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A quantum-mechanical description of Rotating Field Spin Echo

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Abstract – Neutron Spin Echo (NSE) is a technique for quasi-elastic neutron scattering with very high energy resolution. The latter is achieved by comparing the Larmor precession angles of a polarized neutron beam in two well-known magnetic fields before and after scattering in a sample. This “spin encoding” or “Larmor labeling” can be implemented by different technical methods, the most established variants being conventional NSE and Neutron Resonance Spin Echo (NRSE). In this publication we discuss Rotating Field Spin Echo (RFSE), a technique for measurements in the low-resolution domain. The analogy to NRSE is demonstrated by deriving the working principle using the quantum-mechanical method of time-evolution operators, a technique that was developed for NRSE in previous publications. Furthermore, we give an estimation of the inherent upper frequency limit of RFSE based on our theory.

Introduction. – Neutron Spin Echo (NSE) [1] and Neutron Resonance Spin Echo (NRSE) [2] are spectroscopy methods in neutron science for investigating quasi- and inelastic scattering with high energy resolution down to the sub-\(\mu\)eV range. The high resolution is achieved by comparing the Larmor precession angles of the neutron spin in two well-known magnetic fields before and after scattering in a sample. For quasi-elastic scattering the decrease in polarization compared to the empty beam then provides information about the intermediate scattering function \(S(Q,t)\) characterizing the sample.

Spin echo instruments that achieve the above-mentioned “Larmor encoding/decoding” before and after the sample to detect changes in kinetic energy of the scattered neutrons can be implemented in different ways. Unlike conventional spin echo instruments, NRSE uses radio-frequency resonance flipper coils inside a zero-field region to produce the required Larmor precession. For more details about the technical implementation of NRSE we refer to existing literature, e.g. [3–6].

For technical reasons, usually oscillating fields are used in NRSE resonance flippers. The oscillation can be described as a composition of two counter-rotating field components, one of which induces the spin flip. Rotating Field Spin Echo (RFSE), however, is based on coils producing truly rotating fields. Those coils are mounted inside a magnetic shielding providing a field-free space between them, just like for NRSE. As we will see in the following, RFSE can be treated as a special case of NRSE but aims at a different range of energy resolution.

Theoretical description of RFSE. – The technique of spin echo with rotating magnetic fields has first been proposed and simulated in [7] and [8] using an idealized approach. Here we want to take a different approach and base our theoretical description of Rotating Field Spin Echo on the theory existing for Resonance Spin Echo. We start by sketching a classical picture to illustrate why they are equivalent and how the limiting factors of RFSE can be understood as an intrinsic detuning of its NRSE counterpart. Subsequently we will derive the quantum-mechanical description of RFSE using time-evolution operators.

Classical picture. The most simple way of implementing an RFSE coil is to cross two flat layers of windings and phase-shift the sinusoidal currents through them by \(\pi/2\)
of the field can be defined by the directions of the point in the plane, it does not require a zero-field region between the coils but can be proposed in [9] where the static field is pointing in beam direction.

Fig. 1: Magnetic-field configuration inside a flipper coil for NRSE (left) and RFSE (right).

with respect to each other. This setup produces a field

\[
B_{\text{rot}}(t) = \begin{pmatrix} 0 \\ B_y \sin(\omega t) \\ B_z \sin(\omega t + \frac{\pi}{2}) \end{pmatrix} \approx \mu_0 \frac{N}{D} \begin{pmatrix} 0 \\ I_y \sin(\omega t) \\ I_z \cos(\omega t) \end{pmatrix},
\]

where \( \mu_0 = 1.257 \cdot 10^{-6} \text{Vs/(Am)} \) is the magnetic field constant, \( N \) the number of turns per winding layer and \( D \) its length. Inside the coil \( B_{\text{rot}} \) is rotating in the \( y-z \) plane, i.e. perpendicularly to the beam path which shall point in the \( x \)-direction (fig. 1). The sense of rotation of the field can be defined by the directions of the currents \( I_{y,z} \).

