# MIEZE Spectrometer RESEDA 

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## 1 Introduction: The neutron

The neutron is interesting tool for the investigation of matter in a non-destructive way due to its large penetration depth which stems from its lack of electric charge. The energy $(E)$ of a neutron corresponds to a temperature $(T)$ and is related to its velocity $(v)$ via:

$$
E_{n}=\frac{3}{2} k_{B} T=\frac{1}{2} m_{n} v^{2}
$$

where $k_{B}$ is the Boltzmann constant and $m_{n}$ the neutron mass. Following Einstein and de Broglie, the corresponding frequency and wavelength are given by:

$$
\nu=\frac{E_{n}}{h} \quad \lambda=\frac{h}{m_{n} v}
$$

with Planck's constant $h$.
The energy of the neutrons available for measurements, is determined by the production and moderation processes. Reactor neutrons are produced by nuclear fission as fast neutrons ( $E_{n}$ $>1 \mathrm{MeV}$ ) which are then moderated by elastic collisions with atoms belonging to a thermalized medium in order to produce neutrons with appropriate energies In neutron scattering, mainly it cold $\left(0.1 \leq E_{n} \leq 10 \mathrm{meV}, 1 \leq T \leq 120 \mathrm{~K}, 3 \leq \lambda \leq 30 \AA\right)$, thermal $\left(5 \leq E_{n} \leq 100 \mathrm{meV}\right.$, $60 \leq T \leq 1000 \mathrm{~K}, 1 \leq \lambda \leq 4 \mathrm{~A})$ and hot $\left(100 \leq E_{n} \leq 500 \mathrm{meV}, 1000 \leq T \leq 6000 \mathrm{~K}, 0.4 \leq \lambda \leq 1\right.$ A) neutrons are used.

This broad range of energies/wavelengths allows the investigation of a broad spectrum of different phenomena. At the MLZ 18 instruments use cold, 6 use thermal and 2 instruments use hot neutrons.
So-called elastic neutron scattering (i.e. involving no energy transfer between the neutron and the sample) provides the possibility to observe the (magnetic) structure of the studied samples. Inelastic neutron scattering (with non-zero energy transfer) and quasi-elastic scattering (with a small energy transfer compared to the incoming neutron energy) experiments, on the other hand, provide information on the dynamics of the sample (e.g. diffusion, phonon dynamics etc.) by making use momentum and energy conservatiopn:

$$
\begin{align*}
\vec{Q} & =\frac{2 \pi m_{n}}{h}\left(\overrightarrow{v_{i}}-\overrightarrow{v_{f}}\right) \\
\hbar \omega & =\frac{m_{n}}{2}\left(v_{1}^{i}-v_{f}^{2}\right) \tag{1}
\end{align*}
$$

where $v_{i}\left(v_{f}\right)$ is the initial (final) velocity of neutron.
The probability for a neutron to be scattered with a momentum transfer $\vec{Q}$ and energy transfer $\hbar \omega$ is contained in the so-called scattering function $\mathcal{S}(\vec{Q}, \omega)$.

Neutrons are spin $1 / 2$ particles with a magnetic moment of $\mu_{n}=-0.965 \cdot 10^{-26} \mathrm{~J} . \mathrm{T}^{-1}$, which makes it possible to polarize a neutron beam and investigate magnetic effects/interactions in the sample (e.g. ferromagnetism, vortex lattices, skyrmions...). The polarization is commonly defined as

$$
\begin{equation*}
\vec{P}=\frac{n_{+}-n_{-}}{n_{+}+n_{-}} \cdot \vec{u} \tag{2}
\end{equation*}
$$

where $n_{ \pm}$are the number of neutrons with spin parallel $(+)$and antiparallel (-) to the quantization axis $\vec{u}$.

## 2 Neutron Spin Echo

In Neutron Spin Echo (NSE), the spin of the neutron is used to analyze the energy transfer in quasi-elastic scattering experiments. NSE provides a high energy resolution, typically in the $\mu \mathrm{eV}$ to neV range when using cold neutrons. As opposed to other types of spectrometers, this resolution does NOT depend on the wavelength spread in the incident beam, thus the usable neutron flux can be increased by using a neutron beam consisting of a wavelength band, instead of a monochromatic (single wavelength) neutron beam.

### 2.1 Larmor precession



Figure 1: a) Illustration of the Larmor precession phenomenon. The polarization $\vec{P}$ rotates around the static magnetic field $\vec{B}_{z}$ as a function of time, while both amplitudes $P_{\|}$and $P_{\perp}$ are conserved. b) By slowly changing the orientation of $\vec{B}_{z}$, it is possible to adiabatically re-orient the rotation axis of $\vec{P}(t)$ as long as $\partial \theta_{B} / \partial y \ll \omega_{L}$.

