Superfluid helium and cryogenic noble gases as stopping media for ion catchers

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2008

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Experimental techniques

The experimental setup for a cryogenic ion catcher consists mainly of an experimental cell and a cryostat which cools down this cell to the desired temperature. Buffer gas can be let in to the experimental cell independently and it can be filled with the desired buffer gas in order to thermalize and transport ions. In our measurements we used helium (in its cryogenic gas phase and its superfluid phase), argon and neon (in their cryogenic gas phase).

4.1 Cryostat

The cryostat used in this project is of “bath cryostat” type which is designed to contain a sufficient supply of cryogens for a convenient duration of operation without refilling (see Figure 4.1). The refilling interval is determined mainly by factors such as the inherent heat load of the cryostat, the heat load from the electric cabling and the cooling power needed for the experimental cell. The main components of the cryostat are the outer vacuum chamber, the liquid nitrogen reservoir, the liquid helium reservoir, the 77 K heat shield, the 4 K heat shield and the 1 K pot. Most parts of the cryostat except the 1 K pot and the heat shields are made of stainless steel because of its very low heat conductance. The heat shields and 1 K pot are made of copper for a good heat conductance with the cryogen reservoirs and are gold-plated for reflection of heat radiation. The cryostat was custom designed and constructed by Vacuum Specials B.V. (Woerden, Netherlands).

4.1.1 Outer vacuum chamber

The outer vacuum chamber (see Figure 4.1) is at room temperature and its main purpose is to provide vacuum for thermal insulation of the whole system. A vacuum
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Buffer gas line
Outer vacuum chamber
Liquid nitrogen reservoir
Liquid helium reservoir
1 K pot
Experimental cell
4 K heat shield
77 K heat shield
Accelerator beam line port

Figure 4.1: Cryostat.
of $\sim 10^{-7}$ mbar is reached after filling with cryogens. The outer vacuum chamber consists of two parts: (1) the top plate which contains the inlet and outlet ports for the cryogens, the electric cabling ports and the buffer gas port for the experimental cell and (2) the two-piece vacuum chamber, which consists of a cylindrical upper part and a cup-shaped lower part. It is possible to remove the lower part of the chamber or the whole chamber. There are four ports welded on the lower part of the chamber in which three act as vacuum accessory ports (pressure gauges, pumps, venting valves) and the fourth as accelerator beam inlet. All three parts are assembled with rubber O-ring seals and clamps.

### 4.1.2 Liquid nitrogen reservoir

The liquid nitrogen reservoir is an annular stainless steel vessel which can store 7.5 liters (Figure 4.1). This reservoir together with a gold-plated copper heat shield attached to it acts as a heat shield for the inner 4.2 K region. There are three stainless steel tubes connecting the liquid nitrogen reservoir and the top plate of the outer vacuum chamber. These are used as the inlet and outlet ports for the liquid nitrogen. The liquid nitrogen reservoir is wrapped with 25 layers of aluminum-coated polyester foil (NCR2) which acts as super-insulator.

### 4.1.3 Liquid helium reservoir

The liquid helium reservoir is a cylindrical stainless steel chamber which can hold up to 6 liters (Figure 4.1). It is connected to the top plate of the outer vacuum chamber with four stainless steel tubes which act as a support for this reservoir. These tubes are the (1) liquid helium inlet, (2) the exhaust, (3) the needle valve which controls the liquid helium flow from the liquid helium reservoir to the 1 K pot and (4) the pumping line to the 1 K pot which passes through the liquid helium reservoir. These four tubes are heat sunk to the liquid nitrogen reservoir. The liquid helium reservoir is wrapped with 5 layers of aluminum-coated polyester foil (NCR2) as super-insulator. A gold-plated copper heat shield is attached to the liquid helium reservoir to reflect the heat radiation from the 77 K heat shield towards the experimental cell (Figure 4.1).

