Phase Transitions in Quantum Crystals

On Growth of Magnetically Ordered $^3$He
and a Search for Supersolid $^4$He
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On Growth of Magnetically Ordered $^3$He and a Search for Supersolid $^4$He

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Chapter 1

General Introduction
Abstract

This chapter gives a very general overview about the importance of low temperatures and $^3$He in modern physics as well as the motivation which turned into the realization of this thesis. The last section of this chapter is a brief outline of the contents that can be found in the following chapters.
1.1 General Introduction

Many fundamental physical principles, like indistinguishability, the Pauli principle or the Heisenberg uncertainty relation, are manifested most clearly at low temperatures, where the thermal energy of the atoms is comparable to their energy of activation. Few examples are superfluidity of $^4$He below a critical temperature $T_c = 2.17$ K, superfluidity of $^3$He below a critical temperature $T_c = 2.5$ mK, superconductivity in various materials or electronic and nuclear spin ordering. From all chemical elements the two isotopes $^3$He and $^4$He have the lowest boiling point, but also the property of remaining liquid under saturated vapor pressure down to zero temperature. This is a direct consequence of the Heisenberg uncertainty relation, the existence of a confinement results in a finite momentum (due to the small atomic mass, giving a large zero-point energy and the weak interaction between atoms). In order to obtain the solid phase in helium, high pressures (above 25 bars) need to be applied to the system at low temperatures. Helium is also a very pure system at low temperatures because everything else freezes out. Apart from being an impressively remarkable system itself, $^3$He is useful in the study of turbulence and it has interesting analogies with particle physics and cosmology. It has connections with almost any branch of modern theoretical physics.

This thesis concerns the theoretical and experimental study of matter at ultra-low temperatures and the development of techniques to reach and measure at those temperatures. $^3$He is an unique system for studying the growth (and melting) kinetics of crystals. The growth and melting of crystals is accompanied by flows of mass and heat due to the difference in entropy and density of the liquid and solid bulk phases. In ordinary systems these processes govern the growth and melting of the crystals yielding in very slow dynamics. In the case of $^3$He, the melting curve becomes almost flat at ultra-low temperatures because the entropies of the solid and the liquid are almost equal yielding in the vanish of the latent heat. At these temperatures the liquid is a superfluid and the thermal conductivity of the liquid becomes larger. Thus,
the heat generated while growing (or melting) the solid will be carried away very fast and the relaxation times are very short. While other substances need several hours or even days to get into the equilibrium situation in the liquid-solid interface, it takes seconds for $^3$He at very low temperatures. Besides, due to the superfluidity of the liquid, the properties of the interface can be studied since they do not stay masked by the bulk properties of both the liquid and the solid.

1.2 Motivation

Recent experiments on crystal growth and crystal shape in $^3$He at zero magnetic field and $T=0.55$ mK by Tsepelin et al. [1, 2] in Helsinki have revealed many new facets and strong anisotropy in the growth rates. Also experiments by Todoshchenko et al. [3] performed as well in Helsinki revealed that the ratio of growth rates of $<100>$ and $<110>$ facets changes drastically at magnetic ordering in zero field, the so-called Néel temperature ($T_N=0.93$ mK). Very interesting results concerning the growth rate of solid $^3$He have been discovered but still many questions remain unclear. This thesis intends to extend the experimental studies of the shape and growth rate of solid $^3$He to magnetic fields up to $9$ T at temperatures as low as possible.

In the case of $^4$He, a very unusual situation occurs at low temperatures. Due to the superfluid nature of the liquid, the high thermal conductivity of the solid and the small latent heat of solidification, the extremely fast growth rate allows the presence of crystallization waves in the solid/liquid interface below $1$ K [4]. The ultimate goal of the experiments described in this thesis is to observe crystallization waves on solid $^3$He in magnetic fields at ultra-low temperatures (few hundred microkelvins) where the crystallization waves can be excited without excessive damping [5].

1.3 Guideline through this Thesis

Chapter 2 gives an introduction the properties of $^3$He. This is followed by the theory of the crystal growth both from the strong and weak-coupling limits. This chapter ends with a discussion about the equilibrium shape of the crystals.

Chapter 3 introduces the experimental setup. A description of the cryostat (dilution refrigerator and nuclear demagnetization stage) as well as new parts built and assembled in the cryostat needed for the success of the experiments are shown here. At the end, the experimental cell is also presented. These improvements to the cryostat can be also found in the article “Improved Optical Setup to Measure Growth and Melting of Solid $^3$He Below 1 mK in Magnetic Fields up to 9 Tesla” [6].
Chapter 4 presents the experimental method used for the growth of $^3$He crystals and the analyzing techniques followed for the understanding of these measurements. The computer program used for the analysis of the images of the crystals has been published in the paper “Analysis of $^3$He Crystal Images Using a Computer-Generated Wire-Frame” [7]. Results are shown at different magnetic fields and compared with previous results. The last section of the chapter summarizes the results and provides directions for future experiments. All these results can be found in the article “Growth of $^3$He Crystals at Different Magnetic Fields” [8].

Chapter 5 shows the new improvements made on the cryostat after the realization of the experiments described in chapter 4. New heat switches providing better thermal contact between the mixing chamber and the nuclear stage, a more sensitive titanium pressure gauge and an electrically actuated cold valve are presented in this chapter. A paper about the new pressure gauge has been published as “A Small and Very Sensitive Titanium Pressure Gauge for $^3$He Melting Pressure Measurements in Magnetic Field” [9] and another one about the cold valve as “A Magnetically Driven Cold Valve for $^4$He” [10].

Chapter 6 describes the experiments carried out in the Interface Group in the Low Temperature Laboratory in the Helsinki University of Technology during a three months stay. The experiment was about a hot topic at the moment, the possible observation of superfluidity in solid $^4$He. Theory, experiments and discussion are presented in this chapter. These results can be found in the article “Melting Curve of $^4$He: No Sign of the Supersolid Transition down to 10 mK” [11].

1.4 References


Chapter 2

Theory
Abstract

In this chapter an introduction to $^3$He and crystal growth is given. The most relevant properties of $^3$He, both the liquid and the solid phase, as well as their importance for growth of crystals are discussed. Different growth mechanisms and different models explaining the interaction between the solid and liquid interface are presented. The equilibrium shape of the crystals is also presented.
Chapter 2

Theory

2.1 Introduction to $^3$He

Among all the known substances, $^3$He is one of the richest systems with which to do experimental physics. It is a very complex substance, remaining liquid even at zero temperature, showing different types of superfluidity, but also it has connections with almost any branch of modern theoretical physics.

2.1.1 Properties of $^3$He

From all chemical elements $^3$He has the lowest boiling point, but also the property of remaining liquid under saturated vapor pressure down to zero temperature (as well as another stable helium isotope $^4$He). This is due to the small atomic mass, giving a large zero-point energy, and the weak interaction between atoms. $^3$He is also a very pure system at low temperatures because everything else freezes out. Only $^4$He impurities can be present at very low temperatures. In order to obtain the solid phase in $^3$He, due to a large zero point energy, high pressures (between 29 and 35 bars approximately) need to be applied to the system at low temperatures (fig 2.1).

The $^3$He phase diagram has a minimum ($P_{\text{min}}=29.315$ bar, $T_{\text{min}}=319$ mK [1]). The negative slope corresponds to the low temperature side, which is due to the lower entropy of the liquid compared with the solid which is paramagnetic. Making a zoom of the $^3$He phase diagram at very low temperature and high pressure region (exploded view in figure 2.1), a kink in the melting curve can be seen at zero magnetic field. This kink becomes broader when the magnetic field increases. This is the so-called Néel transition. Below this transition solid $^3$He orders magnetically into two different states depending on the magnetic field. Below this point, the melting curve becomes almost flat because the entropies of the solid and the liquid are almost equal and therefore the latent heat vanishes very fast. At low enough temperatures, $^3$He under-
goes a transition into the superfluid phase. The $^3$He atoms have a fermionic behavior, and the pairing can be described in terms of the BCS theory [2]. Superfluidity was first observed by Osheroff et al. [3]. Since the liquid becomes superfluid, the heat generated while growing (or melting) the solid will be carried away very fast and the relaxation times are very short. These conditions cannot be reached with usual crystals and therefore $^3$He is an unique system for studying the growth (and melting) kinetics of magnetically ordered crystals.

### 2.1.2 $^3$He Phase Diagrams in the Presence of a Magnetic Field

Due to its very rich phase diagram, both in the liquid (figure 2.2.a) and solid (figure 2.2.b) state in the presence of an external magnetic field, $^3$He is a very interesting system if magnetic effects want to be studied.
2.1 Introduction to $^3$He

Figure 2.2: a) Phase diagram of liquid $^3$He at melting pressure in the presence of an external magnetic field; b) Phase diagram of solid $^3$He at melting pressure in the presence of an external magnetic field.