The NSE technique is based on the Larmor precession of polarized neutrons. An external magnetic field $(\vec{B})$ will exert a torque on a particle with a magnetic moment. The particle will precess around the magnetic field axis with a precession (Lamor) frequency: $\omega_{L}=\gamma B$, where $\gamma$ is the gyromagnetic ratio of the particle. The neutron gyromagnetic ratio is $\gamma_{n}=2 \mu_{n} / h=2.916$ $\mathrm{kHz} . \mathrm{G}^{-1}$. The Lamor frequency does not depend on the angle between $\overrightarrow{\mu_{n}}$ and $\vec{B}$, as illustrated in Fig. 1 a). Formally, we get for the angular momentum of a single neutron in a magnetic field:

$$
\vec{J}(t)=\left(\begin{array}{c}
J_{\perp}(0) \cdot \cos \left(\omega_{L} t+\varphi_{0}\right) \\
J_{\perp}(0) \cdot \sin \left(\omega_{L} t+\varphi_{0}\right) \\
J_{\|}(0)
\end{array}\right)
$$

where $\varphi_{0}$ is an arbitrary phase at $t=0$. Setting $\varphi_{0}=0$, we easily obtain the rotation angle (or phase) after a time $t$ in a field of amplitude $B$ :

$$
\begin{equation*}
\varphi(v)=\omega_{L} \cdot t=\gamma_{n} B \cdot \frac{L}{v} \tag{3}
\end{equation*}
$$

where $L$ is the field region's thickness and $v$ the neutron velocity. From Eq. (3), we see that the Larmor phase is a direct measure for the neutron velocity.

For a beam of polarized neutrons, this expression can be generalized to describe the polarization of the neutron beam $(\mathrm{P}(\mathrm{t}))$.
The velocity of neutrons used at RESEDA can be obtained from their thermal energy and the neutron mass $m_{n}$ :

$$
v\left[\frac{m}{s}\right]=\sqrt{\frac{2 E}{m_{n}}} \simeq 437.1 \cdot \sqrt{E[\mathrm{meV}]}
$$

leading to

$$
\varphi(v)[\mathrm{rad}] \simeq 41.92 \cdot \frac{B[\mathrm{G}] \cdot L[\mathrm{~m}]}{\sqrt{E[\mathrm{meV}]}}
$$

It is worth noting that non-desired magnetic fields exist in the environment, e.g. the Earth magnetic field $B_{\text {Earth }} \simeq 0.5 \mathrm{G}$. The fields used in spin echo experiments are usually several orders of magnitude stronger than his value.

### 2.2 The Echo signal

NSE uses Larmor precession in two magnetic field regions, one located before $\left(B_{1}\right)$ and one after $\left(B_{2}\right)$ the sample position (see Fig. 2).


Figure 2: Basic principle of a NSE instrument. Arrows indicate neutron spins which precess with the Larmor frequency while travelling through the coils. The area around the sample is a zero magnetic field region.

An initially polarized neutron beam passes through the magnetic fields before and after being scattered by the sample. To maximize the amplitude of the rotative spin component, neutrons spins are initially aligned perpendicular to the magnetic fields (in Fig. $2, \vec{B}_{1,2}$ point in the $z$ direction and the initial beam polarization is directed along $x$ ). Thus, a neutron's spin first precesses inside the magnetic field $\vec{B}_{1}$ with the Larmor frequency $\omega_{L}=\gamma_{n} B_{1}$. The total precession phase of a neutron with velocity $v$ after having passed the first field region of length $L_{1}$ can be written as:

$$
\begin{equation*}
\varphi_{1}(v)=\gamma_{n} \frac{B_{1} L_{1}}{v} \tag{4}
\end{equation*}
$$

If the polarization is analyzed (i.e. projected) in the x -direction, it is calculated, in the general case of a non-monochromatic beam, from the average over all neutrons as follows:


Figure 3: Polarization $P_{x}$ as a function of the flight distance $L$ of neutrons through the apparatus. Numbers correspond to the positions marked on Fig. 2.

$$
\begin{equation*}
P_{x}=\langle\cos \varphi(v)\rangle=\int d v f(v) \cos \left(\gamma_{n} \frac{B_{1} L_{1}}{v}\right) \tag{5}
\end{equation*}
$$

where $f(v)$ is the distribution of neutron velocities. In general $f(v)$ is shaped like a triangular pulse around a center wavelength $\lambda_{0}$. It can be seen from figure (Fig. 3) that as the neutrons travel throught he spectrometer the polarization first decreases as a function of length, and then increases again, due to the reversal in magnetic field direction.
As equation (4) shows, $\varphi$ can either be varied by scanning $B_{1}$ or $L_{1}$. In a real NSE spectrometer, it is usually more practical to scan the magnetic field amplitude while the length of the precession regions is kept constant.

