The Basic Physics of Neutron Scattering Experiments

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Introduction.

In preparing the present lecture I have tried to complement the other contributions to this introductory course by a discussion of the basic physical principles behind the well-established but also developing practice of neutron scattering experiments. Keeping in mind all the various kinds of instruments and techniques which will be covered in the lectures to follow, I did not try to give a list of all the tools and methods used in these instruments. I will invoke a few examples only, either to illustrate the physical principles or to give an idea of the variety, importance or magnitude of various phenomena. The most intriguing aspect of these principles is that in the technique used in neutron scattering experiments contradicting quantum mechanical and classical considerations are inextricable mixed in a fashion which can rightly be called a paradox. For example, while in wave mechanics a very well defined momentum state of a particle has to be extended in real space, classically treated neutron beams are the less spread out in space the more the neutron velocity is well defined. In recent years it has been argued, that this paradox can/should be resolved by treating quantum mechanically not only the microscopic interaction of the neutron with matter in the sample, but its whole evolution within the more than macroscopic instrument. I see the evolution of neutron scattering experimental techniques in another direction: in the capability of taking into account more and more important and sometimes decisive finer details by using more and more realistic mathematical models of the evolution of the neutrons from birth do death, eventually passing by the sample and being scattered more than one times. Working with such numerical "virtual instruments" we will have to go far beyond notions like resolution function, convolution etc, and actually eliminate a large number of approximations currently in use. In view of expecting for the future more precision and detail in the description of all that goes on, I found it compelling to re-examine the ground rules neutrons follow in our instruments and samples and to take another look at approximations which might or might not prove to be precise enough.

1) Neutron scattering experiments: particles and waves

A neutron scattering experiment in most general common terms consist of determining the probability for a neutron in a given initial state, fully characterized by its velocity vector **v** and the direction of its spin vector **S** (which we will define to be a unit vector pointing into a well defined direction) to end up in another state characterized by **v'** and **S'** upon having hit a sample. Some will be tempted to find this definition unsatisfactory: it seems to ignore the quantum mechanical nature of both the particle propagation and the spin. In fact it does not and this will be our first subject to examine: why can the propagation of neutrons through a spectrometer be treated classically while the interaction with the sample must be described by quantum mechanics. This paradoxical approach is the basis of all neutron scattering work. Before getting

tempted of trying to generalize this "unsatisfactory sloppiness" think of the following. In landing on the moon the computers controlling the spacecraft used Newtonian mechanics, and ignored both special and general relativity. Actually upgrading the computer power to take into account exactly the real but tiny relativistic corrections (an error of trajectory of less then 1 m by the time of reaching the Moon) might have delayed the success of the mission by years.

The classical description of the flight path of a neutron consists of assuming that it is geometrically point-like and it is subject to forces, such as gravity. Its spin can also be described by a classical vector, following the classical equation of motion of an inertial momentum under the effect of a torque $\mu \times \mathbf{B}$, where $\mathbf{B}$ is the magnetic field and $\mu = \mu \mathbf{S}$ is the magnetic moment of the neutron. Here μ=-1.913 nuclear magneton, and the – sign indicates that the magnetic moment and the inertial (spin) moment of the neutron point in opposite directions. It can be shown that this torque is fully accounted for by the Zeeman potential energy $-\mu \mathbf{B}$ [1]. This lead us to the Hamiltonian

$$H = \tfrac{1}{2} m \mathbf{v}^2 + mgh(\mathbf{r}) + V(\mathbf{r}) + \mathbf{S}\, \mathbf{I}(\mathbf{r}) - \mu \mathbf{B}(\mathbf{r}) \qquad (1)$$

which also exactly describes the interactions of the neutron while inside a material (if one neglects tiny relativistic effects occurring in very high electric fields near the nuclei and the by now undetected eventual electric charge and electric dipole moment of the neutron). Here the strong nuclear interaction with each of the nuclei in the sample is described by the Fermi pseudo-potential V (practically a delta function for each nucleus, which can also have complex values), which also can have a spin dependent part $\mathbf{I}$ for nuclei with non-zero spin, and the magnetic field $\mathbf{B}$ includes both the fields of macroscopic magnets and the microscopic fields produced by magnetic atoms inside a material. (Contrarily to the common misconception, the neutron does not directly interact with the magnetization of a sample, it only sees the $\mathbf{B}$ field produced by the magnetization.)

