Tridimensional magnetism in Superconducting Infinite-Layer $PrNiO_2$ studied with Resonant Inelastic X-ray Scattering

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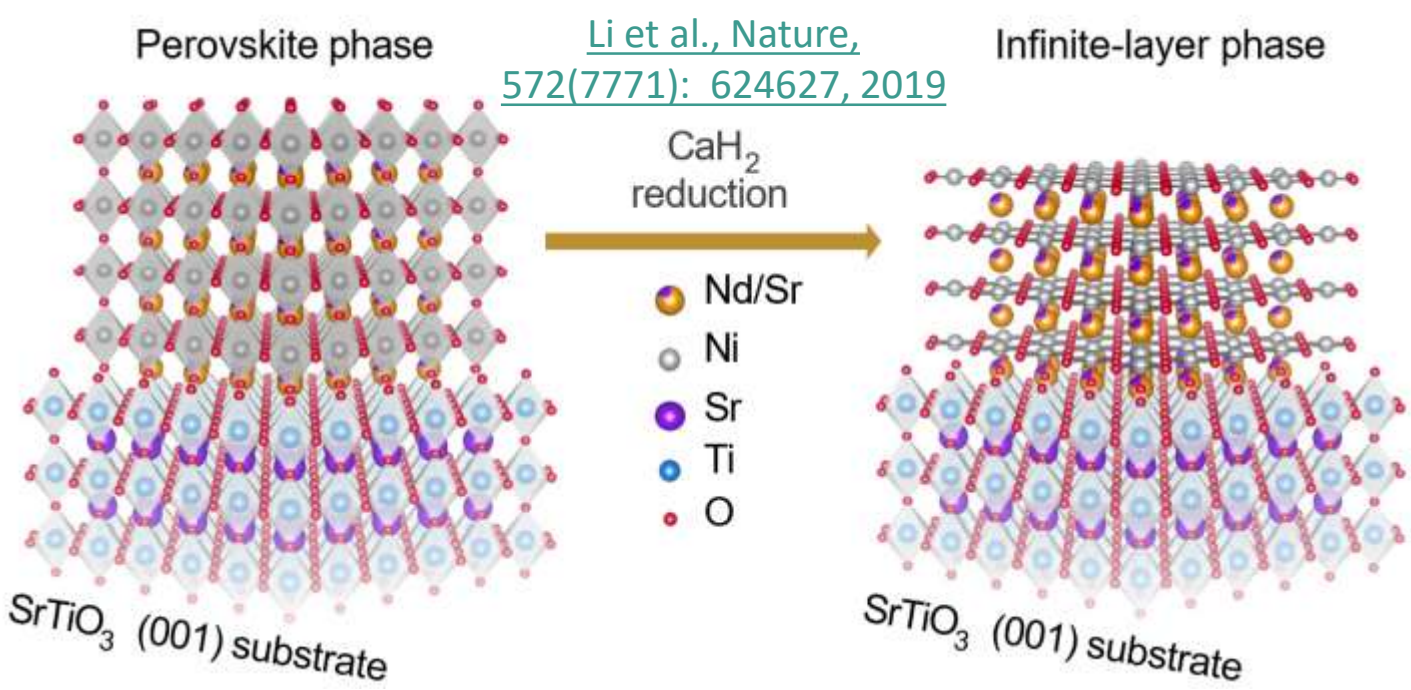
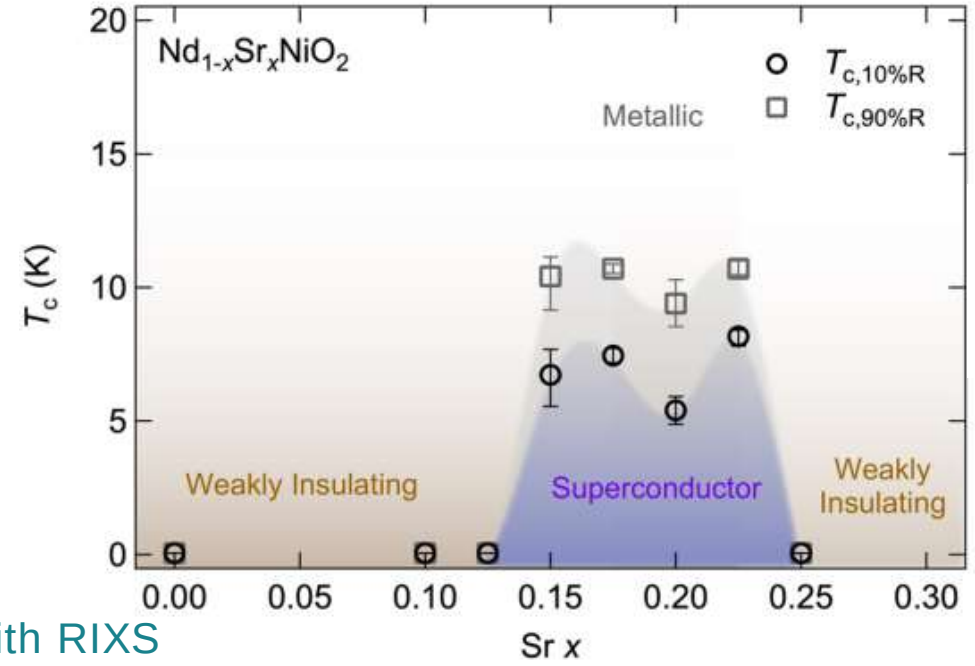
Infinite-Layer nickelates

- Closest High-T Superconductors analogous to cuprates (2019)
- Infinite-Layer structure strongly enhances electron-electron correlation
- Ni^{1+} ($3d^9$) well mimics Cu^{2+}; proposed phase diagram similar to cuprates
- Two main differences with respect to cuprates

