Investigations of phonons in enabled by 2 orders of magnitude increase of the throughput of a new IXS instrument

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Funding: DOE

Collaborators/References

Programming:

D. Parshall, Irada Ahmadova (CU)

Neutron scattering:

A. Merritt, D. Parshall, (CU) D. Abernathy (SNS)

Samples:

Th. Wolf, (Karlsruhe Institute of Technology)

M. Greven, (University of Minnesota)

Experiments at ARCS

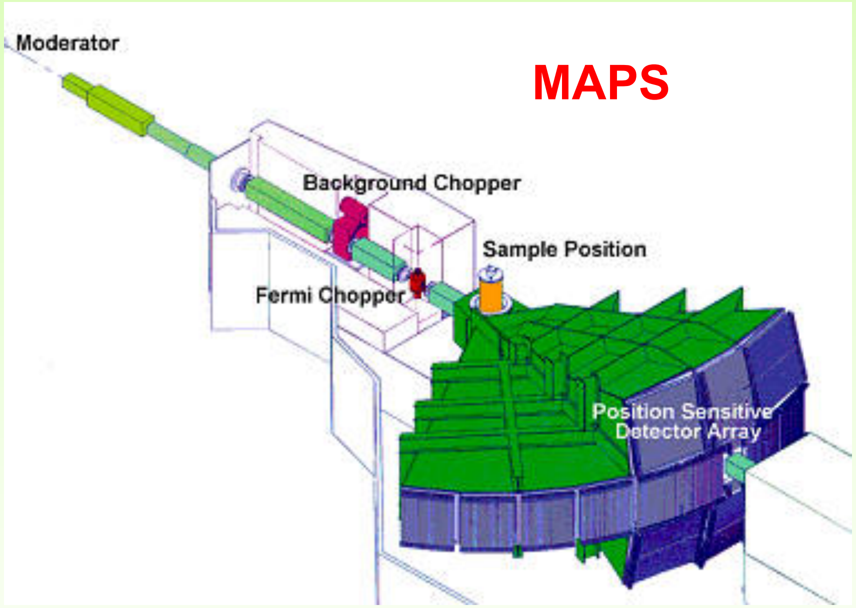
D. Parshall et al., Phys. Rev. B 89, 064310 (2014)

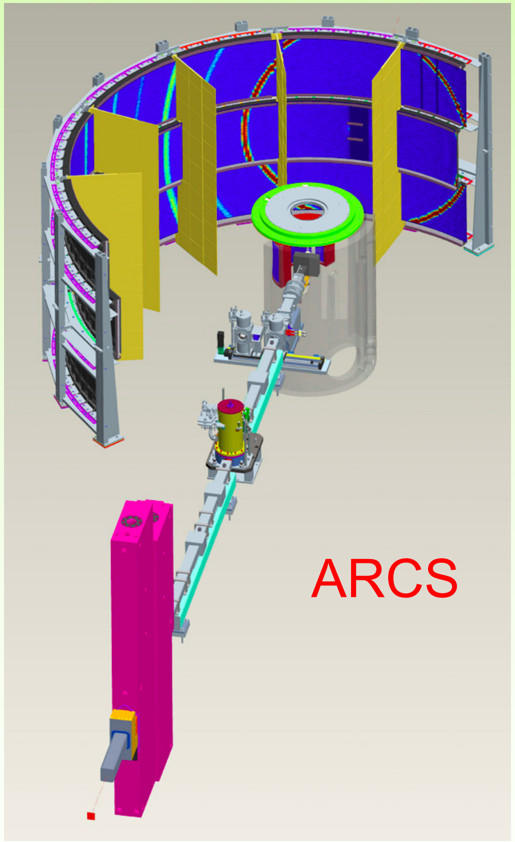
Outline

- Measurements of lattice dynamics on chopper spectrometers at the SNS
- Multizone Phonon Refinement (MPR) explained on the example of $BaFe_2As_2$
- Advantages of MPR
- New results of the new background subtraction algorithm
- Advantages and disadvantages of being able to use x-rays.

New Pulsed Sources

The entire spectrum is measured at once





MAPS (ISIS) and ARCS (SNS) chopper spectrometers with position sensitive detectors at ISIS

SEQUOIA, HYSPEC, CNCS at the SNS are similar

What we measure with chopper spectrometers 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

Neutrons bounce off nuclei and magnetic moments so structure means positions of atoms or magnetic moments

Neutron scattering from a 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

Types of Atomic Lattice Dynamics

- Incoherent lattice vibrations/molecule rotations/diffusion

Polarons, rotational diffusion of large molecules (e.g. C_{60}), dynamic stripes, etc. Hydrogen diffusion in fuel cells, etc.

- Normal modes (Phonons): focus of talk

elastic properties, ferroelectricity, electron-phonon coupling (my group’s specialty), atomic lattice contribution to specific heat and thermal conductivity, negative thermal expansion, isolating phonons from magnetic fluctuations, structural phase transitions, etc.

Every dataset from the SNS is overcomplete typically by a factor of 10 to 100!!!