The classical solution of this equation (with the Fermi pseudo potential taken as an average of local atomic configurations) describes the trajectory of a neutron, including slight changes of direction (i.e. refraction) and velocity due to the potential energy terms, e.g. gravity over a long flight path or on entering materials. The only phenomenon limiting the validity of classical trajectories is the Stern-Gerlach effect: if the macroscopic magnetic field gradients are strong enough, the particle trajectories will split in direction and/or absolute velocity according to the two spin states "up" and "down" of the spin ½ neutron. This can be a measurable effect when we combine the use of superconducting magnets with extreme beam collimation or monochromaticity. The exact treatment of this situation is to follow the trajectories of the "up" and "down" spin states separately [1]. With this slight extra complexity this extremely rare practical case is thus also covered by the analysis which follows, where we will exclude Stern-Gerlach splitting effects.

In light optics where depending on the situation to be described one either uses geometrical optics or wave optics. With particle beams one is best advised to use the particle or the wave picture with the same opportunism. In neutron scattering experiments this does mean that the same neutron will be alternatively described as wave or particle along its adventures from being born in a nuclear reaction to its death in our detector (which works by detecting secondary

particles emitted after neutron absorption) or in some other absorbing material part of the experiment or to its escape from the realm of our considerations. The escaped neutrons can become mixed to the air as an extremely dilute gas (with still some 10^4 neutrons impinging on a 1 m^2 surface from any direction in experimental halls), end up in somebody else's detector as annoying background or absorbed by some material somewhere afar. Few neutrons will have the chance to die by the natural decay with about 15 min halftime, other dangers hit on a much shorter time scale. In air mean free path of thermal neutrons is about 200 m for absorption primarily by the nitrogen (which amounts to about 100 ms lifetime) and by about an order a magnitude shorter for scattering collisions which are responsible for mixing the neutrons to the air as another gas.

To determine the right method to use in following a particle along its destiny starts with the characterization of its source. All neutrons sources emit incoherent radiation as do all light sources with the exception of lasers. This means that the wave functions associated with particles emitted at different times and positions have no correlation in their phases. Being a fermion, there is no danger for the neutron to ever become produced by lasers. The high energy, several MeV neutrons originating from some nuclear reaction (in practice fission or spallation processes and secondary follow ups) are slowed down to thermal energies by collisions with nuclei in the containers called moderators, which are the actual sources of the low energy (<1 eV) neutrons used in scattering experiments in condensed matter. Thus, similarly to incandescent light sources, there is absolutely no coherence between the events that launch the various neutrons towards the beam lines. The initial wave function of the neutron is a matter of speculation, no experiment could ever been conceived to determine it, and thus no theory treated this irrelevant question. My best guess on the basis of very general quantum mechanical hand waving is that every neutron must be born with a huge uncertainty in energy and momentum, since the nuclear processes are very localized in space and time. This is a marked difference compared to monochromatic but incoherent light sources, such as Na vapor lamps. In this latter case the emitted radiation has a very well defined energy i.e. a narrow distribution of wavelengths. As a consequence, it makes sense to talk of a natural coherence length of the emitted light, which is just the inverse of the width of the wavenumber distribution and amounts to some 30 cm for the famous, historical Na vapor lamps. In contrast incandescent light sources emit light with coherence length that has never been determined, for the good reason that it is quite likely shorter than the wavelength. The same is true for neutron sources and all discussion in the literature about a natural coherence length of the neutrons, either longitudinal or transversal has no physical ground whatsoever.

We all know from our years at high school (eventually less and less, since lasers are squeezing out any other source of light from laboratory practices) that coherent radiation necessary for showing observable wave properties in the form of interference effects can be produced by the use of incoherent light sources too, as the founding fathers did it in the last century. All one has to do, is to use a good monochromator (which would only transmit a very narrow wavelength band) and/or a good collimator (e.g. distant narrow slits, as in the famous Young experiment). The first will introduce a longitudinal and the latter one a lateral (transversal) coherence, with coherence length defined by the inverse of the widths of the distributions of the corresponding components of the wavenumber. The Heisenberg uncertainty relation does not contain more physics than just this trivial property of "normal", Gaussian distribution of spatial or temporal frequencies, i.e. the inverse relation between coherence length and the width of the wavenumber

distribution. Using distributions with sharper edges than a Gaussian, one can observe correlation lengths apparently longer than the width of the wavenumber distribution, which is a very popular subject called "squeezed states" of radiation.

