Introduction to Inelastic X-ray Scattering

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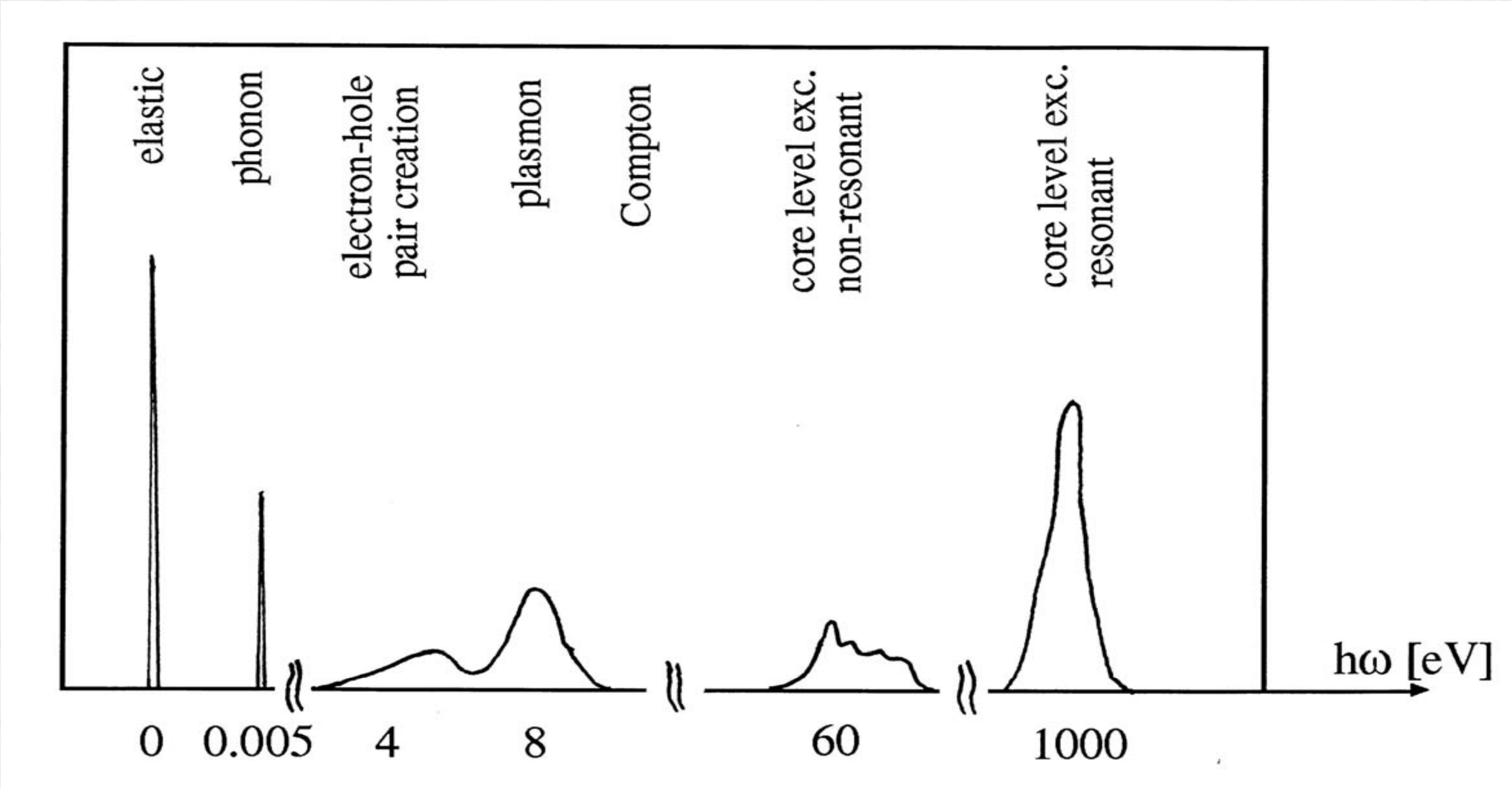
OUTLINE