The complicated broken symmetry state characterizing the superfluidity leads to the existence of several different superfluid phases. When liquid $^3$He is cooled down and reaches a temperature of approximately 2.5 mK at zero magnetic field, it goes from the normal state to the superfluid $A$-phase. If it is cooled down further, then it goes from the $A$-phase to the $B$-phase at about 1.9 mK. If an external magnetic field is applied, there is another superfluid phase appearing, the so-called $A_1$-phase. More superfluid phases exist when $^3$He is confined into restricted geometries.
On the other hand, when solid $^3$He is cooled down at zero magnetic field, it undergoes a transition into the magnetically ordered state at $T_N=0.93$ mK. It goes from the paramagnetic ($PM$) phase to the so-called $U2D2$ phase. The $U2D2$ phase (or low-field phase) is an antiferromagnetic phase with two planes of spins pointing up and two planes of spins pointing down in sequence. When a magnetic field of about 450 mT is applied, the solid goes from the $U2D2$ to the $CNAF$ (canted normal antiferromagnetic) phase. The $CNAF$ phase (or high-field phase) is a normal antiferromagnetic phase, but with the spins slightly tilted from their normal position with a significant magnetization.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{phase_diagram.png}
\caption{Phase diagrams of liquid and solid $^3$He at melting pressure in the presence of an external magnetic field drawn to the same figure. Black lines denote liquid phases and grey lines solid phases.}
\end{figure}

In figure 2.3 the phase diagrams of liquid and solid $^3$He are plotted one on top of each other. The region around 500 mT is especially interesting because two phase transitions take place as a function of the magnetic field, one in the solid, from the $U2D2$ phase to the $CNAF$ phase, and another in the liquid, from the superfluid $B$-phase to the superfluid $A$-phase. Crystals can be grown from the same phase of the liquid ($B$-phase) into two different phases in the solid ($U2D2$ and $CNAF$), and from the two different superfluid phases ($A$ and $B$-phase) into the $CNAF$ solid phase. Thus the crystals can be grown from the same liquid phase into two different solid phases and from two different liquid phases into the same final solid phase, giving the possibility of studying the properties of both liquid and solid in the growing process.
2.2 Introduction to Crystal Growth

2.2.1 Growth Mechanism

Growing a crystal means that atoms from the liquid have to attach to the nucleation sites in the solid. In order to attach these atoms to the solid, a driving force needs to be applied. This driving force is the difference in the chemical potential between the liquid and the solid. This difference in the chemical potential consists of two terms, one is due to the difference in temperature between the solid and the liquid and the other one is due to the overpressure applied for the growth of the crystal:

\[ \Delta \mu = \Delta \mu_P + \Delta \mu_T. \]  

(2.1)

A scheme about the growth mechanism is shown in figure 2.4.

\[ \Delta \mu = \Delta \mu_P + \Delta \mu_T \]

(2.1)

Figure 2.4: Growth mechanism scheme. See text for details.

However, in the region where the experiments were carried out (between \(T_N\) and 0.5 \(T_N\)), since the liquid is in the superfluid state while the crystal is grown, the heat generated at the interface will be rapidly taken away by the liquid, allowing the solid and the liquid to be in thermal equilibrium. Therefore, the temperature dependent driving force term can be neglected. The driving force will depend only on the applied overpressure needed to grow the crystal.

The effective growth coefficient \(k_{\text{eff}}\), which is the quantity to be determined, can be defined as [4]

\[ k_{\text{eff}} = \frac{v_f}{\Delta \mu_P} \]  

(2.2)

where the chemical potential difference (per unit mass) between the liquid and the solid can be expressed as

\[ \Delta \mu_P = \left( \frac{\rho_s - \rho_l}{\rho_s \rho_l} \right) \Delta P \]  

(2.3)
with $\rho_s$ and $\rho_l$ as the molar densities of the solid and liquid respectively, and where $\Delta P$ is the pressure change in the system with respect to the equilibrium melting pressure.

The effective growth coefficient has two contributions [5], one due to bulk effects (bulk growth coefficient $k_{\text{bulk}}$), which takes into account the contribution due to the heat transport and strongly depends on the geometry of the experimental cell, and one due to the interface (intrinsic growth coefficient $k_{\text{int}}$), which in case of faceted growth is determined by spiral growth of elementary steps,

$$\frac{1}{k_{\text{eff, growth}}} = \frac{1}{k_{\text{bulk, growth}}} + \frac{1}{k_{\text{int, growth}}}.$$  \hspace{1cm} (2.4)

Note that the bulk term always lowers the effective growth coefficient. In an ordinary liquid-solid system, the bulk term can be expressed as [6]

$$\frac{1}{k_{\text{bulk, growth}}} = \frac{\rho_s}{T} \frac{Z_1 Z_s}{Z_1 + Z_s} L^2$$  \hspace{1cm} (2.5)

where $Z_1$ and $Z_s$ are the thermal impedances of the liquid and the solid respectively and $L$ is the latent.

In the case of solid-superfluid $^3$He, the solid thermal impedance is much larger than the superfluid thermal impedance $Z_s \gg Z_s$, so the bulk term can be reduced to

$$\frac{1}{k_{\text{bulk, growth}}} = \frac{\rho_s}{T} Z_1 L^2.$$  \hspace{1cm} (2.6)

Since the latent heat vanishes very fast below the Néel transition, the bulk growth coefficient can then be neglected. Then, the effective growth coefficient equals the intrinsic growth coefficient, which is the one which has all the physical information about the growth properties.

Actually there is a method to check whether the last statement is true. In the same way that the effective growth coefficient has been defined (equation 2.4), the effective melt coefficient can be defined as follows

$$\frac{1}{k_{\text{eff, melt}}} = \frac{1}{k_{\text{bulk, melt}}} + \frac{1}{k_{\text{int, melt}}}.$$  \hspace{1cm} (2.7)

The ratio between $k_{\text{eff, growth}}$ and $k_{\text{eff, melt}}$ can be defined as

$$\eta = \frac{k_{\text{eff, growth}}}{k_{\text{eff, melt}}}.$$  \hspace{1cm} (2.8)

If $\eta$ is much smaller than one, it means that melting is faster than growing, which means that the bulk terms are negligible and the effective growth (or melt) coefficient is actually the intrinsic one.
2.2.2 Equilibrium Shape of the Crystals

Crystals surrounded by a liquid reach the equilibrium shape when the free energy of the interface between the two phases is minimal. The condition for the equilibrium at a certain point of the interface is the Herring equation [7]

\[ \Delta P \left( \frac{\rho_s}{\rho_l} - 1 \right) = \left( \alpha + \frac{\delta^2 \alpha}{\delta \phi_1^2} \right) \frac{1}{R_1} + \left( \alpha + \frac{\delta^2 \alpha}{\delta \phi_2^2} \right) \frac{1}{R_2} \] (2.9)

where \( \Delta P \) is the pressure of the bulk liquid calculated from the nominal equilibrium pressure for a planar interface, \( \rho_s \) and \( \rho_l \) are the densities of the solid and liquid phases respectively, \( R_1 \) and \( R_2 \) are the two radii of the crystal surface and \( \phi_1 \) and \( \phi_2 \) are the corresponding angular coordinates.

The surface stiffness tensor is defined as

\[ \gamma \equiv \alpha \delta_{\mu \nu} + \frac{\delta^2 \alpha}{\delta \phi_\mu \delta \phi_\nu}. \] (2.10)

The equilibrium shape of a crystal would be a perfect sphere (if no gravity is present), if the surface tension of the crystal would be isotropic. The surface tension of a crystal is anisotropic due to the different crystallographic orientations existing inside the crystal. Landau showed that at \( T = 0 \) the surface tension of a crystal \( \alpha \) is continuous but with a cusp at every rational orientation (Miller indices) [8]. The presence of facets is a direct result of this unusual behavior of the surface tension. The equilibrium facet size is proportional to the surface tension cusp for a given orientation. A cusp in \( \alpha \) means infinite surface stiffness that yields to zero curvature, in other words, a facet.

Since there is an infinite number of different crystallographic orientations, the surface of an ideal crystal should be covered with an infinite number of facets. This is called the devil’s staircase phenomenon [9, 10]. Landau neglected the surface fluctuations, both thermal and quantum, in his analysis. Fisher and Weeks [11] showed that quantum fluctuations do not destroy the faceting of the crystal and that even at zero temperature any crystal should have many different facets on its surface.

When temperature increases, the thermal fluctuations can hide the cusp of the surface tension for a certain crystallographic orientation, meaning that the facet disappears. Each type of facet has its own roughening transition temperature \( T_R \), which is expressed as [11, 12]

\[ T_R = \frac{2}{\pi k_B} \frac{d^2}{\sqrt{\gamma_\parallel \gamma_\perp}} \] (2.11)

where \( k_B \) is the Boltzmann’s constant, \( d \) is the height of an elementary step on a facet and \( \gamma_\parallel \) and \( \gamma_\perp \) are the principal components of the surface stiffness for a given surface orientation. The two components of the surface stiffness have to be measured...
at a temperature above but close to the roughening temperature of the facet.