### 4.1.4 1 K pot

The 1 K pot is a gold-plated copper chamber attached to the liquid helium reservoir with three carbon fiber bars (Figure 4.1). It is the part of the cryostat system where the lowest temperature is attained. Liquid helium can be let in to the 1 K pot from the liquid helium reservoir via a needle valve and can be pumped away through a pumping line which passes through the liquid helium reservoir (Figure 4.1). The 1 K pot is cooled by lowering the vapor pressure inside it. A roots pump with $500 \text{ m}^3 \text{ h}^{-1}$ pumping capacity is employed to pump on the 1 K pot. The pumping
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is controlled by a manual valve on top of the pump. The lowest temperature attained by this cryostat is 0.92 K; the lowest long term stable temperature achieved is $\sim 1.00$ K. The experimental cell is attached to the 1 K pot with an indium seal and screws (see Figure 4.2a). A steel capillary which acts as buffer gas inlet to the experimental cell passes alongside the liquid helium reservoir and through the 1 K pot to pre-cool the buffer gas before it enters the experimental cell (Figure 4.1).

4.1.5 Electric cabling

Twelve cryogenic coaxial cables (Type SR - Lake Shore Cryotronics) and 16 0.1 mm diameter twisted pair insulated copper wires connect the experimental cell to the outside world. These cables start from vacuum feed-throughs on the top plate and are heat sunk on different temperature stages before they reach the low temperature region. The coaxial cables go all the way to the experimental cell. The twisted pair insulated copper wires end on sockets attached to the bottom of the liquid helium reservoir. Connections from these 4.2 K sockets to the 1 K pot sockets are made with thin twisted pair insulated manganin wires. The temperature sensors and the heater are connected with the temperature controller using these 1 K pot sockets.

4.1.6 Helium consumption

The liquid helium consumption of the cryostat is due to the evaporation in the liquid helium reservoir because of the heat load from the 80 K reservoir by radiation and conduction via ports and because of evaporation in the 1 K pot needed to maintain the temperature of the experimental cell. The heat load due to radiation loss $Q_{\text{rad}}$ is given by

$$Q_{\text{rad}} = A_s \times q_{\text{rad}} \times E \times \left( \frac{1}{1 + n} \right),$$

(4.1)

where $A_s$ is the size of the radiating surface (m$^2$), $q_{\text{rad}}$ is the black-body radiation heat load per unit area (2 W m$^{-2}$ between 80 K and 4.2 K and 20 $\mu$W m$^{-2}$ between 4.2 K and 1 K), $E$ is the emissivity of the surface 0.1 (for a polished metal surface) and $n$ is the number of layers of super-insulation. The heat load due to conduction loss $Q_{\text{con}}$ is

$$Q_{\text{con}} = \frac{A_c \times q_{\text{con}}}{L},$$

(4.2)

where $A_c$ is the cross-sectional area of the conducting material, $q_{\text{con}}$ the conduction per unit length (0.317 W mm$^{-1}$ between 80 K and 4.2 K and $0.6 \times 10^{-3}$ W mm$^{-1}$ between 4 K and 1 K, for stainless steel), and $L$ the length of the conducting material.

The total heat load on the liquid helium reservoir due to radiation is 0.033 W and due to conduction is 0.15 W. The radiation heat load on the 1 K pot and experimental cell are very small (2 $\mu$W) because they are facing the 4.2 K shield. The conduction heat load is contributed by the helium input line to the experimental
4.2 Experimental cell

The experimental cell is made of a copper cylinder. Its top part is attached to the 1 K pot. The bottom is closed with a copper plate with an indium seal (see Figure 4.2a). Electric connections to the experimental cell are made via stycast feed-throughs in the bottom plate. A steel capillary running from the cryostat top plate to the experimental cell via the 1 K pot acts as a pumping port and a buffer gas inlet for the experimental cell. A cernox temperature sensor is attached to the bottom plate in order to achieve an accurate temperature control of the experimental cell.