General Introduction
Experimental and theoretical background

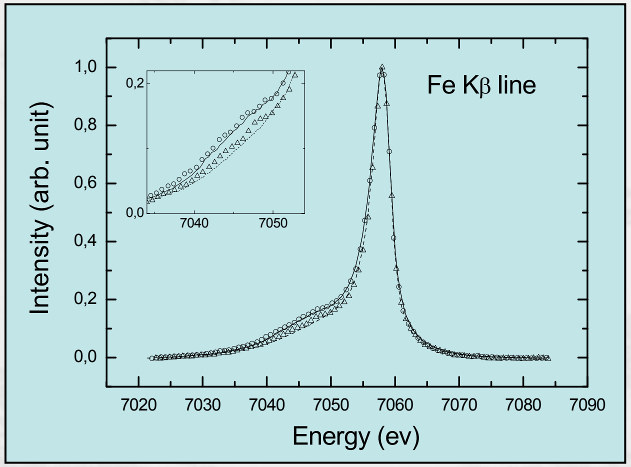
IXS from phonons
- Neutrons versus X-rays
- Examples

Schematic inelastic x-ray spectrum

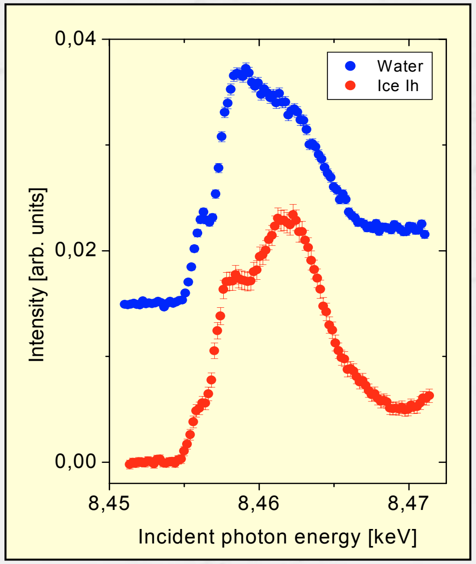


Energy Transfer [eV]