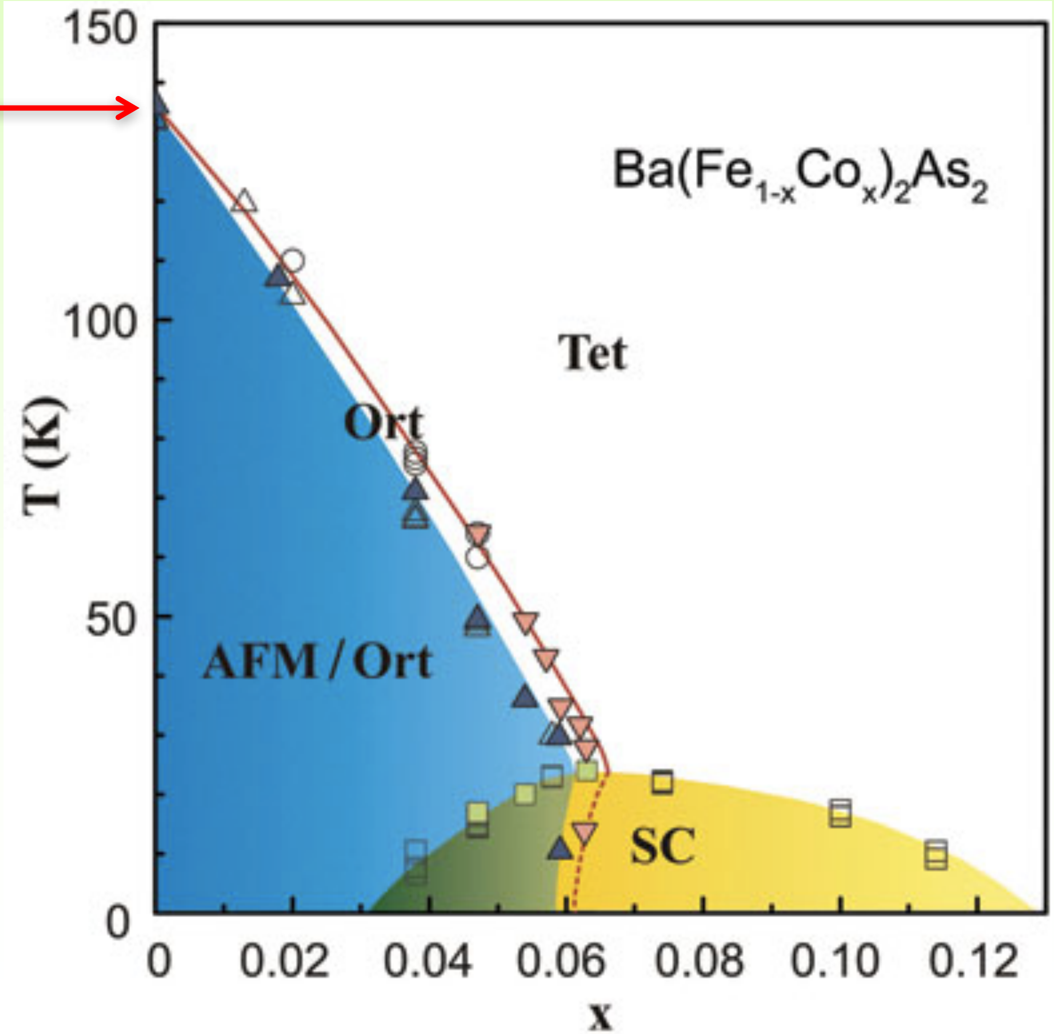
How the data handled now: Typically one finds one or two zones that are “the best” and ignore the rest of the data.

We found:

When it comes to measuring phonons, data quality can be improved by at least an order of magnitude if the entire dataset is used for a new data analysis technique invented in my group: Multizone Phonon Refinement (MPR); Many problems considered unsolvable become solvable.

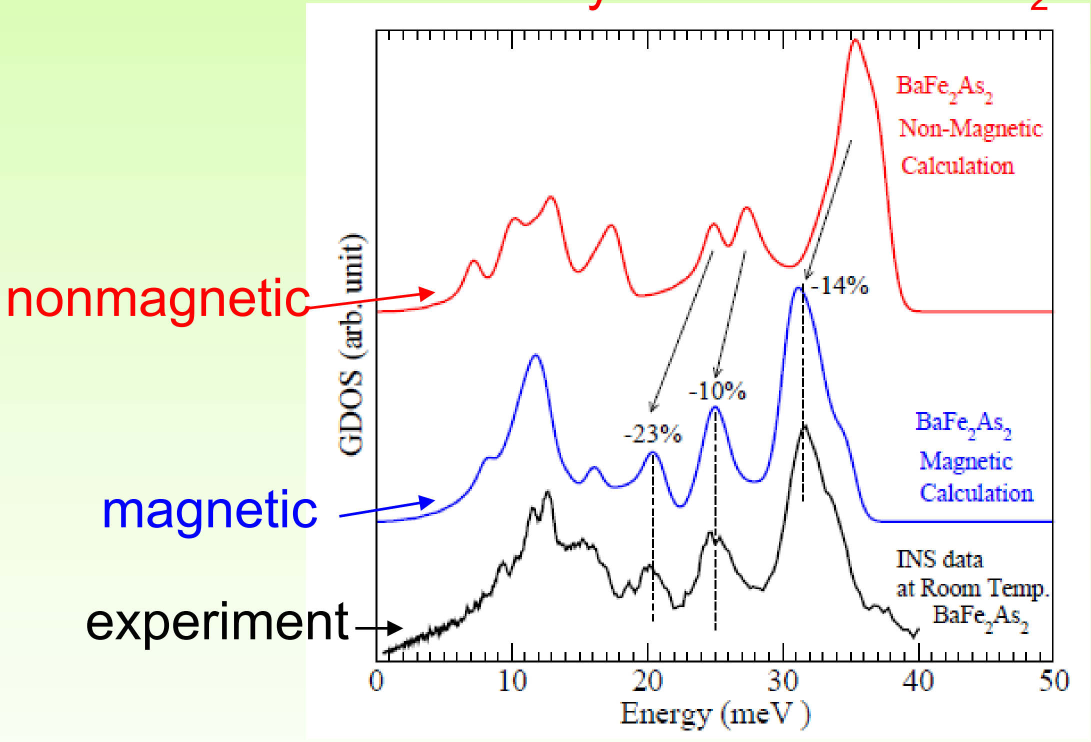
Example: Does magnetic ordering transition have an effect on highest-energy optic phonons in $SrBa_2As_2$?

