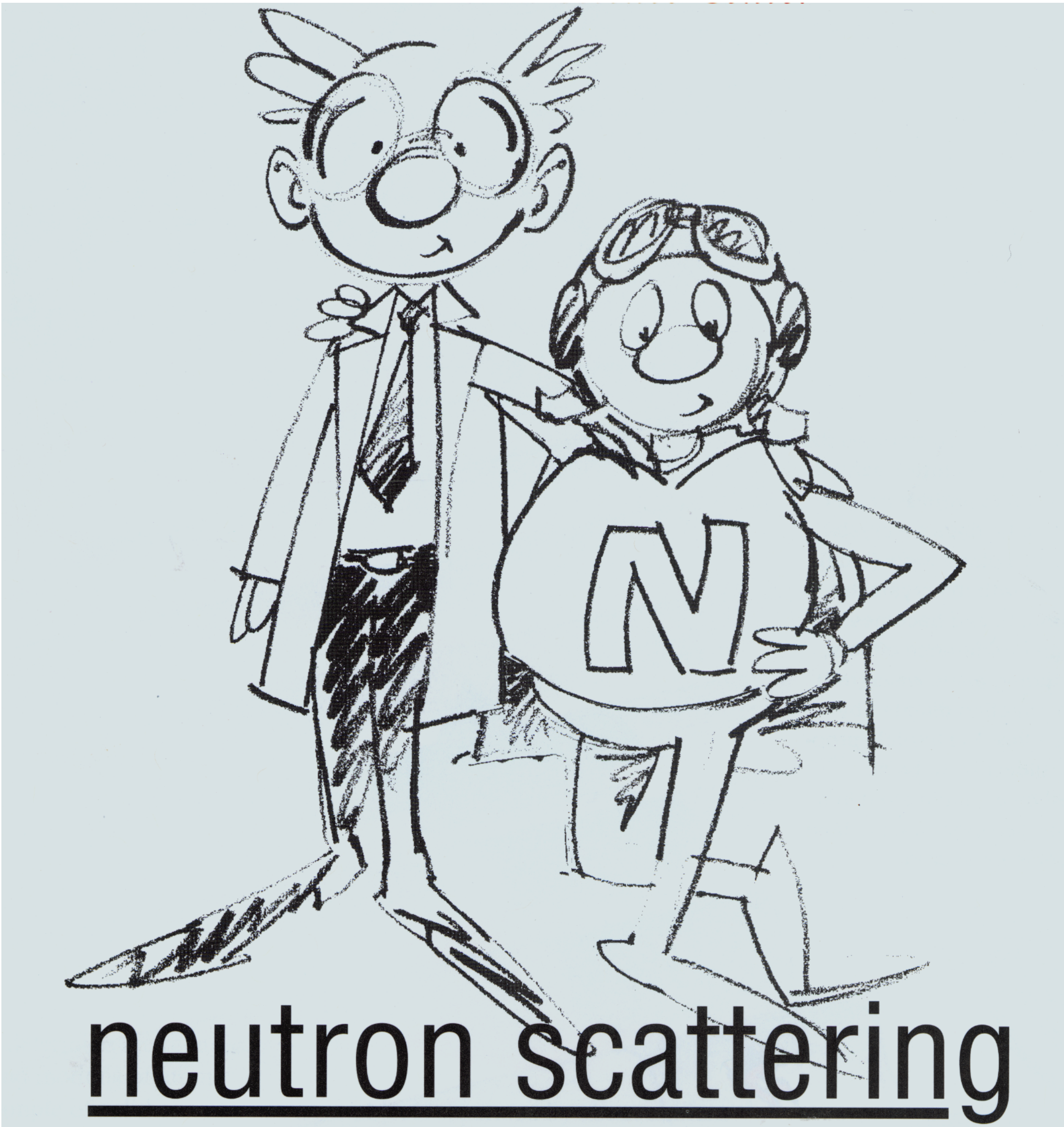


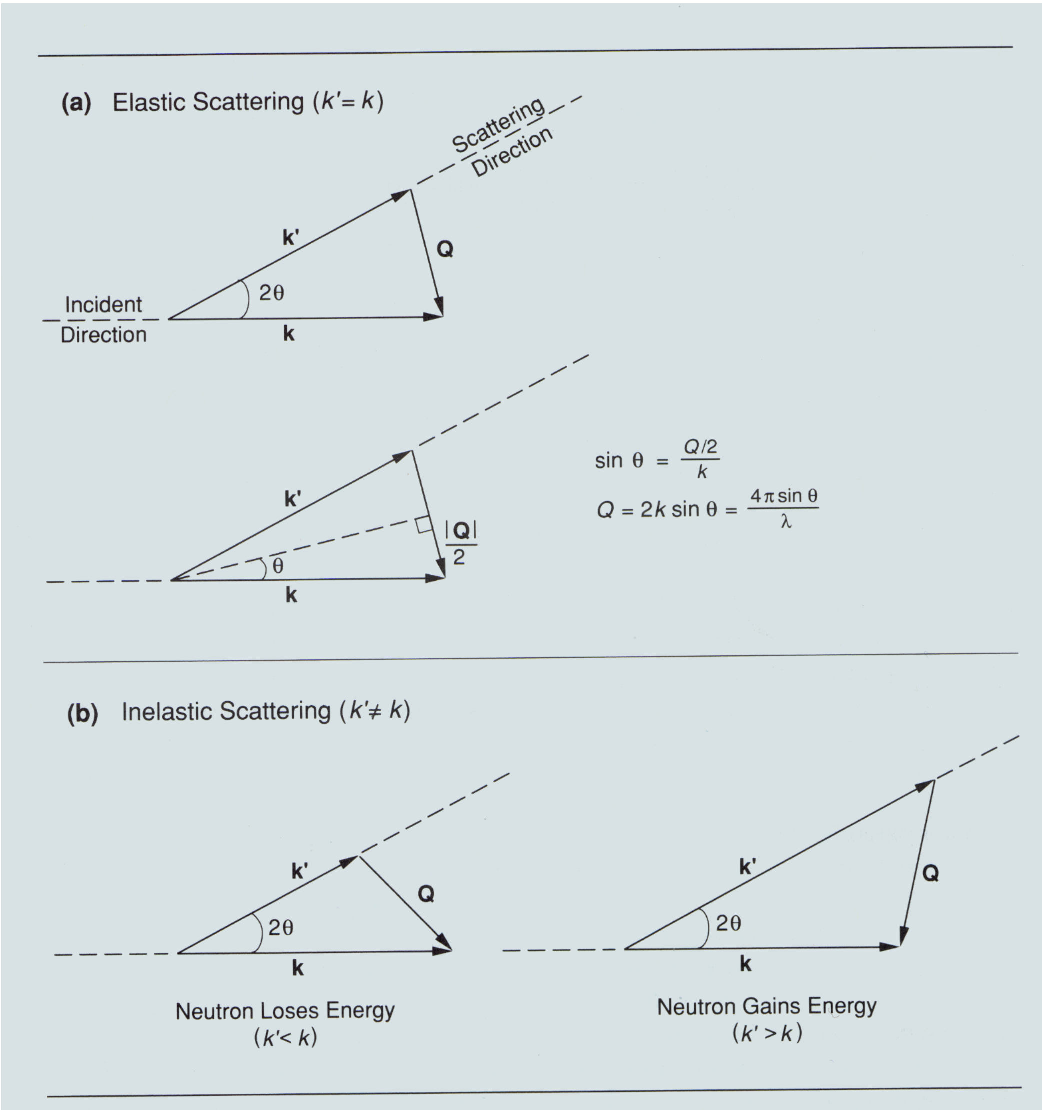
Inelastic Scattering

by

Roger Pynn

Indiana University

The Neutron Changes Both Energy & Momentum When Inelastically Scattered by Moving Nuclei



inelastic scattering

Scattering in which exchange of energy and momentum between the incident neutron and the sample causes both the direction and the magnitude of the neutron's wave vector to change.

The Elastic & Inelastic Scattering Cross Sections Have an Intuitive Similarity

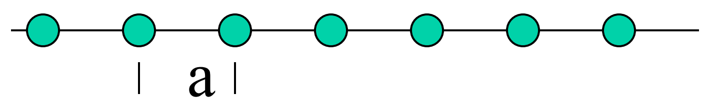
- The intensity of elastic, coherent neutron scattering is proportional to the spatial Fourier Transform of the Pair Correlation Function, $G(r)$ I.e. the probability of finding a particle at position r if there is simultaneously a particle at r=0

- The intensity of inelastic coherent neutron scattering is proportional to the space *and time* Fourier Transforms of the *time-dependent* pair correlation function function, $G(r,t)$ = probability of finding a particle at position r *at time t* when there is a particle at r=0 and *t=0*.

- For inelastic *incoherent* scattering, the intensity is proportional to the space and time Fourier Transforms of the *self-correlation* function, $G_s(r,t)$ I.e. the probability of finding a particle at position r at time t when *the same* particle was at r=0 at t=0

Diffraction from a Frozen Wave

- Recall that $S(\vec{Q}) = \frac{1}{N}\left|\sum_k e^{i\vec{Q}.\vec{r}_k}\right|^2$

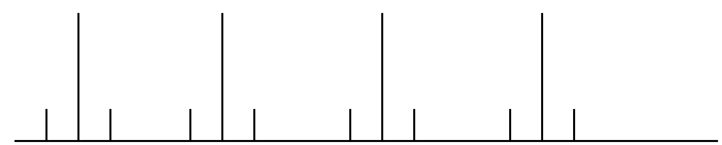


- We know that for a linear chain of “atoms” along the x axis, $S(Q_x)$ is just a series of delta function reciprocal lattice planes at Q_x = n2p/a, where a is the separation of atoms

What happens if we put a "frozen" wave in the chain of atoms so that the atomic positions are $x_p = pa + u\cos kpa$ where p is an integer and u is small?

$$S(Q) = \left|\sum_p e^{iQpa} e^{iQu\cos kpa}\right|^2 \approx \left|\sum_p e^{iQpa}(1 + iQu[e^{ikpa} + e^{-ikpa}])\right|^2$$

$$\approx \left|\sum_p e^{iQpa} + iQu[e^{i(Q+k)pa} + e^{i(Q-k)pa}]\right|^2$$



so that in addition to the Bragg peaks we get weak satellites at $Q = G \pm k$

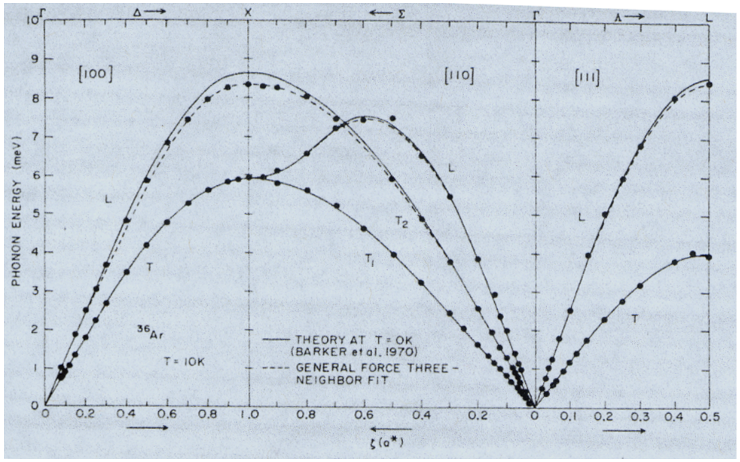
What Happens if the Wave Moves?

