Neutron Compton Scattering

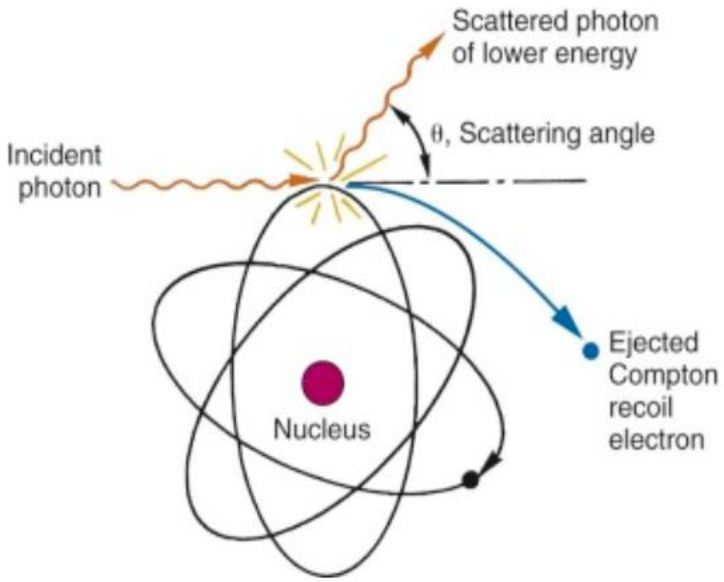
Roberto Senesi

Università degli Studi di Roma “Tor Vergata”, Dipartimento di Fisica and Centro NAST

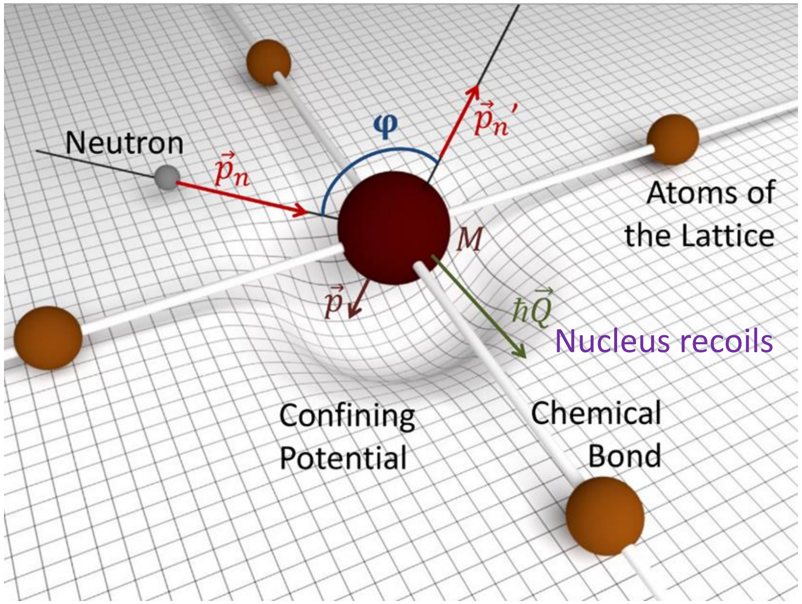
CNR- IPCF Sezione di Messina

Centro Fermi

Neutron Compton Scattering (also known as) Deep Inelastic Neutron Scattering



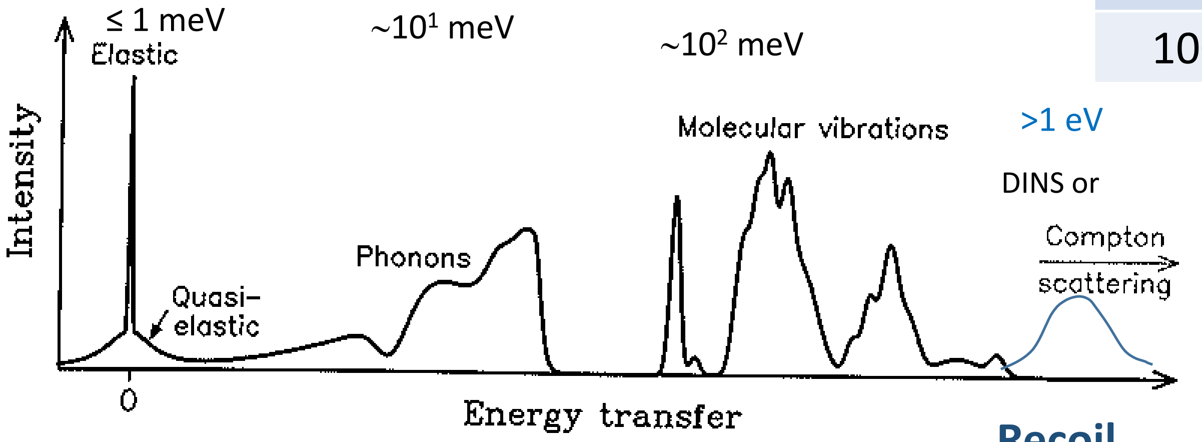
Compton Scattering (photon)



Neutron Compton Scattering

Neutron Compton Scattering
also known as
Deep Inelastic Neutron Scattering

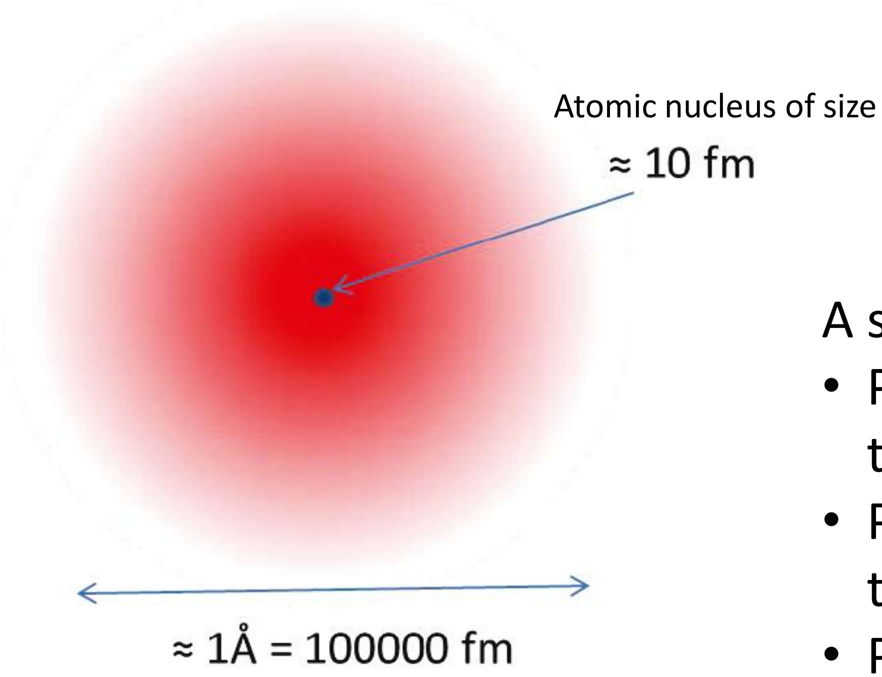
Energy [eV]	Wave length [Å]
0.4	0.45
1	0.29
10	0.09
20	0.06
50	0.04
100	0.03



Recoil excitations

Adapted from: "Elementary Scattering Theory For X-ray and Neutron Users" D.S. Sivia OUP (2011)

Neutron Compton Scattering



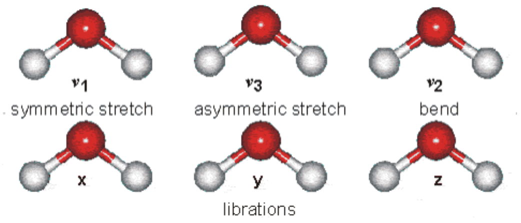
Electron cloud in an atom

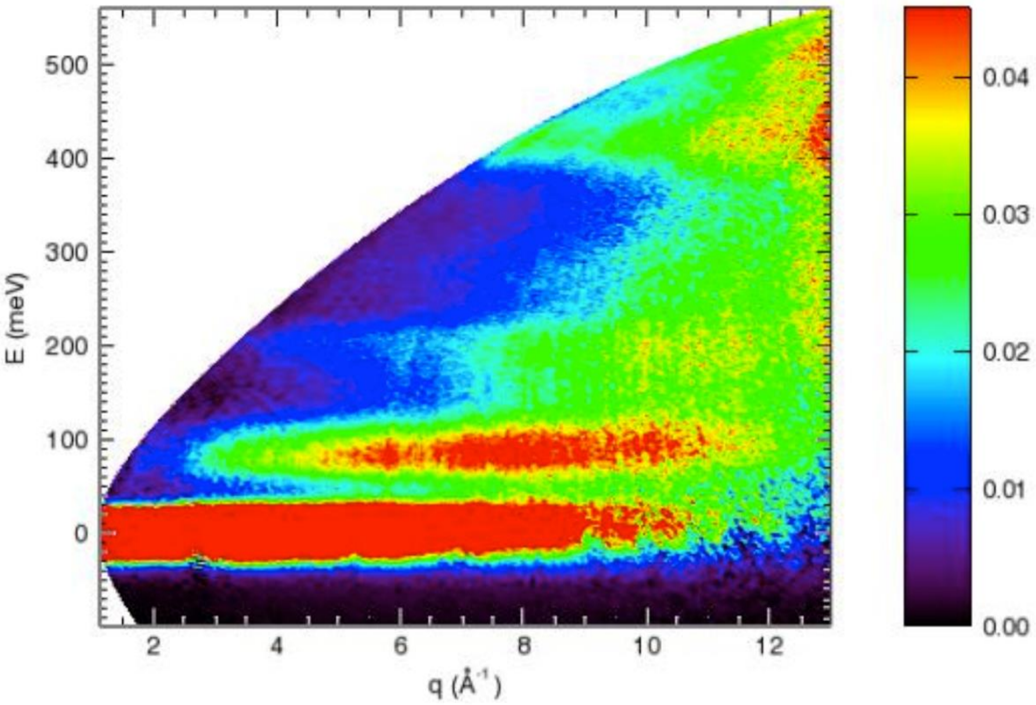
Energy [eV]	Wave length [Å]
0.4	0.45
1	0.29
10	0.09
20	0.06
50	0.04
100	0.03

