Neutron Scattering

with Examples from Cuprate Superconductors
Part 2

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CES center for emergent superconductivity

Coherent magnetic scattering

Amplitude for magnetic scattering $= pS$

$$p = \left(\frac{\gamma r_0}{2}\right) g f(\mathbf{Q})$$

$S = \text{Spin}$

$$\frac{\gamma r_0}{2} = 0.2695 \times 10^{-12} \text{ cm}$$

$r_0 = e^2/m_e c^2$

$\gamma = 1.913$

$g \approx 2$

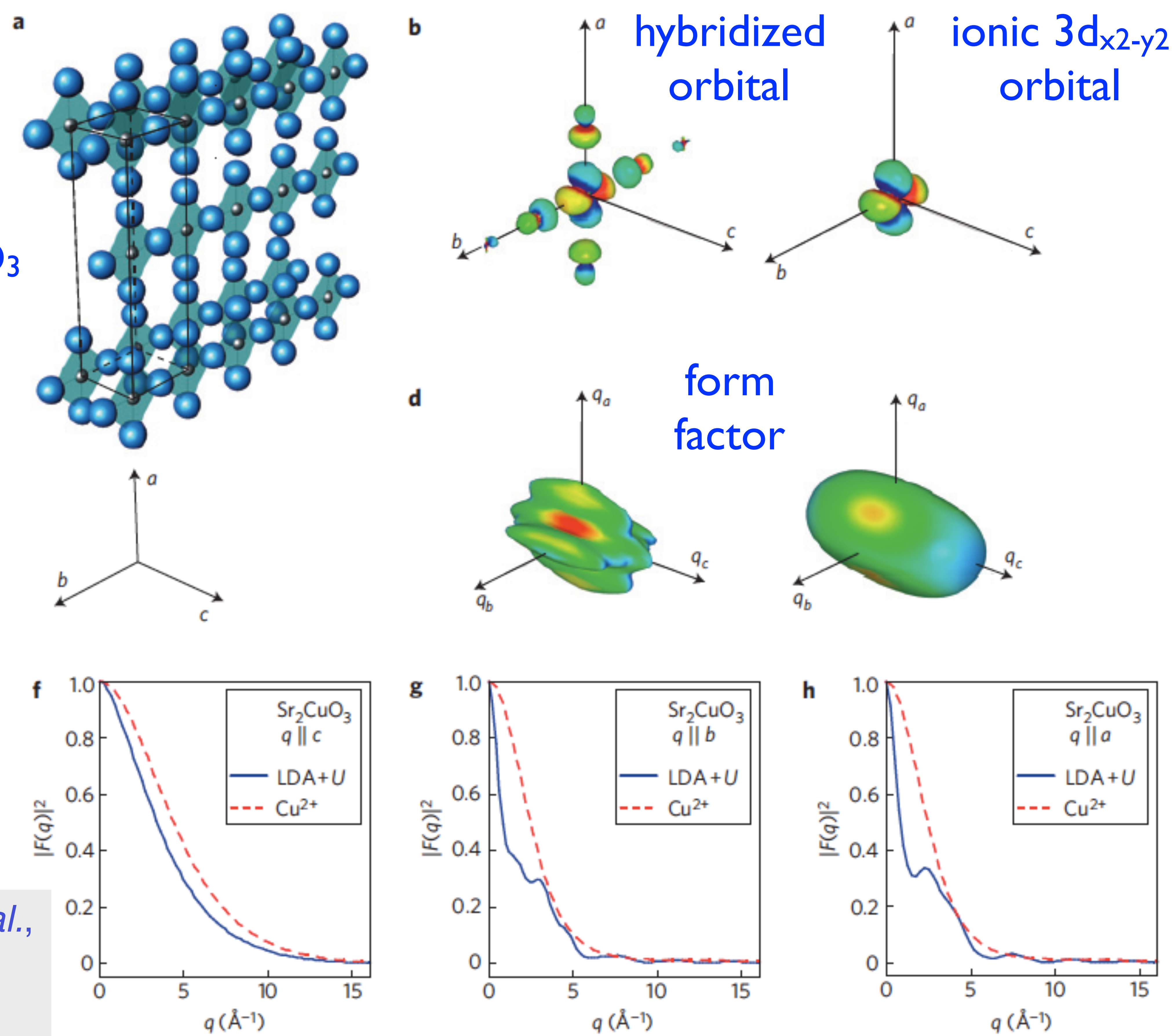
Form factor

$$f(\mathbf{Q}) = \int \rho_s(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r}$$

$$f(0) \equiv 1$$

Form factor in cuprates

Sr_2CuO_3



Walters *et al.*, Nat. Phys. (2009)

Differential cross section with magnetic terms

$$\left.\frac{d^2\sigma}{d\Omega_f dE_f}\right|_{\mathbf{s}_i\rightarrow\mathbf{s}_f} = \frac{k_f}{k_i}\sum_{i,f} P(i)\left|\langle f|\sum_l e^{i\mathbf{Q}\cdot\mathbf{r}_l} U_l^{\mathbf{s}_i\mathbf{s}_f}|i\rangle\right|^2 \delta(\hbar\omega + E_i - E_f)$$

neutron spin operator x 2

$$U_l^{\mathbf{s}_i\mathbf{s}_f} = \langle\mathbf{s}_f|b_l - p_l\mathbf{S}_{\perp l}\cdot\boldsymbol{\sigma} + B_l\mathbf{I}_l\cdot\boldsymbol{\sigma}|\mathbf{s}_i\rangle$$

magnetic interaction vector

nuclear spin operator

neutron spin state

$$\begin{aligned}\mathbf{S}_\perp &= \hat{\mathbf{Q}}\times(\mathbf{S}\times\hat{\mathbf{Q}})\\ &= \mathbf{S} - \hat{\mathbf{Q}}(\hat{\mathbf{Q}}\cdot\mathbf{S})\end{aligned}$$

$$|\mathbf{S}_\perp|^2 = \sum_{\alpha,\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha\hat{Q}_\beta)S_\alpha^* S_\beta$$

Magnetic scattering

$$\frac{d^2\sigma}{d\Omega_f dE_f} = \frac{N}{\hbar}\frac{k_f}{k_i} p^2 e^{-2W} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(\mathbf{Q},\omega)$$

Dynamical structure factor

$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, e^{-i\omega t} \sum_l e^{i\mathbf{Q}\cdot\mathbf{r}_l} \langle S_0^\alpha(0) S_l^\beta(t)\rangle$$

Instantaneous correlations

$$S^{\alpha\beta}(\mathbf{Q}, t=0) = \int_{-\infty}^{\infty} d\omega S^{\alpha\beta}(\mathbf{Q},\omega)$$

Sum rule

$$\int_{-\infty}^{\infty} d\omega \int_{\mathrm{BZ}} d\mathbf{Q}\, S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{(2\pi)^3}{3v_0} S(S+1)\delta_{\alpha\beta}$$

Polarized-beam scattering

- The component of $S_\perp$ that is perpendicular to the incident neutron polarization flips the neutron spin
- If we spin-polarize the incident neutron beam, and analyze the polarization of the scattered beam, we can separate "spin-flip" and "non-spin-flip" scattering
- Polarization analysis is expensive in terms of intensity
- I'm not going to discuss this

Magnetic diffraction

$$\left.\frac{d\sigma}{d\Omega_f}\right|_{\mathrm{coh}}^{\mathrm{el}} = N_m \frac{(2\pi)^3}{v_m} \sum_{\mathbf{G}_m} \delta(\mathbf{Q} - \mathbf{G}_m) |\mathbf{F}_M(\mathbf{G}_m)|^2$$

$$\mathbf{F}_M(\mathbf{G}_m) = \sum_j p_j \mathbf{S}_{\perp j} e^{i\mathbf{G}_m \cdot \mathbf{d}_j} e^{W_j}$$

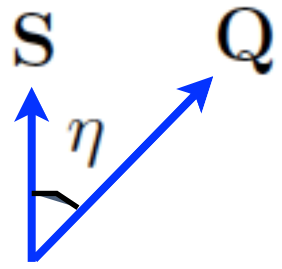
Collinear spins

$$\mathbf{F}_M = \mathbf{S}_\perp \tilde{F}_M \qquad \tilde{F}_M = \sum_j p_j e^{-W_j} e^{i\mathbf{Q}\cdot\mathbf{d}_j}$$

Average over domains

$$\langle |\mathbf{F}_M(\{hkl\})|^2 \rangle = \langle |\mathbf{S}_\perp|^2 \rangle |\tilde{F}_M(\{hkl\})|^2$$

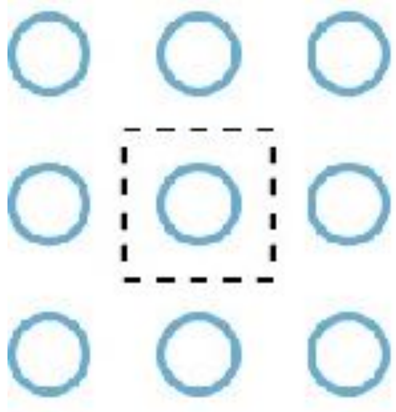
$$\langle |\mathbf{S}_\perp|^2 \rangle = S^2(1 - \langle \cos^2 \eta \rangle)$$

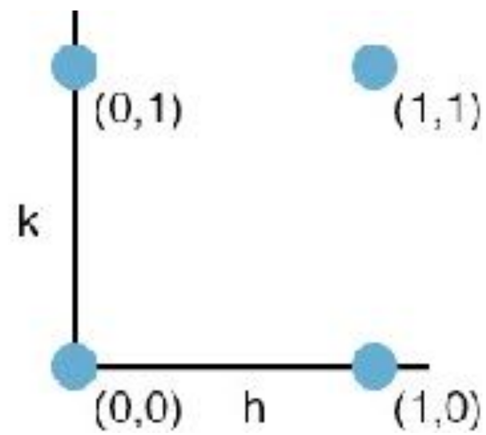


Antiferromagnetic order doubles unit cell

Real space | Reciprocal space

Crystal structure

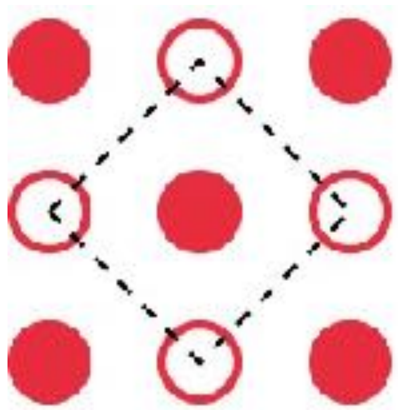


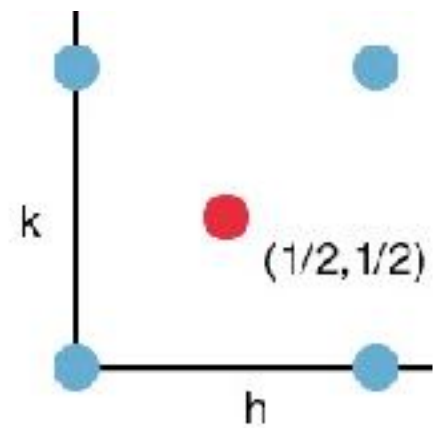


Q = (h, k, l)

in units of ($2\pi/a, 2\pi/b, 2\pi/c$)

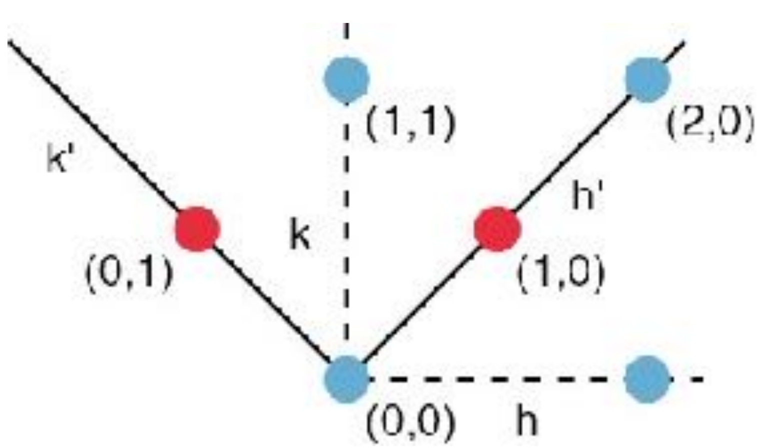
Magnetic structure





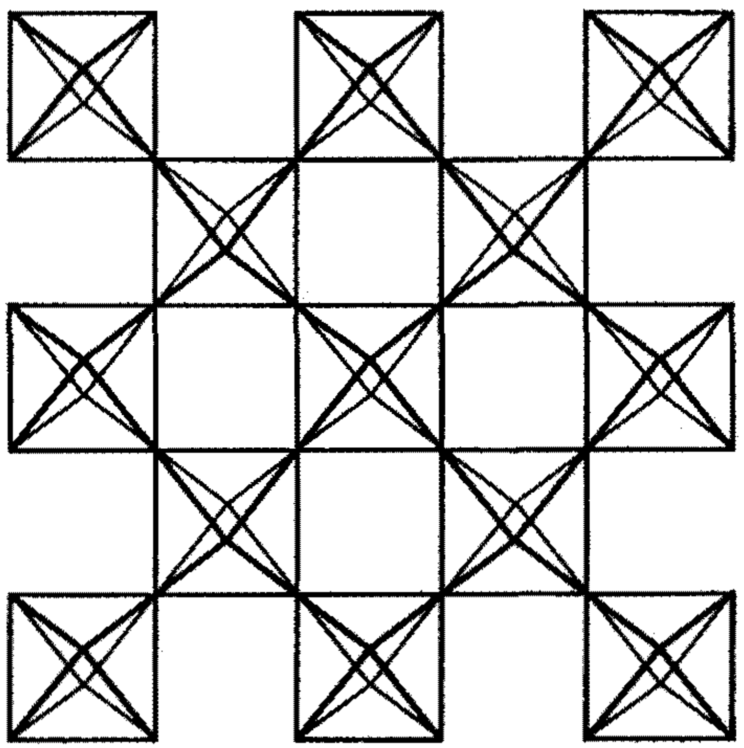
Octahedral tilt pattern in LTO is same as AF order; however, structure factor for tilted octahedra at Q = (1/2,1/2,L) is zero if L = 0.

