CENTRAL EUROPEAN TRAINING SCHOOL
ON NEUTRON SCATTERING

23 –28 Apr 2023

BUDAPEST NEUTRON CENTRE (BNC)

THREE AXES SPECTROMETRY

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O V E R V I E W

Lecture is addressed to newcomers in the field

(few formulae, focus on concepts rather than technical details)

- Introduction to Inelastic Neutron Scattering (INS)
- Relation to other methods
- Three Axes (Triple Axis) Spectrometer
- Examples (taken from experiments)
- Formal and technical aspects (Appendix)

ELASTIC VS. INELASTIC SCATTERING / TAS

DISTRIBUTION of INSTRUMENTS in 19 MAJOR NEUTRON LABS (~ Feb. 2011)
[vs. Non-Resonant Inelastic X-ray Scattering with High Resolution (~ meV)]

	ILL (F)	ISIS (GB)	LLB (F)	FRM2 (D)	HZB (D)	SINQ (CH)	BNC (H)	REZ (CZ)	NIST (US)	ORNL (US)	SNS (US)	LANSCE (US)	CNBC (CDN)	DUBNA (RU)	GATCHINA (RU)	JAERI (JPN)	J-PARC (JPN)	HANARO (KOR)	OPAL (AUS)	Neutron Instrum. Major Labs Total	Neutron Instruments (Estimated World Total)	Synchr Beamlines (Estimated World Total)
Powder Diffraction	6	9-11	7	3	8	4	2	3	3	2	3	5	2	5	3	3	5	2	4	~78 (23%)		
Single Crystal Diffraction	8	1	3	2	2	1				1	1	1			1	1		1		~23 (7%)		
Special. Diffraction	2			2	1							1				1	1			~8 (2.4%)		
(U)SANS	2-3	3-4	5	4	3	3	1	1	3	2	1		1	1	3	4	1	1	3	~43 (13%)		
Reflectometry	3	5	2	3	2	3	2		2		2	2	1	2	2		1	1	1	~34 (10%)		
Specialised Instruments	5-6		1	5	5	6	7	4	6	1				1	3	2	3	1	1	~52 (15%)		
Inelastic TAS	7		5	3	1-2	4	2		4	3			3		1	8		1	2	~44 (13%)		
Inelastic TOF	4	7-8	1	2	1	1	1		1		3	2		2	1	2	4		1	~34 (10%)		
Inelastic Other (Spin Echo, etc.)	5		1	4	1-2	1			3		2				1	1			1	~21 (6%)		
TOTAL	43	26	25	28	25	23	15	8	22	9	12	11	7	11	15	22	15 (9)	7	13 (7)	337	~ 350	>600
INELASTIC	16	8	7	9	4	6	3	-	8	3	5	2	3	2	3	11	4	1	4	99	~ 100	~ 5
% INELAST.	37%	30%	28%	32%	16%	26%	20%	-	36%	33%	42%	18%	43%	18%	20%	50%	27%	14%	31%	~30%	~29%	~1 %

ELASTIC VS. INELASTIC SCATTERING / TAS

DISTRIBUTION of INSTRUMENTS in 17 MAJOR NEUTRON LABS (~ Oct 2021)
[vs. Non-Resonant Inelastic X-ray Scattering with High Resolution (~ meV)]

	ILL (F)	ISIS (GB)	FRM2 (D)	SINQ (CH)	BNC (H)	REZ (CZ)	NIST (US)	ORNL (US)	SNS (US)	ESS (S) 2024	DUB NA (RU)	PIK (RU) 2024	JAERI (JPN)	J-PARC (JPN)	HAN ARO (KOR)	OPAL (AUS)	SNS (CHI-NA) >2021	Neutron Instrum. Major Labs Total	Neutron Instruments (IAEA World Total of 26 countries)	Synchr Beamlines (Estimated World Total)
Powder Diffraction	6	12	3	4	3	3	2	4	5	4	8		3	6	5	4	6	~78 (22%)		
Single Crystal Diffraction	7	1	2	1				1	1				1	1	1			~16 (5%)		
Special. Diffraction	2		2						1	1			1				1	~8 (2.3%)		
(U)SANS	3-4	4	4	3	2	1	5	2	2	2	1		4	1	3	3	2	~43 (12%)		
Reflectometry	4	4	3	3	2		4		2	2	3			2	3	2	2	~36 (10%)		
Specialised Instruments	6-7	6-7	5	6	9	3	10	2	2	1	4		2	4	3	2	4	~70 (20%)		
Inelastic TAS	8		3	4	2		4	4					8		3	2		~38 (11%)		
Inelastic TOF	3	7-8	2	1			1		5	4	3		2	4	1	1	4	~38 (11%)		
Inelastic Other (Spin Echo, BS, etc.)	5		4	1			3		2	1			1	2		1	1	~21 (6%)		
TOTAL	45	37	28	22	18	7	29	13	20	15	19	20	22	20	19	15	20	349	~ 370	> 1000
INELASTIC	16	8	9	4	2	-	7	4	5	5	3	5	11	6	4	4	5	98	~ 105	< 10
% INELAST.	36%	22%	32%	18%	11%	-	24%	31%	25%	33%	16%	25%	50%	30%	21%	27%	25%	~28%	~28%	~1 %

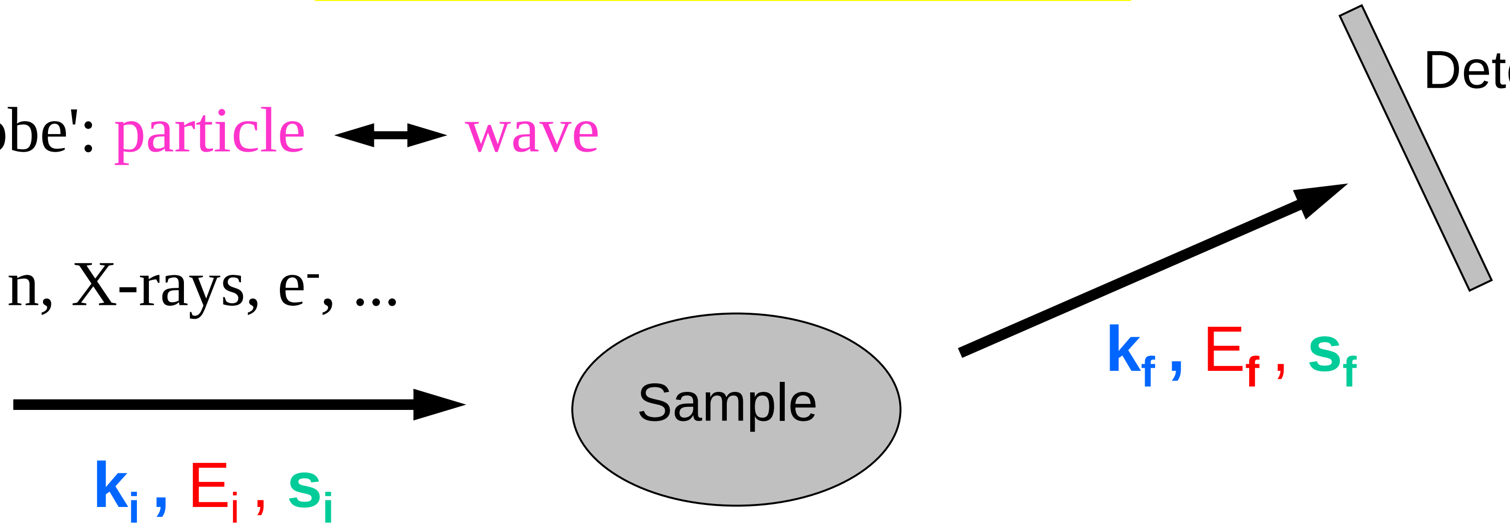
INELASTIC NEUTRON SCATTERING (INS)

General distribution of instruments fairly stable
(at least over the last decade):

- ~ 100 neutron spectrometers,
  i.e. ~30% of all instruments at neutron labs,
  are designed to study inelastic scattering
- ~ 45 Triple Axis Spectrometers (nearly one half of INS)
- non-resonant inelastic scattering with meV resolution
  still largely absent at synchrotons
  (exception: ~5 instruments at ESRF, APS, SPring-8,
  further instruments planned)
- provides in-depth information on a huge range of phenomena in physics, chemistry, biology, materials science, geosciences …

SCATTERING PROCESS

'probe': particle ↔ wave



	initial	final
momentum	$\mathbf{p} = \hbar\, \mathbf{k_i}$	$\mathbf{p} = \hbar\, \mathbf{k_f}$
energy = f (|**p**|)	$E = \hbar\, \omega_i$	$E = \hbar\, \omega_f$
spin (polarization)	$\mathbf{s_i}$	$\mathbf{s_f}$

$E_i \neq E_f \;\rightarrow\;$ INELASTIC SCATTERING

‚Natural‘ variables for describing a scattering process

Momentum transfer $\Delta \mathbf{p} = \hbar\,(\mathbf{k}_i - \mathbf{k}_f) = \hbar\,\mathbf{Q}$

Energy transfer $\Delta E = E_i - E_f = \hbar(\omega_i - \omega_f) = \hbar\,\omega$

Spin and magnetic scattering is not discussed in the following !

$\Delta \mathbf{p}$ and ΔE contain all information !

Energy transfer and momentum transfer are not independent :

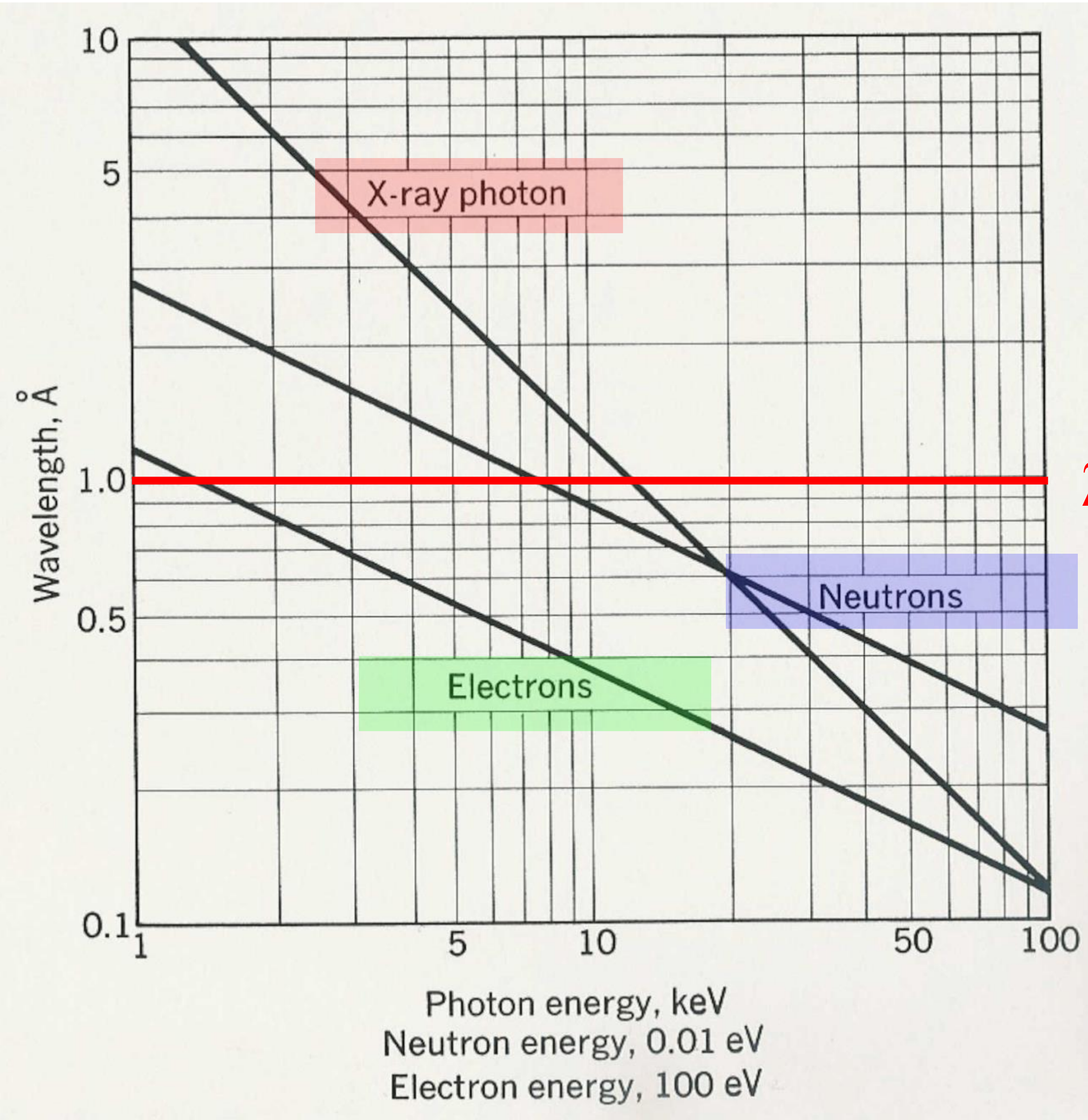
Photon ($m = 0$): $E = p\,c = \hbar\,k\,c$

$\rightarrow \Delta E = \hbar\,c\,(k_i - k_f)$

Neutron, electron ($m \neq 0$): $E = p^2 / (2m) = (\hbar\,k)^2 / (2m)$

$\rightarrow \Delta E = \hbar^2\,(k_i^2 - k_f^2) / (2m)$

RELATION ENERGY-WAVELENGTH for PHOTONS, NEUTRONS, and ELECTRONS



Note different slopes for particles with

$m = 0$ (photon)

$m \neq 0$ (neutron, electron)

λ = 0.1 nm (typical value)

$\lambda = 0.1\ \text{nm} = 1\text{Å} \equiv$

12.4 keV photon

78 meV neutron

144 eV electron

- Generally, the scattering process can be described by a so-called ‚scattering function' $S(\mathbf{Q}, \omega)$

- In many applications only the momentum transfer $\mathbf{Q}$ is analyzed, $S(\mathbf{Q}, \omega) \rightarrow S(\mathbf{Q})$, so that only part of the information provided by the scattering experiment is made use of.

  For example :

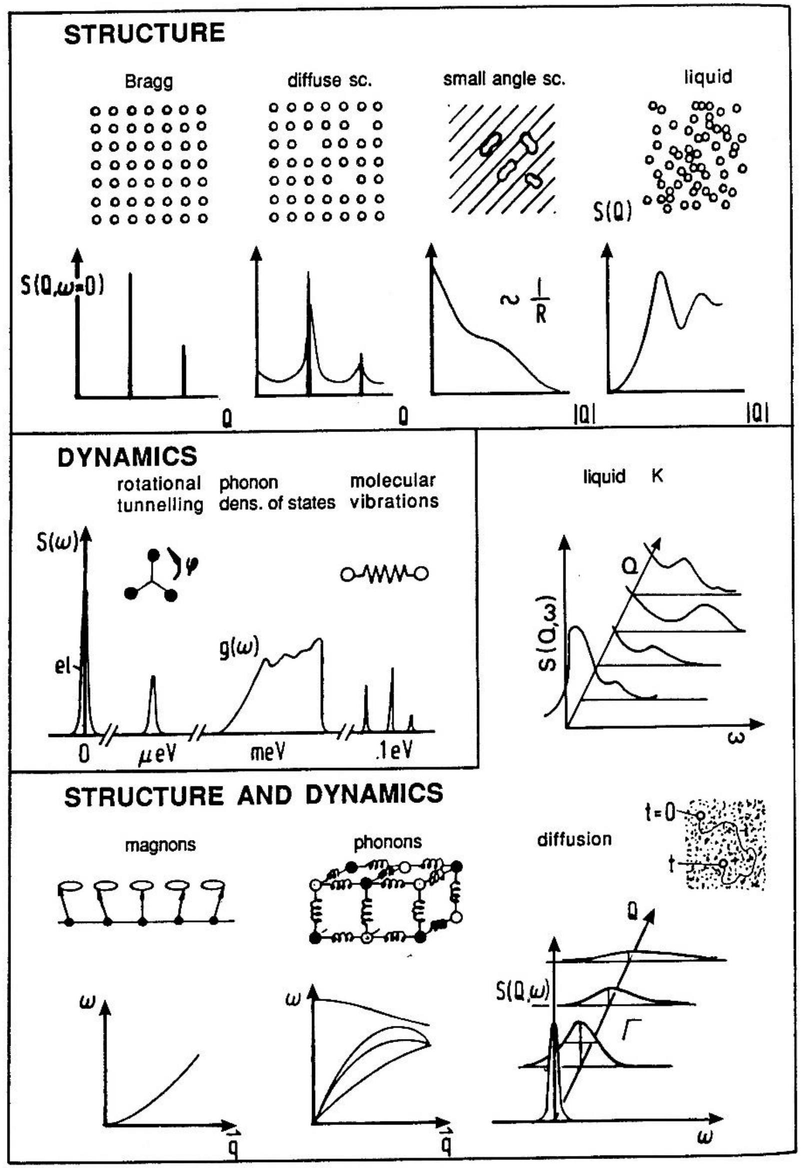
  - X – ray scattering (nearly always)
  - Neutron diffraction
  - Small angle neutron scattering

- The investigation of inelastic processes requires analysis of the energy transfer

Note: energy and momentum transfer $\hbar\,\omega$ and $\hbar\,\mathbf{Q}$ are frequently discussed without taking into account the constant $\hbar$. In practical units, therefore, the momentum transfer usually has the dimension of a reciprocal length while the energy transfer can be represented as an energy, a frequency or a temperature.

S(Q)

ELASTIC



S(ω)

INELASTIC

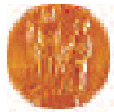
S(Q,ω)

INELASTIC

The scattering function S(Q, ω) in the experimental study of various physical systems

Nobel Prizes related to neutrons

The Nobel Prize in Physics 1935



James Chadwick

"for the discovery of the neutron"

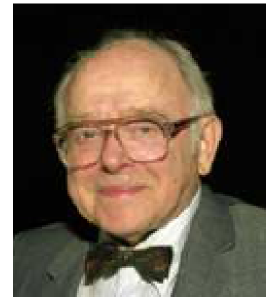
The Nobel Prize in Physics 1994

"In simple terms,
Clifford G. Shull
has helped answer the question of **where atoms are**,

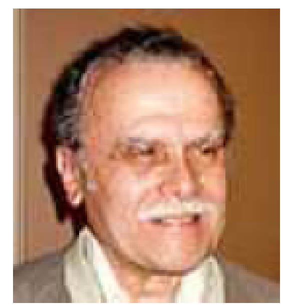
and

Bertram N. Brockhouse the question of **what atoms do**",
(Nobel citation)

"for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter"



"for the development of the neutron diffraction technique"



"for the development of neutron spectroscopy"

O V E R V I E W

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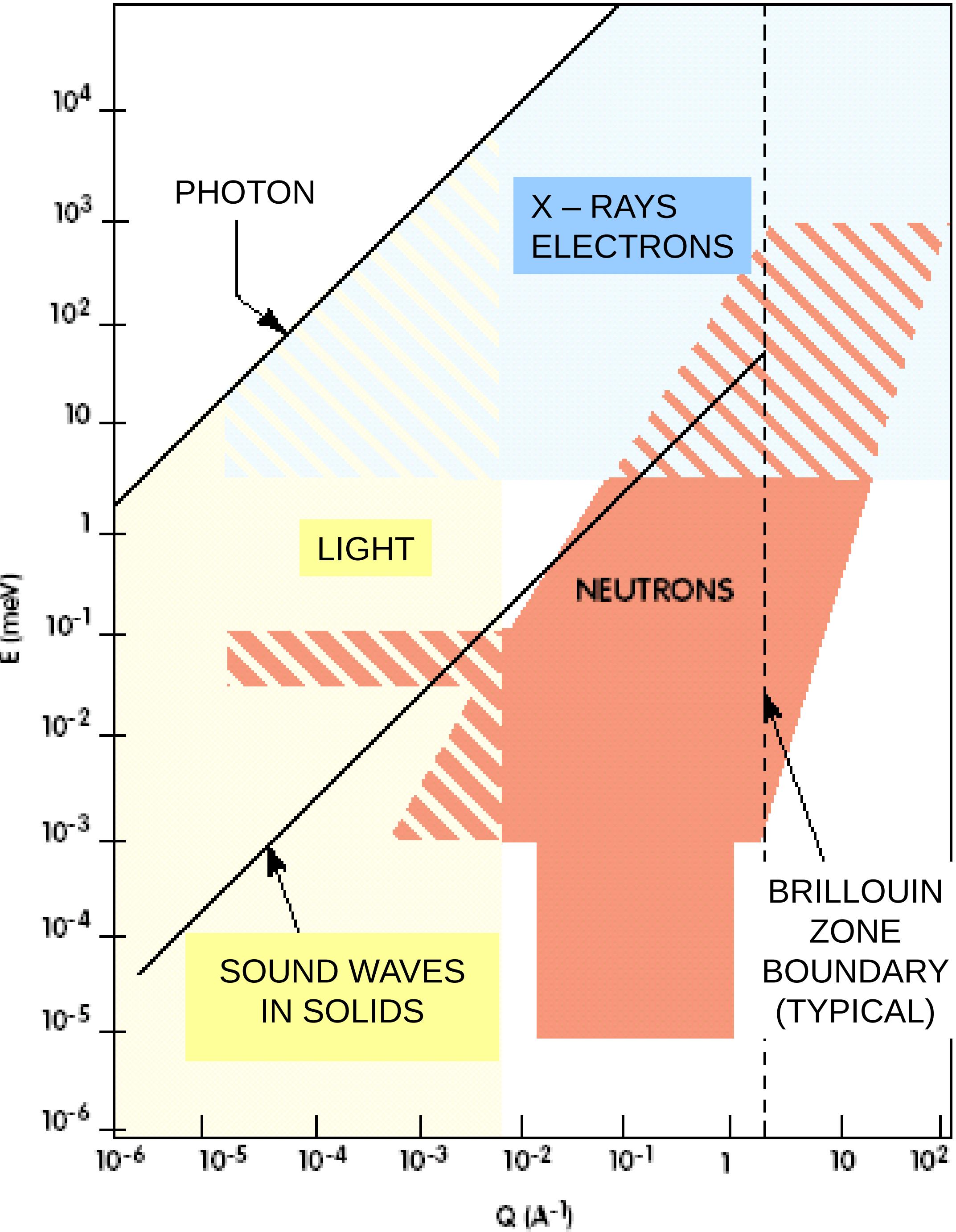
- Examples (taken from experiments)
- Formal and technical aspects

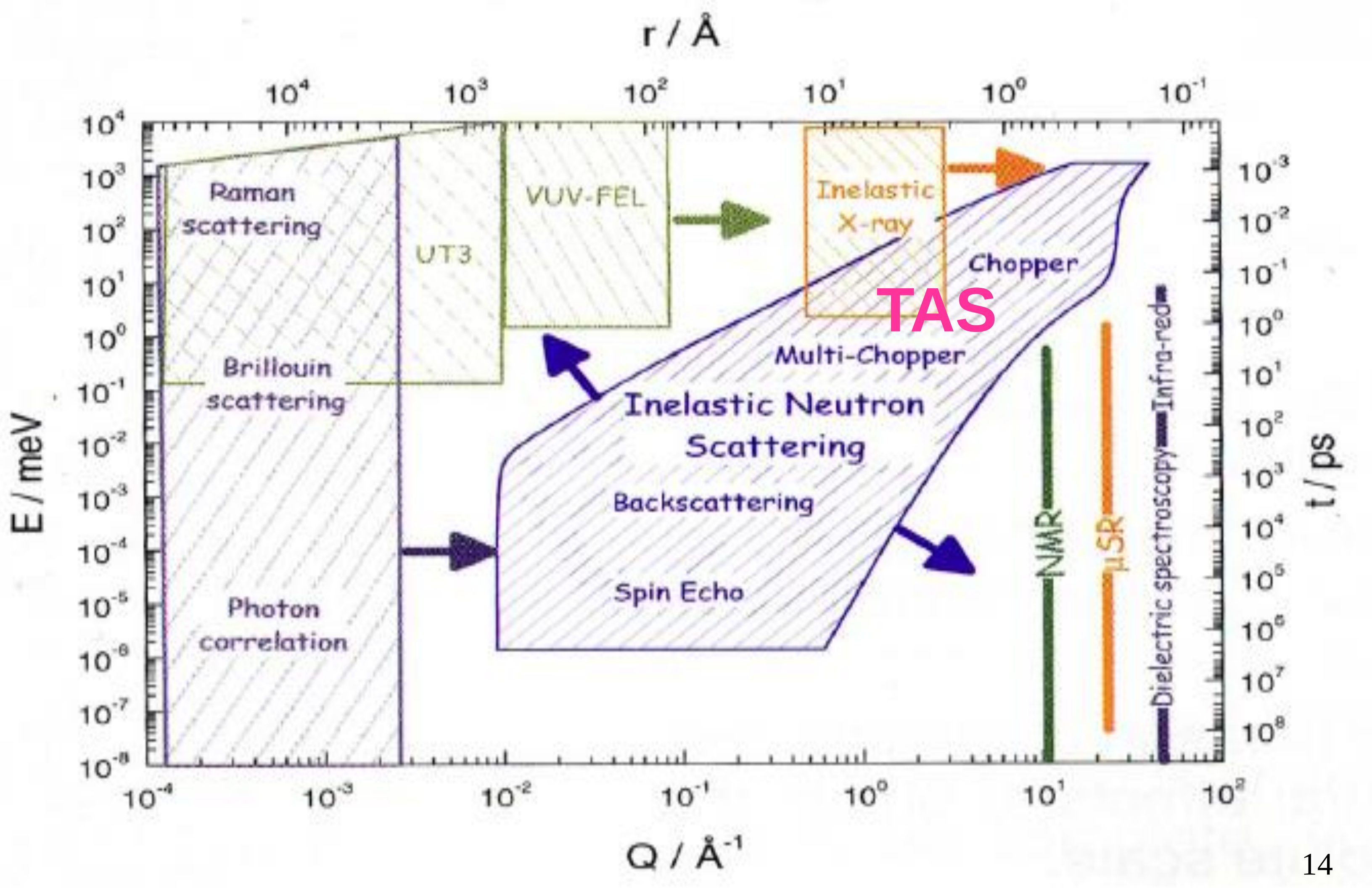


Transfer of **ENERGY** and **MOMENTUM** accessible with various probes

Photons
Neutrons
Electrons

E = ħ c Q (slope=1)
$E = \hbar^2/(2m)\, Q^2$ (slope=2)
define maximum energy and momentum transfer for photons and neutrons