A scattering regime

- Probing **small length scales**: smaller than atom-atom distances
- Probing **large energy transfers**: larger than largest atomic vibration energies
- Probing **large wave vector transfers**: larger than Brillouin zone wave vectors

How to reach Neutron Compton Scattering regime- measurement on ice polycrystal sample:





Excitation of stretching

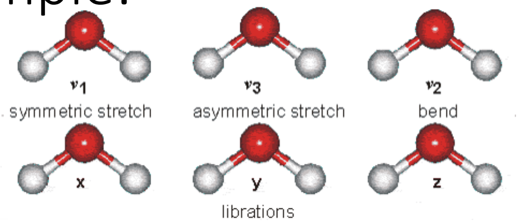
Excitation of bending

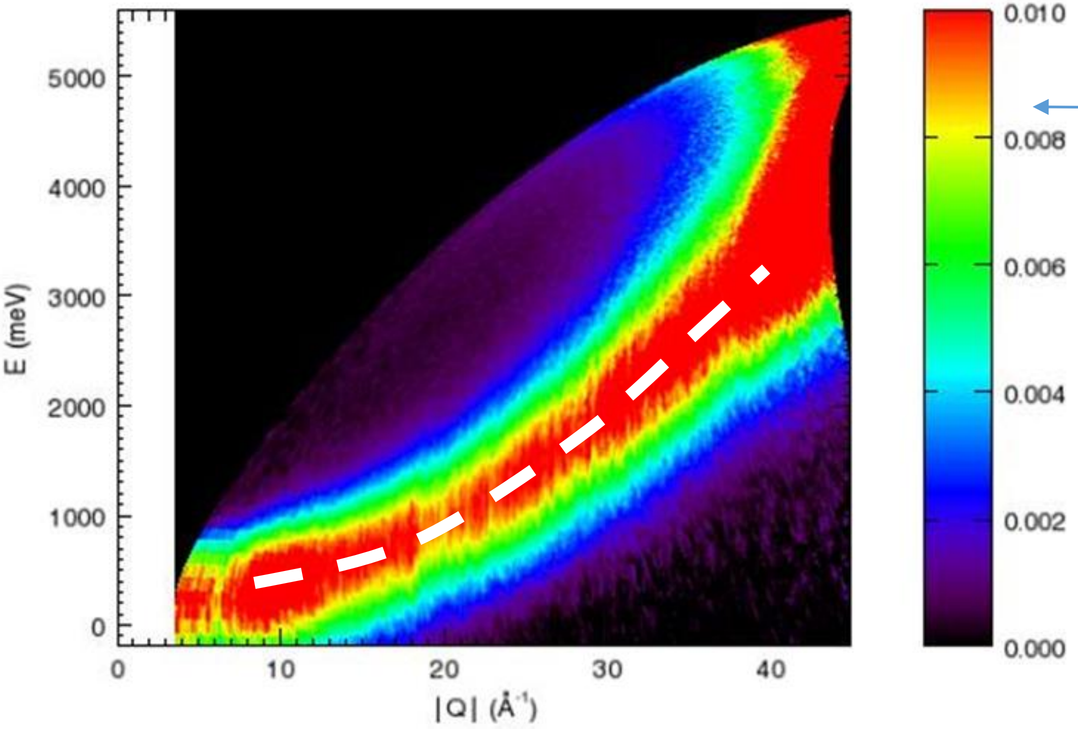
Excitation of libration /rotation

Elastic

INELASTIC NEUTRON SCATTERING MEASUREMENT

How to reach Neutron Compton Scattering regime- measurement on ice polycrystal sample:





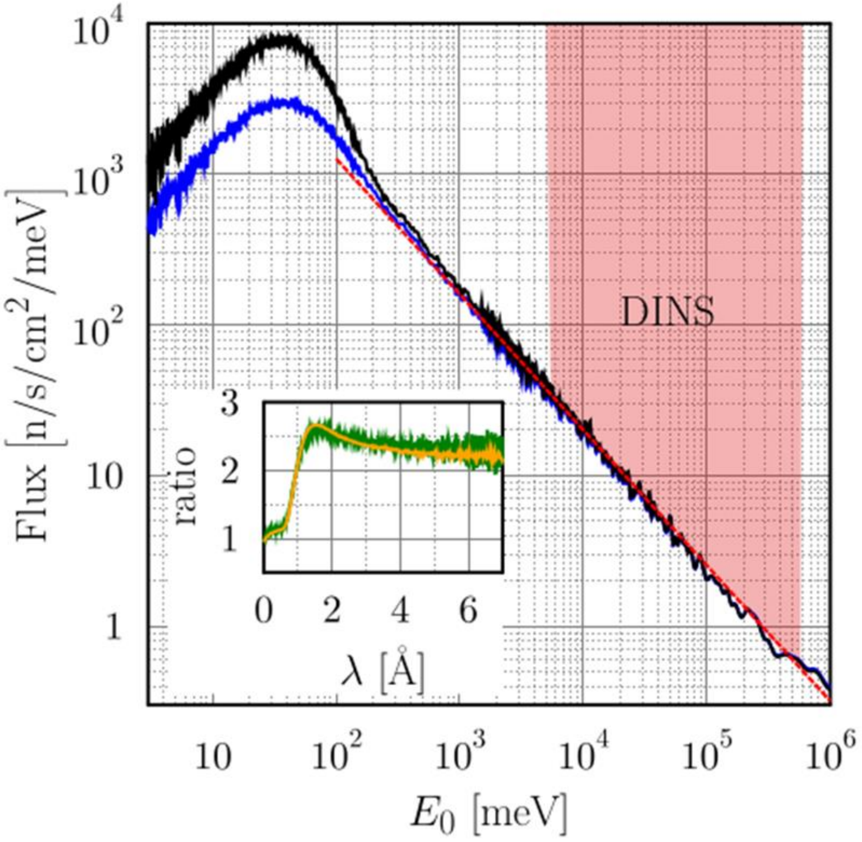
Only one excitation is measured: recoil of the nucleus struck by the neutron

$$\hbar\omega = E = \frac{\hbar^2 Q^2}{2M} \equiv \hbar\omega_r$$

Scattering intensity centred on the recoil line

NEUTRON COMPTON SCATTERING MEASUREMENT

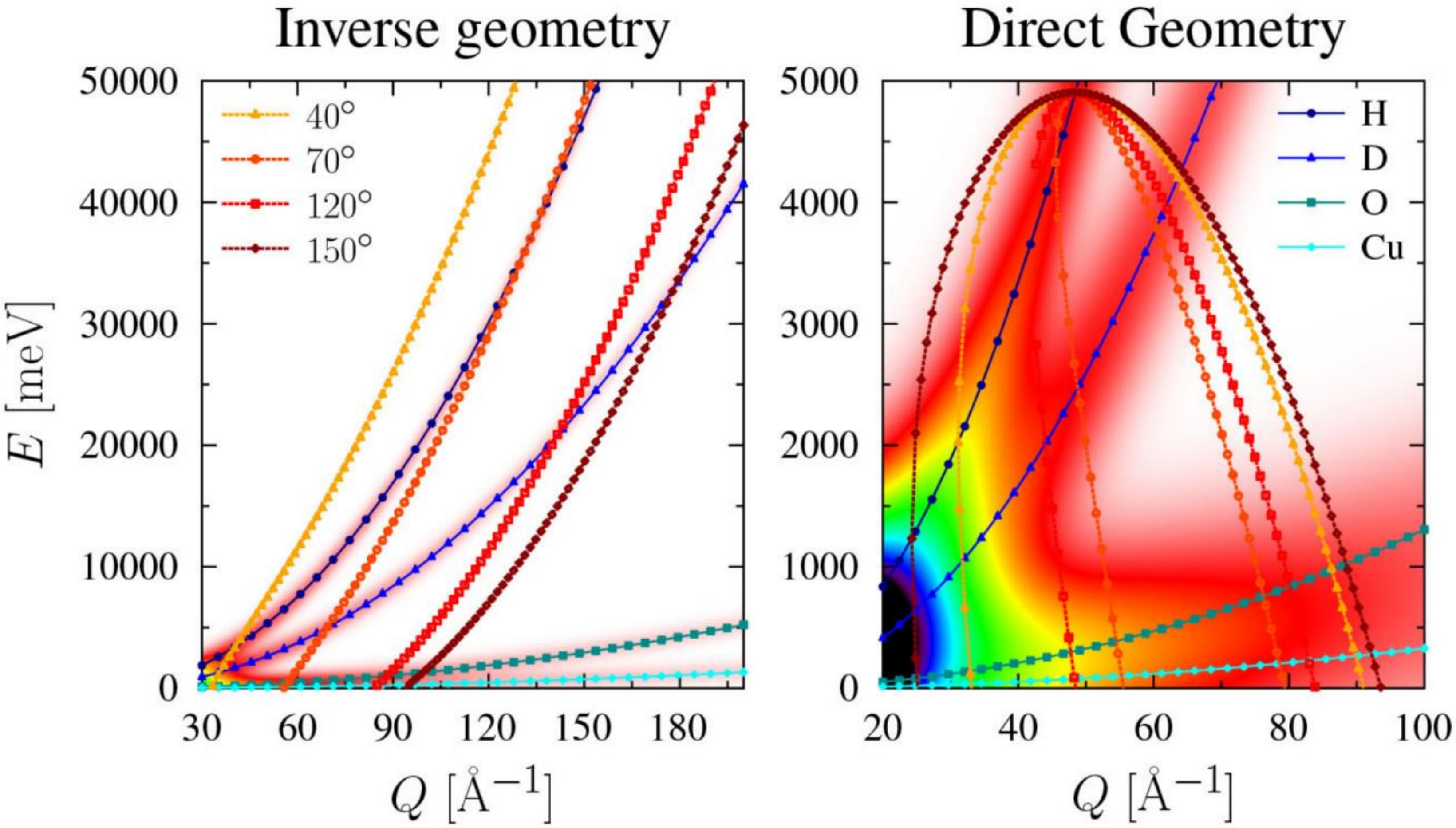
Where to reach Neutron Compton Scattering regime:
Mainly VESUVIO at ISIS, MARI, MAPS, SEQUOIA,…