By now we have arrived to the point to answer the question we have started with: why can we characterize neutron states in scattering experiment by well defined velocities, as in classical mechanics. Geometrical optics is valid, if interference effects are suppressed by the fact that the geometrical dimensions of the beam are much larger than the coherence lengths. Let us consider the example of the lateral coherence produced by collimation. In the so called small angle scattering (SANS) experiments one produces a very well collimated neutron beam by two small diaphragms (typically 1 cm in diameter) at a large distance (typically 10 m), called pin hole geometry. In the experiment we will observe by a large (0.2 – 1 m^2) area detector the fraction of the scattered neutrons close to the original beam direction, within some 1 – 5°. For neutrons with $\lambda = h/mv \approx 6$ Å de Broglie wavelength, for example, the above 1 cm/10 m = 0.001 rad collimation will lead to a lateral width of the wavenumber $k = 2\pi/\lambda$ distribution of $0.001 \times 2\pi/6$ Å ≈ 0.001 $Å^{-1}$. This mean that the lateral coherence length is about 1000 Å= 10^{-5} cm. This is really much smaller than the 1 cm pinholes defining the lateral dimensions of the beam, so we are perfectly right to describe the neutron propagation by the classical point like trajectories. In contrast, with 0.03 mm diameter pin holes the lateral coherence length would be just equal to the beam width, and we should turn to the use of de Broglie wave description of the neutron propagation in our ultra high resolution SANS instrument. This can be the practical situation in synchrotron X-ray radiation experiments, where the source brightness is many many orders of magnitude superior to the neutron case. Such small pinholes are fully out of question for neutrons though: the 10^{10} fold reduction of beam intensity impinging on the sample would leave us with 1 neutron arriving every 15 min and one scattered neutron detected about every day.

As far as lateral coherence is concerned, SANS comes to "closest" to the wave optics limit, and this still means 5 orders of magnitude away. Considering the longitudinal coherence we arrive to similar conclusions. Here we have to consider the degree of monochromatization of the beam. In the so called back scattering spectroscopy (BS) Bragg reflection on perfect crystals (usually Si) at nearly 180° scattering angle (within a few tenth of a degree), one can obtain a wavelength distribution of 10^{-4} relative width at about 6 Å average wavelength, i.e. about 1 μm longitudinal coherence length, corresponding to about 2 ns neutron travelling time. Interference effects would again only matter if one would cut/modulate the beam in packets shorter (i.e. faster) than the above values. Choppers used to cut out neutron pulses from a continuous beam in order to determine the neutron velocity by observing the neutron time-of-flight over a well defined distance of a few meter, have characteristic opening times of a few μs at best.

Thus we conclude that on the length and time scales on which neutron beams can be shaped, modulated, analyzed in an experiment, i.e. beam cross sections on the cm scale in order to maintain reasonable beam intensities and beam pulses on many μs scale (limited both by the speed of mechanical devices and by the need to not to cut out too many neutrons), the neutron motion within the spectrometer simply reduces to classical flight of point-like particles with infinitely well defined magnitude and direction of the velocity. At this stage I will not go in more detail how various spectrometers look like and how they function, this will be discussed at great length in the talks to follow. We just can keep in mind, that indeed, an ideal neutron scattering

experiment is to observe the probability for the sample to change the initially perfectly defined neutron velocity **v** into a perfectly defined scattered neutron velocity **v'**. Similar analysis of the spin behavior shows [2] that it exactly follows the classical equation of motion determined by the variation of the magnetic field **B** as seen by the point like neutron along its trajectory: **B=B(r(t),t)**, where **r(t)** is the position of the neutron at time t.

One apparently paradoxical aspect of the situation merits a few more minutes of attention. One would think, that infinitely well defined classical particle velocity in the de Broglie wave picture corresponds to an infinitely extended plane wave, and it is therefore incompatible with the description as a point like particle. More careful quantum mechanical analysis, however, does not substantiate this contradiction [2]. In wave mechanical reality, the particles are described as wave packets containing plane wave components with a distribution of wavenumbers **k**. If this wave packet evolves in an environment, where according the above analysis geometrical optics is valid, this evolution will be just that of an ensemble of point like classical particles with a classical velocity distribution identical to the wavenumber distribution of the wave packet as defined by the de Broglie relation $m\mathbf{v}=\hbar\mathbf{k}$. Most remarkably, the well known expansion of wave packets of non-zero mass particles simply corresponds to the fact that in the equivalent ensemble of classical particles some particles go faster and than others and leave them more and more behind. The same applies in the lateral direction too, and if one wants to compute the quantum mechanical particle density in the wave packet at any given time it is sufficient to evaluate the classical particle density in the equivalent classical ensemble at the same time. This is an interesting and very useful symmetry: under conditions where wave optic effects are visible, the precise quantum mechanical behavior is given by the quantum superposition of the initial **k** eigenstates, while under conditions of validity of geometrical optics it is given by the ensemble of the corresponding classical trajectories. Both superpositions are incoherent in practice which means that one has never succeeded to maintain a well defined phase relation (coherence) between the various initial **k** components of wave packets in a fermion beam of particles with allegedly identical initial states. Thus even when interference is observed, it only involves interference of one initial **k** eigenfunction with itself within the **k** distribution of the wave packet.

