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Interaction Neutron/Matter

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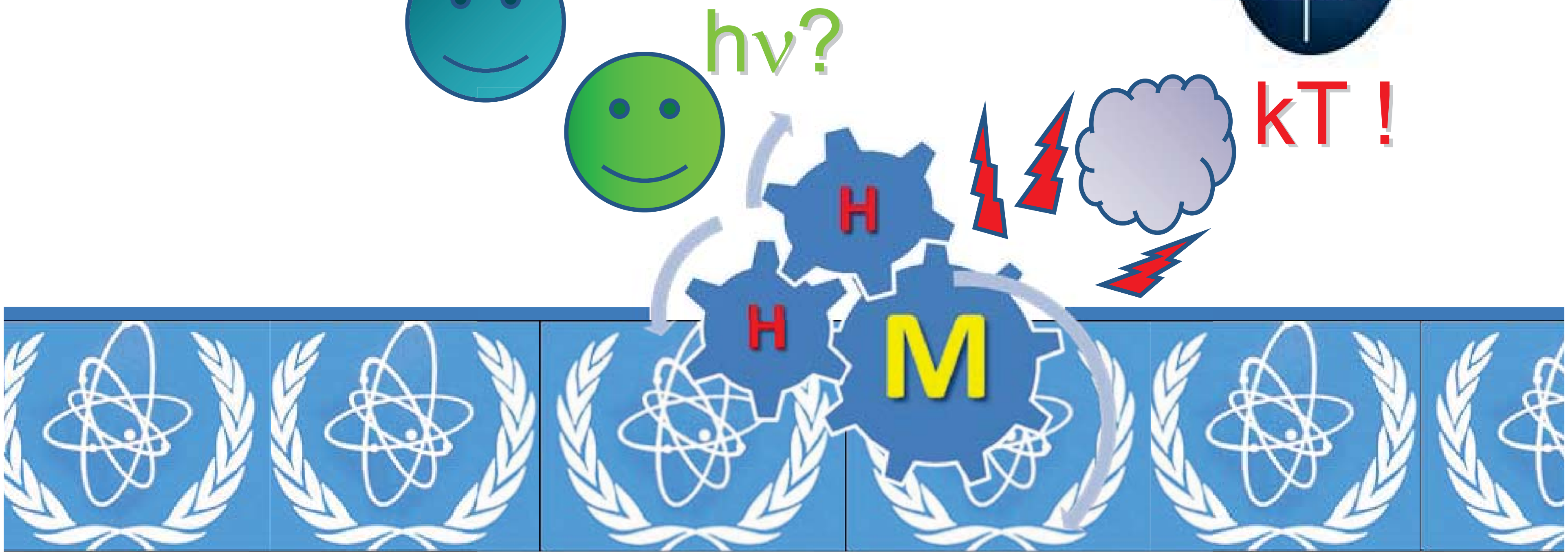
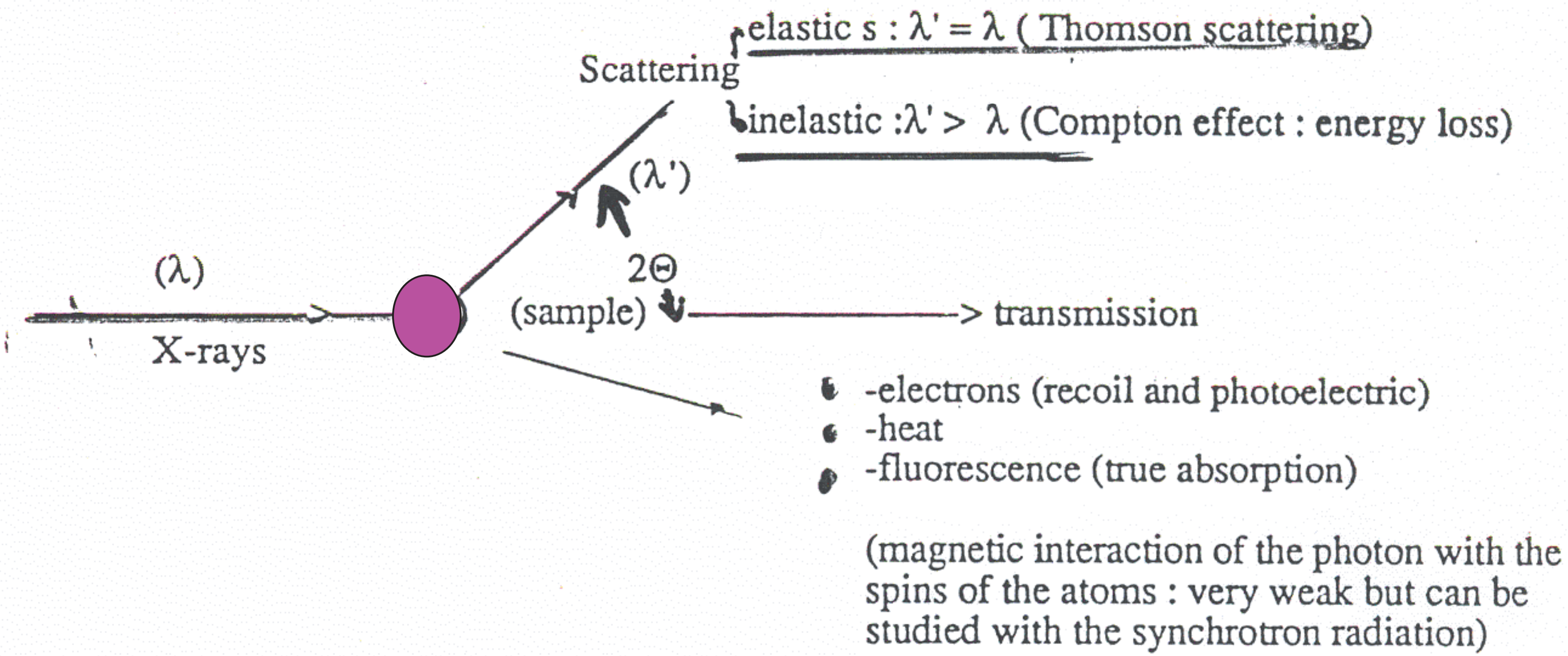