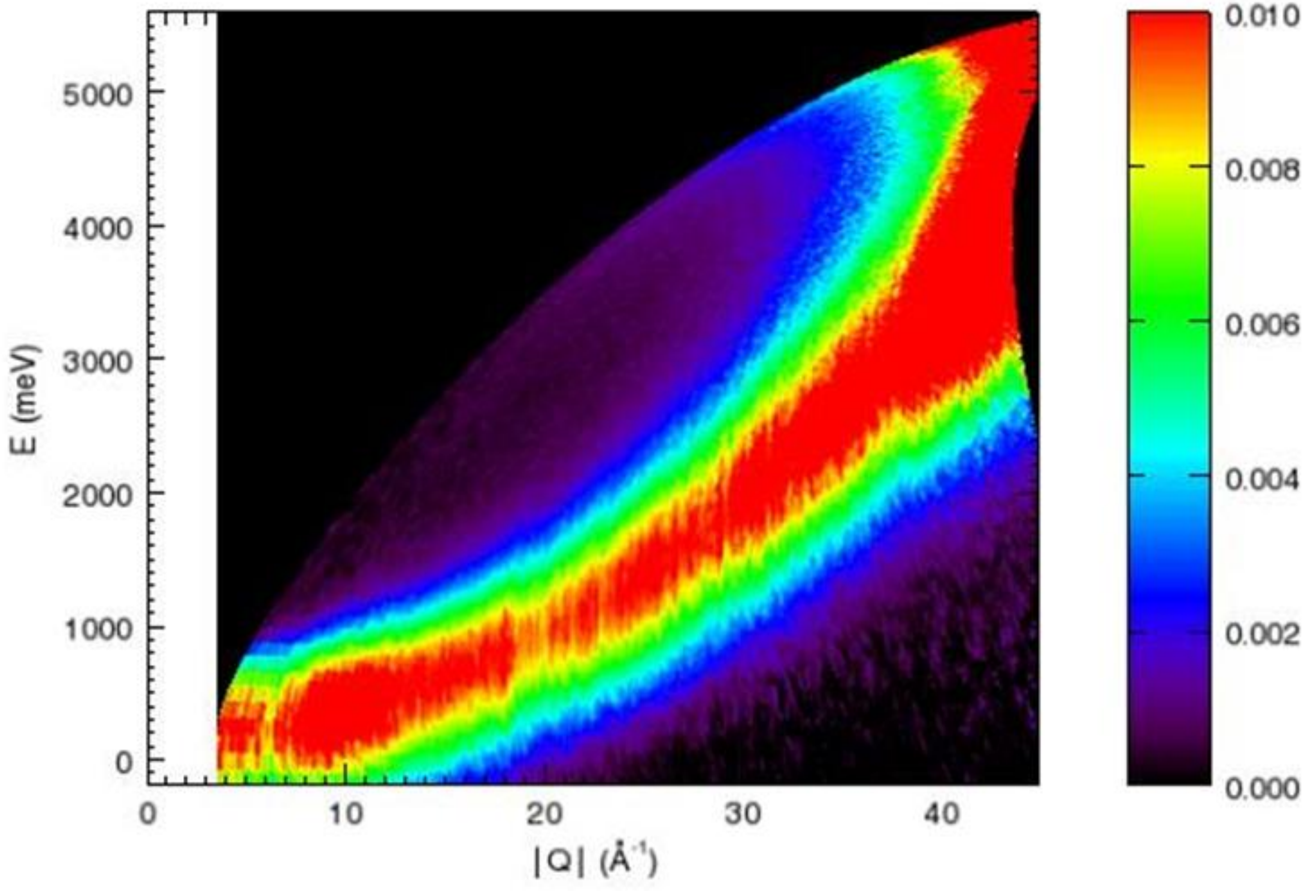
eV spectrometers

At pulsed sources, by selecting the initial (final) energy of the neutron with a resonant foil, one can evaluate the final (initial) energy using the time-of-flight technique, thereby obtaining the energy transfer.

C Andreani, M Krzystyniak, G Romanelli, R Senesi, and F Fernandez-Alonso; Advances in Physics, (2017)

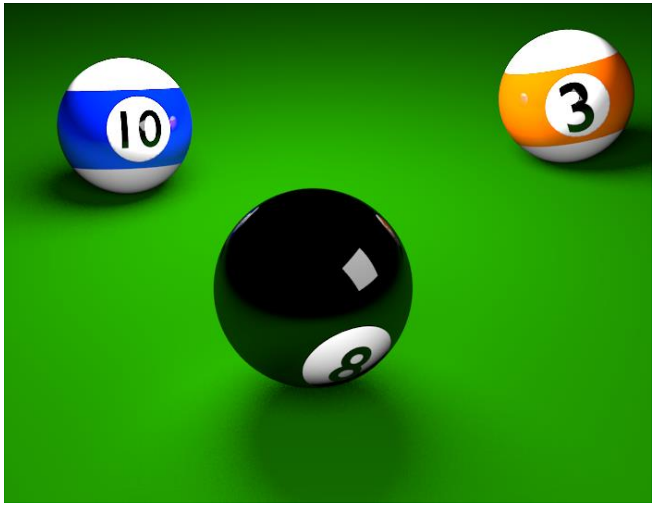


The scattering intensity (centred at the recoil) is a line with a shape! (and not just a narrow line…)

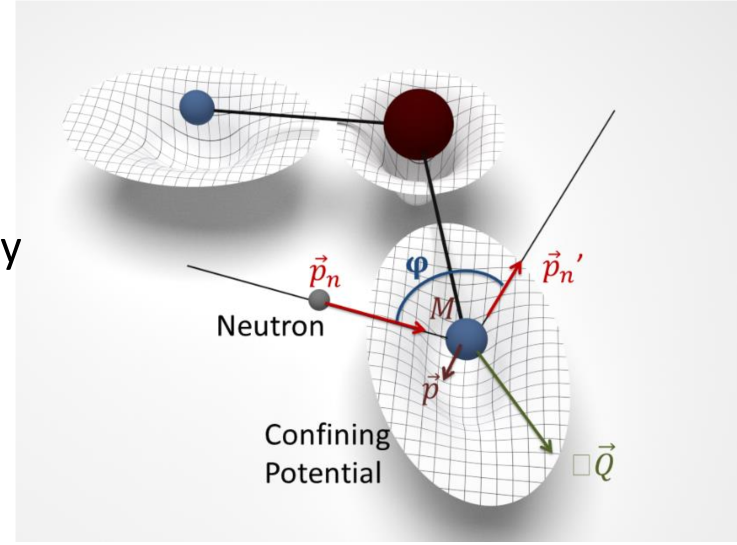


$$\hbar\omega = E = \frac{\hbar^2 Q^2}{2M} \equiv \hbar\omega_r$$

Neutron Compton Scattering is described within the Impulse Approximation



The atom struck by the neutron is not at rest



$$\mathbf{p_n} - \mathbf{p'_n} = \hbar\mathbf{q}$$

Change of neutron momentum in the scattering process

$$\hbar\omega = \frac{(\mathbf{p_n} - \mathbf{p'_n})^2}{2M} + \frac{(\mathbf{p_n} - \mathbf{p'_n})\cdot\mathbf{p}}{M}$$

Change of neutron energy in the scattering process

Only within the Impulse Approximation!

Neutron Compton Scattering is described within the Impulse Approximation

We (experimenters) are able to detect neutrons, their wave vector and energy transfer using the detection systems of the instruments

$$\hbar\omega = \frac{(\mathbf{p_n} - \mathbf{p_n'})^2}{2M} + \frac{(\mathbf{p_n} - \mathbf{p_n'})\cdot\mathbf{p}}{M}$$

Example:

A sample containing 10^21 atoms
Counted for 10 hours on VESUVIO at ISIS (10^7 n/cm^2/s)

- The experiment will measure about 10^6 momenta, $\mathbf{p}$, of individual atoms in the sample.
- That is, a measurement (sampling) of the MOMENTUM DISTRIBUTION

$$\mathrm{n}(\mathbf{p})$$

Derivation of the Impulse Approximation (details available as appendix)

