Neutron and x-ray scattering studies of superconductors

lecture 3: x-ray scattering from cuprates

- resonant inelastic x-ray scattering from magnons and paramagnons
- resonant elastic x-ray scattering from charge density waves
- CDW in cuprate superlattices
- electron-phonon interaction

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Eliashberg theory

ARPES dispersion

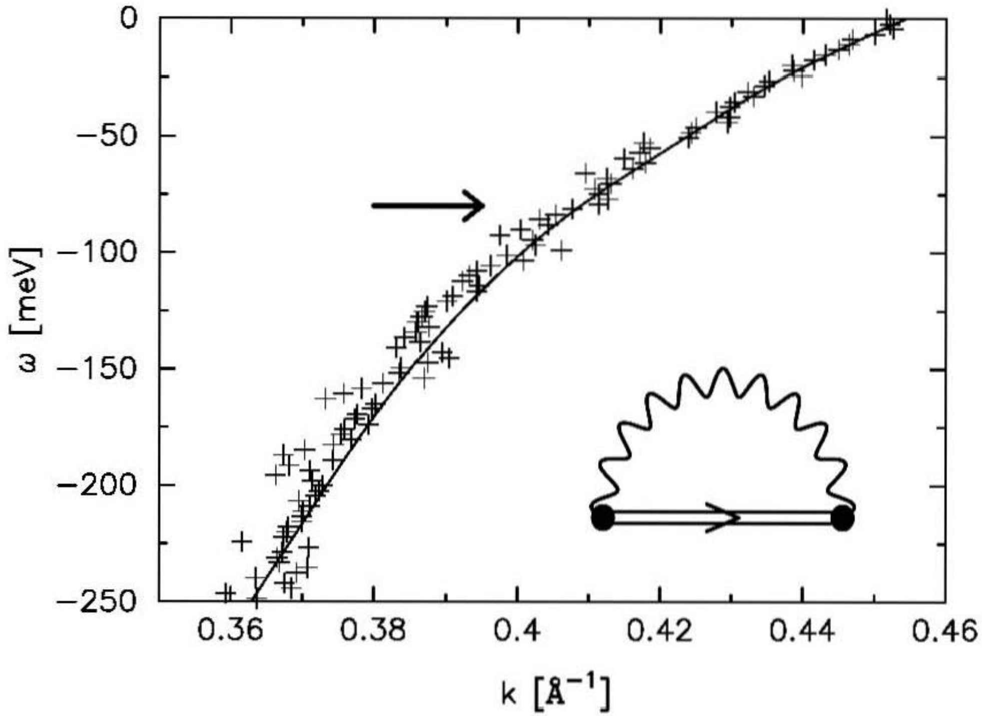
- compare INS & ARPES spectra of underdoped $YBa_2Cu_3O_{6.6}$ (T_c = 62 K)

  → **spin-fermion coupling constant**

- mean-field superconducting gap equation

  → **T_c ~ 170 K**

Dahm et al., Nature Phys. 2009



spin fluctuations have sufficient strength to mediate high-temperature superconductivity

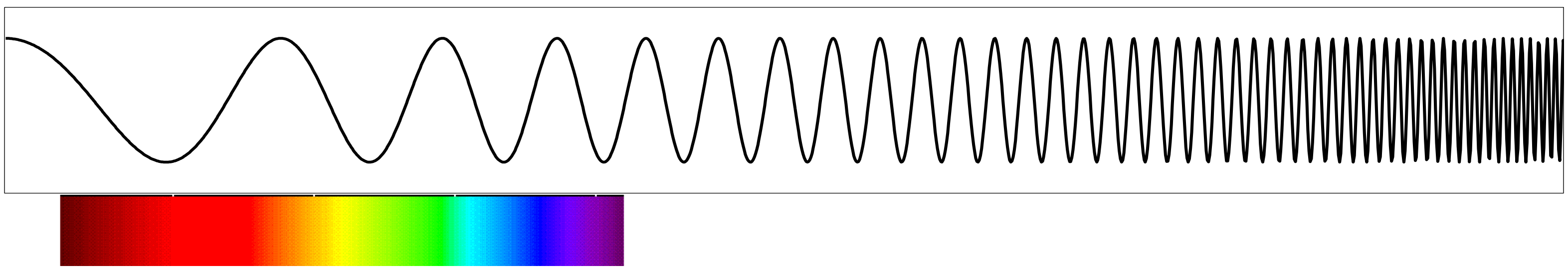
… but what about optimal doping?

→ use x-rays to detect high-energy excitations

X-rays

max. transferred momentum	$Q \ll \pi/a$ $Q \sim 0$ 'zone center'		$Q \sim \pi/a$ $Q \sim 1^{st}$ BZ	$Q \gg \pi/a$ $Q \sim$ several BZ	

electromagnetic spectrum

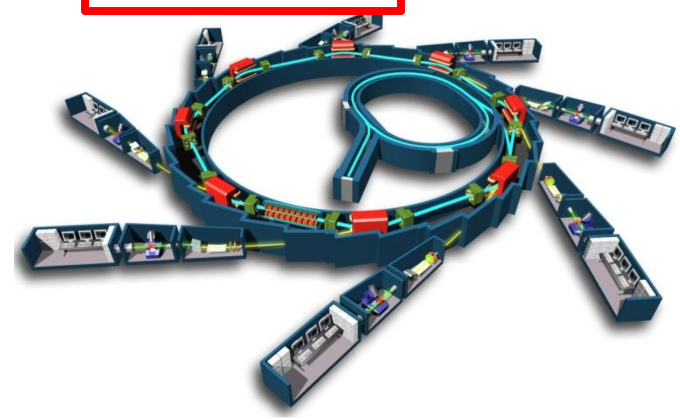


wavelength	10^4 Å	10^3 Å	10^2 Å	10 Å	0.1 Å
energy	1 eV	10 eV	100 eV	1 keV	100 keV

Raman scattering

$\boxed{\text{soft x-ray RIXS}}$

hard x-ray
IXS

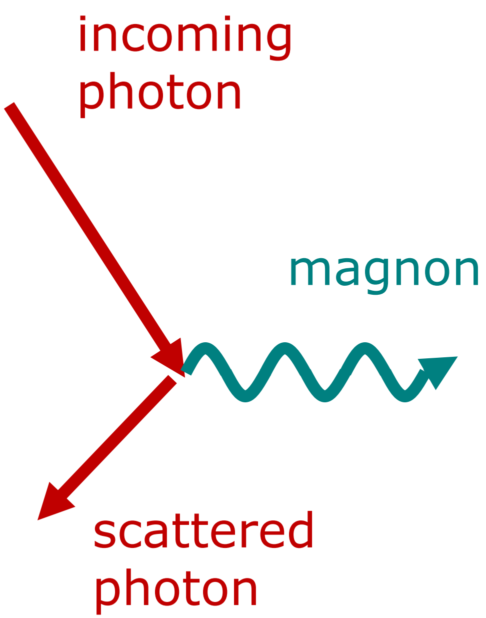


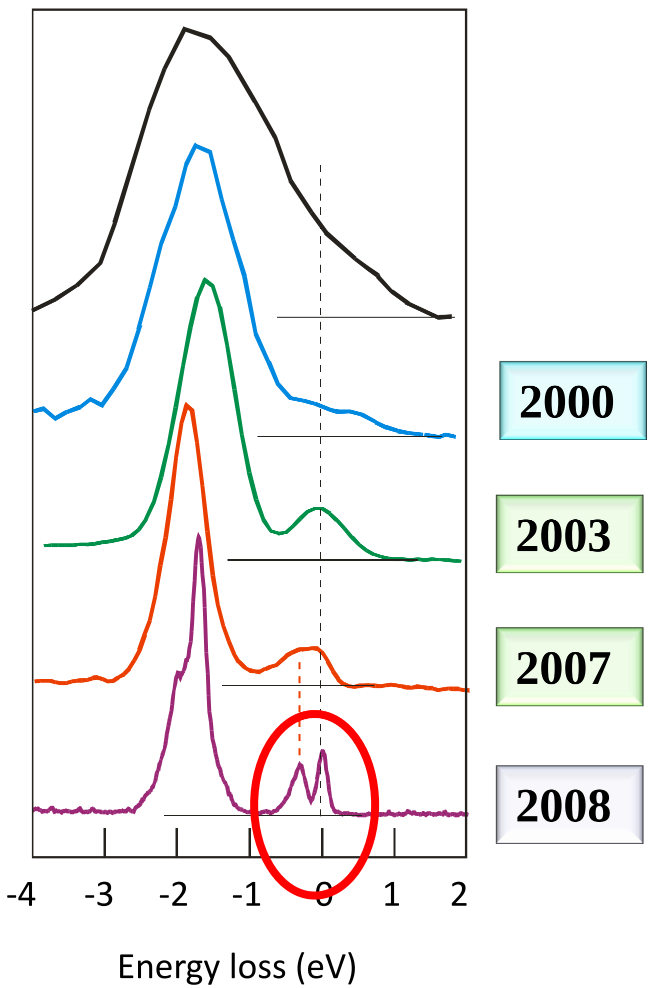
Resonant inelastic x-ray scattering (RIXS)

triple-axis spectrometry with soft x-rays

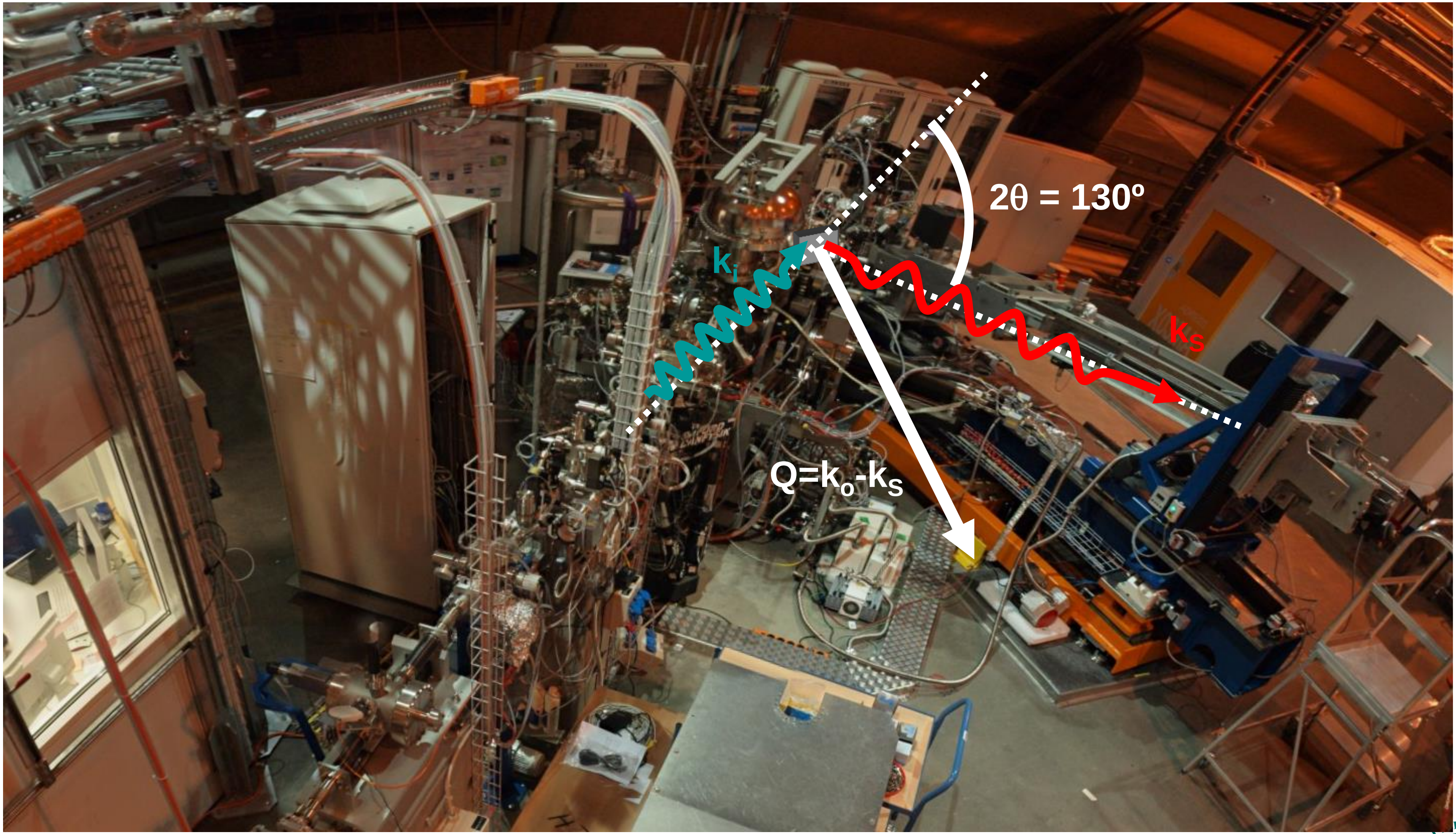
order-of-magnitude increase in in energy resolution
L. Braicovich, G. Ghiringhelli (Univ. Milano)

La_2CuO_4
Cu 2*p* → 3*d*
photon energy ~ 931 eV





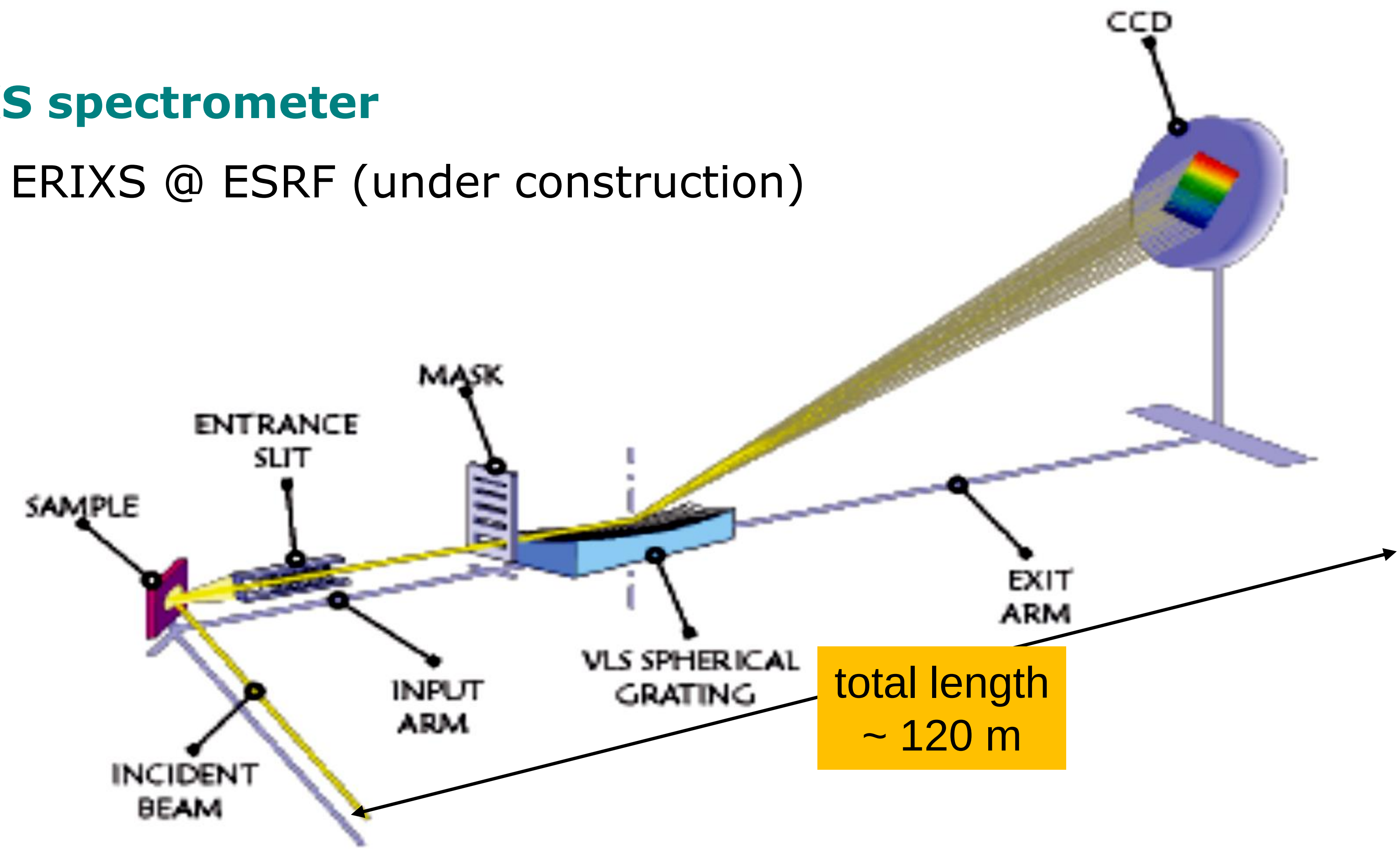
SAXES Spectrometer @ Swiss Light Source



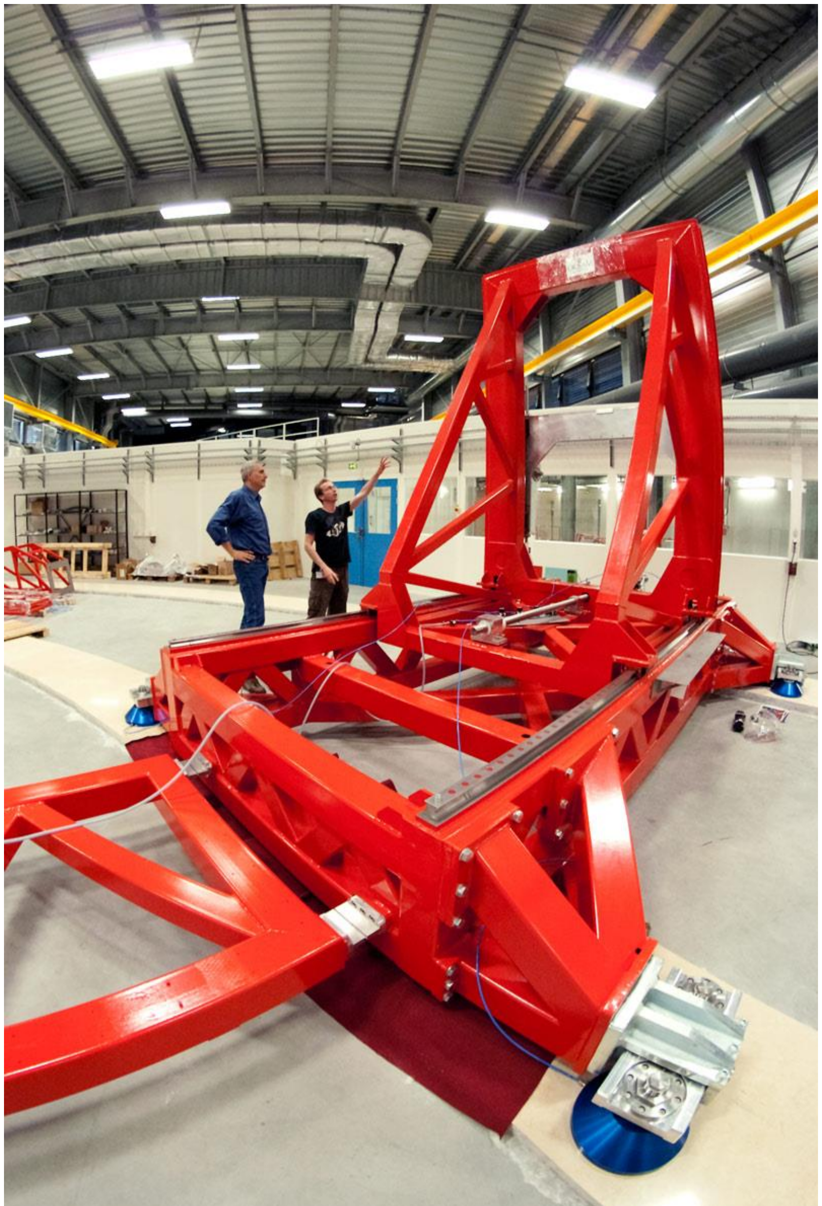
Resonant inelastic x-ray scattering (RIXS)