1) $\Delta > U$: Mott insulators
2) RE role: self-doping



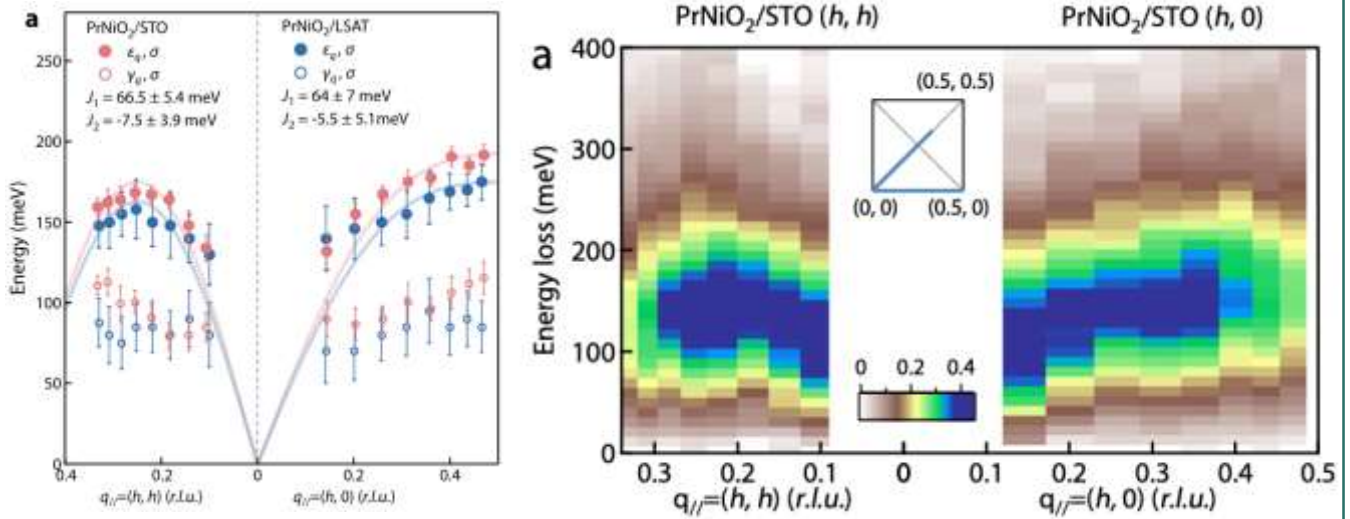


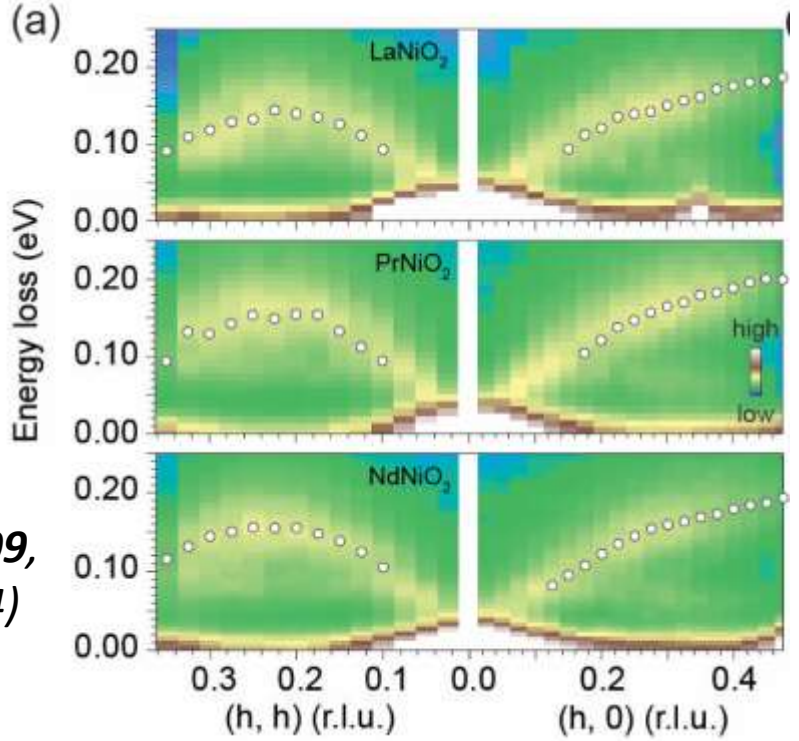
Li et al., Phys. Rev. Lett. 125, 027001 (2020)

IL nickelates magnetism – literature examples

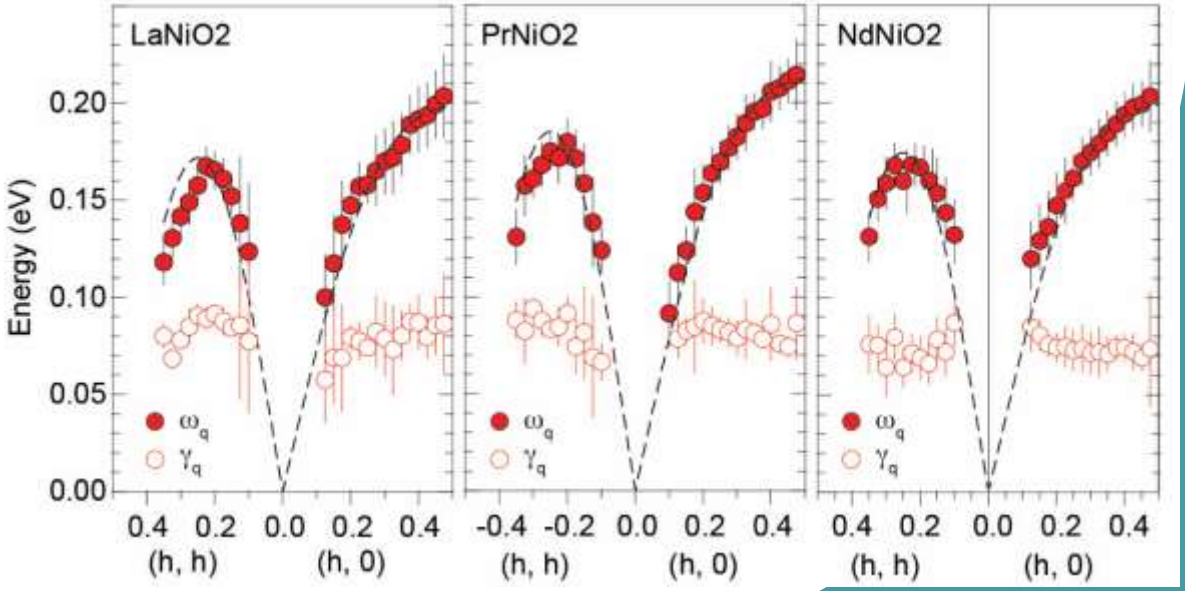
- No long-range AFM order
- Short-range correlations still enable propagation of spin waves (paramagnons)
- So far, only 2D models fitting (Linear Spin Wave theory)

Gao et al. Nat. Comm. 15.1 (2024): 5576.

