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Charge fluctuations and electron phonon coupling studied by very high resolution RIXS

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IXS 2019 – Inelastic X-ray Scattering – Stony Brook – 26 June 2019



Summary

- CDW in cuprates outside the pseudogap: overdoped Bi2201
- CDW in cuprates outside the pseudogap: (Nd/Y)123
- e-ph interaction measured by RIXS



**Ying Ying
Peng**



**Lucio
Braicovich**



**Nick
Brookes**



**Riccardo
Arpaia**



**Roberto
Fumagalli**



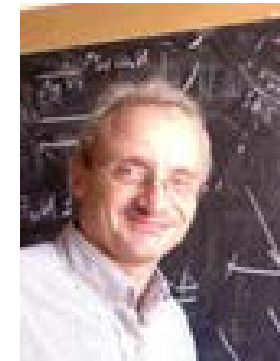
**Matteo
Rossi**



**Carlo
Di Castro**



**Tom
Devereaux**



**Marco
Grilli**



**Jeroen
van den Brink**

Acknowledgements



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CHALMERS

Politecnico di Milano
§now at Beijing U.

ESRF

Chalmers University

Roma La Sapienza

Cottbus

Max Planck Institut,
Stuttgart

KIT, Karlsruhe

CNR/SPIN Napoli

IoP, CAS, Beijing

IFW Dresden
U. Tennessee

Stanford

GG, Lucio Braicovich, Ying Ying Peng[§], Roberto Fumagalli, Matteo Rossi, Marco Moretti Sala, Leonardo Martinelli, Mattia Pagetti, Greta Dellea, Michele Conni, Giovanni De Vecchi

Nick Brookes, Flora Yakhou, Kurt Kummer, Davide Betto,
Andrea Amorese, Christian Morawe and many others

Riccardo Arpaia, Eric Andersson, Floriana Lombardi

Marco Grilli, Sergio Caprara, Carlo Di Castro

Goetz Seibold

Matteo Minola, Bernhard Keimer, Emilie Lefrançois,
Hakuto Suzuki, Martin Bluschke

Matthieu Le Tacon

Marco Salluzzo, Gabriella De Luca

Y. Ding, X. J. Zhou

**Jeroen van den Brink,
Steven Johnston**

**Wei-Sheng Lee, Laura Chaix, Makoto Hashimoto, Z.X. Shen,
Tom Devereaux, Chunjin Jia, Brian Moritz**

N.B. Brookes , F. Yakhou-Harris, K. Kummer, A. Fondacaro, J.C. Cezar, D. Betto, E. Velez-Fort, A. Amorese, G. Ghiringhelli, L. Braicovich, R. Barrett, G. Berruyer, F. Cianciosi, L. Eybert, P. Marion, P. van der Linden, L. Zhang, [NIM A 903, 175 \(2018\)](#)

The merits of ERIXS at ID32

Resolving power: 40,000
at 1 keV
3 times better than
previous record

Collimating Mirror to
increase horizontal
acceptance (20 mrad)

Two CCD

Upgrade of the
optics
in 2020
**Combined
resolving power
up by 30%**

Ref: Kurt Kummer, Nick
Brookes

detection,
CCD thanks to
ated beam, for
resolution [1]

mirror,
ure
on of
tons [2]

eter,
for true diffraction quality scanning



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[1] A Amorese, C Langini,
G Dellea, K Kummer, NB
Brookes, L Braicovich, G
Ghiringhelli, *NIM A* **935**,
222 (2019)

[2] L. Braicovich, M.
Minola, G. Dellea, M. Le
Tacon, M. Moretti Sala, C.
Morawe, J.-Ch. Peffen, R.
Supruangnet, F.
Yakhou, G. Ghiringhelli,
and N. B. Brookes, *Rev.*
Sci. Instrum. **85**, 115104
(2014)

N.B. Brookes, F. Yakhou-Harris, K. Kummer, A. Fondacaro, J.C. Cezar, D. Betto, E. Velez-Fort, A. Amorese, G. Ghiringhelli, L. Braicovich, R. Barrett, G. Berruyer, F. Cianciosi, L. Eybert, P. Marion, P. van der Linden, L. Zhang, *NIM A* **903**, 175 (2018)



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Other contributions from ERIXS@ID32 in this conference



Ying Ying Peng

Evolution of the magnetic excitations in high T_c cuprates
next talk

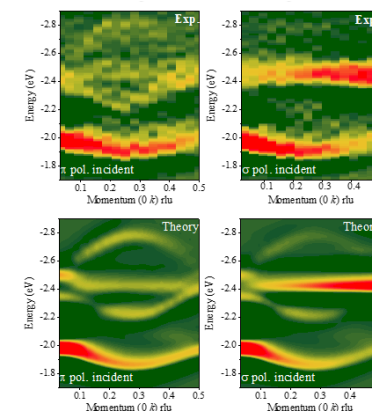
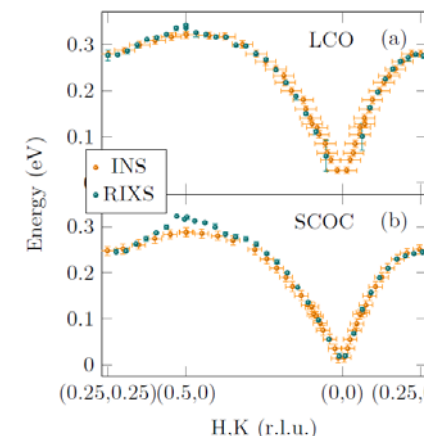


Davide Betto

Bimagnon tail in the magnetic excitations of $Sr_2CuO_2Cl_2$ and La_2CuO_4
Poster PT-4

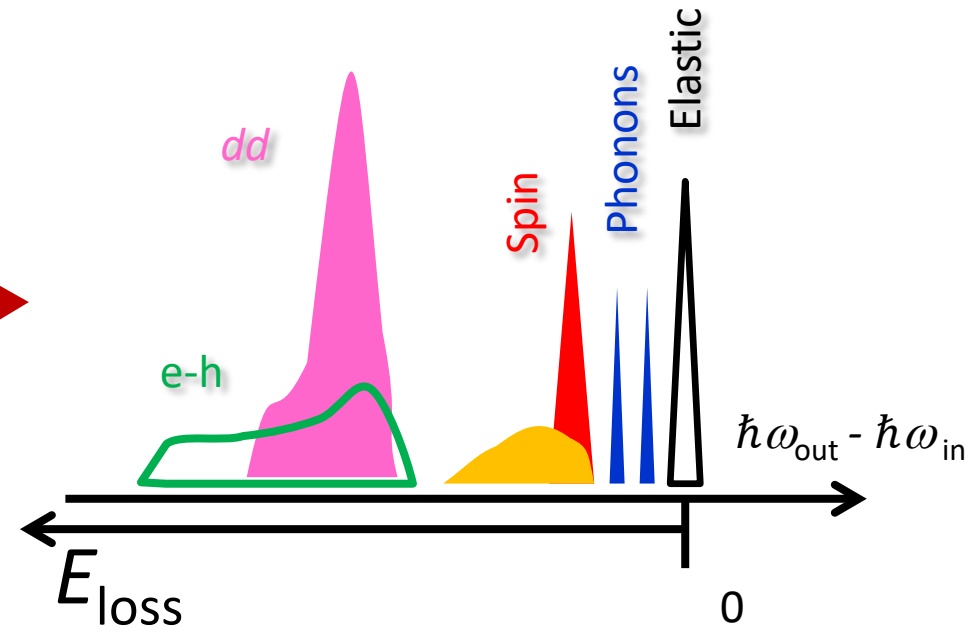
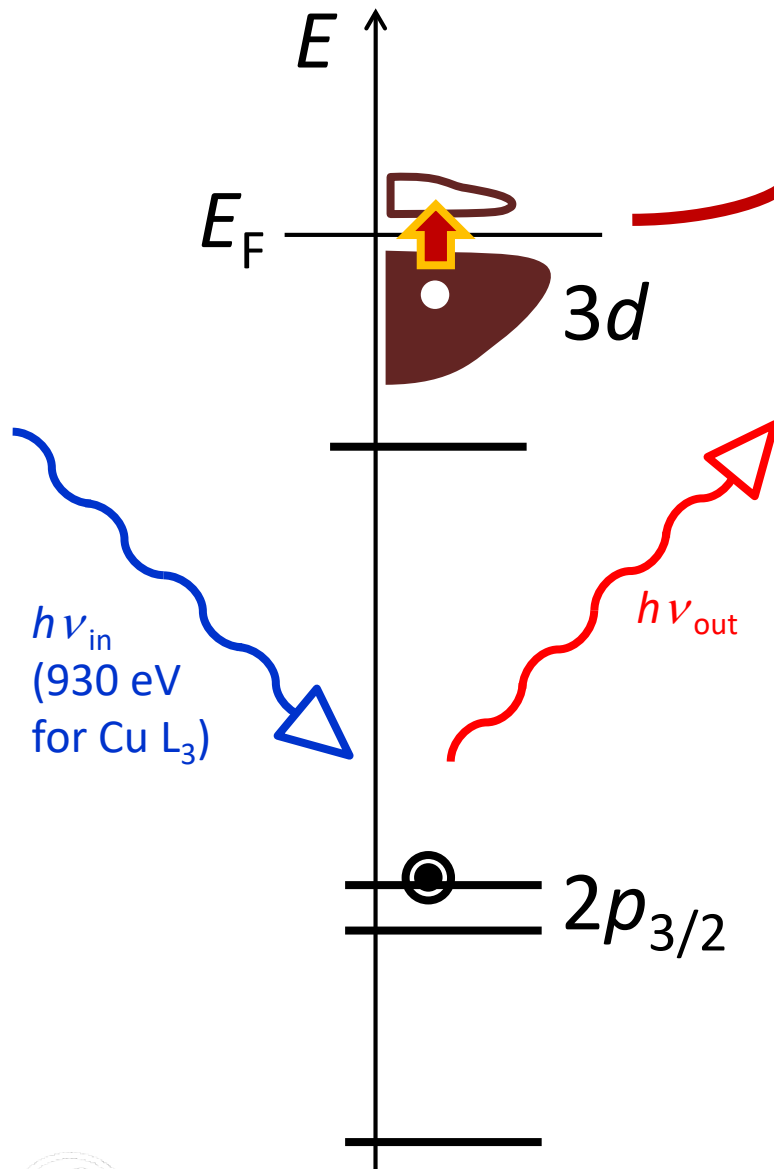
Roberto Fumagalli

Dispersing orbital excitations in the quasi-1D and 2D cuprates
Poster PT-15



and talks by **Kurt Kummer, Andrea Amorese, Hu Miao, Wei Sheng Lee**

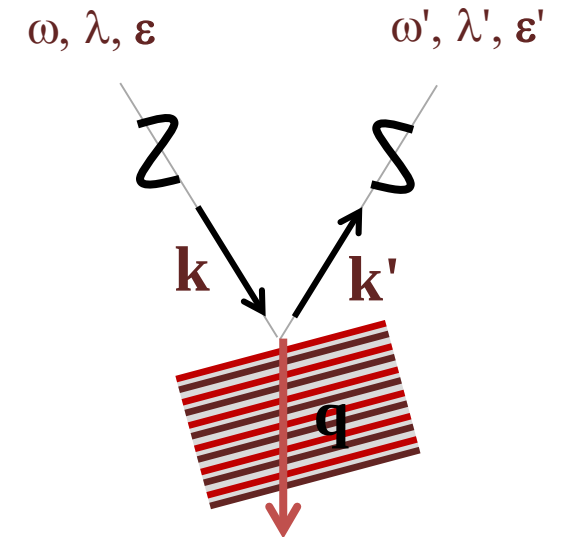
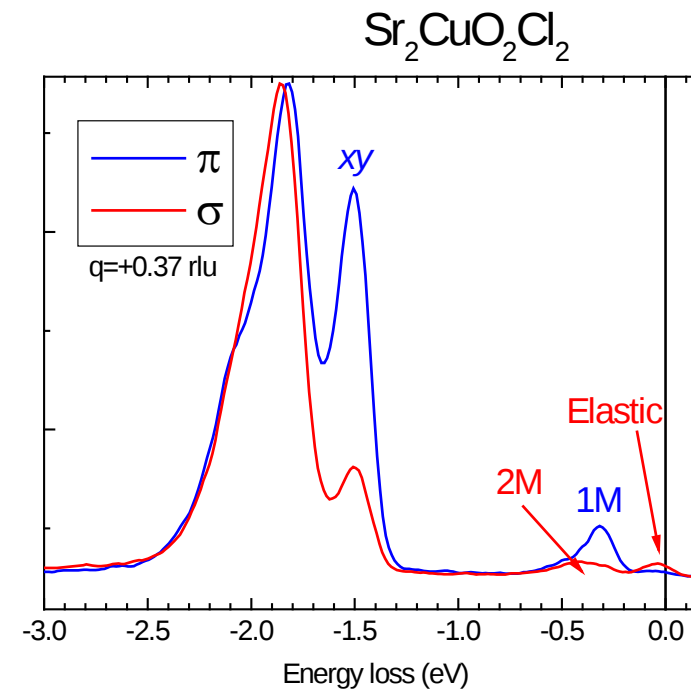
Cu L₃ RIXS



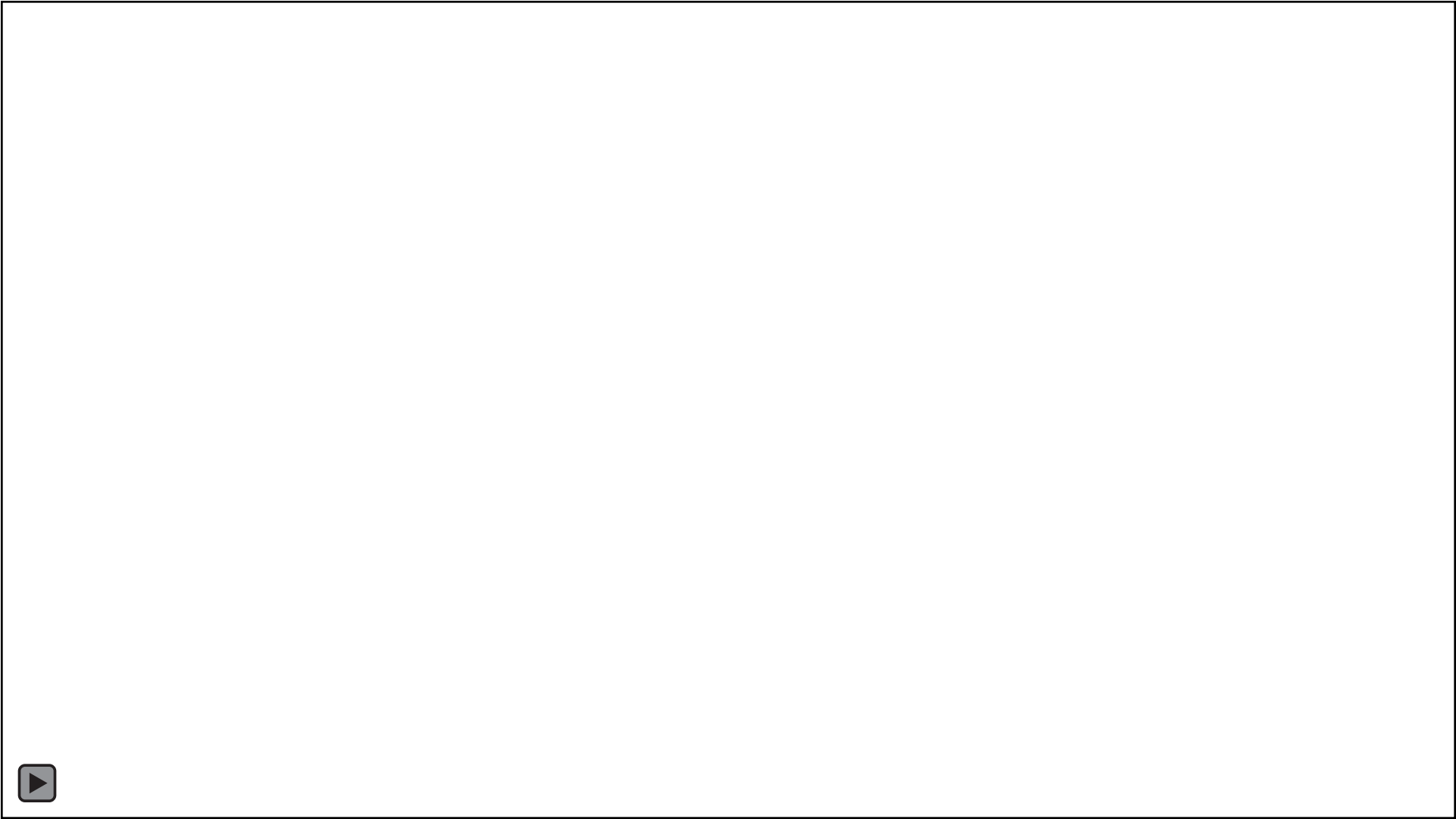
$$\Omega = \omega - \omega'$$

$$\mathbf{q} = \mathbf{k}' - \mathbf{k}$$

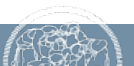
$$\epsilon, \epsilon': \text{polarization}$$



High Tc Superconducting cuprates

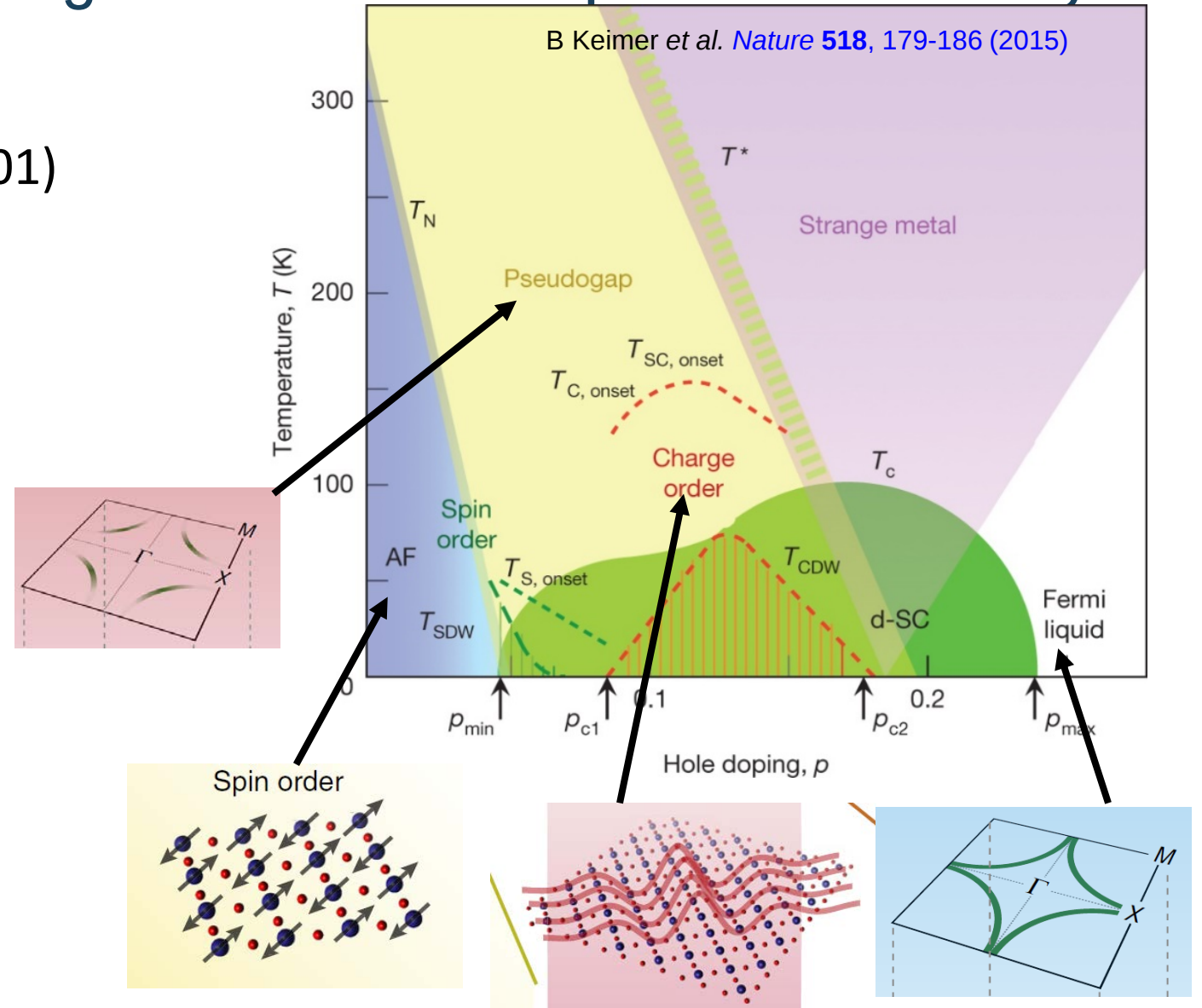
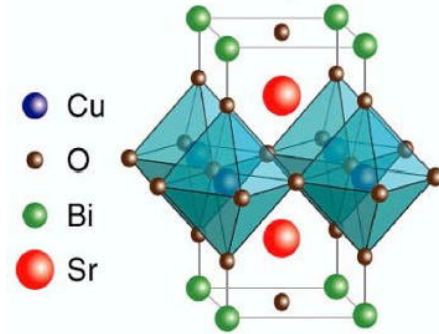
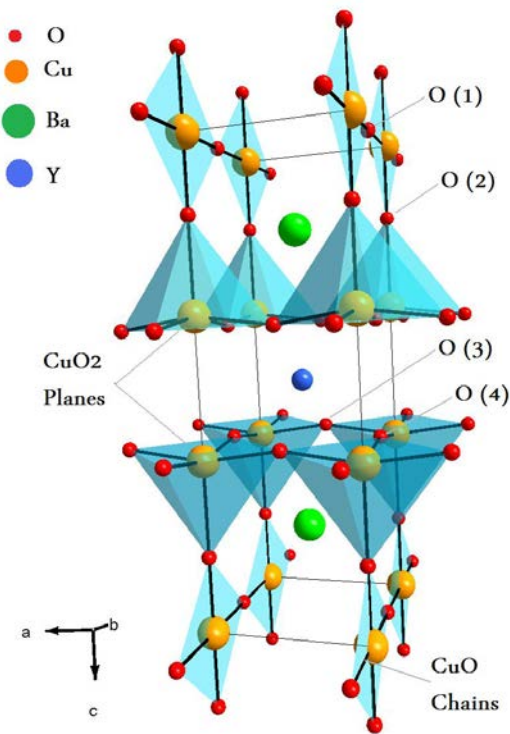


Thanks to Claudio Mazzoli

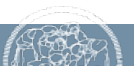


Cuprates: Magnetism, Charge Order and Superconductivity

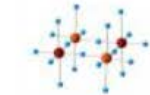
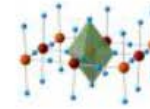
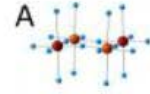
$\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ (YBCO) $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta}$ (Bi2201)



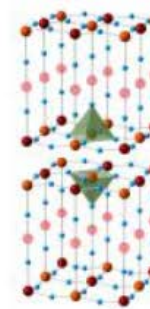
J. Pelliciari and R. Comin *Nature Materials* **17**, 661 (2018)



