NEUTRON DEPOLARISATION IN MAGNETIC MATERIALS

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Abstract

The neutron depolarisation technique is based on the change of polarisation of a polarised neutron beam in three dimensions after transmission through magnetic substances. This change yields the mean domain size, the mean square direction cosines of the domain magnetisations and the mean magnetisation. The method is complementary to other neutron scattering techniques with respect to the size of the inhomogeneities to be studied as well as the dynamic range accessible. The principles of the technique will be explained in some detail and demonstrated with a number of applications.

1. Introduction

The application of neutron depolarisation started already in 1941 by Halpern and Holstein [1] theoretically and Burgy et al in 1950 experimentally [2]. Contrary to neutron scattering and small angle neutron scattering (SANS) the method has never developed into a widespread application. At present, neutron depolarisation is exploited at a few places in the world. Without aiming to be all-embracing, I would like to mention the work by Drabkin, Okorokov et al [3,4] and the theoretical work by Maleyev, Toperverg et al [5] in Leningrad and also by Rauch [6] and Badurek et al [7] in Vienna and all references therein. The range of sizes probed by neutron depolarisation is between 0.02 μm and macroscopic dimensions, i.e. complementary to and overlapping SANS. In the range above 0.1 μm the application of SANS fails by a lack of sufficient resolution. However, the application of the neutron depolarisation technique is confined to magnetic phenomena and enables one to determine magnetic inhomogeneities as domain size or correlation length of the local magnetisation, the mean square direction cosines of the domain magnetisation directions and the mean magnetisation vector [8] and their time dependence. Further it should be noted that in a ND experiment one single intensity measurement determines a correlation length while in a SANS experiment the whole momentum dependence is needed for this information.

In the next sections the neutron depolarization technique will be treated and an overview will be given about the applications, without aiming to be all embracing. A few applications in materials research will be discussed in more detail.

2. Interaction of neutron spin with magnetic induction

Because of the magnetic moment of the neutron the magnetic interaction of the neutron spin with a magnetic field is described by the Hamiltonian \( \hat{H} = -\hat{\mu} \cdot \mathbf{B} \) where \( \hat{\mu} = \gamma \mu_s \hat{s} \) and \( \hat{s} = \frac{\hbar}{2} \hat{\sigma} \). Using basic quantum mechanics the time derivative of the moment operator can be described by,

\[
\frac{d\hat{\mu}}{dt} = -\frac{i}{\hbar} \left[ \hat{\mu}, \hat{H} \right]
\]
The square brackets indicate the commutator of the two included operators. Working out this equation leads exactly to the classical Larmor equation of precession of the spin operator around the magnetic induction \( \mathbf{B} \). The average of this operator can be defined as the classical polarisation vector \( \mathbf{P} = \langle \hat{\mathbf{\sigma}} \rangle \) and is described by,

\[
\frac{d\mathbf{P}}{dt} = \gamma (\mathbf{P} \times \mathbf{B})
\]

(2)

The solution of this equation is the Larmor precession of the polarisation vector around the field \( \mathbf{B} \). Although according to quantum mechanics only one spin component can be measured at a certain time, all three components can be determined successively without any problem. The Larmor precession frequency is given by \( \omega = \gamma |\mathbf{B}| \). Except for the construction of polarisation rotators this precession is the basic principle of the neutron depolarisation technique and also of the neutron spin echo technique. Both techniques make use of the proportionality of the interaction time \( t \) of the neutron with any field and the product of interaction length \( l \) and neutron wavelength \( \lambda \) \(( t = \nu = ml/\hbar )\). With the interaction time also the Larmor precession angle \( \phi = \omega t \) is determined. Another essential feature of these techniques is the use of neutron polarizers for polarisation and analysis of the polarisation.

In case of a interaction with a time dependent field the solution of equation (3) can be approximated by successive iteration. In formula,

\[
\mathbf{P}(t) = \mathbf{P}(0) + \gamma \int dt [\mathbf{P}(0) \times \mathbf{B}(t)] + \gamma^2 \int dt dt' \left[ [\mathbf{P}(0) \times \mathbf{B}(t)] \times \mathbf{B}(t') \right] + \cdots
\]

(3)

where the third term on the right hand side is assumed to be much smaller then the first two terms. The first two terms describe the normal Larmor precession around the average magnetic induction while the third represents the deviations from the average precession, which however is still not depolarisation. After averaging this term over the beam cross-section with different \( \mathbf{B}(t) \) paths this leads to depolarisation of the beam.

