NOTES ON
FUNDAMENTALS OF
NEUTRON INTERACTION WITH MATTER

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INTRODUCTION

The neutron as a tool for research contributes to the understanding and development of almost all aspects of basic and applied sciences. This is despite the relative inaccessibility of neutron sources and the fact that even the most intense sources today provide only relatively weak beams of neutrons.

The use of neutron techniques began in the West as a parasitic activity in nuclear reactors, initially designed to operate as irradiation facilities or test beds for the development of nuclear technology. During the '50s and '60s, research based on the use of neutron beams involved only a small number of scientists, most of them belonging to the reactor operating staff. In subsequent decades, the enormous potentialities of these techniques could be realized through the massive participation of scientists from the academic and industrial communities, who opened the way to the very vast range of current applications.

This is due to the unique combination of properties that thermal neutrons possess. Since neutrons do not ionize the matter with which they interact, they can penetrate deep into it and explore its properties unaffected by surface effects. Likewise, through inelastic dispersion processes or by nuclear reactions, neutrons can create distortions or modifications on the volume of the irradiated sample, while due to their intrinsic dipole magnetic moment, it allows transparent access to the study of the magnetic properties of materials. Finally, it is a unique feature of thermal neutrons their ability to simultaneously explore the structure and dynamics of condensed matter, having an associated wavelength comparable to interatomic distances and a kinetic energy in the range of excitations characteristic of atomic motions.

Although the theoretical foundations of neutron scattering techniques are developed in texts and review publications [1-3], we wish to present here a compact body of expressions aimed at supporting and explaining the basic or most frequently used forms for the interpretation of experimental results. The formulation presented here, considers only

the nuclear scattering of neutrons for condensed matter, but it should be borne in mind that experiments designed for the study of magnetic properties of materials currently play a role of equivalent importance to those.

1. BASIC THEORY

1.1. General

The neutron has no net electric charge, and its electric dipole moment is too small to be measured, even with the most sensitive techniques. This implies that it can penetrate deep into matter, since it must not overcome the Coulombian barrier that charged particles face. Neutrons are scattered by interaction potentials due mainly to nuclear forces, whose intensity depends on the type of nuclide and not in monotonic form of its atomic number. This means that some light elements, such as hydrogen (practically transparent to X-rays), can be strong neutron scatterers. This neutron penetrating power allows its use even in situations in which the beam must first pass through a container, as in the case of a fluid, or a sample that must be maintained under extreme conditions of pressure or temperature.

The energy of thermal neutrons is of the same order as many of the typical excitation energies of condensed matter. Thus, if the neutron interacts inelastically creating or annihilating an excitation, its energy change will be a significant fraction of what it initially possessed, then the measurement of its final energy will provide detailed information about the energies of the excitations in matter and, therefore, of the interatomic forces. On the other hand, the magnetic moment of neutrons allows to investigate the magnetic structure of the material, as well as the characteristic energies of its excitations.

The basic scattering experiment (**Figure 1.1**) consists of impinging a collimated beam of neutrons on a sample (*scattering system*). This system consists of a set of atoms that can form a crystal, an amorphous solid, a liquid or a gas. As a result of the different types of measurements that are made on the scattered neutrons, the corresponding *cross sections* are obtained.

Suppose a monoenergetic incident beam of neutrons of energy *E*. The *incident flux* (Φ) is the number of neutrons that falls per unit time and unit area perpendicular to the beam. Given the angles θ and φ, the *double differential cross section*

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\ , \tag{1}$$

is defined as the number of neutrons scattered per unit time at the solid angle $\mathrm{d}\Omega$ in the direction (θ, φ) with final energies between E' and E'+d*E'*, per unit solid angle and energy.

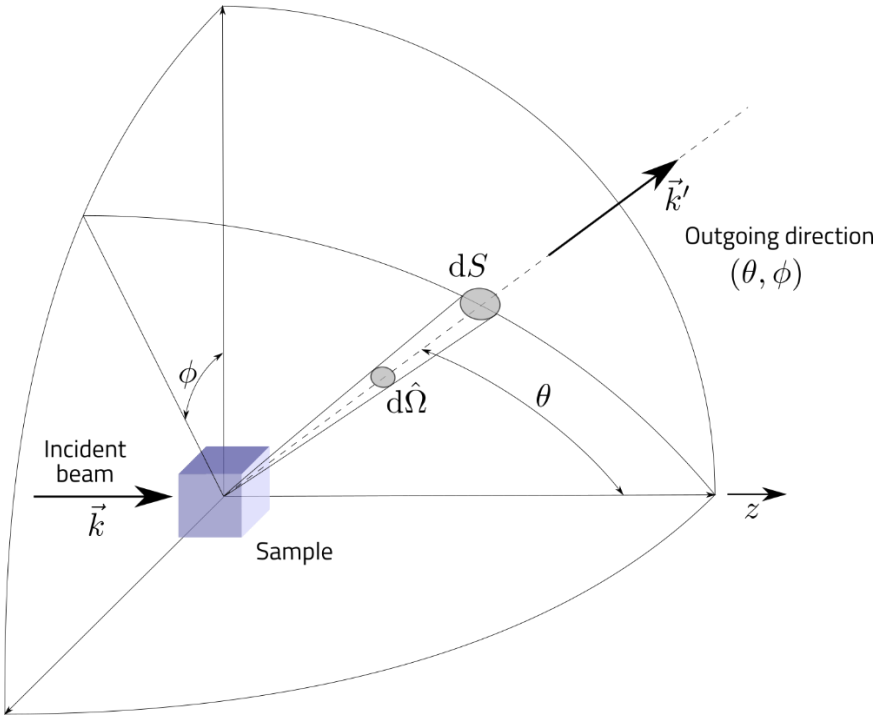

Figure 1.1: Basic scheme of a scattering experiment

If the final energy of the emerging neutrons of the system is not analyzed, it is useful to define the *differential cross section* as the number of neutrons scattered per unit of time and per unit solid angle in dΩ around the direction (θ, φ). This magnitude is obtained from Eq. (1.1) by integration on final energies

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \int \frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \mathrm{d}E'} \mathrm{d}E' \,. \tag{2}$$

For the case in which the direction of the emerging neutrons is not analyzed, the *energy-transfer kernel* (or simply *transfer kernel*) is defined as the number of scattered neutrons, per unit of time and per unit of energy, with energies between E' and E'+d*E'*, which is obtained by integrating the Ec. (1.11) over all angles

$$\frac{\mathrm{d}\sigma}{\mathrm{d}E'} = \int \frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega \, \mathrm{d}E'} \mathrm{d}\Omega \,. \tag{3}$$

Finally, the *total cross section* is defined as the total number of neutrons scattered per unit time in any direction and with any energy, and is obtained by integrating Eq. (1.2) on the angles of dispersion or Eq. (1. 3) on final energies

$$\sigma_{\text{tot}} = \int \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\mathrm{d}\Omega = \int \frac{\mathrm{d}\sigma}{\mathrm{d}E'}\mathrm{d}E' \quad . \tag{4}$$

The basic problem of scattering theory is to obtain theoretical expressions for the cross sections, which are the quantities that are measured directly in scattering experiments. Since experimental cross sections are expressed by scattering center, the above definitions should be divided by the total number of atoms or molecules in the scattering system.

Scattering by a fixed nucleus

To give a physical interpretation to the effective section, consider the case of dispersion by a single nucleus whose position is fixed. The nuclear forces that cause the scattering process have a range of 10^{-14} to 10^{-15} m, which is much less than the wavelength of thermal neutrons (on the order of 10^{-10} m). In these circumstances, scattering comes entirely from s-waves, i.e. with a spherically symmetrical angular distribution.

Placing the origin of the coordinate system in the scattering nucleus, at an instant of time the incident neutrons are represented by a wave function $\psi_{\text{inc}} = e^{i\boldsymbol{kr}}$, which is a plane wave of amplitude 1, indicating that there is the same probability of finding a neutron with momentum ***k*** anywhere in space. This wave is represented in **Figure 1.2** by flat wavefronts. In fact, the amplitude of this wave should be fixed so that it gives a probability of finding a neutron at the ***r*** position that is consistent with the number of neutrons present in the beam being used.

Since the scattering is spherically symmetric, at point ***r*** the wavefunction of the scattered neutrons is written as

$$\psi_{\text{sc}} = -\frac{b}{r}\exp(ikr) \quad , \tag{5}$$

where *b* is a constant, independent of angles θ, ϕ. The spherical wavefronts of the scattered neutron are represented in **Figure 1.2** by circles exiting the nucleus. The minus sign is arbitary, chosen so that positive values of *b* correspond to repulsive potentials. The factor 1/*r* takes into account the fact that the intensity of the scattered neutron beam decreases with the inverse of the square of the distance to the source.

The magnitude *b* in ψ_{sc} is called *scattering length* and, according to it, two types of nuclei can be identified. For the first type, *b* is complex and varies rapidly with neutron energy. Most nuclei are of the second type, for which the composite nucleus does not form in an energy state near an excited state. The scattering length is independent of the energy of the neutron and its imaginary part is small, so that the interaction does not

involve a significant probability of absorption. To this situation we will restrict ourselves, considering *b* as a real magnitude.