Interested in the effect of the magnetic order on optic phonons



Implication for phonons

Phonon density of states of $BaFe_2As_2$

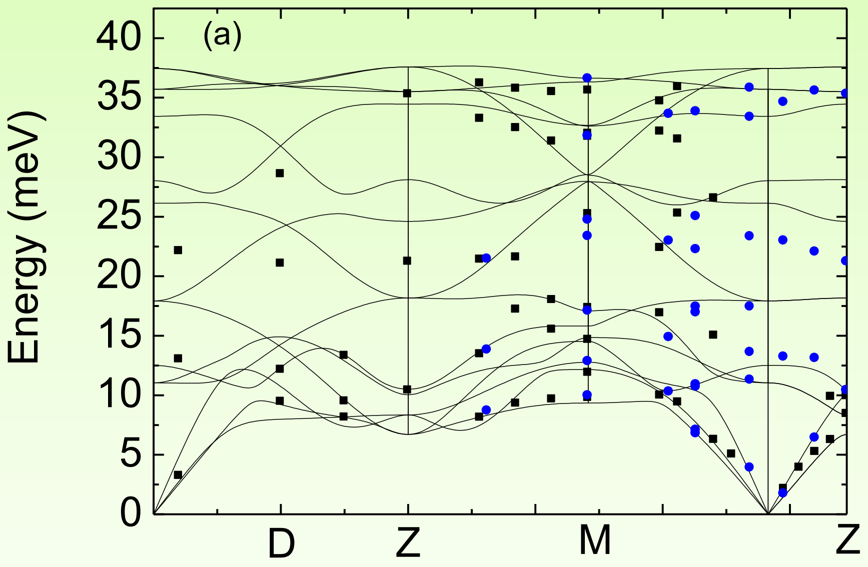


T. Yildirim, Physica C

Frequencies of some phonons strongly depend the Fe magnetic moment.

Magnetic calculation for $BaFe_2As_2$ agrees much better with experiment.

Interested in 5 phonon branches calculated to be around 35meV



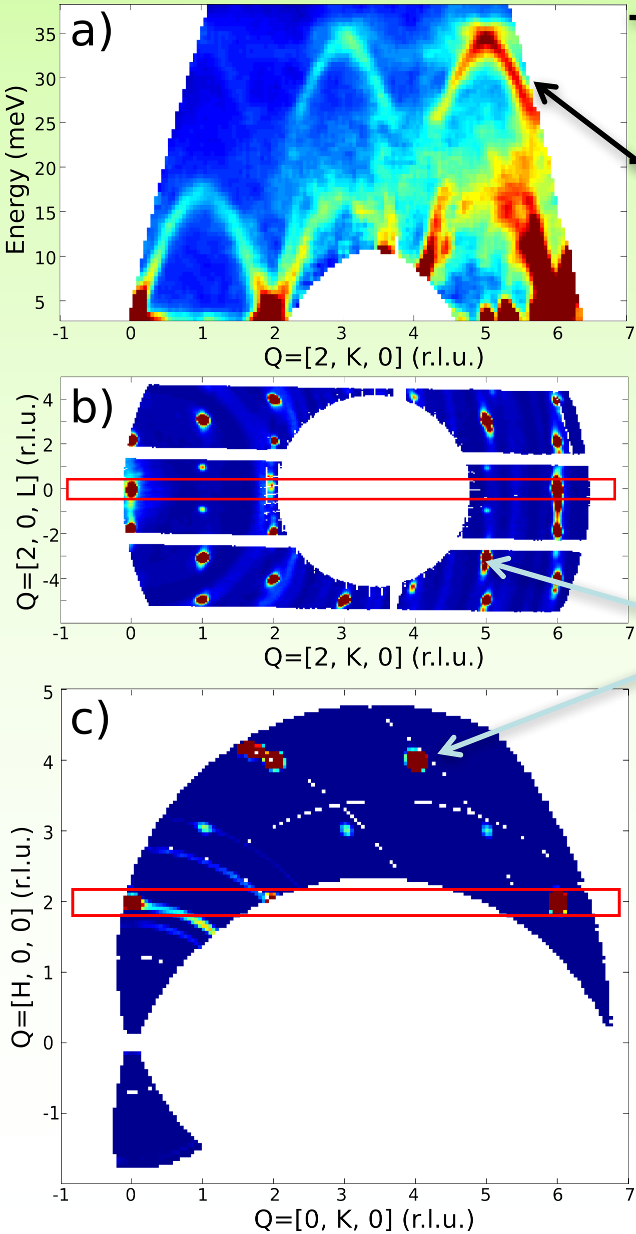
Only two observed in previous IXS experiments (HERIX)

Would like to know what is the temperature-dependence of all these branches: Chopper spectrometers measure everything so this should be possible at the SNS on ARCS or SEQUOIA

Problem: Branches are much closer than the experimental resolution of neutron scattering instruments

Multizone phonon refinement gets around this obstacle

What the neutron data look like



Energy region of interest

Phonons: Where are the 5 branches? Kind of a mess.

