Seminar I
THERMAL NEUTRON SCATTERING BY BOUND ATOMS

Abstract
Why thermal neutron scattering cross sections differ from cross sections at higher neutron energies and target nuclei can no longer be considered free is explained, as is why this is relevant to reactor theory. Wave mechanical scattering of neutrons is introduced and processes governing the changes to thermal neutron scattering cross sections such as the reduced mass effect, coherent and incoherent scattering as well as inelastic scattering effects are analyzed in terms of simple models, such as scattering off a system of two rigidly fixed nuclei or inelastic scattering by a harmonically bound nucleus. An introduction to a more general theory due to Leon Van Hove is given and why its use is limited is explained. Because of the limited usefulness of the theory, the next step in reactor calculations will be the study of propagation of uncertainties due to uncertainties in thermal neutron cross sections.

Author: Gregor Jecl
Mentor: doc. dr. Luka Snoj

Ljubljana, May 2018
Contents

Introduction 1

1 Motivation 1

2 Essential concepts 3

2.1 Wave mechanical scattering and s-wave scattering 3

2.2 Scattering length 5

2.3 The Born approximation 5

3 Cooperative effects in the thermal region 6

3.1 Reduced mass effect 6

3.2 Coherent scattering 7

3.3 Incoherent scattering 9

3.3.1 Nonidentical nuclei 9

3.3.2 Nonconstant internuclear spacing 10

3.4 Inelastic effects 10

4 Van Hove theory 12

Conclusion 13

References 14

Introduction

In 1938, shortly after the 1932 discovery of the neutron by Chadwick, a process had been observed by Otto Hahn and Fritz Strassman in which radioelements were formed from uranium, which had been bombarded by neutrons. In 1939 Lise Meitner and Otto Frisch theoretically explained the process and dubbed it “Nuclear fission” [1].

It was theorised and later confirmed that such a breakup of an atom of uranium would lead to the release of a large amount of energy. The first nuclear reactor was built in 1942 by a team led by Enrico Fermi in Chicago and the first man-made self-sustaining chain reaction was initiated. Data from January 1st 2017 indicates nuclear power accounted for 18.5% of total electricity production of OECD countries and for about 37% of Slovenia’s total electricity production [2].

In this seminar a number of specific physical processes concerning interactions of neutrons with energies less than, or on the order of, 1 eV (termed “slow neutrons” or “thermal neutrons”) is presented. First some motivation is given, followed by how and why the cross sections in this energy interval (the so-called “thermal region”) are different, which are the relevant processes responsible for these differences, and how they affect reactor operation. Afterwards some essential concepts are introduced and in the next section specific effects on neutron cross sections are investigated in more detail. Finally, a more general theory is introduced, but without going into detail.

1 Motivation

The main reason interactions between nuclei and neutrons are different at low incident neutron energies i.e. several eV and below, is that at high incident energies the nucleus can be assumed
to be isolated and free, i.e. not bound in a molecule, liquid or solid.

During the process of thermalization (slowing down of fast fission neutrons to thermal energies) neutron energy decreases in elastic and inelastic scattering events with moderator nuclei. Because targets commonly encountered by neutrons in nuclear reactors are either solid (graphite, beryllium, uranium oxide, uranium metal, zirconium hydride, polyethylene, teflon) or liquid (water, heavy water) the effects of binding of atoms in a molecule become more important and at some point neutrons can no longer be assumed to interact with free nuclei because their kinetic energies are no longer sufficiently large that chemical bonds could be neglected.

Neutrons with energies $E \lesssim 1$ eV have De Broglie wavelengths of $\lambda_D \gtrsim 0.3 \text{ Å}$ which are of the order of interatomic spacing in crystals or molecules and thus interference effects can arise. In reality, because the nuclei are in some sort of a spatial array (of a high degree of regularity in crystalline solids and a lesser one in liquids), the scattered neutron wave functions interfere with one another and can add coherently leading to significantly different differential cross section than would be obtained from individual scattering events, and can greatly influence the flux and heat distributions in the reactor core. This is of high significance in reactor design because it affects reactor operation and also because “hot spots” of neutron or heat flux could cause damage in the reactor core, e.g. in the fuel elements, or could contribute to premature wear of core materials [1,3].

Because the molecule or solid is bound by chemical forces whose binding energy is also of the order of 1 eV, the incident neutron energy, a new type of inelastic scattering can occur, which involves changes in the energy levels of the target material. Energy exchange between the neutron and the nucleus is quantized and can occur only through excitation of discrete vibrational or rotational energy levels [1]. This property of slow neutrons has been used extensively to study atomic motion but in reactor physics understanding inelastic (and elastic) scattering is essential to understanding the process of moderation, and by extension, in determining the neutron spectrum i.e. the spatial and energy distribution of neutrons in the reactor [3,4].