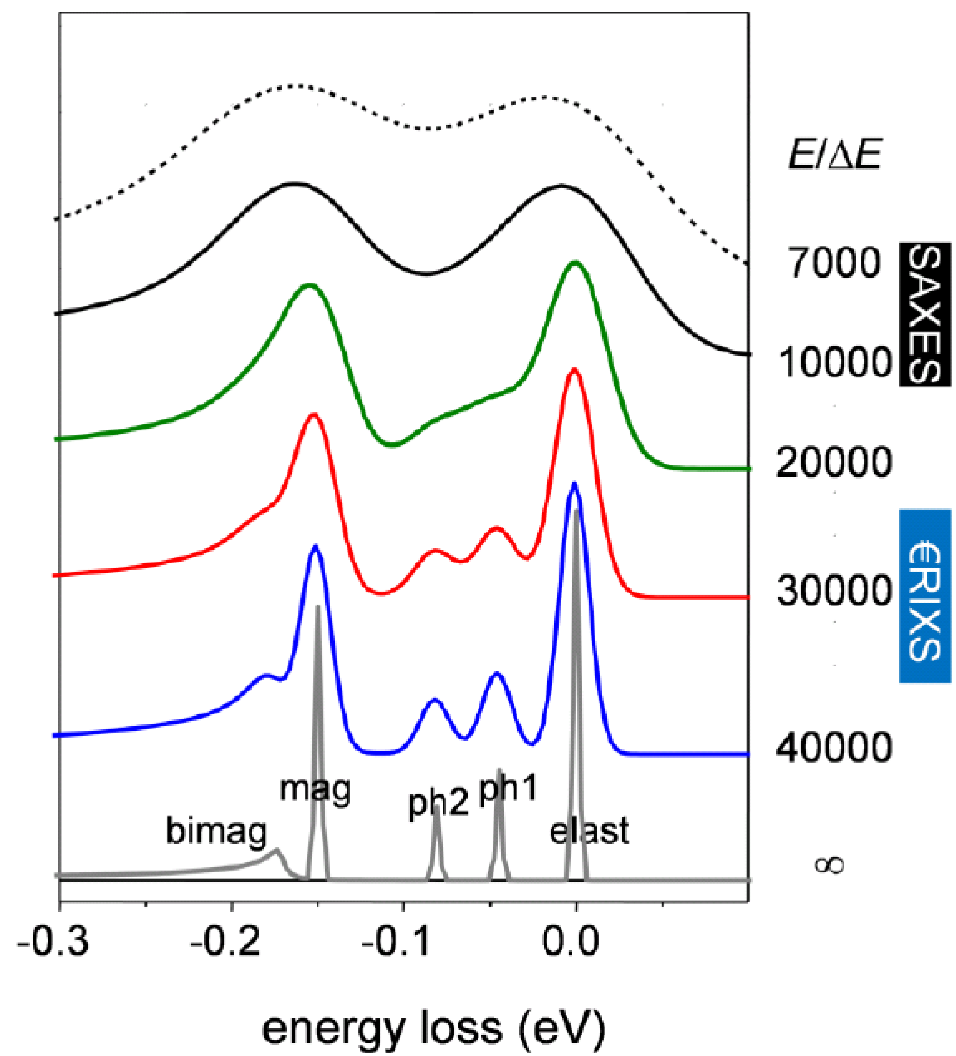
RIXS spectrometer

e.g. ERIXS @ ESRF (under construction)



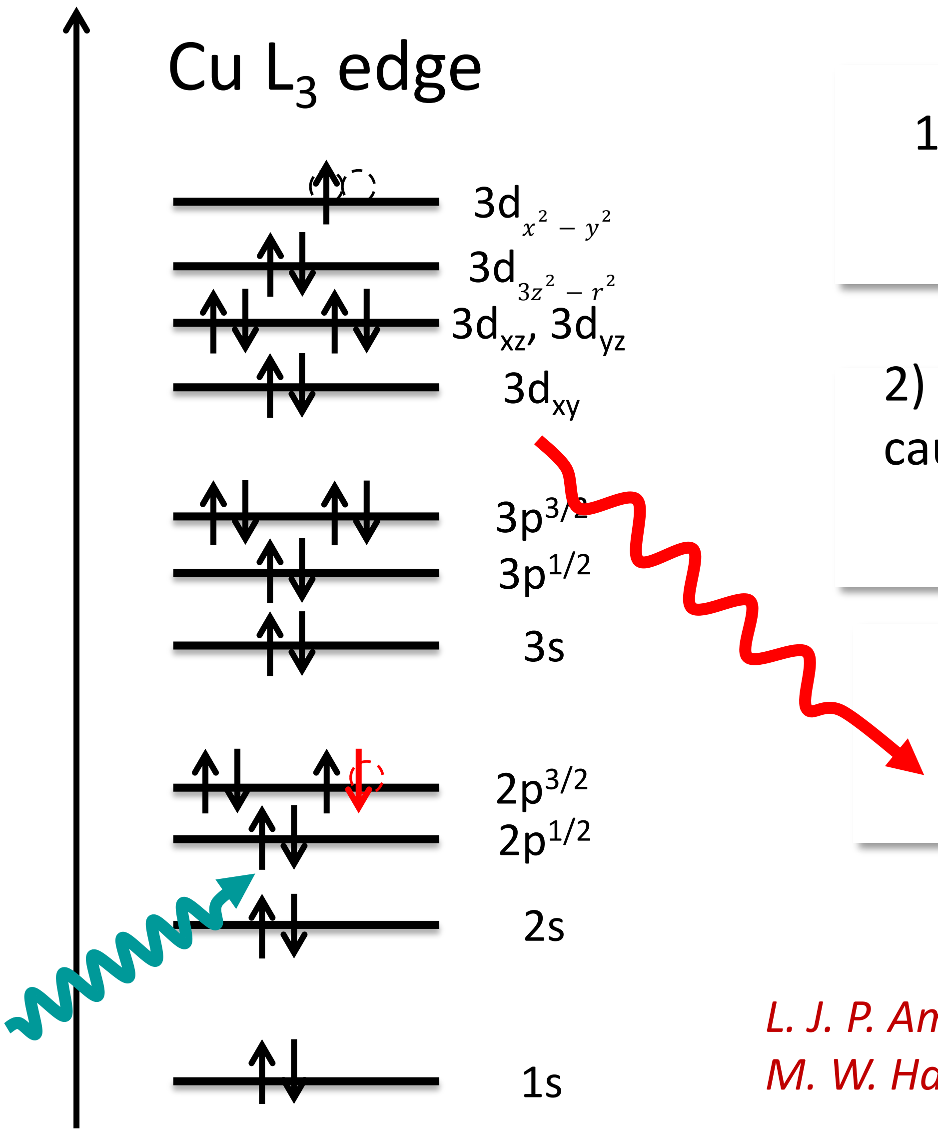
ERIXS Spectrometer @ ESRF





für Festkörperforschung

RIXS from La_2CuO_4



1) Resonant absorption at the Cu L_3 edge

$2p^6 3d^9 \longrightarrow 2p^5 3d^{10}$

2) Spin-Orbit coupling of the core hole can cause a spin flip

$$\vec{L} \cdot \vec{S} = LzSz + (L^+ S^- + L^- S^+)/2$$

3) Optical decay of the core hole

$2p^5 3d^{10} \longrightarrow 2p^6 3d^{9*}$

L. J. P. Ament et al., PRL 2009
M. W. Haverkort, PRL 2010

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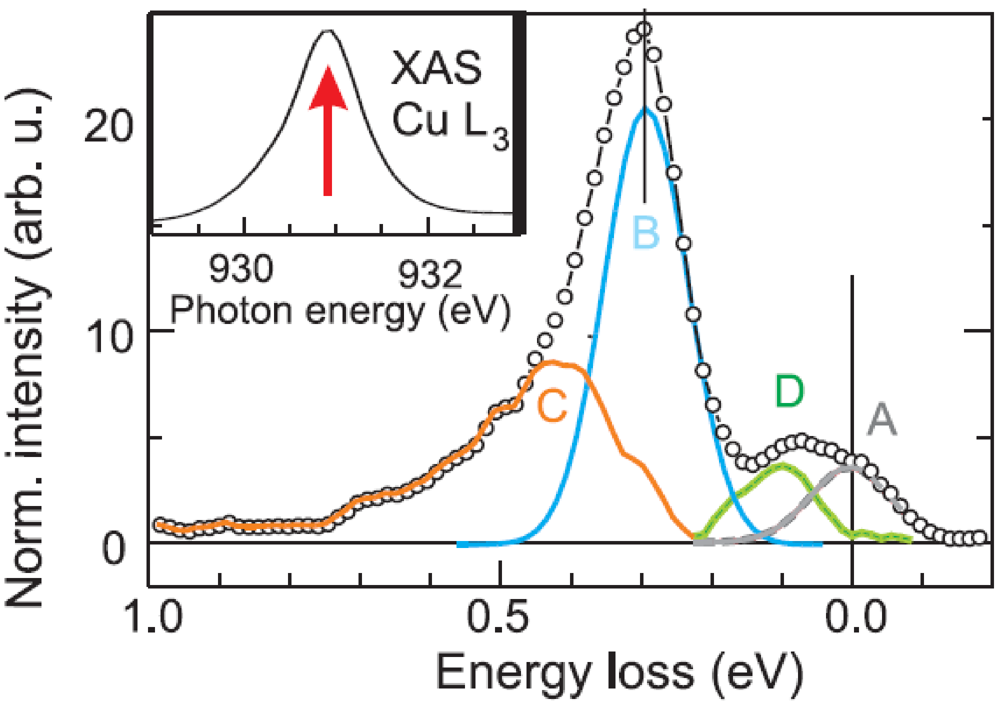
RIXS from La_2CuO_4

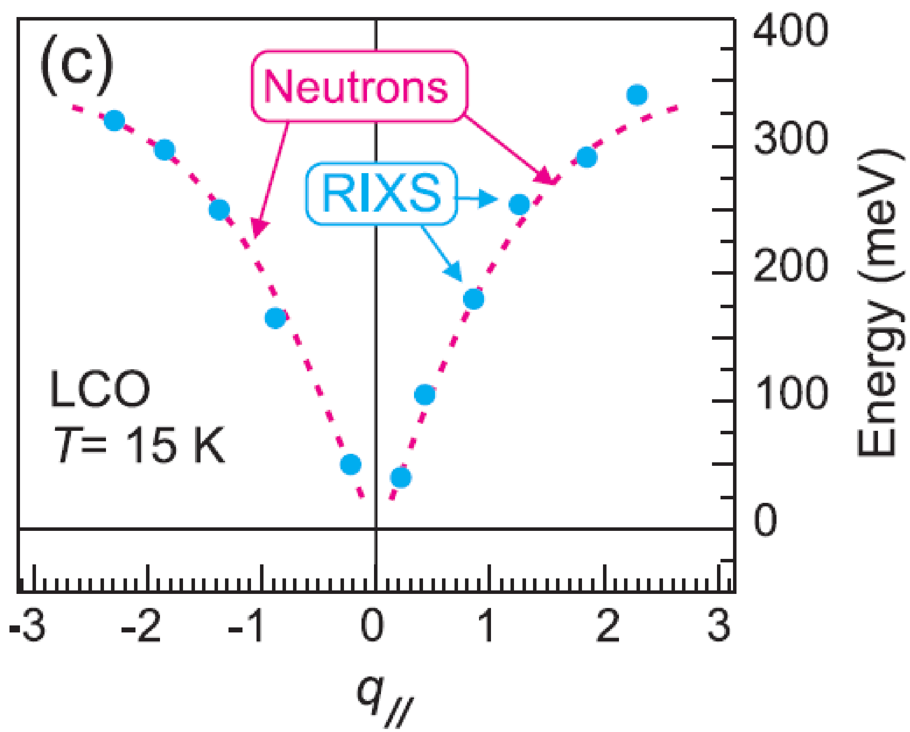
RIXS spectrum

multi-magnons   single magnons   phonons & defects



dispersion of single-magnon excitations

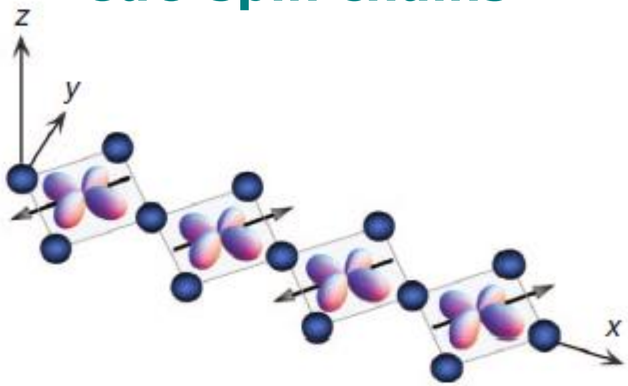
excellent agreement with neutrons



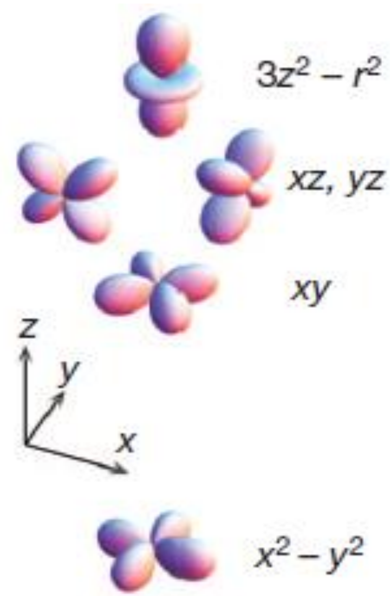
Braicovich et al., PRL 2009, 2010

RIXS from Sr_2CuO_3

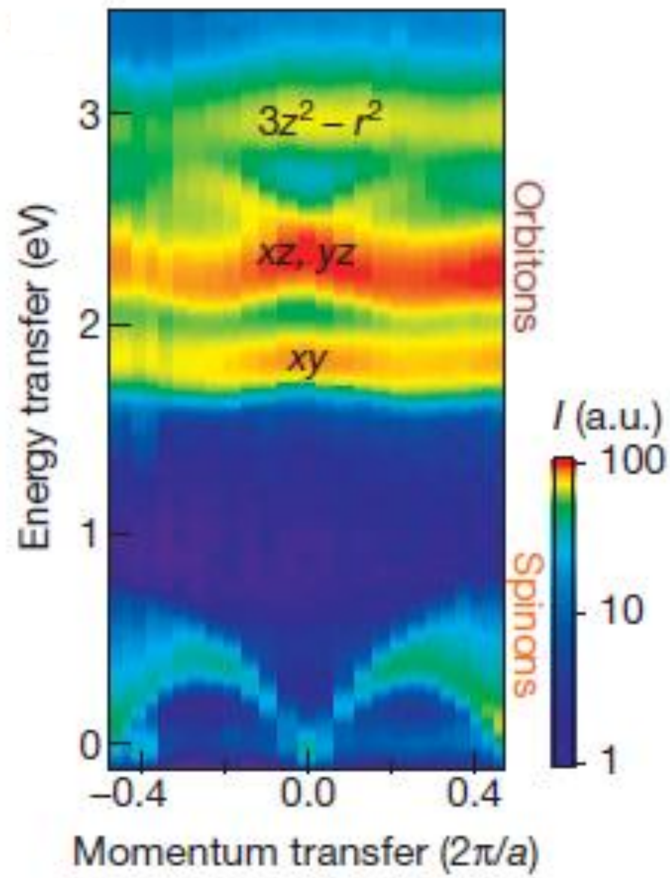
CuO spin chains



orbitally non-degenerate
similar to 2D cuprates



Schlappa et al. Nature 2012



cuprates: spin-orbital separation, low-energy excitations are spin-only

orbitally degenerate systems
e.g. iron pnicides: mixing of spin and orbital excitations at low energies

RIXS theory

reminder: neutrons

magnetic scattering cross section completely understood

theorists can focus on calculating S(Q,ω) ~ χ‘‘(Q,ω)

polarization factor

$$\frac{d^2\sigma}{d\Omega\, dE} = (\gamma r_0)^2 \frac{k_f}{k_i} N \left|F(\mathrm{Q})\right|^2 e^{-2W} \sum_{\alpha\beta} \boxed{(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)} S^{\alpha\beta}(\mathrm{Q},\omega)$$

$$S^{\alpha\beta}(\mathrm{Q},\omega) = \frac{1}{2\pi\hbar} \int \sum_l e^{i\mathrm{Q}\mathrm{r}_l} \boxed{\left\langle S_0^\alpha(0) S_l^\beta(t) \right\rangle} e^{-i\omega t} dt$$

spin-spin correlation function

Is there an equivalent expression for RIXS?

RIXS theory

RIXS cross section

Haverkort, PRL 2010

$$\frac{d^2\sigma}{d\Omega\, dE} \propto \chi''(\mathbf{Q},\omega)$$

$$\chi(\omega,\mathbf{Q}) = -\imath \int_0^\infty e^{\imath\omega t}\langle i|R^{\varepsilon_i\varepsilon_o}_{\omega_i,\mathbf{Q}}(t)^\dagger R^{\varepsilon_i\varepsilon_o}_{\omega_i,\mathbf{Q}}(t=0)|i\rangle dt$$

ε_i ε_o = incident, outgoing photon polarization

$$R_j^{\varepsilon_i\varepsilon_o} = \sigma^{(0)}\varepsilon_i\cdot\varepsilon_o^* + \frac{\sigma^{(1)}}{s}\varepsilon_i\times\varepsilon_o^*\cdot S + \frac{\sigma^{(2)}}{s(2s-1)}\left(\varepsilon_i\cdot S\,\varepsilon_o^*\cdot S + \varepsilon_o^*\cdot S\,\varepsilon_i\cdot S - \frac{2}{3}\varepsilon_i\cdot\varepsilon_o^* S^2\right)$$

X-ray absorption

X-ray magnetic circular dichroism

X-ray magnetic linear dichroism

vanishes for S = ½

caution for larger S e.g. iron pnictides!

RIXS theory

RIXS from cuprates

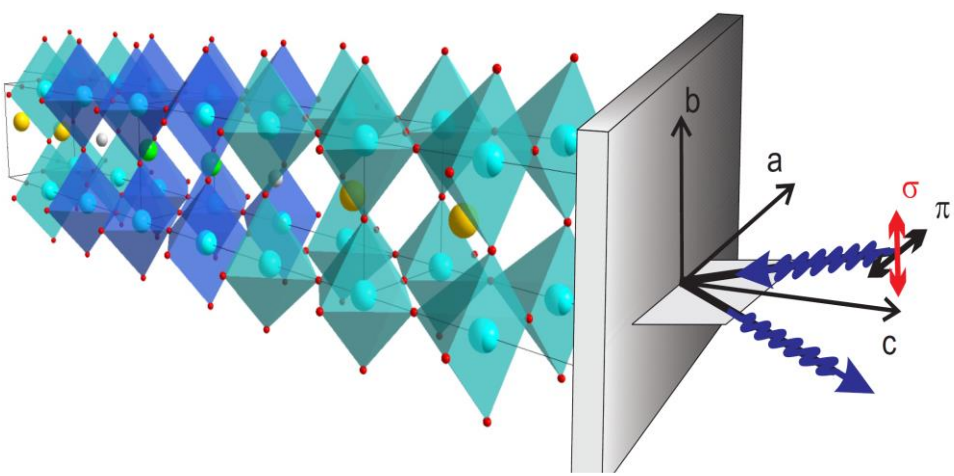
$$R^{\varepsilon_i\varepsilon_o}_{\omega_i,\mathbf{Q}} = \boxed{\sigma^{(0)}\varepsilon_i \cdot \varepsilon_o^*} + \boxed{\frac{\sigma^{(1)}}{s}\varepsilon_o^* \times \varepsilon_i \cdot S_{\mathbf{Q}}}$$

charge spin

$$\chi(\omega,\mathbf{Q}) = -\imath\int_0^\infty e^{\imath\omega t}\langle i|(\varepsilon_o^* \times \varepsilon_i \cdot S_{\mathbf{Q}}(t))^\dagger(\varepsilon_o^* \times \varepsilon_i \cdot S_{\mathbf{Q}}(t=0))|i\rangle$$