Accessible (Q, E) range



Energy units in use for inelastic scattering:

1 meV ≡ 1.602×10^{-22} J ≡ 8.066 cm^{-1} ≡

≡ 0.2418×10^{12} Hz ≡ 11.605 K

[$E = \hbar\,\omega = h\,\nu = h\,c\,E/\lambda = k\,T$ → dispensing with constants gives ω, ν, 1/ λ, or T]

1 Joule =		6.2415×10^{21} meV	1.5092×10^{21} THz	7.243×10^{22} K	5.034×10^{22} cm^{-1}
1 meV =	1.602×10^{-22} J		0.2418 THz	11.605 K	8.066 cm^{-1}
1 THz =	6.626×10^{-22} J	4.1357 meV		47.97 K	33.36 cm^{-1}
1 Kelvin =	1.38065×10^{-23} J	0.08617 meV	0.2085 THz		0.695 cm^{-1}
1 cm^{-1} =	1.9865×10^{-23} J	0.1240 meV	0.02998 THz	1.439 K	

Neutrons: **E [meV] = 2.072 k^2 [Å^{-2}] ≈ 2 k^2 [Å^{-2}]**

X-rays: **E [keV] = 1.973 k [Å] ≈ 2 k [Å]**

Example: λ **= 2 Å (k = 3.14 Å^{-1}) gives for**

Neutrons: E= 20.5 meV or 237 K **X-rays: E = 6.2 keV or 7.2×10^7 K**

(typical values for condensed matter)

COMPARISON OF TECHNIQUES
(cf. typical Brillouin zone dimensions: 10 nm^{-1})

Typical excitation (phonons): $\nu \leq$ 10 THz, $k \leq$ 10 nm^{-1}

[Remember: EM Radiation: $E = p\,c = \hbar\,k\,c$, $\Delta E = \hbar\,c\,(k-k')$, $\Delta\mathbf{k} = \mathbf{k}-\mathbf{k'}$]

Light (Raman, Brillouin scattering): E ~ 500 THz, k ~ 1×10^{-2} nm^{-1}
→ ΔE ok, Δk=Q by 10^3 too small

Infrared: E, k are at least 10 x smaller than for visible light
→ same problem

Ultrasound: $\nu \leq$ 100 MHz, $k \leq 10^{-4}$ nm^{-1} → both ΔE and Δk insufficient

→ The above techniques work only in the long wavelength limit at the center of the Brillouin zone

X-rays: ν ~ 1×10^{6} THz, k ~ 20 nm^{-1} → ΔE, Δk are suitable
but: energy resolution < 10^{-6} required !
(very difficult, yet possible)

Neutrons: ν ~ 20 THz, k ~ 50 nm^{-1} → **excellent probe**

CONSEQUENCES FOR INELASTIC SCATTERING / 1

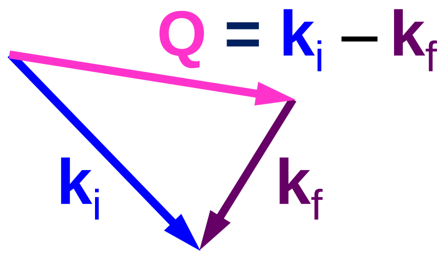
Both energy and momentum have to be conserved.

Momentum: $\hbar\,(\mathbf{k}_i - \mathbf{k}_f) = \hbar\,\mathbf{Q}$

From a geometrical point of view it is equivalent to consider

$$\mathbf{k}_i - \mathbf{k}_f = \mathbf{Q}$$

which can be visualized as the so-called *scattering triangle*.

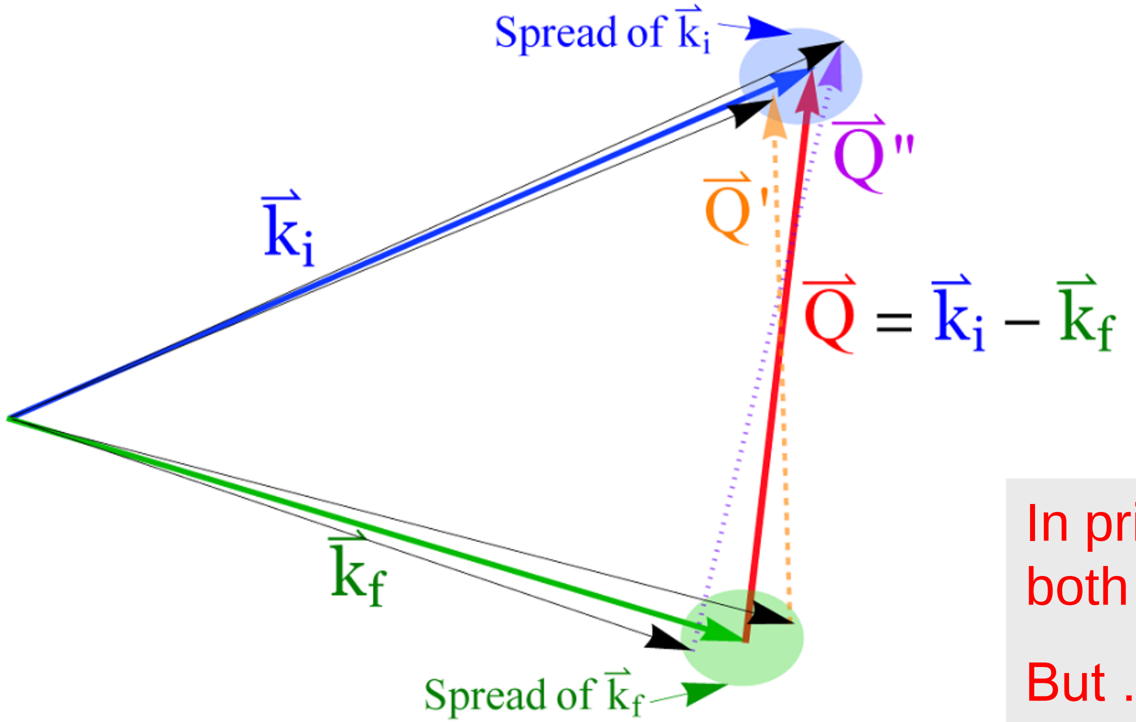


Note: sometimes one also can find the definition $\mathbf{k}_f - \mathbf{k}_i = \mathbf{Q}$ depending on whether the position of the neutron or the sample is adopted in describing the scattering process. Of course, either choice is possible (but they should not be mixed). Besides, in calculating actual scattering cross sections usually only the quantity $\mathbf{Q}^2$ is needed.

CONSEQUENCES FOR INELASTIC SCATTERING / 2
SCATTERING TRIANGLE (e.g. in a phonon measurement)

In a real experiment, both the direction and the length of the vectors $\mathbf{k}_i$ and $\mathbf{k}_f$ are not defined exactly but only within certain limits as given by the resolution of the instrument. Therefore, some scatter in the measured momentum transfer **Q** will be observed (e.g. **Q**' and **Q**'').

Determination of **Q** to a certain precision (say, a few %) presupposes that the vectors $\mathbf{k}_i$ and $\mathbf{k}_f$ are defined with sufficient precision.



Typically, the lengths of $\mathbf{k}_i$, $\mathbf{k}_f$ and **Q** all are of the same order ~5Å^{-1} (= 50 nm^{-1}).

Therefore, in defining $\mathbf{k}_i$ and $\mathbf{k}_f$ a precision of ~10^{-2} would be generally sufficient.

In principle, this holds for both neutrons and X-rays!

But …

CONSEQUENCES FOR INELASTIC SCATTERING / 3

… consider the energy transfer! If we want to measure a typical phonon energy of 20 meV with a precision of 5%, we need a resolution of 1 meV.

Neutrons: Typical neutron energies in phonon measurements are in the range 10 -100 meV. Therefore, in defining E_i and E_f a precision of $\sim 10^{-2}$ will be generally sufficient in accordance with the required precision of momentum transfer.

X-rays: Typical energies are ~10 keV. So, in order to resolve the energy transfer to within 1 meV, a precision of $\sim 10^{-7}$ is required!
This is very hard to achieve and has become possible only within the last decade.

In addition, using special techniques the resolution in neutron experiments can be extended from meV down to μeV and even neV which is cleary beyond the reach of X-rays.

→ Inelastic scattering is still largely the domain of neutrons.

O V E R V I E W

- Introduction to Inelastic Neutron Scattering (INS)
- Relation to other methods
- Three Axes (Triple Axis) Spectrometer
- Examples (taken from experiments)
- Formal and technical aspects

THREE AXES SPECTROMETER

- Comparison with other inelastic neutron scattering techniques
- Range of momentum and energy transfer
- Monochromators, analyzers
- Principles of instrument operation
- Thermal, cold and hot instruments
- Elementary excitations (phonons, [magnons])
- Dynamic structure factor
- Lattice dynamical models - simulation

INELASTIC TECHNIQUES / INSTRUMENTS

- **Three Axes Spectrometer**
- Time of Flight Techniques
- Filter Spectrometers
- Backscattering
- Spin Echo Instruments

Resolution: typically several percent of incoming energy

$\Delta E \cdot \Delta\tau \sim \hbar \rightarrow$ time scale !

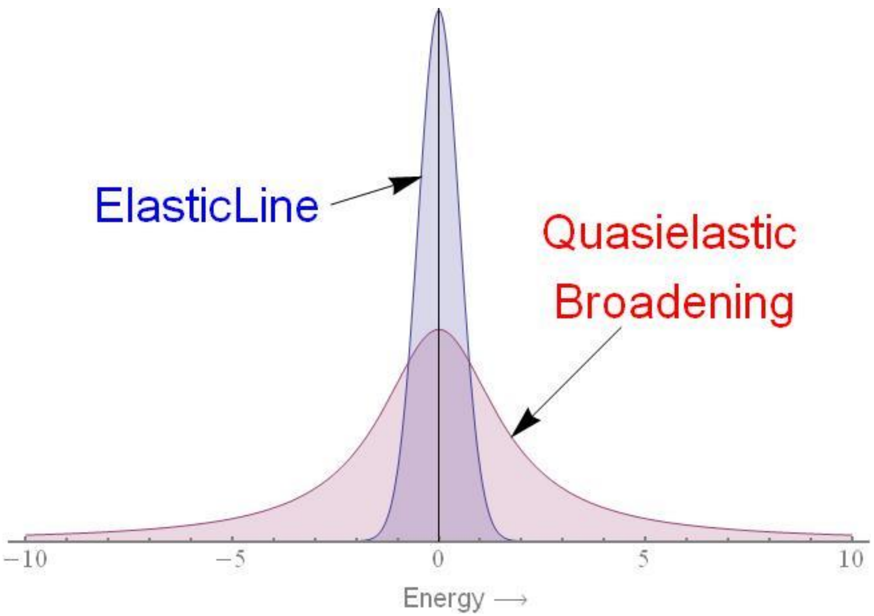
e.g.: $\Delta\tau \sim \hbar / \Delta E \sim \hbar / (\hbar\,\omega) = 1/\omega$

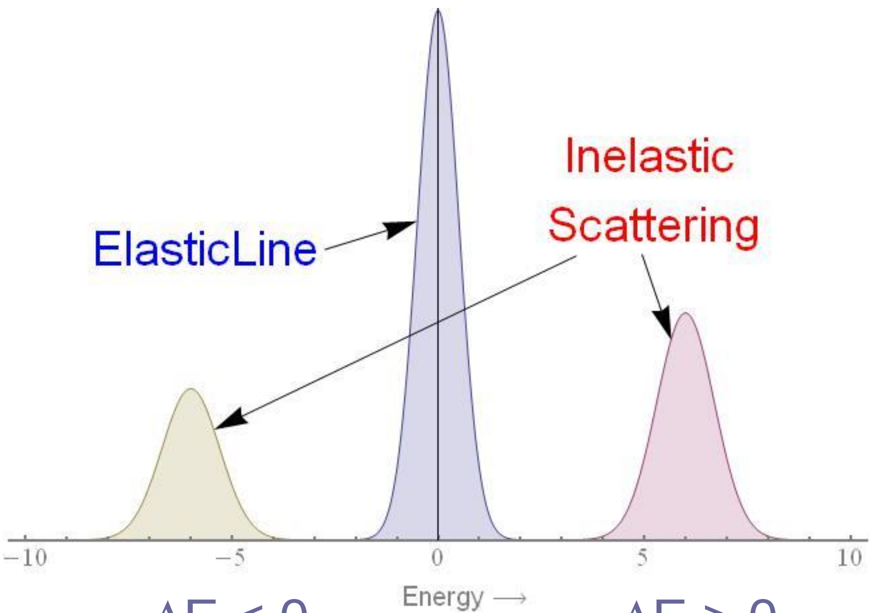
$\omega = 2\pi \cdot 1\ \text{THz} \rightarrow \Delta\tau \sim 1.6 \times 10^{-13}\ \text{s}$

QUASIELASTIC and INELASTIC SCATTERING

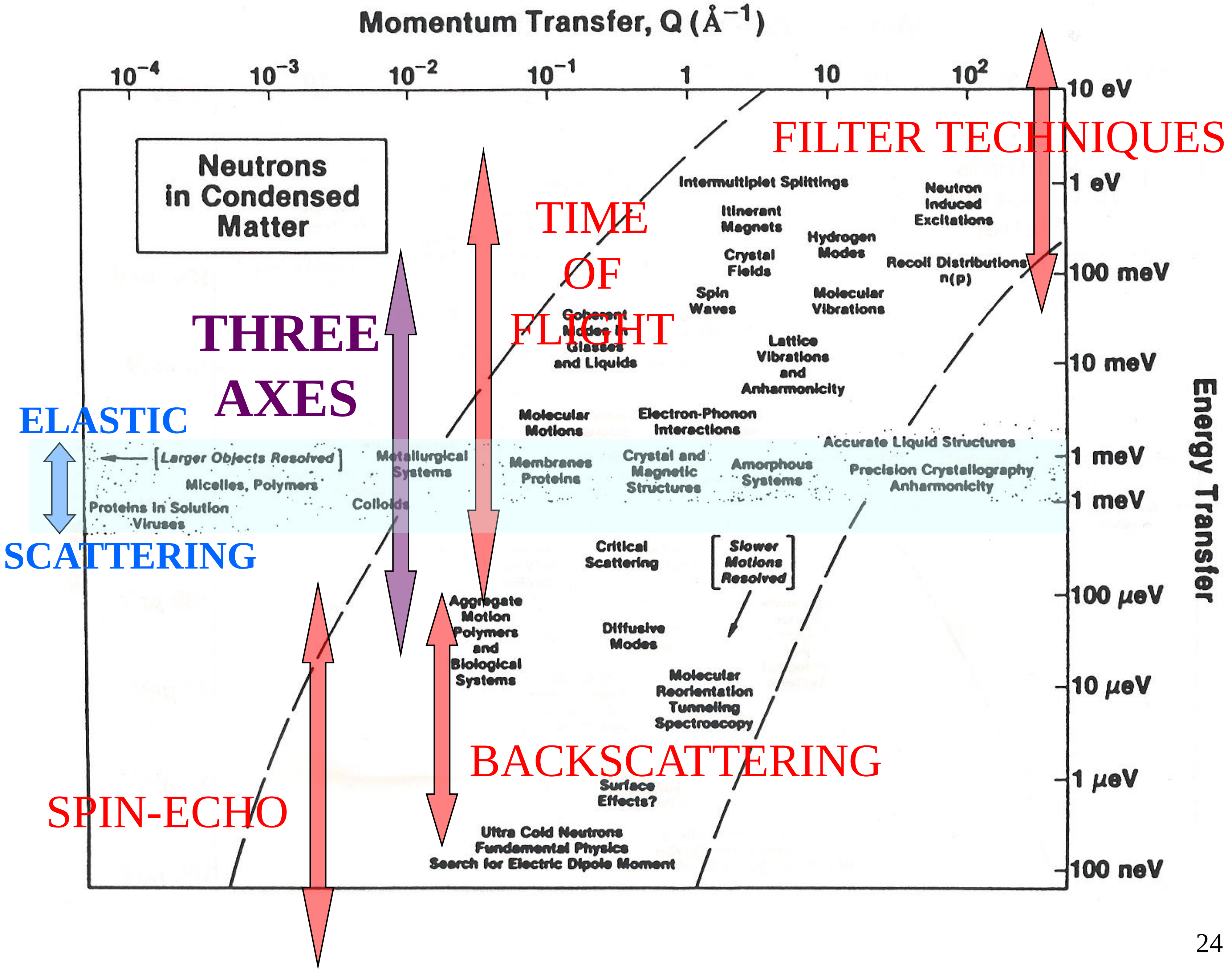
covered by other techniques
(TOF, Backscattering, NSE)

Typical TAS case



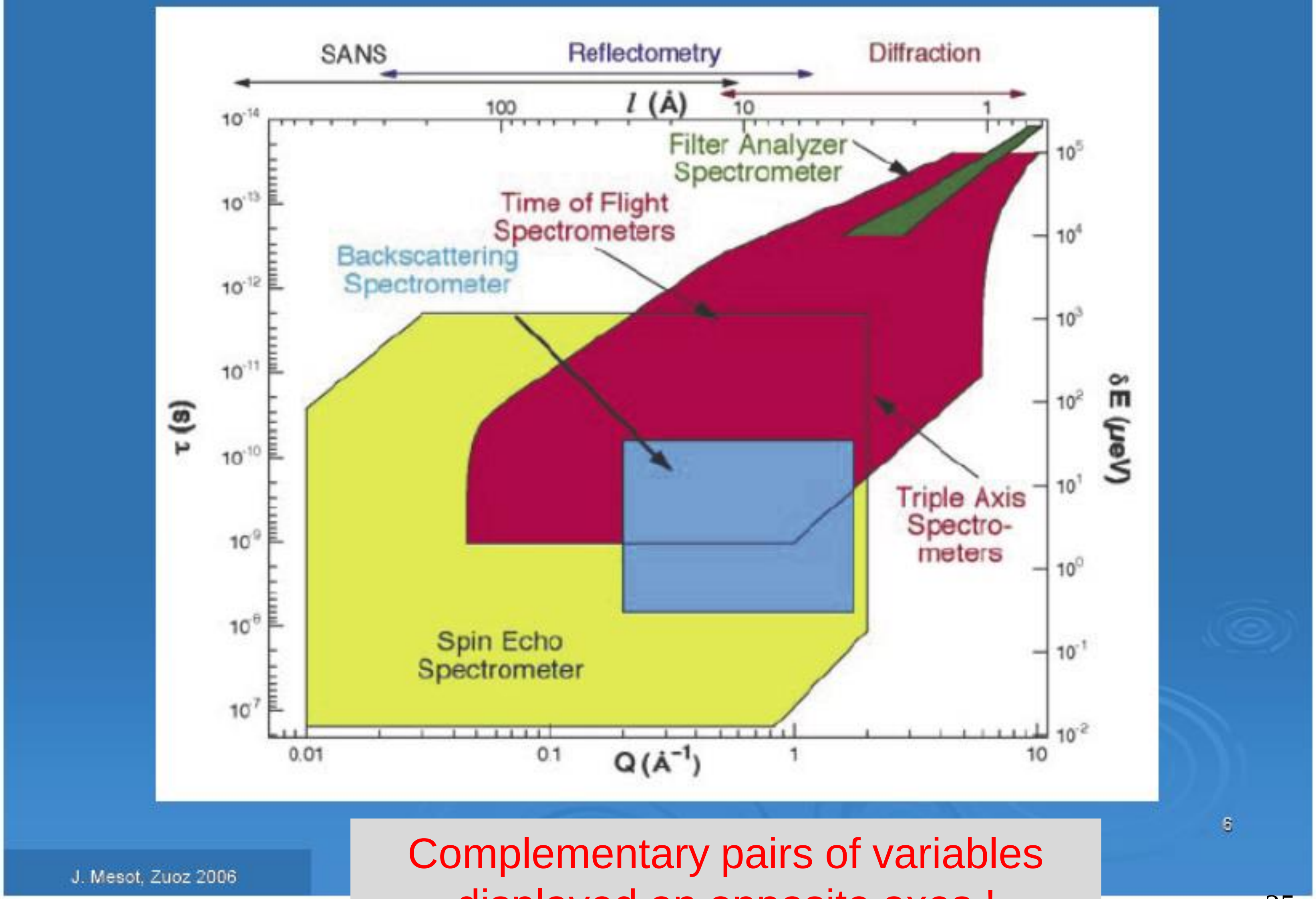


Width of the elastic line usually defined
by the resolution of the instrument

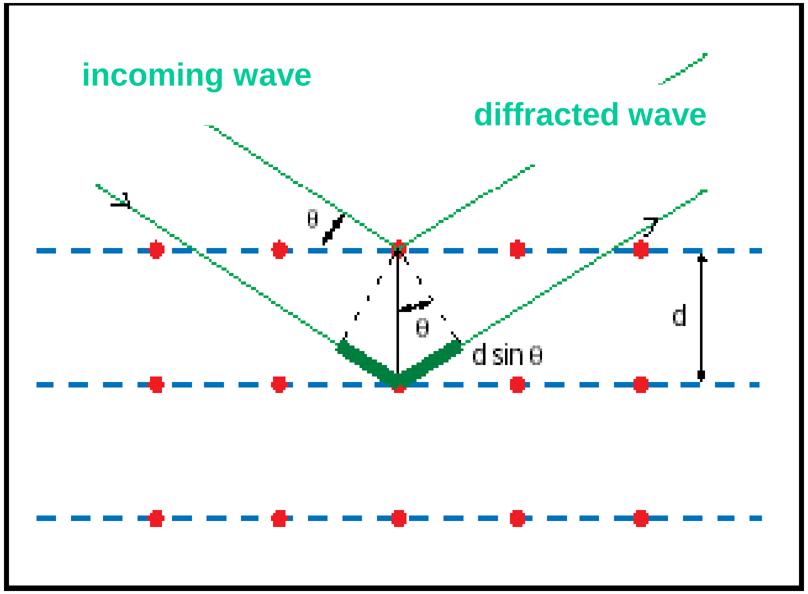


Momentum Transfer, Q (Å⁻¹)
10⁻⁴
10⁻³
10⁻²
10⁻¹
1
10
10²
Neutrons in Condensed Matter
FILTER TECHNIQUES
TIME OF FLIGHT
THREE AXES
ELASTIC SCATTERING
SPIN-ECHO
BACKSCATTERING
Intermultiplet Splittings
Itinerant Magnets
Crystal Fields
Hydrogen Modes
Neutron Induced Excitations
Recoil Distributions n(p)
Spin Waves
Molecular Vibrations
Coherent Modes in Glasses and Liquids
Lattice Vibrations and Anharmonicity
Molecular Motions
Electron-Phonon Interactions
Accurate Liquid Structures
Larger Objects Resolved
Metallurgical Systems
Membranes Proteins
Crystal and Magnetic Structures
Amorphous Systems
Precision Crystallography Anharmonicity
Micelles, Polymers
Proteins in Solution
Viruses
Colloids
Critical Scattering
Slower Motions Resolved
Aggregate Motion Polymers and Biological Systems
Diffusive Modes
Molecular Reorientation Tunneling Spectroscopy
Surface Effects?
Ultra Cold Neutrons Fundamental Physics Search for Electric Dipole Moment
10 eV
1 eV
100 meV
10 meV
1 meV
1 meV
100 μeV
10 μeV
1 μeV
100 neV
Energy Transfer

Range of inelastic instruments



Complementary pairs of variables displayed on opposite axes !