Elastic scattering: Each bright spot is the center of a different Brillouin zone

Main Idea

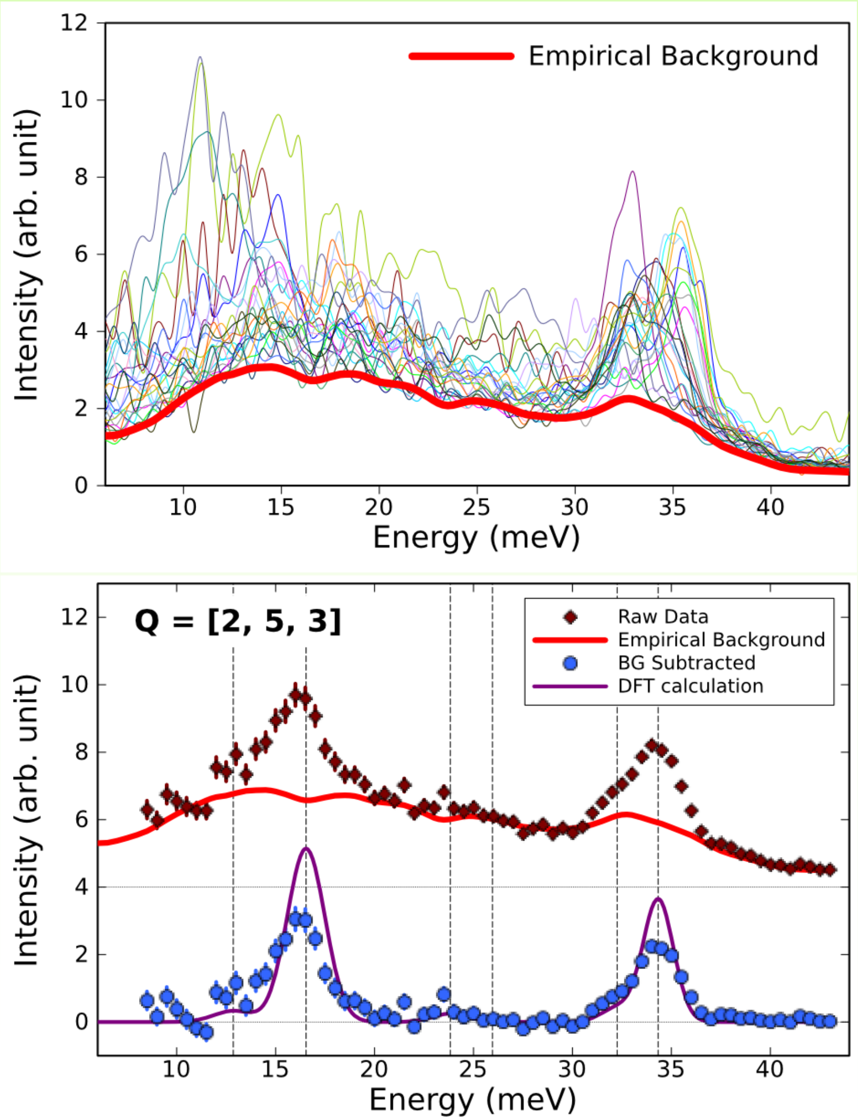
Want phonon frequencies, linewidths and eigenvectors at every reduced reciprocal lattice point q.

Every Brillouin zone contains all phonon peaks, but the intensity of each phonon is different in every zone.

We want to plot intensity as a function of energy at the same reduced q in every zone, and then do a global fit to all these data

But first we need to subtract background.