RIXS can detect single-magnon excitations in crossed polarization

similar to neutrons, but with different polarization factor



dependence on RIXS intensity on

incoming photon polarization allows

separation of spin and charge excitations

polarization **analysis**

planned for ERIXS

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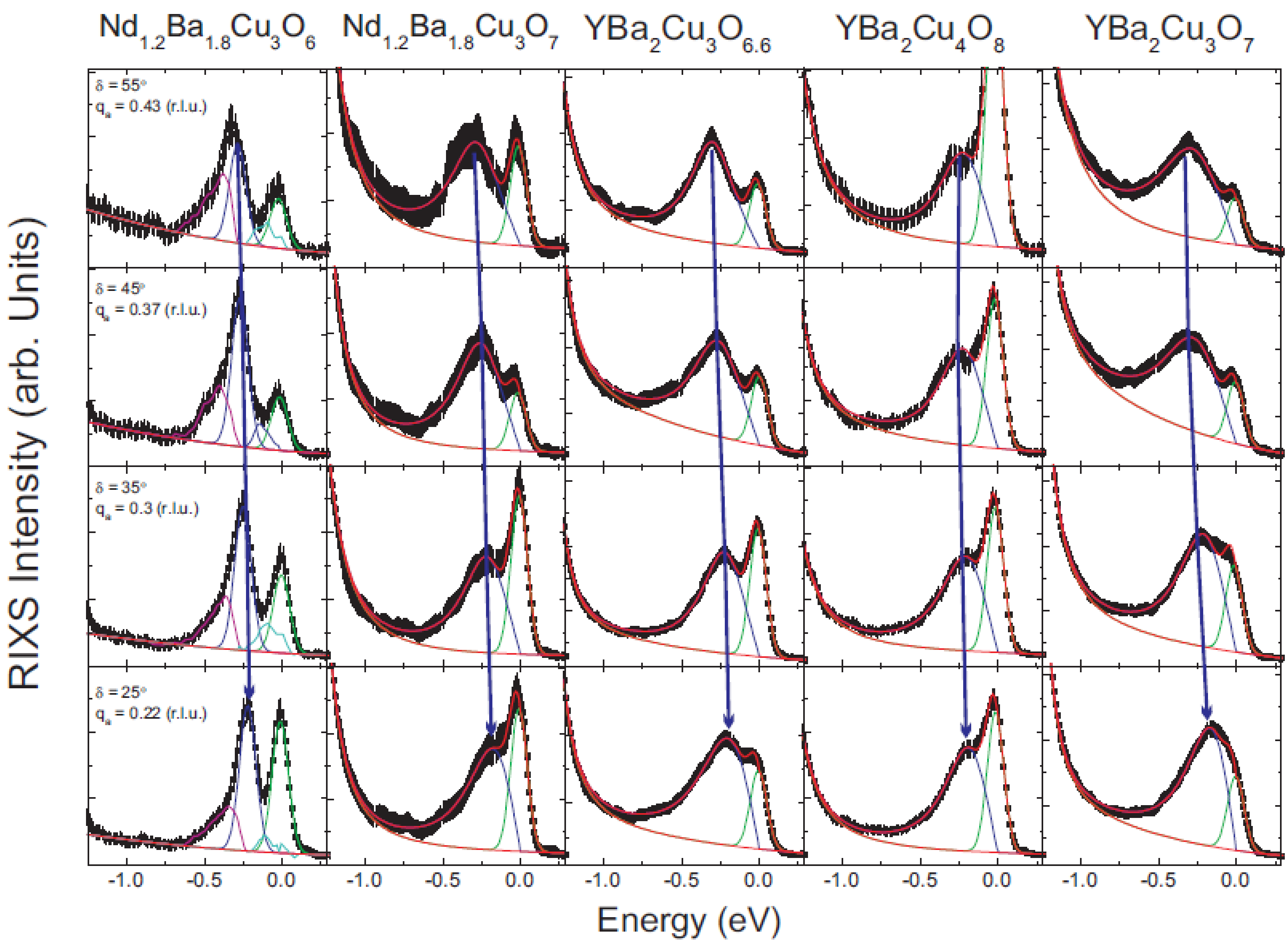
RIXS from doped cuprates



Le Tacon et al.
Nature Phys. 2011

well defined "paramagnon" excitations at all doping levels

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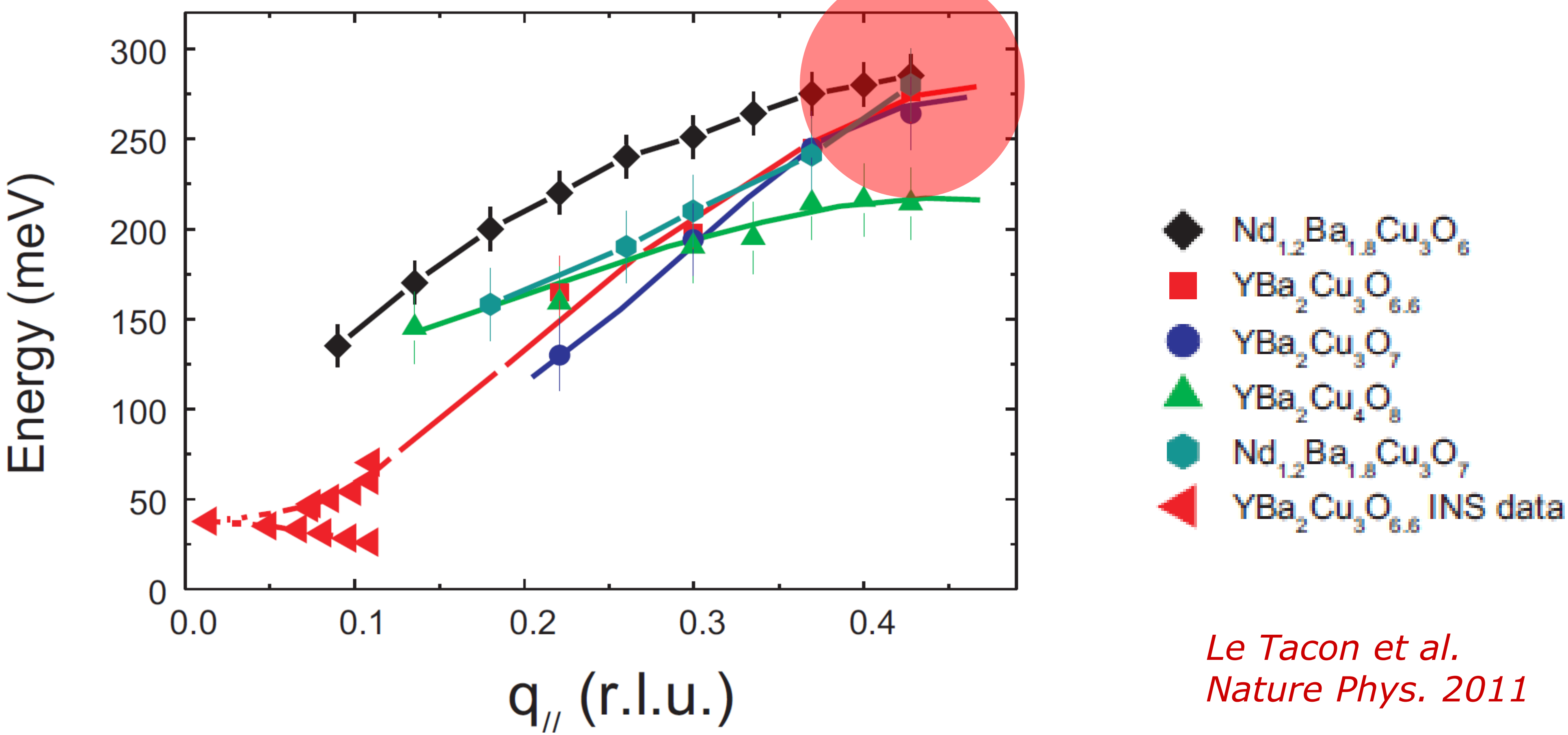
Spin excitations in doped cuprates



Le Tacon et al.
Nature Phys. 2011

- magnon-like quasiparticle excitations observed at all doping levels
- energy-integrated spectral weight conserved upon doping
- **nearest-neighbor spin correlations almost independent of doping**

Highly overdoped $Tl_2Ba_2CuO_{6+x}$

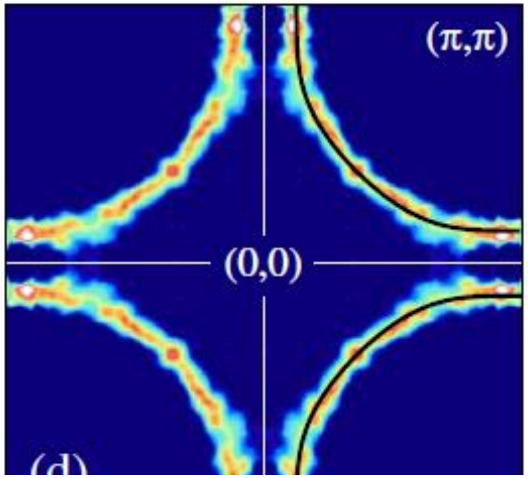
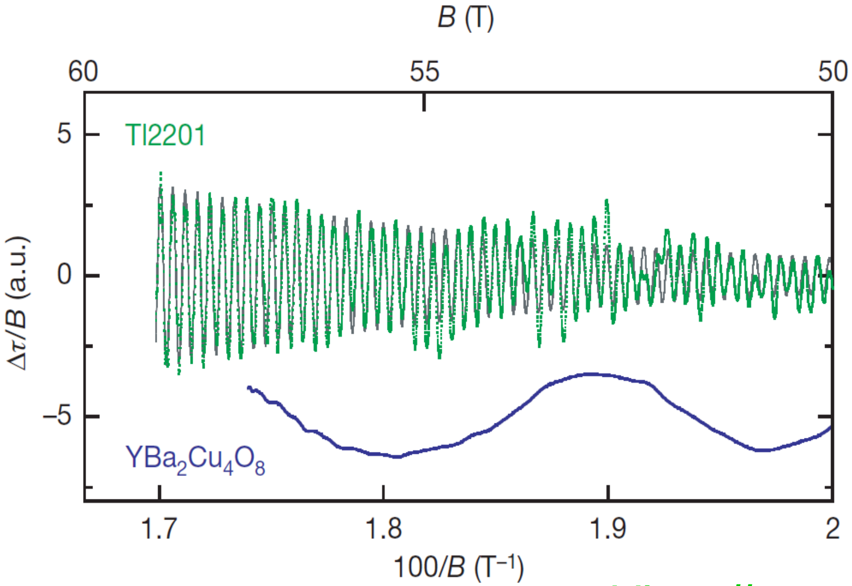


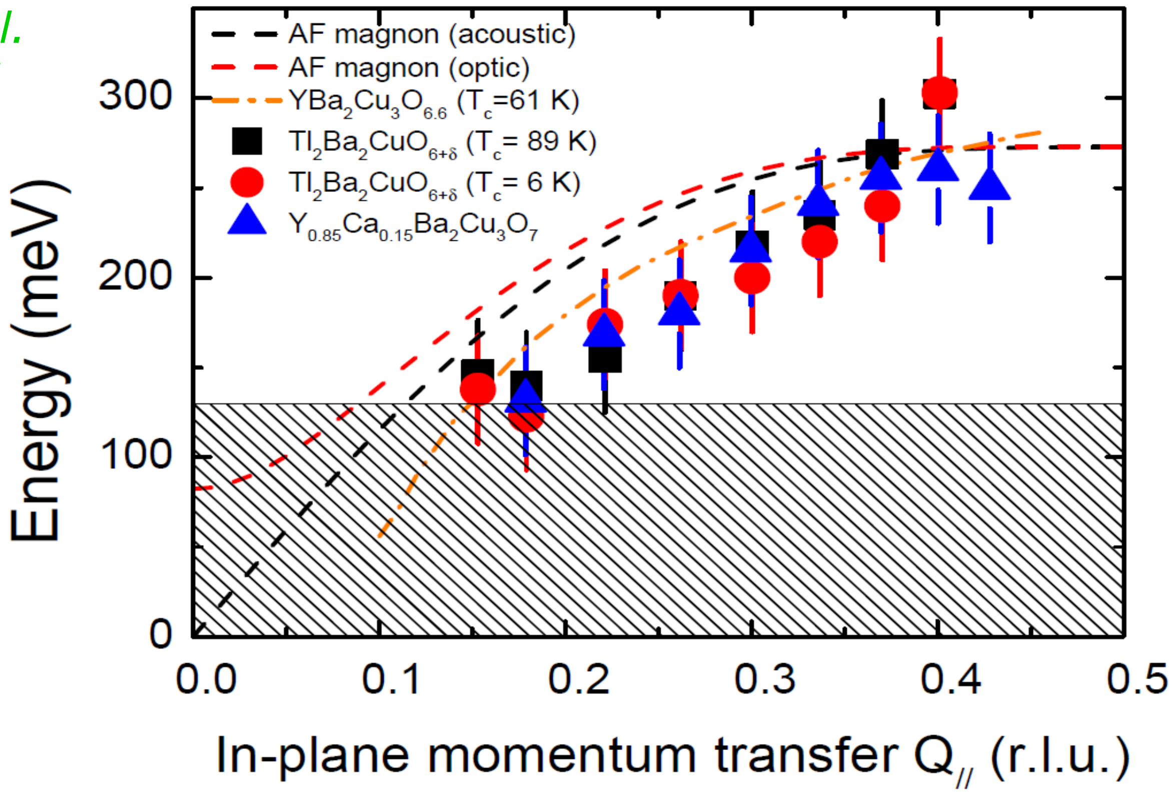
Plate et al.
PRL 2005



Vignolle et al.
Nature 2008

p ~ 0.27, T_c = 6 K

well defined fermionic quasiparticles



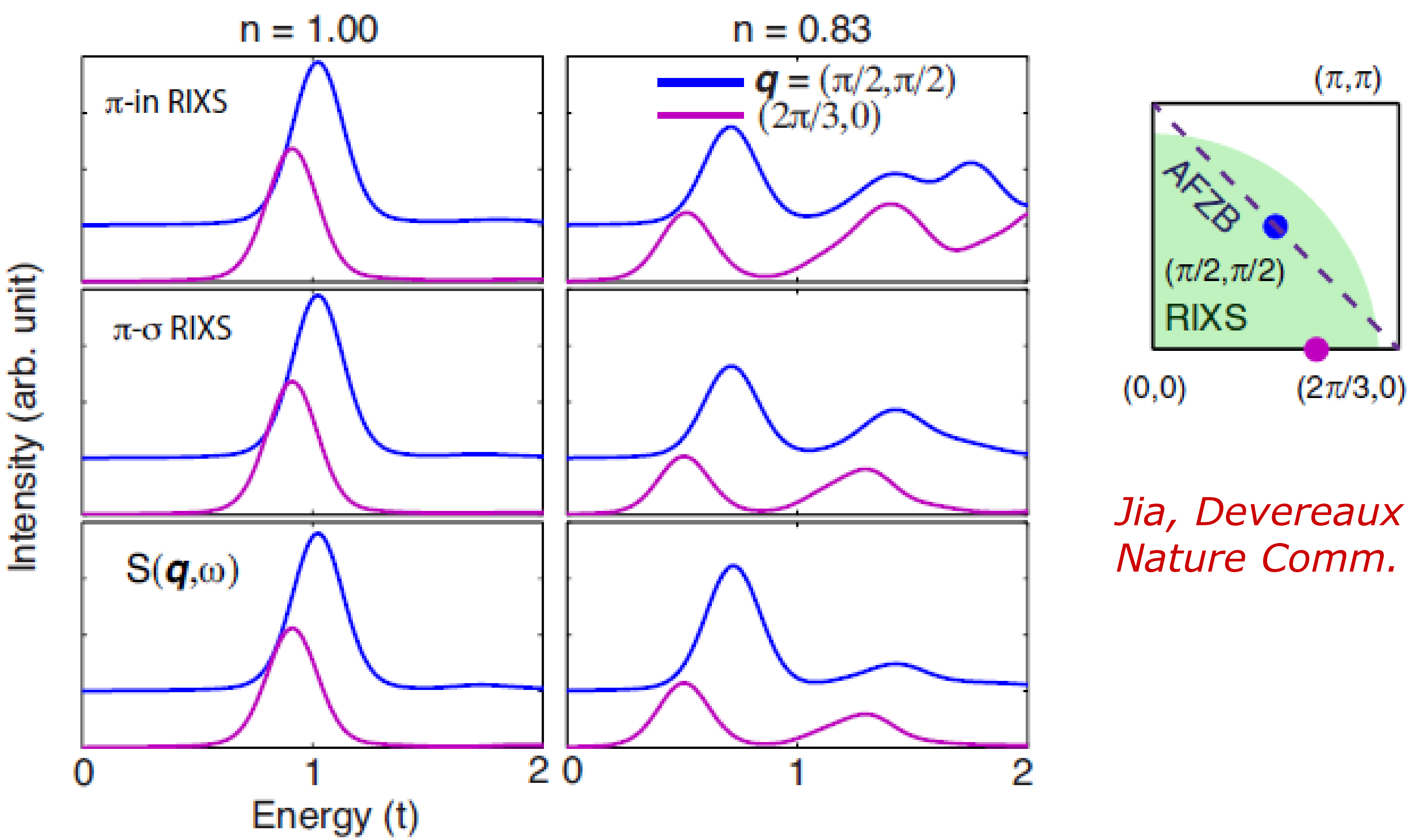
Le Tacon et al.
PRB 2013

well defined paramagnon excitations
→ short-range AF correlations

challenge for theory

RIXS theory

Quantum Monte Carlo calculations of 2D Hubbard model

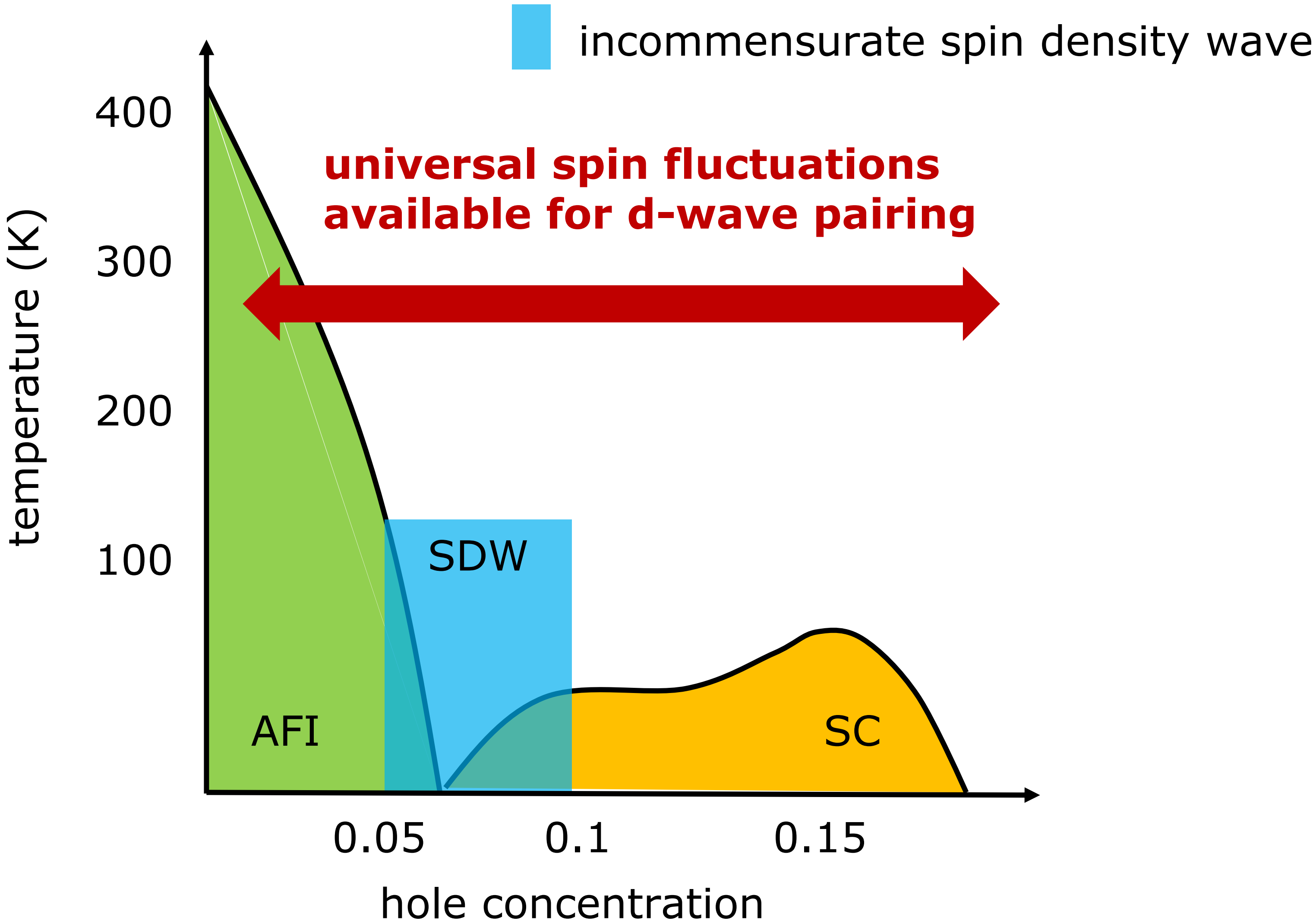


Jia, Devereaux et al.
Nature Comm. 2014

- RIXS cross section proportional to S(Q,ω) even in doped cuprates
- persistence of paramagnon excitations reproduced

