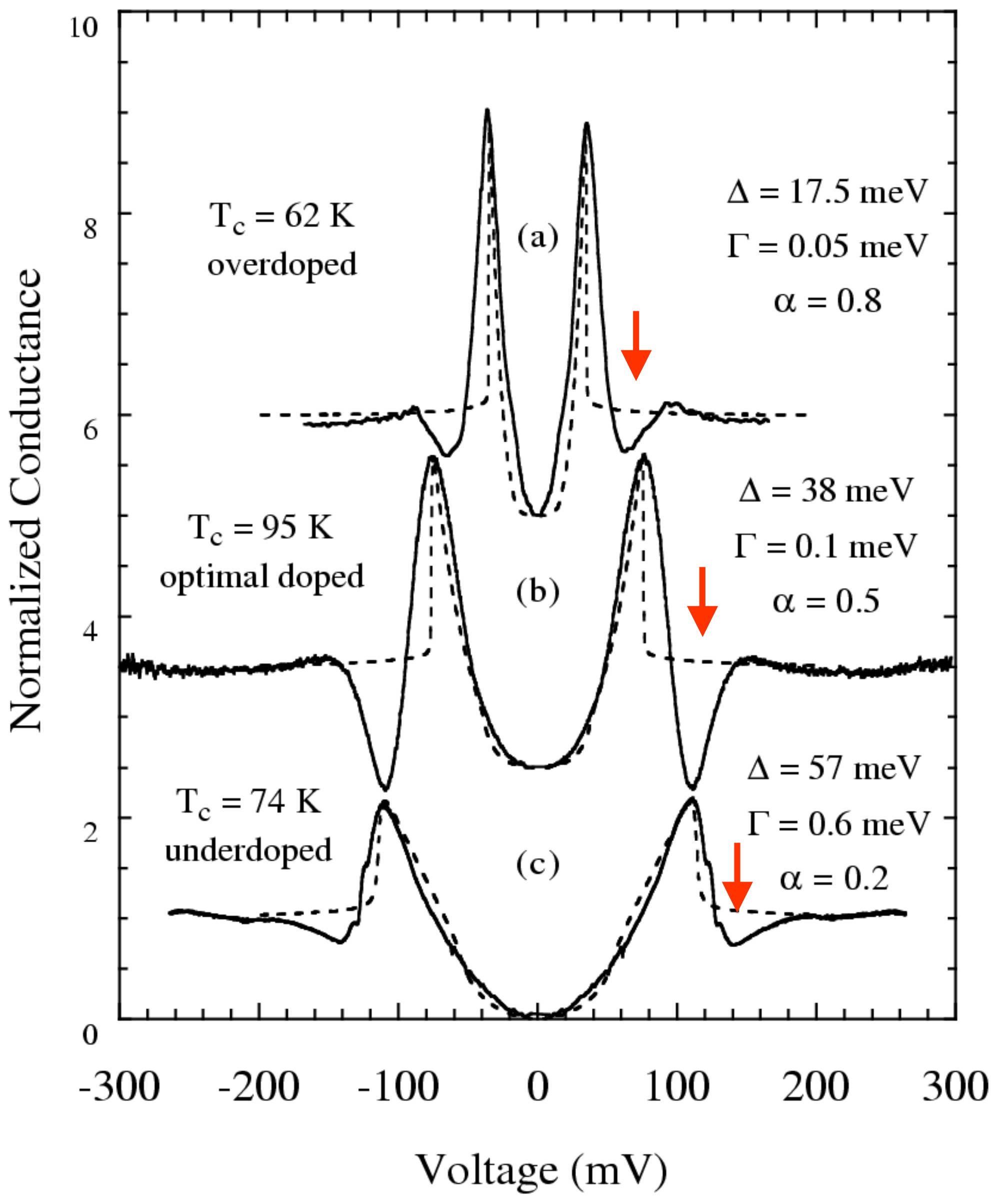




- resonant mode present in two bilayer cuprates with distinct crystal structures and doping mechanisms
- mode energy proportional to T_c in both underdoped and overdoped regimes

H.F. Fong et al., Nature 398, 588 (1999);
H. He et al., Phys. Rev. Lett 86, 1610 (2000)

C-axis tunneling spectra of $Bi_2Sr_2CaCuO_{8+x}$

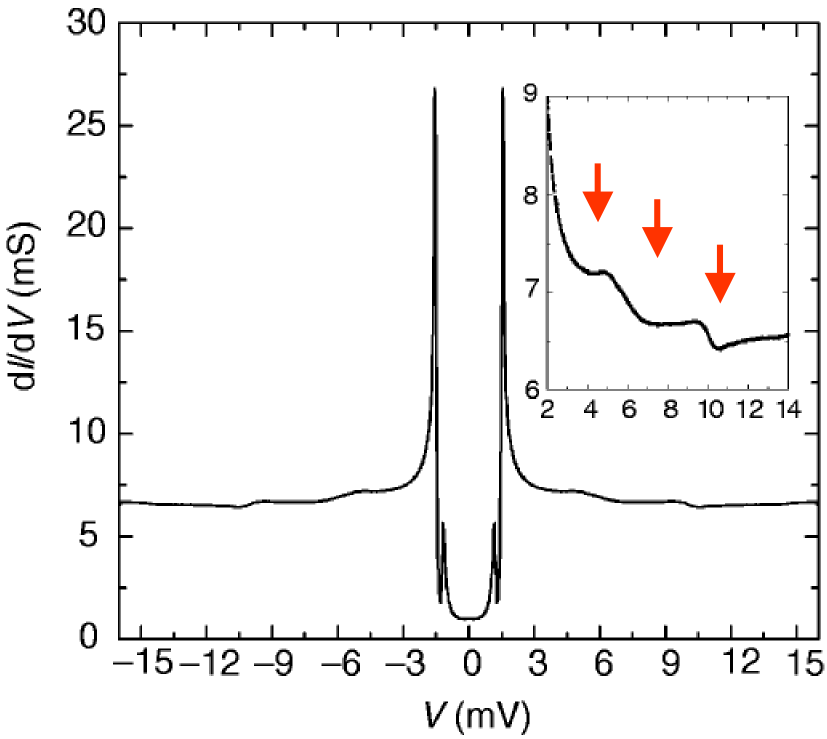


Zasadzinski et al., PRL 2001

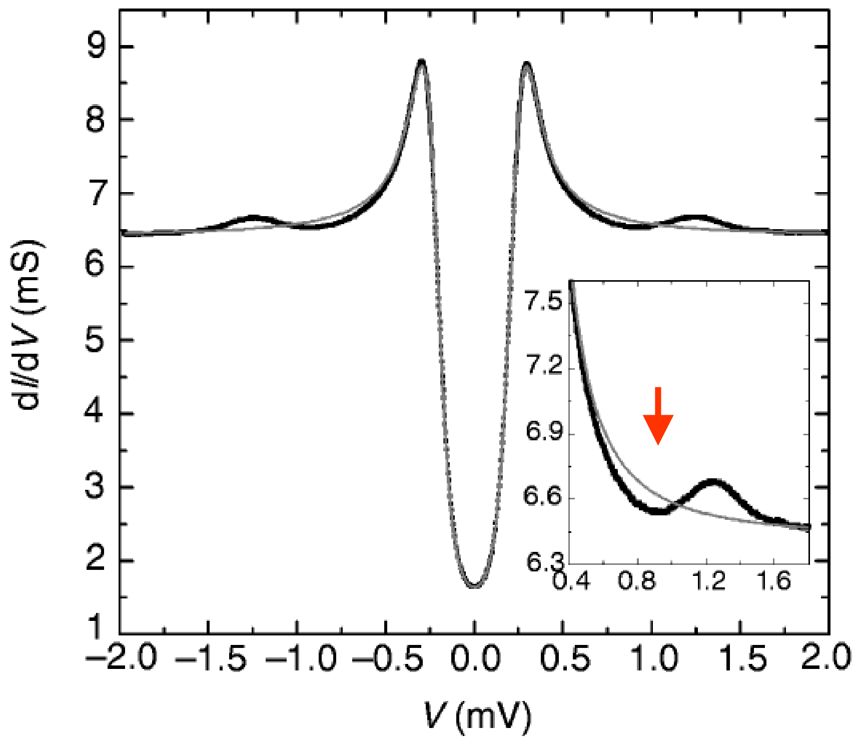
similar peak-dip-hump structure seen in ARPES spectra at $(\pi,0)$

c-axis tunneling matrix element $\sim (\cos k_x - \cos k_y)^2$

UPd_2Al_3 –Al_2O_3 –Pb tunnel junction (T=0.3K)



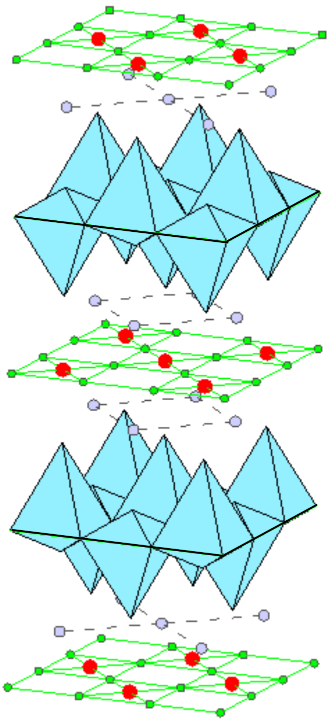
Pb phonons



UPd_2Al_3
spin excitation

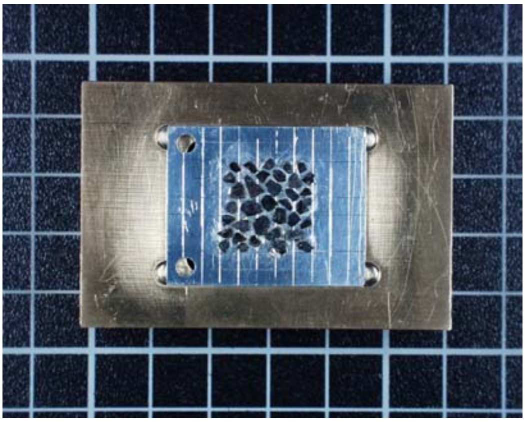
M. Jourdan et al., Nature (99)

