Magnetic and Thermodynamic properties of RNi5 compounds
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Citation for published version (APA):
Kayzel, F. E. (1997). Magnetic and Thermodynamic properties of RNi5 compounds UvA
4 Experimental

4.1 High-field magnetisation measurement

4.1.1 The 40 and 30 tesla pulsed field magnet
The high-field magnetization measurements have been performed in the High Field Installation of the University of Amsterdam [Gersdorf et al. 1983 and Roeland et al. 1988]. Two resistive magnets with similar design are available. In the oldest configuration magnetic fields up to 40 T can be generated. A newer set-up can make fields up to 30 T. Both coils produce a magnetic field in a semi-continuous way. The total pulse length is of the order of one second (see figures 4.1 and 4.2). The highest field of 40 T can be sustained for 50-100 ms, lower fields can be generated for longer periods.

The magnetic field is generated by a current \( I_{\text{max}} = 8800 \) A through a coil. The magnet coil is wound from hard-drawn insulated rectangular copper wire, cross section 2.9×7.13 mm\(^2\) and 680 windings. The magnet is divided at 1/3 of its diameter by a steel cylinder to reinforce the copper coil. The electric power is taken from a 10 kV line of the municipal grid and rectified through 6 MW thyristors which are controlled by a small computer. The maximum output voltage of the rectifier is about 560 V and the maximum power consumption is 5.8 MW. The computer first calculates the current necessary to generate approximately the desired field based on the known properties of the rectifier and the coil. This calculation is used to ignite the thyristors and produce a field. Then a measurement of the actual field is used to regulate to the desired field. Controlling the current in this way has the advantage of being able to control the field, in every aspect, during a pulse. The maximum deviation of the actual field from the desired constant field is less than about 5 mT, after a settling time of about 20 ms.

The limitations of the pulse are only dictated by the design of the magnet and available power supply. The 40 T coil is cooled to 30 K by liquid neon (supplied by a closed-circuit neon liquefier) after being precooled by liquid nitrogen. The 30 T coil is only cooled by liquid nitrogen. At 27 K the resistance of the 40 T magnet is \( \sim 8 \) m\( \Omega \). During a pulse the magnet can heat up to \( \sim 120 \) K with a resistance increase to 150 m\( \Omega \). After such a pulse the cooling machine needs two hours to take away the heat of the pulse. For lower pulses the cooling time is considerably shorter. The resistance of the 30 T magnet at 77 K is about 35 m\( \Omega \) and can be pulsed up to 120 K or 150 m\( \Omega \). The two constraints of the magnet design are the temperature of the coil and the strength of the wire. The strength of hard-drawn copper wire limits the field to about 45 T. At higher fields the Lorentz force, \( F_{\text{Lorentz}} = \text{field} \times \text{current} \), will break the copper wire. The temperature of the coil restricts the magnet through its ohmic resistance. The voltage is regulated to overcome the resistance, heating of the coil increases the resistance, the voltage has to increase again until the maximum voltage is reached.

The field is measured direct with a pick-up coil and an integrator. The pick-up coil is calibrated by measurement of De Haas-van Alphen oscillations of a silver sample. The oscillations are a result of the quantization of closed electron orbits in a magnetic field. The change in \( 1/H \) through a single period of oscillation, \( \Delta(1/H) \), is determined by a simple
**Figure 4.1** A typical example of a high-field pulse with a linear decreasing field from 38 T down to 3 T (continuous curve and left hand scale). The dashed line represents the response of the sample (a single crystal of NdCo$_5$, oriented along the [001] axis, note the transition around 32 T). The rectangular shaped step in the response signal is a calibration signal.

**Figure 4.2** A typical example of a step-wise high-field pulse with a maximum field of 35 T (continuous curve and left hand scale). The dashed curve represents the integrated response of the pick-up coil system without a sample. The rectangular shaped step in the response signal is a calibration signal.
relation:

\[ \Delta \left( \frac{1}{H} \right) = \frac{2\pi e}{\gamma c \cdot A_e} \]  

(4.1)

where \( A_e \) is any extremal cross-sectional area of the Fermi surface in a plane normal to the magnetic field. The oscillations of Cu, Ag and Au are determined with high accuracy by Coleridge and Templeton [1972]. A calibration of the pick-up coil with these values is regularly performed.