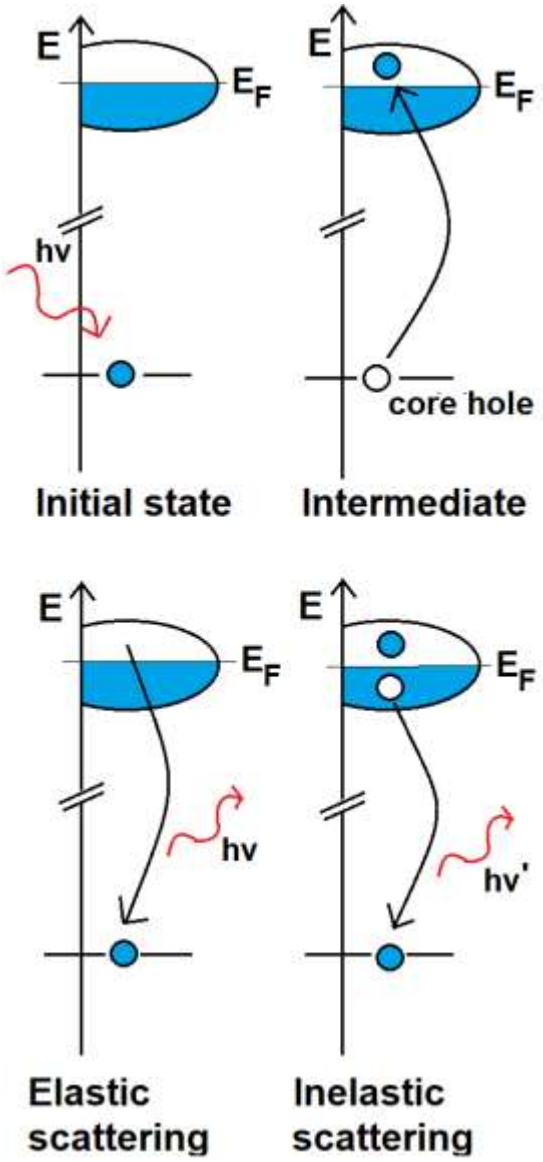


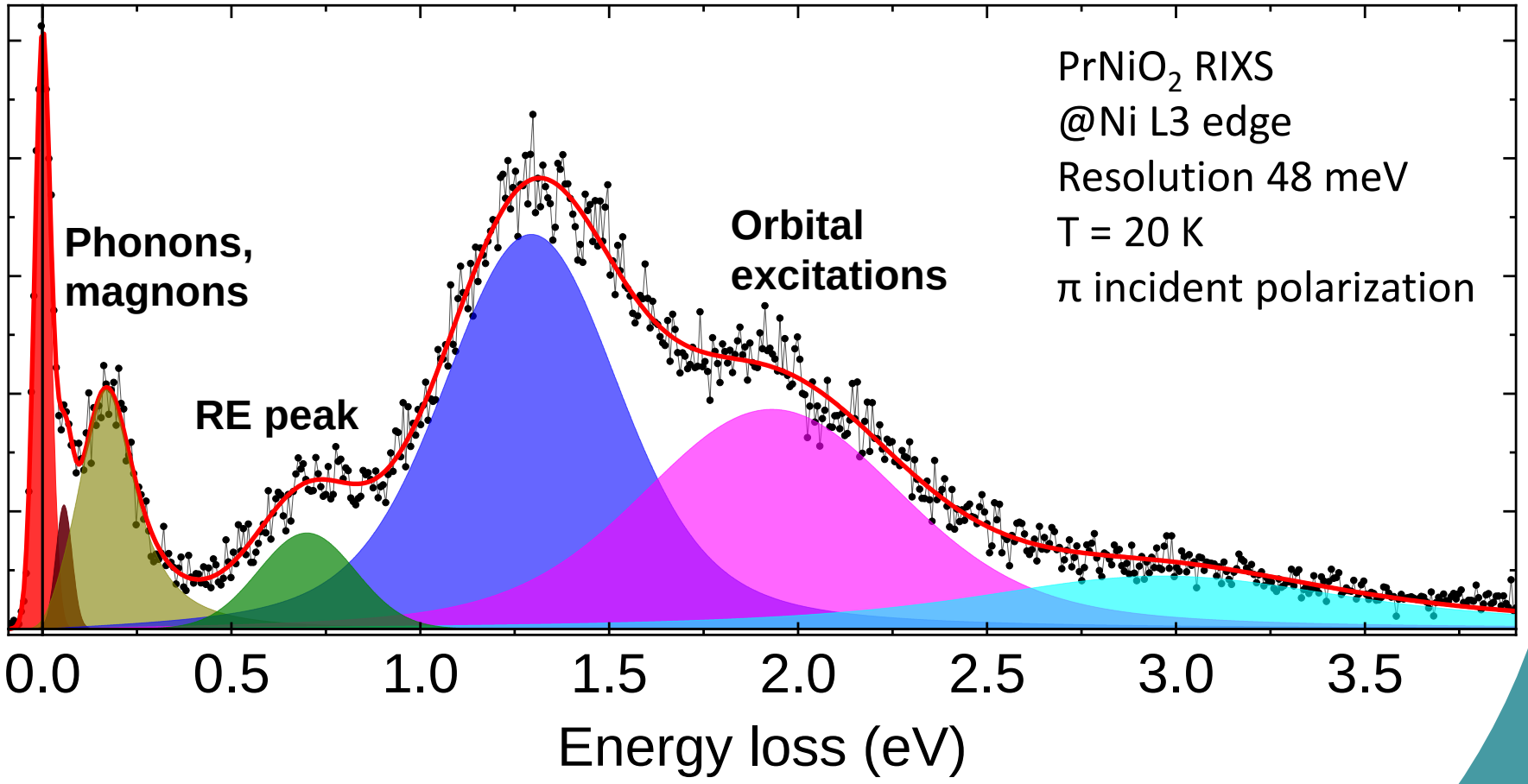


*Rossi et al., Phys. Rev. B **109**, 024512 (2024)*



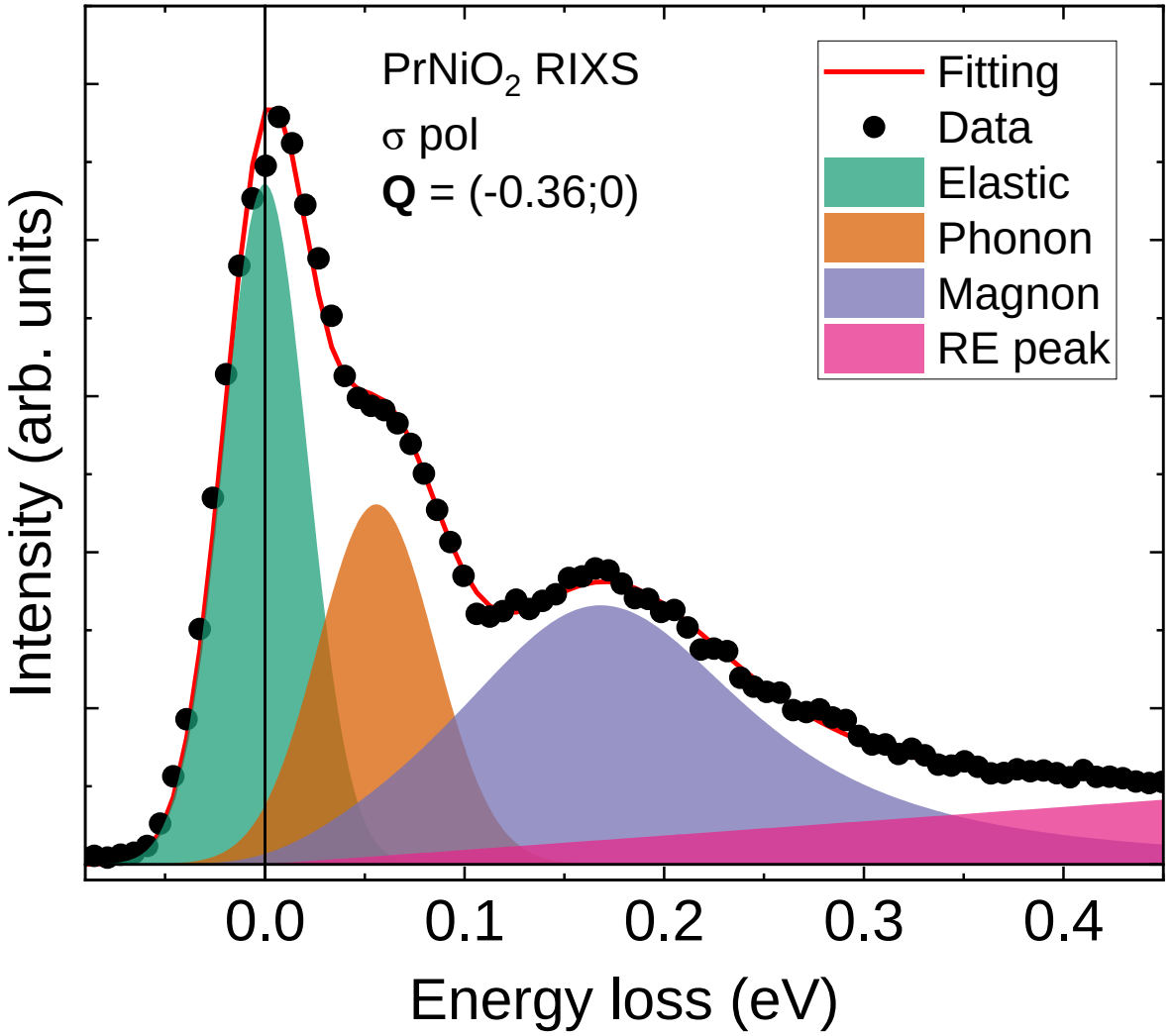
Resonant Inelastic X-ray Scattering (RIXS)





- We work at the Ni L3-edge: 853 eV ca. (soft X-rays)
- Magnon fitting: 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 $\omega_p^2 = \omega_0^2 - \gamma^2$

Sample growth and characterization

- Pulsed-Laser Deposition of precursor perovskite $PrNiO_3$ on STO single crystal substrate, RHEED monitoring
- STO capping layer; topotactic reduction with CaH_2 powder
- Thickness between 15-20 unit cells (~6 nm), Tc between 7 and 11 K