$Tl_2Ba_2CuO_6$

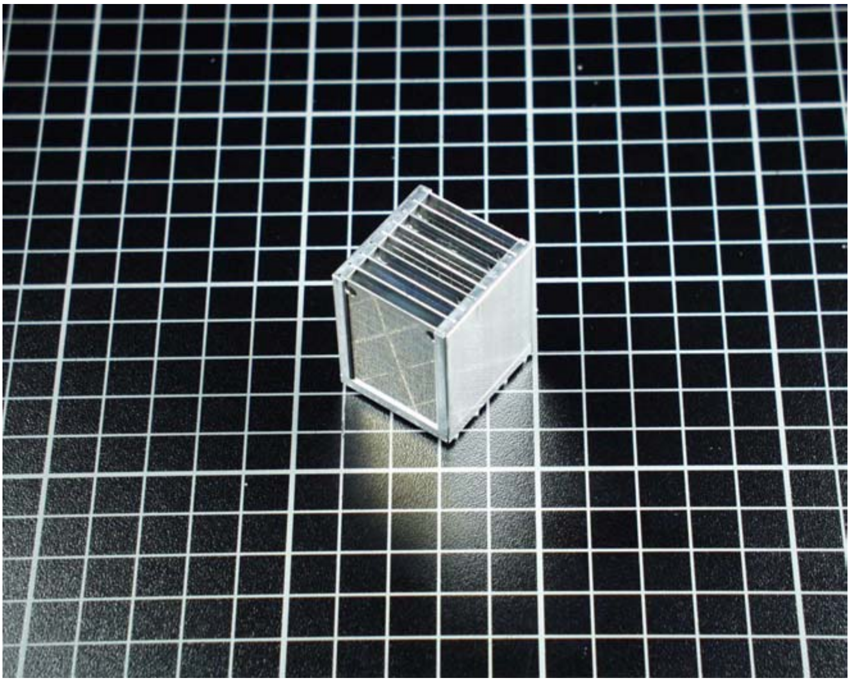


- single layer structure
- tetragonal, unbuckled layers
- high maximum T_c (93K)

Sample Preparation

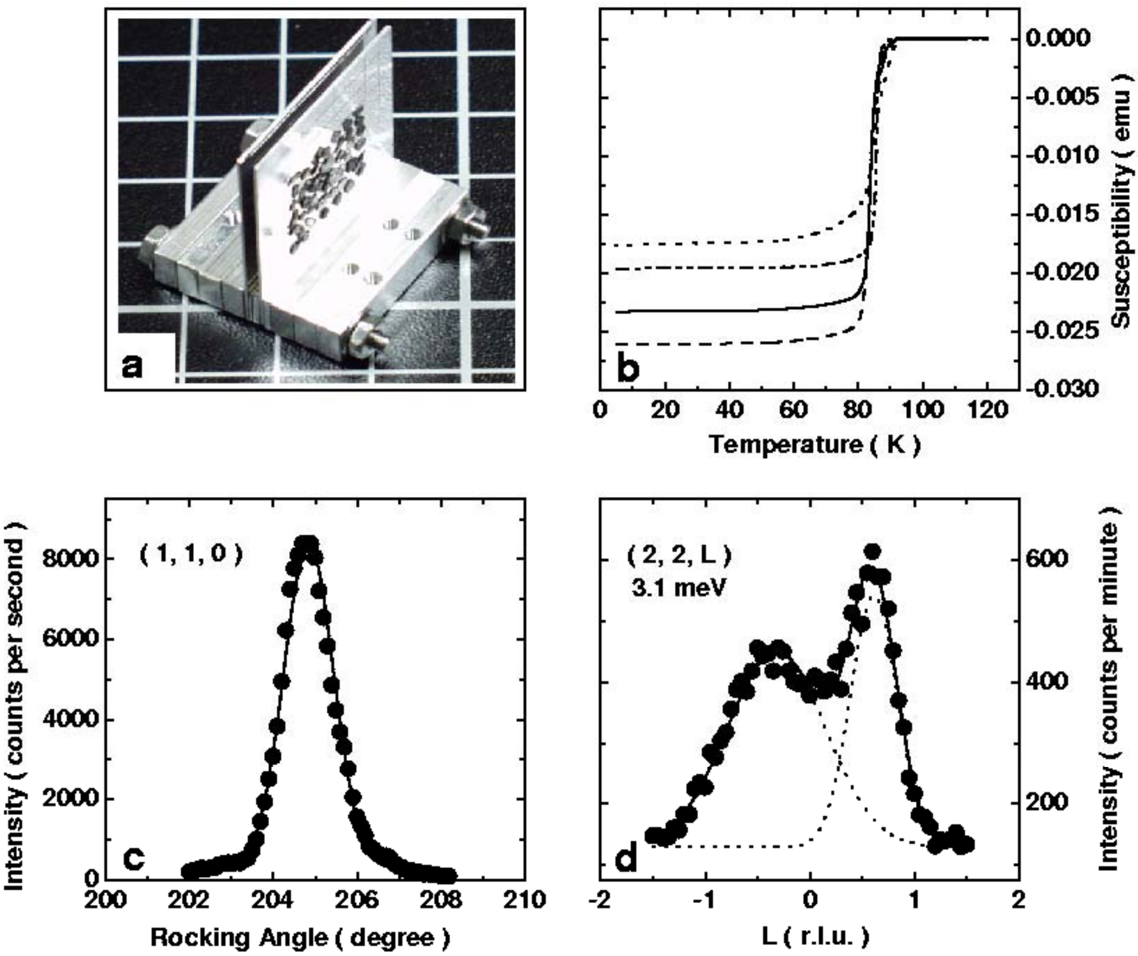


$Tl_2Ba_2CuO_{6+x}$ single crystals



stacked plates with aligned crystals (> 300)

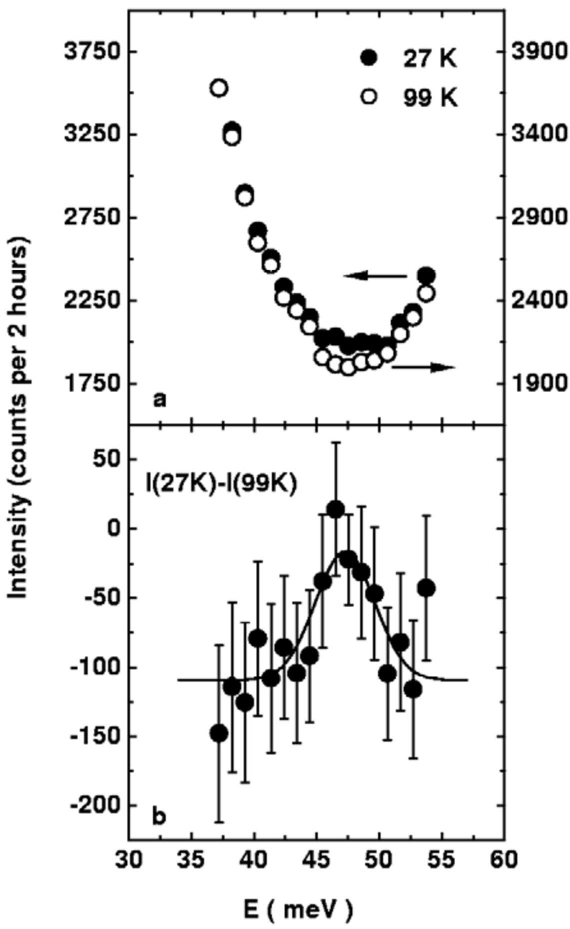
Sample Characterization



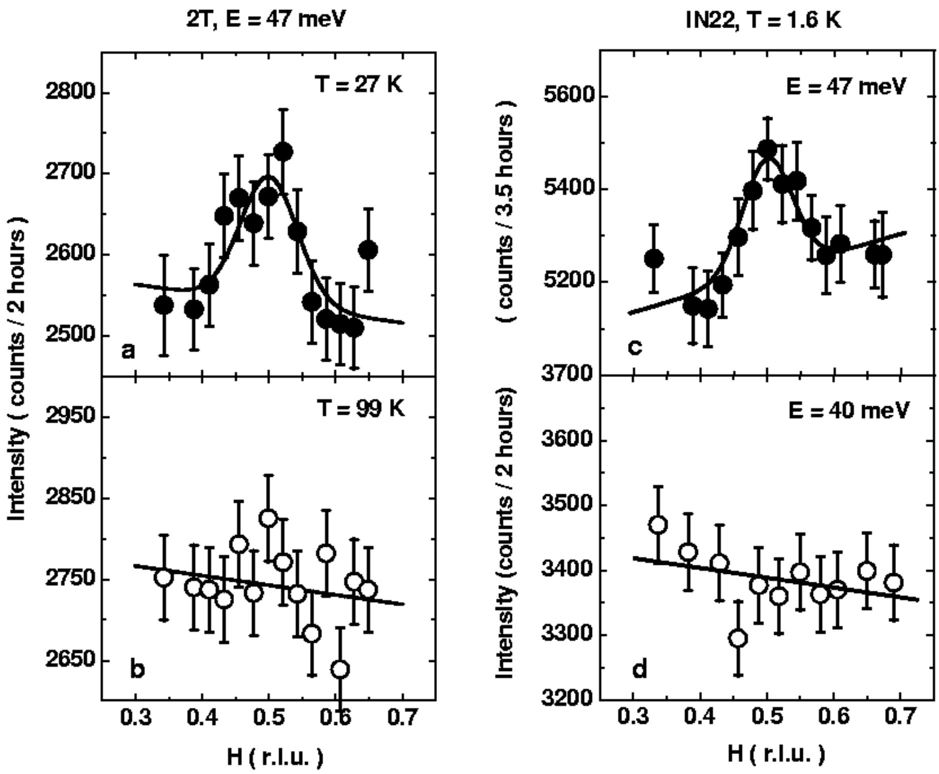
H. He et al., Science 295, 1045 (2002)