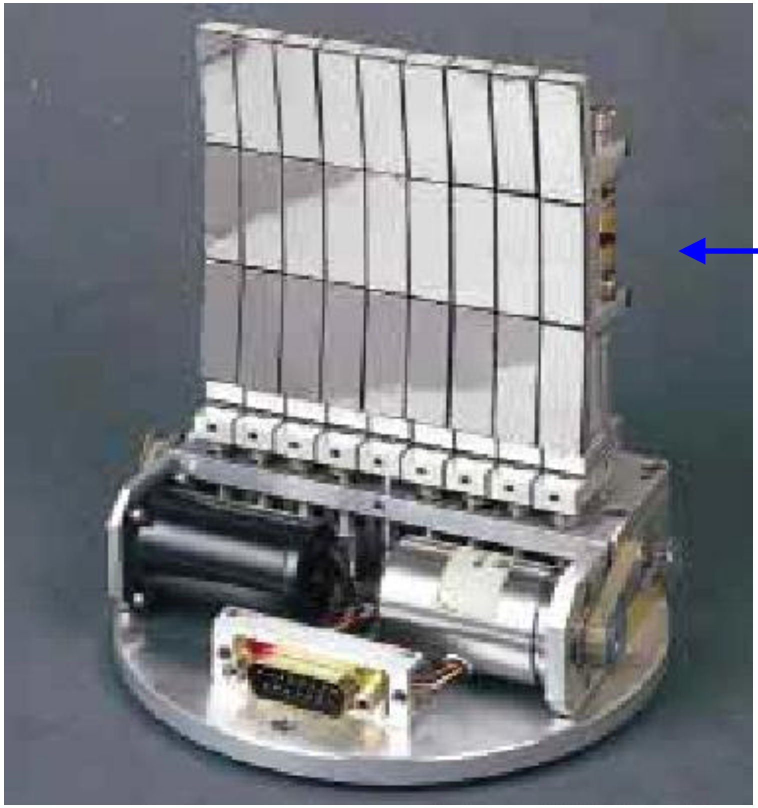


MONOCHROMATORS
and
ANALYSERS

PRINCIPLE:

BRAGG-REFLECTION:

$$2\,d\,\sin\theta = n\,\lambda$$

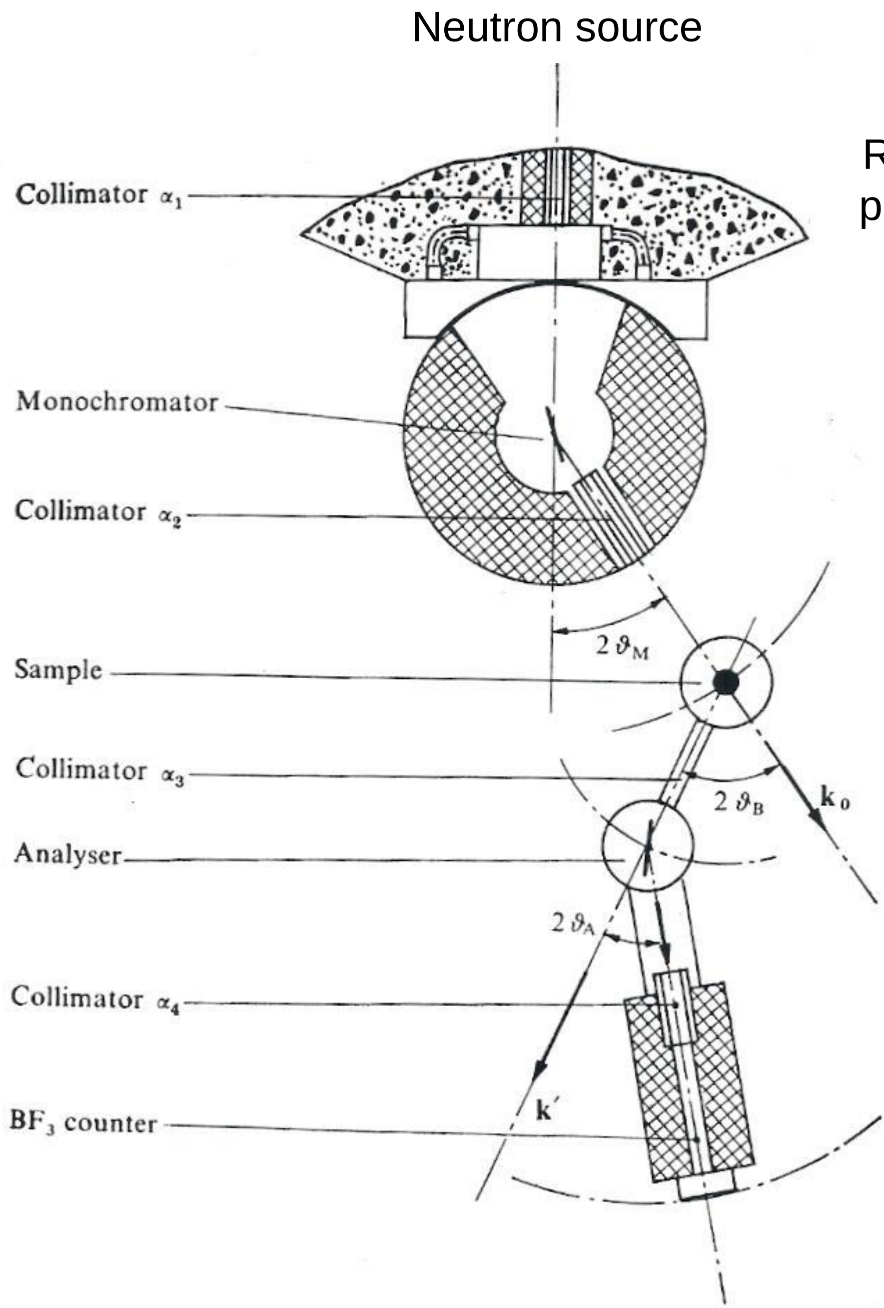


REALISATION:

Array of monocrystalline platelets allowing for focussing

Adjustable curvature as a function of
- sample distance
- sample size
- required resolution
- angle of incident neutron beam

THREE AXES SPECTROMETER FOR MEASURING S (**Q**, ω)

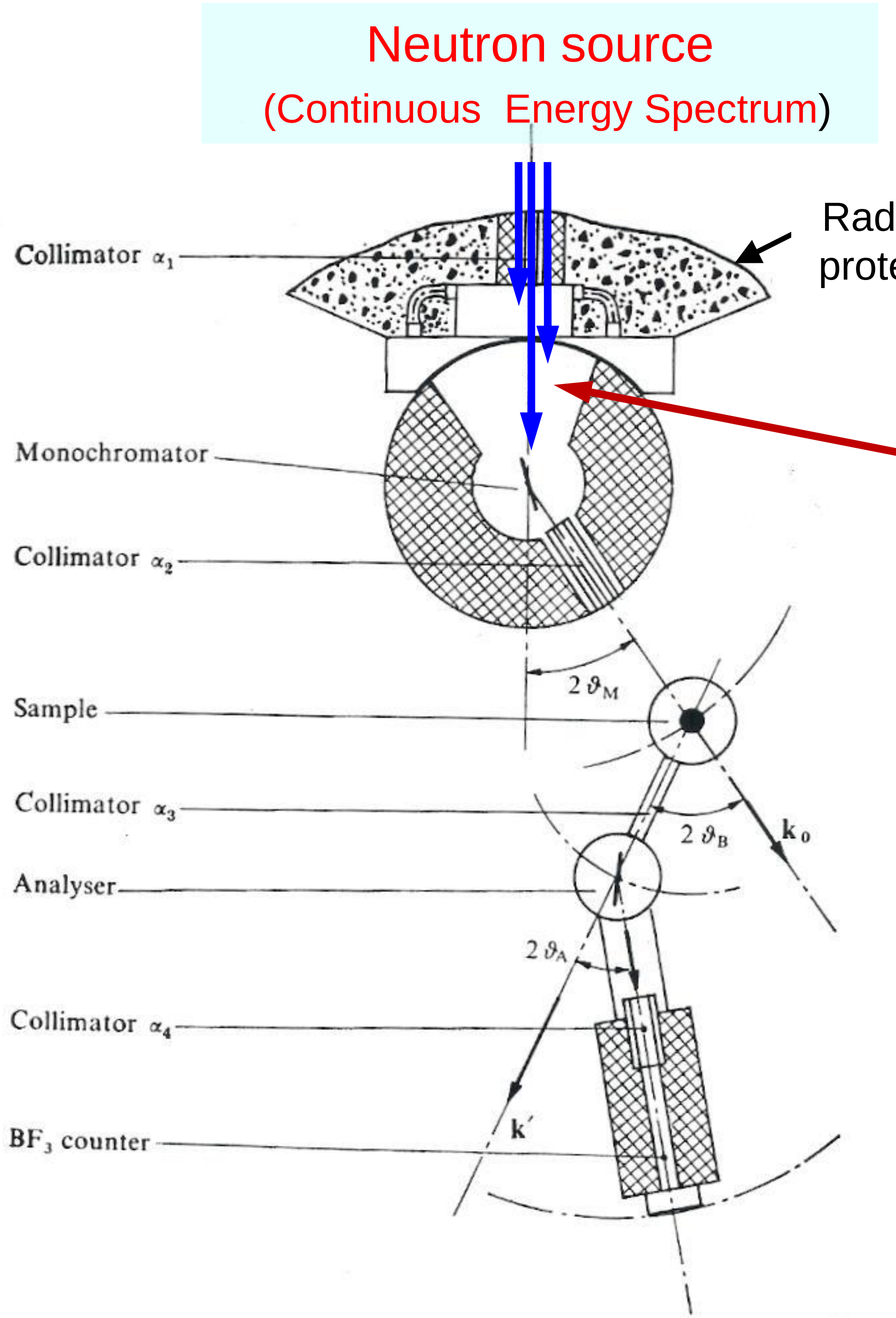


Goal: choose experimental set-up for selected values of

$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$ and

$\hbar\omega = E_i - E_f$

THREE AXES SPECTROMETER FOR MEASURING $S(\mathbf{Q}, \omega)$

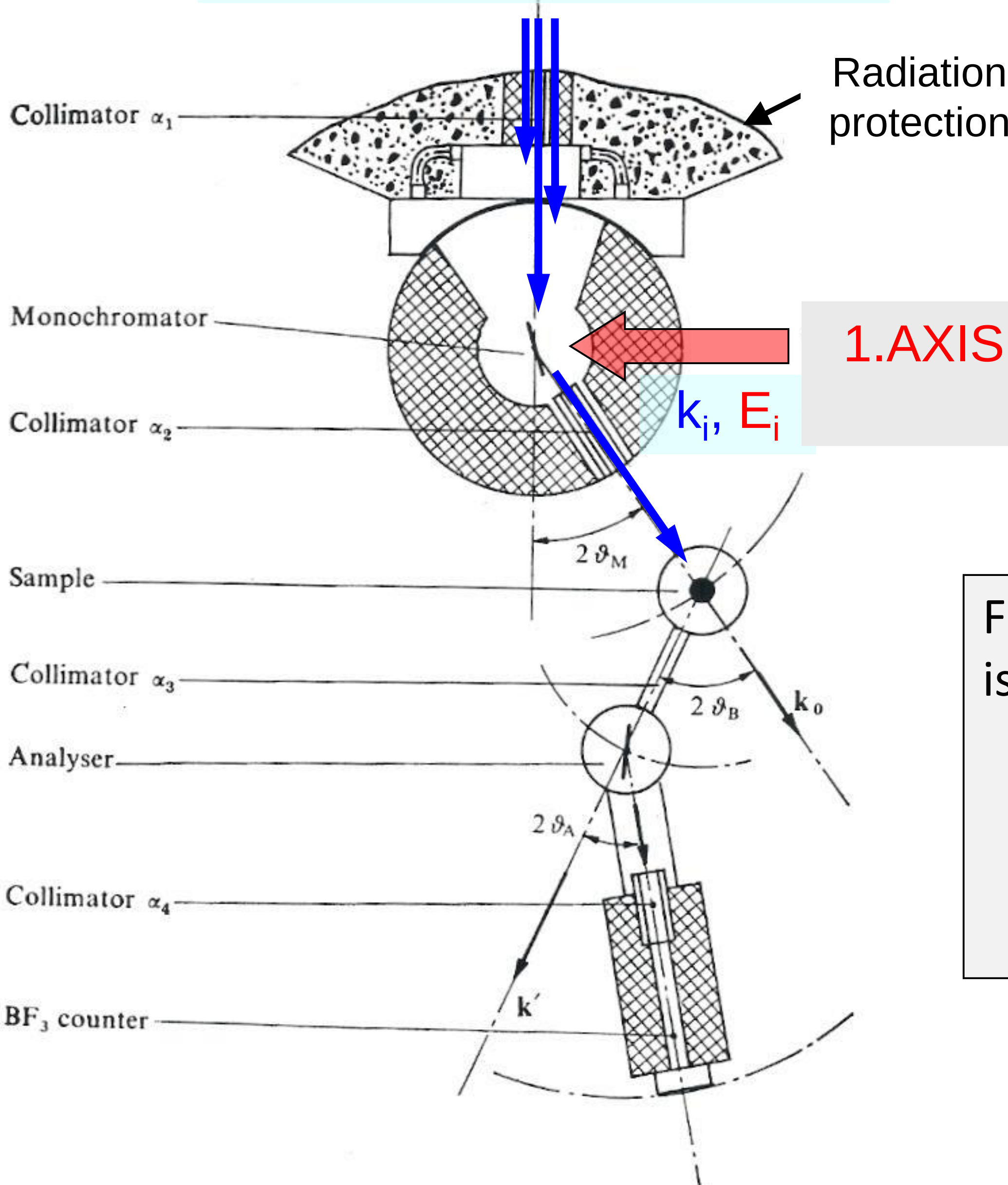


Reactor provides a continuous spectrum of different $\mathbf{k}_i$ (indicated by different lengths of arrows) and corresponding E_i

THREE AXES SPECTROMETER FOR MEASURING $S(\mathbf{Q}, \omega)$

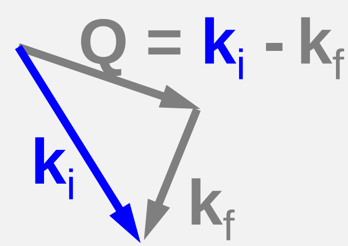
Neutron source

(Continuous Energy Spectrum)

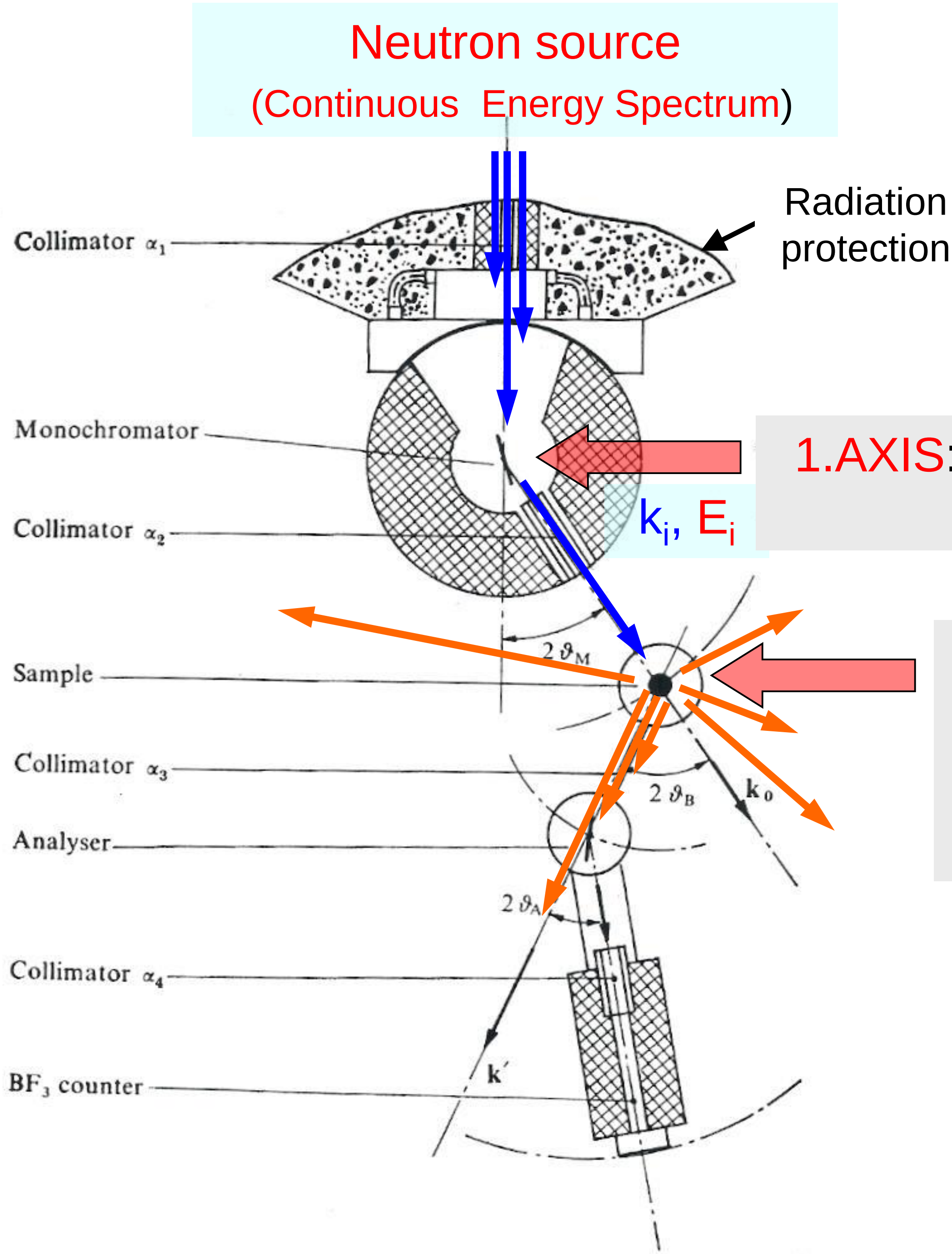


1.AXIS: choice of incident energy E_i ($\rightarrow |\mathbf{k}_i|$)

First side of Scattering Triangle is now defined



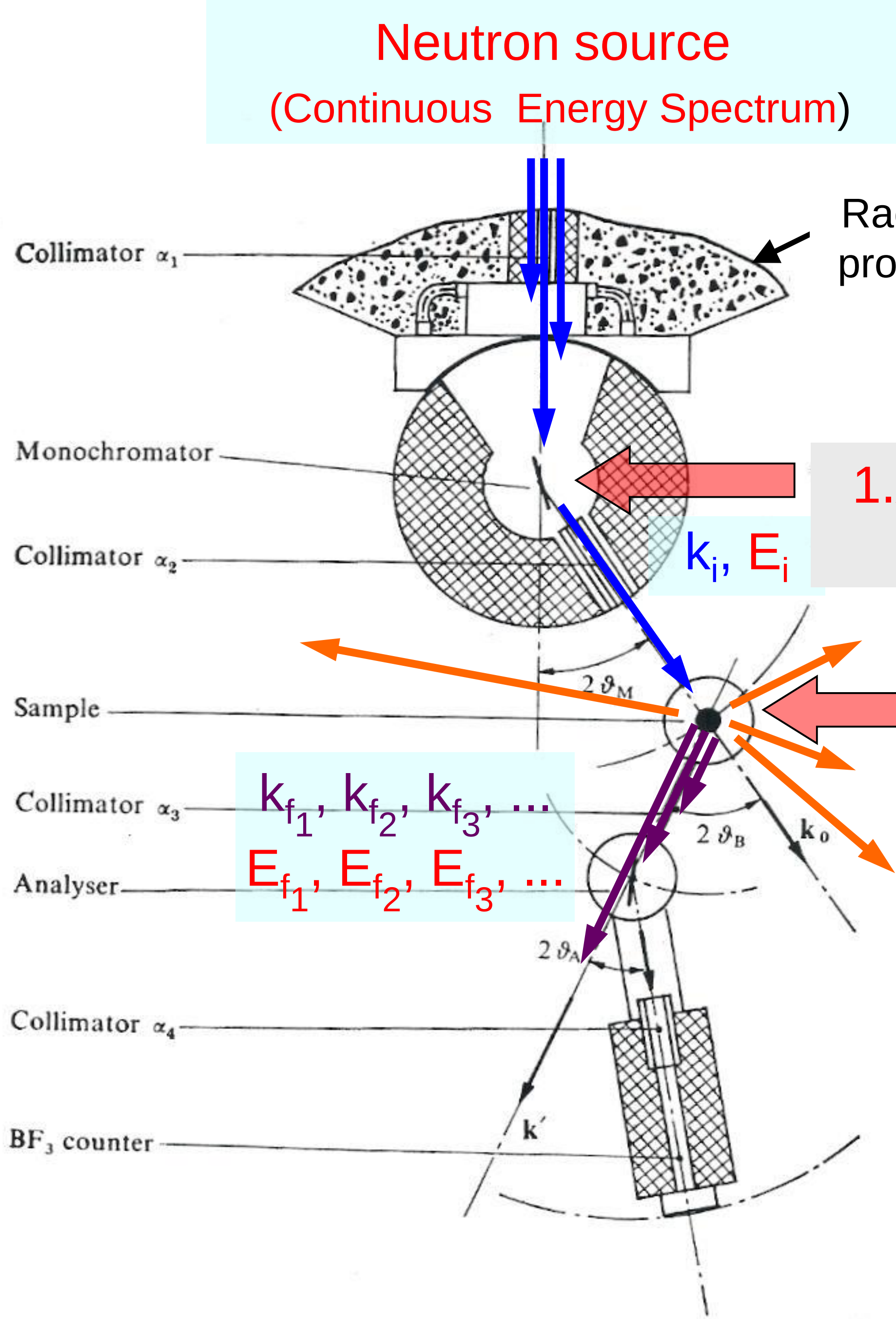
THREE AXES SPECTROMETER FOR MEASURING $S(\mathbf{Q}, \omega)$



1.AXIS: choice of incident energy E_i
($\rightarrow |\mathbf{k}_i|$)

sample position: many scattering processes may take place leading to different E_f and $\mathbf{k}_f$

THREE AXES SPECTROMETER FOR MEASURING S (**Q**, ω)

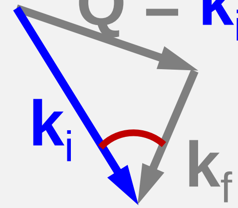


1.AXIS: choice of incident energy E_i ($\rightarrow |\mathbf{k}_i|$)

2.AXIS: choice of angle between $\mathbf{k}_i$ and $\mathbf{k}_f$

First side of Scattering Triangle and the Scattering Angle are now defined

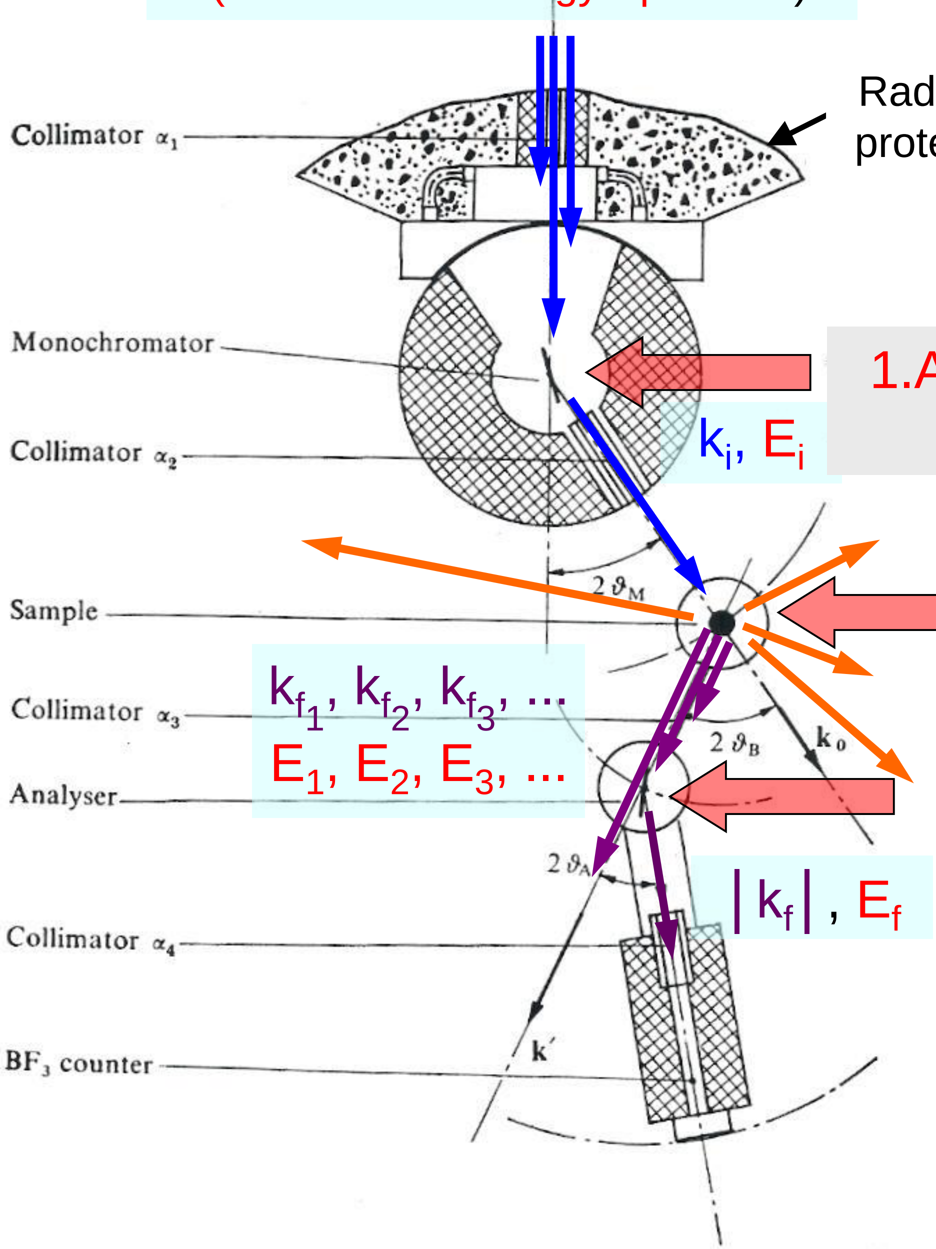
$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$



Neutron source

(Continuous Energy Spectrum)

THREE AXES SPECTROMETER FOR MEASURING $S(\mathbf{Q}, \omega)$



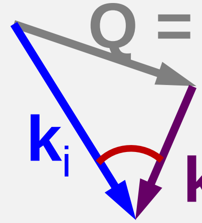
1.AXIS: choice of incident energy E_i $(\rightarrow |\mathbf{k}_i|)$

