Direct observation of the low energy spectrum of dynamic charge stripes in $La_{2-x}Sr_xNiO_4$

Dmitry Reznik

University of Colorado-Boulder

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Collaborators

S. Anisimova, D. Parshall -- CU-Boulder

Daniel Lamago -- Karlsruhe Inst. Of Tech. (KIT), LLB, CEA Saclay

Karol Marty, Mark Lumsden, Jaime A. Fernandez-Baca – HFIR, ORNL

Douglas Abernathy, Barry Wells -– SNS, ORNL

G. Gu, J.M. Tranquada -- BNL

Instruments

1T-- Saclay: first important insights obtained

ARCS – SNS: Comprehensive reciprocal space maps

HB-3 – HFIR: Detailed temperature study

HYSPEC – SNS: Doping dependence

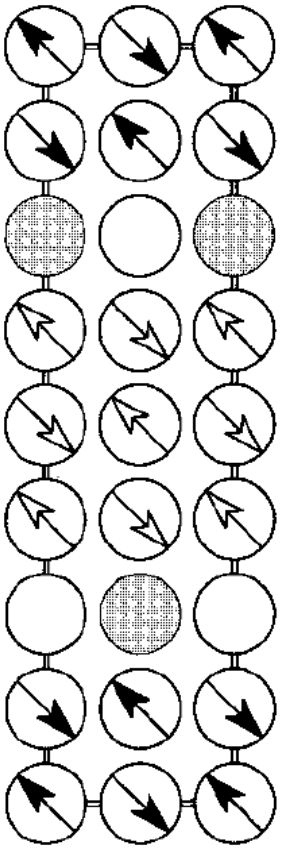
Outline

- What are (dynamic) stripes?
- Spallation Neutron Source
- Spectrum of dynamic charge stripes in the nickelates
- Temperature dependence of dynamic charge stripes
- Comparison with theory
- Conclusions

Reference: S. Anissimova et al., Nature Communications **5**, 3467 (2014)

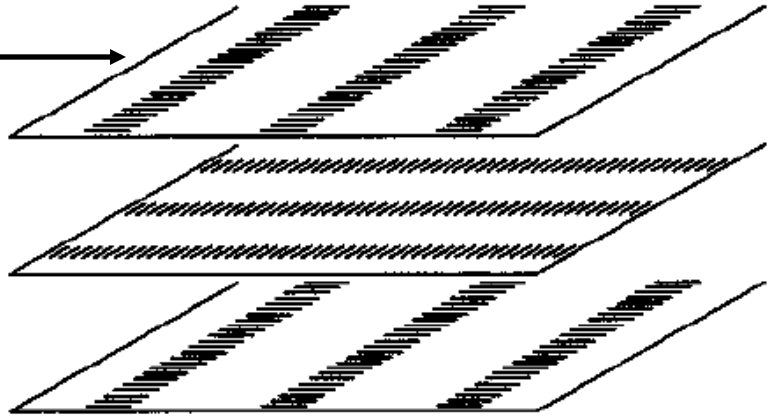
Stripe-Phase Compounds: $La_{1.875}Ba_{0.125}CuO_4$ and $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$

T_s=60K



Spin-charge segregation within a single Cu-O plane

Layer stacking



Lines of charge form a "Wigner crystal". Kinetic energy of the holes and interaction energy of the spins are both optimized

- In cuprates stripe order appears only in a few special compounds
- Key question: what is the role of DYNAMIC stripes? Are they universally present or not?
- Problem: Dynamic CHARGE stripes are very hard to detect.

Both magnetic and charge stripe fluctuations must be observed to make unambiguous conclusions about dynamic stripes. Magnetic stripe spectra in interesting compounds are known. Charge stripe spectra: NOT

Two types of neutron scattering

Neutron spins couple to electronic spins:

Magnetic scattering

Proportional to f(Q) – Magnetic form factor

Decreases with increasing Q

Q – Momentum transfer to the neutron

Neutrons couple to the atomic nuclei:

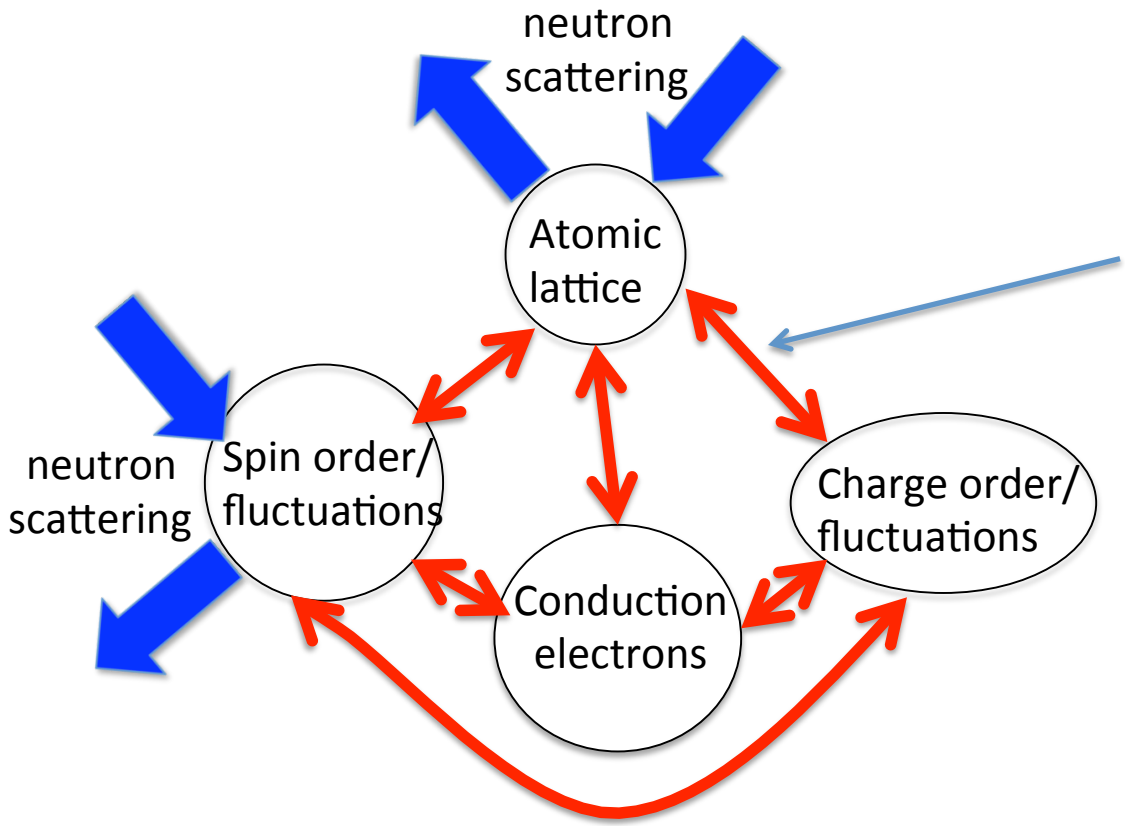
Lattice scattering

Proportional to Q^2

Increases with increasing Q

Magnetic scattering appears at small Q and scattering from the lattice at high Q

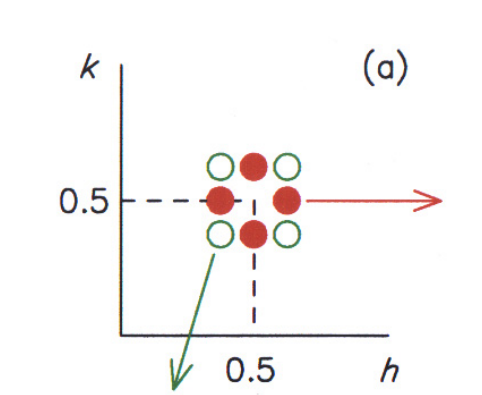
Observing dynamic charge stripes by scattering from atomic lattice vibrations



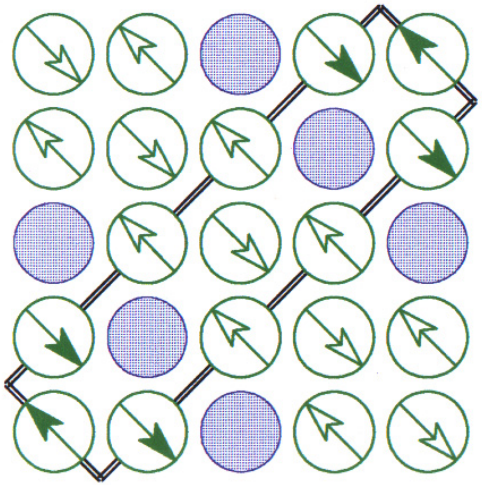
Neutrons do not couple to charge fluctuations directly, but atomic lattice does.

Neutrons scatter from atomic nuclei. We tried to observe fluctuating charge stripes by their signatures in the lattice vibration spectrum

Cuprates vs. nickelates

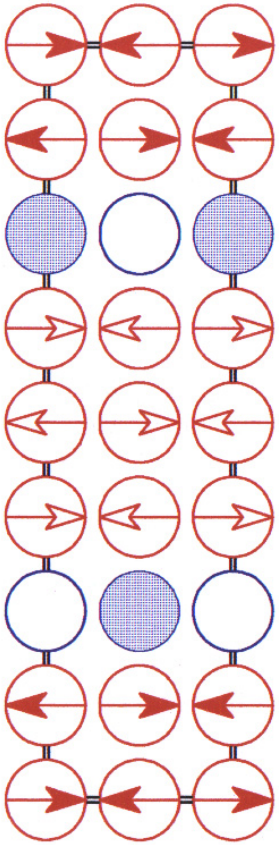


nickelates



(b) NiO_2: $n_h = 0.25$

cuprates



(c) CuO_2: $n_h = 0.125$

J.M. Tranquada, *et. al*, *Nature*, **375**, 1995

45° to Ni-O bonds in nickelates.