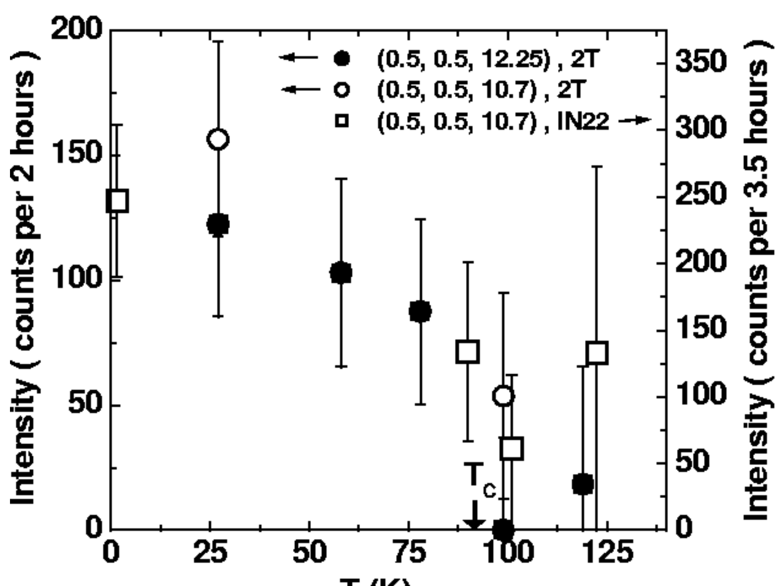
$Tl_2Ba_2CuO_6$

$q = (\pi,\pi)$



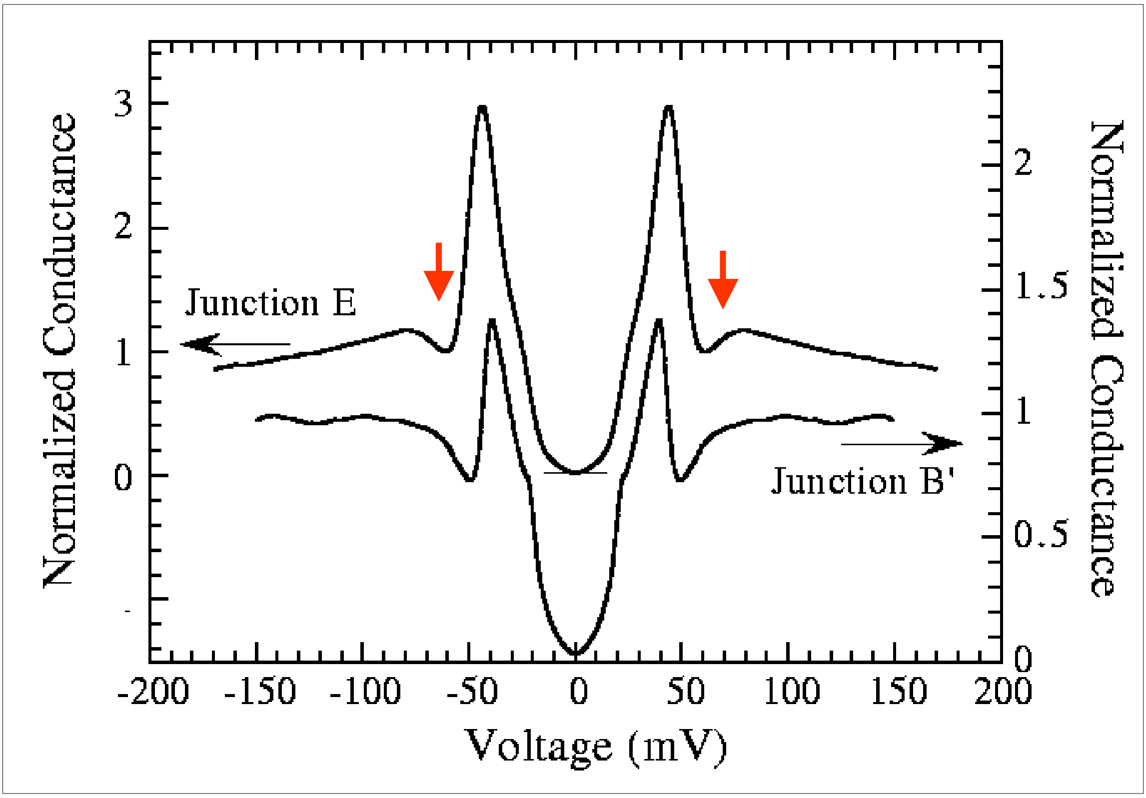
additional intensity at $q = (\pi,\pi)$, $E = 47$ meV
in superconducting state





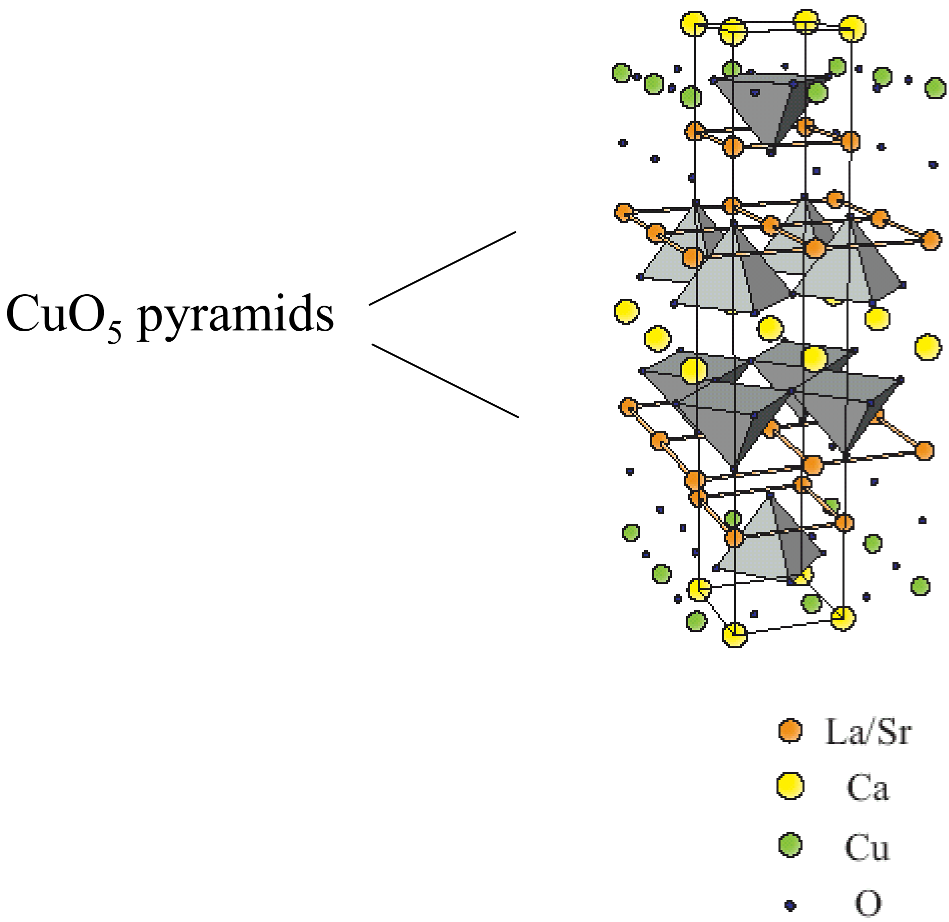
intensity sharply centered at $q_{||} = (\pi,\pi)$, E = 47 meV
independent of $q_\perp$
spectral weight comparable to $YBa_2Cu_3O_7$
disappears above T_C
⇒ **magnetic resonant mode in single-layer cuprate**

Tl-2201 tunnel spectrum



Ozyuzer et al., Physica C 320, 9 (1999)

$La_{2-x}Sr_xCaCu_2O_6$



- double-layer version of $La_{2-x}Sr_xCaCu_2O_6$

- tetragonal crystal structure

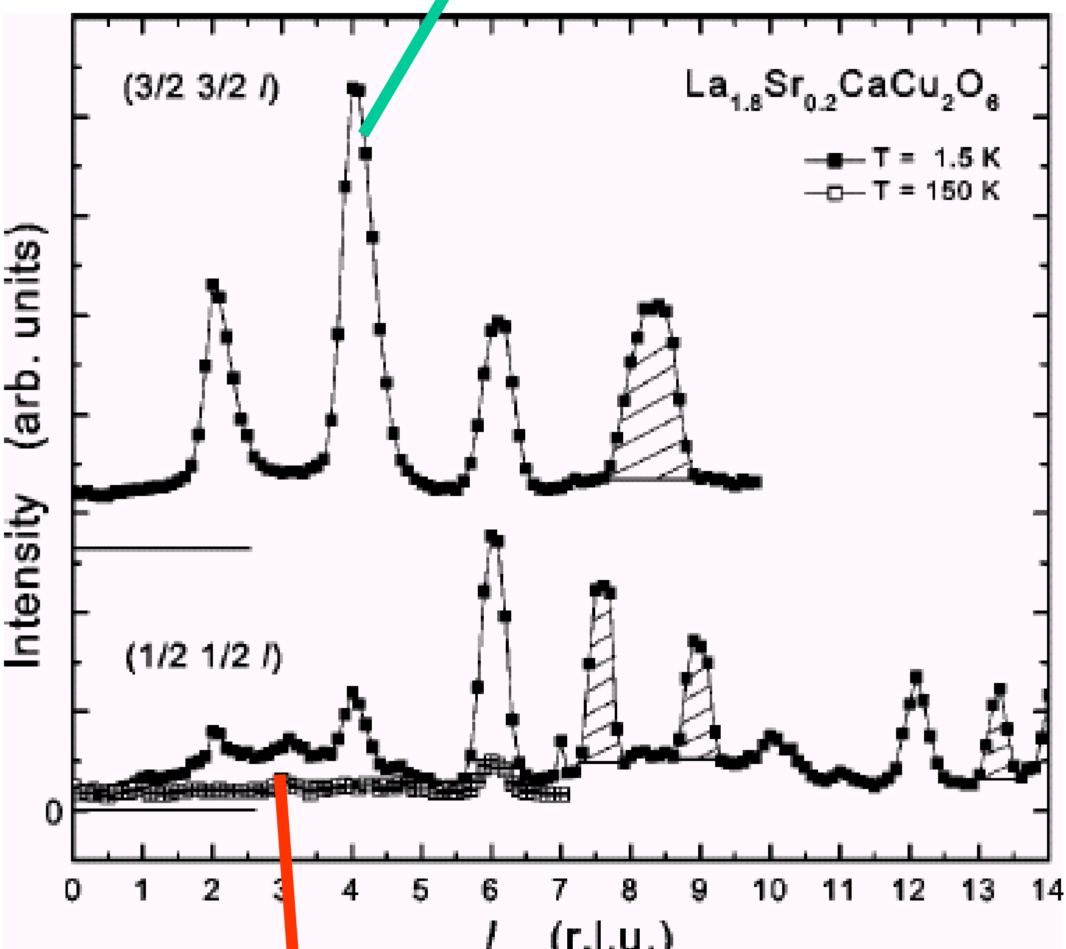
⇒ “simplest bilayer cuprate”