- If the wave moves through the chain, the scattering still occurs at wavevectors G + k and G – k but now the scattering is inelastic

- For quantized lattice vibrations, called phonons, the energy change of the neutron is $\hbar\omega$ where ω is the vibration frequency.

- In a crystal, the vibration frequency at a given value of $\vec{k}$ (called the phonon wavevector) is determined by interatomic forces. These frequencies map out the so-called phonon dispersion curves.

- Different branches of the dispersion curves correspond to different types of motion



phonon dispersion in ^{36}Ar

A Phonon is a Quantized Lattice Vibration

- Consider linear chain of particles of mass M coupled by springs. Force on n'th particle is

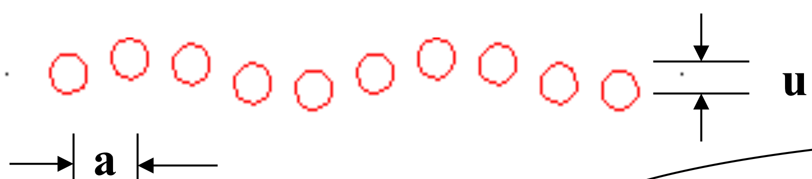
$$F_n = \alpha_0 u_n + \alpha_1 (u_{n-1} + u_{n+1}) + \alpha_2 (u_{n-2} + u_{n+2}) + \ldots$$

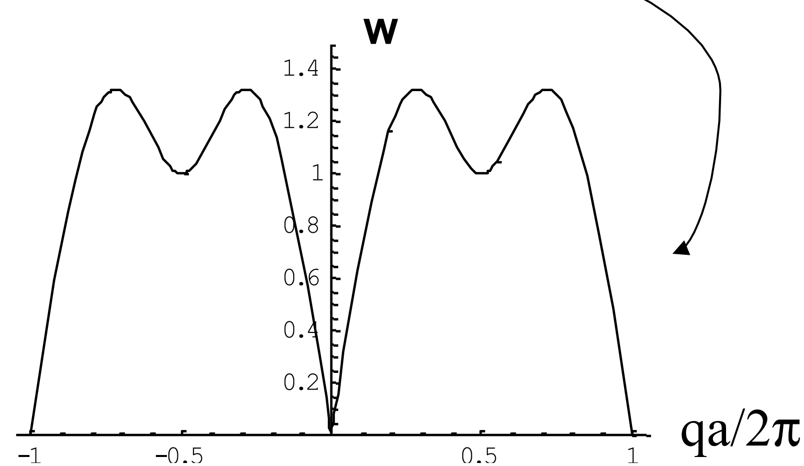
First neighbor force constant

displacements

- Equation of motion is $F_n = M\ddot{u}_n$
- Solution is: $u_n(t) = A_q e^{i(qna - \omega t)}$ with $\omega_q^2 = \frac{4}{M}\sum_\nu \alpha_\nu \sin^2(\frac{1}{2}\nu qa)$

$$q = 0, \pm\frac{2\pi}{L}, \pm\frac{4\pi}{L}, \ldots\ldots \pm\frac{N}{2}\frac{2\pi}{L}$$

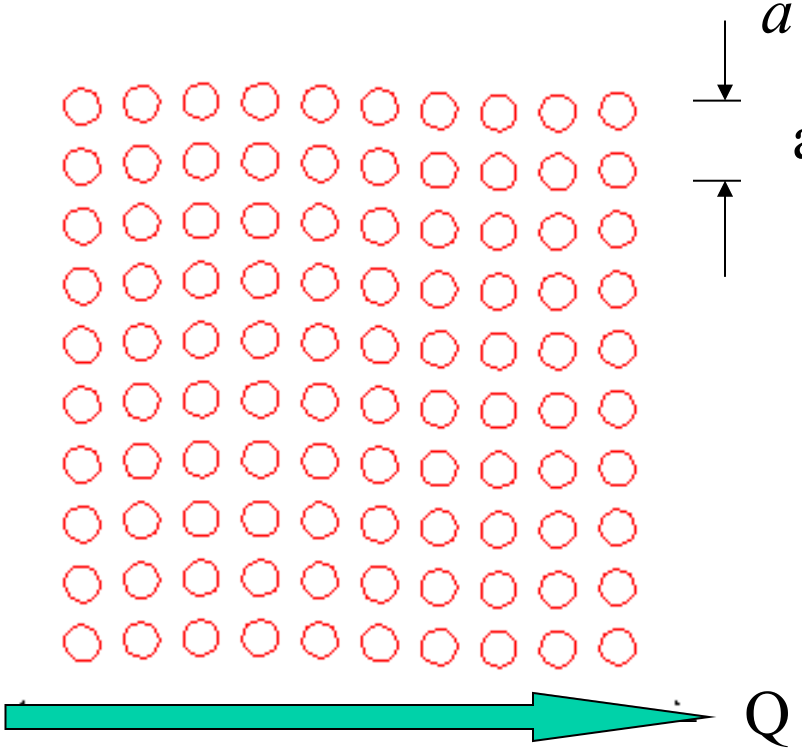




Phonon Dispersion Relation:
Measurable by inelastic neutron scattering

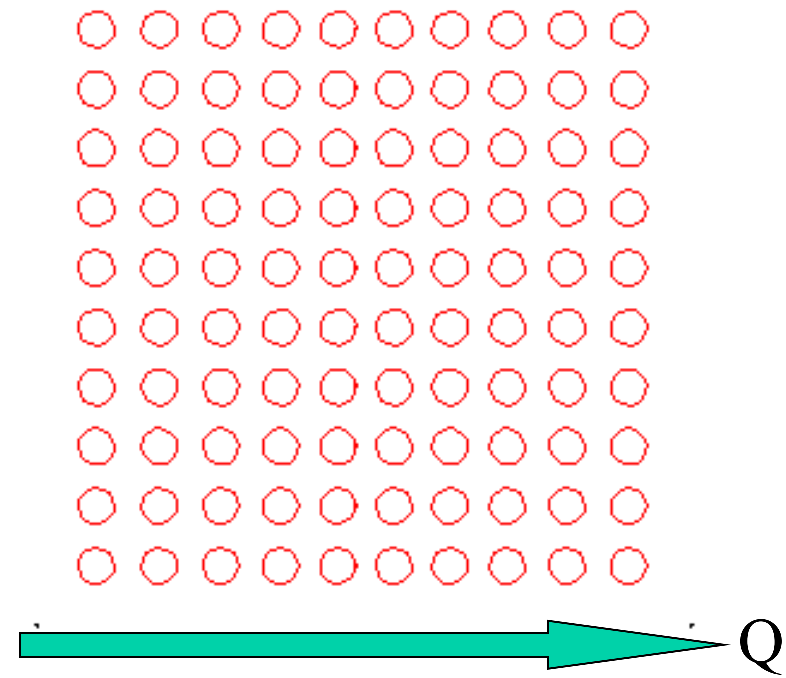
Atomic Motions for Longitudinal & Transverse Phonons

$$\vec{Q} = \frac{2\pi}{a}(0.1,0,0)$$



Transverse phonon

$$\vec{e}_T = (0,0.1,0)a$$

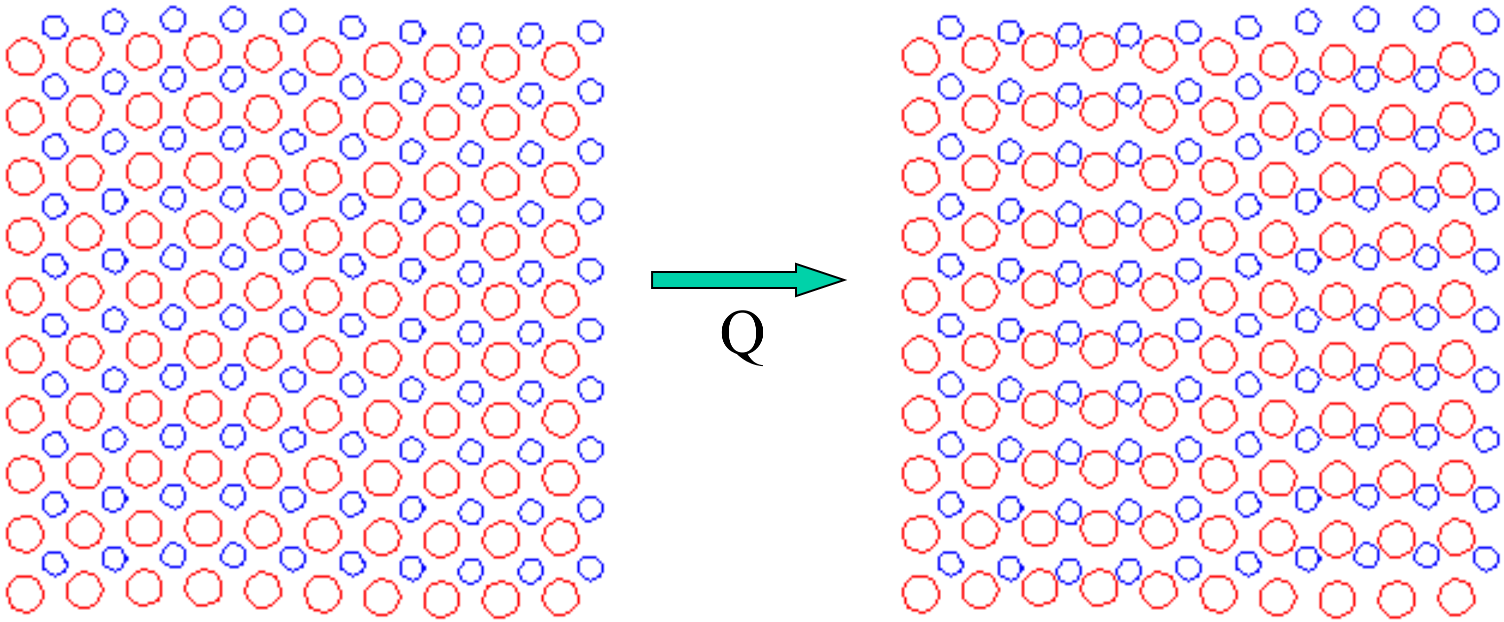


Longitudinal phonon

$$\vec{e}_L = (0.1,0,0)a$$

$$\vec{R}_l = \vec{R}_{l0} + \vec{e}_s e^{i(\vec{Q}.\vec{R}_l - \omega t)}$$

Transverse Optic and Acoustic Phonons



Acoustic

$$\vec{e}_{red} = (0,0.1,0)a$$

$$\vec{e}_{blue} = (0,0.14,0)a$$

Optic

$$\vec{e}_{red} = (0,0.1,0)a$$

$$\vec{e}_{blue} = (0,-0.14,0)a$$

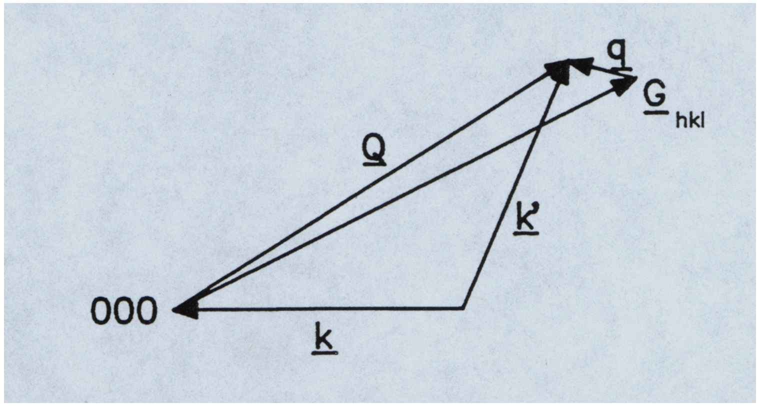
$$\vec{R}_{lk} = \vec{R}_{lk}^{0} + \vec{e}_s e^{i(\vec{Q}.\vec{R}_l - \omega t)}$$