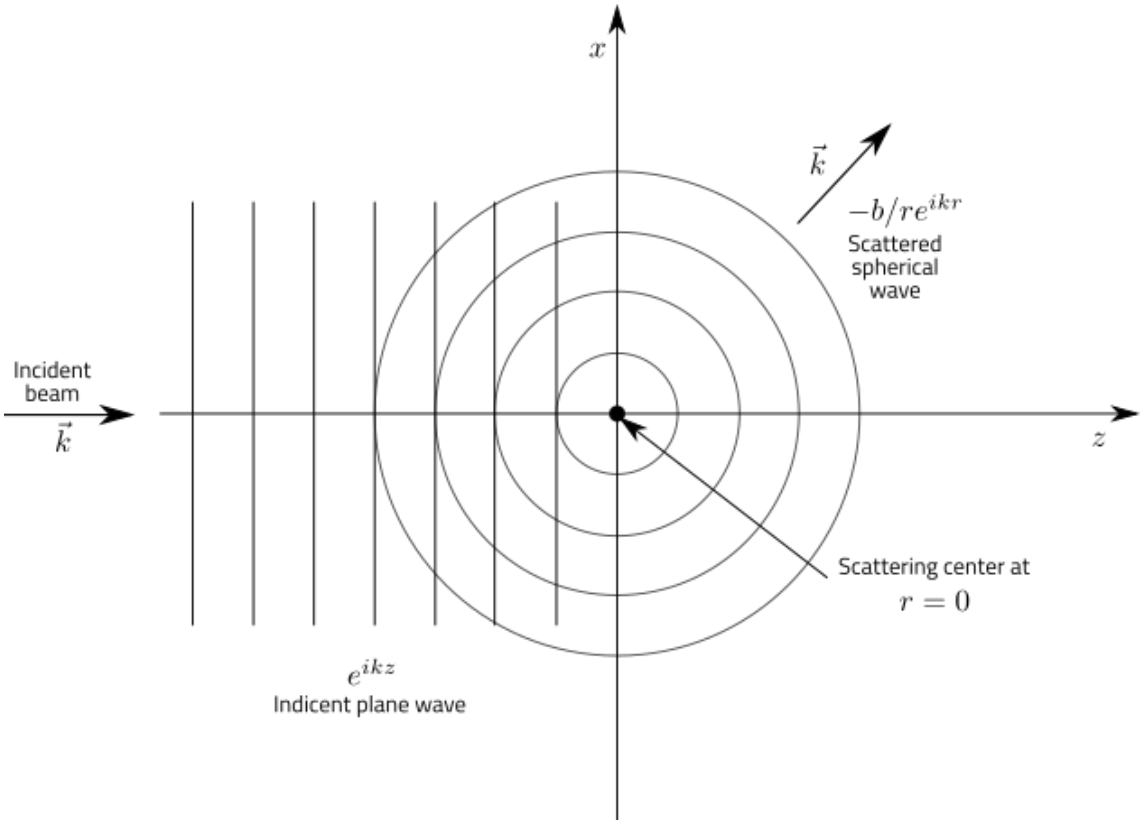

Figure 1.2: A neutron beam striking a fixed nucleus can be represented by a plane wave of unit amplitude. Since the neutron sees the nucleus as a point, scattering is isotropic, so that the scattered wave is spherical (represented by circumferences). As has been assumed a fixed core, the interaction is elastic.

The value of the scattering length depends on the particular nucleus and the spin state of the neutron-nucleus system. If the nucleus has spin I (non-zero), that of the neutron-nucleus system is $I+1/2$ or $I-1/2$, each of these states having its own value of b due to the dependence on the spin of nuclear forces. If the nuclear spin is zero, there is a single scattering length [4].

Using the expressions corresponding to the incident and scattered wave functions, the cross-section $d\sigma/d\Omega$ for the scattering produced by a fixed nucleus is calculated. If v is the speed of the neutrons (the same before and after scattering), the number of neutrons per second passing through the surface dS is

$$v\,\mathrm{d}S\left|\psi_{\mathrm{sc}}\right|^2 = v\,\mathrm{d}S\frac{b^2}{r^2} = v b^2\,\mathrm{d}\Omega \quad , \tag{6}$$

while the incident neutron flux is

$$\Phi = v\left|\psi_{\mathrm{inc}}\right|^2 = v \quad . \tag{7}$$

From the definition of cross section it follows that

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{v b^2 \mathrm{d}\Omega}{\Phi\,\mathrm{d}\Omega} = b^2 \quad , \tag{8}$$

that integrated over all angles allows to obtain for this simple case

$$\sigma_{\mathrm{tot}} = 4\pi b^2 \quad . \tag{9}$$

This result coincides with the section that presents to the incident beam a rigid sphere of radius 2*b*, giving a concrete physical meaning to the scattering length.

Nuclear Scattering

When neutrons are scattered by a general system of particles, the processes can alter the energy and momentum of both the neutron and the scattering system. Scattering is not necessarily elastic as in the case of a fixed nucleus, because atoms have the freedom to move, so they can recoil when they collide with a neutron or, if they are in motion, they can absorb or deliver energy to the neutron.

Let ***k*** be the wave vector of a neutron incident on a scattering system in a state characterized by the index λ, $\psi_{\boldsymbol{k}}$ the wave function of the neutron and χ_λ that of the scattering system. We will assume that the neutron interacts with the system *via* a potential *V* and that it is scattered so that its final wave vector is ***k***', while the final state of the scatterer system is λ'. As shown in **Figure 1.3**, if the coordinate system is placed at some arbitrary point in the scattering system, the positions of the N nuclei of the system are represented by $\boldsymbol{R}_j$ (*j*=1, ... , *N*) and that of the neutron by ***r***.

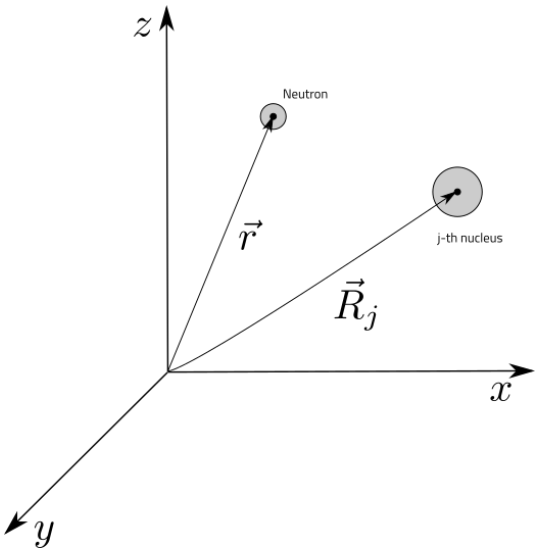

Figure 1.3: Diagram of neutron location and scattering centers.

The differential cross section $(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\lambda\to\lambda'}$ represents the sum of all processes in which the state of the scattering system changes from λ to λ', and the state of the neutron changes from ***k*** to ***k'***. This sum is performed over all values of ***k'*** that lie within the solid angle $\mathrm{d}\Omega$ in the direction (θ,ϕ), keeping constant the values of ***k***, λ and λ'. From the definition of the differential cross section, we have

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\lambda\to\lambda'} = \frac{1}{\Phi}\frac{1}{\mathrm{d}\Omega}\sum_{\substack{\boldsymbol{k}' \\ \text{en } \mathrm{d}\Omega}} W_{\boldsymbol{k},\lambda\to\boldsymbol{k}',\lambda'} \quad , \tag{10}$$

where $W_{\boldsymbol{k},\lambda\to\boldsymbol{k}',\lambda'}$ is the number of transitions per second from state {***k,*** λ} to {***k***',λ'}, and Φ is the incident neutron flux. The summation can be evaluated using *Fermi's Golden Rule*, according to which [1]

$$\sum_{\substack{\boldsymbol{k}' \\ \text{en } \mathrm{d}\Omega}} W_{\boldsymbol{k},\lambda\to\boldsymbol{k}',\lambda'} = \frac{2\pi}{\eta}\,\rho_{\boldsymbol{k}'}\left|\langle \boldsymbol{k}'\,\lambda|V|\boldsymbol{k}\lambda\rangle\right|^2 \quad . \tag{11}$$

In this expression $\rho_{\boldsymbol{k}'}$ is the number of states with momentum ***k'*** in $\mathrm{d}\Omega$ per unit energy. The potential *V* represents the interaction between the incident neutron and the scattering system, and it is the one that causes the transition from the initial states to the final ones. The matrix element is explicitly given by

$$\langle \boldsymbol{k}'\,\lambda'|V|\boldsymbol{k}\lambda\rangle = \int \psi^*_{\boldsymbol{k}'}\,\chi^*_{\lambda'}\,V\,\psi_{\boldsymbol{k}}\,\chi_{\lambda}\,\mathrm{d}\boldsymbol{R}\,\mathrm{d}\boldsymbol{r} \quad , \tag{12}$$

where

$$\mathrm{d}\boldsymbol{R} = \mathrm{d}\boldsymbol{R}_1\,\mathrm{d}\boldsymbol{R}_2\,\mathrm{K}\,\mathrm{d}\boldsymbol{R}_N \quad , \tag{13}$$

and each d***R****j* is a volume element for the j-th nucleus and d***r*** is for the neutron.

We will suppose the neutron and the scattering system in a box, in order to calculate $\rho_{\boldsymbol{k}'}$ and fix the constant of normalization of the neutron wavefunctions, since only those states whose de Broglie wavelengths are periodic in that box will be allowed. The wave vectors of such states form a lattice in space ***k***, and the volume of the unit cell is

$$v_{\boldsymbol{k}} = \frac{(2\pi)^3}{Y} \quad , \tag{14}$$

where *Y* is the volume of the box.

By definition, $\rho_{\boldsymbol{k}'}\,\mathrm{d}E'$ is the number of states in $\mathrm{d}\Omega$ with energy between E' and E'+d*E*', which coincides with the number of wave vectors in the volume element $k'^2\,\mathrm{d}k'\,\mathrm{d}\Omega$. Then

$$\rho_{\boldsymbol{k}'}\,\mathrm{d}E' = \frac{1}{v_{\boldsymbol{k}}}\,k'^2\,\mathrm{d}k'\mathrm{d}\Omega \quad , \tag{15}$$

a result leading to

$$\rho_{\boldsymbol{k}'} = \frac{Y}{(2\pi)^3} k' \frac{m}{\eta^2} \mathrm{d}\Omega \quad . \tag{16}$$

For scattering processes, Fermi's golden rule is equivalent to Born's approximation [Born, 1943], so both the incident and emergent waves are represented as plane waves; then, the matrix element of Eq. (1. 12) is

$$\langle \boldsymbol{k}'\,\lambda' | V | \boldsymbol{k}\lambda \rangle = \frac{1}{Y} \int \exp(-i\boldsymbol{k}'\cdot\boldsymbol{r})\, \chi^*_{\lambda'}\, V \exp(i\boldsymbol{k}\cdot\boldsymbol{r})\, \chi_\lambda \, \mathrm{d}\boldsymbol{R}\, \mathrm{d}\boldsymbol{r} \quad . \tag{17}$$

The incident neutron flux is the product of number density times velocity, i.e.

$$\Phi = \frac{1}{Y} \frac{\eta}{m} k \quad . \tag{18}$$

To obtain the differential effective section, we substitute the Ecs. (1.1111), (1. 16), (1. 17) and (1. 18) in (1. 10), which leads to