In the above treatment only magnetic interaction is assumed. In case of interference of nuclear and magnetic interaction and correlations between the nuclear and magnetic inhomogeneities, new more complicated depolarisation effects can be expected which will not be treated here.

3. Neutron depolarisation

Measuring Method

In this technique use is made of a polariser \( \mathbf{P} \) and an analyser \( \mathbf{A} \) in combination with two polarisation rotators \( \mathbf{R}_1 \) and \( \mathbf{R}_2 \) (see schematic view in fig.1). Each rotator consists of two mutually perpendicular coils in which a magnetic field can be generated of any required size and direction in a plane perpendicular to the neutron beam. In this way the precession of the polarisation vector can be chosen in such a way that any required orientation of the polarisation can be obtained [8].

![Schematic view of the depolarisation set-up. Here \( \mathbf{P} \) and \( \mathbf{A} \) are the polariser and analyser respectively, \( \mathbf{R}_1 \) and \( \mathbf{R}_2 \) polarisation rotators, \( \mathbf{S} \) the sample holder, \( \mathbf{SE} \) a spin echo coil to compensate for large rotation angles in the sample holder and \( \mathbf{D} \) is the neutron detector.](image-url)
In the same way any desired component of the polarisation at the sample position can be rotated to the analysing direction determined by the analyser magnetisation direction. In this way a (3x3) depolarisation matrix can be measured successively that describes the 3-dimensional change of the polarisation vector. To avoid depolarisation by the wavelength spread in the beam, just behind the sample in the sampleholder a spin-echo compensation coil (SE) can be applied to compensate for large rotation angles in the sample, e.g. by strong magnetised samples [9]. The current applied in the SE coil is adjusted to obtain optimal polarisation and doing so the current is a direct measure for the mean magnetisation of the sample. The use of the SE coil creates also the possibility to carry out 3 dim analysis on magnetised media while using a white neutron beam, with the advantage of measuring with very high neutron intensities. All effects together can be described by the formula,

$$P(t) = D P(0)$$  \hspace{1cm} (4)

The matrix $D$ contains now the depolarisation of the sample and the rotation by the magnetisation and spin-echo coil. The matrix is determined from the measured intensities by,

$$D_{ij} = (I_s - I_{ij})/(I_s - I_m)$$  \hspace{1cm} (5)

with $I_s$ and $I_m$ the intensities of the fully depolarised and polarised beam respectively and $I_j$ the intensity of the beam with $j,i = x, y$ or $z$ referring to the direction of polarisation and analysis respectively. The detected neutron pulses are eventually stored in a multichannel in order to measure the time dependence of the depolarisation matrix. The start signal of the latter can be synchronised with a periodic magnetic field or other periodic parameter of the sample. The optimal real time resolution amounts $\sim 5 \mu$s.

**Neutron Depolarisation in systems with known average domain structure.**

In the study of inhomogeneous magnetic media the measured depolarisation matrix $D$ can theoretically be described as the solution of the Larmor equation as described in equation (3), but sometimes the knowledge of the system in study allows one to describe the theoretical depolarisation matrix as a product row of matrices as given in equation (6),

$$P(t) = \prod_{i=1}^{N} D_i P(0)$$  \hspace{1cm} (6)

where some preknowledge of the system can be used to describe the product row of matrices in a number of parameters which can be determined by fitting the measured matrix with the theoretical expression of equation (6). A maximum of 9 parameters can be measured in this way. This approach has been used in the analysis of depolarisation in an amorphous ribbon Fe$_{70}$Ni$_{30}$B$_3$, which consists of three magnetic layers, two surface layers with local magnetic anisotropy perpendicular to the layer and a central layer with easy axis in the plane of the layer [10]. The effect of each of them on the polarisation vector can be described by a simple modified rotation matrix.