Coordinates must rotate by 45° to describe orthorhombic cell

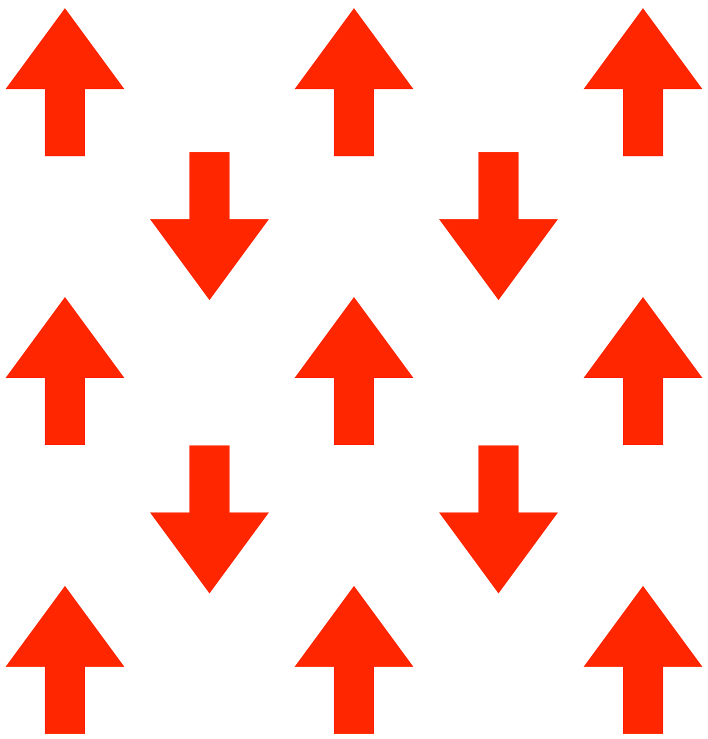


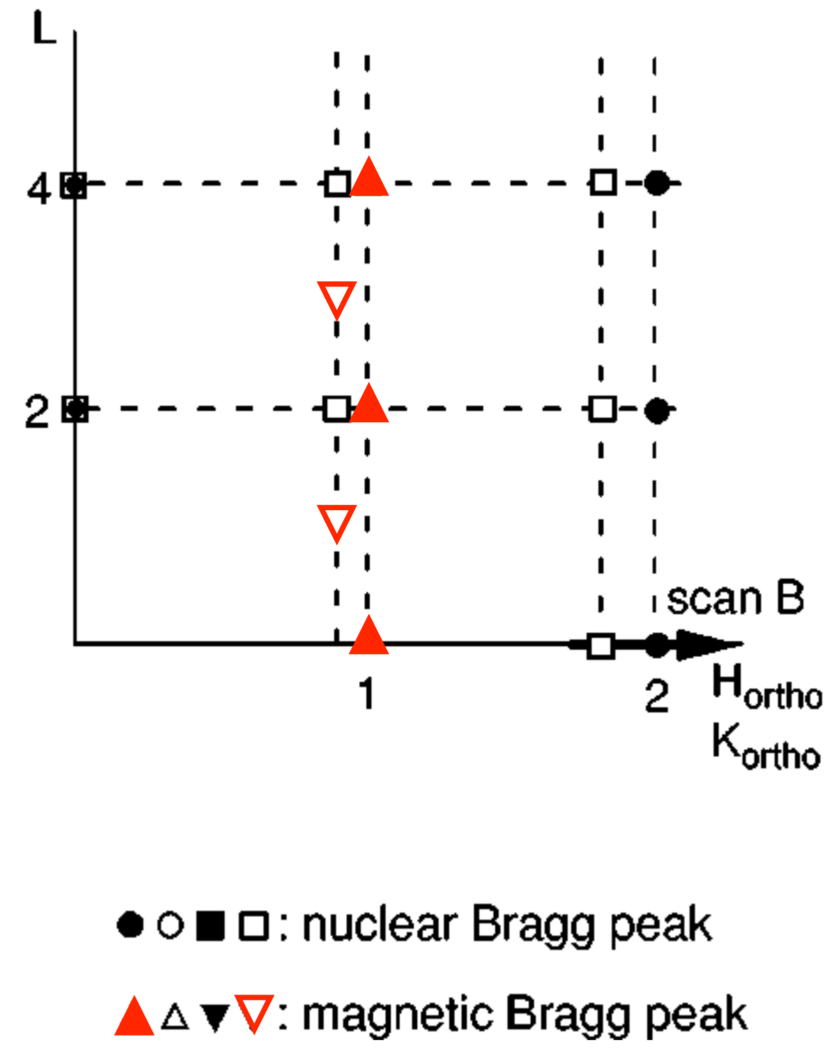
Tilt and spin orders

Octahedral tilts



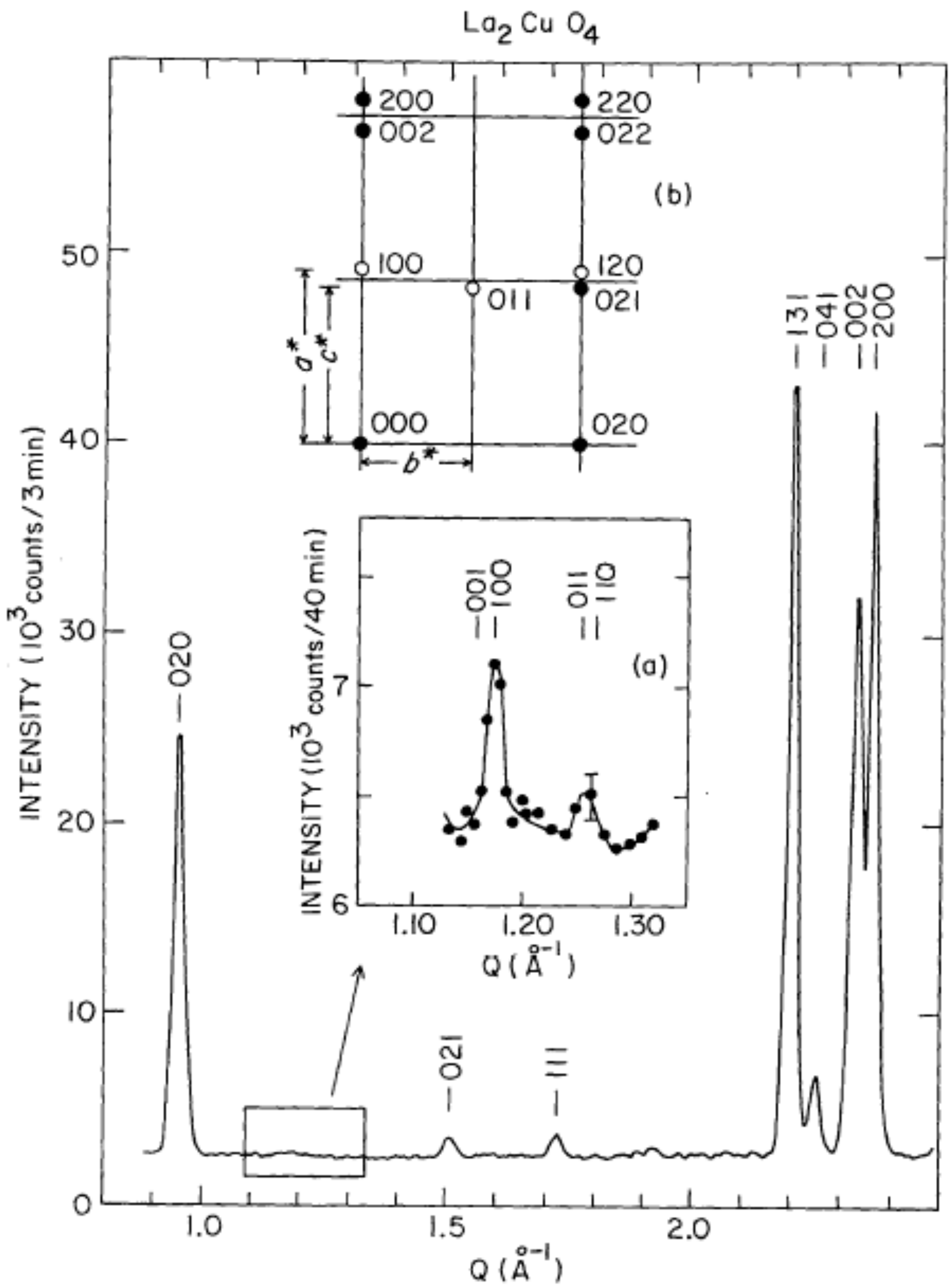
Magnetic moments





Same unit cell within one plane for both, but correlation with second plane in unit cell is different.

First experiments: neutron powder diffraction



La_2CuO_4

Main panel: E_i = 14.7 meV

Insert (a): E_i = 5.1 meV, T = 150 K

Indexing based on *Cmca* space group

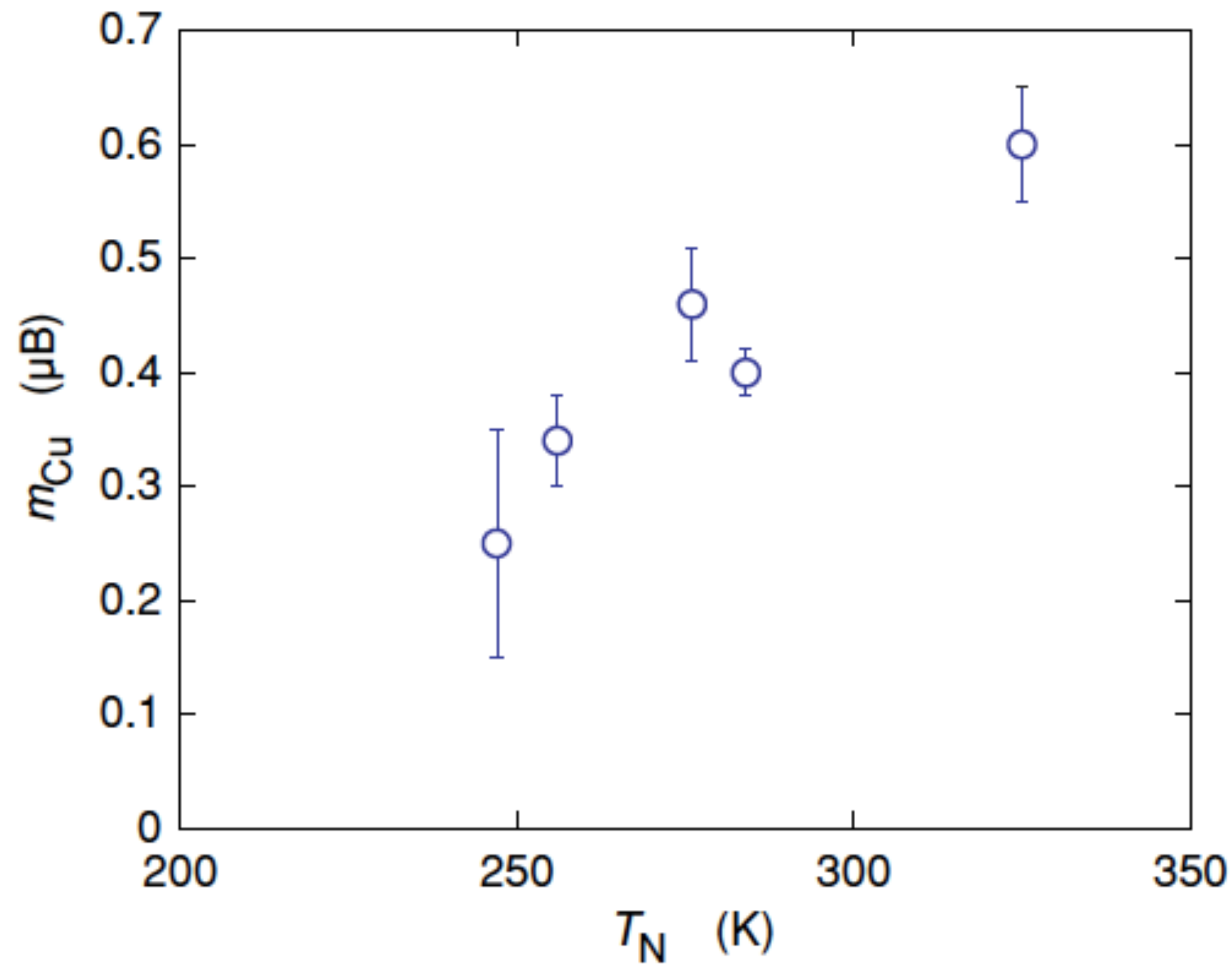
High resolution required to distinguish possible orthorhombic reflections.

First report mistakenly interpreted LTO (021) superlattice peak as due to AF order.

Yang *et al.*, JPSJ **56**, 2283 (1987)

Ordered moment

Compound	T_N (K)	m_{Cu} (μ_B)
La_2CuO_4	325(2)	0.60(5)
$Sr_2CuO_2Cl_2$	256(2)	0.34(4)
$Ca_2CuO_2Cl_2$	247(5)	0.25(10)
Nd_2CuO_4	276(1)	0.46(5)
Pr_2CuO_4	284(1)	0.40(2)
$YBa_2Cu_3O_{6.1}$	410(1)	0.55(3)
$TlBa_2YCu_2O_7$	>350	0.52(8)
$Ca_{0.85}Sr_{0.15}CuO_2$	537(5)	0.51(5)



arXiv:cond-mat/0512115

spin wave theory: $\langle S \rangle = 0.303$

$g \approx 2.2$

$m \approx 0.67\ \mu_B$

Elastic vs. inelastic weight

2D antiferromagnet with S=1/2

Total scattering: $\langle S^2 \rangle = S(S+1) = 3/4$

Elastic weight: $\langle S \rangle^2 = (0.303)^2 = 0.09$

Inelastic weight: $\langle S^2 \rangle - \langle S \rangle^2 = 0.66$

$= 0.88 \langle S^2 \rangle$

Most of the spin scattering is inelastic!
This violates the premise of perturbative spin-wave theory.

Antiferromagnetic spin waves

Assume ordered spins along z

$$\sum_{\alpha,\beta}(\delta_{\alpha,\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(\mathbf{Q},\omega) = \tfrac{1}{2}(1 + \hat{Q}_z^2) S_{\rm sw}(\mathbf{Q},\omega)$$

Linear dispersion

$$\hbar\omega_{\mathbf{q}} = \hbar c q$$

spin-wave velocity

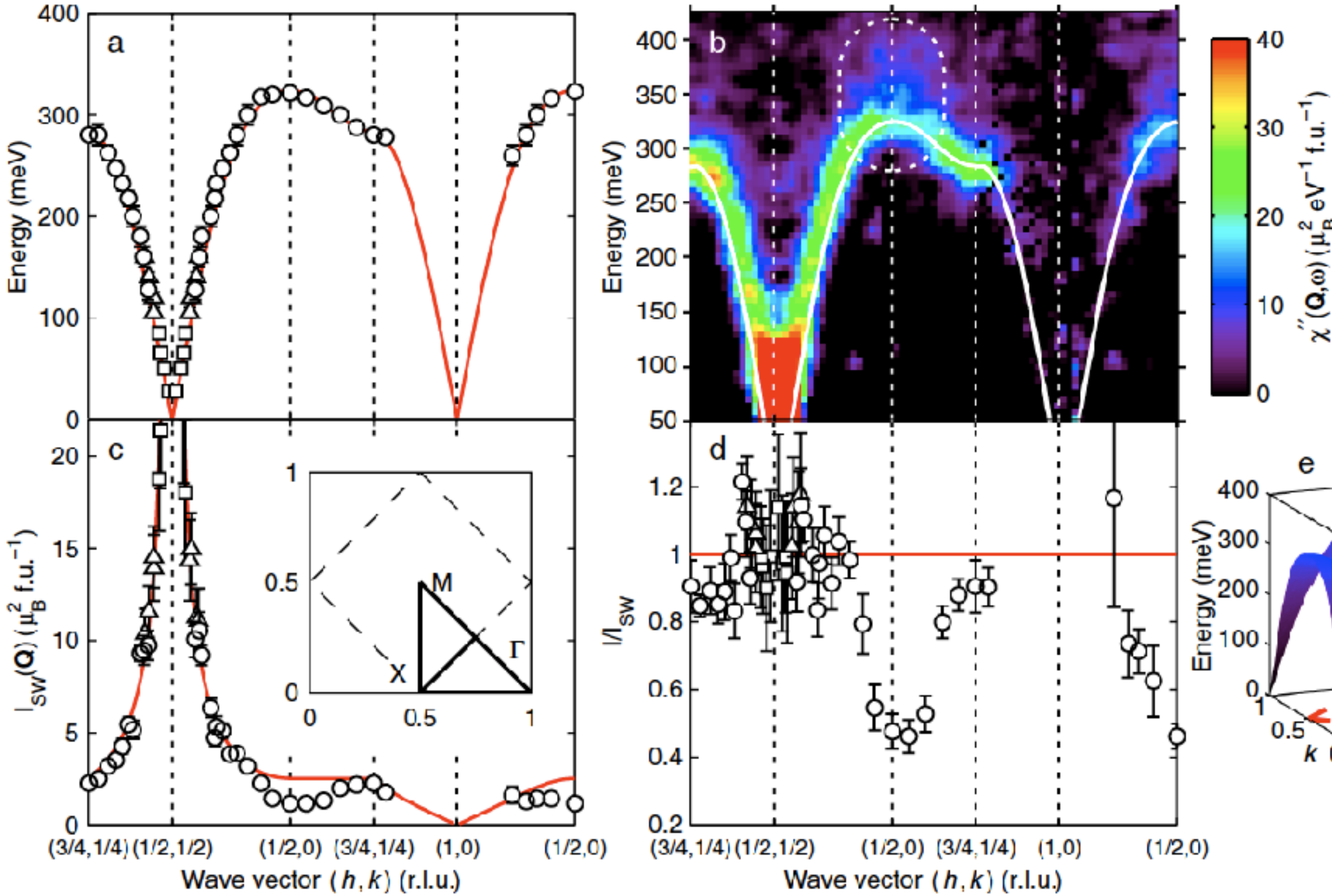
$$c = zJSa/\hbar$$

coordination number (4 for square lattice)

$$S_{\rm sw}(\mathbf{Q},\omega) \; = \; S \sum_{\mathbf{G}_m,\mathbf{q}} \frac{\hbar\omega_0}{\hbar\omega_{\mathbf{q}}} \left[(n_{\mathbf{q}}+1)\delta(\mathbf{Q}-\mathbf{q}-\mathbf{G}_m)\delta(\omega-\omega_{\mathbf{q}}) + n_{\mathbf{q}}\,\delta(\mathbf{Q}+\mathbf{q}-\mathbf{G}_m)\delta(\omega+\omega_{\mathbf{q}}) \right],$$

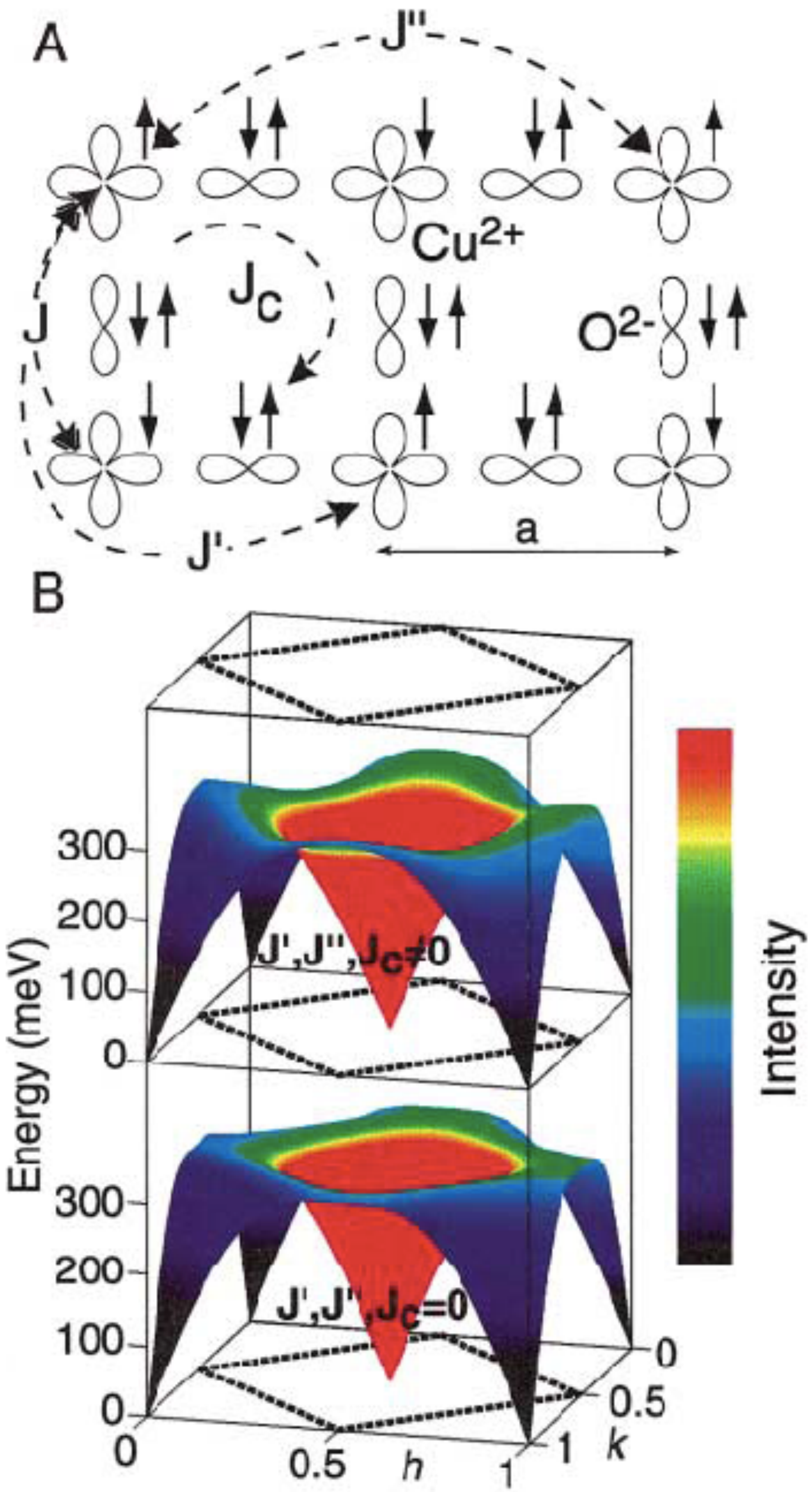
$$\hbar\omega_0 = 2zJS$$

Spin waves in La_2CuO_4



Headings *et al.*, PRL (2010)