Because of different materials found at different parts of the reactor, the neutron spectrum varies in space, i.e. the spectrum is not the same in the reactor core and the surroundings. It also varies significantly in the core itself, most notably because of the control rods, differences in fuel elements and potentially because of pockets of moderator, neutron sources etc [5].

To accurately determine the neutron spectrum, which is the central problem of reactor theory, the neutron transport equation has to be solved. In this equation the neutron populations are divided into groups (intervals) according to their energy and parameters in each group (cross sections for scattering, cross sections for fission, average number of neutrons produced etc.) must be known as accurately as possible to obtain the best possible solution. Uncertainties in these parameters propagate through the transport equation to the neutron spectrum, on which important parameters of the reactor, such as the rate at which various reaction occur, the reactor’s energy production capacity, its stability and safety, depend [6].

Thermal reactors are particularly sensitive to the interactions of neutrons in the thermal region because this is where the bulk of fission reactions occur [1, 7] and because the neutron mean free path, which is proportional to the inverse value of the total cross section (which in turn encompasses the fission cross section), is shortest in this energy region [5]. The latter can be inferred from Figure 1 which shows the energy dependence of the fission cross sections of $^{235}\text{U}$ and $^{238}\text{U}$. 
Figure 1: Comparison of evaluated fission cross sections of $^{235}$U and $^{238}$U [8].

2 Essential concepts

The goal is to study the effects outlined in the preceding section and make predictions on how the scattering cross sections for neutrons of low incident kinetic energy are affected. As was explained, it is necessary to consider the scattering of slow neutrons not as single neutron-nucleus scattering events, but to in some way take into account the binding of atoms into condensed matter. Before such neutron interactions can be studied further, some essential concepts have to be developed.

2.1 Wave mechanical scattering and s-wave scattering

First we consider elastic scattering scattering by a single, isolated, fixed, spinless nucleus. The direction of the incident neutron is taken as the $z$-direction, the nucleus is fixed at the origin and the the coordinate of the neutron is denoted by $r$. The two-body problem reduces to a one-body problem in a interaction potential and because the forces may be regarded as central, the potential depends only on the distance $r$ between the neutron and the nucleus.

The scattering process is described in term of wave mechanics. An incident plane wave moves along the $z$-axis until it encounters a potential on which it scatters and a radial wave (scattered particles) travels away from the scattering center. A plane wave of slightly diminished amplitude (the unscattered particles) continues along the $z$-axis and travels beyond the region containing the potential.

The incident wave is the solution to the Schrödinger equation for a free particle with energy $E$ and wavenumber $k = \sqrt{\frac{2mE}{\hbar^2}}$.

$$\psi_{\text{inc}} = e^{ikz}. \quad (2.1.1)$$

The scattered wave at distances far from the region of interaction, at sufficiently large $r$ when the interaction can be neglected, must represent a free particle moving radially outward. Its energy must be the same as that of the incident particle, its amplitude dependent on the polar angle $\theta$ relative to the $z$-axis (because of symmetry there exists no dependence on $\varphi$) and its magnitude inversely proportional to $r$, since the number of particles must be conserved.
The scattered wave must thus have the form:

$$\psi_{sc} = f(\theta) \frac{e^{ikr}}{r},$$

(2.1.2)

where $f(\theta)$ is the scattering amplitude, a complex quantity [1].

Far from the scattering center the complete wave function is then:

$$\psi \propto e^{ikz} + f(\theta) \frac{e^{ikr}}{r}.$$  

(2.1.3)

The differential cross section can be expressed in terms of the scattering amplitude. The number of neutrons scattered in the radial direction per unit time is

$$r^2 v |\psi_{sc}|^2 = \frac{r^2 v}{V} \left| f(\theta) \frac{e^{ikr}}{r} \right|^2 = v |f(\theta)|^2,$$

where $v$ is the neutron velocity and $V$ is some large (but finite) volume ensuring the normalizability of the wavefunction.

The incident neutron flux is

$$v \frac{|e^{ikz}|^2}{V} = \frac{v}{V}$$

and the differential cross section is the ratio [3,4]:

$$\frac{d\sigma}{d\Omega} = |f(\theta)|^2.$$  

(2.1.4)

Because the forces are central, the angular momentum of the neutron relative to the scattering center is a constant of the motion [3]. It is therefore convenient to expand the wavefunction in a series of “partial waves” which are the eigenfunctions of angular momentum - the Legendre polynomials $P_l$. For the incident wavefunction we obtain [1]

$$\psi_{inc} = e^{ikz} = e^{ikr \cos \theta} = \sum_{l=0}^{\infty} (2l + 1)^{\frac{l}{2}} j_l(kr) P_l(\cos \theta),$$

(2.1.5)

where $j_l(kr)$ is the spherical Bessel function of order $l$ and $i = \sqrt{-1}$.