Stycast rods and spacers are used to build up the electrode configuration inside
the experimental cell. A radioactive ion source, ion trajectory manipulation electrodes, an aluminum ion catcher foil and an α-particle detector are assembled in cylindrical symmetry (see Figures 4.2b and 4.2c).

Once cooled down to the required temperature, helium gas can be let into the experimental cell to have cryogenic gas or superfluid helium as stopping medium for high-energy radioactive ions. Thermalised ions can be transported to the aluminum catcher foil using an electric field provided by the set of guiding electrodes (see Figure 4.2d).

4.2.1 Radioactive source

Many characteristics of ion survival in a stopping medium can be studied using an “open” α-decaying radioactive source; i.e. a thin source without protective layer such that α-decay products can recoil out of the source.

Advantages of this type of studies are: (1) no external high-energy ion beam is required as the α-decay recoil ions simulate an ion beam; (2) absolute efficiencies can be easily determined as the primary source strength is readily measured.

In this work, $^{223}$Ra sources, giving 120 keV $^{219}$Rn recoil ions, were used. We detect the $^{219}$Rn ions via their characteristic α decay. This radioactive decay detection is essentially background-free and sensitive to the decay of a single atom/ion, resulting in very precise measurements over a large dynamic range. $^{223}$Ra sources are prepared from an open $^{227}$Ac source as described in [59]. Details of the decay chain of $^{227}$Ac are given in Figure 4.3 and Table 4.1. A schematic picture of the source preparation set-up is shown in Figure 4.4. The $^{227}$Ac source is placed at the bottom of a chamber filled with $\sim 30$ mbar helium gas. A copper screw is placed a few centimeters above at a negative potential (-150 to -300 V) relative to the $^{227}$Ac source.

$^{223}$Ra ions, recoiling from the $^{227}$Ac source following the α decay of $^{227}$Th, are stopped in the helium gas and transported by the electric field onto the tip of the copper screw. In order to confine the deposition of $^{223}$Ra, a plastic tape is wrapped around the side of the copper screw so that only the tip of the copper screw is visible from the $^{227}$Ac source.

In the first phase of this project, $^{223}$Ra sources where provided by the Department of Physics, University of Jyväskylä, Finland and the Nuclear Physics Institute, University of Mainz, Germany. In Spring 2007 we obtained a 500 kBq $^{227}$Ac source from the International Research Center for Nuclear Material Science, Tohoku University, Japan and set up our own $^{223}$Ra source preparation unit. Taking care of the cleanliness of the system, the maximum recoil collection efficiency of 30% is obtained; resulting in a saturated $^{223}$Ra activity of 75 kBq and thus a $^{219}$Rn recoil ion source strength of 37 500 s$^{-1}$.

In our experiments we mainly consider the α-line from the $^{219}$Rn decay because this isotope is the primary recoil ion from the $^{225}$Ra source. When using an α-decay recoil source, the number of recoils entering the stopping volume and the location
Figure 4.2: (a) Inside view of the cryostat with cell mounted on the 1 K pot. (b) Photograph of the electrode assembly. (c) Cut-away view of the inside structure of the experimental cell. (d) Cross-sectional view of the electrode assembly. Decreasing voltages on the bottom electrode/source, guiding electrodes and catcher foil create the electric field that transports ions from close to the source onto the catcher foil. Dark gray lines indicate the ion trajectories and light gray lines indicate the equipotential lines from ion optics simulations with SIMION [30].
from which they originate is known accurately for the first daughter in the decay chain only. The measured $\alpha$ particle spectra also contains spectral lines from $^{215}\text{Po}$ and $^{211}\text{Bi}$, the other $\alpha$ emitters in the $^{223}\text{Ra}$ decay chain. However, $^{215}\text{Po}$ and $^{211}\text{Bi}$ are difficult to interpret. Being the granddaughter of the $^{223}\text{Ra}$ source, not all $^{215}\text{Po}$ nuclei originate from the source, but from all the places where $^{219}\text{Rn}$ has ended up. Because its half-life of 1.78 ms is comparable to the transport time, it can also decay along its path. The origin of $^{211}\text{Bi}$ recoil ions, being three $\alpha$ decays away from $^{223}\text{Ra}$, is even more uncertain than that of $^{215}\text{Po}$. Moreover, $^{211}\text{Bi}$ is reached via the $\beta$-decay of $^{211}\text{Pb}$ that has a half-life of 36 min; longer than our typical measurement time for a certain setting. This prevents us to deduce any relevant information from the $^{211}\text{Bi}$ line intensities.