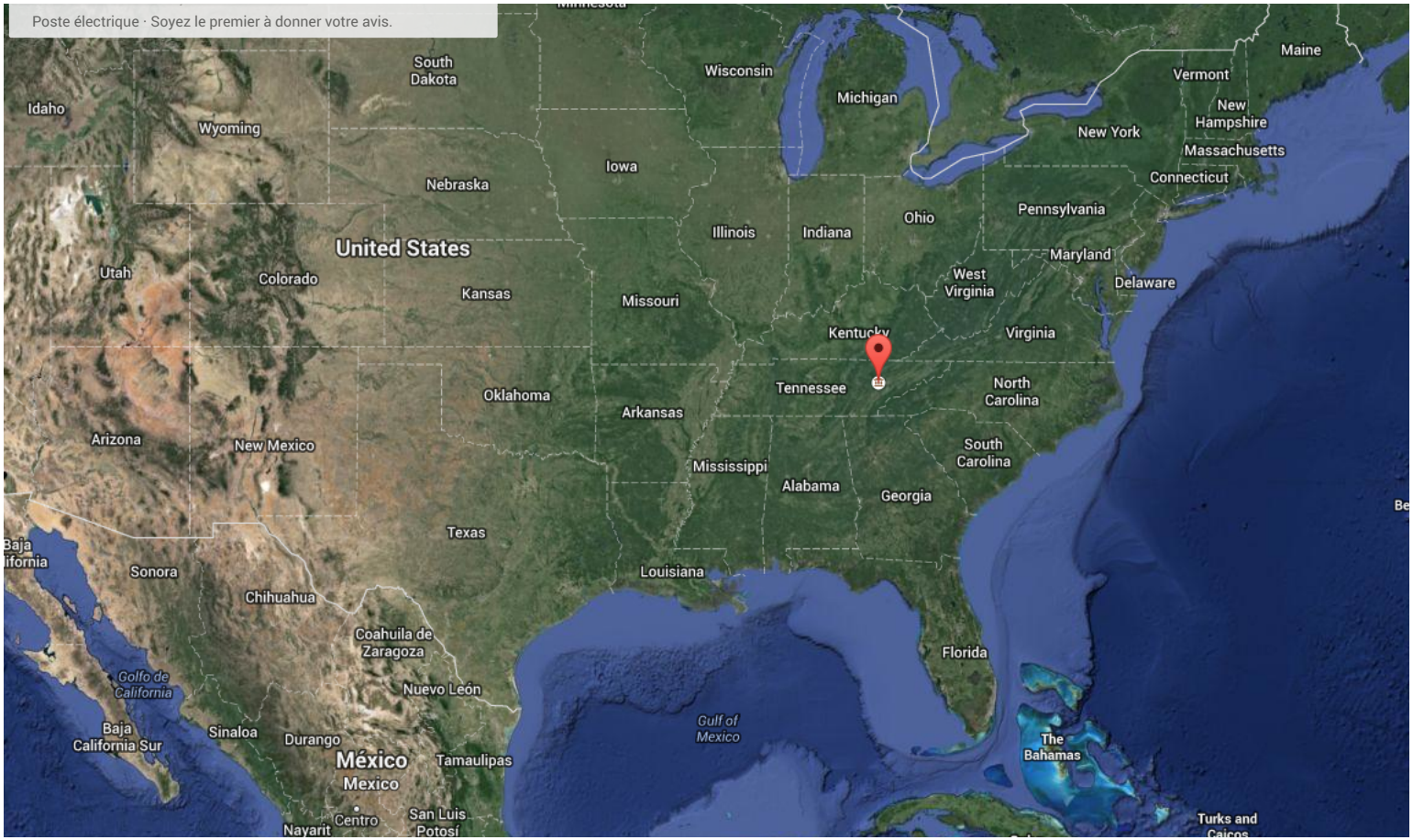
0° to Cu-O bonds in cuprates.

Coupling to the lattice in the nickelates is much stronger, so lattice fluctuations due to fluctuating charges should be easier to observe

Measurements at Spallation Neutron Source (SNS)

Spallation Neutron Source (SNS) built during the last decade takes neutron scattering to the 21st century. It is the most powerful neutron source in the world with unparalleled capabilities