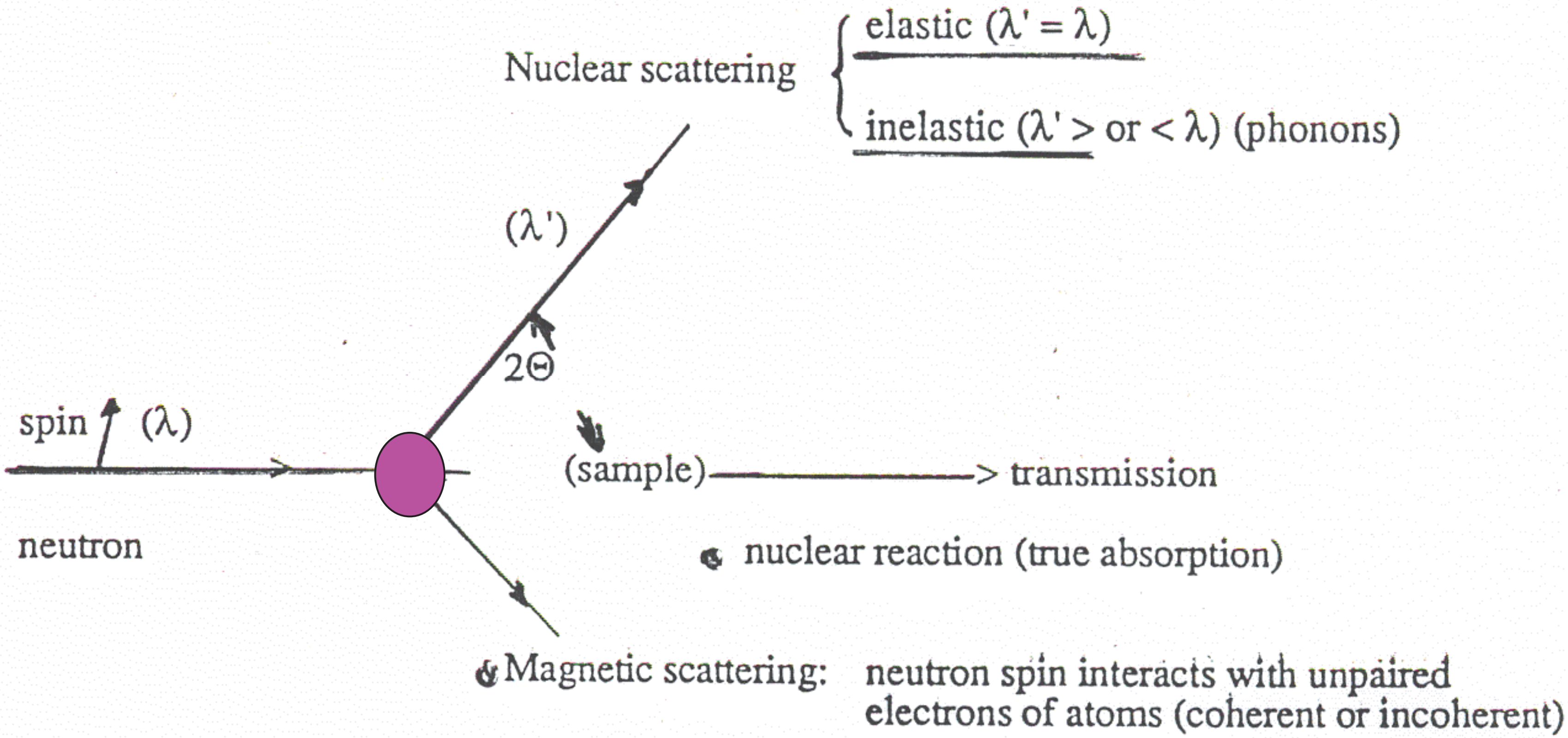
Interaction of a Monochromatic Beam with Matter

As the beam passes through a matter, radiation is removed from the direct beam by a variety of processes summarized below.

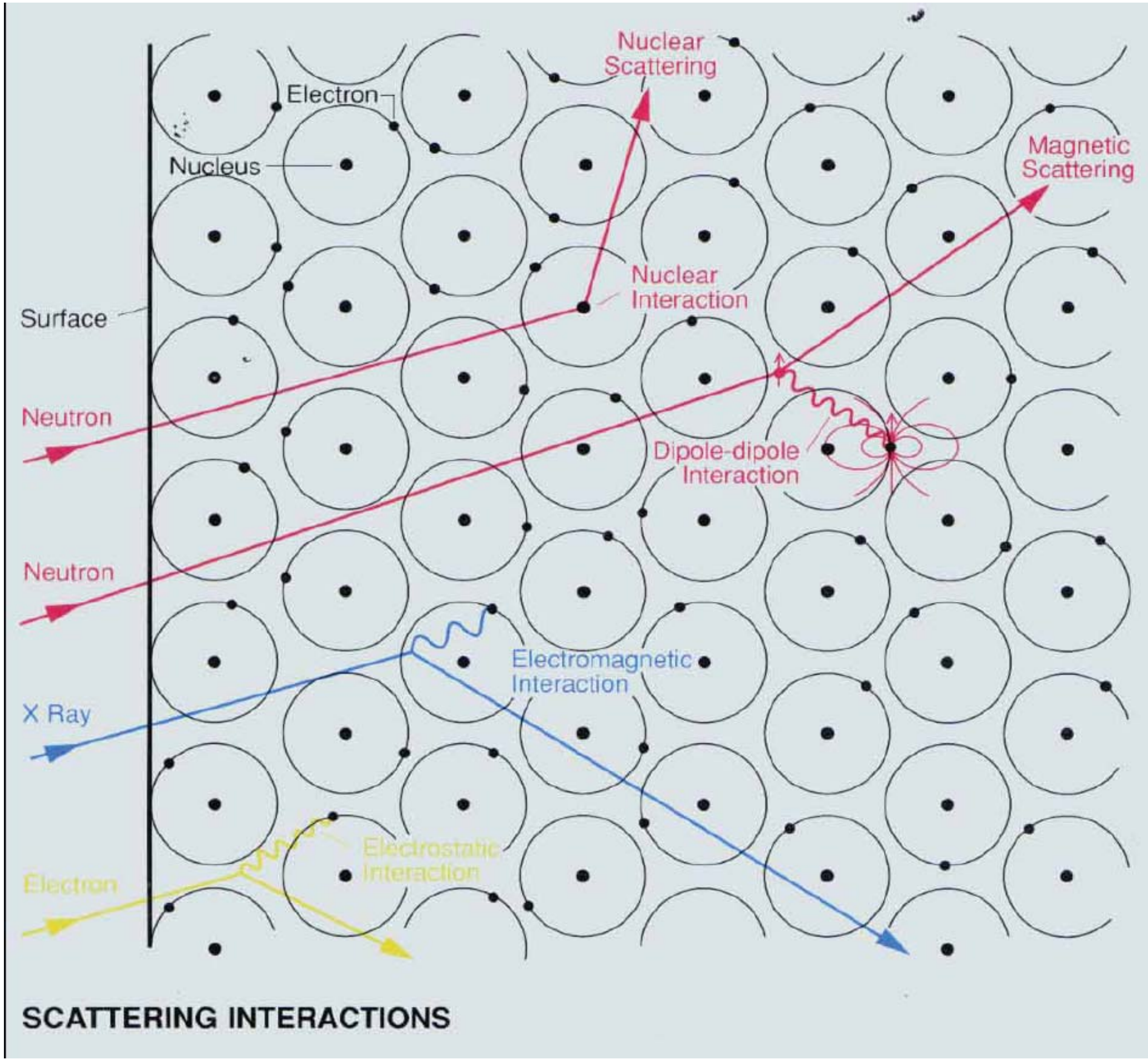
1- X-ray / Matter Interaction processes



2- Neutron / Matter Interaction processes

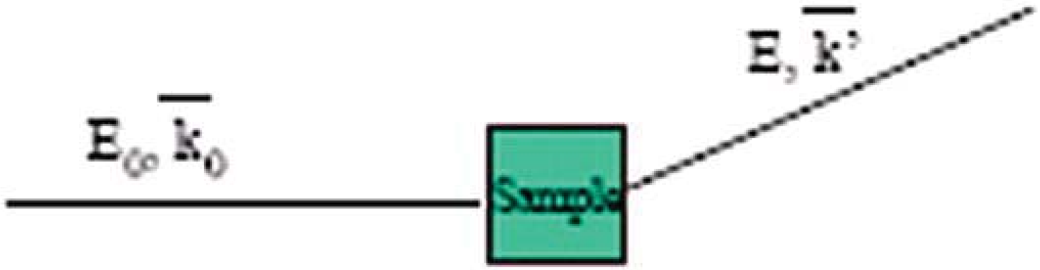


Remark : the scattering effects contribute to the absorption of the incoming beam : the total absorption coefficients take into account the true absorption and the scattering process.(The absorption cross-section).



- Neutrons interact with atomic nuclei via very short range (~fm) forces.
- Neutrons also interact with unpaired electrons via a magnetic dipole interaction.