RESEARCH ARTICLE

ADVANCED MATERIALS
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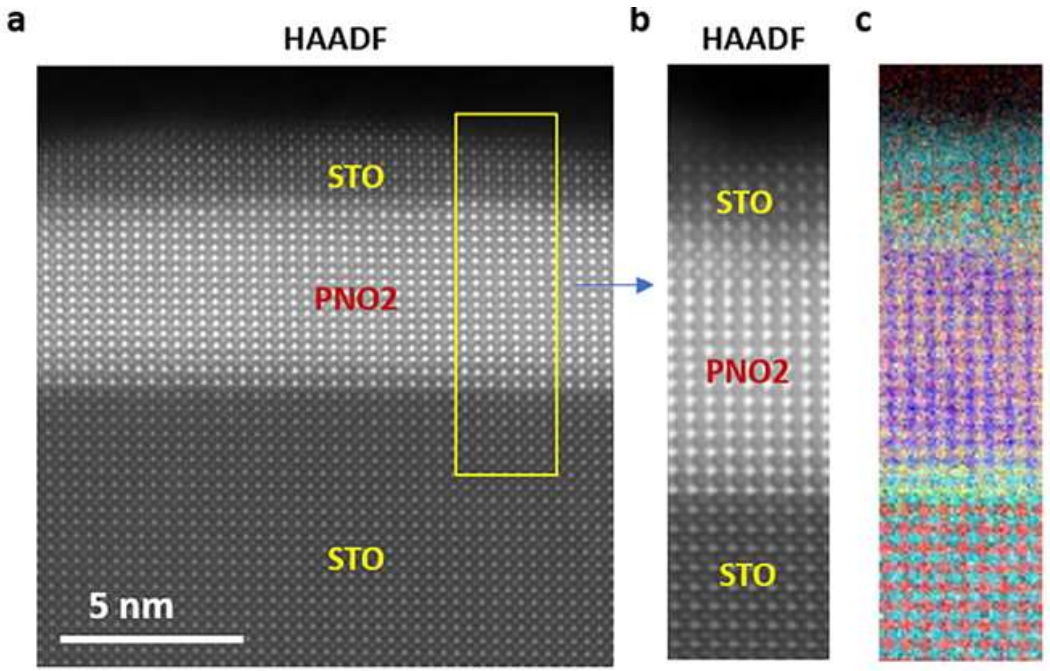
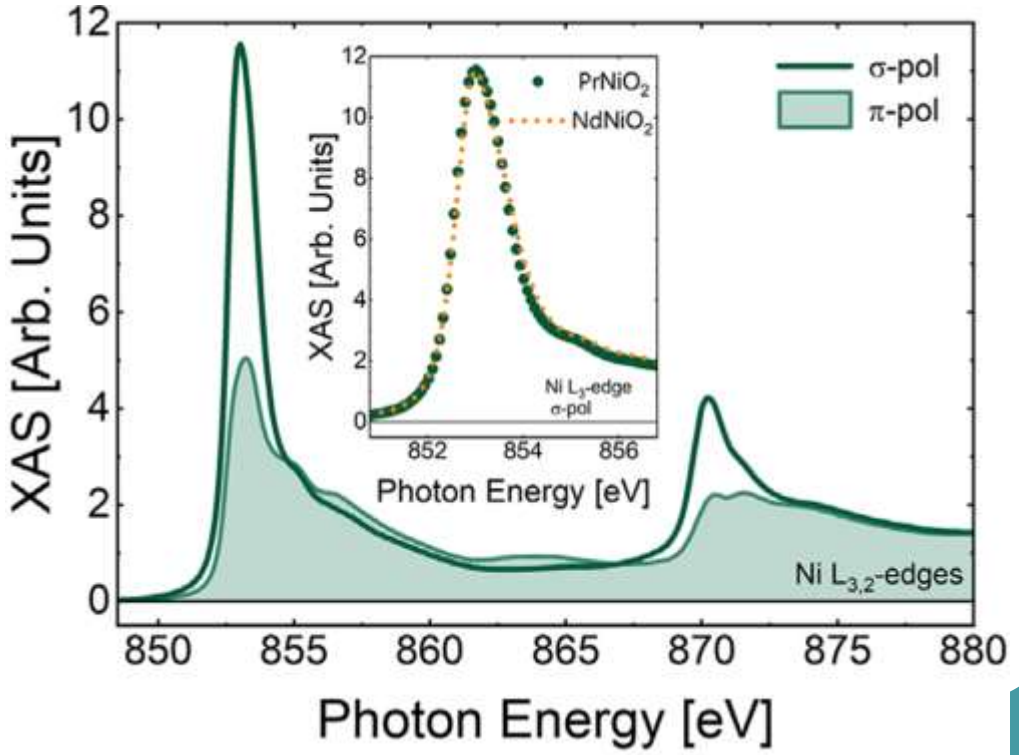
Superconductivity in $PrNiO_2$ Infinite-Layer Nickelates

Hoshang Sahib, Aravind Raji, Francesco Rosa, Giacomo Merzoni, Giacomo Ghiringhelli, Marco Salluzzo, Alexandre Gloter, Nathalie Viart, and Daniele Preziosi**

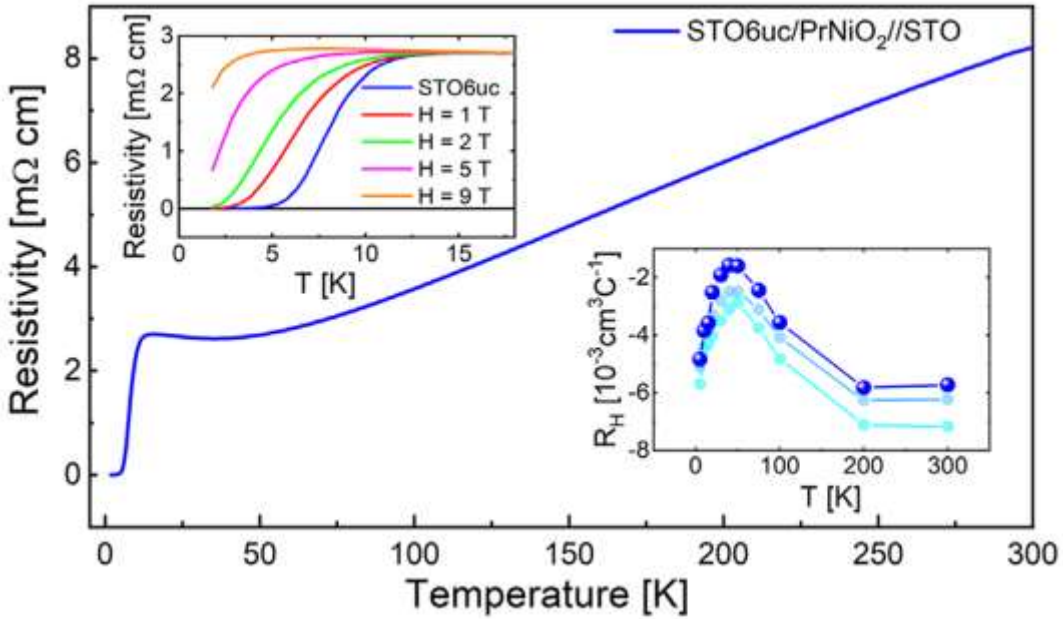
Sahib et al.,
Adv. Mat. 37.16 (2025): 2416187.