Exchange parameters



$$J = 143 \pm 2 \text{ meV}$$

$$J' = J'' = 2.9 \pm 0.2 \text{ meV}$$

$$J_c = 58 \pm 4 \text{ meV}$$

Headings *et al.*, PRL (2010)

Coldea *et al.*, PRL (2001)

Large J requires intermediate coupling

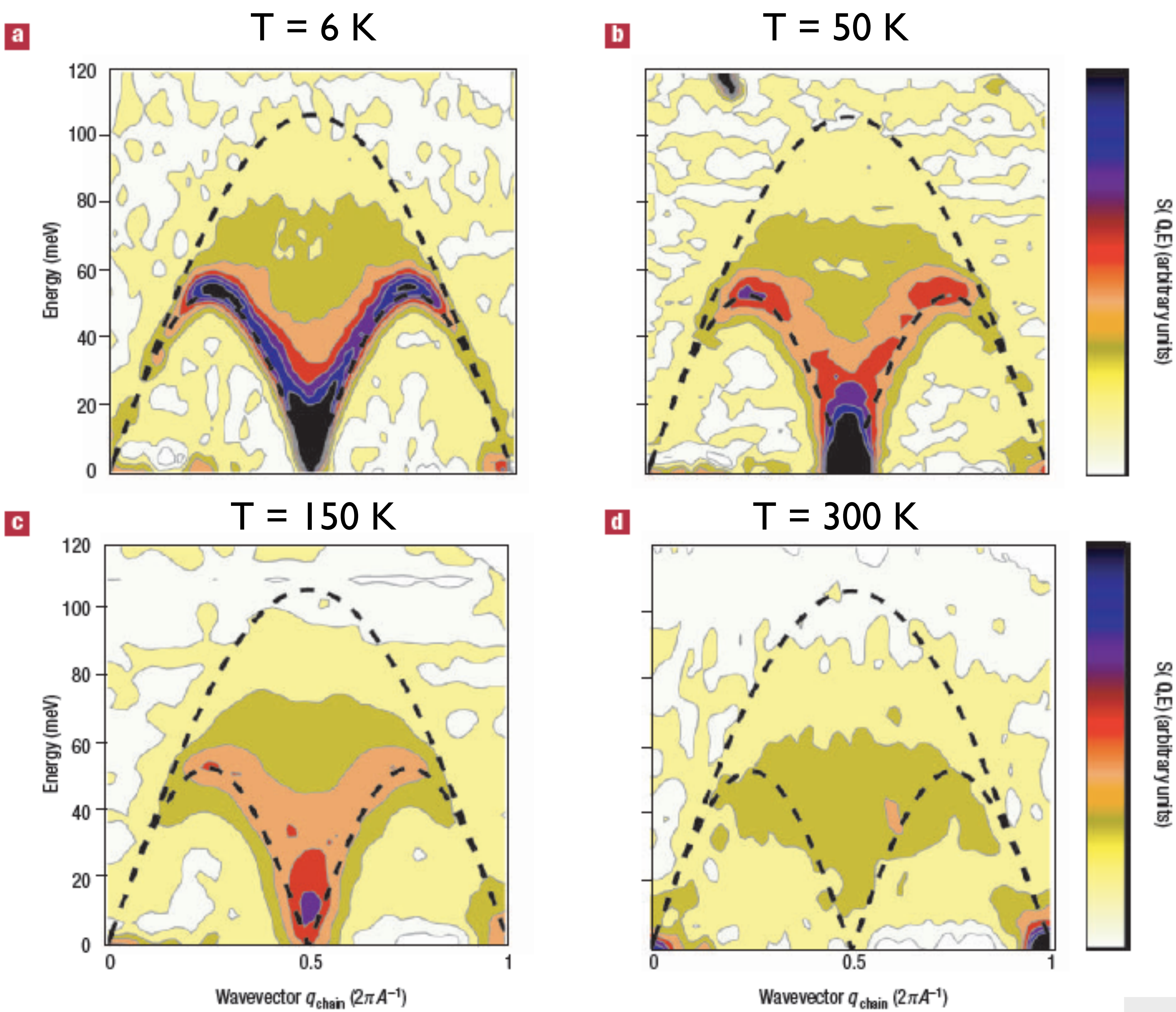
$J = 4t^2 / U$

Suppose $U = 8t$

then $J = t/2$

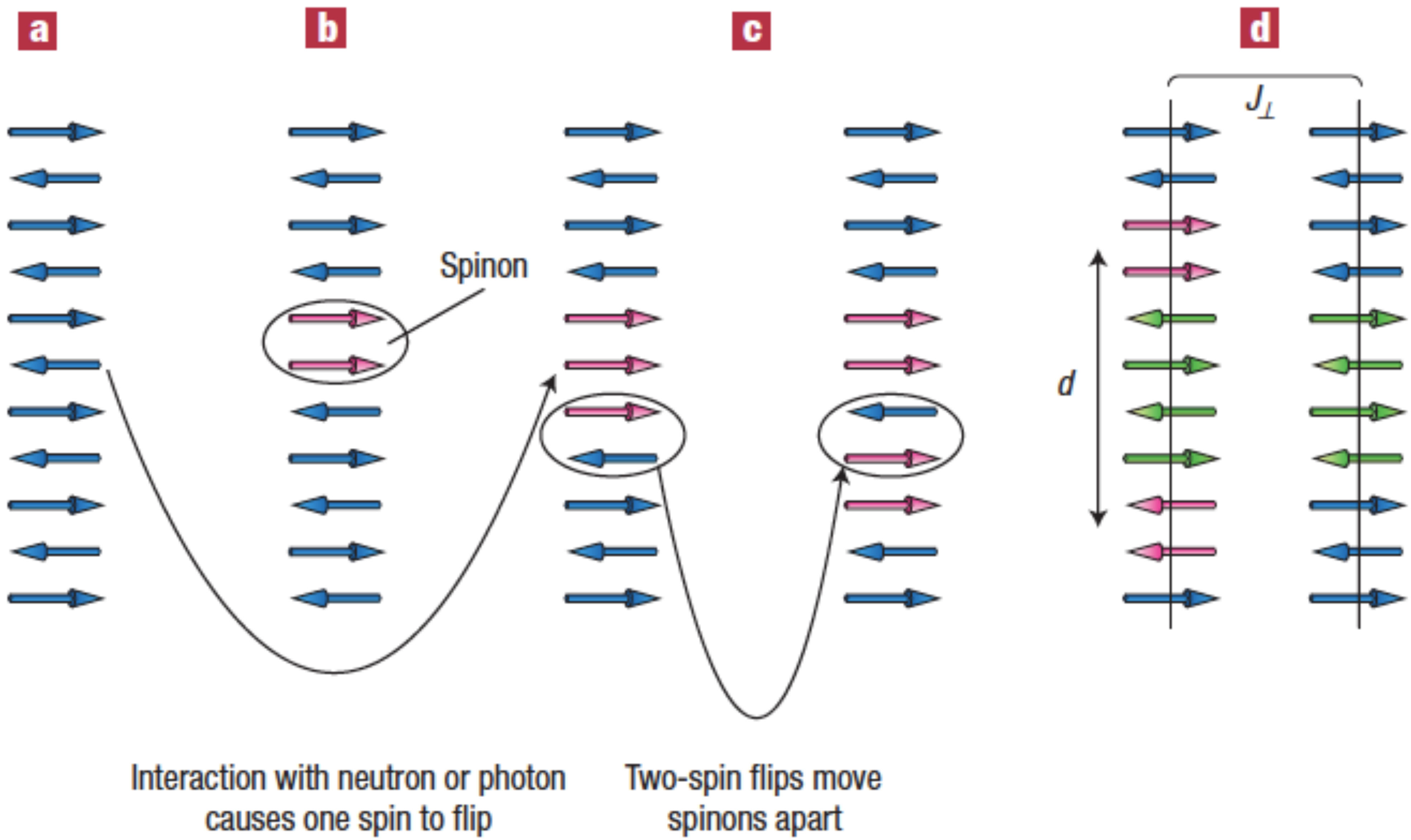
$J = 143$ meV gives $t = 0.3$ eV

$KCuF_3$: S = 1/2 spin chain system



Lake *et al*., Nat. Mat. (2005)

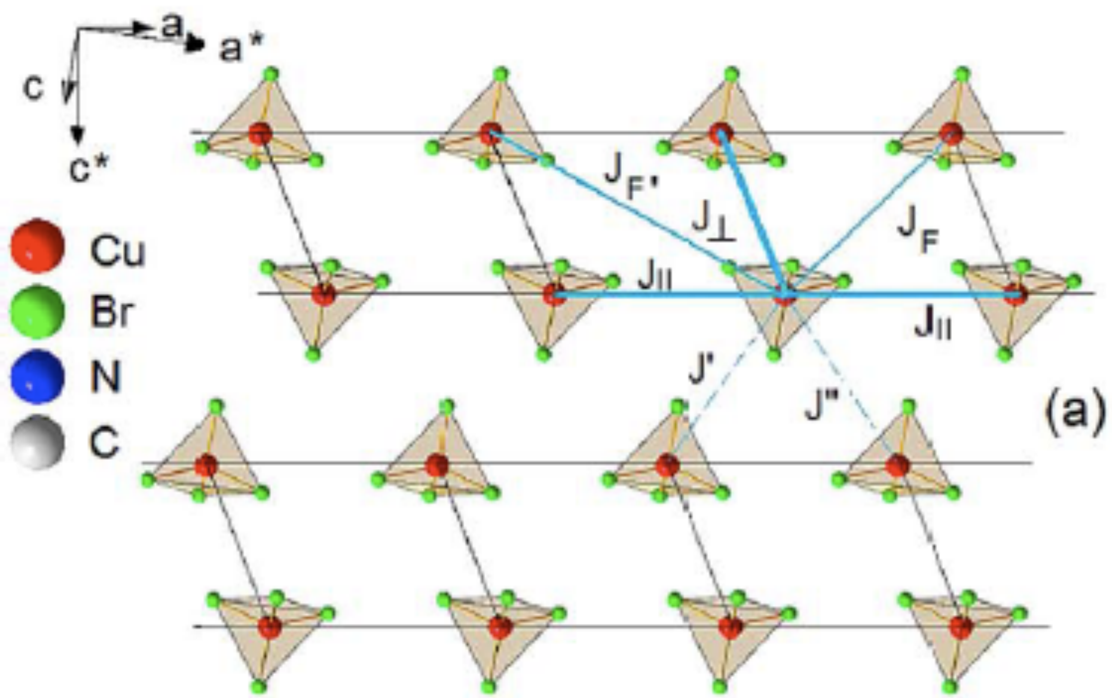
Spinons and the 2-spinon spectrum



Zaliznyak, Nat. Mat. (2005)