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \right)_{\lambda \to \lambda'} = \frac{k'}{k} \left(\frac{m}{2\pi\, \eta^2} \right)^2 \left| \langle \boldsymbol{k}'\,\lambda | V | \boldsymbol{k}\lambda \rangle \right|^2 \quad . \tag{19}$$

This is the cross section for neutrons scattered within $\mathrm{d}\Omega$ in the direction of $\boldsymbol{k}'$, but, as mentioned above, because $\boldsymbol{k}$, λ and λ' are fixed, all scattered neutrons have the same energy, determined by the conservation condition. If E_λ, $E_{\lambda'}$, E y E' are the initial and final energies of the system and neutron respectively, conservation can be expressed in terms of the Dirac function, resulting in δ

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \right)_{\lambda \to \lambda'} = \frac{k'}{k} \left(\frac{m}{2\pi\, \eta^2} \right)^2 \left| \langle \boldsymbol{k}'\,\lambda | V | \boldsymbol{k}\lambda \rangle \right|^2 \, \delta\left(E_{\lambda'} - E_\lambda + E - E' \right) \tag{20}$$

The interaction potential between the neutron and the j-th nucleus has the form $Vj(\boldsymbol{r}-\boldsymbol{R}j)$, so that for the complete system the potential is

$$V(\boldsymbol{r}) = \sum_j V_j(\boldsymbol{r} - \boldsymbol{R}_j) \quad . \tag{21}$$

Using this expression in that of the matrix element yields

$$\langle \boldsymbol{k}'\,\lambda' | V | \boldsymbol{k}\lambda \rangle = \sum_j V_j(\boldsymbol{Q}) \langle \lambda' | \exp(i\boldsymbol{Q}\cdot\boldsymbol{R}_j) | \lambda \rangle \quad , \tag{22}$$

where

$$V_j(\boldsymbol{Q}) = \int V_j(\boldsymbol{x}_j) \exp(i\boldsymbol{Q}\cdot\boldsymbol{x}_j)\, \mathrm{d}\boldsymbol{x}_j \tag{23}$$

is the Fourier transform of the potential,

$$\langle \lambda' | \exp(i\boldsymbol{Q}\cdot\boldsymbol{R}_j | \lambda \rangle = \int \chi^*_{\lambda'} \exp(i\boldsymbol{Q}\cdot\boldsymbol{R}_j)\, \chi_\lambda \, \mathrm{d}\boldsymbol{R} \tag{24}$$

and

$$\boldsymbol{Q} = \boldsymbol{k} - \boldsymbol{k}' \quad . \tag{25}$$

The quantity $\eta \boldsymbol{Q}$ is known as the *momentum transfer vector*.

The next step is to use a specific function for the neutron-nucleus interaction potential. Fermi [1936] showed that the interaction between thermal neutrons and nuclei can be satisfactorily represented by a pseudo-potential, in which all nuclear physics is described by a single parameter: the scattering length. This Fermi *pseudo-potential* is written as

$$V_j(\boldsymbol{r}-\boldsymbol{R}_j)=\frac{2\pi\,\eta^2}{m}b_j\,\delta(\boldsymbol{r}-\boldsymbol{R}_j) \quad , \tag{26}$$

where m is the mass of the neutron and b_j is the scattering length of the j-th nucleus. Thus, for an array of nuclei, such as a crystal, the potential $V(\boldsymbol{r})$ is the sum of the individual neutron-nucleus interactions.

$$V(\boldsymbol{r})=\frac{2\pi\,\eta^2}{m}\sum_j b_j\,\delta(\boldsymbol{r}-\boldsymbol{R}_j) \quad , \tag{27}$$

where the sum is performed over all sites of the crystal.

The scattering length defined in Eq. (1.5) is related to a fixed core and is known as *bound* scattering length. If the nucleus is free the problem must be treated in the center of mass system, and the mass of the neutron must be replaced by the reduced mass of the neutron-nucleus system.

$$\mu=\frac{m\,M}{m+M} \quad , \tag{28}$$

where M is the mass of the nucleus. The scattering length for this process must be replaced by that corresponding to the free atom, called the free scattering length, given by

$$b_f=\frac{M}{m+M}\,b \quad . \tag{29}$$

Inserting Fermi's pseudo-potential into Eq. (1. 23) results

$$V_j(\boldsymbol{Q})=\frac{2\pi\,\eta^2}{m}b_j \quad , \tag{30}$$

that in combination with Ecs. (1.2020) and (1. 22) produces

$$\left(\frac{d^2\sigma}{d\Omega\,dE'}\right)_{\lambda\to\lambda'}=\frac{k'}{k}\left|\sum_j b_j\left\langle\lambda'\middle|exp\left(i\boldsymbol{Q}.\boldsymbol{R}_j\right)\middle|\lambda\right\rangle\right|^2\delta(E_\lambda-E_{\lambda'}+E'-E) \quad . \tag{31}$$

Using the integral form of the Dirac δ distribution, defining the energy change

$$\eta\omega=E-E' \tag{32}$$

and developing the square in Eq. (1. 31) results

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\sum_{j,j'} b_{j'}^{*}\, b_j \langle\lambda'|\exp(-i\boldsymbol{Q}\cdot R_{j'})|\lambda\rangle\langle\lambda'|\exp(i\boldsymbol{Q}\cdot R_j)|\lambda\rangle$$
$$\times\frac{1}{2\pi\,\eta}\int_{-\infty}^{\infty}\exp\left(i\frac{E_{\lambda'}-E_{\lambda}}{\eta}t\right)\exp(-i\omega t)\,\mathrm{d}t \quad . \tag{33}$$

The states $|\lambda\rangle$ and $|\lambda'\rangle$ are eigenfunctions of the Hamiltonian of the scattering system H_0, of eigenvalues E_λ and $E_{\lambda'}$, so that

$$\exp\left(-i\frac{H_0}{\eta}t\right)|\lambda\rangle = \exp\left(-i\frac{E_\lambda}{\eta}t\right)|\lambda\rangle \quad , \tag{34}$$

whereupon the Eq. (1. 33) results

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\frac{1}{2\pi\,\eta}\sum_{j,j'} b_{j'}\, b_j \int_{-\infty}^{\infty}\langle\lambda'|\exp(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j'})|\lambda\rangle$$
$$\times\langle\lambda'|\exp(i\frac{H_0}{\eta}t)\exp(i\boldsymbol{Q}\cdot\boldsymbol{R}_j)\exp(-i\frac{H_0}{\eta}t)|\lambda\rangle\exp(-i\omega t)\,\mathrm{d}t \quad . \tag{35}$$

In a real experiment you cannot measure the cross section for a process in which the scattering system passes from one specific state $|\lambda\rangle$ to another $|\lambda'\rangle$. What is measured is the cross section defined by Eq. (1.1), which is obtained by adding (1. 35final states $|\lambda'\rangle$, keeping the initial state fixed, and then averaging over all possible $|\lambda\rangle$ ones. Using the closure relation, the sum can be performed over the complete set of final eigenstates of the scattering system. The average over all initial states is made by taking into account the probability that the scattering system is in a given state, represented by the Boltzmann distribution

$$p_\lambda = \frac{1}{Z}\exp\left(-\beta E_\lambda\right) \quad , \tag{36}$$

where Z is the partition function

$$Z = \sum_\lambda \exp\left(-\beta E_\lambda\right) \quad , \tag{37}$$

$\beta = (k_B T)^{-1}$, T is the temperature of the system and k_B is the Boltzmann constant. This leads to the following expression

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'} = \sum_{\lambda,\lambda'} p_\lambda\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\frac{1}{2\pi\,\eta}\sum_{j,j'} b_{j'}\, b_j \int_{-\infty}^{\infty}\exp(-i\omega t)\,\mathrm{d}t\times$$
$$\times\left\langle\lambda\left|\exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j'}\right)\exp\left(iH_0 t/\eta\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j\right)\exp\left(-iH_0 t/\eta\right)\right|\lambda\right\rangle \quad , \tag{38}$$

which can be rewritten in terms of the time-dependent Heisenberg operator, $\boldsymbol{R}_j(t)$, defined by

$$\boldsymbol{R}_j(t) = \exp\left(iH_0 t/\eta\right)\boldsymbol{R}_j \exp\left(-iH_0 t/\eta\right) \tag{39}$$

that satisfies relation (because H_0 is an hermitic operator)

$$\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j(t)\right) = \exp\left(iH_0 t/\eta\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j\right)\exp\left(-iH_0 t/\eta\right) \quad . \tag{40}$$

We finally obtain

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'} = \frac{k'}{k}\frac{1}{2\pi\eta}\sum_{j,j'} b_{j'}\,b_j \int_{-\infty}^{\infty}\left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j'}(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j(t)\right)\right\rangle \exp(-i\omega t)\,\mathrm{d}t \quad , \tag{41}$$

where the brackets represent the thermal average.

The Eq. (1.41), called the *master equation,* is the basic expression for the double differential cross section for nuclear scattering, of complex evaluation, except for elementary scattering systems. The properties of the scattering system are contained in the Hamiltonian H_0 and therefore, in the Heisenberg operators and the eigenstates $|\lambda\rangle$.

Coherent and Incoherent cross sections

To obtain a measurable cross section, on Eq. (1.41) remains to average the possible isotope distributions in the system. This average results in

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'} = \frac{k'}{k}\frac{1}{2\pi\eta}\sum_{j,j'}\overline{b_{j'}\,b_j}\int_{-\infty}^{\infty}\exp(-i\omega t)\,\chi_{jj'}(t)\,\mathrm{d}t \quad , \tag{42}$$

where

$$\chi_{jj'}(t) = \left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j'}(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j(t)\right)\right\rangle \quad . \tag{43}$$

This quantity is usually called the *intermediate scattering function,* for reasons that we will discuss later, and represents the thermal and quantum average of the correlation between the particles´ time-dependent position operators.