Phonons – the Classical Use for Inelastic Neutron Scattering

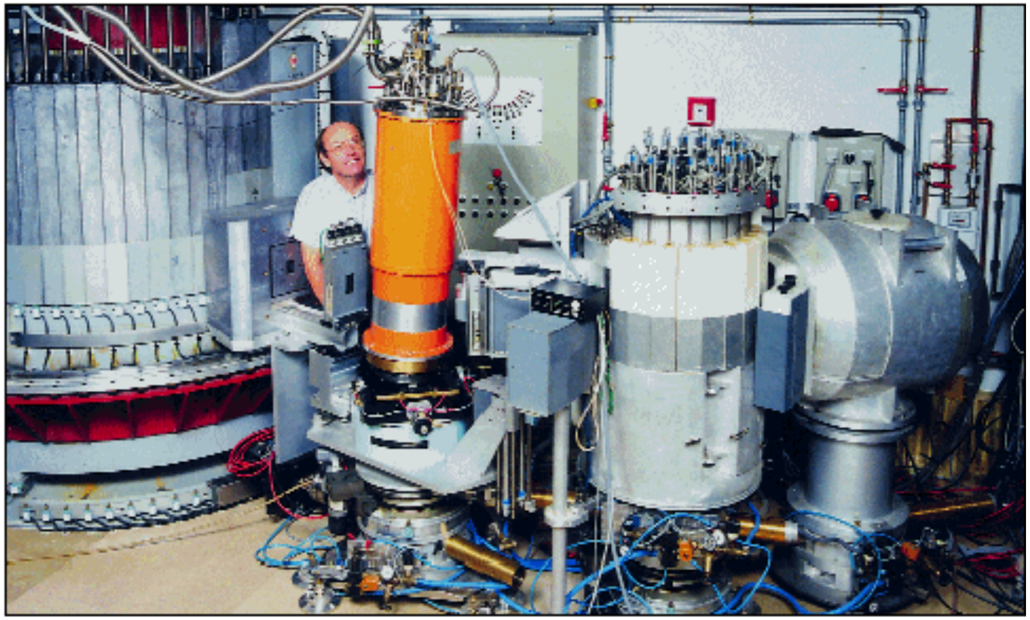
- Coherent scattering measures scattering from single phonons

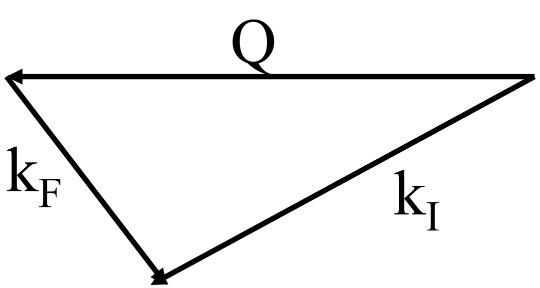
$$\left(\frac{d^2\sigma}{d\Omega dE}\right)_{coh\pm1} = \sigma_{coh}\frac{k'}{k}\frac{\pi^2}{MV_0}e^{-2W}\sum_s\sum_G\frac{(\vec{Q}.\vec{e}_s)^2}{\omega_s}(n_s+\frac{1\pm1}{2})\delta(\omega\mp\omega_s)\delta(\vec{Q}-\vec{q}-\vec{G})$$

- Note the following features:
  - Energy & momentum delta functions => see single phonons (labeled *s*)
  - Different thermal factors for phonon creation (n_s+1) & annihilation (n_s)
  - Can see phonons in different Brillouin zones (different recip. lattice vectors, ***G***)
  - Cross section depends on relative orientation of ***Q*** & atomic motions (***e**$_s$*)
  - Cross section depends on phonon frequency (ω_s) and atomic mass (M)
  - In general, scattering by multiple excitations is either insignificant or a small correction (the presence of other phonons appears in the Debye-Waller factor, W)

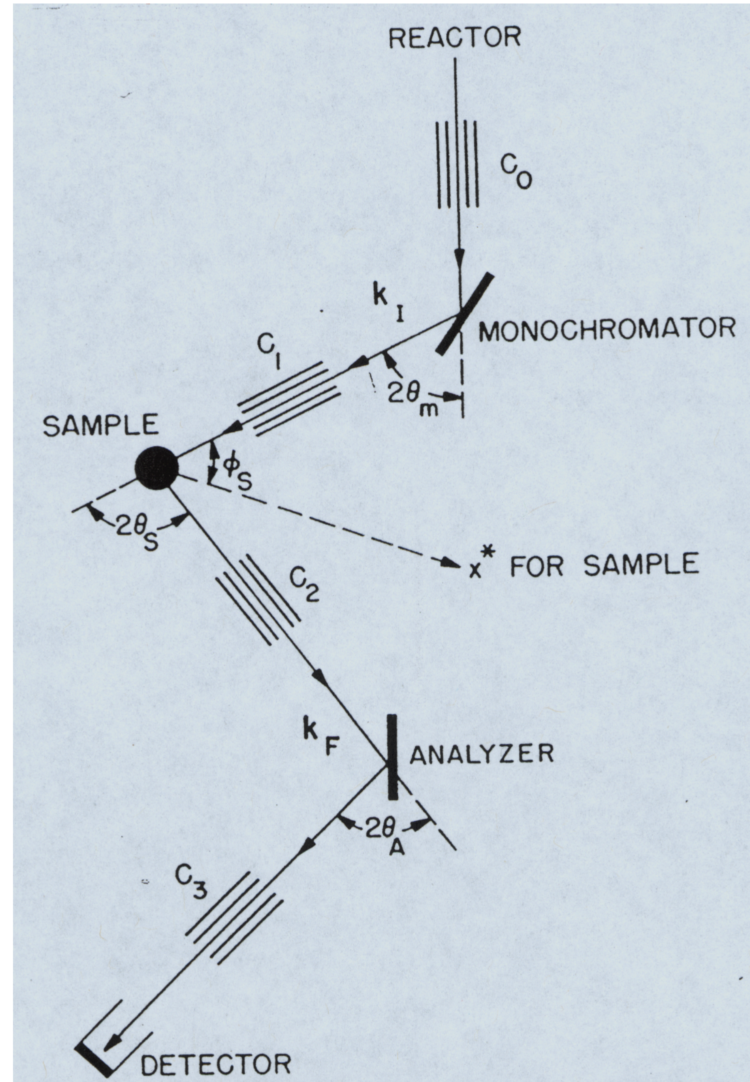


The Workhorse of Inelastic Scattering Instrumentation at Reactors Is the Three-axis Spectrometer



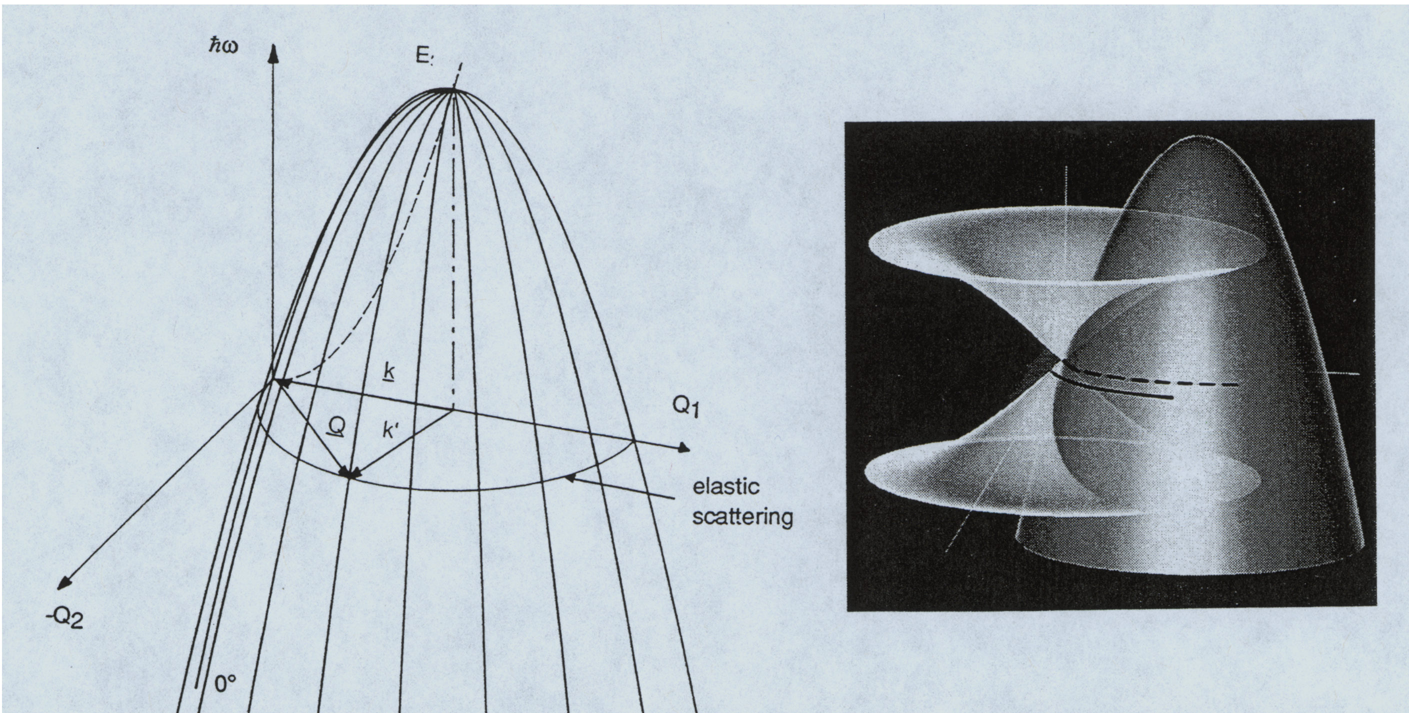


“scattering triangle”