possible controversy: *Benjamin, Demler et al., PRL 2014*

Competing order in $YBa_2Cu_3O_{6+x}$

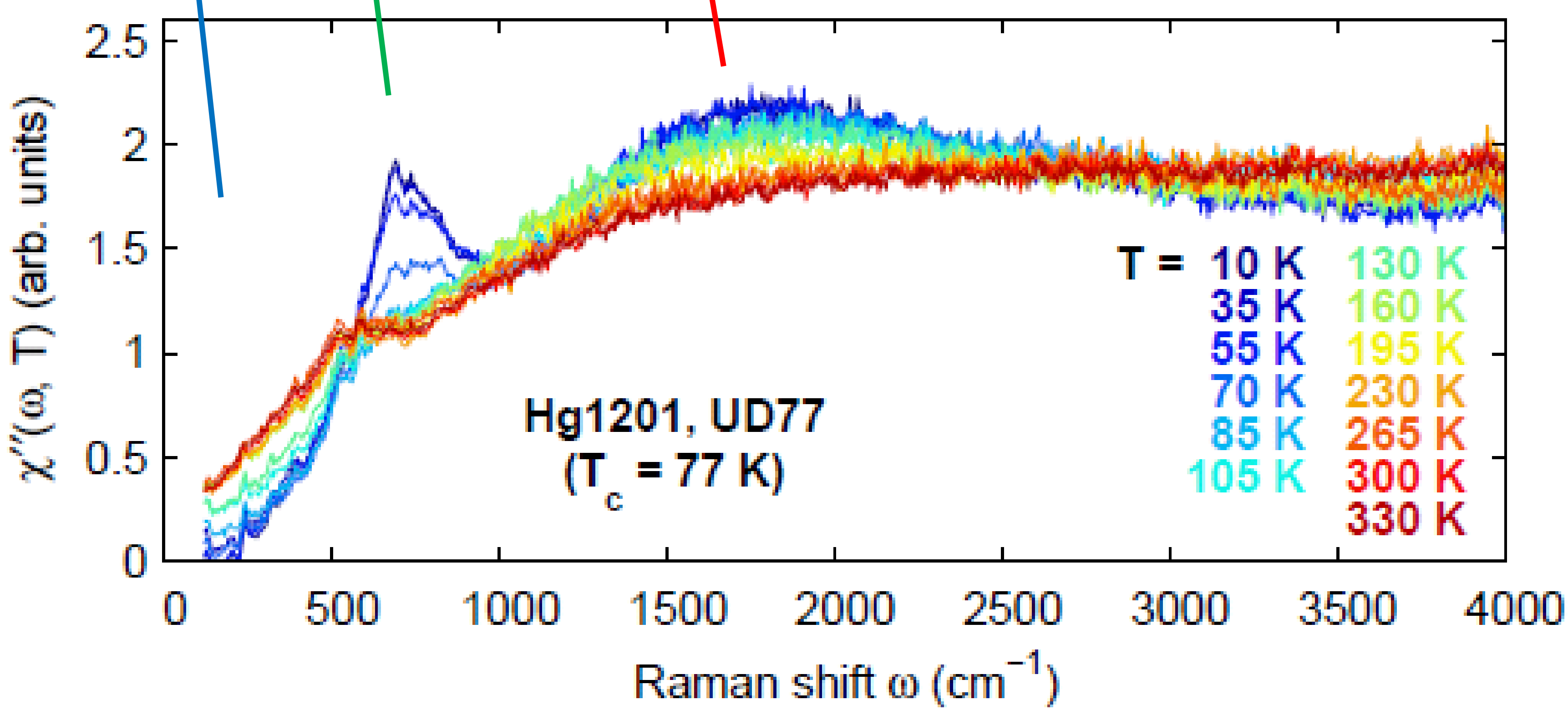


Optical Raman scattering

pseudogap

pair-breaking peak

two-magnon peak
dominated by high-energy magnons



Li et al.
PRL 2012

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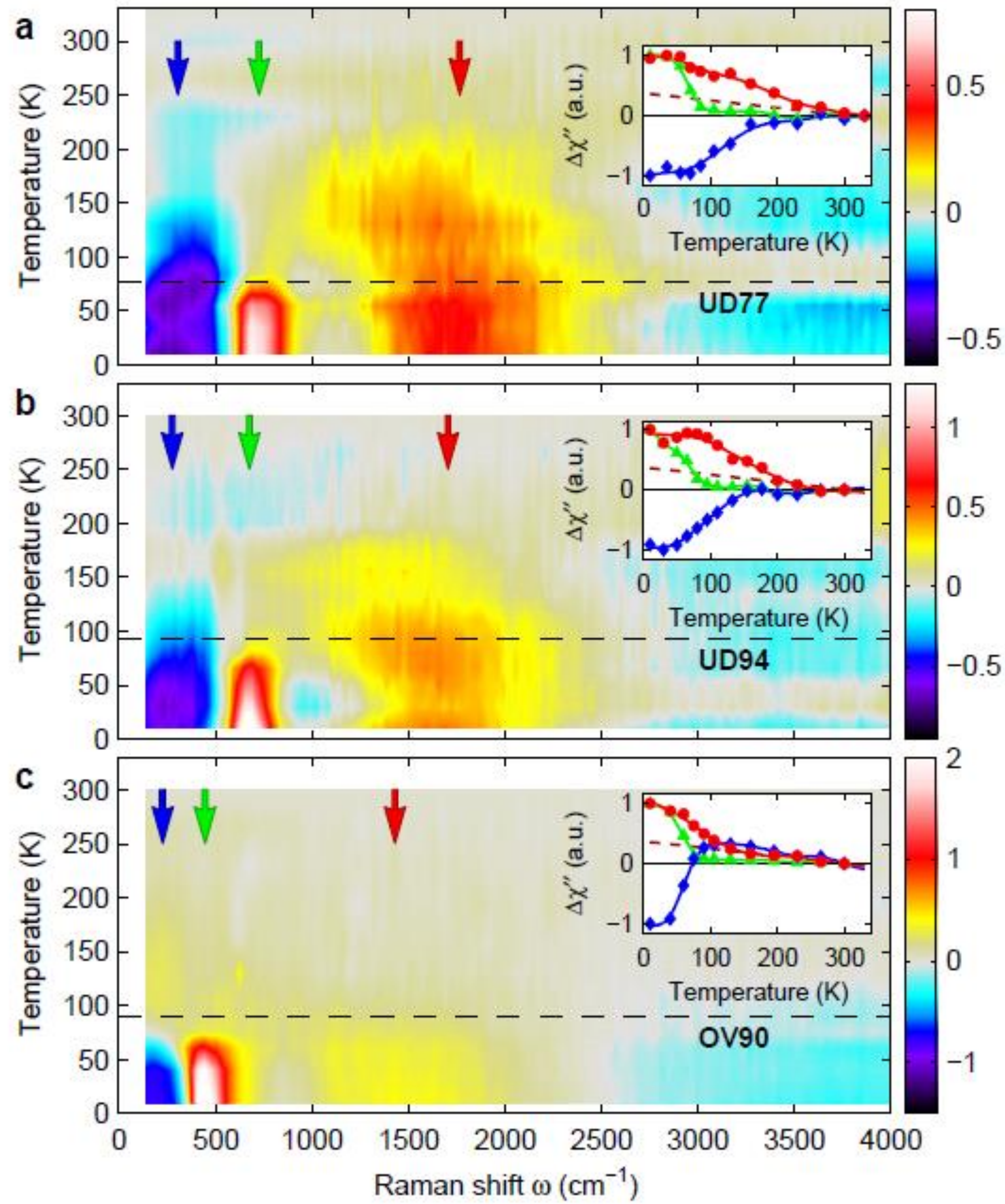
Optical Raman scattering



two-magnon peak shows

shift & intensity enhancement

- at T_c at optimal doping
- above T_c in pseudogap regime

feedback effect

of pairing interaction

on high-energy magnons

analogous to low-energy

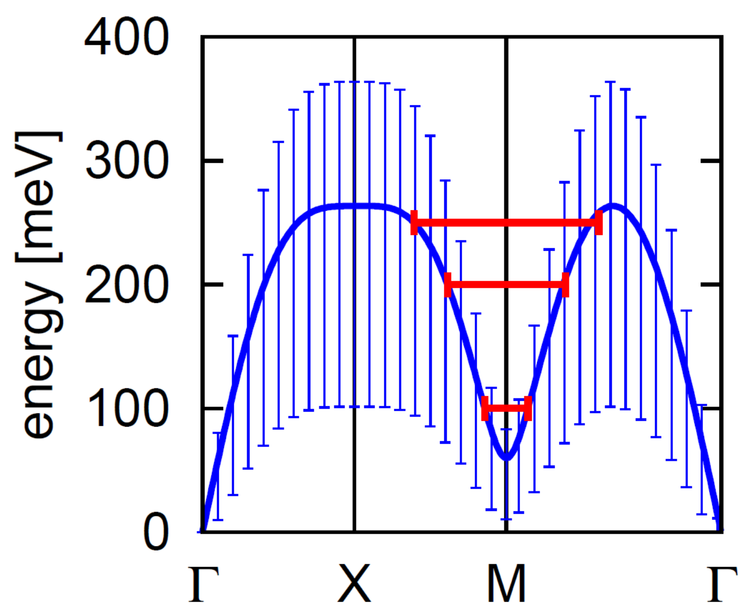
magnetic resonant mode

Li et al.
PRL 2012

Eliashberg theory

complete bosonic spectral function of $YBa_2Cu_3O_7$ (T_c = 90 K)

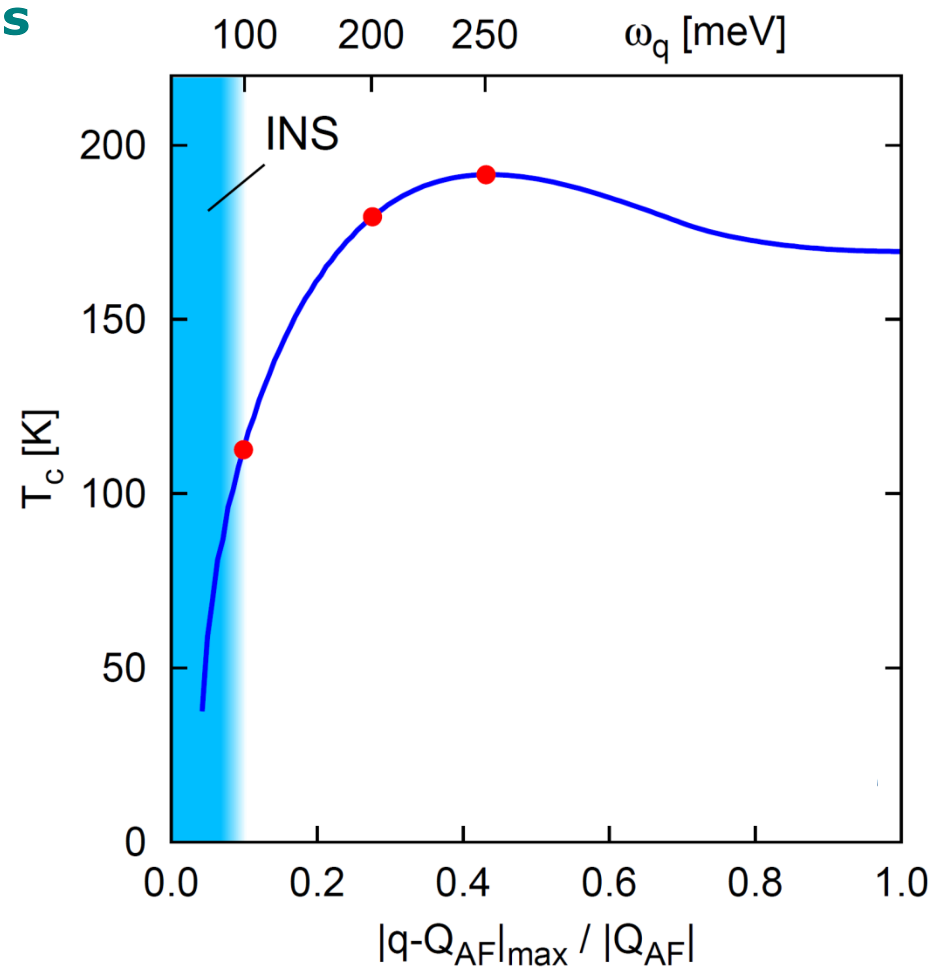
gapped, non-critical spin fluctuations



Le Tacon et al., Nature Phys. 2011

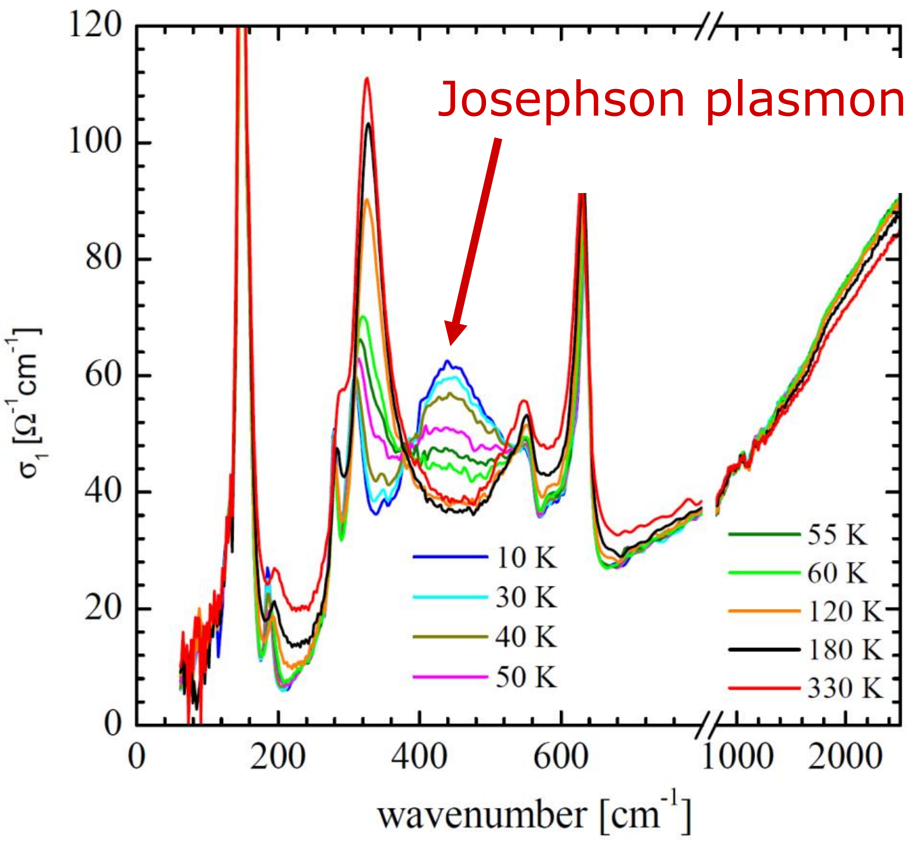
simple mean-field model gives reasonable value for T_c

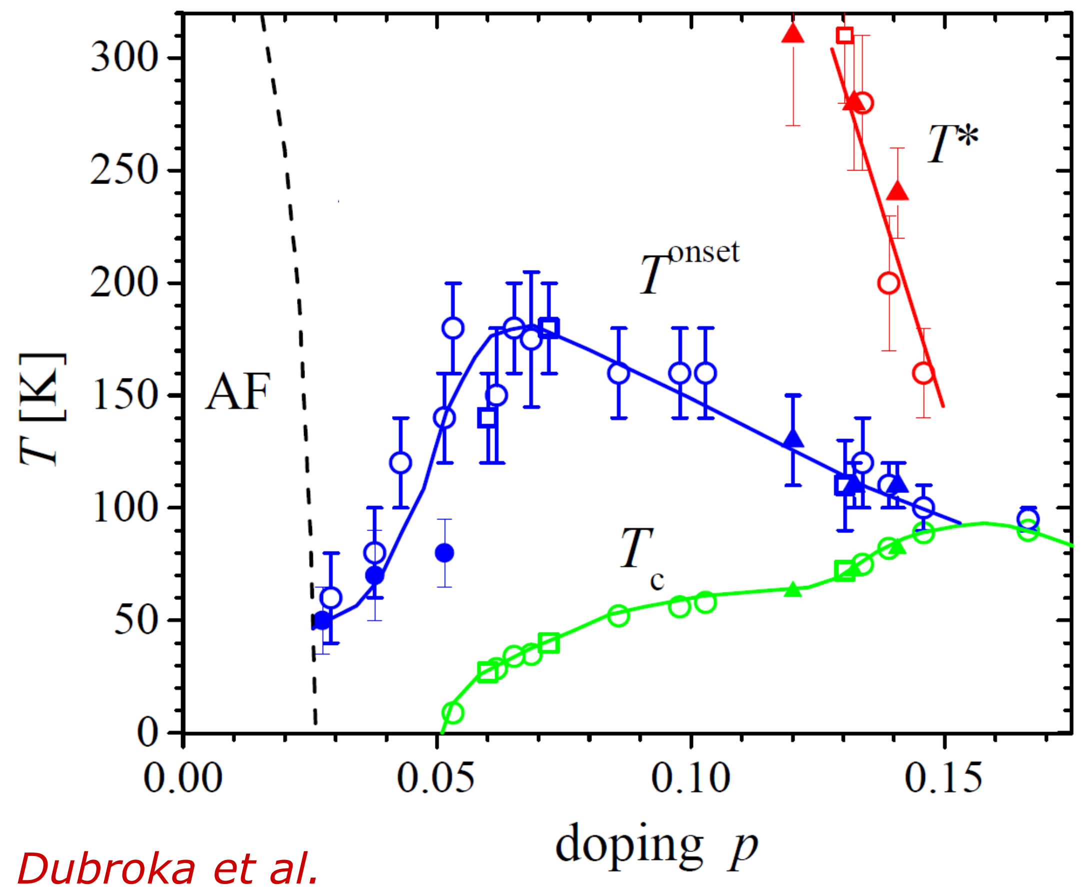
solution of gap equation within t-J model with different momentum cutoffs