After passing the second magnetic field $\vec{B}_{2}$, located downstream of the sample with a magnetic field direction antiparallel to $\vec{B}_{1}$, the final spin phase is:

$$
\begin{equation*}
\varphi(v)=\gamma_{n}\left(\frac{B_{1} L_{1}}{v}-\frac{B_{2} L_{2}}{v}\right) \tag{6}
\end{equation*}
$$

assuming an elastic scattering by the sample (no velocity change before and after scattering). As shown in Fig. 2, the second field leads to a back precession of the neutron spin, denoted by the "-" sign in equation (6). Varying one of the magnetic field integrals ( $B_{1} L_{1}$ or $B_{2} L_{2}$ ) and measuring the polarization for the different field integral values renders the so-called spin echo group. Figure 4 shows such a spin echo group measured at RESEDA. Here, the value of the magnetic field $B_{2}$ is varied, whereas $B_{1}, L_{1}$ and $L_{2}$ are fixed. The center point, where $B_{1} L_{1}=B_{2} L_{2}$, is called the spin echo point. In this situation, the total spin phase of each neutron vanishes $(\varphi(v)=0)$. According to Eq. (5), this naturally leads to $P_{x}=1$. At both sides of this point the polarization decreases.

### 2.3 Inelastic and quasi-elastic scattering

In the case of inelastic scattering, the kinetic energy and, consequently, the velocity of each neutron changes. Eq. (6) then reads:

## spin echo point



Figure 4: Spin echo group measured at RESEDA. The spin echo point corresponds to the situation where the magnetic field integrals and, consequently, the number of precessions in both solenoids are equal (see Eq. (6)). This is where the polarization reaches its maximum value of 1 .

$$
\begin{equation*}
\varphi(v)=\gamma_{n}\left(\frac{B_{1} L_{1}}{v_{1}}-\frac{B_{2} L_{2}}{v_{2}}\right) \tag{7}
\end{equation*}
$$

where $v_{1,2}$ are the neutron velocities before and after scattering, respectively. For quasi-elastic scattering processes, the energy transfer $\hbar \omega$ is small compared to the energy of incident neutrons. In addition, if we assume an energy transfer distribution which is symmetric around $\omega=0$, the average neutron velocity after scattering $\bar{v}_{2}$ equals the average neutron velocity before scattering $\bar{v}_{1}$. Under these circumstances Eq. (7) is still valid and the spin echo point can still be found for $B_{1} L_{1}=B_{2} L_{2}=B L$.
For a small velocity change $\delta v=\left(v_{1}-v_{2}\right) \ll v_{1}$, Eq. (7) can be re-written as:

$$
\begin{equation*}
\varphi(v)=\gamma_{n} B L\left(\frac{1}{v_{1}}-\frac{1}{v_{2}}\right)=\gamma_{n} B L\left(\frac{1}{v_{1}}-\frac{1}{v_{1}-\delta v}\right) \sim-\gamma_{n} B L \frac{\delta v}{v_{1}^{2}} \tag{8}
\end{equation*}
$$

Using the law of energy conservation (Eq. (1)), we get:

$$
\begin{equation*}
\hbar \omega=\frac{1}{2} m_{n} v_{1}^{2}-\frac{1}{2} m_{n}\left(v_{1}-\delta v\right)^{2} \sim m_{n} v_{1} \delta v \tag{9}
\end{equation*}
$$

At the spin echo point, combining Eqs. (9) and (8) thus gives:

$$
\begin{equation*}
|\varphi|=\frac{\gamma_{n} \hbar B L}{m_{n} v_{1}^{3}} \cdot \omega=\frac{\gamma_{n} B L m_{n}^{2} \lambda^{3}}{2 \pi h} \cdot \omega \equiv \tau \cdot \omega \tag{10}
\end{equation*}
$$

where the spin echo (or Fourier) time $\tau$ is the proportionality constant between the energy transfer and the absolute value of each neutron's spin phase. For a given $\tau, \varphi$ is a sensitive measure of $\hbar \omega$. Increasing the Fourier time allows resolving tiny energy transfers, i.e. increasing the energy resolution.