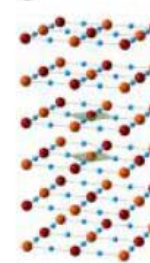
Spin-waves with Cu L₃ RIXS



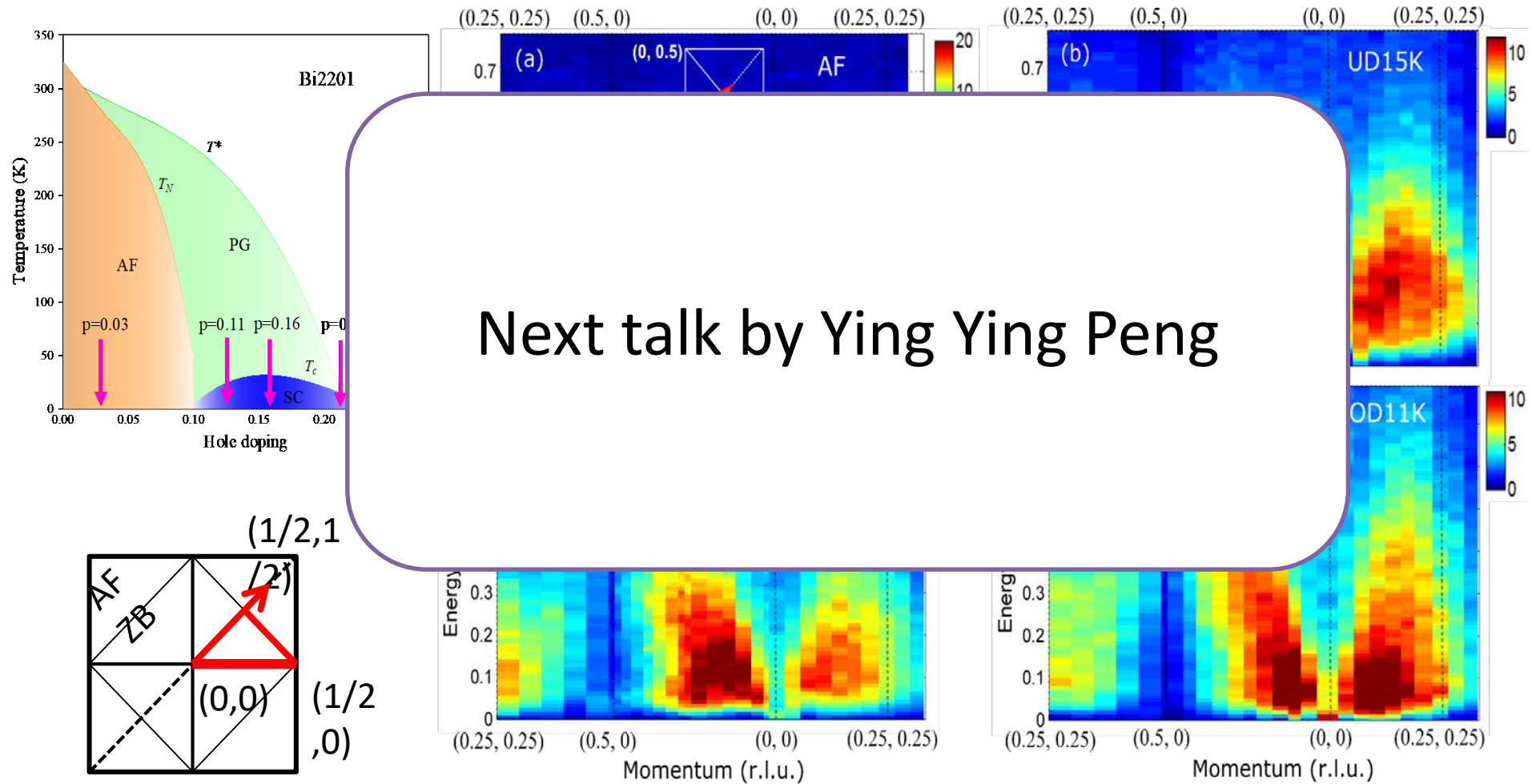
B



C

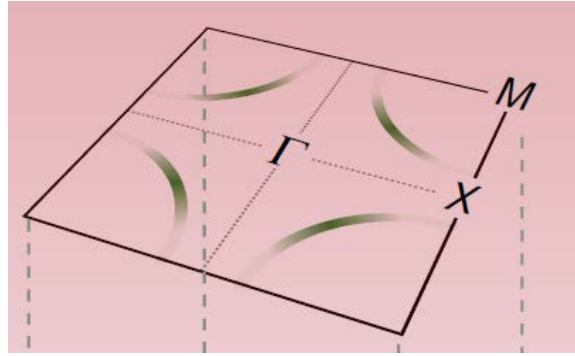
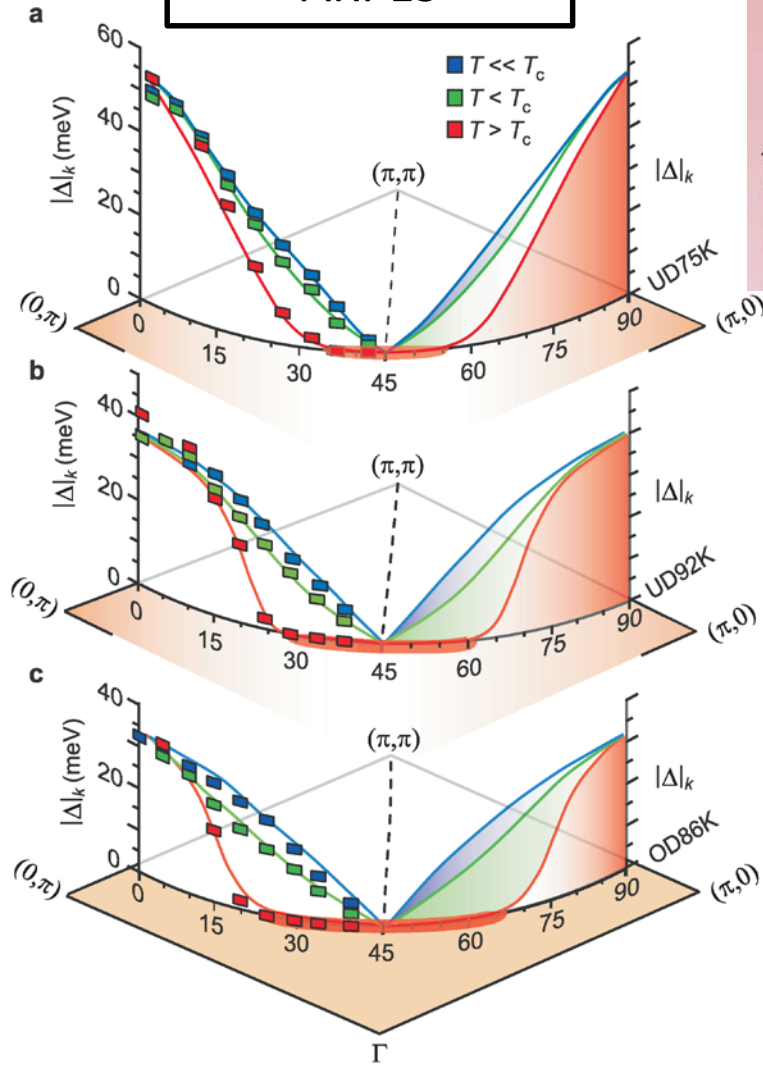


Paramagnons in Bi2201



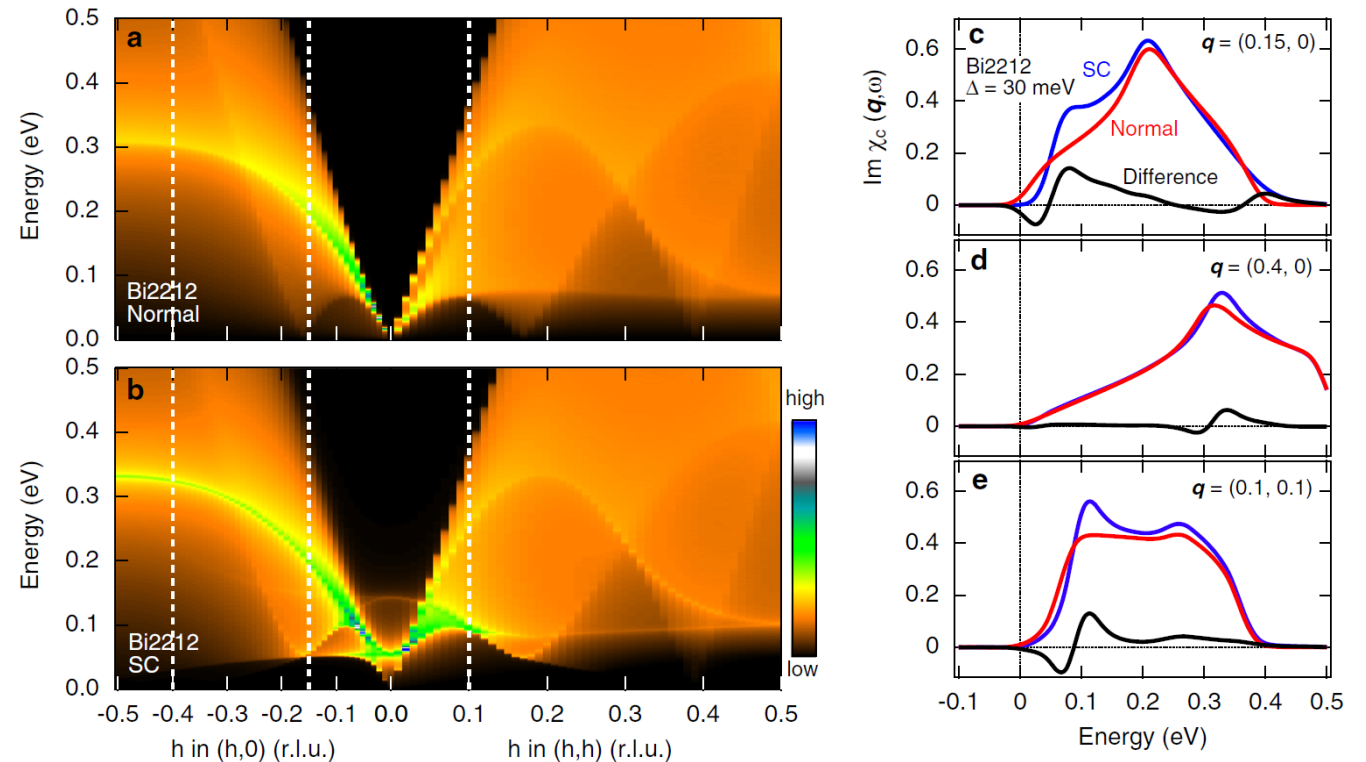
The superconducting gap

ARPES



“RIXS”

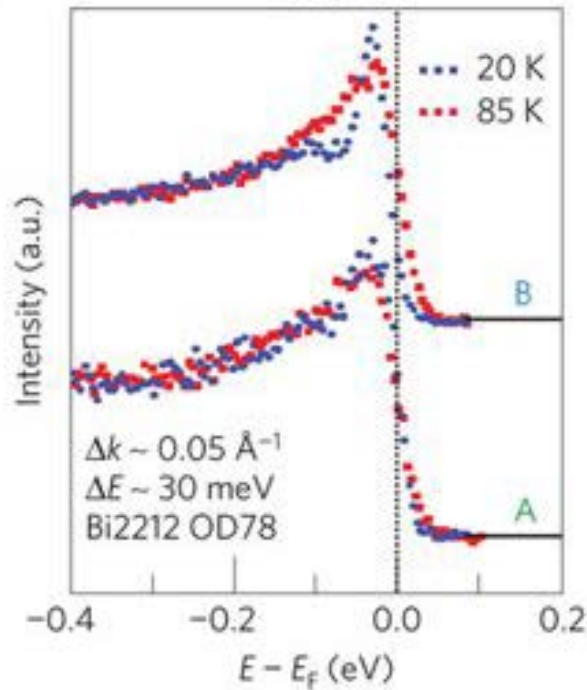
Charge susceptibility:
 $\text{Im } \chi_c(\mathbf{q}, \omega)$



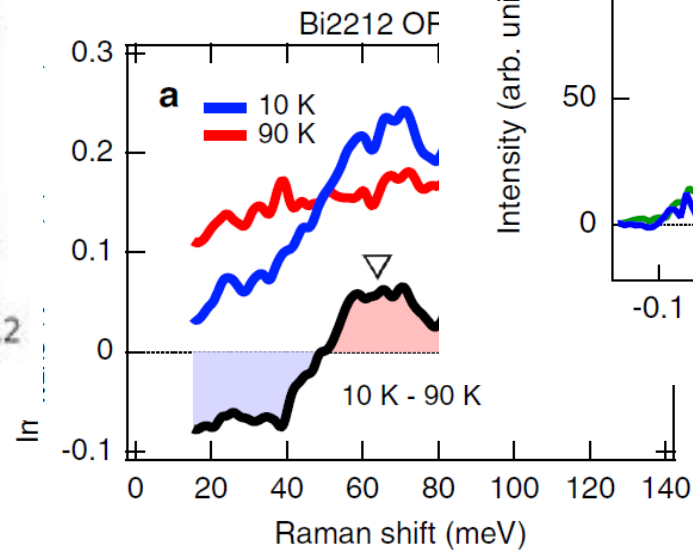
Hakuto Suzuki, Matteo Minola, Yi Lu, Yingying Peng, Roberto Fumagalli, Emilie Lefrançois, Toshinao Loew, Juan Porras, Kurt Kummer, Davide Betto, Shigeyuki Ishida, Hiroshi Eisaki, Cheng Hu, Xingjiang Zhou, Maurits W. Haverkort, Nicholas B. Brookes, Lucio Braicovich, Giacomo Ghiringhelli, Matthieu Le Tacon and Bernhard Keimer, *npj Quantum Materials* **3**, 65 (2018)

The SC gap in RIXS

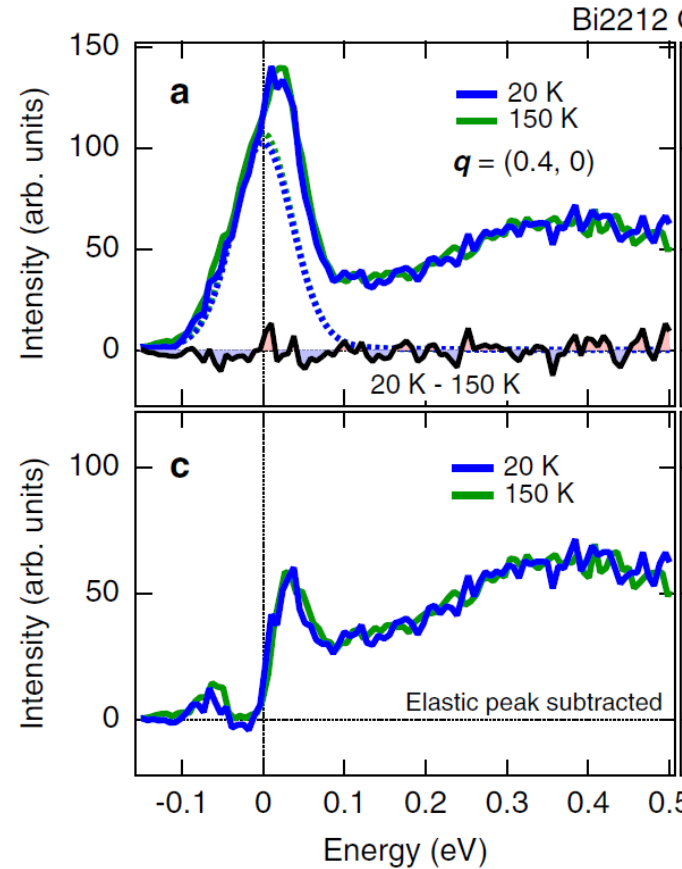
ARPES



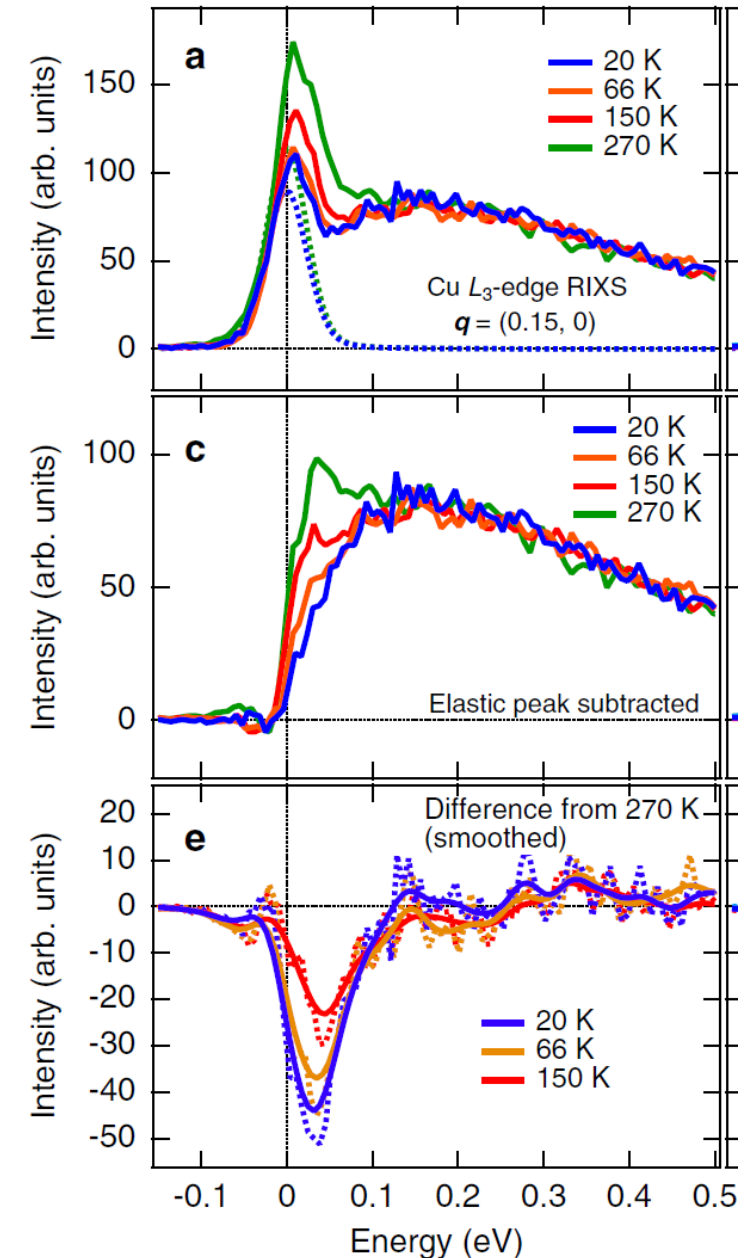
Raman: $q=C$



RIXS: $q \neq 0$

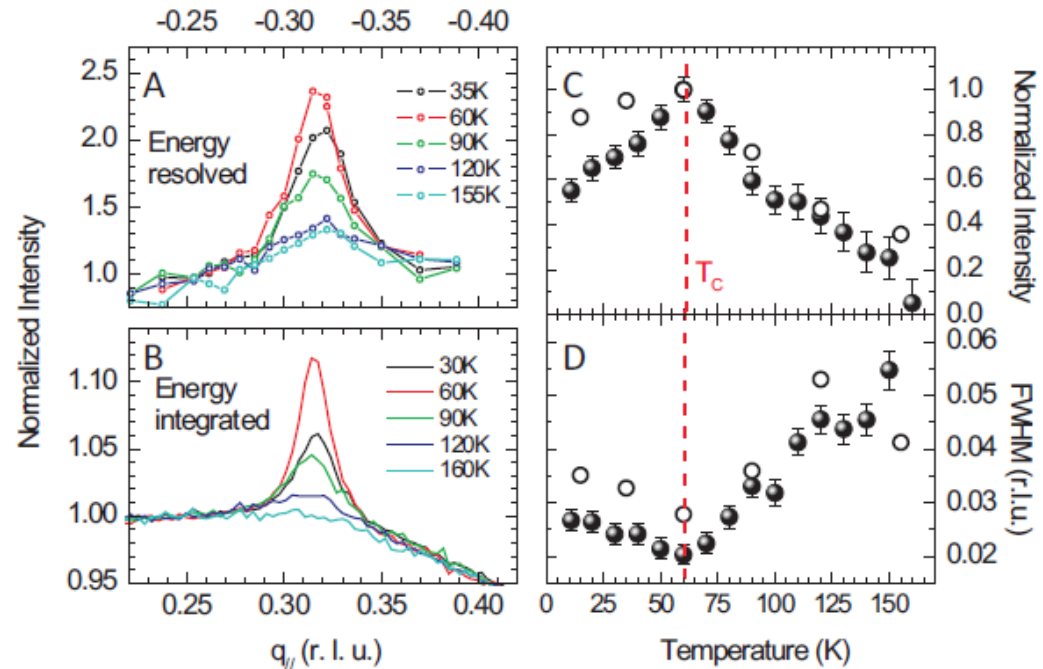
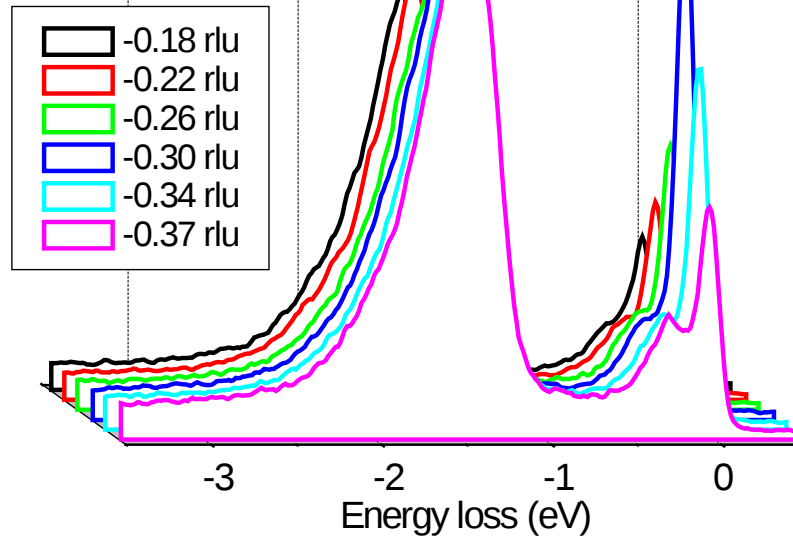


Bi2212 OP90K



First observation of CDW in YBCO with R(I)XS

NBCO $T_c=65K$
V pol, $T=15K$



G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N.B. Brookes, G.M. De Luca, A. Frano, D. G. Hawthorn, F. He, T. Loew, M. Moretti Sala, D.C. Peets, M. Salluzzo, E. Schierle, R. Sutarto, G. A. Sawatzky, E. Weschke, B. Keimer, L. Braicovich, *Science* **337**, 821 (2012)

Other evidences of CDW in YBCO

NMR, Charge modulation
at low T, high field

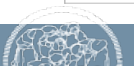
Wu et al. *Nature* **477** 191 (2011)

$L = 0.5$: Doubling of unit cell along c-axis
Field enhancement of the CDW (HXRD)

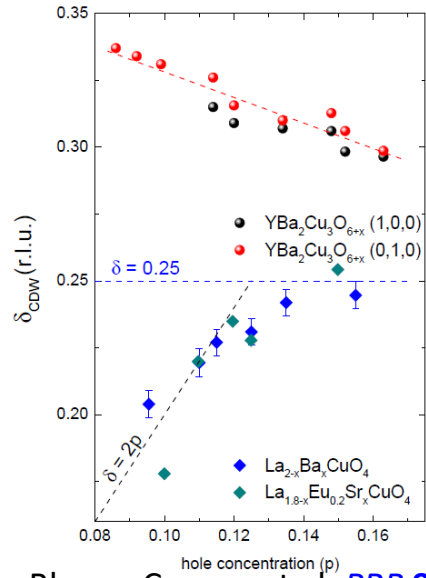
Chang et al. *Nat.Phys.* **8** 871 (2012)

Bi-axial, Static order
under high field.