Magnetism

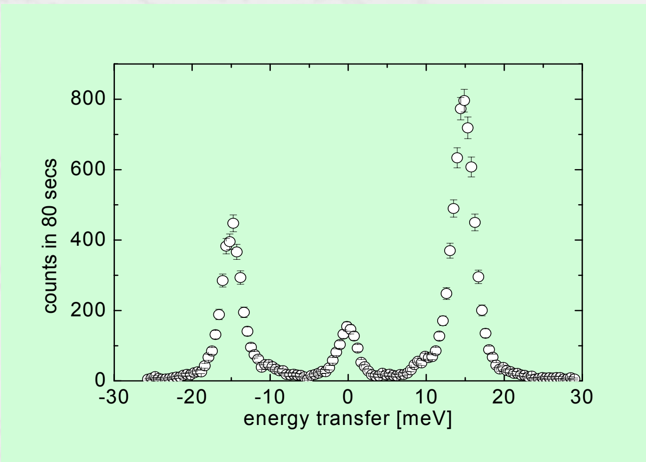


Electronic structure

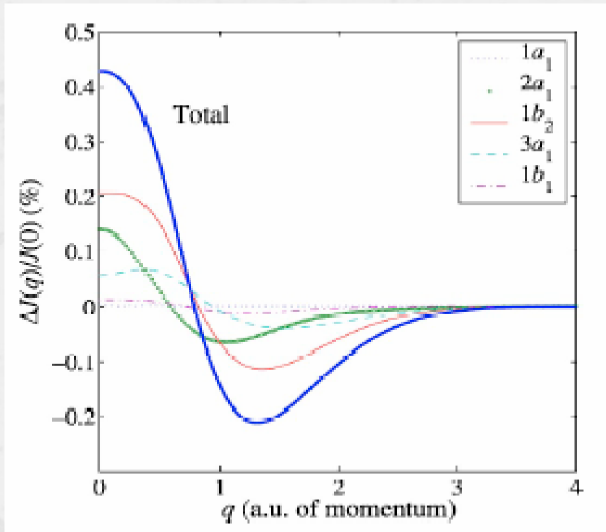


IXS

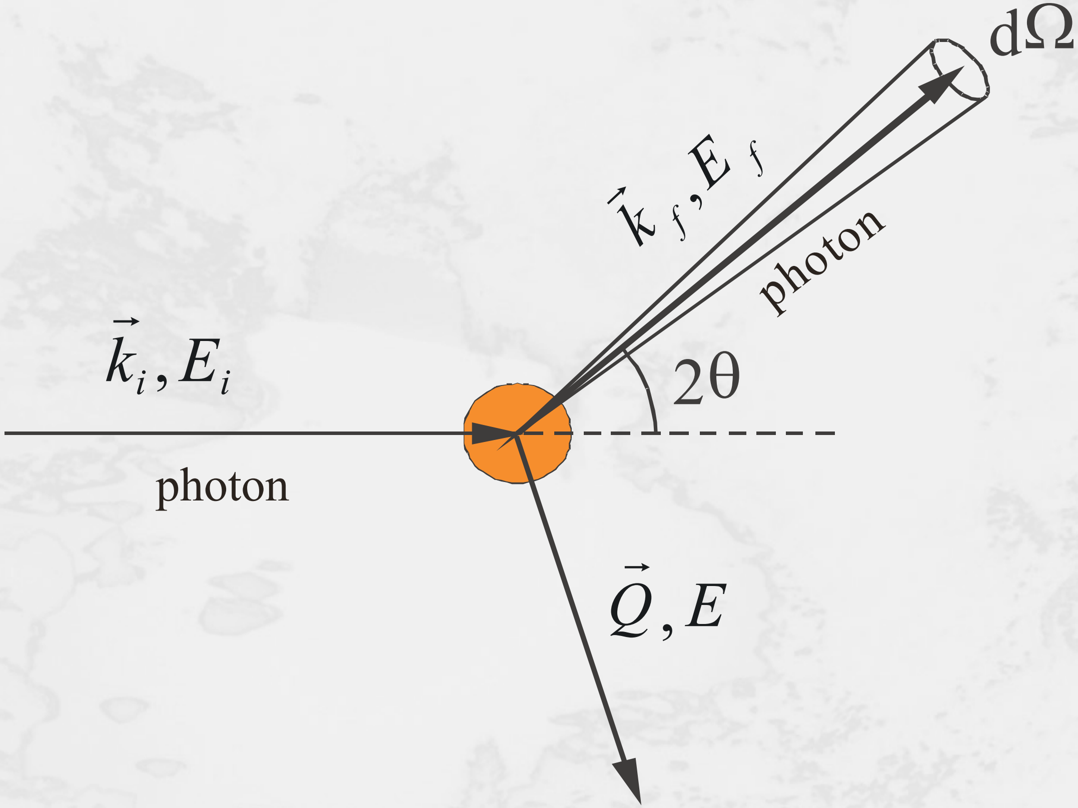
Phonons



Chemical bonding



Scattering kinematics



- Energy transfer: $E_f - E_i = E$
- Momentum transfer: $\vec{k}_f - \vec{k}_i = \vec{Q}$

Theoretical Aspects I

Photon-electron interaction

(neglecting magnetic terms)

$$H_{int} = \frac{e}{m_e c} \sum_j \left(\frac{e}{2c} \vec{A}_j^2 + \vec{A}_j \vec{p}_j \right)$$

A is the vector potential of the electromagnetic field.
p is the momentum operator of the electron.
j is the summation over the electrons of the scattering system.

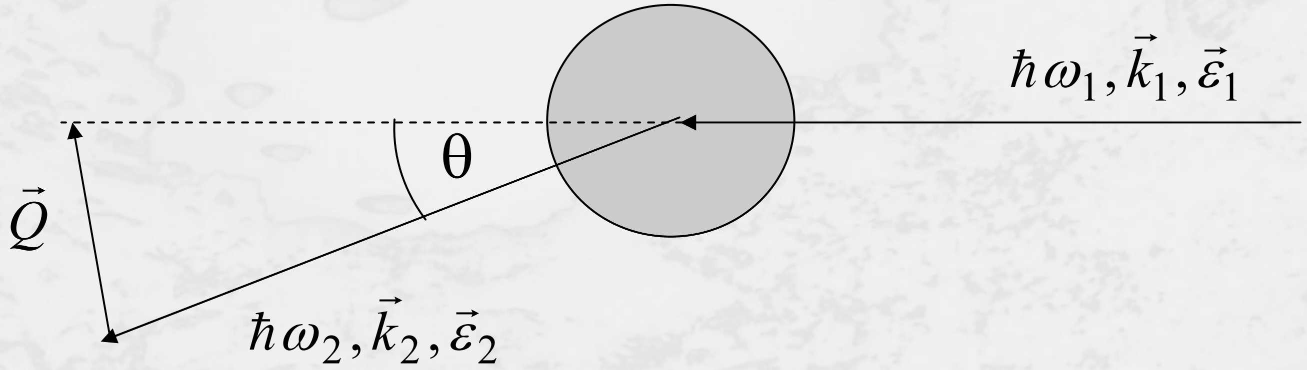
Theoretical Aspects II

2) Scattering of a photon (**A·A** in 1. Order)