The Accessible Energy and Wavevector Transfers Are Limited by Conservation Laws

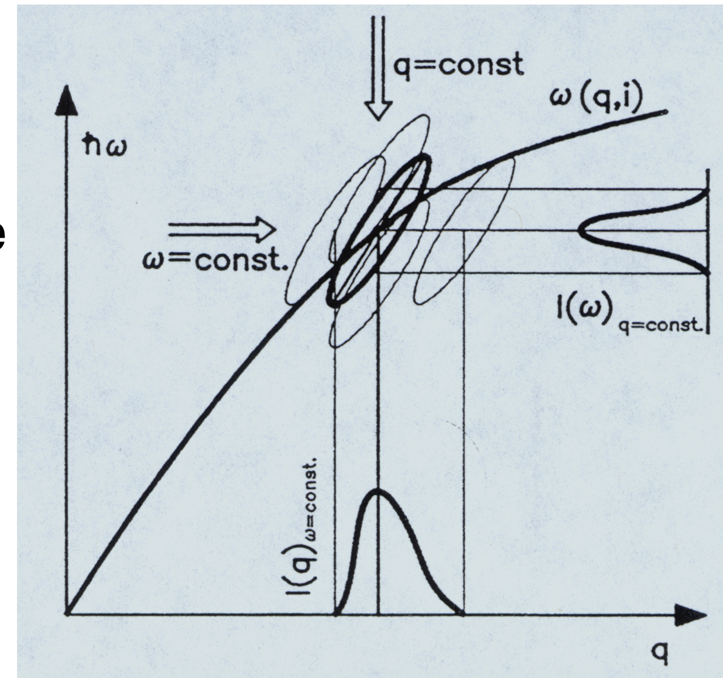
- Neutron cannot lose more than its initial kinetic energy & momentum must be conserved

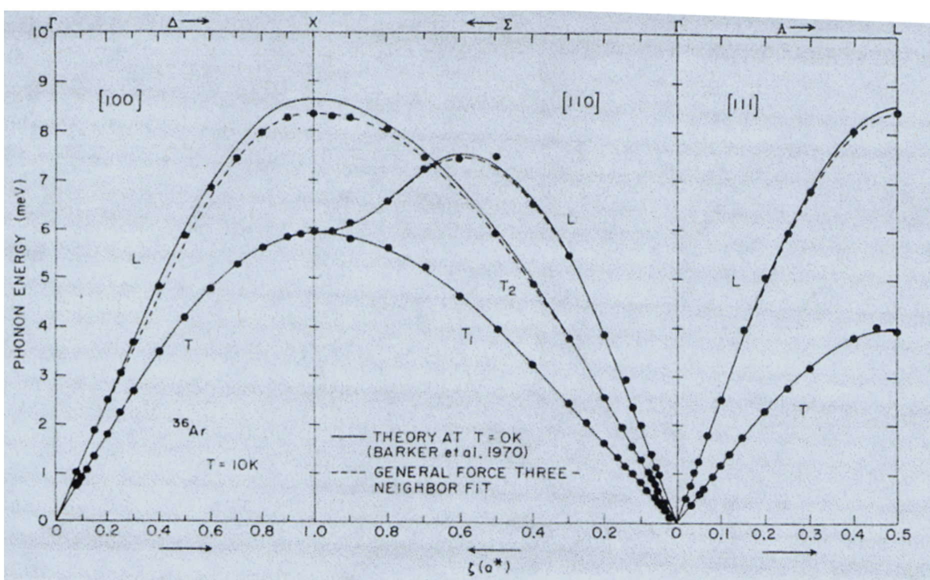


Intersection of the dynamical range surface (paraboloid) with a (rotationally symmetric) dispersion surface. The projection of the lines of intersection into the Q-plane are different for energy gain and energy loss

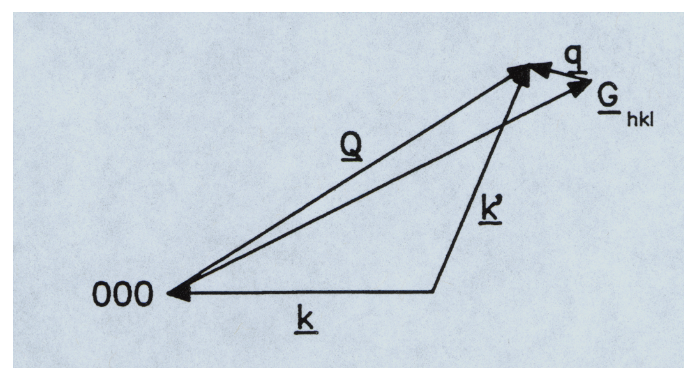
Triple Axis Spectrometers Have Mapped Phonons Dispersion Relations in Many Materials

- Point by point measurement in (Q,E) space
- Usually keep either k_I or k_F fixed
- Choose Brillouin zone (I.e. G) to maximize scattering cross section for phonons
- Scan usually either at constant-Q (Brockhouse invention) or constant-E

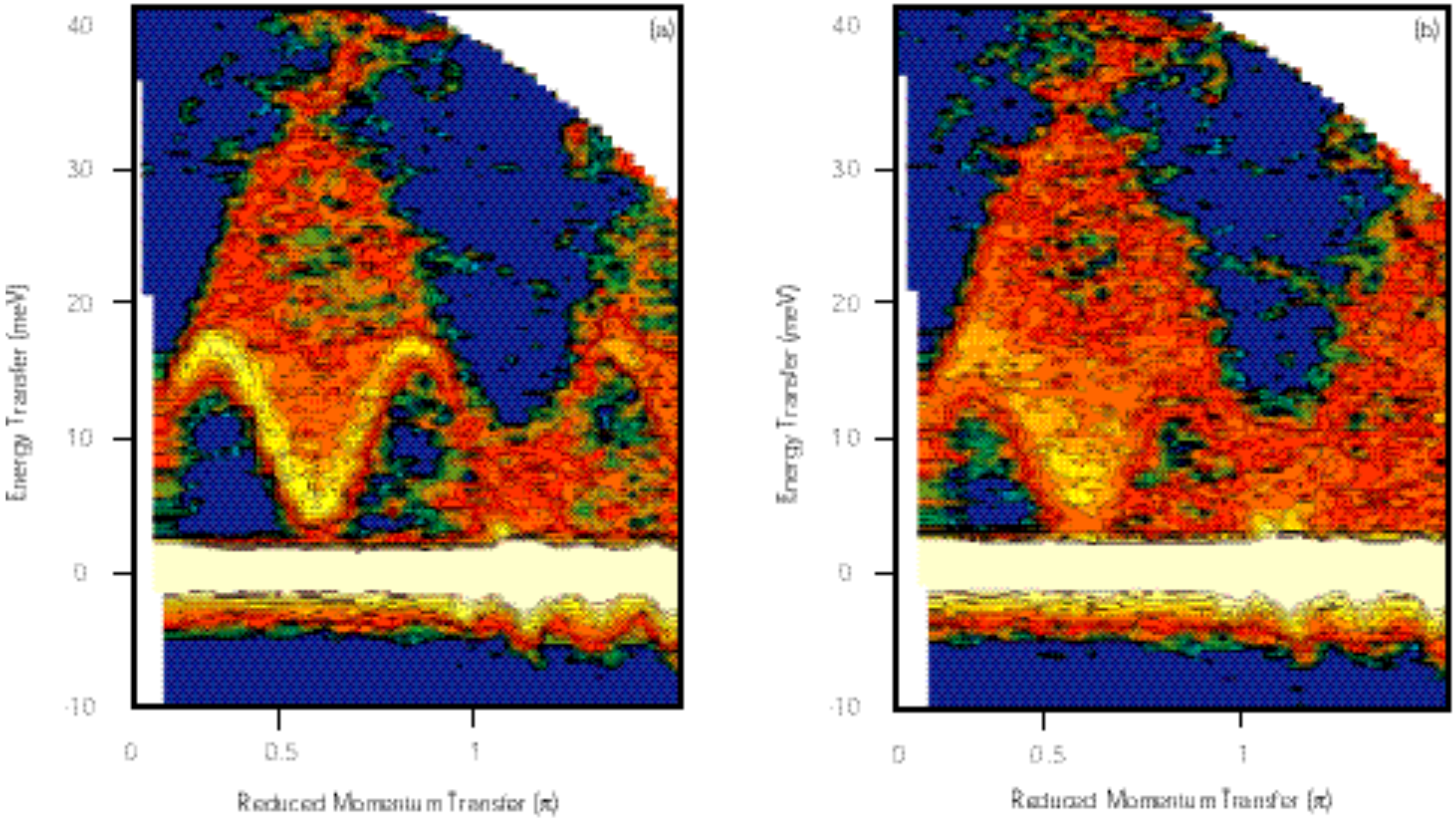




Phonon dispersion of ^{36}Ar



Time-of-flight Methods Can Give Complete Dispersion Curves at a Single Instrument Setting in Favorable Circumstances



$CuGeO_3$ is a 1-d magnet. With the unique axis parallel to the incident neutron beam, the complete magnon dispersion can be obtained

What Use Have Phonon Measurements Been?

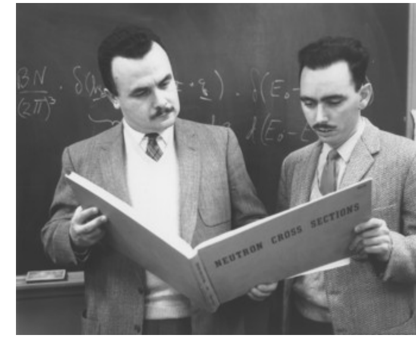
- Quantifying interatomic potentials in metals, rare gas solids, ionic crystals, covalently bonded materials etc
- Quantifying anharmonicity (I.e. phonon-phonon interactions)
- Measuring soft modes at 2^{nd} order structural phase transitions
- Electron-phonon interactions including Kohn anomalies
- Roton dispersion in liquid He
- Relating phonons to other properties such as superconductivity, anomalous lattice expansion etc

PHYSICAL REVIEW VOLUME 128, NUMBER 3 NOVEMBER 1, 1962

Crystal Dynamics of Lead. I. Dispersion Curves at 100°K

B. N. Brockhouse,* T. Arase,† G. Caglioti,‡ K. R. Rao,§ and A. D. B. Woods
Neutron Physics Branch, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada

(Received June 4, 1962)



Brockhouse and Woods

Brockhouse's first 3 axis spectrometer at NRU reactor in 1959

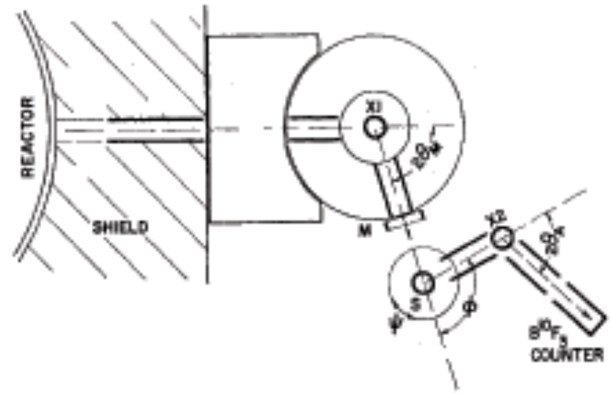
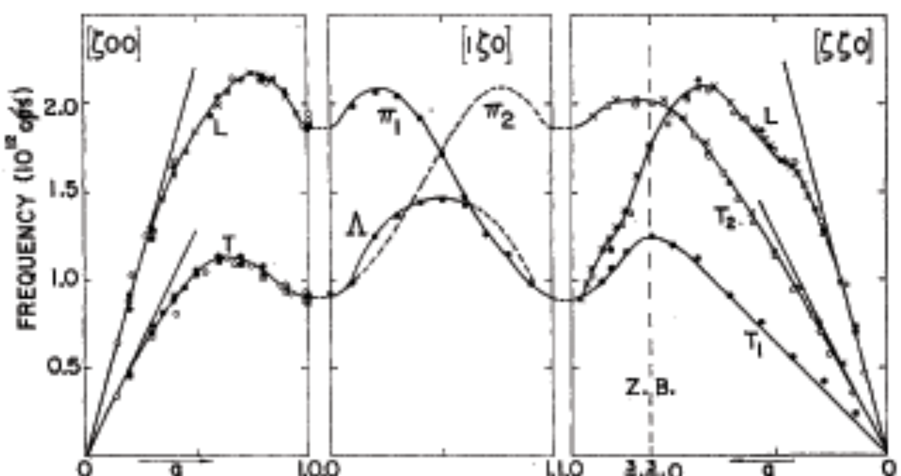
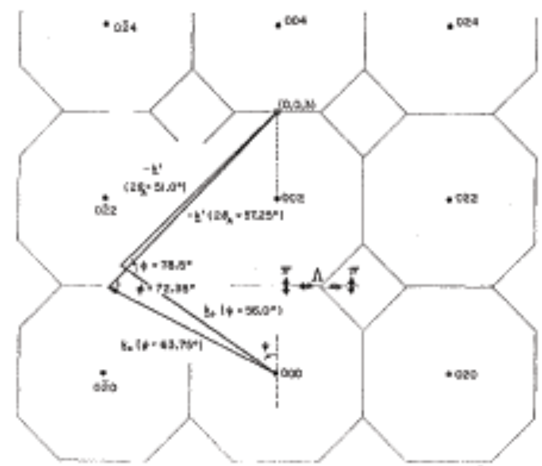


