Resonant soft-x-ray diffraction from electronic order

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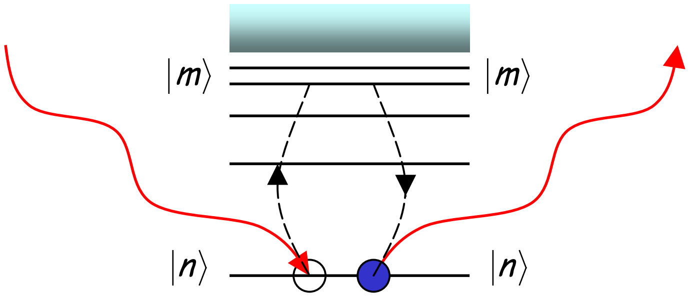
- Resonant soft x-ray diffraction
- Stripe order in $La_{2-x}Sr_xNiO_4$
- Charge and orbital order in Fe_3O_4

Resonant Scattering



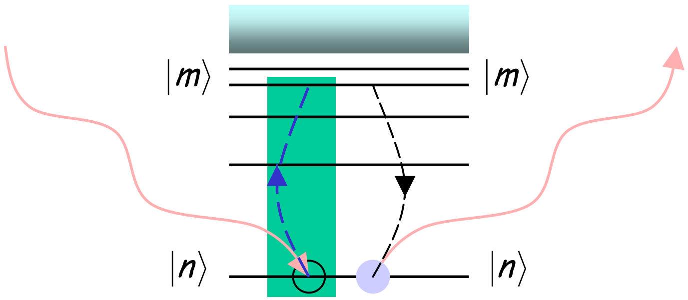
Resonant scattering

- probing the intermediate states
  - element specific
  - sensitive to oxidation state
  - sensitive to orbital orientation
  - sensitive to magnetic moments

\+ probing a certain periodicity

= spectroscopically resolved structure study

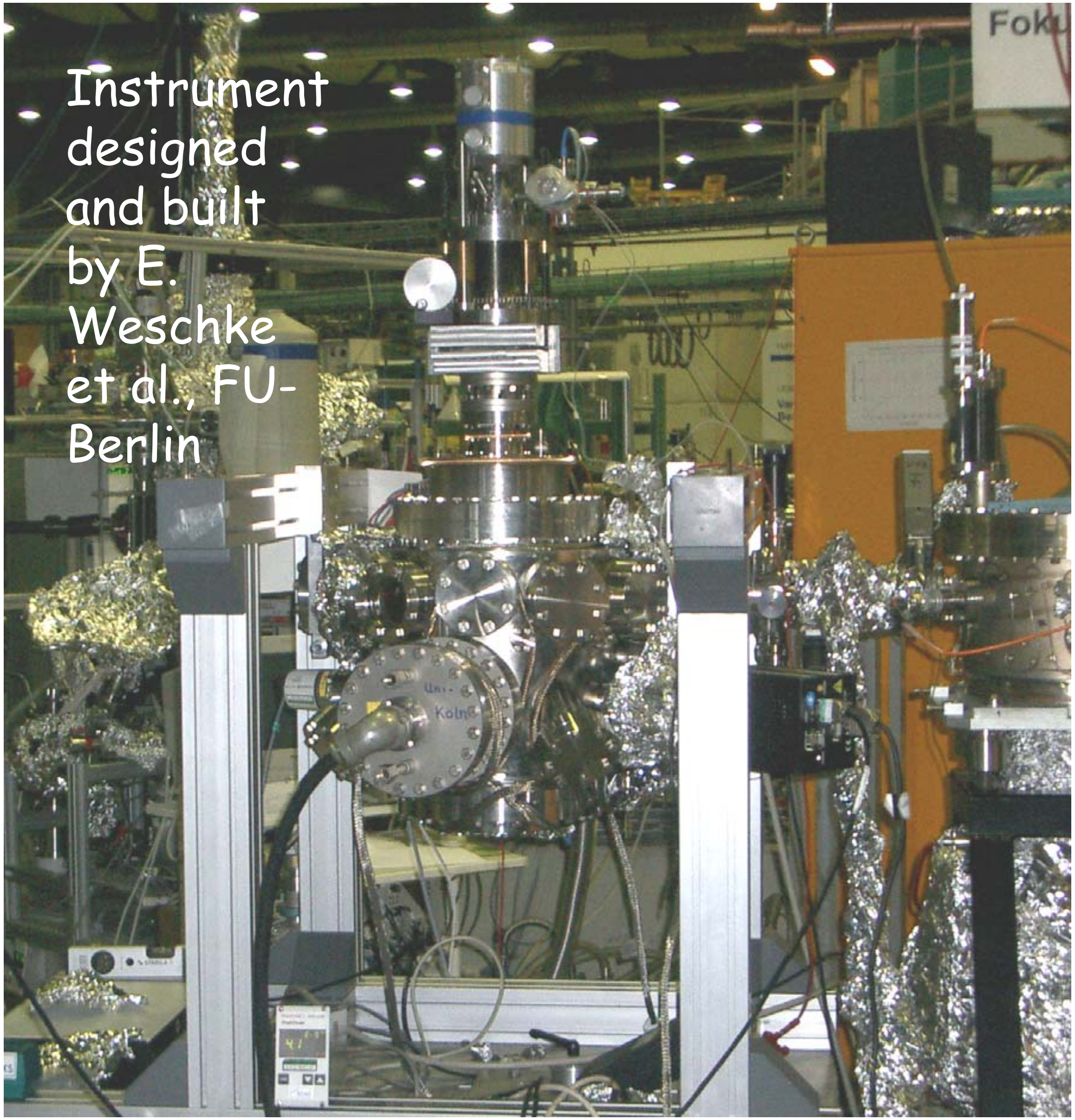
Resonant soft x-ray scattering

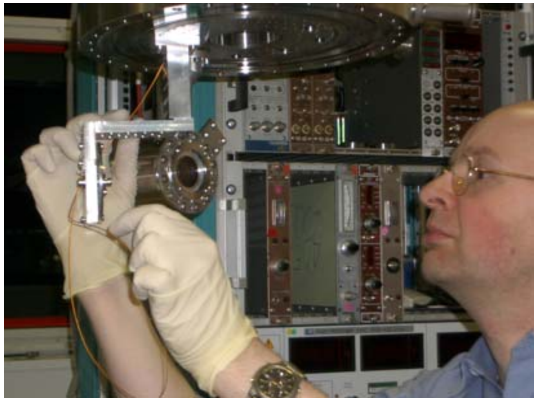


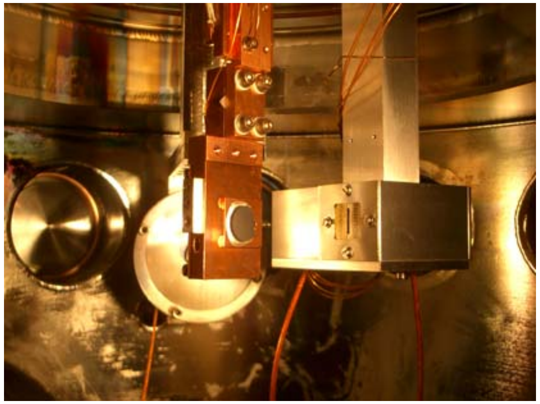
Ion	Excitation	Edge
Transition metals	$2p \rightarrow 3d$	L_2, L_3
Oxygen	$1s \rightarrow 2p$	K
Lanthanides	$3d \rightarrow 4f$	M_4, M_5

- intermediate state determines system properties
- well understood transitions, theoretical treatment available
- very high scattering cross sections
- long photon wavelength -> small accessible momentum space