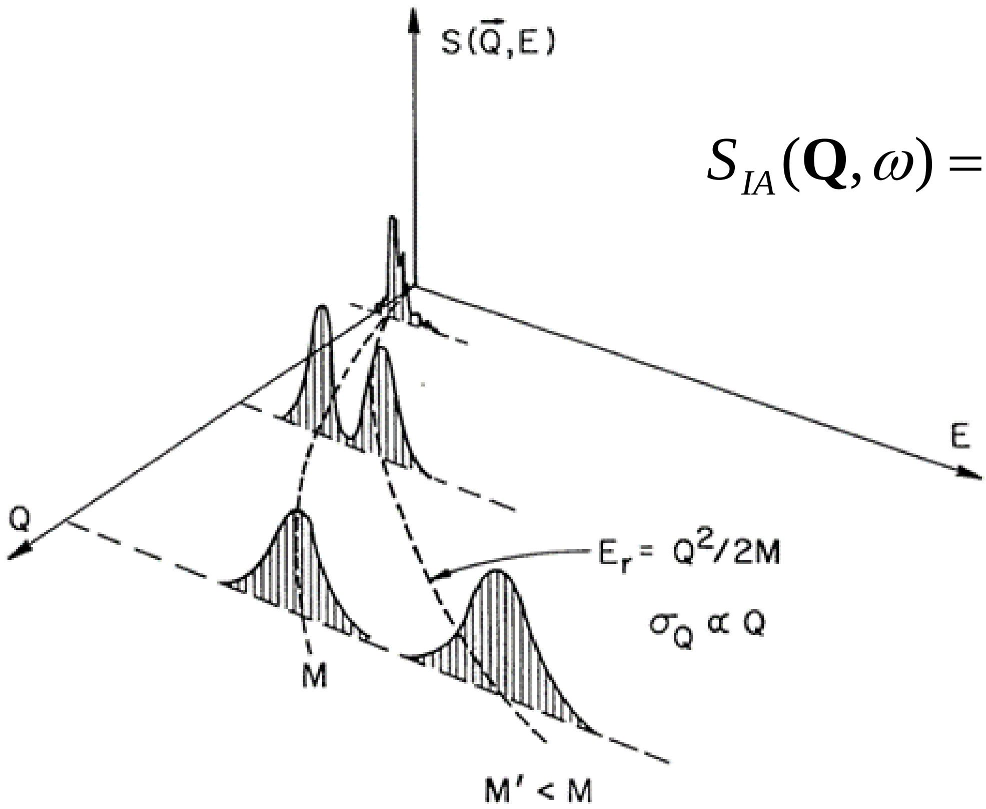
Focus on the short time behaviour of the correlation function $\langle e^{-i\mathbf{Q}\cdot\mathbf{R}_j} e^{i\mathbf{Q}\cdot\mathbf{R}_{j'}(t)} \rangle$

Apply the approximation

$$\mathbf{R}_{j'}(t) = \mathbf{R}_{j'}(0) + \frac{t}{M_{j'}}\mathbf{p}_{j'} + \ldots.$$

RESULT:

$$S_{IA}(\mathbf{Q},\omega) = \hbar \int d\mathbf{p}\; n(\mathbf{p})\; \delta\left(\hbar\omega - \frac{\hbar^2 Q^2}{2M} - \frac{\mathbf{p}\cdot\hbar\mathbf{Q}}{M} \right)$$



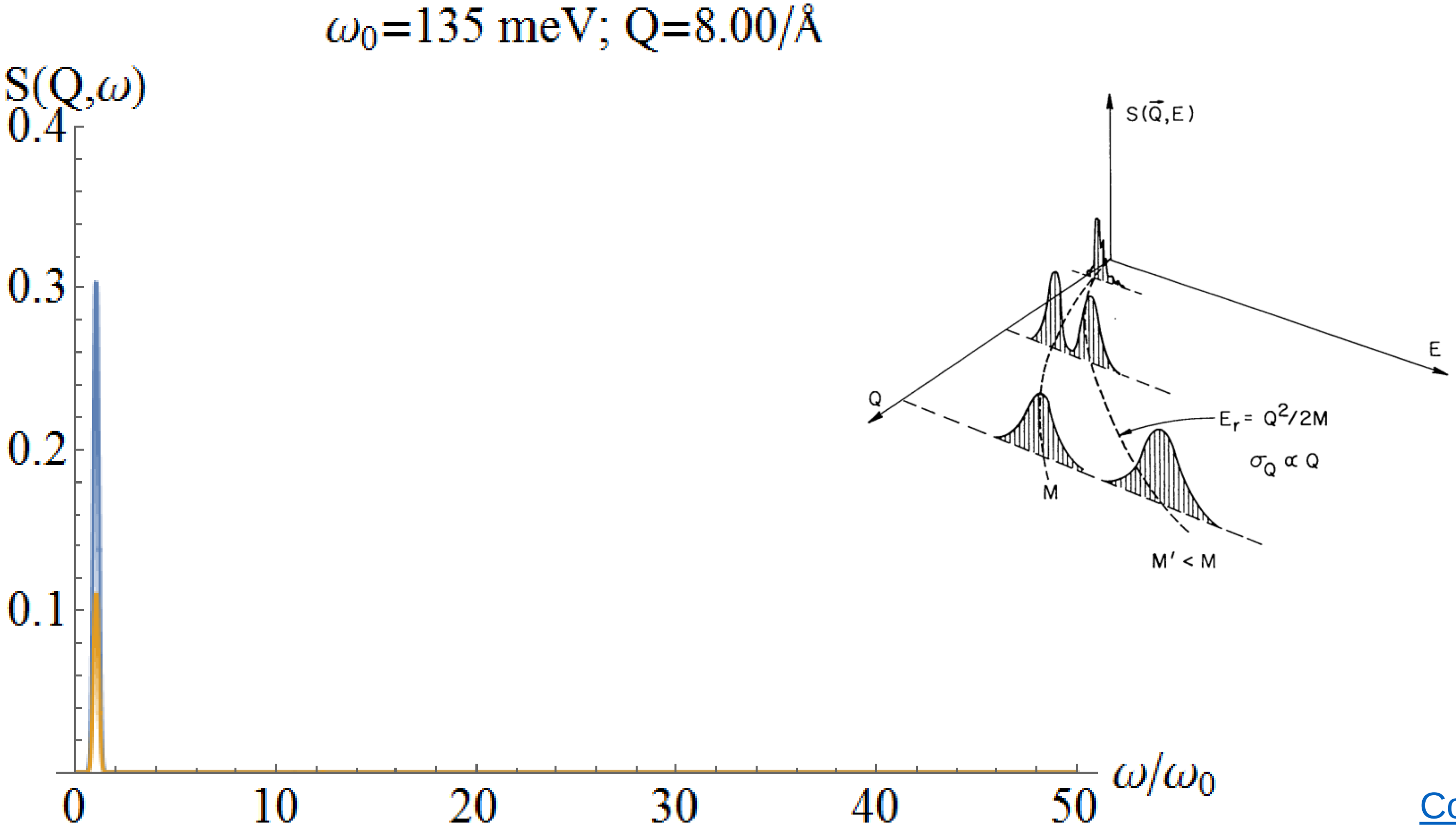
For each mass M in the sample the scattering is centred at a specific recoil peak whose shape is proportional to the shape of momentum distribution

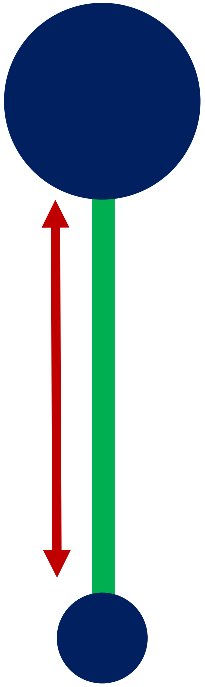
The link to vibrational spectra

(Calculations and graphics courtesy of G. Romanelli)

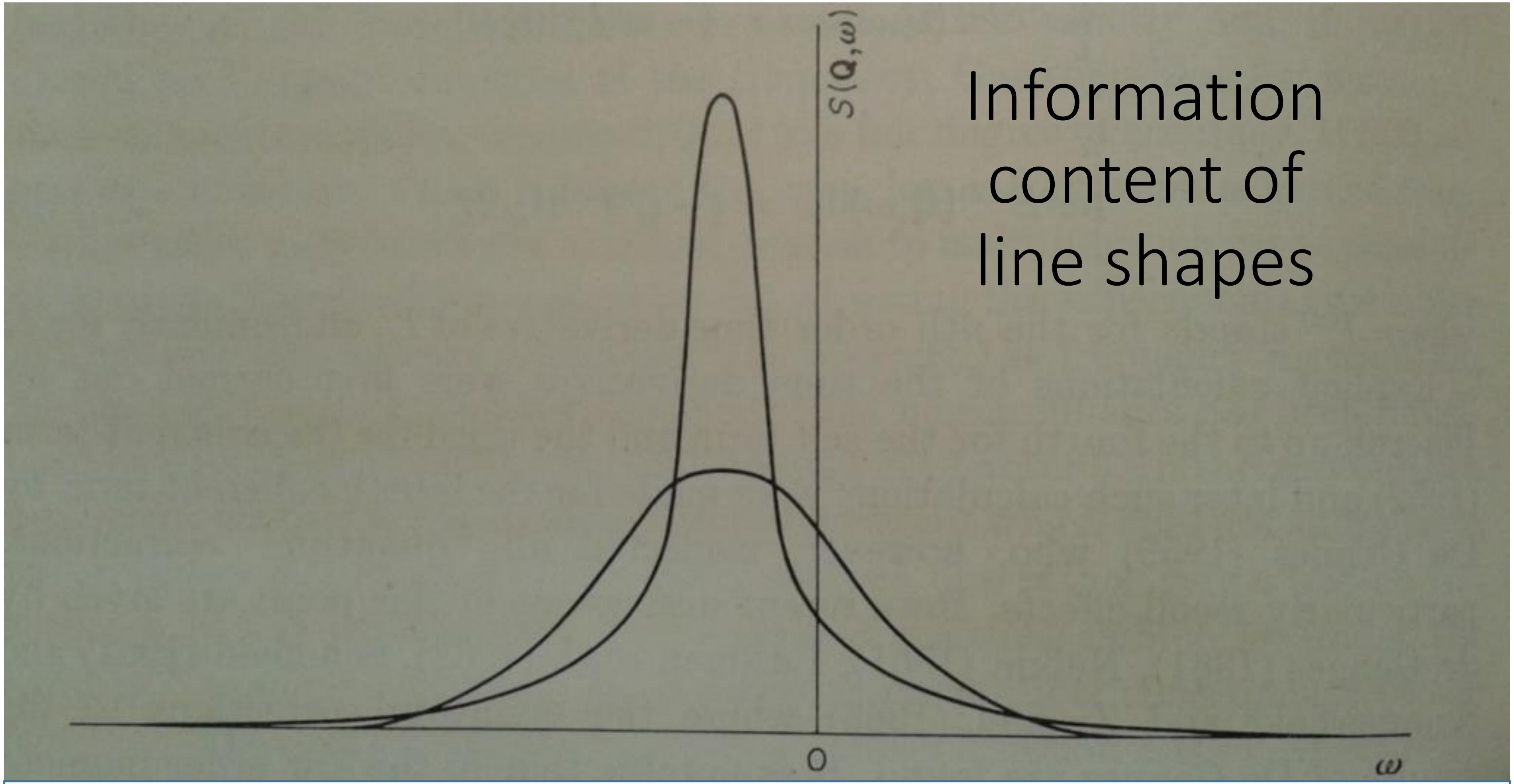
In the approximation of a **harmonic potential**, it is possible to relate the vibrational density of states to the width of the nuclear momentum distribution

The increase of energy and wave vector transfers requires a multi-phonon expansion





GI Watson; J Phys Cond Matt 8 33 (1996)



Information content of line shapes

Are the two curves different or similar?