- $T_{C\,max}$ = 60K after high pressure annealing

$La_{1.8}Sr_{0.2}CaCu_2O_6$ (nonsuperconducting)

two types of ***commensurate*** superstructure reflections:

l=even: nuclear superstructure



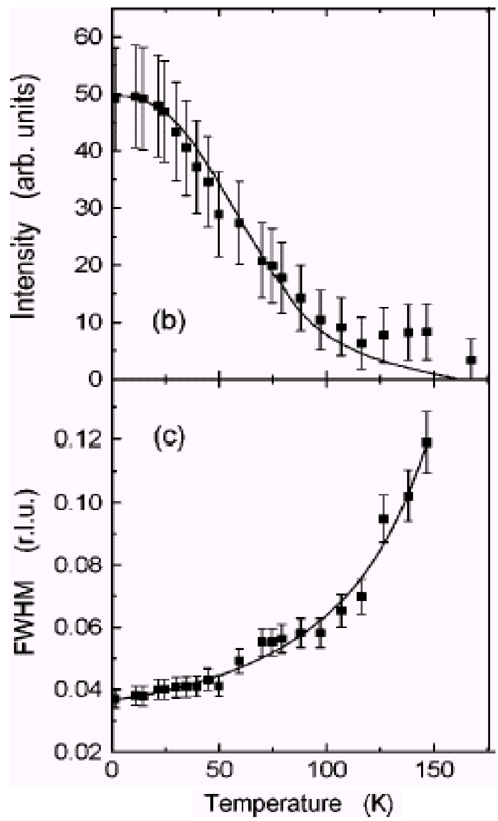
Ulrich et al., PRB65, 220507(R)

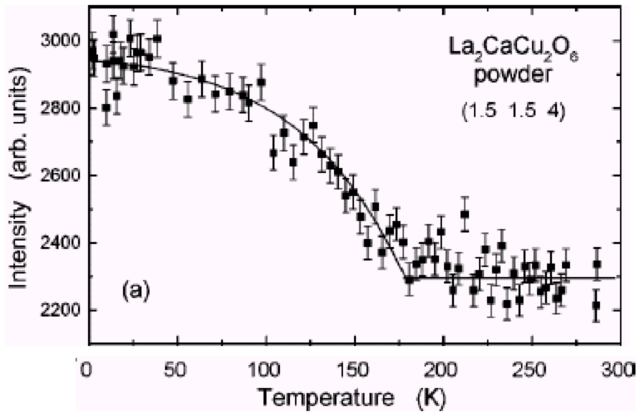
l=odd: magnetic superstructure

Lattice Superstructure

tetragonal-orthorhombic transition, analogous to $La_{2-x}Sr_xCuO_4$

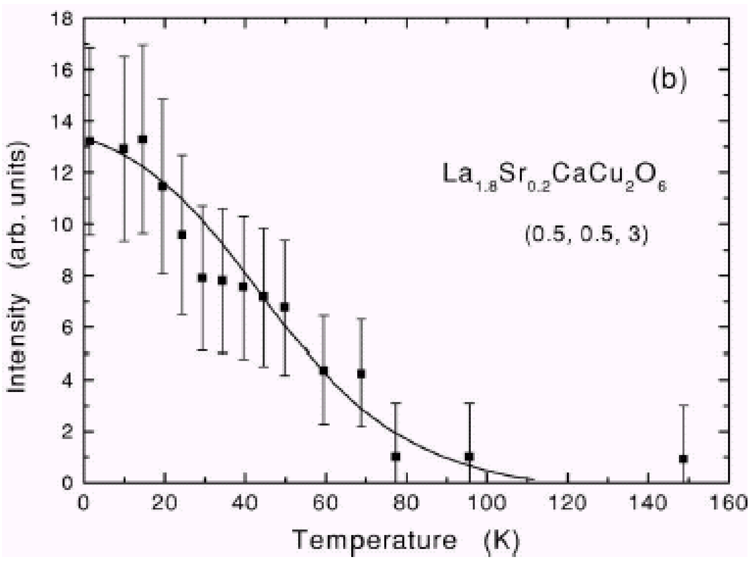
strongly disordered in $La_{1.8}Sr_{0.2}CaCu_2O_6$



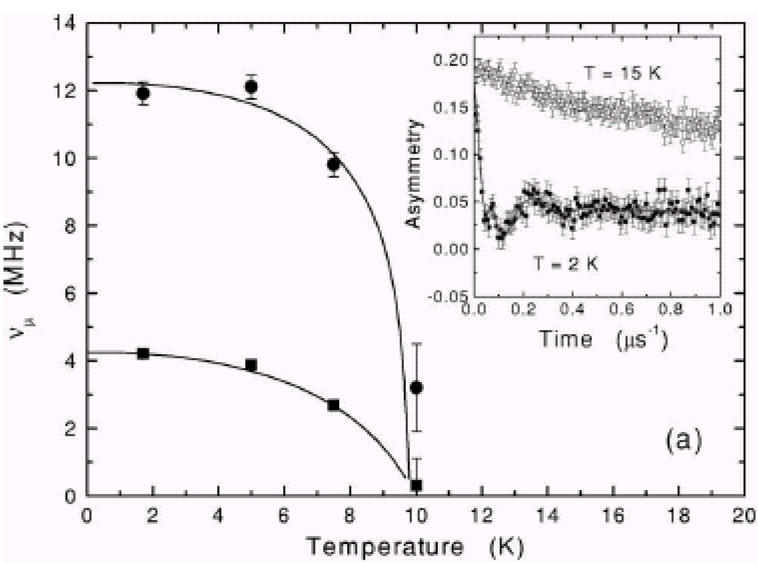


also present in undoped $La_2CaCu_2O_6$
⇒ intrinsic instability of lattice,
not induced by charge localization

Magnetic Superstructure



neutrons
$T_F = 60K$



μSR
$T_F = 10K$

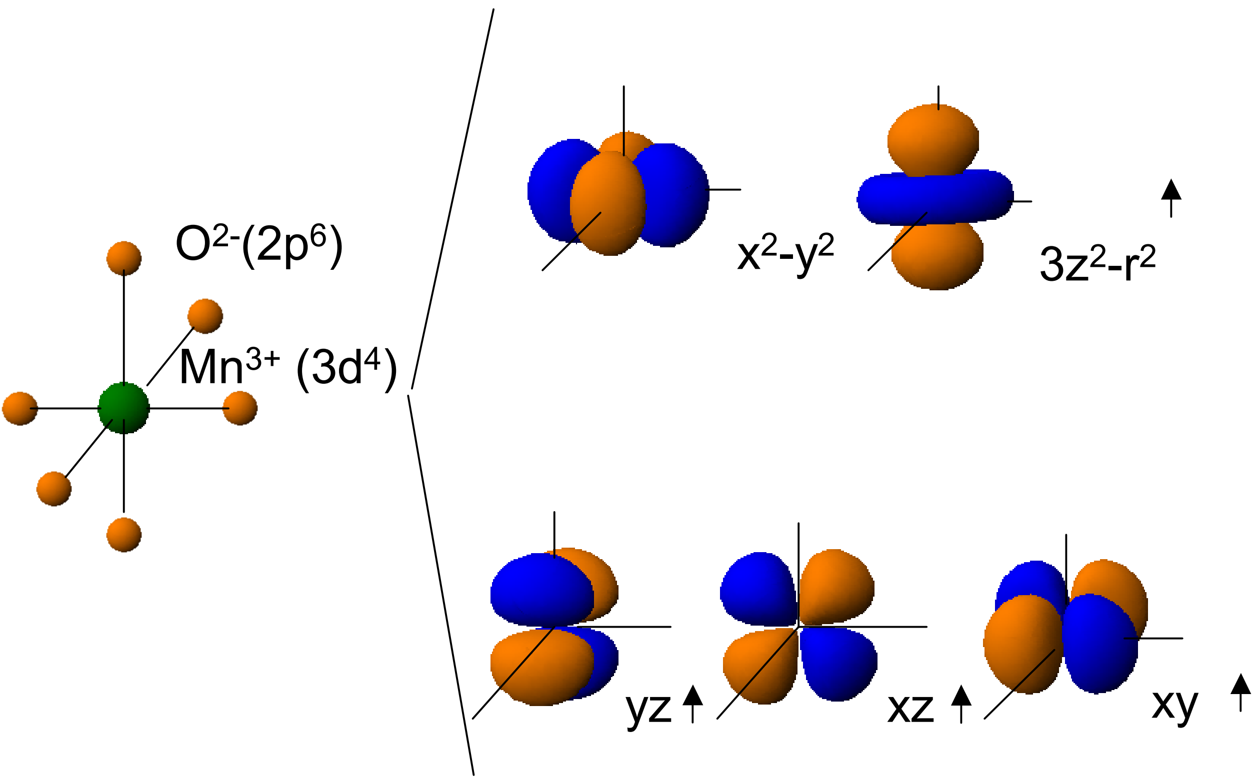
- gradual spin freezing, analogous to underdoped $La_{2-x}Sr_xCuO_4$
- commensurate, despite nominal hole concentration 0.1 per copper