A Typical Neutron Scattering Experiment



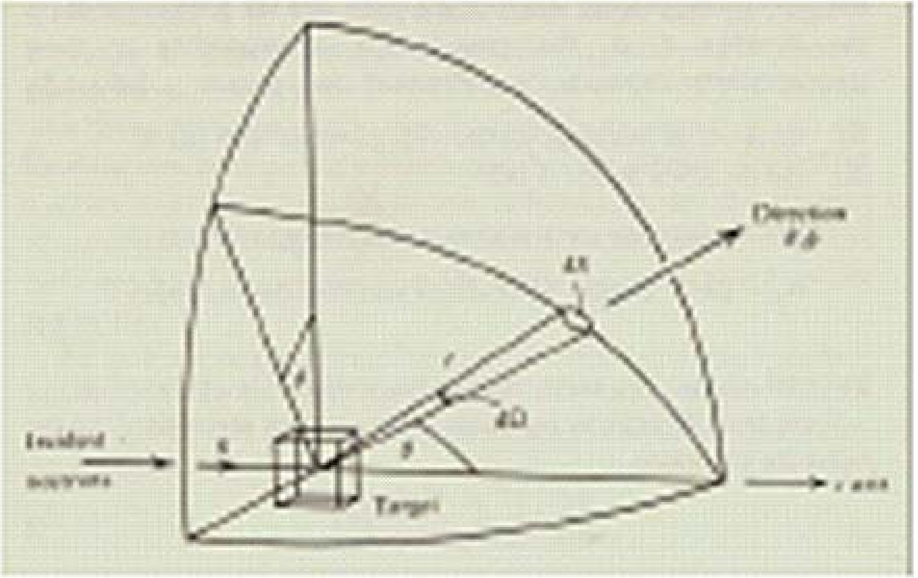
$$\overline{Q} = \overline{k'} - \overline{k_0}$$

$$\hbar\omega = E - E_0$$

$$\text{Momentum} = \hbar k; \quad \text{Energy} = \hbar^2 k^2 / 2m_n$$

- Measure the number of scattered neutrons as a function of Q and ω
- The result is the scattering function S(Q,ω) that depends only on the properties of the sample

What we Measure in an Experiment: the Total or Differential Cross Sections



Φ = number of incident neutrons per cm^2 per second

σ = total number of neutrons scattered per second / Φ

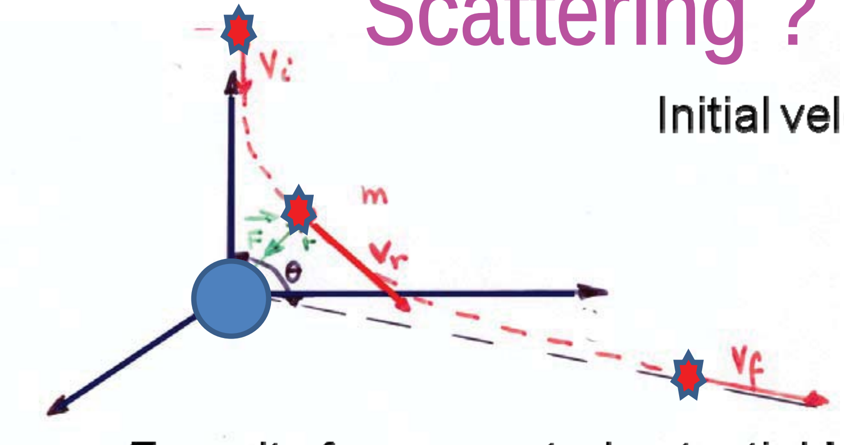
$$\frac{d\sigma}{d\Omega} = \frac{\text{number of neutrons scattered per second into } d\Omega}{\Phi \, d\Omega}$$

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\text{number of neutrons scattered per second into } d\Omega \text{ \& } dE}{\Phi \, d\Omega dE}$$

σ measured in barns:
1 barn = 10^{-24} cm^2

Attenuation = $\exp(-N\sigma t)$
N = # of atoms/unit volume
t = thickness

Scattering ?



Classical point of view

Initial velocity $\boldsymbol{V}_i$, $\boldsymbol{V}_r$ velocity at point $\boldsymbol{r}$, final velocity $\boldsymbol{V}_f$

$\boldsymbol{r}$, distance particle to diffusion centre O

θ, angle of deviation by scattering

O, diffusion centre

m, mass (neutron)

$\boldsymbol{F}$, force applied by O to m

$\boldsymbol{F}$ results from a central potential $\boldsymbol{V}$(r) such as:

$\boldsymbol{F} = \boldsymbol{r}/r \,.\, d\boldsymbol{V}/d\boldsymbol{r}$, the diffusion cross section σ is defined from the particle flux $\boldsymbol{j}$ as

$$\boldsymbol{j}_f \,.\, \boldsymbol{r}/r = \boldsymbol{j}_i\, \sigma/r^2$$

Quantum point of view

$E_i = ½\, m\boldsymbol{v}^2$ (class.) $\rightarrow \boldsymbol{p}^2/2m = -i\,\hbar\, \boldsymbol{V}$ (quant.)

$E_i = -\hbar^2/2m\, \nabla^2 \psi_i$ in vacuum

$E_i = (-\hbar^2/2m\, \nabla^2 + V)\, \psi$ in matter

The incident wave is solved as $\psi_i = \exp i\boldsymbol{k}_i \,.\, \boldsymbol{r}$
with energy $E_i = (\hbar k_i)^2/2m$

In case of elastic scattering

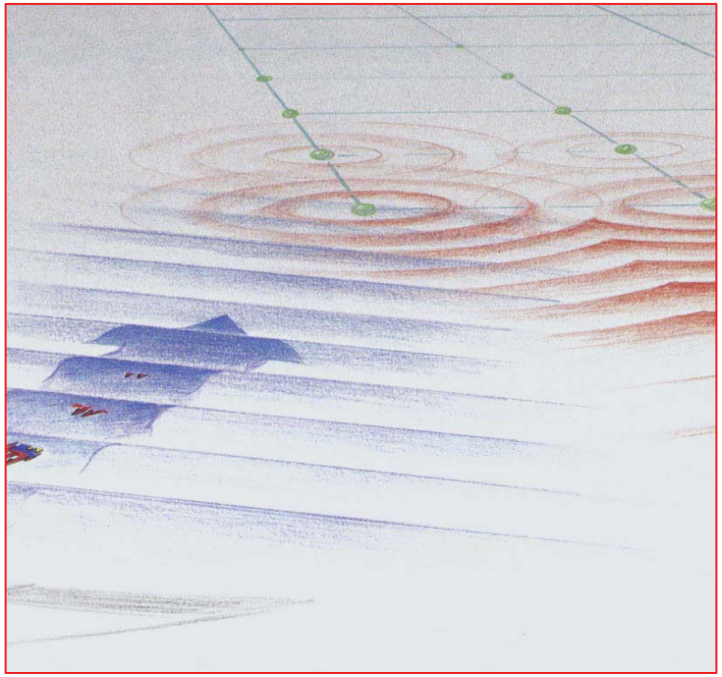
$k_f = k_i$

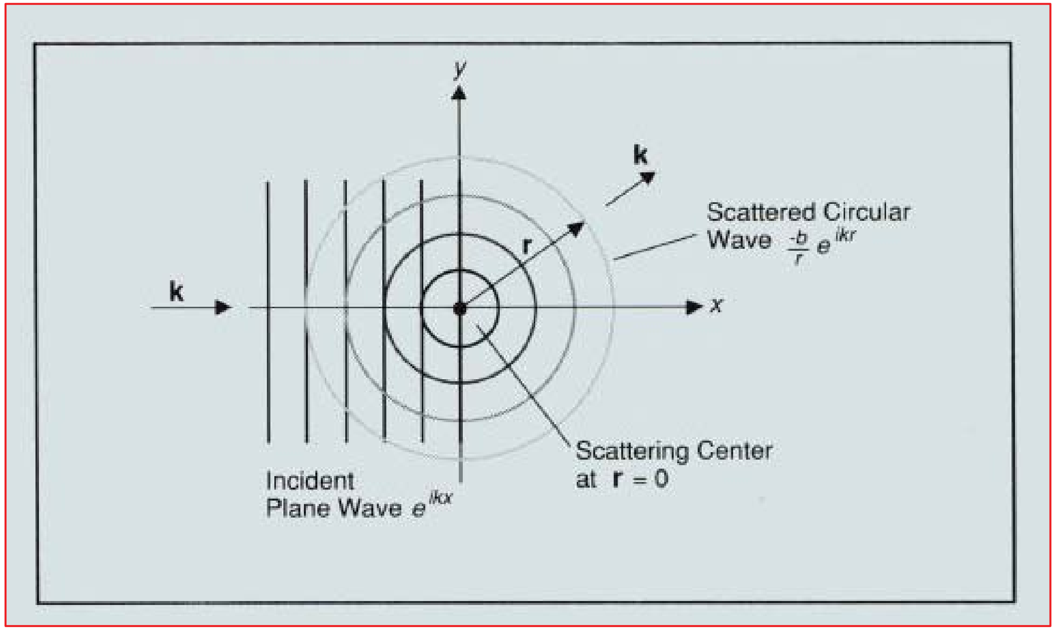
scattering corresponds to ψ, interference state between ψ_i and ψ_f the scattered wave