2. **Types of processes and cross sections**

Neutron propagation in matter can only be described by taking quantum mechanical effects into account, quite to the opposite of what we have seen for neutron trajectories in the free space of collimation paths or time-of-flight tracks for velocity determination. One can distinguish three kinds of processes:

a) Refraction (transmission)
b) Reflection
c) Scattering (diffraction)

a) In a transmission process one can consider the classical particle trajectory as evaluated using eq. (1), with the Fermi and magnetic potentials taken as smooth averages over local atomic densities. (As a matter of fact, this procedure leads to the same result as exact quantum mechanical treatment of considering the modification of the original beam across the sample due to interference with the forward scattered partial waves from each atom.) Thus we will be able to describe the change of neutron beam direction and neutron velocity changes (refraction) as a

simple classical effect of traversing potential walls. One can introduce a neutron optical refractive index analogous to the well-known optical one, which reads

$$n = v/v_0 = 1 - U/mv^2 , \qquad (2)$$

where v and v_0 are the neutron velocities in the media and in vacuum, respectively, and U is the average potential in the media (which can include the internal magnetic fields **B** too for a given direction of the neutron magnetic moment μ). Using this refractive index one can calculate the classical particle trajectories using the formalism of geometrical optics. There is one difference though: the attenuation of the beam intensity due to absorption and scattering effects occurring in a probabilistic manner, as quantum transitions do, do not have classical analogs in particle motion, and have to be phenomenologically included. The neutron refractive indices are very close to unity, typically within 10 to 100 ppm, and with the Fermi potential being most often repulsive, for most materials $n < 1$. With the exception of the case of grazing incident angles, which will be discussed under the keyword reflection, the neutron refraction effects are so small, that they only matter in exceptional very high precision experiments. For a practical approximation in typical neutron scattering work one can assume that the neutron continues its trajectory without noticeable change of direction or velocity but with an attenuation which can be characterized by an attenuation range (depth for 1/e intensity loss) determined by the probability of nuclear absorption and scattering effects. For most solid materials the range amounts from a few mm to a few centimeters, in air it is about 20 m, and for materials used as neutron beam absorbers (e.g. diaphragms, slits, shielding), such as Gd, Cd, B it is but a very small fraction of a mm. Note, that the absorption probability drops rapidly with increasing neutron velocity, and there is no good neutron absorber for neutron energies above 1 eV (velocity above 14000 m/sec).

b) Another geometrical optical effect is the partial or total reflection of the beam on traversing a smooth surface between two media with different refractive indices. This phenomenon is perfectly well described by the optical Fresnel theory, and for n very little less then 1 we find that neutrons coming from the vacuum are totally externally reflected by the surface of the media at grazing incident angles less than a critical angle $\alpha_c = [2(1-n)]^{1/2}$. In view of eq. (2) we find, that the critical angle is inversely proportional to the neutron velocity, and for most materials it amounts to a few tenth of a degree at 1000 m/s neutron velocity, i.e. about 4 Å wavelength and 5 meV energy. Following Fresnel's theory, above the critical angle the probability of neutron reflection R rapidly drops to zero, actually approaching an asymptotic law $R \propto \alpha^{-4}$.

The total reflection effect is a key feature of neutron optics. In particular a neutron wave-guide tube can be built by using smooth glass walls, which act for the neutron beams as optical fibers. A neutron is kept within the guide by a series of total reflections on the walls if the angle between the neutron flight path and the axis of the guide tube does not exceed the critical angle. Compared to optical fibers the critical angle is vanishingly small in neutron guides, thus they only transmit neutrons with propagation direction very close to the tube axis. 200-400 layer interference mirror coating on the internal glass walls of the guide (so called neutron supermirrors) show high reflectivity up to 3 times the critical angle of Ni, the most often used coating material. This allows the guide to transmit more neutrons, e.g. at 4 Å wavelength up to 1.2 ° deviation from the glass tube axis. Ni and more and more supermirror coated guides, including more complex geometries than the simple straight, constant cross section original tubes, play an expanding role

in transporting neutron beams without substantial intensity losses to rather large distances, 50 – 150 m.

The reflection process only exactly follows geometrical optics rules, if the surface between media is perfectly smooth. Fresnel's analysis has shown, that ideal reflection actually requires a flat surface area of the size of the first Fresnel zone at least, which depending on the instrumental configuration and the neutron wavelength will measure about a few tenth of a mm in the direction of the projection of the grazing incident neutron path to the reflecting surface and about an order of magnitude less in the perpendicular direction. Surface roughness within the Fresnel zone will lead to reduced reflection probability and to diffusely scattered (so called non-specular) neutrons around the ideal (specular) direction of reflection. The waviness of the surface over distances larger than the Fresnel zone dimensions results, in contrast, in a blurring of the reflected beam direction, without substantial decrease of the total reflected intensity though.