Soft x-ray diffractometer at BESSY

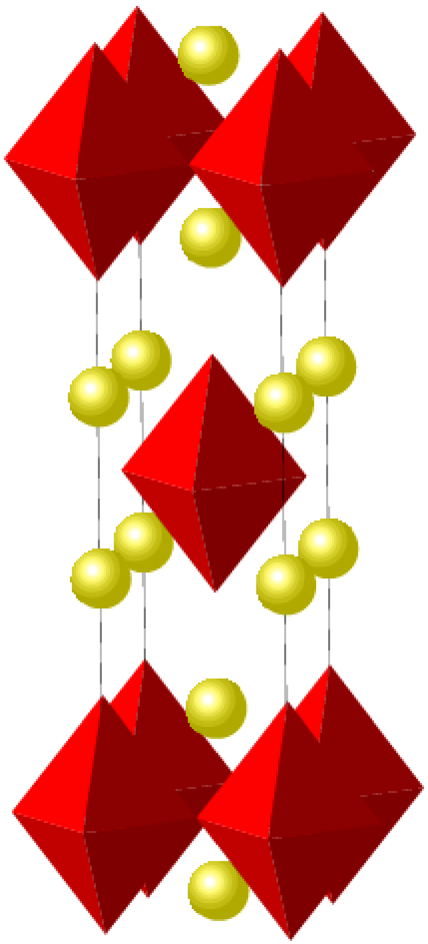


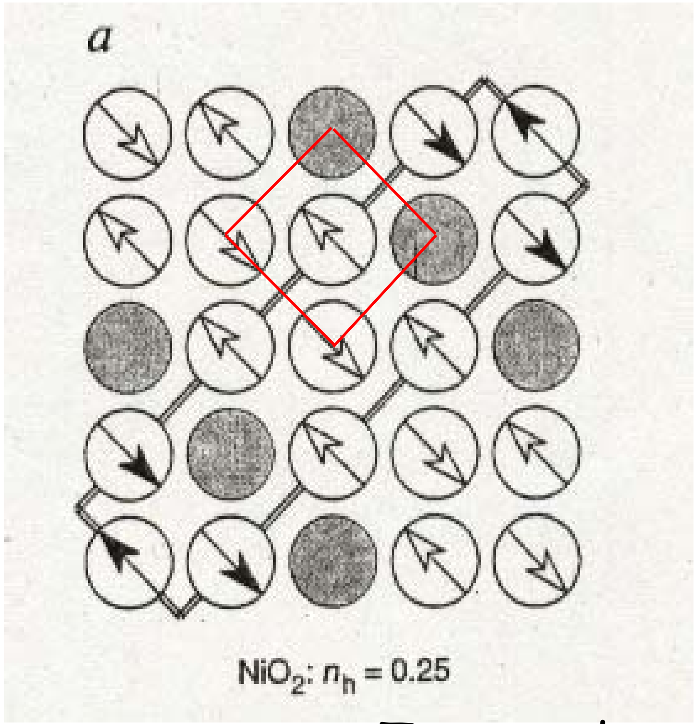


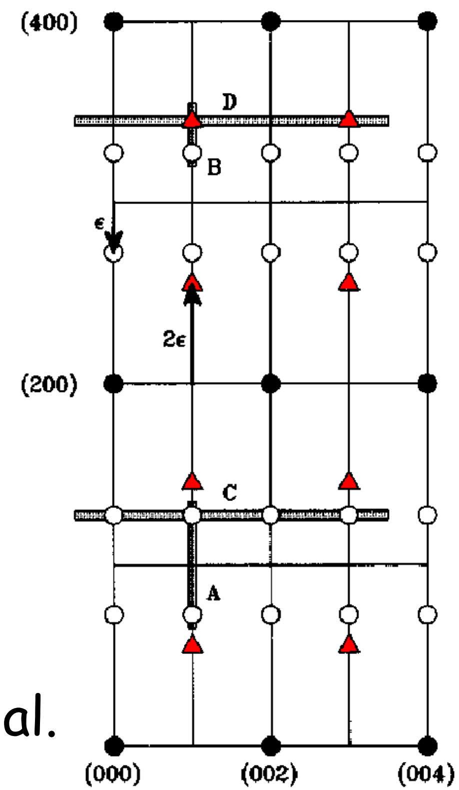


$La_{2-x}Sr_xNiO_4$

- first neutron study of charge order
- isostructural to $La_{2-x}Sr_xCuO_4$
- not superconducting
- stripe phase at low temperatures