A great simplification is possible, if only slow neutrons are considered. In the semiclassical picture, the angular momentum $L$ of a particle is equal to the product of its linear momentum $p$ and the impact parameter $b$. On the other hand the angular momentum is quantized: $L = \hbar \sqrt{l(l+1)}$, $l = 0, 1, 2, \ldots$ and consequently to each value of $l$ corresponds some average effective impact parameter i.e. a minimum distance $b_l = \frac{\hbar \sqrt{l(l+1)}}{p} = \frac{\lambda}{2\pi} \sqrt{l(l+1)}$ to which a particle with angular momentum $L$ approaches the interaction center.

Slow neutrons have wavelengths of the order of nanometers, which is much greater than the region in which the interaction potential is non negative (because neutrons feel only the short range nuclear force) and thus effective impact parameters for $l$, greater than 0, are much larger than the interaction region. This means that in slow neutron scattering only partial waves of $l = 0$ - the “s-waves” are scattered [3].

One of the consequences of this simplification is that the scattered wavefunction has zero angular momentum and the scattering amplitude is independent of angle i.e. the scattering is isotropic [3].
2.2 Scattering length

The scattering length is related to the scattering amplitude in the low energy region and is defined by:

\[ a = - \lim_{k \to 0} f, \quad \text{(2.2.1)} \]

and is a positive quantity for most nuclei, usually on the order of the nuclear radius. If the incident neutron interacts with a nucleus that is a “rigid sphere” of radius \( r_0 \), the wavefunction should vanish at \( r_0 \). For \( k \to 0 \) this holds when \( a = r_0 \), the radius of the rigid sphere (see Eq.(2.1.3)). Neutron-nucleus interactions are in reality not as simple, so the scattering length can be understood as the “radius of an equivalent rigid sphere” [3]. The scattering length can usually be considered to be a real quantity and in is energy independent below 1 eV. The total scattering cross section for a single nucleus is given by \( 4\pi a^2 \) and in subsequent sections the scattering length will be employed in place of the scattering amplitude [1, 3]. Eq.(2.1.3) will thus be expressed in the form

\[ \psi \approx e^{ikz} - a e^{ikr} \frac{r}{r}. \quad \text{(2.2.2)} \]

2.3 The Born approximation

It is possible, using the method of partial waves, to calculate the scattering amplitude \( f \), but this procedure can be laborious, particularly if a large number of partial waves must be considered. In contrast, the Born approximation has a limited range of applicability but is simple to implement. The primary condition for its validity is that within the interaction region the incident wavefunction be much larger than the scattered wavefunction, or in other words, the interaction potential between a particle and the scattering system is regarded as a small perturbation.

In the Born approximation the scattering length is given by

\[ a = -f(\theta) = \frac{m'}{2\pi\hbar^2} \int_{\text{all space}} e^{i(k_0 - k')r'} V(r') \, d^3r', \quad \text{(2.3.1)} \]

where \( m' \) is the reduced mass of the system, \( V(r) \) is the interaction potential, and \( k_0 \) and \( k \) are the incident and scattered wavevectors [1].

This approximation is inapplicable to the calculation of the amplitude of scattering of a slow neutron by a nucleus, because the interaction potential is many times greater than the incident neutron kinetic energy and the wavefunction in the nucleus is nothing like a plane wave. If, however, the scattering length is known experimentally, the approximation can be used and scattering cross sections of atomic systems (crystals, molecules) can be calculated. This is due to the fact that the volume of the nuclei where the wavefunction appreciably differs from the plane wave is negligible compared to the total volume of the atomic system or, put differently, because the scattering length (\( \sim 10 \) fm) is much smaller than the neutron wavelength (\( \sim 1 \) Å) and the interatomic spacing [3]. To this end, Fermi proposed choosing the simplest possible potential in Eq.(2.3.1): a delta function centered on the nucleus:

\[ V(r) = A\delta(r), \quad \text{(2.3.2)} \]

where the constant \( A \) is to be adjusted so that the Born approximation if forced to give the proper free-nucleus scattering length. The full interaction potential of the system is then the sum of such potentials, centered on different nuclei \( R_i \) with constants \( A_i \), where the index \( i \) ranges over all atoms of the system

\[ V(r - R_i) = \frac{2\pi\hbar^2}{m'_i} a_i \delta(r - R_i), \quad \text{(2.3.3)} \]
where $\mathbf{r}$ and $\mathbf{R}_i$ are the position vectors of the neutron and the $i$-th nucleus and $m'_i$ and $a_i$ are the reduced mass and the free-nucleus scattering length of the $i$-th nucleus. This is known as the Fermi pseudopotential [1].