Sample growth and characterization

- No doping, but still resistivity drop, with Tc between 7 and 11 K
- Self-doping-induced superconductivity: no need of Sr substitution
- No sign change in Hall coefficient, unlike conventionally-doped SC samples



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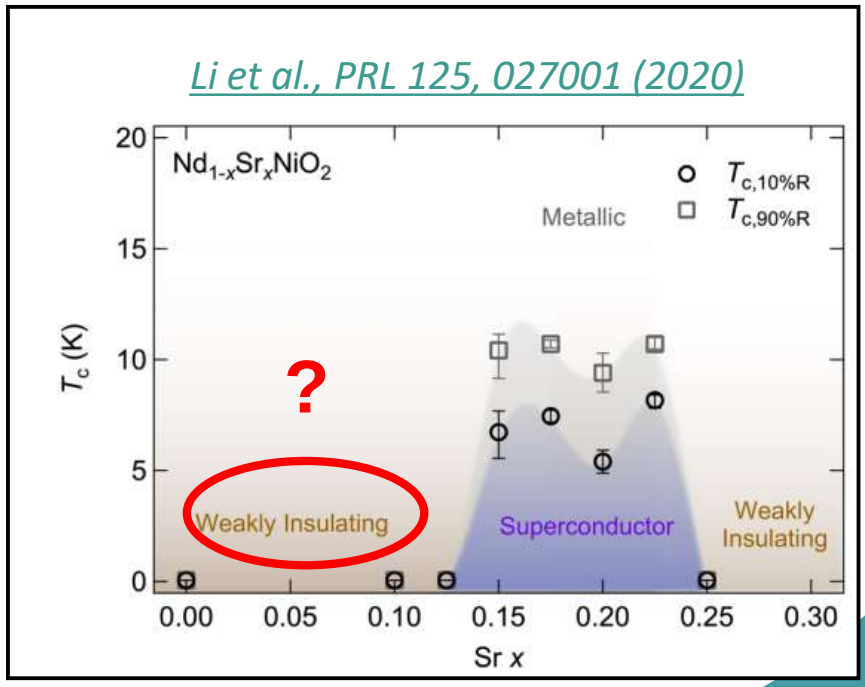
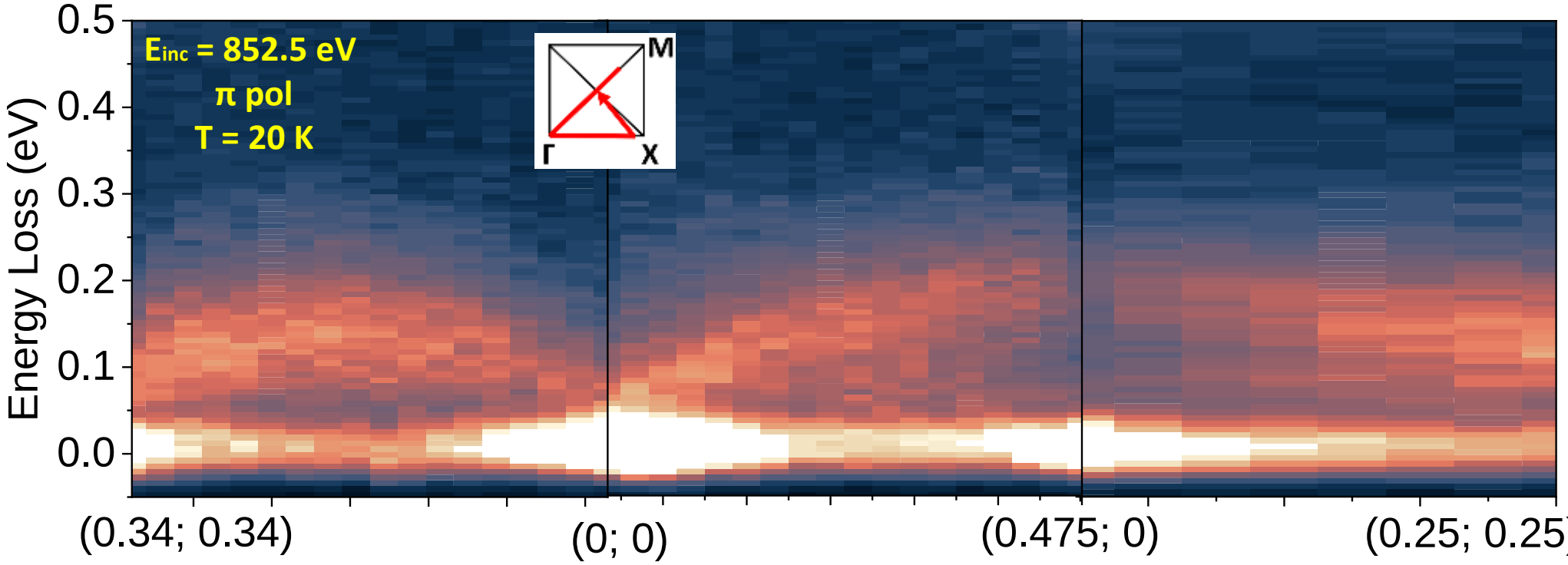
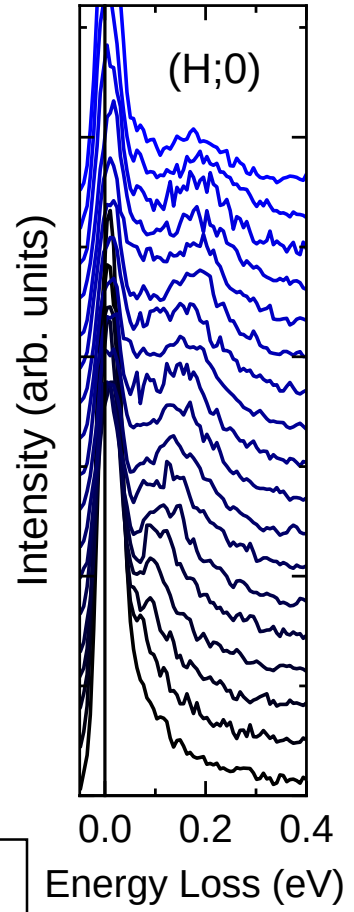