In NRSE flipper coils a rotating field \( B_1(t) \) with angular frequency \( \omega \) is superposed with a highly homogeneous static field \( B_0 \). For zero-field NRSE\(^1\) the technical implementation of a flipper coil requires \( B_0 \) to be perpendicular to the beam path with \( B_1 \) rotating in the orthogonal plane, which means in our case the \( z \)-direction and the \( x-y \) plane, respectively (cf. fig. 1). Two conditions must apply to the fields. The first is

\[
\omega_L = \gamma B_0 = \omega,
\]

which means that \( B_0 \) and \( B_1(t) \) are resonantly tuned, \( \gamma = 1.8325 \cdot 10^8 \text{s}^{-1} \text{T}^{-1} \) is the gyromagnetic ratio of the neutron. The second one concerns the amplitude of the rotating field,

\[
\gamma B_1 \cdot \frac{D}{v} = \gamma B_1 \cdot T = \pi,
\]

and ensures that the neutron spin performs a \( \pi \)-flip around the RF field vector. \( D \) is the length of the field region, \( v \) the neutron velocity, and \( T = D/v \) the flight time through the field region. The neutron beam passing such a coil shall be polarized with respect to \( y \), i.e. \( P_0 = (0, P_y, 0) \). The axis of quantization for spin-up and spin-down is defined by the static field, in this case \( z \).

Comparing the field configuration of NRSE and RFSE (see fig. 1) we recognize that in the latter case the static field is missing and the plane of rotation has been chosen to be the \( y-z \) plane, in contrast to the directions commonly used in zero-field spin echo. However, as we will show in the next section both techniques can be described equivalently using the same mathematical tools.

A common technique to deal with magnetic resonance problems is transferring them into a coordinate system rotating with \( \omega \) where \( B_1 \) is static [10]. This yields an effective field of \( B_0 - \omega/\gamma \) along the \( z \)-axis in the rotating system. In the case of resonance, \( B_0 \) will be cancelled. If the amplitude of \( B_1 \) is tuned such that the polarisation vector performs a \( \pi \)-flip around it, the neutron spins will stay within the \( x-y \) plane after passage of the coil.

For RFSE, however, the transition to the rotating coordinate system will cause an additional field \(-\omega/\gamma \) in the \( x \)-direction, since no resonant static field exists in the laboratory system. The resulting field vector \( B_{eff} = B_{rot} - \omega/\gamma \cdot e_x \) will be pointing out of the \( y-z \) plane, depending on the frequency. Hence, the \( \pi \)-flip will likewise turn \( P_0 \) out of that plane by an angle \( \alpha \). This causes a loss of polarisation, since the amount left for the spin echo measurement is the in-plane component \( P_0 \cos \alpha \) (see, e.g., [3]). This loss is inherent to the technique and the reason why it is limited to lower frequencies, i.e. Fourier times or resolutions, respectively.

Quantum-mechanical description. To quantify the above-sketched classical view of what happens in a rotating field coil, we will switch to the quantum-mechanical picture in which we describe the progression of the spin-up and spin-down states — i.e. the phase they accumulate when passing the instrument — by time-evolution operators. This technique has been developed in [11] and was also used in [5,6].

The fact that in spin-space the \( z \)-axis is a preferential direction is due to the choice of the basis \( |\uparrow \rangle, |\downarrow \rangle \), resulting in the Pauli matrix \( \sigma_z \) being diagonal. But one could perform a basis transformation resulting in the preference of a different quantization axis without changing physics. Knowing this, we will now continue to solve the problem for the rotating field turning not in the \( y-z \), but the \( x-y \) plane (as for NRSE, cf. fig. 1). This allows us to directly apply the existing theory.

The idea is to obtain the quantum-mechanical state after passing the rotating field coils by applying a time-evolution operator describing the magnetic field region to an initial state:

\[
\psi(t) = U(t_1, t_0) \psi_0,
\]

with

\[
\psi_0 = \begin{pmatrix} \psi_0^+ \\ \psi_0^- \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}.
\]

The operator describing the passage through a rotating field coil between times \( t_0 \) (entry coil) and \( t_1 \) (exit coil) can be written as

\[
\text{see eq. (6) on next page}
\]

\[\text{42001-p2} \]
where we introduced the parameters

\[
\delta = \frac{\omega}{\omega_R} \equiv \left[ 1 + \left( \frac{\gamma B_{\text{rot}}}{\omega} \right)^2 \right]^{-1/2},
\]

\[
\varepsilon = \frac{\gamma B_{\text{rot}}}{\omega_R} \equiv \left[ 1 + \left( \frac{\omega}{\gamma B_{\text{rot}}} \right)^2 \right]^{-1/2};
\]

\( \varphi_1 = \omega t_0 \) is the RF phase when the neutron reaches the first coil.

Apparently spin-up and spin-down mix when passing the coil. The spin-flip probability will be more or less significantly smaller than 1, depending on \( \omega_R, \delta \), and \( \varepsilon \).