Although theoretically totally reflecting focussing mirrors could considerably enhance neutron fluxes on small sample areas, and many of the sophisticated optical focussing techniques could also be applied for neutron beams. Due to the disturbning diffuse scattering effects on not ideally smooth surfaces, and to the geometrical problem of handling large beam cross sections at very small grazing incoming beam angles, the development of neutron optical focussing devices only is in its infancy.

c) In contrast to transmission (refraction) and reflection effects, genuine scattering processes cannot be apprehended by geometrical optical methods. In fact here full wave mechanical considerations are required. Since the variation of the Fermi pseudo potential reflects the arrangement of the atoms within the sample, here we are to do with objects in the beam whose characteristic dimensions are much less than the correlation lengths one can typically produce with collimating and monochromating neutron beams, therefore we have to use full quantum mechanical theory. Huygens's principle is a good guide in understanding the basics. Collective scattering effects will occur due to the interference of the spherical waves emitted by all of the nuclei hit by the beam. The experimentally observable quantities also have completely different nature than in the above two cases. Refraction and reflection experiments consist of intercepting the full deflected, attenuated beam, whose geometry is similar to that of the incoming beam we have defined by properly chosen diaphragms, eventually with some blurring due to wavy surfaces. In contrast, in scattering experiments we will have to do with determining the probability distribution of the scattered radiation. This distribution can be observed at three, increasingly demanding level of complexity. If we only are interested in the angular distribution of the scattered neutrons, the experiment is called *elastic*. If we also analyze the velocity dependence of the scattered radiation, we perform *inelastic* scattering work. And if the distribution of the spin direction of the scattered neutrons is also observed, we speak of *polarization analysis*. This latter option can also be performed at different degrees of detail accompanied by different degrees of complexity. One can limit the observation to the component of the spin vector **S** parallel to the magnetic field and, since the neutron spin has the value of ½, the first moment (average value) of any spin component can only determined. But it is also possible to determine the average value of all three spin components. This is called 3 dimensional (3D) polarization analysis, a technique which seems to contradict the notion that only one component of a spin ½ can be simultaneously determined. However, in reality this fundamental

quantum mechanical limitation is only valid for observing one single particle, and not for an average over an ensemble e.g. in a beam. 3D polarization analysis was first pioneered in the 1960's in Leningrad for transmission experiments, it is the basis of the Neutron Spin Echo (NSE) technique and it has been by now developed at ILL for scattering experiments at any scattering angle.

All these scattered beam distribution, of course, are to be related to the distribution of neutron parameters in the incoming beam. Ideally, one would desire to have an incoming been with a perfectly defined velocity **v** and spin **S.** Such a beam would, unfortunately, have zero intensity. Neutron sources always produce a broad Maxwellian distribution of neutron velocities, and selecting a given velocity or spin direction can only be achieved by throwing away all the others. (For spin ½ particles producing a beam with only spins pointing in a preselected direction **S** only means a theoretical loss of 50 %, but the polarizer devices used for selecting the chosen spin state introduce additional undue beam intensity losses which can range from a factor 1.5 at best to 10 or more at worst.) If one ignores parasitical intensity losses, such as finite reflectivity of optical elements or absorption in air or in other materials in the beam (such as vacuum windows), *Liouville theorem* of classical particle mechanics tells us, that the phase space density anywhere in the beam will be the same as in the neutron moderator. The phase space element in a beam can be defined as the product of the collimation solid angle $d\Omega$, the width of energy (or alternatively velocity) distribution dE and the beam cross section. Thus the flux (number of impinging neutrons in unit beam area perpendicular to the beam direction and in unit time) is

$$\varphi = \Phi \eta \, d\Omega \, dE / 4\pi \qquad (3)$$

where Φ is the source flux distribution as a function of energy usually defined for 4π solid angle (which can be looked up in user manuals of neutron centers or calculated if one knows the effective Maxwellian temperature of the source moderator) and $\eta < 1$ describes the intensity losses due to the finite efficiency of the instrument components. The hallmark 1.2×10^{15} thermal flux of the ILL reactor means $\Phi \approx 2\times10^{13}$ neutrons/cm^2 s meV around the peak of the 300 K Maxwellian distribution. Thus e.g. with 0.5° horizontal and 2° vertical collimation and at 1 meV beam monochromaticity (typical numbers for a thermal neutron triple axis experiment) we should ideally end up with 5×10^8 n/cm^{2}s flux on the sample. In actual fact, due to losses in various beam shaping elements, first of all in the monochromator crystal, in reality we rather get 2-5 times less. Since intensity is always a scarce commodity in neutron scattering, one always tries to optimize the incoming beam intensity by choosing $d\Omega$ and dE as large as compatible with the angular or energy resolution required. And polarized neutrons are only used when the unique information they deliver is indispensable.