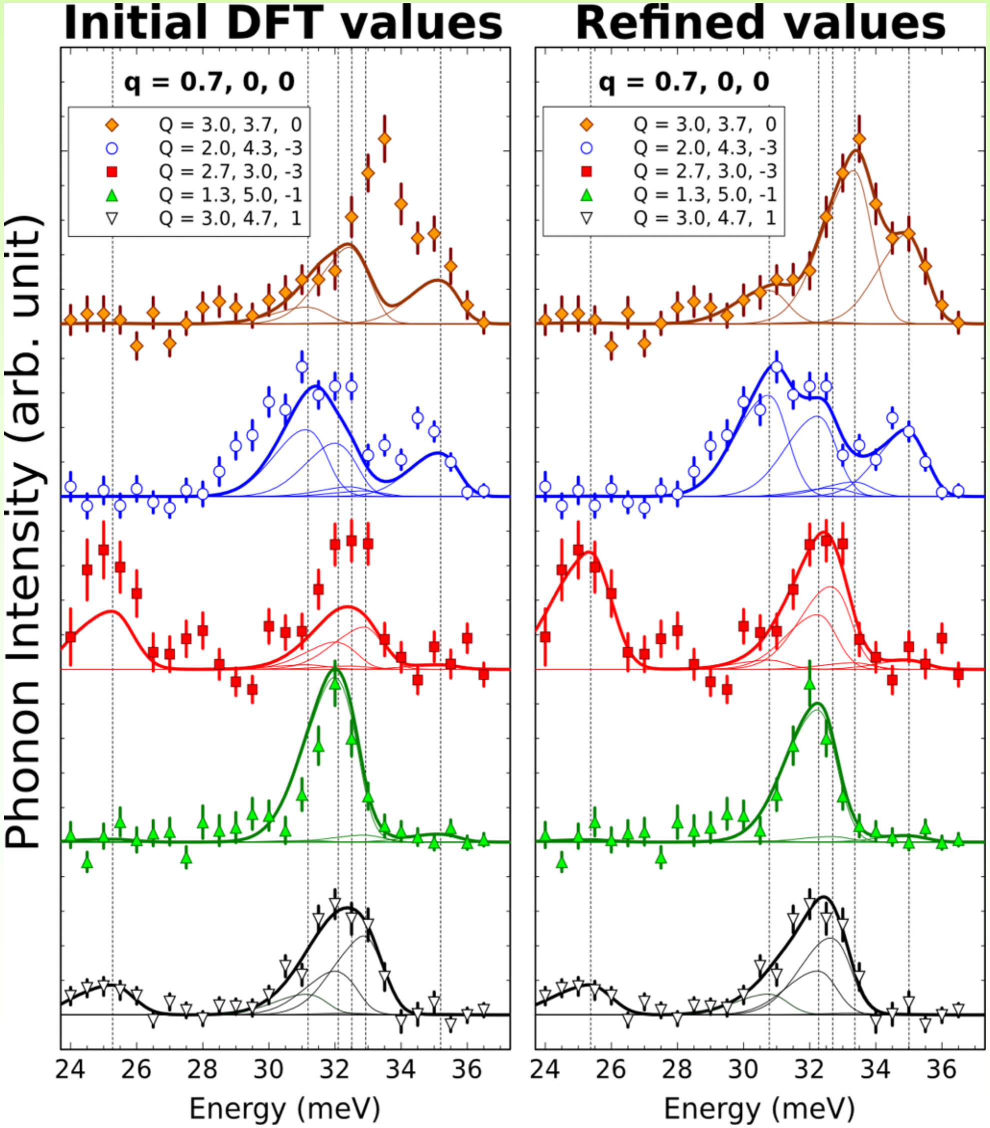
Background Subtraction



1. Divide all data by Q^2
2. Plot many constant Q cuts for a large random set of wavevectors
3. Background as a function of energy is the global minimum of intensity
4. Subtracting this background from raw yields the phonon spectrum

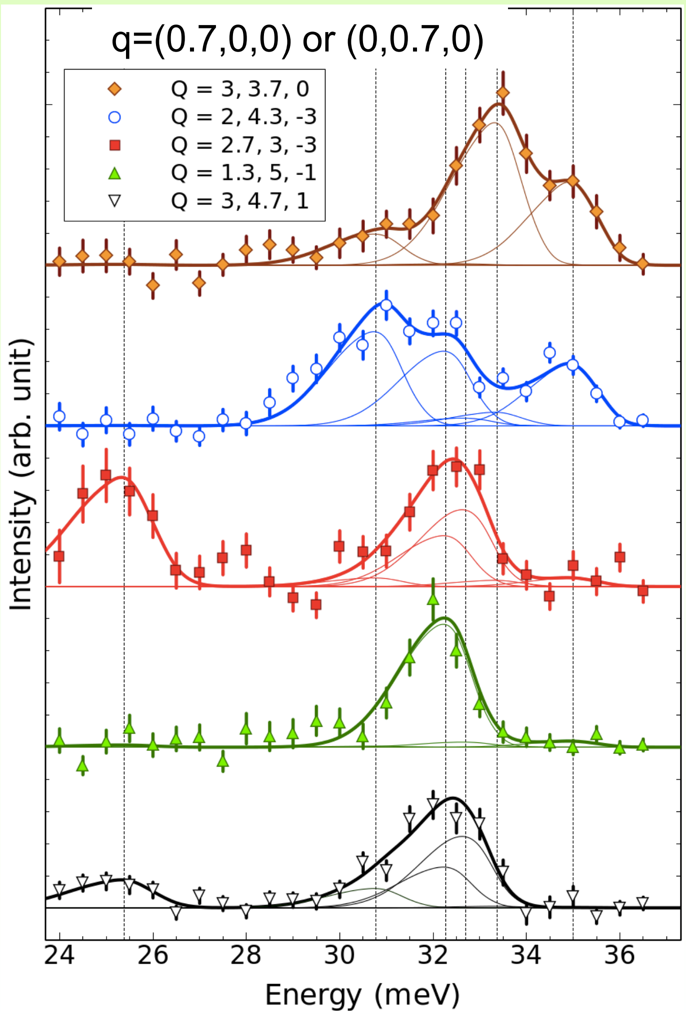
Fitting the phonon spectrum

Fit: Before and after



Starting from calculated spectra, the fit converged rapidly.