### 4.2.2 Electronics and Data Acquisition

The raw data in our experiments are $\alpha$ particle energy spectra. Silicon detectors are used as $\alpha$-energy spectrometers. They function perfectly down to $\sim 1$ K. The silicon detector is a thin slice of boron implanted n-type silicon, forming a rectifying p-n junction. A reverse bias voltage of 50 to 100 V to the p-n junction produces a depletion layer about 100 $\mu$m thick. The electron-hole pairs formed by ionisation when a charged particle traverses the layer are picked up by the electrodes and put in to a charge sensitive preamplifier. The output of the preamplifier is fed into a linear amplifier which amplifies and shapes the pulse, increasing the signal-to-noise ratio in order to optimize the energy resolution. The signal from the linear amplifier is fed into an ADC (analog to digital converter) and then to a MCA (multichannel analyzer) to convert the analog pulses to digital values and add the numerical values in corresponding channel of the spectrum.

### 4.2.3 Spectrum analysis

The experimental data in our measurements are the $\alpha$-decay spectrum of the daughter nuclei of $^{223}\text{Ra}$ collected on the aluminum catcher foil and from the superfluid-vapor interface or the $\alpha$-recoil source (depending on the experimental situation). Thus our typical spectra consist of 2 sets of lines, one from decay on the aluminum catcher foil and an other from the decay at the source or at the interface. Each set of lines consists of 6 lines, three $^{219}\text{Rn}$ lines, two $^{211}\text{Bi}$ lines and a $^{215}\text{Po}$ lines (see Table 4.1).

$$f(x) = \begin{cases} 
A \exp \left[ \frac{dx}{2} \left( \frac{2(x-x_0)+dx}{\sigma^2} \right) \right] & \text{if } x < (x_0 - dx) \\
A \exp \left[ - \frac{1}{2} \left( \frac{x-x_0}{\sigma} \right)^2 \right] & \text{if } x > (x_0 - dx)
\end{cases}$$

In this representation an individual line is defined by four parameters: the position of the line maximum $x_0$, the peak height $A$, the width of the Gaussian $\sigma$ and the dis-
4.2 Experimental cell

Figure 4.3: Decay chain of $^{227}$Ac.