In the bcc lattice, the height of an elementary step is given by

\[ d_{hkl} = \frac{1}{2} a (h^2 + k^2 + l^2)^{-1/2} \]  

(2.12)

where \( a \) is the lattice constant and \( <hkl> \) are the Miller indices of that facet [13].

A well-defined excitation on a facet is an elementary step, a linear defect that separates the facet into two different parts, which differ in height by one atomic layer. This step has a width \( w \) which is defined by its internal structure and the thermal and quantum fluctuations. If the width of the step is small (in the order of the lattice constant \( d \)), the step is strongly localized by the lattice potential (strong-coupling limit, fig. 2.5.a). On the other hand, if the width of the step is large compared with the lattice constant, the step is weakly localized (weak-coupling limit, fig. 2.5.b).

**Figure** 2.5: Elementary step in the a) strong coupling limit and b) weak coupling limit.

**Strong-Coupling Limit**

In the strong-coupling limit (figure 2.5.a) a vicinal surface of a bcc crystal can be considered as a distribution of steps on a primary facet [8]. These primary facets behave as isotropic strings with a linear tension per unit length called the step free energy \( \beta \).

The surface stiffness of a vicinal surface is determined by the value of the step free energy and the interaction between steps at long distances. The most long-ranged are the elastic [14] and the entropic [15] interactions, both depending on the distance as \( 1/x^2 \). In the mK-range the entropic interaction is relatively weak and the elastic interaction energy per unit length \( U \) can be written as [14]

\[ U(x) = \frac{f^2}{E} \left( \frac{d}{x} \right)^2 \frac{2}{\pi} \sigma^2 (1 - \sigma^2) \]  

(2.13)
where \( d \) is the step height, \( E \) is the Young’s modulus, \( \sigma \) is the Poisson ratio, \( f \) gives the strength of the elastic strain produced by the step and \( x \) is the distance between steps (\( x = N d \), where \( N \) is large).

The density of steps and therefore the surface tension depends on the tilted angle of the vicinal surface and this angle can be changed by rotation around the two principal axes, corresponding to the two principal values of the surface stiffness \( \gamma_{\parallel} \) and \( \gamma_{\perp} \) [12]

\[
\gamma_{\parallel} = \frac{2\pi(1-\sigma^2) f^2}{Ed} \frac{1}{N} \tag{2.14}
\]

and

\[
\gamma_{\perp} = \frac{\beta}{d} N \tag{2.15}
\]

This result is valid if the vicinal surface is rough. The roughening transition temperature \( T_{R,N10} \) of a vicinal surface can be calculated using equation 2.16 and the values obtained for \( \gamma_{\parallel} \) and \( \gamma_{\perp} \) from equations 2.14 and 2.15,

\[
T_{R,N10} = \sqrt{\frac{\beta_{100} f d_{R,N10}^2}{E k_B d_{100}}} \tag{2.16}
\]

where \( \beta_{100} \) is the free step energy of the principal step and \( d_{100} \) and \( d_{2N10} \) are the heights of the primary and secondary steps respectively. In the bcc structure \( d_{N10} = a/N \) for an odd \( N \) and \( d_{N10} = a/2N \) for an even \( N \), for \( N \gg 1 \).

The energies of these secondary steps can be calculated using the method by Landau [8], whose calculations were made for the case of Van der Waals interaction between the steps.

Assuming a set of parallel steps with height \( d_{100} \) for the primary facet with all distances between steps equal to \( x \) but one which is slightly larger, \( x + \Delta x \) (see figure 2.6) and an interaction energy between the step \( m \) and \( n \) equal to \( U_{m,n} = 1/x^2 \), the change of the total energy due to the shift of steps by \( \Delta x \) is
\[ \Delta U^+ = \sum_{m>0, n \leq 0} (x_{mn} + \Delta x)^2 - x_{mn}^{-2} = \]
\[ = \sum_{m>0, n \leq 0} -2 \frac{\Delta x}{x_{mn}^3} + 3 \frac{(\Delta x)^2}{x_{mn}^4} = \]
\[ = -\frac{\pi^2}{3} \frac{\Delta x}{x^3} + 3 \zeta(3) \frac{(\Delta x)^2}{x^4} \]  

where \( \zeta(3) = 1.202... \) For the same configuration of steps but a shift \( \Delta x \) of opposite sign, the change of total energy is

\[ \Delta U^- = \frac{\pi^2}{3} \frac{\Delta x}{x^3} + 3 \zeta(3) \frac{(\Delta x)^2}{x^4}. \]  

The sum of these two expressions gives the energy of a pair of secondary steps with opposite sign. The step energy of one of these secondary steps can be obtained taking the interaction energy in the form of equation 2.13

\[ \beta_{N10} = \frac{6}{\pi} \zeta(3) \frac{(1 - \sigma^2)}{E} \frac{f^2}{d_0^2(\Delta x)^2} d_{N10}^2 \]  

where \( d_0 \) is the height of the primary step and \( \Delta x \) is the minimal non-zero shift of the step.

These energies have been calculated for steps parallel to the primary step. For other orientations these energies should be somehow different. This anisotropy will be neglected for the discussion of the experimental data as well as for the anisotropy in the value of \( f \).

As a conclusion from the strong-coupling limit, it can be said that the free step energy of a facet depends on the step height as a fourth power, \( \beta \sim d^4 \).

The step energy is roughly equal to the surface energy of the additional area, \( \beta \sim \gamma_0 d \). In spite of the anisotropy, the ratio \( \beta_0 / \gamma_0 d \) is approximately equal to one.

**Weak-Coupling Limit**

The step energy is expected to be significantly reduced with respect to \( \gamma d \) for the weak-coupling limit (figure 2.5.b). Following Nozières [12], the values of the step energy and the step width can be estimated by

\[ \beta \sim d \sqrt{\gamma V} \]  

(2.20)
where $V$ is the energy barrier which pins the liquid/solid interface to the crystal lattice. If the effective width of the interface $l$ is large, $V$ is exponentially small. Assuming that $l$ is constant for all surface orientations

$$V \sim \gamma \exp \left( -\frac{l}{d} \right)$$  \hspace{1cm} (2.21)

and therefore

$$\beta \sim d\gamma \exp \left( -\frac{l}{2d} \right).$$  \hspace{1cm} (2.22)

This equation is derived for temperatures close to the roughening transition, but it has been proved to be valid also at zero temperature [16].

The surface stiffness of $^3$He crystals has been measured for rough crystals and it was found to be almost isotropic and temperature independent, $\gamma = \gamma_0 \approx 0.06 \text{ erg/cm}^2$ [17]. Approaching the roughening temperature, the surface stiffness increases due to the thermal and quantum fluctuations, although this effect is small in the weak-coupling limit.

There is not much experimental data in $^3$He on the step energy, step width and interface width. There is an indirect estimation by Rolley et al. [17] of the step energy, $\beta_{<110>} \approx 1.2 \times 10^{-11} \text{ erg/cm}$, which gives $w_{<110>} \approx 100d_{<110>} \approx 3 \times 10^{-6} \text{ cm}$.

Another independent indirect estimation of the step energy and step width can be done by calculating the interface width in $^4$He using equation 2.22 and the measured values of $<0001>$ facets [17, 18]. The result is $d/l_{<0001>} = 5.7$. Since $^3$He has a larger zero-point motion, it is reasonable to assume that $d/l_{<110>} = 7...8$, which yields into a step energy of $3...5 \times 10^{-11} \text{ erg/cm}^2$. This value is not far from the estimation by Rolley et al. [17].

In the weak coupling limit, the distribution of the step over a number of lattice spacings reduces the step energy, $\beta_0/\gamma_0d \ll 1$ and $w/d \gg 1$. For the $<0001>$ facet of $^4$He it has been found experimentally that $w \approx 8d$ [17], or even $w \approx 16d$ [19] with $\beta/\gamma d \approx 0.057$ [17]. Due to the larger zero-point motion of $^3$He it has been suggested [16] that in $^3$He the coupling of the interface to the lattice would be even weaker than in $^4$He [20].

### 2.2.3 Growth Velocity of Facets

For the growth of a facet some mechanism is required for the creation of sites in the solid where the atoms from the liquid can stick to. Thermal and quantum fluctuation provide such sites.
If the crystal has no dislocations, the growth of the facet is determined by the nucleation of new atomic layers. This mechanism is highly non-linear with respect to the applied driving force [22, 23].

In the presence of screw dislocations, spiral growth is the main growth mechanism at low temperatures. The spiral growth theory for $^4\text{He}$ crystals [24] is more complicated than the classical spiral growth theory since the effects of step inertia and quantum localization of the steps need to be taken into account. These effects can be neglected in $^3\text{He}$ crystals since the step velocity $v_s$ is restricted to a low critical velocity $v_c$ when the step mobility suddenly decreases. The velocity of a facet can be expressed in the following way

\[
v_f = \frac{\mu d^2}{19\beta} \left( \frac{\rho_s - \rho_l}{\rho_l} \right)^2 K(\Delta P)^2 \tag{2.23}
\]

where $\mu$ is the step mobility, $d$ is the height of the elementary step of the facet, $\beta$ is the step energy, $\rho_s$ and $\rho_l$ are the molar densities of the solid and liquid $^3\text{He}$ respectively, $K$ is the number of steps produced by one dislocation, and $\Delta P$ is the applied overpressure.