3 Cooperative effects in the thermal region

Having introduced the necessary formalism in the last section, attention can be turned to the investigation of slow neutron cross sections. In this section three cooperative phenomena (so called because they result from cooperation of more than one scattering center) will be presented in more detail.

3.1 Reduced mass effect

Because the scattering length $a$ is proportional to the reduced mass of the neutron-nucleus system (Eq.(2.3.1)), and the scattering cross section is given by $4\pi a^2$ (see section 2.2), it is in fact proportional to the square of the reduced mass of the system: $\sigma_S \propto m'^2$.

If a slow neutron were to interact with a free nucleus, the scattering cross section would be given by $\sigma_S = \sigma_{\text{free}} = 4\pi a^2$, the “free atom scattering cross section”, and would be constant in the absence of near-by resonances because the scattering length is constant at low energies. If, on the other hand, the atoms are bound in a molecule or crystal, the measured cross section would be $\sigma_{\text{free}}$ only if the incident neutron energies were high enough (above $\sim 1$ eV) that the chemical bonds could be ignored and the atoms effectively free.

If the neutron energy is lower than the binding energy of the atoms in the molecule or crystal, the atoms are, because of chemical bonds between them, unable to rebound freely, and the effective mass of the scattering center increases. The measured scattering cross section would behave as:

$$\sigma_S = \sigma_{\text{free}} \left( \frac{m'_{\text{eff}}}{m'_{\text{free}}} \right)^2,$$

where $m'_{\text{free}}$ is the reduced mass of the free atom and neutron system and $m'_{\text{eff}}$ is the effective reduced mass of the system:

$$m'_{\text{free}} = \frac{m_n M}{m_n + M}, \quad m'_{\text{eff}} = \frac{m_n M_{\text{eff}}}{m_n + M_{\text{eff}}}.$$

The latter is a function of incident neutron energy because the effective mass of the target atom $M_{\text{eff}}$ is a function of energy and varies between the molecular or crystal mass $M^*$ at very low energies, where the molecule or crystal recoils as a whole because it acts as a rigid system and the atomic mass $M$ at high energies, where the atom acts as though free [1].

In the low-energy limit the scattering cross section is called the “bound atom cross section”

$$\sigma_b = \lim_{E \to 0} \sigma_S(E) = \sigma_{\text{free}} \left( \frac{1 + \frac{m_n}{M}}{1 + \frac{m'_n}{M'}} \right)^2,$$

and its ratio to the free atom cross section depends on the mass of the scattering atom. The importance of the reduced mass effect declines with increasing atom mass. For heavy atoms it is nearly unimportant because the reduced mass is practically equal to the neutron mass, as is the reduced mass of a heavy atom bound in an even heavier molecule, and so the ratio is unity.
For lighter atoms on the other hand, the effect can be quite pronounced. This is best illustrated
by hydrogen $^1\text{H}$ in five cases: atomic hydrogen gas, molecular hydrogen gas, water, paraffin and
a molecular crystal. Ratios of the cross sections are given in Table 1. The bound atom cross
section of hydrogen varies between 1.78 and 4 times that of the free hydrogen, depending on
the mass of the molecule in which it is bound. The cross section for scattering by hydrogen and
compounds in which hydrogen is bound is, therefore, four times larger at low energies than at
energies much higher than the chemical bond [3].

<table>
<thead>
<tr>
<th>Scatterer</th>
<th>Molecular mass</th>
<th>$\sigma_b/\sigma_{free}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$\text{H}_2$</td>
<td>2</td>
<td>$(4/3)^2 = 1.78$</td>
</tr>
<tr>
<td>$\text{H}_2\text{O}$</td>
<td>10</td>
<td>$(20/11)^2 = 3.31$</td>
</tr>
<tr>
<td>$\text{C}<em>{22}\text{H}</em>{46}$</td>
<td>310</td>
<td>$(620/311)^2 = 3.98$</td>
</tr>
<tr>
<td>Crystal</td>
<td>$\infty$</td>
<td>4</td>
</tr>
</tbody>
</table>

Table 1: Bound and free atom cross section ratios for hydrogen and hydrogen compounds [1].

3.2 Coherent scattering

Slow neutrons have wavelengths of the order of the interatomic spacings in molecules and crys-
tals i.e. a few Å and therefore coherence effects caused by interference of waves, scattered from
various nuclei become detectable, particularly in crystalline solids, but to a lesser extent also in
liquids.