Table 4.1: Isotopes in the $^{223}$Ra decay chain and their properties.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay mode</th>
<th>Notation used</th>
<th>Half life</th>
<th>Energy [keV]</th>
<th>Branching Ratio [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{223}$Ra</td>
<td>α</td>
<td></td>
<td>11.435(4) d</td>
<td>5539.8(9)</td>
<td>9.20(20)</td>
</tr>
<tr>
<td></td>
<td>α</td>
<td></td>
<td></td>
<td>5606.7(3)</td>
<td>25.7(5)</td>
</tr>
<tr>
<td></td>
<td>α</td>
<td></td>
<td></td>
<td>5716.2(3)</td>
<td>52.6(13)</td>
</tr>
<tr>
<td></td>
<td>α</td>
<td></td>
<td></td>
<td>5747.0(4)</td>
<td>9.20(20)</td>
</tr>
<tr>
<td>$^{219}$Rn</td>
<td>α</td>
<td>Rn III</td>
<td>3.96(1) s</td>
<td>6425.0(10)</td>
<td>7.5(6)</td>
</tr>
<tr>
<td></td>
<td>α</td>
<td>Rn II</td>
<td></td>
<td>6552.6(10)</td>
<td>12.9(6)</td>
</tr>
<tr>
<td></td>
<td>α</td>
<td>Rn I</td>
<td></td>
<td>6819.1(3)</td>
<td>79.4(10)</td>
</tr>
<tr>
<td>$^{215}$Po</td>
<td>α</td>
<td>Po</td>
<td>1.781(4) ms</td>
<td>7386.1(8)</td>
<td>99.99977(2)</td>
</tr>
<tr>
<td>$^{211}$Pb</td>
<td>β⁻</td>
<td></td>
<td>36.1(1) min</td>
<td>1373 (end-point)</td>
<td>100</td>
</tr>
<tr>
<td>$^{211}$Bi</td>
<td>α</td>
<td>Bi II</td>
<td>2.14(2) min</td>
<td>6278.2(7)</td>
<td>16.19(14)</td>
</tr>
<tr>
<td></td>
<td>α</td>
<td>Bi I</td>
<td></td>
<td>6622.9(6)</td>
<td>83.54(14)</td>
</tr>
</tbody>
</table>
Experimental techniques

Figure 4.4: Schematic diagram of $^{223}\text{Rn}$ source preparation setup.

tance from the position of the line maximum $x_0$ to the junction point $dx$ (Figure 4.7) [91]. The two sets are either mingled or separated depending on the difference in energy loss between $\alpha$-emission from the source or the superfluid helium-vapor interface and the foil. The energy loss depends on the helium gas density and the distance travelled.

Table 4.2 shows an example of the typical energy loss experienced by alpha particles in our experiment. Figure 4.5 shows the channel number of identifiable peaks as function of their emitted energies. A linear relation is observed between the channel numbers and the $\alpha$ energies of an individual set. Two linear fits are obtained for the two sets of lines. Thus four parameters, the slope and offset of each linear fit can describe the positions of all 12 $\alpha$ peaks. The offset of the fit corresponds to the energy loss in the buffer gas and the aluminum catcher foil. The slope of the fit corresponds to the energy per channel.

Analyzing an $\alpha$-decay spectrum involves the deduction of the total number of $\alpha$ counts contributed by the decay of the individual species. When determining the areas of the individual lines it is necessary to know the correct line shape. To test various functional forms, the intense well-isolated $^{215}\text{Po}$ line is used. When fitted with a Gaussian profile we observe a significant deviation at its low energy side (Figure 4.6). The line shape is accurately described by a combination of a Gaussian with a single exponential tail at the low-energy side which joins the gaussian such
Figure 4.5: A typical energy calibration plot. The channel numbers of identifiable peaks from each set are plotted as function of the emitted $\alpha$-energies. A linear relation between the channel numbers and emitted $\alpha$-energies is observed for an individual set.

Table 4.2: Example of the typical energy loss experienced by alpha particles in our experiments at temperatures 1.2 K and 1.6 K. $\alpha$-particles emitted from the superfluid surface have to travel through 61 mm helium vapor, 0.81 $\mu$m aluminum foil and another 3 mm helium vapor. $\alpha$-particles emitted from the foil have to travel through 0.81 $\mu$m aluminum foil and 3 mm helium vapor. $E_\alpha$ is the emitted $\alpha$ energy, $E_s^\alpha$ is the detected energy of $\alpha$ particles emitted from the superfluid surface, $E_f^\alpha$ is the detected energy of the $\alpha$ particles emitted from the catcher foil and $\Delta E$ is the energy difference between $\alpha$-particles emitted from catcher foil and superfluid surface. $\Delta E$ at 1.6 K is larger than $\Delta E$ at 1.2 K due to the larger vapor pressure at 1.6 K.