For very small frequencies (\( \omega \to 0 \)) the parameters tend to \( \delta \to 0 \) and \( \varepsilon \to 1 \). If we further tune \( B_{\text{rot}} \) to meet eq. (3), i.e. \( \gamma B_{\text{rot}} D/v = \gamma B_{\text{rot}} T = \pi \) (and hence \( \omega_R T \to \pi \)), the diagonal elements of the operator in eq. (6) vanish, resulting in a “perfect” \( \pi \)-flip of the polarization vector, which means a complete population inversion of the two spin states. This ideal RFSE scenario corresponds to the NRSE case described above (cf. eqs. (2) and (3)), and we would obtain

\[
\psi_1 = \frac{-i}{\sqrt{2}} e^{-\frac{i}{2} \mathcal{H}_0 (t_1-t_0)} \left( e^{-i(\varphi_1 + \frac{\varphi_2}{2})} e^{i(\varphi_1 + \frac{\varphi_2}{2})} \right). \tag{10}
\]

The accumulated spin phase after the first coil would then be \( 2\varphi_1 + \omega D/v \). For finite frequencies the parameters \( \delta \) and \( \varepsilon \) reflect the (relative) “detuning” and the (relative) “tuning” of the coil, respectively, i.e. the deviation from the ideal NRSE case.

The second coil shall be separated from the first by a distance \( L_1 \). Like for an NRSE instrument, we have zero field between coil 1 and 2, and therefore, no additional phase is accumulated. The state entering the second coil at time \( t_1 = t_0 + D/v \) shall be denoted by \( \psi_{12} \). After passing it, we can calculate the spin state \( \psi_{2} \) at time \( t_2 = t_1 + D/v = t_1 + L_1/v \) (i.e. after one arm of an RFSE instrument):

\[
\text{see eq. (11) on next page}
\]

In case of very small frequencies (\( \delta \to 0, \varepsilon \to 1, \omega_R T \to \pi \)) we obtain

\[
\psi_2 = e^{-\frac{i}{2} \mathcal{H}_0 (t_2-t_0)} \frac{1}{\sqrt{2}} \left( \begin{array}{c} -e^{-i(\varphi_2-\varphi_1)} \\ e^{i(\varphi_2-\varphi_1)} \end{array} \right) \tag{12}
\]

where we substituted \( \varphi_2 = \varphi_1 + L_1/v \), since the rotating fields are phase-locked and consequently have a constant phase relation.

The accumulated spin phase after the second coil, \( 2(\varphi_2 - \varphi_1) = 2\omega L_1/v \), does not depend on the absolute time, but only on the flight time \( L_1/v \) between the coils. This is the expected NRSE result. If no sample is present it is cancelled in the second arm of the spectrometer, thus —under ideal conditions— restoring the initial polarization (“spin echo”).

**Derivation of the field tuning condition for RFSE.** For real rotating field coils a polarization loss will occur due to the lack of a static field (which is in resonance with the rotating field frequency) and the finite thickness of the coil.
\[ \psi_2 = U(t_2, t_1) \psi_{12} = e^{-i \frac{1}{2} \mathcal{H}_0 (t_2 - t_0)} \]

\[ \times \left( -e^{-i(\varphi_2 - \varphi_1)} e^{2} \sin^2 \left( \frac{\varphi_0}{2} T \right) + e^{-i \omega T} \left[ \cos \left( \frac{\varphi_0}{2} T \right) + i \delta \sin \left( \frac{\varphi_0}{2} T \right) \right] \psi_0^+ - i \varepsilon \sin \left( \frac{\varphi_0}{2} T \right) \left[ e^{-i \varphi_2} \left[ \cos \left( \frac{\varphi_0}{2} T \right) - i \delta \sin \left( \frac{\varphi_0}{2} T \right) \right] + e^{-i (\omega T + \varphi_1)} \left[ \cos \left( \frac{\varphi_0}{2} T \right) + i \delta \sin \left( \frac{\varphi_0}{2} T \right) \right] \psi_0^- \right) \]

\[ \left( e^{i(\varphi_2 - \varphi_1)} e^{2} \sin^2 \left( \frac{\varphi_0}{2} T \right) - e^{i \omega T} \left[ \cos \left( \frac{\varphi_0}{2} T \right) - i \delta \sin \left( \frac{\varphi_0}{2} T \right) \right] \psi_0^- \right) \]. \quad (11)

Therefore, the spin encoding will work reasonably only up to a certain frequency limit.