Conclusions

- progress in understanding influence of lattice type on spin dynamics
- commensurate magnetic resonant mode generic to all cuprates with high T_C
- disorder and/or striped phase responsible for suppression of resonant mode and superconductivity in LaSr-based cuprates

Orbital degeneracy

Example: $LaMnO_3$

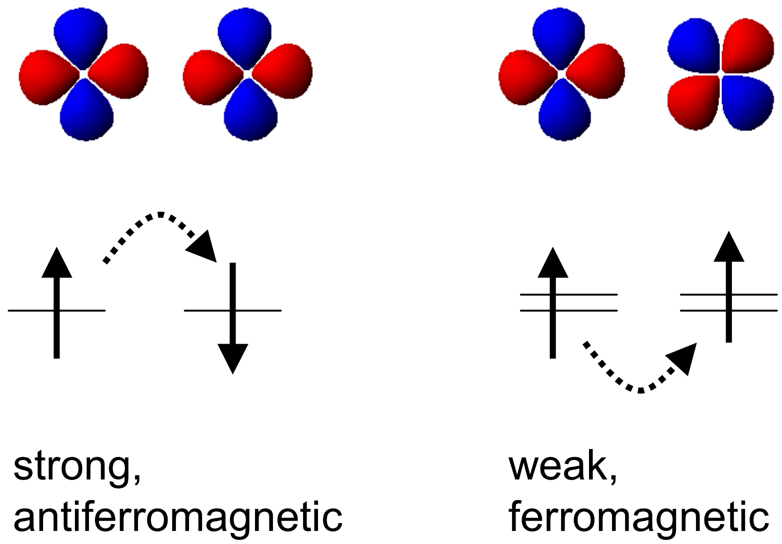


Superexchange Interaction

$$H = \sum_{ij} [J_{ij}(\tau_i, \tau_j) S_i \bullet S_j + K_{ij}(\tau_i, \tau_j)]$$

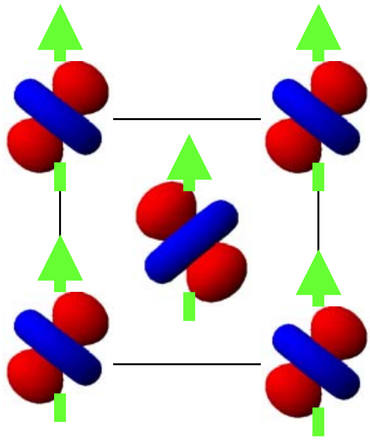
τ_i, τ_j orbital pseudospin,
same commutation relations as S_i, S_j

example:

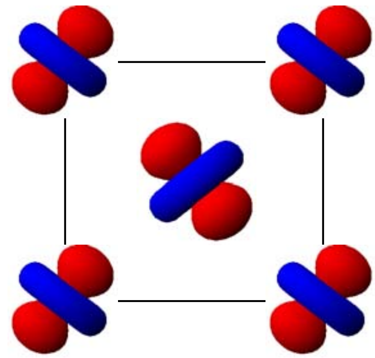


Goodenough-Kanamori Rules

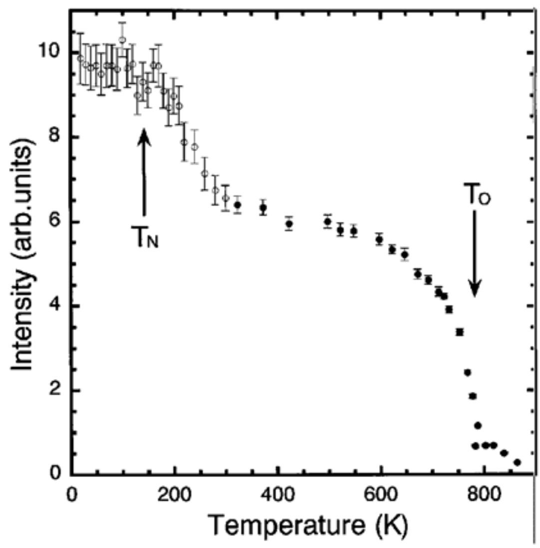
$LaMnO_3$ (S=2, e_g orbitals)



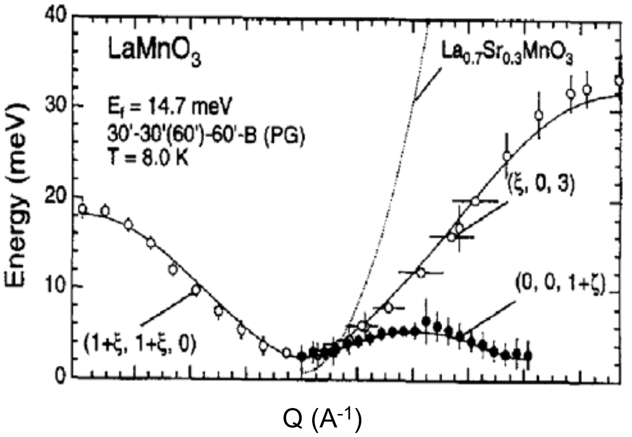
$T<T_N$: magnetic order



$T<T_O$: orbital order
locks in exchange interactions



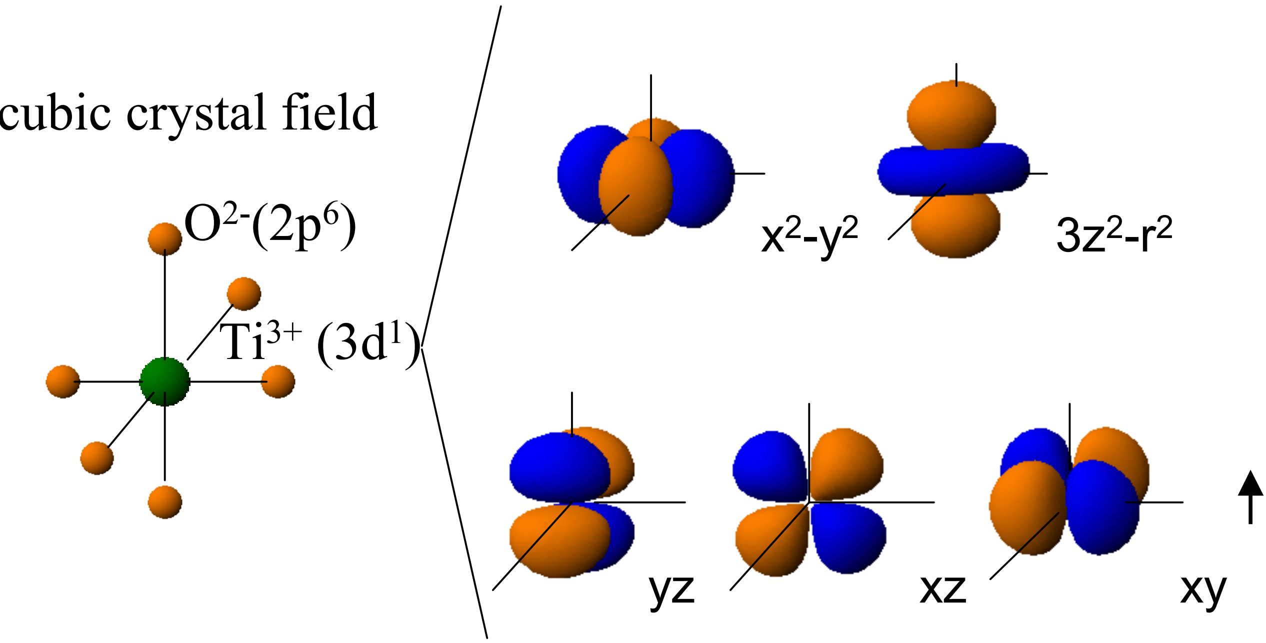
Murakami et al.



$T<T_N$: anisotropic spin wave spectrum
reflecting orbital ordering pattern

Hirota et al.

$3d^1$ System: Titanates



Mechanisms to lift orbital degeneracy

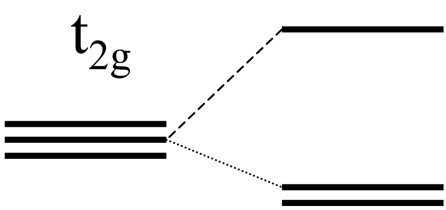
- Jahn-Teller effect
- superexchange
- spin orbit coupling

Jahn-Teller distortions in t_{2g}-systems $\Delta d \sim 0.05$ Å
compare to e_g-systems (manganites): $\Delta d \sim 0.25$ Å

Jahn-Teller energy scale $\propto \Delta d^2$
much smaller than in manganites

$LaTiO_3$: no observable Jahn-Teller distortion

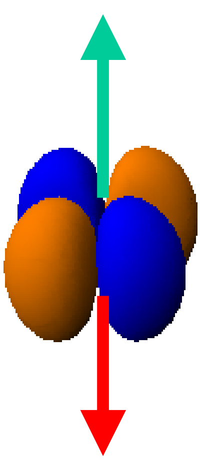
spin-orbit coupling
$\lambda \sim 20$ meV



$|xz\uparrow\rangle + i\,|yz\uparrow\rangle - i\,|xy\downarrow\rangle$

$|xz\uparrow\rangle + i\,|yz\uparrow\rangle + 2i\,|xy\downarrow\rangle$
$|xz\uparrow\rangle - i\,|yz\downarrow\rangle$

- orbital ordering with **complex** combinations of wave functions
- **unquenched** orbital angular momentum