### 4.1.2 Magnetization measurements

The magnetization is measured by an induction method. In principle, the voltage induced in a coil by the time variation of the magnetization is electronically integrated to get the magnetization itself. The induction voltage is proportional to

\[ \frac{\partial B}{\partial t} = \frac{\partial}{\partial t} \mu_0 (H+M) \]  

(4.2)

In order to obtain the magnetization one has to remove the contribution of \( \partial H/\partial t \) from the output signal as well as possible. The easiest way to do this is by combining the measuring coil with a compensation coil. This would be a coil wound in the opposite sense, which measures only the contribution of the field. This is satisfactory for slowly varying fields, but not sufficient for transient fields. During fast changing fields eddy currents are generated in the metallic parts of the magnet assembly resulting in stray fields which change the field configuration both in space and in time. It was shown by Gersdorf et al. [1967] that a coil consisting of two confocal concentric ellipsoidal coils has the property, if designed with correct dimensions, to generate no external field while the interior field is homogeneous. There is no mutual inductance with an external magnet, and moreover the coupling with a magnetic moment placed at the interior is independent of the position of the sample. A next to best approximation is an elaborated system of four solenoids which has been designed to be insensitive to a field configuration consisting of the first three orders of spherical harmonics as well as to the position of the sample inside the coil. Accuracy in machining prevents the exact construction of the calculated coils, especially the large zero-order contribution has to be compensated by an external signal which is derived from a simple pick-up coil system. The remaining signal is due to imperfect adjustment of this compensation and to non-linear magnetic response of the coil system itself.

Internal strains in the construction, especially after thermal cycling, will alter the delicate balance between the coils and result into an irreproducible signal, proportional to the field. Subtraction of signals obtained after long time intervals (due to long cooling periods of the magnet) may therefore be erroneous in this respect. But because this error is known to be proportional to the field one can usually correct for it, or reduce it by averaging several runs.

The output signal of the coil system is electronically integrated, digitalized and stored. The main problem is to interpolate properly the zero of the integrator during the pulse. The uncertainty introduced in this way is the main factor determining the overall accuracy of the magnetization measurements; expressed in magnetic moment the precision is \( 2 \times 10^{-5} \) Am\(^2\).

A special pulse shape uses an auxiliary field prior to the actual high field pulse and also before the integrator is set free. If now, at the end of the pulse, the auxiliary field is switched off, and a ferromagnetic sample is simultaneously pulled out of the measuring coil system, a very accurate
measurement of the ferromagnetic moment is possible. Pulling out of the sample eliminates remanence as an uncertain factor. After each pulse, the magnetization and the field are calculated and the result of the same pulse without a sample subtracted.

The system has been calibrated with a known magnetic moment, namely the zero field saturation magnetization of pure nickel.

The sample is introduced into the system in a teflon sample holder. The maximal available space for a sample is 3 mm diameter and a total length of 25 mm in order to stay within a 1% homogeneity of the field. The total system is immersed in the cryogenic liquid to prevent heating by eddy currents. In this way measurements from 1.35 to 4.2 K are a standard method.

Magnetization measurements in this thesis were primarily done on spherical single-crystalline specimens. These samples were orientated with Laue back scattering technique within 1° along a crystallographic axis. After orientation the samples were glued with strain gauge glue and cast into transparent styecast 1266 [Emmerson & Cuming] between two non-magnetic cylinders (celleron) to fix the sample orientation during the pulse. If more accuracy is required an empty sample holder and a celleron plus styecast sample is measured and subtracted from the pulse with sample.

4.2 Low-field magnetization measurements

Low-field magnetization measurements, in fields up to 7 T, have been performed in a moving sample magnetometer. This system is based on a set of oppositely wound pick-up coils within a superconducting solenoid. The sample is moved between the centres of the pick-up coils. By integration of the induced voltage the magnetization of the specimen can be determined.

The temperature in sample space can be changed from 4.2 K to about 300 K. Two different sample holders are available: one of perspex and a beryllium-copper one. The latter is used to apply a hydrostatic pressure, up to 5.5 kbar, to the sample [Buis 1979, Frings 1984].

4.3 Specific-heat measurements

4.3.1 Introduction
In this section the specific-heat method and the used equipment is described. There are several ways of determining the specific heat of intermetallic compounds at low temperatures. These methods can be divided in two main measuring techniques:
– the adiabatic method [see eg Morin and Maita 1963],
– the dynamic (non-adiabatic) method.
The latter method can be divided into several sub-methods, eg the continuous heating method [Ashworth and Steeple 1969, Schutz 1974, Junod 1979], the relaxation method [Bachman et al. 1972], the ac-method (ac calorimetry) [Sullivan and Seidel 1968].
The choice of a suitable method depends on several conditions. The most important conditions are: the thermal isolation of the sample from the environment, the importance of relative or absolute accuracy, the heat capacity and the thermal conductance of the sample measured.

