POLITECNICO MILANO 1863 | DIPARTIMENTO DI FISICA

Infinite-layer nickelate superconductors studied with Resonant Inelastic X-ray Scattering

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PolimiX group

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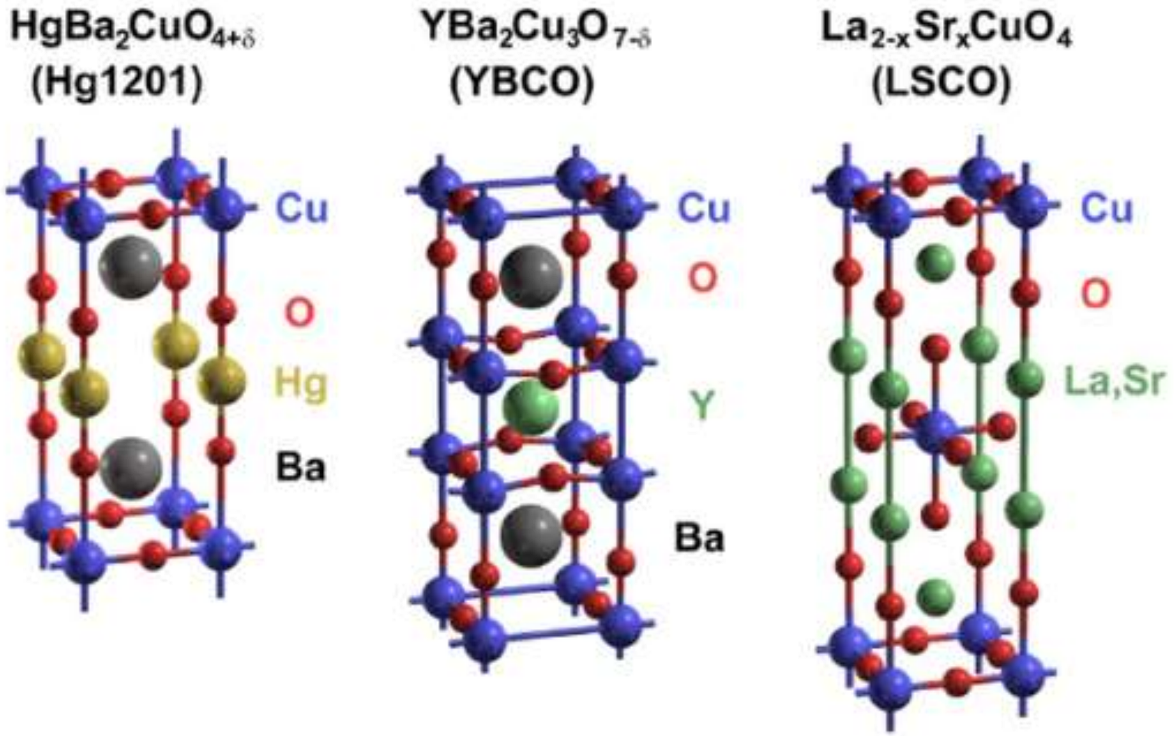
ESRF

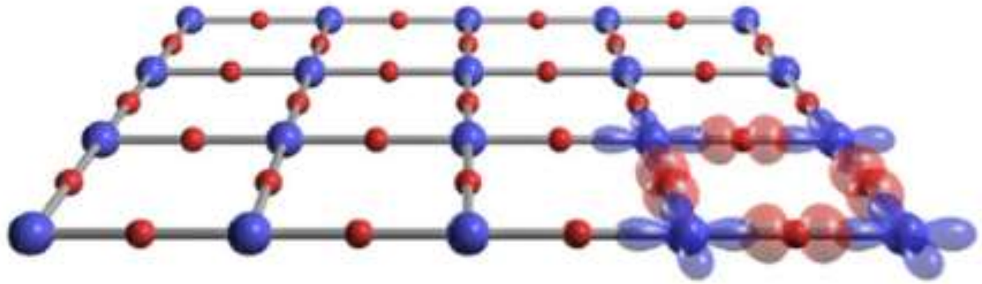
CHALMERS UNIVERSITY OF TECHNOLOGY

IPCMS Institut de Physique et Chimie des Matériaux de Strasbourg

JAGIELLONIAN UNIVERSITY IN KRAKOW

Cuprates





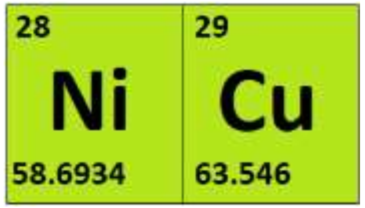
Barišić, N. et al., Proceedings of the National Academy of Sciences of the United States of America. 110. 10.1073/pnas.1301989110.

- Highest superconducting T_C at atmospheric pressure
- CuO_2 planes amplify electron-electron interactions
- Cu^{2+} has $3d^9$ configuration (S = ½), but <u>not</u> conductive
- Planar square Cu-O coordination: CF distortion
- Tc around 90 K

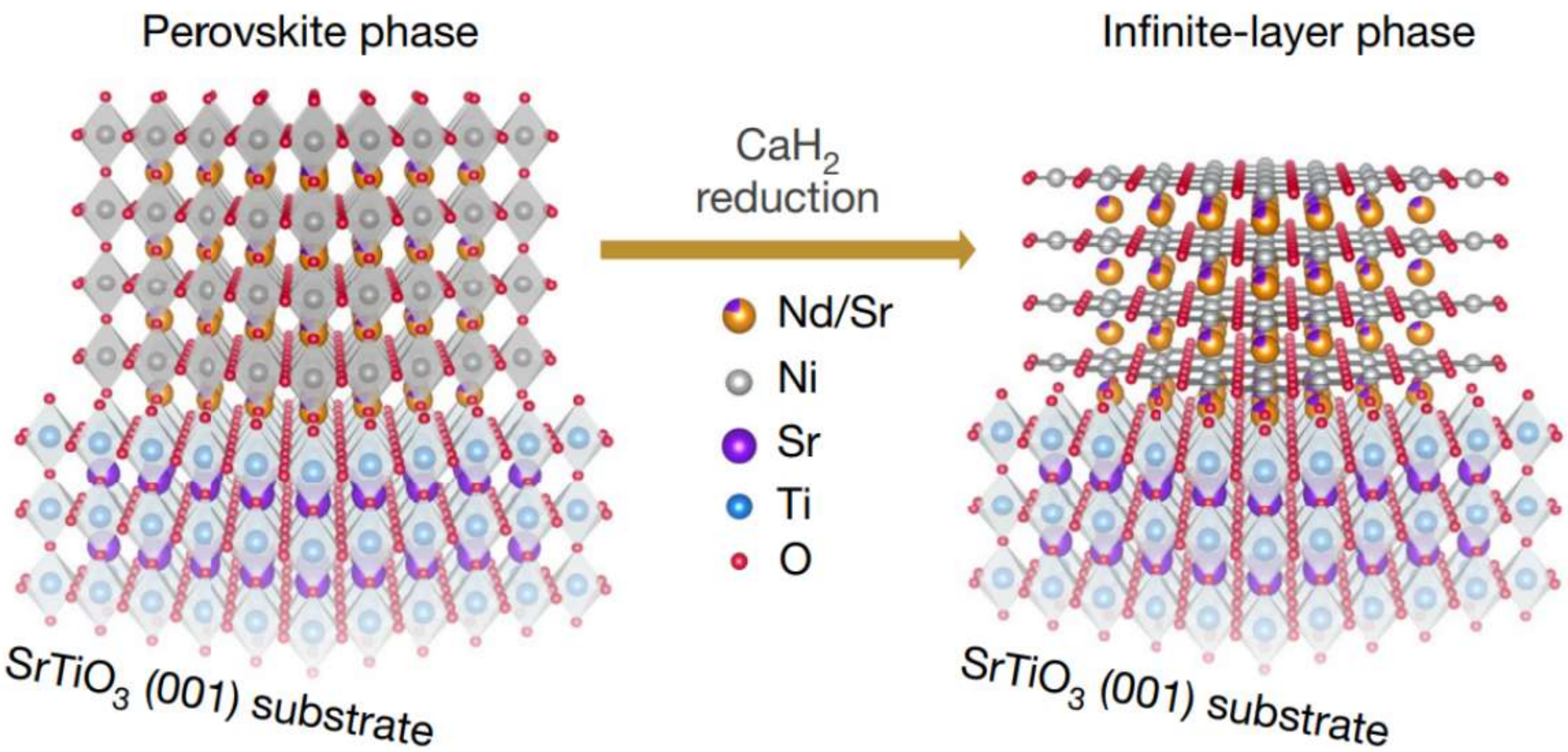
$$d_{x^2-y^2}$$

Chen et al., Physical Review B, 87 (2013)