Where it is

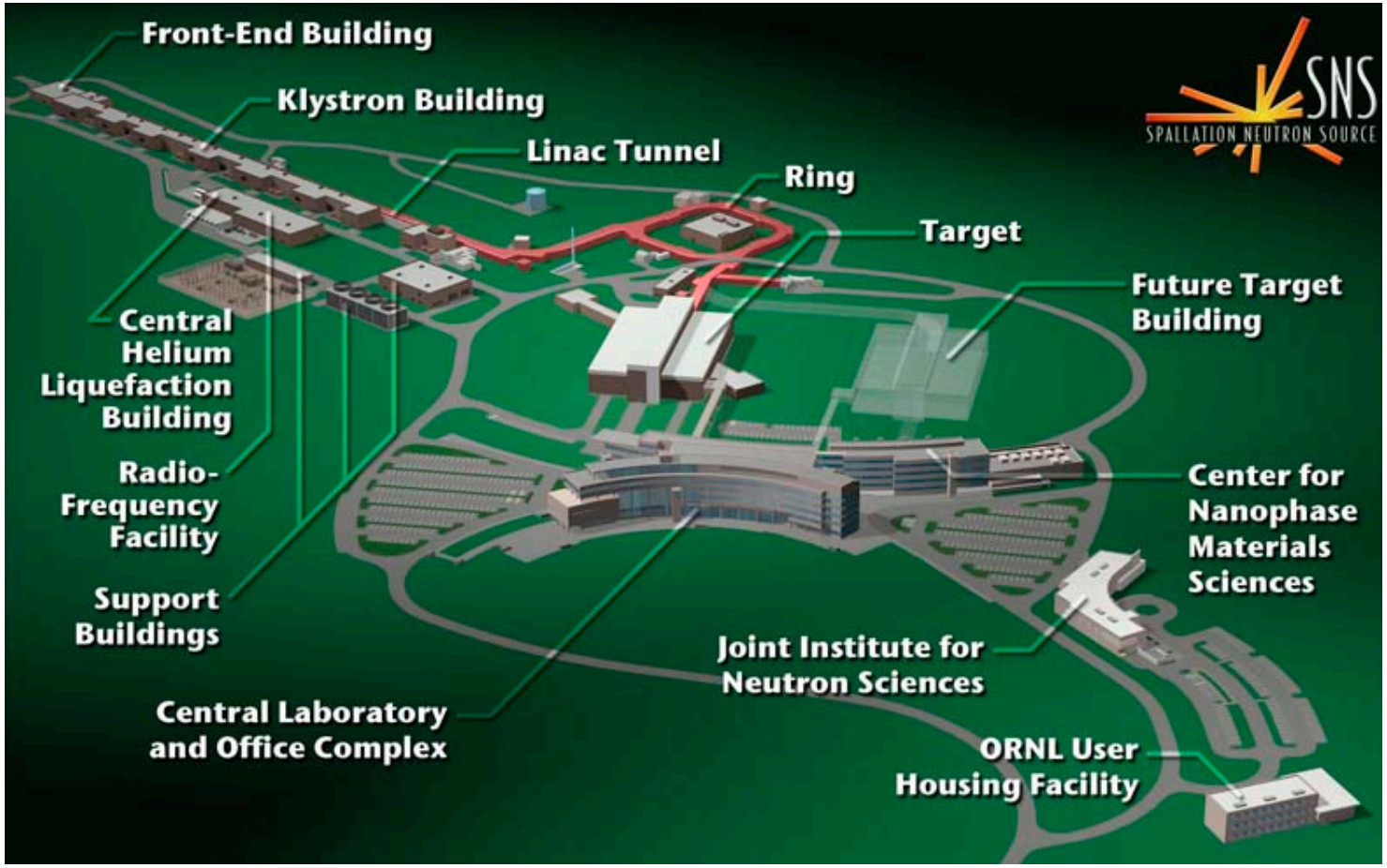


At Oak Ridge National Lab about 40 miles from Knoxville, Tennessee

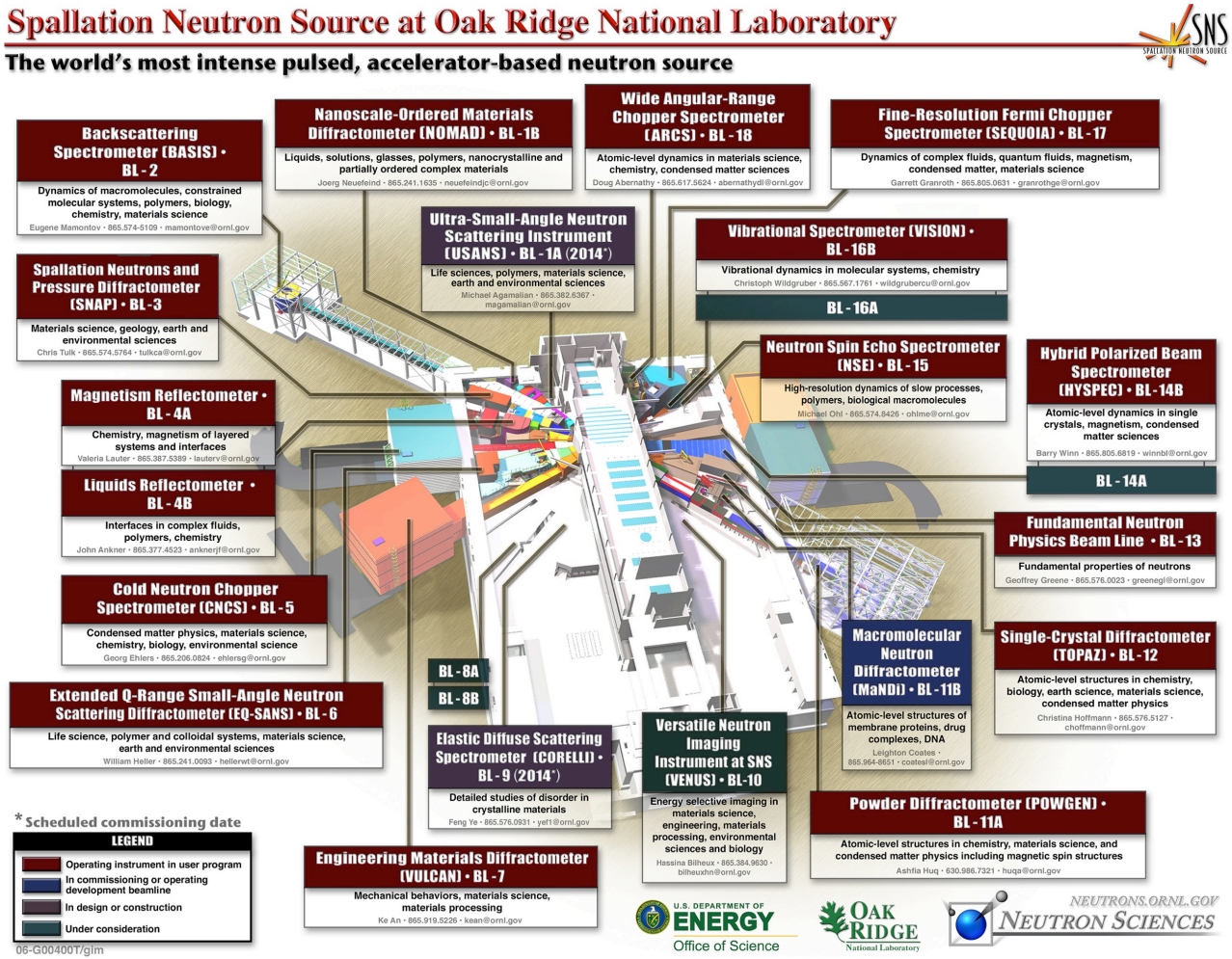
Site before it was built

What the SNS looks like today

SNS Schematic

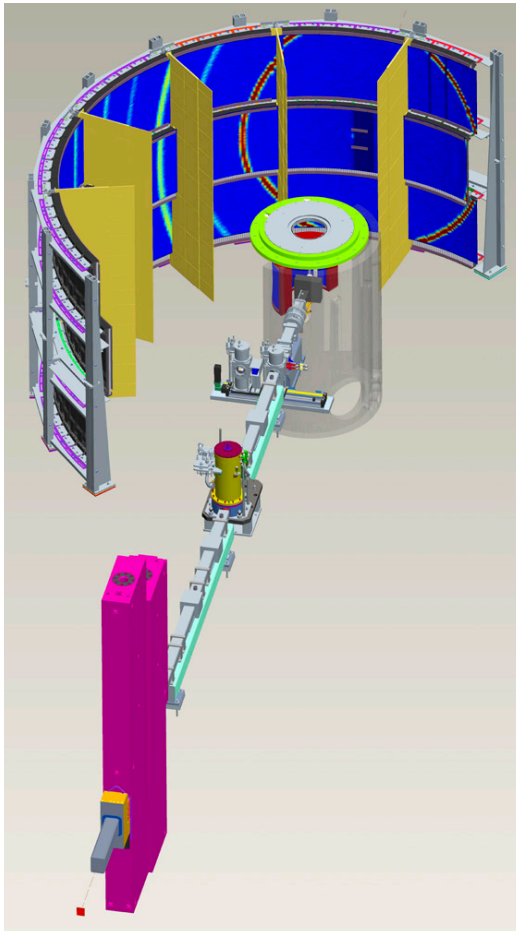


Instruments

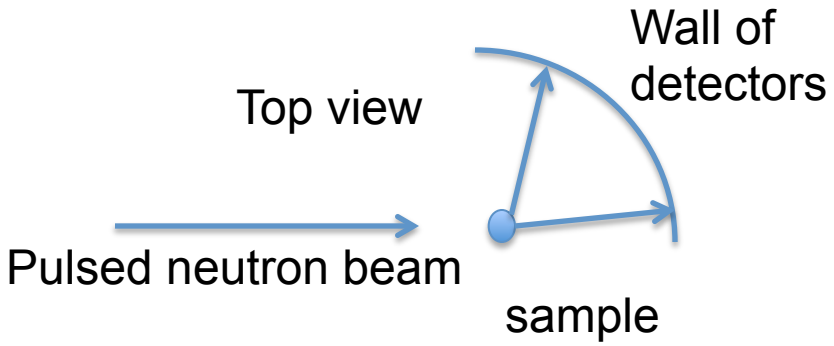


Pulses of neutrons freed by the spallation process are slowed down in a moderator and guided through beam lines to highly specialized instruments for conducting experiments.

Measurements on the ARCS time-of-flight spectrometer at the Spallation Neutron Source



Energy transfer measured by neutron time-of-flight. Sample rotates to map out a 4D chunk of reciprocal space

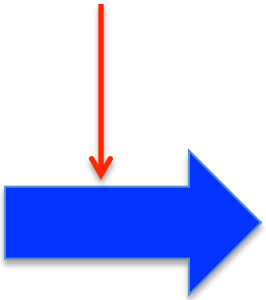


What we measure with chopper spectrometers

4 at the SNS

Diffraction in 4D (space + time)

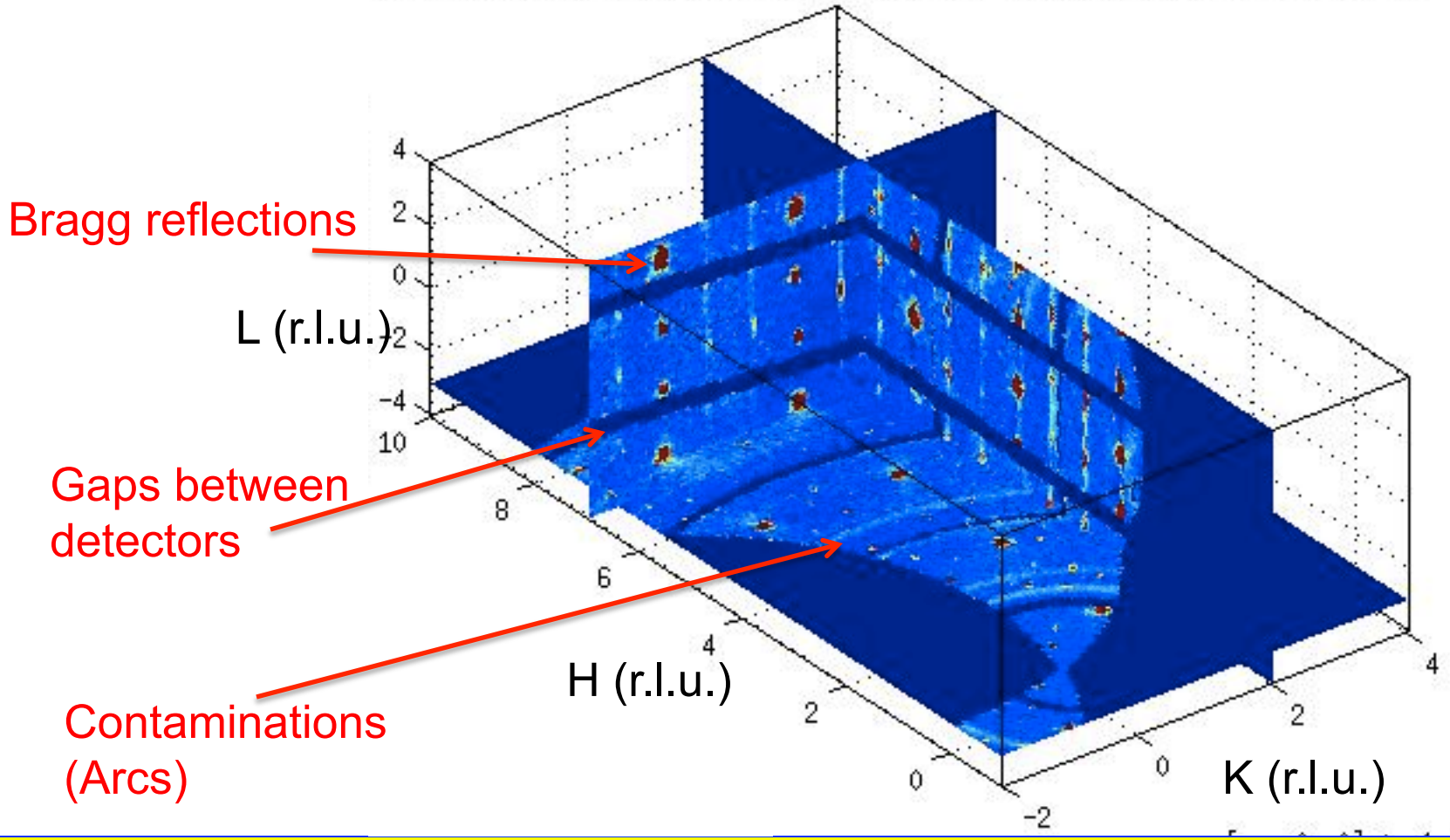
Time-dependent structure in real space (x,y,z,t)



Reciprocal space (q_x, q_y, q_z, ω) is Fourier Transform of real space structure

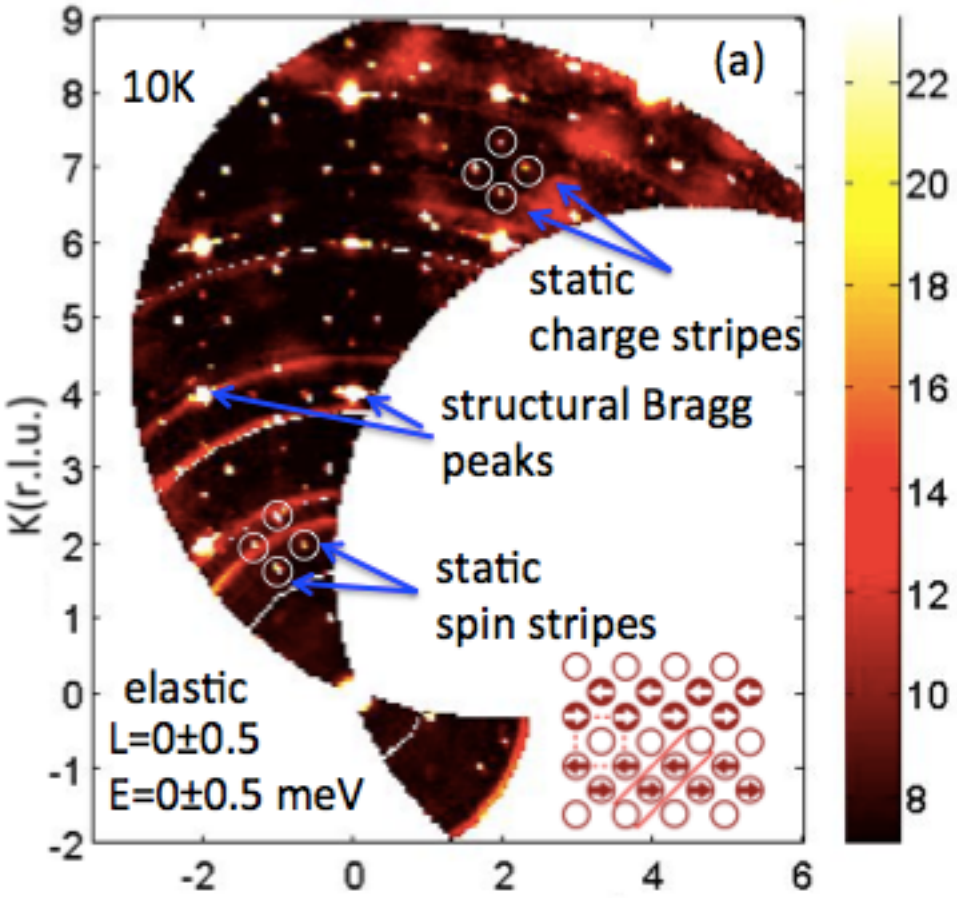
"Structure" means positions of atoms or magnetic moments

Reciprocal space for E=0



3D cut through measured portion of the 4D reciprocal space. Note periodic structure of the Bragg peaks and of weaker features

A closer look: Elastic scattering, 10K



Brightest spots: Bragg peaks
Weak regular dots: Stripes
Everything else: Contamination

Magnetic stripes at low Q
"Charge" peaks at high Q.