In any practical situation all the specific-heat measurements have a relaxation component in their measuring curve, even the adiabatic ones (then referred to as ‘semi-adiabatic’), see figure
4.4. In the case of an ideal adiabatic measurement it has an infinite relaxation time. A known amount of heat is added to the system, heats it up to a higher temperature, and the whole system should stay at this temperature. A more realistic behaviour is presented in figs. 4.3.a and b. In a relaxation measurement the total system is heated until a steady state situation exists. Then the heater is turned off and the whole system relaxes back to the surrounding temperature.

In the next paragraphs the adiabatic method and the relaxation method will be discussed.

4.3.2 Adiabatic measurement

Adiabatic calorimetry is based on the heat-pulse method and is considered as one of the most accurate heat capacity measuring methods. A sample is isolated very well from its surroundings. In the ideal case no heat exchange takes place and the method is really adiabatic. In a simplified model the thermometer - sample link and heating device - sample link are considered ideal and without any thermal resistance. Then the thermometer heat capacity and the heater heat capacity do not contribute to the system.

At time $t_2$, indicated in fig. 4.3.b, a constant power $P$ is switched on. The constant heating, up to time $t$, means that a known quantity of energy, $\Delta Q = P(t_1 - t_2)$, is added to the system. The corresponding temperature increase, $\Delta T$, taken at time $(t_3 + t_2)/2$, is determined by extrapolation. The heat capacity, $C$, is then calculated as

$$C = \lim_{\Delta T \to 0} \frac{\Delta Q}{\Delta T} \approx \frac{\Delta Q}{\Delta T}$$  \hspace{0.5cm} (4.3)

Assumed is that during the heating the resistance of the heater-wire does not change and the temperature step is sufficient small to allow for the approximation.

In adiabatic methods at low temperatures a heat switch is needed to cool the sample. In many cases this is a complicated mechanical construction. In a practical situation there will be always a small heat leak, and the method is called semi-adiabatic. In our equipment the sample plus addenda (sapphire plate, resistances for heater, thermometers, grease) is attached thermally to its environment by their wiring allowing us to cool the sample to the lowest temperatures without...
The advantage of the adiabatic measuring method is high, absolute and relative, accuracy. A disadvantage is the need for large samples to keep the error limited. Heat leaks through the electrical wires, necessary for temperature measurement and heating, give the lower limit for the mass of the sample. The thermal conductance of these wires will determine the relaxation time of the system and with this the steepness of the cooling curve, the temperature line $t_3 - t_4$ in figure 4.3.b. In measurements with a small sample the contribution of the heat capacity of the sapphire plate will be of the same order of magnitude as the heat capacity of the specimen. Also the thermal conductance is less as the contact area becomes smaller and the adhesive (Apiezon N) is relatively thick. This all leads to strong curvatures and overshoot in the cooling part of the measuring curve (shorter relaxation time of the system) and a linear approximation of that part of the curve, used in extrapolating to the mid-point of the heating, $(t_2 + t_3)/2$, is no longer allowed. The measurement can then no longer be taken as adiabatic and different analysing techniques are necessary to extract the heat capacity of the sample. A measured specific heat curve with adiabatic conditions is shown in figure 4.4.

If the curvature is too strong, an extended measurement of the cooling period is taken to determine the relaxation time with an exponential fit. In the measuring computer programme a first estimate for the end-temperature is obtained by extrapolation of the preheating temperature dependence up to the time at which the last data were recorded. Then, with a least squares method, an exponential fit is made and the end-temperature is adjusted until a best fit is attained. This exponential fit for the period $t_3$ to $t_4$ is extrapolated to the mid-point of the heating and $\Delta T$ is determined.

Figure 4.4  Measured curve under semi-adiabatic conditions.

a heat switch.
4.3.3 Relaxation measurement

In order to cope with all possible samples and a wide temperature range the possibility to record relaxation curves has been incorporated into the measuring software. In certain measuring conditions, especially small samples or low heat capacity, the relaxation method has to be used. This method was first introduced by Bachman et al. [1972].

Let us first assume a perfect thermal contact between sample, heater and thermometer. This system is connected by a relative weak link to a heat sink. In fig 4.5a, $k$ is the heat leak conductance. During the heating of the sample the environment is kept at constant temperature, $T_0$ (with a separate thermometer and regulating circuit). A constant power, $P$, is supplied to the sample. After some time, the sample will reach a constant temperature, $T_0+\Delta T$, since the heat per

\[
P = \frac{P}{k}.
\]

Now if, at $t = 0$, the heater is switched off, the temperature of the sample will decay exponentially to the surrounding temperature with a relaxation time $\tau = C/k$. [Bachmann et al. 1972]. The cooling curve is given by

\[
T_s(t) - T_0 = \Delta T e^{-\tau t}.
\]

Hence, the heat capacity of the sample plus addenda can be determined by:

\[
C = \frac{P}{\Delta T}.
\]

Precise measurement of the power applied, $P$, is straightforward. In practice, accurate determination of the temperature step, $\Delta T$, and the relaxation time $\tau$ from the measured data is more complex.