The final NSE signal polarization $P_{N S E}$ is given by (using Eqs. (5) and (8), ):

$$
\begin{equation*}
P_{N S E}=\frac{\int d \omega \mathcal{S}(\vec{Q}, \omega) \cos (\omega \tau)}{\int d \omega \mathcal{S}(\vec{Q}, \omega)}=\frac{\mathcal{S}(\vec{Q}, \tau)}{\mathcal{S}(\vec{Q}, \tau=0)} \tag{11}
\end{equation*}
$$

Eq. (11) describes the basic principle of NSE data evaluation. In this expression, the denominator is a normalization which is applied to account for possible changes of the beam polarization by the scattering process itself. In total, $P_{N S E}$ is proportional to $\mathcal{S}(\vec{Q}, \tau)$, the cosine Fourier transformation of the scattering function $\mathcal{S}(\vec{Q}, \omega)$. In order to obtain full information on the characteristics of $\mathcal{S}(\vec{Q}, \omega), P_{N S E}$ has to be measured for several $\tau$-values. In general, quasi-elastic processes lead to a decay of the beam polarization $P_{N S E}(\tau)$. We also note that, in practice, the determination of $P_{N S E}$ at the spin echo point is accomplished by slight variations of the field integrals $B_{1} L_{1}$ or $B_{2} L_{2}$ and fitting of an appropriate theoretical model to the measured values.

### 2.4 Example

Imagine moving particles inside a liquid sample. Each scattered neutron may lose or gain energy through its interaction with such a particle. This will result in different velocities for the incoming and outgoing neutrons. The strength of the NSE technique is to detect the small energy changes of each individual neutron. To illustrate this fact, we assume a monochromatic neutron beam, with neutron velocity $v_{1}$ and an incoming spin angle $\phi_{i}$. Let us further assume, that the particles inside our liquid scatter $50 \%$ of neutrons such that they experience a velocity loss of $\Delta v\left(v_{2,1}=\right.$ $\left.v_{1}-\Delta v\right)$ while the other $50 \%$ experience a velocity gain of $\Delta v\left(v_{2,2}=v_{1}+\Delta v\right)$. This leads to a polarization smaller than 1 . Neutrons of the first sort $\left(v_{2,1}=v_{1}-\Delta v\right)$ will be slowed down, and therefore spend a longer time in the applied magnetic field, and reach a higher number of precessions, leading to a final angle $\phi_{f, 1}>\phi_{i}$. While neutrons of the second sort ( $v_{2,1}=v_{1}+$ $\Delta v)$ will be too fast to reach the initial spin angle $\phi_{i}\left(\phi_{f, 2}<\phi_{i}\right)$.

To be more realistic, we can assume for our sample a scattering function, $\mathcal{S}(\vec{Q}, \omega)$, with a Lorentzian lineshape, i.e.

$$
\begin{equation*}
\mathcal{S}(\vec{Q}, \omega)=\frac{\Gamma}{\Gamma^{2}+\omega^{2}} \tag{12}
\end{equation*}
$$

Using Eq. (11) gives for the measured signal an exponential decay of the polarization as a function of $\tau$ :

$$
\begin{equation*}
P_{N S E}=\langle\cos \varphi\rangle=e^{-\Gamma \tau} \equiv e^{-\tau / \tau_{0}} \tag{13}
\end{equation*}
$$

From the $\tau$ dependence of $P_{N S E}$, it is then possible to determine the linewidth parameter $\Gamma$ of the scattering process and, conversely, its lifetime $\tau_{0}=\Gamma^{-1}$.

## 3 Neutron Resonance Spin Echo Methods

### 3.1 Neutron Resonance Spin Echo (NRSE)

### 3.1.1 The NRSE Coils



Figure 5: a) A radiofrequency spin flipper as used on RESEDA. b) Decomposition of an oscillating field into two counter-rotating components.

In Neutron Resonance Spin Echo (NRSE) Spectroscopy, the long solenoids used in "classical" NSE are replaced by a pair of so-called radiofrequency spin flipper coils:

- The core of the RF flipper, consists of the RF coil which creates an oscillating field in the $(z, x)$-plane which can be described as the superposition of two counter-rotating field components:

$$
B_{o s c}(t)=\frac{B_{r f}}{2} \cdot \exp \left(i \omega_{r f} t\right)+\frac{B_{r f}}{2} \cdot \exp \left(-i \omega_{r f} t\right)
$$

- The $B_{0}$ coil(s) are arranged in Helmholtz configuration around the RF coil and create a static magnetic field $B_{0}$ in the $y$-direction (along the neutron flight path)
- To prevent the field of the $B_{0}$ coils to leak too far outside the coils and cause inhomogeneities the cut-off coils are installed around the $B_{0}$ and RF coils. The cut-off coils are mounted in Helmoltz configuration as well, however their field points in the opposite direction as the one of the $B_{0}$ coil.