The step mobility has not yet been measured in $^3\text{He}$. Below the Néel transition, the step mobility can be estimated taking into account the two main processes which contribute to the step resistivity $1/\mu$, the scattering of the magnons in the solid with the moving step and the scattering of the quasiparticles in the superfluid with the moving step.

Assuming specular (100%) reflection at the interface and neglecting anisotropy in the magnon velocity, the magnon contribution to the step resistivity $1/\mu_m$ can be estimated using the scattering cross section [25] as

\[
\frac{1}{\mu_m} \approx \frac{(k_B T)^4 d^2}{w \hbar^3 c^4} \tag{2.24}
\]

where $k_B$ is the Boltzmann’s constant, $w$ is the step width (expected to be few lattice constants $a$ as in $^4\text{He}$ [12, 26]), $\hbar$ is the Plank constant, and $c$ is the magnon velocity.

On the other hand, the scattering cross section of the quasiparticles is very difficult to calculate since their wavelength is very short. Nevertheless, this scattering cross section can be estimated as $\sigma \sim d^2/w$ [25]. The contribution $1/\mu_{qp}$ due to the quasiparticles is

\[
\frac{1}{\mu_{qp}} \sim \frac{\hbar^4 d^2}{w} \exp(-\Delta/T) \tag{2.25}
\]

where $k_F$ is the Fermi wave vector and $\Delta$ is the superfluid energy gap of $^3\text{He}-B$. This is only valid at low step velocities, $v_s < k_B T/\hbar k_F$. If the step velocity is higher,
then $1/\mu_{qp}$ decreases [24, 21].

The growth velocity of facets is described by equation 2.23 until the step velocity reaches certain critical velocity $v_c$, and the step mobility suddenly decreases [27] due to a Cherenkov-type creation of excitations. The step velocity can then be expressed by

$$v_s = \mu \Delta P$$  \hspace{1cm} (2.26)

In this regime the growth velocity does not depend on the step mobility anymore and the facet growth velocity becomes linear dependent on the overpressure $\Delta P$. The expression for the facet growth velocity is the following

$$v_f = \frac{d^2v_c}{2\pi\beta} \left( \frac{\rho_s - \rho_l}{\rho_s} \right) K \Delta P.$$  \hspace{1cm} (2.27)

Generally speaking, $K$ depends on the Burgers vector of the dislocation, and in the simplest case $K = 1$. In absence of reliable data on characteristics of dislocations in our crystals we assume in the following $K = 1$.

In $^3$He the lowest critical velocities are the magnon velocity $c$ and the pair breaking velocity $v_{pb}$. At zero magnetic field both are approximately 8 cm/s [28, 29]. The effect of the magnons and quasiparticles on the step mobility depends on the strength of their coupling to the step motion.

From equation 2.26 it is possible to estimate the overpressure at which the facet velocity goes from a quadratic to a linear behavior. The step velocity equals the magnon critical velocity ($c = 8$ cm/s) and the mobility of the step can be considered to be the one for the magnons (equation 2.24),

$$\Delta P = \frac{v_s}{\mu} = \frac{c}{\mu_m} = \frac{(k_B T)^4 d^2}{\hbar^3 c^3}$$  \hspace{1cm} (2.28)

This yields to an overpressure of approximately 100 $\mu$bar. Below this value of the overpressure, the facet velocity may have a quadratic behavior (equation 2.23), but above it, the facet velocity may depend linearly on the applied overpressure (equation 2.27).

For magnons rather strong coupling is expected since the moving step directly touches and interacts with the spins close to the interface while for quasiparticles, due to their large momentum and the large value of the effective width $w$ of the step, the coupling becomes significant only at higher step velocities.
2.3 References


Chapter 3

Experimental Setup
Abstract

In this chapter the experimental setup used for the study of $^3$He crystals below the Néel transition while applying magnetic fields up to 9 T is described. The cryostat has been developed for many years and in this section only the most recent changes will be discussed. It consists of a dilution refrigerator in combination with a nuclear demagnetization stage. For carrying out our experiments we have optical access to the experimental cell.
Chapter 3

Experimental Setup

3.1 The Dilution Refrigerator

The dilution refrigerator was built by G.A. Vermeulen and G. Frossati [1] and it has been modified slightly over the years [2, 3].

The dilution refrigerator has a lowest stable temperature around 5 mK with a cooling power of approximately 20 μW at 10 mK. It is used for precooling the nuclear demagnetization stage, the experimental cell and the helium required for the experiment. An schematic drawing of the inner vacuum can of the cryostat is shown in figure 3.1.

The impedances of the 1K-pot have been changed since the 1K-pot was getting blocked at low temperatures. Before changing the impedances, they were flushed with benzine, acetone and ethanol and then pumped for a whole night, but the problem persisted. There are two capillaries connecting the 1K-pot with the main bath, with rather different impedances. The large impedance is used for the normal operation of the dilution refrigerator while the small one is only opened by means of a needle valve while condensing the mixture. Cu-Ni capillary \( \varnothing 0.3 \times 0.5 \) mm and 140 cm length was used for the large impedance, obtaining a value for the impedance of \( Z_{\text{large}} = 7.0 \times 10^9 \) cm\(^{-3}\). The same capillary was used for the small impedance but with a shorter length, 50 cm, obtaining a value of \( Z_{\text{small}} = 2.5 \times 10^9 \) cm\(^{-3}\). After the installation of the new capillaries, the 1K-pot did not get blocked anymore.

3.2 The Optical Setup

The optical system is basically the one developed by Marchenkov et al. [3, 4], but with some modifications [5].

In the original design all parts (light source, CCD camera, lenses, thermal filters and cold mirrors) were inside the vacuum can of the dilution refrigerator. The light
for the image was produced by a LED, which was thermally anchored to the 1K-pot. At present the light comes from outside into the cryostat through a 200 \( \mu \text{m} \) multimode optical fiber. In this way it is possible to change the light source, being able to simply change from LED to laser makes adjusting the optics much easier.

A change to the beam expander was also made. In the old design [3] the end of the optical fiber served as a point source and the parallel beam was formed by a set of negative and positive lenses and a long collimating tube. However, it was found out that the light sensitivity was not uniform all over the beam. The old beam expander was changed by a simpler design, using just a convex lens with at one side the end of the optical fiber in its point of focus. In this way a parallel beam with a diameter equal to 4 mm and an uniform intensity distribution was created.
Further, all the lenses, thermal filters and cold mirrors were replaced for new ones because the old ones were scratched due to frequent cleaning. Figure 3.2 shows an image at room temperature, where a set of wires was placed at the position of the sample cell.

![Image of wires at different positions](image)

**Figure 3.2:** A picture at room temperature made with the present optical setup. At the position of the sample cell a grid of wires with different diameters was placed at different positions with respect to the point of focus. On cooling down it was possible to observe the wires on focus getting blurred while the wires close to the CCD were sharper, i.e. the point of focus was moving.

Because the wires were placed at different positions along the optical path, it was possible to observe that the focal point was moving on cooling down. In fact, below LN$_2$ temperatures the point of focus is about 7 mm closer to the CCD camera than
at room temperature. Contraction in the optical setup and changes in the dielectric properties of the lenses during cooling changes the position of the point of focus. The quality of the images at low temperature considerably improved when adjusting the optics 7 mm out of focus and closer to the CCD at room temperature. Still, on cooling down the images get sometimes somehow blurred (looking out of focus), but it has not been possible to find out the cause of this behavior.

### 3.3 The Copper Nuclear Demagnetization Stage

The nuclear demagnetization stage is made of a massive oxygen free high conductivity copper rod with a purity of 99.99% (4 N) with some slits cut in order to avoid heating by eddy currents [6]. Its total mass is approximately 4 kg, which corresponds with about 60 mol. Details about the construction and testing of the nuclear stage can be found in the thesis by P. van der Haar [6] and A. Marchenkov [3]. A schematic drawing of the nuclear stage is shown in figure 3.3.

There is a $^3$He melting curve thermometer mounted on the nuclear demagnetization stage. At the bottom of the nuclear stage a silver rod is attached and at the end of this silver rod the experimental cell is clamped. The thermal contact between the experimental cell and the nuclear stage is good since the minimum in the $^3$He melting curve in the experimental cell can be seen after one minute that the pressure gauge on the nuclear stage has passed it.

### 3.4 The Heat Switches

The nuclear stage needs first to be precooled by the dilution refrigerator and thermally isolated when the demagnetization is started. For this purpose, two parallel heat switches are used.

Normally a heat switch consists of a superconductor, which can be forced to go from the normal to the superconducting state and vice versa. When the heat switch is in the superconducting state, the thermal conductivity is negligible, but it is increased by many orders of magnitude when it goes to the normal phase [7].