The above mentioned expression is based on the assumption that the sample plus addenda can be considered as one single thermal unit, and that the thermal response of this thermal unit is much

**Figure 4.5**  a) Schematic heat-flow model for relaxation method with a single relaxation time $\tau$ (after Bachman et al. [1972]). A similar model can be made for the thermometer and heater. b) Heat-flow model for sample and substrate exhibiting a "lumped $\tau$ effect". [after Shepperd et al. 1984].
higher than that of the wires. Often this condition is not met: the thermal conductance between
the sample and the sample holder is small (see figure 4.5b). Especially when the sample is very
small, or has a very low heat capacity, the relative contribution of the thermal conductivity of the
sample-substrate bond contributes considerably.

As an approximation, two exponentials are used to describe the relaxation curve:

\[ T_s(t) - T_0 = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2} \]  \hspace{1cm} (4.6)

where

\[ A_1 + A_2 = \Delta T \]  \hspace{1cm} (4.7)

In case \( A_1, \tau_1, A_3 \) and \( \tau_2 \) all can be determined, the heat capacity \( C \) is then calculated with
[Shepherd et al. 1984]:

\[ C = k_b \frac{A_1 \tau_1 + A_2 \tau_2}{A_1 + A_2} \]  \hspace{1cm} (4.8)

Notice that \( k_b \) can be determined separately (\( k_b = P/\Delta T \) in the heating stage). In the case that \( \tau_2 \)
is an order of magnitude smaller then \( \tau_1 \) and \( A_2 \) is much smaller than \( A_1 \), then \( \tau_2 \) and \( A_2 \) cannot be
resolved accurately enough. In this case, we find from eq. 4.8, knowing that \( A_2 \tau_2 \ll A_1 \tau_1 \) and \( A_1 + A_2 = \Delta T \), the heat capacity

\[ C \approx k_b \frac{A_1 \tau_1}{\Delta T} \]  \hspace{1cm} (4.9)

One obtains the amplitude \( A_1 \) and relaxation time \( \tau_1 \) by subtracting \( T_0 \) from the \( T_s \) values,
collected during the decay, and fitting a straight line through \( \ln(T_s(t) - T_0) \). To avoid the \( \tau_2 \)-effect,
the curve should be fitted only after a time \( \approx 7 \times \tau_2 \) or \( \approx 0.7 \times \tau_1 \) from the begin of the relaxation

![Figure 4.6](image-url)

**Figure 4.6** Typical curve in a measurement of the specific heat with the relaxation
method.

RNi₅ compound
In figure 4.6 a measured relaxation specific heat curve is shown.

4.3.4 The specific-heat set-up
The high-temperature specific-heat insert is designed to measure the heat capacity in the temperature range from 1.4 - 300 K. Two shields are used to screen the sapphire plate with thermometer, heater, sample and addenda from the surroundings. Using a carbon-glass thermometer and a temperature controller, the temperature of the inner shield is measured and kept constant at temperature $T_0$ (the temperature at which the adiabatic measurement is started) within $0.1\%$ of $T_0$. At higher temperatures, typically sample temperatures above 150 K, the outer shield is heated with a constant power to remove a part of the temperature difference between the surrounding ($T_{he} = 4.2$ K) and the sapphire plate ($T>150$ K). The temperature of the sapphire plate and sample is measured with two thermometers; a thin film platinum resistance calibrated from 20 K to 300 K and a ruthenium oxide thin film resistance calibrated from 1.35 K to 25 K. The whole setup is computer controlled to simplify the measurement. A more detailed description of the specific heat set-up is given in Kim-Ngan [1993].

4.4 µSR measurements

4.4.1 Introduction to the technique
In this paragraph we give only a short introduction to the µSR technique. For further reading there are a number of publications e.g. a publication by Chappart [1984], a review article by Cox [1987], a textbook by Schenk [1985] or the thesis of Dalmas de Reotier [1990] which all give a more detailed description of the technique.

The µSR spectroscopy (µSR is the acronym of the English words "muon spin rotation" or "muon spin relaxation" or "muon spin resonance" or "muon spin research") is basically the implantation of polarized muons in a lattice. The spin of this particle will, at its position, only interact with the local magnetic field. The muon acts then as if it were a microscopic magnetic field meter. This information about the local magnetic field can be obtained by studying the anisotropic decay.