- non-resonant

3) Scattering of a photon (**p·A** in 2. Order)

- resonant scattering
- absorption followed by emission



Theoretical Aspects III

Lowest order perturbation theory (Fermi's Golden Rule):

$$w_{i\to f} = \frac{2\pi}{\hbar}\left|\langle F|H_{int}|I\rangle + \sum_N \frac{\langle F|H_{int}|N\rangle\langle N|H_{int}|I\rangle}{E_N - E_I}\right|^2$$

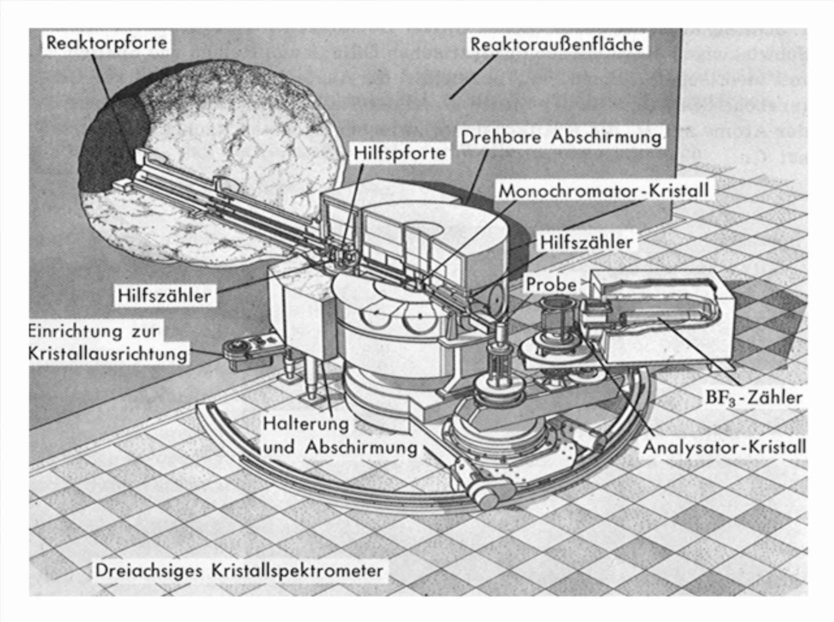
$$\delta\left(E_F - E_I - \hbar\omega\right)$$

Double differential cross section

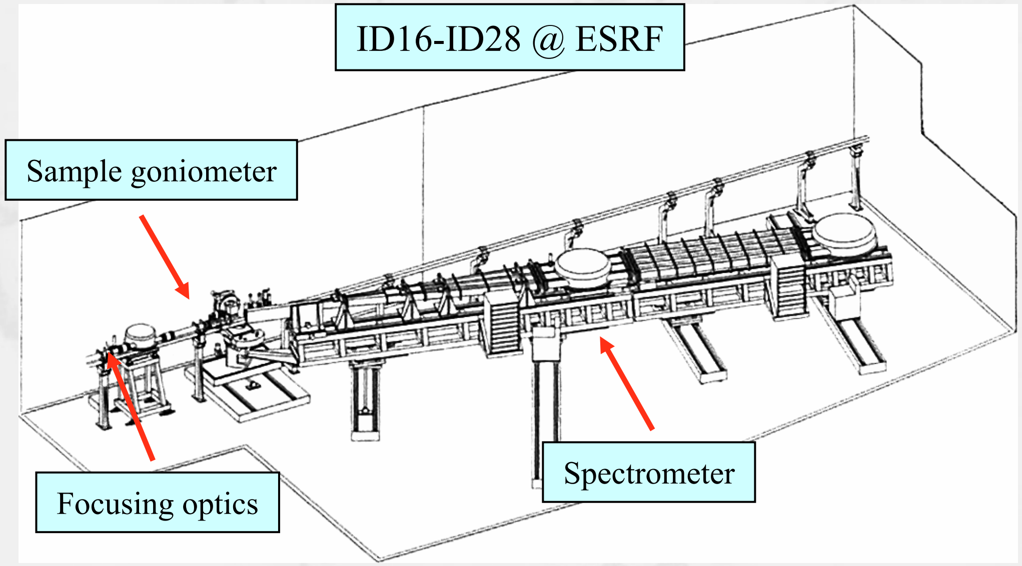
$$\frac{d^2\sigma}{d\omega_2 d\Omega} = \sum_F w_{i\to f}\ \rho\left(E_f\right) I_0^{-1}$$

$w_{i\to f}$ is the transition probability, $\rho(E_f)$ is the density of final states and I_0 the incident flux.