Information content of $\mathbf{n(p)}$

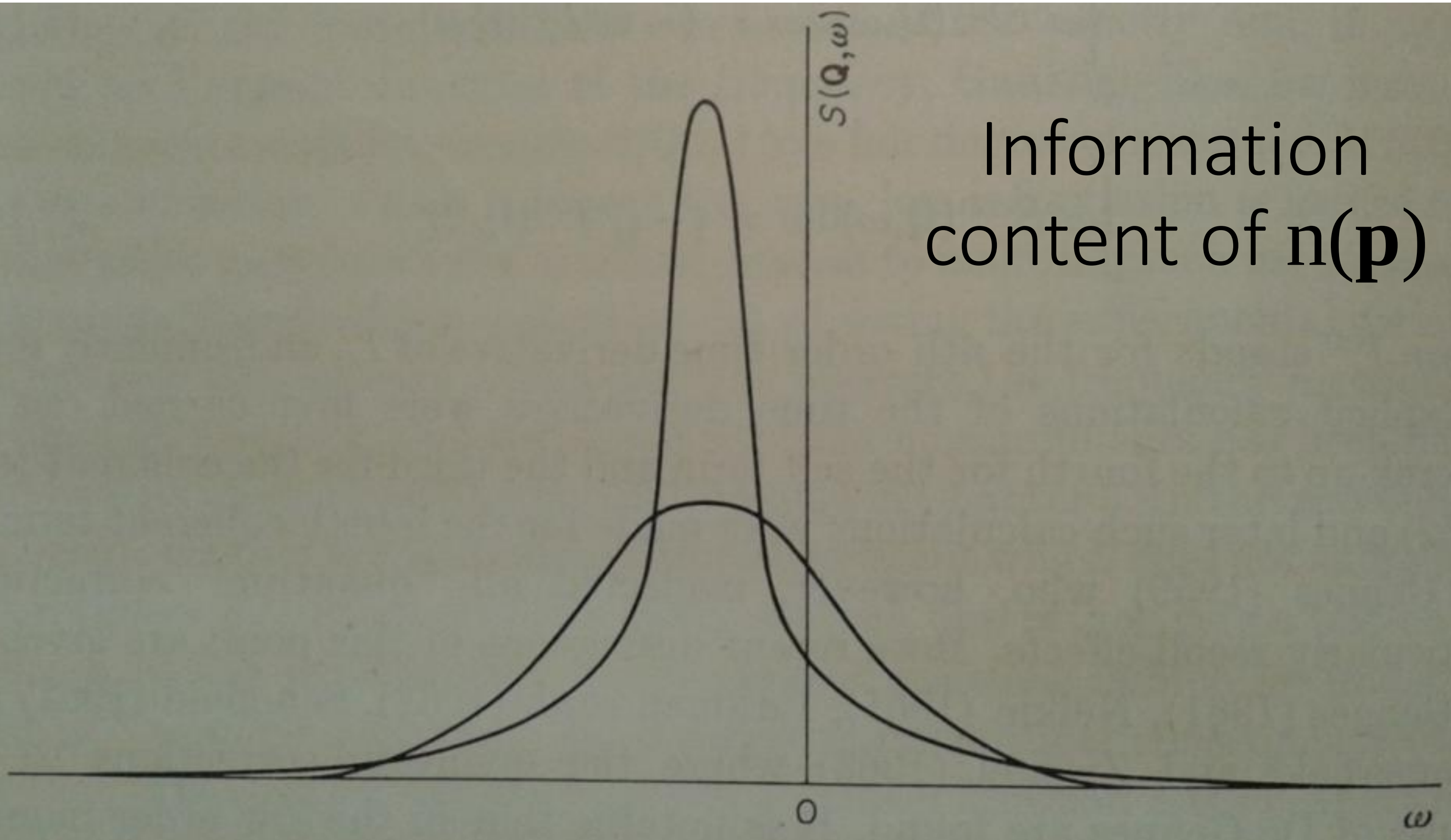


FIG. 7.1. Two different curves are given which have the same zero, first and second moments.

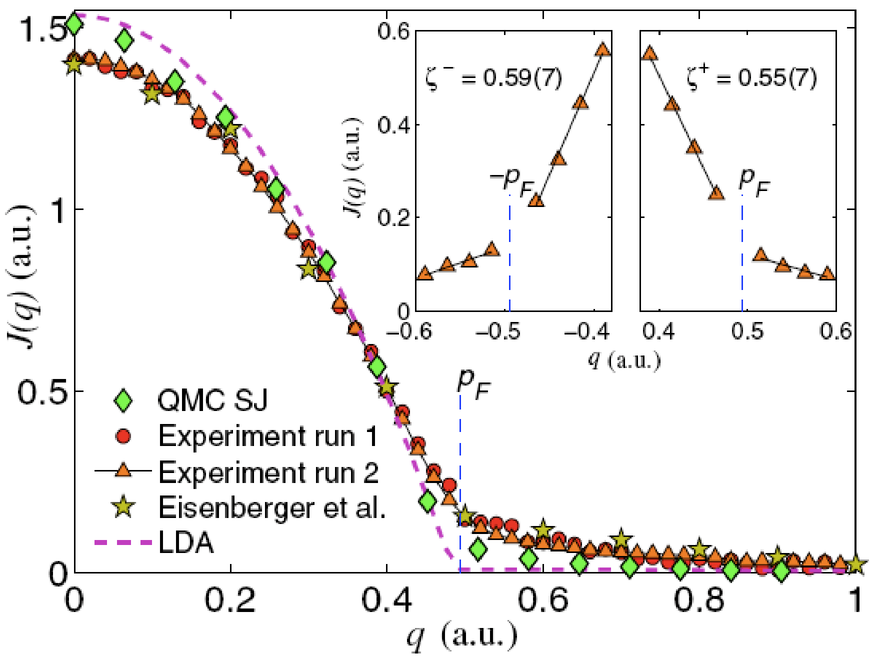
$$\langle E_K \rangle = \frac{\langle p^2 \rangle}{2M} = \int d\ \mathbf{p}\ p^2\ n(\mathbf{p})$$

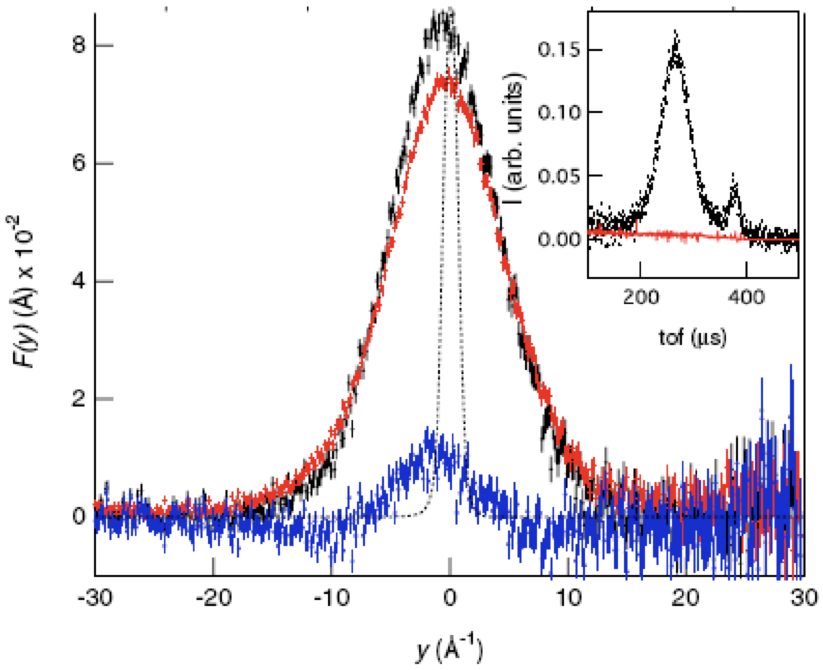
Atomic mean kinetic energy!
Dominated by quantum effects
of ZERO POINT MOTION

How do we measure momentum distributions and kinetic energies of..?

Electrons, using ID16 at ESRF

Protons, using VESUVIO at ISIS





Compton profile of Na; p- resolution≈ 0.04 a.u. ≈ 13% FWHM

S. Huotari et al, PRL 105, 086403 (2010)

$$q = \mathbf{p}\cdot\hat{Q}$$

Neutron Compton profile of water; p-resolution≈14%

A. Pietropaolo et al, PRL 100, 127802 (2008)

$$y = \frac{\mathbf{p}}{\hbar}\cdot\hat{Q}$$

TITI LVCRETI CARI DE RERVM NATVRA LIBER SECVNDVS

….Quod quoniam constat, ni mirum nulla quies **est** reddita corporibus primis…

….As it is well assessed, no rest is given to prime bodies (*atoms*) …

Restless motion which persists to absolute zero..