Leboeuf et al. *Nat.Phys.* **9** 79 (2013)



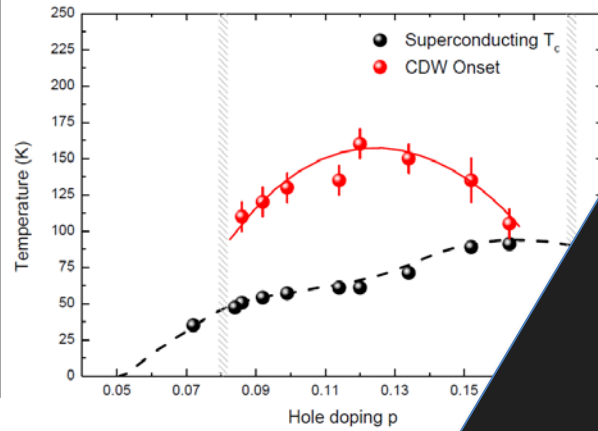
Ubiquitous CDW in cuprates



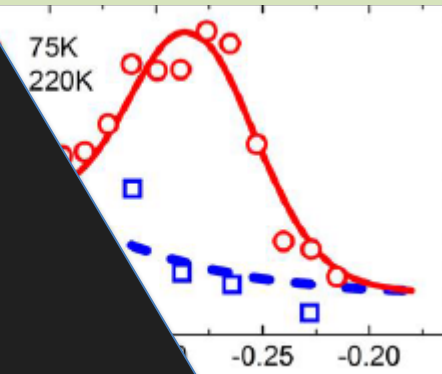
Blanco-Canosa et al. *PRB* **90**, 054513 (2014)

Huecker et al. *PRB* **90**, 054514 (2014)

YBCO

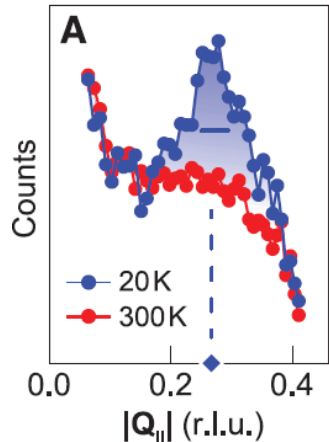


Hg1201 underdoped



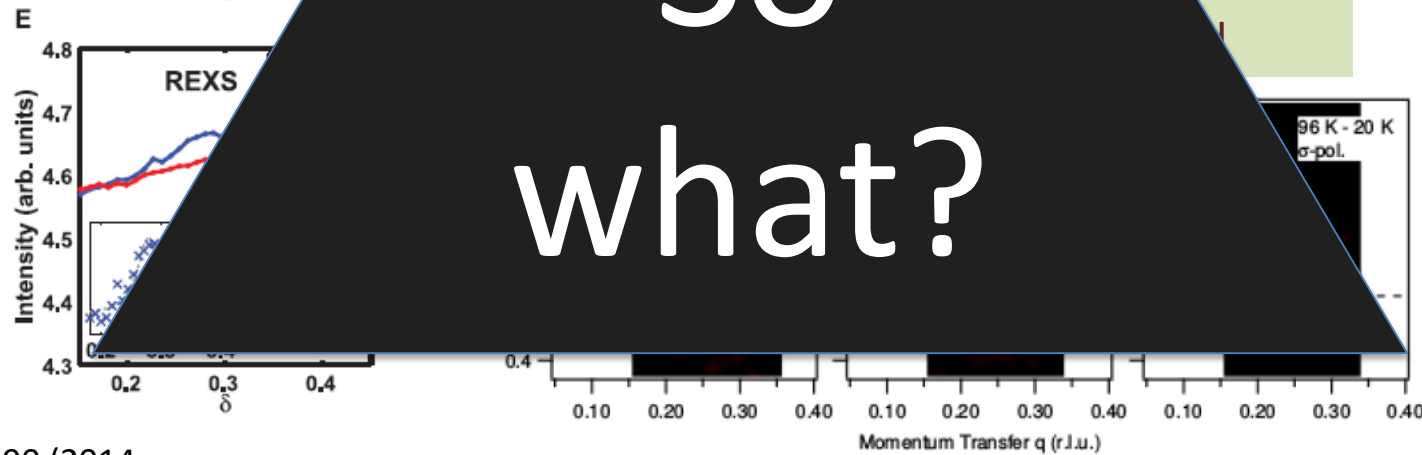
PRB **90**, 054515 (2014)

Bi2201 and Bi2212 underdoped



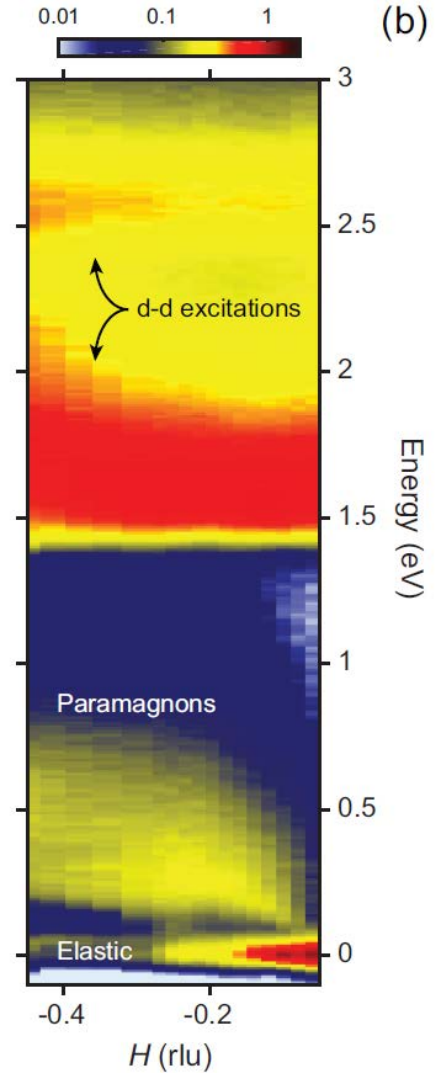
R. Comin et al, *Science* **343**, 390 (2014);

Eduardo H. da Silva Neto et al, *Science* **343**, 393 (2014)



M. Hashimoto, G. Ghiringhelli et al, *PRB Rapid* **89** 220511 (2014)

NCCO e-derdoped

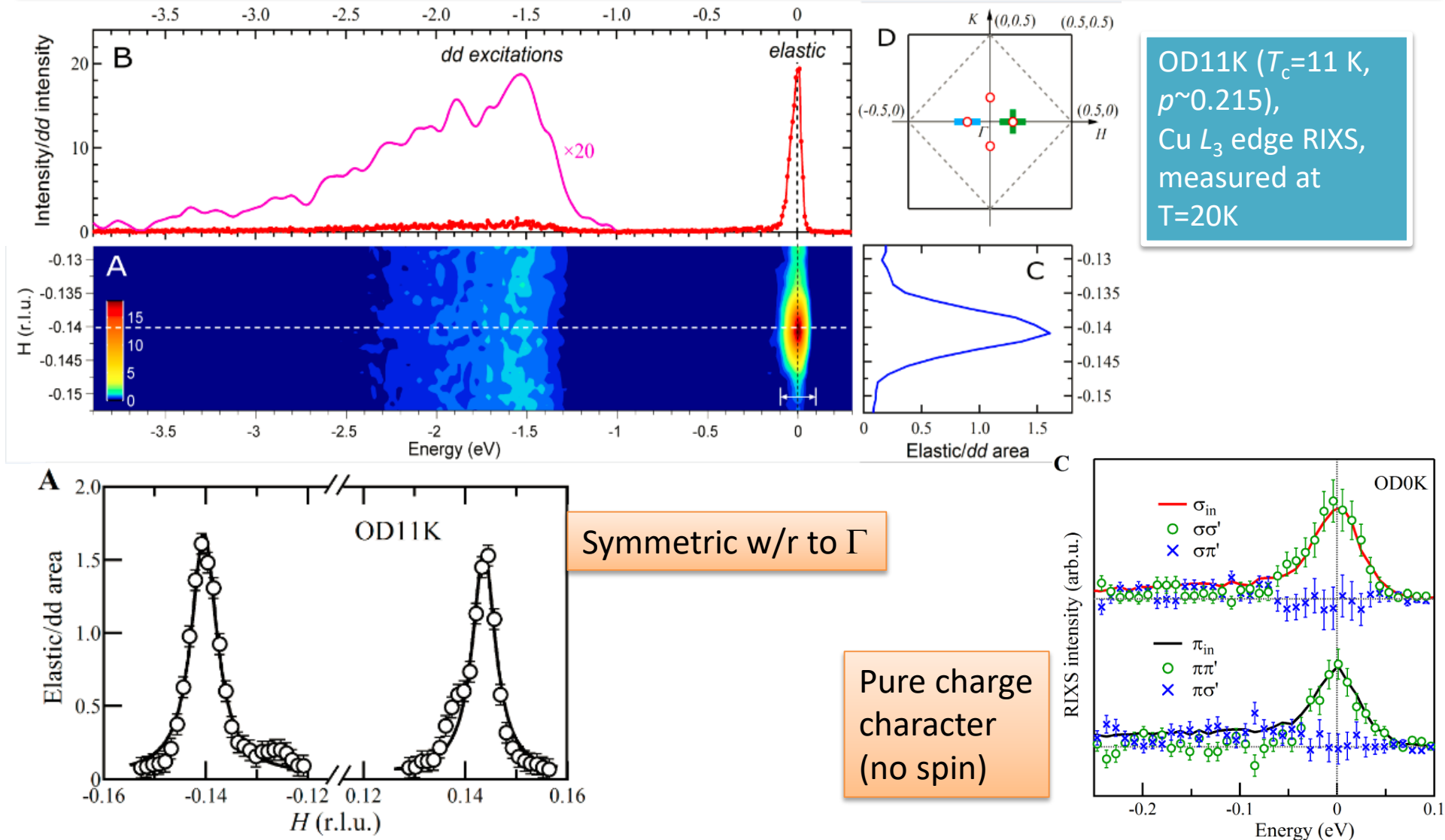


M. E. H. da Silva Neto et al, *PRB* **98** 161114 (2018)

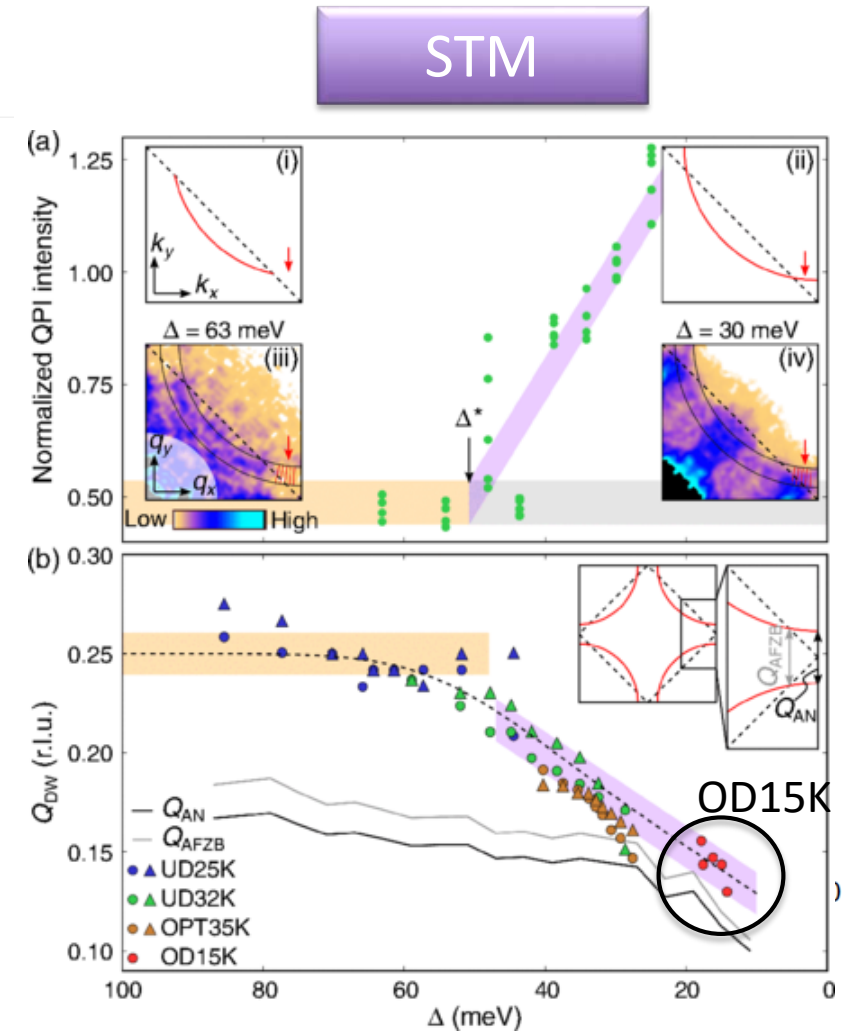
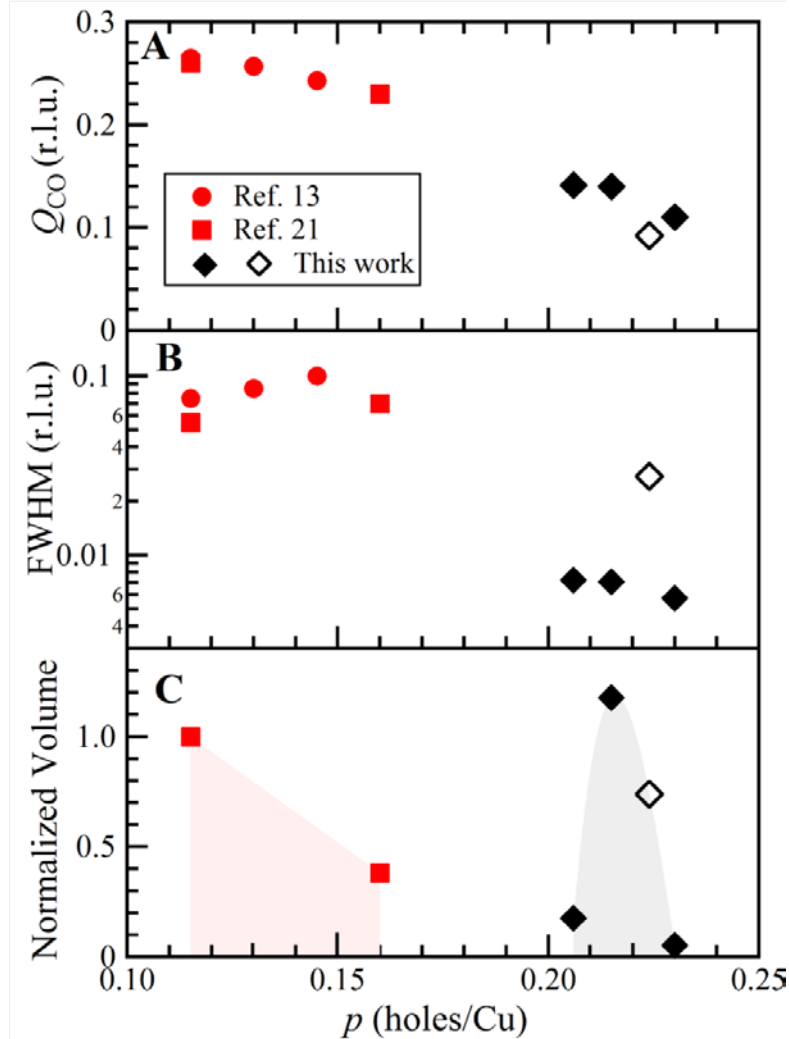
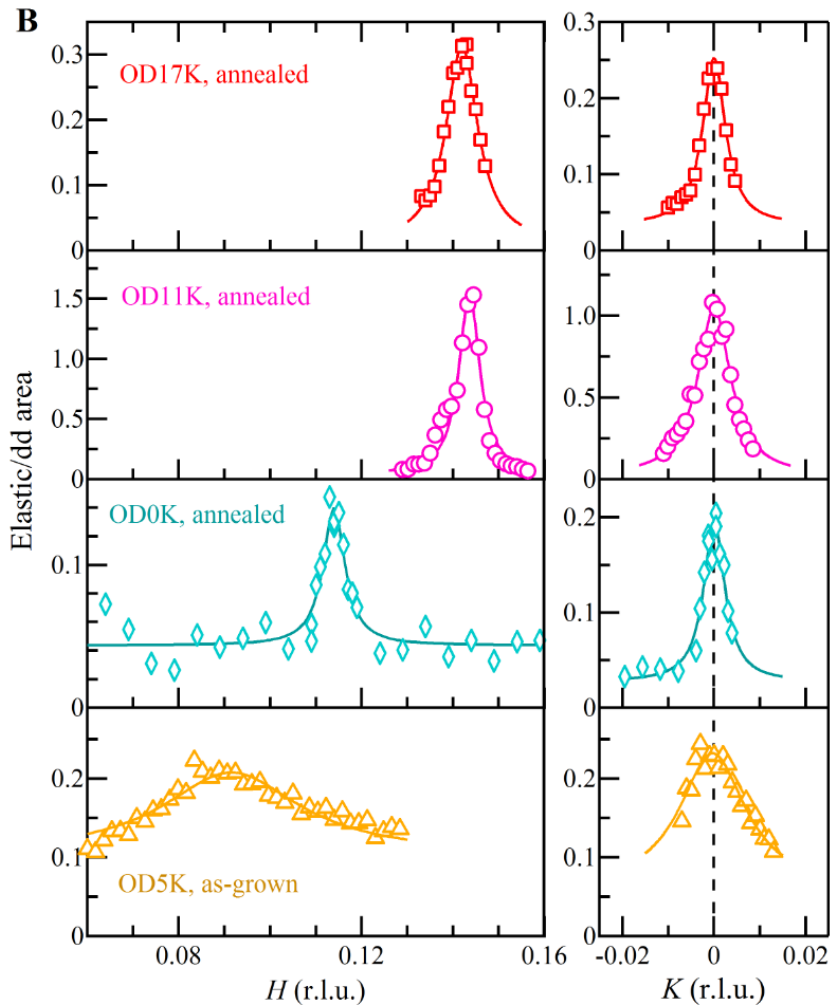
So
what?

Overdoped $(\text{Bi,Pb})_{2.12}\text{Sr}_{1.88}\text{CuO}_{6+\delta}$

Unexpected observation of a very intense and sharp peak in pseudo-tetragonal (1,0) direction



Re-entrant CDW in OD Bi2201



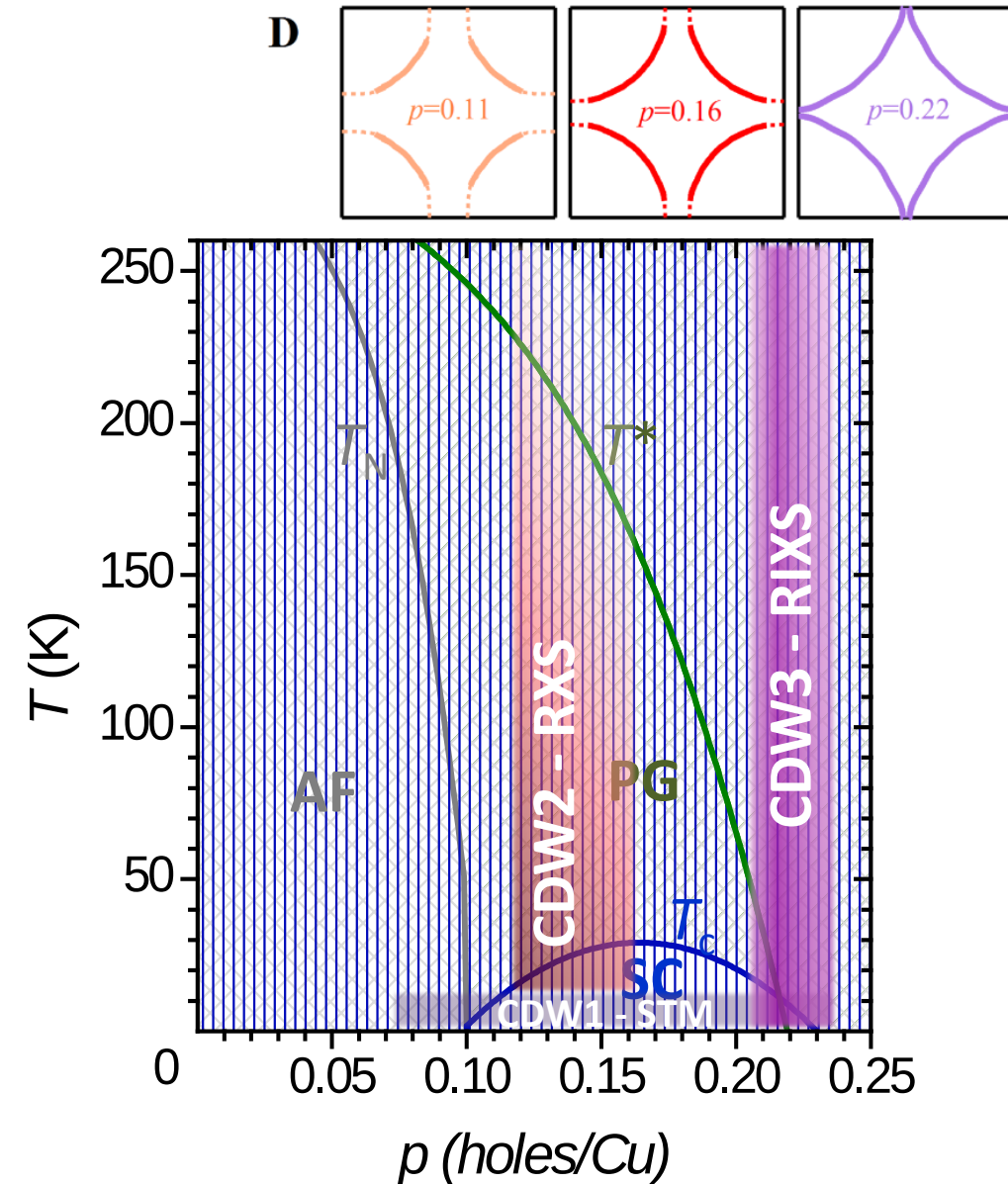
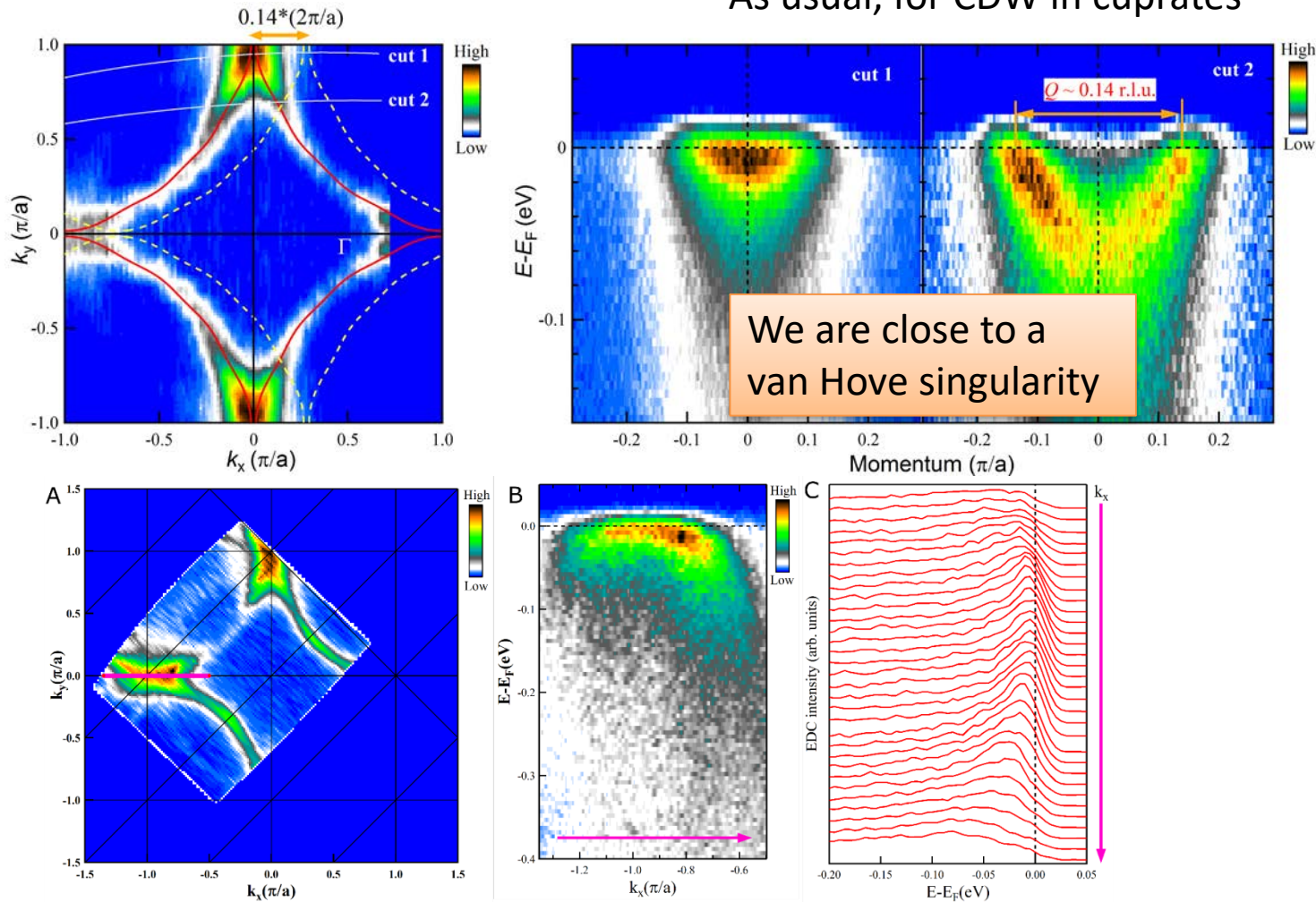
Y. Y. Peng, R. Fumagalli, Y. Ding, M. Minola, S. Caprara, D. Betto, M. Blüschke, G. M. De Luca, K. Kummer, E. Lefrançois, M. Salluzzo, H. Suzuki, M. Le Tacon, X. J. Zhou, N. B. Brookes, B. Keimer, L. Braicovich, M. Grilli, G. Ghiringhelli, *Nature Materials* **17**, 697 (2018).