Each Bragg peak corresponds to 1 Brillouin zone:
A typical dataset contains 10-100 Bragg peaks.

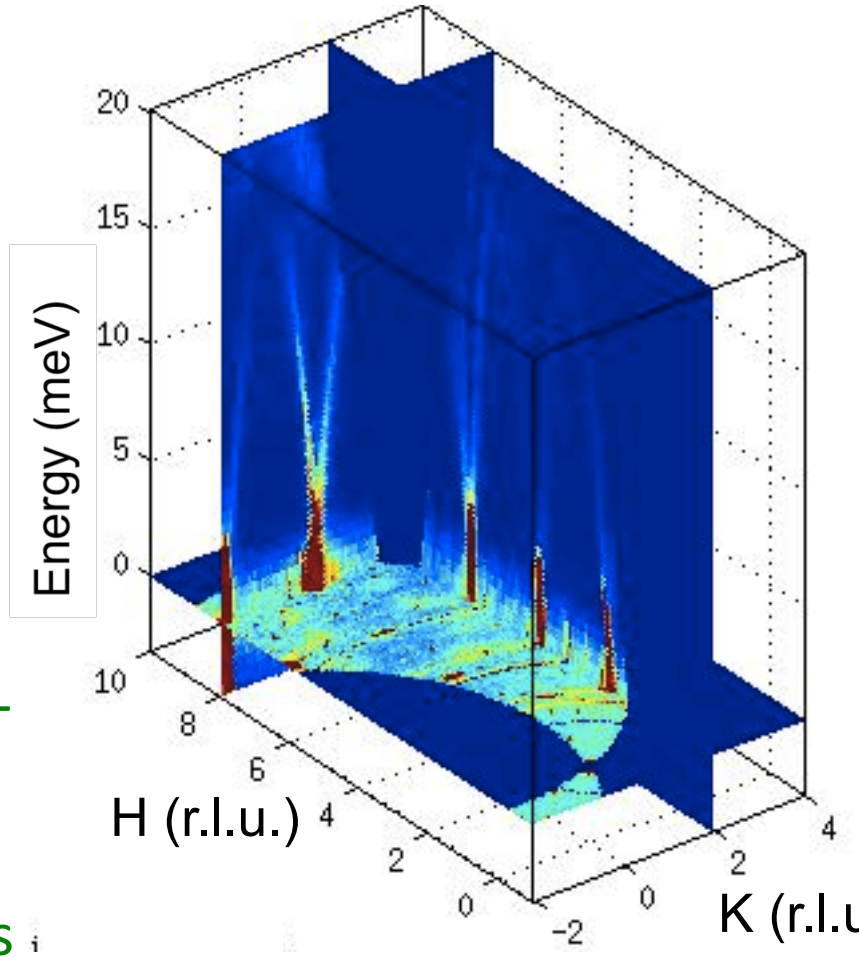
What is at nonzero energy?

Acoustic phonons (sound waves) emanate from every Bragg peak

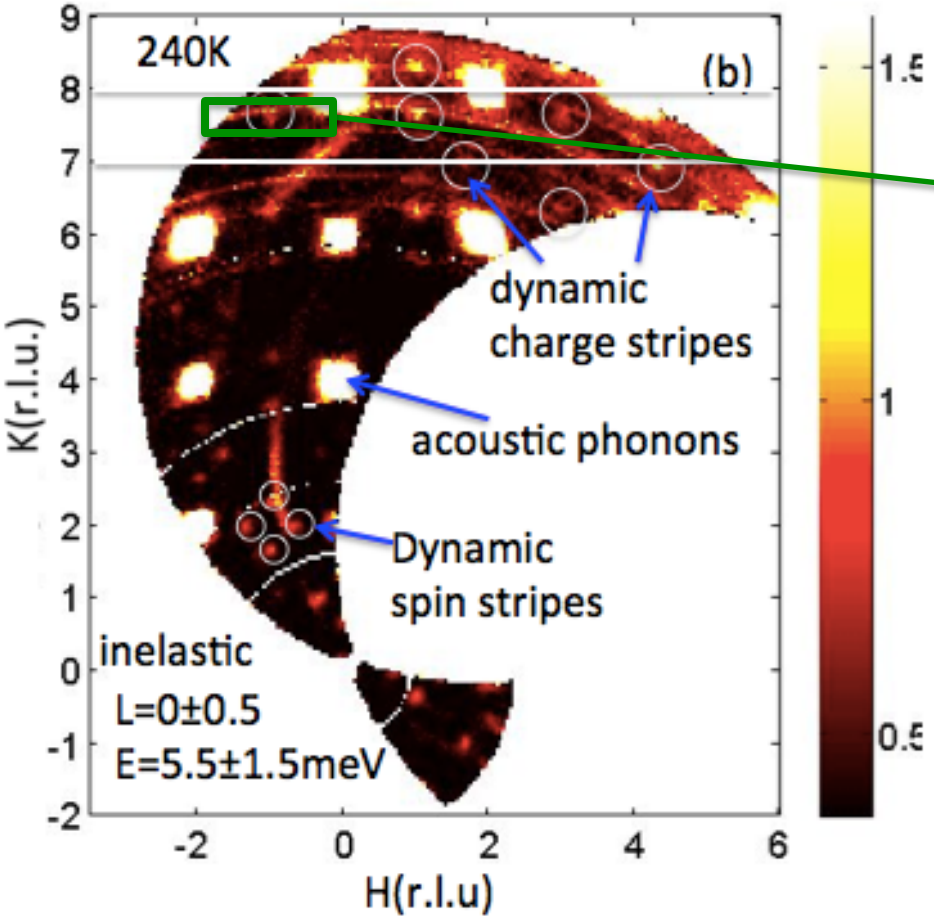
Nuclei in a solid act as (damped) coupled harmonic oscillators

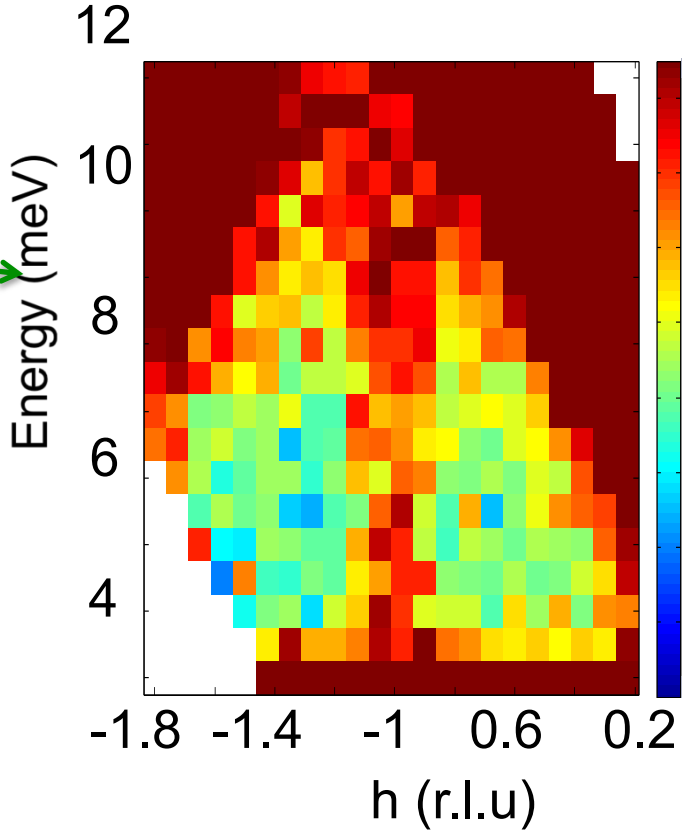
Quantization produces phonons: bosons with quantum numbers **q** (crystal momentum) an E (Energy). E=f(**q**) is a dispersion relation

If N is the number of atoms in the UNIT CELL, for each **q** there are 3N phonons with usually different Energies (dispersions) and vibrational characters