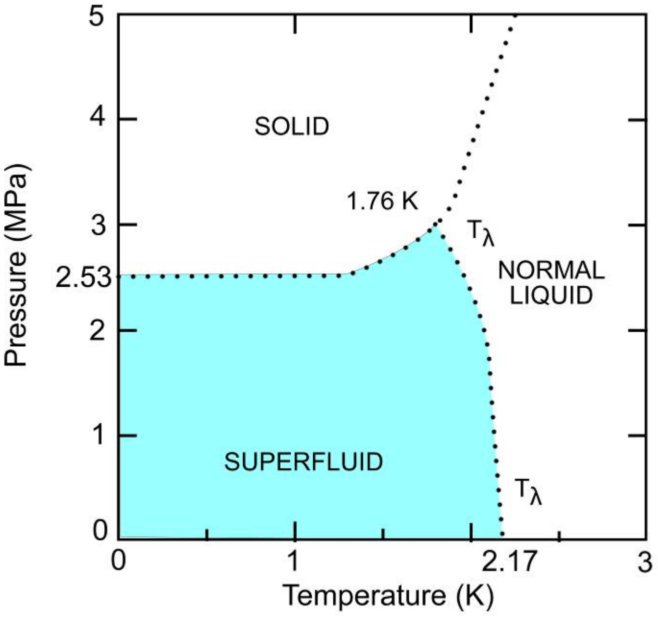
Fig. 1. Schematic drawing of the apparatus.

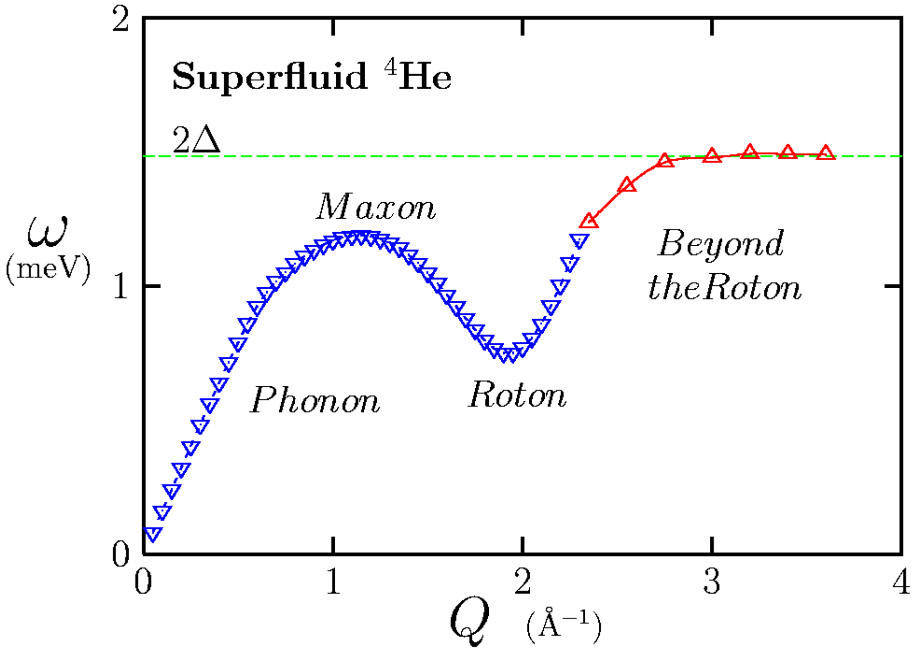




The constant-Q method

Roton Minimum in Superfluid ^{4}He was Predicted by Landau





Neutron scattering studies of structural phase transitions at Brookhaven*

G. Shirane

Brookhaven National Laboratory, Upton, New York 11973

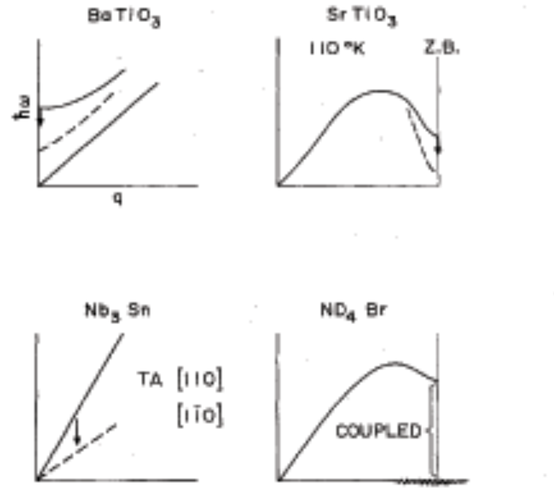


FIG. 3. Typical soft mode (arrows) phase transitions studied at Brookhaven. These represent temperature-dependent phonon dispersion relations $\hbar\omega$ vs q.

Gen Shirane

Tormod Riste

$SrTiO_3$ looked like a simple mean-field displacive phase transition described by a soft-mode theory until Riste discovered the Central Peak

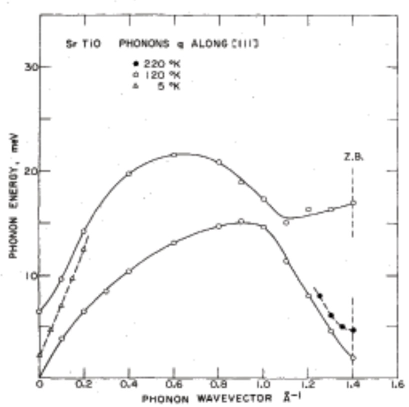
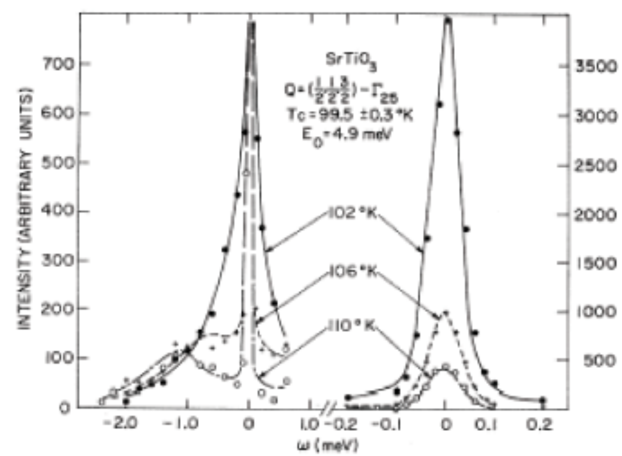


FIG. 6. Temperature-dependent phonon modes in $SrTiO_3$ measured by Shirane and Yamada (1969). The 110°K transition is caused by the soft mode at the zone boundary. Soft mode near the origin is due to incipient ferroelectricity.



The Neutron Scattering Society of America

www.neutronscattering.org

Press Release May 1, 2006

Dr. Taner Yildirim

is the recipient of the

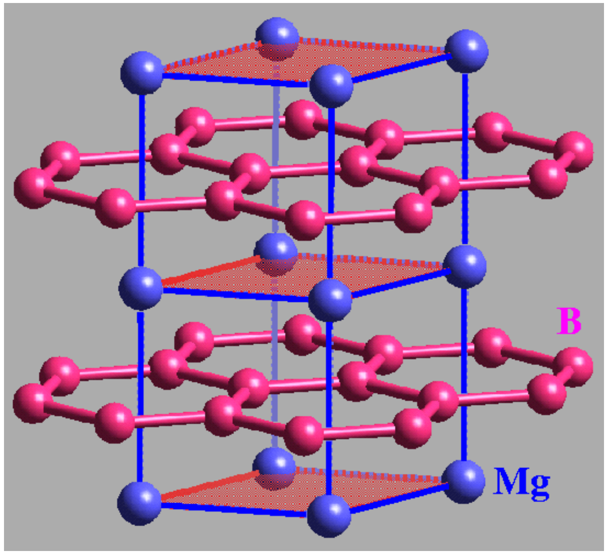
2006 Science Prize

of the Neutron Scattering Society of America with the citation:

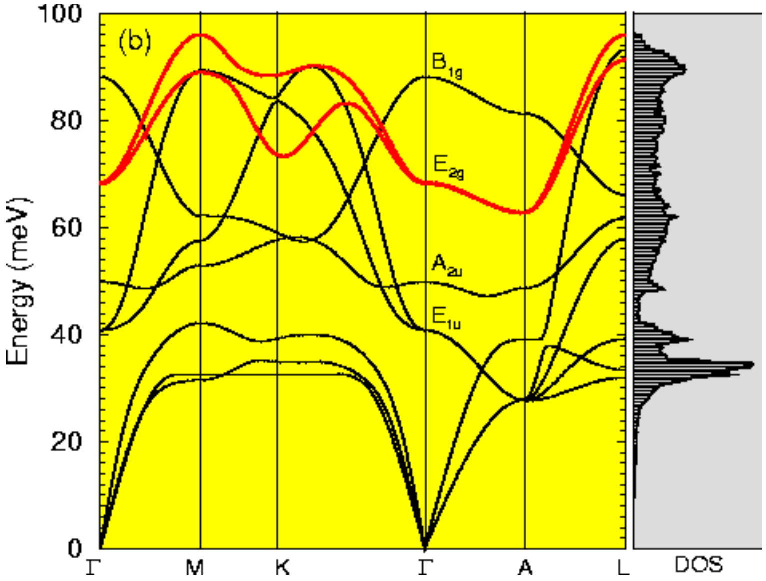
"For his innovative coupling of first principles theory with neutron scattering to solve critical problems in materials sciences"

MgB_2 Superconducts at 40K. Why?