Tatiana A. Webb, Michael C. Boyer, Yi Yin, Debanjan Chowdhury, Yang He, Takeshi Kondo, T. Takeuchi, H. Ikuta, Eric W. Hudson, Jennifer E. Hoffman, and Mohammad H. Hamidian
Phys. Rev. X **9**, 021021 (2019)

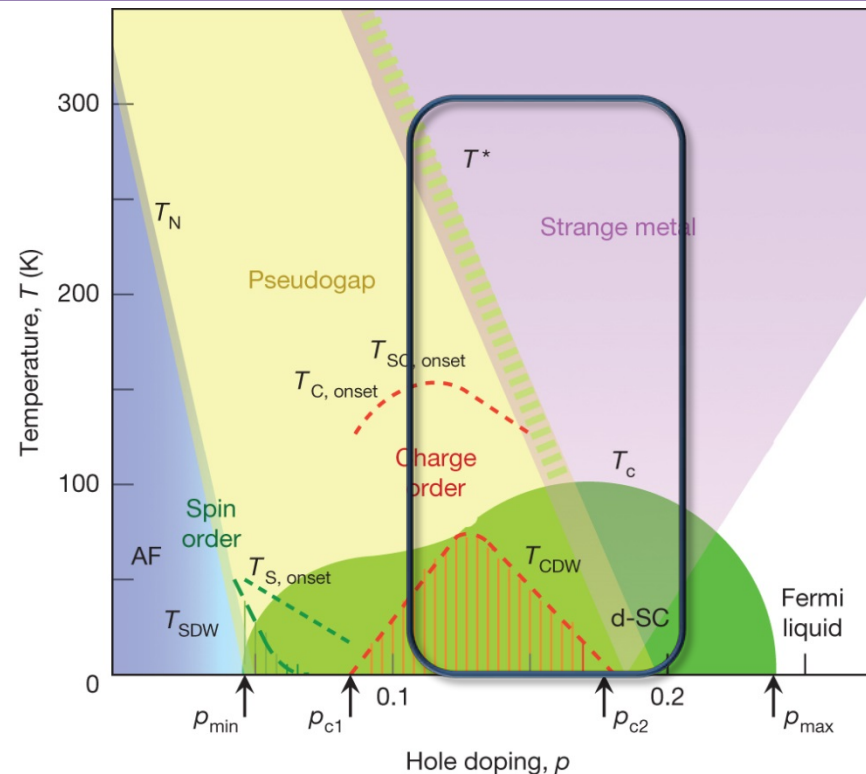
CDW in OD Bi2201: the role of electronic structure

ARPES on OD11K ($T_c=11$ K, $p\sim 0.215$), at $T=20$ K: no Fermi surface nesting or folding*

* As usual, for CDW in cuprates



Taking a better look at CDW, at “high” T , in (Y,Nd)BCO

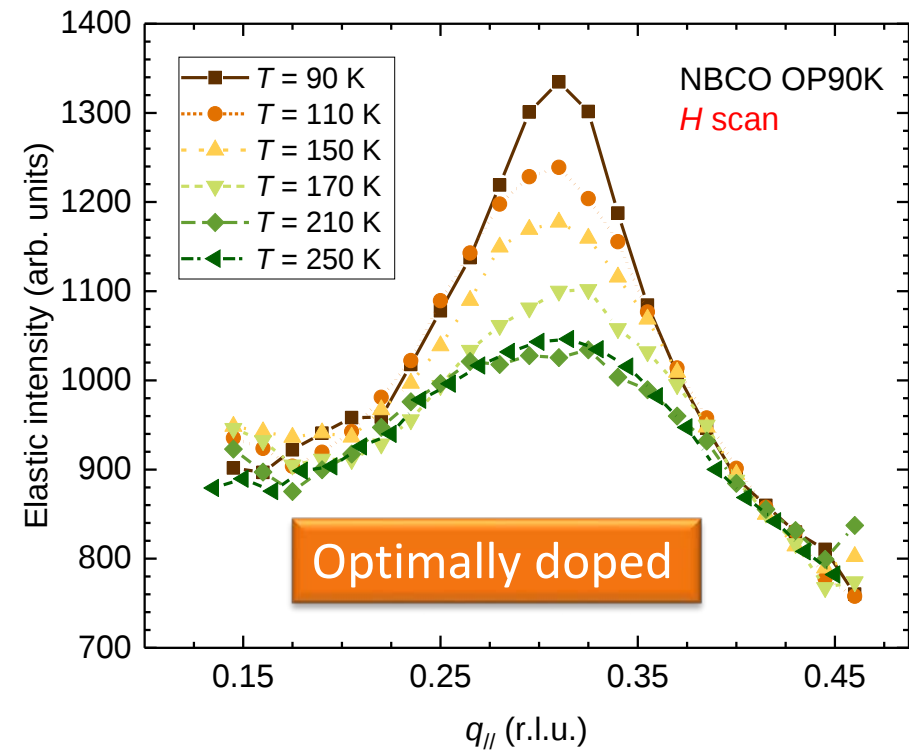
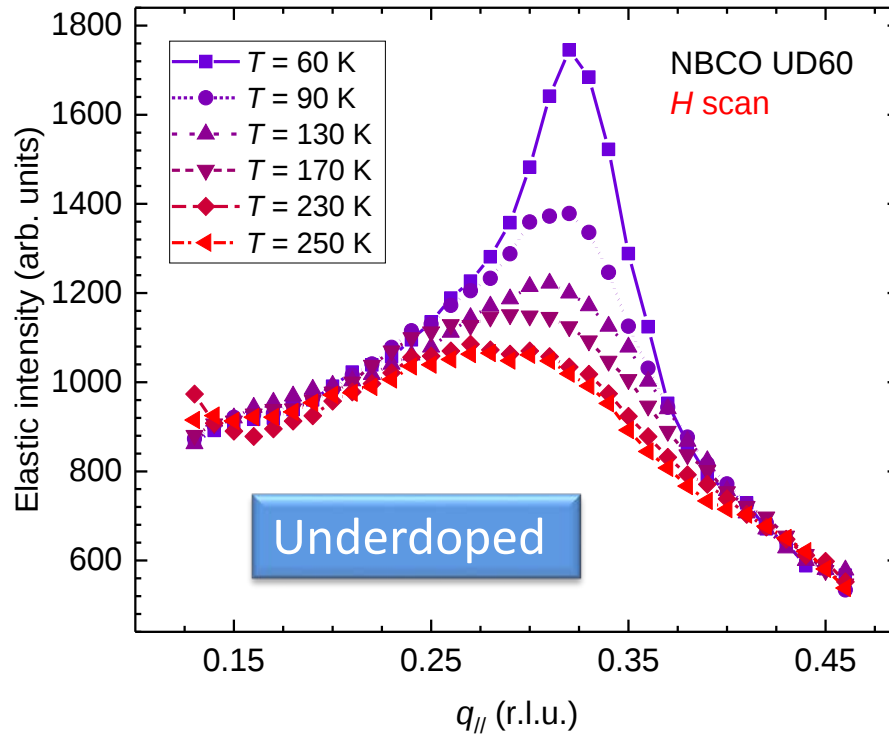
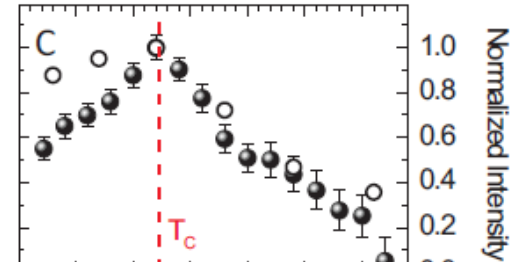
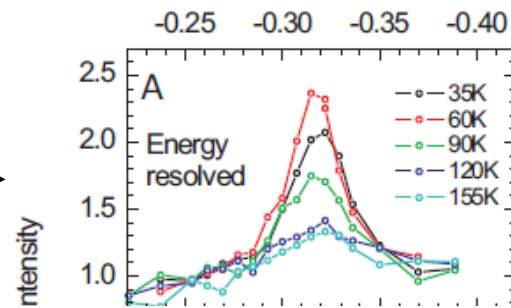
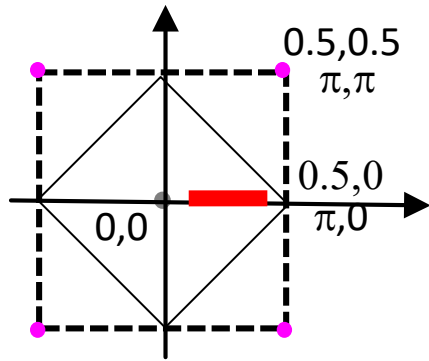


Samples

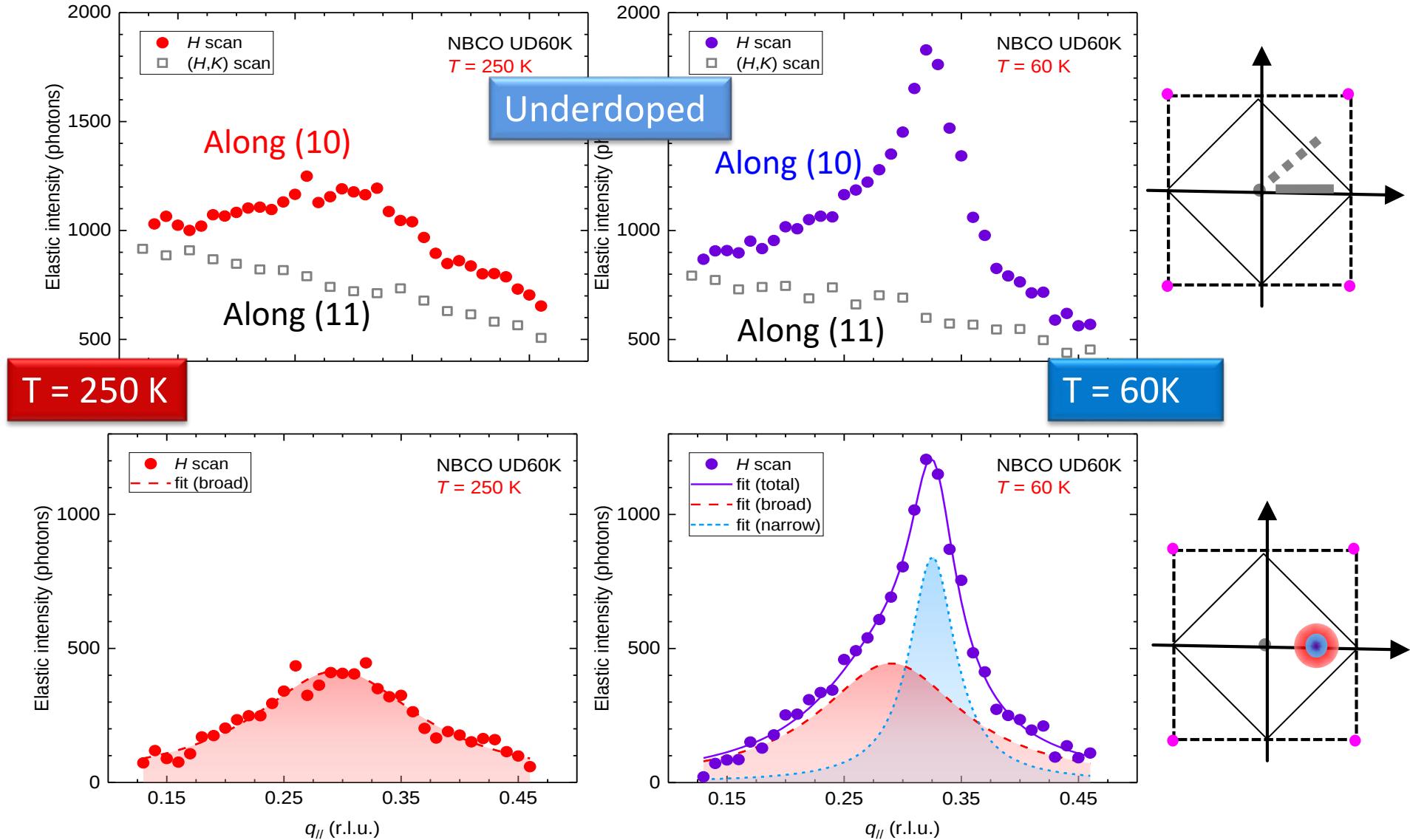
Epitaxial films grown
in CNR/SPIN Napoli
and Chalmers
(M Salluzzo, GM De
Luca, R Arpaia, E
Andersson, F
Lombardi)

R. Arpaia, S. Caprara, R. Fumagalli, G. De Vecchi, Y.Y. Peng, E. Andersson, D. Betto, G. M. De Luca, N. B. Brookes, F. Lombardi, M. Salluzzo, L. Braicovich, C. Di Castro, M. Grilli, G. Ghiringhelli, [arXiv1809.04949](https://arxiv.org/abs/1809.04949)

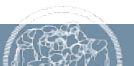
What is the real T_{CDW} ?



Not just one CDW peak



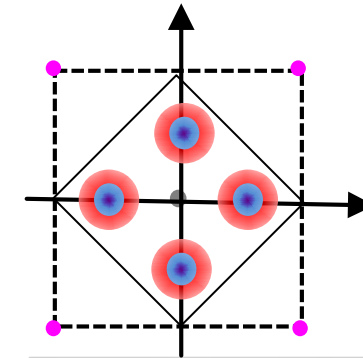
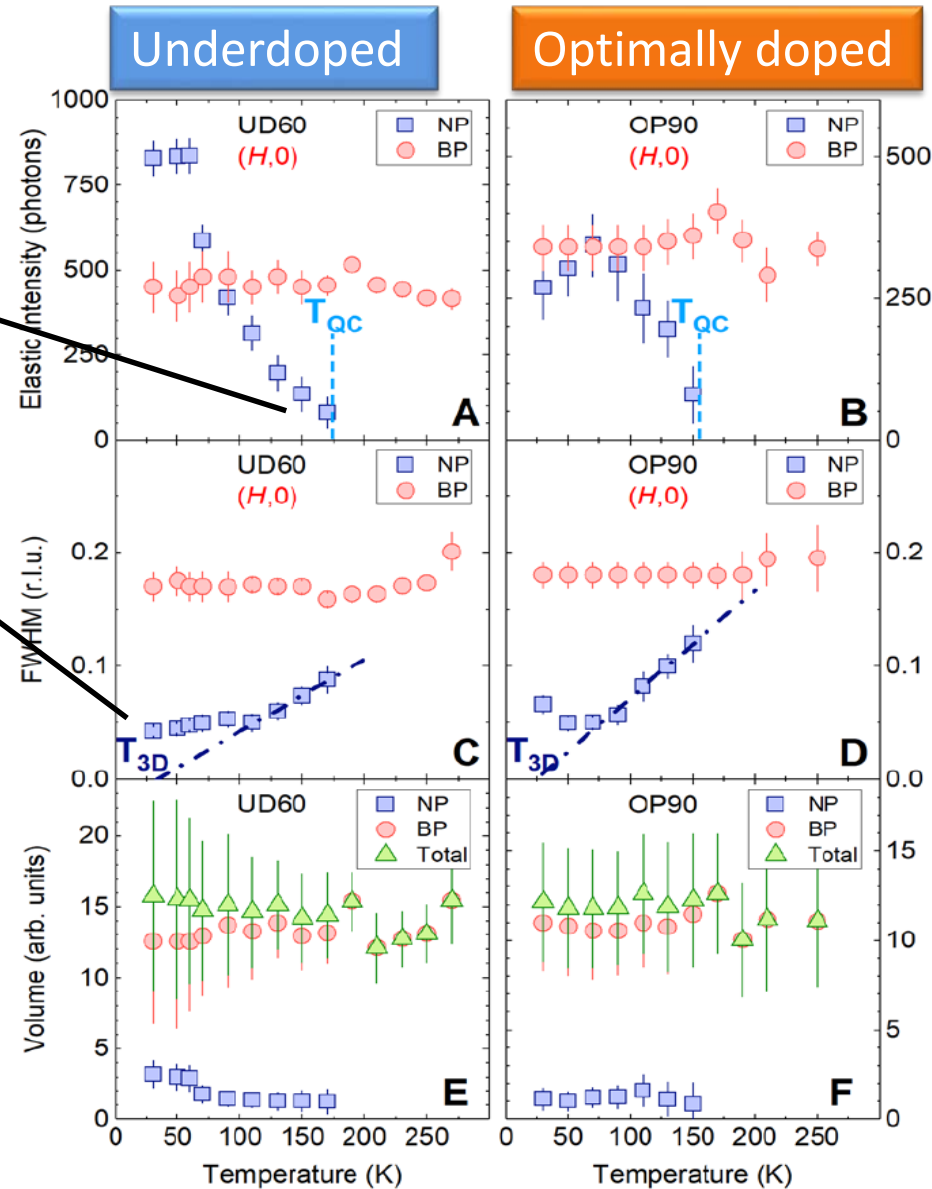
R. Arpaia, S. Caprara, R. Fumagalli, G. De Vecchi, Y.Y. Peng, E. Andersson, D. Betto, G. M. De Luca, N. B. Brookes, F. Lombardi, M. Salluzzo, L. Braicovich, C. Di Castro, M. Grilli, G. Ghiringhelli, [arXiv1809.04949](https://arxiv.org/abs/1809.04949)



Narrow Peak (NP) and Broad Peak (BP)

$T_{QC} = T_{CDW}$
Onset of NP

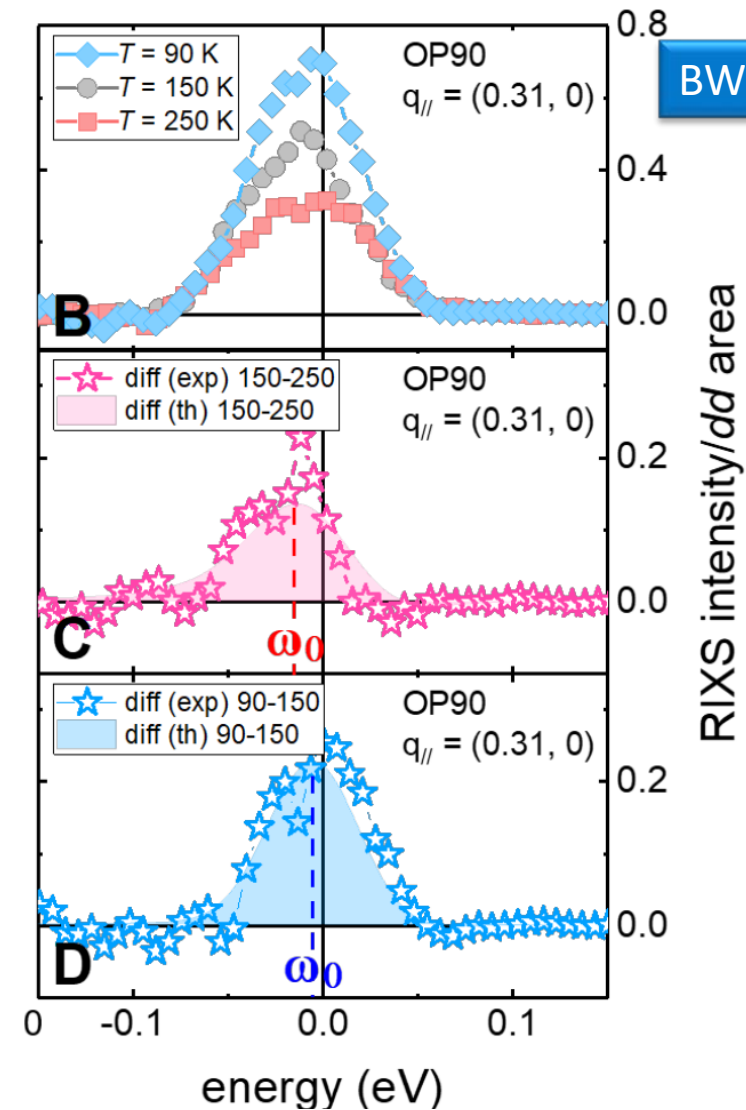
T_{3D}
Critical T for 3D
order
without SC