Under the assumption that there is no correlation between the *b* values of different nuclides, the average indicated in Eq. (1.42) can be calculated as

$$\overline{b_{j'}\,b_j} = \begin{cases} \overline{b}^2 & j' \neq j \\ \overline{b^2} & j' = j \end{cases} \quad , \tag{44}$$

The cross section is then divided into two terms

$$\begin{aligned}\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'} = {} & \frac{k'}{k}\frac{1}{2\pi\eta}\overline{b}^2\sum_{j,j'}\int_{-\infty}^{\infty}\exp(-i\omega t)\,\chi_{jj'}(t)\,\mathrm{d}t \\ & + \frac{k'}{k}\frac{1}{2\pi\eta}\left(\overline{b^2}-\overline{b}^2\right)\sum_{j}\int_{-\infty}^{\infty}\exp(-i\omega t)\,\chi_{jj}(t)\,\mathrm{d}t \quad .\end{aligned} \tag{45}$$

The first term of this equation is known as the *coherent* scattering cross section and the second as the *incoherent* scattering cross section. In coherent scattering processes, the

neutron wave interacts with the entire scattering system as a unit, so that waves scattered by different nuclei interfere with each other. This type of scattering depends on the relative distances between the constituent atoms, giving information about the structure of the materials. Coherent elastic scattering gives information about the equilibrium structure, while coherent inelastic scattering gives information about the collective motions of atoms, such as those produced by vibrational waves or phonons in a crystal lattice. In incoherent scattering processes, the neutron wave interacts independently with each nucleus of the system, so that waves scattered by different nuclei do not interfere. Incoherent scattering is due to the interaction of the neutron wave with the *same* atom, but in different positions and at different times, thus providing information about dynamics of the system.

We rewrite the expressions for the coherent and incoherent cross sections, respectively, as

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh}} = \frac{\sigma_{\mathrm{coh}}}{4\pi}\frac{k'}{k}\frac{1}{2\pi\,\eta}\sum_{j,j'}\int_{-\infty}^{\infty}\exp(-i\omega t)\;\chi_{jj'}(t)\,\mathrm{d}t \tag{46}$$

and

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{inc}} = \frac{\sigma_{\mathrm{inc}}}{4\pi}\frac{k'}{k}\frac{1}{2\pi\,\eta}\sum_{j}\int_{-\infty}^{\infty}\exp(-i\omega t)\;\chi_{jj}(t)\,\mathrm{d}t\ , \tag{47}$$

where

$$\sigma_{\mathrm{coh}} = 4\pi\,\bar{b}^2 \tag{48}$$

and

$$\sigma_{\mathrm{inc}} = 4\pi\,(\overline{b^2} - \bar{b}^2)\ . \tag{49}$$

The unit normally used for σ is the barn, equivalent to $10^{-28}\,\mathrm{m}^2$.

The scattering lengths b_j represent, within the Fermi pseudopotential approach, the nuclear interaction of incidend neutrons with the nuclei that conform the sample material. As such interaction is governed by the nuclear potential, that is different for different nuclei present in the scatterer, the b_j will vary randomly not only for the atomic species but also for the isotopes. Moreover, because of the spin-dependence of the nuclear force, the scattering length will also depends on the relative orientation of the neutron spin and the nuclear spin (if different from zero). The two possible interactions, parallel and antiparallel will have associated scattering lengths $b^{(+)}$ and $b^{(-)}$, respectively, that combine with their corresponding statistical weights.

Although presented separately, coherent and incoherent scattering processes do not occur independently, but are different manifestations of the observed total scattering.

Some systems behave more or less coherently, according to the variety of nuclides present and their scattering lengths. It must be emphasized that the cross sections involve three essentially different factors: one (σ) related to the strength of the neutron-nuclei interaction, a second *k´/k* which is a kinematical factor, and the third (correlation functions) that depends solely on the characteristics of the scattering system. Thus, neutron scattering measurements yield direct information on the structural and dynamical properties of the sample material.

Correlation functions

The explicit forms of the coherent and incoherent intermediate scattering functions are, from Eqs.(1. 42) and (1.43)

$$\chi_{\mathrm{coh}}(\boldsymbol{Q},t)=\frac{1}{N}\sum_{j,j'}\left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j'}(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_{j}(t)\right)\right\rangle \tag{50}$$

and

$$\chi_{\mathrm{inc}}(\boldsymbol{Q},t)=\frac{1}{N}\sum_{j}\left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j}(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_{j}(t)\right)\right\rangle \ , \tag{51}$$

where *N* is the total number of scattering units in the system.

Their Fourier transforms with respect to *t* are, respectively, the *coherent* and *incoherent scattering functions* (or *dynamic structure factors,* or *scattering laws*):

$$S_{\mathrm{coh}}(\boldsymbol{Q},\omega)=\frac{1}{2\pi\,\eta}\int_{-\infty}^{\infty}\exp(-i\omega t)\ \chi_{\mathrm{coh}}(\boldsymbol{Q},t)\,\mathrm{d}t \tag{1.52}$$

and

$$S_{\mathrm{inc}}(\boldsymbol{Q},\omega)=\frac{1}{2\pi\,\eta}\int_{-\infty}^{\infty}\exp(-i\omega t)\ \chi_{\mathrm{inc}}(\boldsymbol{Q},t)\,\mathrm{d}t \ , \tag{52}$$

so that the double differential scattering cross section can be expressed as

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}=\frac{1}{4\pi}\frac{k'}{k}N\left(\sigma_{\mathrm{coh}}\,S_{\mathrm{coh}}(\boldsymbol{Q},\omega)+\sigma_{\mathrm{inc}}\,S_{\mathrm{inc}}(\boldsymbol{Q},\omega)\right) \ . \tag{53}$$

We will review some analytic properties of the scattering functions. Due to the Hermiticity of the position operators the following equality holds:

$$S(\boldsymbol{Q},\omega)=\ S^{*}(\boldsymbol{Q},\omega) \tag{1.55}$$

that means that $S(\boldsymbol{Q},\omega)$ is real, which is evident because it is proportional to the observable scattering cross section. Also,

$$S(-\boldsymbol{Q},-\omega)=\exp(-\beta\hbar\omega)\,S(\boldsymbol{Q},\omega) \tag{1.56}$$

known as *the principle of detailed balance*. This property expresses the microscopic

reversibility of the scattering process, and it is very useful from a practical point of view as it permits to generate the complete scattering function from its calculation over only part of the *(**Q**,ω)* plane.

The moments of *S(**Q**,ω)* produce a set of consistency checks for theoretical models. We define the n-th moment of the scattering function

$$S_n(\mathbf{Q}) = \eta \int_{-\infty}^{\infty} d\omega \, (\eta\omega)^n \, S(\mathbf{Q},\omega) = \left(\frac{\eta}{i}\right)^n \frac{\partial^n}{\partial t^n} \, \chi(\mathbf{Q},t) \, \Big|_{t=0} \tag{1.57}$$

where the last equality follows from the form of Eq. (1.52).

Then, we obtain the *Zero Moments*

$$S_0(\mathbf{Q}) = \chi(\mathbf{Q},0) = \frac{1}{N} \left| \Sigma_l \, \exp\left(i\mathbf{Q}.\mathbf{R}_l\right) \right|^2 \tag{1.58}$$

and

$$S_{0,inc}(\mathbf{Q}) = \chi_{inc}(\mathbf{Q},0) = 1 \tag{1.59}$$

that reveal the structure factor in the coherent case and the normalization condition for the incoherent component.

The *First Moment* results

$$S_1(\mathbf{Q}) = \frac{\eta^2 Q^2}{2M}$$

where *M* is the scatterer mass. This is a general result, valid for any system as long as the interactions amongst its components depend solely on their positions and not on their moments.

These correlation functions, scattering laws and intermediate functions contain all the information about the structure and dynamics of the scattering system. Such information is therefore obtained directly in the measurement of a double differential cross section.

2. SCATTERING IN SIMPLE SYSTEMS

Nuclear scattering in gases

Consider the simplest case of a scattering system composed of a single free atom of mass *M* and at rest in the laboratory system.

Because it is at rest, the atom is in the eigenstate is $|0\rangle$ of the momentum operator,

whose eigenvalue is *p*=0. Although this situation does not correspond to a state of thermodynamic equilibrium, it is the one assumed in the theory of neutron moderation when the energy of the neutron is such that the thermal and chemical bonding effects of the scattering system can be neglected. From the relation (1.40) and defining the operator

$$H_j(\boldsymbol{Q}) = \exp\left\{-i\boldsymbol{Q}\cdot\boldsymbol{R}_j\right\} H_0 \exp\left\{i\boldsymbol{Q}\cdot\boldsymbol{R}_j\right\} \quad , \tag{2.54}$$

the intermediate scattering function (1.51) is rewritten as

$$\chi_{\text{inc}}(\boldsymbol{Q},t) = \langle 0|\exp\left\{-iH_0(\boldsymbol{p}+\eta\boldsymbol{Q})t/\eta\right\} \exp\left\{iH_0(\boldsymbol{p})t/\eta\right\}|0\rangle \quad , \tag{2.55}$$

where the Hamiltonian corresponds to that of a free particle, i.e.

$$H_0(\boldsymbol{p}) = \frac{p^2}{2M} \quad . \tag{2.56}$$

These expressions lead to the following intermediate scattering function

$$\chi_{\text{inc}}(\boldsymbol{Q},t) = \exp\left\{ i\frac{\eta Q^2}{2M}t \right\} \quad , \tag{2.57}$$

whose Fourier transform produces the scattering law

$$S_{\text{inc}}(\boldsymbol{Q},\omega) = \frac{1}{2\pi\,\eta}\int_{-\infty}^{\infty} \mathrm{e}^{-\frac{it}{\eta}\left(\eta\omega-\frac{\eta^2Q^2}{2M}\right)}\mathrm{d}t = \delta\left(\eta\omega-\frac{\eta^2Q^2}{2M}\right) \quad . \tag{2.58}$$

Thus, the effective double differential section results in

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\text{inc}} = \frac{\sigma_{\text{inc}}}{4\pi}\frac{k'}{k}\delta\left(\eta\omega-\frac{\eta^2Q^2}{2M}\right) \quad . \tag{2.59}$$

To obtain the energy transfer nucleus, this expression is integrated over the solid angle, i.e.

$$\begin{aligned}\left(\frac{\mathrm{d}\sigma}{\mathrm{d}E'}\right)_{\text{inc}} &= \frac{\sigma_{\text{inc}}}{4\pi}\frac{k'}{k}\int_{\Omega}\delta\left(\eta\omega-\frac{\eta^2Q^2}{2M}\right)\mathrm{d}\Omega = \frac{\sigma_{\text{inc}}}{2}\frac{k'}{k}\int_{-1}^{1}\delta\left(\eta\omega-\frac{\eta^2}{2M}(k^2+k'^2-2kk'\,\mu)\right)\mathrm{d}\mu \\ &= \frac{\sigma_{\text{inc}}A}{4E}\int_{\eta\omega-\eta^2/2M\,(k-k')^2}^{\eta\omega-\eta^2/2M\,(k+k')^2}\delta(z)\,\mathrm{d}z\end{aligned} \tag{2.60}$$

where *A*=*M*/*m* and μ represents the cosine of the scattering angle. Finally, the following result is obtained for the energy-transfer kernel

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}E'}\right)_{\text{inc}} = \begin{cases} \dfrac{\sigma_{\text{inc}}A}{4E} & \left(\dfrac{A-1}{A+1}\right)^2 E < E' < E \\ 0 & \text{en el resto} \end{cases} \quad . \tag{2.61}$$

The total cross section is obtained by integrating the energy-transfer kernel over all energies, and results in

$$\sigma_{\mathrm{inc}}(E) = \sigma_{\mathrm{inc}}\left(\frac{A}{A+1}\right)^2 \quad , \tag{2.62}$$

which is the so-called *total cross section σ of the free atom*.