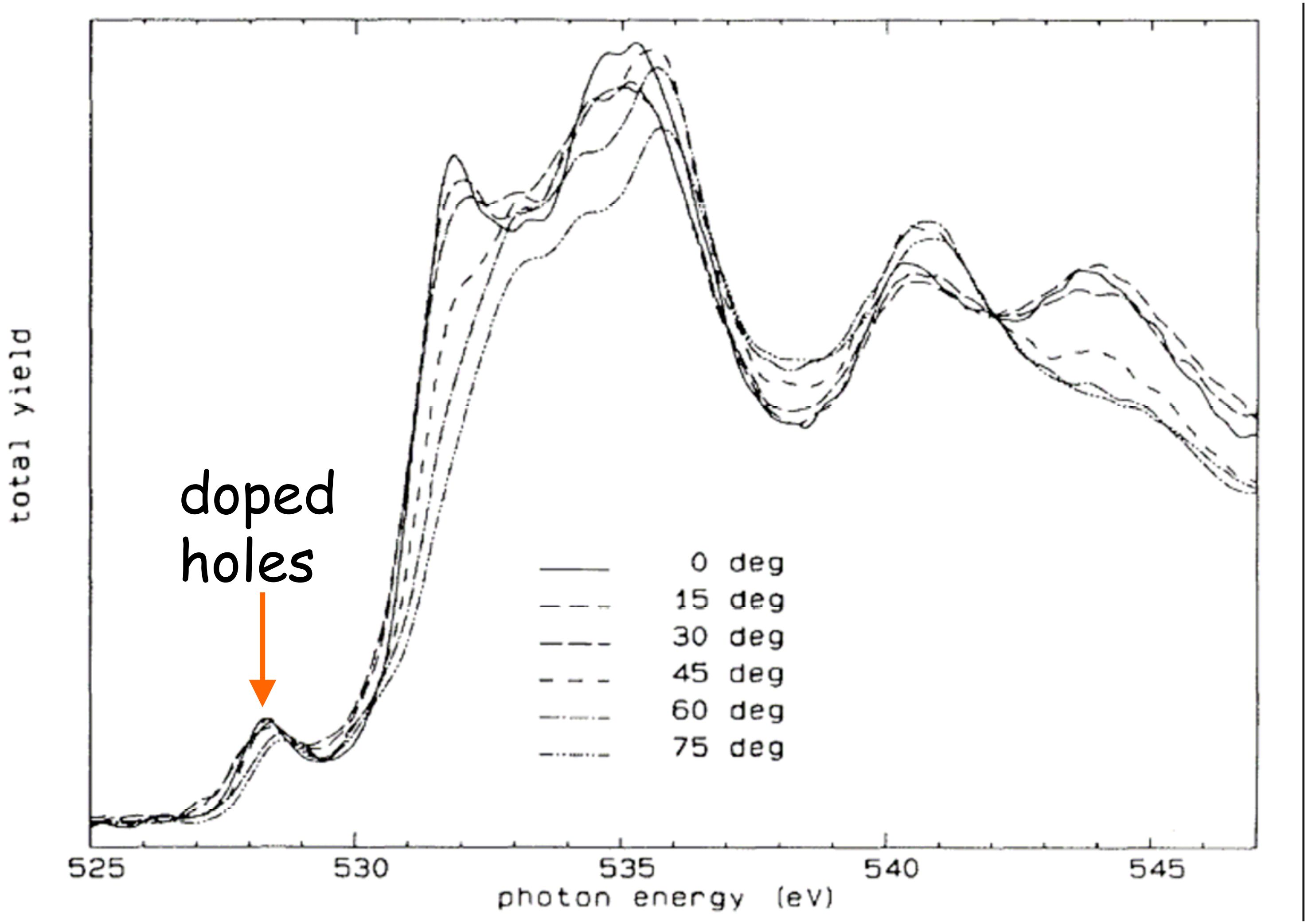




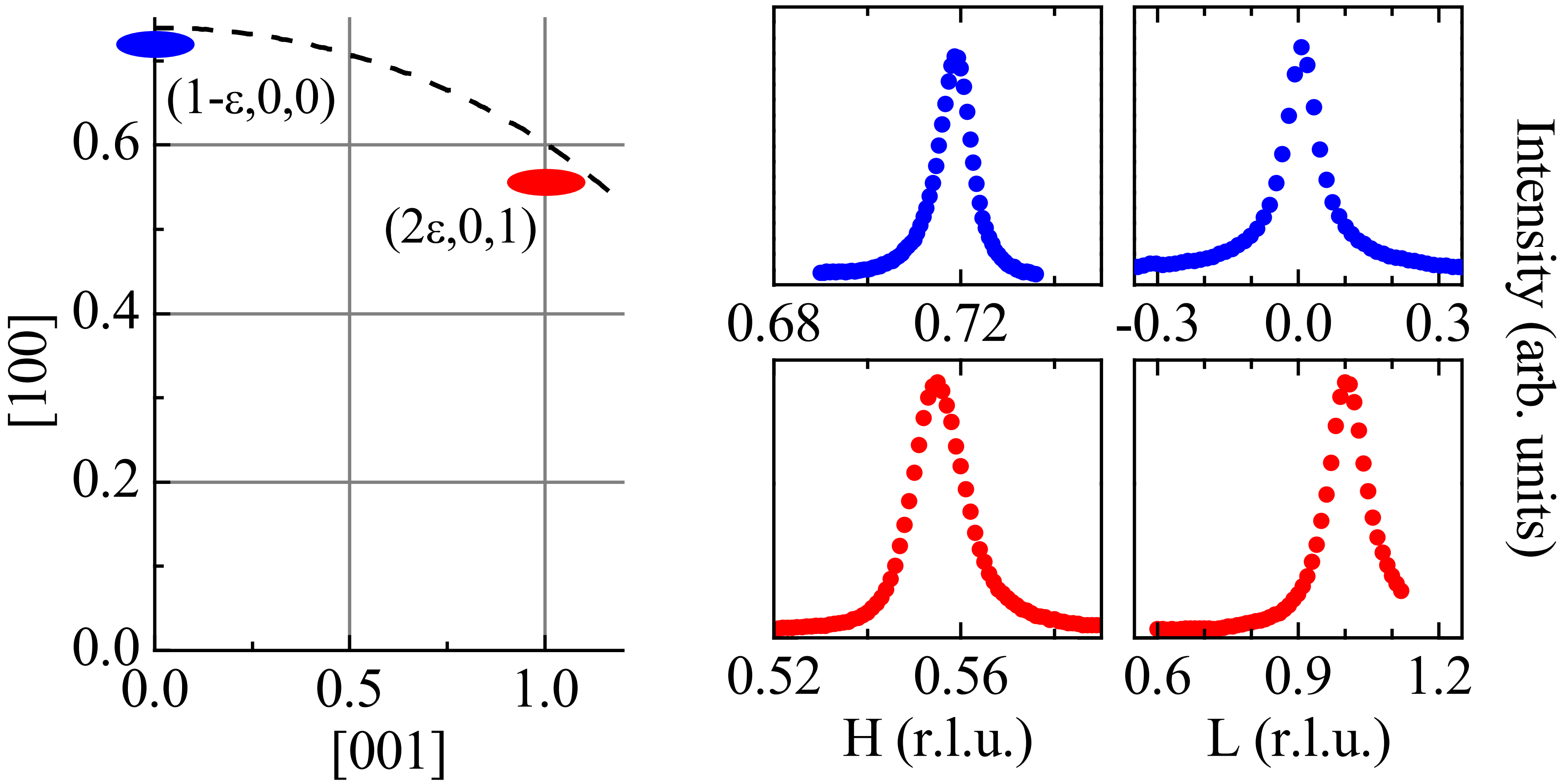
Tranquada et al.

Symmetry of the doped holes ?

O-K (1s->2p) XAS, P. Kuiper et al., PRB 44, 4570 (1991)