The neutrons entering the RF spin flipper coil start to rotate around $\vec{B}_{y}$ with the Larmor frequency $\omega_{L}=\gamma_{n} B_{y}$. In addition to this motion, as soon as they enter the RF flipper, the neutron spins start to precess around the $\vec{B}_{r f}$ component which rotates in the same direction they do. The frequency of the oscillating field is adjusted to be in resonance with $\omega_{L}$, i.e.:

$$
\omega_{r f} \equiv \omega_{L}
$$

thus justifying the name NRSE. This additional motion is called Rabi oscillation, in complete analogy with Nuclear Magnetic Resonance (NMR) (see Fig. 6 a)). After a flight time $t$ and a precession angle of $\omega_{R} t=\pi$, the neutrons leave the magnetic field of the coil with their magnetic
moment effectively flipped with respect to the initial direction of $\vec{B}_{r f}$. This is the so-called resonant $\pi$-flip. After traversing a flight path of length $L$, the neutrons enter a second NRSE coil which produces identical static and oscillating fields which lead to a second $\pi$-flip for the spins.


Figure 6: a) The spin magnetic moment $\vec{\mu}_{n}$ is initially in the $(z, x)$-plane. In the rotating coordinate system, the spin stays still in the absence of a rf field. If we add a field $\vec{B}_{r f}$, rotating with a frequency which matches the Larmor frequency of the static field, $\vec{\mu}_{n}$ precesses around $\vec{B}_{r f}$ with a Rabi frequency $\omega_{R}$. After a half-twist, it is back in the $(z, x)$-plane, mirrored with respect to $\vec{B}_{r f}$. b) If $\vec{\mu}_{n}$ is initially parallel to $\vec{B}_{y}$, the effect of the spin flipper is simply to flip it with respect to $\vec{B}_{y}$.

Considering the case where the initial spin direction is parallel to $\vec{B}_{y}$ (Fig. $6 \mathbf{b}$ )): in this case, the opening angle of the Larmor precession around the static field is 0 . However, the precession around $\vec{B}_{r f}$ still takes place as discussed above and the angle between $\vec{\mu}_{n}$ and $\vec{B}_{r f}$ is now exactly $\pi / 2$. The neutron spin performs a $\pi$-rotation around the axis given by the radiofrequency field, which corresponds to a spin flip from up to down (or vice versa) with respect to $\vec{B}_{y}$.

### 3.1.2 NRSE signal and setup

Since RESEDA is operated as a longitudinal NRSE, the following considerations will be done in this configuration. Combining two resonant spin flippers at a distance $L$ from each other, as is done in NRSE results in the same final neutron spin phase as for one single NSE solenoid (length $L$ ), when the magnitude of the precession field $\vec{B}_{z}$ is doubled. The phase of the neutron spin after traversing the first coil (A) is given by:

$$
\begin{equation*}
\varphi_{A}(v)=2 \omega_{r f} \cdot t_{A}+\omega_{r f} \cdot \frac{d}{v}-\Phi_{0} \tag{14}
\end{equation*}
$$

where $\Phi_{0}$ is the starting phase (which can be set to 0 here), $t_{A}$ is the time at which the neutrons enter the first flipper and $d$ its thickness. After flying for a distance $L$, the neutrons enter the second NRSE coil (B) and the final phase is:

$$
\begin{align*}
\varphi(v) & =\varphi_{B}(v) \\
& =2 \omega_{r f} \cdot\left(t_{A}+\frac{L}{v}\right)+\omega_{r f} \cdot \frac{d}{v}-\varphi_{A}(v) \\
& =2 \omega_{r f} \cdot \frac{L}{v} \\
& =2 \gamma_{n} B_{z} \cdot \frac{L}{v} \tag{15}
\end{align*}
$$

This gives us an additional factor two as compared to the "classical" NSE technique, as mentioned above.
An advantage of NRSE is the compactness of the precession devices which reduces the effect of field inhomogeneities. A complete NRSE setup consists, consequently, of four NRSE coils, two in each spectrometer arm, with small guide fields in between (Fig. 7). Varying the field integral to recover the spin echo signal can be done either by changing the distance between one pair of NRSE flipper or by scanning the RF field frequency in one arm.