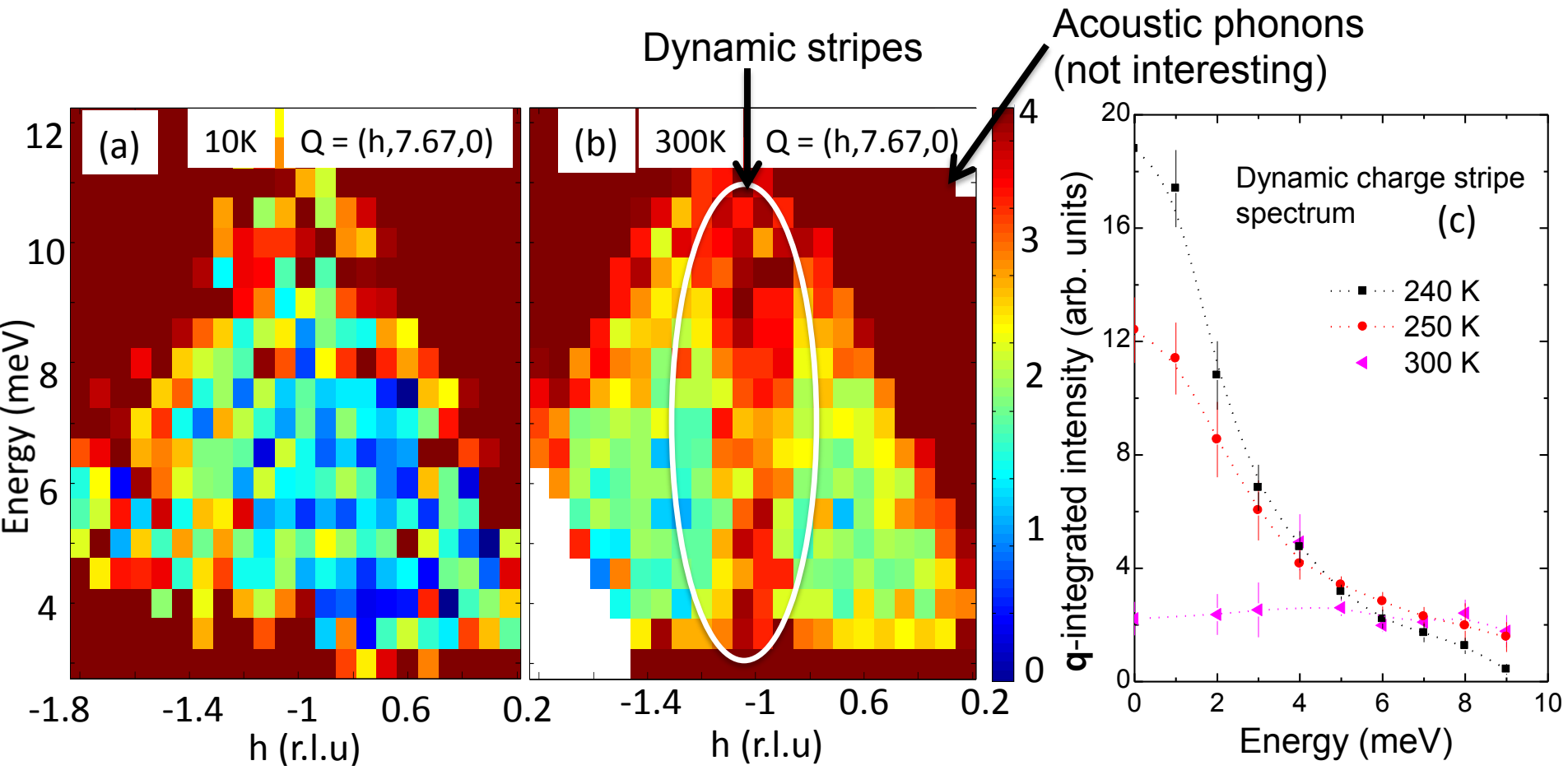
Dynamic Stripes at 240K





Brightest spots: Phonons
Weak regular dots: Dynamic stripes
Everything else: Contamination

Temperature-dependence



Dynamic charge stripes disappear at low T

Near T_c quasielastic scattering dominates

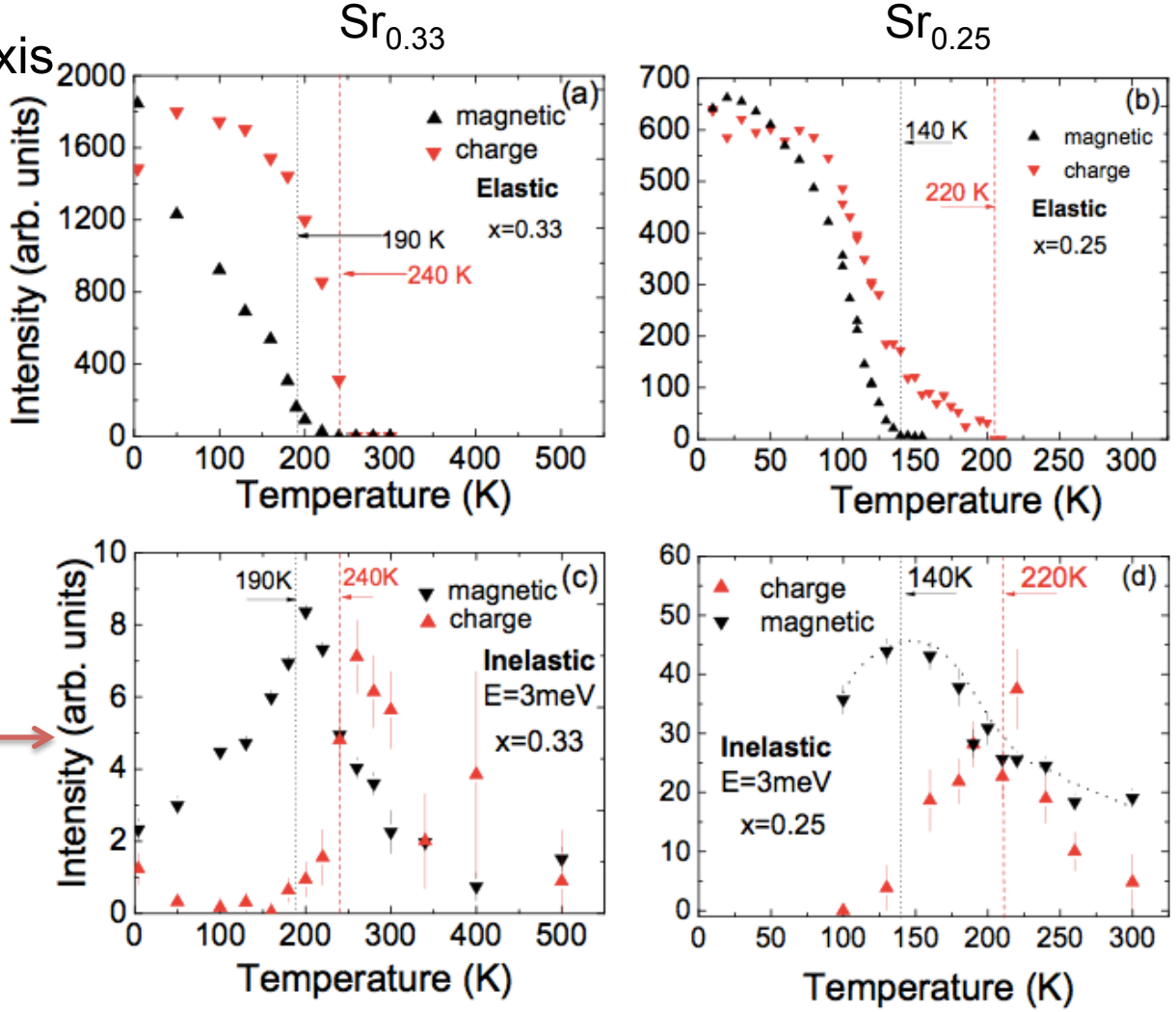
Far from T_c the spectrum is flat.