ψ_1 is a spherical wave:

$\psi_1 = \exp i\, \boldsymbol{k.r}/r\ f(\boldsymbol{k}_f,\boldsymbol{k}_i)$

So $\boldsymbol{Q} = \boldsymbol{k}_f - \boldsymbol{k}_i = 2\pi \boldsymbol{H}$, f(θ),





Far from the scattering centres (in sample), waves should be considered as plane waves

Born Approximation

One considers $\boldsymbol{V}(\boldsymbol{r})$ as a small perturbation, so $\psi(\boldsymbol{r})$ = exp i $\boldsymbol{kr} + \partial\psi$ with $\partial\psi << 1$

Using the Green function formalism (that is given by $G_{\boldsymbol{k}0}(\boldsymbol{r})$ (e i$\boldsymbol{k}_0$.r)/4$\pi\boldsymbol{r}$)

when $\boldsymbol{r}$ goes to large values, on obtains :

$\partial\psi \rightarrow m/2\pi h^2 \int \boldsymbol{V}(\boldsymbol{r}) \exp i (\boldsymbol{k} - \boldsymbol{k}').\boldsymbol{r}' \, d\boldsymbol{r}' (\exp i\, \boldsymbol{k}.\boldsymbol{r})/r$ where $k' = k\, \boldsymbol{r}/r$

$\partial\psi \rightarrow (\exp i\, \boldsymbol{kr})/r\, f(\boldsymbol{k}, \boldsymbol{k}') = \exp i\, \boldsymbol{kr})/r\, f(\theta)$

The scattered wave amplitude is proportional to V($\boldsymbol{Q}$), the Fourier transform of the interaction potential, and besides:

$$\sigma(\theta) = f(\theta)^2$$

$^{1}_{0}n$: spin ½, no electrical charge ↔ wave function

Atom

nucleus : nuclear spin I
electron : electronic spin s_e

} interaction potentials V
cross sections

$\Psi = \Psi_i + \Psi_f$ → spherical scattered wave $\Psi_f = \exp ikr/r \, f(k_f,k_i)$
↘ plane incident wave $\Psi_i = \exp ik_i r$

$f(k_f,k_i)$ has dimension of length
~ scattered amplitude ≈ cross section

Comparisons

electron diffraction V is a coulombian potential
XRD V is electromagnetic (Maxwellian)
neutron V are dipolar (I, s_e)
no coulombian interaction
very short range nuclear forces = strong penetration

Amplitudes

$\boldsymbol{V}_N(\boldsymbol{r}) \rightarrow$? - the nuclear potential has a strongly short efficiency distance.

$V_N(\boldsymbol{r}) = 2\pi h^2 / m\, \delta(\boldsymbol{r})$ so, $\int \delta(\boldsymbol{r})$ expi $\boldsymbol{q}\, \boldsymbol{r}$ dr = 1 then $f_N(\boldsymbol{k}, \boldsymbol{k}') =$ constant = **b**

$\boldsymbol{V}_M(\boldsymbol{r}) \rightarrow$? - the magnetic dipole – dipole interaction : $\overset{1}{\underset{0}{n}}$. e- for which potential is

$\boldsymbol{V}_M(\boldsymbol{r}) = (\boldsymbol{M}_n \times \boldsymbol{V}) \cdot (\boldsymbol{M}_e \times \boldsymbol{V}) \cdot (1/r)$ leads to

$$f_N(\boldsymbol{k}, \boldsymbol{k}') = 2m/h^2\, (\boldsymbol{M}_n \times \boldsymbol{e}) \cdot (\boldsymbol{M}_e \times \boldsymbol{e}) \quad \text{with} \quad \boldsymbol{e} = \boldsymbol{Q}/Q$$

Since the magnetic electron density is far to be punctual, so it must be F.T.

$f_e(\boldsymbol{q})$, depends on the scattering vector (Bragg angle)

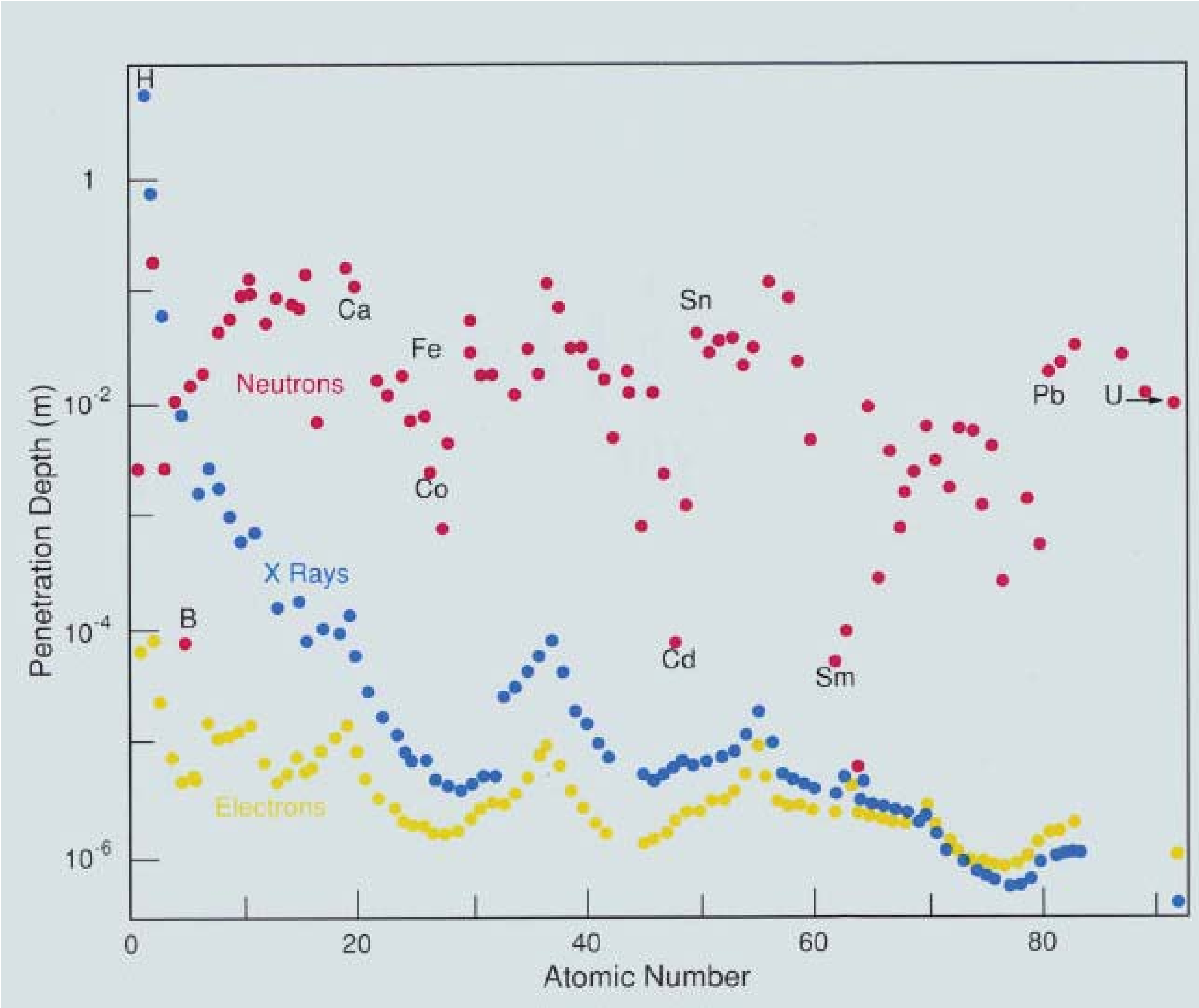
Remark 1: If $\boldsymbol{M}_e$ is // to $\boldsymbol{e}$ there is no magnetic scattering !