NP has critical
behavior
BP is almost
constant

BP is much larger in
integrated
amplitude than NP

Energy resolved measurements: charge density fluctuations



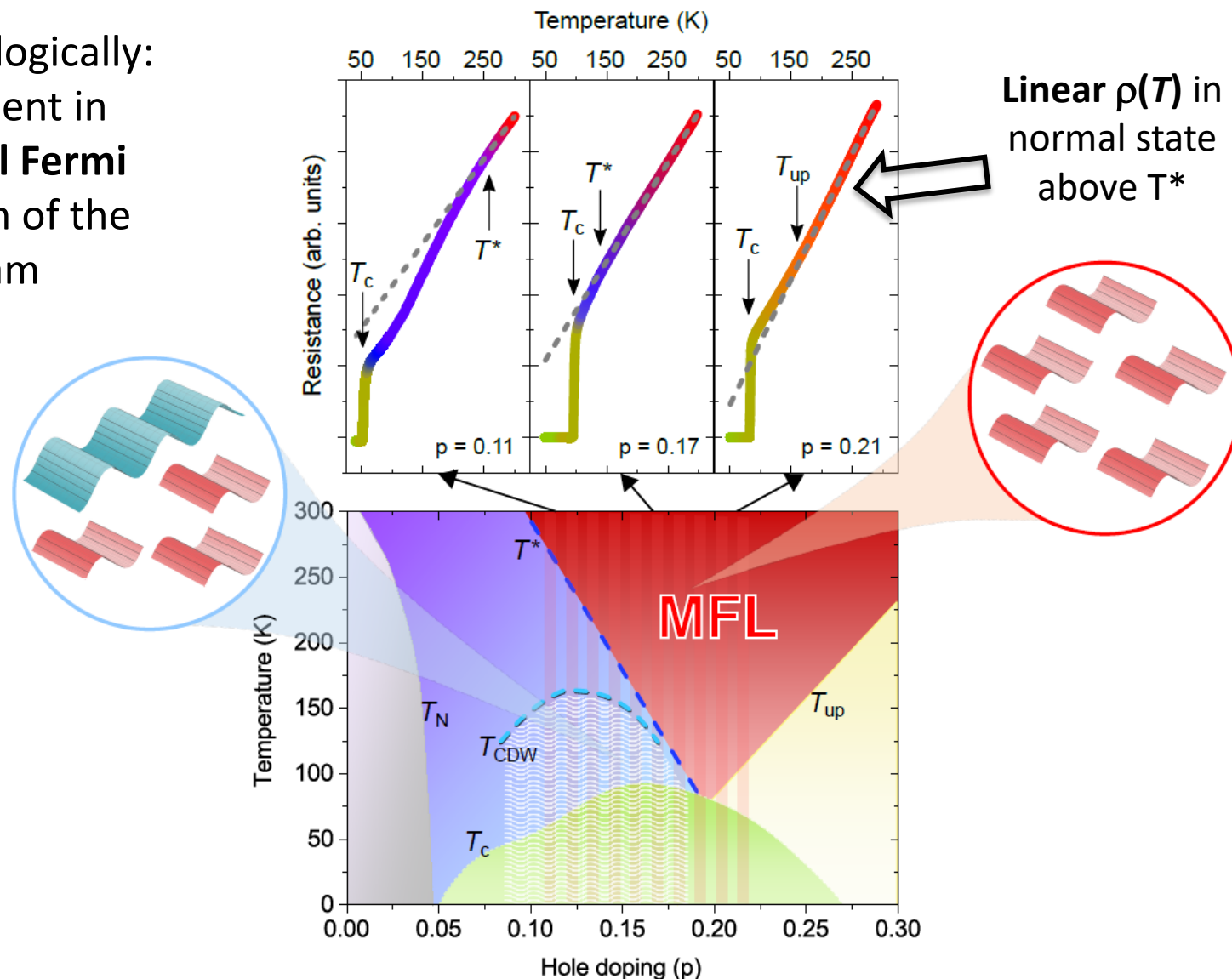
CDF and Marginal Fermi Liquid picture

CDW, T dependent

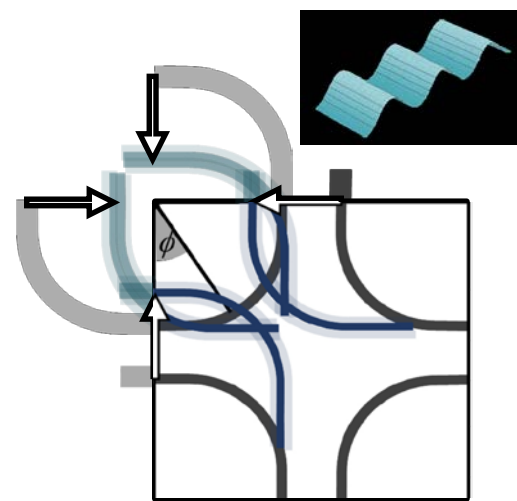


CDF,
quasi T independent

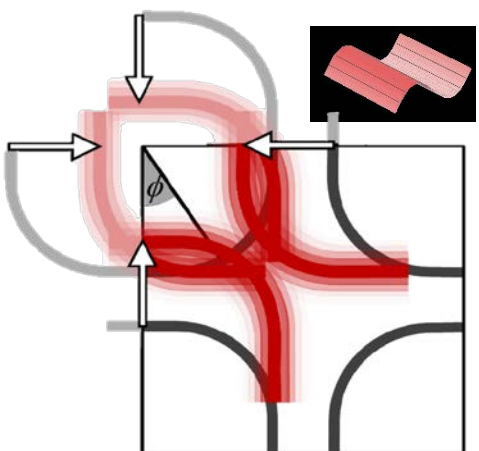
Phenomenologically:
CDF are present in
the **Marginal Fermi Liquid**
region of the
phase diagram



Scattering from CDF can lead to linear $\rho(T)$

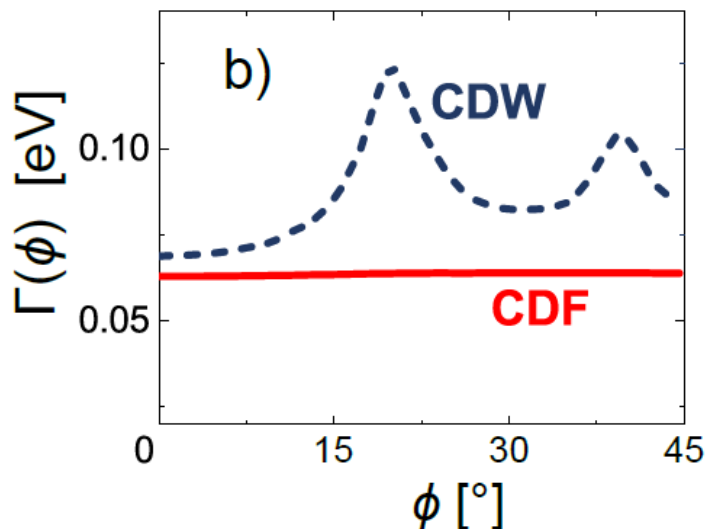


CDW scattering

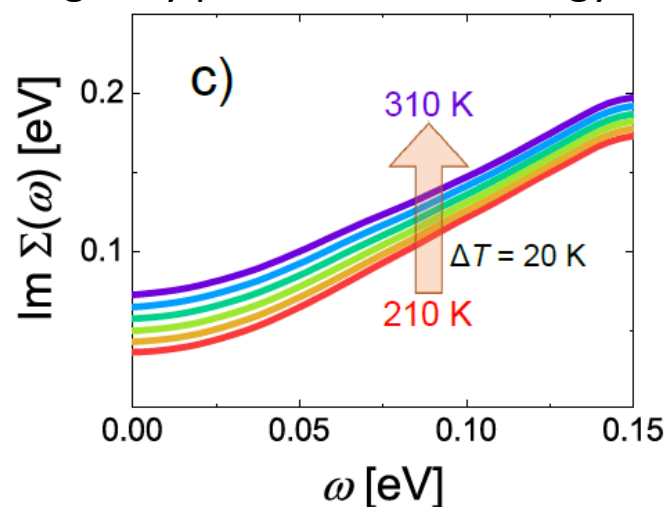


CDF scattering

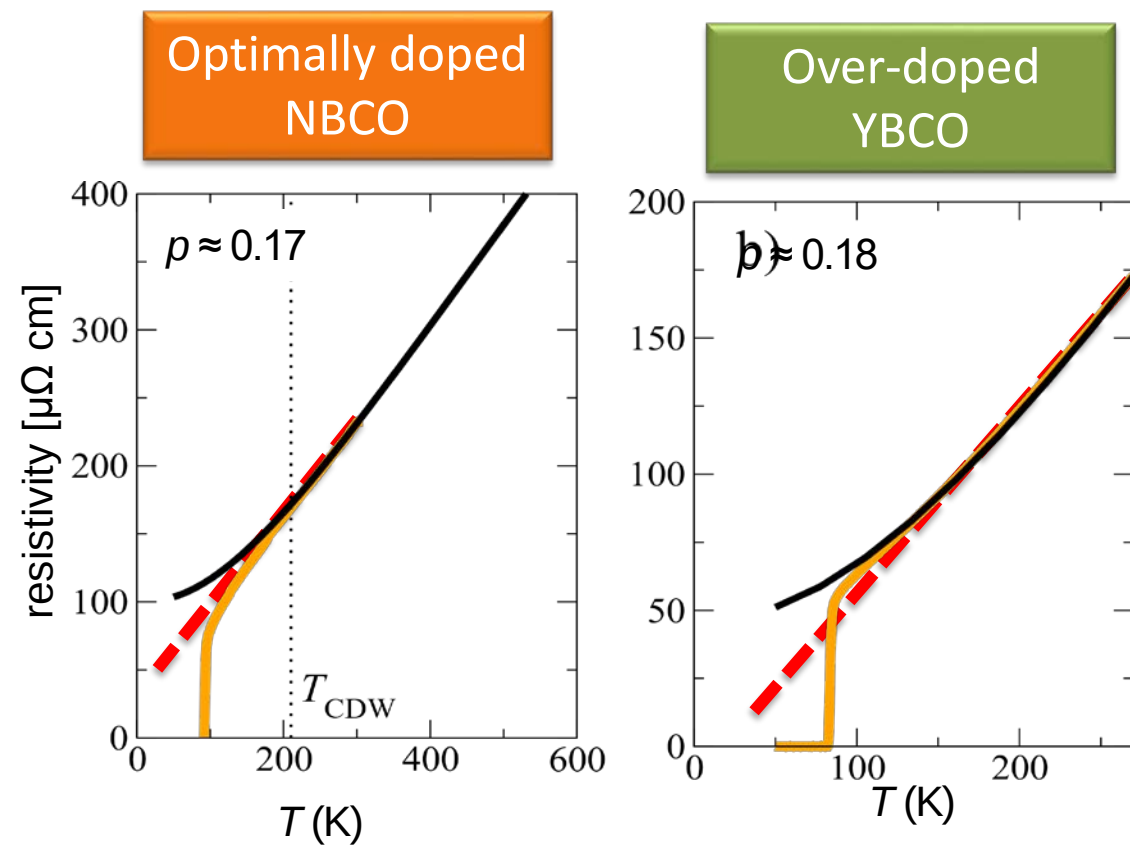
Scattering rate: $\Gamma = \text{Im } \Sigma(\phi, T, \omega = 0)$



Imaginary part of el. self-energy Σ

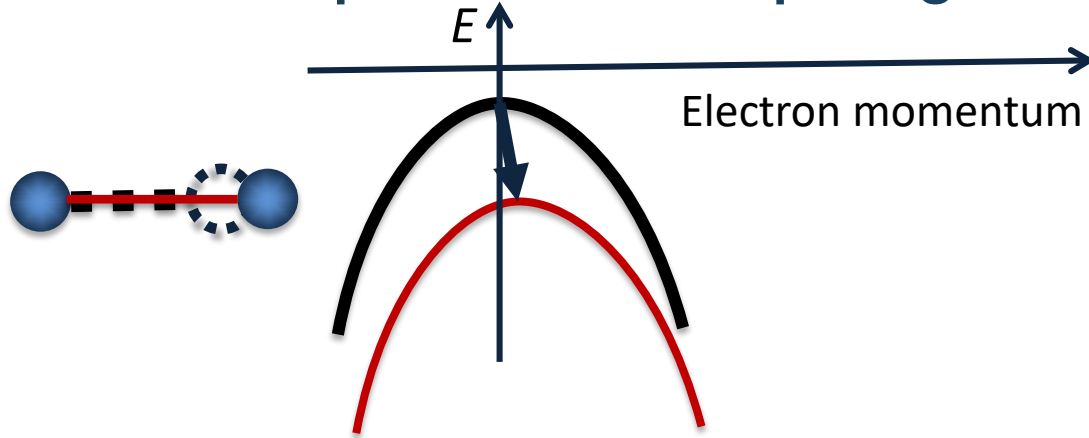


Resistivity of OP and OD samples is quantitatively reproduced



G. Seibold, R. Arpaia, Y.Y. Peng, R. Fumagalli, L. Braicovich, C. Di Castro, M. Grilli, G. Ghiringhelli, and S. Caprara, [arXiv:1905.10232](https://arxiv.org/abs/1905.10232)

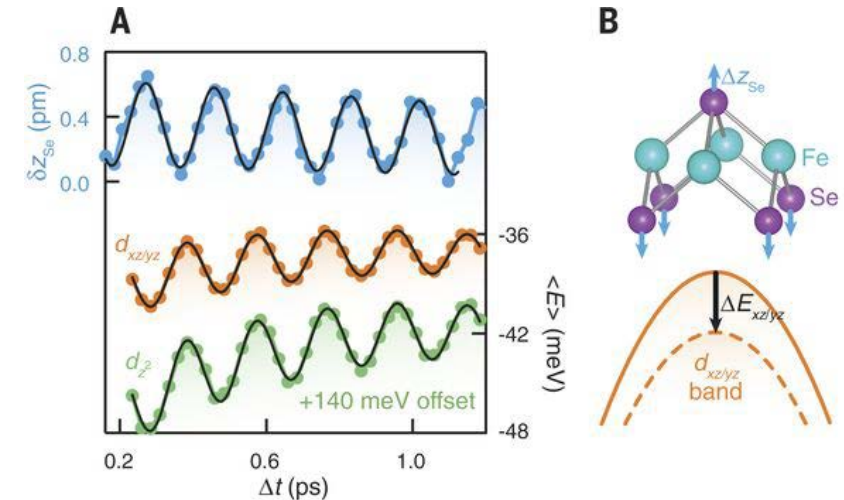
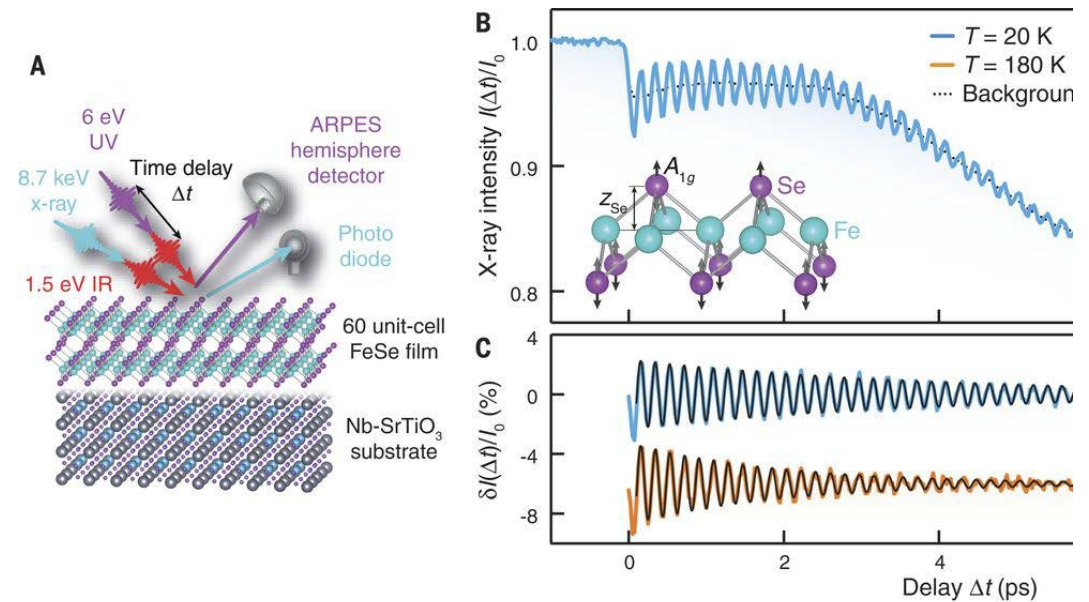
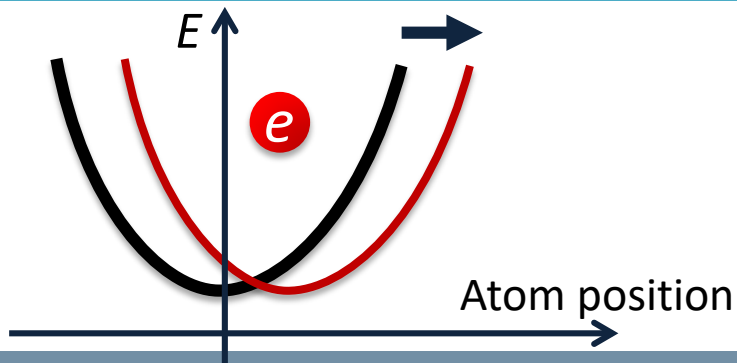
Electron phonon coupling



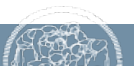
How $\mathcal{H}$ is modified by an atomic displacement

$\equiv$

How lattice is deformed by the presence of a valence electron

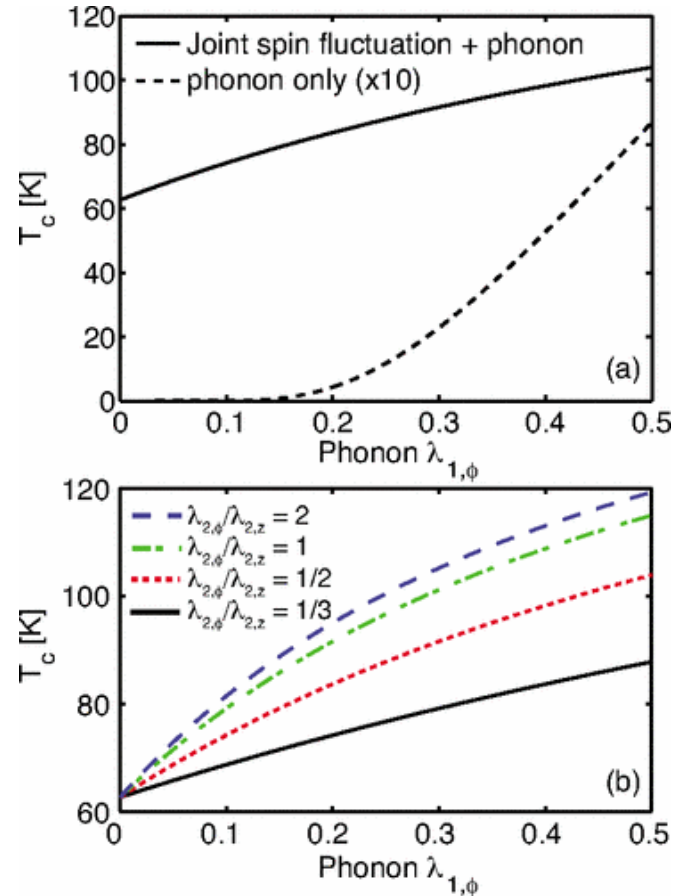


S. Gerber, S.-L. Yang, D. Zhu, H. Soifer, J. A. Sobota, S. Rebec, J. J. Lee, T. Jia, B. Moritz, C. Jia, A. Gauthier, Y. Li, D. Leuenberger, Y. Zhang, L. Chaix, W. Li, H. Jang, J.-S. Lee, M. Yi, G. L. Dakovski, S. Song, J. M. Glownia, S. Nelson, K. W. Kim, Y.-D. Chuang, Z. Hussain, R. G. Moore, T. P. Devereaux, W.-S. Lee, P. S. Kirchmann, and Z.-X. Shen, *Science* **357**, 71(2017)

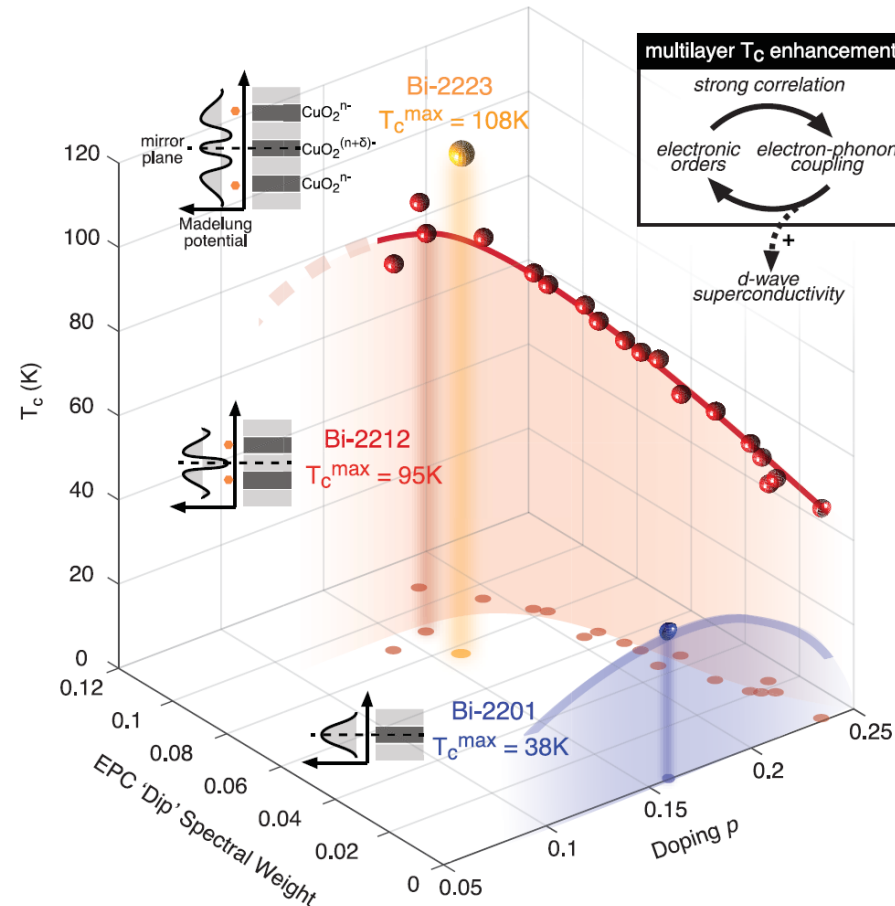


Electron phonon coupling (EPC) in cuprates

EPC: cooperative effect of phonons in SC pairing in cuprates

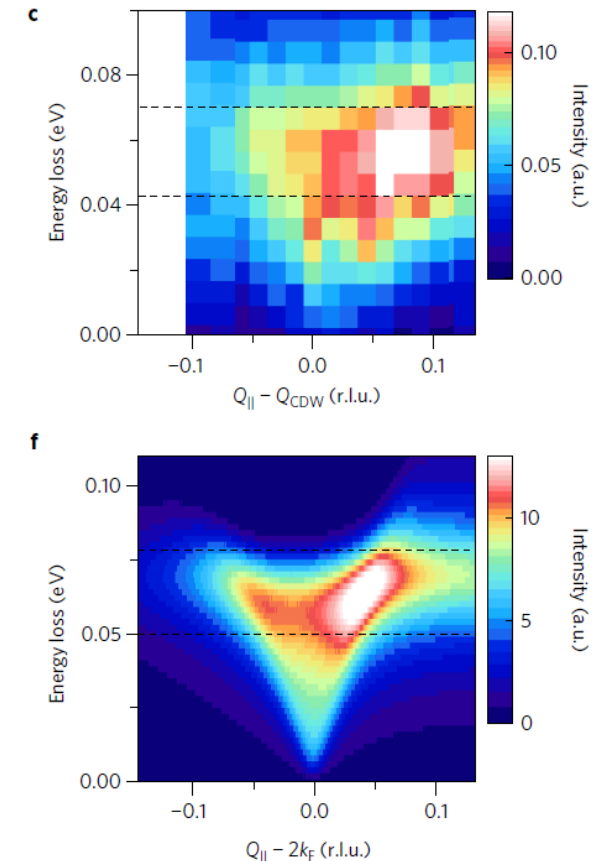


S. Johnston, F. Vernay, B. Moritz, Z.-X. Shen, N. Nagaosa, J. Zaanen, and T. P. Devereaux
Phys. Rev. B **82**, 064513 (2010)