Detailed Temperature Dependence

Measured on triple-axis spectrometer

Static stripe order parameter →

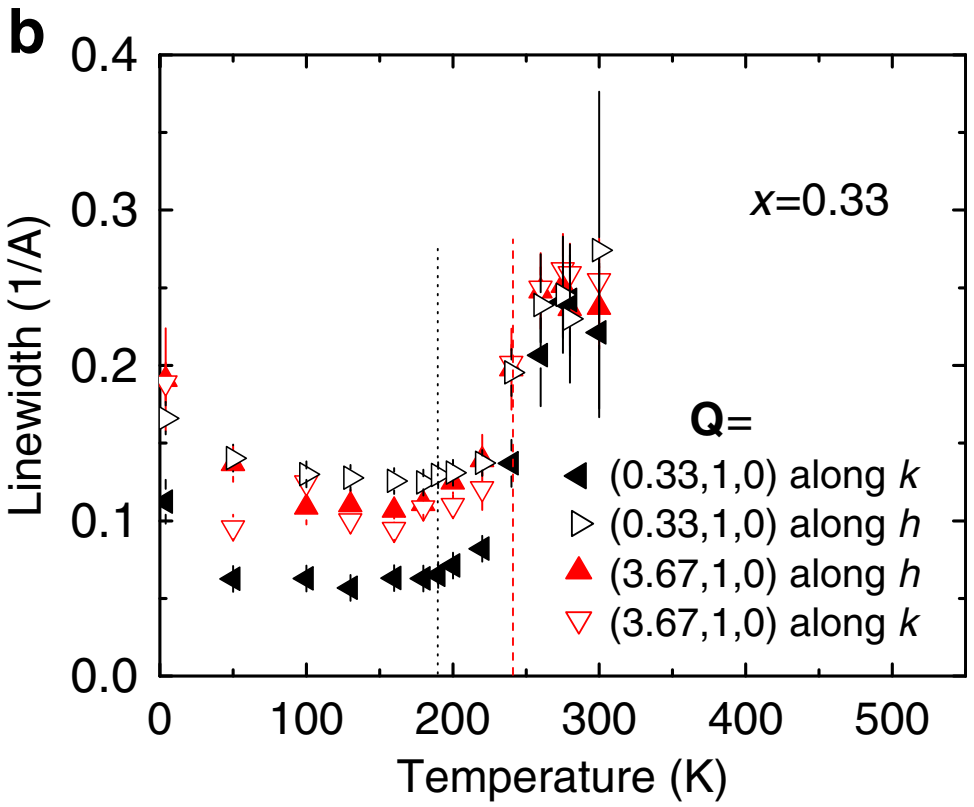
Dynamic stripe order parameter →



Static stripes disappear at charge/spin ordering transitions

Maximum of *dynamic* stripes at the ordering transitions

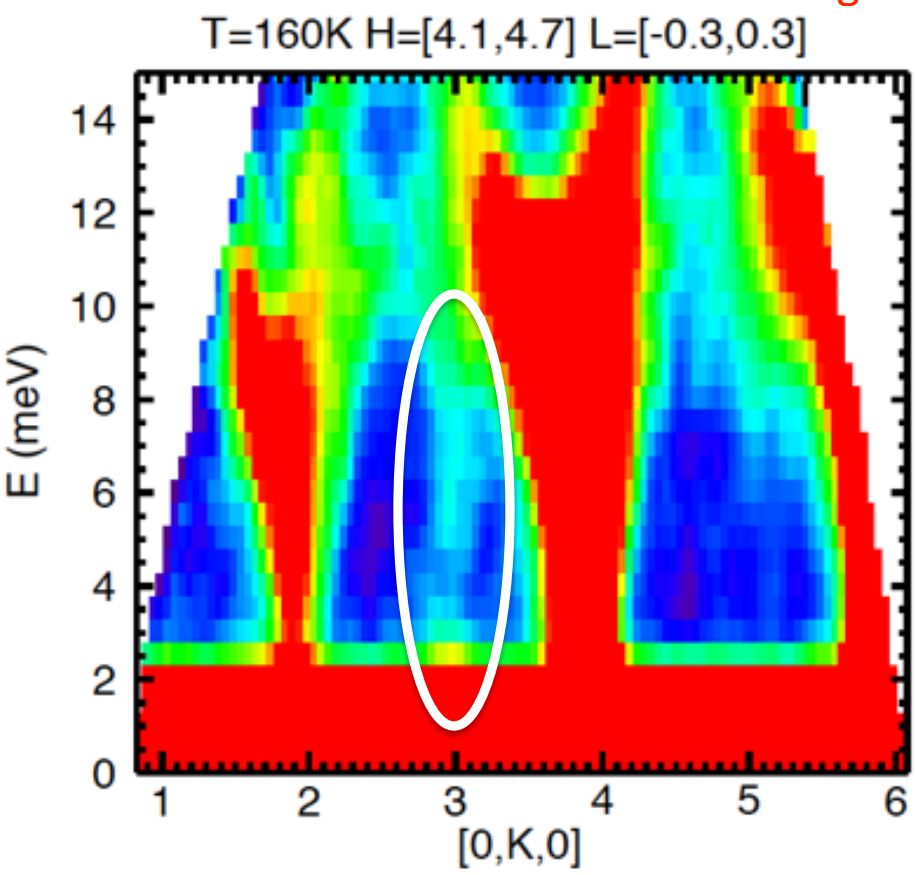
Temperature-dependent linewidth

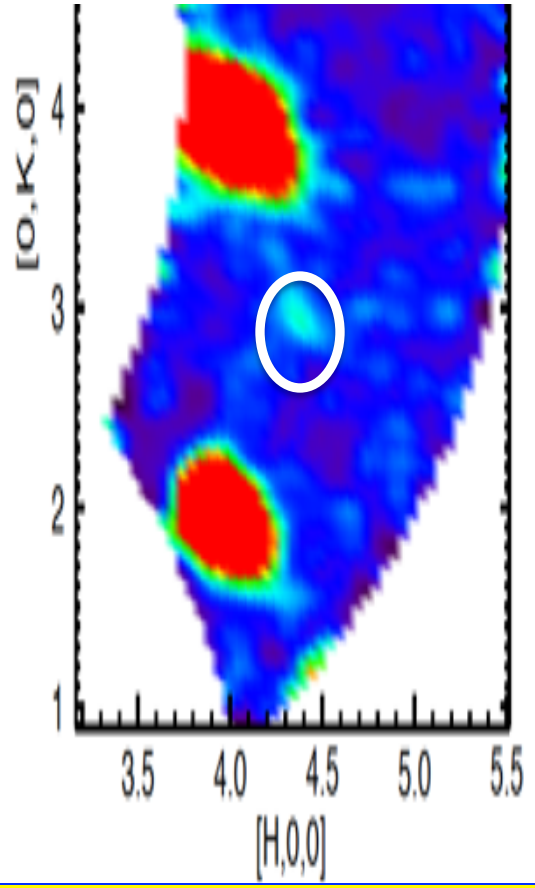


Linewidth in **q**-space jumps at the charge ordering transition

Dynamic stripes in underdoped $La_{1.75}Sr_{0.25}NiO_4$

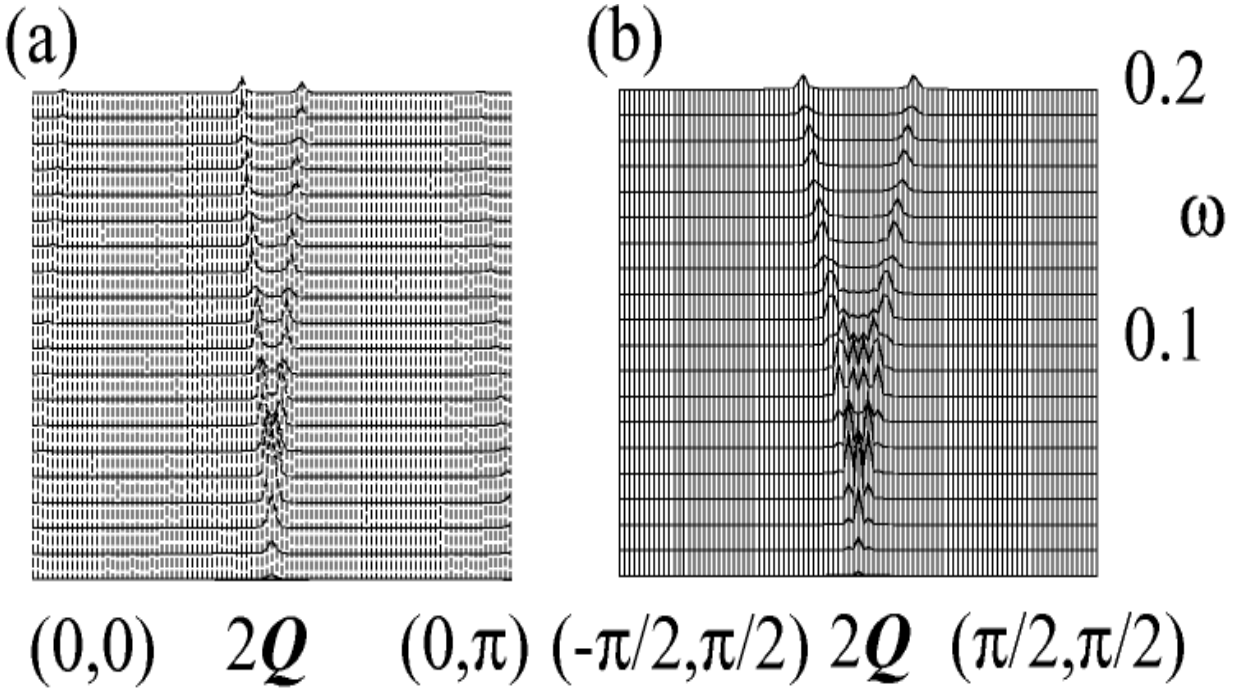
New data from HYSPEC $q_{charge}=(2x,0,1)=(0.5,0,1)$;





Dynamic charge stripes appear are similar to x=0.33, but seem to be anisotropic in the h-k plane. Data analysis in progress

Stripe collective mode predicted at low T



Kaneshita et al. predicted collective mode due to stripes.

We observe something similar but near T_c

Conclusions:

- Clear signatures of dynamic charge stripes are identified at low energies in $La_{1.67}Sr_{0.33}NiO_4$ and in $La_{1.75}Sr_{0.25}NiO_4$
- Dynamic stripes are fluctuations associated with the stripe-ordering transition

Key questions:

Does dynamic stripe amplitude becomes enhanced or suppressed with the reduction of the ordering temperature when x is different from 1/3?

Do cuprates show quantum criticality associated with an incipient stripe ordering transition?

Why does the charge signal at 3meV disappear when spins order?

Neutron scattering from a crystal (with periodic lattice)

From momentum to crystal momentum

- Crystal lattice forms a periodic potential; The reciprocal lattice is its Fourier transform.
- Momentum, no longer a good quantum number, is replaced by crystal momentum with a conservation law: **k**'=**k**+**K**, where K is any reciprocal lattice vector
- Reciprocal space is now split into Brillouin zones with nonequivalent **k**s filling each zone.

Every excitation shows up in every zone but with different scattering intensity

Interactions of dynamic stripes with phonons

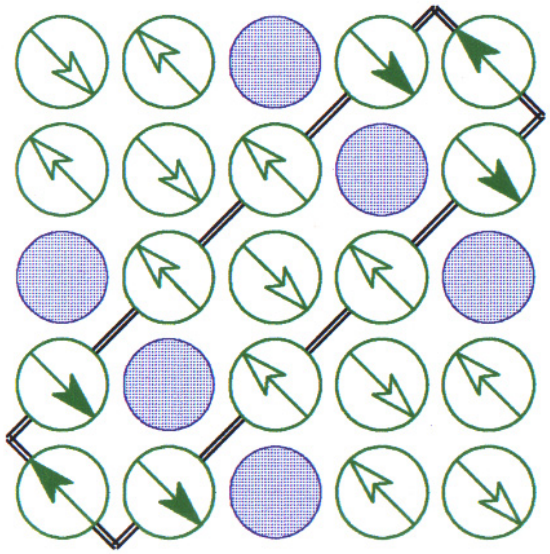
Phonons with atoms vibrating in the a-b plane do not show any obvious signs of interactions with dynamic stripes. (e.g broadening, softening at the ordering wavevector, or anticrossing). No data for c-axis polarized phonons

Lattice vibrations that accompany charge fluctuations are not phonons in the sense that they are not specific vibrational eigenmodes of the crystal lattice.

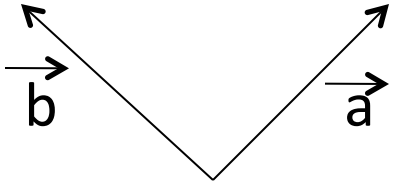
May be similar to incoherent quasielastic scattering observed in some manganites

We measured dynamic charge stripes in nickelates by neutron scattering

- $La_{2-x}Sr_xNiO_4$, x = 1/3, 1/4



Orthorhombic notation:



|a|=|b|=5.4Å

If x is doping

q_{charge}=(2x,0,1);
q_{spin}=(1-x,0,1)

Where in reciprocal space to look for stripes in nickelates?

For x=1/3: $q_{charge}=(0.67,0,1)$ $q_{spin}=(0.67,0,1)$ the same !