Superconducting fluctuations in underdoped YBCO

c-axis optical conductivity

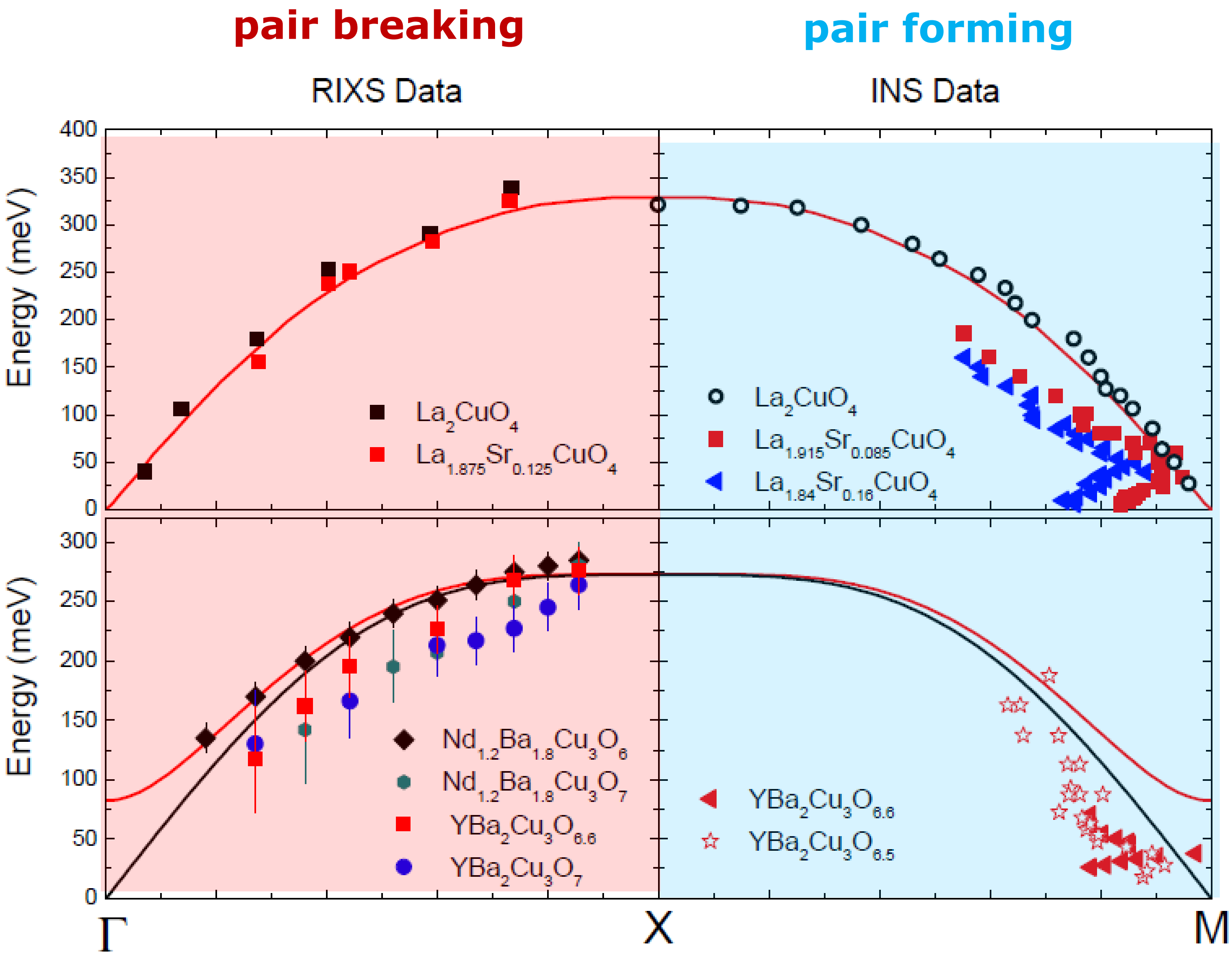




Dubroka et al.
PRL 2011

superconducting fluctuations observed up to at least 180 K

Eliashberg theory



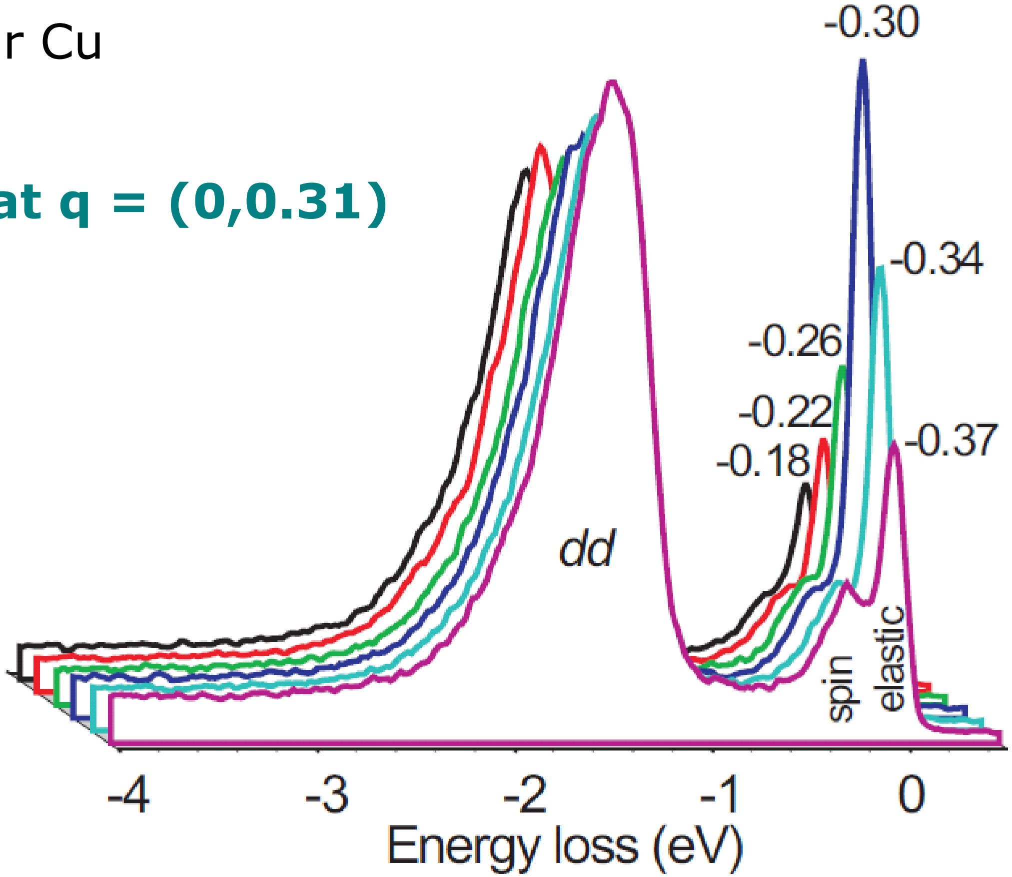
overdoped LSCO
excitations at
q ~ (π,π) disappear
→ reduction of T_c

Wakimoto et al.
PRL 2007

Charge density wave

resonant inelastic x-ray scattering on underdoped $YBCO_{6.6}$

photon energy tuned to L-edge of planar Cu



resonant elastic scattering (REXS) at q = (0,0.31)

polarization dependence indicates charge correlations

Ghiringhelli et al., Science 2012
Achkar et al., PRL 2012
Blanco-Canosa et al., PRL 2013

consistent with NMR

Wu et al., Nature 2011; Nature Com. 2013

confirmed with hard x-rays

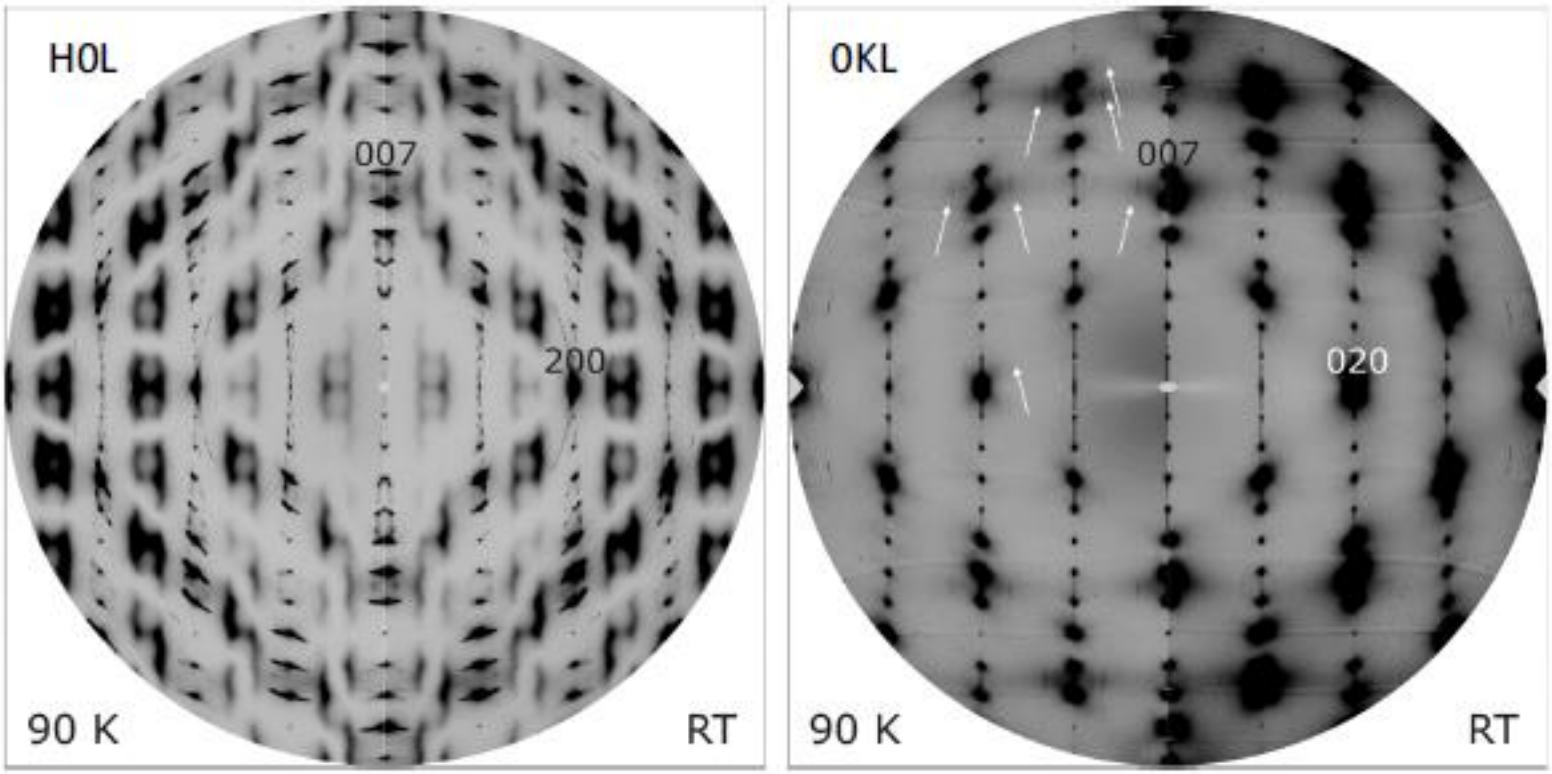
Chang et al., Nature Phys. 2012; Blackburn et al., PRL 2013

Charge density wave

hard x-ray diffraction pattern

short-range superstructure from oxygen dopant order

superposed on signatures of electronic charge order



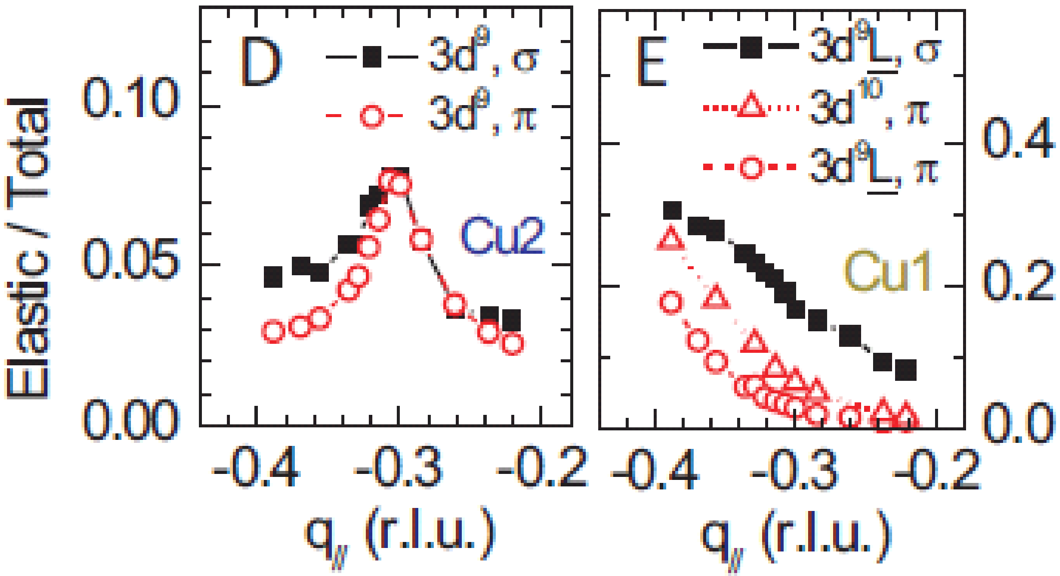
Le Tacon et al.
Nature Phys. 2014

Site-selective charge correlations

REXS signal

not present at resonance of chain Cu

→ only planar Cu atoms involved

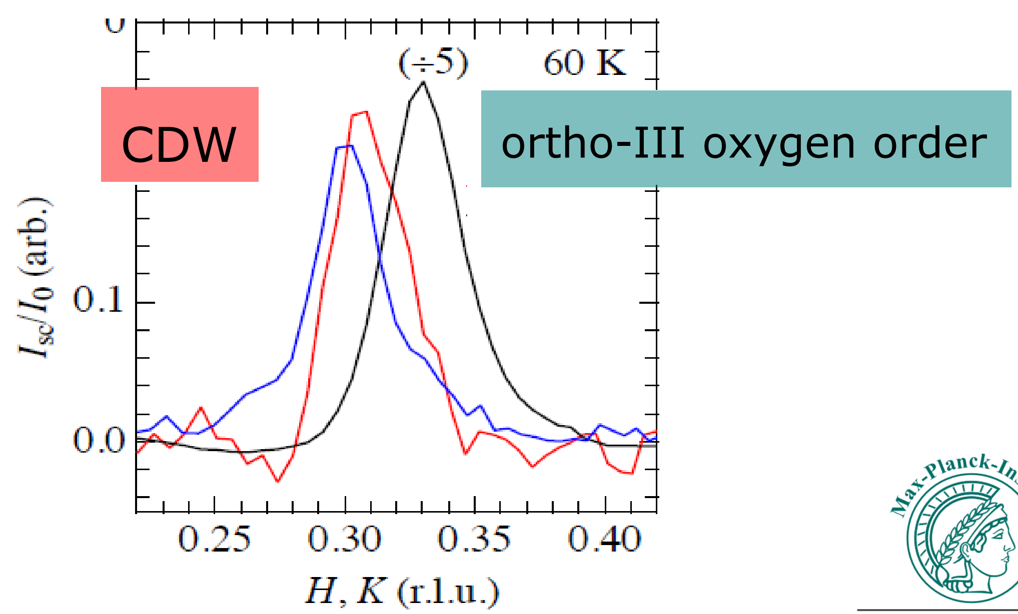


comparison of planar REXS signal and NREXS from chains

→ REXS is incommensurate

$q = (\xi\ 0\ L)$ with $\xi = 0.31 \neq \frac{1}{3}$

Achkar et al., PRL 2012



CDW temperature dependence

$YBa_2Cu_3O_{6.6}$

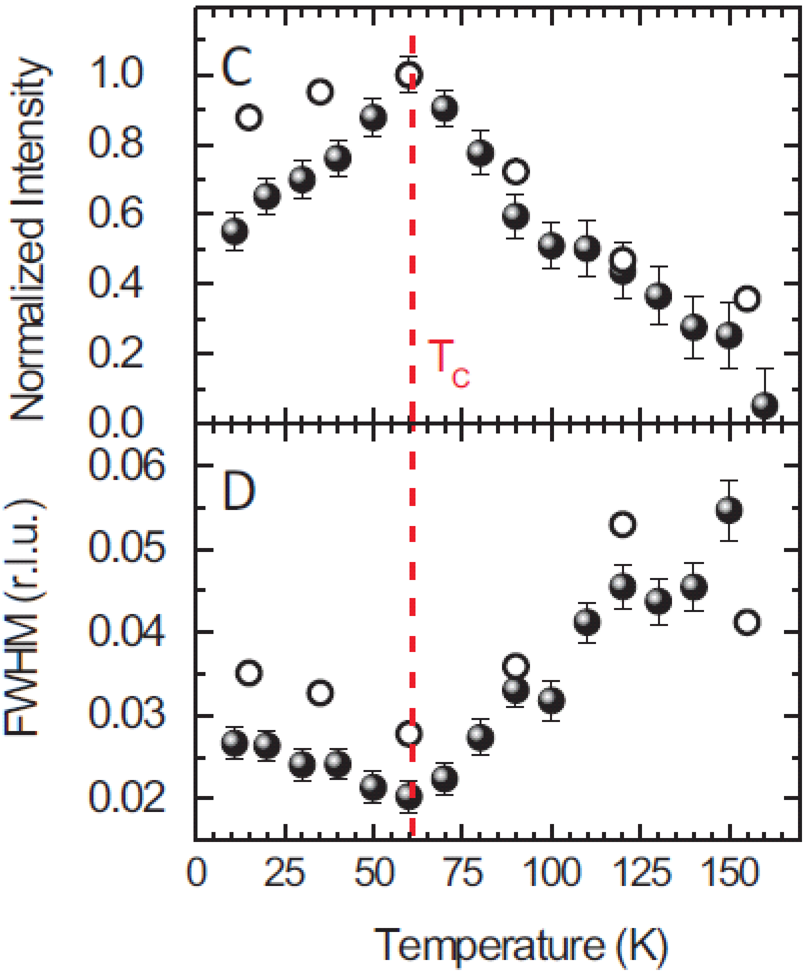
maximum correlation length 16 a

→ **CDW nearly critical**

correlation length suppressed below T_c

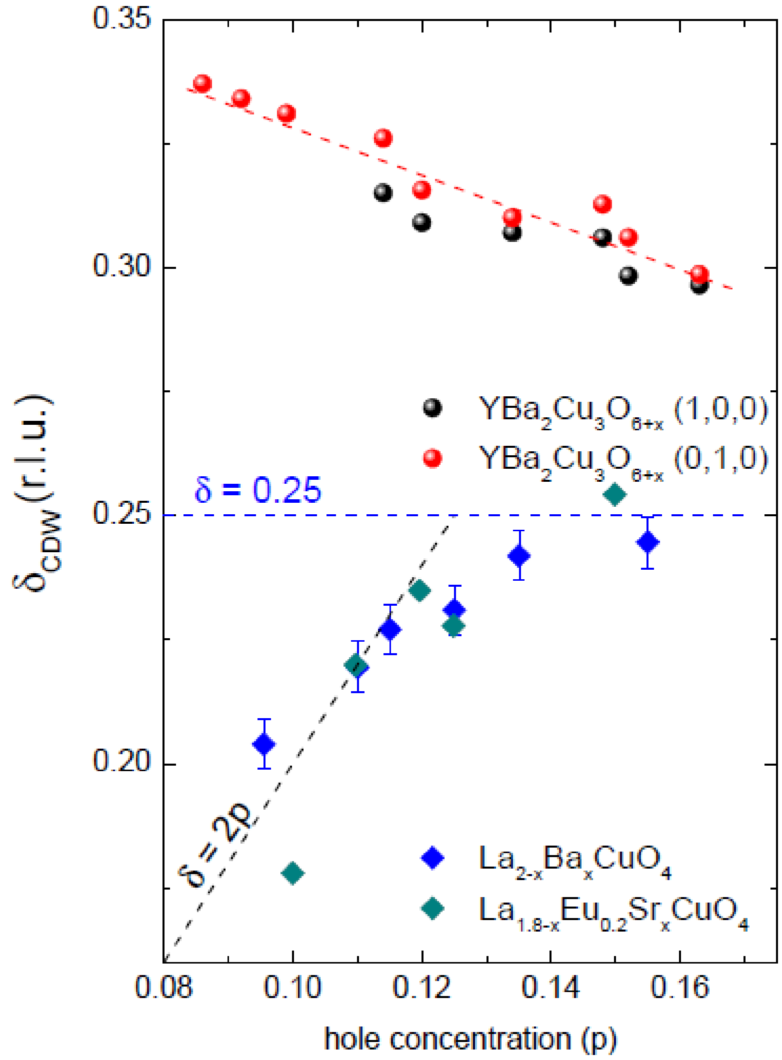
→ **CDW competes with superconductivity**

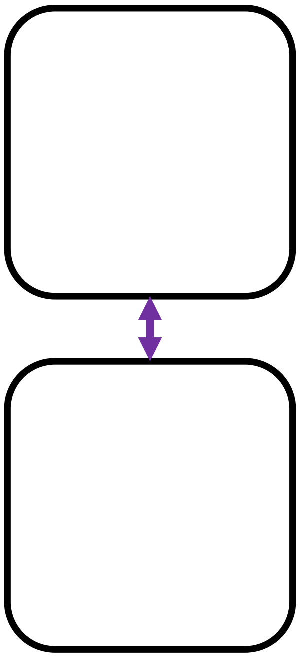
CDW fluctuations reduce T_c



Ghiringhelli et al., Science 2012
Achkar et al., PRL 2012
Blanco-Canosa et al., PRL 2013; arXiv 2014

Doping dependence of CDW wavevector





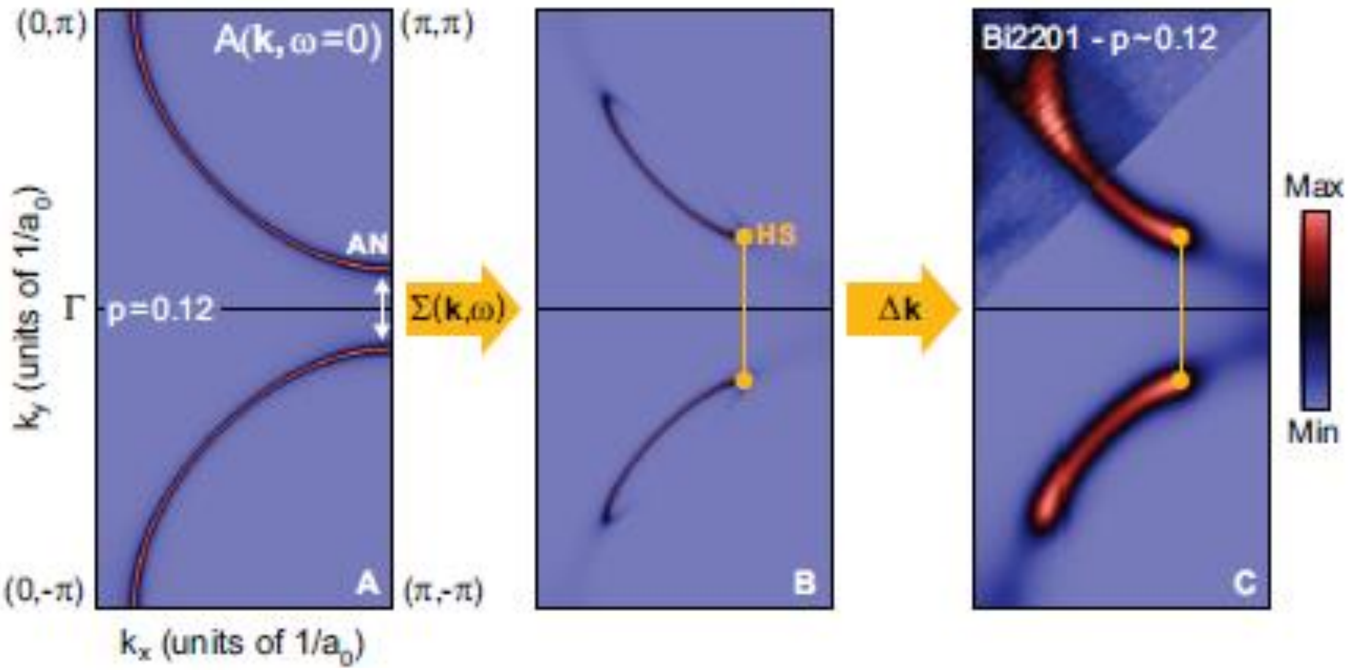
Blanco-Canosa et al. arXiv 2014

- CDW wave vector qualitatively consistent with size of Fermi surface
- different from "stripes" in $La_{2-x}(Sr,Ba)_xCuO_4$