$Nd_{1-x}Sr_xNiO_2$ (NSNO)



- Ni^{1+}/Ni^{2+} mimic Cu^{2+}/Cu^{3+}
- Spin ½, same configuration *$3d^9/3d^8$*; NiO_2 planes
- T_C around 10 K, thickness ~ 10 nm



$Nd_{1-x}Sr_xNiO_3$ $Nd_{1-x}Sr_xNiO_2$

Li et al., Nature, 572(7771): 624627, 2019

- Infinite-layer phase, obtained by apical oxygen deintercalation (topotactic reduction): CaH_2 powder
- Self-doping from RE: nominally undoped samples are not perfectly insulating and may show superconductivity

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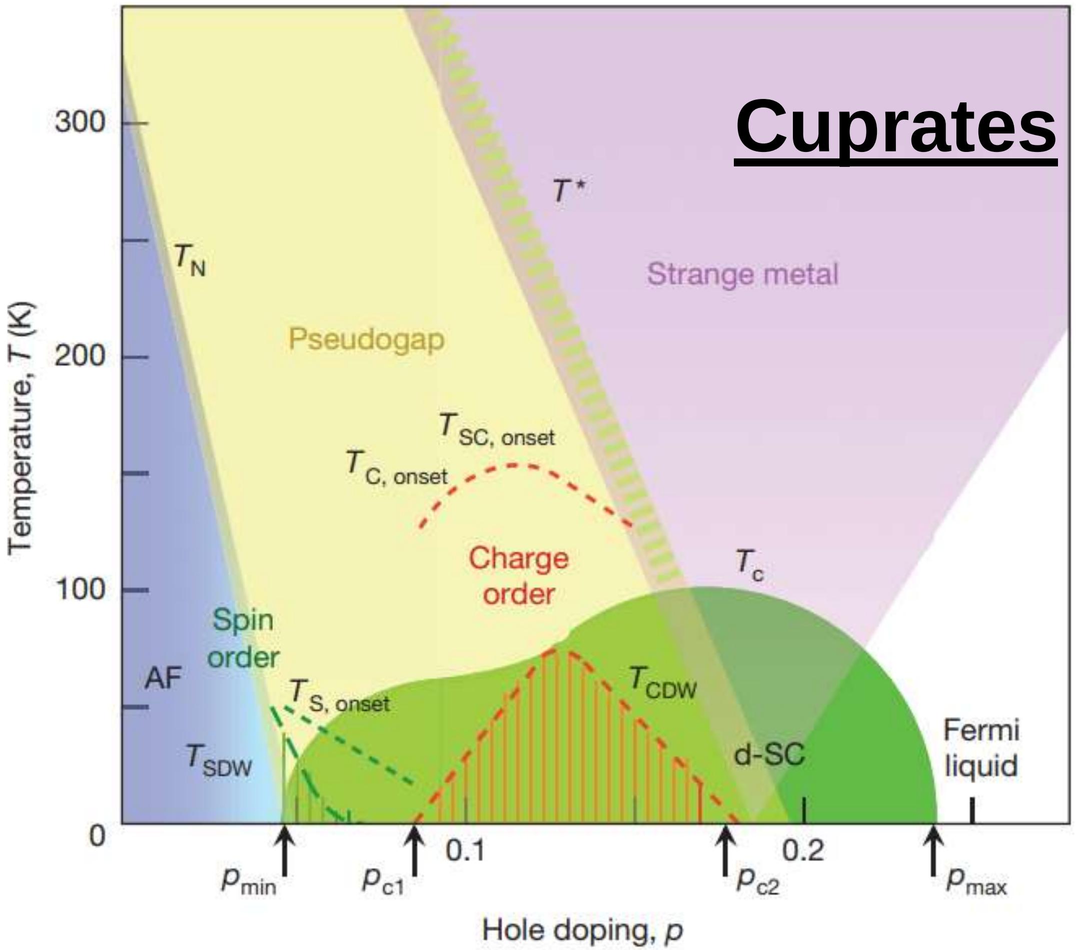
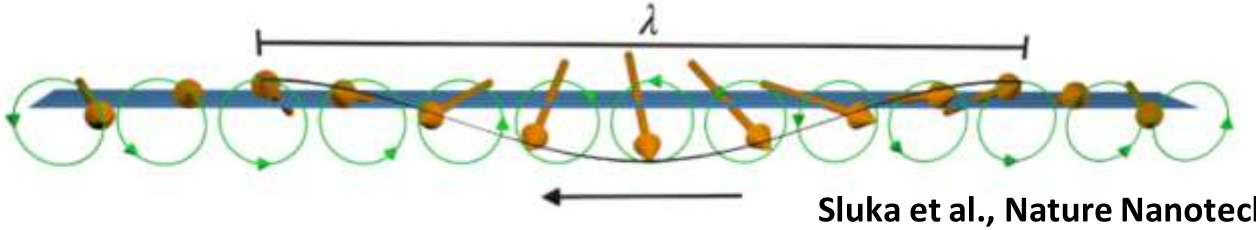
Dr. Daniele Preziosi

Phase diagram

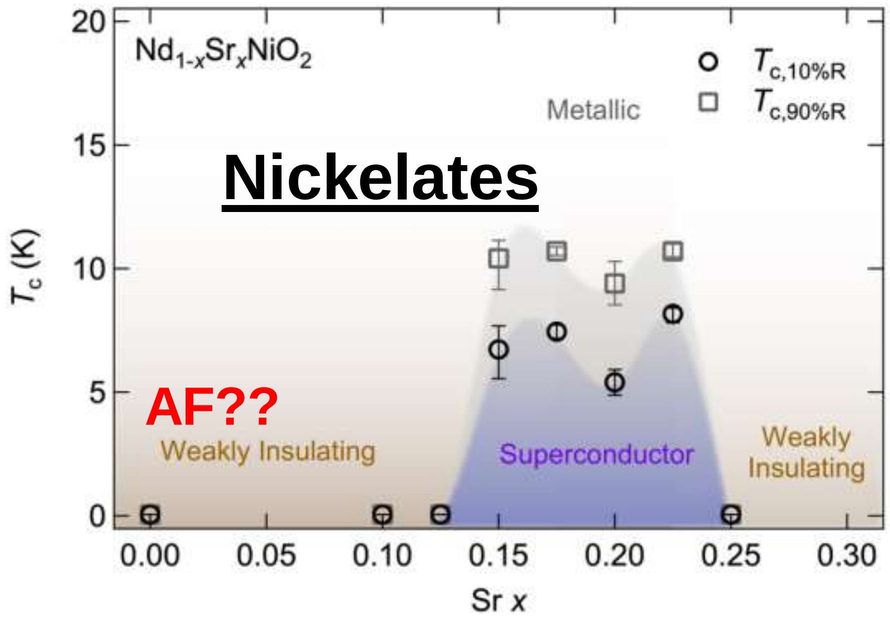


Keimer et al., Nature, 518 (7538): 179186, 2015.



Sluka et al., Nature Nanotechnology. 14. 10.1038/s41565-019-0383-4, 2019

- Many different regimes often competing with each other, e.g. magnetism and superconductivity
- Doping: AF destruction, SC rise
- Magnetic excitations (spin waves) can be an indirect probe of SC

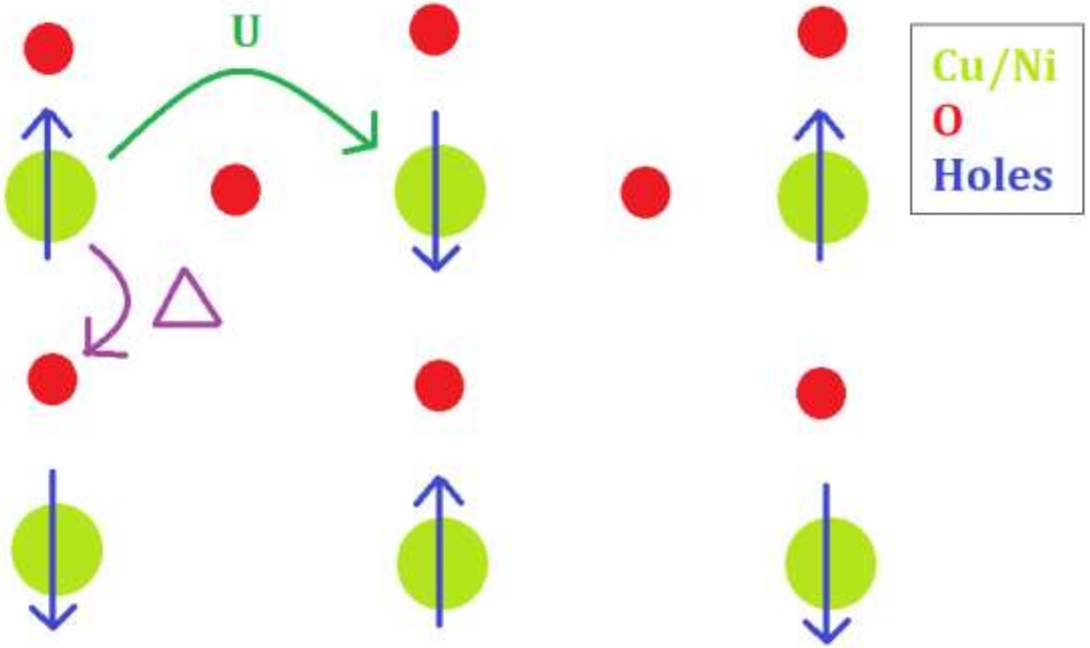


Li et al., Phys. Rev. Lett. 125, 027001 (2020)