The muon, member of the family of the leptons like the electron, is a particle found in 1936 by Anderson and Neddermeyr who studied cosmic rays. The muon has a unitary electric charge; it exists in two forms with opposite charge $\mu^+$ and $\mu^-$. The muon has a mass of 105.6 MeV/c$^2$ which is about $1/9$ of the proton and 200 times a electron mass. The spin of the positive muon, with a value of $\hbar/2$, is parallel to the magnetic moment (anti-parallel in case of the negative muon).

Table 4.1 presents some physical properties of the positive muon compared with the positron and the proton.

The $\mu^+$ decays in the following way:
Table 4.1. Some physical properties of the positive muon compared with a positron and a proton.

<table>
<thead>
<tr>
<th>particle</th>
<th>$e^+$</th>
<th>$\mu^+$</th>
<th>$p^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>rest mass (MeV/c²)</td>
<td>0.511</td>
<td>105.6</td>
<td>938.9</td>
</tr>
<tr>
<td>charge</td>
<td>$e^+$</td>
<td>$e^+$</td>
<td>$e^+$</td>
</tr>
<tr>
<td>Spin (h)</td>
<td>$\frac{1}{2}$</td>
<td>$\frac{1}{2}$</td>
<td>$\frac{1}{2}$</td>
</tr>
<tr>
<td>gyromagnetic ratio</td>
<td>$1.7608 \times 10^{11}$</td>
<td>$8.5161 \times 10^8$</td>
<td>$2.6752 \times 10^8$</td>
</tr>
<tr>
<td>decay time (s)</td>
<td>stable</td>
<td>$2.197 \times 10^{-6}$</td>
<td>stable</td>
</tr>
</tbody>
</table>

\[ \mu^+ \rightarrow e^+ + \bar{\nu}_e + \nu_\mu \quad (4.10) \]

Here $\nu_e$ and $\nu_\mu$ are the electron and muon neutrino. Neutrinos are very hard to detect, so only the positron $e^+$ will be detected. Its kinetic energy is large enough to let the positron escape from the lattice. The emission of the positron is highly anisotropic. The probability of emission is proportional to

\[ [1 + A \cos(\theta)] \quad (4.11) \]

Here, $\theta$ is the angle between the direction of the muon spin at the time of decay and the direction of emission of the positron. The factor $A$, the asymmetry parameter, depends on the

Figure 4.7 Angular distribution of positrons from the $\mu^+$ decay. The radial distance is $1 + A \cos \theta$. Left only positrons of maximum energy are detected ($A = 1$). Right: positrons with all energies are detected ($A = 1/3$).

$R\text{Ni}_3$ compound
Experimental

positional energy and the average of A over all the possible positron energies is 1/3. In practice A is about between 0.20 and 0.24. In figure 4.7 the emission angle dependence is shown in two cases.

### 4.4.2 Implantation of muons in matter

Two muon bundle types are developed in the past years. One is a bundle with surface muons with a small kinetic energy (in order of 4.12 MeV). These are called surface muons because the penetration depth is small (e.g. 0.2 mm in copper). The other has fast muons with typical kinetic energy of 129 MeV. The surface muons have a typical velocity of 0.27c while the fast muons travel with 0.75 - 0.87c.

The following questions can be posed: which process thermalises the muons in the material? Does the muon keep the same polarisation direction during this tempering? In figure 4.8 the thermalization process of the muon in matter is sketched. First the positive muon is slowed down in the lattice via ionisation of atoms and scattering with electrons until it has a energy of ~2-3 keV. This deceleration occurs in an estimated time of 10^{-10} -10^{-9} s. At this stage the muon is capable of forming an atom together with an electron, called muonium. During this time inelastic scattering of muonium atoms and a series of gain and loss of electrons slow the muon down to lower energy (~200 eV) in a time of 5 ×10^{-13} s. Then due to the surroundings of free electrons the muonium will dissociate and lose its electron.

This implantation process described above takes in the order of 0.1-1 ns. Also the primary processes bring into play only electrostatic interactions, which are non spin-dependent. So the time is too short for the muon to have a loss of polarisation.

The muon can diffuse between interstitial sites in a crystal. The $\mu^+$ diffusion may introduce experimental difficulties in studies of magnetic properties of compounds. However, at low temperatures the muon will be localized on one site. At higher temperature we have to take into account the diffusion of the muon through the lattice. The muon is almost always found on

\[
\begin{align*}
\text{bundle of muons} & \quad \Delta t \leq 10^{-10} \text{ s} \\
\downarrow & \\
\text{Retardation of the } \mu^+ \text{.} & \\
\text{Loss of energy by interaction} & \quad E_\mu = 2 \text{ -3 keV} \\
\downarrow & \\
\text{with the electrons.} & \\
\downarrow & \\
\text{Loss of energy of the } \mu^+ \text{ by} & \quad \Delta t \approx 5 \times 10^{-13} \text{ s} \\
\text{interaction with the degenerated} & \\
\text{electron gas. Formation of muonium.} & \quad E_\mu = 200 \text{ eV} \\
\downarrow & \\
\text{Collision of muonium atoms} & \quad 10^{-12} - 10^{-11} \text{ s} \\
\downarrow & \\
\text{Dissociation of the thermalised muonium} & \quad E_\mu \approx 2 \text{ eV}
\end{align*}
\]