Y. He, M. Hashimoto, D. Song, S.-D. Chen, J. He, I. M. Vishik, B. Moritz, D.-H. Lee, N. Nagaosa, J. Zaanen, T. P. Devereaux, Y. Yoshida, H. Eisaki, D. H. Lu, and Z.-X. Shen *Science* **362** 62 (2018)

CDW couple with some optical modes



L. Chaix, G. Ghiringhelli, Y. Y. Peng, M. Hashimoto, Y. He, S. Chen, K. Kummer, N.B. Brookes, B. Moritz, S. Ishida, Y. Yoshida, H. Eisaki, L. Braicovich, Z.-X. Shen, T. P. Devereaux, W.-S. Lee, *Nature Physics* **13**, 952 (2017)

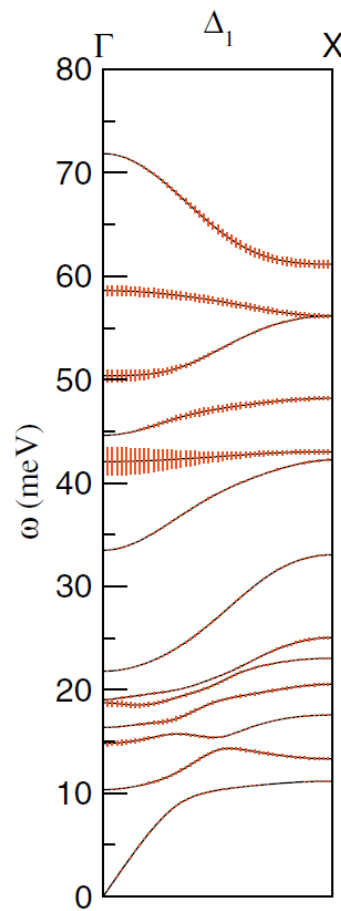
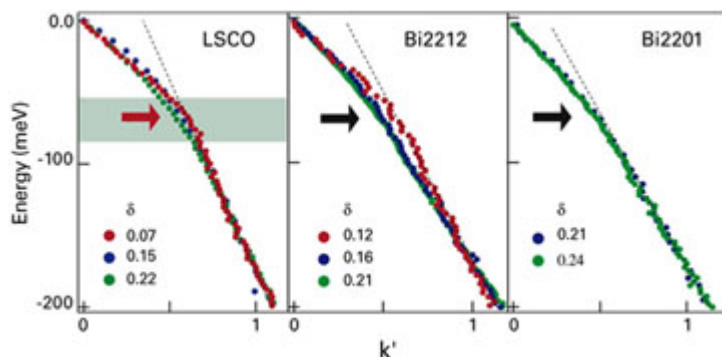
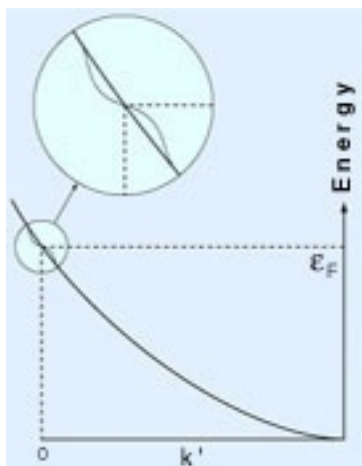
Measuring EPC with RIXS

Large parameter space: electron $\mathbf{k}$, phonon $(\omega, \mathbf{q})$

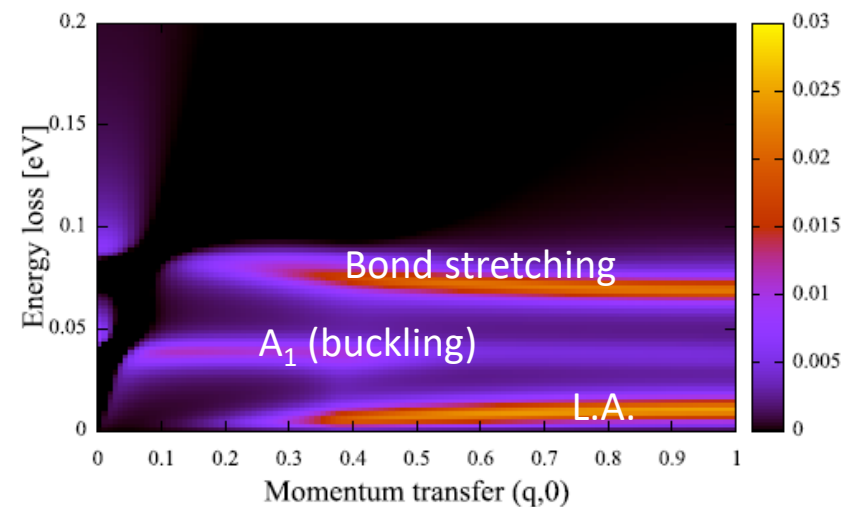
ARPES: kinks related to EPS
 $\mathbf{k}$ and ω can be obtained
 $\mathbf{q}$ is unknown

INS: linewidth related to EPC
 $\mathbf{k}$ is unknown
 ω and $\mathbf{q}$ are measured

RIXS: ph peak intensity prop. to EPC
 $\mathbf{k}$ is partly unknown
 ω and $\mathbf{q}$ are measured



Theoretical phonon intensity in RIXS



L. Pintschovius, *phys. stat. sol. (b)* **242**, 30 (2005)

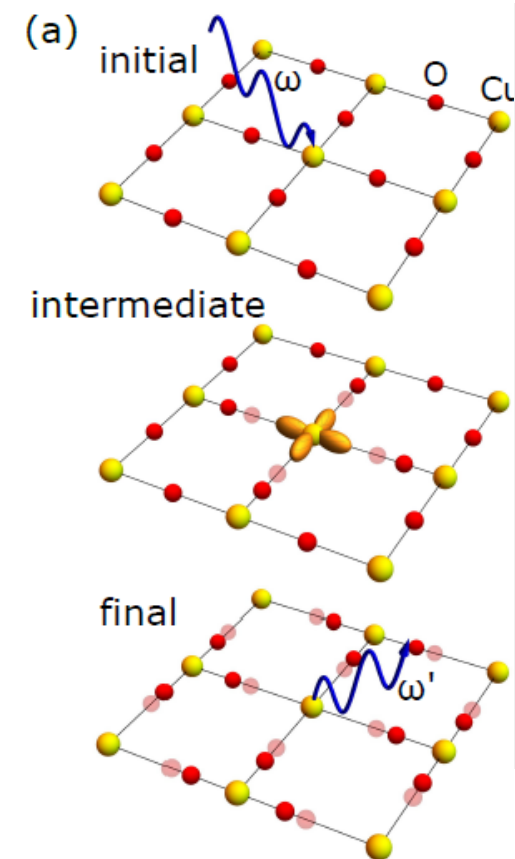
T. P. Devereaux, A. M. Shvaika, K. Wu, K. Wohlfeld, C.-J. Jia, Y. Wang, B. Moritz, L. Chaix, W.-S. Lee, Z.-X. Shen, G. Ghiringhelli, and L. Braicovich, *PRX* **6**, 041019 (2016)

Measuring EPC with RIXS: mechanism

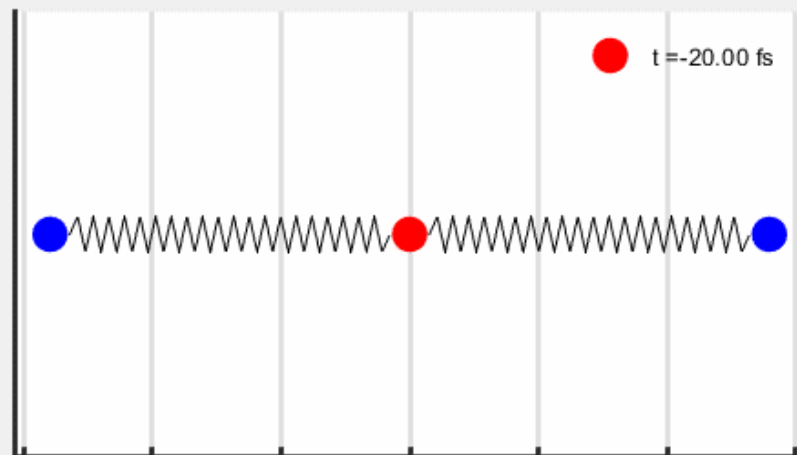
Theory by Ament, van den Brink

Phonon peak intensity grows with:

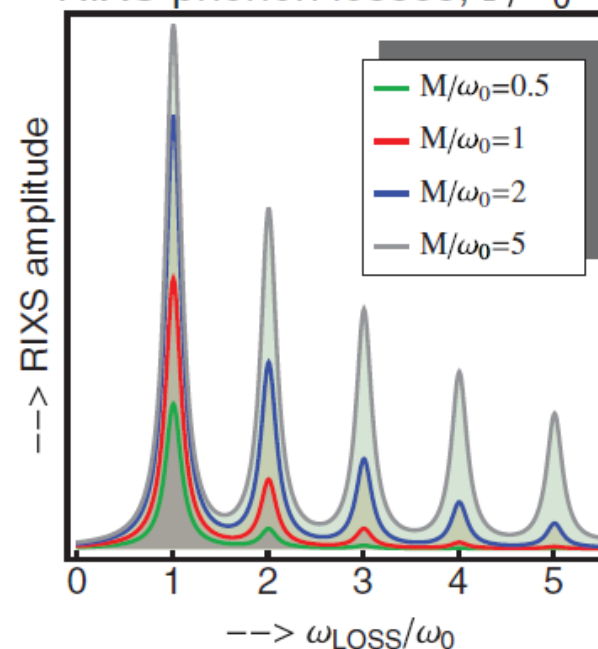
- EPC, i.e., M or $g = (M/\omega_0)^2$
 - Intermediate state lifetime
- 2ph, 3ph peaks*



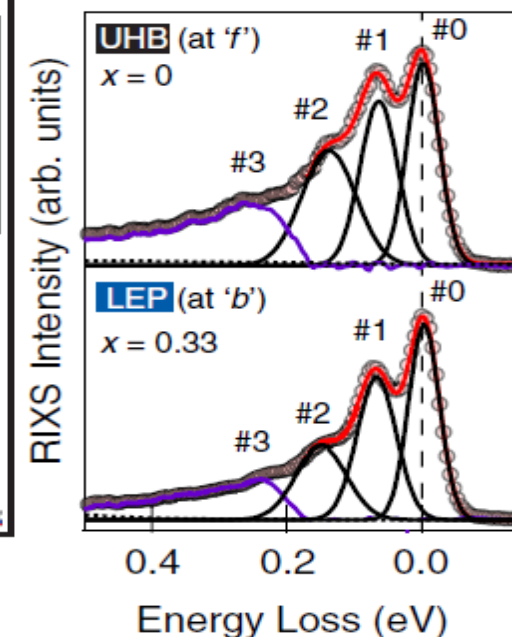
Harmonic oscillator analogy



RIXS phonon losses, $\Gamma/\omega_0=5$



Application to 1D system (O K edge)



Matteo Rossi, Riccardo Arpaia, Roberto Fumagalli, Marco Moretti Sala, Davide Betto, Gabriella M. De Luca, Kurt Kummer, Jeroen van den Brink, Marco Salluzzo, Nicholas B. Brookes, Lucio Braicovich, Giacomo Ghiringhelli, [arXiv:1902.09163](https://arxiv.org/abs/1902.09163), to appear in PRL

L. J. P. Ament, M. van Veenendaal and J. van den Brink, [EPL 95 \(2011\) 27008](https://doi.org/10.1063/1.3600000)

W-S Lee et al, [PRL 110, 265502 \(2013\)](https://doi.org/10.1126/science.1228888)

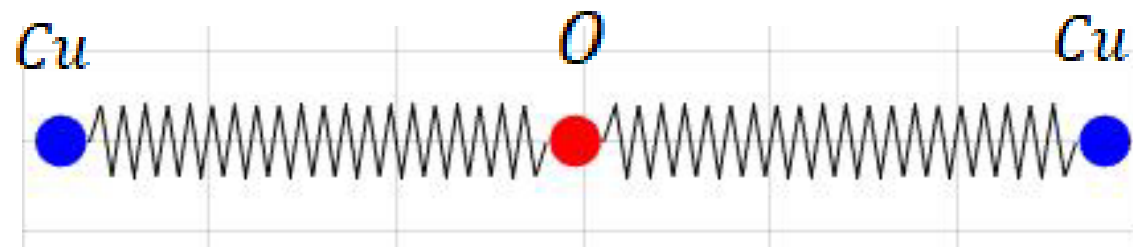


Classical model of EPC in RIXS

Ground state

$$M\ddot{x} = +k_0(\Delta x - x) - k_0(\Delta x + x)$$

Δx is the pre-deformation of springs: irrelevant in ground state, relevant in intermediate state

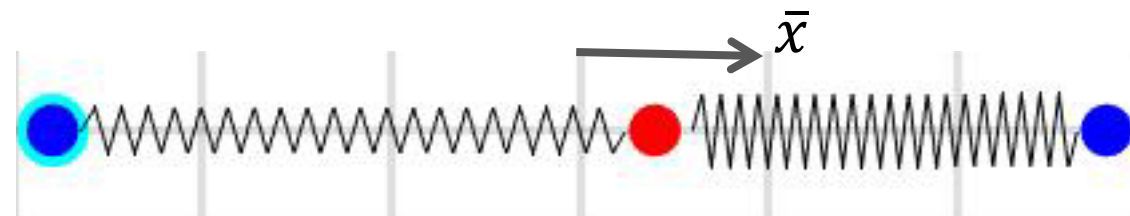


$$\omega_0 = \sqrt{\frac{2k_0}{M}}$$

Intermediate state

New equilibrium position $\bar{x} = \frac{k_{ep}}{2k_0 - k_{ep}} \Delta x$

$$M\ddot{x} = +k_0(\Delta x - x) - (k_0 - k_{ep})(\Delta x + x)$$

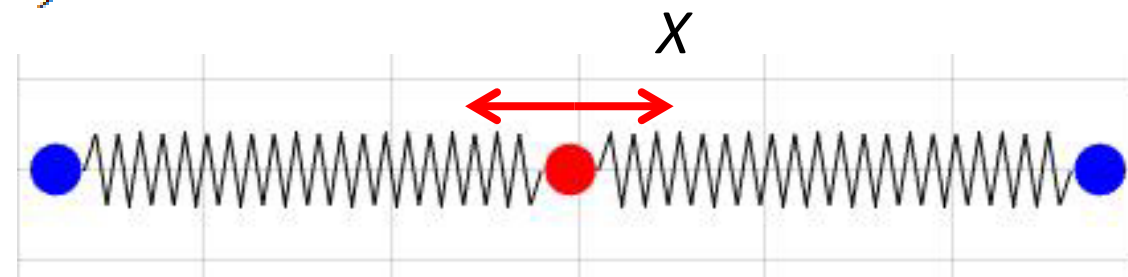


$$\omega_1 = \sqrt{\frac{2k_0 - k_{ep}}{M}}$$

Final state

$$x(t) = X \sin(\omega_0 t + \phi)$$

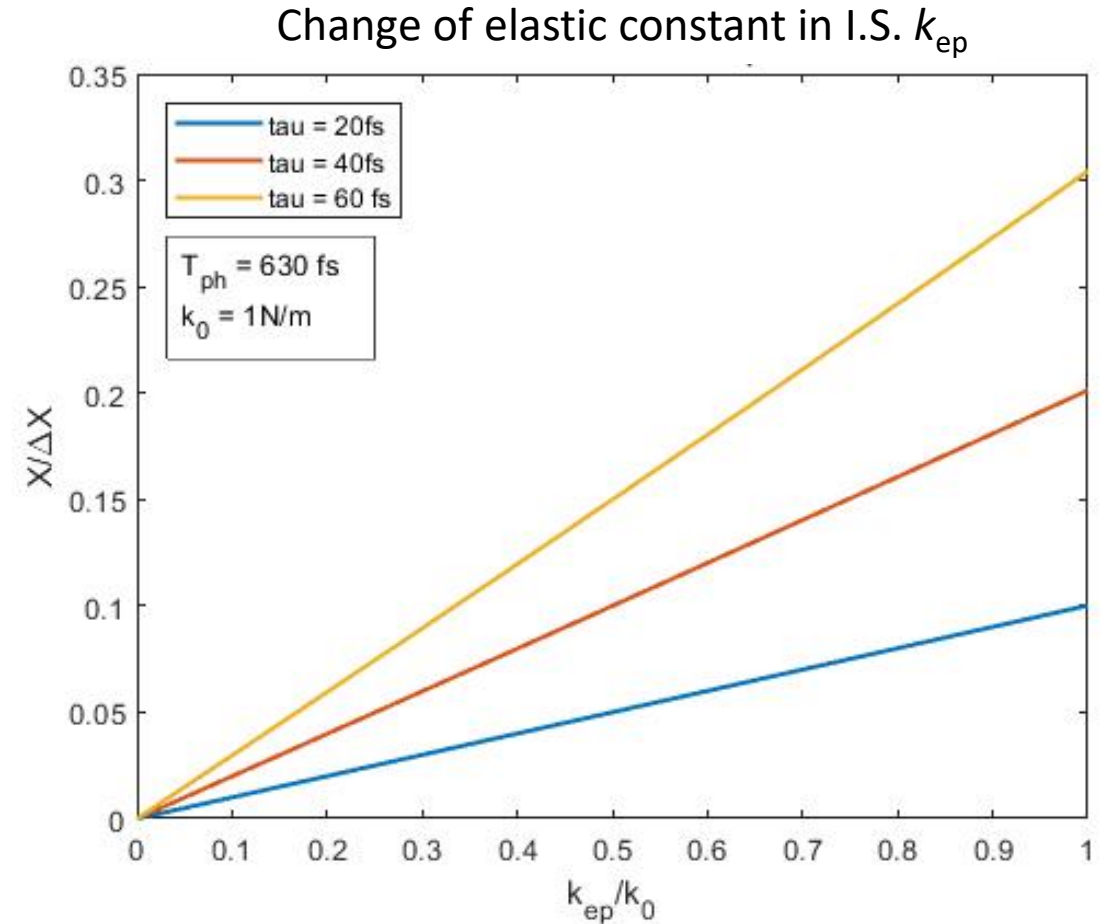
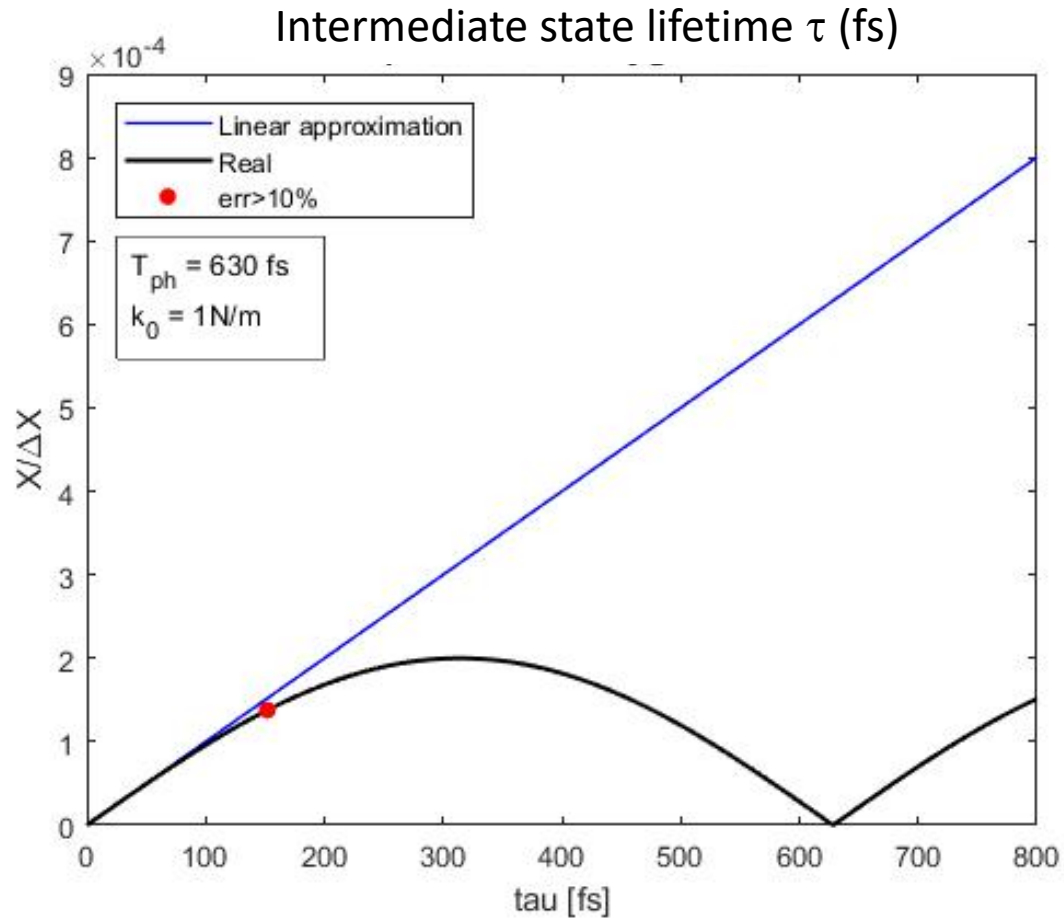
$$X^2 = \bar{x}^2 \left(\left(\frac{\omega_1}{\omega_0} \right)^2 \sin^2(\omega_1 \tau) + (1 - \cos(\omega_1 \tau))^2 \right)$$



$$\omega_0 = \sqrt{\frac{2k_0}{M}}$$

$$\frac{X}{\Delta X} = \frac{k_{ep}}{2k_0 - k_{ep}} \sqrt{\left(\left(\frac{\omega_1}{\omega_0} \right)^2 \sin^2(\omega_1 \tau) + (1 - \cos(\omega_1 \tau))^2 \right)}$$