It is obvious that the sample does not care about what kind of experiment we have decided or are equipped to perform. We might do an elastic scattering experiment while most of the scattering is inelastic. Or we might assume that we have do to with small angle scattering while in much of what we observe is refraction or reflection on interfaces. Or, a constant worry in all scattering experiments is the contribution from multiple scattering processes, which make data interpretation ambiguous or erroneous. Under certain ideal conditions the distribution of scattered neutrons can be described in terms of scattering cross sections. The angular distributions can be described by the differential cross section $d\sigma/d\Omega$, whose integral over all scattering angles in 4π

gives the total scattering cross section σ. The absorption of the neutrons by nuclear reactions in the sample can be accounted for by the absorption cross section σ_a which, similarly to σ, describes the fraction of neutrons removed by this process from the beam traversing the sample. The total attenuation of the beam intensity corresponds to the sum of these two cross sections. For inelastic scattering the double differential cross section $d^2\sigma/d\Omega\, dE'$ can be used, which gives the distribution of scattered neutrons both as function of scattering angle and final neutron energy E'. In polarization analysis experiments transitions from one initial neutron spin state to a final one can be described by introducing a number double differential cross sections for each of the various spin state transition. For example the $z \rightarrow x$ cross section will describe the angular and energy distribution of scattered neutrons with spin parallel to the +x axis, when the incoming neutron spin state was parallel to the +z axis.

The key point with the use of cross section formalism is that the scattering probability in a homogeneous and homogeneously illuminated sample must be proportional to the sample volume. Most commonly cross sections are given for one chemical formula unit of the sample (e.g. for one molecule, if applicable). The meaning of the cross sections σ and $d\sigma/d\Omega$, for example, is expressed by their relation to the total scattered beam intensity I or the beam intensity dI scattered at various scattering angles into solid angle $d\Omega$:

$$I = \varphi\, N\, \sigma\,, \qquad dI = \varphi\, N\, d\sigma/d\Omega\,, \tag{4}$$

where N is the number of formula units in the sample, and both intensities are expressed in neutrons/sec units. This means in particular that each formula unit of the sample removes by scattering from the incoming beam the number of neutrons impinging in 1 second on a cross sectional surface σ. Next we will examine the conditions for a sample to produce scattering effects proportional with the number of atoms in the sample, in agreement with eqs. (4), which turns out to be rather non-trivial.

3. Validity of the cross section approach

Theoretical analysis of the neutron (and similar) scattering phenomena in condensed matter by van Hove in the 1950's has shown, that the differential neutron scattering cross sections are directly related to space-time correlation functions describing the detailed atomic behavior of the matter. A particularly important feature of this theoretical finding is that the so called scattering function $S(q,\omega)$ does not directly depend on the incoming and outgoing neutron velocities **v** and **v'**, (i.e. 6 parameters) but only on the following 4 parameter combinations

$$\hbar\mathbf{q} = m\mathbf{v}' - m\mathbf{v} \tag{5}$$

$$\hbar\omega = \tfrac{1}{2}m\, v^2 - \tfrac{1}{2}\, m\, v'^2 \tag{6}$$

Here **q** is the called momentum transfer and $\hbar\omega$ the energy transfer. (Note that in some textbooks the energy transfer is defined with the opposite sign, and in scattering events it can take both

negative and positive values.) The relation between the scattering function and the double differential cross sections is:

$$d^2\sigma/d\Omega\, dE' = S(q,\omega)\, v'/v$$

Thus in neutron scattering work one has a large degree of liberty of choosing the incoming neutron beam parameters in order to achieve optimal experimental conditions. A best known example is the use of incoming neutron wavelengths $\lambda > 4.6$ Å in many diffuse and small angle scattering experiments: above this critical wavelength Aluminum, the most often used material for beam windows, sample holders, cryostat and furnace parts has no more active Bragg reflection, and thus spurious multiple scattering effects involving the sample environment are vastly reduced. This choice is of course excluded, if the neutron velocity $v = 2\pi\hbar/m\lambda$ is not large enough to achieve the high **q** values or energy transfers we are interested in, c.f. eqs. (5) and (6).