**Figure 4.8** Slackening of the $\mu^+$ in a metal after Brewer et al.[1975].

\[\text{RNI}_5 \text{ compound}\]
interstitial sites, only in irradiated samples containing many vacancies the muon may occupy a lattice position.

4.4.3 μ^SR spectroscopy

To summarize, the μ^SR technique is based on two different experimental facts. Firstly, a muon is produced with its polarization antiparallel to its momentum and hence, the muon is naturally polarized. Secondly, the muon decay is anisotropic and its anisotropy is directly related to the μ^ spin direction mentioned above. Therefore, it is possible to follow the evolution of a muon in a sample.

In the DC μ^ beam set-up, the polarized muon beam is aimed at the sample. Each muon passes the initial detector(s) and if there is (almost) no simultaneous signal at the detector behind the target, then a muon has been stopped in the sample. A muon event requires that within a certain time interval after the μ^ stop, a positron is detected. This event is called data gate. Each implanted muon starts a clock and triggers a data gate signal of about 10 μs length (few muon lifetimes). During the data gate no other muon is accepted in order to avoid coincidences. In this way a rate-versus-time histogram is made. The data gate limits the number of events (stopped muons), to the order of 5×10^4 μ^/s.

In a pulsed μ^ beam set up, when the muon burst in the pulsed beam is sufficiently short, compared to the muon lifetime, and the separation between the pulses is long compared to the muon life time, then it can be used as a collective start signal for all incoming muons of the pulse. In this way the limitation of the number of events of the DC method is avoided.

The number of detected muons, N, is given in a law of exponential decay observed for radioactive particles of mean lifetime τμ (=2.197 μs).

\[ N_e(t) = N_0 e^{-t/\tau_\mu} [1 + acos\theta] \quad (4.12) \]
The muon spins in the incoming beam are polarized in the direction of the beam. The positron detector (e\(^+\) are detected hence the subscript) is making an angle \(\theta\) with the trajectory of the incoming beam and hence with the muon spin. The desintegration anisotropy (eq. 4.1) has been taking into account. \(N_0\) and \(a\) are effective parameters derived from respectively \(N\) and \(A\) and representing the experimental conditions. To complete the calculation we have to integrate over all possible energies of the positron. Implicitly, we assumed that all muon spins remain polarized. In reality, the spin interacts with the local magnetic moment at the site where it is implanted (stopped). Possibly they can lose their initial polarisation. Now, we define \(P_\alpha(t)\) as the polarisation of the beam, at time \(t\), projected in the direction, \(\alpha\), at which the detector is placed. It is the average value of the ensemble of muons with their spin projected in the direction \(\alpha\). With a change in notation (eq. 4.1) of \(N_\alpha(t)\) to \(N_\alpha^{e^+}(t)\) we get

\[
N_\alpha^{e^+}(t) = N_0 e^{-\alpha t} [1 + a P_\alpha(t)]
\]  

(4.13)

Presenting \(\mu\)SR experiments one often shows only \(a P_\alpha(t)\). The information in \(N_\alpha^{e^+}(t)\) adds only the radioactive decay of the muon and conceals more or less the asymmetry, \(a P_\alpha(t)\). Thus only \(a P_\alpha(t)\) is presented. This is accomplished by taking two detectors with the same characteristics placed opposite to each other. We have one forward detector

\[
N_F^{e^+}(t) = N_0 e^{-\alpha t} [1 + a P_F(t)]
\]

and the backward detector \((\theta = 180^\circ)\)

\[
N_B^{e^+}(t) = N_0 e^{-\alpha t} [1 + a P_B(t)]
\]

(4.15)

Using the property that \(P_F(t) = -P_B(t)\) we arrive at the ratio

\[
R(t) = \frac{N_F^{e^+}(t) - N_B^{e^+}(t)}{N_F^{e^+}(t) + N_B^{e^+}(t)} = a P_F(t)
\]  

(4.16)

yielding directly the muon spin depolarization function.

**Different experimental geometries.**

The muon spin precesses in a magnetic field. Two experimental geometries can be used. Either, the applied field is perpendicular or, it is parallel to the initial beam polarization, the **transverse** and **longitudinal** field geometry, respectively.