In order to provide a descriptive picture for the following calculations, we switch from the SU(2) representation exploited above to SO(3) and the polarization picture, which can be done using the density matrix formalism to transform between the two representations. The resulting general expression for the polarization vector for the passage through a resonance flipper coil was given by one of the authors in [6].

Assuming an initial polarization vector \( P_0 = (0, P_{0y}, 0) \) and considering that RFSE coils have no static field \((B_0 = 0)\), we obtain for the polarization after one coil

\[ P(T) = P_{0y} \]

\[ \left( -\frac{1}{2} \sin(\omega T) \left( 1 + \cos(\omega T \delta) - \cos(\omega T) \right) + \delta \cos(\omega T) \sin(\omega T) + \varepsilon \sin(\omega T + 2 \varphi_1) \sin^2 \left( \frac{\varphi_0}{2} T \right) \right) \]

\[ \times \left( \frac{1}{2} \cos(\omega T) \left( 1 + \cos(\omega T \delta) - \cos(\omega T) \right) + \delta \sin(\omega T) \sin(\omega T) - \varepsilon \cos(\omega T + 2 \varphi_1) \sin^2 \left( \frac{\varphi_0}{2} T \right) \right) \]

\[ \varepsilon \delta (\cos(\omega T) - 1) \sin \varphi_1 + \varepsilon \sin(\omega T) \cos \varphi_1 \right). \quad (13) \]

where we used the notation from eqs. (8) and (9).

The “usable amount” of polarization for an actual spinecho measurement is the projection \( P_p \) of vector \( P \) from eq. (13) onto the polarization vector \( P_{\text{NRSE}} \) that corresponds to the perfect NRSE spin flip with both pi-flip and resonance condition fulfilled. This ideal vector after passage of a properly tuned resonance flipper, starting with \( P_0 = (0, P_{0y}, 0) \), reads

\[ P_{\text{NRSE}} = P_{0y} \left( \sin(2 \varphi_1 + \omega T) \right) - \cos(2 \varphi_1 + \omega T) \right). \quad (14) \]

The projection \( P_p \) is collinear to \( P_{\text{NRSE}} \) and its norm is

\[ P_p = \frac{P \cdot P_{\text{NRSE}}}{|P_{\text{NRSE}}|^2} \]

\[ = P_{0y} \left[ \varepsilon^2 \sin^2 \varphi_1 (1 - \cos(\omega T)) + \cos(2 \varphi_1) \cos(\omega T) + \delta \sin(2 \varphi_1) \sin(\omega T) \right]. \quad (15) \]

Equation (15) applies to a specific phase \( \varphi_1 \) that the rotating field vector has with respect to the initial polarization vector. To finally obtain the beam polarization we have to average over all possible phases \( \varphi_1 \), i.e.

\[ P_{p, \text{beam}} = \langle P_p(\varphi_1) \rangle_{\varphi_1} = \frac{1}{2 \pi} \int_0^{2 \pi} P_p(\varphi_1) d\varphi_1 = P_{0y} \varepsilon^2 \sin^2 \left( \frac{\omega T}{2} \right). \quad (16) \]

Equation (16) provides the condition for optimizing the field amplitude \( B_\text{rot} \) for a given frequency \( \omega \). \( B_\text{rot} \) is implicitly contained in the Rabi frequency \( \omega_R \) and the parameter \( \varepsilon \) defined in eq. (9).

Estimation of the upper resolution limits for RFSE.

We shall now estimate the effect quantitatively by evaluating eq. (16) under the assumption of parameters typical for an RFSE setup: a coil thickness of \( D = 0.004 \text{ m} \), an average neutron wavelength of \( \lambda = 10 \text{ Å} \) (which corresponds to \( v \approx 395.6 \text{ m/s} \)), an initial polarization of \( P_{0y} = 0.95 \), and frequencies of \( f = \omega/(2 \pi) = 20 \text{ kHz} \) and 35 kHz. Assuming a coil distance in one spectrometer arm of 1 m that would yield spin echo times of \( \tau \approx 0.26 \text{ ns} \) and 0.45 ns, respectively.

Figure 2 (top) depicts a 3D surface plot of the polarization after one RFSE coil depending on the magnitude and frequency of the rotating field. Figure 2 (bottom) shows cuts through the surface plot at 20 kHz and 35 kHz. \( B_\text{rot} \) can, as said, be derived from eqs. (16) and (9), and it becomes apparent that with increasing frequency, the field magnitude needs to decrease slightly in order to maximize the polarization and hence the performance of the instrument. For our specific example, \( B_\text{rot} \) should be changed from 1.67 mT for 20 kHz to 1.61 mT for 35 kHz to obtain the maximum possible polarization. The latter is reduced from 80.45% (20 kHz) to 56.09% (35 kHz), which constitutes an approximate upper frequency limit up to which measurements can be performed in practice.