Inelastic Scattering from Phonons



Brockhouse (1955)



Burkel, Dorner and Peisl (1987)

Thermal neutrons:

$E_i = 25$ meV

$k_i = 38.5$ nm^{-1}

$\Delta E/E = 0.01 - 0.1$

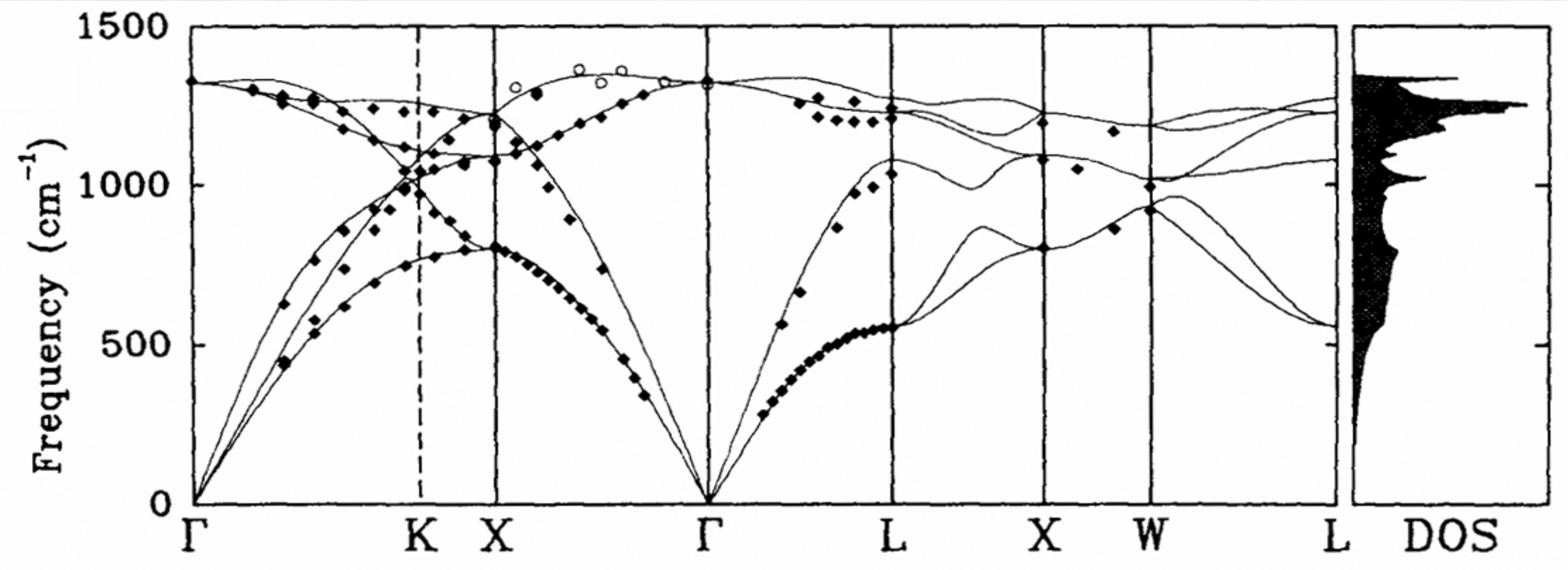
Hard X-rays:

$E_i = 18$ keV

$k_i = 91.2$ nm^{-1}

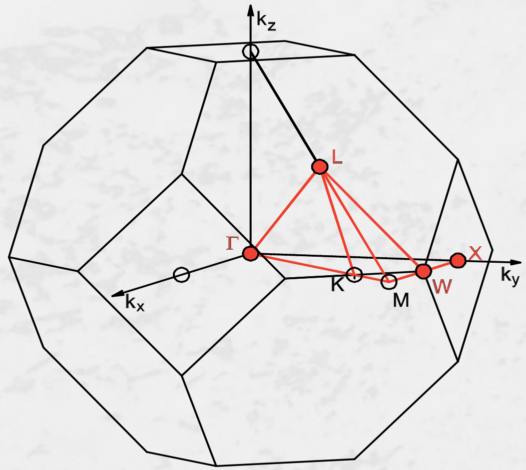
$\Delta E/E \leq 1 \times 10^{-7}$

Phonon dispersion and density of states



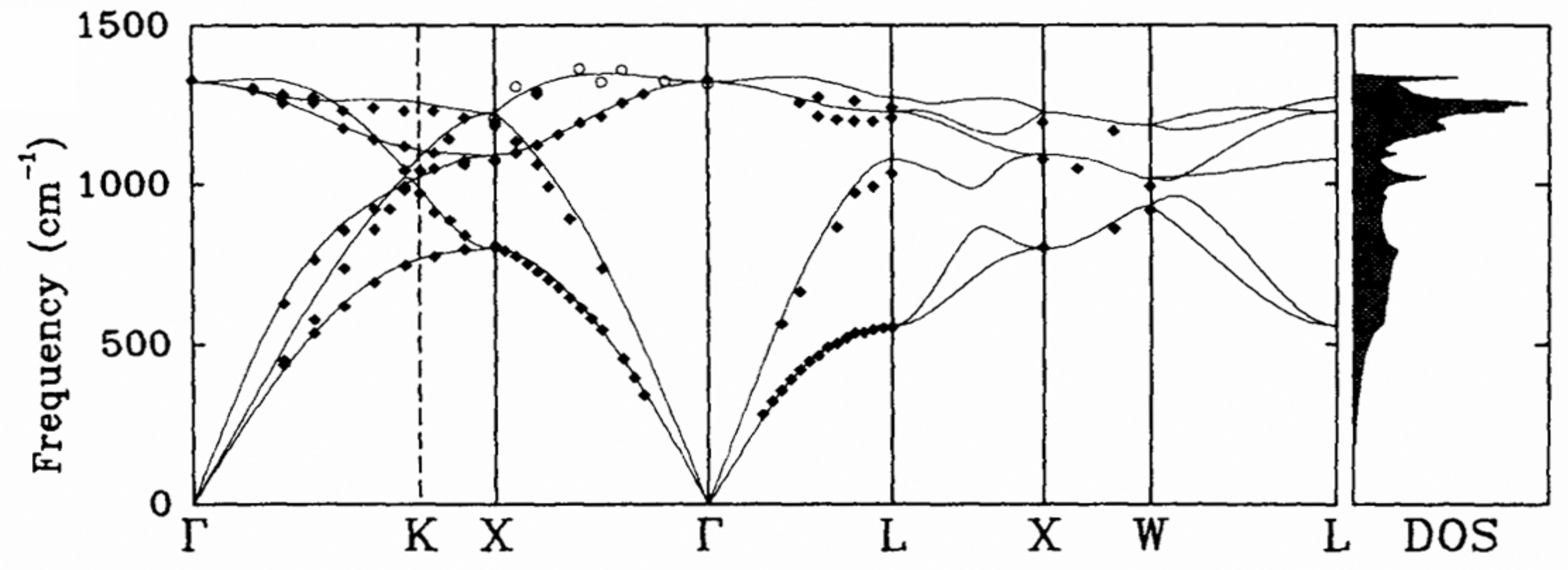
diamond

P. Pavone et al. Phys. Rev. B 48 (1993) 3156



Brillouin zone for fcc structure