- Yildirim did first-principles calculation of phonons in MgB_2 (particulary anharmonicity & electron-phonon interaction) & compared with neutron scattering



Crystal structure is layered

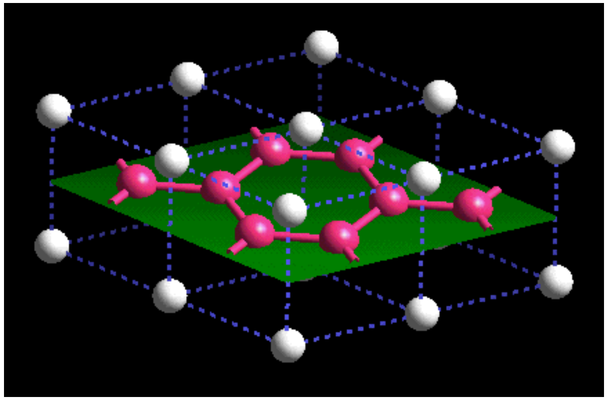


- Optic & acoustic modes separated
- Red modes frequencies dominated by e-p interaction

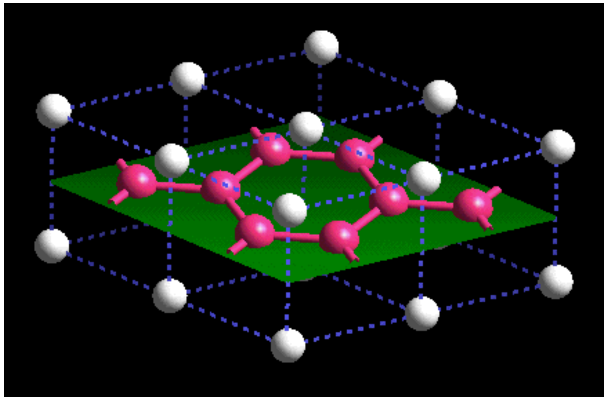
Graphics courtesy of Taner Yildirim

Motions Associated with Zone Center Modes

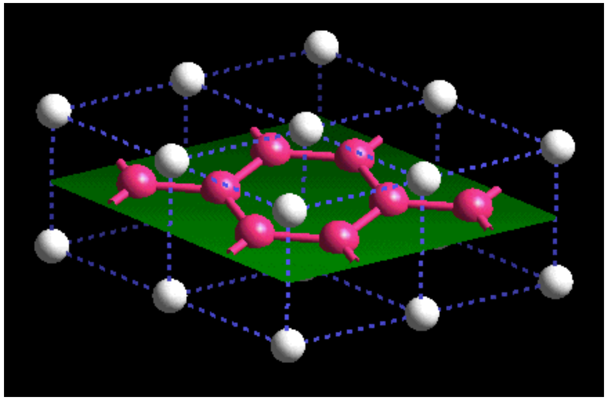
E_{1u}



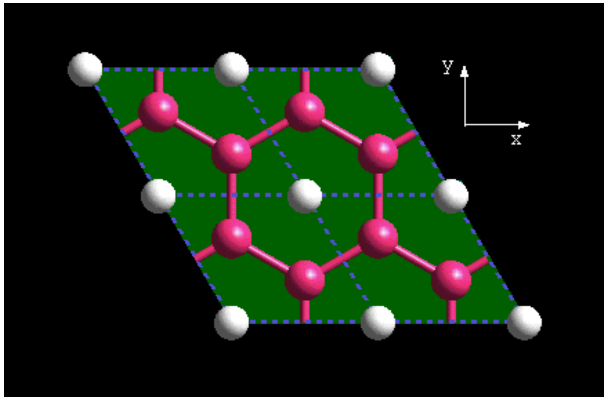
A_{2u}



B_{1g}



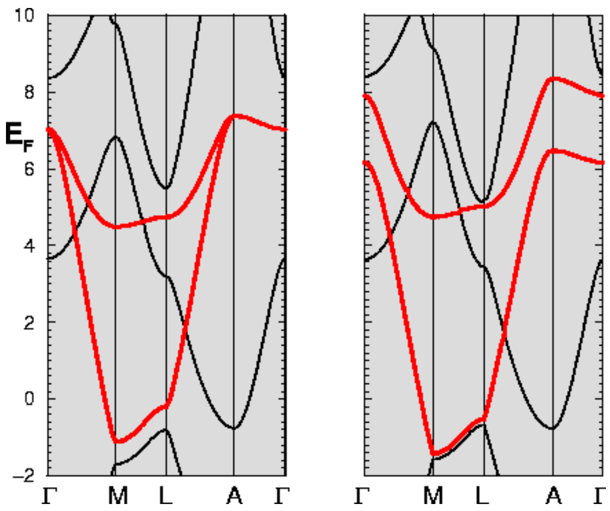
E_{2g}

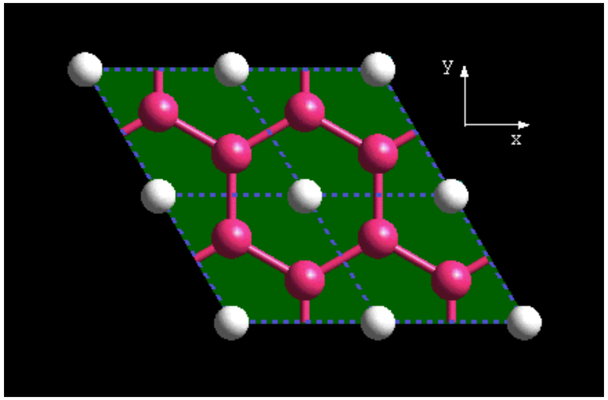


Very anharmonic

The Large Displacements Associated with E_{2g} Cause Large Electron-Phonon Coupling

- Because the effective potential for the E_{2g} mode is shallow and wide, the B atom-motions are large amplitude
- This causes significant overlap of electron shells and significant effects on the band structure close to E_F
- The strong e-p interaction causes the "high" T_c





The Inelastic Scattering Cross Section

Recall that $\left(\dfrac{d^2\sigma}{d\Omega.dE}\right)_{coh} = b_{coh}^2 \dfrac{k'}{k} NS(\vec{Q},\omega)$ and $\left(\dfrac{d^2\sigma}{d\Omega.dE}\right)_{inc} = b_{inc}^2 \dfrac{k'}{k} NS_i(\vec{Q},\omega)$

where $S(\vec{Q},\omega) = \dfrac{1}{2\pi\hbar}\iint G(\vec{r},t)e^{i(\vec{Q}.\vec{r}-\omega t)}d\vec{r}dt$ and $S_i(\vec{Q},\omega) = \dfrac{1}{2\pi\hbar}\iint G_s(\vec{r},t)e^{i(\vec{Q}.\vec{r}-\omega t)}d\vec{r}dt$

and the correlation functions that are intuitively similar to those for the elastic scattering case:

$G(\vec{r},t) = \dfrac{1}{N}\int\left\langle \rho_N(\vec{r},0)\rho_N(\vec{r}+\vec{R},t)\right\rangle d\vec{r}$ and $G_s(\vec{r},t) = \dfrac{1}{N}\sum_j\int\left\langle \delta(\vec{r}-\vec{R}_j(0))\delta(\vec{r}+\vec{R}-\vec{R}_j(t))\right\rangle d\vec{r}$

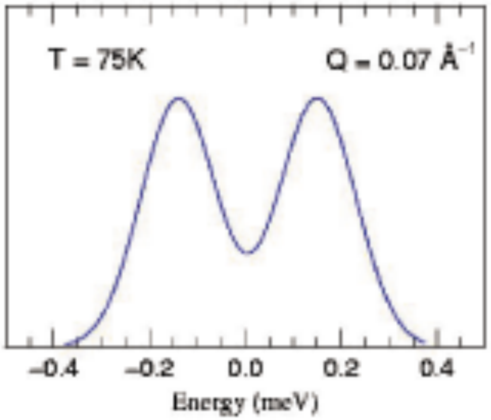
The evaluation of the correlation functions (in which the ρ's and δ - functions have to be treated as non - commuting quantum mechanical operators) is mathematically tedious. Details can be found, for example, in the books by Squires or Marshal and Lovesey.

Examples of S(Q,w) and S_s(Q,w)

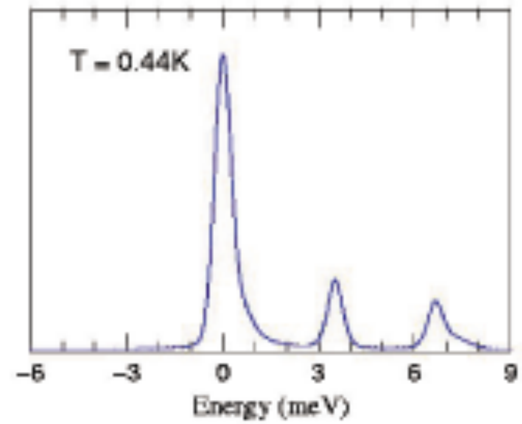
- Expressions for S(Q,w) and S_s(Q,w) can be worked out for a number of cases e.g:
  - Single phonons
  - Phonon density of states
  - Various models for atomic motions in liquids and glasses
  - Various models of atomic & molecular translational & rotational diffusion
  - Rotational tunneling of molecules
  - Transitions between crystal field levels
  - Spin waves and other magnetic excitations such as spinons

- Inelastic neutron scattering reveals details of the shapes of interaction potentials in materials

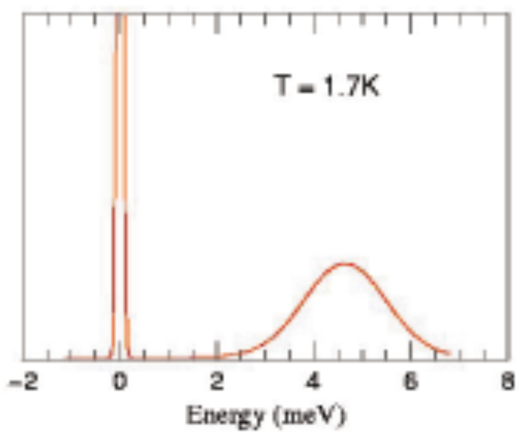
Measured Inelastic Neutron Scattering Signals in Crystalline Solids Show Both Collective & Local Fluctuations*



Spin waves – collective excitations



Crystal Field splittings ($HoPd_2Sn$) – local excitations



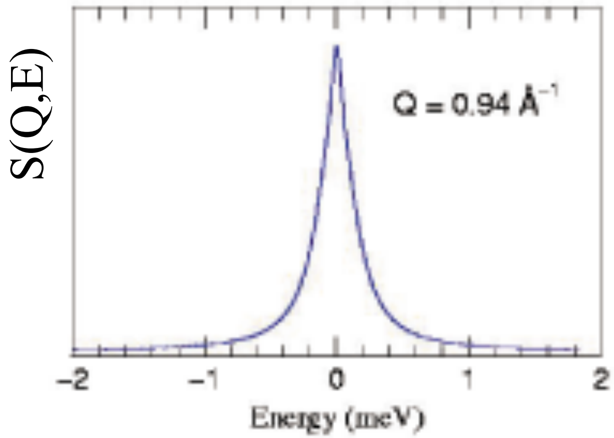
Local spin resonances (e.g. $ZnCr_2O_4$)