In analogy with coherent scattering of light, interference effects in scattering of neutrons can
occur only between waves that describe neutrons having the same energy (wavelength) and
the same spin state (polarization) in the laboratory system. Scattered waves from different
nuclei are independent and of the form of spherical s-waves (Eq.(2.1.2)) and the total scattered
wave at laboratory point $r$ is the sum of the individual waves from all nuclei:

$$\psi_{sc} = \sum_{i=1}^{N} -a_i e^{ik'\cdot|r-R_i|},$$

where $a_i$ is the scattering length of the $i$-th nucleus, $k'$ is the wavevector of the scattered neu-
tron of magnitude $k' = \sqrt{2m_nE'/\hbar}$, $E'$ being the laboratory energy of the scattered neutrons
and $R_i$ is the laboratory position of the $i$-th nucleus.

An important property of coherent scattering is that only the angular distribution of scattered
neutrons is affected i.e. coherent scattering affects only the differential cross section, not the
total cross section, because the number of neutrons is conserved. Illustration of the effect of
coherent scattering on the cross section is done by analysis of a simple system of two identi-
cal, spinless, rigidly fixed nuclei, separated by vector $R_n$, on which a plane wave of the form
Eq.(2.1.1) is incident, as in Figure 2.
Here only the special case for $\varphi = 0$ will be considered corresponding to scattering in the plane of the paper (or screen). The scattered wave is detected in the direction $(r, \theta)$ where the distance $r$ is much greater than the separation between the nuclei. If the origin of the coordinate system is fixed on the first nucleus, the scattered wave from this nucleus has the form

$$\psi_{s,1} = -ae^{ik' \cdot r}/r,$$

(3.2.2)

where $a$ is the bound atom scattering length (given by the square root of Eq.(3.1.3)), $k'$ the outgoing wavevector and $r$ the vector to the detector. The scattered wave function from the second nucleus at the detector is given by a similar expression, but in which the phase difference between the two scattered waves must be taken into account. From Figure 2

$$k_{AB} + k'_{BC} = k_r \cdot R_n - k' \cdot R_n = -q \cdot R_n,$$

where $q = k' - k$ is the momentum transfer vector.

The second scattered wave is then

$$\psi_{s,2}(r, \theta) = -ae^{ik' \cdot r - iq \cdot R_n}/|r + R_n| \approx -ae^{ik' \cdot r - iq \cdot R_n}/r,$$

(3.2.3)

where the approximation was made that the separation between the nuclei is much smaller then the distance to the detector and thus the inverse-$r$ term is nearly unaffected. The total scattered wavefunction can now be written in the form

$$\psi_s = \psi_{s,1} + \psi_{s,2} = -ae^{ik' \cdot r}/r [a + a e^{-iq \cdot R_n}].$$

(3.2.4)

The bracketed part of Eq.(3.2.4) can be thought of as an "effective" scattering length of the system, and by Eq.(2.1.4) the differential cross section per nucleus can be written as

$$\frac{d\sigma_S}{d\Omega} = \frac{1}{2} |a + ae^{-iq \cdot R_n}|^2 = a^2 [1 + \cos(q \cdot R_n)],$$

(3.2.5)

where the factor one-half is introduced because there are two nuclei.

If only elastic scattering occurs, the vector $k'$ and $k$ are of the same magnitude and the dot product in Eq.(3.2.5) simplifies to $q \cdot R_n = kd \cos \theta = 2 \pi d \cos \theta / \lambda$, where $d$ is the magnitude of $R_n$ an $\theta$ is the scattering angle. Two limiting cases can be studied: at high energies $kd \gg 1$ the cosine in Eq.(3.2.5) oscillates rapidly for minuscule differences of scattering angle. In this case a detector would, because of its finite size and solid angle relative to the scattering center, see only the average, which is zero.

The differential scattering cross section in this case is $\frac{d\sigma_S}{d\Omega} \simeq a^2$ and the total cross section is

$$\sigma_s = \int_0^{\pi/2} \frac{d\sigma_S}{d\Omega} d\Omega = 4\pi a^2.$$

(3.2.6)
In the low energy limit $kd \ll 1$, the differential cross section is $\frac{d\sigma_S(\theta)}{d\Omega} \simeq 2a^2$ and the total cross section is

$$\sigma_S = 8\pi a^2.$$ (3.2.7)

The cross section per nucleus in the low energy limit is seen to have twice the value it would have if the nucleus were isolated. In the intermediate region the cross section is a function of the scattering angle $\theta$ and plots for different values of $d/\lambda$ are show in Figure 3.

The conclusion is similar to that of the previous section: neutron scattering cross section is higher because of the effects of binding. As the neutron energy decreases, the neutron wavelength increases and the neutron can no longer “see” the structure of the molecule or crystal on the nuclear scale and feel the potentials of the isolated nuclei making up the molecule. Instead, the neutron “sees” the target as a sum of the individual scattering lengths of the constituent nuclei and the cross section for scattering increases. Even though the amplitude of the incident neutron wave is (because the scattering length is small $\sim 10$ fm) almost unchanged in a scattering event with a single nucleus, extinction of the incident neutron beam becomes significant if the neutrons are of sufficiently low energy and there are a multitude of scattering centers [1].