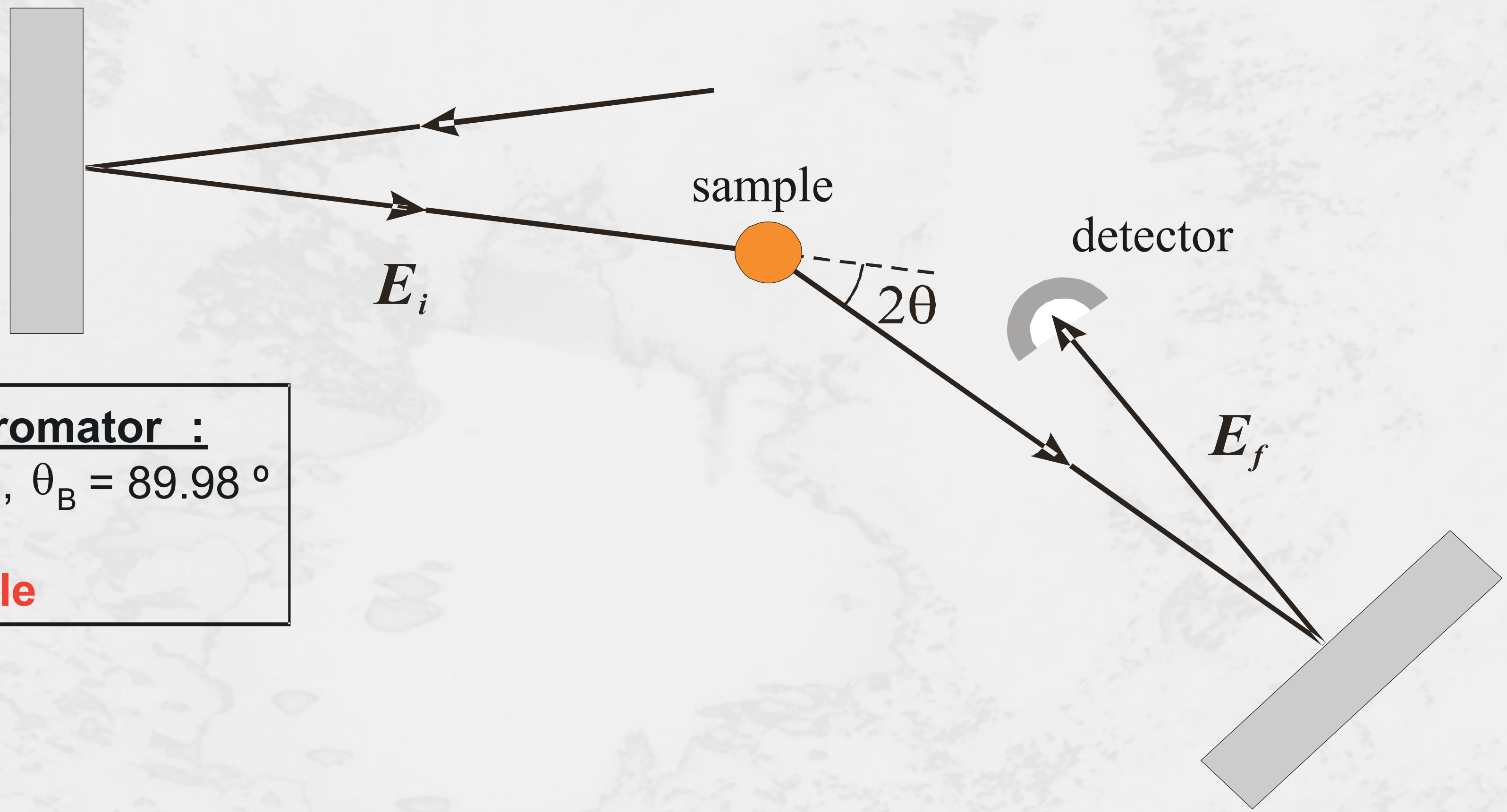
Information derived from IXS/INS data



- Sound velocities
- Elasticity
- Interatomic force constants (potential)
- Dynamical instabilities (phonon softening)
- Anharmonicity
- Phonon-electron coupling

- Thermodynamics (C_V, S_V, θ_D, …)