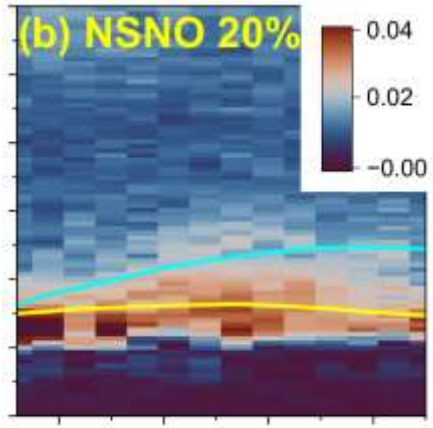
RIXS maps – $PrNiO_2$



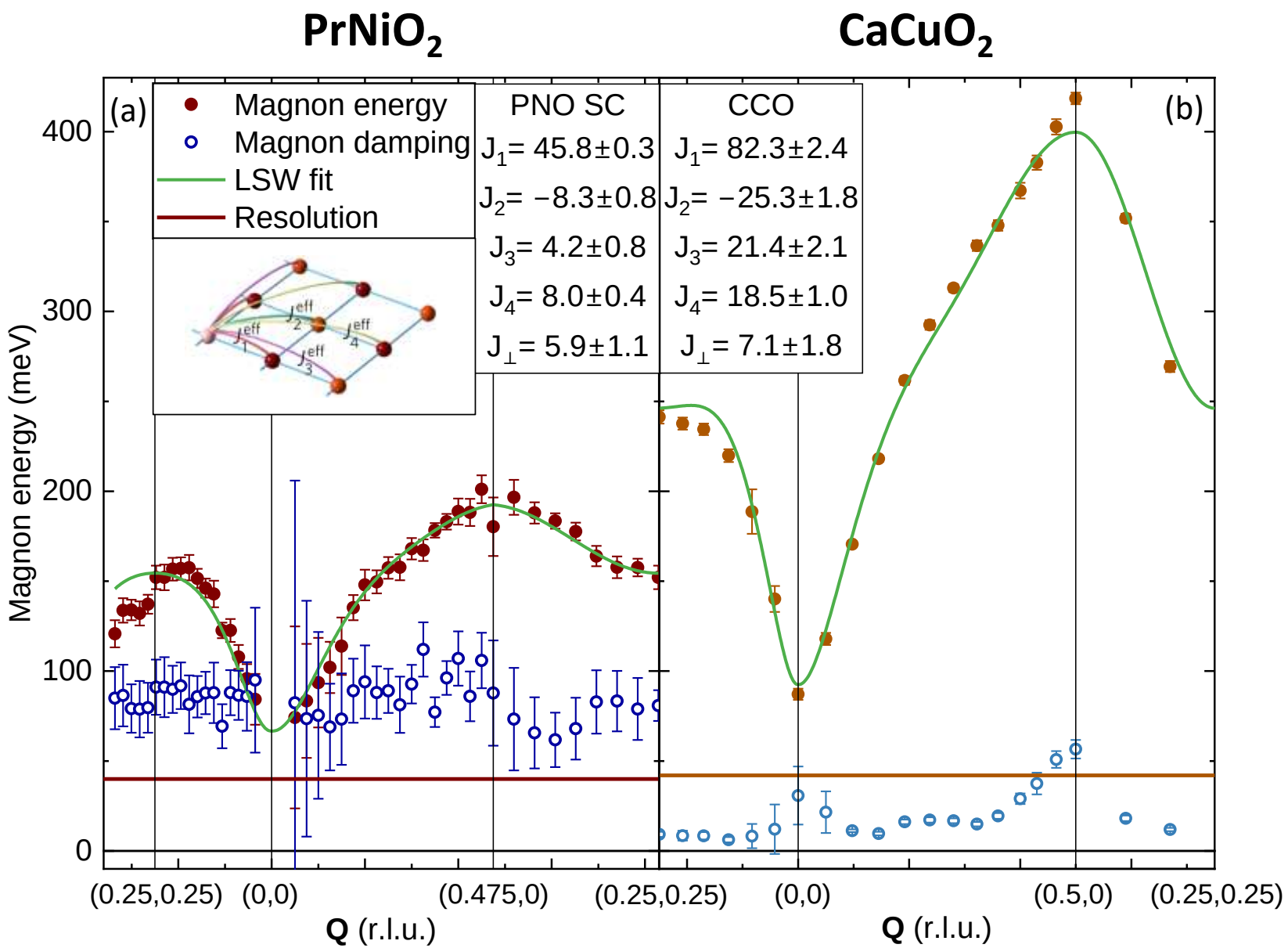


- Very clear dispersions, high quality of the samples
- Expected magnetic bandwidth: *200 meV* ca., similar to other IL nickelates
- Magnetic exchange is still strong right above Tc

Rosa et al. *Physical Review B* 110.22 (2024): 224431.



Dispersion relations



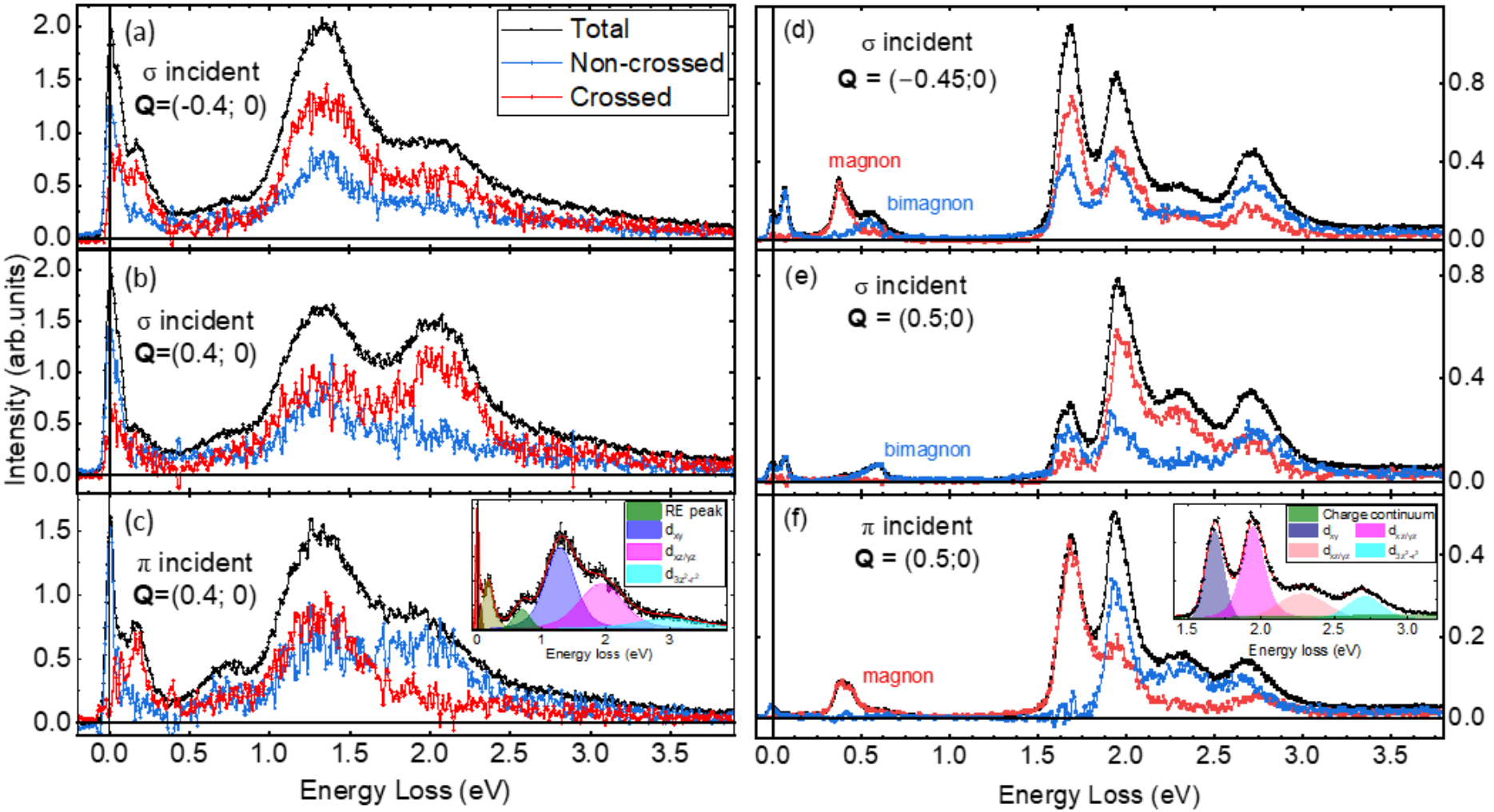
- Fitting of the dispersions with SpinW software (Matlab)
- Implemented *J*s up to fourth nearest-neighbor, plus an interlayer exchange J
- Repeated for CCO data, for comparison: PNO shows a larger degree of tridimensionality $\mathbf{J_\perp / J_1}$
- Possible explanation: opposite role of strain: $a_{CCO} < a_{STO} < a_{PNO}$
- Anomaly on the left points, toward the Bragg peak at (0.5;0.5)

Polarization-resolved RIXS

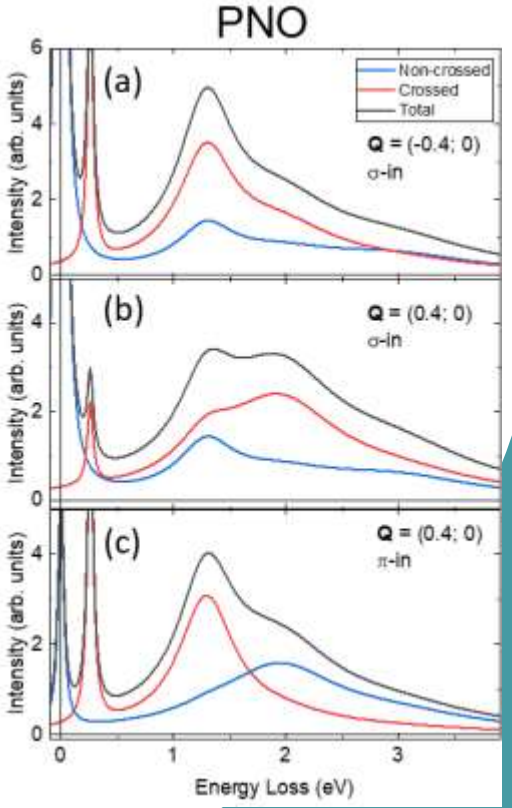
- Confirmed magnetic nature of the dispersive peak
- In CCO, four peaks instead of expected three