### 3.3 Incoherent scattering

In an experiment, the part of the cross section that shows angular dependence would be identified as the “coherent cross section” and the constant background on which it lies would be identified as the “incoherent cross section”, but theoretically the difference lies in how the different scattered waves and scattering lengths are summed.

Any violations of the conditions imposed in the previous section will result in some degree of incoherent scattering of which there are three fundamental sources: scattering off unidentical nuclei, scattering off a system of nonconstant internuclear spacing and scattering off nuclei of nonzero spin [1]. Only the first two will be described in this seminar.

#### 3.3.1 Nonidentical nuclei

Targets made of nonidentical nuclei (different chemical elements or different isotopes) have more than one scattering length and neutrons scattered from it will partly scatter incoherently. Considering a similar case of two nuclei as in section 3.2 but with different scattering lengths, an analogue of Eq.(3.2.5) can be written

$$\frac{d\sigma_S}{d\Omega} = \frac{1}{2} |a_1 + a_2 e^{-i\mathbf{q} \cdot \mathbf{R}_n}|^2 = \frac{1}{2} [a_1^2 + a_2^2 + 2a_1a_2 \cos (\mathbf{q} \cdot \mathbf{R}_n)].$$ (3.3.1)

In this case, as the cosine ranges from $-1$ to $+1$ the cross section no longer ranges from 0 to 1, but from $(a_1 - a_2)^2/2$ to $(a_1 + a_2)^2/2$. The minimum would appear in the experiment as a constant isotropic background.

In general the differential cross section per nucleus can be written in the form

$$\frac{d\sigma_S}{d\Omega} = \frac{1}{N} \left| \sum_{n=1}^{N} a_n e^{-i\mathbf{q} \cdot \mathbf{R}_n} \right|^2,$$ (3.3.2)

and can be factored into two terms: one independent of the scattering angle and one dependent on it. The result is given by

$$\frac{d\sigma_S}{d\Omega} = \left\{ \langle a^2 \rangle - \langle a \rangle^2 \right\} + \frac{1}{N} \left\langle a \right\rangle^2 \left| \sum_{n=1}^{N} e^{-i\mathbf{q} \cdot \mathbf{R}_n} \right|^2,$$ (3.3.3)
where $\langle a \rangle = \sum_{n=1}^{N} a_n$ is the average value of all the scattering lengths and $\langle a^2 \rangle = \sum_{n=1}^{N} a_n^2$ is the average of the squared scattering length [1,3].

Equation (3.3.3) is the basis for the definitions of the two different types of scattering. The first part in curly brackets is defined as the incoherent cross section because it is independent of angle and the second part, the coherent cross section, is just the generalization of Eq.(3.2.5) from the last section. The term coherent scattering originates from the fact, that in the second part of Eq.(3.3.3) the phases are summed coherently, meaning summed before being squared. It is of note that if all the scattering lengths were identical, then $\langle a^2 \rangle = \langle a \rangle^2$, the incoherence would disappear and the cross section would be completely coherent.

The total cross section is commonly expressed in terms of two cross sections, corresponding to coherent in coherent scattering,

$$\sigma_S = \sigma_{coh} + \sigma_{inc},$$

(3.3.4)

where in terms of scattering lengths the total cross section is written as

$$\sigma_S = 4\pi \langle a^2 \rangle,$$

(3.3.5)

the coherent cross section as

$$\sigma_{coh} = 4\pi \langle a \rangle^2,$$

(3.3.6)

and the incoherent cross section as the difference of the two

$$\sigma_{inc} = 4\pi \left[ \langle a^2 \rangle - \langle a \rangle^2 \right].$$

(3.3.7)

### 3.3.2 Nonconstant internuclear spacing

Coherence is known to diminish because of the violation of constancy of internuclear spacings, the chief example of the validity of this assertion being monoatomic gasses or amorphous solids, in which the atoms are randomly distributed and coherence effects nonexistent. In crystalline solids, however, coherence effects arise even if the constancy condition is somewhat violated e.g. because of defects.

Even in a perfect crystal incoherence still exists because of thermal motion of the atoms about their equilibrium positions. Because the time scale of atomic motion is much shorter than the time of observation, the positions of atoms/nuclei in a crystal are no longer delta functions but rather distributions.

Formally this is taken into account by modification of the expression for the differential cross section, Eq.(3.3.2), by introducing a probability distribution for the position of the nucleus

$$\frac{d\sigma_S(\theta, \varphi)}{d\Omega} = \frac{1}{N} \sum_{n=1}^{N} a_n \left| \int e^{-i\mathbf{q} \cdot \mathbf{R}_n} P_n(\mathbf{R}_n) \ d^3R_n \right|^2,$$

(3.3.8)

where $P_n(\mathbf{R}_n)d^3R_n$ is the probability of finding the $n$-th nucleus in volume element $d^3R_n$ about point $\mathbf{R}_n$ and is in fact the absolute square of the wavefunction of the $n$-th nucleus, and depends on the details of the potential binding the atom to its lattice site [1].