In the original design the switching element of each heat switch consisted of four tin foils. After seven years of operation the performance of the two tin heat switches had deteriorated a lot. During a typical magnetization of the nuclear stage, a ramp to 5 T in approximately two hours, the heat was so poorly taken out that it warmed up over 250 mK.
Due to ‘tin disease’ most of the foils were completely ruined. In one of the heat switches all foils were cut and we think that the earlier background heat leak of 50 nW to the nuclear stage [8] was caused by mechanical vibrations which made the two parts ring against each other.

The design of the new aluminium heat switches is shown in figure 3.4. Tin has the advantage that it can be directly soldered to the copper heat links, but it has the problem that the ‘tin disease’ would certainly arise again. The aluminium and the copper were joined together with the technique of diffusion welding. We decided to use a bulk piece of aluminium (purity 6N) [9] instead of the foils because it is
Thermal contact to nuclear stage

Copper heat link

Aluminum switching element

Copper heat link

Thermal contact to mixing chamber

Figure 3.4: Schematic drawing of the new heat switch.

easily pressed in between the copper heat links and gives a large thermal conductivity in the normal state. Calculations showed that the phonon conductivity in the superconducting state is still negligible in this configuration when the mixing chamber temperature is below 10 mK. Because the foils allow for deformation on cooling down but a bulk piece of aluminium does not, cuts in the copper link were made. Now the whole assembly can work as a spring to accommodate differential thermal contraction.

Before assembling the copper parts were annealed for 92 hours at 940 °C in an oxygen atmosphere of $1 \times 10^{-4}$ mbar. Then the copper-aluminium-copper sandwich was pressed together at 1200 bar and under pressure clamped together in a stainless steel holder. For diffusion welding [10] this assembly was put back in the vacuum oven for one hour at 500 °C.
The performance of the new heat switches is much better. The zero field background heat leak to the nuclear stage is now as small as 3 nW and only starts to increase if the mixing chamber is warmed up higher than 50 mK, as can be seen in figure 3.5. From this we can conclude that the background heat leak of 3 nW to the nuclear stage is not due to phonon conductivity through the aluminium piece. Furthermore the temperature of the nuclear stage does not increase above 40 mK if magnetized to 5 T in two hours, with heater switches in the normal metal state.

![Figure 3.5: The temperature of the nuclear stage as a function of time while the temperature of the mixing chamber was raised from 7 mK at 17:30 h to 55 mK at 18:40 h and then lowered again. The arrows indicate the time interval during which the mixing chamber was above 50 mK.](image)

**Figure 3.5:*** The temperature of the nuclear stage as a function of time while the temperature of the mixing chamber was raised from 7 mK at 17:30 h to 55 mK at 18:40 h and then lowered again. The arrows indicate the time interval during which the mixing chamber was above 50 mK.

### 3.5 The Experimental Cell

A schematic view of the experimental cell is shown in figure 3.6. The upper (Pomeranchuk) part of the cell is from an older experimental cell made by van Rooijen [8].
The Kapton membrane is made of eight layers of 12.5 µm Kapton foil which are glued together with Stycast 1266. The silver body of the heat exchanger in the cell is covered by a Ag-Pt sinter with a surface area of about 30 m². The volume just above the pressure gauge is filled up with a quartz cylinder in order to decrease the dead volume in the cell.

The optical part of the body of the cell is made out of Araldite. On the side two Suprasil windows (φ6 × 1 mm) are glued into holders of chalk (grey) Araldite with Stycast 2850FT. Just above the volume which is in view a vibrating wire is mounted. It is a semicircular loop of 3.5 mm made from a φ60 µm NbTi wire. In a magnetic field of 2 T, it has a resonance frequency of about 4 kHz. At the superfluid $A_1$ and $A_2$ transitions it shows sharp changes in the resonance frequency and amplitude.

In the upper volume of the cell we installed a sapphire capacitive pressure gauge as developed by Griffioen and Frossati [11]. In the pressure range of interest ($29 < P < 35$ bar) it has a sensitivity of about 0.5 pF/bar.
On the bottom of the cell we glued a 120 Ω strain gauge, which is used as a heater to nucleate the crystals by applying a small current pulse. The strain gauge is made of a thin film resistor (with a surface area of a few mm$^2$) which is deposited on a Kapton foil. Because it is very flat it can be mounted on the bottom of the cell, which is just below the field of view, while one can still see almost the whole lower part of the crystal.

3.6 References

Chapter 4

Results and Analysis
Abstract

This chapter explains the measurements carried out at different magnetic fields, how they were done and the analyzing techniques used for their understanding. It also includes a comparison with the results found at zero magnetic field by Tsepelin et al. [1, 2]. In the last section all the results are discussed and further experiments for deeper understanding are proposed.
Chapter 4

Results and Analysis

4.1 Crystal nucleation

Nucleating a single $^3$He crystal is a very challenging operation. Since the slope in the $^3$He melting curve is negative at low temperatures, the crystal nucleates in the warmest spot of the cell. Once the crystal is nucleated and has grown for a while, this is not the warmest spot of the cell anymore since the growth of the crystal locally cools the surroundings down (Pomeranchuk effect). Therefore, a second crystal may nucleate at the new warmest spot of the cell. Nevertheless, below the Néel transition, where the latent heat is almost zero and the liquid is superfluid, this operation is relatively easy.

The pressure trace of one nucleation can be seen in figure 4.1. The nucleation procedure of a crystal was done as follows. First of all, all the solid $^3$He in the experimental cell was melted until there was only liquid in the cell (figure 4.1.a). The liquid $^3$He was then slowly pressurized increasing the pressure of the $^4$He side of the Pomeranchuk cell with a constant mass flow (figure 4.1.b). When the $^3$He pressure was above the melting pressure (but still without solid nucleated), a heat pulse of 2.5 nW for 2-3 seconds at the bottom of the cell was applied inducing the nucleation of solid and the consequence drop of $^3$He pressure (figure 4.1.c). Once the crystal has been nucleated, it was still grown at constant overpressure until it was in the optical view (figure 4.1.d). After this, the crystal was melted till an apparently smooth round seed was left ready to be measured.

4.2 Analysis Technique

Once two dimensional projections of the crystals have been taken in the experiment, the next aim is to convert them to a three dimensional body. In order to come to any conclusion about the exact shape, going from two to three dimensions, additional information about the object is needed. It is known that $^3$He crystalizes in the bcc (body centered cubic) structure. Therefore the angles between different facets types...
From this, together with the recorded picture of the crystal, the expressions from the rotated normal vectors can be derived [3, 4]. However, these equations have to be solved numerically by the Newton-Raphson algorithm, resulting in an unpractical equation. Besides, this calculation is very sensitive to small errors made in extracting the data from the recorded image.

An alternative method was therefore used [5]. This method has the same assumptions, but the shape of the crystal is found by simulation instead of calculation. A wire-frame containing $<110>$, $<100>$ and/or $<211>$ planes is built up and compared with the image of the crystal. It is easy to extend the program in such a way that more types of facets are included in the fitting process.

The projection of the wire-frame is constructed in the following way. First, the normal vectors of the planes that correspond to the Miller indices indicated by the user are scaled by an user-determined scaling factor. Then all the vectors are written into a two dimensional array. After that, the points where three planes meet are calculated by the Gauss algorithm and the points that lie outside are deleted. Then the program searches for pairs of points that have two planes in common, drawing
Figure 4.2: This figure shows the computer program [5] that generates a wireframe of a bcc crystal and how it is used for the analysis of the data.

The analysis of a single crystal can be followed in figure 4.2. First, a bitmap image of the \(^3\)He is loaded and a crystal wire-frame calculated as described before is drawn on the screen and place on top of the image (figure 4.2.a). This wire-frame can be rotated along its own axes and along the optical axis and it can be translated over the screen vertical and horizontally. Facets can be placed further from or closer to the origin of the crystal and different types of facets can be included or excluded. The simulation process takes place on the screen. Obviously, the first guess of the crystal orientation remains as a personal choice and one has to decide which facet correspond to which Miller indices, but it provides an interactive trial and error method to fix the wire-frame and the image of the crystal (figure 4.2.b). Once the image is perfectly fitted by the wire-frame, another image of the crystal, after some growth time, is loaded into the program (figure 4.2.c). In the analysis of this new picture, the origin and
the orientation of the wire-frame are kept the same, but the length from the origin to each facet is changed until the crystal is again perfectly fitted (figure 4.2.d). The length change of the normal vector will be a quantity in pixels and, as the real-space dimensions of the image are known, the velocity of each facet can be determined by the pixel/real-space ratio.

Nevertheless, there is a fundamental uncertainty in the program inherent to the process of making a projection of the crystal. Namely, the 2D wire-frame image does not change if one lets each facet of the real crystal to grow (or melt) by \( v \cos \phi_i \Delta t \), where \( \phi_i \) is the angle between the normal to the \( i \)-facet and the optical axis \( z \). Such a process would correspond to the shift of the whole crystal by \( v \Delta t \) along \( z \) (see figure 4.3). This degree of freedom is out of control the method used. It means that a term \( v \cos \phi_i \) may be added to the measured velocity of each facet without further change of the wire-frame image, and the value of \( v \) is only limited by the condition that the total velocity of each facet is positive.