One important feature of neutron scattering studies of condensed matter is that the interaction between neutrons and matter is weak, and the van Hove theory of cross section, based on first order Born approximation, applies with a great precision. This implies that the neutron scattering probability in the sample is small compared to 1. Another way to put this is the condition that the scattering cross section of the sample is much smaller than its real, geometrical cross section. The total cross section of an atom varies randomly from one nuclear species to another (it can be found in tables) with $\sigma_a=10^{-23}$ cm^2 being a typical value. In a sample the spherical waves emitted in the scattering process by the individual atoms (the diameter of the scattering object, the nucleus, being much smaller than the neutron wavelength) interfere with each other, producing the final scattering pattern. Since the scattered intensity scales with the square of the scattered wave amplitude, the total scattering cross section for N atoms, which scatter in constructive interference (e.g. Bragg reflection in a compact perfect monocrystralline grain, or small angle scattering in the nearly forward direction on a precipitate), is $N^2\sigma_a/N^{2/3}$. The denominator here takes into account that the relative angular width of scattering range, within which each atom scatters in phase, scales as the inverse of the number of atoms along an edge of the grain (e.g. angular width of a Bragg peak). The diameter of the grain will be about $(N\, V_a)^{1/3}$, where V_a is the average volume per atom and it amounts to some 10^{-23} cm^3. Thus we find that the total scattering cross section of our grain will exceed its geometrical cross section for $N > V_a/\sigma_a^{2/3} \approx 10^8$. In other words, one can only expect that the a scattering process can be characterized by a cross section if the distance within which the atoms scatter coherently in the process we are investigating is not more than typically a few tenth of a μm. For weak scattering processes, such as inelastic effects. this distance could be allowed to be longer, but it is unlikely that the coherence length of dynamic processes really becomes that large.

From the point of view of the experimental procedure this implies, that it only makes sense to derive a cross section from the observed intensities (c.f. eqs. (4)) if we are can convince ourselves that much less than 10^8 atoms (or more for weak scattering phenomena) participate coherently in the process. In particular, above this limit the scattered intensity will not be proportional to the number of scattering atoms, and the very definition of the cross sections breaks down. The interpretation of data obtained under such conditions requires more complex procedures, such as exactly solving the Schroedinger equation of neutron motion in extended perfect crystals (dynamical diffraction theory). There are several practical consequences of this limitation of the

cross sections approach. For example, reliable powder diffraction experiments can only be performed in fine grained samples. Or in small angle scattering for objects approaching the μm size the Born approximation breaks down, and refraction and/or reflection effects on the surface of the objects become dominant. Often it is just the neutron scattering experiment which allows us to determine the correlation length within the sample, and to verify if the results can be expressed in terms of a cross section or not. The theoretical formulae used to calculate cross sections are usually based on first Born approximation and do not reflect this limitation of the formalism.

4. Experimental procedures: calibration and background

Using eqs. (4), or double differential similar ones, for the absolute determination of cross sections is not as straightforward as these equations would suggest. For example the incoming beam flux might not be exactly known, it might not be homogeneous over the whole sample cross section, the efficiency of the neutron detectors is less than 100 % and not exactly known or not homogeneous etc. Therefore absolute intensity measurements are always performed relative to a standard sample with a well-known cross section, volume and density. Pure (H free) Vanadium is a favorite choice. It scatters perfectly elastically (E = 0, cf. eq. (6)) and isotropically, except for small angle scattering ($q < 0.1$ Å^{-1}), the cross section is fairly well known and its absorbs moderately. It is important that the standard must have the same geometry as the sample and it must intercepts exactly the same part of the beam. For practically used V sheet thicknesses (0.5 – 1 mm), however, the absorption becomes not fully negligible and it may make the scattering somewhat anisotropic depending on the form of the sample. In addition multiple scattering also occurs, i.e. a neutron after having already been scattered in V undergoes another scattering process before leaving the material, which can also make the scattering anisotropic. These perturbations are not important in usual precision work, but may deserve attention in some cases.

Calibration by standards also is crucial for determining the resolution of instruments. Thus diffractometers are easily calibrated by well-known powder samples of low absorption. For inelastic scattering experiments the calibration is more delicate. Only elastically scattering standards are well known enough, e.g. V, and one can thus easily determine the instrumental energy transfer resolution around E = 0. For other energy transfers the resolution can more or less reliably determined by extrapolation using a mathematical model of the spectrometer.