Figure 7: Schematic of a complete LNRSE setup consisting of four NRSE spin flippers, two in each spectrometer arm, up- and downstream of the sample. The rotating field $\vec{B}_{r f}$ is in resonance with the static field $\vec{B}_{y}$. As in NSE, a NRSE setup is completed by a polarizer in front of the apparatus and a spin analyzer behind the second precession region.

### 3.2 MIEZE

Classical NSE and NRSE have one big disadvantage. It is very difficult to measure under depolarizing conditions (magnetic samples, applied magnetic field) or strongly incoherent scattering samples (materials containing large amounts of Hydrogen). The Modulation of IntEnsity with Zero Effort (MIEZE) method, developed by R. Gähler, R. Golub and T. Keller, resolves this shortcoming by transforming the rotational phase of the spin into an intensity modulated signal using an analyzer.
This is possible due to the fact that the complete spin phase manipulation, which is done by the resonant flippers, happens before the sample. A schematic of a LMIEZE setup is shown in figure 8.

The starting point, is the same as for NSE or NRSE. Analogously to NRSE we can write the spin phase after the first RF flipper as:

$$
\begin{equation*}
\varphi_{1}(v)=2 \omega_{r f} \cdot t_{1}, \tag{16}
\end{equation*}
$$

with $\mathrm{t}_{A}$ the time of arrival at the first resonant flipper. The neutron arrives at the second flipper at a time $t_{B}=t_{A}+L_{1} / v$, where $\mathrm{L}_{1}$ is the distance between the two flippers and $v$ is the neutron velocity. The spin phase after the second flipper can then be written as:


Figure 8: Schematic of a complete LMIEZE setup. The primary spectrometer arm is the same as for LNRSE, with two big exceptions: 1) the spin analyzer is moved in front of the sample, to convert the spin phase into an intensity modulated signal before the sample, to avoid depolarization of the beam by sample and/or sample environment; 2) The two resonant spin flippers are operated at different frequencies.

$$
\begin{equation*}
\varphi_{2}(v)=2 \omega_{r f 1} \cdot t_{2}-\varphi(v)_{1}=2 \cdot\left(\omega_{r f 2}-\omega_{r f 1}\right) \cdot t_{1}+2 \omega_{r f 2} \frac{L_{1}}{v} \tag{17}
\end{equation*}
$$

From here on, the neutron passes through the analyzer and hits the detector after a distance $L_{2}$ behind the second flipper at a time $t_{D}=t_{1}+\frac{L_{1}+L_{2}}{v}$. We will use this to replace $t_{1}$ in equation 17:

$$
\begin{equation*}
\varphi_{B}(v)=2 \cdot\left(\omega_{r f 2}-\omega_{r f 1}\right) \cdot\left(t_{D}-\frac{L_{1}+L_{2}}{v}\right)+2 \omega_{r f 2} \frac{L_{1}}{v} \tag{18}
\end{equation*}
$$

From now on we will write: $\left(\omega_{r f 2}-\omega_{r f 1}\right)=\Delta \omega$; A polychromatic beam would lead to a loss of phase information, if any velocity dependent terms remained in equation 18. Therefore, to recover the full signal at the detector, we are placing the detector such that all velocity dependent terms are zero:

$$
\begin{equation*}
0=-2 \Delta \omega\left(L_{1}+L_{2}\right)+2 \omega_{r f 1} L_{1} \tag{19}
\end{equation*}
$$

From this we can recover the MIEZE condition:

$$
\begin{equation*}
\frac{\Delta \omega}{\omega_{r f 1}}=\frac{L_{1}}{L_{2}} \tag{20}
\end{equation*}
$$

The phase at the detector then becomes:

$$
\begin{equation*}
\varphi_{D}(v)=2 \cdot\left(\omega_{r f 2}-\omega_{r f 1}\right) \cdot t_{D} \tag{21}
\end{equation*}
$$

From this a sinusoidal intensity signal in time

$$
\begin{equation*}
I\left(t_{D}\right) / I_{0}=1 / 2\left(\cos \left(2\left(\omega_{B}-\omega_{A}\right) t_{D}\right)+1\right) \tag{22}
\end{equation*}
$$

is obtained, hence the name MIEZE (Modulation of IntEnsity by Zero Effort).
When we assume an energy transfer $\hbar \omega$ to a sample, placed at a distance $L_{\text {SD }}$ upstream of the detector, the arrival time at the detector is changed by:

$$
\begin{equation*}
\Delta t_{D}=L_{\mathrm{SD}}\left(\frac{1}{v}-\frac{1}{\sqrt{v^{2}+\frac{2 \hbar \omega}{m_{n}}}}\right) \tag{23}
\end{equation*}
$$