PNO

CCO

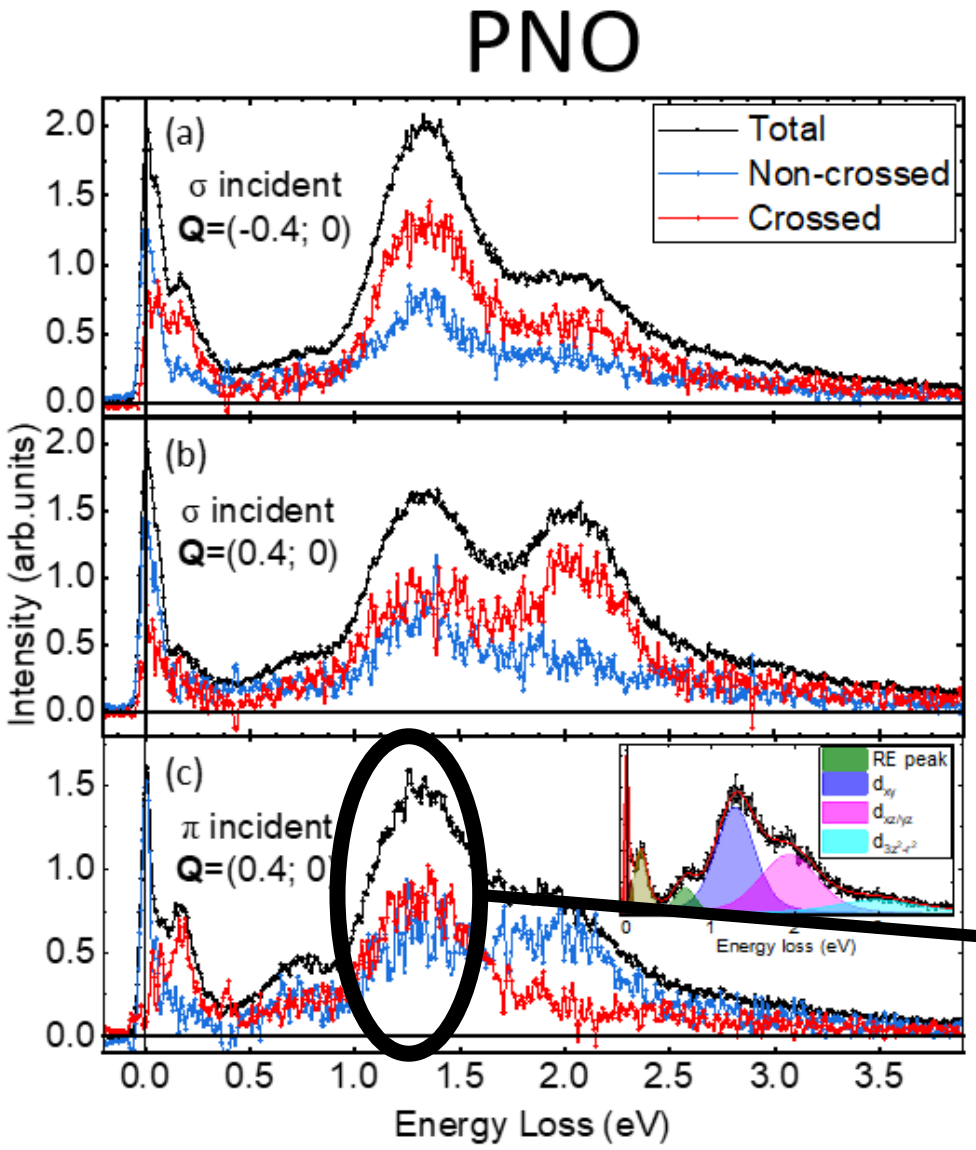


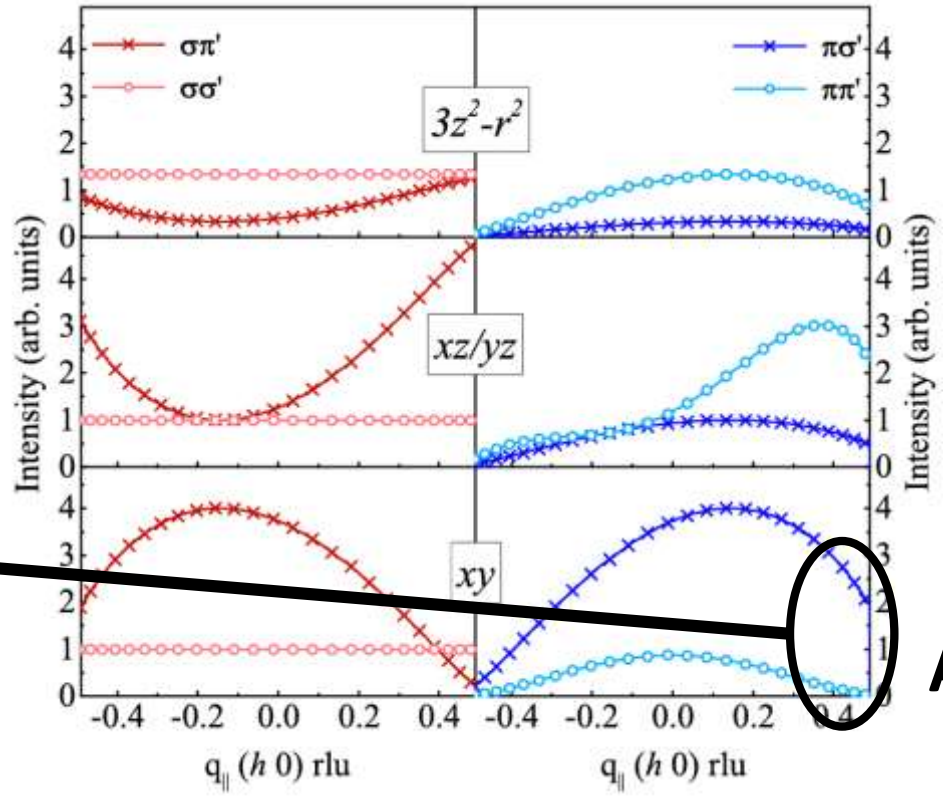
Single-ion:



Polarization-resolved RIXS

- General agreement with single-ion cross-section calculations
- DIscrepancy: *dxy* in H polarization
- *dxy* orbital appears to be more hybridized with Pr *5d* states