2.AXIS: choice of angle between $\mathbf{k}_i$ and $\mathbf{k}_f$

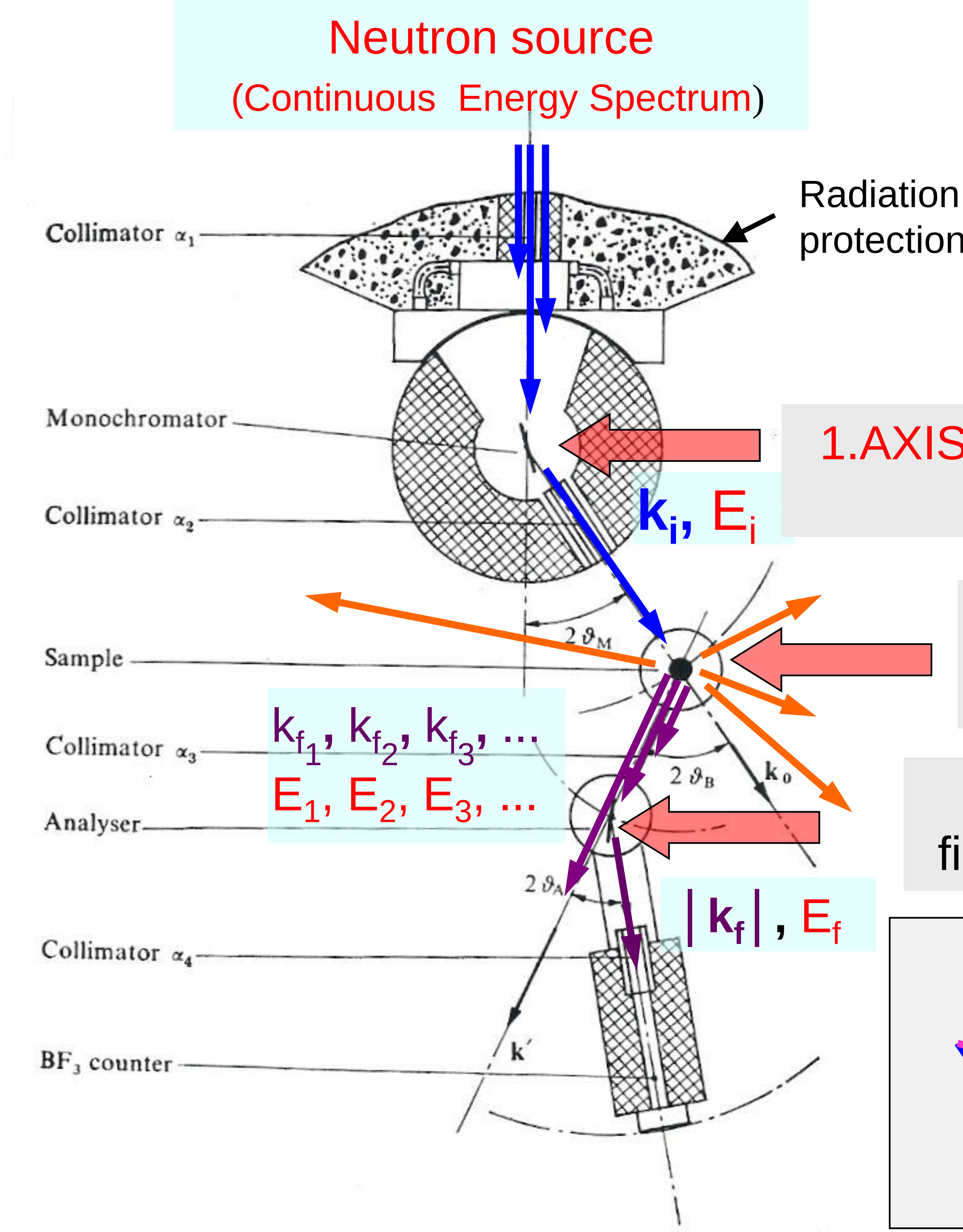
3.AXIS: choice of final energy E_f $(\rightarrow |\mathbf{k}_f|)$

Second side of Scattering Triangle is now defined

$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$



THREE AXES SPECTROMETER FOR MEASURING $S(\mathbf{Q}, \omega)$



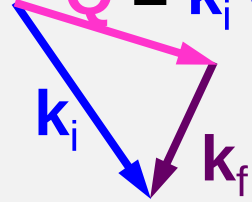
1.AXIS: choice of incident energy E_i ($\rightarrow |\mathbf{k}_i|$)

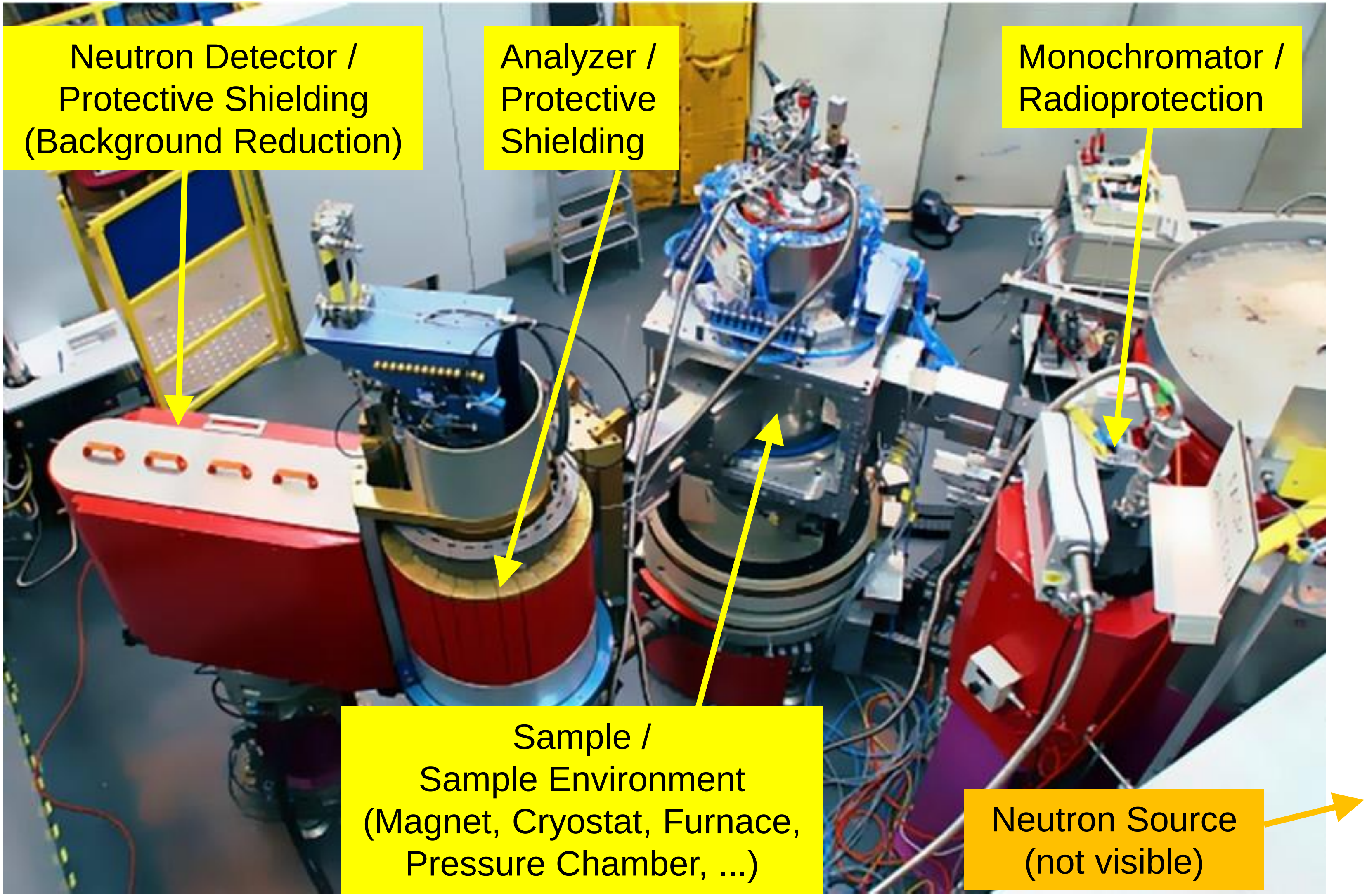
2.AXIS: choice of angle between $\mathbf{k}_i$ and $\mathbf{k}_f$

3.AXIS: choice of final energy E_f ($\rightarrow |\mathbf{k}_f|$)

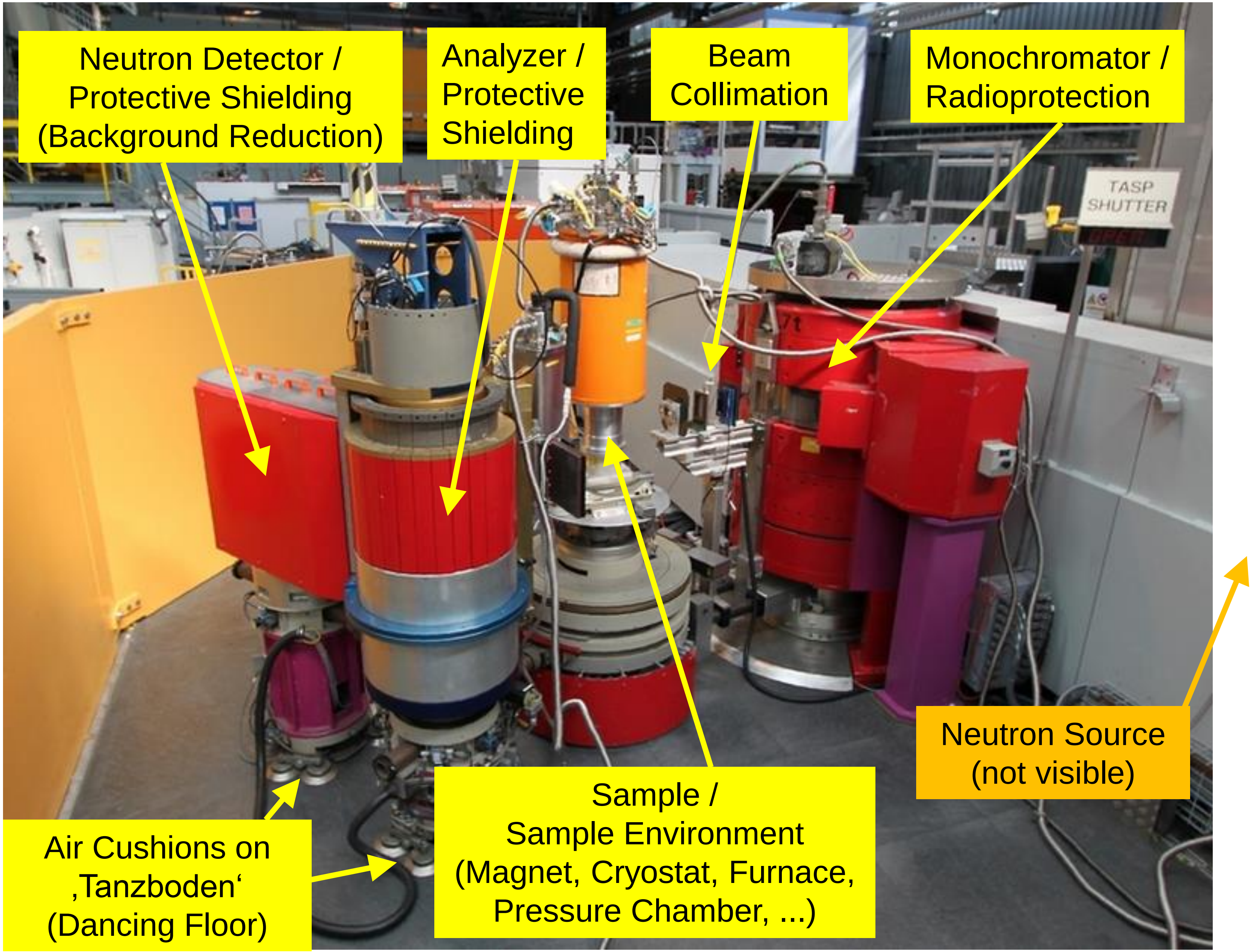
SCATTERING TRIANGLE

$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$





Three Axes Spectrometer TASP / SINQ



Three Axes Spectrometer TASP / SINQ

Most important advantage of triple axis spectrometers (TAS) over other instruments:

Access to S (**Q**, ω) for **arbitrary** combinations of **Q** and ω

Most of the basic elements of TAS were developed already 1950-60 by B.Brockhouse (Nobel prize 1994) and his group at the research reactor at Chalk River (Canada).

Today, typically 10-20% of the instruments at sources providing a continuous flux of neutrons (mostly nuclear reactors) are TAS.

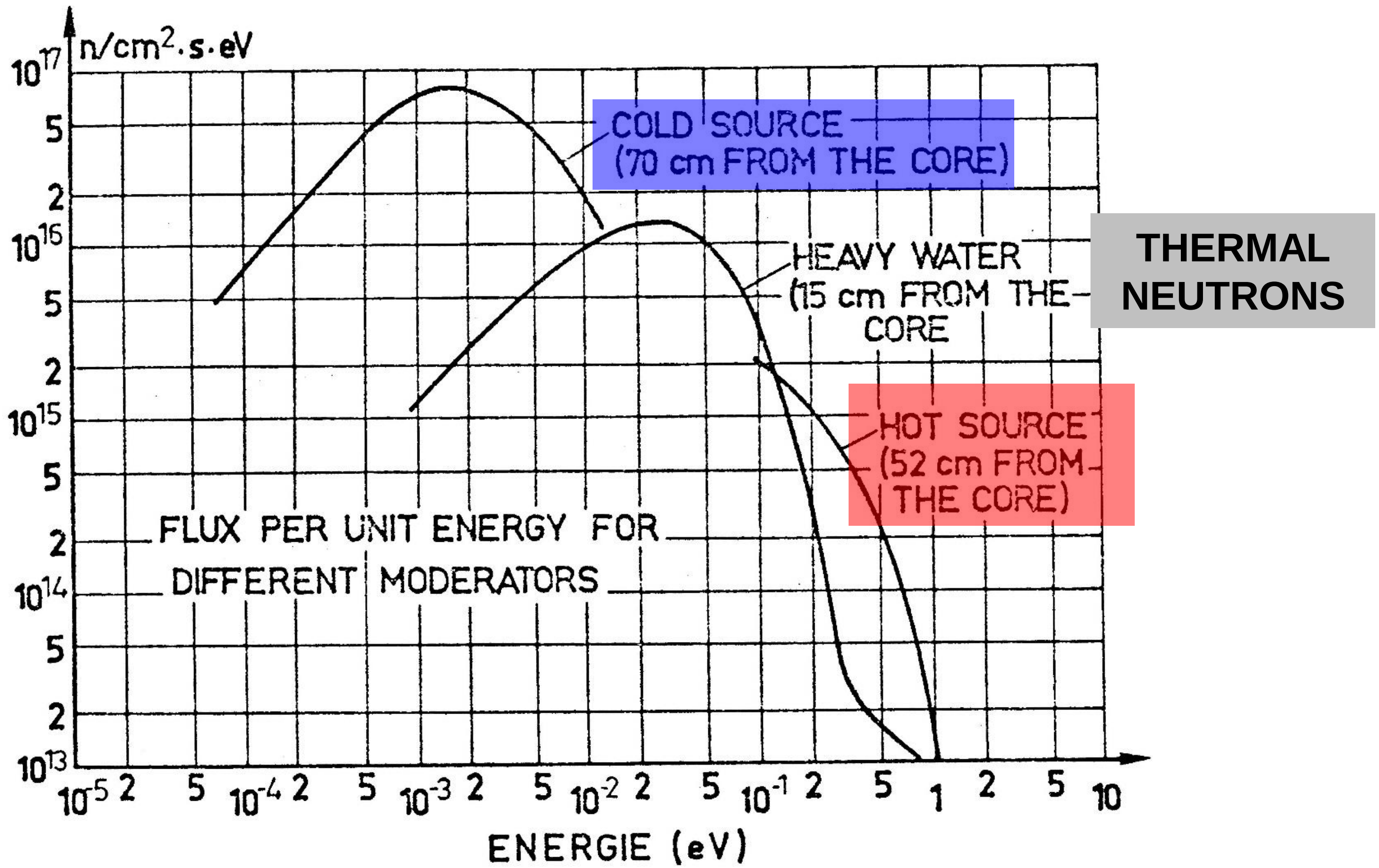
At pulsed sources (essentially spallation sources) Time-of-Flight techniques (TOF) are more appropriate.

CURRENT TECHNICAL DEVELOPMENTS with RELEVANCE FOR TAS

(not covered in this presentation)

- Computer simulation of neutron sources, neutron transport and instrument design already during planning (optimisation of use of produced neutrons)
- Focussing devices (higher neutron flux on samples)
- Remote control of experiments including sample environment (cryogenics, furnaces, pressure, magnetic field, ...)
- Multi-analyser arrangements (higher data acquistion rates)
- Computer simulation of experiments during planning and in real-time taking into account both instrument and sample
- Multi-analyser arrangements
- Event-based data acquisition systems

ILL Grenoble 58 MW



Neutron flux as a function of energy

Thermal spectrum

Cold and Hot source

>> Cold, Thermal, Hot Instruments

Inelastic scattering processes can bring about an energy loss or an energy gain of the scattered neutron. In the first case energy is transferred from the neutron to the sample, in the second case from the sample to the neutron.

The **Q**-ω range accessible in a neutron scattering experiment (more precisely: the accessible range of momentum transfer ħ**Q** and of energy transfer ΔE according to the creation or annihilation of an excitation ħω follows from the following consideration:

Energy-momentum relation: $\Delta E = \hbar\omega = \hbar^2(k_i^2 - k_f^2)/(2m)$

For the quantities k_i and k_f this yields

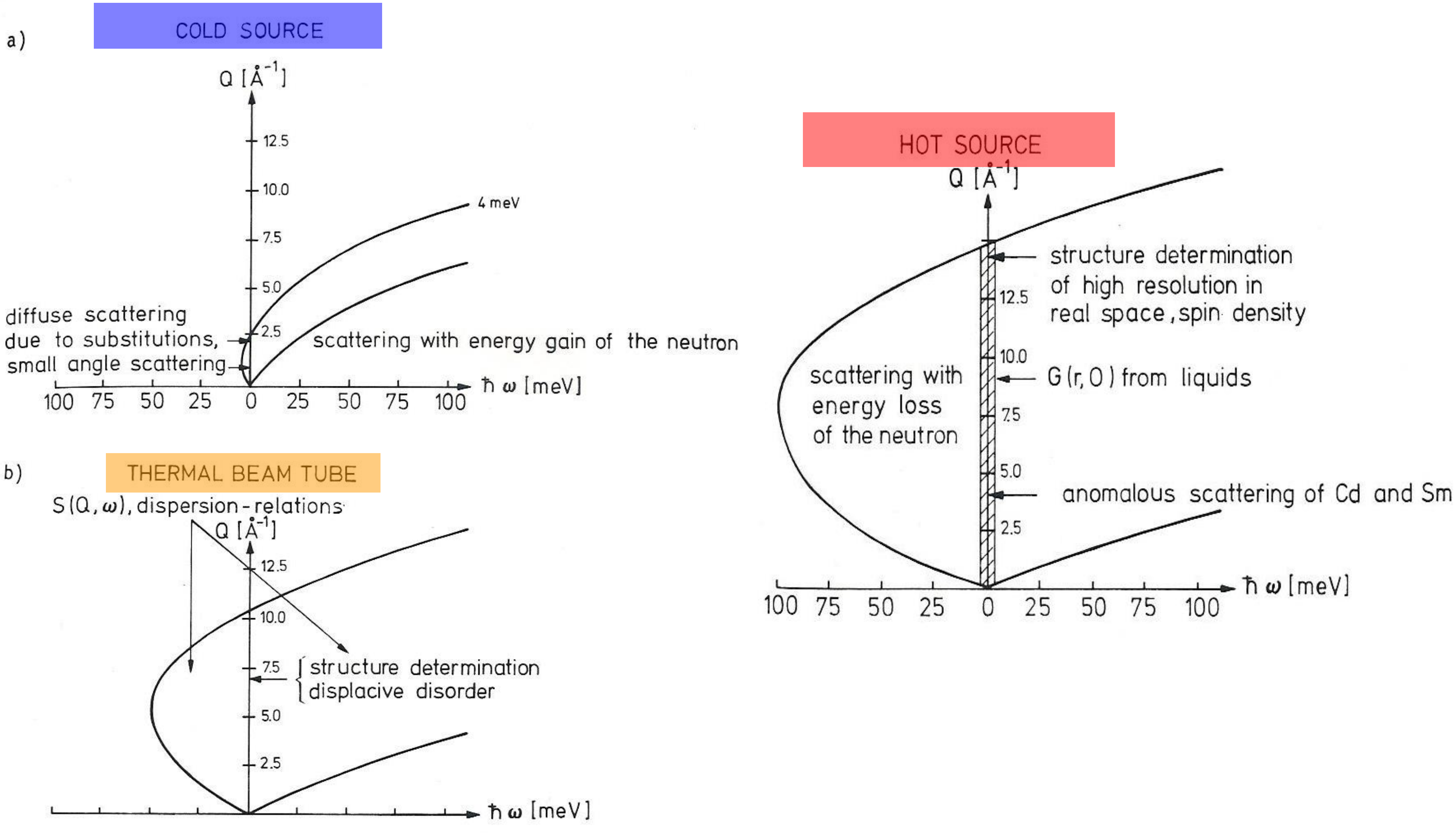
$$k_i > k_f: \quad k_f = \sqrt{(k_i^2 - 2m\omega / \hbar)}$$

$$k_i < k_f: \quad k_f = \sqrt{(k_i^2 + 2m\omega / \hbar)}$$

In addition the following inequality must hold for k_i and k_f :

$$k_i - k_f \leq |\mathbf{k_i} - \mathbf{k_f}| \leq k_i + k_f$$

By these conditions the accessible **Q** - ω range is completely defined.



Accessible energy and momentum range for experiments with (a) cold, (b) thermal and (c) hot neutrons

WHERE DO PHONONS PLAY A ROLE ?

In the Harmonic approximation the knowledge of the dispersion relations, eigenvectors and density of states permits to calculate macroscopic quantities such as

- lattice specific heat
- Debye temperature
- elastic moduli

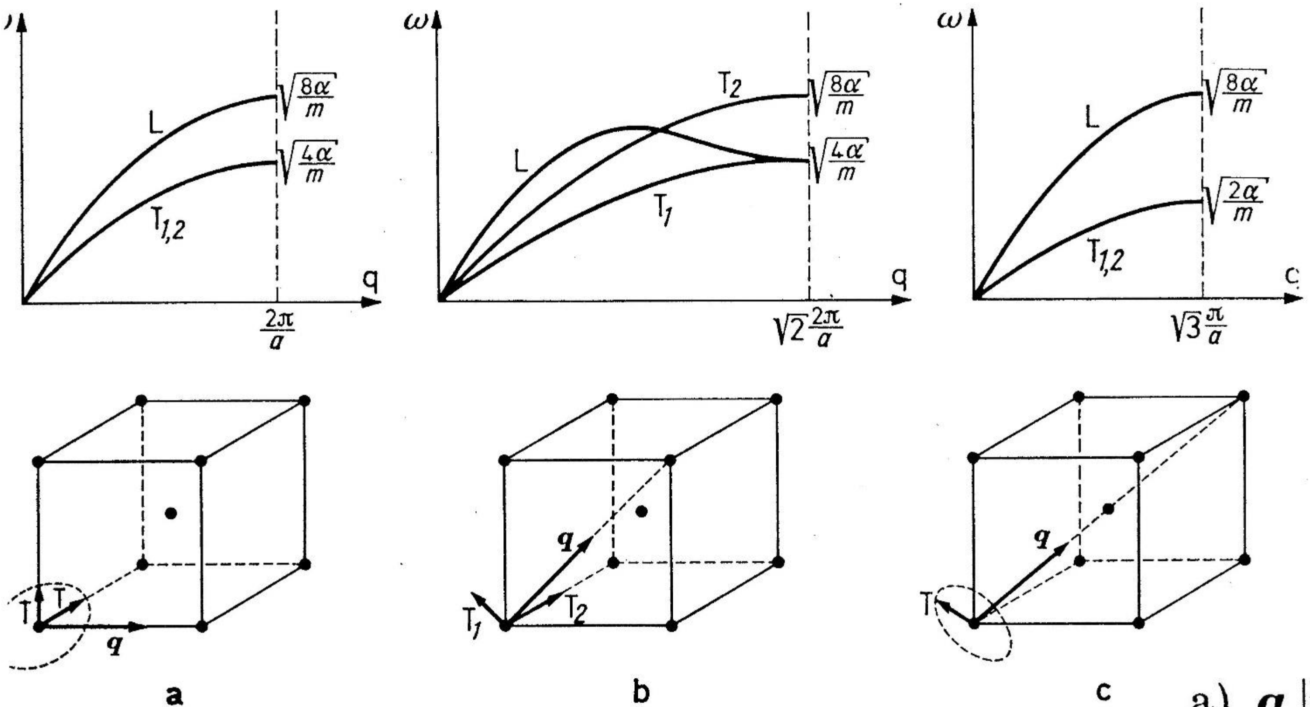
The study of Anharmonic effects reflected by phonon energy shifts and finite linewidths contributes to our understanding of macroscopic properties such as

- heat conduction
- thermal expansion.