We now consider the case where the disperser system is a gas of free particles. In this situation the mass atom *M* is one of a set of such atoms, which are in thermodynamic equilibrium at a temperature *T*. Using the free particle Hamiltonian and performing the thermal average indicated in (1.51), the intermediate scattering function is obtained.

$$\chi_{\mathrm{inc}}(\boldsymbol{Q},t) = \exp\left\{\frac{\eta Q^2}{2M}\left(it - \frac{k_{\mathrm{B}}T}{\eta}t^2\right)\right\} \quad , \tag{2.63}$$

whose Fourier transform leads to the double differential cross section

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{inc}} = N\frac{\sigma_{\mathrm{inc}}}{4\pi}\frac{k'}{k}\frac{1}{\sqrt{2\pi}\,\Lambda}\exp\left\{-\frac{\left(\eta\omega - \eta^2Q^2/2M\right)^2}{2\,\Lambda^2}\right\} \quad , \tag{2.64}$$

where

$$\Lambda^2 = \frac{\eta^2Q^2}{M}k_{\mathrm{B}}T \quad , \tag{2.65}$$

Integrating over the solid angle, and after an extensive calculation found in textbooks, the energy-transfer kernel is obtained.

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}E'}\right)_{\mathrm{gas}} = N\frac{\sigma_{\mathrm{inc}}A}{8E}\left\{\mathrm{erf}\left(\eta x'_{\mathrm{g}} - \rho x_{\mathrm{g}}\right) \pm \mathrm{erf}\left(\eta x'_{\mathrm{g}} + \rho x_{\mathrm{g}}\right) + \exp\left(x_{\mathrm{g}}^2 - x'^2_{\mathrm{g}}\right)\left[\mathrm{erf}\left(\eta x_{\mathrm{g}} - \rho x'_{\mathrm{g}}\right) \mu\, \mathrm{erf}\left(\eta x_{\mathrm{g}} + \rho x'_{\mathrm{g}}\right)\right]\right\} \quad , \tag{2.66}$$

where $x_{\mathrm{g}}^2 = E/k_{\mathrm{B}}T$, $x'^2_{\mathrm{g}} = E'/k_{\mathrm{B}}T$, $\eta = (A+1)/2\sqrt{A}$, $\rho = (A-1)/2\sqrt{A}$ and the upper sign corresponds to case $E' \le E$ and the lower sign to case $E' \ge E$.

The total cross-section of a gas is obtained by integrating the energy-transfer kernel over the final energies, which leads to

$$\sigma_{\mathrm{inc}}(E) = N\sigma_{\mathrm{inc}}\left(\frac{A}{A+1}\right)^2\left\{\left(1 + \frac{1}{2Ax_{\mathrm{g}}^2}\right)\mathrm{erf}(\sqrt{A}x_{\mathrm{g}}) + \frac{1}{\sqrt{\pi A}x_{\mathrm{g}}}\exp(-Ax_{\mathrm{g}}^2)\right\} \quad . \tag{2.67}$$

Nuclear scattering in crystals

Let now be a Bravais crystal with only one atom per unit cell, where each site is determined by a vector $\boldsymbol{l}_j$. Allowing thermal movement of the j-nucleus, its instantaneous position will be

$$\boldsymbol{R}_j = \boldsymbol{l}_j + \boldsymbol{u}_j \quad , \qquad (2.68)$$

where $\boldsymbol{u}$ j is the displacement from the equilibrium position and $\boldsymbol{l}_j$ is constant (**Figure 2.1**).

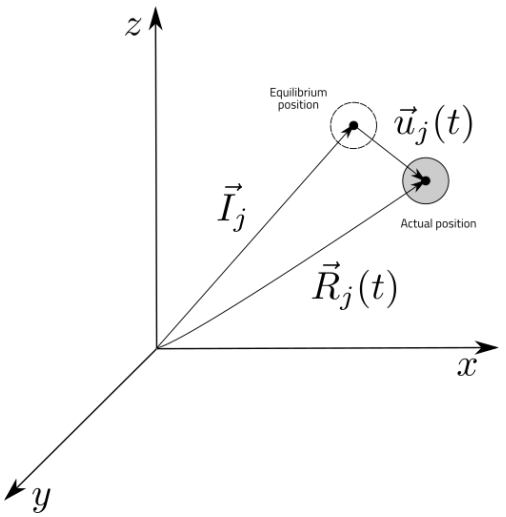

Figure 2.1: Position of the j-th nucleus of the crystal.

For a Bravais crystal, the correlation between the positions of the nuclei j and j' depends only on $\boldsymbol{l}_j - \boldsymbol{l}_{j'}$. Thus, in the expression of the intermediate coherent scattering function given by Eq. (1.50), the sum over j is the same for each value of j', so that j'=0 can be assumed, resulting in

$$\chi_{\mathrm{coh}}(\boldsymbol{Q},t) = \frac{1}{N}\sum_{j,j'}\left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_{j'}(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j(t)\right)\right\rangle$$
$$= \sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j)\left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{u}_0(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{u}_j(t)\right)\right\rangle \quad . \qquad (2.69)$$

Making a similar reasoning for the intermediate incoherent scattering function, Eq. (1.51), where each term in j is equal to the term j = 0, one obtains

$$\chi_{\mathrm{inc}}(\boldsymbol{Q},t) = \frac{1}{N}\sum_{j}\left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{R}_j(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{R}_j(t)\right)\right\rangle$$
$$= \left\langle \exp\left(-i\boldsymbol{Q}\cdot\boldsymbol{u}_0(0)\right)\exp\left(i\boldsymbol{Q}\cdot\boldsymbol{u}_0(t)\right)\right\rangle \quad . \qquad (2.70)$$

In these expressions $\boldsymbol{u}_j(t)$ is the Heisenberg operator for $\boldsymbol{u}_j$.

Assuming that interatomic forces are linear with displacements, these can be expressed as sums of displacements due to normal modes of oscillation, i.e.

$$\boldsymbol{u}_j = \left(\frac{\eta}{2MN}\right)^{1/2}\sum_s \frac{\boldsymbol{e}_s}{\sqrt{\omega_s}}\left\{a_s \exp(i\boldsymbol{q}\cdot\boldsymbol{l}_j) + a_s^{\dagger}\exp(-i\boldsymbol{q}\cdot\boldsymbol{l}_j)\right\} \quad , \qquad (2.71)$$

where $\boldsymbol{q}$ is the wave vector of the mode, p its polarization index (p=1, 2, 3) and s represents the pair of indices $\boldsymbol{q}$, p. The angular frequency and the mode polarization vector are ω_s and $\boldsymbol{e}_s$, respectively, and M is the mass of the nucleus, assumed equal for

all sites. The annihilation and creation operators of the mode s, are a_s y $a_s^\dagger$. The sum is performed on the *N* values of $\boldsymbol{q}$ in the first Brillouin zone and on the three values of *p*.

Using Heisenberg's annihilation and creation operators in Eq.(2.72),

$$a_s(t) = \exp\left(iH_0 t/\eta\right) a_s \exp\left(-iH_0 t/\eta\right) = a_s \exp(-i\omega_s t) \tag{2.72}$$

and

$$a_s^\dagger(t) = \exp\left(iH_0 t/\eta\right) a_s^\dagger \exp\left(-iH_0 t/\eta\right) = a_s^\dagger \exp(i\omega_s t) \quad , \tag{2.73}$$

one obtains $\boldsymbol{u}_j(t)$, and finally the product

$$\boldsymbol{Q}\cdot\boldsymbol{u}_j = \left(\frac{\eta}{2MN}\right)^{1/2} \sum_s \frac{\boldsymbol{Q}\cdot\boldsymbol{e}_s}{\sqrt{\omega_s}} \left\{ a_s \exp(i\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t) + a_s^\dagger \exp[-i(\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t)] \right\} \quad . \tag{2.74}$$

With the expressions obtained for the intermediate scattering functions, the coherent cross section is rewritten as

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh}} = \frac{\sigma_{\mathrm{coh}}}{4\pi}\frac{k'}{k}\frac{N}{2\pi\eta}\sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j)\int_{-\infty}^{\infty}\left\langle \exp U \exp V\right\rangle \exp(-i\omega t)\,\mathrm{d}t \quad , \tag{2.75}$$

where

$$U = -i\boldsymbol{Q}\cdot\boldsymbol{u}_0(0) = -i\sum_s \left\{ g_s a_s + g_s a_s^\dagger \right\} \quad , \tag{2.76}$$

$$V = i\boldsymbol{Q}\cdot\boldsymbol{u}_j(t) = i\sum_s \left\{ h_s a_s + h_s^* a_s^\dagger \right\} \quad , \tag{2.77}$$

$$g_s = \left(\frac{\eta}{2MN}\right)^{1/2} \frac{\boldsymbol{Q}\cdot\boldsymbol{e}_s}{\sqrt{\omega_s}} \tag{2.78}$$

and

$$h_s = \left(\frac{\eta}{2MN}\right)^{1/2} \frac{\boldsymbol{Q}\cdot\boldsymbol{e}_s}{\sqrt{\omega_s}} \exp\left\{ i(\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t) \right\} \quad . \tag{2.79}$$

We proceed in a similar way with the incoherent cross section, obtaining

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{inc}} = \frac{\sigma_{\mathrm{inc}}}{4\pi}\frac{k'}{k}\frac{N}{2\pi\eta}\int_{-\infty}^{\infty}\left\langle \exp U \exp V_0\right\rangle \exp(-i\omega t)\,\mathrm{d}t \quad , \tag{2.80}$$

where

$$V_0 = i\boldsymbol{Q}\cdot\boldsymbol{u}_0(t) \quad , \tag{2.81}$$

has the same definition as *V*, but for *j*=0.