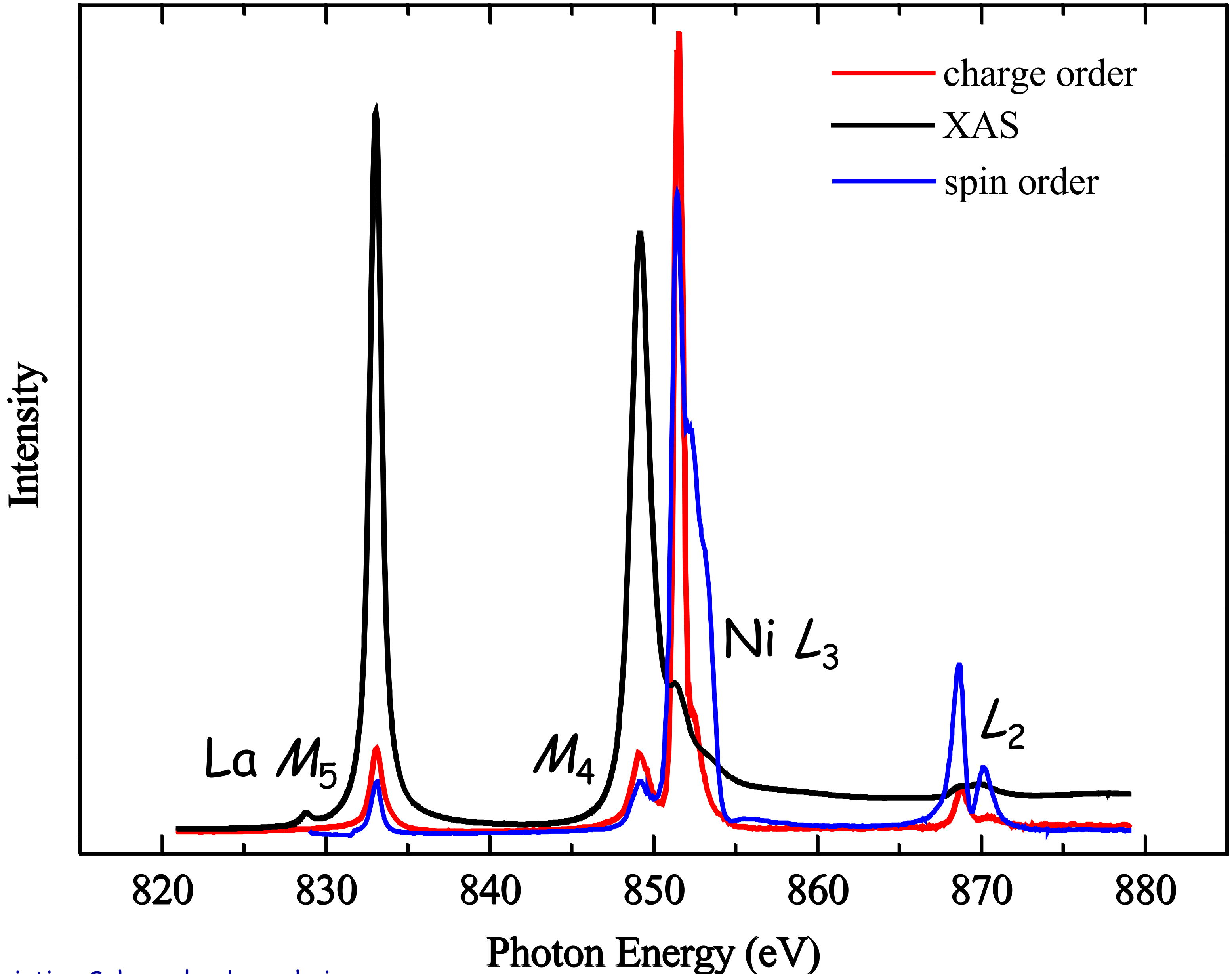


Well-developed order



	ξ_a (Å)	ξ_c (Å)
spin	300	50
charge	200	40

Energy dependence



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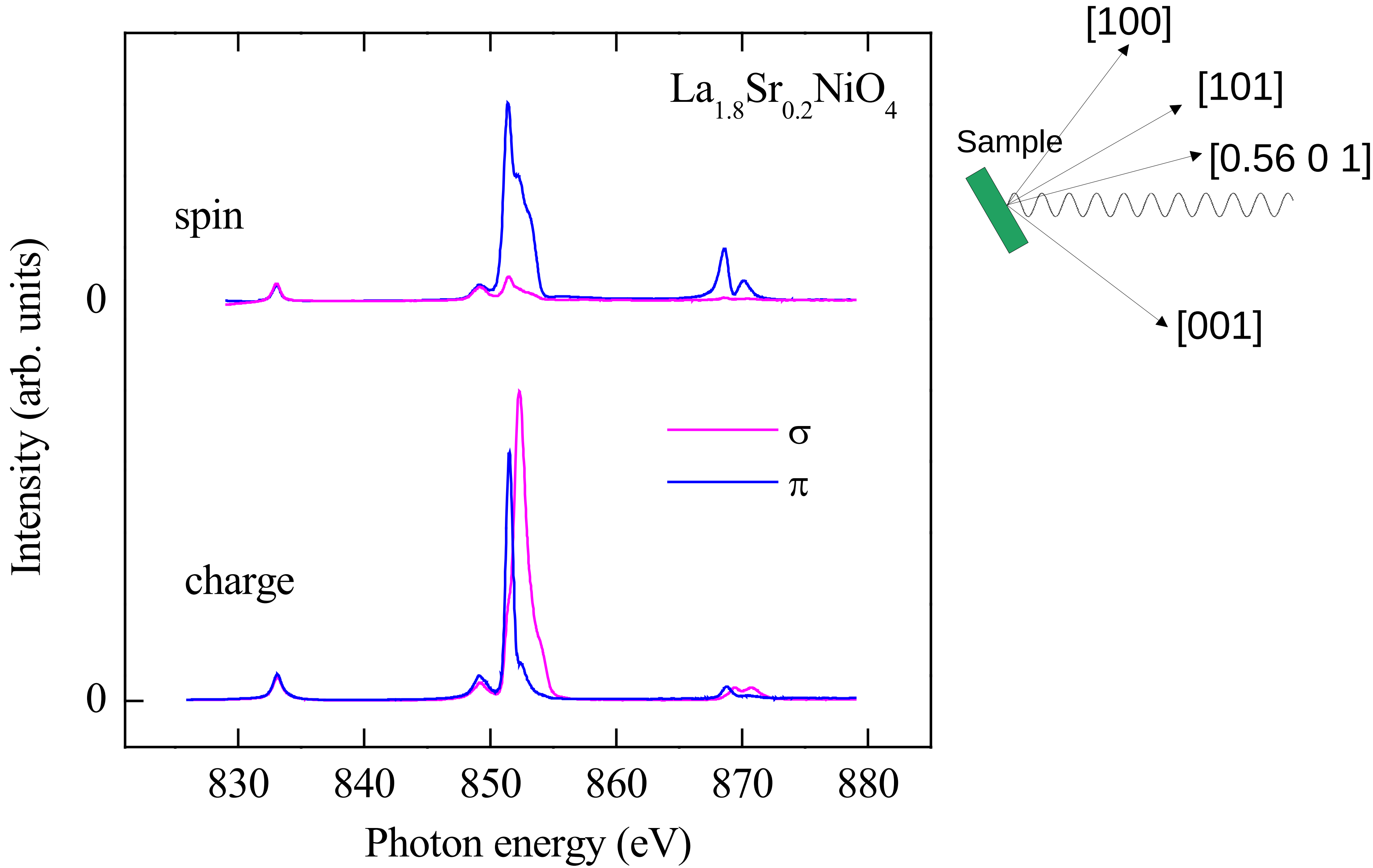
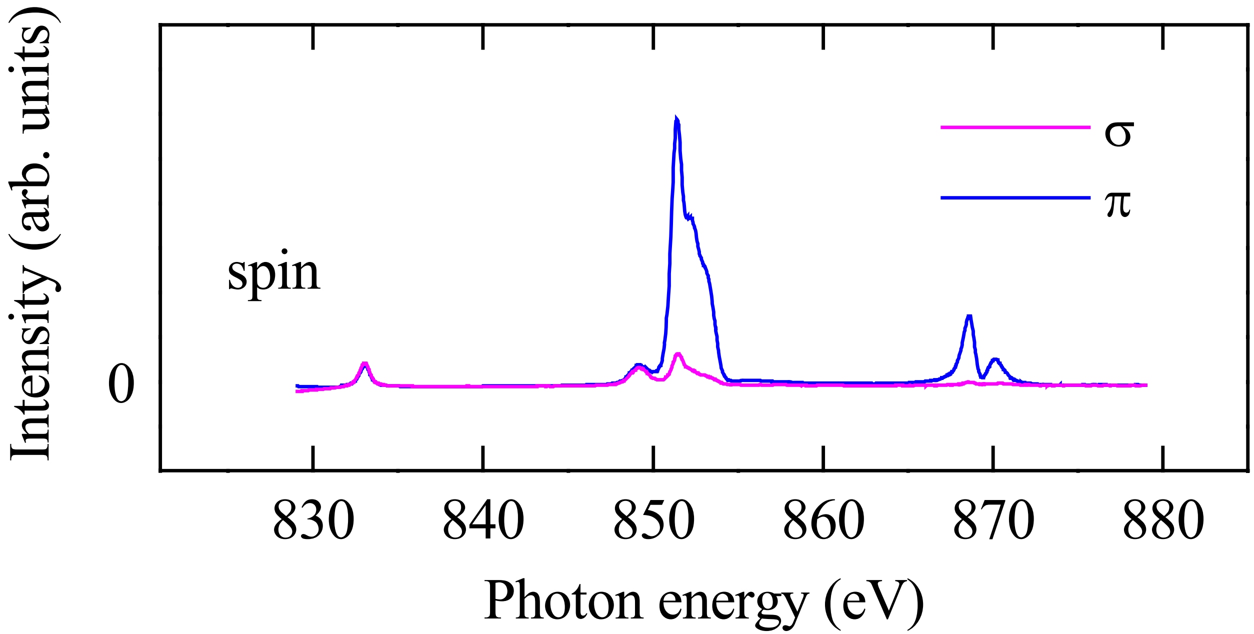
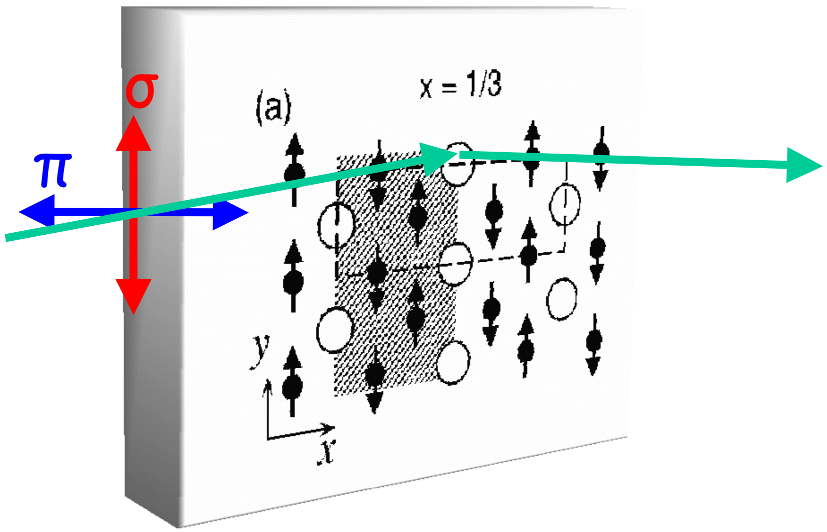
Polarization dependence