**Neutron Depolarisation in systems with random magnetic correlations.**

More generally the theoretical depolarisation matrix is found by solving eq.(3) for the case of zero net magnetisation in the small angle approximation which is here equivalent with the Born approximation in scattering theory. This leads to [11],

$$P' = P - \frac{cL}{W} \iiint d^3 r \int_0^z \left[ (\mathbf{B}(x) \cdot \mathbf{B}(x')) P - (\mathbf{P} \cdot \mathbf{B}(x')) \mathbf{B}(x) \right] + \ldots$$  \hspace{1cm} (7)
In this expression the integrals over \( t \) and \( t' \) have been replaced by the integrals over the position coordinate \( x \) and \( x' \). The terms \( B(x) \) and \( B(x') \) are short hand notation of \( B(x,y,z) \) and \( B(x',y,z) \). Moreover the averaging over the cross-section of the beam and one integral over \( x \) have been combined in the integral over a small part \( L_w \) of the sample volume \( W \). From this expression the elements of the depolarisation matrix \( D \) can be found,

\[
D_{ij} = \delta_{ij} \left( 1 - cL_w \xi^0 \right) + cL_w \alpha^0_{ij}
\]  

(8a)

with

\[
\alpha^0_{ij} = \frac{1}{W} \int W^3 r^z \int x' B_i(x)B_j(x') \, dz_0
\]

(8b)

\[
\xi^0 = \sum_i \alpha^0_{ii}
\]

More generally in a sample magnetised in the y-direction, the depolarisation matrix under certain conditions is given by the matrix elements,

\[
D_{xx} = D_{zz} = \cos \phi \exp \left[ -cL_s \left( \xi^0 - \alpha^0_{xx} \right) \right]
\]

\[
D_{xz} = -D_{zx} = \sin \phi \exp \left[ -cL_s \left( \xi^0 - \alpha^0_{xz} \right) \right]
\]

\[
D_{yy} = \exp \left[ -cL_s \left( \xi^0 - \alpha^0_{yy} \right) \right]
\]  

(9)

with \( L_s \) the total sample thickness in the neutron direction and

\[
\alpha^0_{ij} = \frac{8\pi^4}{W} \int Q^3 \Delta B_i(q) \Delta B_j(-q) \, d^2 q
\]

\[
\xi^0 = \frac{8\pi^4}{W} \int Q^3 \Delta B(q) \cdot \Delta B(-q) \, d^2 q
\]

\[
\Delta B(q) = \frac{H_0}{(2\pi)^3} \int W^3 \Delta B(r) e^{iqr} \, d^3 r
\]

\[
\Delta B(r) = [q' \times \Delta M(r) \times q']
\]

and \( \phi = \langle B > L_s c_1 \)

with \( c_1 = 4.7 \times 10^{14} \, \lambda \, m^{-2} \, T^{-1} \) and \( c = c_1^2 / 2 \)

Expressions (9) have been derived in the Larmor approach assuming isotropic depolarisation in the xz-plane. In the scattering approach it is not possible to derive these expressions for a magnetised sample because the Born approximation is not valid in the case of large continuous phase rotation in passing the sample. Using the Eikonal approach [12,13,14] it appears possible to derive the depolarisation formulae also in the magnetised case, from which the intuitive Larmor approach for this case can be verified.