On the microscopic level it provides information on, e.g.,

- higher terms of the interatomic potential
- the mechanisms underlying various phase transitions
- superconductivity
- phonon scattering processes

MODEL OF DISPERSION RELATIONS IN SIMPLE CUBIC LATTICE (1 ATOM)



RECIPROCAL SPACE

REAL SPACE

a) $\boldsymbol{q} \parallel [100]$, $\{q_{x^*}, q_{y^*}, q_{z^*}\} = q\,\{1, 0, 0\}$, $q \leqq 2\pi/a$,

b) $\boldsymbol{q} \parallel [101]$, $\{q_{x^*}, q_{y^*}, q_{z^*}\} = q\{1, 0, 1\}$, $q \leqq \sqrt{2} \cdot 2\pi/a$,

c) $\boldsymbol{q} \parallel [111]$, $\{q_{x^*}, q_{y^*}, q_{z^*}\} = q\{1, 1, 1,\}$, $q \leqq \sqrt{3}/2 \cdot 2\pi/a$.

- General case: N atoms in unit cell
  → 3N phonon branches
- 'Pure' polarization (longitudinal, transverse)
  along directions of high symmetry
- Degeneracy possible

Example: Phonon Dispersion Curves for KBr
(Model calculation using simple Rigid Ion Model)

<u>Long wavelength limit:</u>

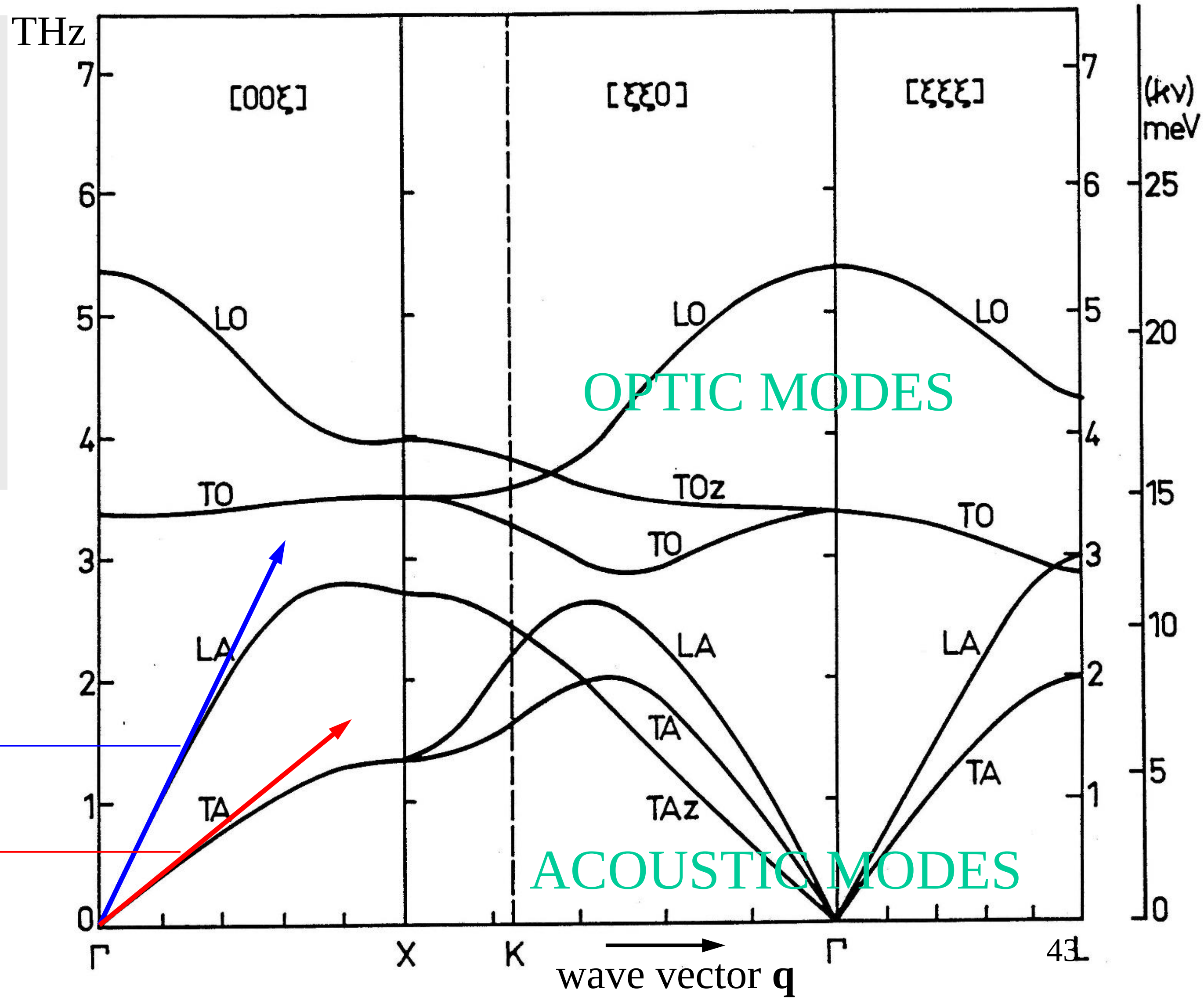
slopes of acoustic modes give sound velocities

→ elastic moduli

$v_{sound} = \omega/q = \nu \cdot \lambda$

$\rho\, v_{long}^{2} = c_{11}$

$\rho\, v_{trans}^{2} = c_{44}$



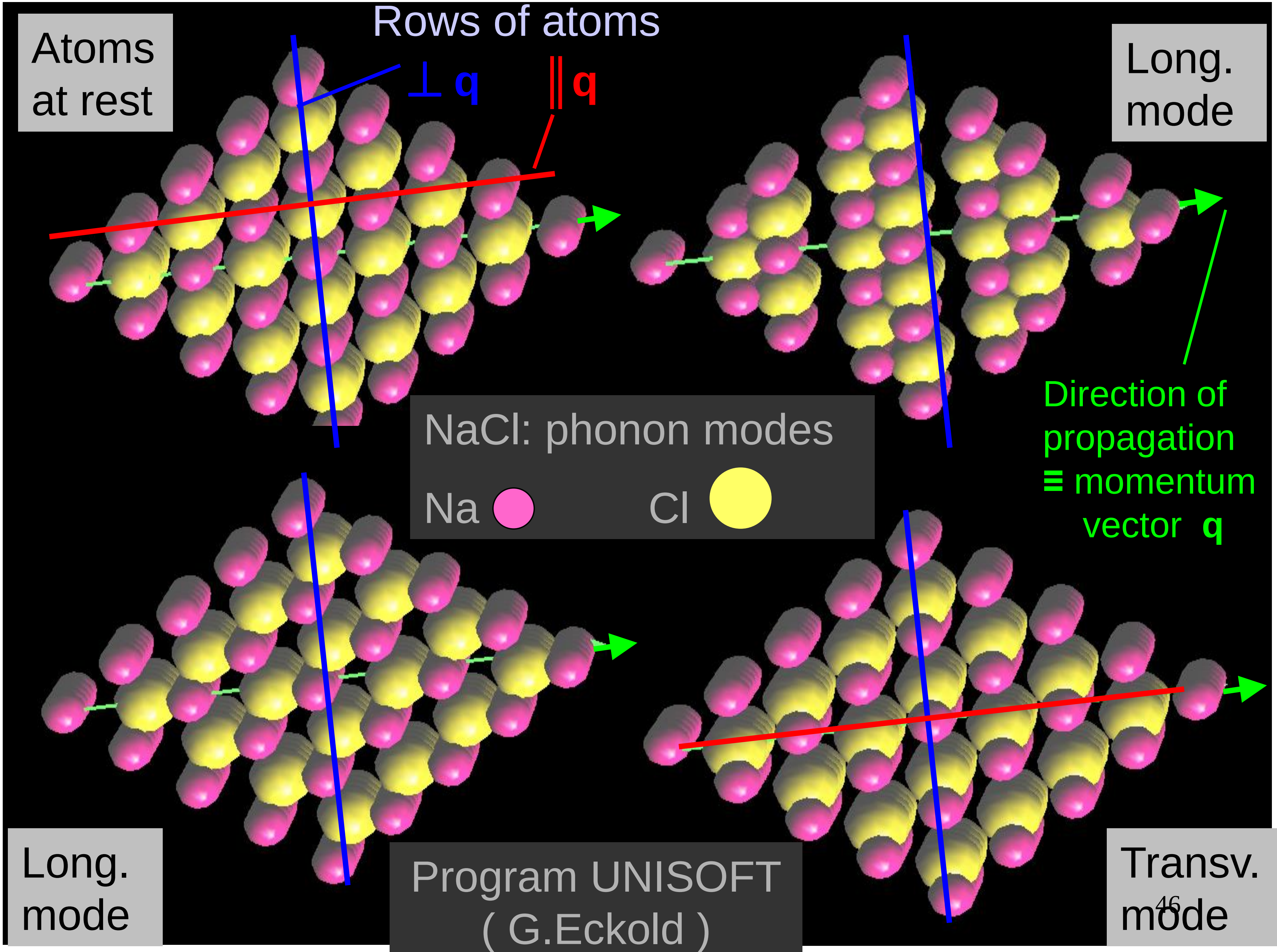
Dynamic Structure Factor for coherent one-phonon scattering

$$S^{\pm}_{coh}\left(\vec{Q},\ \omega_s\right) \sim \left| \sum_{j=1}^{r} b_{coh,j} \exp\left[-\frac{1}{2} Q^2 \langle u_j^{\,2} \rangle\right] \frac{\vec{Q} \cdot \vec{e}_{s,j}}{\sqrt{M_j}} \exp\left[-i\,\vec{Q} \cdot \vec{R}_j\right] \right|^2 \times \frac{1}{\omega_s} \left(\frac{1}{\exp\left[\frac{\hbar\,\omega_s}{k\,T}\right] - 1} + \frac{1}{2} \pm \frac{1}{2} \right) \delta\left(\varepsilon \pm \hbar\,\omega_s\right)$$

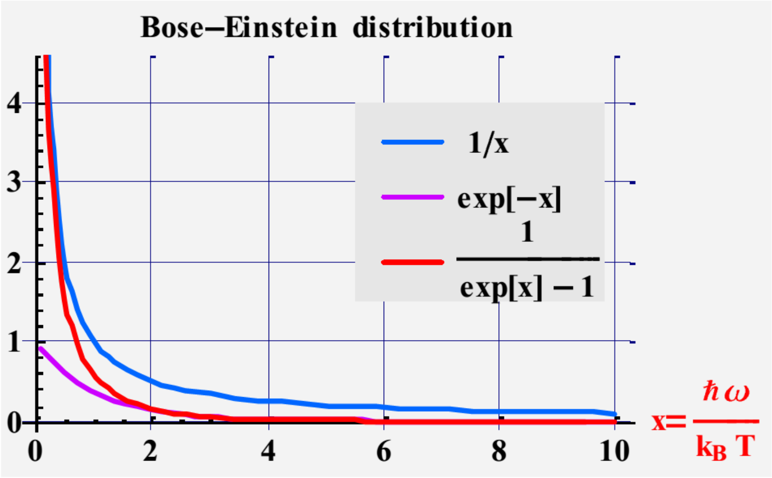
- summation over all atoms: $\sum_{j=1}^{r}$
- coherent scattering length: $b_{coh,j}$
- Debye-Waller factor: $\exp\left[-\frac{1}{2} Q^2 \langle u_j^{\,2} \rangle\right]$
- root mean square displacement: $\langle u_j^{\,2} \rangle$
- momentum transfer: $\vec{Q}$
- phonon polarisation vector: $\vec{e}_{s,j}$
- atomic mass: M_j
- phase factor ≠ Bragg diffraction !: $\exp\left[-i\,\vec{Q} \cdot \vec{R}_j\right]$
- atomic position: $\vec{R}_j$
- phonon frequency: ω_s
- Bose distribution function for phonons: $\frac{1}{\exp\left[\frac{\hbar\,\omega_s}{k\,T}\right] - 1}$
- Phonon creation (+) or annihilation (-): $\pm \frac{1}{2}$
- Energy conservation: $\delta\left(\varepsilon \pm \hbar\,\omega_s\right)$

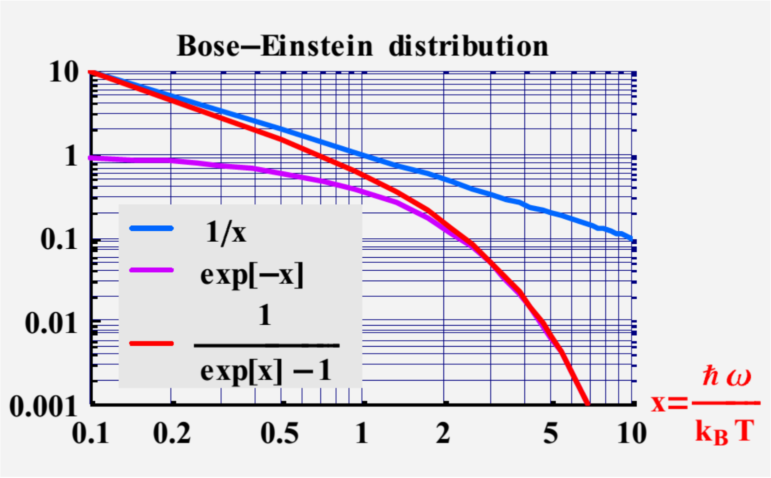
Simulation of lattice vibrations using the program UNISOFT (Götz Eckold, Uni Göttingen)

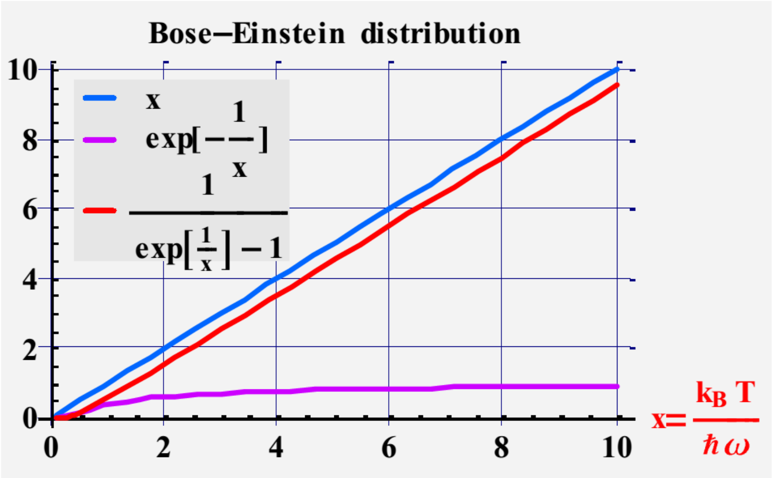
- Polarisation and eigenvectors
- Phonon dispersion curves
- Dynamical structure factors
- Density of states
- Specific heat

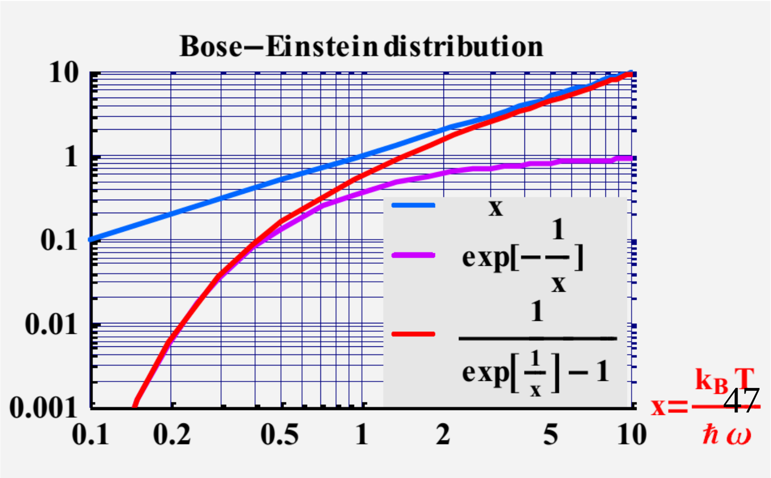

Rows of atoms
Atoms at rest
⊥ q
∥ q
Long. mode
Direction of propagation ≡ momentum vector q
NaCl: phonon modes
Na
Cl
Long. mode
Program UNISOFT (G.Eckold)
Transv. mode

- The average number of phonons with frequency ω excited at a temperature T is given by the ***Bose-Einstein distribution function***
- For small x, i.e. for small phonon frequencies ω and/or for large temperatures T, the function approaches 1/x
- For large x, i.e. for large phonon frequencies ω and/or for small temperatures T, the function approaches exp[-x]
- The scattering cross section for phonon annihilation (i.e. the neutron gains energy in the scattering process) is proportional to the number of excitedphonons, i.e. the Bose-Einstein distribution
- The scattering cross section for phonon creation (i.e. the neutron loses energy in the scattering process) is proportional to the number of excited phonons, i.e. the Bose-Einstein distribution, multiplied by exp(ħω/kT)









OVERVIEW

- Introduction to Inelastic Neutron Scattering (INS)
- Relation to other methods
- Three Axes (Triple Axis) Spectrometer
- Examples (taken from experiments)
- Formal and technical aspects

EXAMPLES

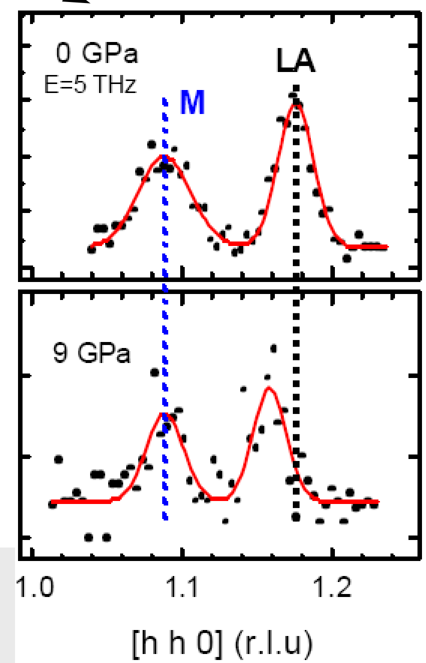
partly didactic / partly current research

1. PHONON / MAGNON DISPERSION in IRON under HIGH PRESSURE investigated with NEUTRONS …
2. … and SYNCHROTRON RADIATION
3. INTRAMOLECULAR MODES in C_{60}
4. ANHARMONICITY: MODE GRÜNEISEN PARAMETERS in RbBr
5. HYDROGEN in METALS: LOCAL MODES
6. SPIN EXCITATIONS in Fe-PNICTIDES
7. RATTLER MODES in THERMOELECTRIC MATERIALS
8. SPIN DYNAMICS in $BaFe_{1.85}Co_{0.15}As_2$
9. ELASTIC SCATTERING

PHONON / MAGNON DISPERSION IN IRON UNDER HIGH PRESSURE

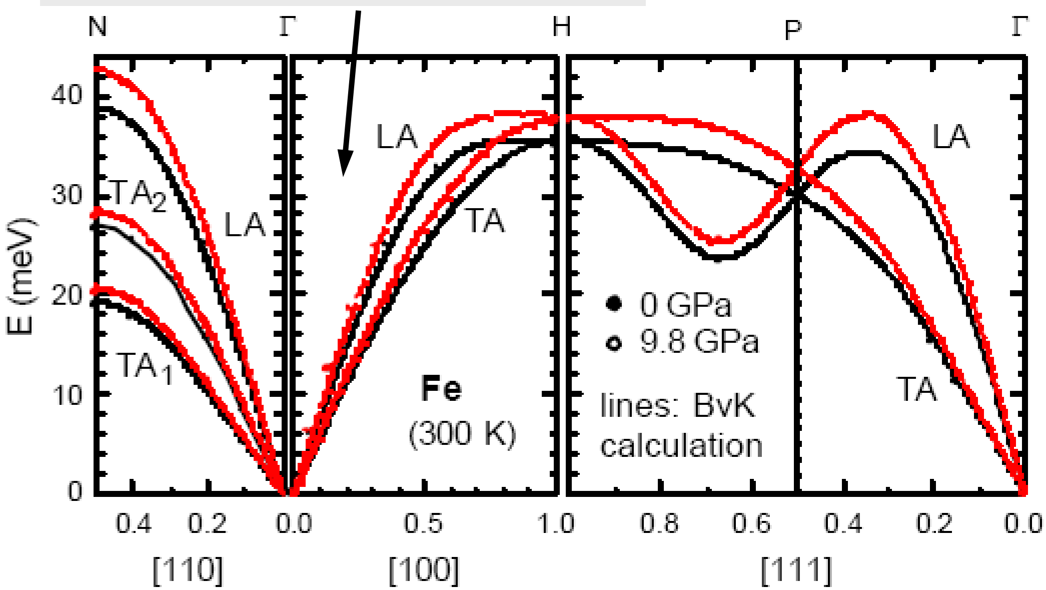
LLB: S.Klotz, M.Braden, PRL **85** (2000) 3209

Comparison of changes in magnon and phonon peaks with pressure

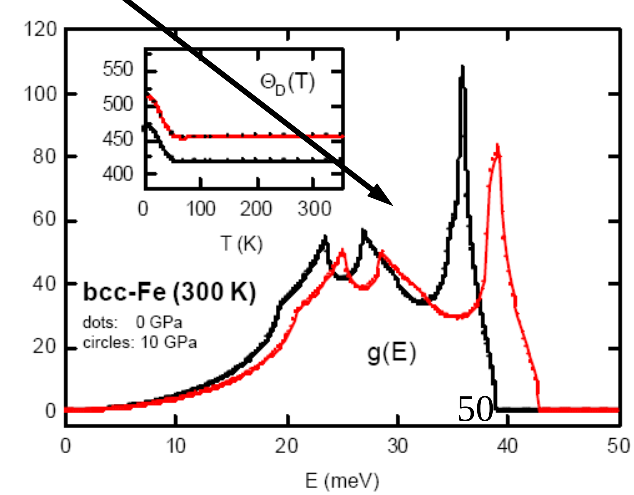


- <u>Interest:</u> iron under extreme conditions important for geophysical models of the earth
- Phase transition bcc → hcp at ~ 11GPa
  Are there precursor phenomena like in other *bcc* metals ?
- Two surprises: -- stiffening of <u>all</u> phonon branches
  -- magnon dispersion does not change

phonon dispersion relations



phonon density of states



PHONON DISPERSION IN IRON UNDER HIGH PRESSURE with SYNCHROTRON RADIATION (for comparison)

ESRF: G.Fiquet et al., Science **291** (2001) 468

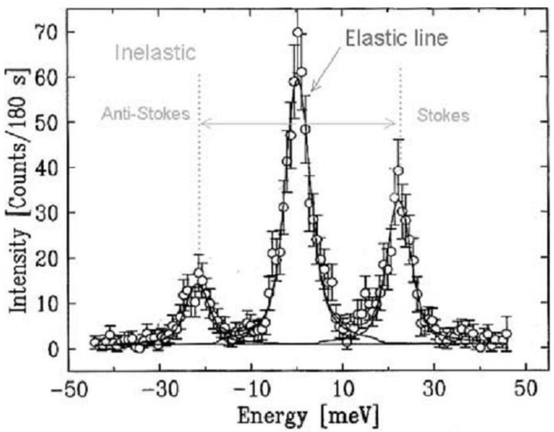
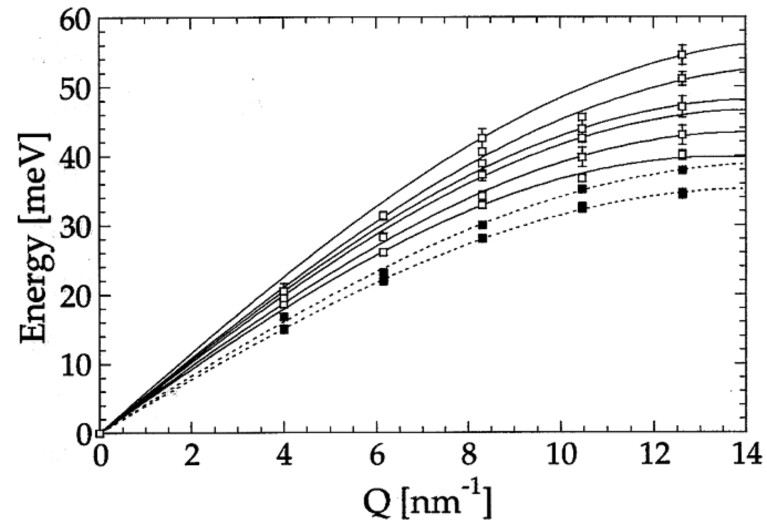
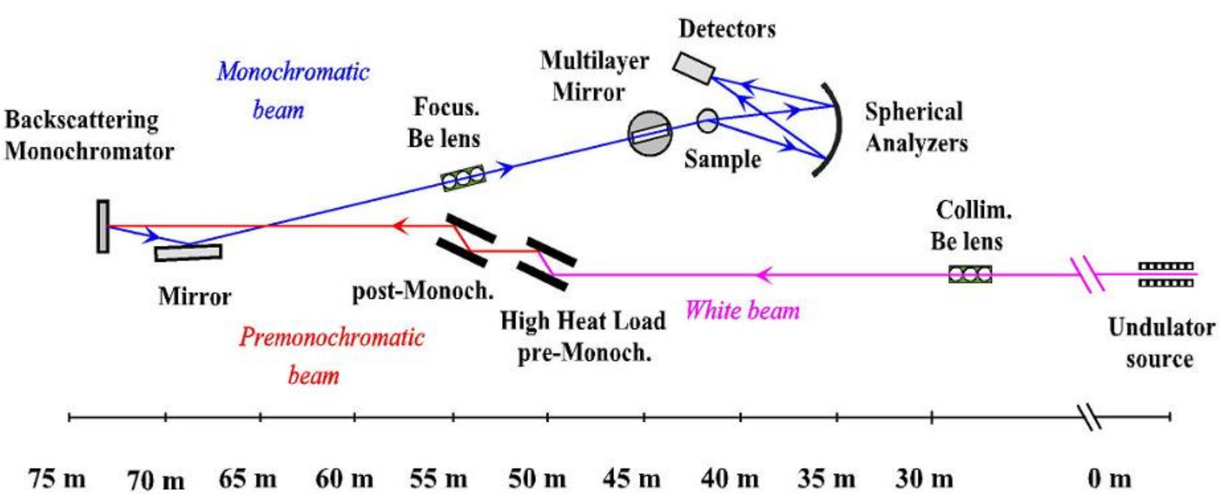


Fig. 3. Example of an inelastic X-ray scattering pattern obtained from a bcc iron foil at ambient conditions using the (8 8 8) monochromator reflection, providing an energy resolution of 5.5 meV. Scanning time is 5 h. $Q = 6.16\,\mathrm{nm}^{-1}$. The experimental data (open circles) are plotted with their error bars along with the corresponding fits.