**Transverse geometry.**

In the **transverse** geometry all the muons experience the same external field perpendicular to the initial polarisation. This is often called muon spin **rotation**. The muon precesses in a transverse field. One measures the Larmor angular frequency

\[
\omega = \gamma \mu_0 H
\]

(4.17)

in which \(\mu_0 H\) is the field experienced by the muon at its position and \(\gamma\) is the muon gyromagnetic ratio. Due to this field the whole angular distribution pattern (eq.4.1) rotates with \(\omega\) around \(\mu_0 H\), leading to a periodic modulation of \(N(t)\). In zero field, and also in the longitudinal mode, there is no angular frequency \(\omega\) since there is no precession.

In equation (4.1) we have assumed that all implanted muons feel the same external magnetic field. Since all muons penetrate the sample with an identical spin orientation, the spin ensemble precesses in phase with the unique angular frequency \(\omega\) during its entire lifetime. In that case no
depolarization (or dephasing) is expected. Remark that in the DC $\mu^+$ beam the muon ensemble never exists simultaneously in time as there is always only one muon present at the time in the sample. Now, we want to take into account the microscopic environment of the muon. The atoms of the sample will always have some dipole moment, which gives rise to a very small field superimposed to the external field. Suppose the orientation of the magnetic moments is random, then this extra field adds or opposes the external field depending on the muon site and location. Although this extra field maybe three orders of magnitude smaller than the applied field it has a profound effect on the $\mu^+$SR signal. It results in a distribution of frequencies and one observes a progressive dephasing of the muon spin ensemble, the oscillations are damped. This is a static origin of dephasing. An other reason for the depolarization of the muon spins can be of dynamic origin. Magnetic materials produce strong magnetic moments at the muon sites. A slow relaxation of these moments during the muon lifetime adds a fluctuation to the total magnetic field and leads to a depolarization of the muon spins. Another dynamic effect is observed when the muon diffuses through the material, jumps from site to site during its lifetime. The muon feels then different local fields and depolarization has to be expected. If the precession time is longer than the average time a muon stays at one site, then the muon will feel only a mean field and the dephasing is diminished. So in the dynamic approach the $\mu^+$SR depolarization function $P(t)$ will be damped. The exponential damping can be presented as

$$P(t) = e^{-\lambda t}$$  \hspace{1cm} (4.18)

with $\lambda$ the damping rate.

In the static case the depolarization is assumed to result from a continuous and isotropic gaussian distribution of static fields with a second moment $\Delta^2/\gamma^2_\mu$ along the external field

$$P(\vec{B}, t) = \frac{\gamma_\mu}{\sqrt{2\pi}\Delta_\chi} \exp \left( - \frac{\gamma^2_\mu B^2 x}{2\Delta_\chi^2} \right)$$  \hspace{1cm} (4.19)

and the depolarization function takes the form

$$P_x(t) = \exp (-\frac{1}{2} \Delta^2_\chi t^2)$$  \hspace{1cm} (4.20)

In the dynamic case a random fluctuation of the local fields as seen by the muon is assumed. Now the shape of the depolarization function depends on the fluctuation rate, on the correlation time $\tau_c$ of the fluctuation compared with the field distribution. In the case of rapid fluctuations ($\tau_c \ll 1$) a Lorentzian form is assumed and the depolarization function can be written as

$$P_x(t) = \exp (-\frac{1}{2} \Delta^2_\chi \tau_c t)$$  \hspace{1cm} (4.21)

**Longitudinal geometry.**

In the longitudinal geometry the applied field is along the beam direction. That means that the applied magnetic field is parallel to the initial muon polarisation. This configuration is known as muon spin relaxation. In contrast to the transverse geometry the longitudinal geometry allows a broad spectrum of applied magnetic fields, from 0.01 mT to fields that exceed the now possible fields. The exponential damping can be measured in the longitudinal geometry. At zero external magnetic field and a gaussian distribution one obtains the formula for the depolarization

$$RNi_5 \text{ compound}$$
\[ P_{\tau}(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2) \exp(-\frac{1}{2} \Delta^2 t^2) \]

For small \( t \) values \( P_{\tau}(t) \) follows a gaussian curve \( e^{-\Delta^2 t^2} \), \( P_{\tau}(t) \) drops to a minimum at \( t \Delta^2 = 1+(1+\Delta^2)^2 \) and shows later recovery to \( 1/3 \). This can be understood intuitively if one considers the random distribution of internal fields which implies that on the average \( 1/3 \) of the \( \mu^+ \) will not precess since for them the internal field and the initial polarisation are parallel.

In case of fast electron spin dynamics (and no diffusion) above the magnetic ordering temperature, \( P_{\tau}(t) \) has the same form as \( P_{\tau}(t) \).