For x=1/4: $q_{charge}=(0.5,0,1)$ $q_{spin}=(0.75,0,1)$

We looked in Brillouin zones next to the zone centers at Q=(H,K,L=1), H,K-odd.

e.g. Zone center at Q=(7,1,1): stripe order is at Q=(7.67,1,0), (6.33,1,0), (7,1.67,0),(7,0.33,0)

This allowed us to make measurements with L=0, i.e. without tilting the crystal

Magnetic form factor determination

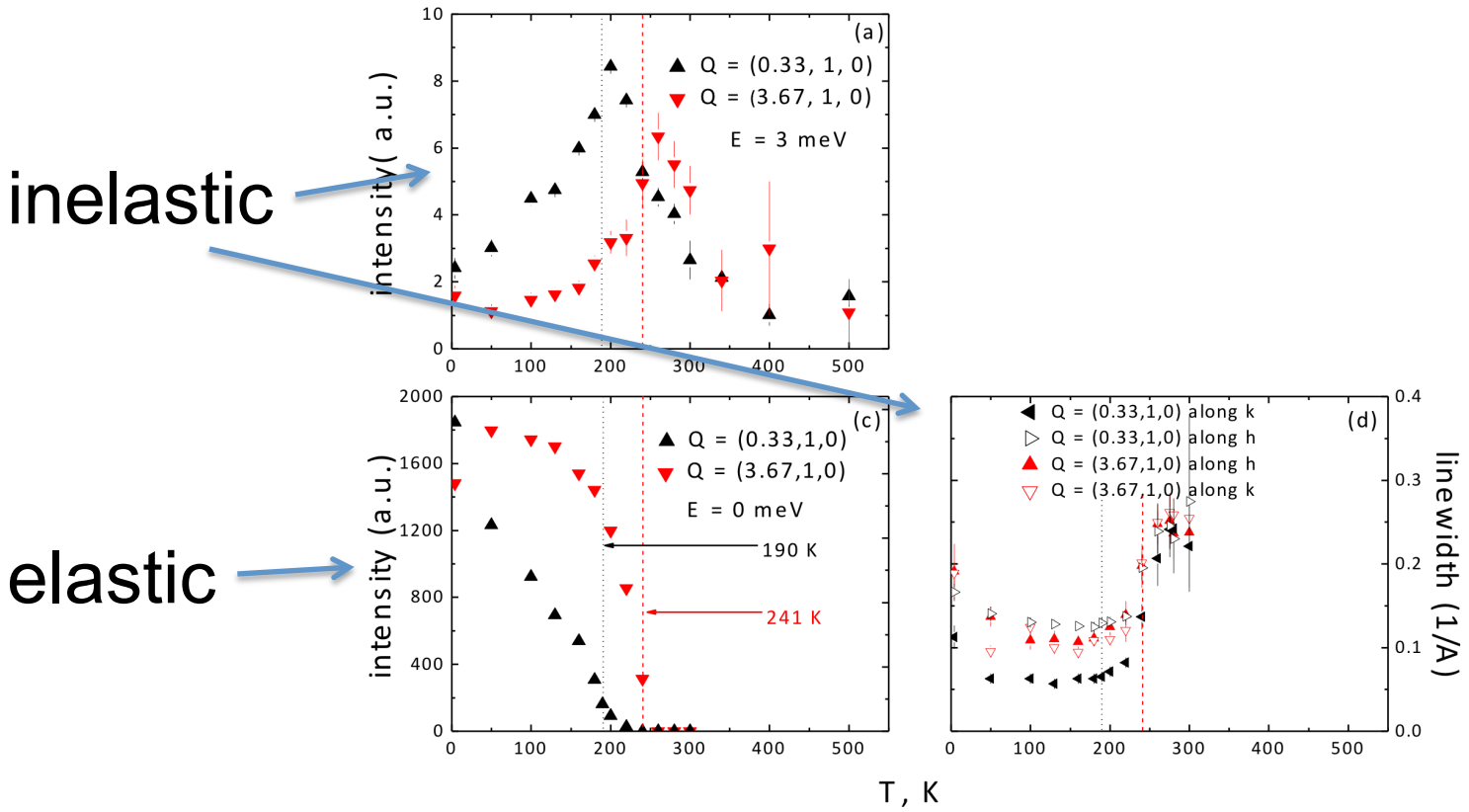
Q=(3.67,1,0) is not big enough to kill magnetic scattering completely, so it is difficult to separate magnetic and nuclear contributions without knowing the magnetic form factor $F^2(Q)$

We measured a sample with x=0.45 where magnetic and charge fluctuations should appear at different wavevectors:

Magnetic: q=(1-0.45,0,1)=(0.55,0,1)

Charge: q=(0.95,0,1)

Intensity at 3meV vs. Temperature



At low Q the signal peaks near magnetic transition. At high Q– near charge ordering T_c. Gradually die off at high T. Linewidth jumps above T_c

Where in reciprocal space to look for stripes in nickelates?

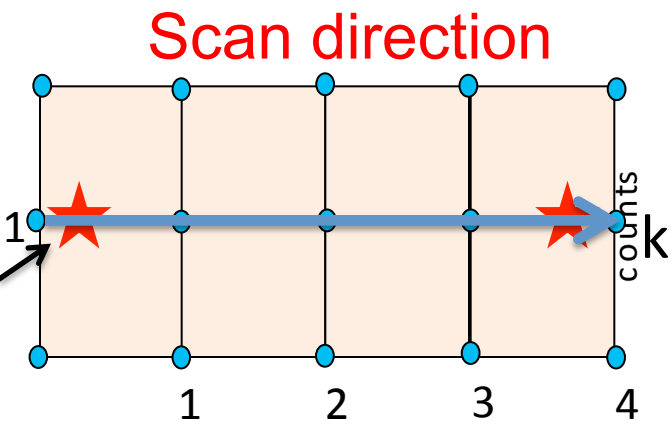
For x=1/3: $q_{charge}=(0.67,0,1)$ $q_{spin}=(0.67,0,1)$ the same !

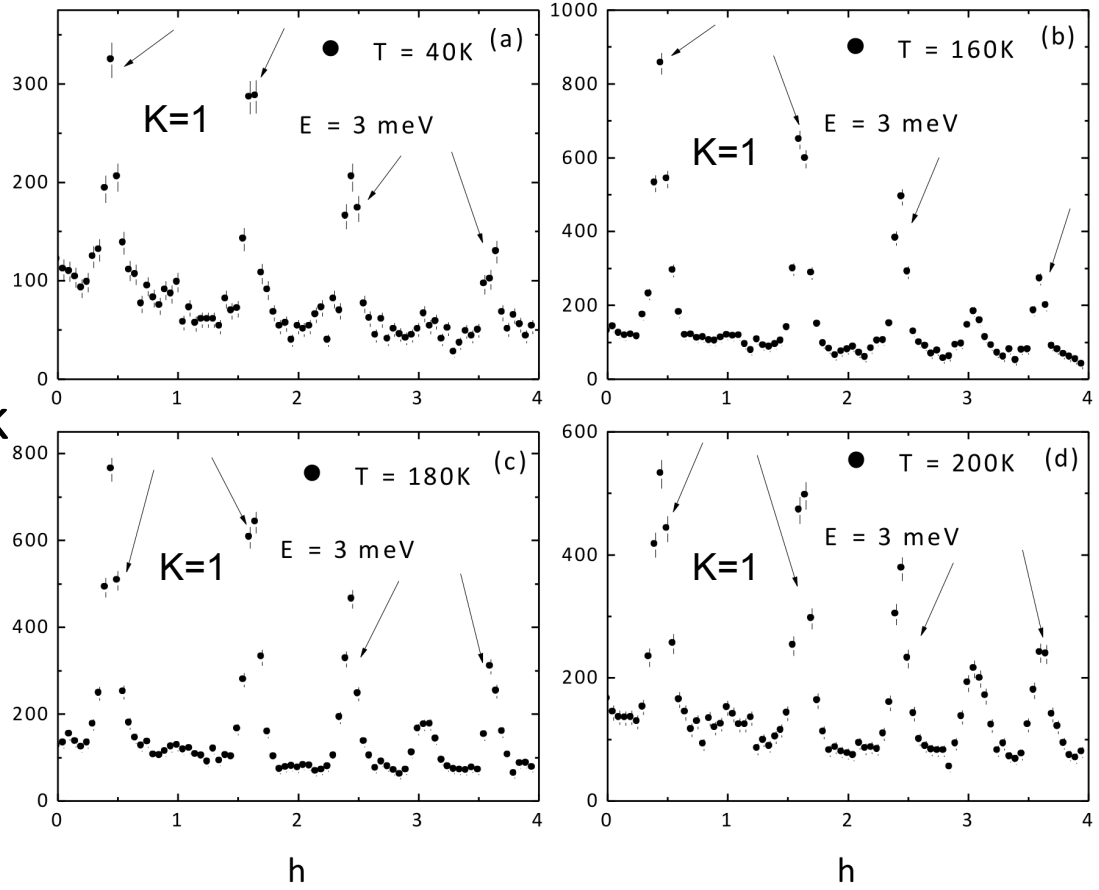
We looked in Brillouin zones next the zone centers at Q=(H,K,L=1), H,K-odd.

e.g. Zone center at Q=(7,1,1): stripe order is at Q=(7.67,1,0), (6.33,1,0), (7,1.67,0),(7,0.33,0)