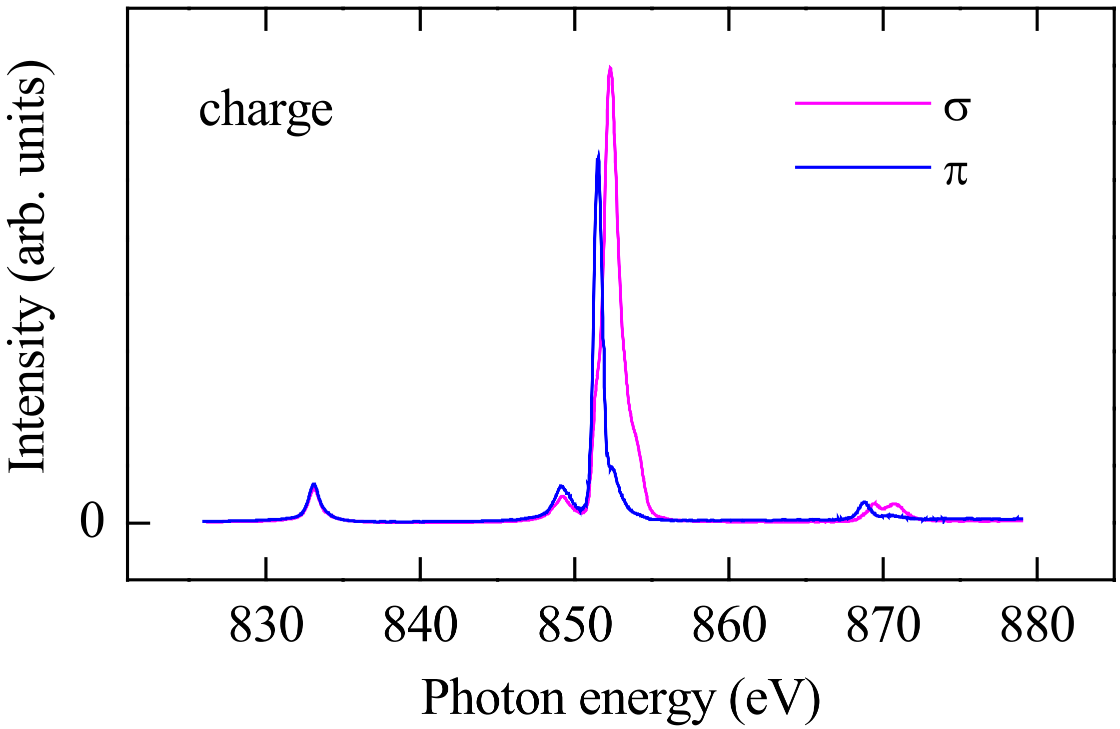
Polarization dependence

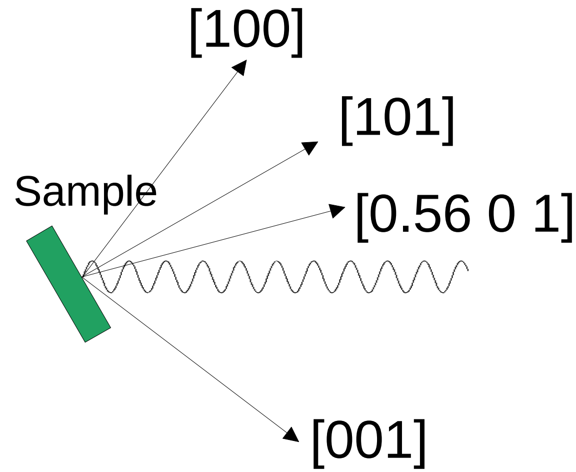


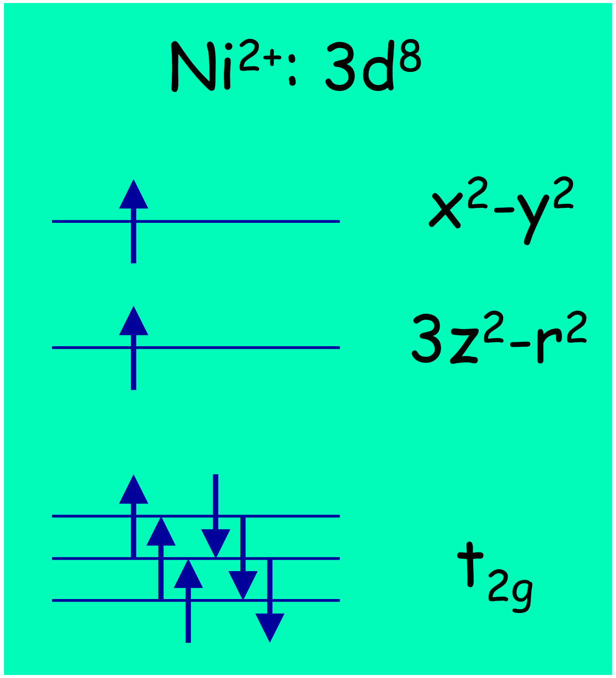


$$f_{mag} \propto \left(\hat{\varepsilon}' \times \hat{\varepsilon}\right) \cdot \vec{m}$$

Polarization dependence

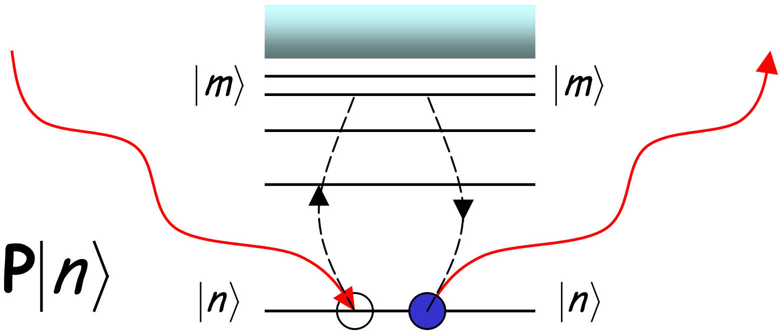






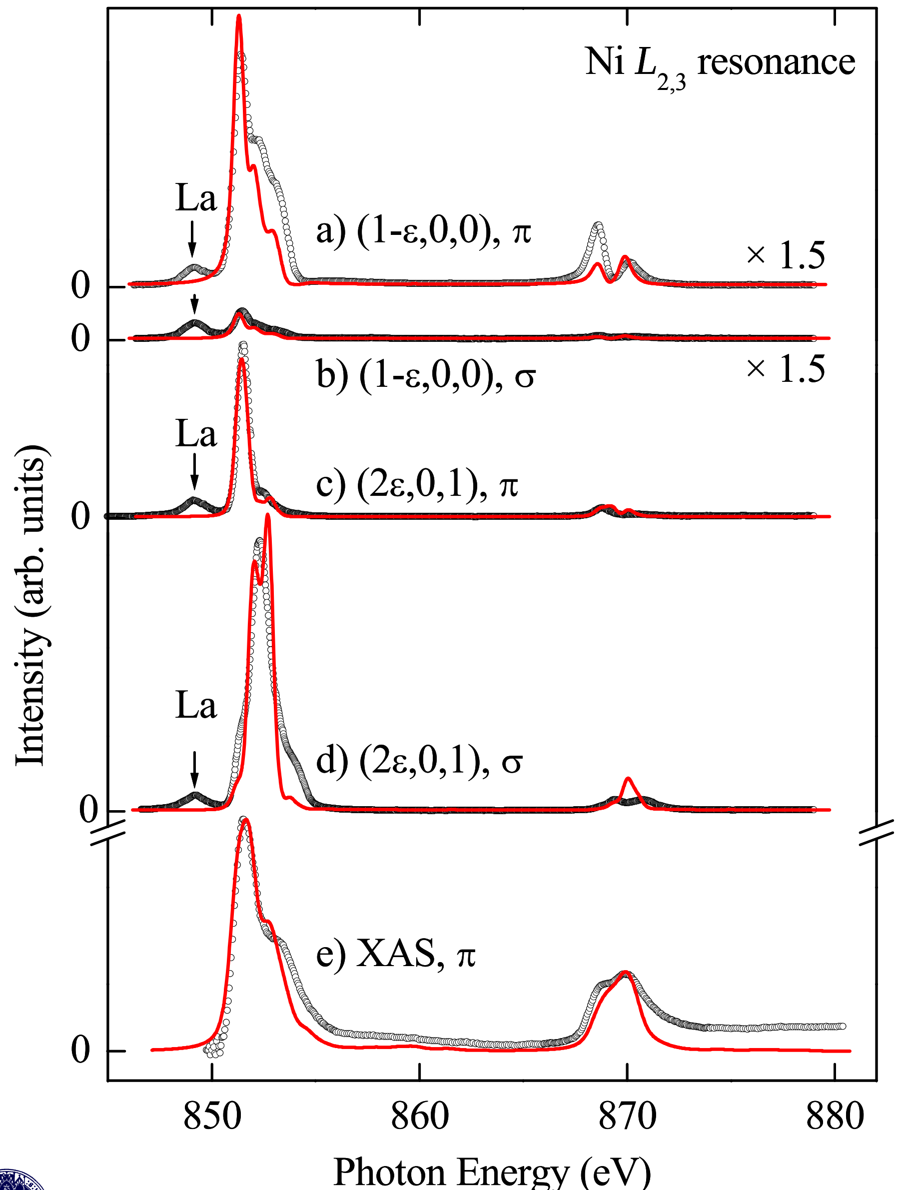
Cluster calculation

$$F_n(\omega) = k^2 \langle n|\mathbf{P} \frac{1}{E_n - \hbar\omega - H_m + i\Gamma_m/2} {}^{\dagger}\mathbf{P}|n\rangle$$



- excited state is strongly localized → NiO_6-cluster calculation possible
- full atomic multiplet calculation
- |n>, E_n: 3*d* - 3*d* Coulomb, exchange, spin-orbit interaction, 3*d* - O-2*p* hybridization
- H_m: 2*p* core-hole - 3*d* Coulomb and exchange interaction, 2*p* spin-orbit interaction