Hubbard model

Useful to describe the behavior of doping holes

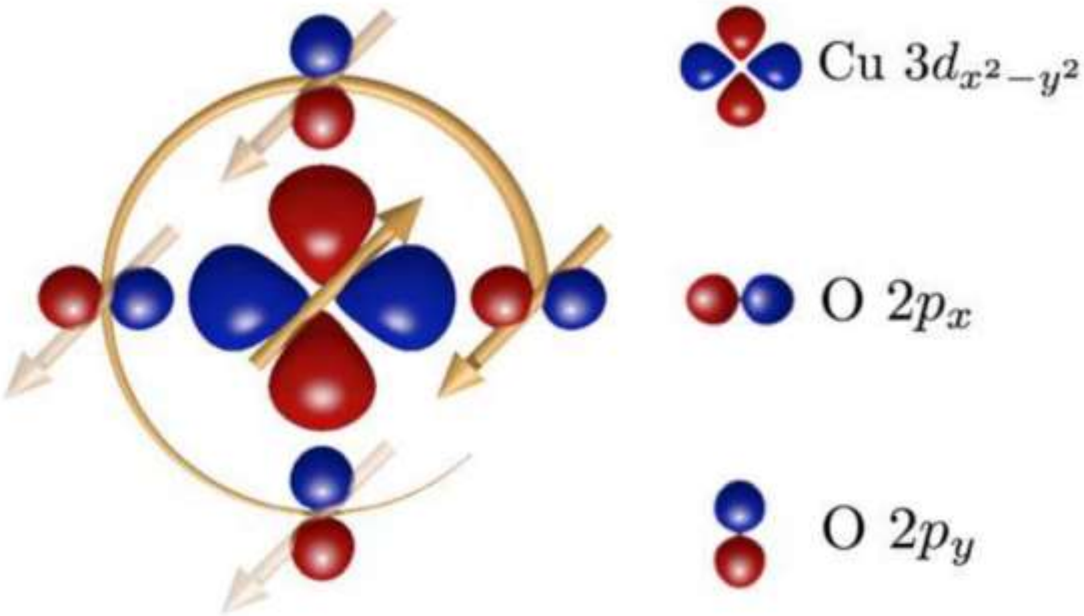
U = Mott-Hubbard energy of hole
Δ = Charge-transfer energy of hole



Materials classification according to these:
Zaanen et al., Physical review letters, 55(4):418, 1985

In cuprates, usually *Δ* < *U*: hole transfer to the ligand is favored (*3d⁹L̲*)

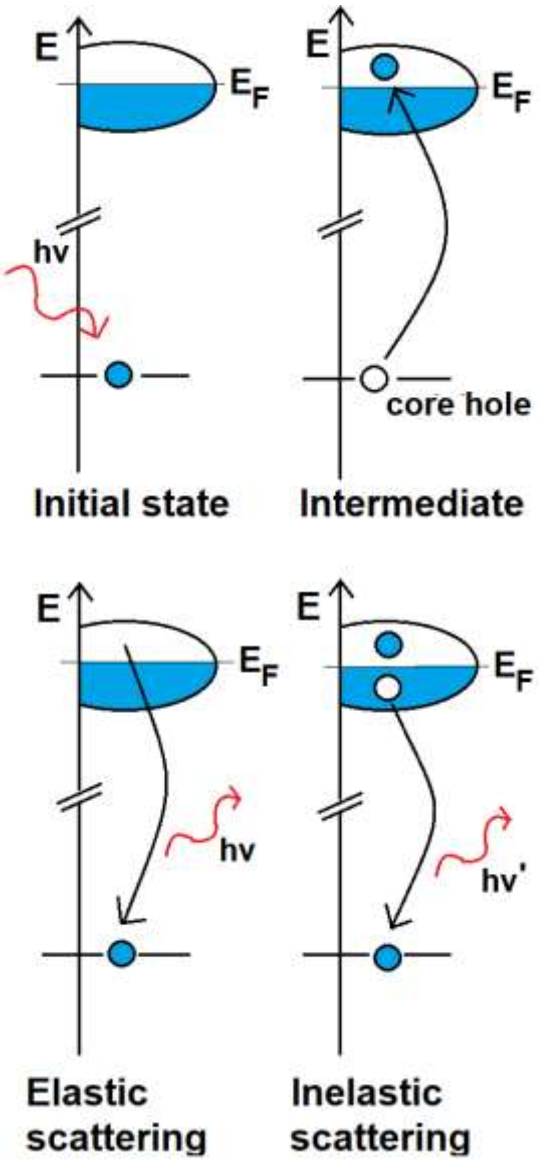
Hole delocalization along neighbors:



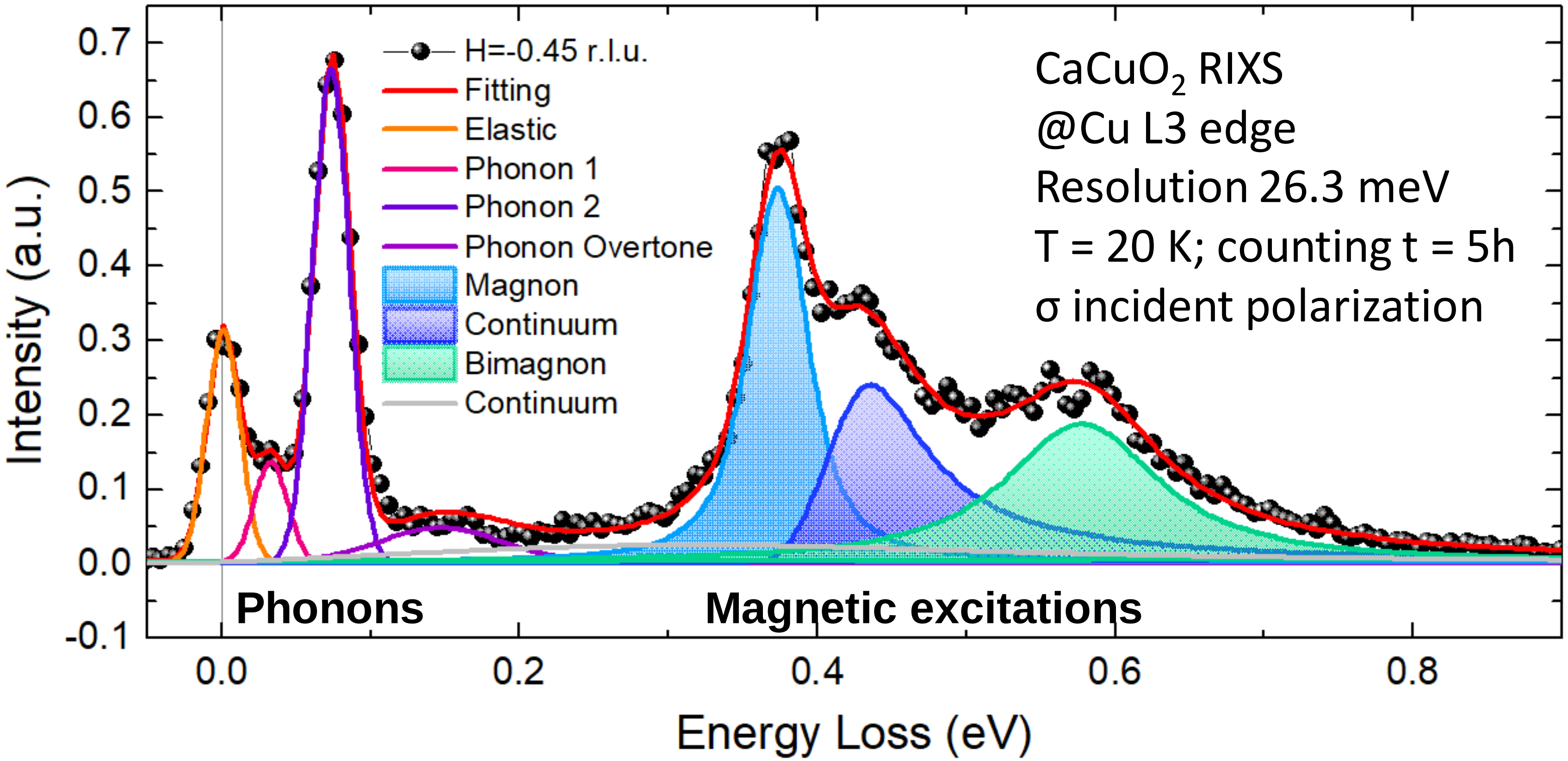
Zhang-Rice singlet
Chen et al., Physical Review B, 87 (2013)