Since the commutator [*U*, *V*] is a scalar, the thermal average can be written as

$$\left\langle \exp U \exp V\right\rangle = \left\langle \exp(U+V)\right\rangle \exp\left\{\tfrac{1}{2}(UV - VU)\right\} \tag{2.82}$$

and *<u>for a harmonic oscillator</u>* it can be proved that

$$\left\langle \exp(U+V)\right\rangle = \exp\left\{\tfrac{1}{2}\left\langle (UV-VU)^2\right\rangle\right\} \quad . \tag{2.83}$$

On the other hand, *U* is proportional to the component in the direction $\boldsymbol{Q}$ of the

displacement of the atom from the origin to time zero, while V is the corresponding quantity for the atom j at time t. Since the origin of times is arbitrary and for a Bravais crystal all atoms are equivalent, it is evident that

$$\langle U^2 \rangle = \langle V^2 \rangle \quad . \tag{2.84}$$

From these three equations it can be shown that

$$\langle \exp U \exp V \rangle = \exp\langle U^2 \rangle \exp\langle UV \rangle \quad , \tag{2.85}$$

thus the expressions for the coherent and incoherent differential cross sections are

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\, \mathrm{d}E'} \right)_{\mathrm{coh}} = \frac{\sigma_{\mathrm{coh}}}{4\pi} \frac{k'}{k} \frac{N}{2\pi\,\eta} \exp\langle U^2 \rangle \sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j) \int_{-\infty}^{\infty} \exp\langle UV \rangle \exp(-i\omega t)\, \mathrm{d}t \tag{2.86}$$

and

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\, \mathrm{d}E'} \right)_{\mathrm{inc}} = \frac{\sigma_{\mathrm{inc}}}{4\pi} \frac{k'}{k} \frac{N}{2\pi\,\eta} \exp\langle U^2 \rangle \int_{-\infty}^{\infty} \exp\langle UV_0 \rangle \exp(-i\omega t)\, \mathrm{d}t \quad , \tag{2.87}$$

where the corresponding results have been used for V_0.

In the above expressions, an exponential factor is recognized, which is generally denoted as

$$\exp\{-2W\} = \exp\langle U^2 \rangle = \exp\left\langle -\left\langle \{\boldsymbol{Q}\cdot\boldsymbol{u}_0(0)\}^2 \right\rangle \right\rangle \quad , \tag{2.88}$$

known as *the Debye-Waller factor*.

From Eq. (2.76), the Debye-Waller factor results in

$$2W = \sum_\lambda p_\lambda \sum_{s,s'} g_s g_{s'} \langle \lambda | (a_s + a_s^\dagger)(a_{s'} + a_{s'}^\dagger) | \lambda \rangle = \sum_\lambda p_\lambda \sum_s g_s^2 \langle \lambda | a_s a_s^\dagger + a_s^\dagger a_s | \lambda \rangle \quad . \tag{2.89}$$

If n_s is the occupancy number of the s-th oscillator, for the state $|\lambda\rangle$ the matrix element is

$$\langle \lambda | a_s a_s^\dagger + a_s^\dagger a_s | \lambda \rangle = 2n_s + 1 \quad . \tag{2.90}$$

The form of p_λ (Eq. (1.36)) allows every term in the sum over s to be averaged independently over $|\lambda\rangle$, so that

$$2W = \frac{\eta}{2MN} \sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s} \langle 2n_s + 1 \rangle \quad . \tag{2.91}$$

For <u>*cubic crystals*</u> the mean value of $(\boldsymbol{Q}.\boldsymbol{e}_s)^2$ is directly $Q^2/3$, then

$$2W = \frac{\eta}{2MN} \frac{Q^2}{3} \sum_{\boldsymbol{q},p} \frac{\langle 2n(\omega_p(\boldsymbol{q})) + 1 \rangle}{\omega_p(\boldsymbol{q})} = \frac{\eta}{2MN} \frac{Q^2}{3} \frac{V_c}{(2\pi)^3} \sum_p \int \frac{\langle 2n(\omega_p(\boldsymbol{q})) + 1 \rangle}{\omega_p(\boldsymbol{q})} \mathrm{d}\boldsymbol{q} \quad , \tag{2.92}$$

where V_c is the volume of the crystal and the dependence with the wave vector and polarization modes has been made explicit. Even for non-cubic crystals these expressions are good approximations.

Since the number of degrees of freedom is of the order of 10^{23}, it is possible and convenient to express the sum over the oscillation modes as an integral over the frequencies, using the density of normal modes *Z(ω)*, or phonon density of states (DOS), defined so that *Z(ω)dω* is the fraction of the normal modes between ω y ω+dω. Then

$$\frac{V_c}{3N}\sum_p \int \frac{f(\omega_p(\boldsymbol{q}))}{(2\pi)^3}\,\mathrm{d}\boldsymbol{q} = \int Z(\omega)\, f(\omega)\,\mathrm{d}\omega \quad , \tag{2.93}$$

where, by comparison, it follows that

$$Z(\omega) = \frac{V_c}{3N}\sum_p \int \frac{\delta\left(\omega - \omega_p(\boldsymbol{q})\right)}{(2\pi)^3}\,\mathrm{d}\boldsymbol{q} \quad . \tag{2.94}$$

So, we get the following expression for the Debye-Waller factor

$$2W = \frac{\eta Q^2}{2M}\int_0^\infty \langle 2n(\omega)+1\rangle \frac{Z(\omega)}{\omega}\,\mathrm{d}\omega \quad , \tag{2.95}$$

where the number $n(\omega)$, has the expression

$$\langle n(\omega)\rangle = \left[\exp(\beta\eta\omega) - 1\right]^{-1} \quad , \tag{2.96}$$

which is the average boson occupation number.

In the case of the incoherent double differential cross section, for the factor $\exp\langle UV_0\rangle$ an analysis like that performed with the Debye-Waller factor can be made, now starting from the Ecs. (2.7776) and (2. 77) for the case $\boldsymbol{I}_j$=0. Then

$$\begin{aligned}\langle UV_0\rangle &= \sum_\lambda p_\lambda \sum_{s,s'} g_s g_{s'} \langle\lambda|(g_s a_s + g_s a_s^\dagger)(h_{s'} a_{s'} + h_{s'}^* a_{s'}^\dagger)|\lambda\rangle \\ &= \sum_\lambda p_\lambda \sum_s \langle\lambda| g_s h_{s'} a_s a_s^\dagger + g_s h_{s'}^* a_s^\dagger a_s|\lambda\rangle \\ &= \frac{\eta}{2MN}\sum_\lambda p_\lambda \sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s}\left\{\mathrm{e}^{-i\omega_s t}\langle\lambda|a_s a_s^\dagger|\lambda\rangle + \mathrm{e}^{i\omega_s t}\langle\lambda|a_s^\dagger a_s|\lambda\rangle\right\} \quad .\end{aligned}$$

Using the properties of the annihilation and creation operators, the indicated matrix elements have expressions related to the occupation number, so that

$$\langle UV_0\rangle = \frac{\eta}{2MN}\sum_\lambda p_\lambda \sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s}\left\{\mathrm{e}^{-i\omega_s t}\langle n_s+1\rangle + \mathrm{e}^{i\omega_s t}\langle n_s\rangle\right\} \quad .$$

Again, for cubic crystals and replacing the sum by an integral over the frequencies, one obtains

$$\langle UV_0\rangle = \frac{\eta Q^2}{2M}\int_0^\infty \frac{Z(\omega)}{\omega}\left\{\mathrm{e}^{-i\omega t}\langle n(\omega)+1\rangle + \mathrm{e}^{i\omega t}\langle n(\omega)\rangle\right\}\mathrm{d}\omega \quad .$$

If the function is defined

$$\gamma(t) = \int_{-\infty}^{\infty} \frac{Z(\omega)}{\omega}\, n(\omega)\,\mathrm{e}^{-i\omega t}\,\mathrm{d}\omega \quad ,$$

and symmetry properties of the occupancy number are used for harmonic oscillators and

state density, we can write it as

$$\gamma(t) = \int_0^\infty \frac{Z(\omega)}{\omega} \left\{ [n(\omega)+1]\, \mathrm{e}^{i\omega t} + n(\omega)\, \mathrm{e}^{-i\omega t} \right\} \mathrm{d}\omega \quad ,$$

It clearly follows that *the intermediate incoherent scattering function* has the following expression

$$\chi_{\mathrm{inc}}(\boldsymbol{Q},t) = \exp\left\{ \frac{\eta Q^2}{2M} \left[\gamma(t) - \gamma(0) \right] \right\} \quad ,$$

which allows to obtain the differential cross section section through the Ecs. (1.53) and (1.54).

The Phonon Expansion

A crystal of N atoms has 3*N* normal modes, whose quantum numbers determine the initial state $|\lambda\rangle$ of the crystal. In a generic scattering process, the state of the crystal changes to $|\lambda'\rangle$, characterized by another set $n_1', n_2', \mathrm{K}\ , n_{3N}'$ of quantum numbers. Scattering processes can be classified according to changes in those quantum numbers.

Elastic processes: All quantum numbers remain unchanged, that is, $n_i' = n_i$ for all *i* from 1 to 3*N*.

One-phonon processes: All quantum numbers remain unchanged, except one corresponding to the oscillator α, which changes by one unit, i.e. $n_i' = n_i$ for all i $\neq\alpha$ and . $n_\alpha' = n_\alpha \pm 1$

Two-phonon processes: All quantum numbers remain unchanged, except for two corresponding to the oscillators α and β, which change by one unit, i.e. for all i $\neq\alpha$ or β, and $n_\alpha' = n_\alpha \pm 1,\ n_\beta' = n_\beta \pm 1$

Similarly for processes of three, four and more phonons.