Fig. 3. LA phonon dispersion curves of iron at different pressures. Lines represent the results of the fit of Eq. 1. Solid symbols and dashed lines stand for measurements carried out on the bcc phase at 0.2 and 7 GPa. Open symbols and solid lines correspond to the pattern recorded on the hcp structure of iron at 19, 28, 45, 55, 64, and 110 GPa from bottom to top, respectively. The energy position of the phonons could be determined within 3% (error bars).

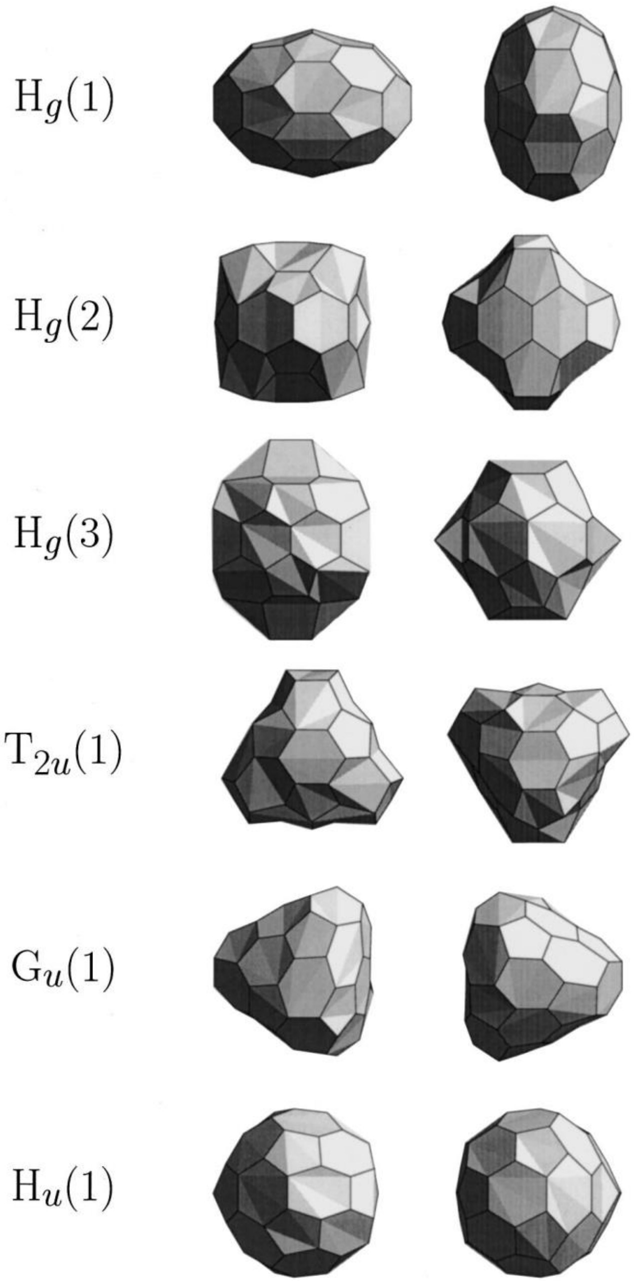


ESRF, ID28
Inelastic Beamline



Monochromator: Si (n,n,n)

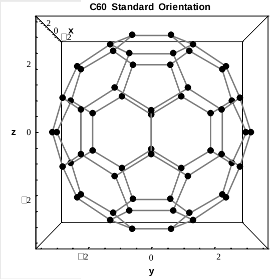
Bragg angle 89.98°,
T control in the mK range,
E ~ 13 – 25 keV
ΔE/E ~ 10^{-7} – 10^{-8},
Photon flux after Mono
10^{9} – 10^{11} / s



INTRAMOLECULAR MODES

C_{60} molecule:

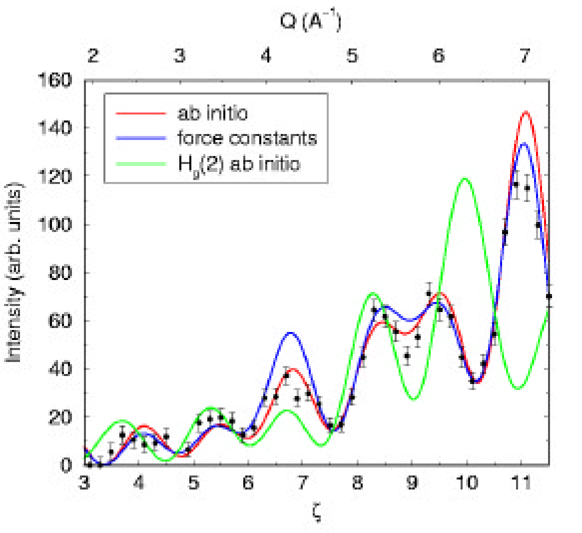
174 intramolecular modes, icosahedral symmetry
→ 46 distinct modes (33-195 meV)
10 Raman active
4 Infrared active
32 'silent' modes



solid state: weak van der Waals interaction
→ internal modes only weakly influenced

- study of frequencies and eigenvectors (displacement patterns)
- comparison with 10 different models (ab initio, phenomenological)
- single crystal + powder

LLB: R.Heid, L.Pintschovius, PRB **56** (1997) 5925

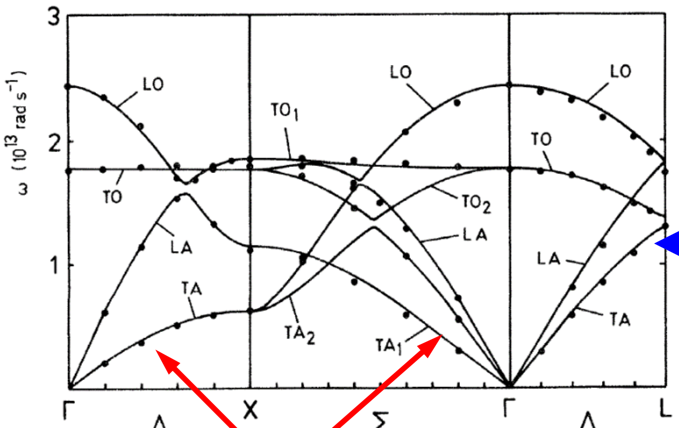


single crystal data
E=33meV, $H_g(1)$mode

Anharmonicity: mode Grüneisen parameters
Anomalous thermal expansion in RbBr

<u>Interest:</u> explain macroscopic behaviour in terms of microscopic properties

- Thermal expansion is due to the anharmonicity of the interatomic potential. At low temperatures only low-energy phonons are excited.
- Therefore the anharmonic potential contributes to thermal expansion only along those directions which correspond to displacements induced by low-frequency modes !



phonon dispersion curves

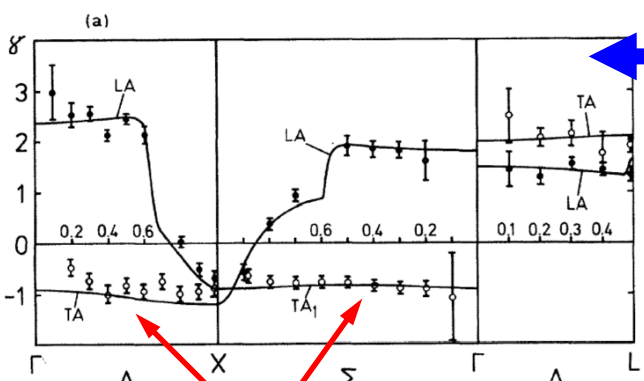
Dominating excitations at low T

determine frequency changes under hydrostatic pressure

mode Grüneisen parameters

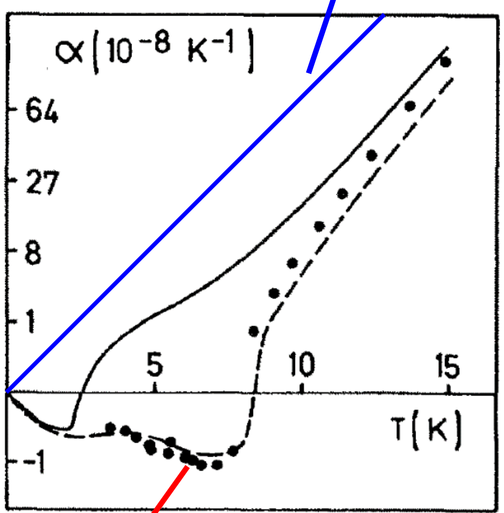
$$\gamma\begin{bmatrix}\vec{q}\\ j\end{bmatrix} = -\left[\frac{d\ln\omega(\vec{q},j)}{d\ln V}\right]_T$$

(dimensionless quantities)



branches with negative mode-γ

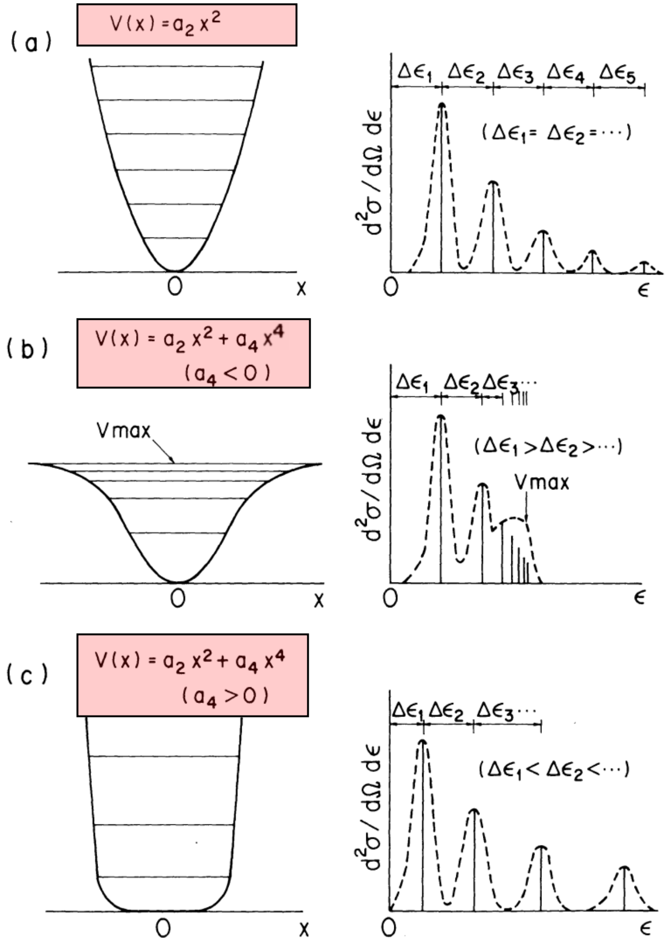
usual behaviour at low T:
Thermal expansion $\alpha \sim T^3$



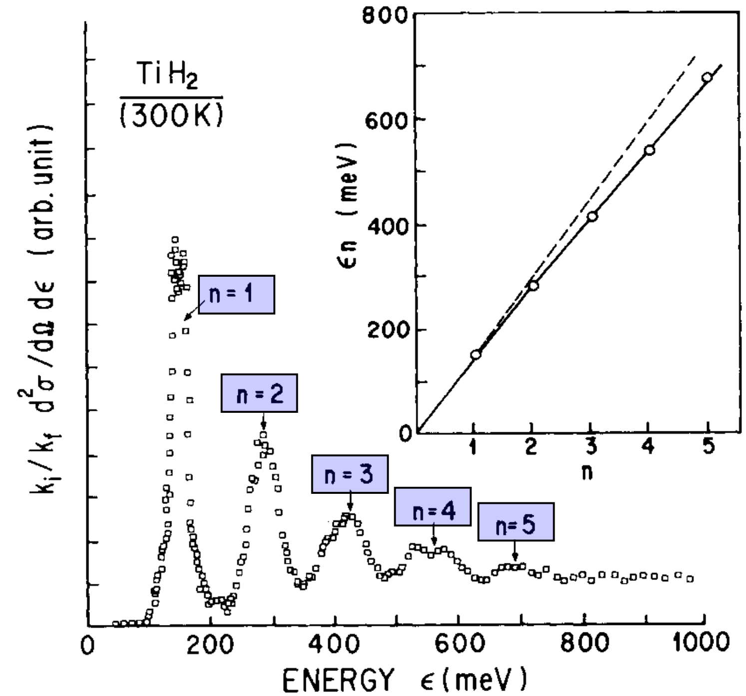
Anomaly: negative thermal expansion

Risø: G.Ernst et al., PRB 29 (1984) 5805

Hydrogen in metals - Local Modes



Hydrogen atoms (protons) occupy interstitial sites



Three types of hydrogen potential

(a) Harmonic potential

(b) Trumpet-like potential

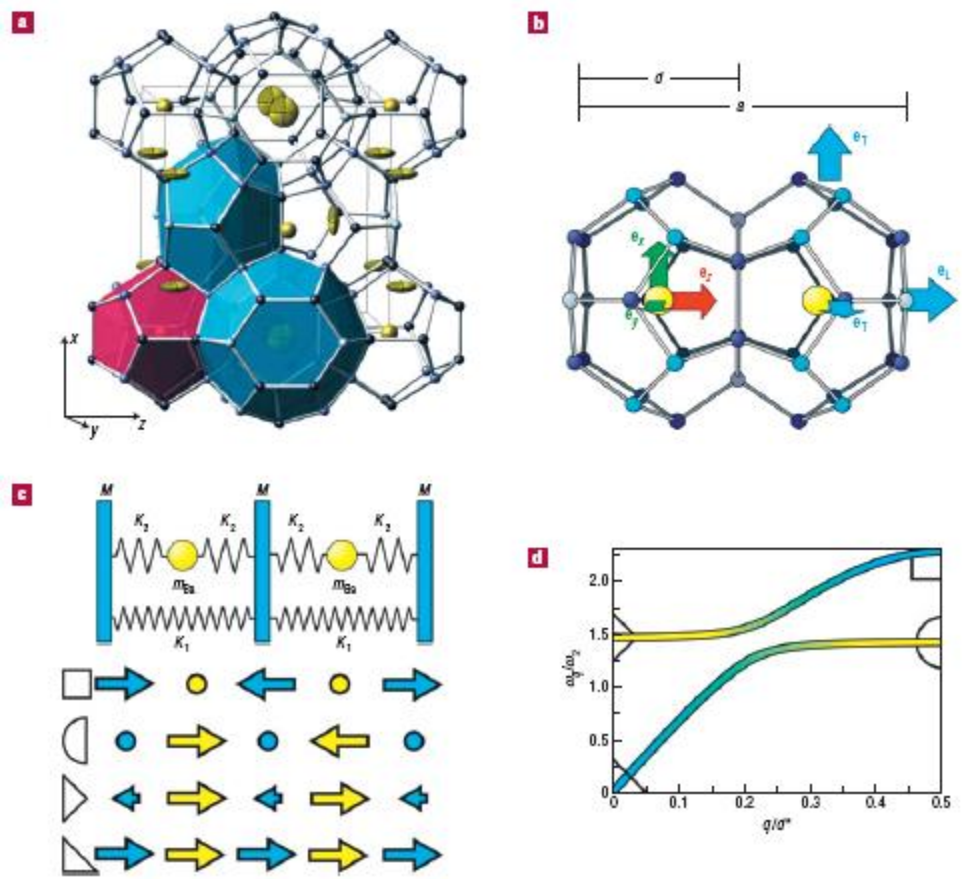
(c) Well-like potential

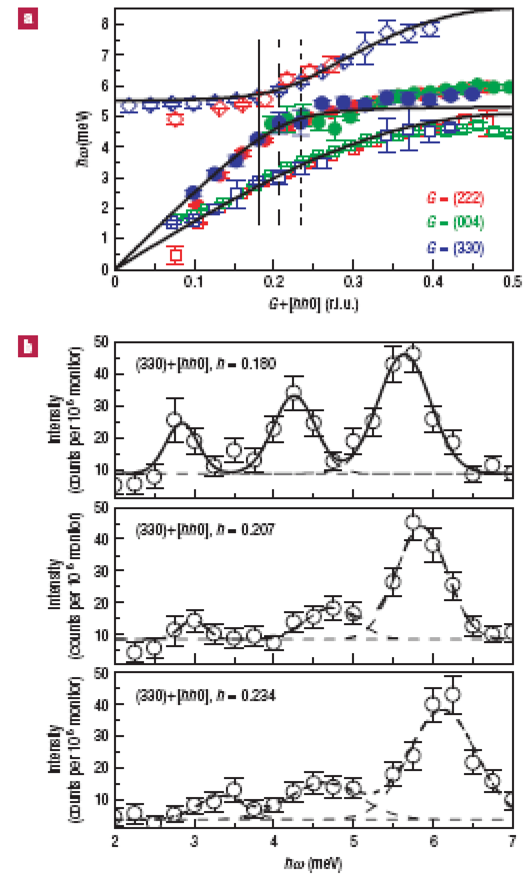
[S.Ikeda, J.Phys.Soc.Japan **65** (1987) 565]

Energy spectrum of local modes in TiH_2 (powder sample, **Time of Flight measurement !**)

Rattler Modes in Thermoelectric Materials

Interest: combination of high electrical and low thermal conductivity
Concepts: ‚phonon glass – electron crystal‘ , rattling modes in cage structures
Potential applications: use of waste-heat, novel refrigerators

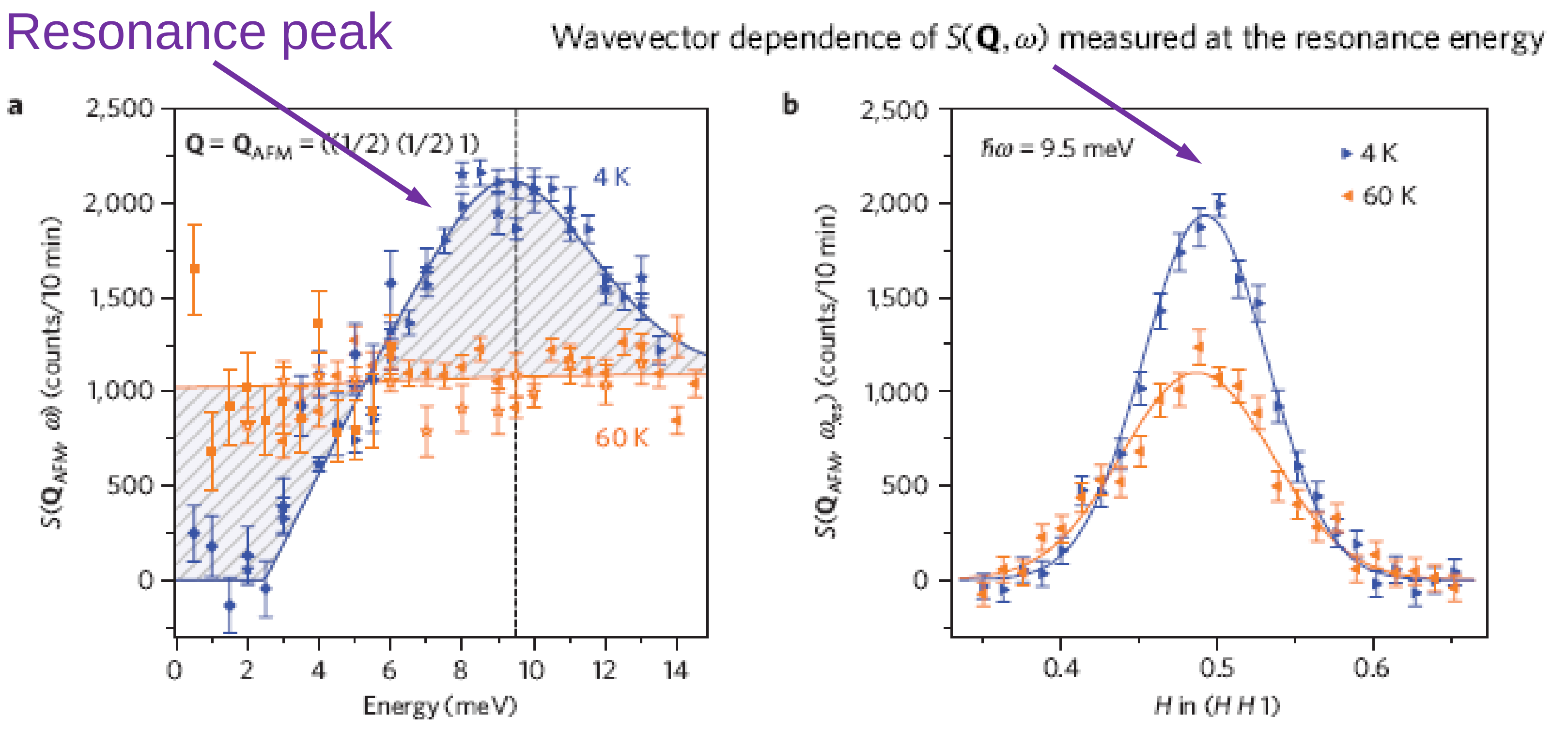




Avoided crossing of acoustic mode of cage structure and flat rattling mode of guest atom in $Ba_8Ga_{16}Ge_{30}$
[PSI: M.Christensen, Nature Materials **7** (2008) 811]

Spin Dynamics in $BaFe_{1.85}Co_{0.15}As_2$

Interest: understanding of pairing mechanism in unconventional (high T_c) superconductors. Existence of magnetically mediated Cooper pairing ?

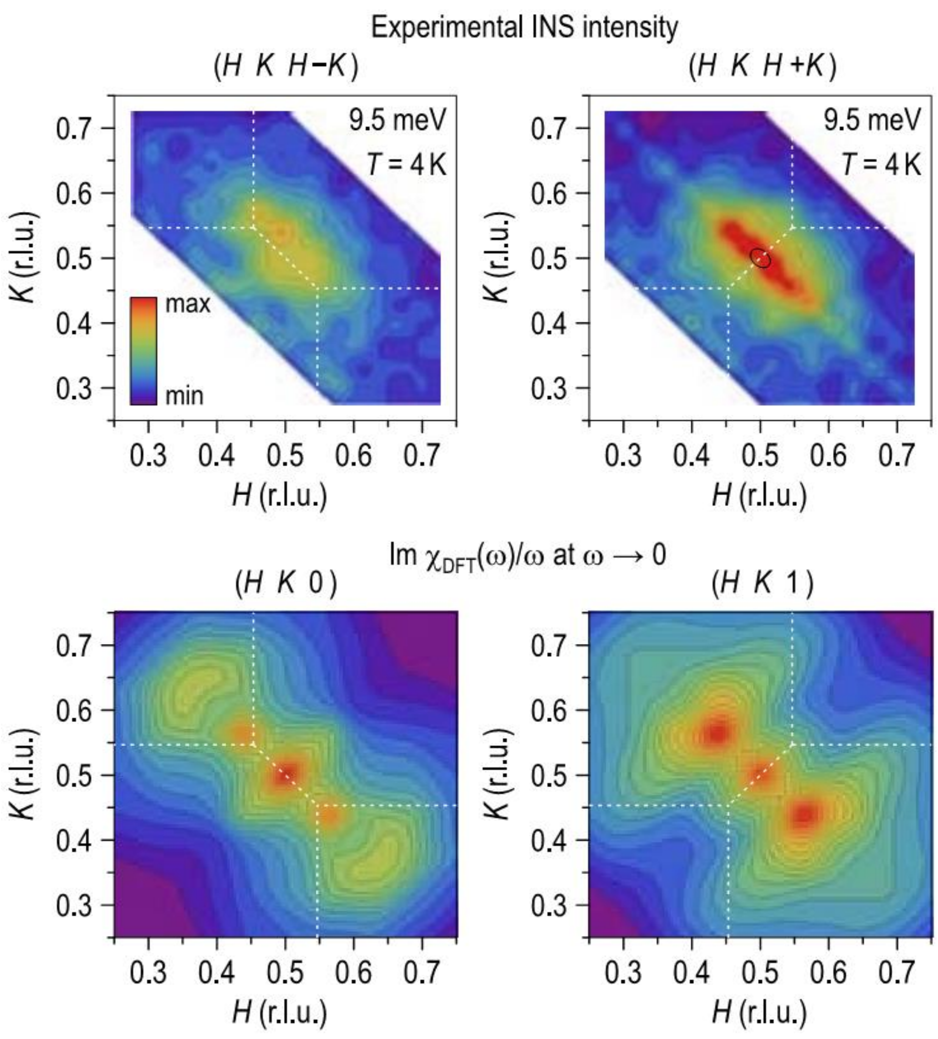


Spin excitations in the vicinity of the AFM wavevector Q_{AFM}, in the superconducting ($T = 4$ K) and the normal state ($T = 60$ K).