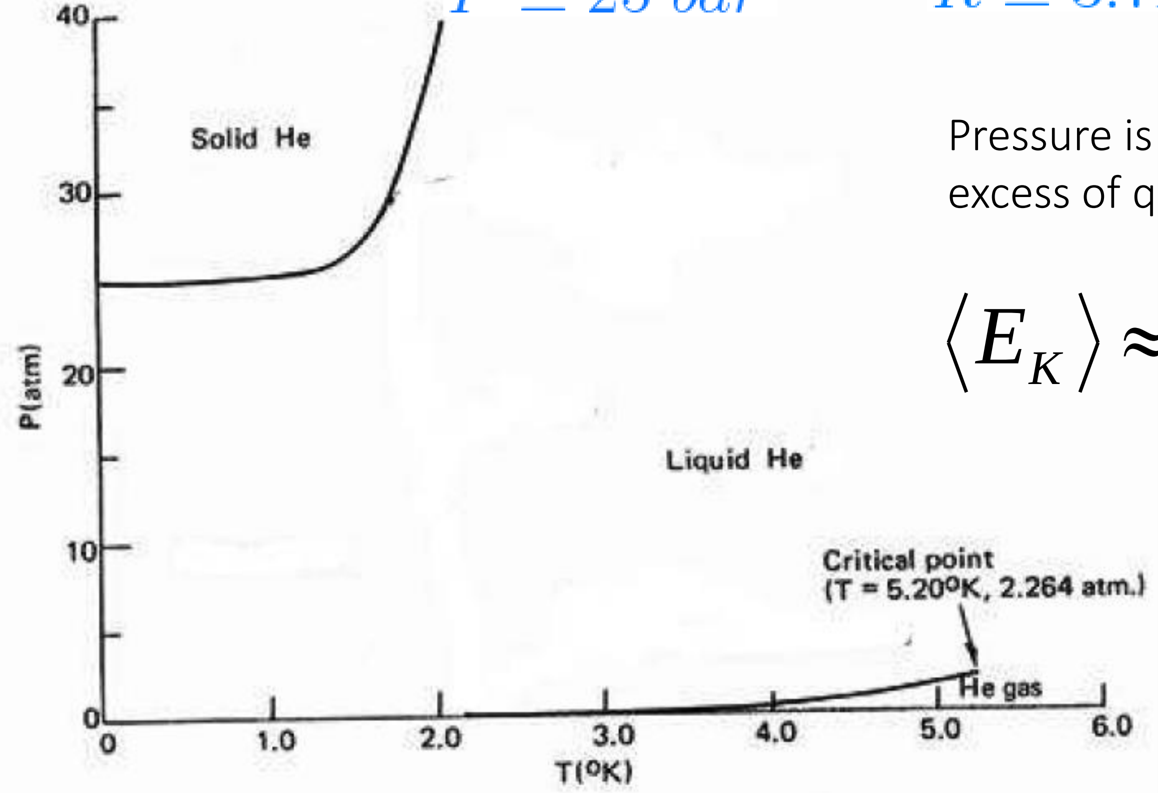
QUESTION: why solid helium does not exist by simply lowering the temperature?

Solid Helium at T= 0 K?

$$G(P,R) = \frac{\hbar^2}{2M_{He}(R-\sigma)^2} + \frac{1}{2}zv(R) + PR^3$$

-Gibbs free energy, as a funtion of interatomic distance R, and pressure P has a minimum at

$$P \simeq 25\ bar \qquad R \simeq 3.7\text{Å}$$



phase diagram of He

Pressure is needed to overcome the excess of quantum kinetic energy

$$\langle E_K \rangle \approx \frac{\hbar^2}{2M_{He}(R-\sigma)^2}$$

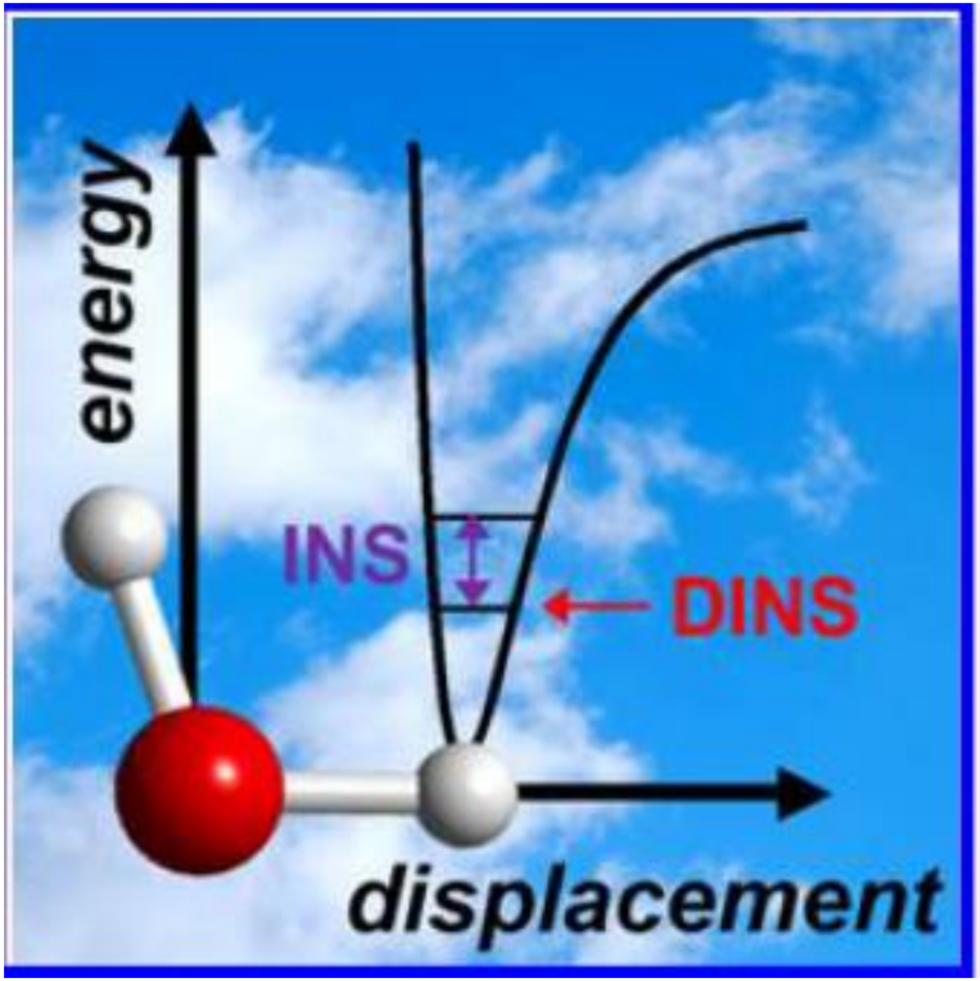
Relevance of measurements of momentum distributions and kinetic energies

- The quantum liquids and solids: helium (3 and 4), hydrogen (and deuterium), neon, lithium,…
- Protons and oxygen in water/ice: competing quantum effects on melting, metastable phases (supercooled , amorphous ice, - *do not drink heavy water, please…*)
- Protons on the surface of proteins: tunnelling or not?
- Water protons around DNA: balancing changes in enthalpy of hydration?
- Changes in kinetic energies during the setting of cements
- Atoms and molecules constrained in nm boundaries, protons in double well potentials…

- C Andreani, M Krzystyniak, G Romanelli, R Senesi, and F Fernandez-Alonso; "Electron-volt neutron spectroscopy: beyond fundamental systems", Advances in Physics, (2017)

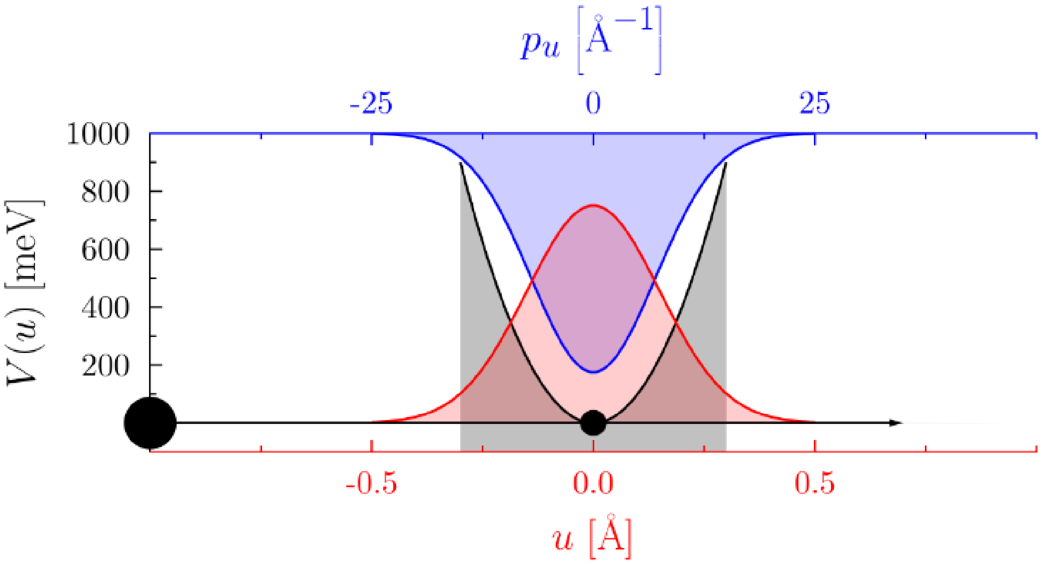
- C Andreani, R Senesi, M Krzystyniak, G Romanelli, F Fernandez-Alonso, «Atomic quantum dynamics in materials research» , Experimental Methods in the Physical Sciences 49, 403-457 (2017).