Conversely, for NSNO it is ***Δ* > *U***: strong hole localization on the metal

Resonant Inelastic X-ray Scattering (RIXS)

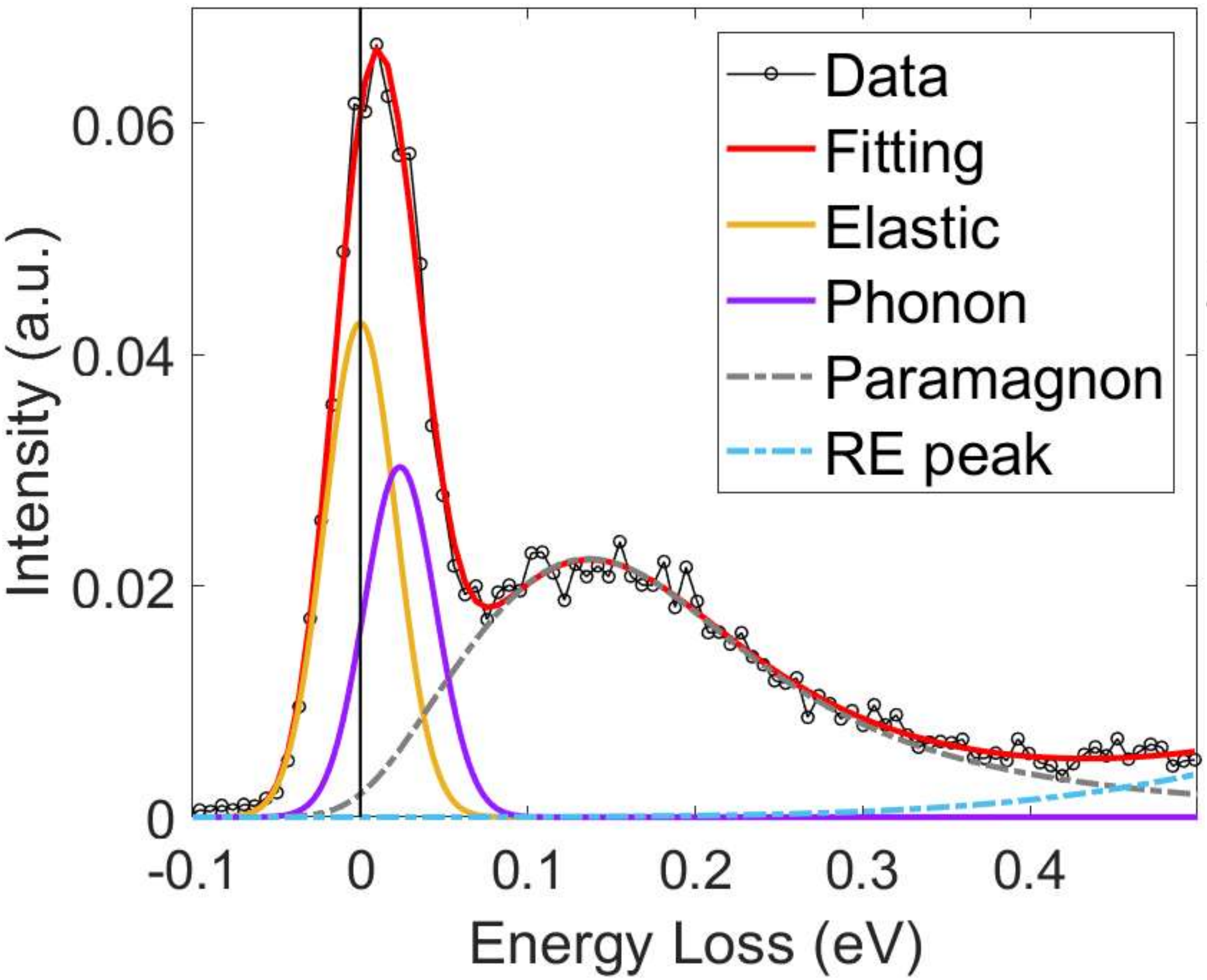


Martinelli et al., Physical Review X, 12(2):021041, 2022



- We work at the L3-edge of Cu and Ni: 900 eV ca. (soft X-rays)
- Magnon fitting for cuprates: Damped Harmonic Oscillator susceptibility

Damped Harmonic Oscillator (DHO) fitting



Paramagnon fit (DHO):

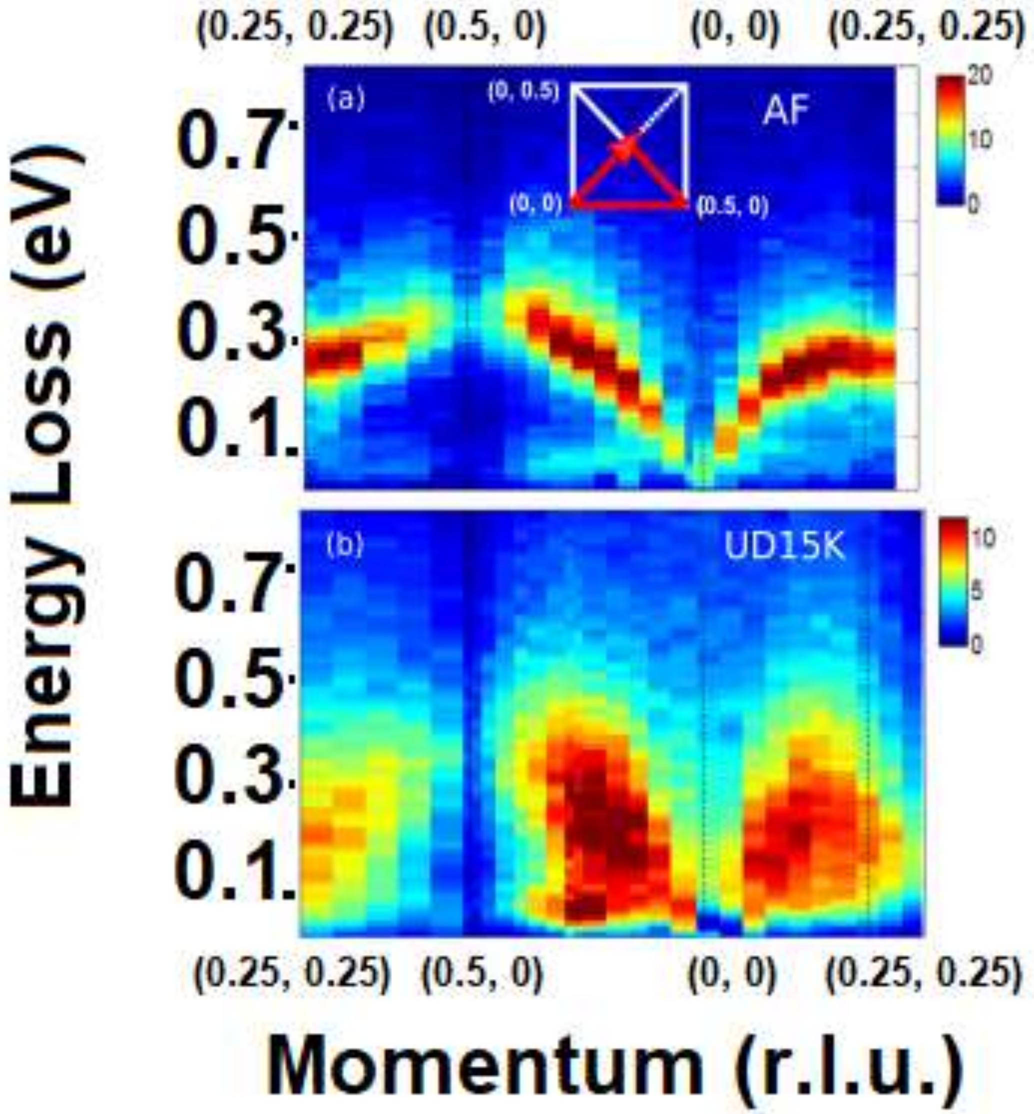
$$\chi''(\omega) = \frac{A_2 \gamma \omega}{(\omega_0^2 - \omega^2)^2 + 4\gamma^2\omega^2}$$