S = 1/2, two-leg spin ladder

$(C_5D_{12}N)_2CuBr_4$

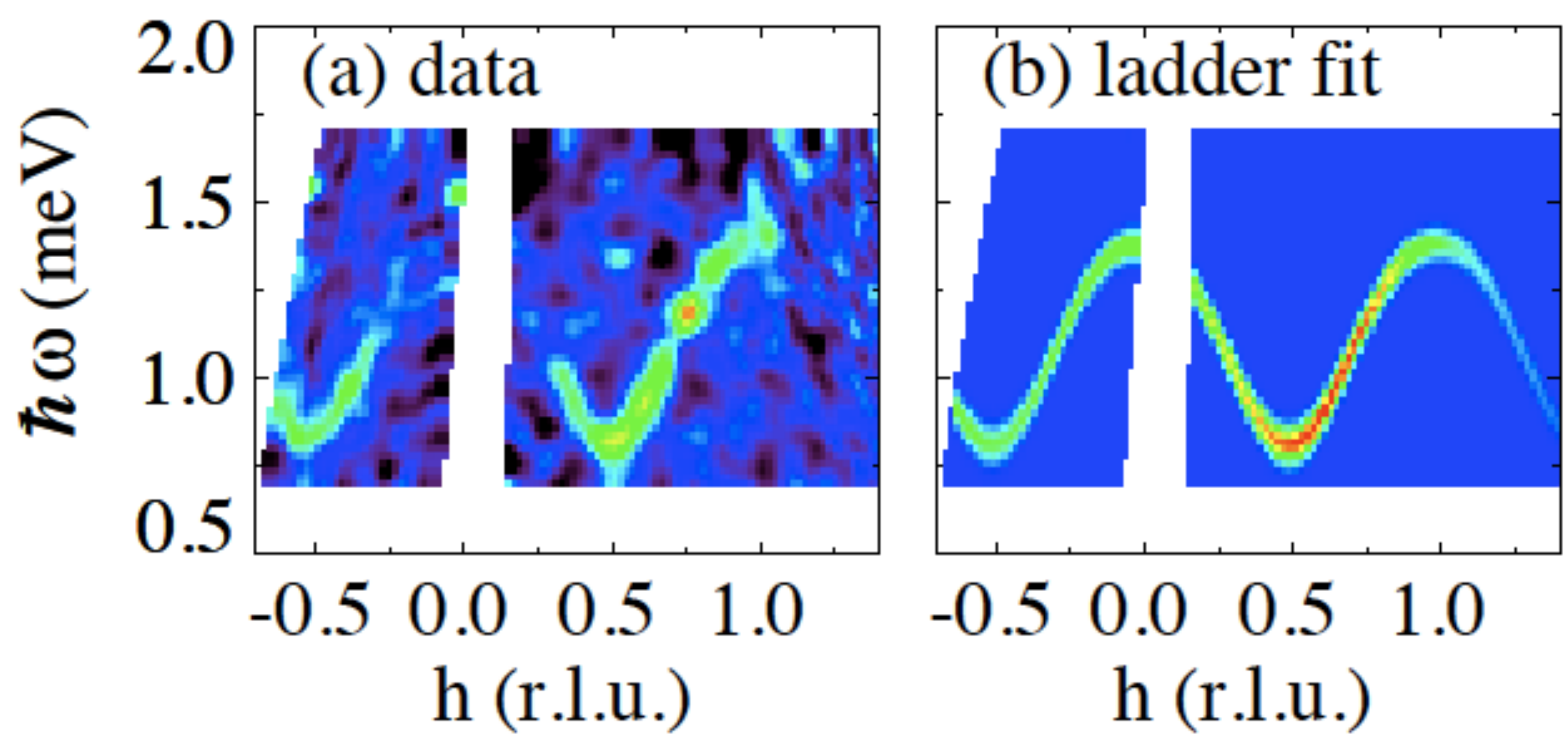


ground state: singlets on rungs

excited state: triplet can disperse along ladder

J_{rung} = 1.09 meV

J_{leg} / J_{rung} = 0.29

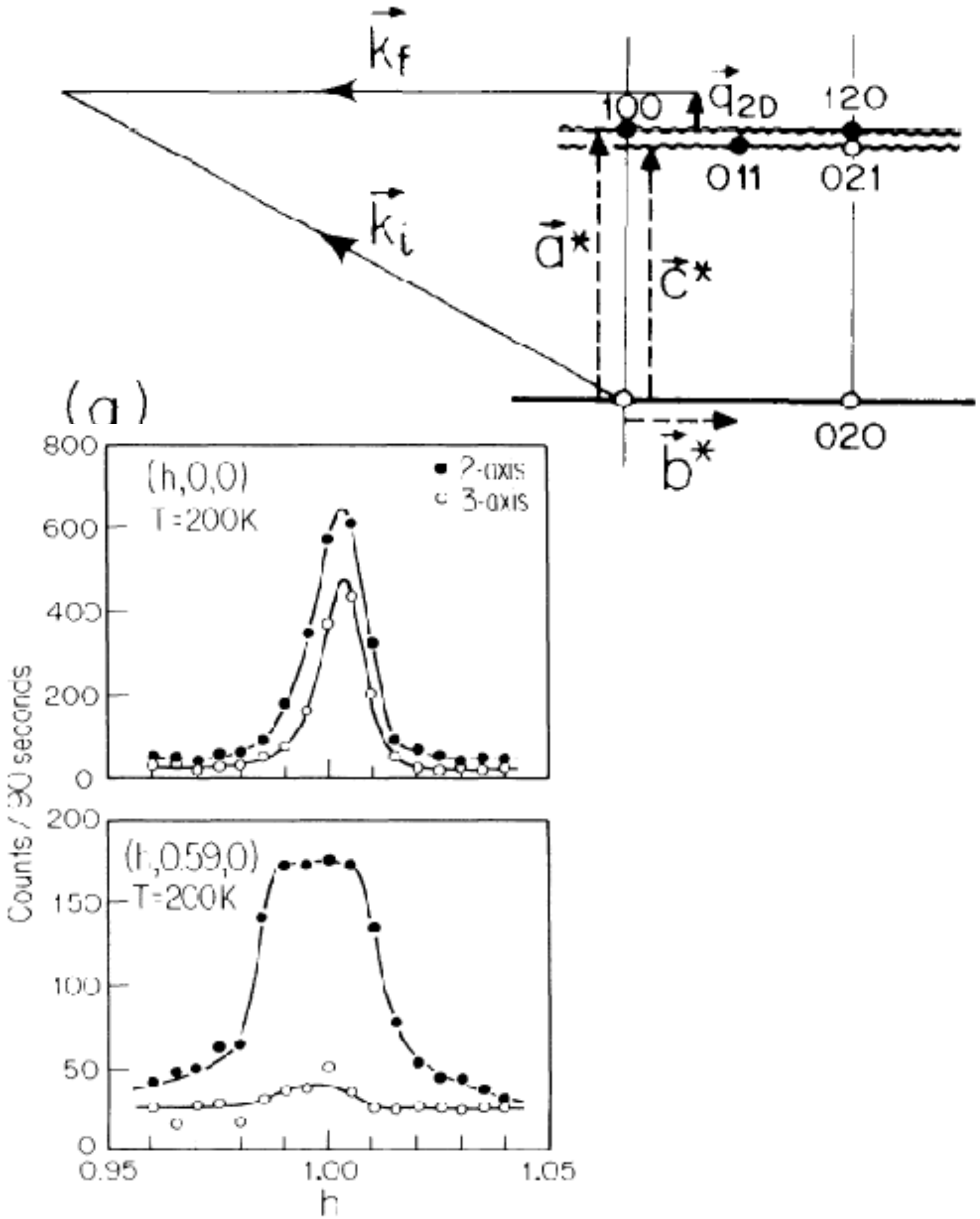


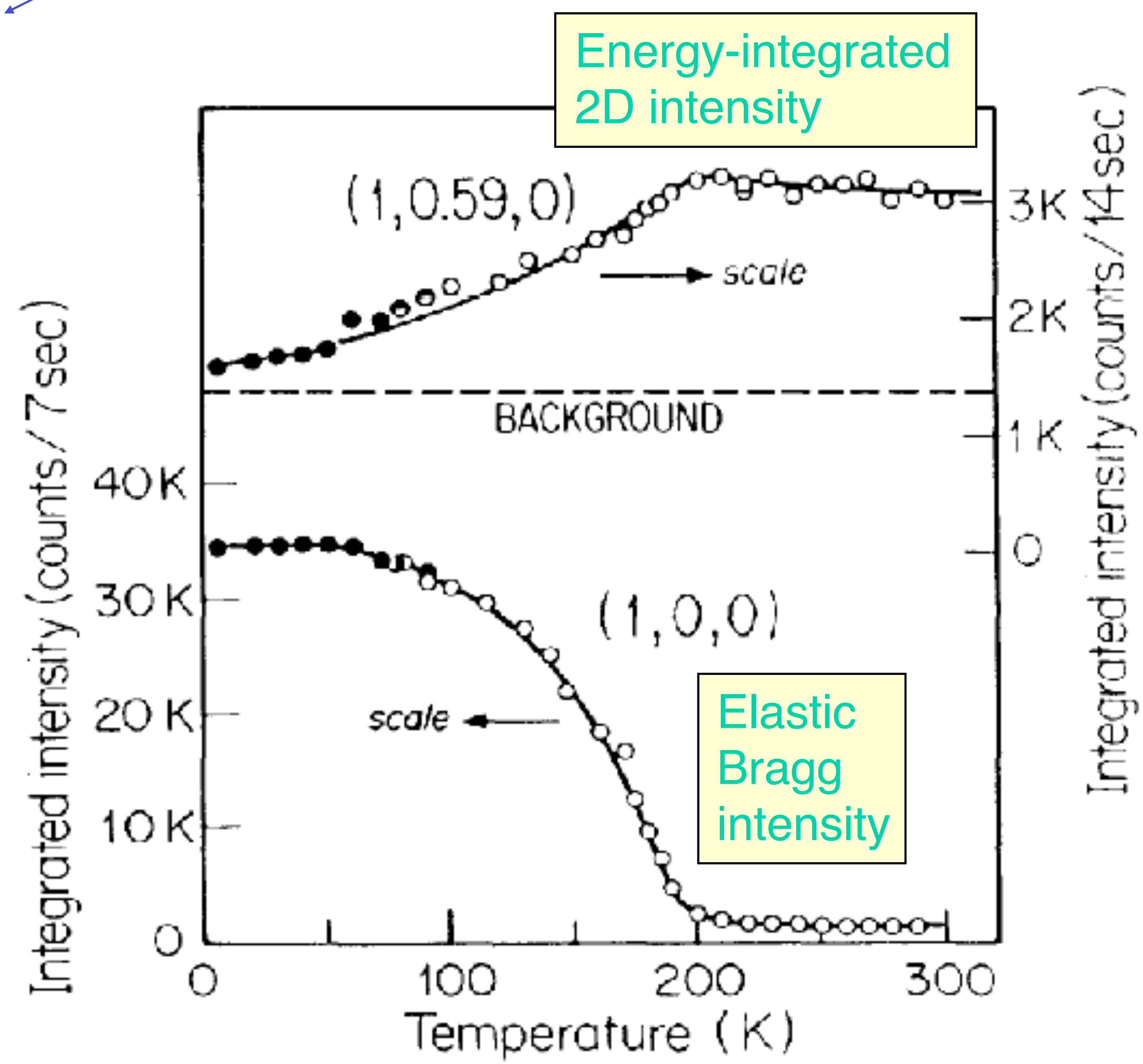
Savici *et al.*, PRB (2009)

Magnetic critical scattering

Neutron scattering on single crystal La_2CuO_4 Shirane *et al.*, PRL **59**, 1613 (1987)

Rods of scattering from 2D spin correlations





Cu spins maintain 2D correlations to high T

$S(\mathbf{q}_{2D}) \sim 1 / [(\mathbf{q}_{2D})^2 + \xi^{-2}]$

ξ = spin-spin correlation length

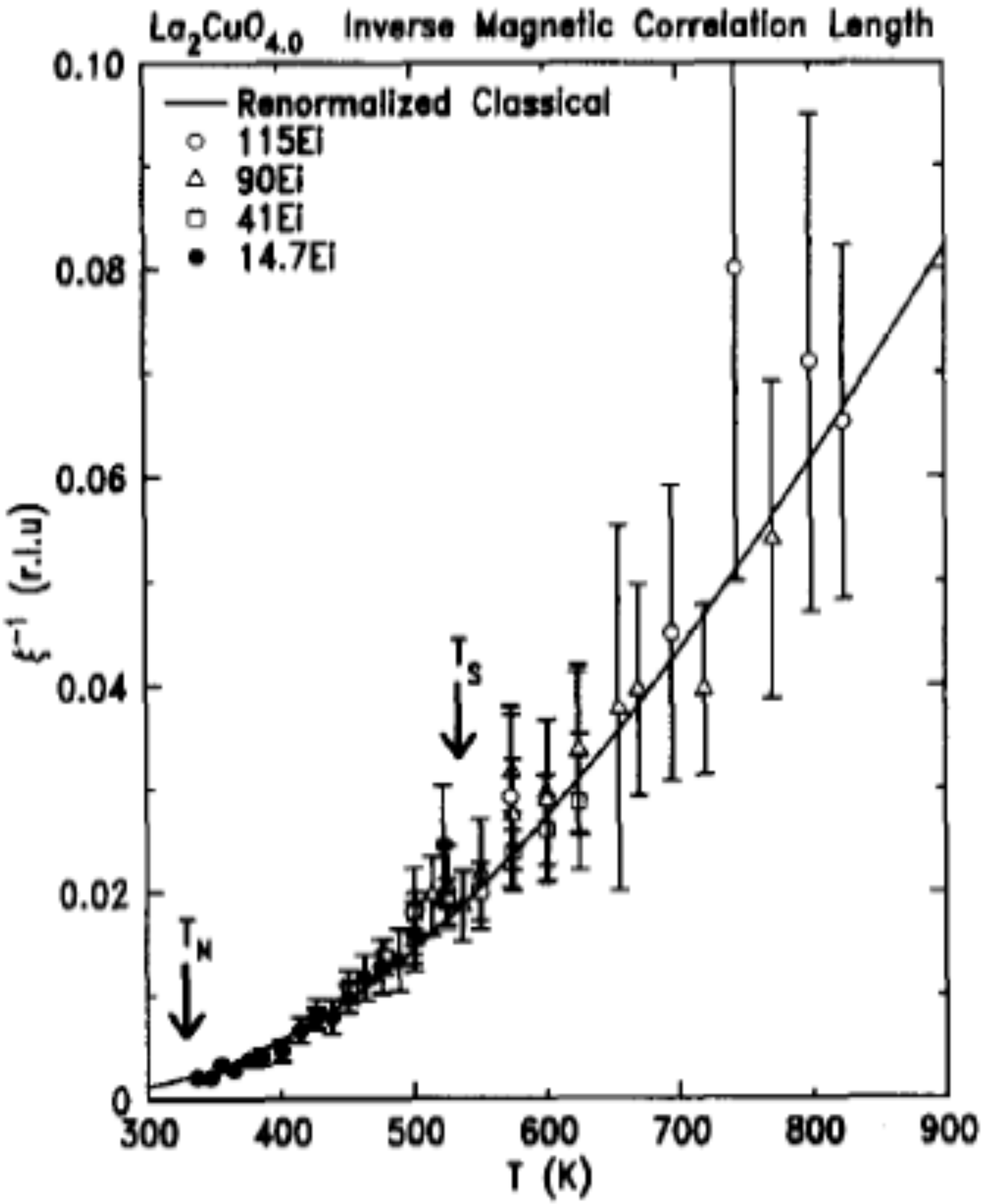
$\xi^{-1} \sim \exp(-\alpha J/T)$

J = 135 meV ~ 1500 K

Theory:

Chakravarty, Halperin,+Nelson, PRB **39**, 2344 (1989)

Hasenfratz+Niedermayer, Phys. Lett. B **268**, 231 (1991)



Expt: Birgeneau *et al.*, JPCS **56**, 1913 (1995)

Single CuO_2 layer should order AF at T=0

ξ(T) is consistent with Renormalized Classical behavior and not Quantum Disordered

Chakravarty *et al.*, PRL (1988)

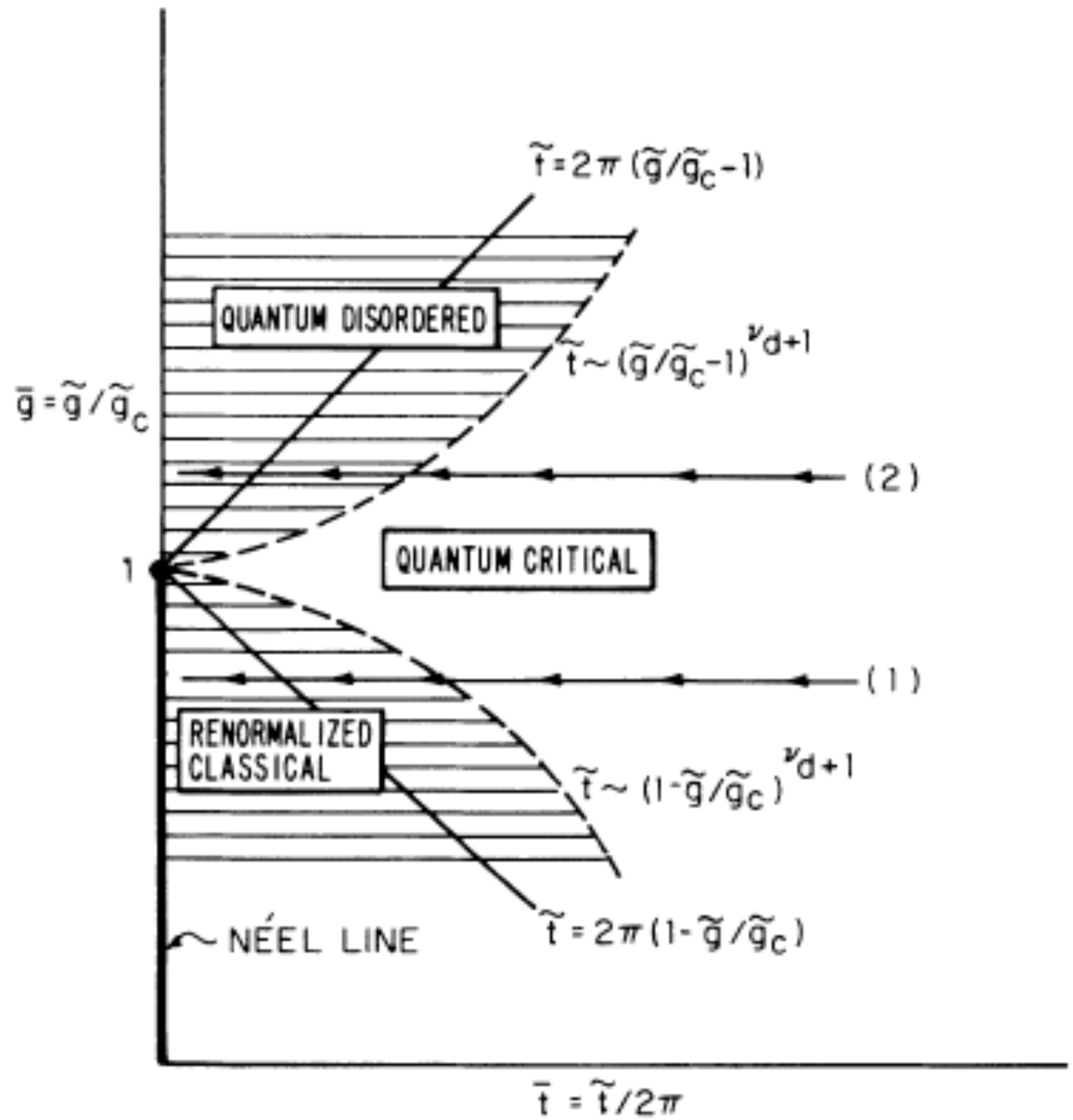
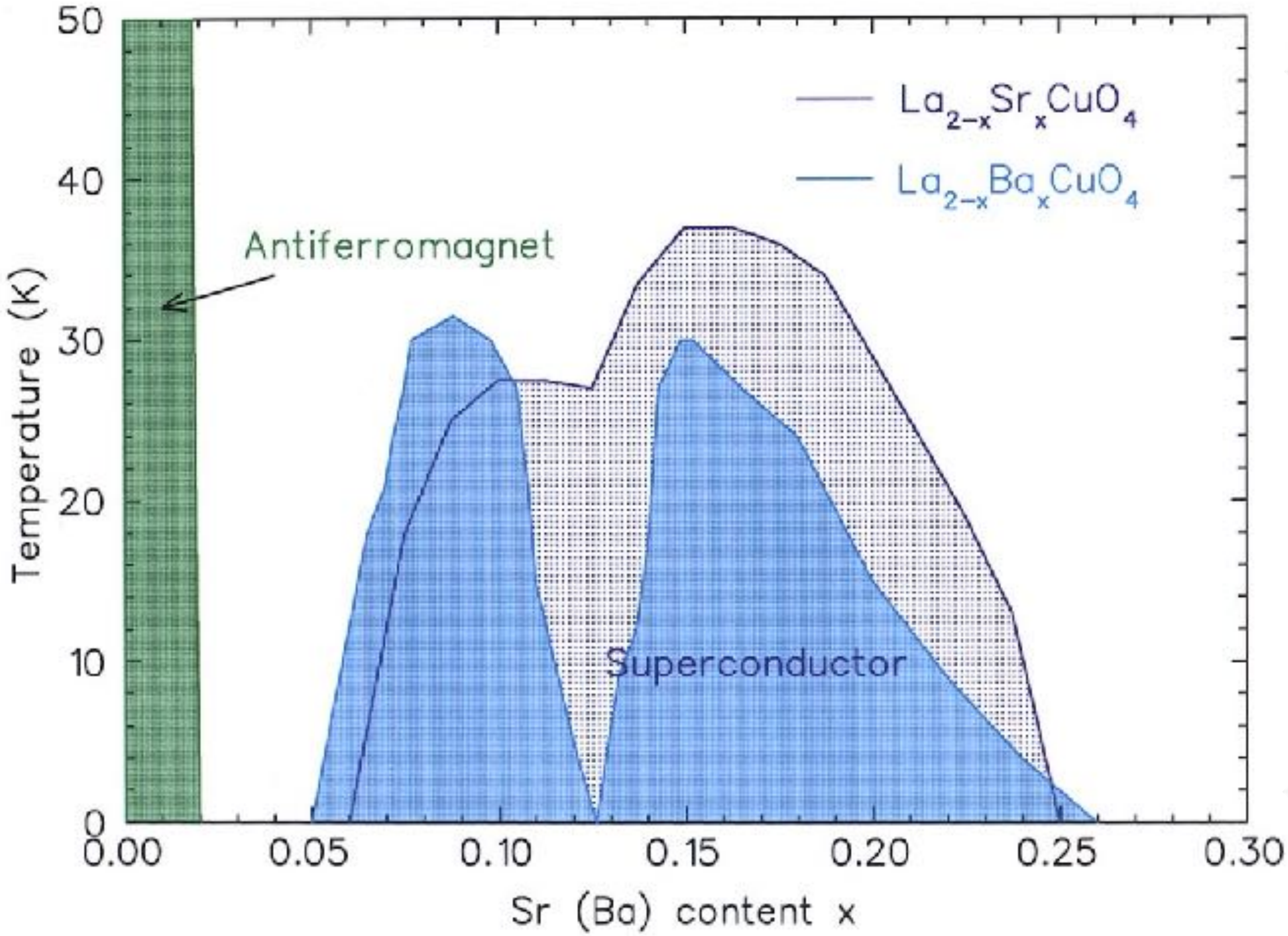
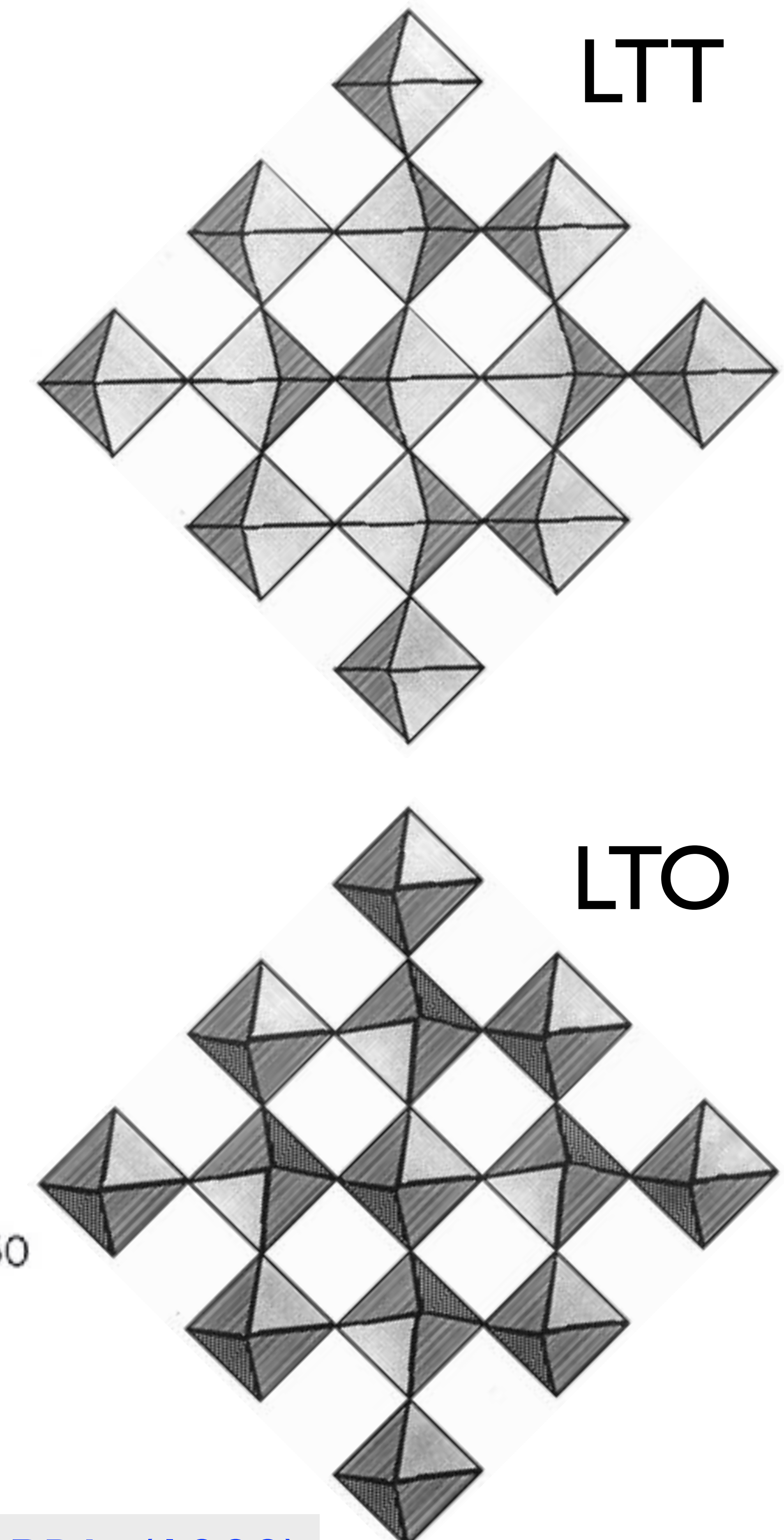