Phonons at the same reduced q but in different zones



Nearest zone center

3,3,0

2,5,-3

2,3,-3

2,5,-1

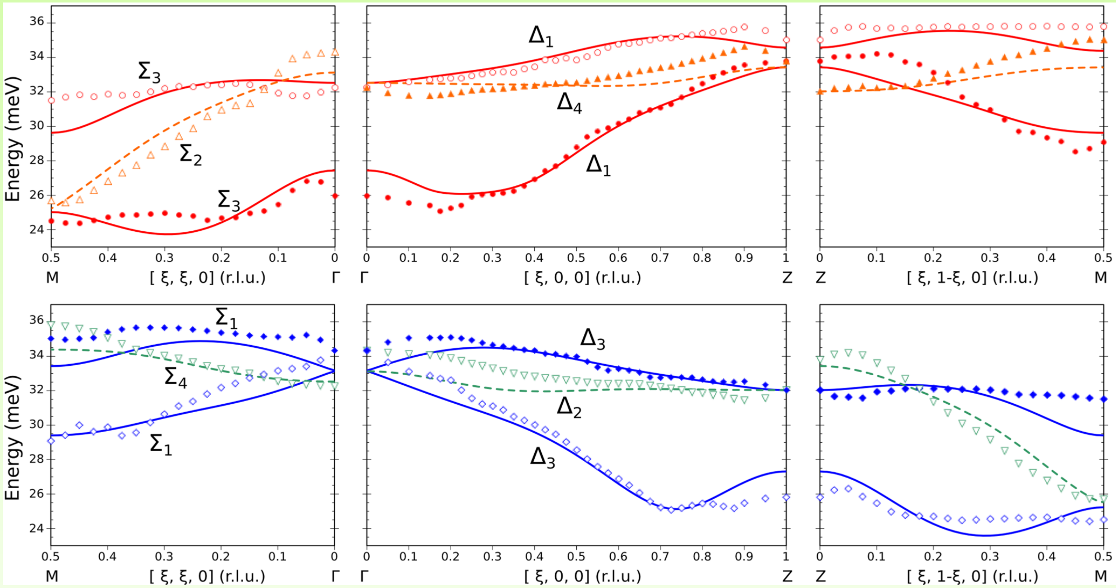
3,4,1

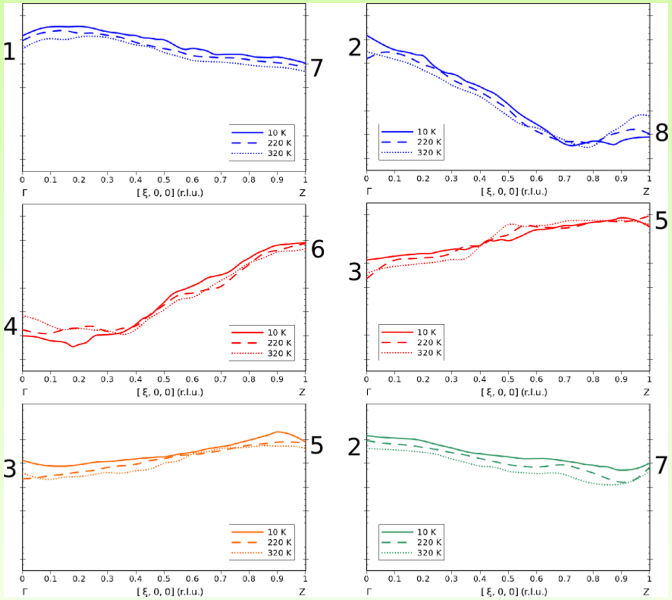
Same phonons in each curve, but with different intensities.

We fit all these curves + many more from other zones while *constraining the peak positions and linewidths to be the same in each curve* while letting intensities vary.

Such a fit allows us to determine where all phonons are much more accurately than from fitting any single curve.

Final result compared with DFT





The agreement with DFT is good. No significant anomalies

No observable temperature-dependence

Later weak T-dependence was observed in one phonon on a detwinned sample (Baron group)

Magnetic ordering transition in twnned $SrFe_2As_2$ at 200K does not noticably influence spectra of high-energy optic phonons

MRP allowed us to get around limits set by spectrometer resolution and resolve phonon branches that were very close together

Using MPR to find all phonons branches and improve statistics

New work on $HgBa_2CuO_4$ (Hg1201) in progress

New code allows fitting arbitrary datasets (any number of Brillouin zones and any number of phonon peaks. (limited by computer speed). No need to start with DFT calculations.

The background subtraction scheme that worked for Ba122 does not work here because the background depends on |Q| due to the phonon structure factor and dispersions in Al sample holder. Entirely new algorithm was developed for background subtraction

Problem: What are the phonon frequencies and lifetimes at a given reduced wavevector q.

1. Generate constant Q slices in every zone for every Q that correspond to q (about 100)
2. Calculate and subtract background
3. Manually select the files that will be used in the fitting (about 20)
4. Perform multizone fitting

Background subtraction

Background has a powder spectrum, i.e. depends on |Q| only

Background determination procedure for a given wavevector **Q**:

Starting from the wavevector of interest move away a random angle away to a wavevector that has the same length but different direction.

Do a constant Q cut there and draw a smooth line through the data

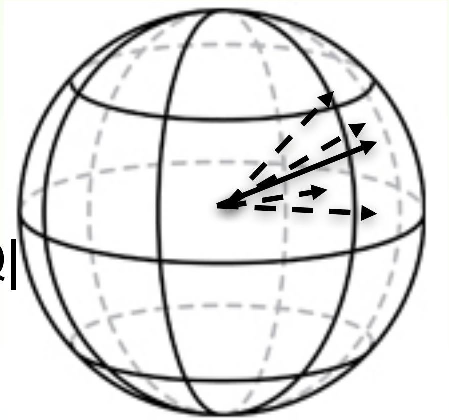
Go to another wavevector of the same length but different direction

Do a constant Q cut there and draw a smooth line through the data

Repeat ~20 times.