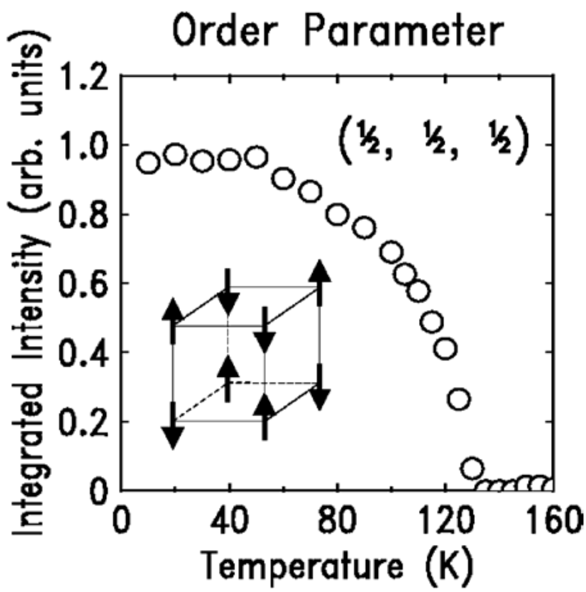


orbital moment
(locked into crystal lattice)

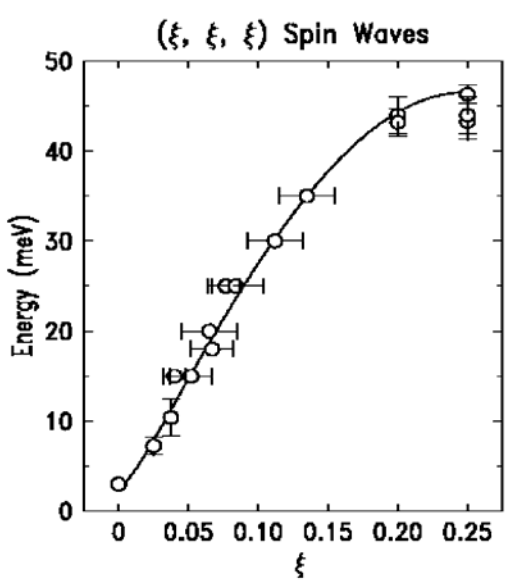
spin moment
(coupled to orbital moment
by spin-borbit interaction)

Expect **strongly anisotropic** spin dynamics
(large anisotropy gap in spin wave spectrum)

Neutron scattering from Insulating $LaTiO_3$



- G-type antiferromagnetic order inconsistent with all electronic structure calculations
- small ordered moment ($0.45\mu_B$)

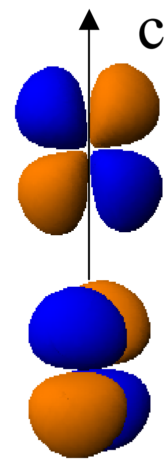


isotropic spin waves

Keimer et al., PRL 2000

Theory of $LaTiO_3$

$\Delta d < 0.01$ Å ⇒ negligible Jahn-Teller coupling



Superexchange Hamiltonian:
two active orbitals on every bond

for c-axis bond:

$$H_{SE} = J_{SE} \sum (S_i \cdot S_j + \tfrac{1}{4})(\tau_i \cdot \tau_j + \tfrac{1}{4} n_i n_j)$$

$$J_{SE} = \frac{t^2}{U}$$

$S = \tfrac{1}{2}$; $\quad \tau = \tfrac{1}{2}$ in xz, yz subspace

$\langle \mathrm{n} \rangle = \tfrac{2}{3}$

also need to consider Hund's rule,
spin-orbit interactions

Theory of $LaTiO_3$

Orbital liquid model (Khaliullin & Maekawa)

correlated spin-orbital fluctuations:
(spin singlet)×(orbital triplet) ⇔ (spin triplet)×(orbital singlet)
⇒ orbital order obliterated

consequences:
with fixed parameter J = 15.5 meV from neutron scattering

- ordered moment 0.5 μ_B
- spatially isotropic spin dynamics
- spin gap 3 meV

} as observed

- continuum of fermionic orbital excitations   not yet observed

Conventional orbital order (Imada et al., Khomskii et al.)

special linear combination of orbitals
⇒ subtle crystallographic distortions   not yet observed

$YTiO_3$

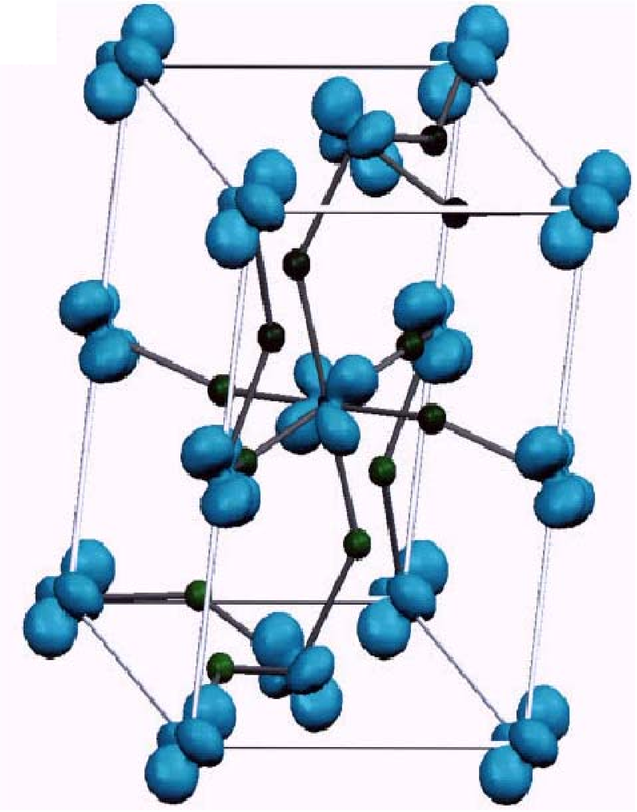
- smaller O-Ti-O bond angle than $LaTiO_3$
  weakened superexchange

- spin ferromagnetism as predicted by
  electronic structure calculations

Orbital order:

$$\left|\psi\right\rangle_{1,3} = c_1\left|yz\right\rangle \pm c_2\left|xy\right\rangle$$

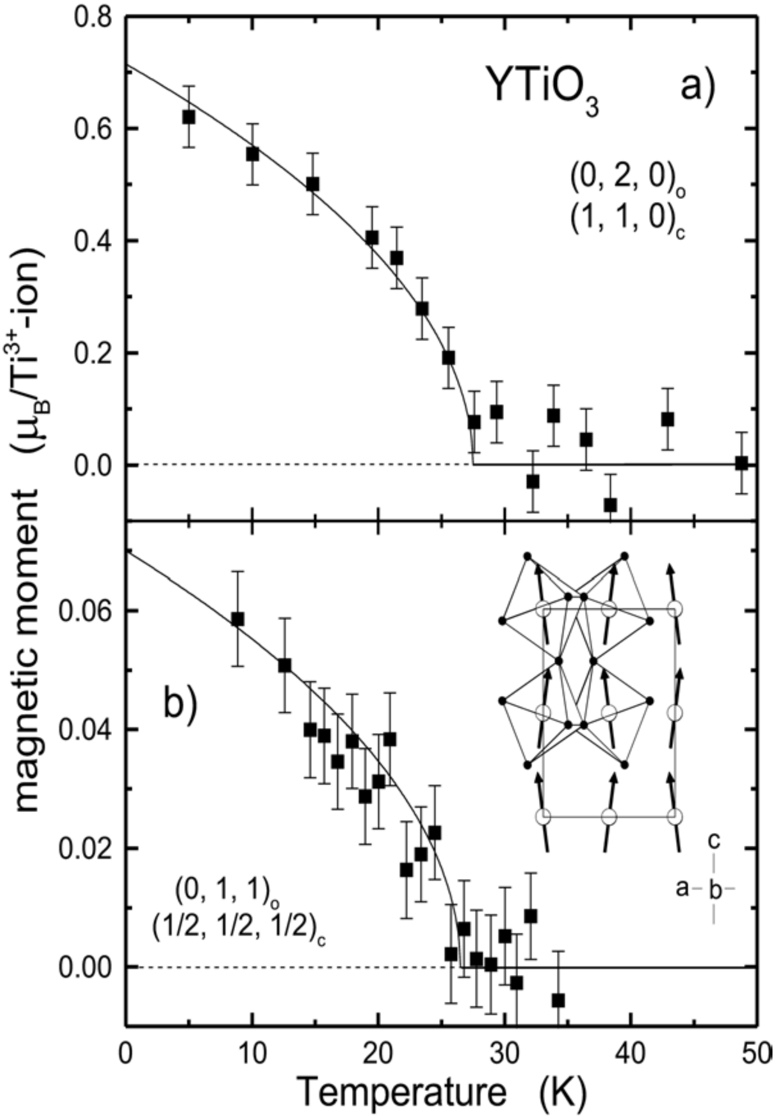
$$\left|\psi\right\rangle_{2,4} = c_1\left|xz\right\rangle \pm c_2\left|xy\right\rangle$$



Theory:
Sawada & Terakura
Mizokawa et al.
...

Experiment:
Akimitsu et al. (neutrons)
Itoh et al. (NMR)
...