CDW and Fermi arcs

pseudogap at antinodes

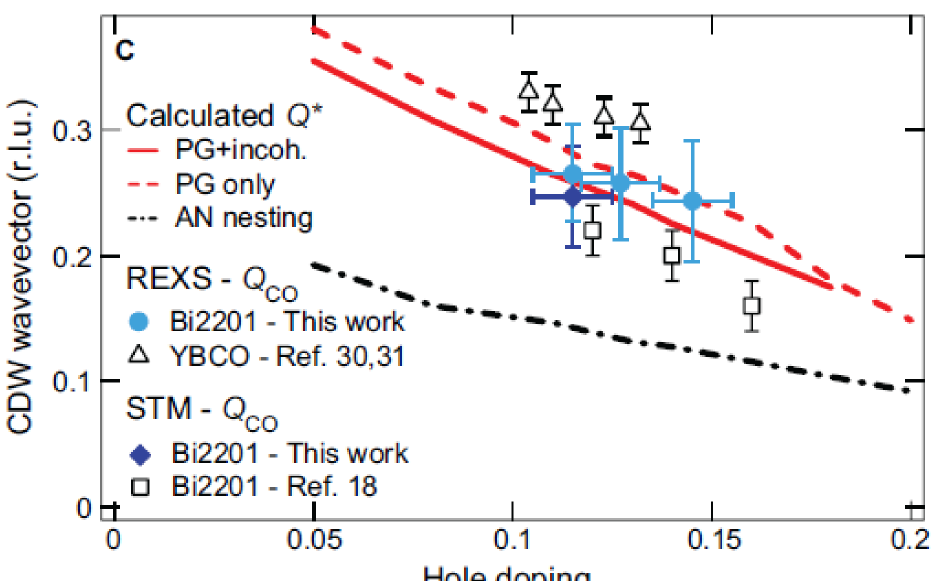
→ **"Fermi arcs"**



distance between Fermi arc tips

matches CDW wave vector

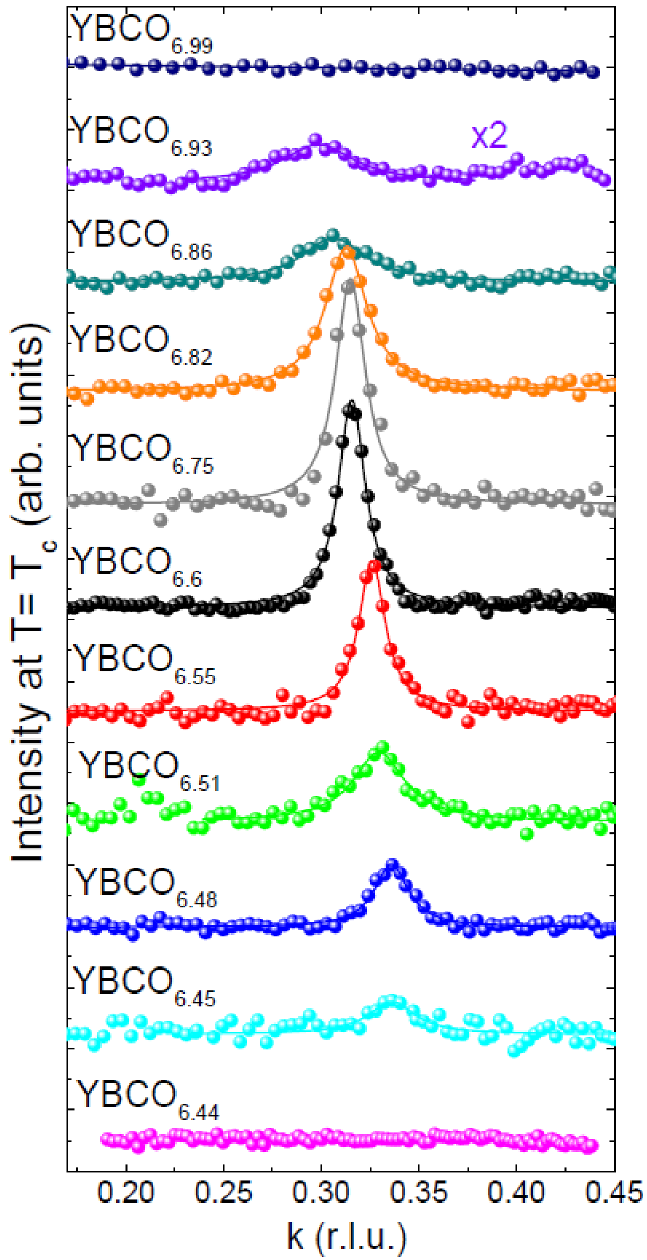
Comin et al., Science 2014



Doping dependence of CDW amplitude

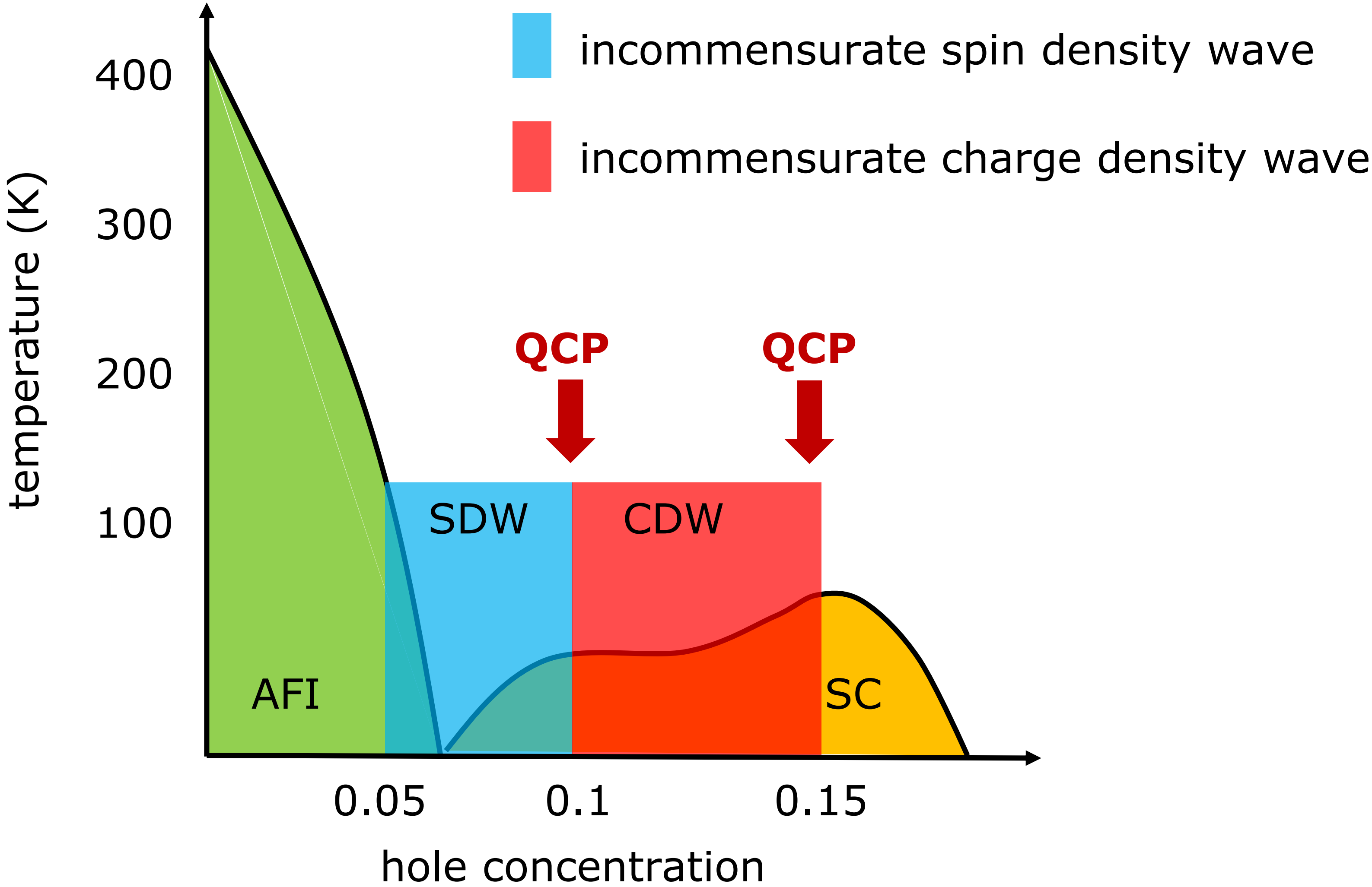
- CDW amplitude maximum at p ~ 0.12
- CDW vanishes for p ~ 0.08, 0.16

→ **two quantum critical points?**

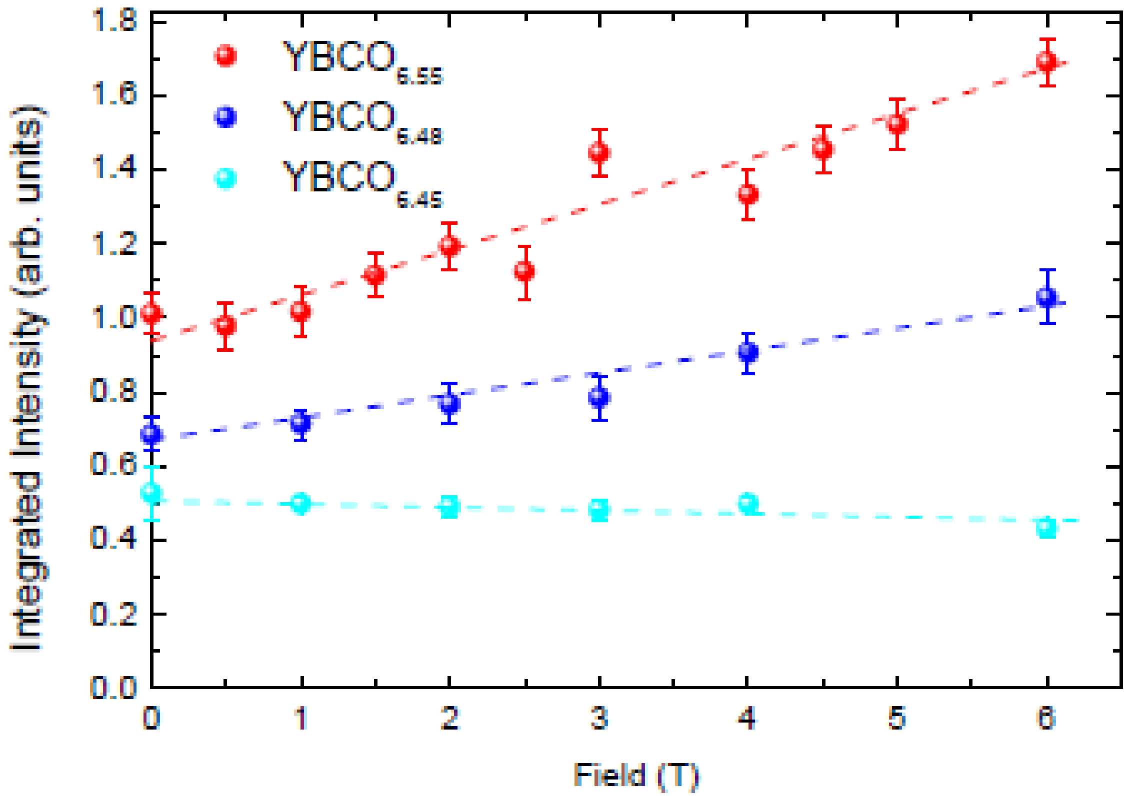


Blanco-Canosa et al.
arXiv 2014

Competing order in $YBa_2Cu_3O_{6+x}$



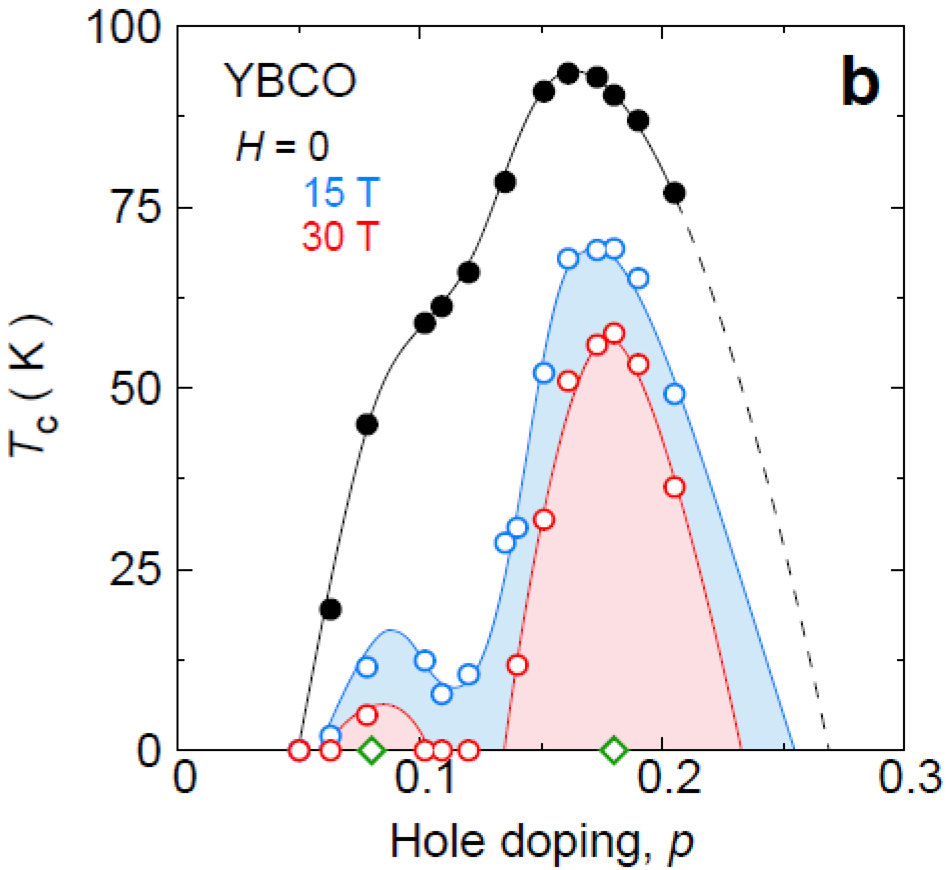
Magnetic field dependence



Blanco-Canosa et al.
PRL 2013, arXiv 2014

magnetic field weakens superconductivity
enhances CDW correlations

Magnetic field dependence



Grisonanche et al.
Nature Comm. 2014

transport experiments in high magnetic fields

superconducting dome splits into two domes

centers coincide with CDW quantum critical points

Are quantum-critical CDW fluctuations important for high-T_c ?

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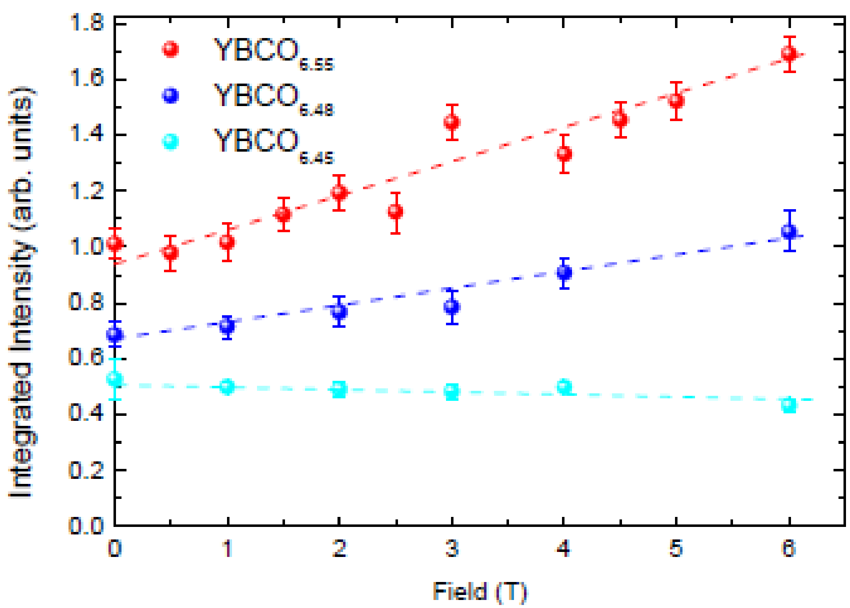
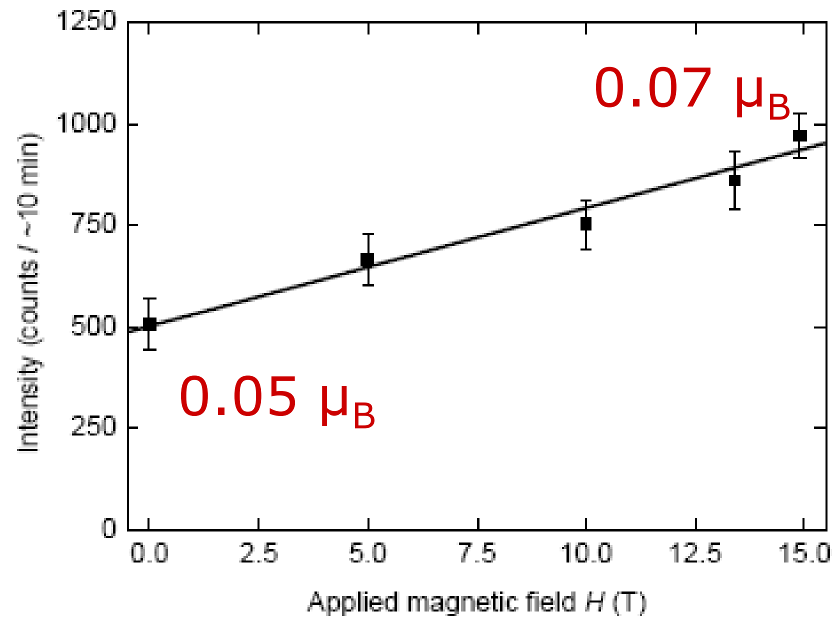
Magnetic field dependence