Results



one single model describes all three spectra

- hole mainly on in-plane oxygen ions -> strong 2d character
- hole-rich: 7.9 *d*-electrons, hole-poor: 8.2
- afm coupling between Ni and hole spins (like Zhang-Rice singlet)

very similar to cuprates

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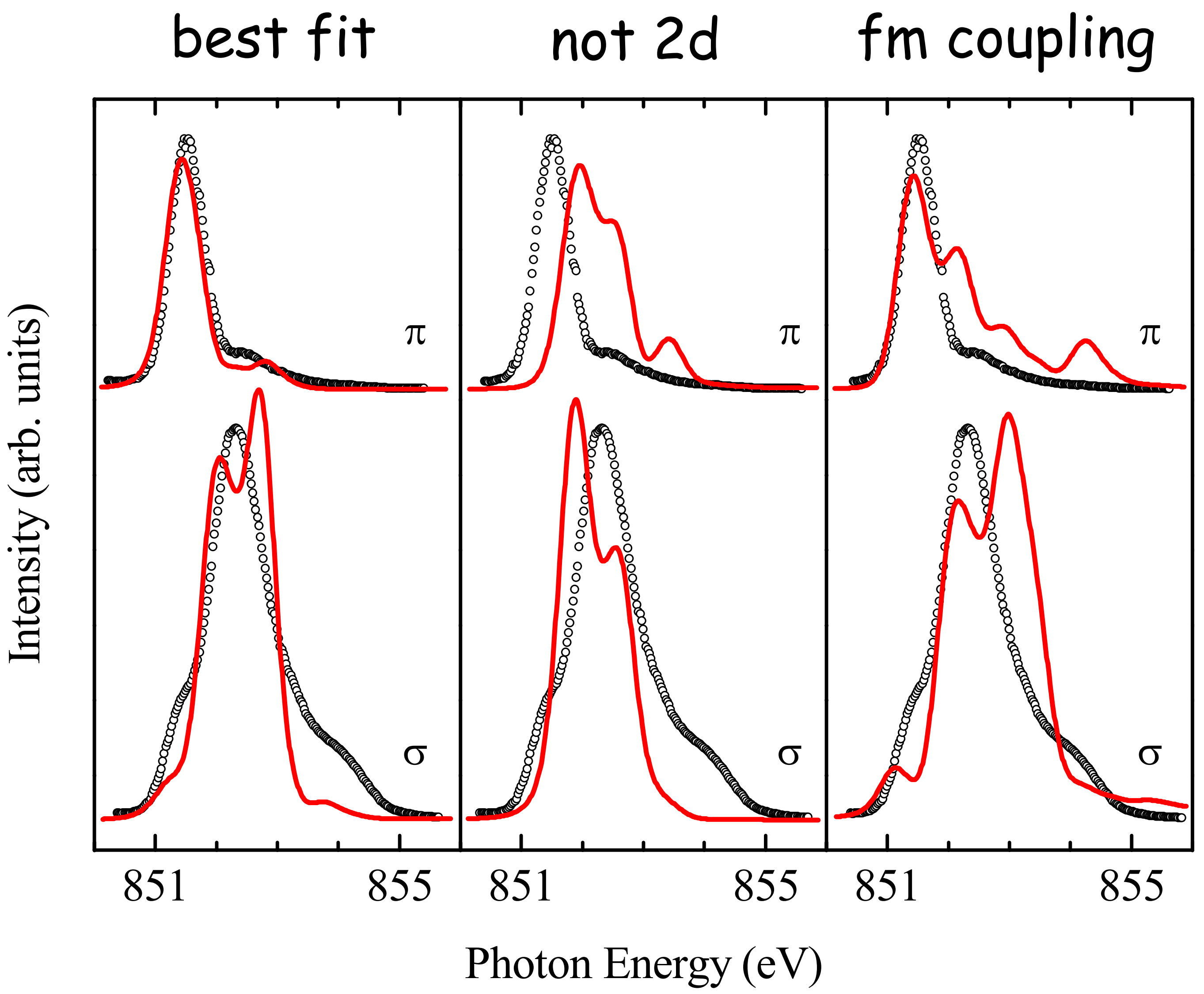
Sensitivity



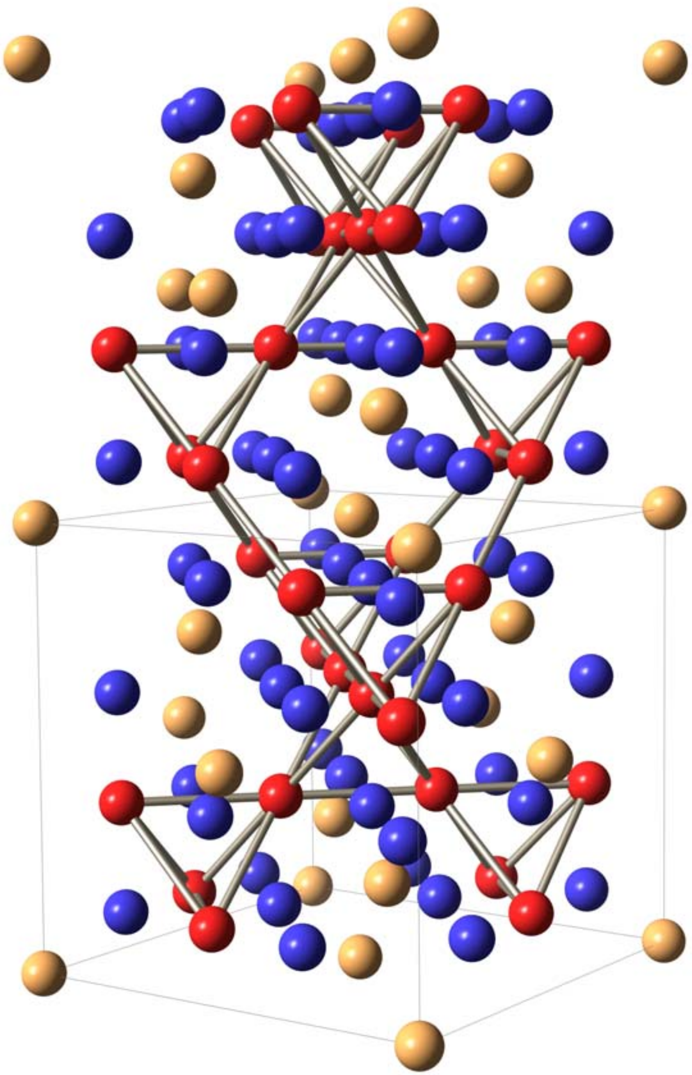
Summary so far

- direcly observed charge ordering in $La_{1.8}Sr_{0.2}NiO_4$
- scattering contrast due to difference in the electronic state of the Ni ions
- spectroscopic information about the ordered part of the system
- realistic microscopic theory available
- 2-dimensional character of the doped holes
- holes reside mainly on the oxygen ligands and couple antiferromagnetically to the canted Ni spin

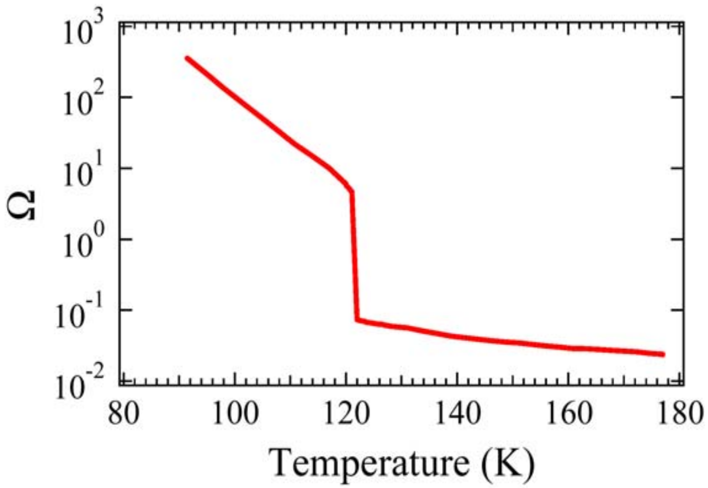
PRL 95, 156402 (2005)

Magnetite (Fe_3O_4)

- **Inverse spinel structure above 122 K**
  - 1/3: tetrahedral (A-site) Fe^{3+}
  - 2/3: octahedral (B-site) Fe^{3+}, Fe^{2+}
- A-site ↓ , B-site ↑ , $T_C \sim 860K$