This allowed us to make measurements with L=0, i.e. without tilting the crystal

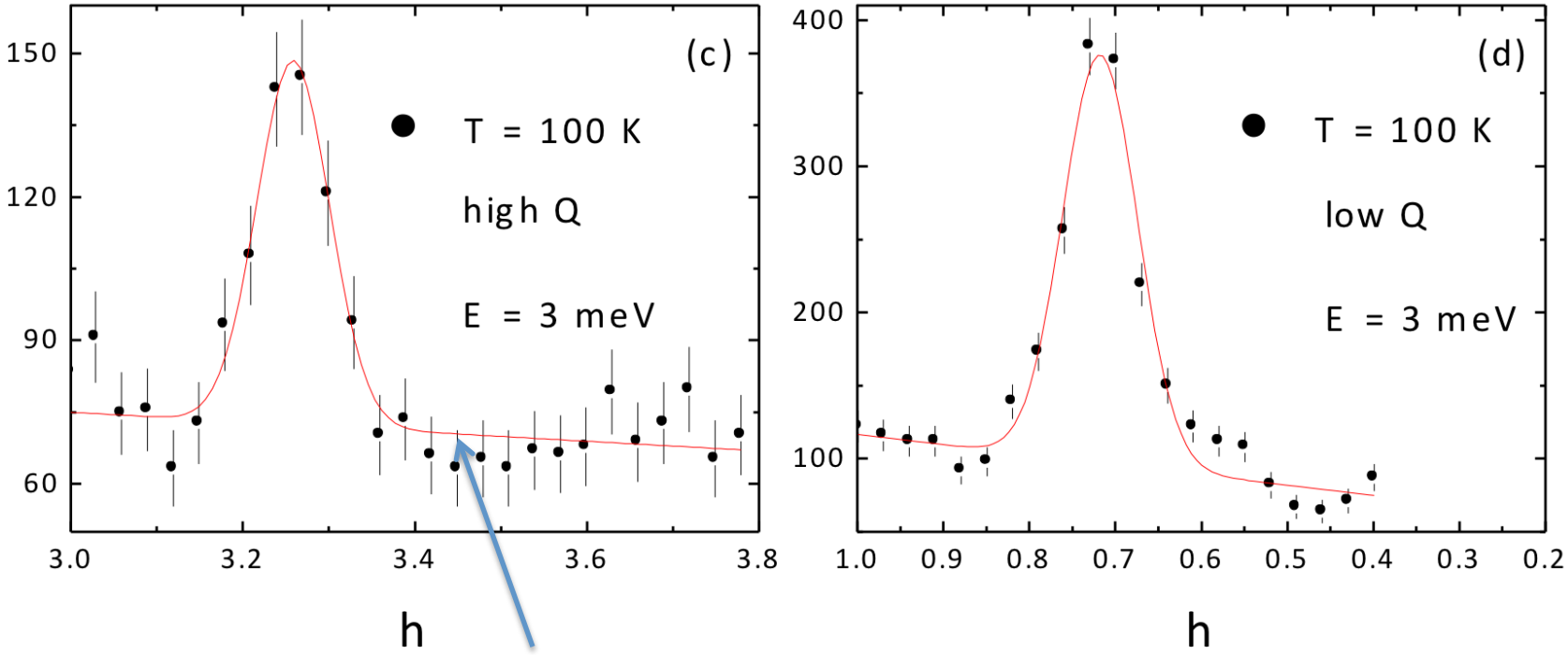
Magnetic scattering for x=0.45





Ratio of the peak intensity at (3.55 1 0) divided by intensity at (0.45 1 0) is about 3.4. This is the ratio of magnetic form factors squared between low Q and high Q.

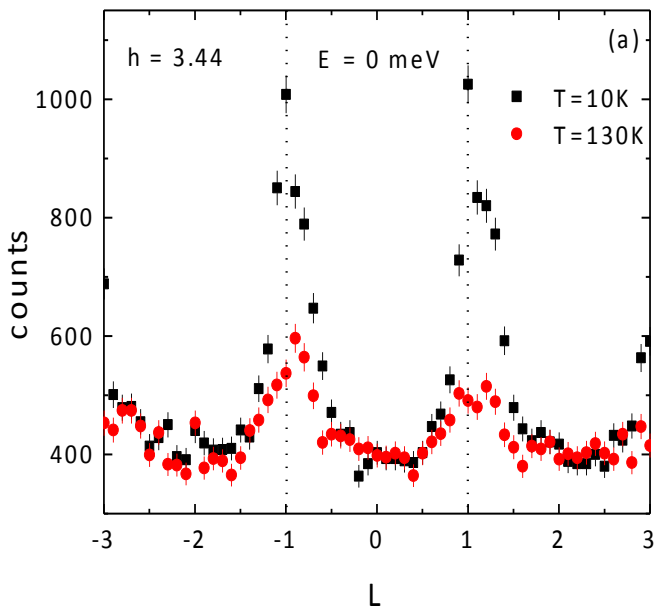
Magnetic form factor



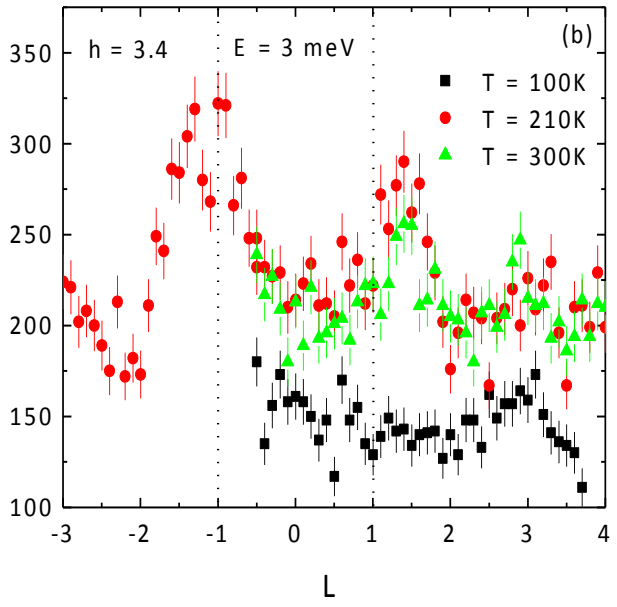
100K: Magnetic peaks only
Here we can determine the ratio of the magnetic form factors to be 3.6

Finding L with the biggest structure factor

elastic



inelastic

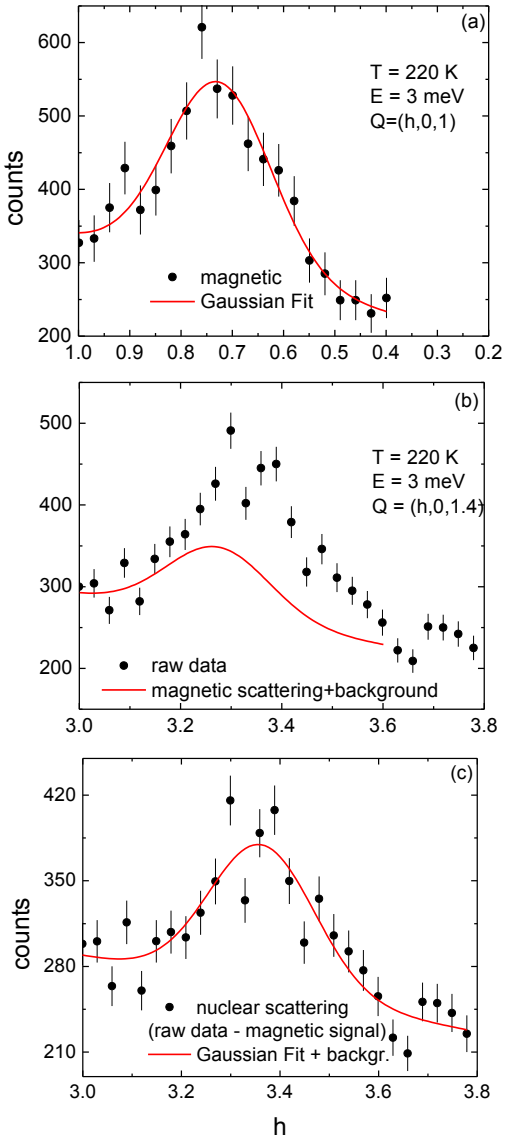


Static sripes at Q=(3.44,0,±1)

Inelastic signal peaks at L=1.4

We made further inelastic measurements at L=1.4

220K data



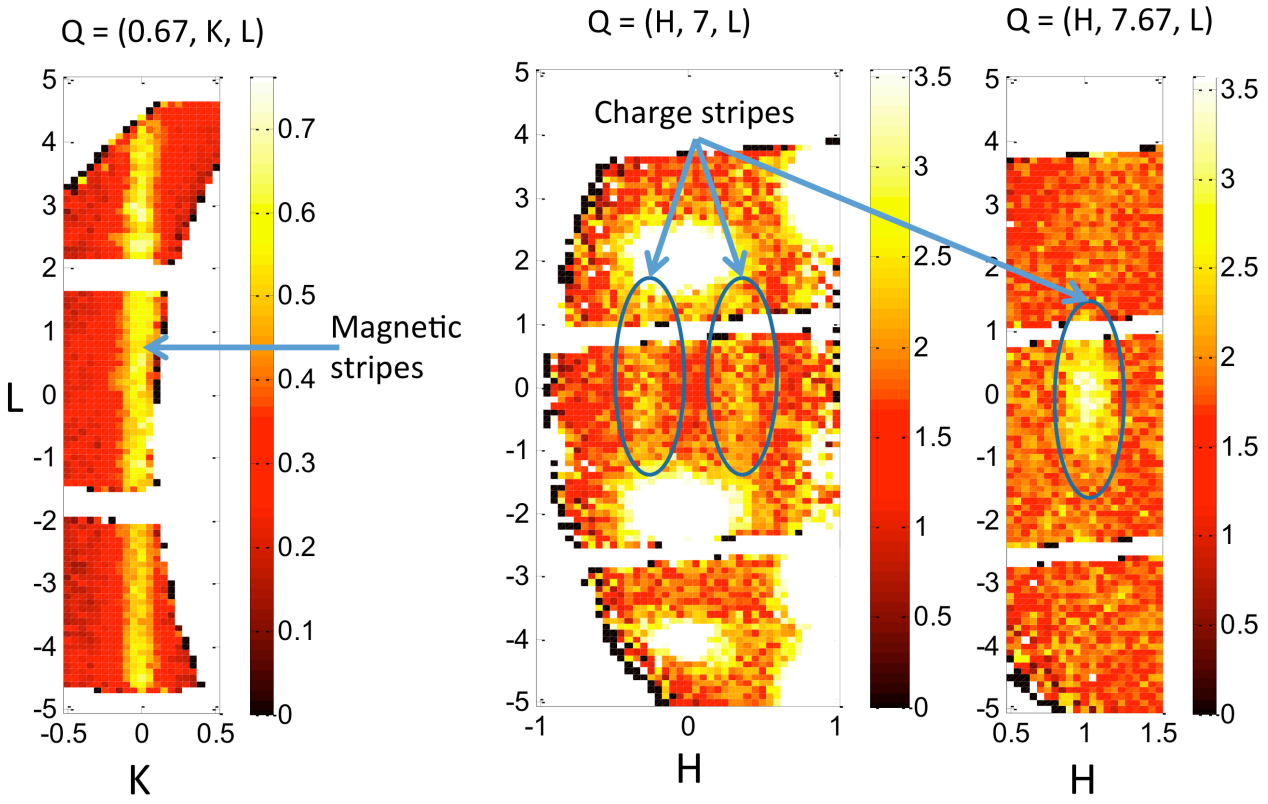
Low Q: All scattering is magnetic

High Q: Line - Magnetic scattering = low Q/3.4.

High Q: Data – magnetic scattering = Lattice scattering

Clear lattice fluctuations at high Q in addition to magnetic scatteringl

L-dependence



Magnetic stripes: L-independent, i.e. there are no interlayer correlations

Charge stripes: Also no interlayer correlations but complex structure factor