The development in phonons is obtained if the factor exp$\langle UV\rangle$ in Eq (2.8786) is developed in Taylor series. ($\langle exp\ UV_0\rangle$ in Eq. (2. 87)), i.e.

$$\exp\langle UV\rangle = 1 + \langle UV\rangle + \frac{1}{2!}\langle UV\rangle^2 + \mathrm{K} + \frac{1}{p!}\langle UV\rangle^p + \mathrm{K} \quad ,$$

where the *pth* term produces the cross section for all *p* phonon processes. Thus, the first term gives the elastic cross section; the following $\langle UV\rangle$ term gives the cross section for all processes of a phonon in whichα it is, in turn, each of the 1 to 3*N* and each n_α increases or decreases by one unit; the term (1/2!) $\langle UV\rangle^2$ gives the cross section for all two-phonon processes in which the combination $\alpha\beta$, is, in turn, each of the 3N (3N-1)/2

combinations of two oscillators taken from 3*N* and, for each combination, n_β and n_α varies by one unit, and so on.

Elastic coherent scattering

Replacing exp⟨*UV*⟩ by 1 in Ec. (2.8786) we obtain

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh}} = \frac{\sigma_{\mathrm{coh}}}{4\pi}\frac{k'}{k}\frac{N}{2\pi\,\eta}\exp\langle U^2\rangle \sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j)\int_{-\infty}^{\infty}\exp(-i\omega t)\,\mathrm{d}t \quad ,$$

So that, if the energy change of the neutron $\eta\omega$ and the integral form of the δ, are taken into account, it turns out

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh}} = \frac{\sigma_{\mathrm{coh}}}{4\pi}N\exp\langle U^2\rangle \sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j)\,\delta(\eta\omega) \quad ,$$

situation that corresponds to elastic scattering. This expression can be easily integrated to obtain the differential cross section dσ/dΩ, keeping the energy *E* of the incident neutron fixed:

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{coh\ el}} = \int_0^{\infty}\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh\ el}}\mathrm{d}E' = \frac{\sigma_{\mathrm{coh}}}{4\pi}N\exp\langle U^2\rangle \sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j) \quad .$$

Finally it results

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{coh\ el}} = \frac{\sigma_{\mathrm{coh}}}{4\pi}N\frac{(2\pi)^3}{v_0}\exp(-2W)\sum_{\boldsymbol{\tau}}\delta(\boldsymbol{Q}-\boldsymbol{\tau}) \quad ,$$

where the sum over the lattice was performed in the standard form, v_0 is the volume of the unit cell of the crystal and $\boldsymbol{\tau}$ is a vector of the reciprocal lattice. The latter equation indicates that scattering can only occur when the momentum transfer vector $\boldsymbol{Q}$ coincides with a vector of the reciprocal lattice $\boldsymbol{\tau}$, which is Bragg's Law.

For Bravais crystals with base, the following expression is obtained for the coherent elastic differential cross section

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{coh\ el}} = N\frac{(2\pi)^3}{v_0}\sum_{\boldsymbol{\tau}}\delta(\boldsymbol{Q}-\boldsymbol{\tau})\left|F_{\mathrm{N}}(\boldsymbol{Q})\right|^2 \quad ,$$

where

$$F_{\mathrm{N}}(\boldsymbol{Q}) = \sum_{\boldsymbol{d}}\bar{b}_{\boldsymbol{d}}\exp(i\boldsymbol{Q}\cdot\boldsymbol{d})\exp(-W_{\boldsymbol{d}})$$

is known as the *nuclear structure factor of the unit cell*, the sum is performed over all the sites of the base (each characterized by the vector $\boldsymbol{d}$ that locates it with respect to the site of the network), the mean scattering length refers to that magnitude for each site of the base and $W_{\boldsymbol{d}}$ is given by

$$W_{\boldsymbol{d}} = \frac{\eta}{4 M_{\boldsymbol{d}} N} \sum_s \frac{(\boldsymbol{Q}\cdot \boldsymbol{e}_{\boldsymbol{d},s})^2}{\omega_s} \coth\left(\tfrac{1}{2}\beta\eta\omega_s\right)$$

or

$$W_{\boldsymbol{d}} = \frac{\eta Q^2}{4 M_{\boldsymbol{d}}} \int_0^{\omega_m} \coth\left(\tfrac{1}{2}\beta\eta\omega\right) \frac{Z(\omega)}{\omega}\, \mathrm{d}\omega \quad ,$$

Valid for a cubic crystal, where the sum over the normal modes has been replaced by the integral over the frequencies, and the occupation number has been used.

Coherent scattering of a phonon

Returning to scattering for a Bravais crystal, from Eq. (2.86) the coherent cross section of a phonon is obtained by replacing exp⟨UV⟩ by ⟨ UV⟩. From the Ecs. (2.7776) and (2. 77)

$$\langle \lambda | UV | \lambda \rangle = \sum_{s,s'} \langle \lambda | (g_s a_s + g_s a_s^\dagger)(h_{s'} a_{s'} + h_{s'}^* a_{s'}^\dagger) | \lambda \rangle \quad .$$

The matrix elements in the summation are zero except for the terms where the annihilation and creation operators appear in the same normal mode, then

$$\langle \lambda | UV | \lambda \rangle = \sum_s g_s h_s^* (n_s + 1) + g_s h_s n_s \quad .$$

From this expression, and from the Ecs. (2.7978) and (2. 79), we have

$$\langle UV \rangle = \sum_s g_s h_s^* \langle n_s + 1 \rangle + g_s h_s \langle n_s \rangle$$

$$= \frac{\eta}{2MN} \sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s} \left[\exp\{-i(\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t)\} \langle n_s + 1 \rangle + \exp\{i(\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t)\} \langle n_s \rangle \right] \quad ,$$

producing the cross section

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh\,1\,ph}} = \frac{\sigma_{\mathrm{coh}}}{4\pi} \frac{k'}{k} \frac{1}{4\pi M} \exp(-2W) \sum_j \exp(i\boldsymbol{Q}\cdot\boldsymbol{l}_j) \sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s}$$

$$\times \int_{-\infty}^{\infty} \left[\exp\{-i(\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t)\} \langle n_s + 1 \rangle \right.$$

$$\left. + \exp\{i(\boldsymbol{q}\cdot\boldsymbol{l}_j - \omega_s t)\} \langle n_s \rangle \right] \exp(-i\omega t)\, \mathrm{d}t \quad .$$

Using the integral form of the Dirac delta and solving the sum over the lattice, each of the two terms in the above expression can be written as

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh}\,\pm 1\,\mathrm{ph}} = \frac{\sigma_{\mathrm{coh}}}{4\pi} \frac{k'}{k} \frac{(2\pi)^3}{v_0} \frac{\eta}{2M} \exp(-2W)$$

$$\times \sum_s \sum_{\boldsymbol{\tau}} \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s} \begin{Bmatrix} \langle n_s + 1 \rangle \\ \langle n_s \rangle \end{Bmatrix} \delta(\eta\omega \,\mu\, \eta\omega_s)\, \delta(\boldsymbol{Q} \,\mu\, \boldsymbol{q} - \boldsymbol{\tau}) \quad .$$

Due to those δ distributions that appear in this expression, for the dispersion process to

occur the two conditions must be met:

$$\eta\omega = \pm\eta\omega_s \qquad \text{and} \qquad \boldsymbol{Q} = \boldsymbol{\tau} \pm \boldsymbol{q}$$

Analyzing the situation corresponding to the sign (+), the first condition indicates that the energy of the neutron decreases by an amount equal to the energy of a phonon of the s-th normal mode; this process, in which a phonon is created, is known as *emission or creation of phonons*. The second condition can be considered as an expression of momentum conservation, since $\eta\boldsymbol{Q}$ it is the change of the momentum of the neutron, while $\eta(\boldsymbol{\tau} + \boldsymbol{q})$ it is the momentum delivered to the crystal. Similarly, the situation corresponding to the sign (-) can be analyzed, where in this process a phonon is annihilated and is known as a*bsorption* or *annihilation of phonons*.

As can be seen from the expression (2.96 tends to 1 and $\langle n_s \rangle$ tends to zero, so that the phonon absorption cross section tends to zero, a predictable result since at this limit all oscillators are in their ground state.

For crystals with more than one atom per lattice site, the expression for the cross section generalizes as follows

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\mathrm{coh}\,\pm 1\,\mathrm{ph}} = \frac{k'}{k}\frac{(2\pi)^3}{2v_0}\sum_s\sum_{\boldsymbol{\tau}}\frac{\eta}{\omega_s}\left|\sum_{\boldsymbol{d}}\frac{\bar{b}_{\boldsymbol{d}}}{\sqrt{M_{\boldsymbol{d}}}}\exp(-W_{\boldsymbol{d}})\exp(i\boldsymbol{Q}\cdot\boldsymbol{d})(\boldsymbol{Q}\cdot\boldsymbol{e}_{\boldsymbol{d},s})\right|^2 \times\left\{\begin{matrix}\langle n_s+1\rangle\\ \langle n_s\rangle\end{matrix}\right\}\delta(\eta\omega\,\mu\,\eta\omega_s)\,\delta(\boldsymbol{Q}\,\mu\,\boldsymbol{q}-\boldsymbol{\tau}) \quad . \tag{2.97}$$

Multiphonon coherent scattering

There are two main reasons why there is interest in learning about multiphonon cross sections. The first is that in many scattering experiments it is useful to be able to estimate the background, in order to discount it and isolate the most interesting scattering processes; The second is that as the energy of incident neutrons increases, the cross section is increasingly controlled by multiphonon terms.

From Eq. (2.86) the coherent cross section for p phonons is obtained by replacing exp $\langle UV\rangle$ by $\langle UV\rangle^p$ /p! and can be easily shown to contain two δ functions giving rise to the following equations

$$\eta\omega = \sum_{n=1}^{p}(\pm\eta\omega_{s_n}) \tag{2.98}$$

and

$$\boldsymbol{Q} = \boldsymbol{\tau}\sum_{n=1}^{p}(\pm\boldsymbol{q}_n) \quad . \tag{2.99}$$

Both conditions must be met for this type of scattering to occur, where the neutron

creates or annihilates a phonon in *p* different normal modes.