Local Fluctuations – energy does not depend on Q

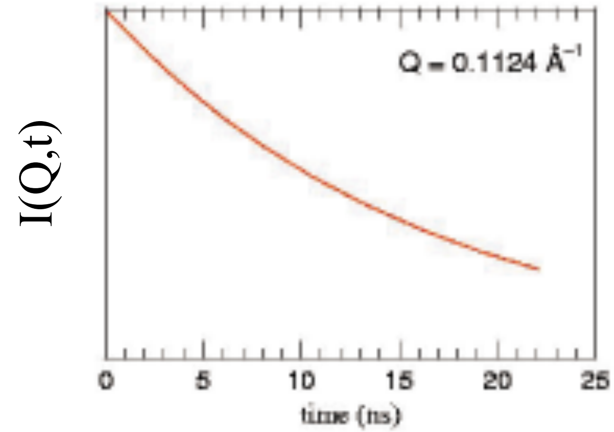
Collective fluctuations – energy is usually Q-dependent

* The following 5 viewgraphs contain material supplied by Dan Neumann, NIST

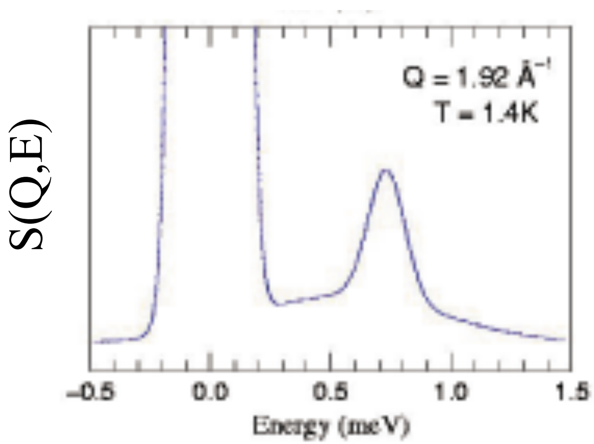
Measured Inelastic Neutron Scattering Signals in Liquids Generally Show Diffusive Behavior



"Simple" liquids (e.g. water)



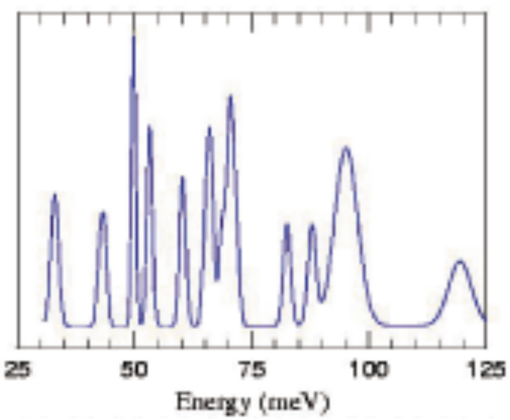
Complex Fluids (e.g. SDS)



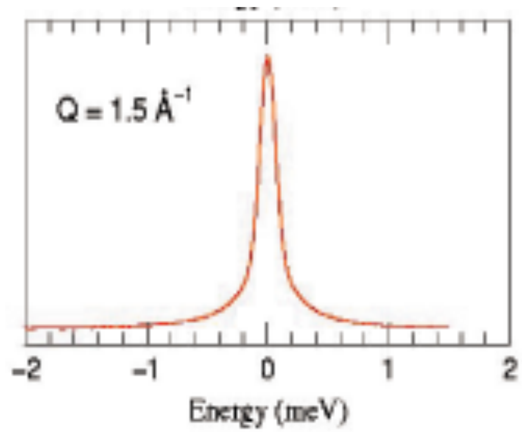
Quantum Fluids (e.g. He in porous silica)

Diffusive motion is usually measured using the incoherent neutron scattering cross section and is manifested by a spectral peak centered at E = 0 – so-called quasielastic scattering.

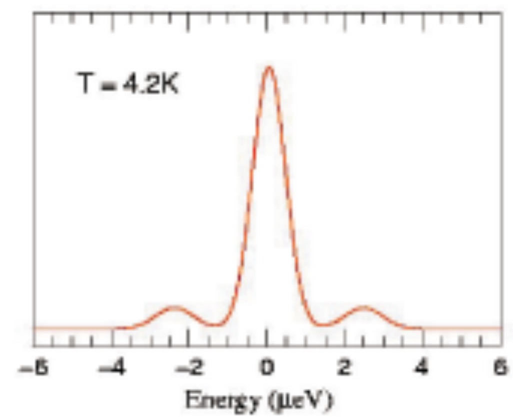
Measured Inelastic Neutron Scattering in Molecular Systems Span Large Ranges of Energy



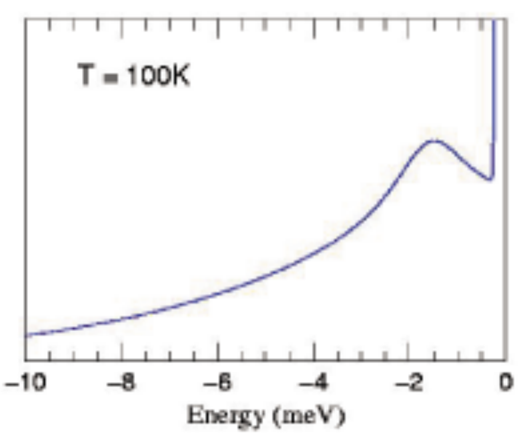
Vibrational spectroscopy (e.g. C_{60})



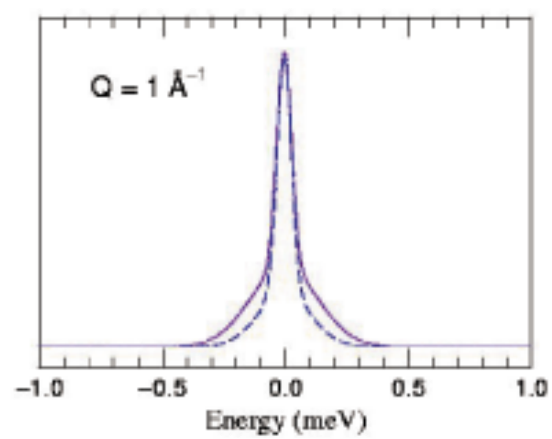
Molecular reorientation (e.g. pyrazine)



Rotational tunneling (e.g. CH_3I)



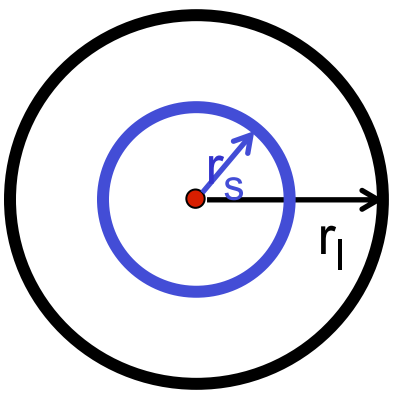
Polymers

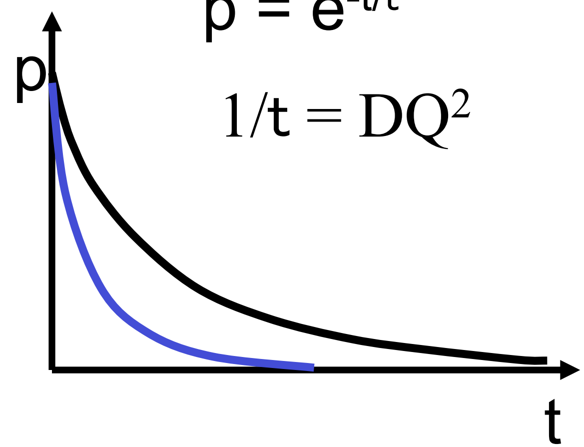


Proteins

Quasielastic Neutron Scattering

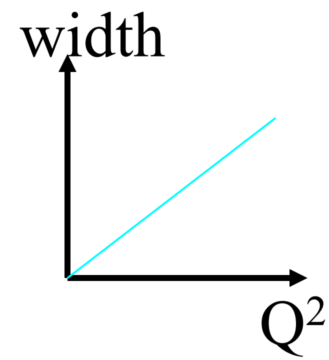
- For a single diffusing particle, the probability, p, of finding it within a sphere around its starting position looks like….





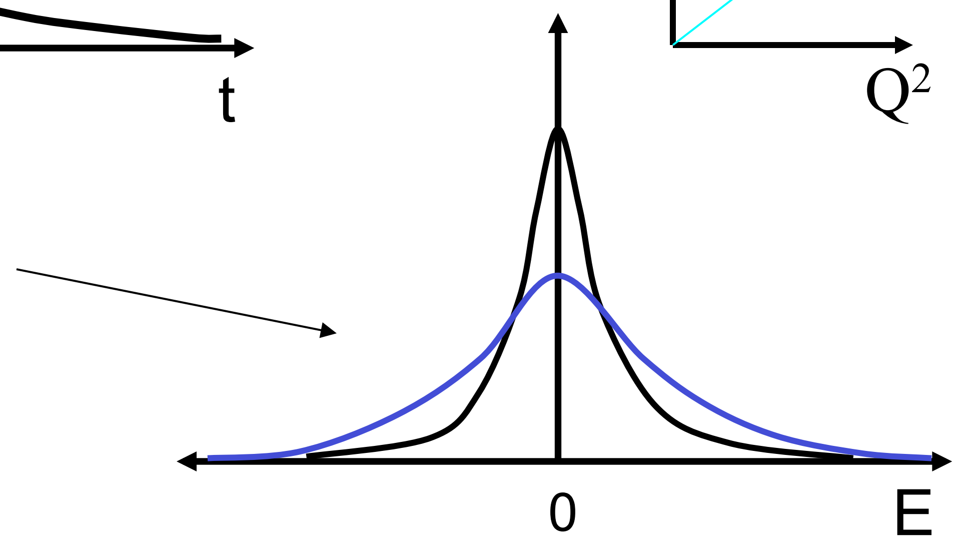
$p = e^{-t/t}$

$1/t = DQ^2$



- $S_{inc}(Q,E)$ is the time Fourier transform of this probability

$$S_{inc}(Q,E) = \frac{\hbar}{\pi} \frac{DQ^2}{(\hbar DQ^2)^2 + E^2}$$



Quasielastic Neutron Scattering

- If there is a finite probability that a particle occupies its initial position as t -> ∞ the scattering will include an elastic component