Experimental IXS set-up

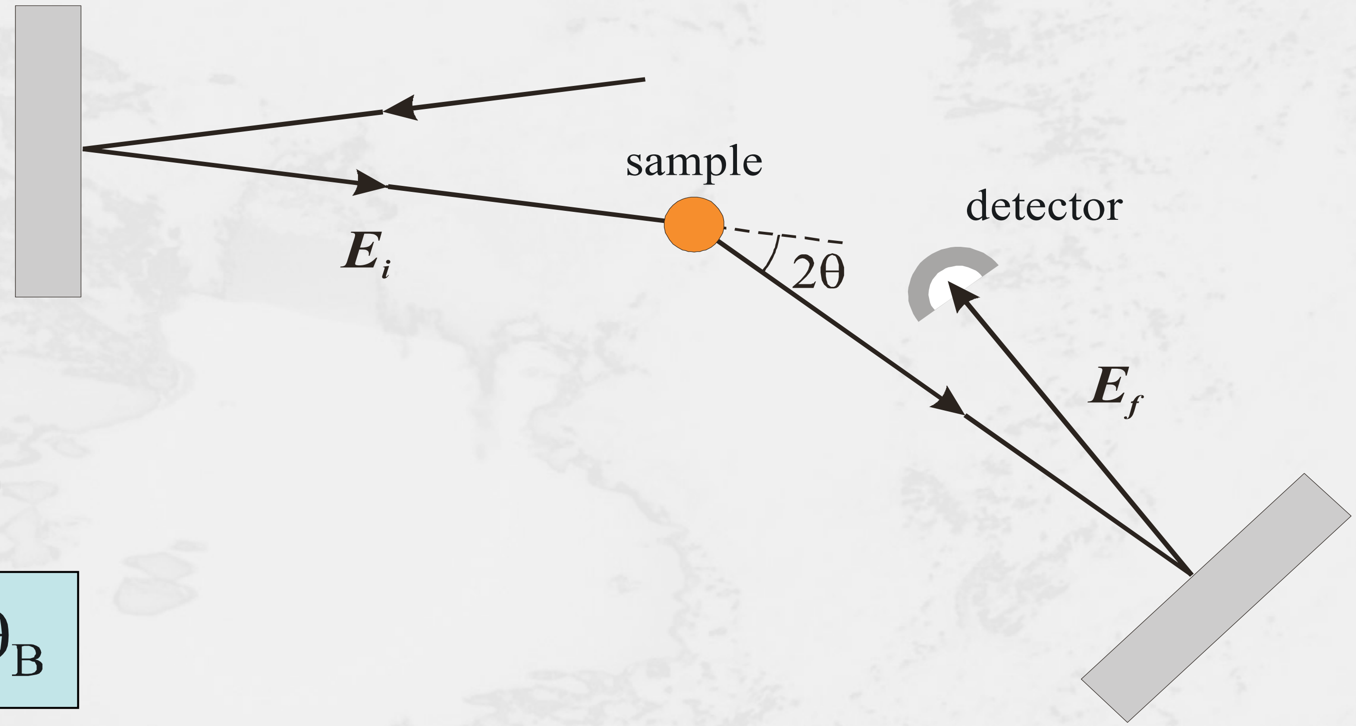


Monochromator :
Si(n,n,n), θ_B = 89.98 º
n=7 - 13
λ_1 **tunable**

Spot size: 30 x 60 μm^2 (H x V)

Analyser :
Si(n,n,n), θ_B = 89.98 º
n=7 - 13
λ_2 **constant**

IXS set-up: energy scanning



$\mathbf{Q} = 4\pi/\lambda\cdot\sin(\theta)$

$\lambda = 2\cdot d(T)\ \sin\theta_B$

$\Delta d/d = \mathbf{\Delta E}/E = -\alpha(T)\cdot\Delta T$ ($\alpha=2.58\cdot10^{-6}$ at RT)

IXS versus INS

IXS

$$\frac{\partial^2\sigma}{\partial E\partial\Omega} = r_0^2\,\frac{k_1}{k_2}(\vec{\varepsilon}_1\cdot\vec{\varepsilon}_2)f(Q)^2\;S(\vec{Q},E)$$

- no correlation between momentum- and energy transfer
- ΔE/E = 10^{-7} to 10^{-8}
- Cross section ~ Z^2 (for small Q)
- Cross section is dominated by photoelectric absorption (~ $\lambda^3 Z^4$)
- no incoherent scattering
- small beams: 100 μm or smaller

INS

$$\frac{\partial^2\sigma}{\partial E\partial\Omega} = b^2\,\frac{k_1}{k_2}S(\vec{Q},E)$$

- strong correlation between momentum- and energy transfer
- ΔE/E = 10^{-1} to 10^{-2}
- Cross section ~ b^2
- Weak absorption => multiple scattering
- incoherent scattering contributions
- large beams: several cm

Why X-rays?

- Small sample volumes: $10^{-4} – 10^{-5}$ mm^3
- No kinematic limitations: E independent of Q

- Novel, exotic materials in very small quantities
- Previously unexploited Q,E region in disordered system
- Very high pressures > 1Mbar

An untypical IXS scan

Diamond; Q=(1.04,1.04,1.04)

Stokes peak:
phonon creation
energy loss

Anti-Stokes peak:
phonon annihilation
energy gain