Relevance of measurements of momentum distributions and kinetic energies



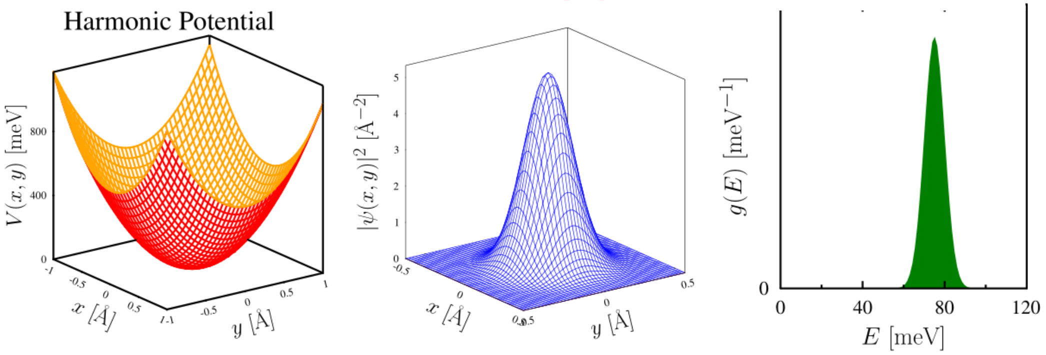
- Neutron Compton Scattering (Deep Inel. Neutron. Scattering- DINS): sensitive to ground state kinetic energy
- Inelastic Neutron Scattering (INS): sensitive to the energy difference between ground and first excited states

The shape of the potential determines the shape of the nuclear momentum distribution.

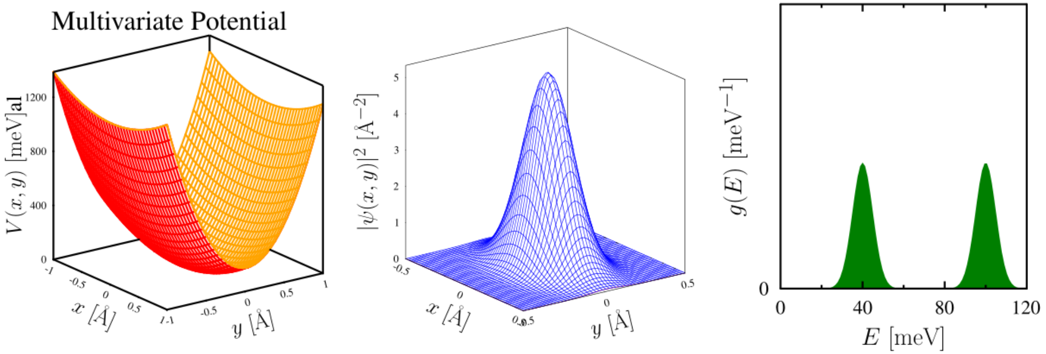


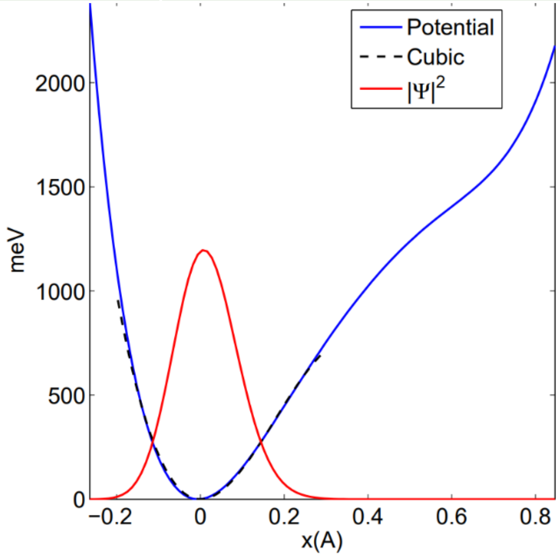
Potential	Momentum distribution
Isotropic	Gaussian
Anisotropic	Multivariate
Anharmonic	Gauss-Hermite expansion

Harmonic Potential



Multivariate Potential

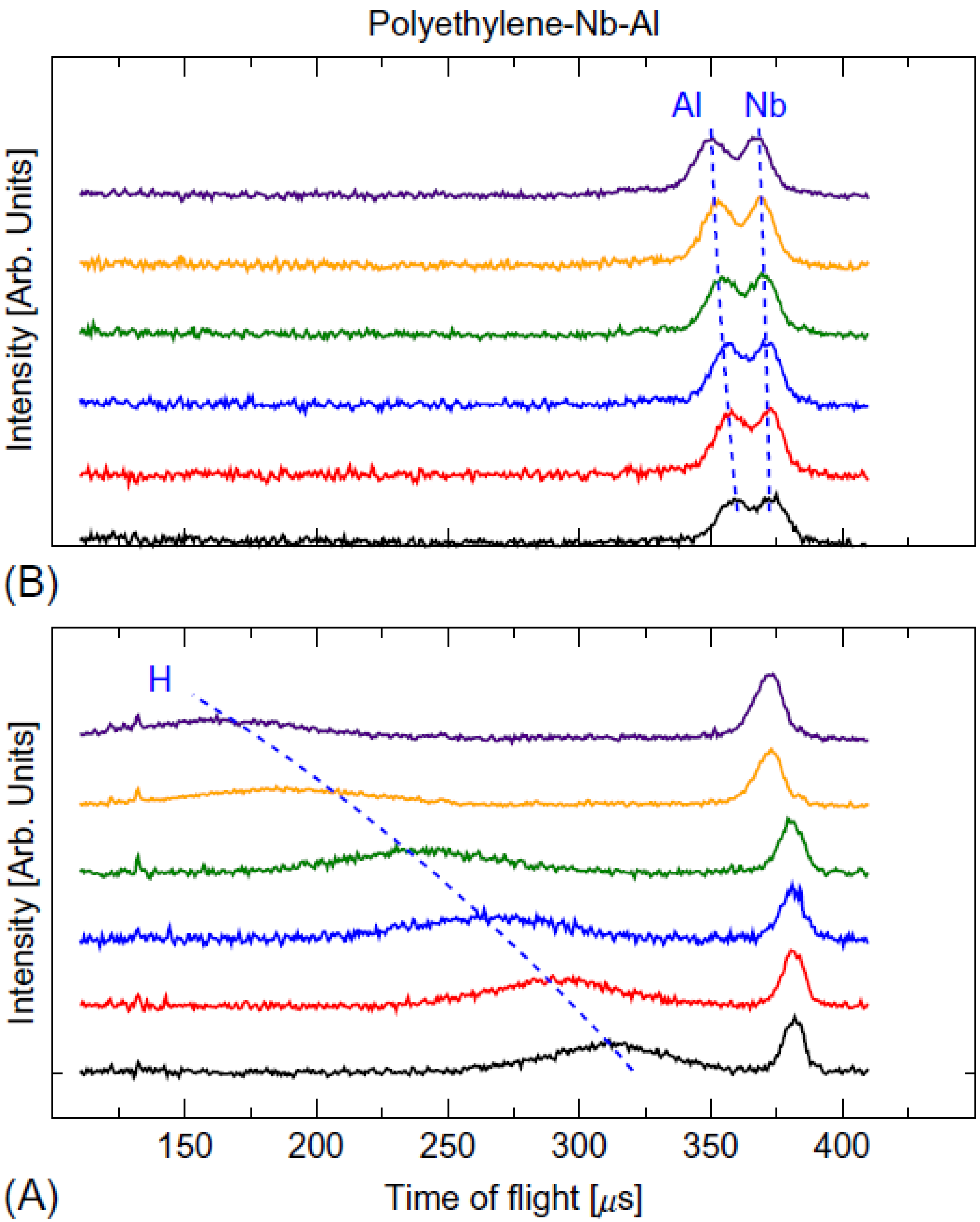




C Andreani, R Senesi, M Krzystyniak, G Romanelli, F Fernandez-Alonso; Rivista del Nuovo Cimento, 291-340 (2018)

L Lin, JA Morrone, R Car, and M Parrinello; Phys Rev B 83, 220302(R) (2011)

How the data look alike?