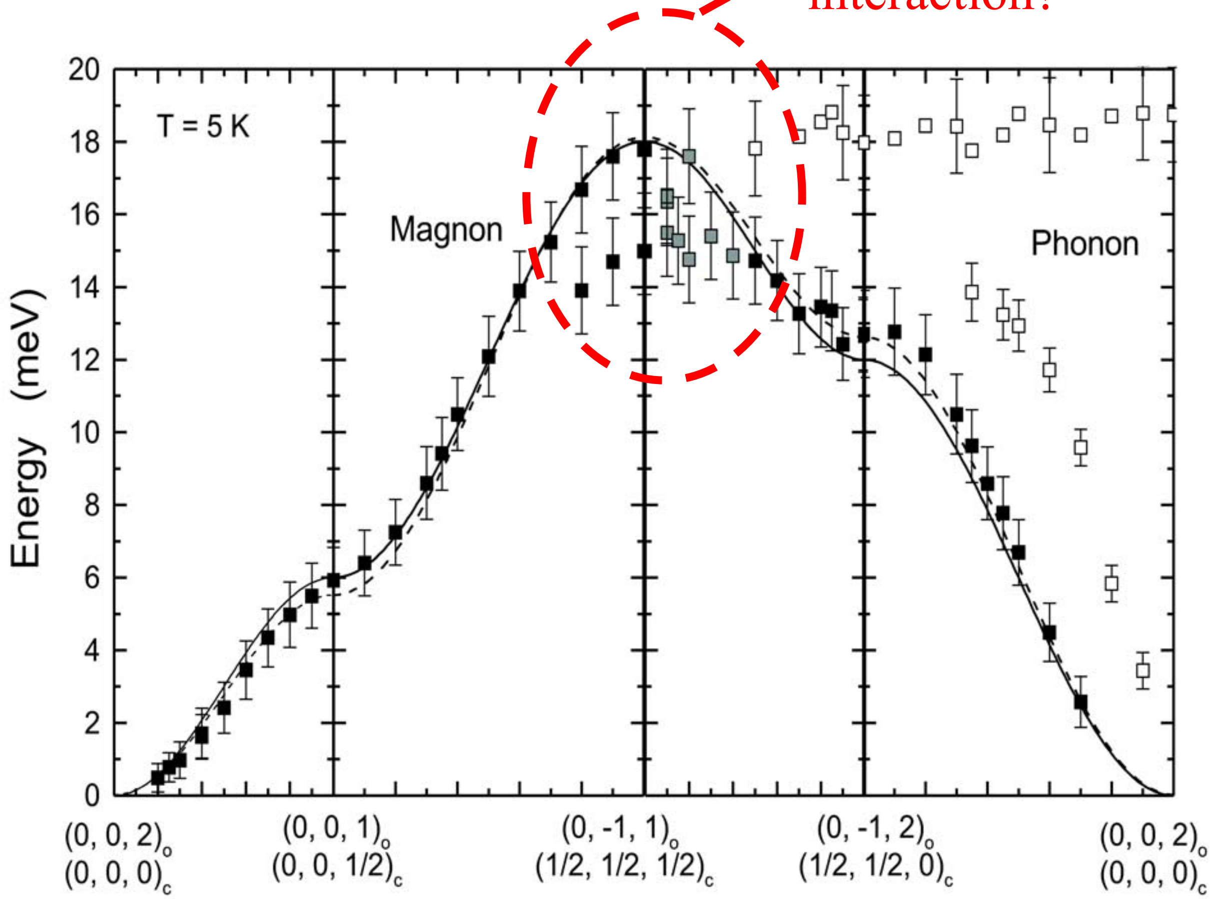
Elastic Neutron Scattering from $YTiO_3$



ferromagnetism with G-type and A-type antiferromagnetic canting, canting angle $\phi \sim 5^o$

Inelastic Neutron Scattering from $YTiO_3$

magnon-phonon interaction?



—— fit to Heisenberg model with **isotropic** ferromagnetic exchange

$E = 6SJ\,(1 - \gamma_q)$ $J = 3.0$ meV

- $J_c = J_{ab} = J$ identical within ~ 5%
- T_C calculated from J consistent with experiment
- magnon gap < 0.2 meV

Calculation of Exchange Parameters

orbital state :

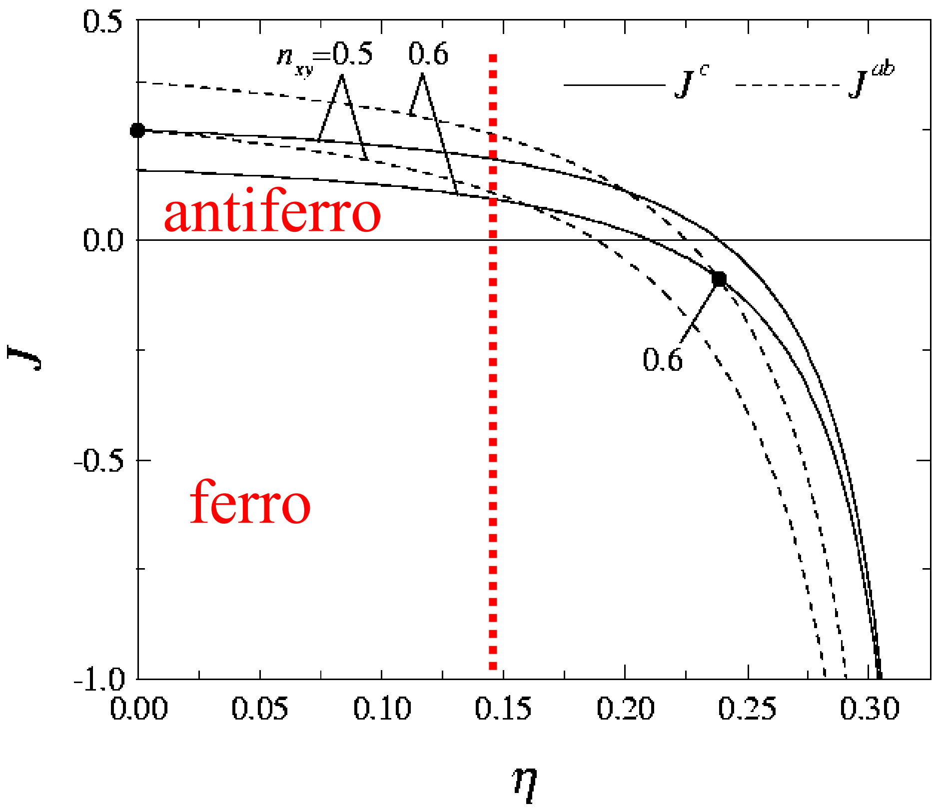
$$|\psi\rangle_{1,3} = c_1|yz\rangle \pm c_2|xy\rangle$$

$$|\psi\rangle_{2,4} = c_1|xz\rangle \pm c_2|xy\rangle$$

$$n_{xy} = c_2^2 \sim 0.5$$

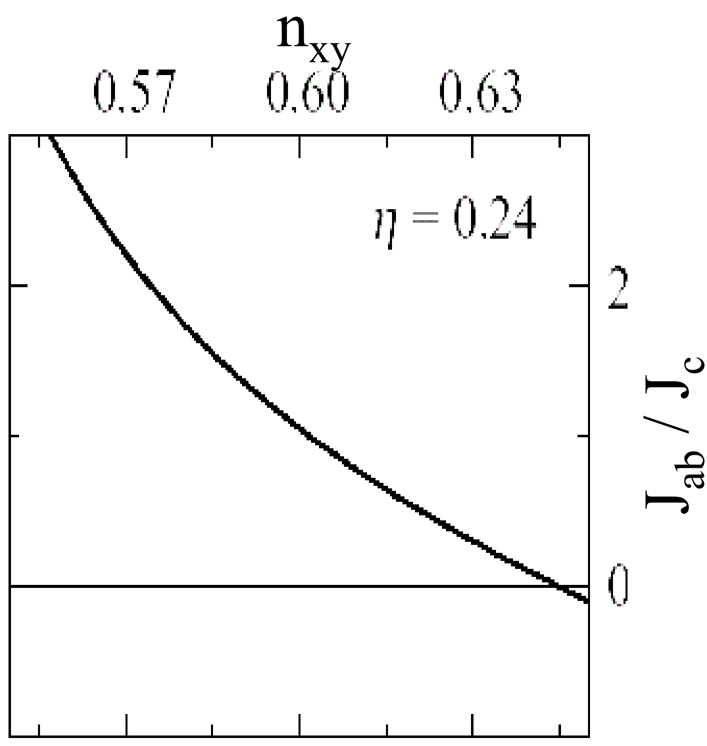
$$\eta = J_H / U \sim 0.15$$

from experiment



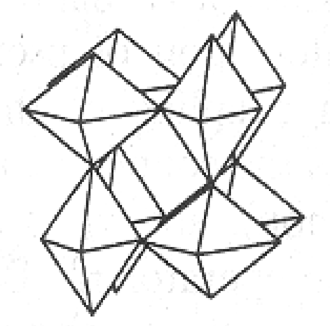
isotropic ferromagnetism only for $\eta > \eta_{exp}$

Calculation of Exchange Parameters

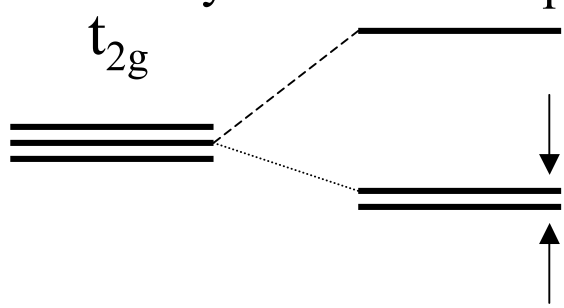


spin Hamiltonian highly sensitive to orbital state
spatially isotropic ferromagnetism requires coincidence

Estimate of magnon gap Δ



elongated octahedra



$$\Delta = J_{SE}\left(\frac{\lambda}{D}\right)^2, \quad J_{SE} \sim 0.1J$$

$$\text{canting angle } \phi = \frac{J_{SE}}{3J}\frac{\lambda}{D}$$

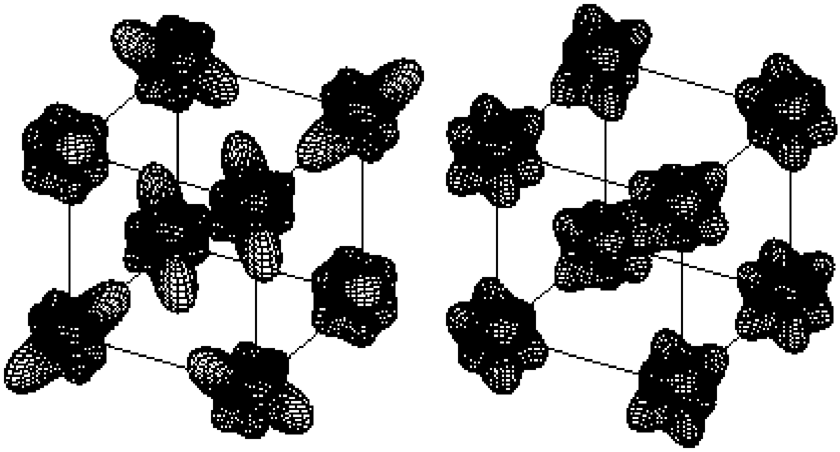
$\Big\}$ D > 200 meV

Large orbital splitting of unknown origin required to explain small magnon gap

⇒ fine tuning on several levels required to reconcile orbitally ordered state with measured magnetic dynamics

New Orbitally Ordered States

derived from superexchange model with spin ferromagnetism imposed (Okamoto & Khaliullin)



- reduced anisotropy due to strong orbital quantum fluctuations

- naturally explains spatially isotropic magnon dispersions, small magnon gap

- need to check compatibility with neutron and NMR form factors

- ordering pattern cannot explain observed lattice distortions, but magnetic dynamics not strongly altered when experimental lattice distortion is included

  ⇒ different origin of lattice distortions?
  different ordered states?