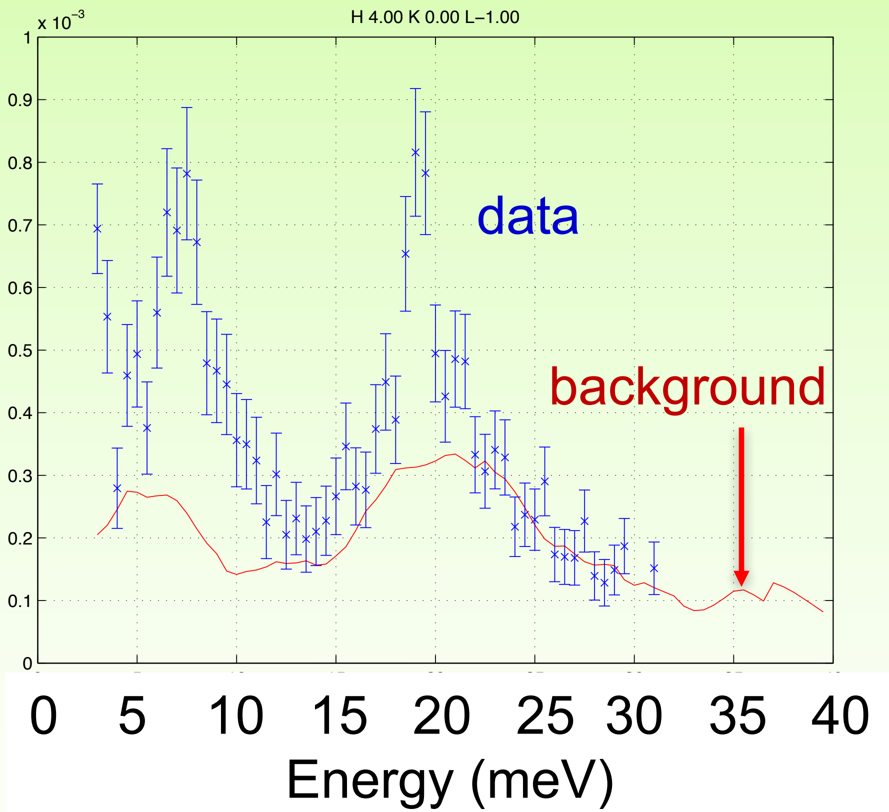
The background is the minimum of these 20 smooth curves.

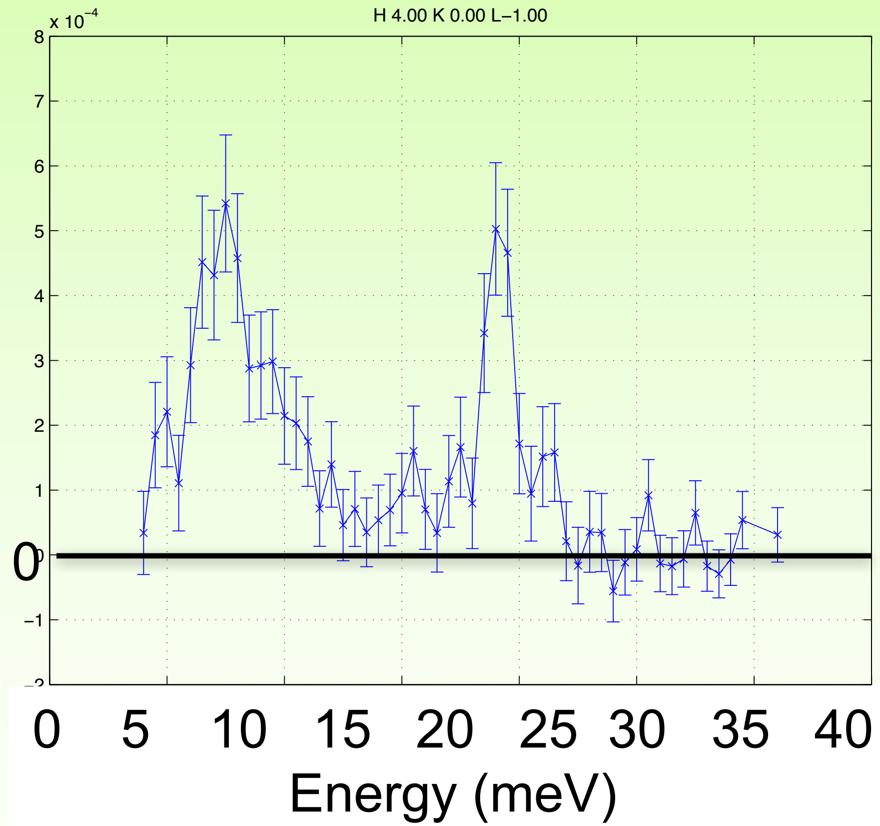
Radius = |Q|



Background subtraction

zone center Q=(4, 0 , 10)





Clean peaks after background subtraction.

Note that the broad component of the peak at 18meV disappears after background subtraction.

Manually select files that will be fitted and guess peak positions

We ended up with ~ 90 datasets (one for each wavevector contained in the measured $S(Q,\omega)$. Majority of the datasets have no discernible peaks after background subtraction. Only ~20 have identifiable phonon peaks and these were manually selected.