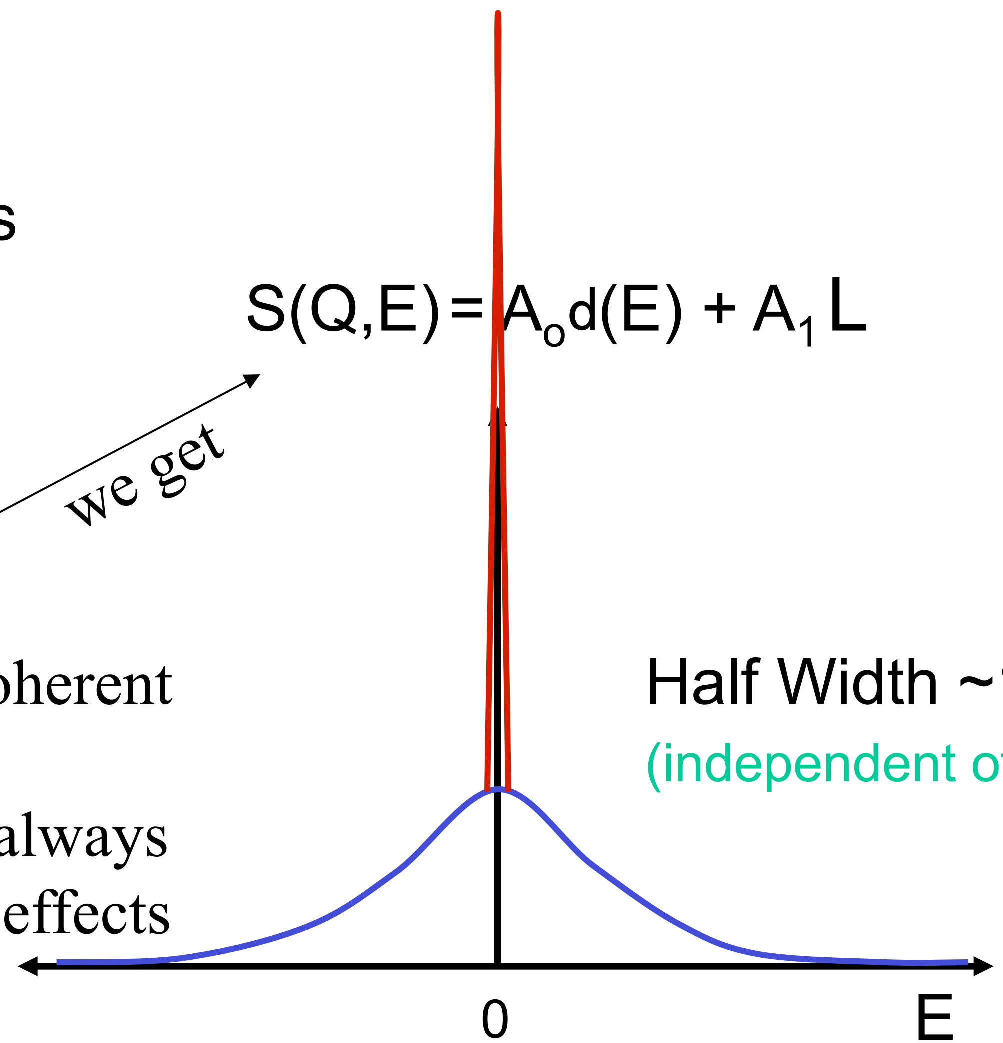
- For example, if two sites may be occupied and

$$p_1 = ½ + ½ e^{-2t/t}$$

$$p_2 = ½ - ½ e^{-2t/t}$$

we get

$$S(Q,E) = A_o d(E) + A_1 L$$

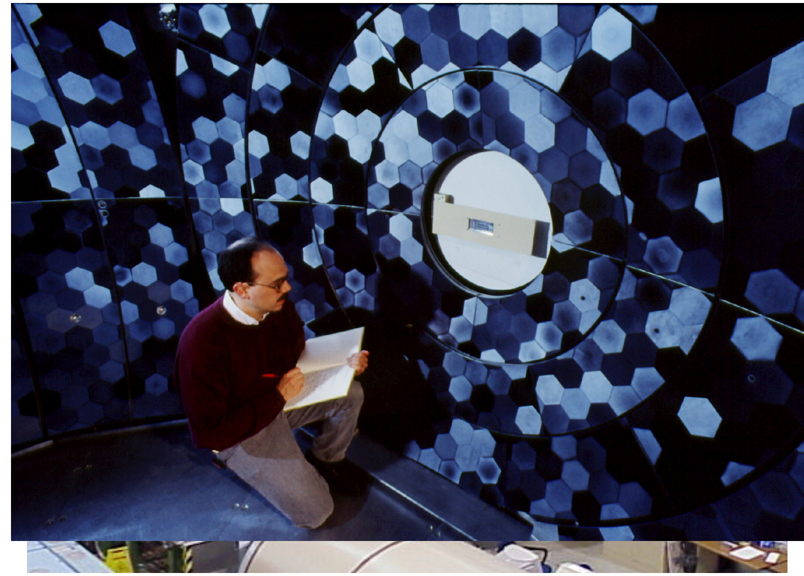


A_0 is called the Elastic Incoherent Structure Factor (EISF)

Note that the d-function is always broadened by instrumental effects

Spectrometers for Measuring Quasielastic Scattering

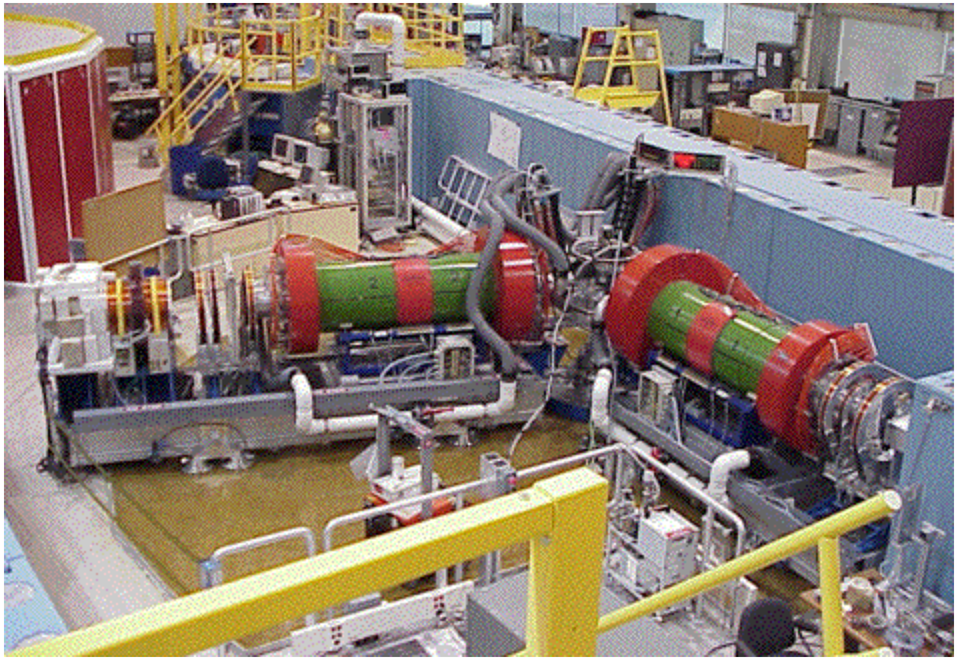
Chopper spectrometer with pulsed mono-chromatic incident neutron beam and time-of-flight energy analysis
0.01 < DE < 0.1 meV for cold neutrons
1 < DE < 10 meV for thermal neutrons



Backscattering spectrometer with poly-chromatic incident beam and energy analysis by crystal analyzer
0.001 < DE < 0.1 meV for cold neutrons

Note (1) that the value of the energy resolution, DE, sets the minimum observed width of spectral line and (2) that the good energy resolution of backscattering is obtained at the expense of poor Q resolution

Another Way to Measure Quasielastic Scattering: Neutron Spin Echo



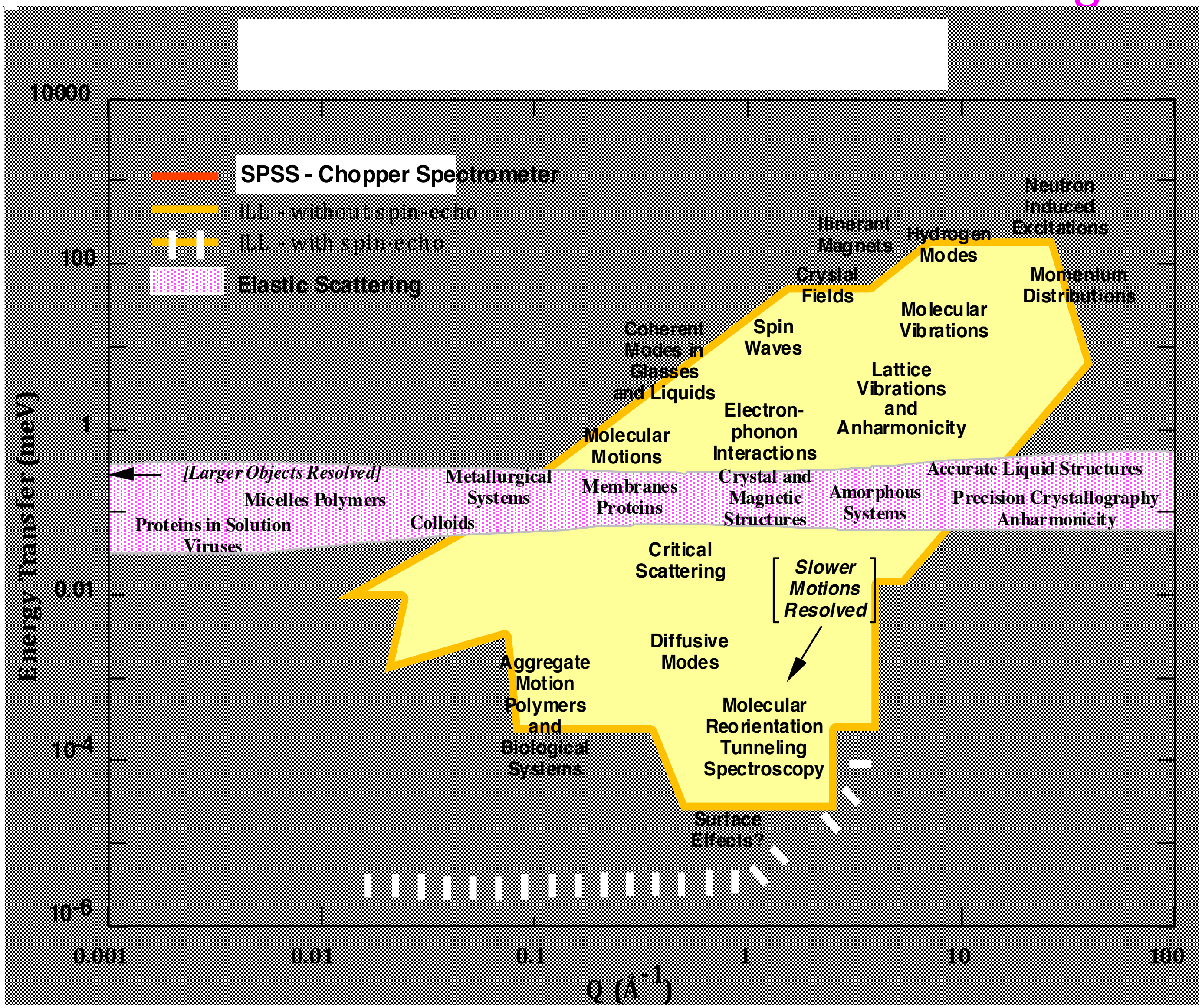
NSE measures the energy Fourier transform of S(Q,E)

It is easier to measure coherent Scattering with NSE

$0.00001 < \Delta E < 0.001$ meV
i.e. times between ns and ms

- NSE works by using the precession of a neutron's magnetic moment (spin) in a field as a "clock" to measure the neutron's speed.
- The neutron spins undergo many ($\sim 10^5$ turns) in the green solenoid magnets above.
- In effect the spins are "wound up" in the first field and "unwound" by the same amount in the second field if the scattering by the sample (between the solenoids) is elastic.
- If the scattering is inelastic, the exact unwinding (or echo) is suppressed and the polarization of the neutron beam at the echo position is a measure of the inelasticity of the scattering.

Much of the Scientific Impact of Neutron Scattering Has Involved the Measurement of Inelastic Scattering



Energy & Wavevector Transfers accessible to Neutron Scattering