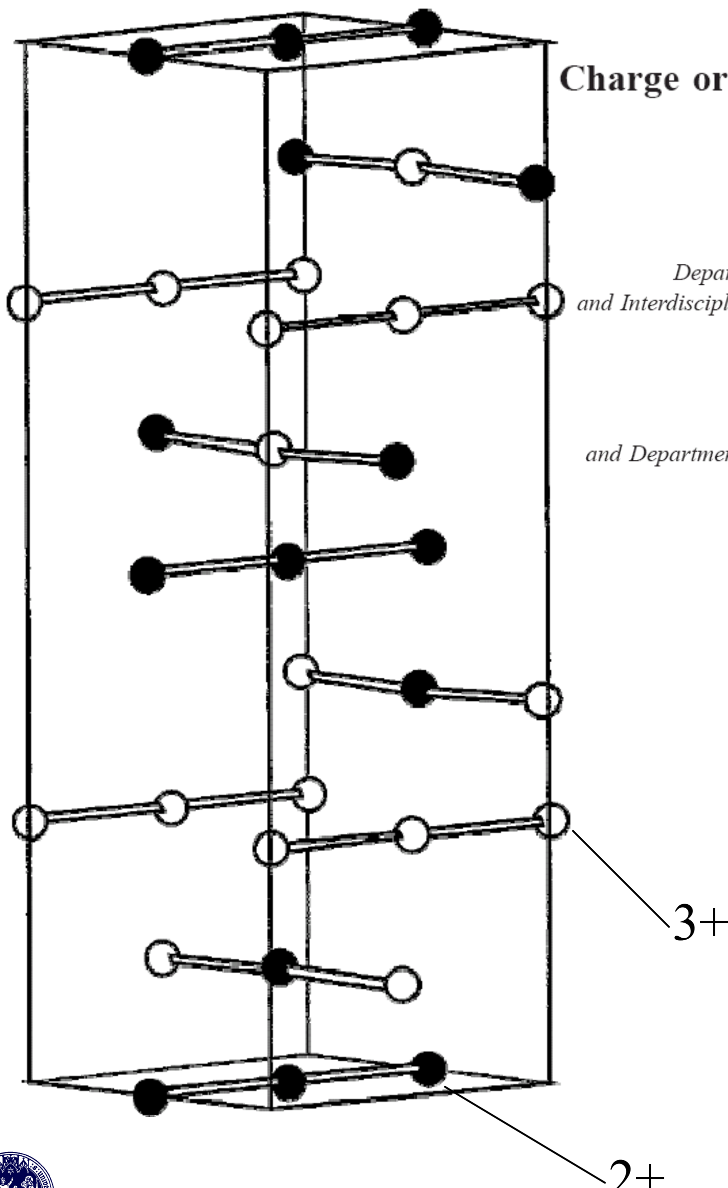


- **Verwey transition , T_V = 122 K**



Electronic structure of the low-temperature phase still unknown

Proposed structure



PHYSICAL REVIEW B **66**, 214422 (2002)

Charge ordered structure of magnetite Fe_3O_4 below the Verwey transition

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(Received 9 August 2002; published 31 December 2002)

Oxygen *K*-edge resonant diffraction

PRL 96, 096401 (2006) PHYSICAL REVIEW LETTERS week ending 10 MARCH 2006

Charge-Orbital Ordering and Verwey Transition in Magnetite Measured by Resonant Soft X-Ray Scattering

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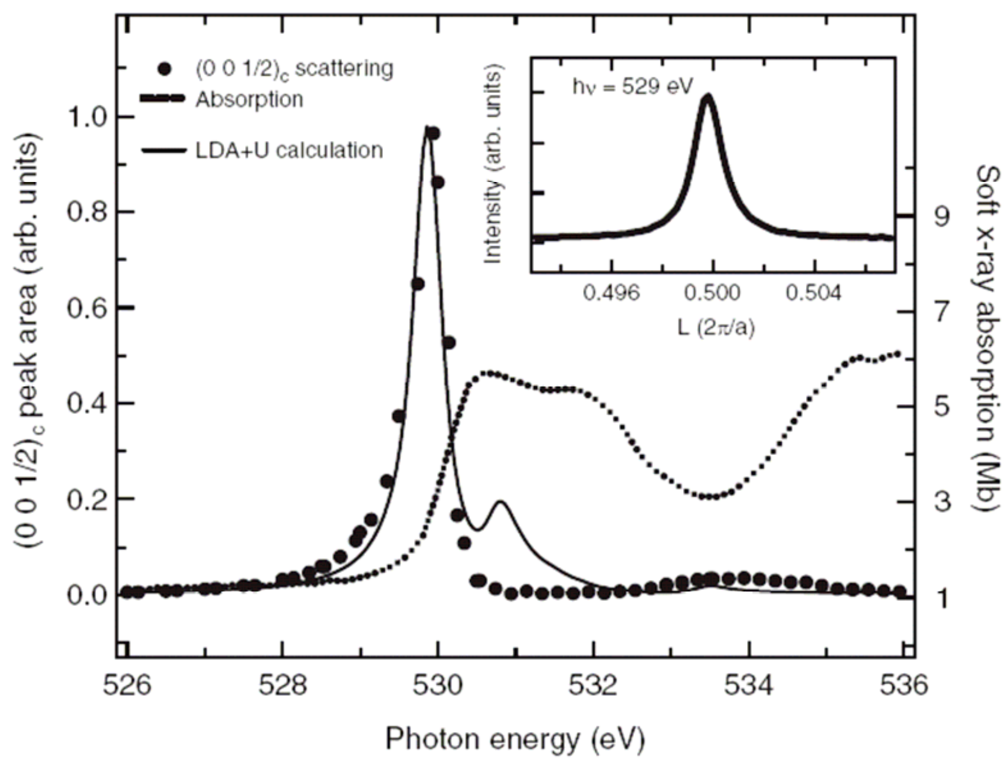
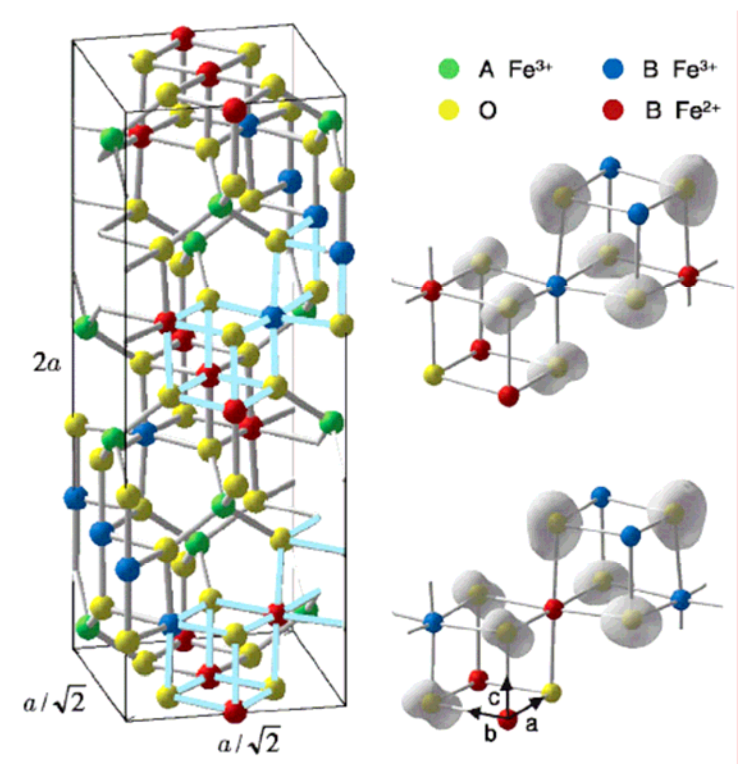
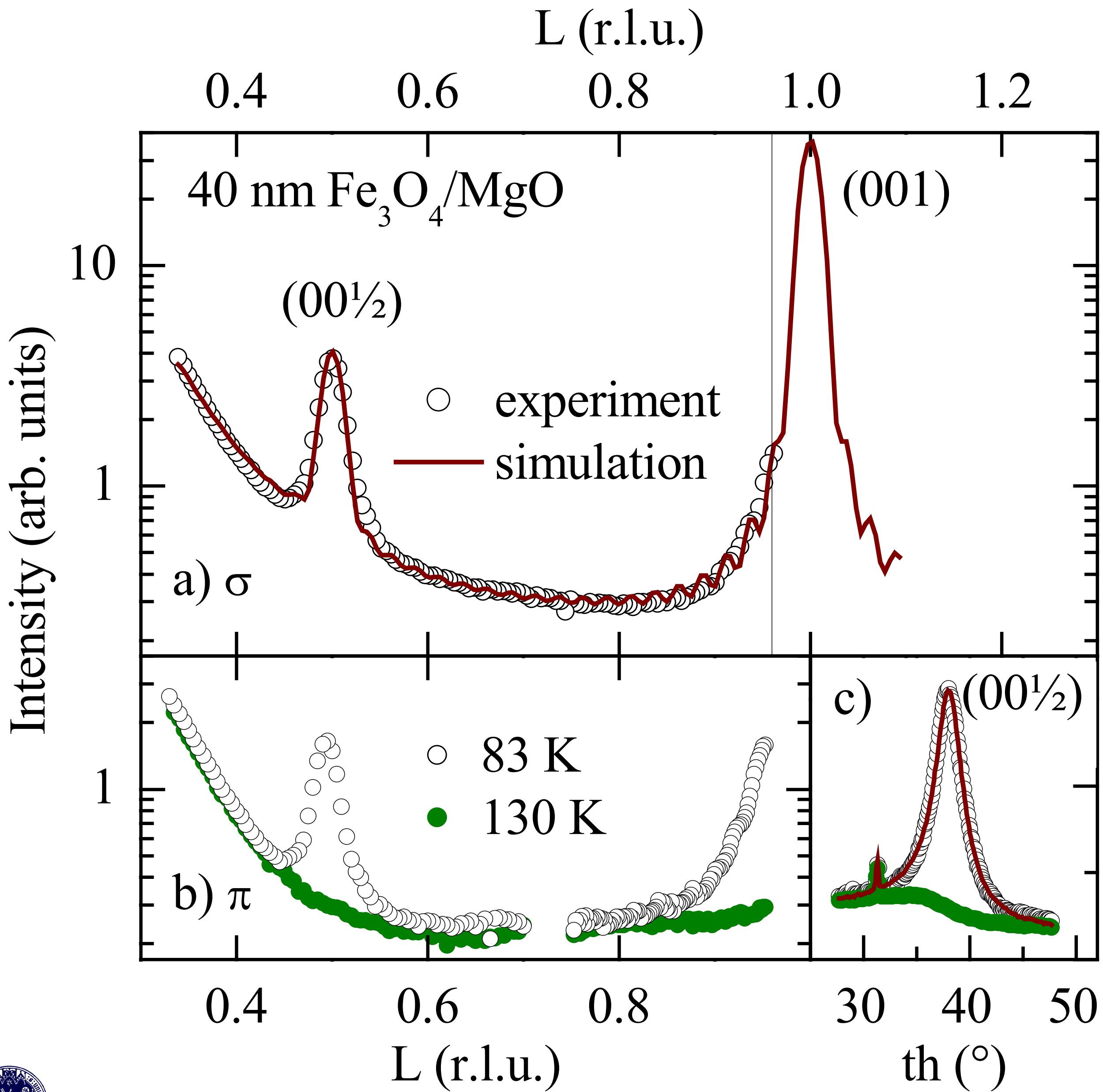