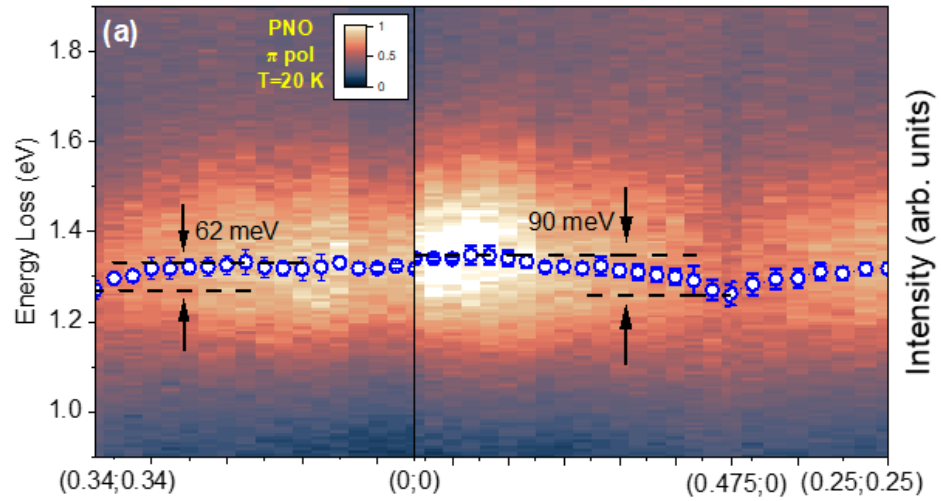


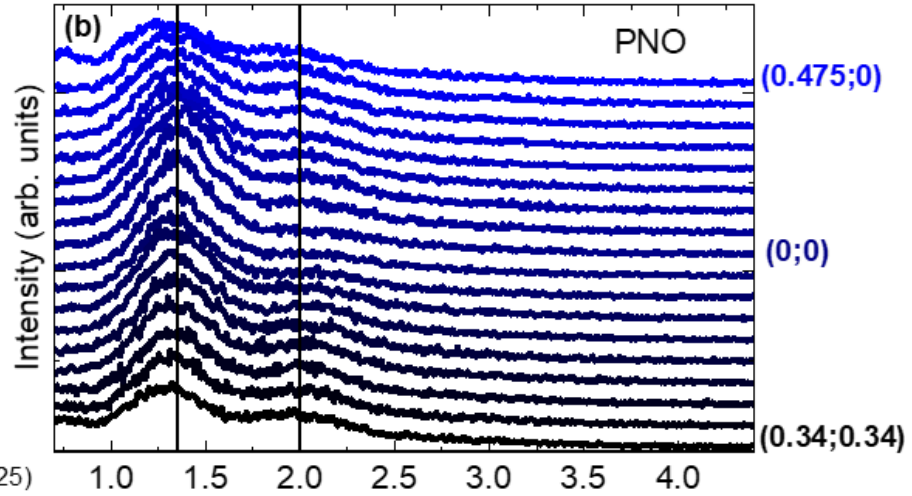
Anomaly

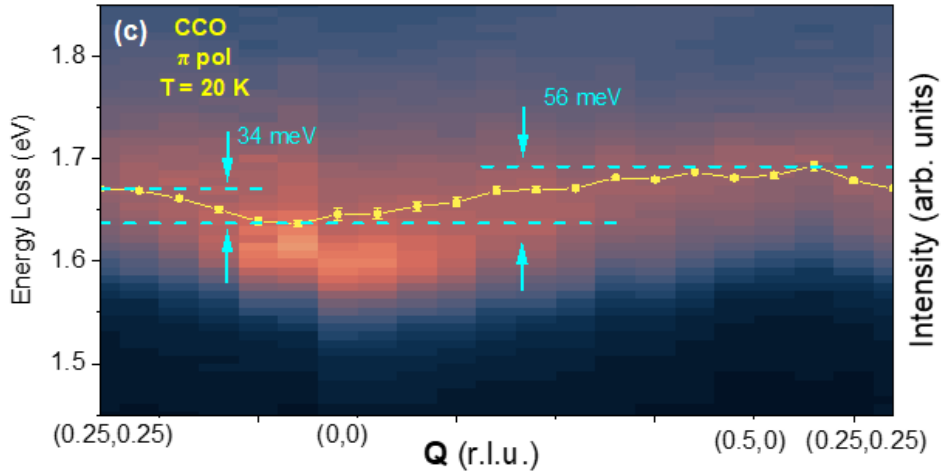
Fumagalli et al., *Physical Review B* 99.13 (2019): 134517

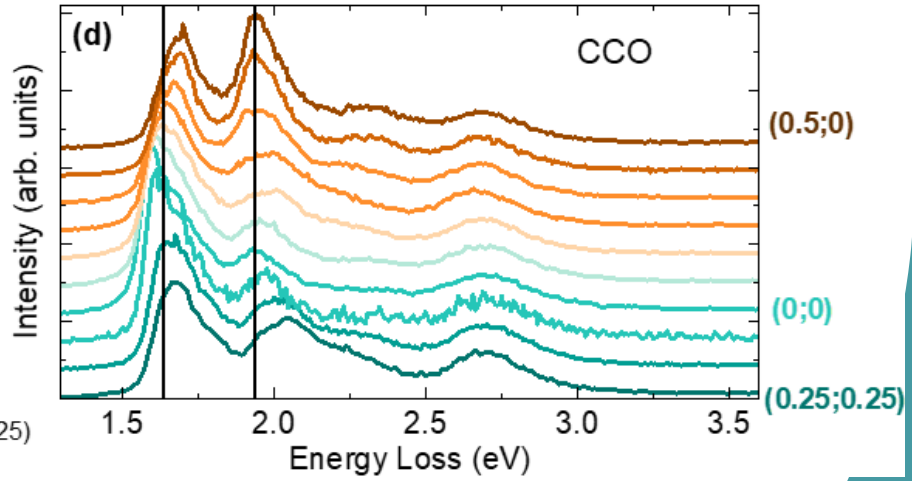
Orbital dispersion

- Evidence of dispersion of orbital peak
- Opposite trend to CCO, similar instead to 1D models
- In 2D systems like CCO, spin-orbital separation was attributed to next-nearest neighbor exchange
- Our findings might require a different explanation







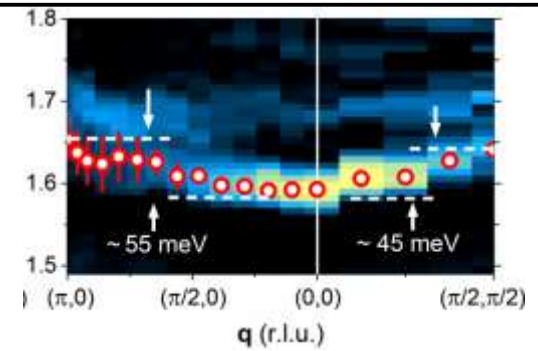


d_{xy} dispersion:

[meV]	PNO	CCO
(H;0)	90±9	56±11
(H;H)	62±6	34±11

$CaCuO_2$

Martinelli et al., Physical Review Letters 132.6 (2024): 066004.



Conclusions

- We recently reported a superconducting state in a nominally undoped $PrNiO_2$ sample, with Tc between *7* and *11 K*
- The most reasonable explanation is based self-doping from Pr pockets of states close to the Fermi level
- RIXS measurements revealed the presence of relatively sharp, dispersing magnetic excitations at a temperature right above Tc. This hints at a possible coexistence of the two orders
- Opening of a gap at the gamma point reveal the tridimensional nature of magnetism in IL nickelates, so far ignored in existing literature. Retrieved a $J_{\perp}$*~6 meV* for PNO
- Polarization-resolved RIXS confirms previous assignments of orbital peaks. We performed an extensive comparison with CCO cuprate, showing better agreement of PNO with single-ion calculations
- We furthermore reported a *~90 meV* dispersion in momentum for the first *dd* peak. Collective nature of dd peaks constitutes an evidence of spin-orbital separation
- Necessity of revising the IL nickelates phase diagram

Acknowledgments

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- Leonardo Martinelli (now at University of Zurich)

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Samples:

IPCMS Institut de Physique et Chimie des Matériaux de Strasbourg

- Daniele Preziosi
- Hoshang Sahib

TOR VERGATA UNIVERSITÀ DEGLI STUDI DI ROMA

- Daniele Di Castro

Thank you!

Easy-plane anisotropy fitting

(La2NiO4)

Biało, Izabela, et al., *Communications Physics* 7.1 (2024): 230.