Within the spin echo approximation, i.e. $\hbar \omega \ll \frac{m_{n}}{2} v^{2}(\delta v \ll v)$, this can be simplified to

$$
\begin{equation*}
\Delta t_{D} \approx \frac{L_{\mathrm{SD}} \hbar \omega}{m_{n} v^{3}} \tag{24}
\end{equation*}
$$

The probability of the scattering process is given by the scattering function $S(Q, \omega)$, analogously to classical NSE. Integration over the whole energy spectrum shows, that the intensity is reduced according to

$$
\begin{equation*}
I\left(t_{D}\right) / I_{0}=\frac{1}{2} \int\left(\cos \left(2 \Delta \omega\left(t_{D}+\Delta t_{D}\right)\right)+1\right) S(Q, \omega) \mathrm{d} \omega \tag{25}
\end{equation*}
$$

The symmetry of $S(\omega)$ allows to rewrite the intensity modulation (25).

$$
\begin{align*}
I\left(t_{D}\right) / I_{0} & \left.=\frac{1}{2} \int\left(\cos \left(2 \Delta \omega t_{D}\right)+\cos \left(2 \Delta \omega \Delta t_{D}\right)\right)+1\right) S(\omega) \mathrm{d} \omega \\
& =I_{\text {instr }}+C \\
& \left.=I_{\text {instr }}+\frac{1}{2} \int\left(\cos \left(2 \Delta \omega \Delta t_{D}\right)\right)\right) S(\omega) \mathrm{d} \omega \tag{26}
\end{align*}
$$

The contrast $C$ can be identified with the cosine Fourier transform, just like the polarization in NSE, and simplifies to

$$
\begin{equation*}
C=\int \cos \left(\omega \tau_{M I E Z E}\right) S(q, \omega) \mathrm{d} \omega \tag{27}
\end{equation*}
$$

under the spin echo approximation. For this, the MIEZE time

$$
\begin{equation*}
\tau_{M I E Z E}=\frac{2 \Delta \omega L_{S D} \hbar}{m_{n} v^{3}}=\frac{m_{n}^{2}}{h^{2}} L_{S D} 2 \Delta f \lambda^{3} \tag{28}
\end{equation*}
$$

was introduced. Analogous to NSE, the wavelength goes in with cubic power, emphasizing the benefit of high wavelengths, where neutron flux can be traded for better energy resolution. The field integral $B L$ of conventional spin echo (equation 10) is replaced by the frequency difference in the resonant flipper coils $\Delta f$ and the sample detector distance $L_{S D}$.

## 4 RESEDA

### 4.1 The instrumental setup



Figure 9: a) A typical velocity selector with helical wings made of absorbing material. Adjusting its rotation speed allows to select neutrons wavelength. b) The V-cavity polarizer installed on RESEDA.

RESEDA is a Neutron Resonant Spin Echo spectrometer with two secondary arms, one to perform NRSE experiments and one for MIEZE experiments. The arms are enumerated as 0 (incoming beam), 1 (NRSE) and 2 (MIEZE). A schematic of RESEDA, with the most important components shown is depicted on Fig. 10. RESEDA is located at the end station of the curvedneutron super mirror guide NL5-S, and uses the upper third of this guide, a section measuring $36 \times 36 \mathrm{~mm}^{2}$. The guide is fed by cold moderated neutrons from the FRM II research reactor which possess a Maxwellian spectrum with a usable neutron flux between $3.5 \AA$ and $20 \AA$. In front of the coils and flippers, the beam preparation is accomplished, i.e. the neutron wavelength and the spin polarization are selected. A velocity selector (see Fig. 9, a)) makes it possible to smoothly vary the wavelength used for the experiment. The beam is spin polarized by a V-cavity polarizer ${ }^{1}$ (Fig. 9, b)) consisting of magnetic layers with different refractive indices for spin up and spin down states, respectively. The spin up neutrons are reflected out of the direct beam and absorbed in the polarizer walls, so that only one spin component is transmitted to the instrument. Thus, we obtain a polarized beam, which corresponds to $n_{-} \simeq 1$ and $n_{+} \simeq 0$ in Eq. (2).