Trends in classical oscillator model



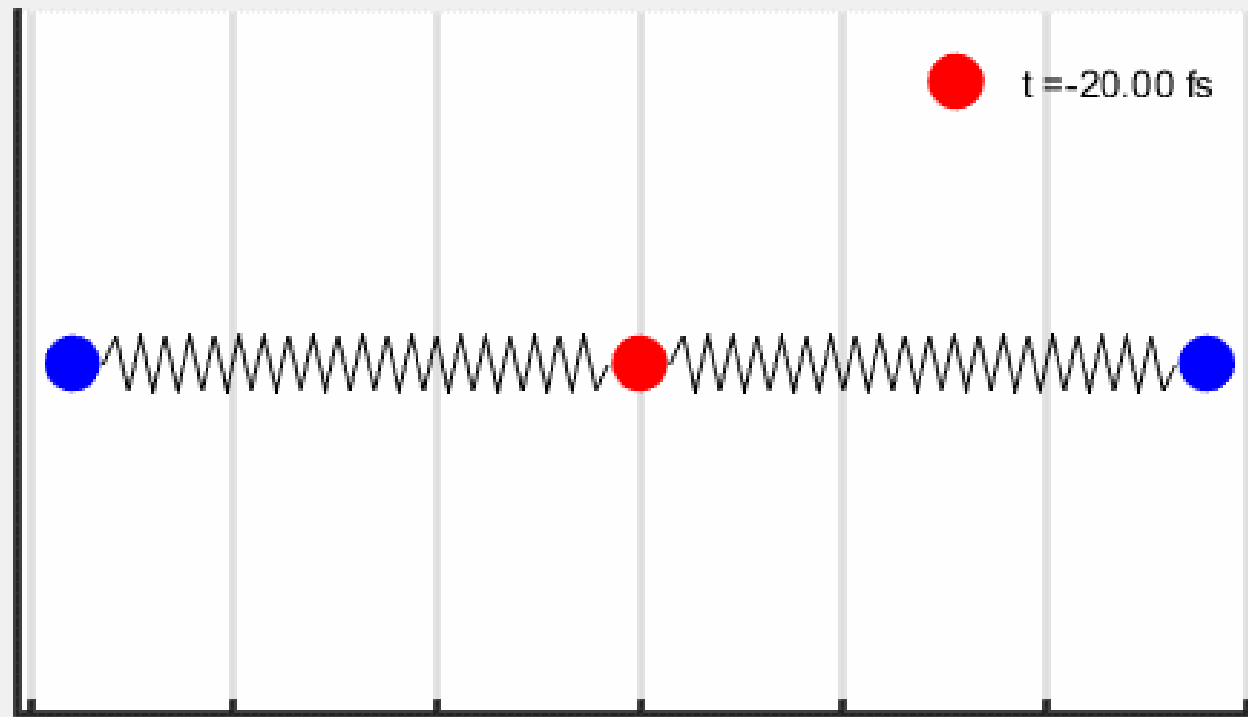
Amplitude of oscillations:

$$\frac{X}{\Delta X} = \frac{k_{ep}}{2k_0 - k_{ep}} \sqrt{\left(\left(\frac{\omega_1}{\omega_0} \right)^2 \sin^2(\omega_1 \tau) + (1 - \cos(\omega_1 \tau))^2 \right)}$$



The movie

Classical harmonic oscillator analogy

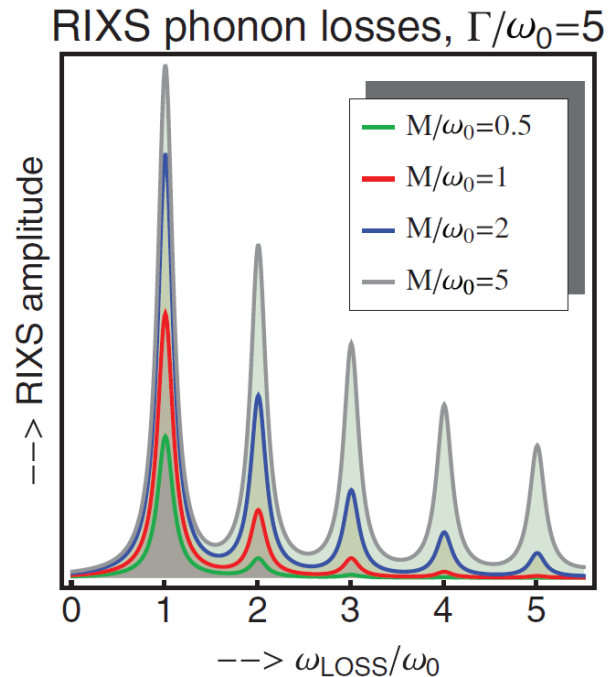


Leonardo Martinelli, GG et al



Determining the electron-phonon coupling strength from Resonant Inelastic X-ray Scattering at transition metal L-edges

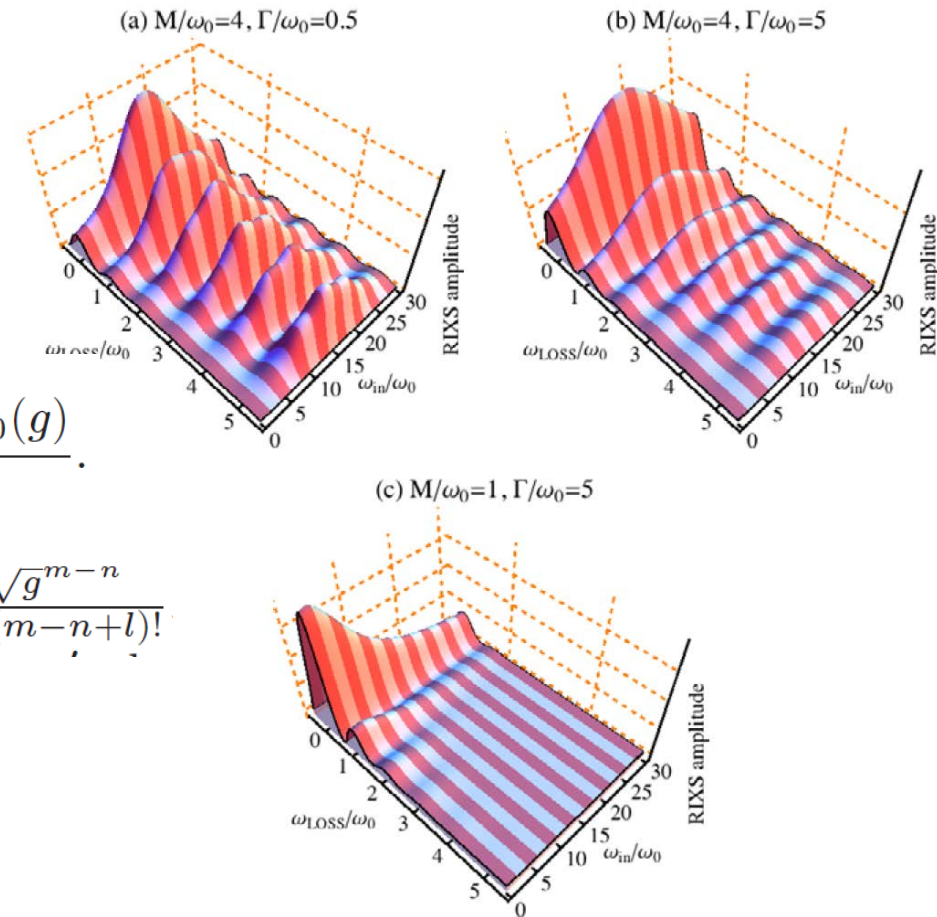
L. J. P. AMENT^{1,2}, M. VAN VEENENDAAL^{3,4} and J. VAN DEN BRINK²



$$A_{\mathbf{q}}^{\text{Einstein}} = \sum_{n=0}^{\infty} \frac{B_{\max(n',n),\min(n',n)}(g) B_{n0}(g)}{z + (g - n)\omega_0}.$$

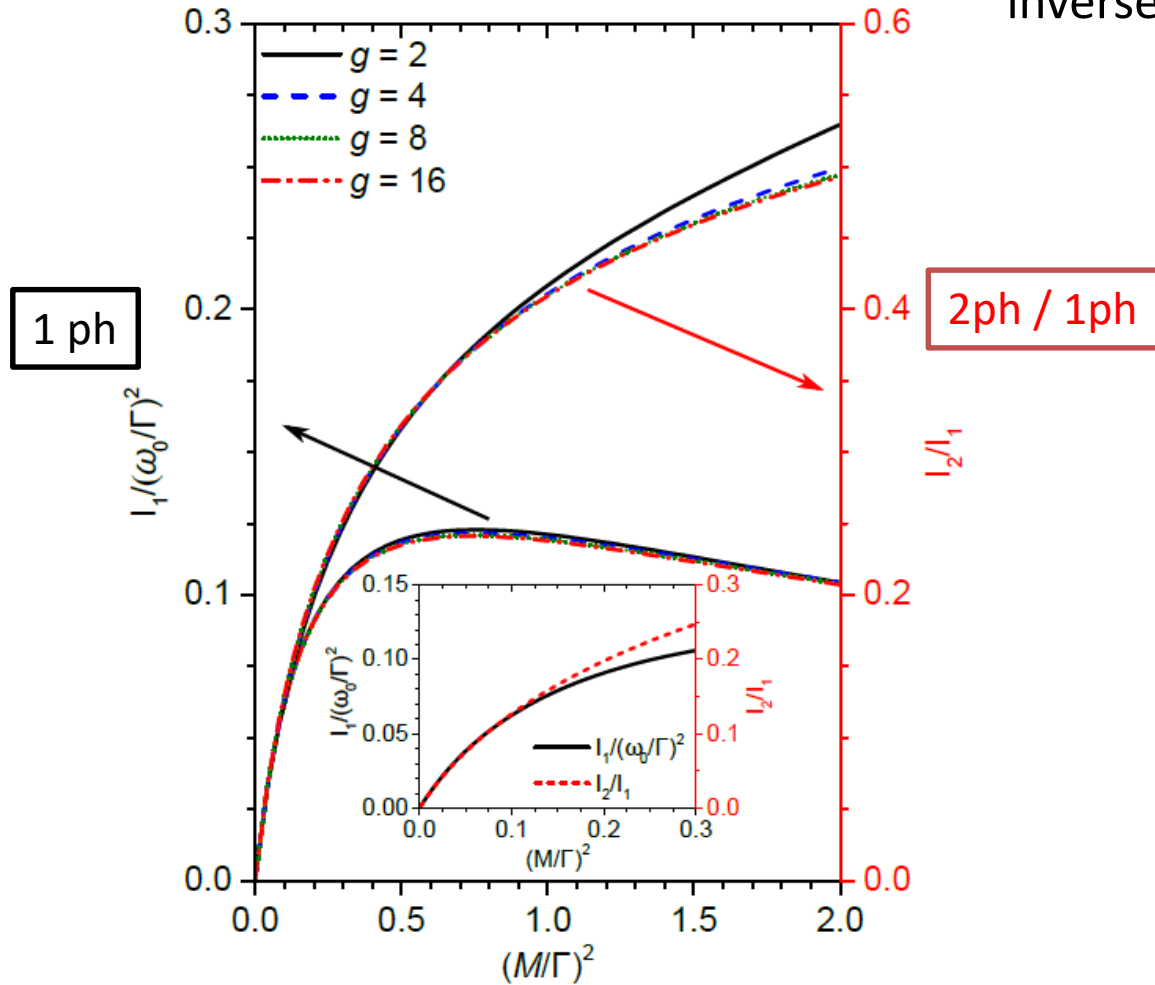
$$B_{mn}(g) = (-1)^m \sqrt{e^{-g} m! n!} \sum_{l=0}^n \frac{(-g)^l \sqrt{g^{m-n}}}{(n-l)! l! (m-n+l)!}.$$

$$g = M^2 / \omega_0^2.$$

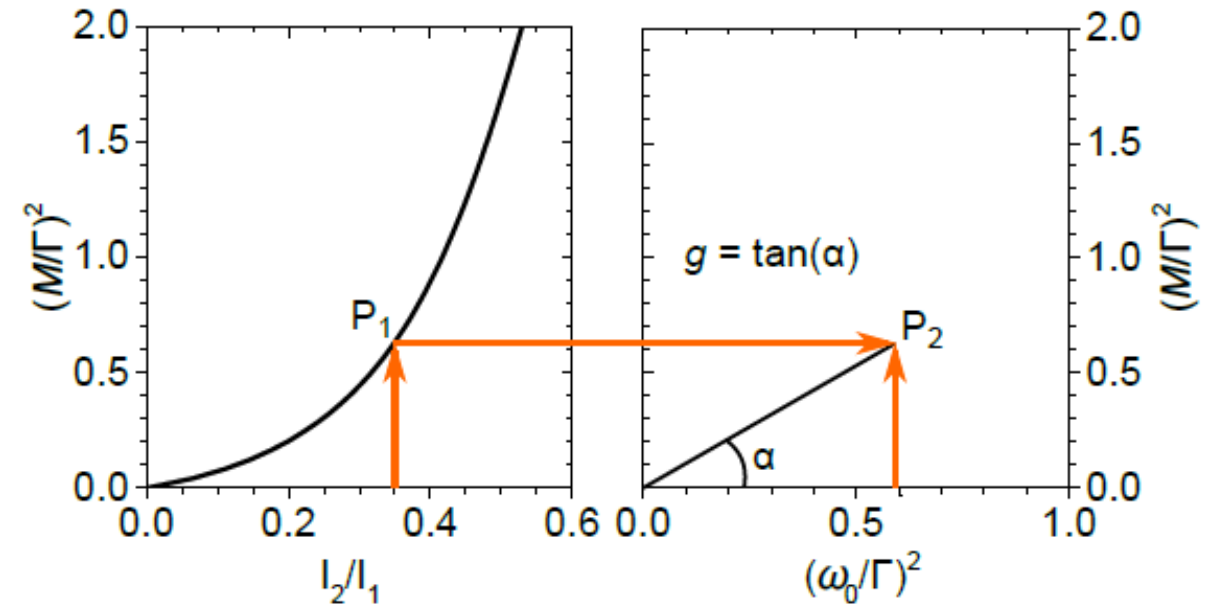


Scaling laws

We scale all parameter on Γ , the inverse of intermediate state lifetime



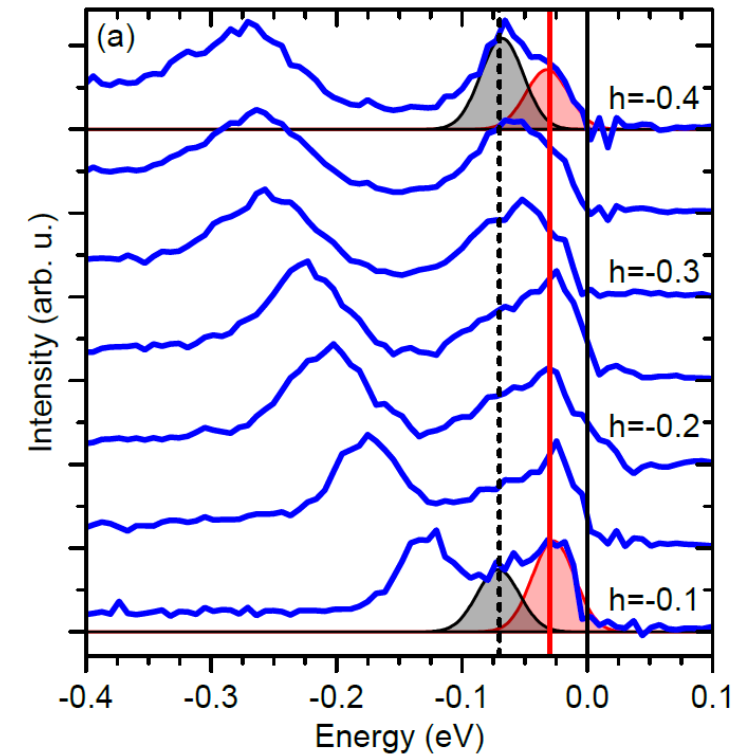
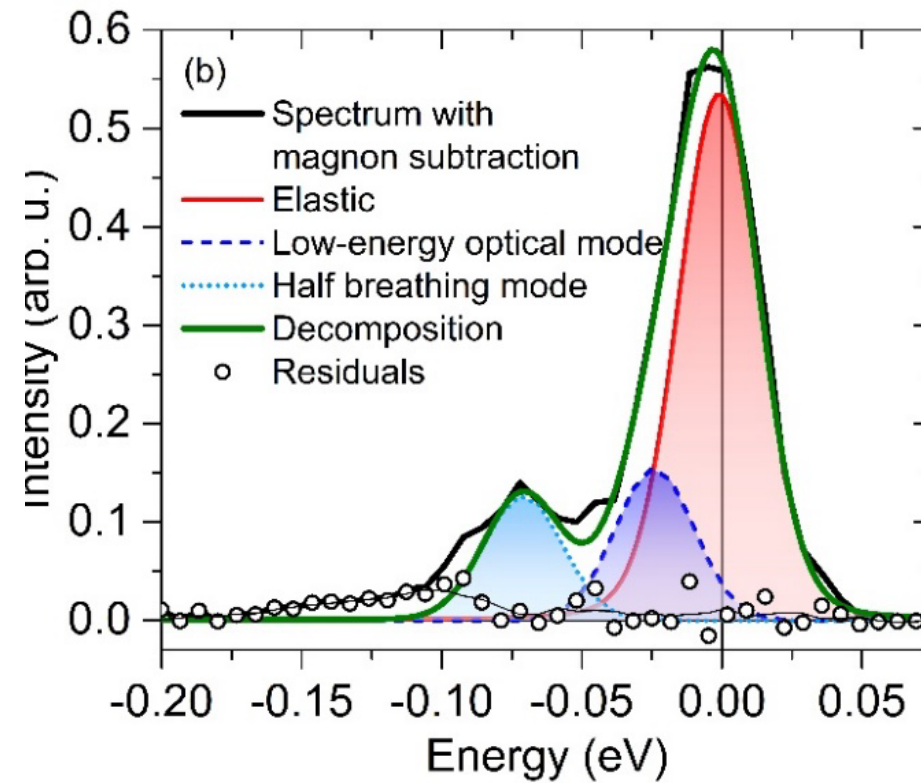
If one can measure I_2/I_1 , one can get $g = M^2/\omega_0^2$ by knowing ω_0 and Γ



Lucio Braicovich, Matteo Rossi, Roberto Fumagalli, Yingying Peng, Yan Wang, Riccardo Arpaia, Davide Betto, Gabriella M. De Luca, Daniele Di Castro, Kurt Kummer, Marco Moretti Sala, Mattia Pagetti, Giuseppe Balestrino, Nicholas B. Brookes, Marco Salluzzo, Steven Johnston, Jeroen van den Brink, Giacomo Ghiringhelli, [arXiv:1906.01270](https://arxiv.org/abs/1906.01270)



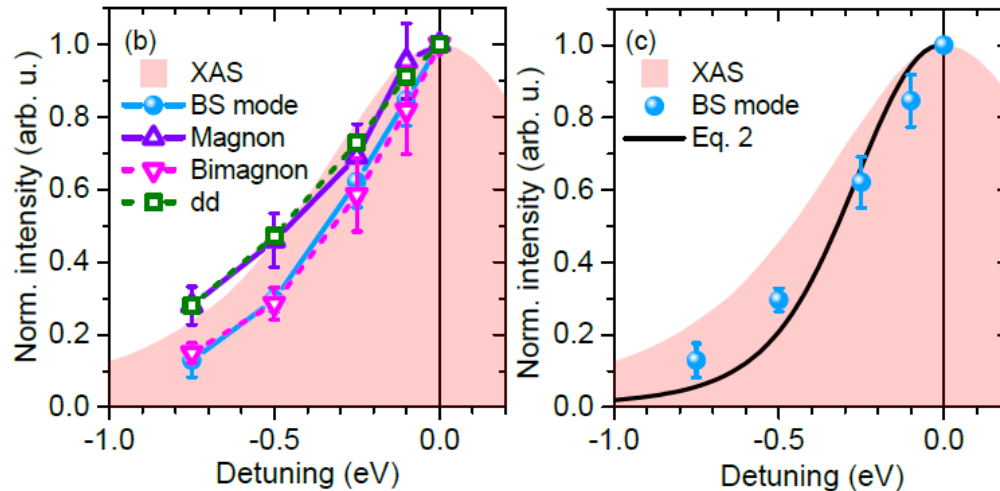
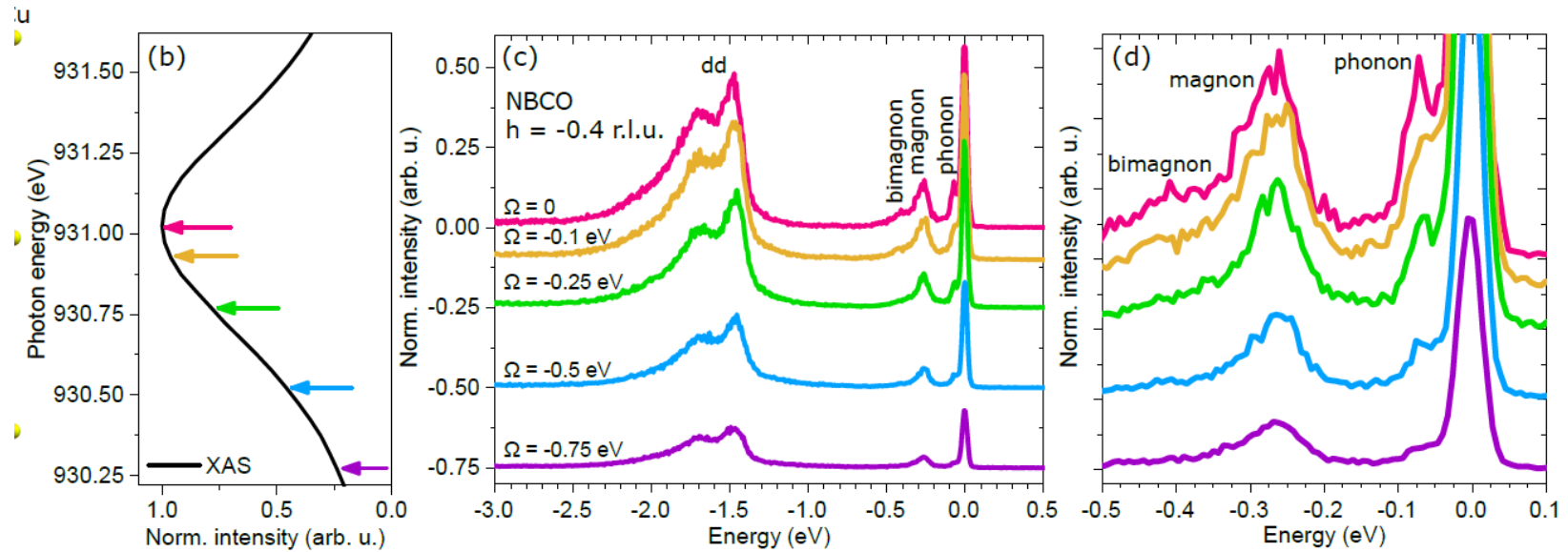
At Cu L₃ it does not work directly: no *2ph* peak



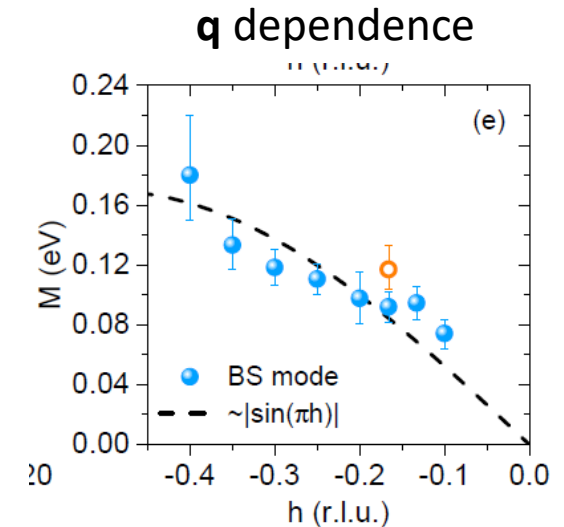
We need alternative methods to measure M

New method to obtain M from RIXS data: detuning

By detuning the incident x-ray phonon energy with respect to max of XAS, the effective lifetime decreases, reducing the phonon peak intensity more than XAS is reduced.