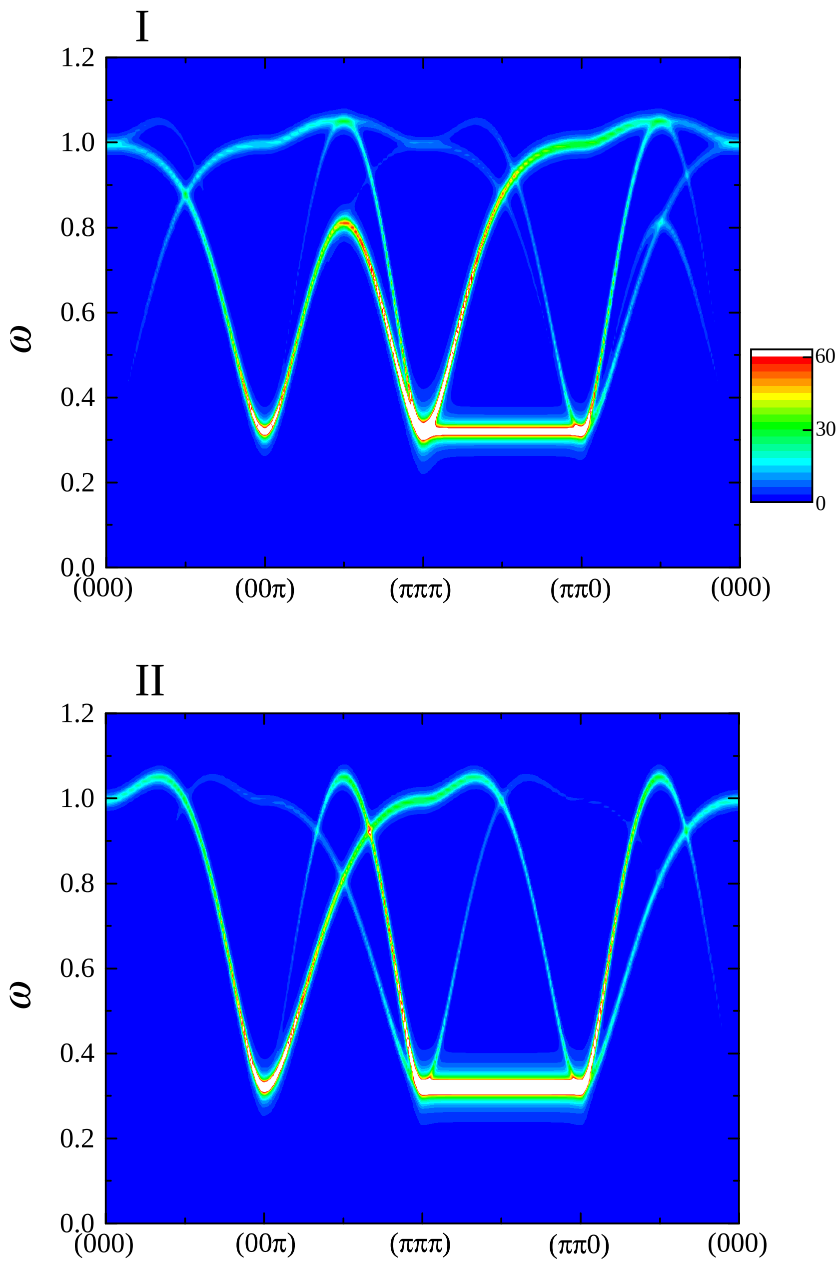
Octahedral Distortions in Non-JT Systems

	θc	θab	ΔL=l−s	ΔL/L (%)
SmAlO3*	159	161	0.003	0.16
GdAlO3	157	157	0.008	0.43
HoAlO3	153	152	0.022	1.14
YAlO3	152	152	0.026	1.36
YAlO3#			0.020	1.05
YTiO3	144	140	0.060	2.94
YFeO3$	144	145	0.033	1.64

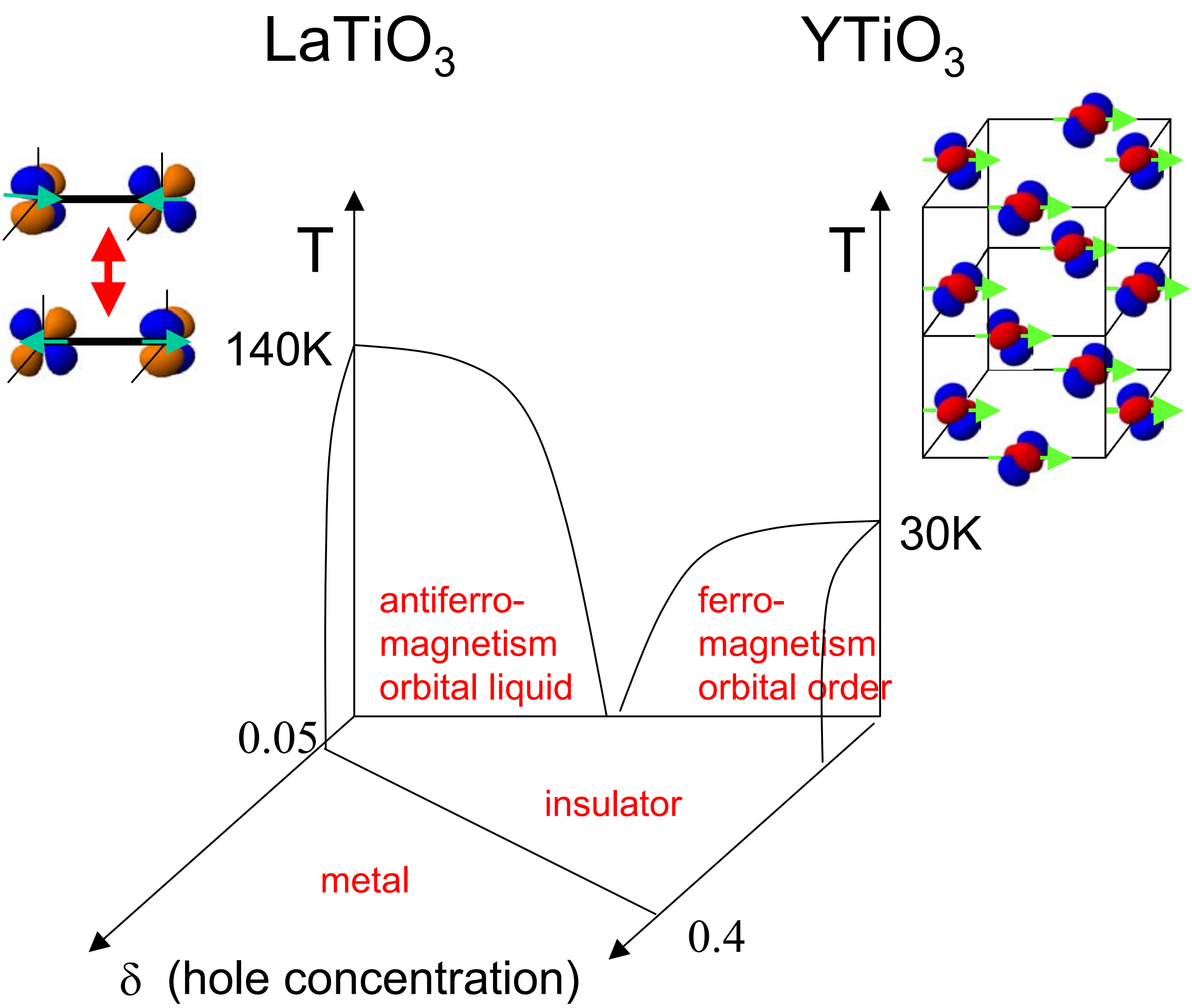
Douglas du Boulay, PhD Thesis (The Univ. of Western Australia, 1996)
*Marezio et al. J. Sol. Stat. Chem., 4, 11 (1972)
#Diehl & Brandt, Mat. Res. Bull., 10, 85 (1975)
$Marezio et al. J. Sol. Stat. Chem., 6, 23 (1971)

Explicit theory for $YTiO_3$ incorporating orbital zero-point fluctuations explains gapless, isotropic spin wave spectrum (Okamoto & Khaliullin)

Prediction: **Orbiton Dispersions**



$(La,Y)TiO_{3+\delta}$ Phase Diagram



Tokura et al.

Conclusions

Spin excitations in copper oxides

→ Novel collective spin excitations

→ Key role for mechanism of
high temperature superconductivity

Spin excitations in titanium oxides

→ Interplay with orbital dynamics
$\Rightarrow$ orbital liquid ?