### 3.4 Inelastic effects

Molecular liquids and crystalline solids, i.e. almost all reactor materials, consist of atoms bound together by interatomic forces and thus contain energy levels. In the energy range below $\sim 1$ eV these are generally discrete and separated by $\sim 0.1$ eV. Above $1$ eV they begin to overlap and above $\sim 10$ eV, the binding energy of a typical atom in a molecule or crystal, they become
continuous. Interactions, in which the energy levels of the target are affected, are called inelastic scattering. Neutrons can gain or lose energy only in amounts, determined by intervals between levels in the target, hence interactions of slow neutrons with these materials depend critically on the energy level distributions i.e. on the frequency spectra of target molecules or crystals, the calculation of which is the problem of condensed matter physics.

Energy levels in molecules and crystals are of three basic types: electronic, vibrational and rotational, the latter two being most strongly affected by neutron collisions.

To explain how vibrational levels affect the cross section, a diatomic molecule is considered, in which one atom is much heavier than the other, for example the hydrogen atom in hydrates of heavy metals. This is the well known problem of the quantum mechanical harmonic oscillator and the energy levels are equidistant and given by $E_n = \hbar \omega (n + 1/2)$, where the quantum number $n$ takes integer values and $\omega$ is the oscillation frequency. A quantum of energy is thus $\hbar \omega$ and a neutron can gain or lose some integer multiple ($n$) of this quantum in an inelastic scattering event with the molecule [1].

If the incident neutron energy is less than the energy quantum, transmission of energy to the molecule is impossible. Similarly, transmission of energy from the molecule to the neutron is impossible, if the molecule is in its ground ($n = 0$) state.

In Figure 4 the total scattering cross section for a harmonically bound hydrogen atom in the ground state is plotted and the importance of the discrete energy levels is illustrated.

The cross section exhibits characteristic “bumps” with minima near neutron energies that are multiples of $\hbar \omega$, which are due to the disappearance at these points of one of the inelastic scatterings for some $n$. If a neutron has energy of a little less than $n\hbar \omega$, the transition of the hydrogen atom to state $n$ is impossible, but all other transitions for $n' < n$ are possible. If, however, the neutron has energy of a little more than $n\hbar \omega$, the transition to $n$ becomes possible.

An example of a compound in which hydrogen can be accurately regarded as harmonically bound is zirconium hydride ZrH$_{1.5}$. The measured versus calculated cross sections are shown in Figure 5.

Because the scattering cross section for hydrogen is much larger than that of zirconium, the measured cross section is almost completely that of hydrogen. The experimental curve exhibits the characteristic minima from the position of which the oscillation frequency can be determined. The calculated and measured cross section are in relatively good agreement [3].

---

Figure 4: Inelastic scattering from state $n' = 0$ to state $n' = n$ (dashed line) and total (full line) cross sections of harmonically bound hydrogen in its ground state. The reason for the $1/E$ dependence of the elastic scattering cross section is that the plot is in the laboratory system, in which this dependence is typical. Also shown is the reduced mass effect, because of which the cross section is four times larger than that for a single, free hydrogen nucleus at energies close to zero. Adapted from [3].
Figure 5: Fit of the total measured scattering cross section of hydrogen obtained by subtracting the constant cross section of Zr from the total cross section of ZrH (solid line), data point of the measurement (dots) and the theoretical curve for harmonically bound hydrogen with $\hbar \omega = 0.137$ eV (dashed line). Adapted from [3].

The illustration of the effects of the rotational levels on cross section is possible by analysis of the rigid rotator, but will not be done here. The energy eigenvalues are given by $E_J = \frac{\hbar^2}{2I} J(J + 1)$, where $I$ is the moment of inertia of the system and $J$ the rotational quantum number. Unlike vibrational levels, rotational levels are not equidistant and are much closer together and even though the mechanism of how they would affect the scattering cross section is in principle similar, the cross section itself would look completely different.

Of course, actual molecules are not as simple: not all molecules are diatomic, the vibrational motion is not purely harmonic, rotation is only approximately rigid, and their frequency spectra, and especially those of crystals, are much more complicated.

4 Van Hove theory

Even though in this seminar the effects on scattering cross sections were, for clarity, presented one by one by means of the most simple of models, a more general theory of slow neutron scattering in which these effects are automatically contained exist, first developed by Leon Van Hove, in which the Born approximation and the Fermi pseudopotential are used to derive expressions for double differential cross sections. The central part of the theory is the “scattering law” $S(\alpha, \beta)$, known in condensed matter as the “dynamic structure factor”.