H-dependence of CDW



Blanco-Canosa et al. PRL 2013
see also Backburn et al., PRL 2013

H-dependence of SDW for YBCO$_{6.45}$



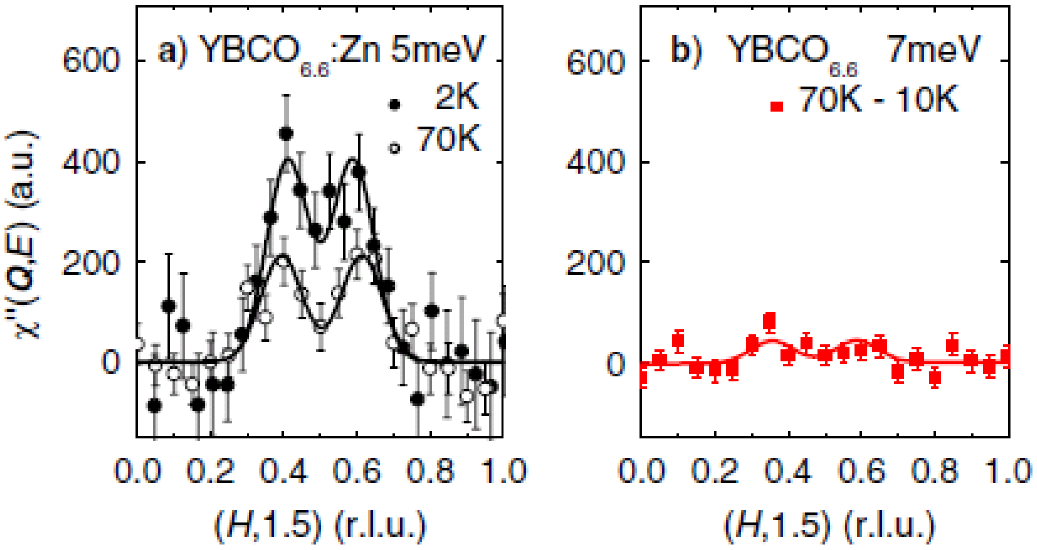
Haug et al. PRL 2009

three-phase competition between SDW, CDW, dSC for p ~ 0.08

Spinless impurities

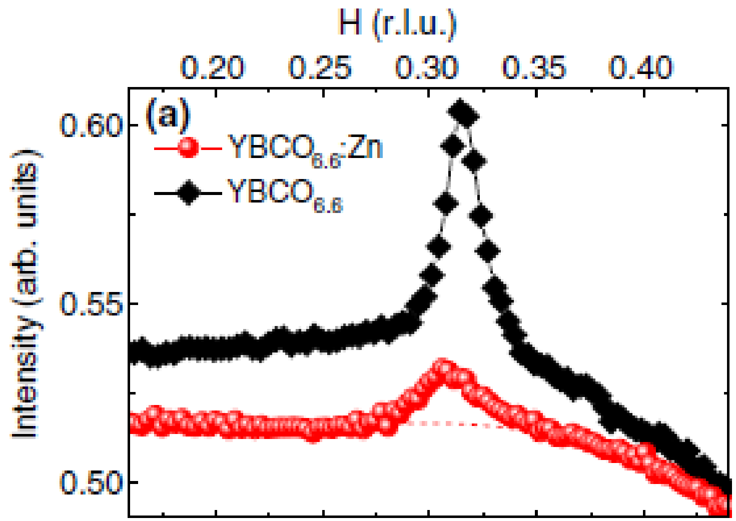
substitution of 2% Zn^{2+} (S=0) for planar Cu^{2+} (S= ½) in $YBCO_{6.6}$

enhances IC-SDW in $YBCO_{6.6}$



Suchaneck et al., PRL 010

degrades IC-CDW



Blanco-Canosa et al., PRL 2013

three-phase competition between SDW, CDW, superconductivity

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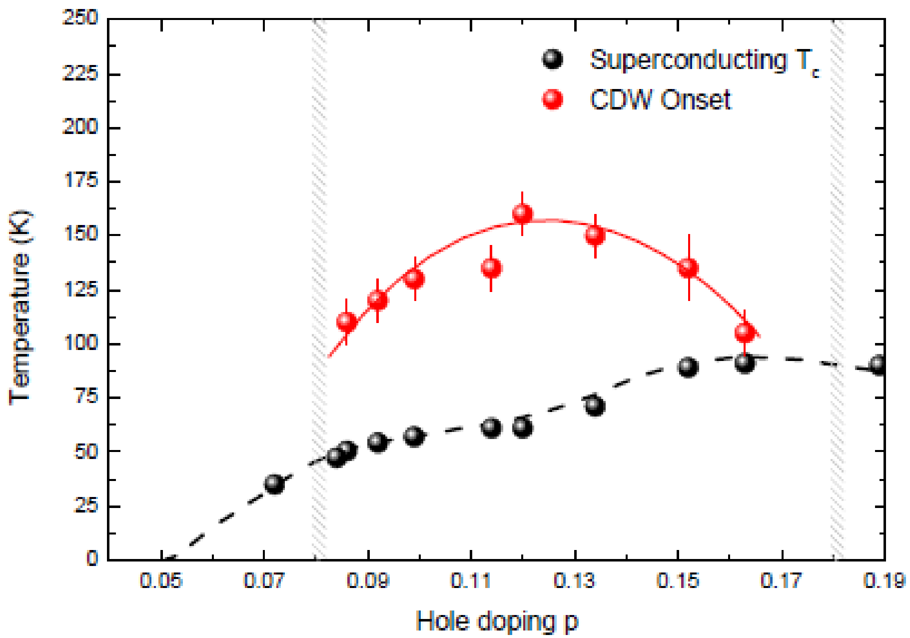
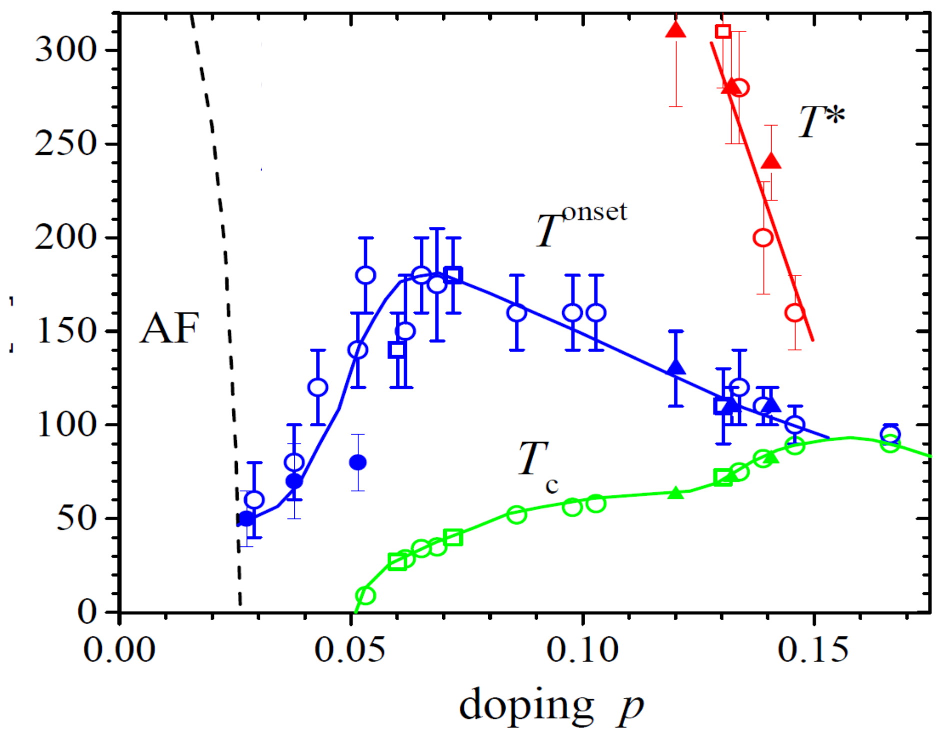
Doping dependence of CDW onset temperature

onset of CDW



Blanco-Canosa et al., arXiv 2014

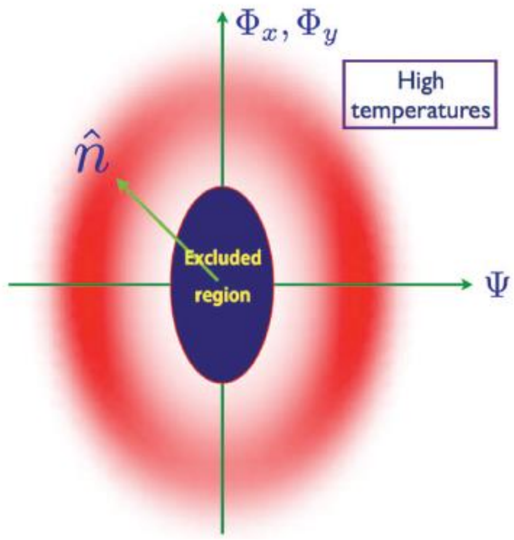
onset of superconducting fluctuations

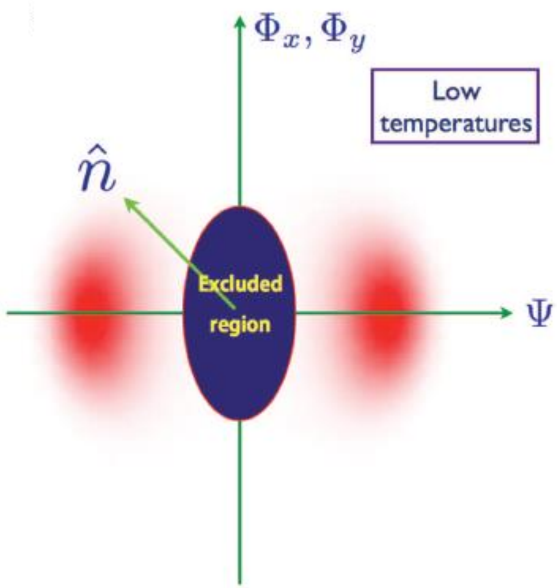


Dubroka et al., PRL 2011

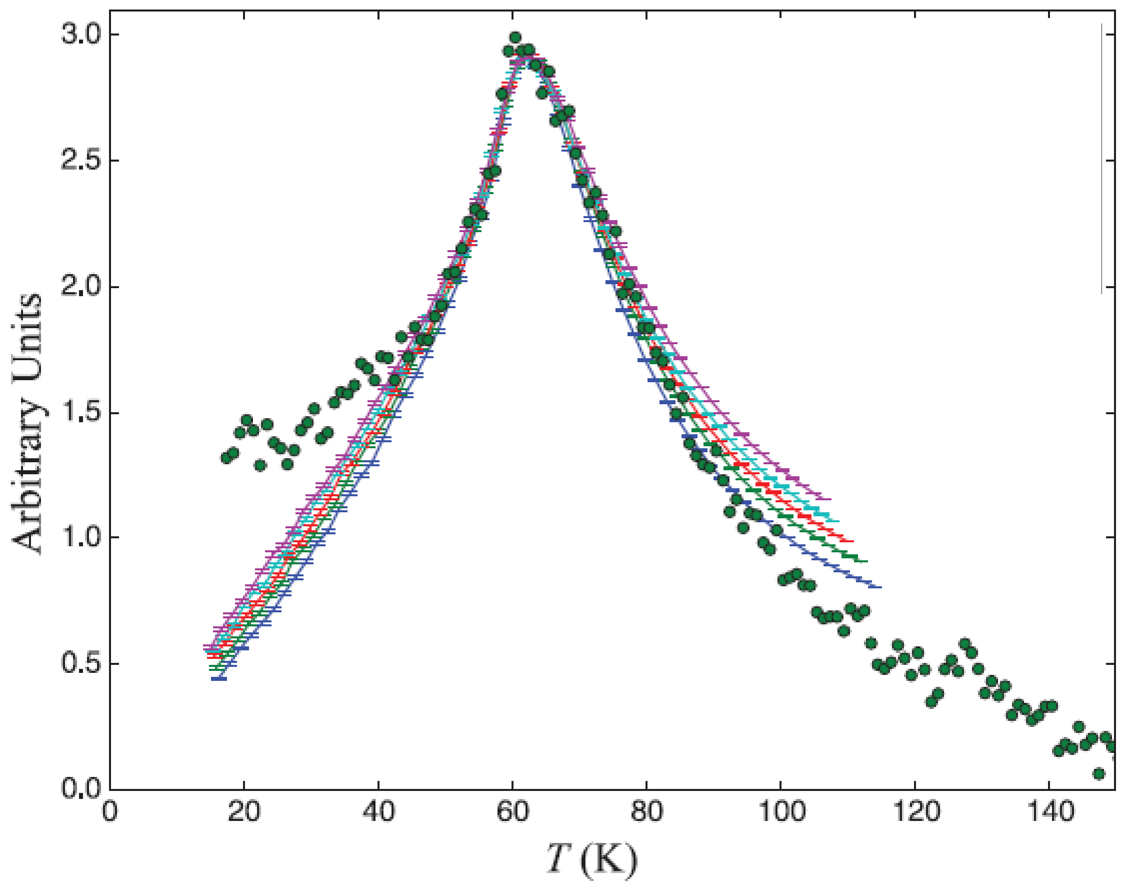
combined CDW & superconducting fluctuations in pseudogap regime

Combined CDW-dSC fluctuations



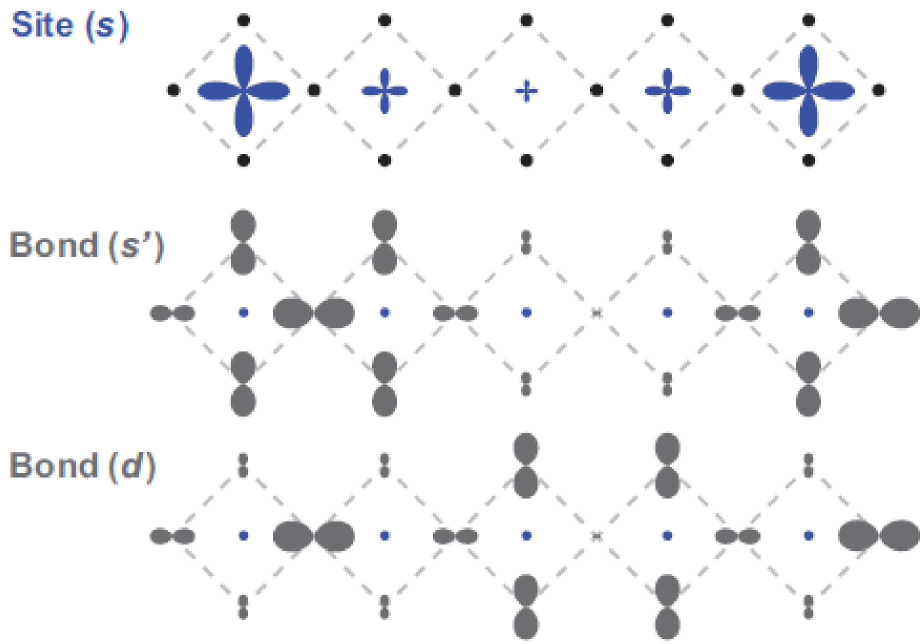


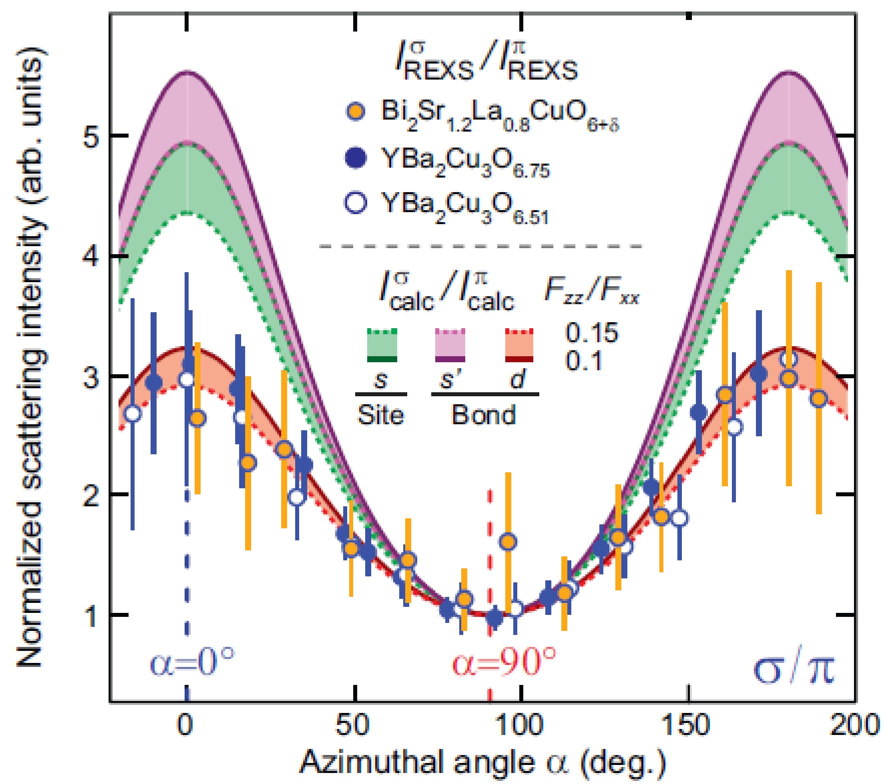
fit of CDW intensity versus temperature in multi-component order parameter model



Hayward et al., Science 2014

CDW displacement pattern





polarization dependence of resonant scattering intensity
indicates bond CDW with d-wave displacement pattern

Comin et al., arXiv 2014

für Festkörperforschung

Doping dependence of CDW anisotropy

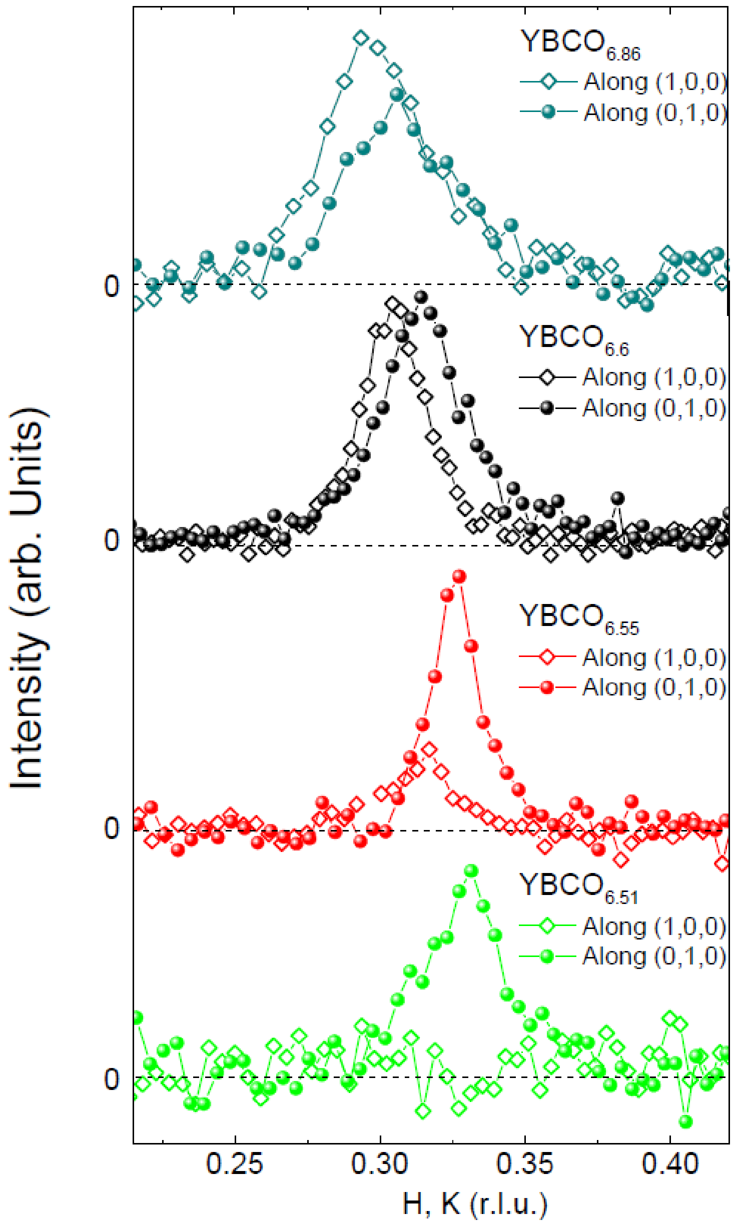
- p < 0.12: strong uniaxial anisotropy
- p ~ 0.12: nearly isotropic
- p > 0.12: anisotropy switches sign