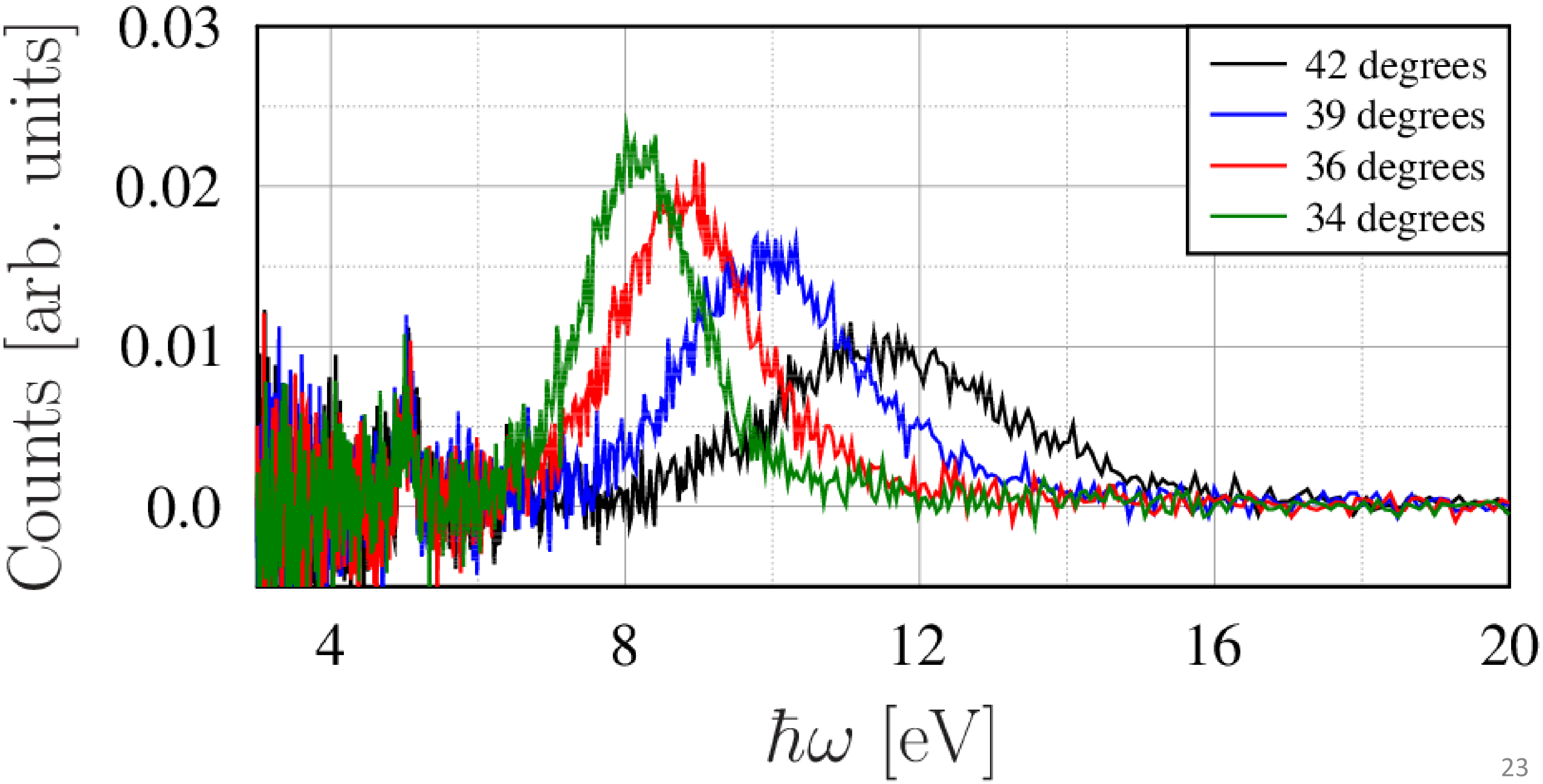
ToF spectra from a sample composed of three superimposed polyethylene, niobium, and aluminum slabs

Mass separation capability. For each atomic mass in the sample

$$M_i$$

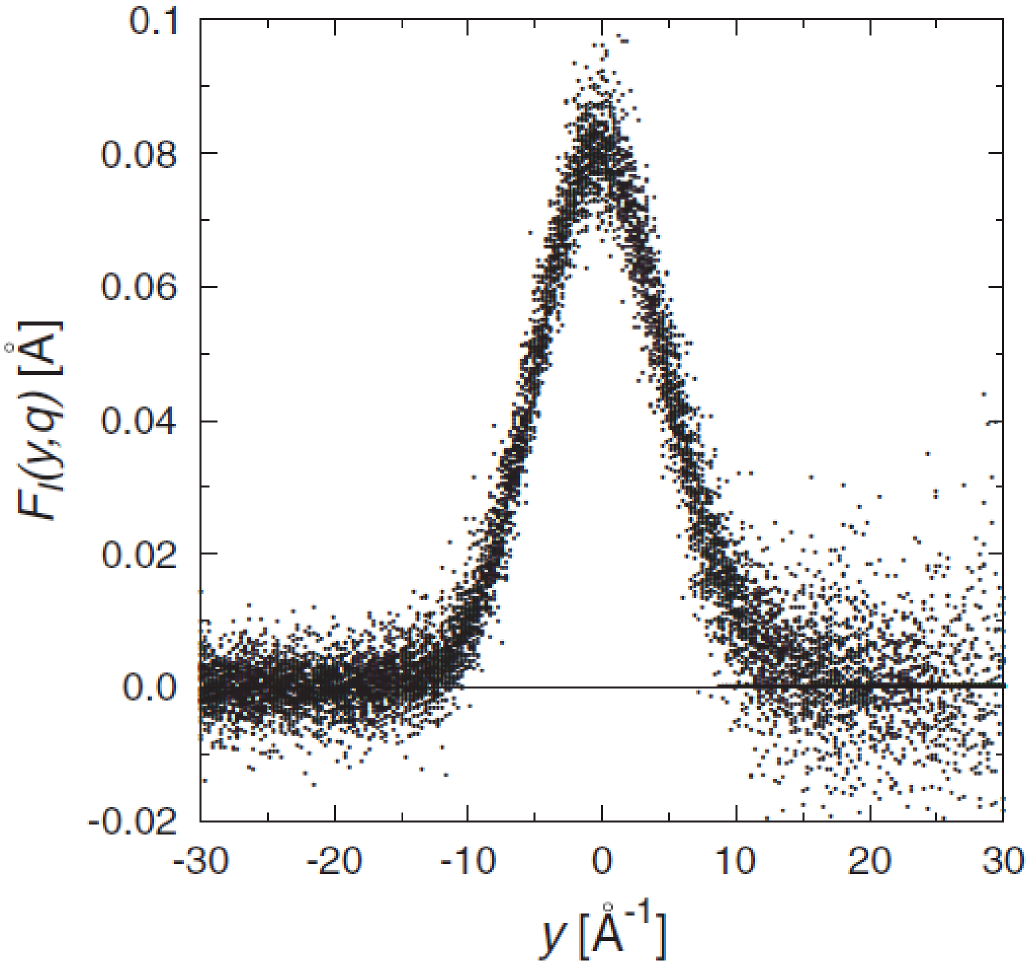
$$\frac{\hbar^2 Q^2}{2M_i}$$

Example of spectra converted into energy transfer



Example of spectra converted into atomic momentum (wave vector) units

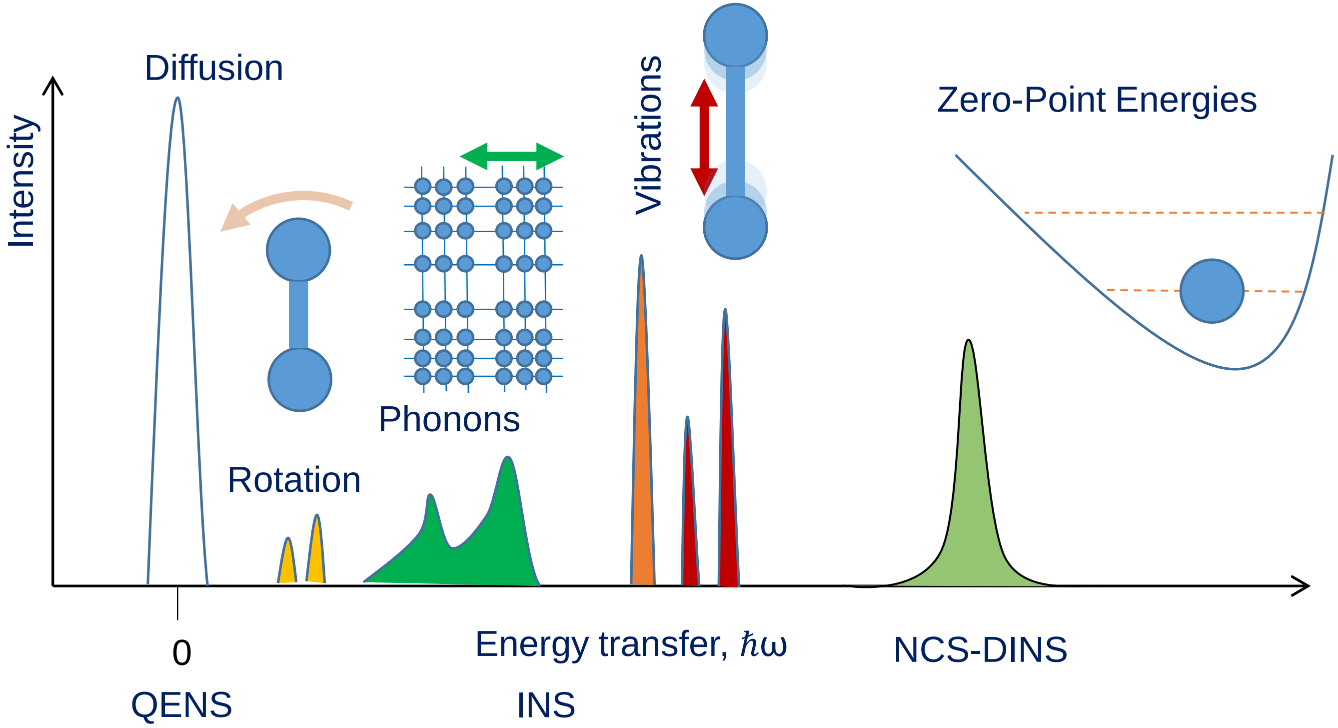
$$y = \frac{M}{\hbar^2 Q}\left(\hbar\omega - \frac{\hbar^2 Q^2}{2M}\right) = \frac{1}{2}\mathbf{p}\cdot\hat{Q}$$



All spectra scale and collapse into the Neutron Compton Profile

Inelastic neutron scattering probes different motions and processes depending on the magnitude of the energy transfer.

$$Q\Delta x \sim \frac{1}{2} \qquad \omega\Delta t \sim \frac{1}{2}$$



Energy [eV]	Wavelength [Å]
0.4	0.45
1	0.29
10	0.09
20	0.06
50	0.04
100	0.03

TOR VERGATA
UNIVERSITÀ
DEGLI STUDI
DI ROMA

Consiglio Nazionale
delle Ricerche

Tutorial

14:00-15:30 today

A project to identify unknown chemical components in a sample by inspecting its neutron Compton spectra in energy transfer

Data representative of an experiment carried out on VESUVIO