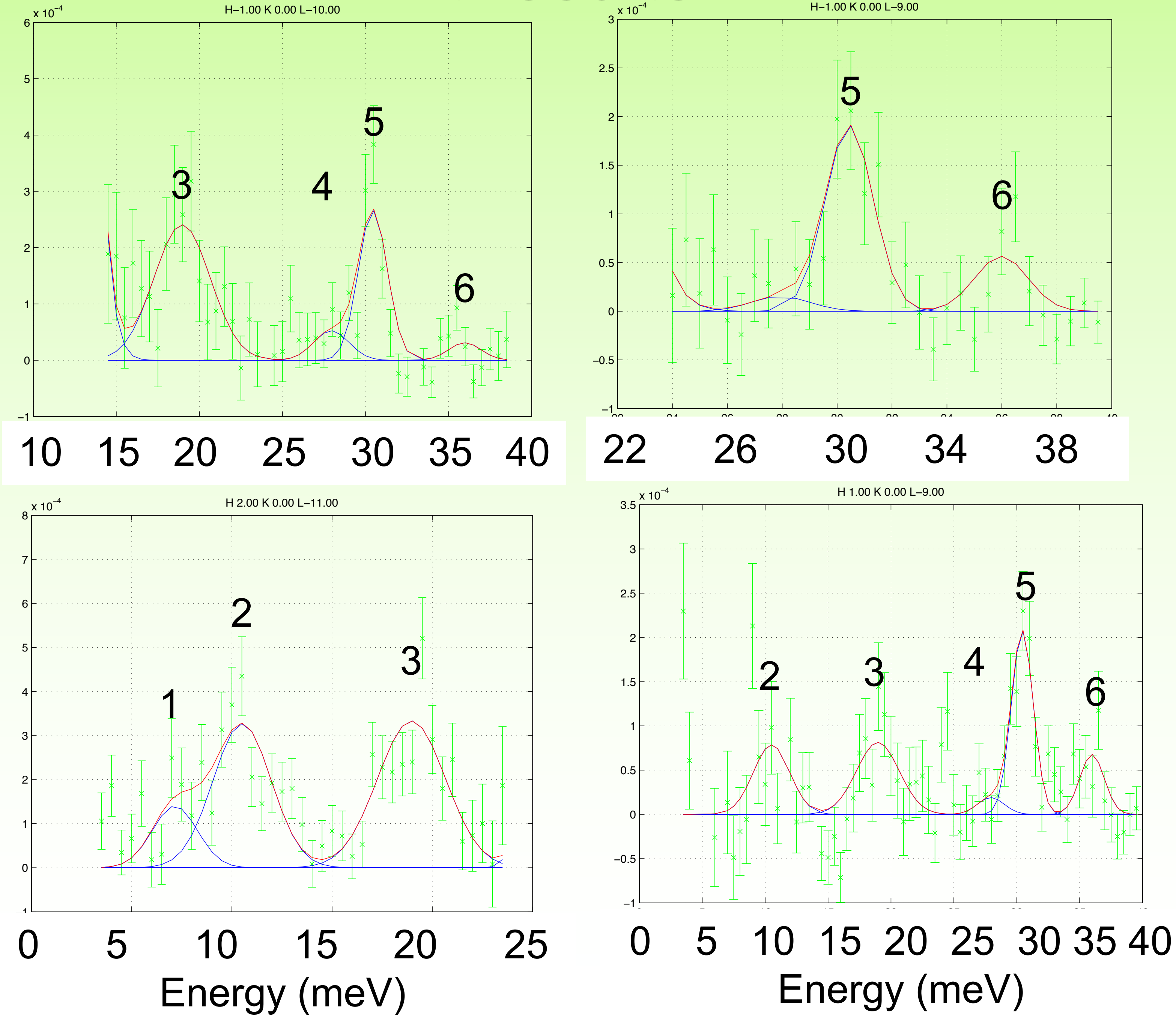
Peak positions guessed based on visual inspection of the 20 datasets and entered into a text file.

Multizone fitting of the 20 datasets as before

The main source of uncertainty: Background determination

Accurate eigenvector determination is impossible, because there is a chance that a part of the peak was included in the background.

Fit results


H−1.00 K 0.00 L−10.00
3
4
5
6
10 15 20 25 30 35 40
H−1.00 K 0.00 L−9.00
5
6
22 26 30 34 38
H 2.00 K 0.00 L−11.00
1
2
3
0 5 10 15 20 25
Energy (meV)
H 1.00 K 0.00 L−9.00
2
3
4
5
6
0 5 10 15 20 25 30 35 40
Energy (meV)

Output

peak positions

linewidths

```
7.1555  10.5121 18.9597 27.9403 30.4441 35.9807
1.5867  2.0552  2.4031  1.5021  1.2344  1.3690

H 1.00 K 0.00 L-9.00
2.430938e-09    7.854154e-05    8.137369e-05    1.902531e-05
2.077175e-04    6.777959e-05


H 2.00 K 0.00 L-11.00
1.399424e-04    3.269418e-04    3.332818e-04    1.157936e-01
1.002177e-06    1.000000e-06


H-1.00 K 0.00 L-10.00
9.570378e-07    9.542006e-03    2.406922e-04    5.254494e-05
2.661487e-04    3.116029e-05


H-1.00 K 0.00 L-9.00
1.000000e-06    1.000000e-06    3.370253e-03    1.526843e-05
1.909012e-04    5.649075e-05
```

amplitudes for each zone

Neutron vs. x-ray

Neutron

Advantages

Penetrate deep into the bulk

Couple to the lattice and magnetic degrees of freedom

Very high resolution energy (down to 1 μeV) “easy”

Gaussian resolution function

Multichannel detection allows mapping large regions of reciprocal space

Disadvantages

Background often high and not flat

Big samples required

High energy-low Q impossible

Spurious peaks often appear

Certain isotopes absorb neutrons

Poor q-resolution

Flux-limited

X-ray

Advantages

Background well behaved

Small samples may be measured

High q-resolution

Resolution ellipsoid not tilted

Almost no background

Electronic Charge excitations may be explored

Large range of Q-E can be measured the same monochromator

No spurious peaks

Disadvantages

Not enough beamtime available!!!

Lorentzian energy resolution function

Very high energy resolution difficult

Sensitive to surface contamination

Does not couple to electronic spins

Weak signal from vibrations involving light atoms

Flux limited: reciprocal space mapping impossible

What if x-rays had X100 intensity?

In this case mapping reciprocal space becomes feasible.

Now with the well behaved background the main source of uncertainty disappears and it will become possible to extract phonon amplitudes and, therefore, eigenvectors from the data. Also, data analysis becomes simpler and will take less computer time.

Will be possible to do MPR on small samples.

Disadvantage: Hard to see light atoms.

This would be a huge step in measuring lattice dynamics in single crystals

Need Large Q Range!!!