After the polarizer the neutron spins are adiabatically rotated such that they point along the flight direction. Afterwards, the neutrons enter the first precession zone with the first two resonant spin flippers. What happens behind the first precession zone, depends on the measurement mode:

- for LNRSE measurements a $\pi$ flipper reverses the neutron polarization, the neutrons interact with the sample, pass the second precession zone. Afterwards, the spins are again adiabatically rotated, go through the analyzer and are detected in a ${ }^{3} \mathrm{He}$ detector.
- for LMIEZE the spins are adiabatically flipped after the first precession zone, after which the spin phase is transformed into an intensity modulated signal using an analyzer. After interacting with the sample and traversing a region of length $\mathrm{L}_{S} D$ they are detected by a ${ }^{1} 0 \mathrm{~B}$ CASCADE detector.

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Figure 10: Sketch of the NRSE/MIEZE spectrometer RESEDA.

### 4.2 Typical Applications

When operated as a MIEZE spectrometer, data may be recorded readily in depolarizing sample environments, such as very large magnetic fields. Likewise, depolarizing substances, such as ferromagnets or superconductors, strong incoherent scattering materials containing, for instance, hydrogen may be studied. RESEDA's LNRSE option represents a set-up akin a conventional NSE spectrometer, therefore being perfectly suited for typical problems from the field of soft matter and glass transitions. Summarized, the typical applications of RESEDA are:

- dynamics and diffusion of polymer melts
- dynamics of water in porous media
- diffusion processes in ionic liquids
- dynamics of soft biological systems
- freezing of glasses and spin glasses
- critical magnetic fluctuations
- melting of superconducting vortex lattices
- dynamics at quantum phase transitions
- dynamics of magnetized thin foils
- emergent excitations in quantum magnets


## 5 Preparatory exercises

1) Read these instructions carefully.
2) Calculate the energy and velocity of the neutrons which are accessible at RESEDA (cold source at $T=25 \mathrm{~K}$ ). Compute the time they need to pass the spectrometer arms given that the velocity selector to detector distance $\simeq 8 \mathrm{~m}$.
3) Look up the typical value of the Earth magnetic field. Calculate the precession frequency of neutron spins in this field. Conclude, with 2), on the phase they would accumulate while traveling from velocity selector to detector.
4) Assuming a gaussian wavelength distribution of $13 \%$ full width at half maximum (FWHM) as provided by the selector, calculate the envelope of the spin echo group.
5) Calculate the spin phase $\varphi_{1}(v)$ assuming a field integral $B L=100$ G.m and a neutron wavelength of $5 \AA$. Calculate the final spin phase $\varphi(v)$ if the neutron is scattered inelastically with an energy transfer $\hbar \omega=0.01 \mathrm{meV}$.
6) Using the same field integral and neutron wavelength as in 5), calculate the polarization in the case of quasielastic scattering with $\mathcal{S}(\vec{Q}, \omega)$ given by a Lorentzian function with a linewidth $\Gamma=0.01 \mathrm{meV}$.
7) Do you expect the Diffusion coefficients for $\mathrm{H}_{2} \mathrm{O}$ and $\mathrm{D}_{2} \mathrm{O}$ to be the same? why?/why not? Look up the Diffusion coefficients for water $\left(\mathrm{H}_{2} \mathrm{O}\right)$, heavy water $\left(\mathrm{D}_{2} \mathrm{O}\right)$ as well as 1 molar and 0.1 molar solutions of NaCl in water, at room temperature.

## 6 Experimental procedure

For this practical session you will be determining the translational diffusion constant of 0.1 and/or 1 molar solutions of NaCl in ultra pure water and compare it to the values reported in literature. For this $S(Q, \tau)$ needs to be measured for a minimum of 3 different Q values.

Firstly, you will have a guided tour on the instrument to see all the different components, especially the precession devices. You will then go to the chemical laboratory to prepare the solutions. Afterwards, you will mount the sample onto the sample stick.

The experiment will then involve the following steps:

- Sample alignment
- Stabilizing the temperature in a heater cell.
- Check of the instrumental setup (e.g. slits, cryostat, measurement angles, neutron wavelength, etc.)
- Measuring $S(Q, \tau)$
- Resolution using powdered carbon, in the same cuvette as the water sample.

The quasi-elastic data will be acquired by means of a scan overnight. The length and number of measurements will have to be adjusted to the available time. Thus, it may be necessary to use some measurements of the preceding groups. Then, you will analyze the data using the new MIEZEPY tool (Instructions on how to use the Software are in the Appendix). You will then fit the part of $S(Q, \tau)$ which corresponds to the translational diffusion of water, using an exponential decay and use the parameters to determine the diffusion coefficient.

Do not hesitate to ask questions!

## Suggested reading

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https://mlz-garching.de/instrumente-und-labore/spektroskopie/reseda.html

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[^0]:    ${ }^{1}$ http://www.swissneutronics.ch/products/polarizing-devices.html