Remark 2: $\boldsymbol{M}_e = Me_\perp + Me_{//}$ leads to $(\boldsymbol{M}_n \times \boldsymbol{e}) \cdot (\boldsymbol{M}_e \times \boldsymbol{e}) = Mn \cdot Me_\perp$.

$$f_N(\boldsymbol{k}, \boldsymbol{k}') = e^2 m_0.c^2\gamma\ \sigma_n \cdot \sigma_{e\perp} \cdot f_e(\boldsymbol{Q})$$

$$e^2 m_0.c^2\gamma = 0.538\ 10^{-12}\ \text{cm} = 5.38\ \text{f (Fermi)}$$

Penetration Depth



Note for neutrons:

- **H/D difference**
- **Cd, B, Sm…**
- **no systematic A dependence**

Coherent / Incoherent

If, two waves are coherent, amplitudes add,

If two waves are incoherent, intensities add.

e.g. A_1 and A_2 with a phase difference $\Delta\Phi$ (whatsoever)

$I = (A_1 + A_2 + \cos\Phi)^2 = A_1^2 + 2\, A_1.A_2 \cos\Delta\Phi + A_2^2 \cos\Delta\Phi^2$

$<\cos\Delta\Phi> = 0$; $<\cos^2\Delta\Phi> = 1/2$ and $<I> = I_{inc.} = A_1^2 + 1/2\, A_2^2$

Suppose $A_1 = 1$ and $A_2 = \varepsilon$ and $\Delta\Phi$ = either 0 or π; (= control of coherence),

one defines $I^+ = I(0) = 1 + \varepsilon + \varepsilon^2$ and $I^- = I(\pi) = 1 - \varepsilon + \varepsilon^2$

$\Delta I = I^+ + I^- = 4\varepsilon$, exemple for $\varepsilon = 0.05$, ΔI leads to compare $1.25\ 10^{-3}$ to $2\ 10^{-1}$

Nuclear coherency

$I \leftrightarrow 2n+1$ states

$I + 1/2 \rightarrow b^+$

$\overset{0}{\underset{1}{n}} - I_N$: the total angular momentum has only 2 states

$I - 1/2 \rightarrow b^-$

So $b = b_{coh.} + b_{inc.}\, I \,.\, s_N$, then $b_{coh.} = b^+ (I+1)/(2I+1) + b^-\, I/(2I+1)$

$b_{inc.} = (b^+ - b^-)/(2I+1)$

$<I \,.\, s_N> = 0$; $<(I \,.\, s_N)^2> = I(I+1)$

So, $\sigma_{coh.} = 4\,\pi(p^+b^+ + p^-b^-)^2$ and $s_{inc.} = 4p\, p^+p^-\, (b^+ - b^-)^2$

with $p^+ = (I+1)/(2I+1)$ and $p^- = I/(2I+1)$

$F_N(\boldsymbol{H}) = b_1 \exp 2\pi i\, \boldsymbol{H.r}_1 + b_2 \exp 2\pi i\, \boldsymbol{H.r}_2$ A_1 and A_2 are not correlated, so :

$<F(\boldsymbol{H})^2> = b_1 coh.^2 \exp 2\pi i\, \boldsymbol{H.r}_1 + b_2 coh.^2 \exp 2\pi i\, \boldsymbol{H.r}_2 + b_{1inc.}{}^2\, I_1(I_1+1) + b_{1inc.}{}^2\, I_1(I_1+1)$

The incoherent scattering does not depend on any scattering vector ***H*** (at elastic collision for nuclear scattering processes)

Magnetic coherency

Considering the polarisation of a neutron beam before and after scattering, 4 states can be found for the scattered amplitudes:

$U{+}{+} = b_{coh.} + p\,\mu_{\perp z} + b_{inc.}\, I_z$

$U{+}{-} = p\,(\mu_{\perp x} - \mu_{\perp y}) + b_{inc.}\,(I_x + I_y)$

$U{-}{-} = b_{coh.} + p\,\mu_{\perp x} - b_{inc.}\, I_z$

$U{-}{+} = p\,(\mu_{\perp x} + \mu_{\perp y}) + b_{inc.}\,(I_x - I_y)$

where p is the amplitude for magnetic scattering

→ coherent magnetic scattering is always non spin-flip

→ scattering by electronic moment $\boldsymbol{\mu}$ may be spin-flip or non spin-flip depending on the relative orientation of $\boldsymbol{\mu}$, of polarisation $\boldsymbol{I}$ (→ ***oz***) and scattering vector ***Q***.

→ polarised neutron allows polarisation analysis

→ separation between coherent and nuclear spin incoherent scatterings

→ magnetic structures :

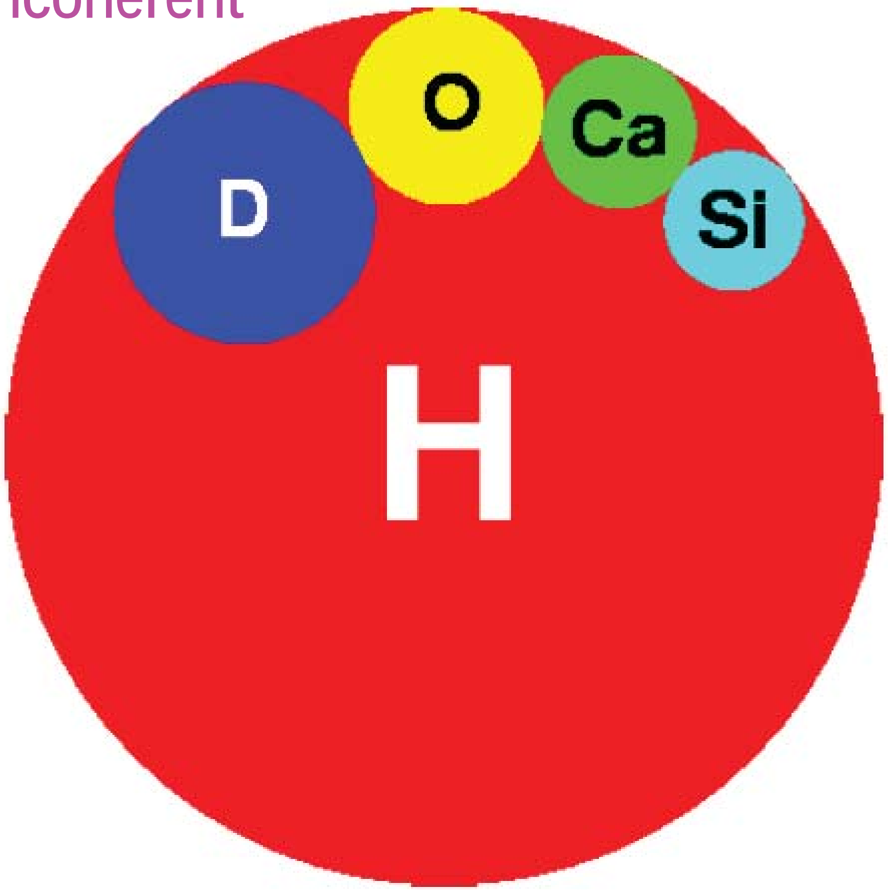
→ $\boldsymbol{I} \,//\, \boldsymbol{Q}$ – separation of nuclear and magnetic lines