40 nm Magnetite (Fe_3O_4) / MgO



peak broadening in thin film -> (001/2) and (001) observable

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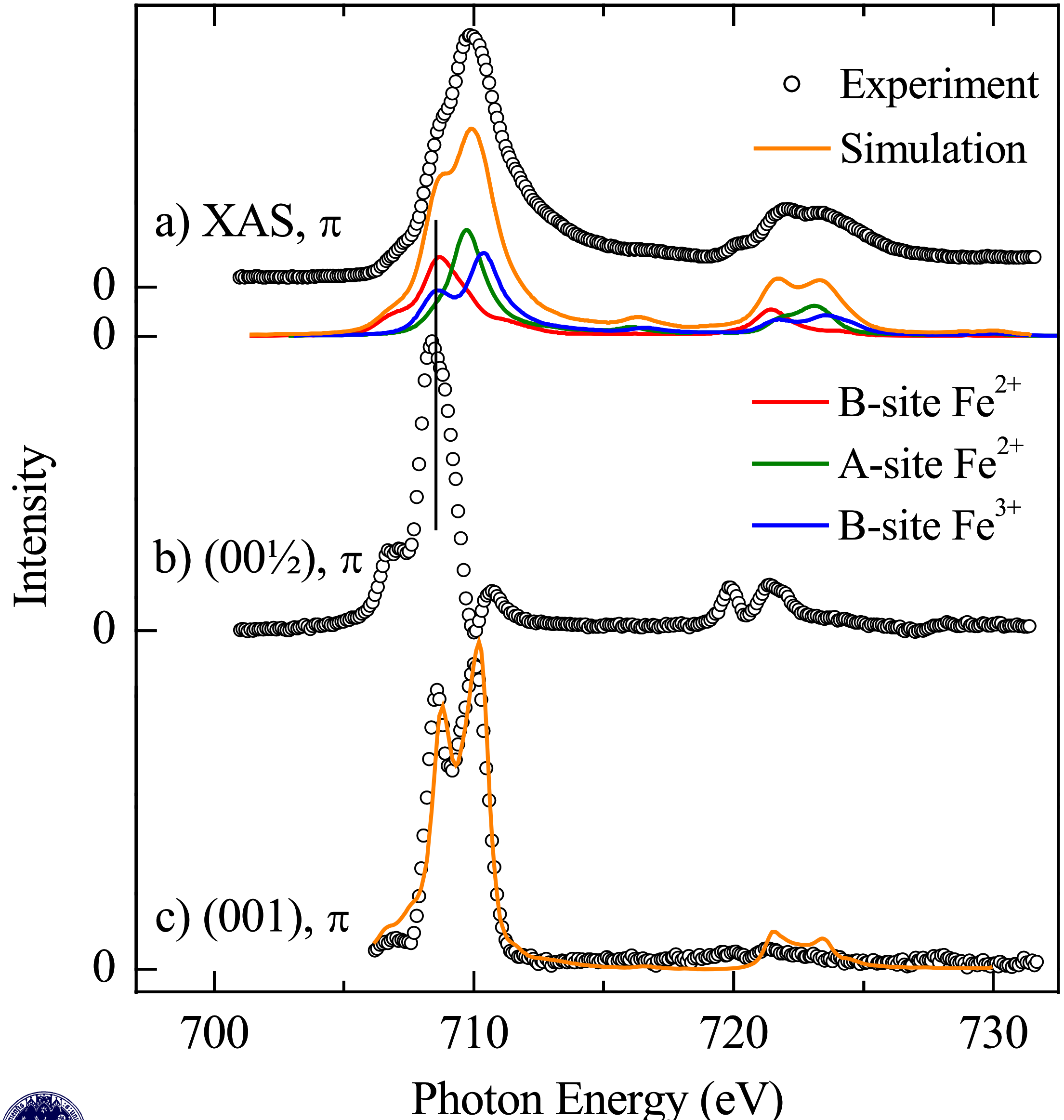
40 nm Magnetite/MgO



(001) resonates at both B-site absorption maxima: charge order

(001/2) resonates only at the 2+ B-site absorption maximum: only orbital degree of freedom

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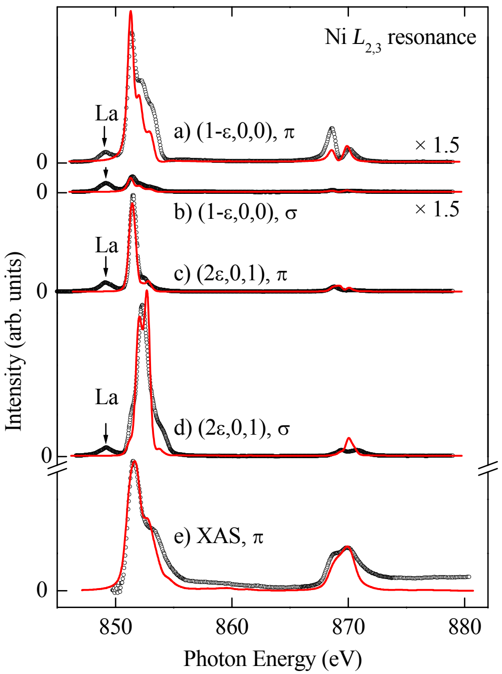
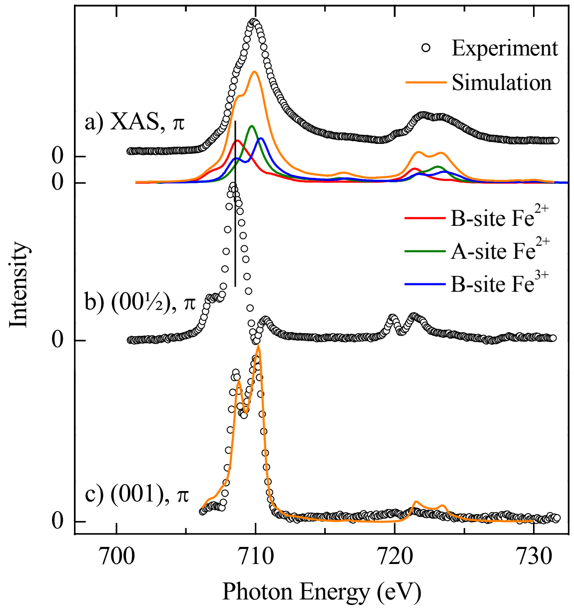
Conclusion

- experimental approach for spatial modulations of the electronic state
- particularly powerful at transition-metal $L_{2,3}$ and rare-earth $M_{4,5}$ resonances -> microscopic theory
- charge and spin order in $La_{1.8}Sr_{0.2}NiO_4$ similar to cuprate
- charge and orbital order in Fe_3O_4 described by different modulation vectors





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