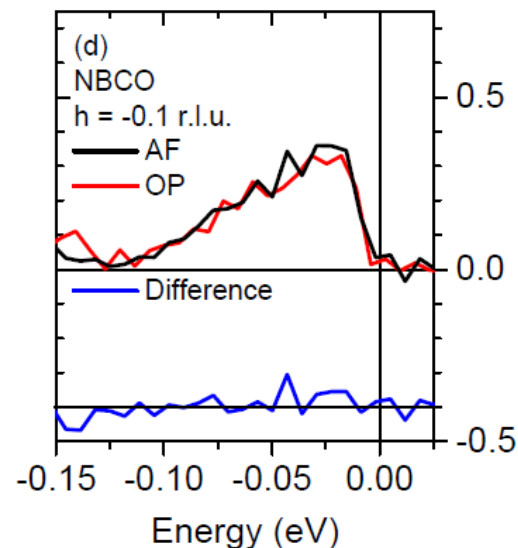
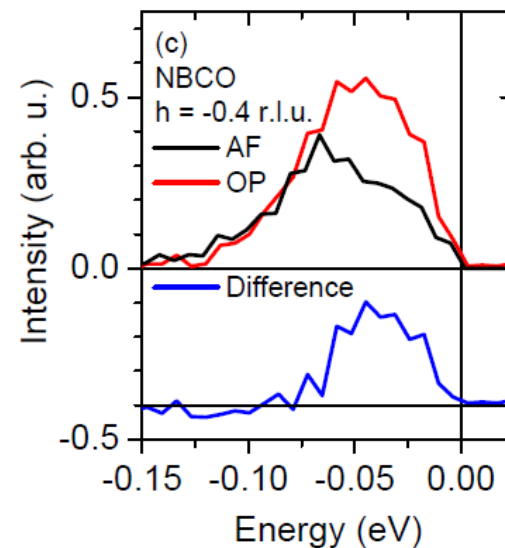
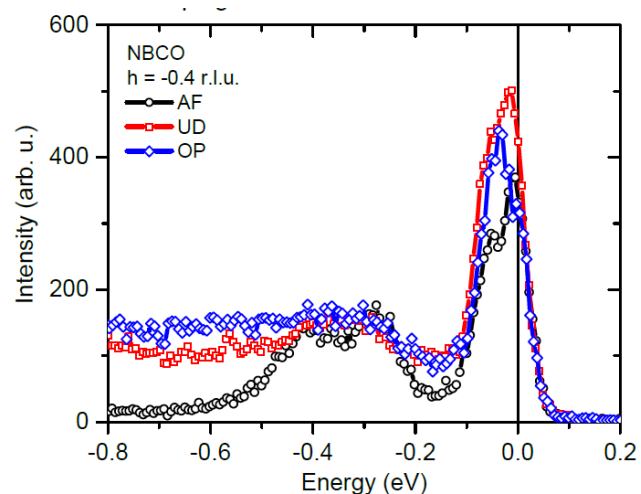
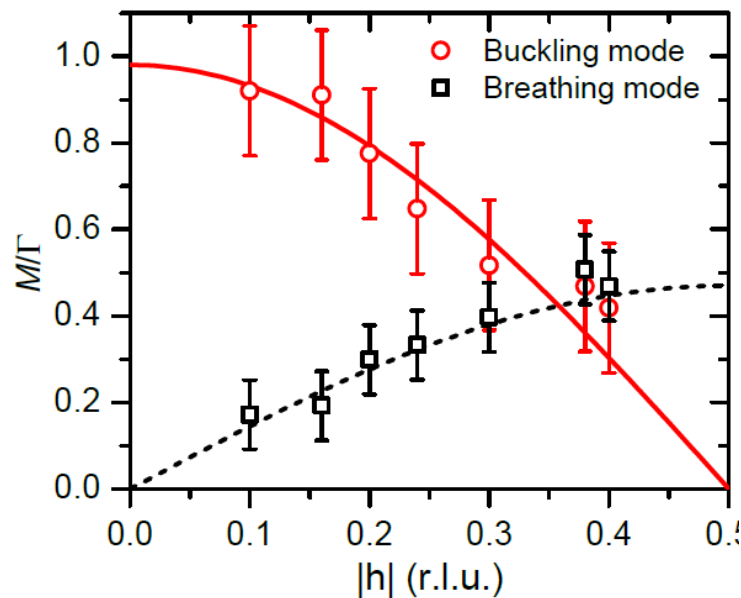
Fitting to simple equation allows estimate of $M = 160$ meV at $(0.40, 0, L)$



EPC in NBCO

Doping dependence: interplay with CDW at large q in (10)

q dependence



Lucio Braicovich, Matteo Rossi, Roberto Fumagalli, Yingying Peng, Yan Wang, Riccardo Arpaia, Davide Betto, Gabriella M. De Luca, Daniele Di Castro, Kurt Kummer, Marco Moretti Sala, Mattia Pagetti, Giuseppe Balestrino, Nicholas B. Brookes, Marco Salluzzo, Steven Johnston, Jeroen van den Brink, Giacomo Ghiringhelli, [arXiv:1906.01270](https://arxiv.org/abs/1906.01270)

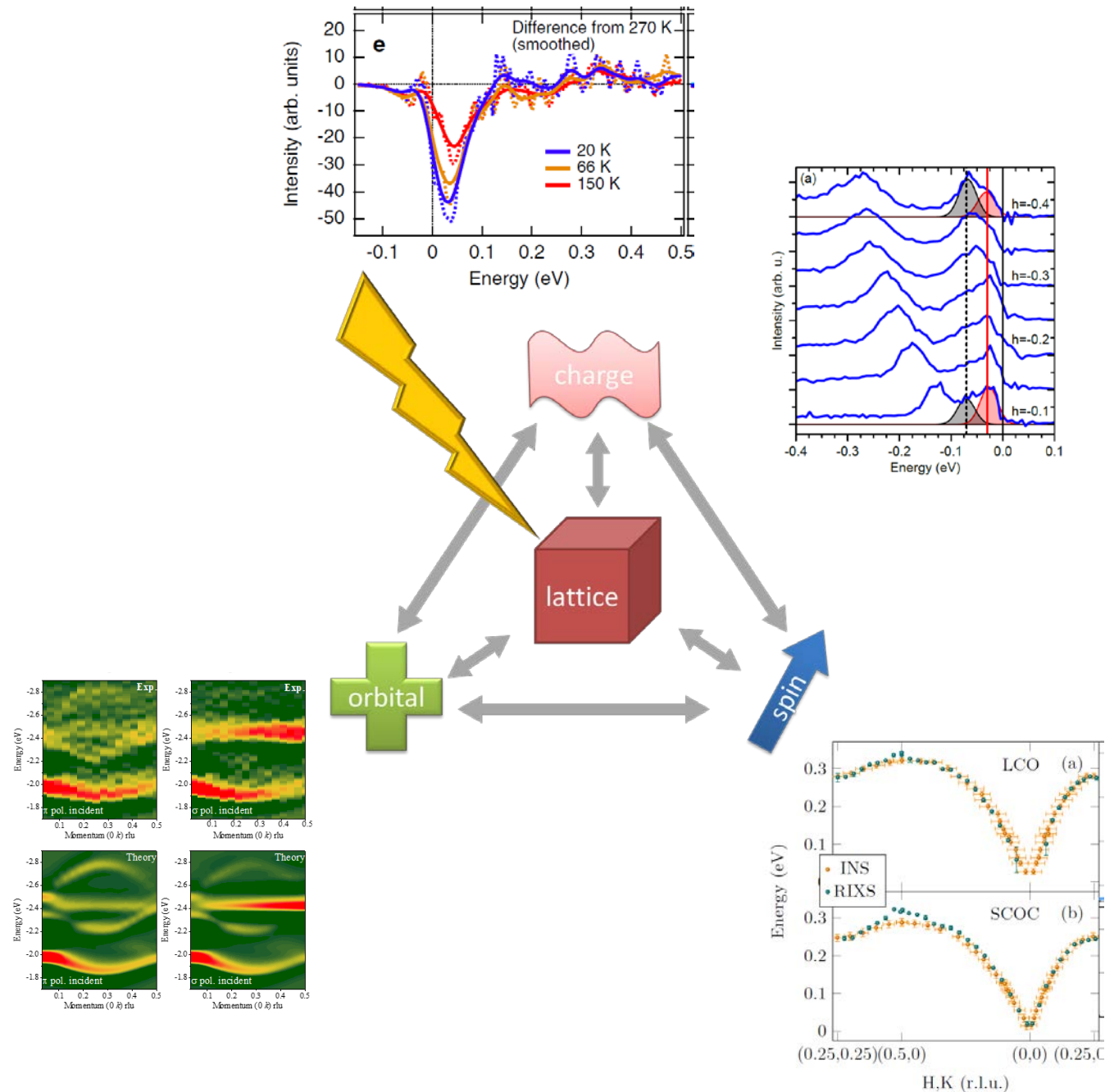


Conclusions

RIXS can really measure spin, charge, orbital and lattice excitations

Better resolution and higher efficiency still needed

Adding the time variable will be challenging but useful to further probe interconnections



Fluctuating CDW might provide unified picture

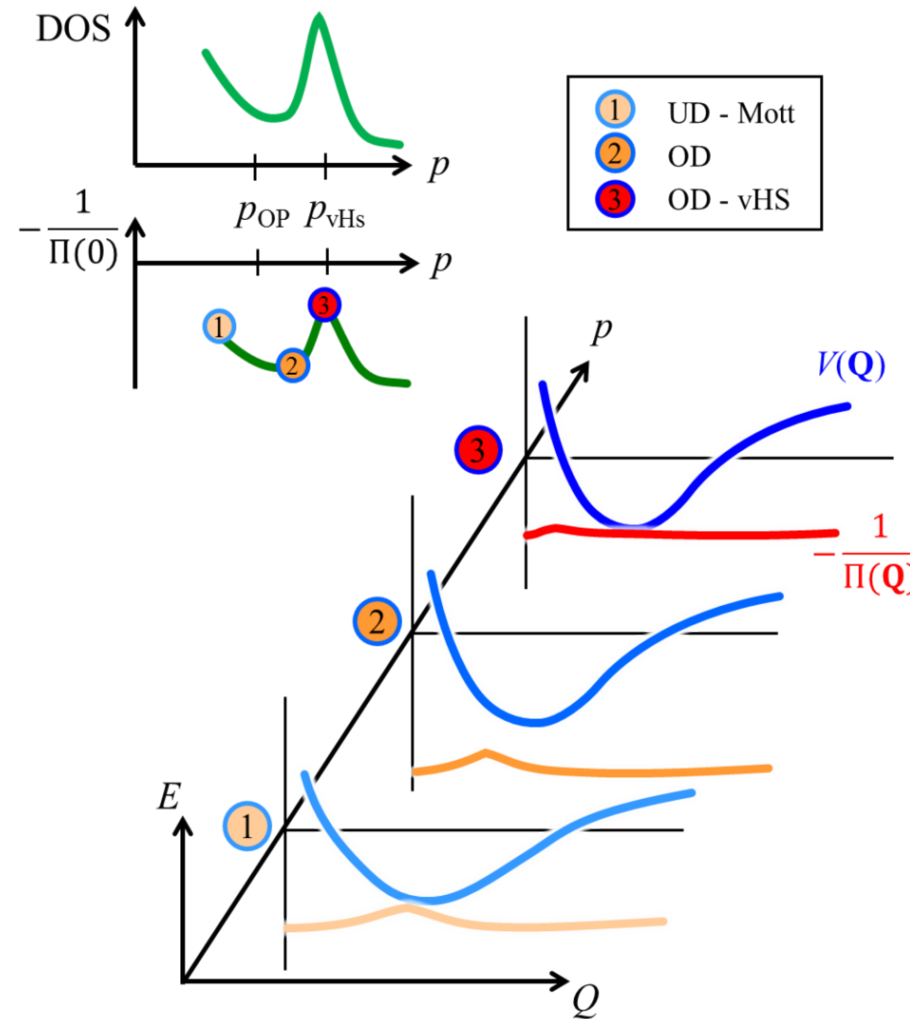
Fermi liquid, RPA:

$$\chi(\mathbf{Q}) = \frac{\Pi(\mathbf{Q})}{1 + V(\mathbf{Q})\Pi(\mathbf{Q})}$$

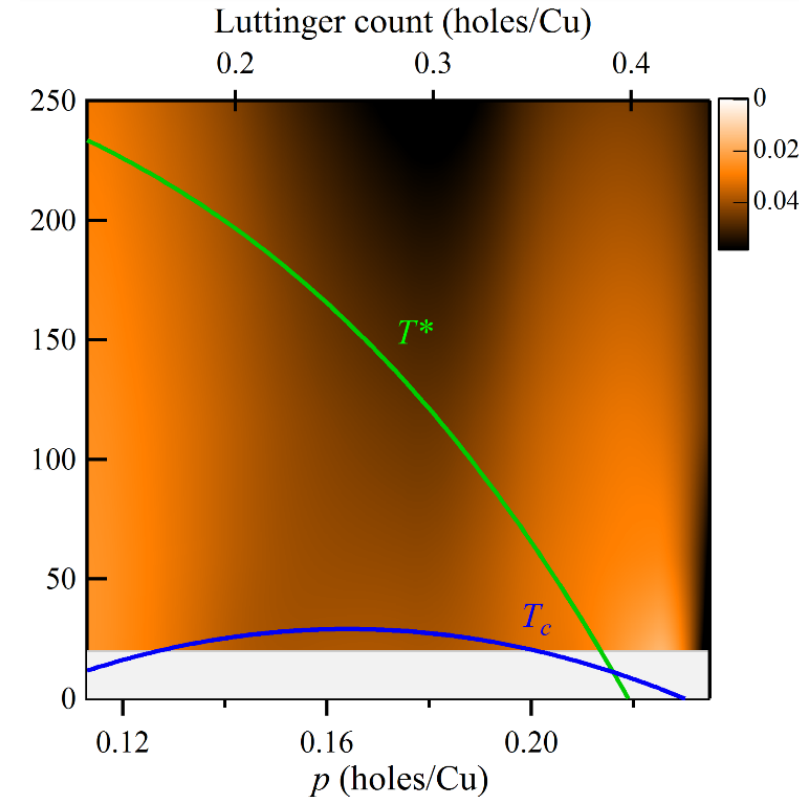
$$V(\mathbf{Q}_{co}) = -\frac{1}{\Pi(\mathbf{Q}_{co})}$$



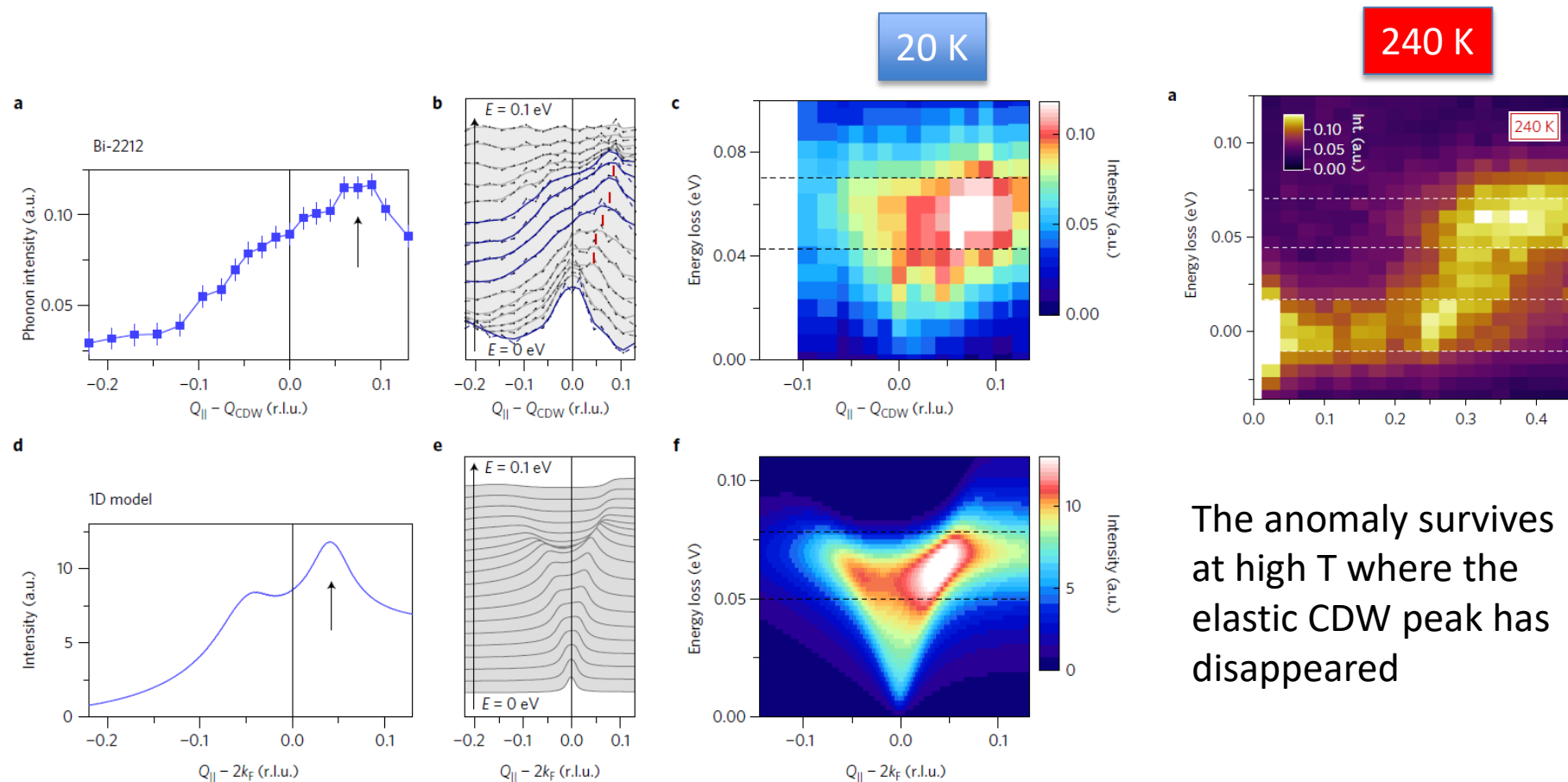
$$\Pi(0) \propto \text{DOS}$$



The fluctuating CDW scenario seems compatible with our discovery

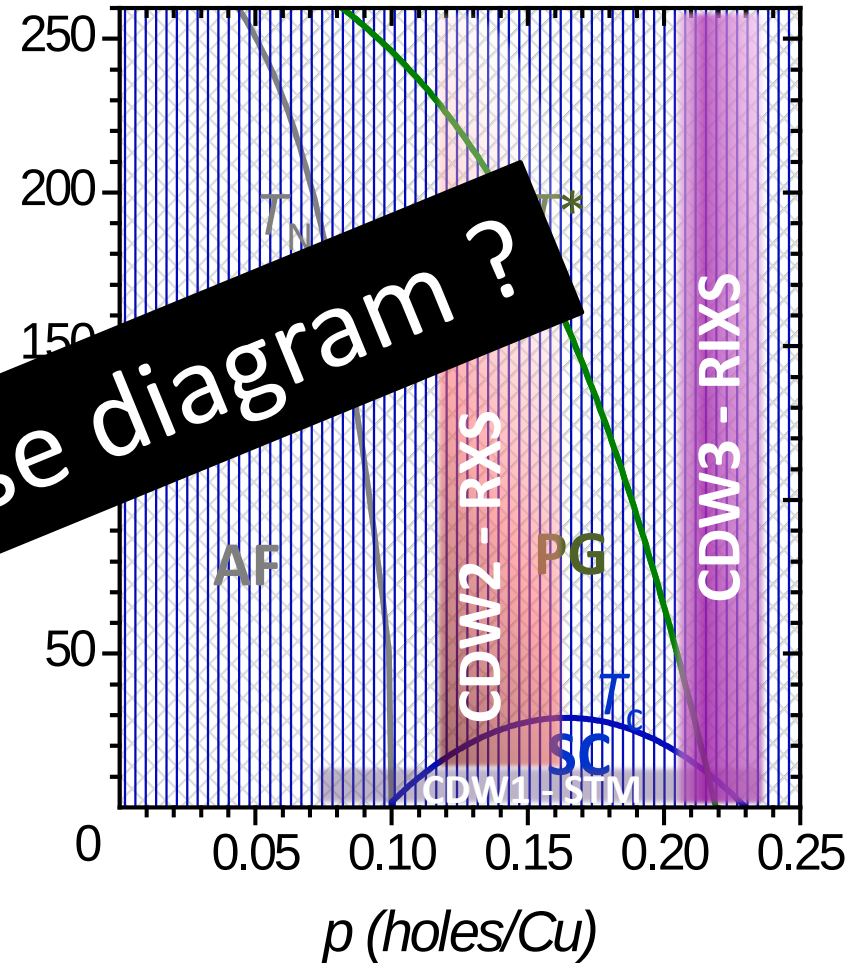
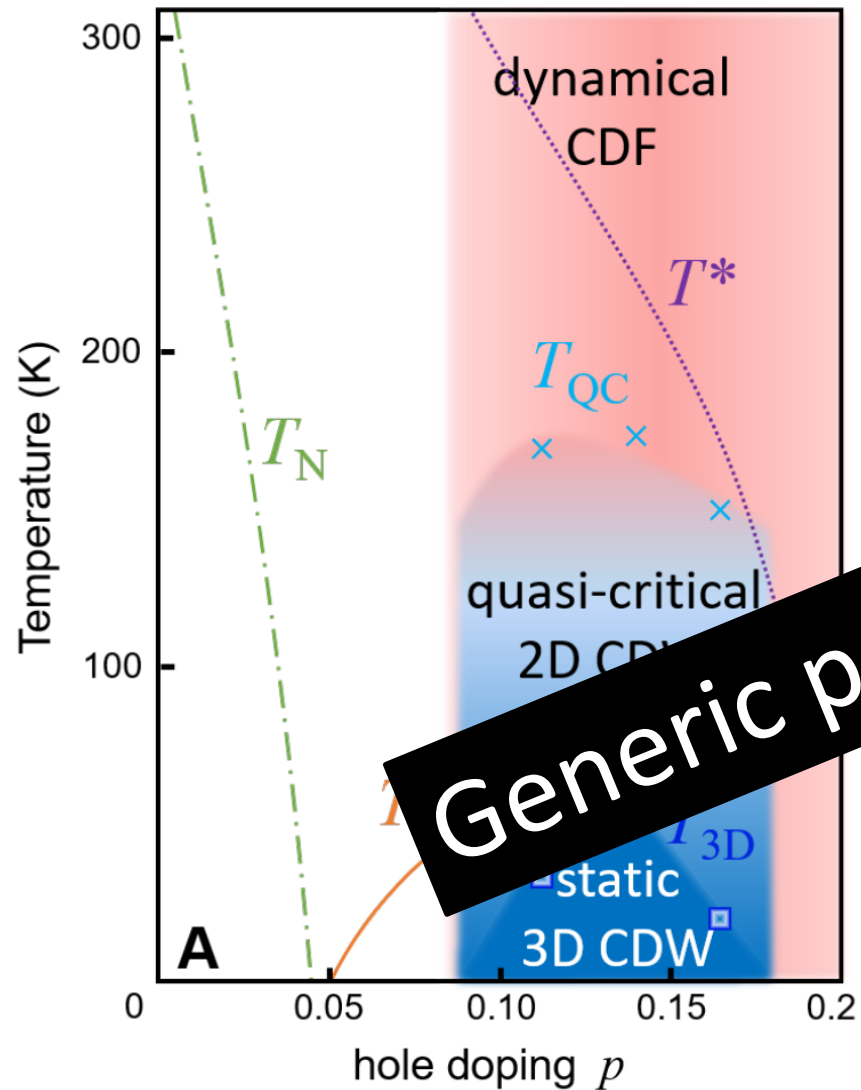


Phonons in RIXS reveal CDW excitation



L. Chaix, G. Ghiringhelli, Y. Y. Peng, M. Hashimoto, Y. He, S. Chen, K. Kummer, N.B. Brookes, B. Moritz, S. Ishida, Y. Yoshida, H. Eisaki, L. Braicovich, Z.-X. Shen, T. P. Devereaux, W.-S. Lee, *Nature Physics* **13**, 952 (2017)

Conclusions on CDW and CDF



Generic phase diagram ?