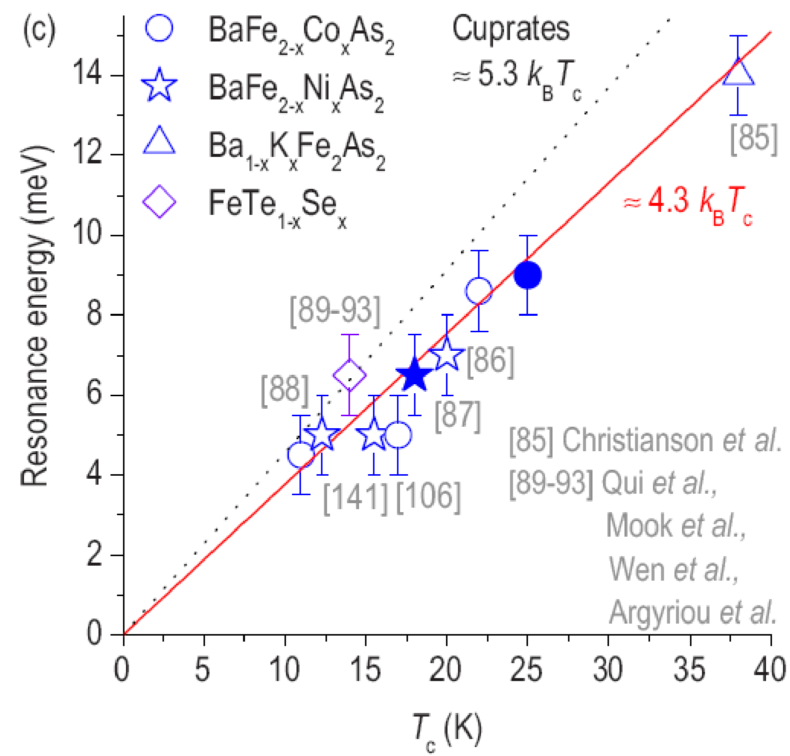
Important step towards theoretical understanding of superconductivity in iron arsenides [FRM2, LLB: D.S.Inosov, Nature Physics 2010]

Asymmetric spin-excitation spectra in Fe-pnictides

J.T.Park et al., Phys. Rev. B **82** (2010) 134503



The magnetic resonant mode is the most prominent signature of superconductivity in the spin excitation spectrum of several unconventional superconductors

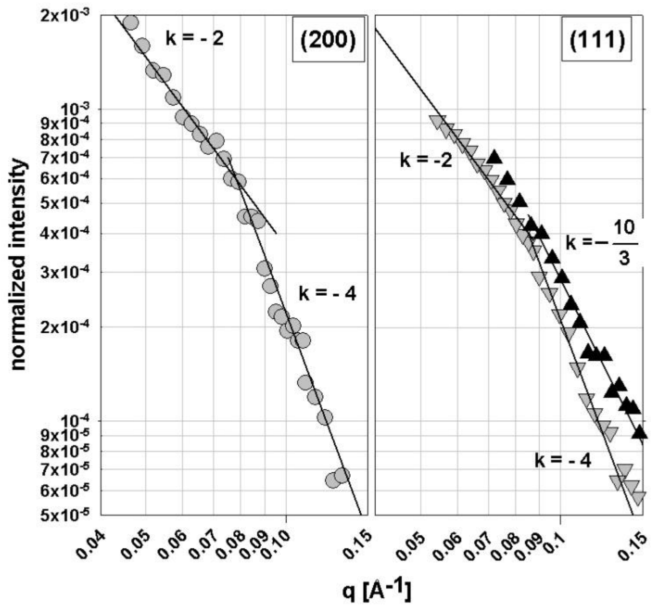


Resonance energy region: Experiment vs. results from DFT calculations

Resonance energy vs T_c for different Fe-based superconductors.

Distortion scattering in Li_2O

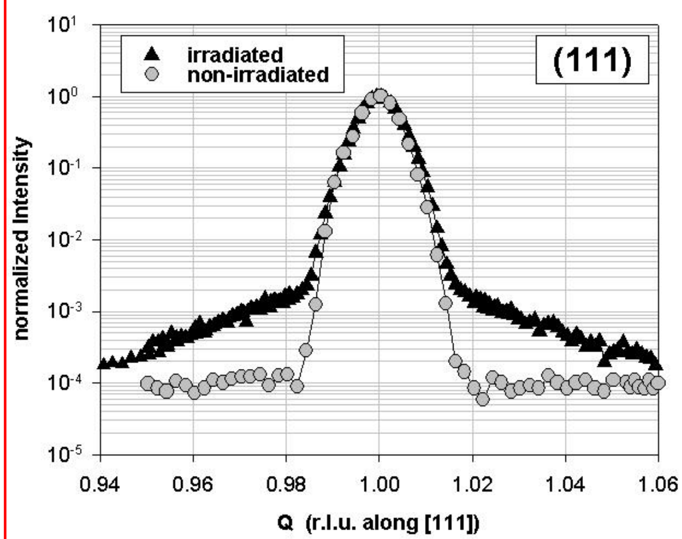
[G.Krexner et al., PRL **91** (2003) 135502 (LLB)]

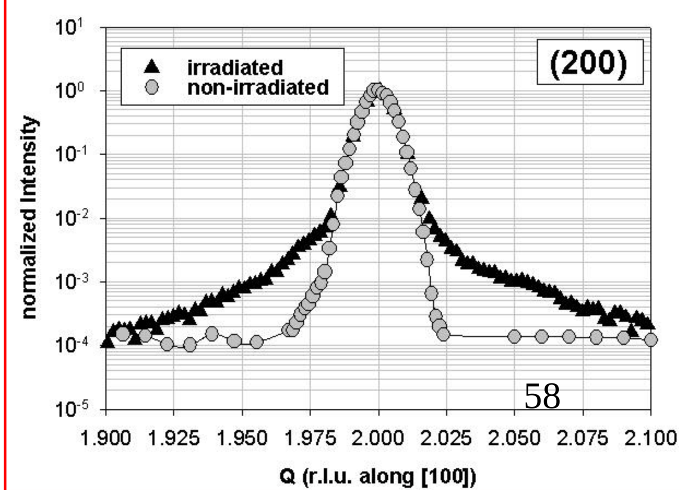


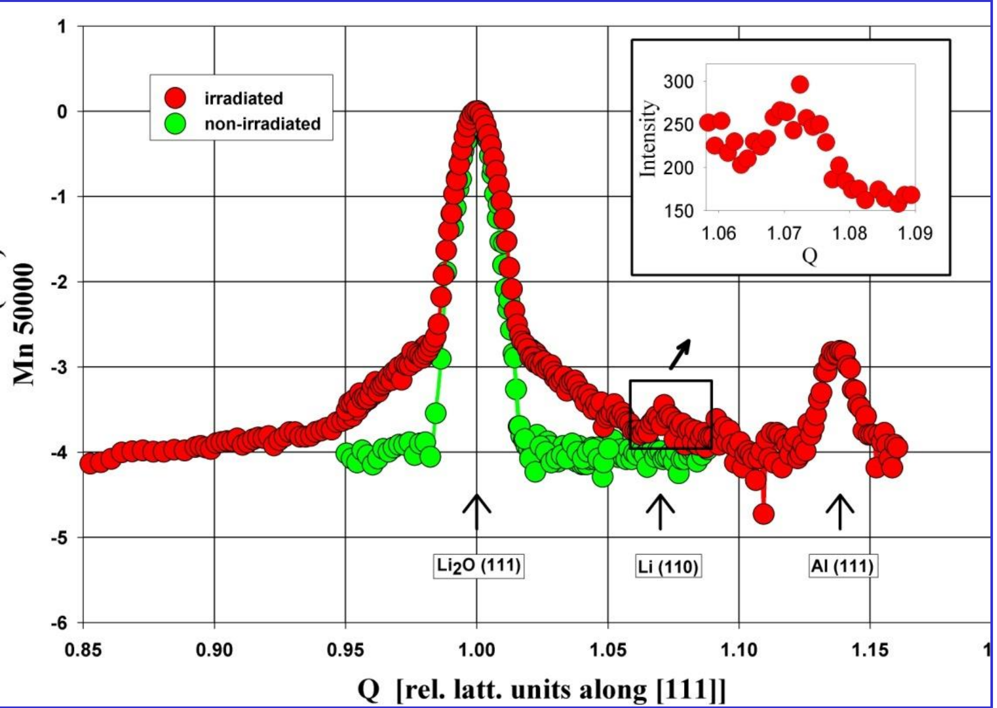
Formation of metallic Li colloids in irradiated samples (size < 10 nm) gives rise to distortion scattering

Requires excellent
Q resolution
and very
low background

→ TAS required







Triple axis spectrometers (in particular cold TAS) are **not only** used for the investigation of **inelastic scattering !**

Elastic scattering is studied **as well** if

- the discrimination of inelastic scattering is important
- good resolution and low background is required

Examples are

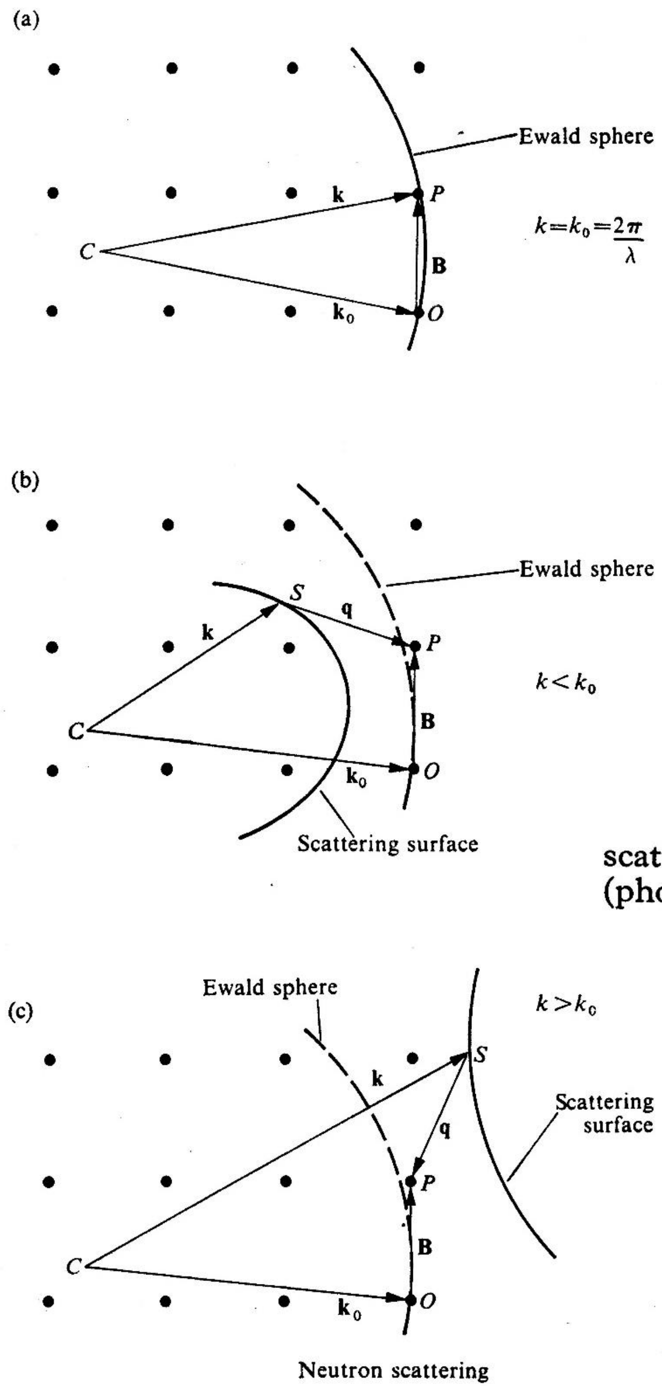
- diffuse scattering (e.g. due to defects)
- distortion scattering around Bragg peaks
- short – range order
- neutron holography (needs thermal or hot TAS)

O V E R V I E W

- Introduction to Inelastic Neutron Scattering (INS)
- Relation to other methods
- Three Axes (Triple Axis) Spectrometer
- Examples (taken from experiments)
- Formal and technical aspects

FORMAL and TECHNICAL ASPECTS

- The scattering geometry
- Resolution function
- Correlation functions (van Hove)
- Phonon density of states
- KBr : a detailed example



REPRESENTATION OF INELASTIC SCATTERING PROCESSES IN THE RECIPROCAL LATTICE

Reciprocal lattice diagrams representing (a) zero-order (Bragg) scattering of neutrons, (b) first-order scattering of neutrons with loss of energy (phonon emission), (c) first-order scattering of neutrons with gain of energy (phonon absorption) (after Willis 1969).

SCATTERING GEOMETRY FOR THE MEASUREMENT OF TRANSVERSE AND LONGITUDINAL PHONONS

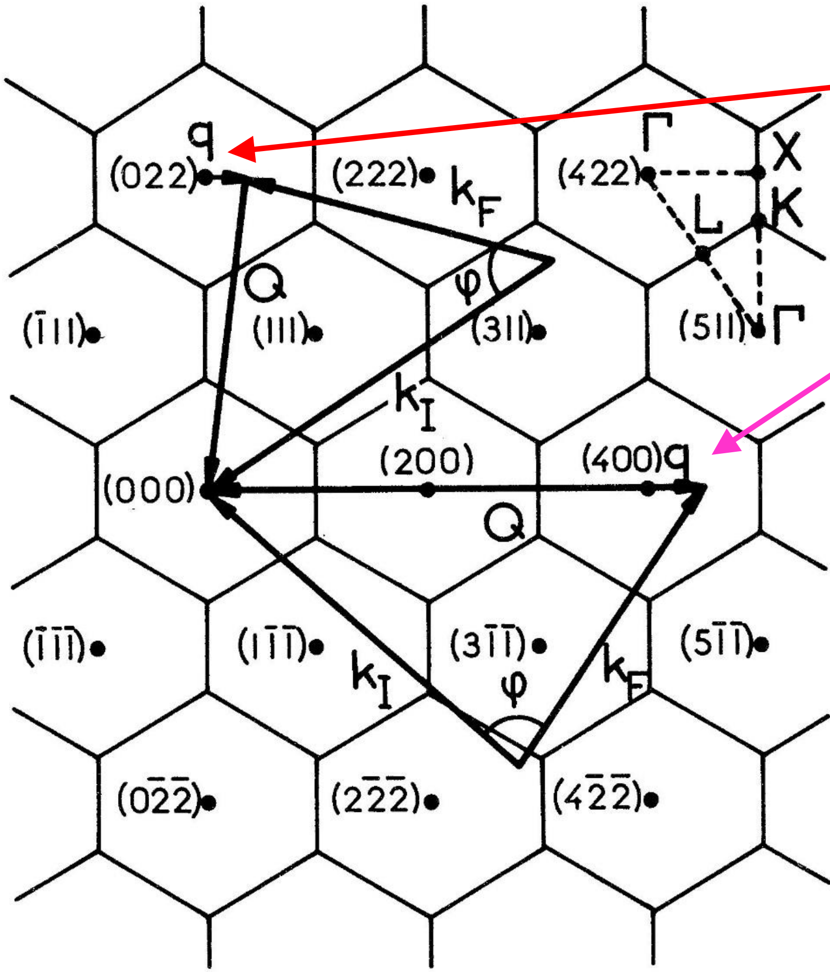


Fig. 8. Reciprocal lattice of a fcc structure with the [01$\bar{1}$] direction perpendicular to the experimental plane. Some conventional symbols for symmetry points are given in the upper right. Scattering diagrams for transverse phonons (top) and for longitudinal phonons (bottom) in [100] direction are inserted. $\underline{k}_I$ and $\underline{k}_F$ are the incoming and scattered wavevectors of the neutron, φ the scattering angle, $\underline{Q}$ the momentum transfer of the neutron, and $\underline{q}$ the phonon wavevector

EFFECTS due to FINITE RESOLUTION (1)

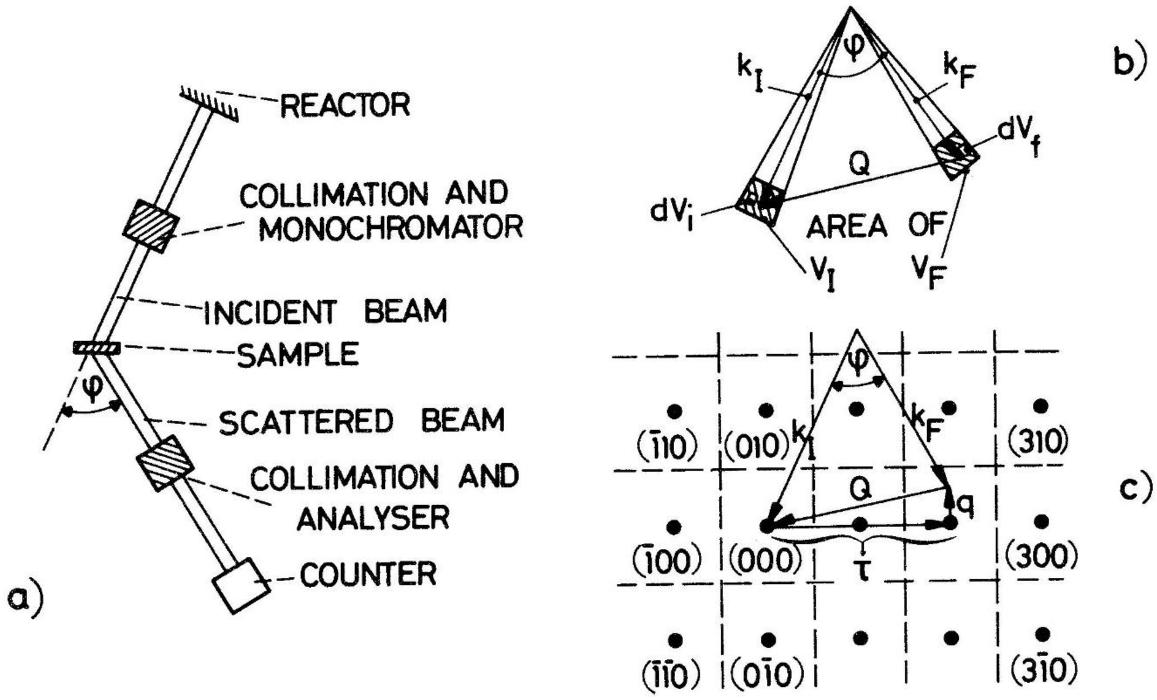


Fig. 1a-c. Inelastic neutron scattering: (a) path of neutrons in real space with "black boxes" for the determination of neutron energy before and after scattering; (b) corresponding distribution of neutrons V_I and V_F in reciprocal space around the mean wave vectors k_I and k_F; (c) momentum transfer Q of the neutron in relation to the reciprocal lattice of the sample (vectors τ) and the phonon wave vector q. (DORNER and COMES, 1977)

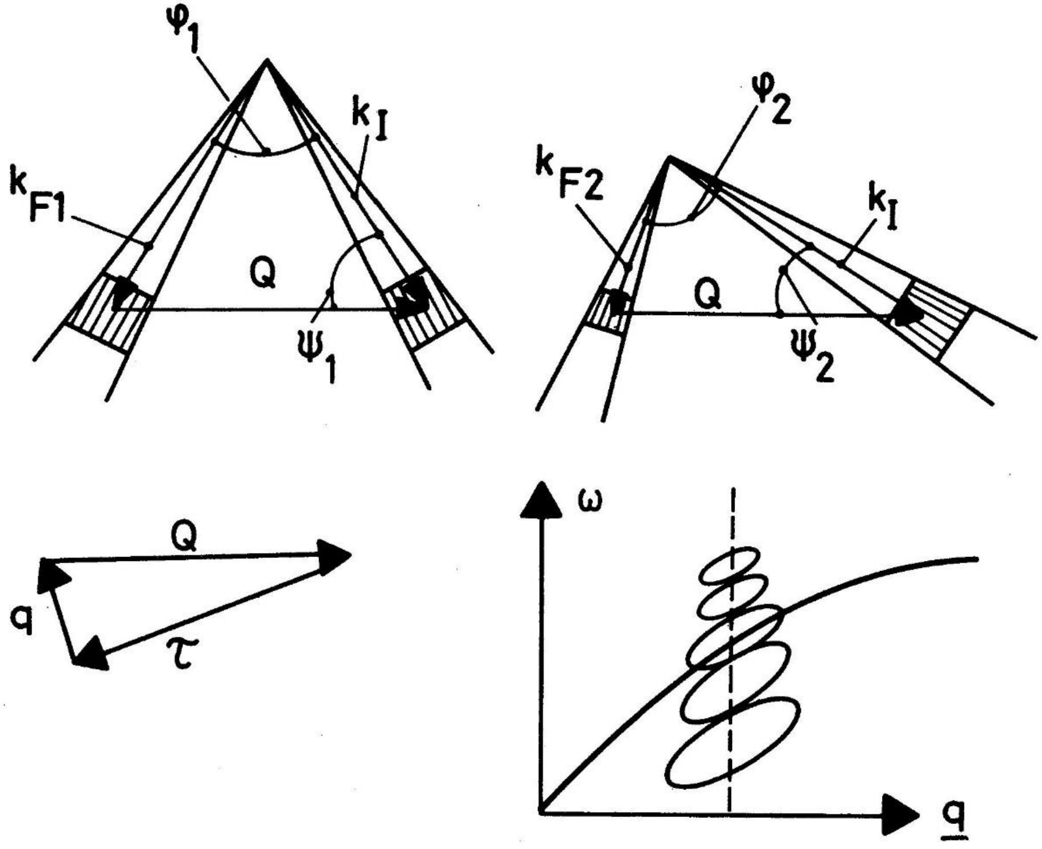
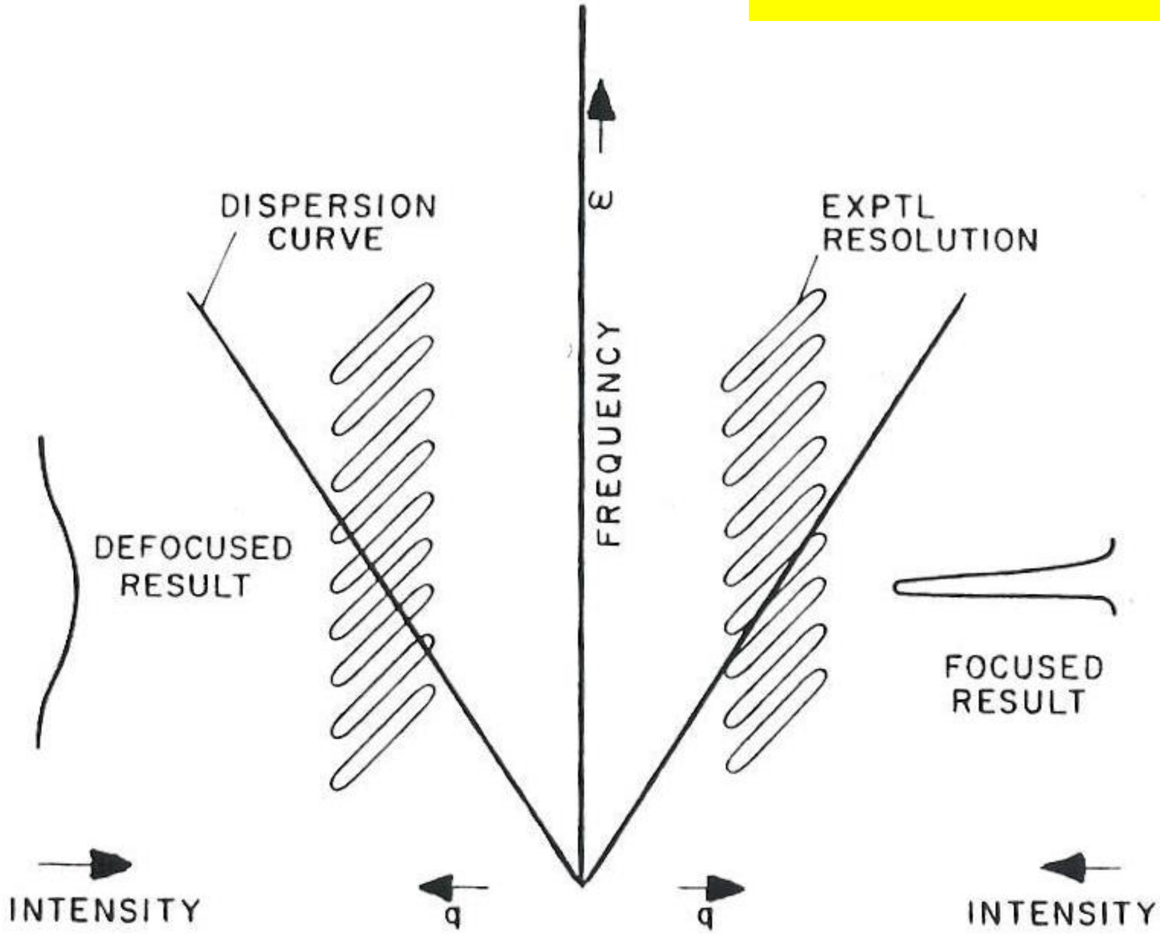


Fig. 4. Q-constant scan with k_I fixed. φ and ψ are scattering angle and sample orientation. The hetched areas give the distributions of k_i around k_I and of k_f around k_F. $\underline{q}$ is the phonon wavevector. In $\underline{q}$-ω space a constant Q scan is drawn with varying resolution. (DORNER, 1976)

EFFECTS due to FINITE RESOLUTION (3)



INFLUENCE OF INSTRUMENTAL RESOLUTION ON THE MEASUREMENT OF PHONON DISPERSION CURVES

EFFECTS due to FINITE RESOLUTION (4)