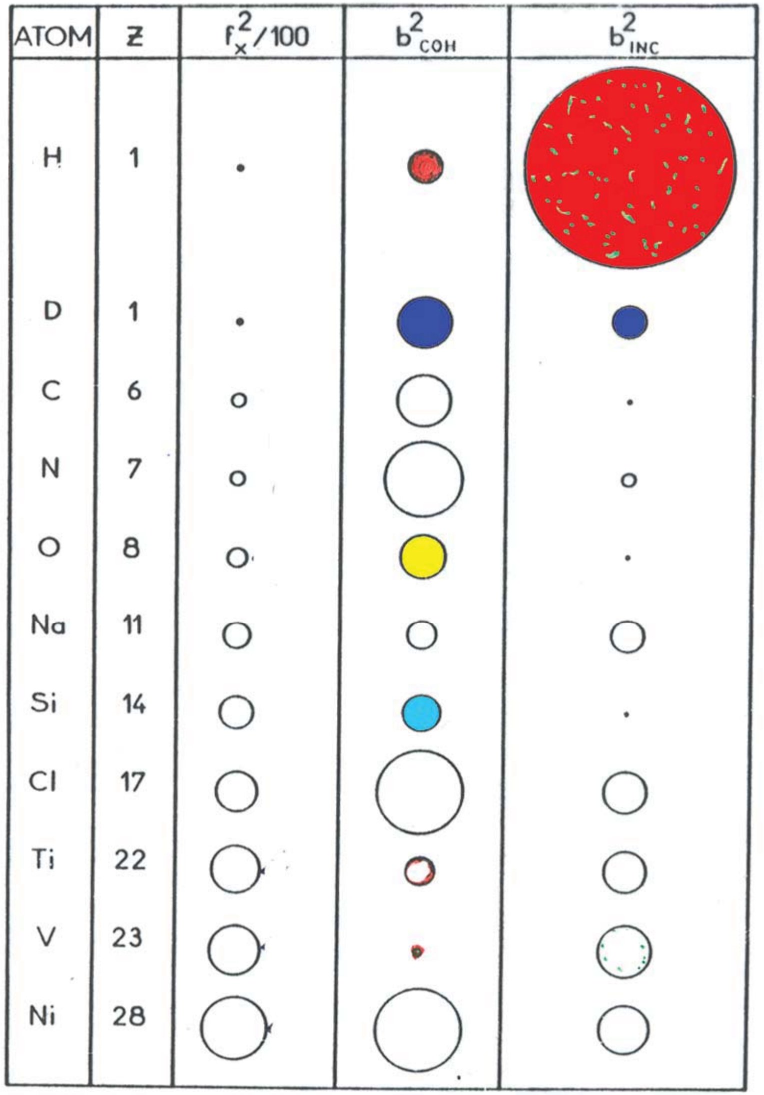
→ $\boldsymbol{I} \perp \boldsymbol{Q}$ – reveals spin flip contributions (non collinear systems)

→ incoherent paramagnetic scattering

On some scattering neutron cross sections

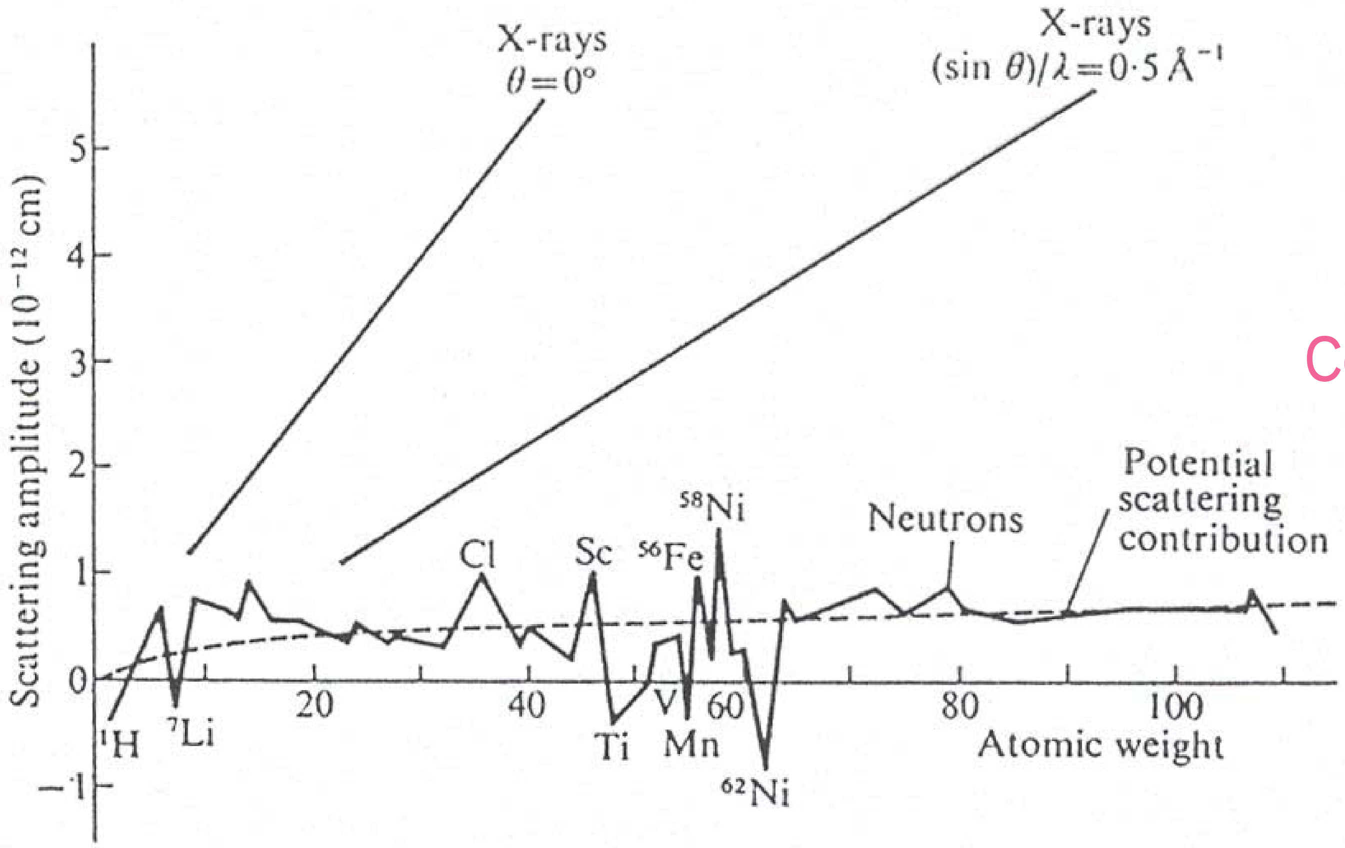
Incoherent





Comparaison des sections efficaces de diffusion cohérente et incohérente des neutrons avec celles de diffusion cohérente des rayons X pour une sélection d'atomes.