ω_0 = undamped frequency

γ = damping

Peak at $\omega_p^2 = \omega_0^2 - \gamma^2$

Magnetic excitations : cuprates



$(Bi,Pb)_2(Sr,La)_2CuO_{6+\delta}$

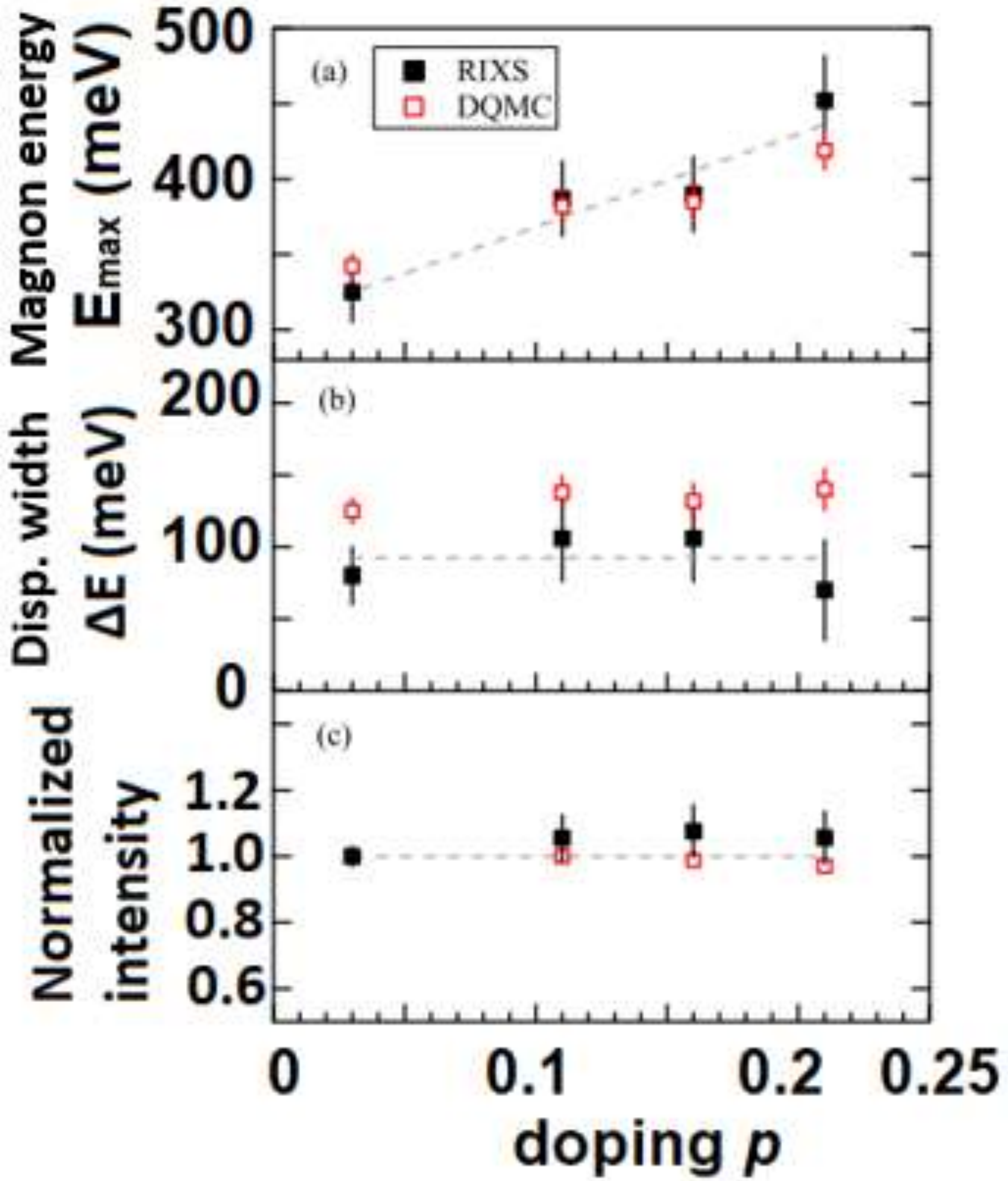
ω_p

γ

T = 20 K

AF: p = 0.03

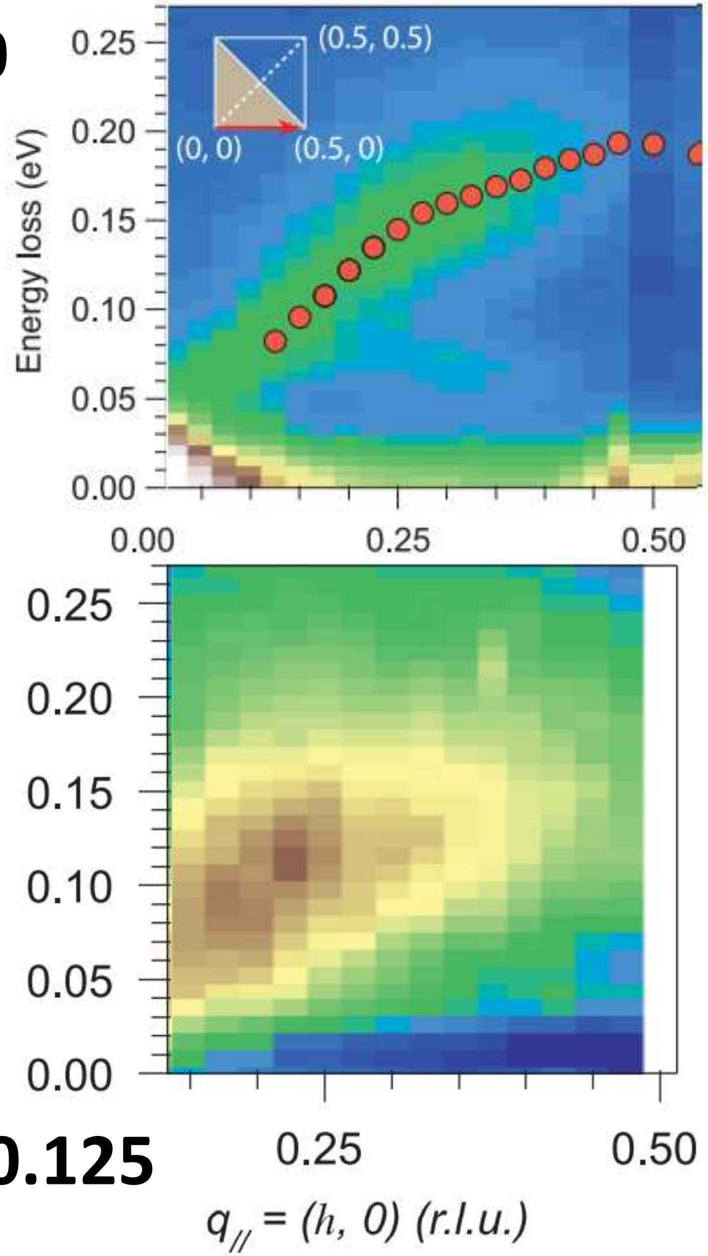
UD: p = 0.11



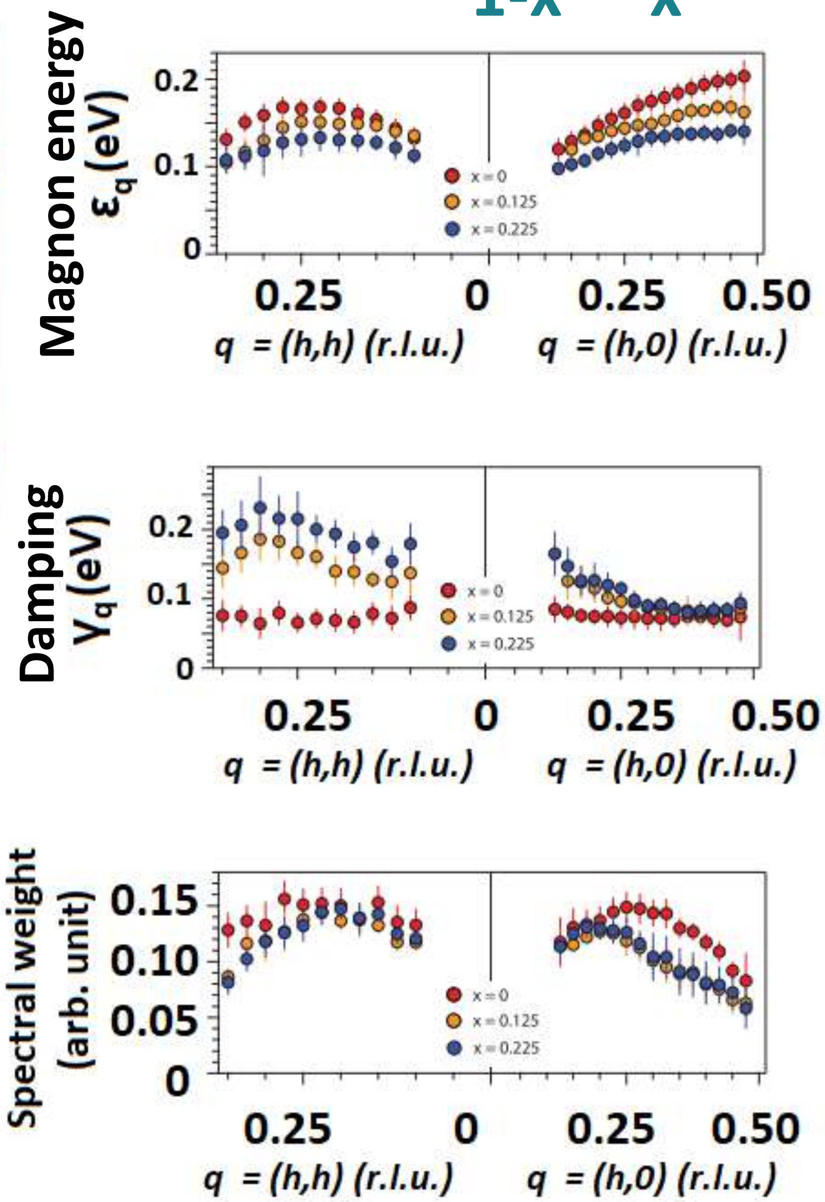
Peng et al., Physical Review B 98, 144507 (2018)