FIG. 1. Crossover phase diagram for $d=2$. $\nu_{d+1}=0.7$ for $d=2$. $\tilde{g}_c$ is the critical point of the $(d+1)$-dimensional non-linear σ model.

1/8 problem

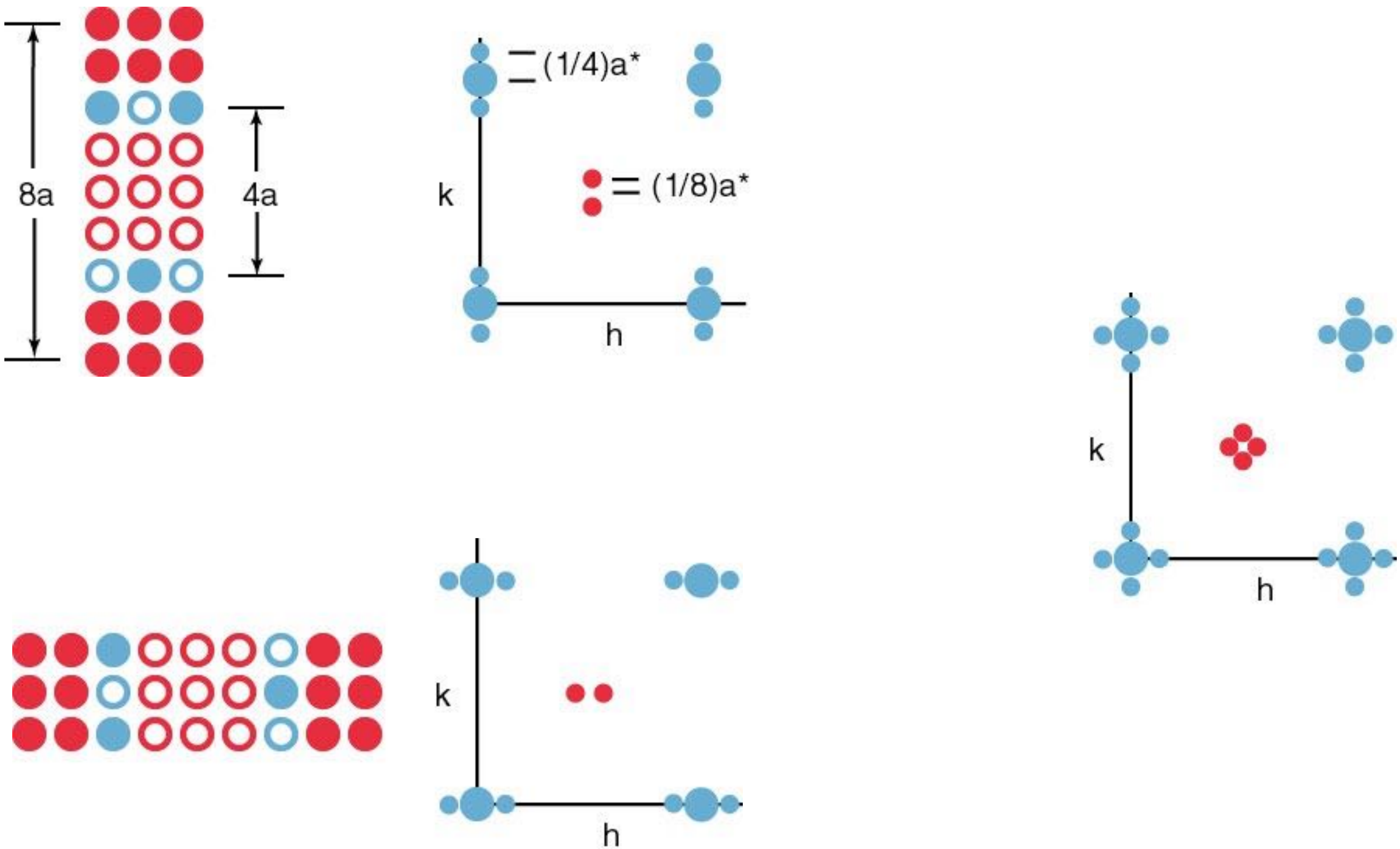


Moodenbaugh *et al.*, PRB (1988)

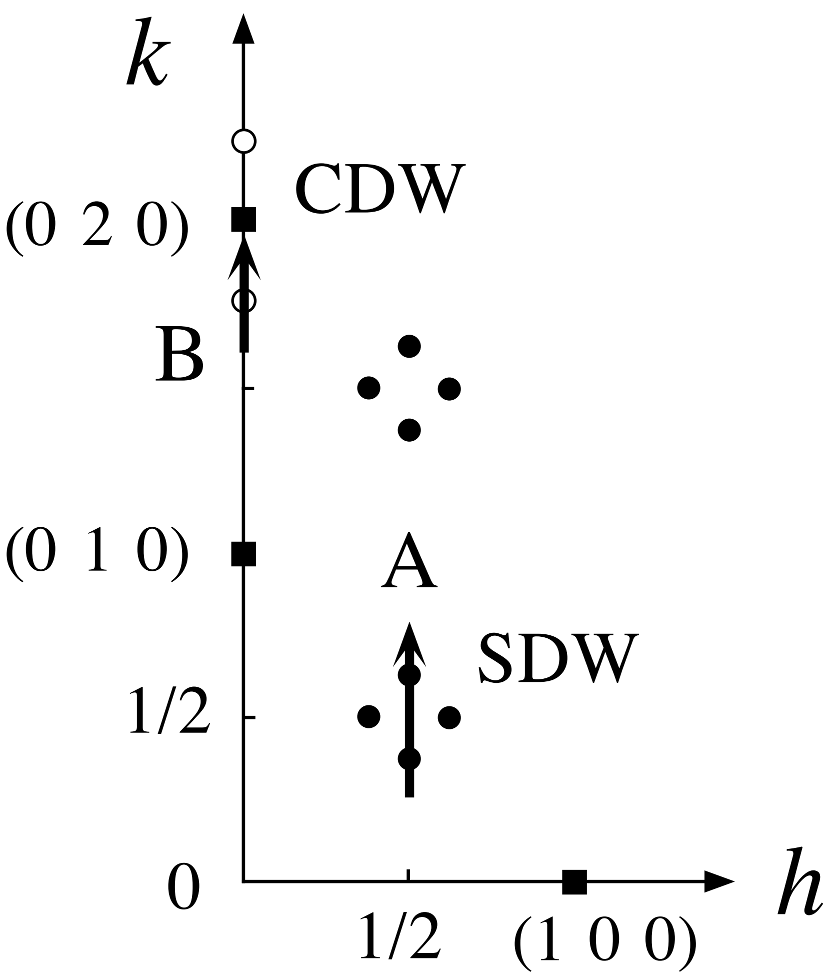


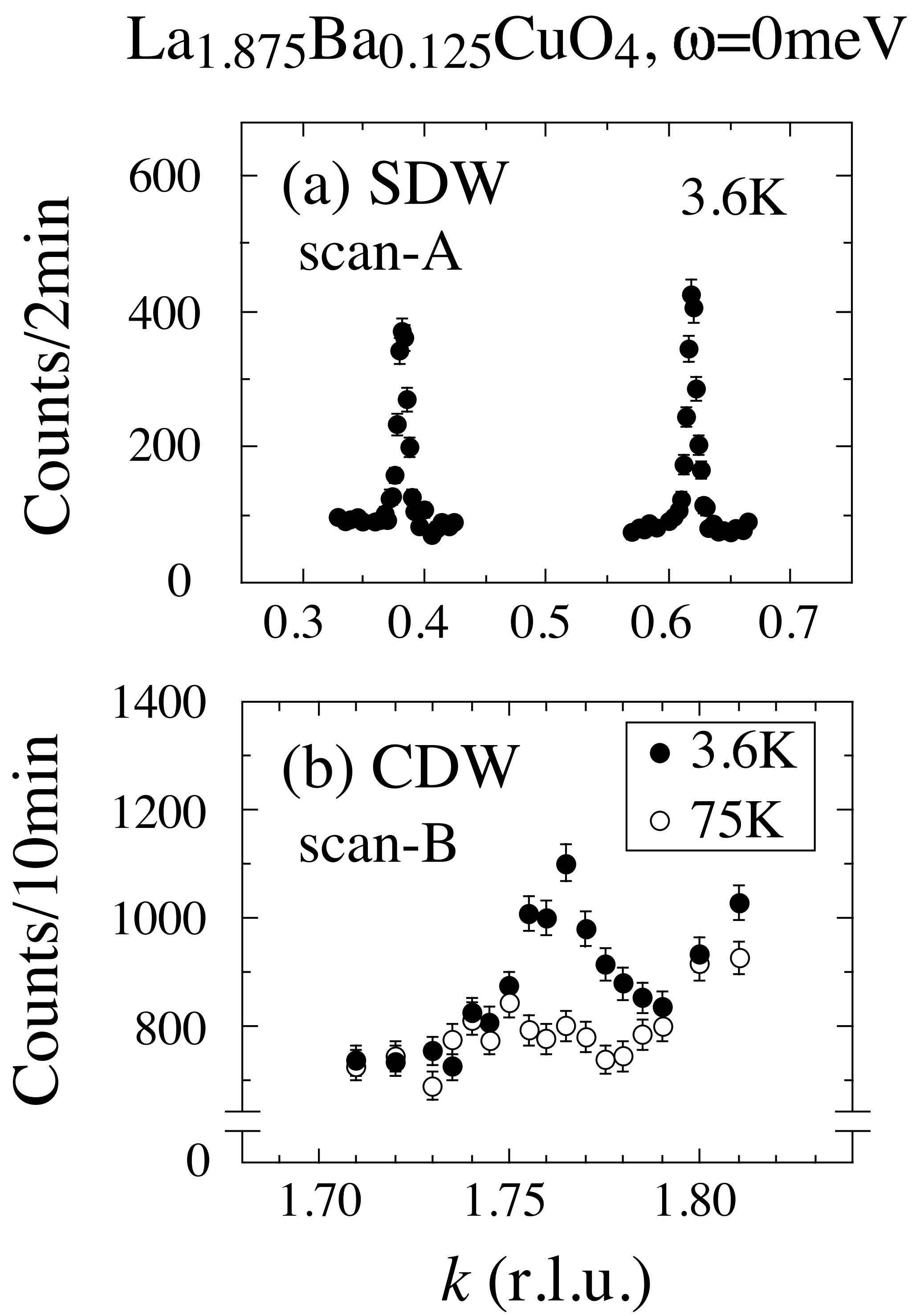
Axe *et al.*, PRL (1989)