Another important instrumental parameter, the background, can also only be determined by measurement. In all experiments many neutrons reach the detector which do not come from scattering events in the sample, but rather from neutrons scattered by the air, by other spectrometer components or by neighboring experiments. These latter ones are particularly dangerous, since they can change with time, depending on what goes on at the neighbours. The other main problem with the background is that it is modified by the presence of the sample. On the one hand side, absorption by the sample attenuates the intensity in many neutron trajectories contributing to the background. This effect can be relatively easily taken care of. One determines the background in the exact experimental configuration in the absence of the sample (i.e. for the empty sample holder, if there was one) and with the sample replaced by a total neutron absorber (e.g. Cd) of the same size and shape as the sample and placed at the same position. Interpolating between these two background data, i.e. between 0 and 100 % absorption for the value of the

sample absorption gives the best guess. Note, that since a beam traversing the sample is also attenuated by the total scattering cross section, the absorption is often better determined by calculation based on tabulated absorption cross sections for all atomic species involved. On the other hand side, the presence of the sample can also increase the background by the mechanism of multiple scattering. For example, neutrons scattered by the sample towards cryostat, sample holder and other spectrometer parts, can be scattered by these objects into the detector. This is one of the reason why one has to keep the total scattering probability in the sample low, in any case less then 10 %. The other reason, of course, is that multiple scattering within the sample can mess up the data interpretation directly too. These processes are very hard to predict, and one has to make all possible effort to minimize them by placing neutron absorbers everywhere it is possible. In view of the potential of background enhancement by sample scattering, it is the best bet to only consider beam attenuation trough the sample by true absorption as a background reducing process, as described above. Nevertheless, background always remains a most uncertain feature in any neutron scattering experiment, except when it is completely negligible compared to the signal.

5. Advanced trends: simulation based data evaluation.

It has been mentioned in the previous chapter, that the contribution to the neutron background of multiple scattering effects between sample and its environment cannot be safely determined. Unless we have to do with small corrections, the same applies to multiple scattering effects within the sample. It is relatively simple and straightforward to calculate the attenuation of the neutron beam due absorption in the sample taking exactly into account both before scattering and after scattering intensity losses. The effect of beam attenuation by scattering is, however, a completely different matter: the neutrons removed from the beam are still around, and might still reach the detector by changing directions again in another scattering process. Furthermore, since the multiply scattered neutrons can have an extended length, zig-zag trajectory within the sample, they are subject to a much more complex influence of the true absorption. The effects of finite spectrometer resolution (e.g. angular or energy) is another phenomenon, which can become difficult to evaluate, in particular in view of all kinds of inevitable inhomogeneities, such as the variation of the spectrometer response across the incoming beam cross section.

Early signs of considerable future progress can, however, be observed in recent years for the treatment of these complex issues. The emerging new approaches are based on computer based Monte-Carlo simulation of the entire neutron trajectories from entering the spectrometer to detection, including multiple scattering events in the sample and eventually in its immediate environment including back and forth scattering between sample and surrounding. (Although, it is very difficult to collect enough information on the scattering processes in objects around the sample, such as cryostats etc.) Modeling all these events is based on the basic physics of neutron propagation we have considered in detail in the previous chapters. In particular:

a) neutrons can be treated as point-like particles following classical trajectories between events of interaction with matter, either the sample or parts of the instrument,
b) refraction and total or partial reflection effects also can be described by classical trajectories of point-like neutrons, as determined by geometrical optical considerations, if one takes additionally into account the absorption attenuation of the beam inside matter,

c) scattering events, which can be accounted for by the usual quantum mechanical scattering cross section formalism, are localized within about a μm, i.e. they happen in point-like regions in comparison of the size of the samples, beam cross section and various other spectrometer parts,
d) between these extremely localized scattering events the neutron propagation within the sample again corresponds to classical trajectories connecting well defined points, with added attenuation due to true nuclear absorption.

Using a detailed and geometrically exact computer model of the spectrometer allows us to best reproduce and correct for most spurious effects, first of all multiple scattering in the sample. Simulating the background, in particular the crucial influence of the presence of the sample on the background will remain elusive for quite some time to come. On the other hand, this kind of "virtual spectrometer" approach also offers the best, most reliable way to determine the influence of the instrumental response on the data. Instead of ambiguous deconvolution procedures (where they apply) we will routinely reproduce the actually measured spectra for all kinds of model cross sections, and identify those models that are compatible with the observed data as they come from the instrument. Computers do not think, and computational models do not replace clear physical and analytical understanding of what is going. But they are badly needed in a complex environment, such as a neutron scattering experiment, to evaluate in a quantitative fashion all consequences of our understanding of the processes. Nearly every time I went through such a process, the computer surprised me with some quantitative detail I did not foresee, but often I could or should have foreseen. This experience is likely to have a quite general validity.

[1] F. Mezei, Zeeman energy, interference and Neutron Spin Echo: A minimal theory, Physica **B151** (1988) 74
[2] F. Mezei, Coherent approach to neutron beam polarization, in: imaging Processes and Coherence in Physics, M. Schlenker et al., editors (Spinger Verlag, Heidelberg, 1980) 114