Magnetic excitations : $Nd_{1-x}Sr_xNiO_2$ (NSNO)

x = 0



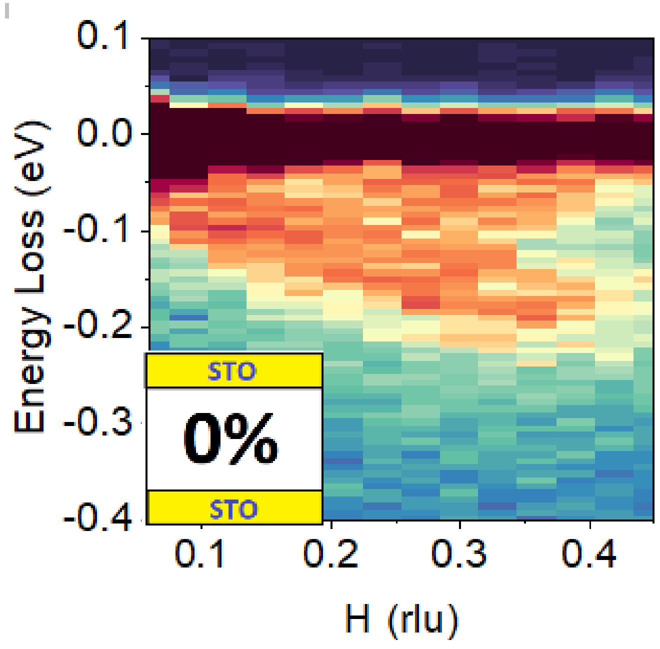
x = 0.125

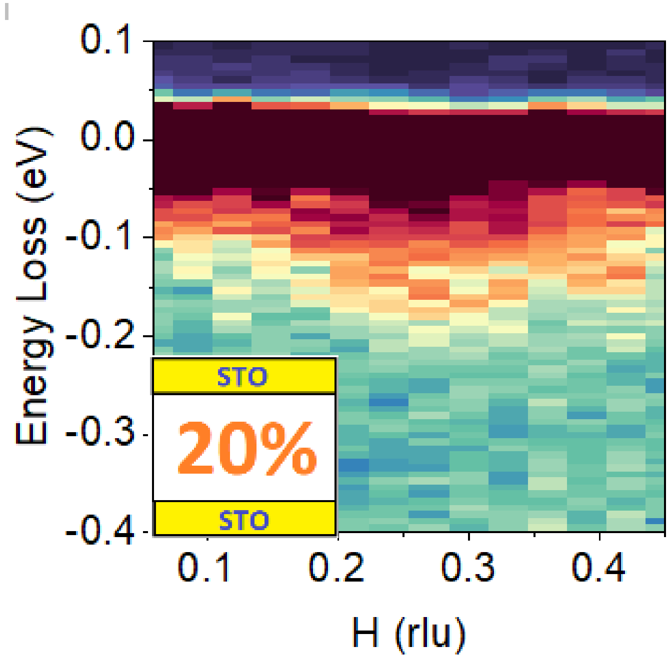


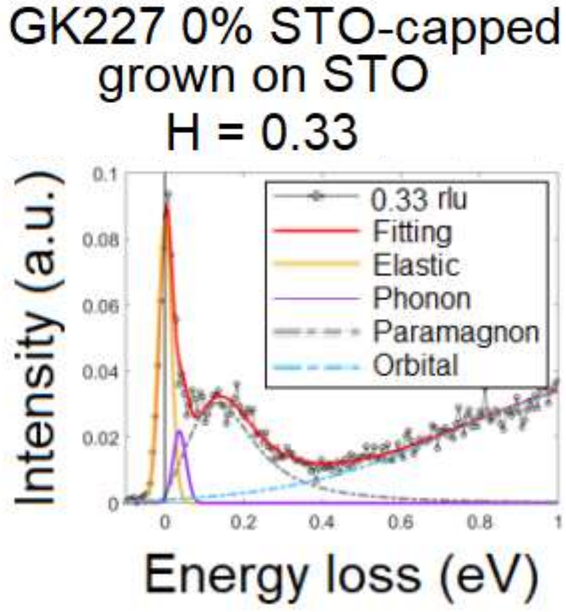
- Excitations compatible with spin-½ AFM magnons on a square lattice
- As in cuprates, overdamping upon doping
- Unlike in cuprates, energy softening and mild decrease in spectral weight
- Attributed to strong localization of doping holes on Ni sites

Lu et al., Science 373, 213–216 (2021)

Magnetic excitations - NSNO



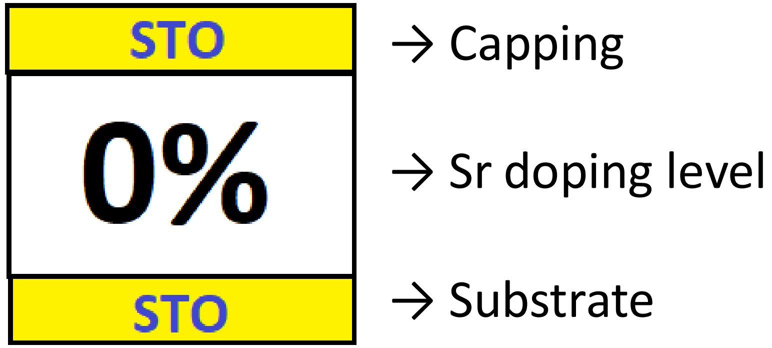




$Nd_{1-x}Sr_xNiO_2$

- The samples were realized by Dr. Daniele Preziosi and Guillaume Krieger (Institut de Physique et Chimie des Matériaux, Strasbourg, France) by RHEED-monitored PLD
- T = 20 K, resolution 39 meV ca.
- Incident energy: Ni^{1+} L3 edge
- π incident polarization