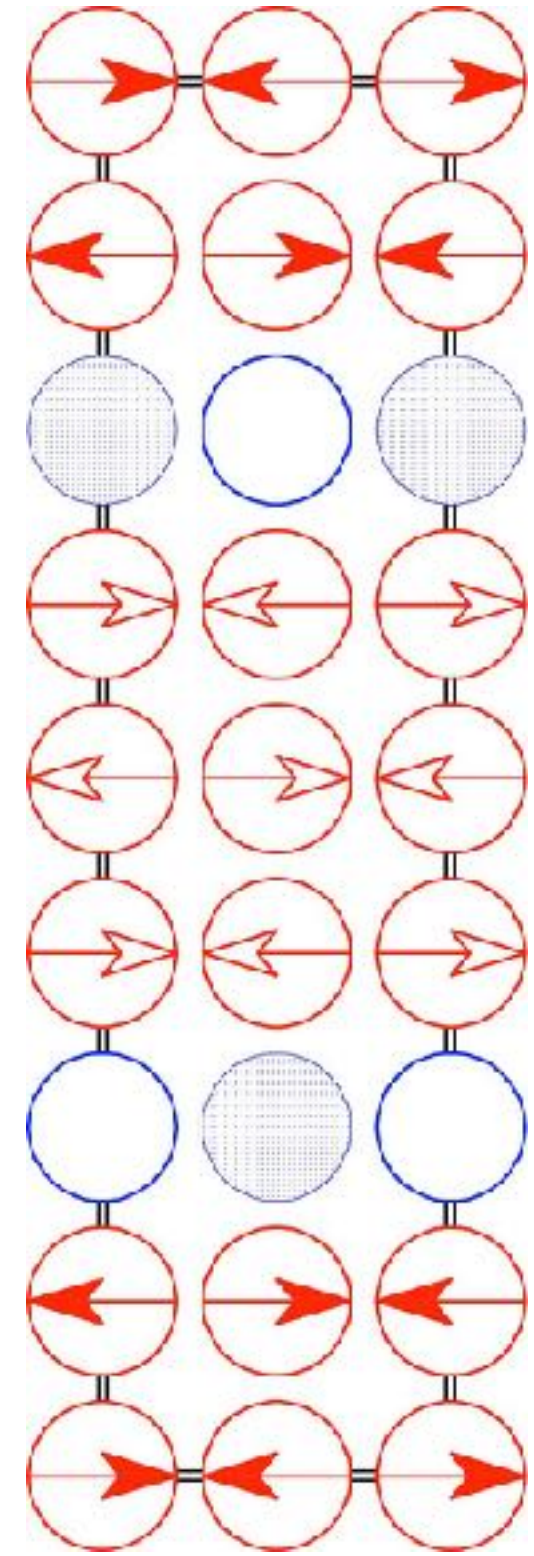
Stripes and superlattice peaks



Spin and charge stripe order

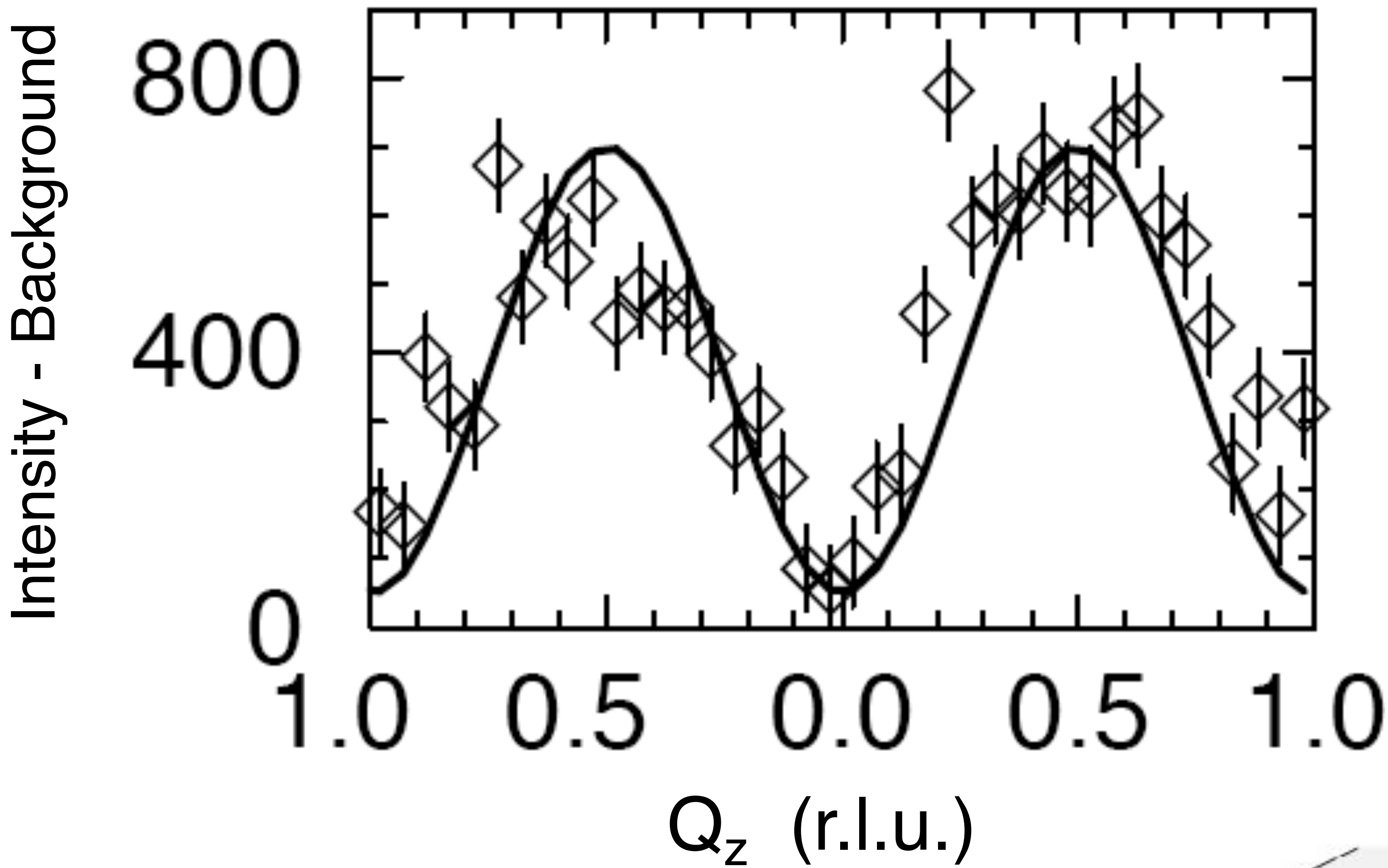






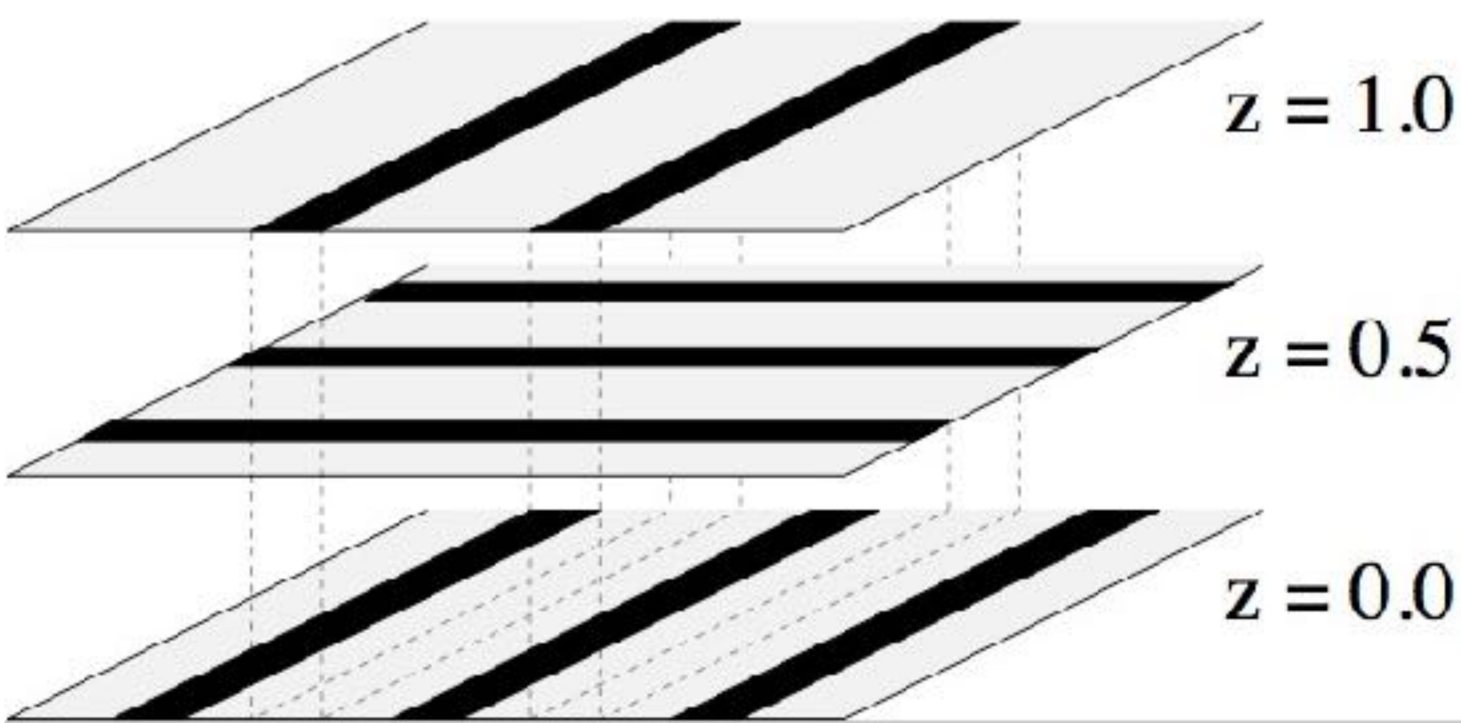
Fujita *et al.*, PRB (2004)

Charge order: stripes, not checkerboard



Hard (100-keV) X-ray diffraction study of $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ shows that there are

4 CuO_2 layers per supercell



Only makes sense in terms of stripe order!

Zimmermann *et al.*, Europhys. Lett. (1998)

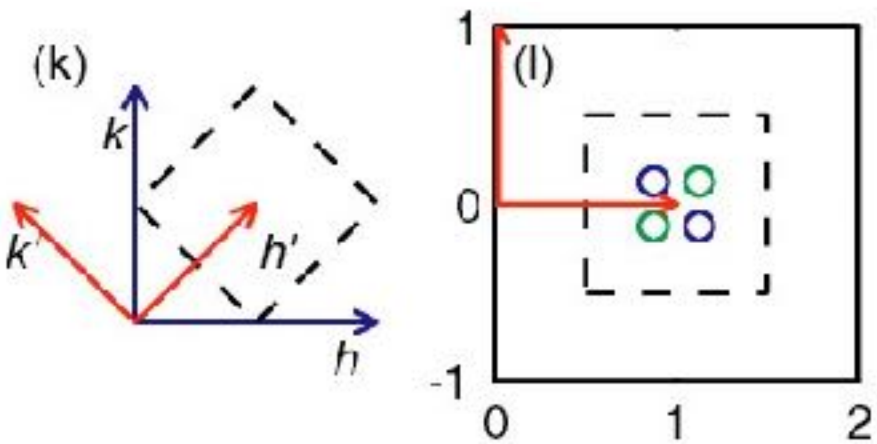
Constant-energy slices through magnetic scattering

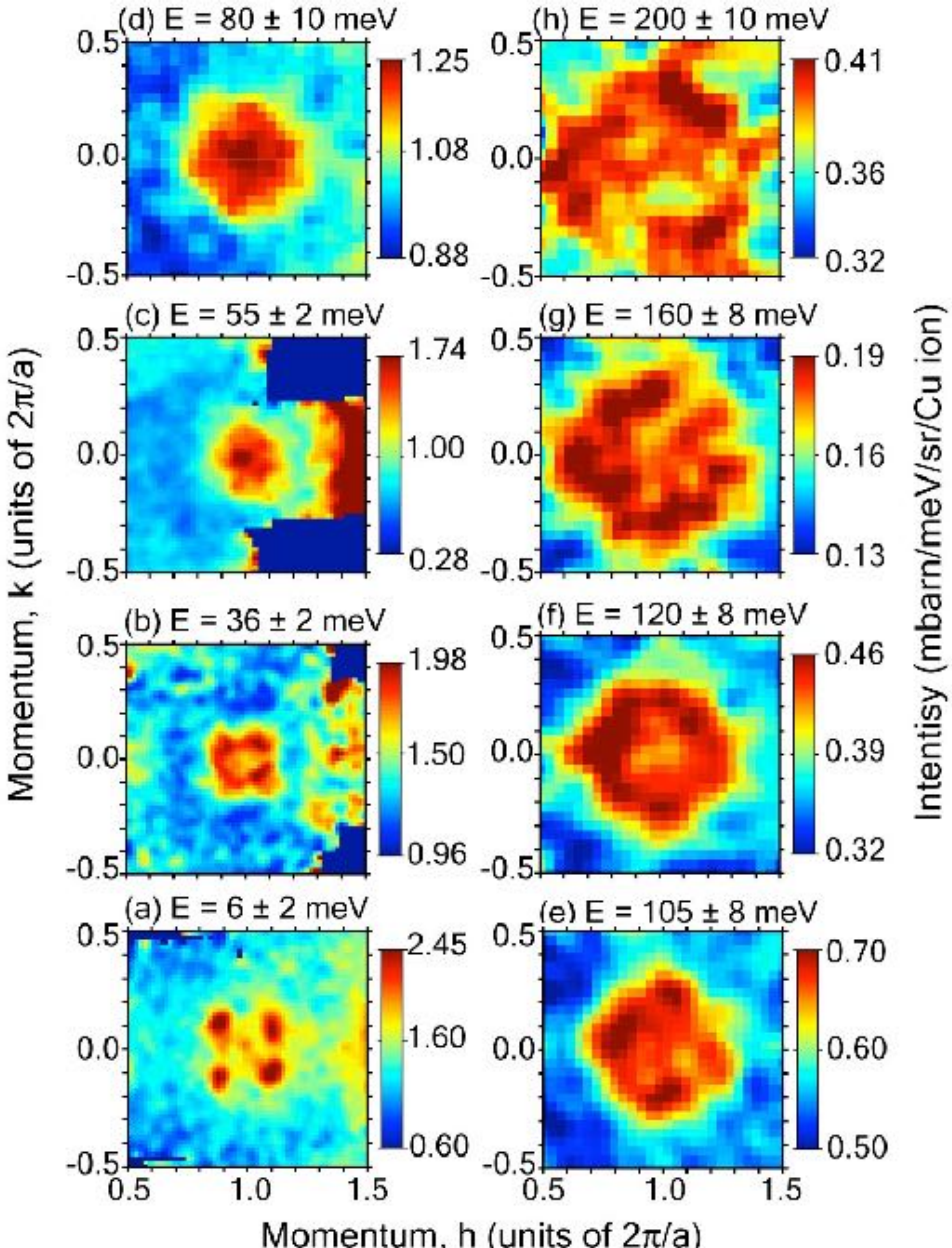
Stripe-ordered
$La_{1.875}Ba_{0.125}CuO_4$

T = 12 K

$T_c < 6$ K

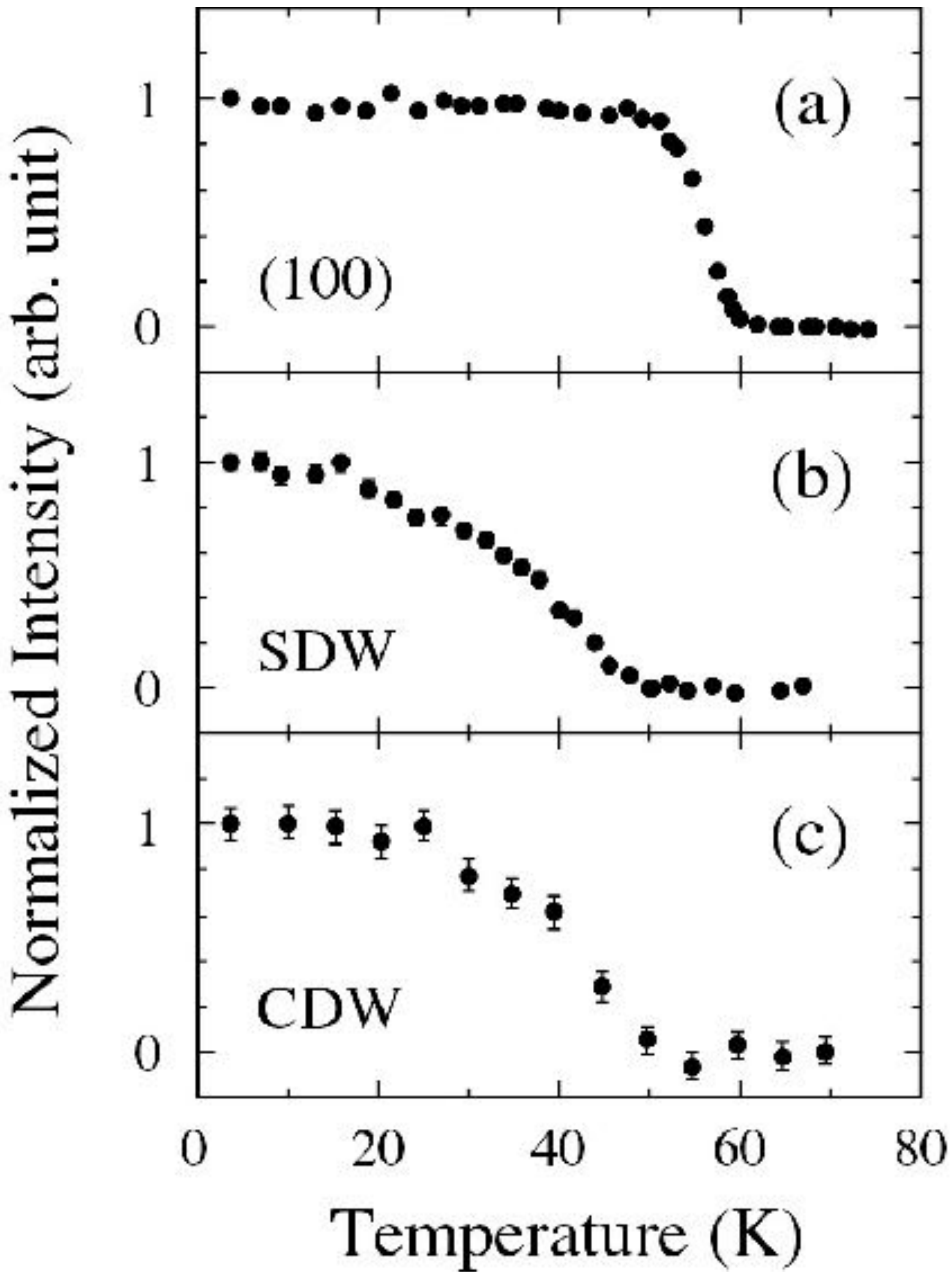
JMT *et al.*, Nature (2004)