In expressions (10) \( q \) is a vector in the 2 dimensional reciprocal space \( Q \) perpendicular to the beam direction and \( q' \) its unit vector. The expressions (10) can be found from
equations (8) by using the Fourier representation of the magnetic induction fluctuation. This representation has the advantage that stray fields from a certain magnetisation distribution are included already in the expressions by means of the vector expression of $\Delta \mathbf{B}(\mathbf{r})$ in eq.(10) which contains no field contribution any more. The latter is the result of applying Maxwell’s equations on $\mathbf{B}(\mathbf{r})$. A more rigorous treatment of the depolarisation in terms of neutron scattering theory shows that the reciprocal vector $\mathbf{q}$ can directly be identified with the elastic neutron momentum transfer vector. The correlation length $\xi^0$ of eq.(10) is in scattering terms nothing else than the total magnetically scattered intensity, $\xi^0$ of eq.(10) determines in this way the smallest correlation length which can be determined with this method. From equation (10) it appears that ND can be compared directly with SANS results. The correlation parameter $\xi^0$ can be identified with the fluctuations of $\mathbf{B}(\mathbf{t})$ around the average $\langle \mathbf{B} \rangle = \mathbf{mB}$. These fluctuations can be approached by $\xi^0 = \mathbf{B}^2/(1-m^2)\delta$, where $\delta$ is a correlation length over which the magnetic fluctuations are more or the less homogeneous. In ferromagnetic domain structures $\delta$ can be identified with the mean domain size. The ratio $\gamma = \alpha^2/\xi^0$ gives the mean square direction cosines of the fluctuations along $i= x, y$ and $z$ axis. A trivial correlation in the magnetic sample imposed by the Maxwell laws $\text{div} \mathbf{B} = 0$ and $\text{rot} \mathbf{H} = 0$ and translated in Fourier space by $\mathbf{B}(\mathbf{r}) = [\mathbf{q}' \times [\mathbf{M}(\mathbf{r}) \times \mathbf{q}']]$ in equation (11), causes that also in further isotropic systems not $\gamma_x = \gamma_y = \gamma_z = \gamma$ is measured but $\gamma_x = \gamma_y = \gamma_z = \gamma$ if $x$ is the transmission direction of the neutron beam.

**Polarisation rotation in magnetised media.**

From equation (10) the precession angle $\varphi$ is proportional to the $\langle \mathbf{B} \rangle$ where $\langle \mathbf{B} \rangle$ is spatially defined within the margins of the sample thickness $d$. What is the situation in case $\langle \mathbf{B} \rangle$ is directed parallel to the neutron beam? In this case also precession occurs in the demagnetisation fields of the sample and one may wonder what is actually measured in the Larmor precession. For this purpose one may consider the line integral $\int \mathbf{J}_d \mathbf{dS}$ along a neutron path. Neglecting the contributions in infinity one can easily derive,

$$\int_{-\infty}^{\infty} \mathbf{J}_d \mathbf{dS} = \int_{S} \mu_0 \frac{\mathbf{J}}{S} \text{rot} \mathbf{M} dS = \mu_0 \int_{S} \mathbf{M}_d \mathbf{dx} = \mu_0 \mathbf{M} d$$

which means that also in this case without currents ($\mathbf{H} = 0$) the measured rotation angle corresponds to the magnetisation in product with the thickness of the sample and the effects of the demagnetisation fields cancel.

### 4. Application of ND in magnetic materials

The ND has been applied in numerous subjects in the course of the years. Some of them are listed below. As an example the application in HTS YBACU will be discussed in more detail.

**Thin Films**

In thin magnetic films neutron depolarisation can be applied in studying the domain structure perpendicular as well as parallel to the film [15]. In this respect neutron depolarisation is also complementary to neutron reflectometry which studies the atomic and magnetic structure of the film perpendicular to the surface [16]. The application range of the reflection technique is typically from atomic dimensions up to 0.1 $\mu$m perpendicular to the film, while the range of the neutron depolarization starts above 0.01 $\mu$m, however in all directions.

**Amorphous ribbons under stress**

Amorphous ribbons appear to be consist of a layered domain structure, caused by a stress transition from compressive to tensile stresss going from the surface to the
centre of the ribbon. With 3 dim. ND it appears possible to analyse this domain structure in detail and by doing this as a function of applied stress on the ribbon, it appears possible to scan the stress distribution in the ribbon [10]. Also from the wave length dependence of 1 dim. ND with the polarisation making an angle with the average magnetisation of the ribbon, similar although less informative results can be obtained [17].

**Small Particle Systems, [11]**

In these systems the magnetic correlation between particles in remanent state and field dependent can be determined, together with the mean direction cosines of the local particle magnetisation directions and the average magnetisation [18]. Also the field history and effect of particle shapes appear important on the correlations [11]. The measured parameters are important for characterisation of the microstructure in magnetic recording materials and ferrofluids.