An extra assumption is therefore needed. It is reasonable to assume that facets of the same type are growing with the same velocity. Then the “optical axis” contribution can be determined unambiguously, provided that at least two different facets of the same type but with different \( \cos \phi_i \) are present in the crystal image. Moreover, this assumption can be checked directly if more than two such a facets are present.

**Figure** 4.3: Scheme illustrating the fundamental uncertainty of the data analysis. See text for details.
All crystal facets were analyzed with and without this assumption. Most of them showed a difference in velocity lower than ten per cent. There were only few crystals of which the velocities were dramatically different after applying this assumption. These crystals were not used in the following discussion. Therefore, the assumption that each type of facet grows with the same velocity is valid.

Once the velocity of each facet is known, only the applied overpressure during growth of the crystal needs to be determined. When the crystal is in equilibrium, that is without growing or melting, it is at the melting pressure. When the crystal is being grown (or melted), the pressure jumps up (or down). Subtracting these two pressures, the overpressure (or underpressure) applied for the growth (or melt) is obtained. As can be seen in figure 4.4, during the same growth experiment, different overpressures were obtained. Each of these overpressures correspond to a different growth velocity. Thus, in the same growth sequence, different overpressures and velocities were obtained.

![Figure 4.4](image.png)

**Figure 4.4**: The applied overpressure for growing the crystal is obtained from the melting pressure. Black points correspond to the $^3$He pressure and grey points to the $^4$He pressure. This crystal was grown at zero magnetic field and a temperature of 0.65 mK. See text for details.

It is also very important to know that there is only one crystal present in the experimental cell. In case there would be a second crystal out of the optical view, it would be impossible to determined unambiguously the velocities with which each
crystal is growing (or melting) because it is not known which crystal is growing (or melting). The amount of solid, or the solid fraction, can be calculated with the following expression [6]

\[ x_s = \frac{v_l}{\Delta v_{sl}} [\alpha + \kappa] \left( P_3 - P_{3m} \right) - \alpha \left( P_4 - P_{4m} \right) \]  

(4.1)

where \( v_l \) is the liquid molar volume at the pressure \( P_3 \), \( \Delta v_{sl} \) is the difference between molar volumes of the liquid and the solid, \( \kappa = \kappa_l \simeq \kappa_s \) is the compressibility of liquid (solid) \(^3\)He and \( P_{3m} \) and \( P_{4m} \) are the \(^3\)He and \(^4\)He pressures when the solid is nucleated. The elasticity of the cell \( \alpha \) is equal to

\[ \alpha = -\frac{\kappa_l}{d(P_3 - P_4)} \]  

(4.2)

The elasticity of the cell is obtained from the slope of \( P_3 \) versus \( P_3 - P_4 \) during the compression in the all-liquid state (see figure 4.5).
4.3 Experiments at $B=0$ T

In this section the experiments carried out at zero magnetic field are described. This data is compared with the results found by Tsepelin et al. [1, 2]. At this magnetic field, the liquid is in the superfluid $B$-phase and the solid is magnetically ordered in the $U2D2$ phase.

Since the experimental cell was cooled down for first time, the main aim of the experiments at zero magnetic field was to check whether it was possible to cool $^3$He down below the Néel transition and grow crystals at these temperatures, as well as compare the results with the measurements by Tsepelin et al. [1, 2].

In the experiments at zero magnetic field only the $\langle 110 \rangle$ and $\langle 100 \rangle$ facets were seen while Tsepelin et al. [2] had seen up to eleven different facets. However, in previous experiments in Leiden by Wagner et al. [7] also the $\langle 211 \rangle$ facet was seen. The reason for this significant difference in the amount of different types of facets might be the different analyzing techniques. While in Leiden direct images of the crystals are used, Tsepelin et al. used an interferometer in the cryostat and the phase shift technique, obtaining a higher sensitivity [8].

The same seed was kept for one day, growing faceted crystals and melting them several times per day. A typical growth of a crystal is shown in figure 4.6. It was grown from an apparently smooth rounded seed and immediately after applying overpressure to the cell, the crystal started growing and got its faceted growth shape, since the temperature was far below the roughening temperature of many facet types. The slowest growing facets determined the shape of the crystal very quickly.

The results of the $\langle 110 \rangle$ and $\langle 100 \rangle$ facets are shown in figure 4.7. Black points correspond to the $\langle 110 \rangle$ facet while the grey points correspond to the $\langle 100 \rangle$ facet. Various slopes can be seen in figure 4.7. These different slopes could be explained in terms of the error in the measurement of the applied overpressure. In our experiment overpressures down to 0.1 mbar were measured. The pressure gauge had an accuracy of 0.05 mbar, which gave an considerable error to the measurements. However, this difference of the facet velocities is too big for being only caused by the error measurement of the applied overpressure. It might be possible that this difference is due to real difference of facet velocities.

The solid fraction was calculated for each crystal grown showing that the amount of solid in the cell was always around 1% at the end of the growth. The volume of the crystal seen in the optical view can be estimated since the light beam had a diameter of 3.5 mm and the volume of the cell was also known. Calculating the ratio between the volume of the crystal with the volume of the cell and comparing it with the solid fraction, it can be concluded that there was only one crystal present in the cell during each growth sequence.
The effective growth coefficient $k_{\text{eff}}$ and the step energy $\beta$ were calculated for each facet of each crystal, and compared with the step energy measured by Tsepelin et al. [2] (as it can be seen in table 4.1). The step energies measured by Tsepelin et al. [2] and the averaged step energy found in this work ($\beta_{<110>}=(7.2 \pm 2.5) \times 10^{-10}$ erg/cm and $\beta_{<100>}=(2.3 \pm 1) \times 10^{-10}$ erg/cm) are of the same order of magnitude.
4.3 Experiments at $B=0$ T

**Figure 4.7:** Velocities of the different facets plotted versus the applied overpressure below the Néel transition at $B=0$ T. Black points correspond to $<110>$ facet and light grey to $<100>$ facet. Solid lines connecting points are drawn to guide the eyes.

**Table 4.1:** Effective growth coefficients (s/m) and step energies (erg/cm) for the different facets seen at zero magnetic field.

<table>
<thead>
<tr>
<th>Facet</th>
<th>Crystal</th>
<th>$k_{eff}$ $(10^{-5})$</th>
<th>$\beta$ at $B=0$ T $(10^{-10})$ (this thesis)</th>
<th>$\beta$ at $B=0$ T $(10^{-10})$ Tsepelin et al. [2]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt;110&gt;$ a)</td>
<td>1.3 ± 0.4</td>
<td>11 ± 3</td>
<td>6.6</td>
<td></td>
</tr>
<tr>
<td>$&lt;110&gt;$ b)</td>
<td>4 ± 1</td>
<td>3.6 ± 0.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;110&gt;$ c)</td>
<td>2 ± 1</td>
<td>7 ± 3.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;100&gt;$ a)</td>
<td>2.3 ± 0.7</td>
<td>3 ± 1</td>
<td>1.6</td>
<td></td>
</tr>
<tr>
<td>$&lt;100&gt;$ b)</td>
<td>5 ± 2</td>
<td>1.4 ± 0.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;100&gt;$ c)</td>
<td>3 ± 2</td>
<td>2.4 ± 1.6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
4.4 Experiments at $B=2$ T

In this section the experiments carried out at a magnetic field of 2 T are described. At this field, the liquid is in the superfluid $A$-phase and the solid orders into the $CNAF$ phase.

A typical growth process of a crystal at $B=2$ T is shown in figure 4.8. It was grown from an apparently smooth rounded seed and immediately after applying overpressure to the cell, the crystal started growing and large facets became visible. Note that the shape of the crystal is different from the crystal grown at zero magnetic field in figure 4.6. Also remarkable is the significant change in time to grow a ‘large’ crystal. At $B=2$ T more facets can be seen and the crystal grows much faster.

At this magnetic field ($B=2$ T) three different facet types were observed. Their velocities can be seen in figure 4.9. These crystals were analyzed as explained earlier. The solid fraction was calculated for each crystal grown, being around 1% after the growth. It can be concluded that there was only one crystal present in the cell during each growth sequence. First thing to notice is that the facet velocity has become larger for the $<110>$ and $<100>$ facets and that also the $<211>$ is now seen, while it was not observed at zero magnetic field.

The $<110>$ facet is the slowest one in the absence of magnetic field and at $B=2$ T it is very surprising that it is not the slowest one anymore. Since it has the smallest interplanar distance, it was expected to be also the slowest facet also in the presence of a magnetic field. Although the solid is magnetically ordered in a different phase, still the crystal symmetry is roughly the same, so not such a big change was expected.