Legend

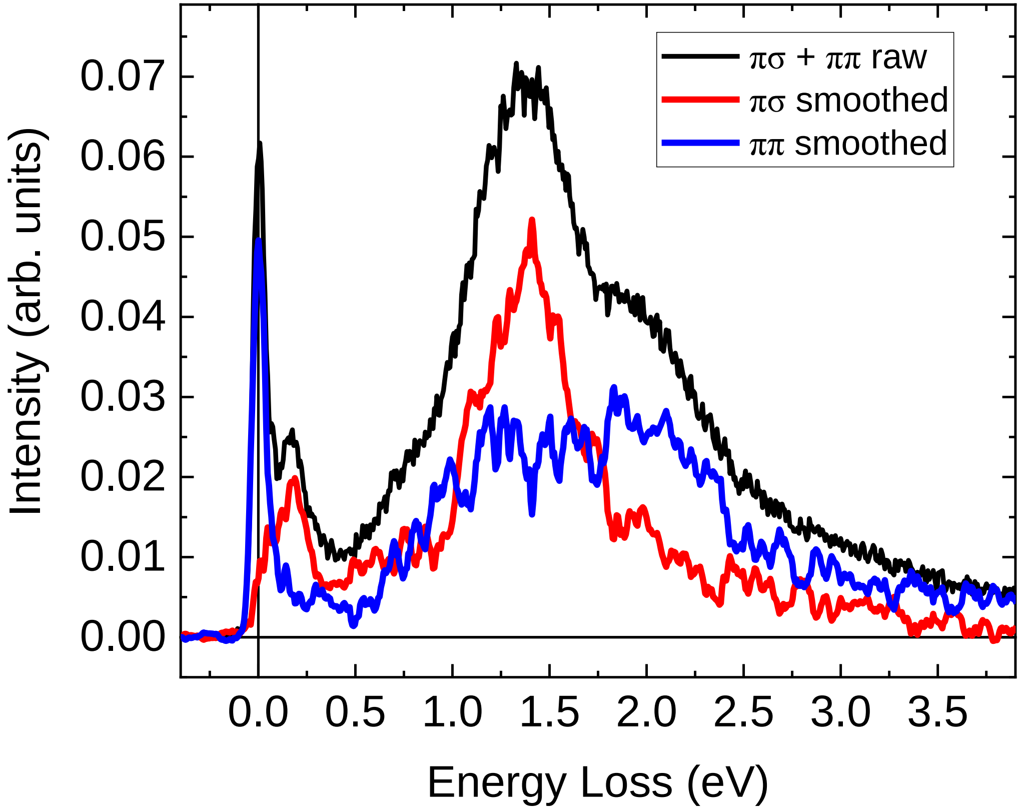


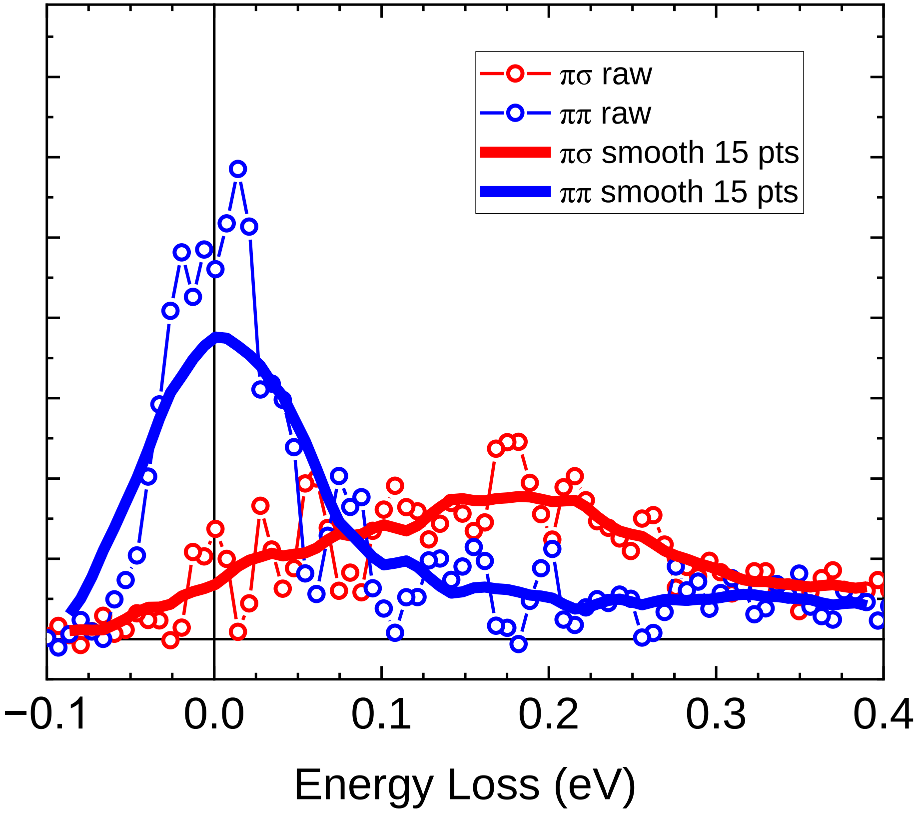
- Clearly dispersing feature @ 100-200 meV, which disappears with doping

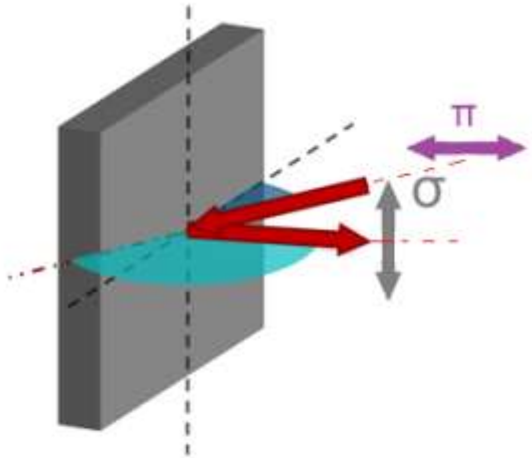
Polarimeter analysis

Undoped NNO

- ESRF (ID32 beamline) is currently the only facility in the world allowing such an analysis
- Graded multilayer mirror, with Rσ ≠ Rπ

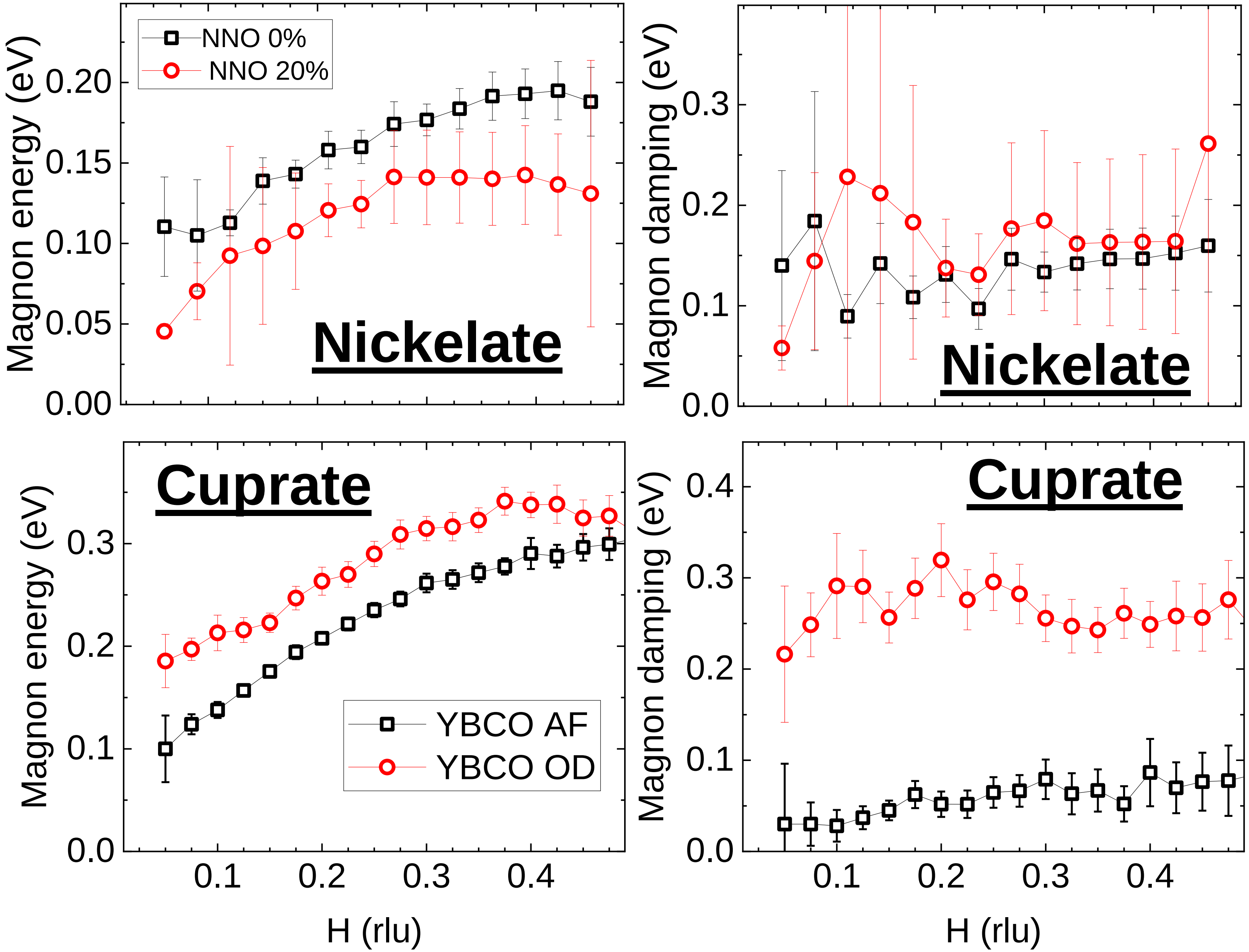


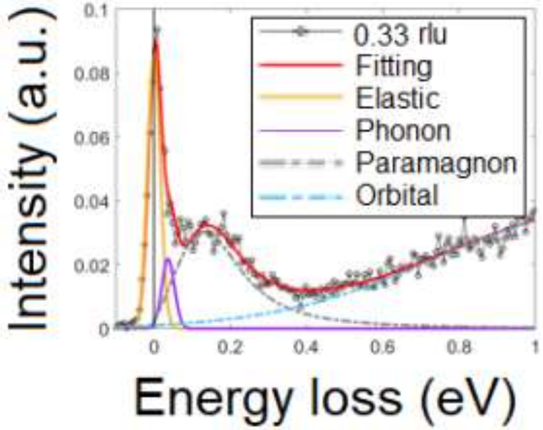




- Spin-flip nature is confirmed: polarization direction of incident light is rotated
- Disentanglement from underlying continuum