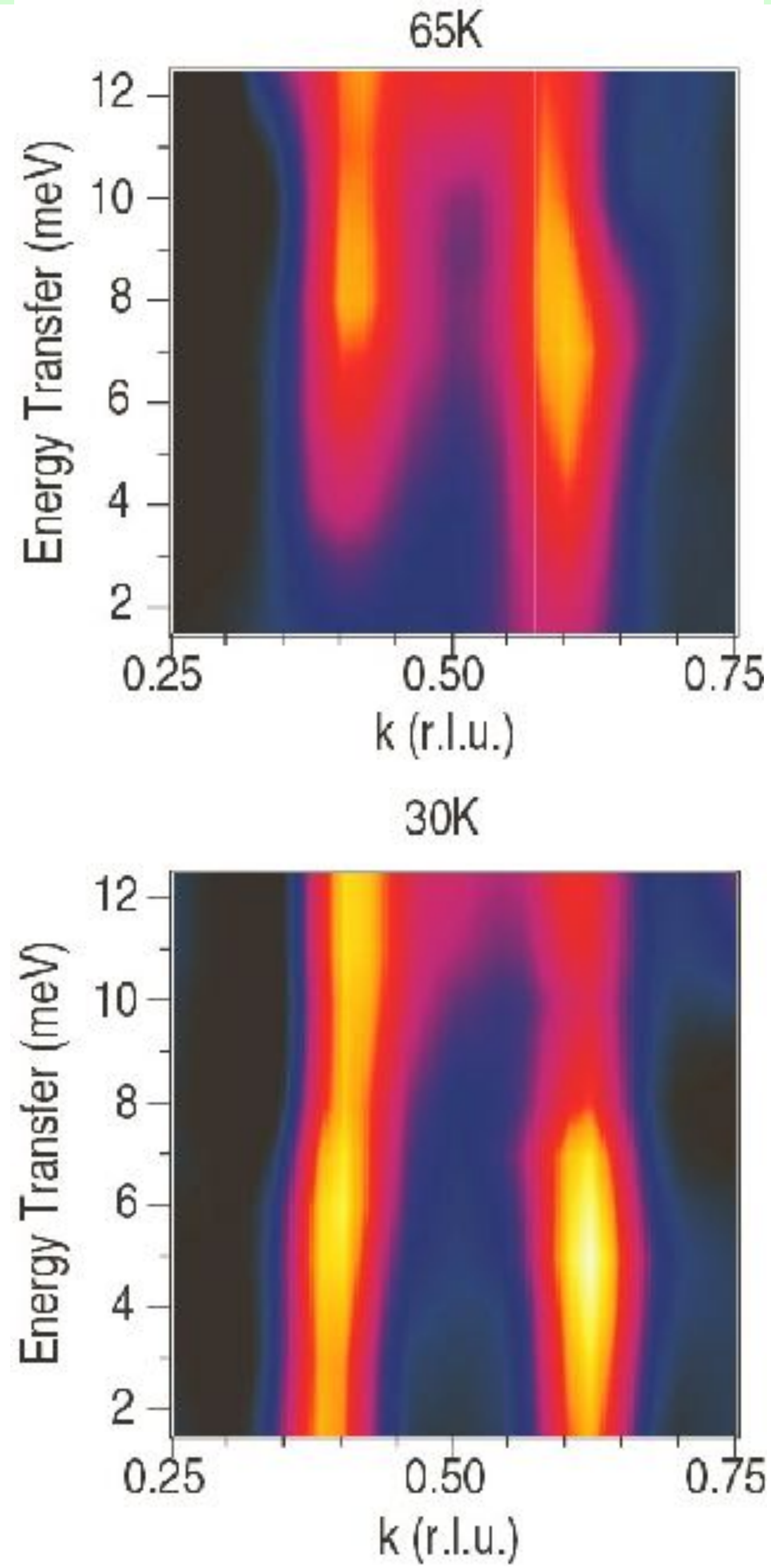




$La_{1.875}Ba_{0.125}CuO_4$

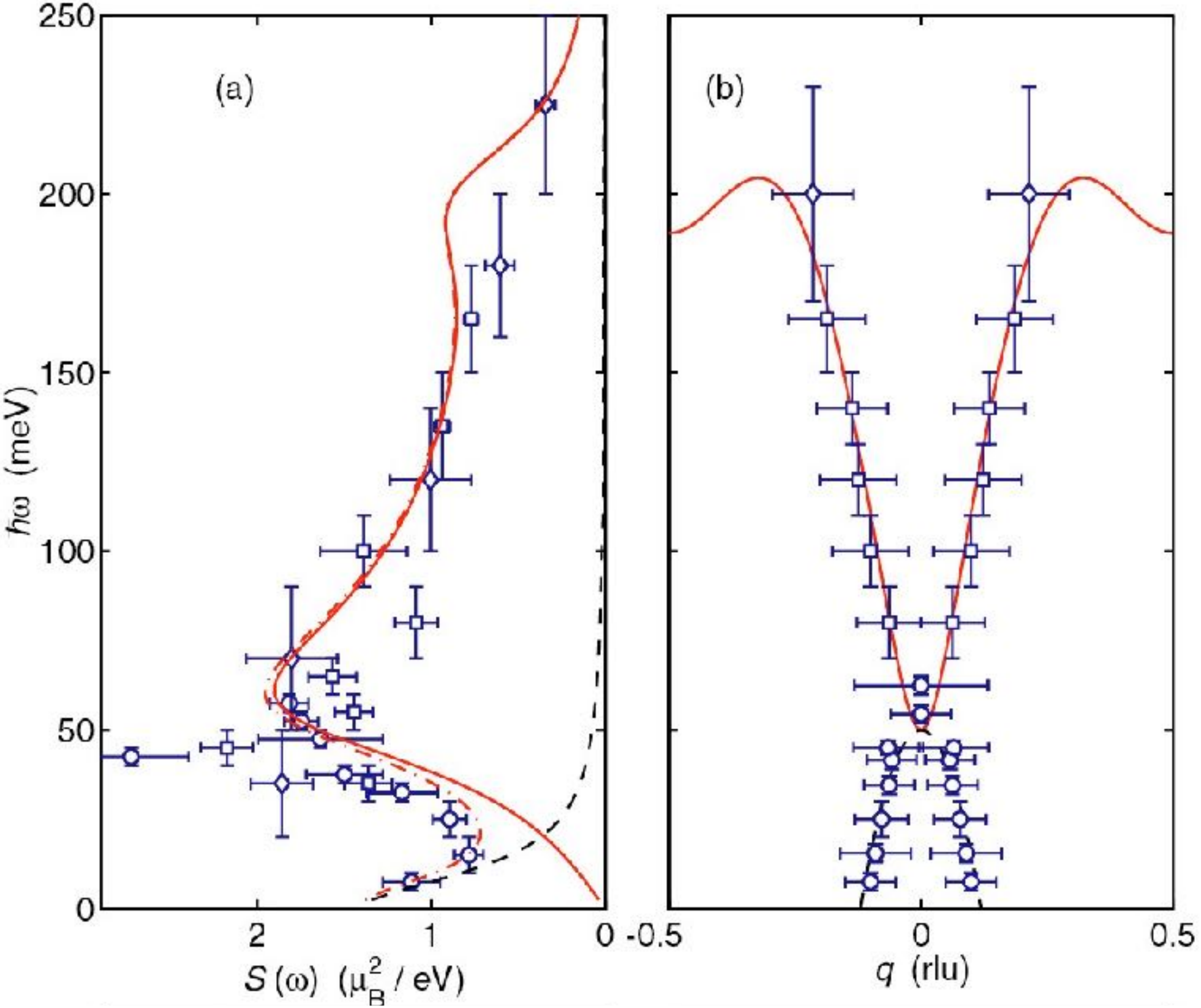
Fujita *et al.*, PRB (2004)



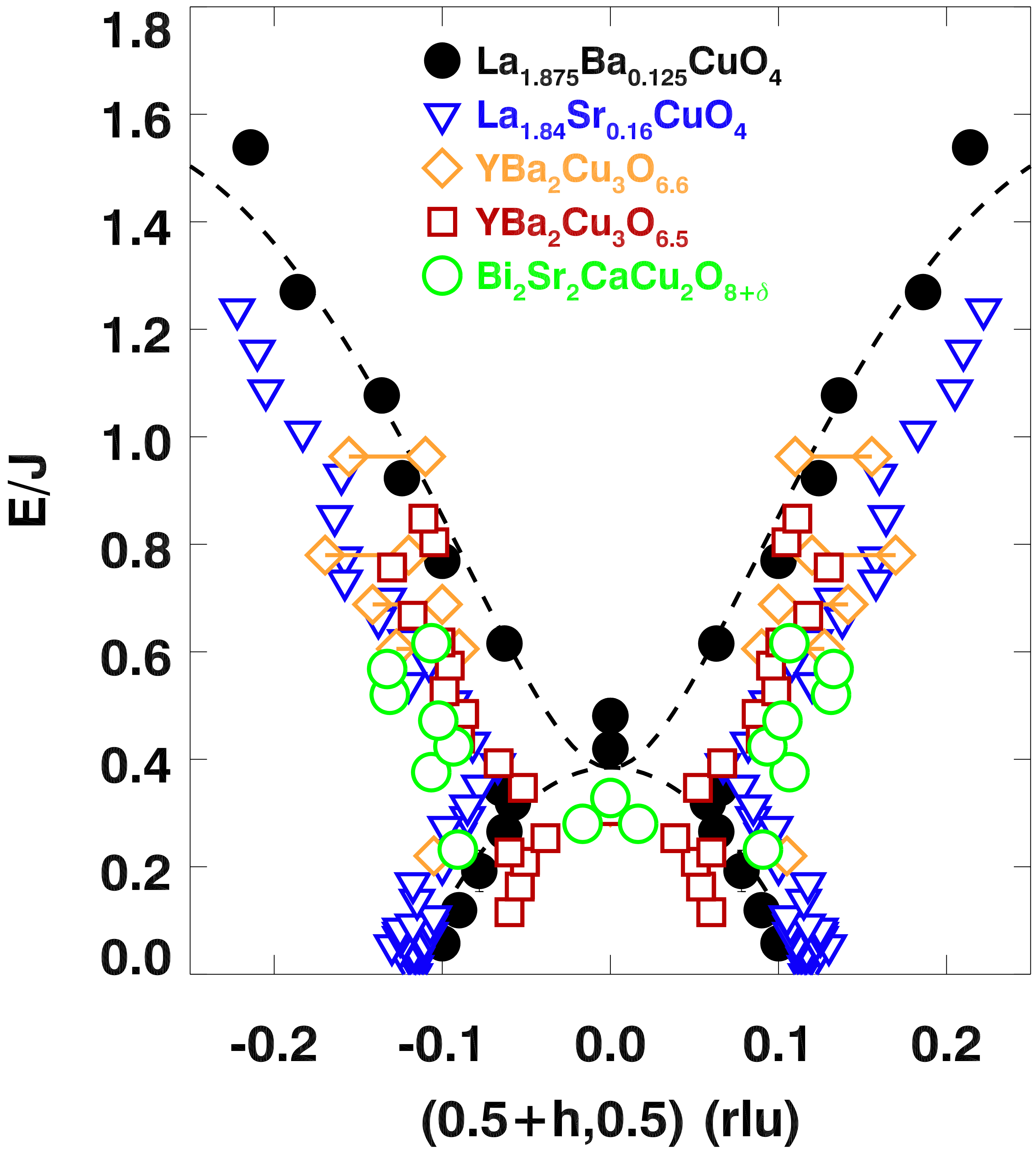


χ''

Spectral weight and dispersion



Universal magnetic spectrum



JMT *et al.*, Nature (2004)

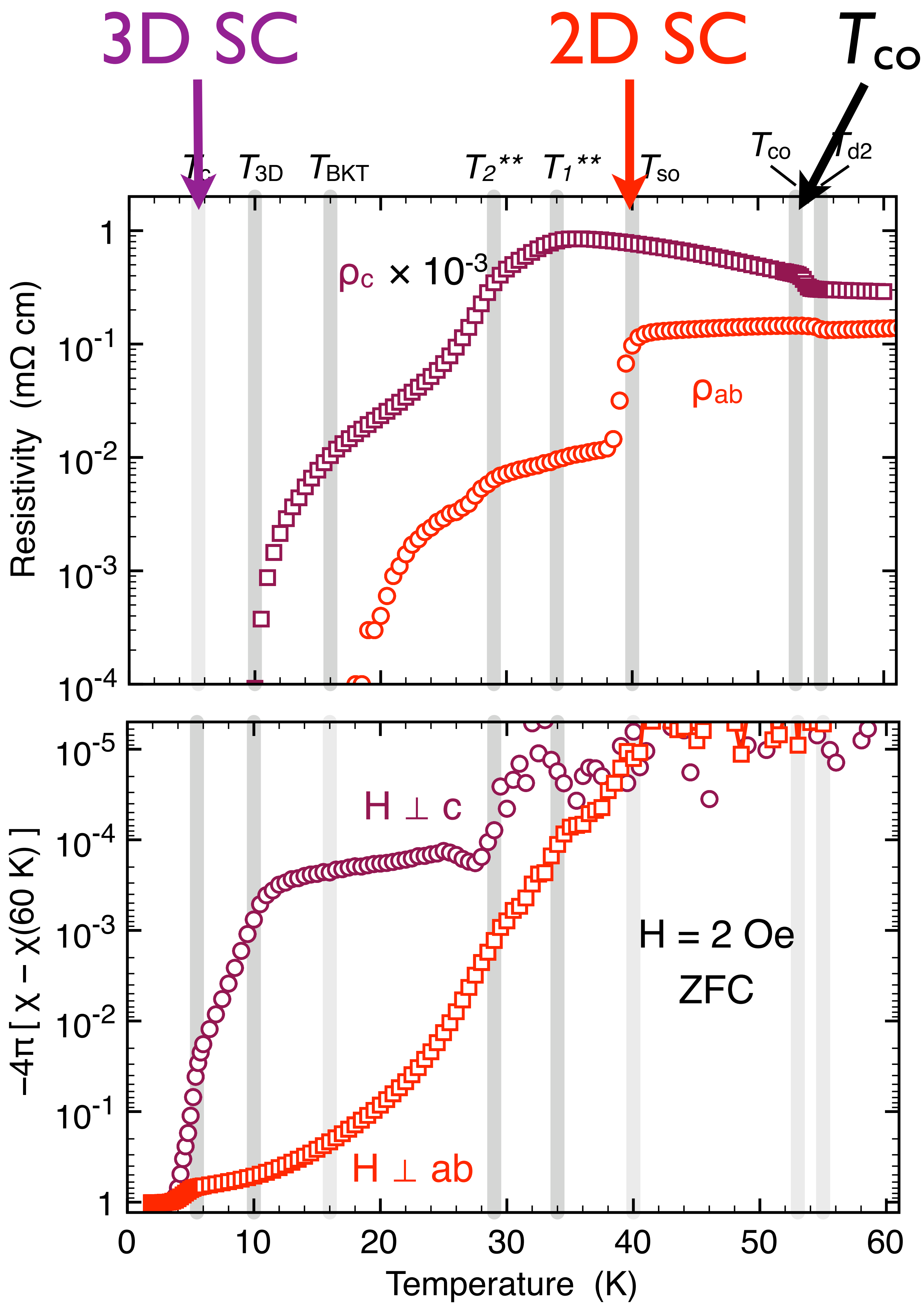
Vignolle *et al.*, Nat. Phys. (2007)

Hayden *et al.*, Nature (2004)

Stock *et al.*, Phys. Rev. B (2005), (2010)

Xu *et al.*, Nat. Phys. (2009)

LBCO x=1/8



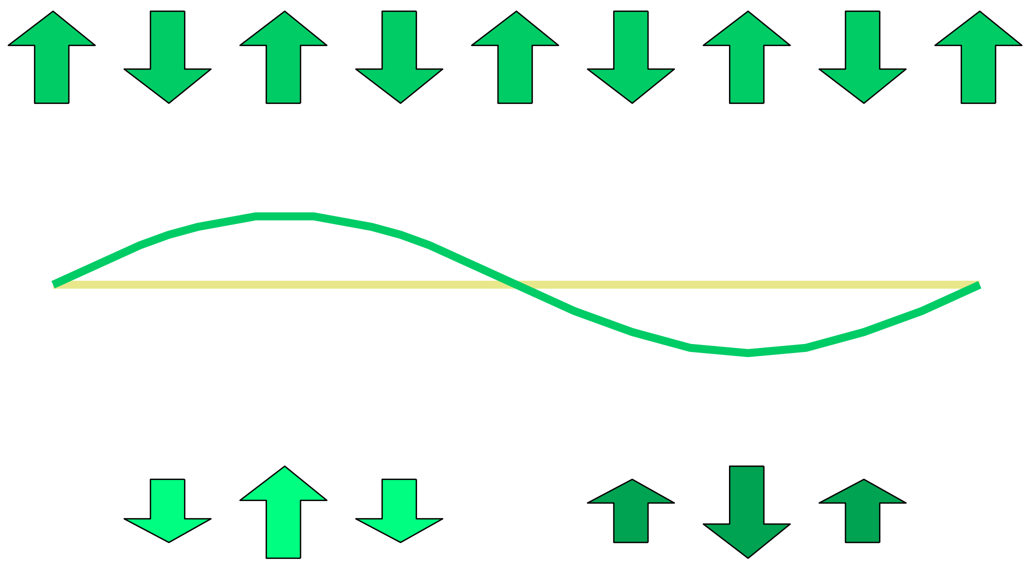
Stripe order:

- compatible with 2D SC at 40 K
- *frustrates* interlayer Josephson coupling

Q. Li *et al.*, PRL (2007)
JMT *et al.*, PRB (2008)

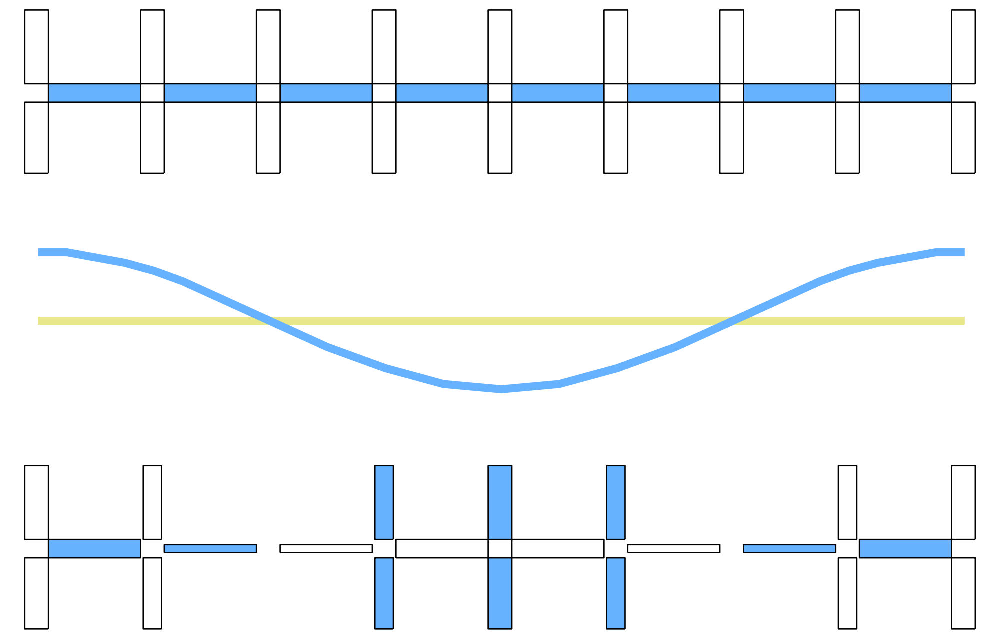
Intertwined Orders

Antiferromagnetism

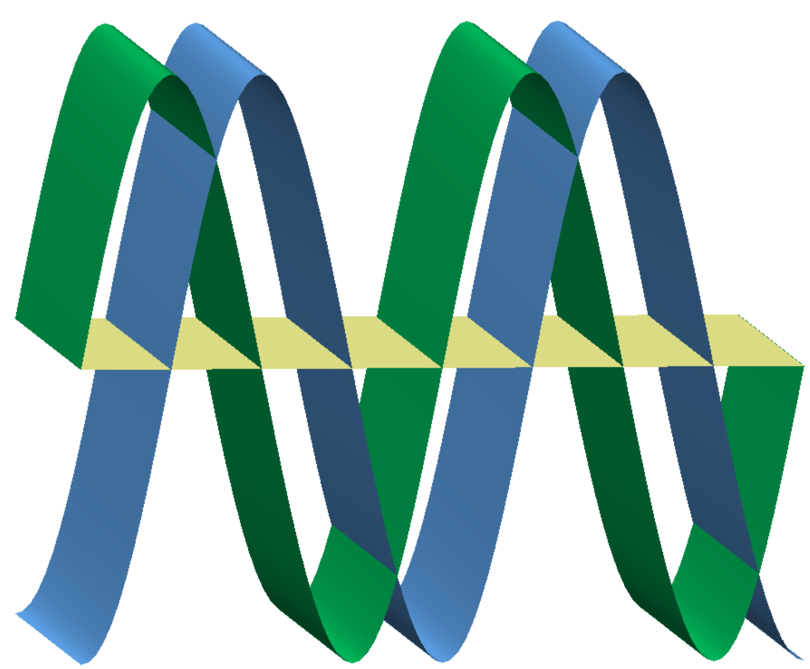


Spin Stripes

Uniform d-wave Superconductor

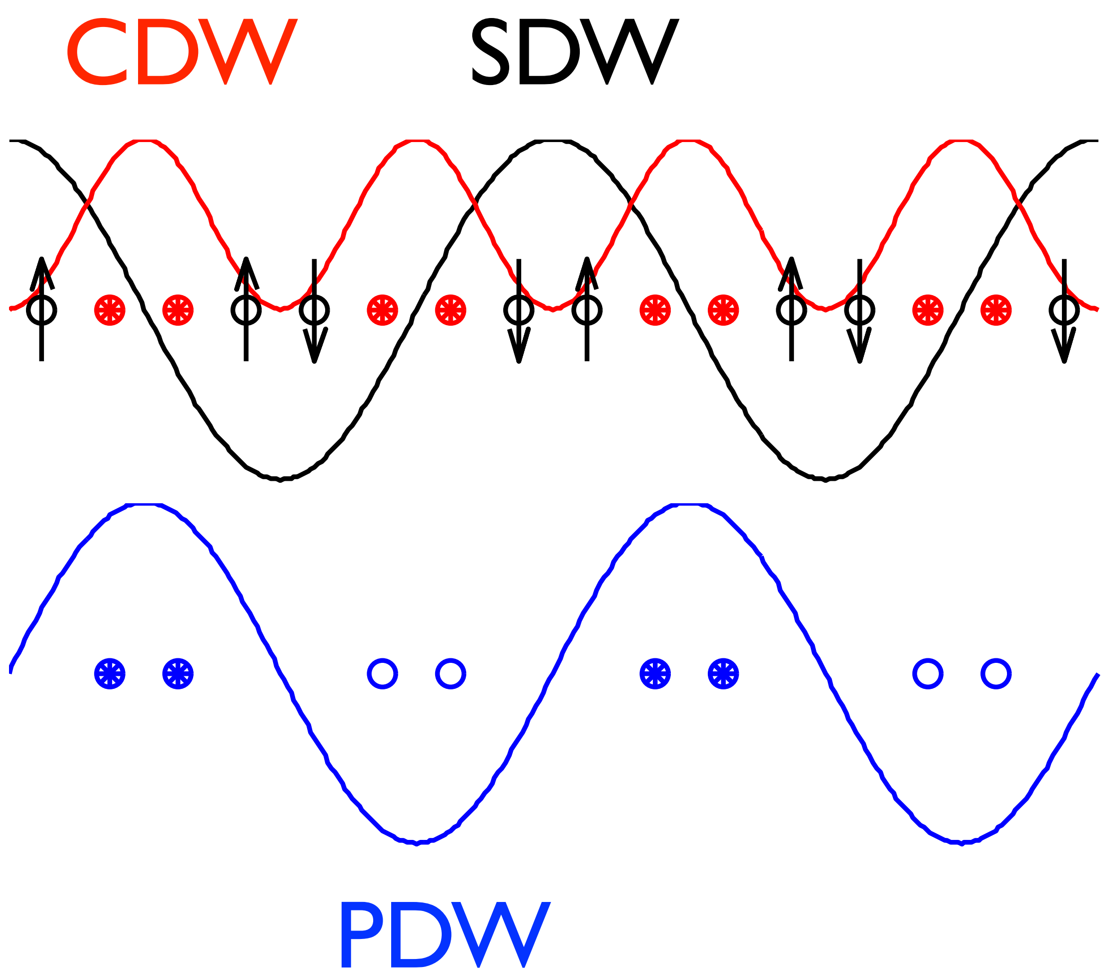


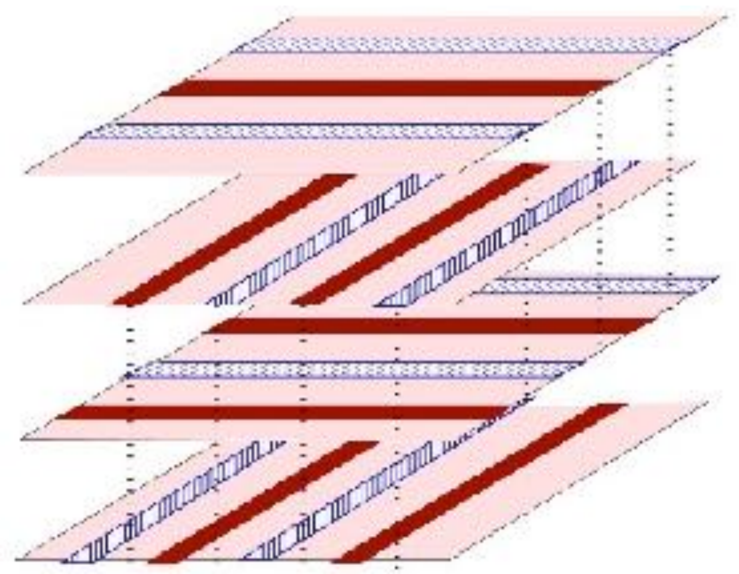
Pair Density Wave



Intertwined antiferromagnetism and superconductivity

2D SC and Pair-Density-Wave Superconductor



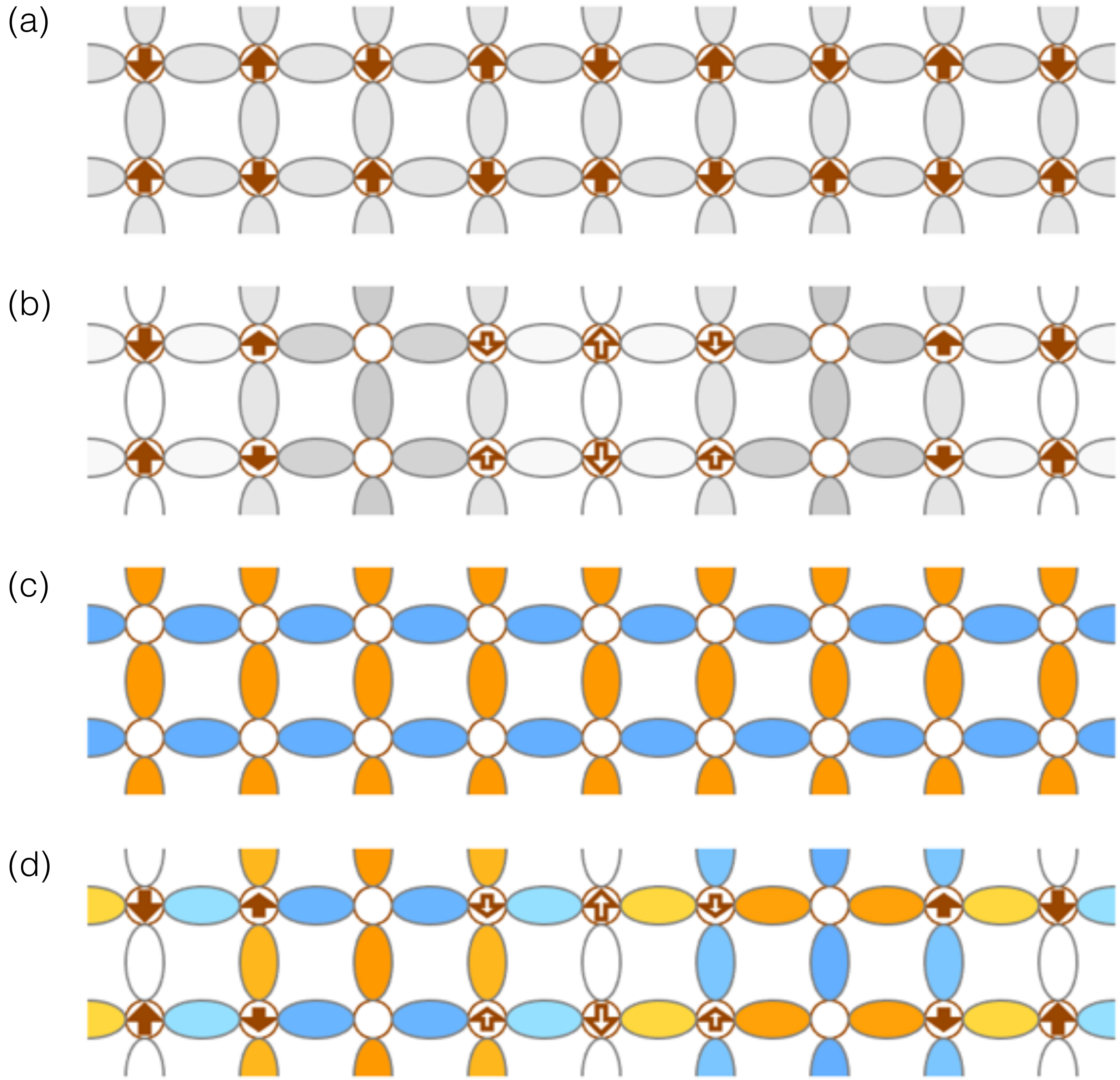


Frustration of interlayer coupling:
Himeda *et al.*, PRL (2002)
Berg *et al.*, PRL (2007)

P.A. Lee, PRX (2014)

Intertwined superconductivity
and antiferromagnetism

Intertwined orders



AF order

SDW+CDW

d-wave SC

Pair density wave
+SDW+CDW

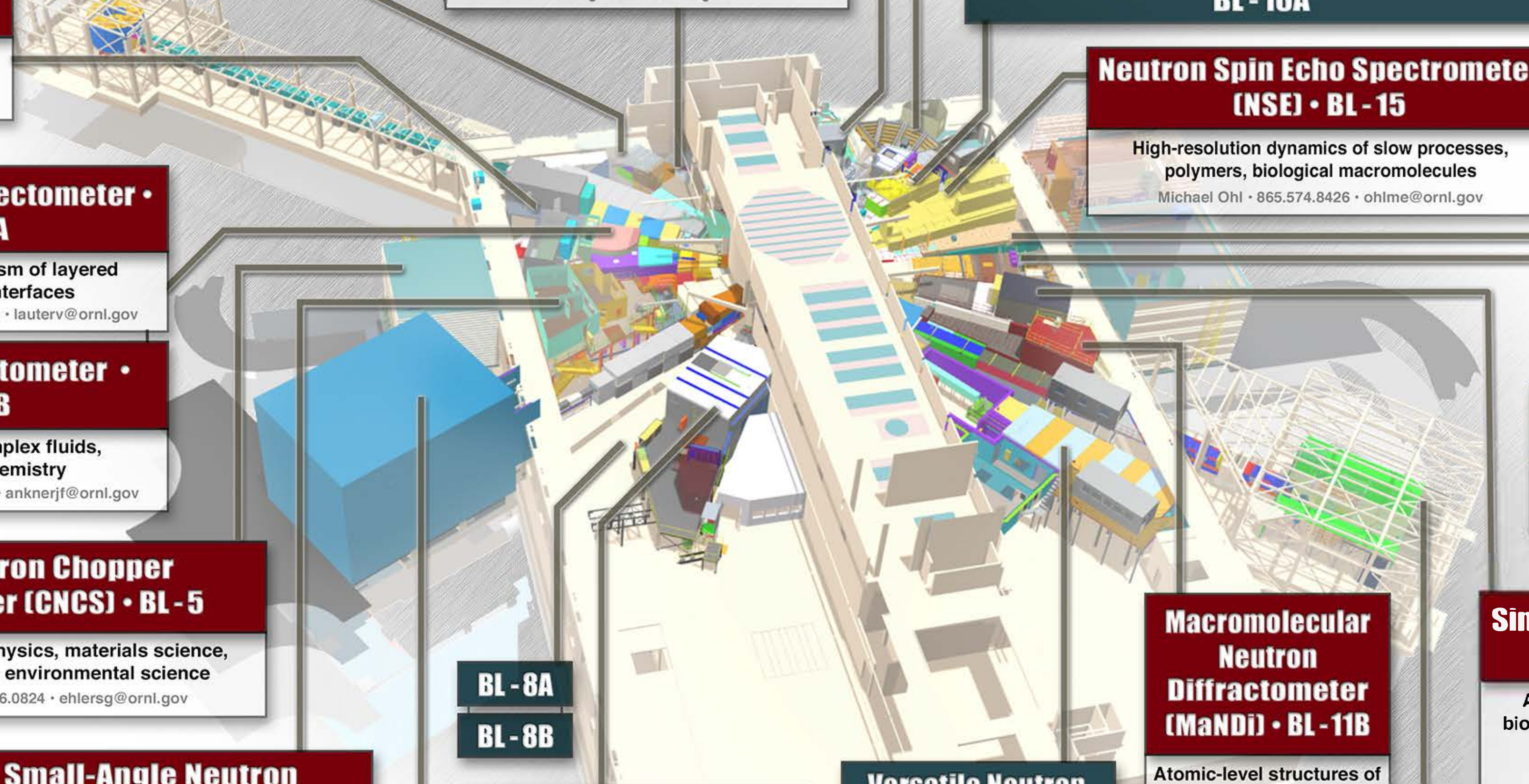
Fradkin et al., RMP (2015)

Spallation Neutron Source

OAK RIDGE National Laboratory | SPALLATION NEUTRON SOURCE

World's most intense pulsed, accelerator-based neutron source

NEUTRONS.ORNL.GOV



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Nanoscale-Ordered Materials Diffractometer (NOMAD) • BL-1B
Liquids, solutions, glasses, polymers, nanocrystalline and partially ordered complex materials
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Wide Angular-Range Chopper Spectrometer (ARCS) • BL-18
Atomic-level dynamics in materials science, chemistry, condensed matter sciences
Doug Abernathy • 865.617.5624 • abernathydl@ornl.gov

Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA) • BL-17
Dynamics of complex fluids, quantum fluids, magnetism, condensed matter, materials science
Matthew Stone • 865.202.6898 • stonemb@ornl.gov

Ultra-Small-Angle Neutron Scattering Instrument (USANS) • BL-1A
Life sciences, polymers, materials science, earth and environmental sciences
Michael Agamalian • 865.382.6367 • magamalian@ornl.gov

Vibrational Spectrometer (VISION) • BL-16B
Vibrational dynamics in molecular systems, chemistry
Yongqiang Cheng • 865.5576.9064 • chengy@ornl.gov

BL-16A

Spallation Neutrons and Pressure Diffractometer (SNAP) • BL-3
Materials science, geology, earth and environmental sciences
Chris Tulk • 865.574.5764 • tulkca@ornl.gov

Neutron Spin Echo Spectrometer (NSE) • BL-15
High-resolution dynamics of slow processes, polymers, biological macromolecules
Michael Ohl • 865.574.8426 • ohlme@ornl.gov

Hybrid Spectrometer (HYSPEC) • BL-14B
Atomic-level dynamics in single crystals, magnetism, condensed matter sciences
Barry Winn • 865.805.6819 • winnbl@ornl.gov

BL-14A

Magnetism Reflectometer • BL-4A
Chemistry, magnetism of layered systems and interfaces
Valeria Lauter • 865.387.5389 • lauterv@ornl.gov

Liquids Reflectometer • BL-4B
Interfaces in complex fluids, polymers, chemistry
John Ankner • 865.377.4523 • anknerjf@ornl.gov

Fundamental Neutron Physics Beam Line • BL-13
Fundamental properties of neutrons
Geoffrey Greene • 865.576.0023 • greenegl@ornl.gov

Cold Neutron Chopper Spectrometer (CNCS) • BL-5
Condensed matter physics, materials science, chemistry, biology, environmental science
Georg Ehlers • 865.206.0824 • ehlersg@ornl.gov

BL-8A

BL-8B

Macromolecular Neutron Diffractometer (MaNDi) • BL-11B
Atomic-level structures of membrane proteins, drug complexes, DNA
Leighton Coates • 865.964-8651 • coatesl@ornl.gov

Single-Crystal Diffractometer (TOPAZ) • BL-12
Atomic-level structures in chemistry, biology, earth science, materials science, condensed matter physics
Christina Hoffmann • 865.576.5127 • choffmann@ornl.gov

Extended Q-Range Small-Angle Neutron Scattering Diffractometer (EQ-SANS) • BL-6
Life science, polymer and colloidal systems, materials science, earth and environmental sciences
William Heller • 865.241.0093 • hellerwt@ornl.gov

Elastic Diffuse Scattering Spectrometer (CORELLI) • BL-9
Detailed studies of disorder in crystalline materials
Feng Ye • 865.576.0931 • yef1@ornl.gov

Versatile Neutron Imaging Instrument at SNS (VENUS) • BL-10
Energy selective imaging in materials science, engineering, materials processing, environmental sciences and biology
Hassina Bilheux • 865.384.9630 • bilheuxhn@ornl.gov

Powder Diffractometer (POWGEN) • BL-11A
Atomic-level structures in chemistry, materials science, and condensed matter physics including magnetic spin structures
Ashfia Huq • 630.986.7321 • huqa@ornl.gov

Engineering Materials Diffractometer (VULCAN) • BL-7
Mechanical behaviors, materials science, materials processing
Ke An • 865.919.5226 • kean@ornl.gov

*Scheduled commissioning date

LEGEND
- Operating instrument in user program
- In design or construction
- Under consideration

15-G00337A/gim

The Spallation Neutron Source is a facility of Oak Ridge National Laboratory, managed by UT-Battelle for the US Department of Energy.

HYSPEC

HYbrid SPECtrometer

BL14B at the SNS (ORNL)

Time-of-flight with area detector

Polarization analysis

NIST Center for Neutron Research