In the Born approximation, the coherent and incoherent double differential scattering cross sections (differential cross section per unit interval of neutron outgoing energy) are given by

$$
\frac{d^2\sigma}{d\Omega dE_f}^{coh} = \frac{k_f}{k_i} \sum_{\tau} \sum_{i=1}^{N} a_i^{coh} \left< \tau | e^{-i\mathbf{q} \cdot \mathbf{R}_i} | \tau_0 \right> ^2 \delta \left( E_\tau - E_{\tau_0} + \frac{\hbar^2}{2m} (k_f^2 - k_i^2) \right), \quad (4.0.1a)
$$

$$
\frac{d^2\sigma}{d\Omega dE_f}^{inc} = \frac{k_f}{k_i} \sum_{\tau} \sum_{i=1}^{N} \left< \tau \left| e^{-i\mathbf{q} \cdot \mathbf{R}_i} \right| \tau_0 \right> ^2 \delta \left( E_\tau - E_{\tau_0} + \frac{\hbar^2}{2m} (k_f^2 - k_i^2) \right), \quad (4.0.1b)
$$

where the neutron initial and final wavenumbers as well as the final energy are labeled by the indices $f$ and $i$, and $\tau_0$ and $\tau$ are the initial and final state of the scattering system [3].

Whereas the differential cross section contains information about the space correlations between the atoms in a sample, the double differential cross section contains information about both space and time correlations [4]. The incoherent part of the double differential cross section depends only on the correlation between positions of the same nucleus at different times and is zero, if there is no incoherence in the sample (section 3.3), while the coherent part depends on
the correlation between positions of different nuclei at different times [1, 4]. The cross sections can be rewritten in terms of the dynamic structure factor, which was done by Van Hove, and in the simplest case where incoherence is absent, the double differential cross section is

\[
\frac{d^2\sigma}{d\Omega dE_f} = \langle a \rangle^2 \frac{k_f}{k_i} S(q, \omega),
\]

(4.0.2)

where \( q \) and \( \omega \) are the momentum and energy change of the neutron and \( S(q, \omega) \) is the four dimensional Fourier transform of the time-dependent pair-correlation function \( G(r, t) \) given by [1, 4]

\[
S(q, \omega) = \frac{N}{2\pi\hbar} \int e^{i(q \cdot r - \omega t)} G(r, t) \, d^3r \, dt.
\]

(4.0.3)

The meaning of the function \( G(r, t) \) for a classical system is, that it represents the probability that if there was a particle at the origin at time \( t = 0 \), the same or any other particle will be found at some later time \( t \) at some position \( r \) [4]. In other words, it is the average density distribution at a time \( t + t' \) as seen from the point where a particle passed at time \( t \) [9].

In practice, the dynamic structure factor is written in a dimensionless form \( S'(q, \omega) = (k_B T) e^{-\frac{\hbar \omega}{2k_B T}} S(q, \omega) \) and the double differential cross section is expressed in the form

\[
\frac{d^2\sigma}{d\Omega dE_f} = \langle a \rangle^2 \frac{k_f}{k_B T} e^{-\frac{\beta}{2}} S'(\alpha, \beta),
\]

(4.0.4)

where \( S'(\alpha, \beta) \) is called “the scattering law”, \( \alpha = \left[ E_i + E_f - 2\sqrt{E_i E_f} \cos \theta \right] / Ak_B T \) and \( \beta = \hbar \omega / k_B T \) are the dimensionless momentum and energy exchange and \( A \) is the “mass of the principal scatterer” (the principal scatterer of a system of scatterers is chosen to be the one with the highest cross section) in neutron mass units [1].

Unfortunately, the theory is of limited use in reactor theory, because the calculated cross sections are not of sufficient accuracy to preclude the need for their measurement. Even though rigorous, the theory depends on the determination of the scattering law, which is the Fourier transform of the space-time correlation function, which depends on details of the dynamic structure, some of which are unknown [1].

**Conclusion**

Effect on neutron scattering cross section which have been described in this seminar are due to the fact that at low incident neutron energies the scatterer nuclei can no longer be considered free, but the effects of binding have to be taken into account. In scattering events such low energy neutrons exchange momentum not only with a single nuclei, but with molecules, their wavefunctions can interfere with those of other neutrons instead of scattering isotropically and their energy changes in quanta instead of continuously. As a result, cross sections in the low energy region are higher, but modeling of thermal cross sections is difficult because the effect are complex and simultaneous. Estimating the uncertainties of model calculations and the propagation of these uncertainties to uncertainties in reactor physical parameters calculated by using the neutron transport equation is the next big challenge in reactor physics.
References