4.4.4 Experimental muon set-ups

\( \mu^+ \)SR spectroscopy can be performed in Canada (TRIUMF at Vancouver), Japan (KEK/BOOM at Tokyo), Russia (at Dubna) and two laboratories in Western Europe. One of these laboratories is the Rutherford Appleton Laboratory (RAL), located at Chilton near Oxford in the United Kingdom. Its muon facility shares the proton accelerator with the neutron spallation source called ISIS. The other European muon source is located at the Paul Scherrer Institute (PSI) at Villigen Switzerland. We have performed measurements at ISIS and PSI.

ISIS produces a double pulse of protons at a repetition rate of 50 Hz. In the double pulse, each pulse is 70 ns wide (FWHM) and the centres of the pulses are separated by 325 ns. Only a fraction (a few percent) of the proton beam necessary to produce neutrons is used for the production of muons. Graphite targets inserted in the proton beam before the main (neutron production) target generate muon pulses with similar time structure as the main proton beam. The muons are guided by the magnetic fields in the beam-line to three experimental areas where they are implanted in the material for muon spectroscopic studies. Two of these areas contain scheduled instruments. MuSR(from 1987) and EMU(from 1993). The third area is used as a test beam. MuSR and EMU have similar performance except that EMU is optimised for longitudinal magnetic field studies and is not available for transverse fields. As the source is pulsed, there is no background during the measurements, when the muons are decaying. Hence, especially low damping rates between 0.005 MHz and 10 MHz can be measured in the longitudinal mode.

In PSI a continuous source produces muons. With this conventional continuous source higher damping rates can be determined. Frequencies (hyperfine fields) between 0.1 MHz and 60 MHz can be measured.

4.5 Inelastic Neutron Scattering measurements

Neutron scattering is for us an additional tool to study condensed matter on a microscopic basis. The wave-length of thermal neutrons is comparable to interatomic distances in solids and the energy change of thermal neutrons due to inelastic scattering processes is of the same order of magnitude as the initial energy. These two features together with the fact that the neutron is a neutral particle makes it an excellent measuring probe [Marshall and Lovesey 1971]. In cooperation with P.C.M. Gubbens and C.F. de Vroege of the Technical University of Delft we have measured the inelastic neutron scattering spectrum of polycrystalline ErNi\(_5\) at 20 K and 60 K. A good introduction in the experimental details of inelastic neutron scattering in the study of solid state can be found in Sköld and Price (1986).

In the Interfacultair Reactor Instituut of the University of Delft (IRI), the rotating crystal spectrometer RKS 2 has been used to make time-of-flight (TOF) measurements. A TOF analyser
is suited to measure simultaneously the scattering angle ($\phi$) and the energy transfer ($\hbar\phi$) of the monochromatic pulse-train of neutrons. A large, rotating, single crystal is used to monochromate, or select neutrons of a particular wavelength, and simultaneously pulsate the neutron beam. The reflected mean wave length depends on the scattering angle $2\theta$ and monochromator crystal plane spacing $d$ according to Bragg's law, $\lambda = 2d \sin \theta$. Before and after the rotating crystal the beam is collimated. Higher-order reflections are suppressed by two choppers in phase with the rotating crystal (see fig 4.10.). Additional filters remove unwanted orders.

**Table 4.2** Instrument parameters of the rotating crystal spectrometer RKS2 at IRI

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>incident wave length</td>
<td>$0.07 \leq \lambda_0 \leq 0.35$ nm or $5660 \geq v \geq 1130$ ms$^{-1}$</td>
</tr>
<tr>
<td>incident energy</td>
<td>$9 \leq E_0 \leq 170$ meV</td>
</tr>
<tr>
<td>beam surface</td>
<td>$25 \times 100$ mm</td>
</tr>
<tr>
<td>scattering angle</td>
<td>$(2\theta_B - 90^\circ) \leq \phi \leq 2\theta_B$</td>
</tr>
<tr>
<td>detectors</td>
<td>$83 \times ^3$He detectors</td>
</tr>
<tr>
<td>time of flight resolution</td>
<td>$0.02 \leq \Delta t/t \leq 0.05$</td>
</tr>
</tbody>
</table>

**Figure 4.10** Sketch of the rotating crystal spectrometer RKS 2 of the Interfacultair Reactor Institute (IRI). A monochromatic pulsed bundle is created by Bragg scattering at a rotating single-crystal. Two additional single-crystal choppers, rotating in phase with the main rotating crystal, suppress higher order reflections and unwanted other reflections. An extra background reduction is achieved by additional silicon single-crystal filters.

$\text{RNi}_5$ compound