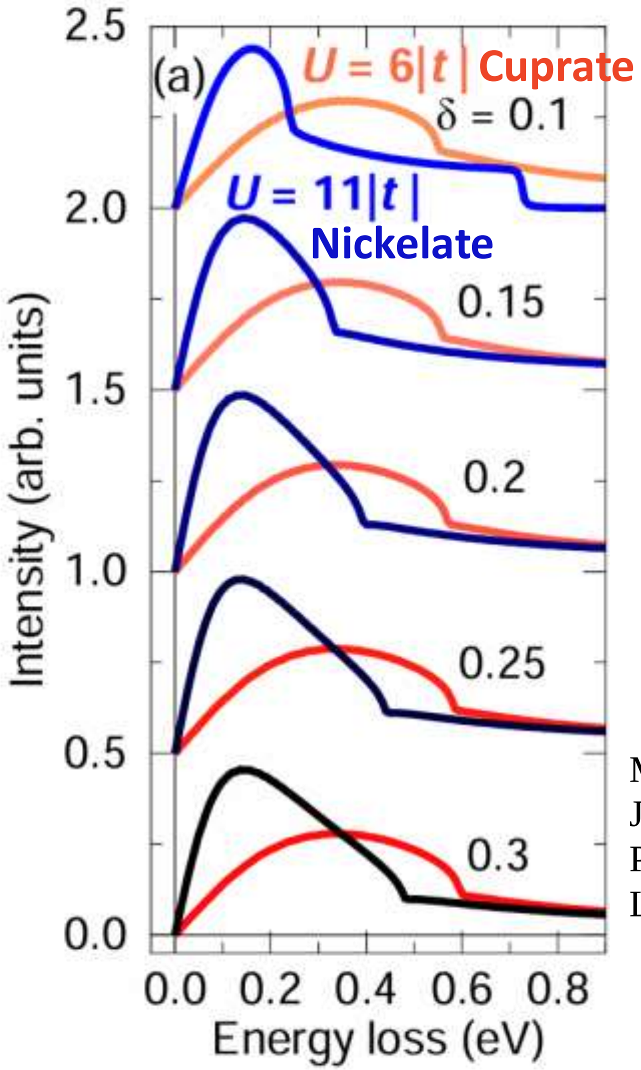
DHO Fitting trends

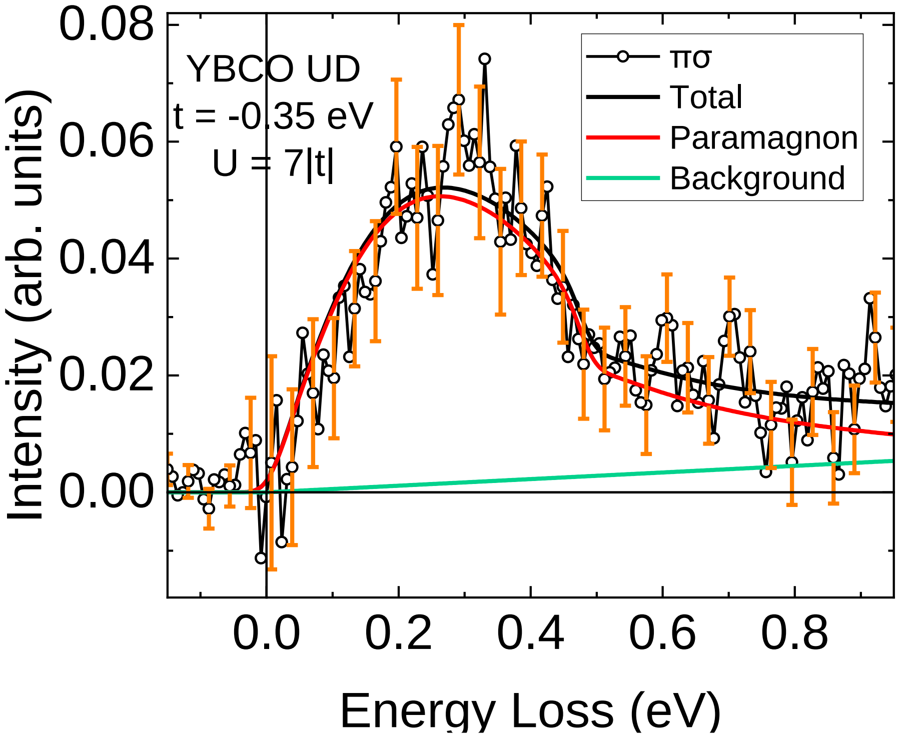


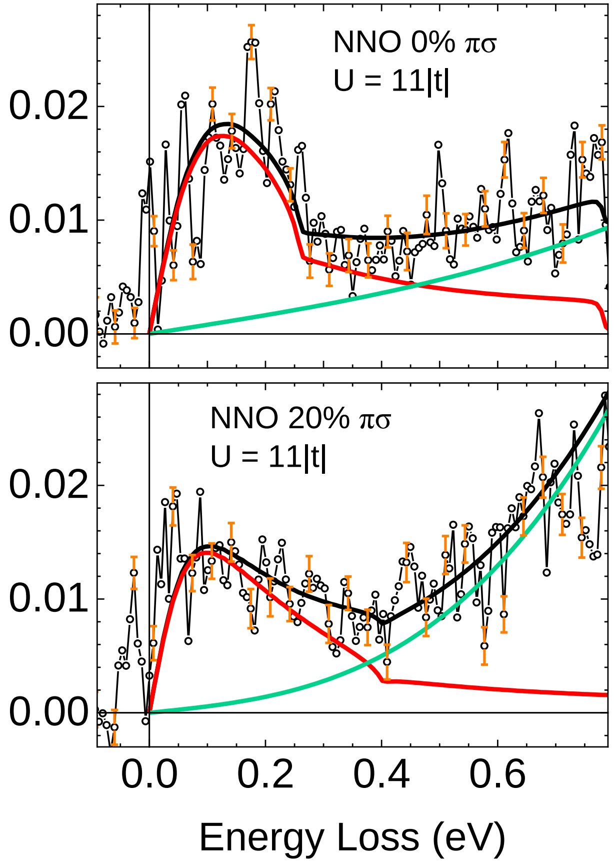


- YBCO recent data (July 2023) for comparison; Tc = 87 K
- Energy softening with doping for NNO is confirmed, in contrast to the cuprate
- Nickelates agreement is quite problematic

New susceptibility model







M. Fidrysiak and J. Spałek, Phys. Rev. B 104, L020510 (2021).

$$J \sim \frac{4t^2}{U}$$

Parameter	Cuprate	Nickelate
U	$7|t|$ = 2.45 eV	$11|t|$ = 4.4 eV
t	-0.35 eV	-0.4 eV
t'/t	0.25	0.25
k_BT	0.35	0.30

A remarkably larger value of *U = 11|t| = 4.4 eV* is found for nickelates with respect to cuprates

Conclusions and discussion

- RIXS analysis of magnetic excitations dependence on doping in nickelate
- Comparison with cuprates: opposite energy behavior is confirmed
- Polarimeter analysis: disentanglement of the peak from the underlying excitations (mainly charge continuum)
- Development of a new, Hubbard-based susceptibility model, allowing a direct tuning of the main energy parameters
- First result: $U = 11|t|$ in nickelates, while $U = 6|t|$ in cuprates. <u>What about Δ ??</u>
- Submitted paper

(available on arXiv):

Spin excitations in $Nd_{1-x}Sr_xNiO_2$ and $YBa_2Cu_3O_{7-\delta}$: the influence of Hubbard U

F. Rosa,[1,*] L. Martinelli,[1,†] G. Krieger,[2,‡] L. Braicovich,[3] N.B. Brookes,[3] G. Merzoni,[1,4] M. Moretti Sala,[1] F. Yakhou-Harris,[3] R. Arpaia,[5,6] D. Preziosi,[2] M. Salluzzo,[7] M. Fidrysiak,[8] and G. Ghiringhelli[1,9,§]

https://arxiv.org/abs/2406.09271

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CHALMERS UNIVERSITY OF TECHNOLOGY

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SPIN

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Dr. Guillaume Krieger

(NSNO samples)