This inversion of growth rate could mean that the change in the facet velocity might be due to the different liquid phase from which the solid grows. While at zero magnetic field the crystal is grown from the superfluid $B$-phase, at $B=2$ T the crystal is grown from the superfluid $A$-phase. The superfluid $B$-phase is isotropic in the $k$-space, resulting in an isotropic pair-breaking velocity. On the other hand, the superfluid $A$-phase has two nodes on the poles of the Fermi sphere, where the pair-breaking velocity goes to zero with a maximum in the equator of the sphere [9].

It is easy to see, however, that this anisotropy is not important in the present case. Indeed, a step moving at velocity $v_s$ radiates excitations by Cherenkov mechanism at angle $\phi$ defined as $v_s \sin \phi = v_{pb} = \Delta(\theta)/p_F$, where $p_F$ is the Fermi momentum (see figure 4.10). On the other hand, the energy gap $\Delta$ depends on angle $\theta$ in $k$-space calculated from vector $\vec{l}$ as $\Delta = \Delta_0 \sin \theta$. As usual, we assume that $\vec{l}$ is normal to the facet, which implies that $\phi = \theta$. We see that the critical velocity $v_{s,c}$ is still determined by $\Delta_0$ for all excitations independently of the radiation angle. Thus, a possible change in the facet velocity at the $A \rightarrow B$ transition could be due to the change of $\Delta_0$. 
4.4 Experiments at $B=2$ T

![Figure 4.8: Typical growth of a crystal at $B=2$ T.](image)

Since the magnon velocity in the solid ($v_m \approx 8$ cm/s) [10] is lower than the pair breaking velocity of the liquid ($v_{pb} \approx 10$ cm/s), the magnon velocity will be used as the critical velocity in the analysis. However, there is a crossover of the magnon and pair breaking velocities around $B=2.5$ T. Above this magnetic field, the magnon velocity becomes larger than the pair breaking velocity. Thus, for magnetic fields higher than $B=2.5$ T the pair breaking velocity will be used as critical velocity in
Figure 4.9: Velocities of the different facets plotted versus the applied overpressure below the Néel transition at $B=2$ T. Black points correspond to $<110>$ facet, light grey to $<100>$ facet and dark grey to $<211>$ facet. Lines connecting points are drawn to guide the eyes.
4.4 Experiments at $B=2\,\text{T}$

The effective growth coefficient $k_{\text{eff}}$ and the step energy $\beta$ were determined for each facet of each crystal. The step energies of different facets seen during the experiment were calculated from equation 2.27 using as a critical velocity the magnon velocity [10] ($v_c \simeq 8\,\text{cm/s}$). These step energies are compared with the ones obtained by Tsepelin et al. [2] at zero magnetic field (see table 4.2).

If the analysis is done in terms of growth with critical step velocities, it would indicate that the averaged step energy at $B=2\,\text{T}$ of the facets $<110>$ and $<100>$ is one order of magnitude smaller than at zero magnetic field ($\beta_{<110>}=(0.57\pm0.25)\times10^{-10}\,\text{erg/cm}$ and $\beta_{<100>}=(0.18\pm0.05)\times10^{-10}\,\text{erg/cm}$), showing that the presence of an external magnetic field indeed affects the growth kinetics of the crystal. The consequences will be discussed later. On the other hand, the step energy of the $<211>$ remains almost unaltered ($\beta_{<211>}=(0.3\pm0.1)\times10^{-10}\,\text{erg/cm}$).
Table 4.2: Effective growth coefficients (s/m) and step energies (erg/cm) for the different facets seen at $B=2$ T and compared with values found at zero magnetic field in this thesis and in the work by Tsepelin et al. [2].

<table>
<thead>
<tr>
<th>Facet</th>
<th>Crystal</th>
<th>$k_{\text{eff}}$ (10$^{-5}$)</th>
<th>$\beta$ at $B=2$ T (10$^{-10}$)</th>
<th>$\beta$ at $B=0$ T (10$^{-10}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;110&gt;</td>
<td>a)</td>
<td>30 ± 12</td>
<td>0.5 ± 0.2</td>
<td>7.2 ± 2.5</td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>b)</td>
<td>26 ± 15</td>
<td>0.6 ± 0.3</td>
<td></td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>c)</td>
<td>28 ± 13</td>
<td>0.5 ± 0.2</td>
<td></td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>d)</td>
<td>20 ± 8</td>
<td>0.7 ± 0.3</td>
<td></td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>e)</td>
<td>30 ± 22</td>
<td>0.5 ± 0.3</td>
<td></td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>f)</td>
<td>23 ± 9</td>
<td>0.6 ± 0.2</td>
<td></td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>a)</td>
<td>50 ± 12</td>
<td>0.14 ± 0.03</td>
<td>2.3 ± 1</td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>b)</td>
<td>45 ± 20</td>
<td>0.16 ± 0.07</td>
<td></td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>c)</td>
<td>59 ± 17</td>
<td>0.12 ± 0.03</td>
<td></td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>d)</td>
<td>30 ± 8</td>
<td>0.24 ± 0.06</td>
<td></td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>e)</td>
<td>44 ± 20</td>
<td>0.16 ± 0.07</td>
<td></td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>f)</td>
<td>30 ± 7</td>
<td>0.24 ± 0.06</td>
<td></td>
</tr>
<tr>
<td>&lt;211&gt;</td>
<td>a)</td>
<td>23 ± 6</td>
<td>0.21 ± 0.05</td>
<td>0.33 [2]</td>
</tr>
<tr>
<td>&lt;211&gt;</td>
<td>b)</td>
<td>17 ± 9</td>
<td>0.3 ± 0.15</td>
<td></td>
</tr>
<tr>
<td>&lt;211&gt;</td>
<td>c)</td>
<td>20 ± 4</td>
<td>0.24 ± 0.05</td>
<td></td>
</tr>
<tr>
<td>&lt;211&gt;</td>
<td>d)</td>
<td>15 ± 4</td>
<td>0.32 ± 0.09</td>
<td></td>
</tr>
<tr>
<td>&lt;211&gt;</td>
<td>e)</td>
<td>15 ± 8</td>
<td>0.3 ± 0.15</td>
<td></td>
</tr>
<tr>
<td>&lt;211&gt;</td>
<td>f)</td>
<td>10 ± 3</td>
<td>0.5 ± 0.1</td>
<td></td>
</tr>
</tbody>
</table>

4.5 Experiments at $B=8$ T

In this section the experiments carried out at a magnetic field of 8 T are described. At this field, the liquid is in the superfluid $A_1$-phase and the solid orders into the $CNAF$ phase. Some experiments were also carried out when the liquid was in the Fermi liquid state.

A typical growth of a crystal at $B=8$ T is shown in figure 4.11. It was grown from an apparently smooth rounded seed and immediately after applying overpressure to the cell, the crystal started growing with large facets visible. Note that the shape of the crystal is different from the crystal grown at zero magnetic field in figure 4.6 as well as the growth time to get a large crystal. At $B=8$ T more facet types can be seen and the crystal grows much faster. The quality of these images is not as good as the zero magnetic field and $B=2$ T since the optical access to the experimental cell got worse during the cool down.
4.5 Experiments at $B=8$ T

Figure 4.11: Typical growth of a crystal at $B=8$ T.

At this magnetic field ($B=8$ T) again three different types of facets were observed below the Néel transition ($T_N=3.1$ mK) and in the superfluid $A_1$-phase ($T_{A_1}=2.7$ mK). The velocities for the three of them can be seen in figure 4.12.

The effective growth coefficient $k_{\text{eff}}$ and the step energy $\beta$ were calculated for each facet of each crystal and shown in table 4.3. The step energies below the $T_{A_1}$ tran-
sition of the different facets seen during the experiment were calculated using as a critical velocity the pair breaking velocity ($v_c \simeq 10$ cm/s). These step energies were compared with the ones obtained by Tsepelin et al. [2] at zero magnetic field (see table 4.3). The step energies of all the facets have increased compared with the step energies found at $B=2$ T.

The averaged step energies of each type of facet have increased compared with the step energies found at $B=2$ T ($\beta_{<110>}=(3 \pm 1)\times10^{-10}$ erg/cm), $\beta_{<100>}=(0.9 \pm 0.3)\times10^{-10}$ erg/cm), $\beta_{<211>}=(1 \pm 0.4)\times10^{-10}$ erg/cm).

The velocity of the $<110>$ facet has been also measured below the Néel transition ($T_N=3.1$ mK), but above the superfluid $A_1$-phase ($T_{A_1}=2.7$ mK), i.e. from the Fermi liquid state, due to a small heat leak to the nuclear stage which was slowly warming the experimental cell. No other facets were seen at these conditions. The $<110>$ facet velocity was measured exactly at the superfluid transition ($T_{A_1}=2.7$ mK) (see Figure 4.12: Velocities of the different facets plotted versus the applied overpressure below the Néel transition of a crystals grown from the superfluid $A_1$-phase at $B=8$ T. Black point correspond to $<110>$ facet, light grey to $<100>$ facet and dark grey to $<211>$ facet. Lines connecting points are drawn to guide the eyes.