consistent with uniaxial modulation

more comprehensive analysis

by Comin, Damascelli *et al.*

Blanco-Canosa et al., arXiv 2014



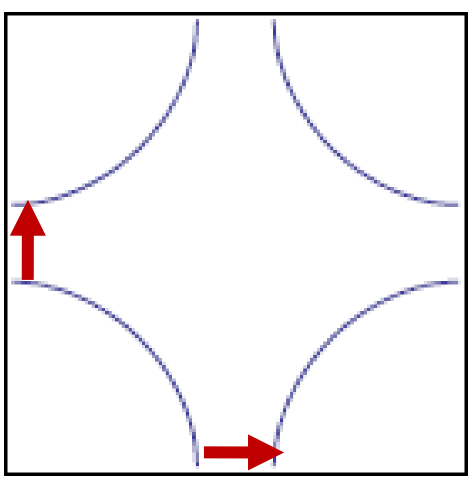
Electron-phonon interaction

ARPES anomalies at antinodes

interpreted as evidence of

electron-magnon and/or

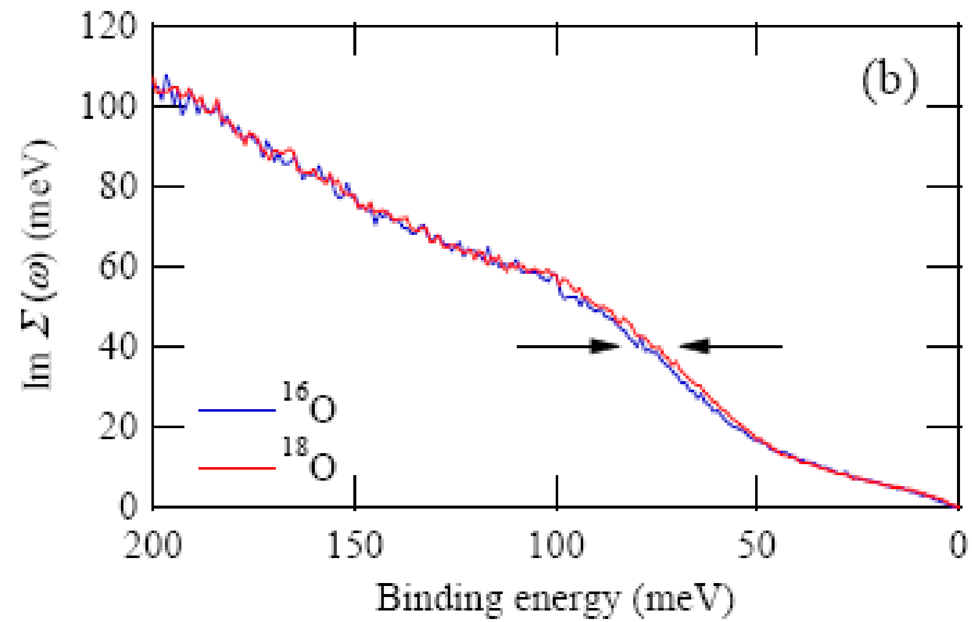
electron-phonon interaction



Cuk et al., PRL 2004

isotope effect

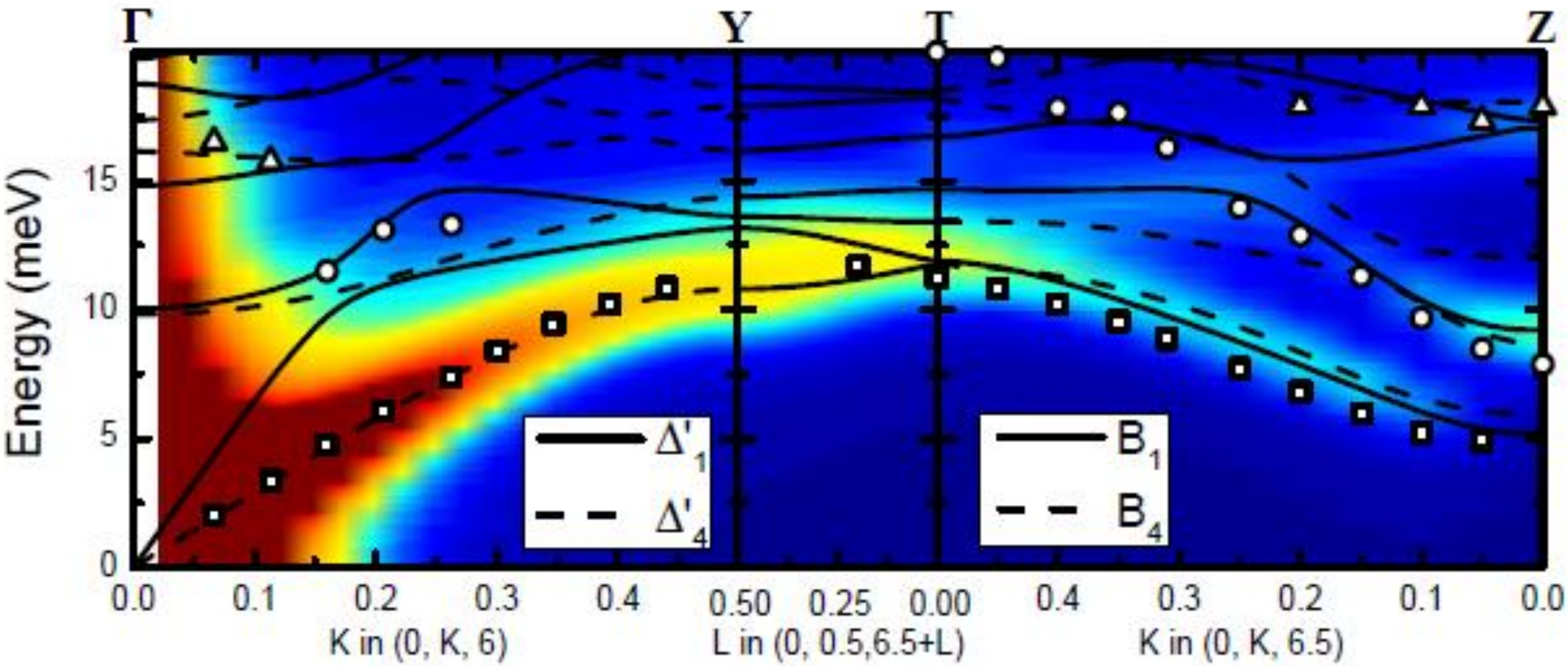
indicates phonon contribution



Iwasawa et al., PRL 2008

CDW dynamics?

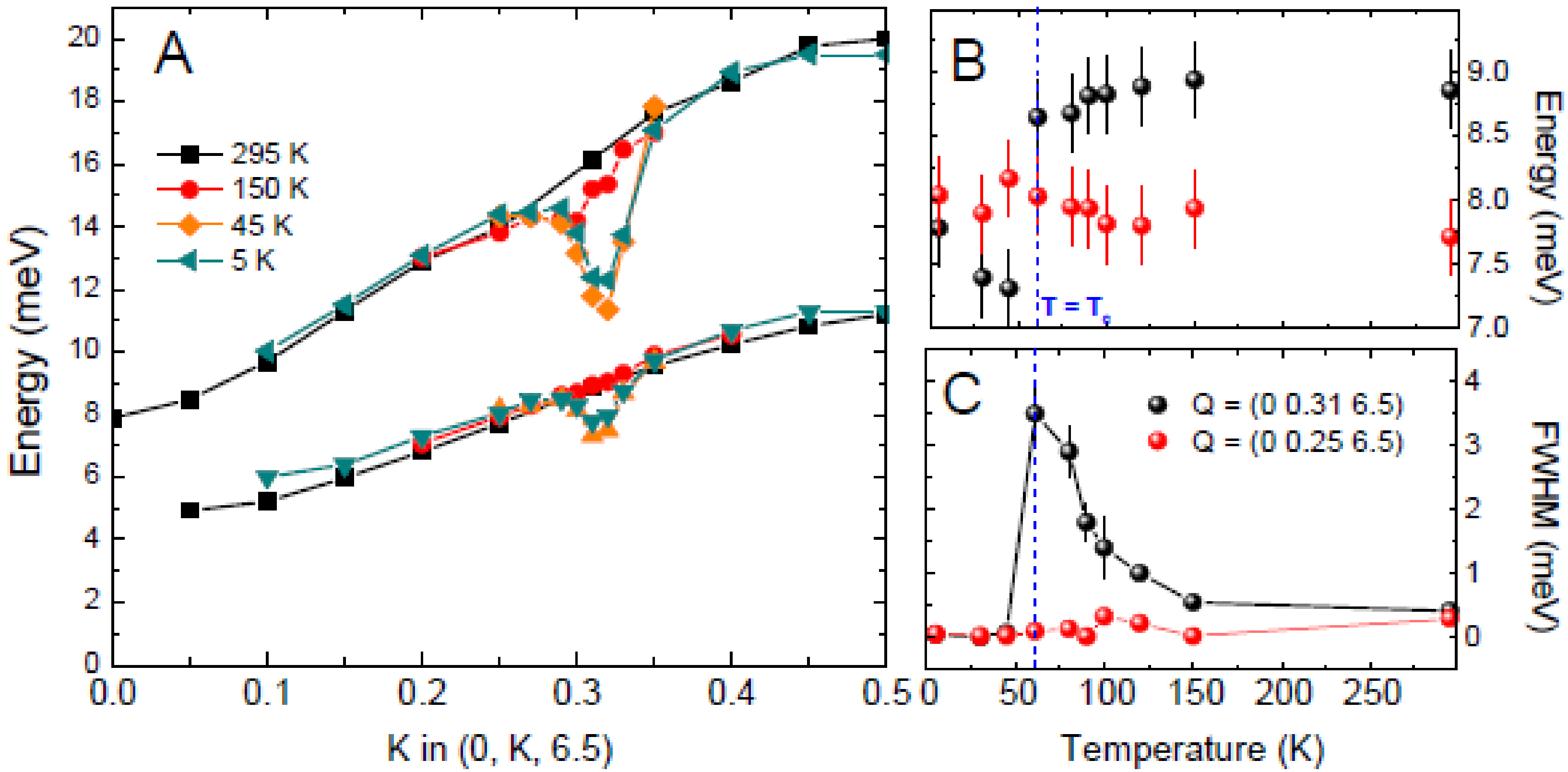
acoustic phonons at room temperature



Le Tacon et al., Nature Phys. 2014

acoustic phonon dispersions & intensities well described by LDA

CDW-induced phonon anomalies



T < T$_c$ giant phonon softening

T > T$_c$ giant phonon linewidth

in narrow range around CDW wavevector

Le Tacon et al.
Nature Phys. 2014

highly anisotropic electron-phonon interaction

favors CDW formation, not superconductivity

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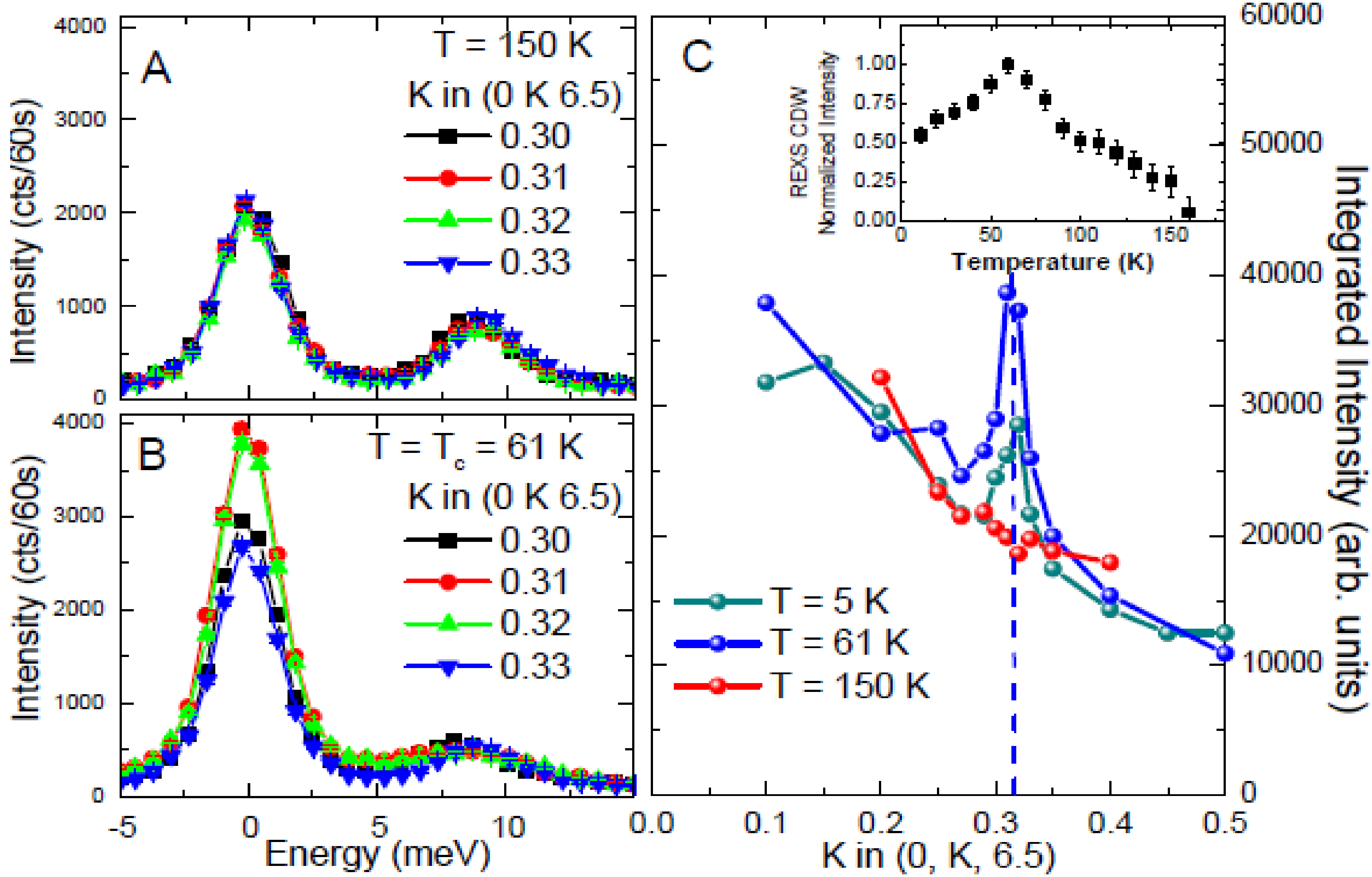
CDW-induced central peak in $YBCO_{6.6}$



elastic central peak (ΔE < 100 μeV)

impurity-nucleated CDW nanodomains below 150 K

Le Tacon et al. Nature Phys. 2014

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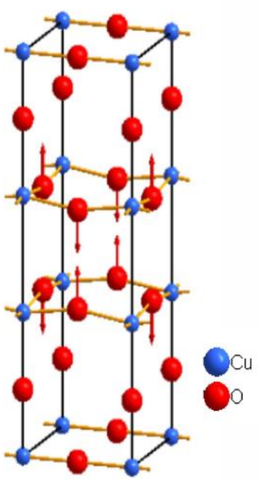
Optical phonons

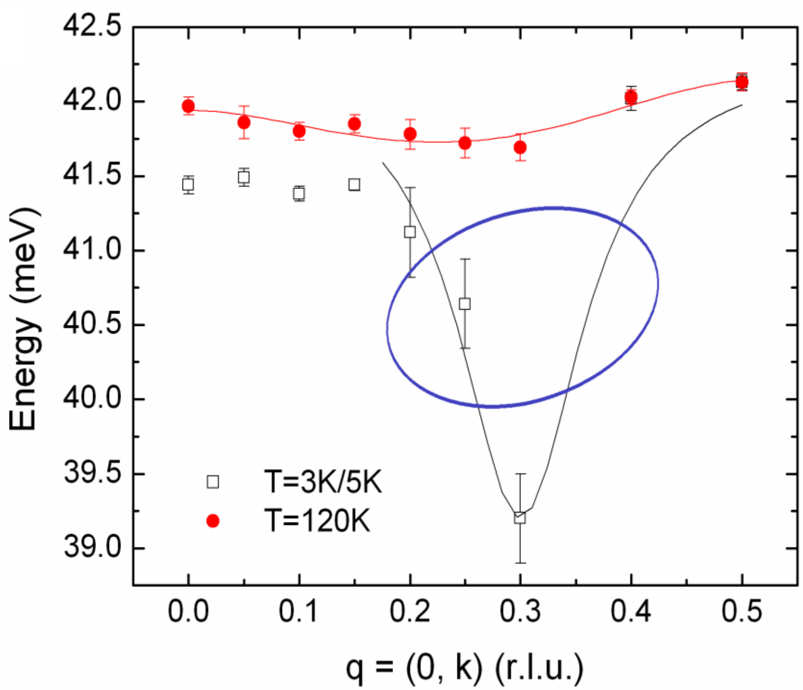
Cu-O bond-bending mode

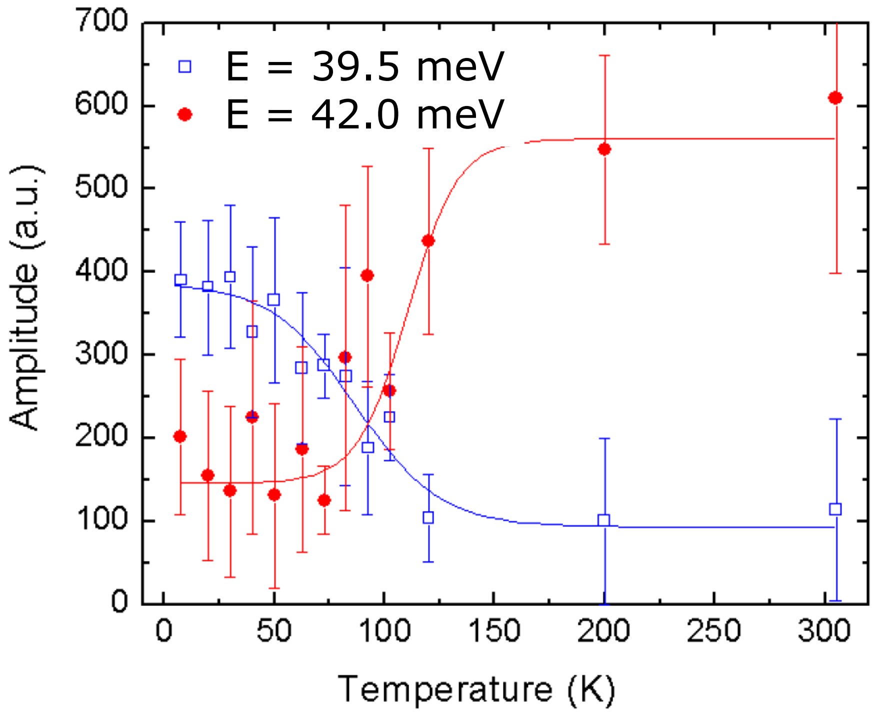
in optimally doped $YBCO_7$

onset of dispersion anomaly

for T ~ T_c





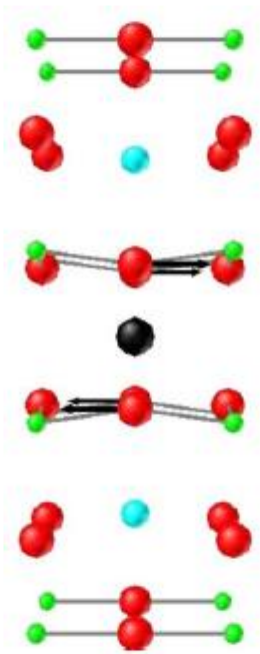


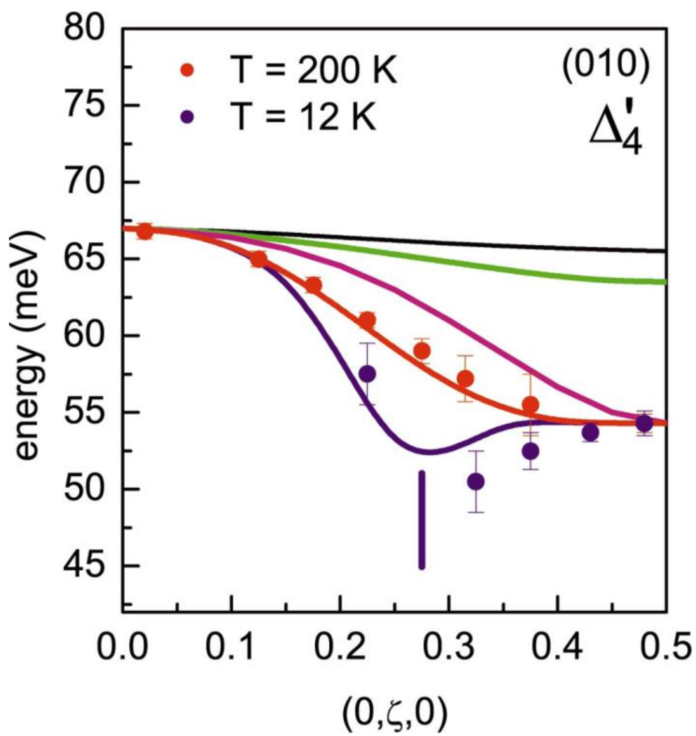
Raichle et al.
PRL 2011

für Festkörperforschung

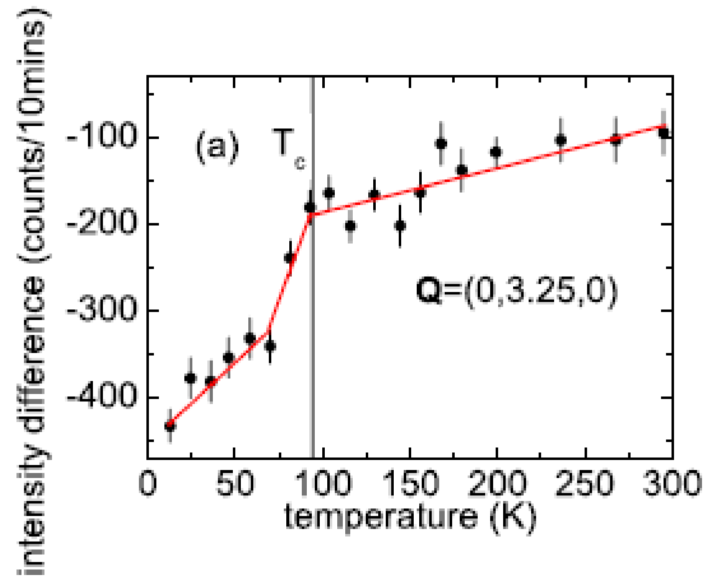
Optical phonons

similar dispersion anomaly for Cu-O bond-stretching mode





Pintschovius et al.
PRB 2004



Reznik et al.
PRB 2008

- fluctuating CDWs at optimaly doping?
- CDW-related Kohn anomalies → antinodal ARPES kinks?

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Summary & Outlook

summary

reasonably comprehensive description of spin and charge correlations in cuprates

key theoretical challenges

- understanding relative stability of SDW, CDW, superconductivity in 2D metals
- quantitative understanding of high-temperature superconductivity
- prediction of higher-temperature superconductors

outlook

- oxide molecular beam epitaxy
- dynamical control