Summary and discussion. – The working principle of RFSE can be understood and theoretically treated as a special case of NRSE. We derived the quantum-mechanical result for the accumulated spin phase after one arm of
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Fig. 2: Polarization after one RFSE coil according to eq. (16), depending on the rotating field magnitude and frequency (top). To optimize the achievable polarization, the magnitude of the rotating field needs to be shifted to smaller values for higher frequencies (bottom). For comparison, the polarization at multiple $\pi$ flips is plotted as well. The gain in polarization in those cases is due to a significantly increased $B_{\text{rot}}$ magnitude, which results in a less pronounced detuning effect.

To optimize the achievable polarization, the magnitude of the rotating field needs to be shifted to smaller values for higher frequencies (bottom). For comparison, the polarization at multiple $\pi$ flips is plotted as well. The gain in polarization in those cases is due to a significantly increased $B_{\text{rot}}$ magnitude, which results in a less pronounced detuning effect.

For an RFSE instrument by using time-evolution operators applied to an initial spinor representing the spin state of the neutron. After parameterizing the result we derived the condition for optimizing the rotating field amplitude for a given frequency, and gave a numerical estimation of the loss of polarization for a typical set of parameters. In its basic NRSE-type setup using standard wire coils this loss is intrinsic to the RFSE technique even under ideal conditions. As the estimation of the maximum frequency in the previous section shows, it is suitable only for lower resolutions, i.e. frequencies up to few 10 kHz.

In order to make RFSE more interesting as a stand-alone technique, it would be desirable to extend the resolution to higher Fourier times. Strategies that might be exploited in order to achieve this include:

- Use of thin magnetized foils as spin flipper (see, e.g., [13], [14] and references therein). Equation (16) implies that if the spin flip happens on a very short path or time scale $T$, the usable coil frequencies can be increased, which in turn results in higher resolutions. In [13] it was demonstrated that such coils work in principle and further research on suitable foil alloys is required.

- The bootstrap technique (see, e.g., [3]). Stacking of $N$ single coils with alternating direction of rotation improves the resolution by a factor $N$. Although this will also lead to a reduction in polarization since more spin flips are performed, compared to operating the coil at a $N$ times higher frequency the bootstrap method would be the one allowing for higher resolutions. A respective feasibility experiment has been performed and will be presented by one of the present authors (NA) and collaborators in a forthcoming publication.

- Adding a resonant static field perpendicular to the rotating field.

This basically turns the coils into resonance spin echo flipper with truly rotating RF fields. Presented in [9] and [15] is a longitudinal NRSE setup including a guide field, while abandoning the zero-field requirement. The rotating fields (instead of linearly oscillating ones normally used in NRSE coils) would also eliminate the Bloch-Siegert shift [5] limitation.

An obvious advantage of the RFSE technique is that the technical effort that must be put into the most crucial parts of an RFSE instrument — the coils and their electronic equipment — is significantly lower compared to NSE/NRSE, and the implementation of the instrument can be realized in a rather compact way. The low-resolution domain cannot be accessed by NRSE due to the aforementioned Bloch-Siegert shift which becomes relevant for frequencies below approximately 25 kHz.

Also for conventional NSE machines very small Fourier times in the sub-ps range to detect fast dynamics are challenging to reach. RFSE may therefore function as the low-resolution mode for standard NSE spectrometers. This could be achieved by coupling coils that turn the polarization from the $x$-direction outside the RFSE zero-field shielding to the $y$-direction inside (cf. fig. 1). Since the maximum Fourier time required from the low-resolution option is about 100 ps (for sufficient overlap with the normal spin echo mode), the overall geometric dimensions of one RFSE arm could be brought down to approx. 0.4 m, based on the parameters from the previous section and assuming $N = 2$ bootstrap RFSE coils. This should allow for mounting the setup around the sample stage of most existing spin echo instruments.

Summarizing, RFSE could fill a gap in both resolution and field of application, i.e. as a low-resolution add-on or in combination with other scattering techniques like, e.g., small-angle neutron scattering instruments or triple-axis

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spectrometers. The details of those and other technical aspects will need to be addressed in future publications.

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