**Time dependent ND**

Also in the dynamics of materials, neutron depolarisation is complimentary with inelastic neutron scattering. However, the dynamics studied with neutron depolarisation are measured in real time in contrast with the scattering techniques. Dynamic experiments are carried out by applying a periodic action to the sample such as a magnetic field or a tension and studying the response of the domain structure on this action by measuring the neutron intensity in periodically triggered time channels [19]. With this technique a time resolution of about 5 $\mu$s can be obtained. Because of the limitation in time scale at short times time dependent neutron depolarisation has up to now only been applied successfully to eddy current limited domain wall movements and to time dependent rotation of induction in high T superconductors and flux creep [20].

The eddy currents have been studied in a ring shaped iron and nickel sample with rectangular cross-section of the ring. By applying a block-shaped magnetic field in the ring by a toroidal coil the magnetisation in the ring is reversing periodically in time, starting by domain nucleation at certain centra at the ring surface, which expand in time until they merge to a more or the less flat domain wall moving to the centre of the ring. From the time dependent Larmor precession of the polarisation around the local magnetisation the wall mobility of the flat wall could be determined [19, 21], form the depolarisation the time dependent flatness of the moving wall [19, 22]. In similar studies in thin FeNi soft magnetic films the repelling forces between expanding domain nuclei, approaching each other from the opposite surfaces could be demonstrated [23].

**Magnetic Phase Transitions**

Because of the sensitivity of the Larmor precession for small magnetic inductions ND has been applied to study the critical behaviour in pure Fe and Ni. It appeared possible to determine the critical exponents $\beta$ and $\gamma$ with very high accuracy close to the critical temperatures of Fe and Ni [24].

The first order phase transition of Austenite - Ferrite Steel is very important in heat treatments of steels. With ND it appears possible to determine from the mean Larmor precession angle the mean fraction of ferrite, because ferrite is ferromagnetic, and from the depolarisation the mean particle size of the ferrite particles transformed [25]. This method appears to achieve quite unique information about this phase transition.

**Superconductors**

By scanning a very narrow neutron beam through a superconductor in a magnetic field, detailed information can be obtained about the magnetic flux penetration into the superconductor [26]. By studying the time dependence of ND after applying a pulse or alternating field, the time dependence of the flux penetration can be followed on millisecond scale [27].
Temperature dependence of ND in high T superconductor YBaCuO.

As an illustration of the application of the depolarization technique, experiments were carried out on a sintered YBa$_2$Cu$_3$O$_x$ sample of size 5.35 x 20.55 x 11 mm$^3$ with a grain size between 1 and 10 $\mu$m. [20]. The sample positioned inside a coil is clamped between a magnetic short circuit to prevent emerging flux from the sample and coil to affect the polarization of the beam outside the sample. The whole is positioned in a cryostat. At low temperature (4K) a pulsed magnetic field of $\sim$ 0.7 T with 0.1 ms duration was applied after which the depolarization matrix was measured with rising temperature. Fig.2 gives the results of the average magnetic induction components $B_x$, $B_y$ and $B_z$ and the correlation parameters $\alpha_{xy}$ and $\alpha_{xy}$ which show the correlations in the magnetic components perpendicular and parallel to the applied field direction.

The different temperature dependence of $\alpha_{xy}$ and $\alpha_{xy}$ is striking and is interpreted as follows. Direct after the field pulse the fluxlines are randomly directed in the half sphere with some remanence in the initial field direction, but in bundles of $\sim$20$\mu$m sizes of the same order as the crystallite size. With increasing temperature the fluxlines start to stretch themselves in the average fluxline direction. Above 20 K all fluxlines are parallel but still inhomogeneously distributed. Above 40 K no measurable depolarization is present and the fluxlines are distributed homogeneously over the sample. Above this temperature the linear T dependence of $\langle B \rangle$ indicates a $(T-T_f)^{1/2}$ dependence of the fluxline spacing.

![Figure 2](image)

**Fig.2** (Left) Remanent magnetic induction as a function of temperature, after zero-field cooling and a field pulse at 4.2 K. (Right) Average of the squared induction fluctuations perpendicular ($\alpha_y$) and parallel ($\alpha_x$) to the initial applied field, plotted as a function of temperature.

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