<table>
<thead>
<tr>
<th>$\alpha$</th>
<th>$E_\alpha$ [keV]</th>
<th>$E_s^\alpha$ [keV]</th>
<th>$E_f^\alpha$ [keV]</th>
<th>$\Delta E$ [keV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{219}$Rn</td>
<td>6819</td>
<td>6540</td>
<td>6702</td>
<td>162</td>
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<td>$^{219}$Rn</td>
<td>6553</td>
<td>6258</td>
<td>6432</td>
<td>174</td>
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<td>6425</td>
<td>6123</td>
<td>6303</td>
<td>180</td>
</tr>
<tr>
<td>$^{215}$Po</td>
<td>7386</td>
<td>7120</td>
<td>7276</td>
<td>156</td>
</tr>
<tr>
<td>$^{211}$Bi</td>
<td>6623</td>
<td>6333</td>
<td>6503</td>
<td>170</td>
</tr>
<tr>
<td>$^{211}$Bi</td>
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<table>
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<th>$E_s^\alpha$ [keV]</th>
<th>$E_f^\alpha$ [keV]</th>
<th>$\Delta E$ [keV]</th>
</tr>
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<tr>
<td>$^{219}$Rn</td>
<td>6819</td>
<td>5416</td>
<td>6654</td>
<td>1238</td>
</tr>
<tr>
<td>$^{219}$Rn</td>
<td>6553</td>
<td>5092</td>
<td>6383</td>
<td>1291</td>
</tr>
<tr>
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<td>$^{215}$Po</td>
<td>7386</td>
<td>6091</td>
<td>7232</td>
<td>1141</td>
</tr>
<tr>
<td>$^{211}$Bi</td>
<td>6623</td>
<td>5178</td>
<td>6454</td>
<td>1276</td>
</tr>
<tr>
<td>$^{211}$Bi</td>
<td>6279</td>
<td>4751</td>
<td>6104</td>
<td>1353</td>
</tr>
</tbody>
</table>
Figure 4.6: A $^{215}$Po $\alpha$-decay line fitted with a Gaussian line shape and a Gaussian with low-energy exponential tail (see Equation 4.3).

Figure 4.7: Gaussian with low-energy exponential tail for different distances $dx$ from its center to the start of its exponential tail. The low-energy tail arises from the energy loss due to multiple scattering.
Figure 4.8: A typical α-particle energy spectrum and its analysis. The solid line in (a) shows the measured spectrum and the fitted spectrum is overlayed in gray. The individual components resulting from the fit for the three α-decaying isotopes originating from both the foil (vertical lines) and the source (black) are shown in the lower parts. Panel (b) shows the residual/sigma of the fit. Here sigma is $\sqrt{\text{counts}}$. (c) shows the histogram representation of the residual/sigma of the fit; it follows a normal distribution with a standard deviation $\sim 1$ and mean $\sim 0$. This shows that our fitting function is a perfect representation of the spectra.
that the function and its first derivative are continuous \[91\]: The total intensity of such a Gaussian-exponential pulse is

\[
I = A \left[ \int_{-\infty}^{x_0 - dx} e^{-\frac{1}{2} \left( \frac{2(t-x_0+dx)}{\sigma^2} \right)} dt + \int_{(x_0 - dx)}^{\infty} e^{-\frac{1}{2} \left( \frac{t-x_0}{\sigma} \right)^2} dt \right].
\] (4.4)

Figure 4.6 shows the fit of such a function to a single \(^{219}\)Po \(\alpha\)-line. We need 48 parameters to describe the total spectrum containing 12 lines (12 for peak positions, 24 for individual line shapes and 12 for individual intensities). As mentioned before only 4 energy calibration parameters (2 for the interface or source and 2 for the aluminum foil) can describe all the peak positions. This reduces the number of free parameters from 48 to 40. Further, by taking the branching ratios of the \(^{219}\)Rn and \(^{211}\)Bi decays (see Table 4.1) into account, the intensities of 6 lines from the same location can be represented by 3 parameters. So for two \(\alpha\)-decay locations 6 parameters describe the intensities of the 12 \(\alpha\)-lines. This reduces the number of free parameters further from 40 to 34. It is reasonable to assume that the \(\alpha\)-lines belonging to the different decay branches of the same isotope decaying at the same position have the same line shape, \(\sigma\) and \(dx\). Thus with 12 shape parameters (6 for the \(\alpha\)-particles from source or interface and 6 for the \(\alpha\)-particles from the catcher foil) we can represent the line shapes of all 12 lines and the number of free parameters is finally reduced to 22.