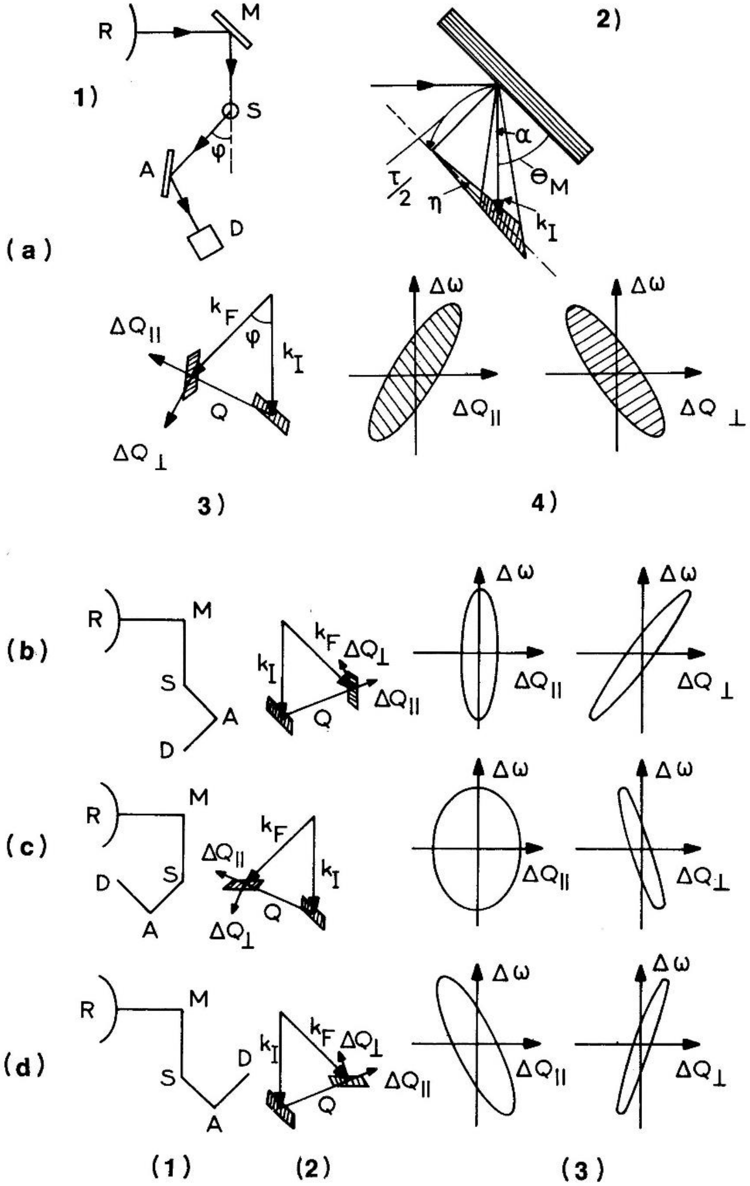


Fig. 6. (a) Resolution or transmission volume of a TAS: (1) path of the neutron beam; (R) reactor, (M) monochromator, (S) sample, (A) analyser, (D) detector; (2) reflection from a single crystal monochromator with mosaic width η and Bragg angle θ_M. The hatched area gives the distribution of $\underline{k}_i$ around $\underline{k}_I$; (3) scattering diagram for (1) in reciprocal space; (4) projections of the resolution. (DORNER, 1976). (b-d) Resolution or transmission volumes of a TAS in different geometries. Diagrams (1,3,4) are as defined in (a)

The differential scattering cross section can be interpreted as the Fourier transformation of a **correlation function** in both space and time (van Hove 1954)

Coherent case: Pair correlation function

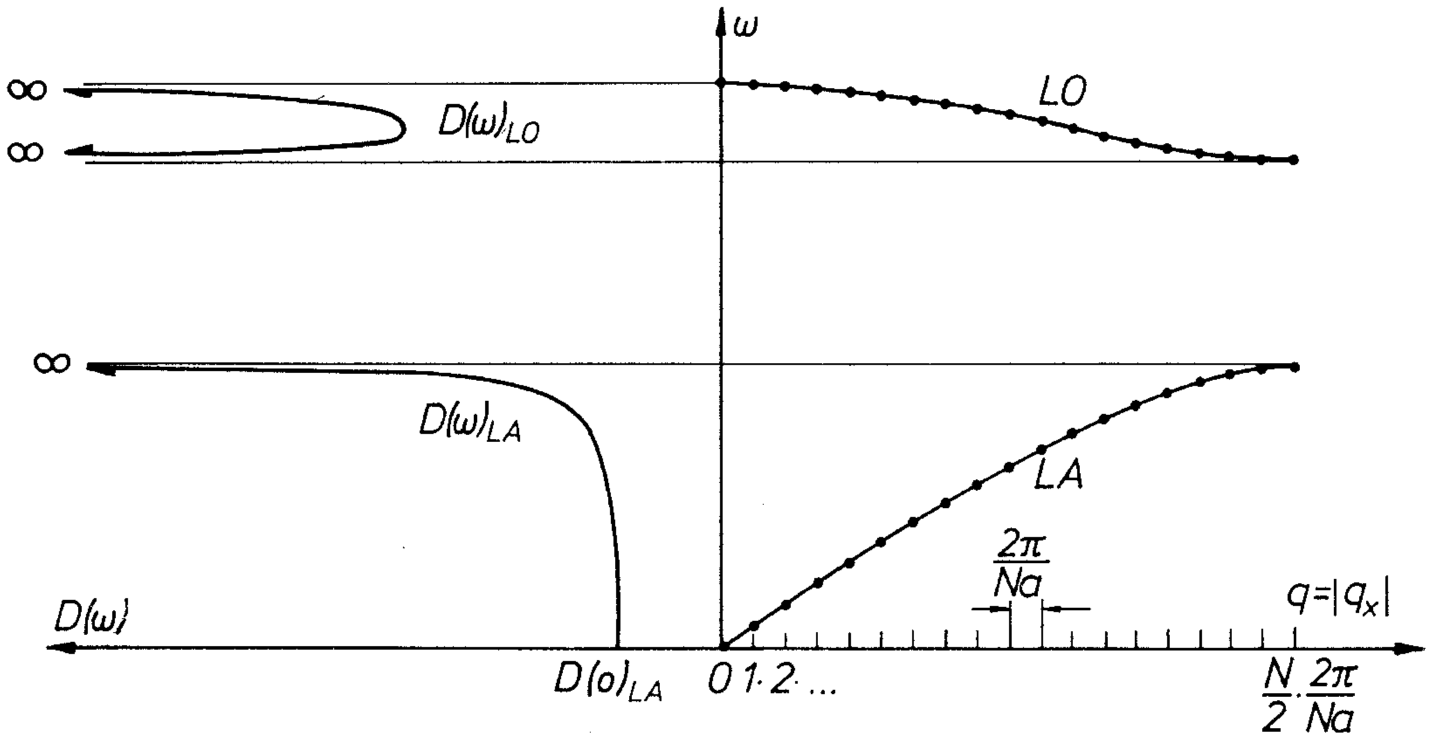
Incoherent case: Auto (Self-) correlation function

$$\left(\frac{d^2\sigma}{d\Omega\, d\varepsilon}\right)_{\mathrm{coh}} = \frac{b^2_{\mathrm{coh}} k}{2\pi\hbar k_0} \int\int dr\, dt e^{i\varkappa r - i\omega t} G(r, t)$$

$$\left(\frac{d^2\sigma}{d\Omega\, d\varepsilon}\right)_{\mathrm{inc}} = \frac{b^2_{\mathrm{inc}} k}{2\pi\hbar k_0} \int\int dr\, dt e^{i\varkappa r - i\omega t} G_s(r, t)$$

SINGULARITIES IN THE DENSITY OF STATES

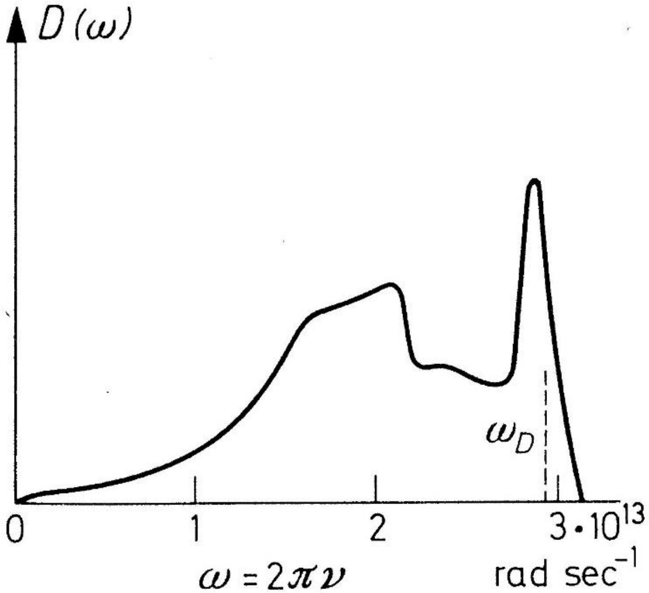
(MODEL CALCULATION)

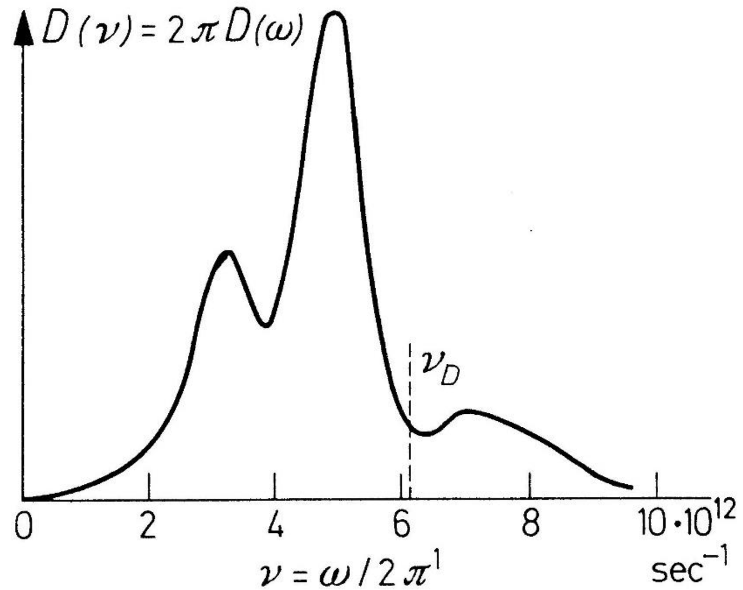


Longitudinal waves in a linear AB-chain

(mass ratio of atoms 3:1)

EXAMPLES of DENSITY OF STATES (DOS)





Ag (fcc)

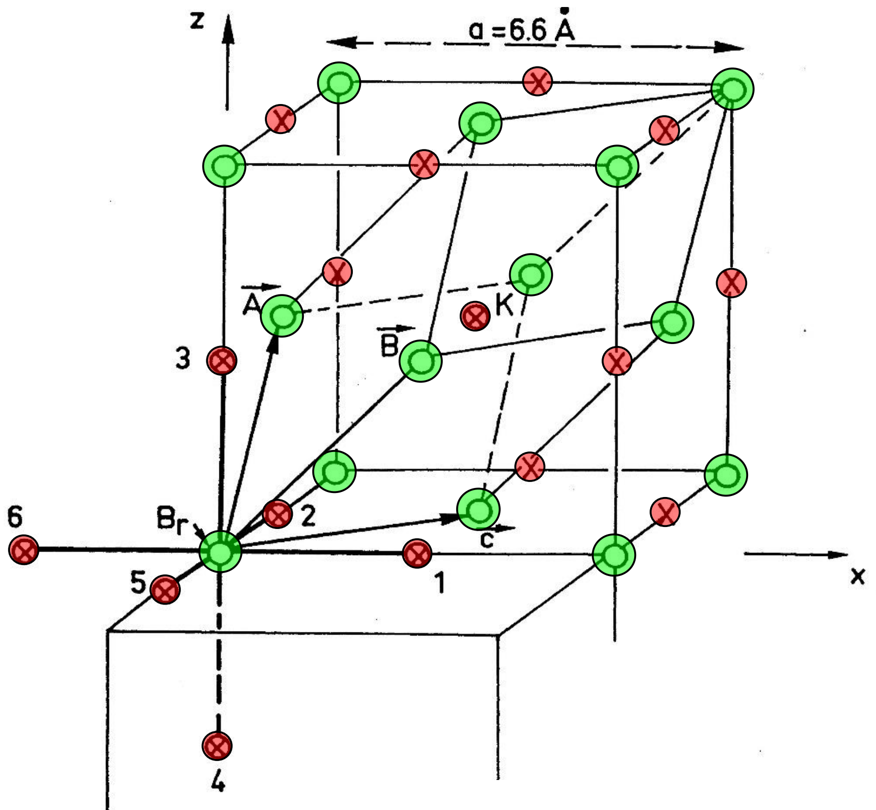
monatomic

NaCl (fcc)

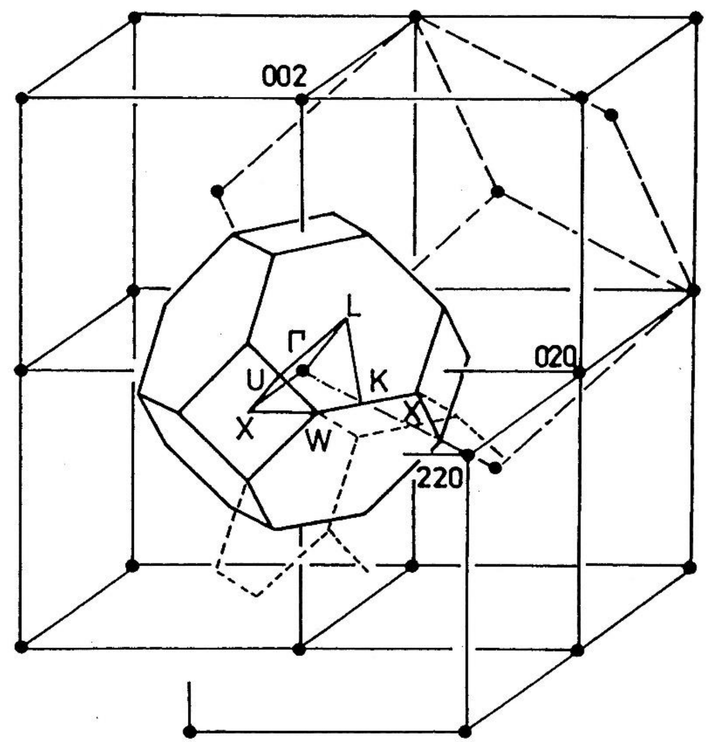
diatomic

Example: Phonon Dispersion Curves in KBr (1)

Unit cell of KBr (real space)



First Brillouin zone of KBr (reciprocal space)



Atomic positions of Br

Atomic positions of K

KBr (2): Model calculation using simple Rigid Ion Model

<u>Long wavelength limit:</u>

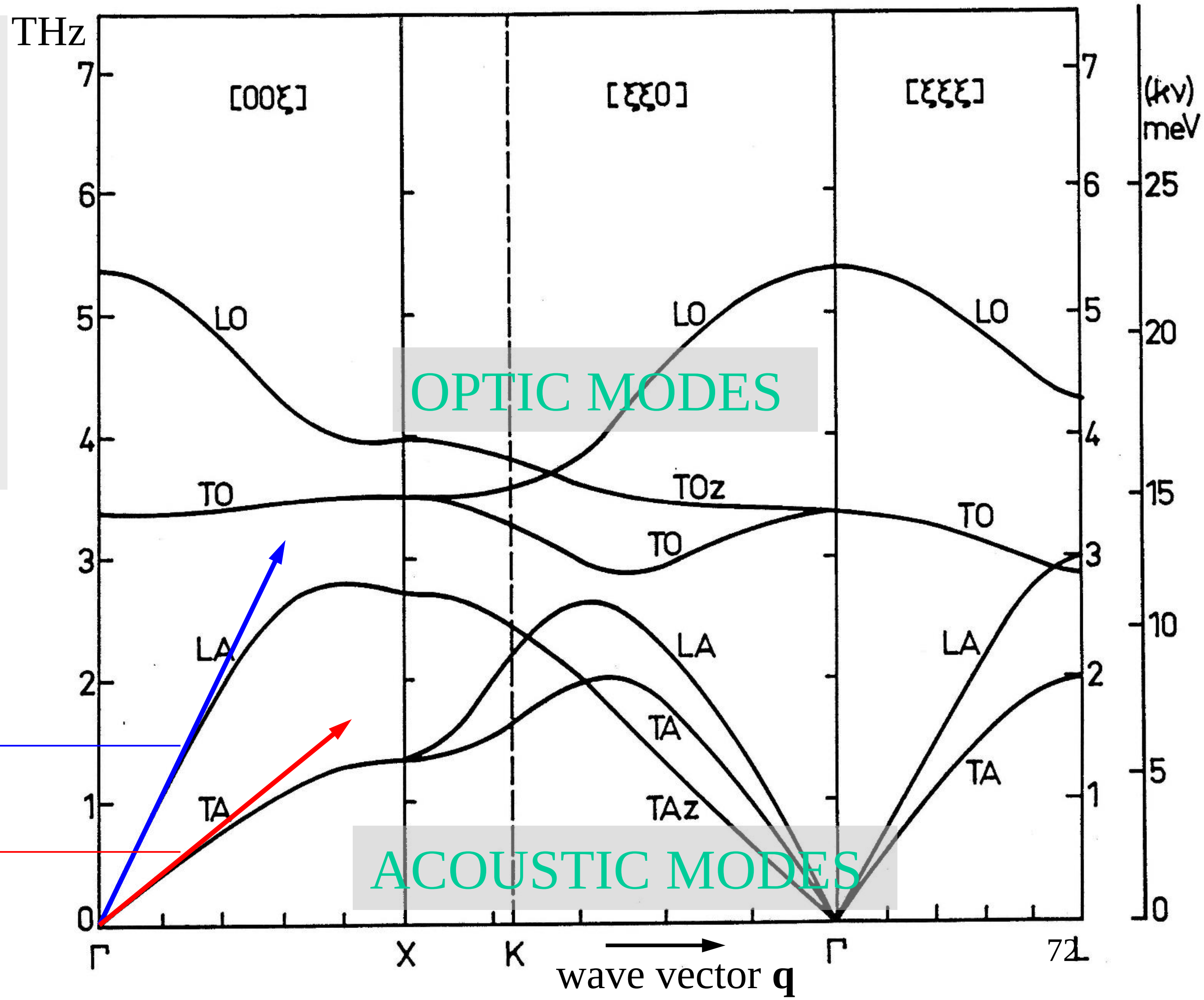
slopes of acoustic modes give sound velocities

→ elastic moduli

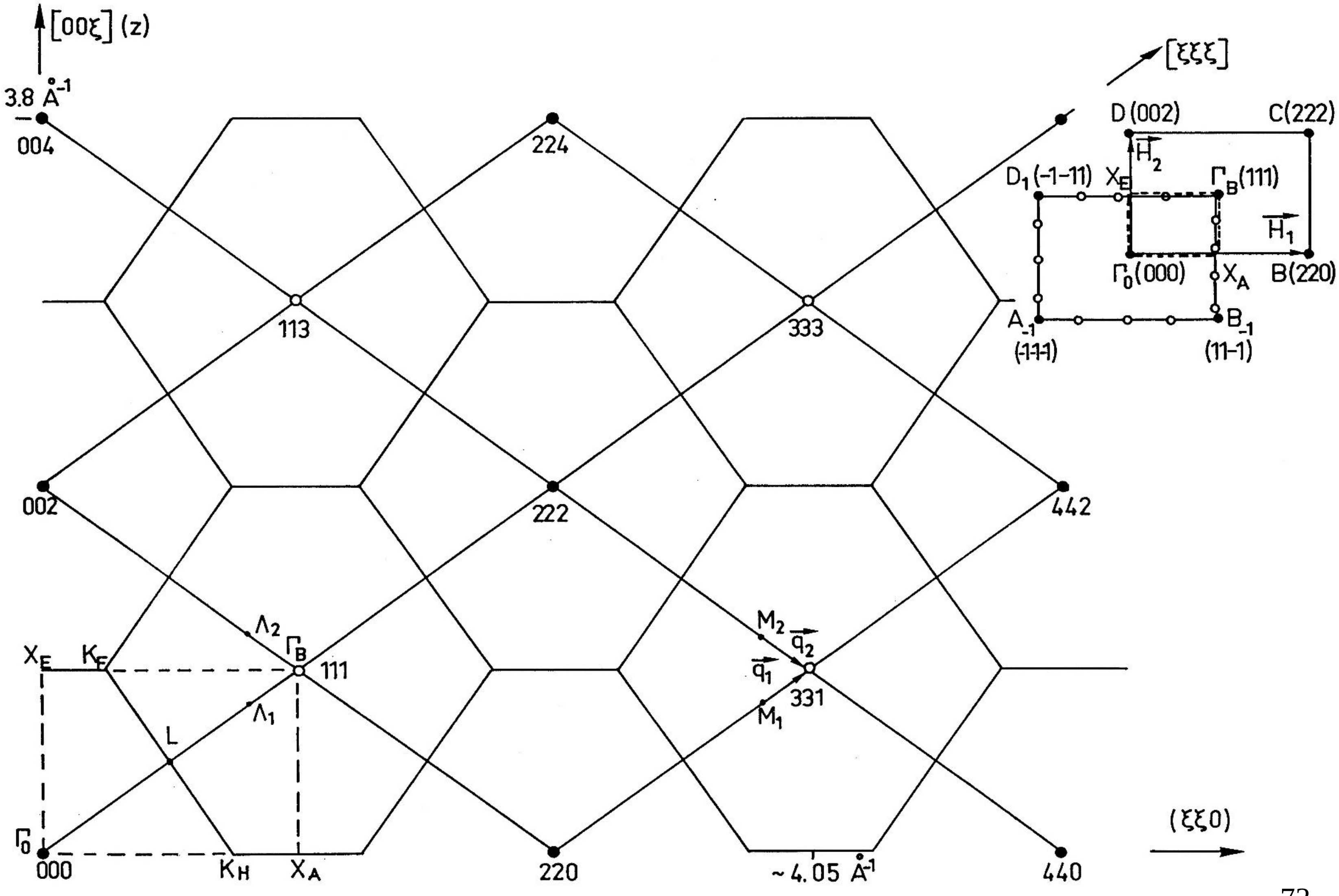
$v_{sound} = \omega/q = \nu\,\lambda$

$\rho\, v_{long}^2 = c_{11}$

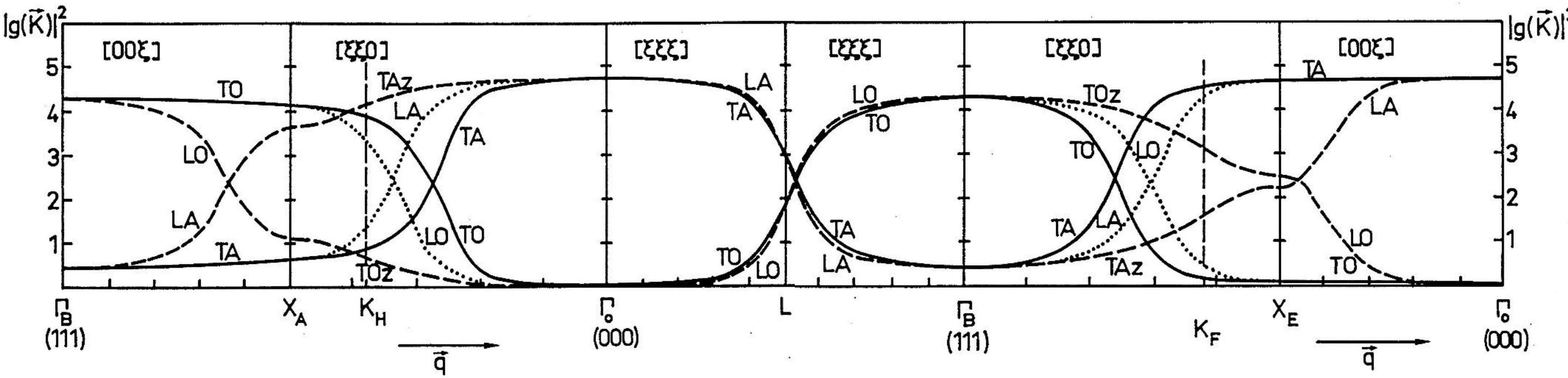
$\rho\, v_{trans}^2 = c_{44}$



KBr (3): (110) plane of reciprocal lattice



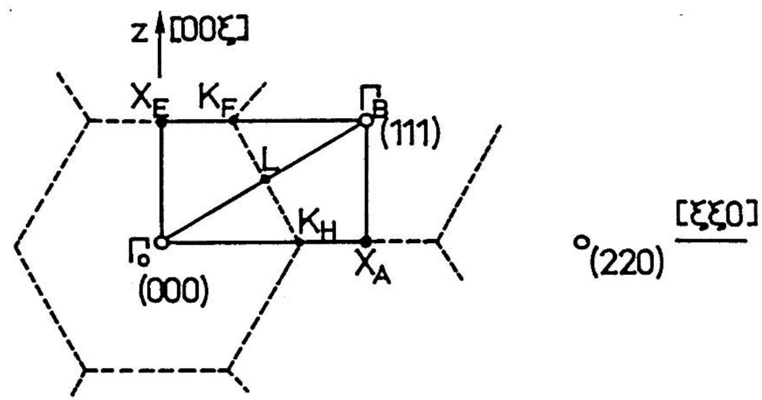
KBr (4): Dynamical structure factors along principal symmetry directions



Positions of the high symmetry points

Γ_o, K_H, X_A, Γ_B, K_F, X_E

in the reduced zone scheme



The lecture was prepared partly using material from the following

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