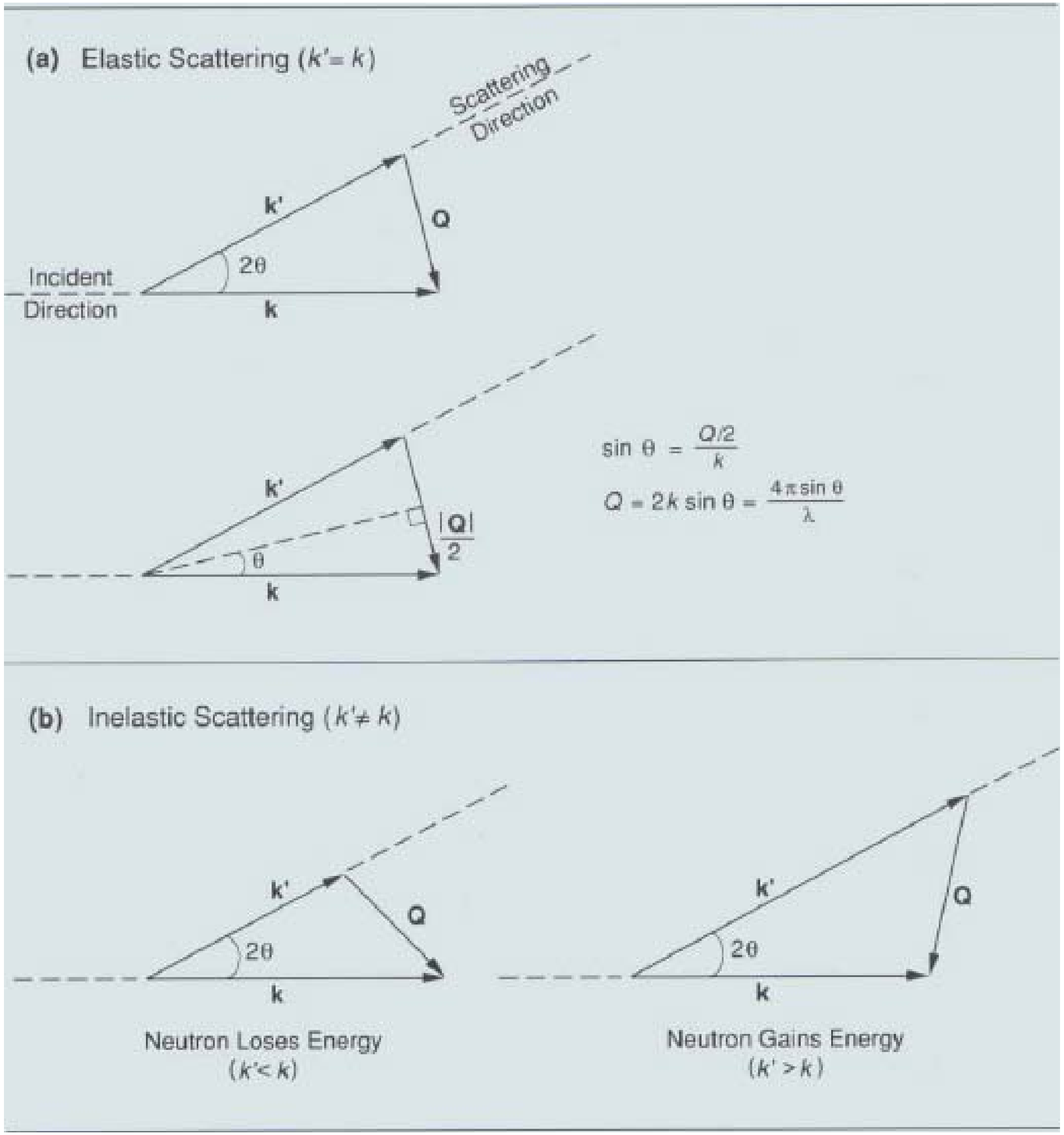
Nuclear Scattering Amplitude



Coherent

Irregular variation of neutron scattering amplitude with atomic weight due to superposition of 'resonance scattering' on the slowly increasing 'potential scattering'; for comparison the regular increase for X-rays is shown. (From *Research* (*London*) 7, 257 (1954).)

Inelastic



Neutrons can
Gain or Lose Energy
in the (Inelastic)
Scattering Process

What we can lean from Inelastic Scattering ?

- Neutron energies ~ meV
  1 meV ~ 8 cm^{-1} ~ 240 GHz ~ 12 K ~ 0.1 kJ/mol ~ ps

- Comparable to the time scales of atomic motions in materials

- Significant fractional changes in neutron energy result from exchanging energy with moving atoms in a material

- Compare:
  – Light: E ~ eV's; λ ~ 100's nm so Q ~ 0; also selection rules
  – X-rays: E ~ keV's; λ ~ 0.1 nm

- Note that cold neutrons (E < 5 meV; λ > 0.4 nm) allow one to observe *both* longer length scales and slower dynamics

Inelastic Neutron Scattering measures Atomic Motions

The concept of a pair correlation function can be generalized:
G(r,t) = probability of finding a nucleus at (r,t) given that there is one at r=0 at t=0
G_s(r,t) = probability of finding a nucleus at (r,t) if the *same* nucleus was at r=0 at t=0
Then one finds:

$$\left(\frac{d^2\sigma}{d\Omega dE}\right)_{coh} = b_{coh}^2 \frac{k'}{k} N S_{coh}(\vec{Q},\omega)$$

$$\left(\frac{d^2\sigma}{d\Omega dE}\right)_{inc} = b_{inc}^2 \frac{k'}{k} N S_{inc}(\vec{Q},\omega)$$

ħQ & ħω are the momentum & energy transferred to the neutron during the scattering process

where

$$S_{coh}(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\iint G(\vec{r},t)e^{i(\vec{Q}.\vec{r}-\omega t)}d\vec{r}dt \quad \text{and} \quad S_{inc}(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\iint G_s(\vec{r},t)e^{i(\vec{Q}.\vec{r}-\omega t)}d\vec{r}dt$$

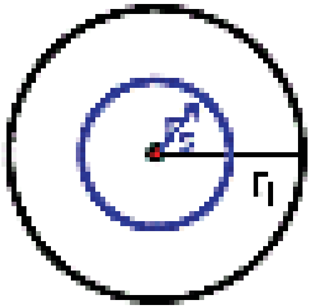
Inelastic coherent scattering measures *correlated* motions of atoms
Inelastic incoherent scattering measures *self-correlations* e.g. diffusion

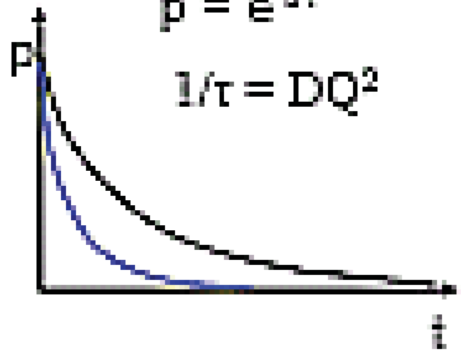
Examples of $S(Q,\omega)$ and $S_s(Q,\omega)$

- Expressions for $S(Q,\omega)$ and $S_s(Q,\omega)$ can be worked out for a number of cases e.g:
  - Excitation or absorption of one quantum of lattice vibrational energy (phonon)
  - Various models for atomic motions in liquids and glasses
  - Various models of atomic & molecular translational & rotational diffusion
  - Rotational tunneling of molecules
  - Single particle motions at high momentum transfers
  - Transitions between crystal field levels
  - Magnons and other magnetic excitations such as spinons
- Inelastic neutron scattering reveals details of the shapes of interaction potentials in materials

Quasielastic Neutron Scattering

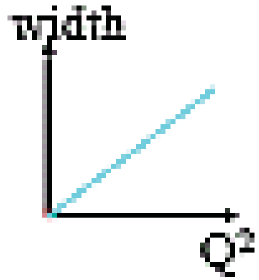
- For a single diffusing particle, the probability, p, of finding it within a sphere around its starting position looks like….