We use a \(\chi^2\) minimization technique in our fitting procedure. The IGOR Pro 6 data analyzing program \[127\] is used to carry out the fitting procedure. The initial values for different parameters are deduced from strong, well-defined lines in the spectrum. Some of the parameters are fixed and others let free to iterate until the \(\chi^2\) minimized values are attained. The number of fixed parameters is then reduced in a step by step manner. For low-statistics spectra we have to take the same shape parameters for every line in each set. Here we assume that the \(\alpha\)-particles emitted from the same region but from different isotopes result in the same line shape. This is often necessary only for \(^{215}\)Po and \(^{219}\)Rn. A typical example of the result of the fitting procedure is shown in Figure 4.8.

### 4.3 Gas density in the experimental cell at low temperature

For the gas catcher measurements the temperature and density are the relevant gas parameters. In practice the temperature and pressure are measured and the density is calculated using the ideal gas law. In the setup used in this work the temperature of the experimental cell is measured directly with sensors attached to
the cell and 1 K pot. The pressure in the cell is however not measured directly. A pressure gauge is mounted at the end of the filling capillary on top of the cryostat. The pressure gauge is thus at room temperature and measures the pressure of the room-temperature end of the filling capillary. In such a situation, i.e. where two volumes at different temperatures are connected, one cannot simply combine the temperature measurement of the volume at one temperature with the pressure measurement of the volume at the other temperature to obtain the density in either volume. This situation is investigated as follows. The cell was filled with gas at room temperature and then closed. The ratio of the pressure of the warm volume at room temperature \( P_w \) to the temperature of the cold volume in the experimental cell \( T_c \) was followed during cooling down. Many such data sets for different gases, starting pressures and sizes of the warm volume are shown in Figure 4.9, where the \( \frac{P_w}{T_c} \) ratio is normalised to the ratio \( \frac{P_0}{T_0} \) before cooling down starts. In the absence of the effect discussed here the ideal gas law predicts that

\[
\frac{P_w}{T_c} = \frac{P_0}{T_0} = 1
\]  

at all temperatures although pressure and temperature are measured at different places. We see a clear deviation from this. The effect does not depend on the type of gas nor the pressure at room temperature within the rather limited range studied here.

The deviation is larger if the warm volume \( V_w \) is bigger relative to the cold volume \( V_c \). The density in the cold volume can be determined from the measured \( T_c \) and \( P_w \). A cold and a warm volume \( V_c \) and \( V_w \) are connected with a capillary of negligible volume. In the setup the capillary to cell volume ratio is a few \( 10^{-3} \). With everything at room temperature \( T_0 \), the ideal gas law holds for the whole system

\[
P_0(V_c + V_w) = nRT_0 ,
\]

where \( P_0 \) is the pressure at room temperature, \( n \) is the amount of gas (in mole) in the system and \( R = 8.31 \text{ J mole}^{-1} \text{ K}^{-1} \) is the universal gas constant. During the experiments the volume \( V_c \) is cooled down to a temperature \( T_c \), the volume \( V_w \) stays at room temperature \( T_0 \). The pressures in both volumes are \( P_c \) and \( P_w \) respectively. This gives the ideal gas law equations in the separate volumes

\[
P_cV_c = n_cRT_c ,
\]

\[
P_wV_w = n_wRT_0 ,
\]

with the total amount of gas unchanged

\[
n = n_c + n_w ,
\]
Experimental techniques

Figure 4.9: Ratio of the warm volume pressure ($P_w$) to the cold volume temperature ($T_c$) as a function of $T_c$ during cooling down, normalised to the ratio before cooling down starts ($P_0/T_0$). Data sets for helium and argon at different starting pressures $P_0$ and warm to cold volume ratios $V_w/V_c$ are shown.