Unlike the coherent scattering case of a phonon, given a wave vector ***k***, it is always possible to find combinations of *p* normal modes such that their values of ***q*** and ω_s satisfy the scattering conditions for any value of *k*' . That is, the conservation of momentum and energy does not impose restrictions as severe as in the elastic case or a phonon. So multiphonon scattering tends to a function that depends smoothly on the angle of scattering and the incident energy.

Incoherent scattering

In the incoherent double differential cross section, given by Eq. (2.87), $\exp\langle UV_0\rangle$ is developed in powers of $\langle UV_0\rangle$ in a totally analogous way to what was done for the coherent cross section. Thus it follows that the p-th term corresponds to a process of *p* phonons.

To calculate the elastic incoherent cross section, the $\exp\langle UV_0\rangle$ is replaced by 1 in the Eq. (2. 87) and proceeds in the way that was done with the coherent contribution, to finally obtain

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\text{inc el}} = \frac{\sigma_{\text{inc}}}{4\pi} N \exp(-2W) \quad .$$

The only dependence of this differential cross section with the direction of dispersion is on the Debye-Waller factor, which depends on ***Q***. At low temperatures scattering is isotropic, since the Debye-Waller factor is close to one.

The incoherent cross section of a phonon is obtained by replacing $\exp\langle UV_0\rangle$ by $\langle UV_0\rangle$ in Eq. (2.8887):

$$\left\langle UV_0\right\rangle = \frac{\eta}{2MN}\sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s}\left[\exp(i\omega_s t)\left\langle n_s+1\right\rangle + \exp(-i\omega_s t)\left\langle n_s\right\rangle\right]$$

and

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\text{inc 1 ph}} = \frac{\sigma_{\text{inc}}}{4\pi}\frac{k'}{k}\frac{\eta}{2M}\exp(-2W)\sum_s \frac{(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2}{\omega_s}$$

$$\times\left[\left\langle n_s+1\right\rangle\delta(\eta\omega-\eta\omega_s)+\left\langle n_s\right\rangle\delta(\eta\omega+\eta\omega_s)\right] \quad .$$

The first term in the bracket corresponds to phonon creation and the second to annihilation.

This means that for fixed **k**, θ, and crystal orientation, there is incoherent scattering of a phonon for a continuous range of values of k', and for a given k' there is scattering of all normal modes whose ω_s satisfies the conservation of energy condition. Therefore, the cross section can be represented in terms of the phonon density of states $Z(\omega)$

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\text{inc}\pm1\text{ ph}} = \frac{\sigma_{\text{inc}}}{4\pi}\frac{k'}{k}\frac{3N}{4M}\exp(-2W)\frac{\left\langle(\boldsymbol{Q}\cdot\boldsymbol{e}_s)^2\right\rangle_{\text{av}}}{\omega}Z(\omega)\left[\coth\left(\tfrac{1}{2}\beta\eta\omega\right)\pm1\right] \quad ,$$

As before, the quantity $\langle(\boldsymbol{Q}.\boldsymbol{e}_s)^2\rangle_{av}$ is the value of $(\boldsymbol{Q}.\boldsymbol{e}_s)^2$ averaged over all modes with frequency ω_s, so that for a cubic crystal it can be replaced by $Q^2/3$, and the incoherent cross section for processes of one phonon is

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'}\right)_{\text{inc}\pm1\text{ ph}} = \frac{\sigma_{\text{inc}}}{4\pi}\frac{k'}{k}\frac{N}{4M}Q^2\exp(-2W)\frac{Z(\omega)}{\omega}\left[\coth\left(\tfrac{1}{2}\beta\eta\omega\right)\pm1\right] \quad .$$

Incoherent approximation

In evaluating coherent and incoherent cross sections for multiphonon processes, two basic difficulties arise. The first is that when the incident energy or temperature of the system increases, the convergence of development into phonons progressively worsens. The second is that, as the number of phonons involved in the scattering process increases, the cross section becomes increasingly difficult to calculate. Since this is particularly true for the coherent cross section, the incoherent approach is used, which consists of replacing it with the corresponding incoherent component. In order not to change the ratio from coherent to incoherent scattering, it must be multiplied by the factor $\sigma_{\text{coh}}/\sigma_{\text{inc}}$.

Sjölander´s Phonon Expansion

For the incoherent intermediate scattering function, in the isotropic (cubic) and harmonic (Gaussian) approximations to represent the motion of the crystal atoms, we obtain

$$\chi_{\text{inc}}(\mathbf{Q},t) = \exp\left\{\frac{\eta Q^2}{2M}\left[\gamma(t)-\gamma(0)\right]\right\}$$

where

$$\gamma(t) = \int_{-\infty}^{\infty}\frac{Z(\omega)}{\omega}n(\omega)\,\mathrm{e}^{-i\omega t}\,\mathrm{d}\omega \quad ,$$

that, using symmetry properties of the occupation number *n(w)* and the phonon density of states $Z(\omega)$ can be written as

$$\gamma(t) = \int_0^\infty \frac{Z(\omega)}{\omega} \left\{ [n(\omega)+1]\, \mathrm{e}^{i\omega t} + n(\omega)\, \mathrm{e}^{-i\omega t} \right\} \mathrm{d}\omega \ ,$$

Because

$$2W = \frac{\eta Q^2}{2M} \int_0^\infty \langle 2n(\omega)+1 \rangle \frac{Z(\omega)}{\omega} \mathrm{d}\omega = \frac{\hbar \boldsymbol{Q}^2}{2M}\ \gamma(0)$$

Expanding the intermediate scattering function, we get

$$\chi(\boldsymbol{Q}, t) = \ e^{-2W} \sum_{p=0}^{\infty} \frac{(2W)^p}{p!} \left[\frac{\gamma(t)}{\gamma(0)}\right]^p$$

where each term represents the interaction of the neutron with *p* phonons.
In the manner of Sjölander (1958), we can define the functions

$$G_p(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega t} \left[\frac{\gamma(t)}{\gamma(0)}\right]^p \ dt$$

Which allow the scattering cross section to be written in the form

$$\frac{d^2\sigma}{d\omega\ dE'} = N\ \frac{\sigma_{inc}}{4\pi}\ \frac{\boldsymbol{k}'}{\boldsymbol{k}}\ e^{-2W} \sum_{p=0}^{\infty} (2W)^p\ \frac{G_p(\omega)}{\hbar p!}$$

This is the phonon expansion for the incoherent double differential cross section, within the cubic and the Gaussian approximations.
The integration limit in the definition of the $G_p(\omega)$ is formally extended to infinity, although the expression under the integral sign differs from zero only within a finite interval, determined by the condition $/\omega/ \leq p\omega_m$ for a given p, where ω_m is the maximum frequency of the normal modes (PDOS).
It is evident from the same definition that the $G_p(\omega)$ are normalized to unity. Also, and independently of the model for the solid

$$G_0(\omega) = \hbar\ \delta(\hbar\omega)$$

Using the convolution theorem for Fourier transforms, an important recurrence relationship is satisfied by the $G_p(\omega)$

$$G_p(\omega) = \int_{-\infty}^{\infty} G_1(-\omega')\ G_{p-1}(\omega' + \ \omega)\ d\omega'$$

We have seen before that the zero-phonon term gives rise to the elastic component, which is completely different for the coherent and the incoherent cross sections. Therefore, these terms are treated exactly, whereas it is usual to approximate the coherent inelastic contribution by the incoherent inelastic contribution. This is the **incoherent approximation** for the inelastic cross section.

The Case of Molecular Solids

An approximate density of states is used to represent the actual dynamics of the molecular solid:

$Z(\omega) = a_s Z_s(\omega) + a_r Z_r(\omega) + a_v Z_v(\omega)$; a_i are relative weights.

Under the assumption of no coupling between modes, the intermediate scattering function is

$$\chi(Q,t) = \chi_s(Q,t).\chi_r(Q,t).\chi_v(Q,t)$$

where each of the factors is further assumed to satisfy the Gaussian approximation

$$\chi_i(Q,t) = \exp\left\{\frac{\eta Q^2}{2M}\, a_i \left[\, \gamma_i(t) - \gamma_i(0) \,\right]\right\} \quad ; \quad \text{i = s, r, v}$$

and the time-dependent mean-square displacement $\gamma(t)$ is given by

$$\gamma(t) = \int_0^\infty \frac{Z(\omega)}{\omega}\left[\{n(\omega)+1\}e^{i\omega t} + n(\omega)e^{-i\omega t}\right] d\omega = \sum_i a_i \gamma_i(t)$$

with $n(\omega)$ being the occupation number. Phonon expansion of the intermediate scattering functions:

$$\chi_i(Q,t) = e^{-2W_i}\left\{\sum_m \frac{(2W_i)^m}{m!}\left[\frac{\gamma_i(t)}{\gamma_i(0)}\right]^m\right\}$$

where $2W_i = \frac{\eta Q^2}{2M_i}\gamma_i(0)$, $\frac{1}{M_i} = \frac{a_i}{M}$ and the associated scattering laws are given by

$$S_i(Q,\omega) = e^{-2W_i}\sum_p \frac{(2W_i)^p}{p!}\frac{G_p^{(i)}(\omega)}{\eta} \quad , \quad G_m^{(i)}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt e^{-i\omega t}\left[\frac{\gamma_i(t)}{\gamma_i(0)}\right]^m \quad .$$

The complete scattering law is calculated as

$$S(Q,\omega) = \frac{1}{2\pi\eta}\int_{-\infty}^{\infty} dt\, \chi(Q,t) e^{-i\omega t} = S_s(Q,\omega) \otimes S_r(Q,\omega) \otimes S_v(Q,\omega)$$

FINAL COMMENTS

We have seen that the cross sections associated with neutron scattering can be expressed essentially as the product of two factors. The first factor, σ , depends on the interaction between the neutron and the individual particles that make up the sample. The second factor, $S(\mathbf{Q},\omega)$, does not depend at all on the neutron properties or the characteristics of the interaction, being exclusively a property of the disperser system determined by its temperature and interactions between the particles that compose it.

From a formal point of view, we can say that the cross section is a measure of the response of the system, which in turn is determined by correlation functions representative of spontaneous fluctuations in the scattering system. This close relationship between the response and the spectrum of fluctuations is a consequence of the fact that neutron scattering involves a weak 'effective' interaction, and the response of the system is then linear, i.e. describable by first-order perturbation theory. Under these conditions, the scattering function contains all the information about the dynamics and structure of the disperser system.

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