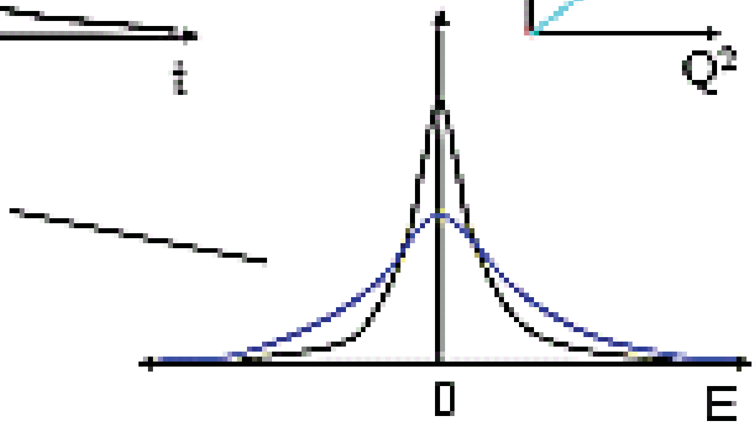
$$p = e^{-t/\tau}$$

$$1/\tau = DQ^2$$



- $S_{inc}(Q,E)$ 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 -> 8 the scattering will include an elastic component

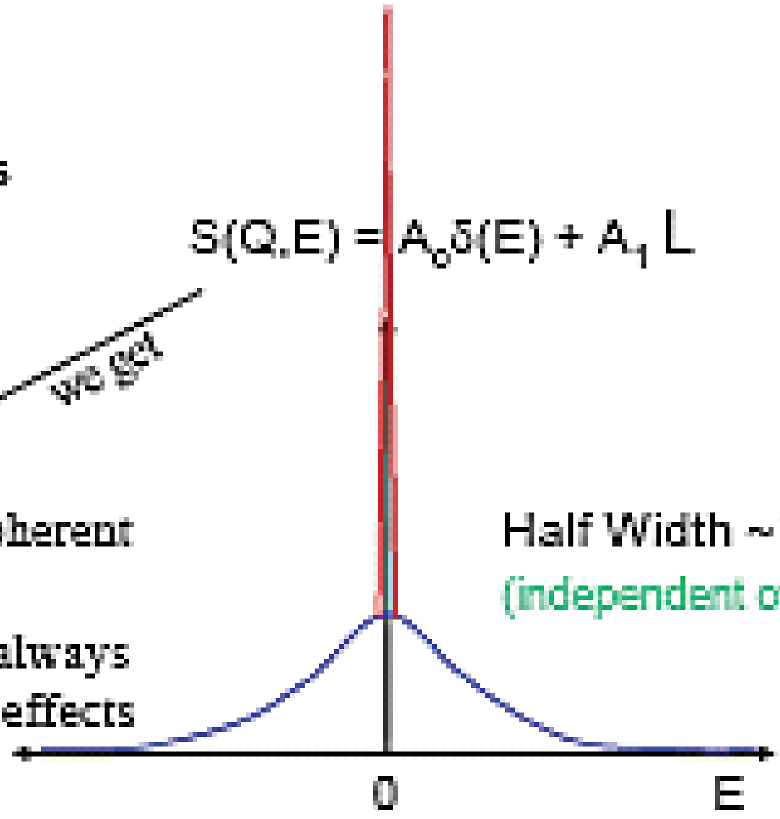
- For example, if two sites may be occupied and

$$p_1 = \tfrac{1}{2} + \tfrac{1}{2}\, e^{-2t/\tau}$$

$$p_2 = \tfrac{1}{2} - \tfrac{1}{2}\, e^{-2t/\tau}$$

we get

$$S(Q,E) = A_0\delta(E) + A_1\, L$$



Half Width ~ $1/\tau$

(independent of Q)

A_0 is called the Elastic Incoherent Structure Factor (EISF)

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

Recall, other interesting techniques as:

SANS (Small Angle Neutron Scattering): size of agregates, grains, particles, domains at µm to nm

Diffuse Scattering: ponctual to extended defects, static or dynamic characters

Reflectometry: thin layer analysis

Neutronography: absorption contrasts

etc…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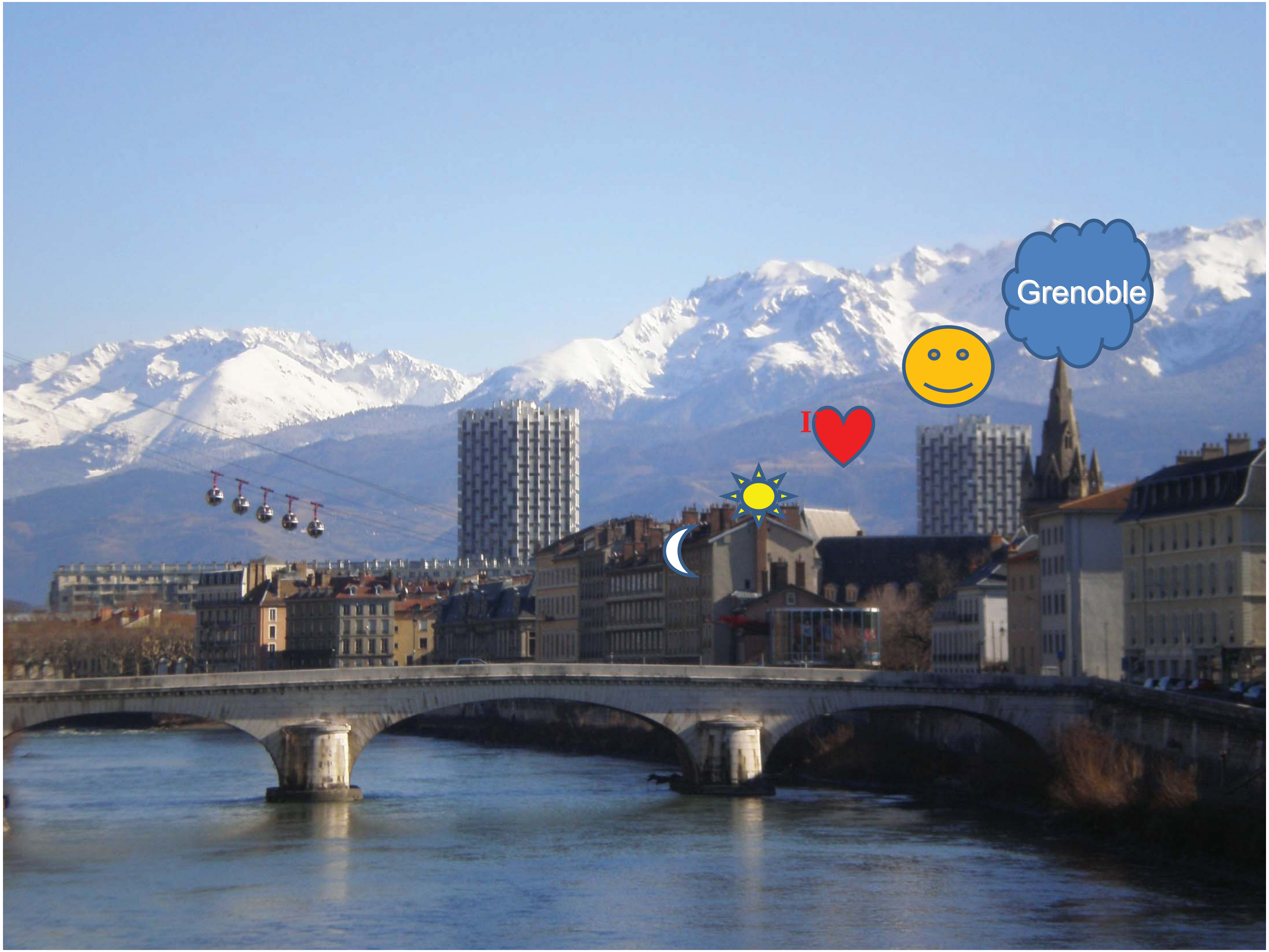
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