where $n_c$ and $n_w$ are the amount of gas (in mole) in the cold and warm volumes. Substituting Equation 4.7 and Equation 4.8 in Equation 4.6 gives

$$P_c = \frac{T_c P_0}{T_0} \left( 1 + \left( 1 - \frac{P_w}{P_0} \right) \frac{V_w}{V_c} \right).$$

(4.10)

For $V_w \rightarrow 0$, Equation 4.10 reverts to what is expected from the ideal gas law: $P_c/T_c$ is constant during cooling down. For non-negligible value of $V_w/V_c$, both $P_w$ and $P_c$ are not what is expected from the ideal gas law. $P_c$ can be calculated from $P_w$, $T_c$ and $V_w/V_c$ and the density in the cold volume can be determined using the ideal gas law applied to this volume

$$\frac{n_c}{V_c} = \frac{P_c}{RT_c}.$$  

(4.11)

The density of the warm volume can be calculated in an analogous manner. Figure 4.10 shows the densities of the cold and warm volumes relative to the density at room temperature for the same measurements represented in Figure 4.9. The density in the warm volume decreases and the density of the cold volume increases. This relationship is plotted as a function of $V_w/V_c$ for a cold volume temperature
4.3 Gas density in the experimental cell at low temperature

Figure 4.10: Density relative to the room temperature value in the warm and cold volumes as a function of the cold volume temperature $T_c$ for different warm to cold volume ratios $V_w/V_c$. The cold volume relative density is extrapolated to 5 K.

Figure 4.11: Cold volume density $n_c/V_c$ relative to its room temperature value at a cold volume temperature $T_c$ of 78 K as a function of warm to cold volume ratio $V_w/V_c$. The result of a linear fit with a fixed offset of $(n_c/V_c)_\text{rel} = 1$ is indicated.
of 78 K in Figure 4.11. Equation 4.11 and 4.10 are used to calculate the density in the cell from the measured values $T_c$, $P_w$ and the known volume ratio $V_w/V_c$.

In summary: when cooling down, the pressure in the warm volume decreases less than the temperature (see Figure 4.9). At the same time, the higher pressure in the warm volume causes the cold volume density to increase (see Figure 4.10). Because these two effects are of similar size, the error made in calculating the cold volume density using the warm volume pressure is small. For $V_w/V_c < 0.5$, this error is less than 5%. In all measurements except the mobility measurements, $V_w/V_c$ was 0.18 or 0.065 and the effect described in this section is not corrected for. The correction is performed for the mobility measurements of $^{219}$Rn ions in noble gases where $V_w/V_c = 0.55$ (see Section 5.3.1).

4.4 Conclusion

An experimental apparatus was designed and constructed in order to perform the experiments outlined later in this thesis. The apparatus has two major parts, the cryostat and an experimental cell. The cryostat is able to stabilize the temperature of the helium cell between 1 K and room temperature. The electrode configuration in the experimental cell is optimized based on the results from ion-optical simulations. An $\alpha$-recoil ion source is an excellent choice to probe the transport and extraction efficiencies of ions in a stopping medium since radioactive decay detection is sensitive to a single atom or ion. This technique provides a near zero background level. The line shape of the $\alpha$-decay is accurately described by a combination of a Gaussian with a single exponential tail at the low energy side. By considering the branching ratios of $\alpha$-decays, assuming the same line shape per isotope and location and using energy calibrations of the observed lines, the number of free parameters required to represent the line shapes of all 12 lines is reduced from 48 to 22.