Figure 4.12: Velocities of the different facets plotted versus the applied overpressure below the Néel transition of a crystals grown from the superfluid $A_1$-phase at $B=8$ T. Black point correspond to $<110>$ facet, light grey to $<100>$ facet and dark grey to $<211>$ facet. Lines connecting points are drawn to guide the eyes.
Table 4.3: Effective growth coefficients (s/m) and step energies (erg/cm) for the different facets seen at $B=8$ T and compared with values found at zero magnetic field in this work and in the work by Tsepelin et al. [2].

<table>
<thead>
<tr>
<th>Facet</th>
<th>Crystal</th>
<th>$k_{\text{eff}}$ ($10^{-5}$)</th>
<th>$\beta$ at $B=8$ T ($10^{-10}$)</th>
<th>$\beta$ at $B=0$ T ($10^{-10}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt;110&gt;$ a)</td>
<td>6 ± 3</td>
<td>3 ± 1.5</td>
<td>7.2 ± 2.5</td>
<td></td>
</tr>
<tr>
<td>$&lt;110&gt;$ b)</td>
<td>8 ± 3</td>
<td>2.2 ± 0.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;110&gt;$ c)</td>
<td>9 ± 2</td>
<td>3.6 ± 0.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;110&gt;$ d)</td>
<td>5 ± 2</td>
<td>4 ± 1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;100&gt;$ c)</td>
<td>12 ± 2</td>
<td>0.7 ± 0.1</td>
<td>2.3 ± 1</td>
<td></td>
</tr>
<tr>
<td>$&lt;100&gt;$ d)</td>
<td>8 ± 3</td>
<td>1.1 ± 0.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$&lt;211&gt;$ c)</td>
<td>9 ± 2</td>
<td>0.8 ± 0.2</td>
<td>0.33 [2]</td>
<td></td>
</tr>
<tr>
<td>$&lt;211&gt;$ d)</td>
<td>5 ± 2</td>
<td>1.2 ± 0.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 4.13.a) and slightly above (figure 4.13.b at $T=2.8$ mK and figure 4.13.c at $T=2.9$ mK), showing some temperature dependence. In figure 4.13.d the velocities below and above the superfluid $A_1$-phase are compared.

The velocity of the $<110>$ facet has been also measured below the Néel transition ($T_N=3.1$ mK), but above the superfluid $A_1$-phase ($T_{A_1}=2.7$ mK), i.e. when the solid was growing from the normal liquid. This experiment was performed while the cell was slowly warming up. No other types of facets were seen at this conditions. The $<110>$ facet velocity was measured exactly at the superfluid transition ($T_{A_1}=2.7$ mK) (see figure 4.13.a) and slightly above (figures 4.13.b at $T=2.8$ mK and 4.13.c at $T=2.9$ mK), showing some temperature dependence. In figure 4.13.d the velocities below and above the superfluid $A_1$-phase transition are compared.

The difference in the $<110>$ facet velocity below and above the superfluid transition is an indication that the liquid plays an important role in the growth mechanism. The explanation for this observation might be that the heat generated at the interface by the growth of the crystal is no longer carried out as fast as when the liquid was in the superfluid phase, yielding to a local heating of the interface. Another possible effect of growing crystals from the normal liquid phase may be that there are no spin currents in the liquid, making the attachment of the atoms from the liquid to the solid slower since the solid is magnetically oriented.

The solid fraction was estimated for each crystal grown showing that the amount of solid in the cell was around 1%. The volume of the crystal seen in the optical view can be estimated since the light beam was 4 mm in diameter in this run. The ratio between the volume of the crystal and the volume of the cell indicates that there was only one crystal present in the cell during each growth sequence.
Chapter 4. Results and Analysis

Figure 4.13: Figures a), b) and c) show velocities of the different facets plotted versus the applied overpressure slightly above the superfluid transition at $B=8$ T and figure d) compares the data below and above the superfluid transition. Black points correspond to $<110>$ facet, light grey to $<100>$ facet, dark grey to $<211>$ facet and black open symbols for the $<110>$ facet above the superfluid transition. Lines connecting points are drawn to guide the eyes.

4.6 Preliminary Experiments at $B=0.8$ T

Experiments at a magnetic field of 0.8 T at a temperature of approximately 1 mK were also done. At this field, the liquid is in the superfluid $A$-phase and the solid magnetically orders into the CNAF phase.

A typical growth of a crystal at $B=0.8$ T is shown in figure 4.14. As in the previous situations, it was grown from an apparently smooth rounded seed and immediately after applying overpressure to the cell, the crystal started growing. More facets were seen compared to a crystal grown at zero magnetic field in figure 4.6. Notice as well that the crystal grows faster.
4.6 Preliminary Experiments at $B=0.8 \, T$

Since the magnon velocity in the solid at this magnetic field [11] is lower than the pair breaking velocity of the liquid ($v_{pb} \approx 10 \, \text{cm/s}$), the magnon velocity will be used as the critical velocity ($v_c \approx 5 \, \text{cm/s}$) in the analysis.

**Figure** 4.14: Typical growth of a crystal at $B=0.8 \, T$. 

Since the magnon velocity in the solid at this magnetic field [11] is lower than the pair breaking velocity of the liquid ($v_{pb} \approx 10 \, \text{cm/s}$), the magnon velocity will be used as the critical velocity ($v_c \approx 5 \, \text{cm/s}$) in the analysis.
Unfortunately, the analysis of the data taken at this magnetic field turned out to be very difficult to analyze since it was very complicated to fit the crystals with the computer program. The computer program only generates \( <110 >, <100 > \) and \( <211 > \) facets. It might happen that more facets than the ones generated by the program were present in this crystal, making the fitting process impossible. Another possible reason might be the fact that only two adjacent \( <110 > \) facets were seen in the crystal. In a normal fitting process, three adjacent \( <110 > \) facets are needed to be completely sure that the assumption each type of facet of a crystal is moving with the same velocity during the growth is valid. Somehow, this could also affect the fitting procedure. Nevertheless, an upper limit for the velocity of the \( <110 > \) facet could be obtained.

The step energy of the \( <110 > \) facet was compared with the one obtained by Tsepelin et al. [2] at zero magnetic field (see table 4.4). Notice that the step energy at \( B=0.8 \) T of the \( <110 > \) facet is almost one order of magnitude smaller that the one obtained by Tsepelin et al. [2] at zero magnetic field, showing that the presence of an external magnetic field indeed affects their behavior as it has been previously mentioned in section 4.4.

<table>
<thead>
<tr>
<th>Facet</th>
<th>( k_{\text{eff}} ) (s/m)</th>
<th>( \beta ) at ( B=0.8 ) T (10(^{-10}))</th>
<th>( \beta ) at ( B=0 ) T (10(^{-10}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( &lt;110 &gt; )</td>
<td>11 ± 5</td>
<td>0.81 ± 0.35</td>
<td>7.2 ± 2.5</td>
</tr>
</tbody>
</table>

The solid fraction was around 1% for each crystal grown at this magnetic field. The light beam diameter was 3.5 mm and therefore, it was possible to estimate the volume of the crystal, showing that there was only one crystal in the growth sequence.

### 4.7 Discussion

This section is a summary of the measurements taken on \(^3\)He below the Néel transition, possible conclusions and open lines to continue these studies.

Crystals have been grown at different magnetic fields \( (B=0 \) T, \( B=0.8 \) T, \( B=2 \) T and \( B=8 \) T) below the Néel transition showing a significant anisotropy for the different facets. The growth rates can vary by more than one order of magnitude depending on the magnetic field.
4.7 Discussion

Table 4.5: The effective growth coefficient obtained in this work compared with the effective melt coefficient obtained by different authors for the $<110>$ facet. The arrow in the ‘Liquid ↔ Solid phases’ column shows whether $k_{\text{eff}}$ was obtained during growing (pointing to the right) or during melting (pointing to the left).

<table>
<thead>
<tr>
<th>B (T)</th>
<th>Liquid ↔ Solid phases</th>
<th>T (mK)</th>
<th>$k_{\text{eff}}$ ($\times10^{-5}$s/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>$B \rightarrow U2D2$</td>
<td>0.65</td>
<td>$2.4 \pm 0.8$</td>
</tr>
<tr>
<td>0</td>
<td>$B \rightarrow U2D2$</td>
<td>0.55</td>
<td>200 [8]</td>
</tr>
<tr>
<td>0.07</td>
<td>$B \rightarrow U2D2$</td>
<td>0.73</td>
<td>50 [13]</td>
</tr>
<tr>
<td>0.2</td>
<td>$B \rightarrow U2D2$</td>
<td>0.7</td>
<td>23 [14]</td>
</tr>
<tr>
<td>0.33</td>
<td>$B \rightarrow U2D2$</td>
<td>0.7</td>
<td>40 [15]</td>
</tr>
<tr>
<td>0.33</td>
<td>$B \rightarrow U2D2$</td>
<td>0.62</td>
<td>60 [15]</td>
</tr>
<tr>
<td>0.8</td>
<td>$A \rightarrow \text{CNAF}$</td>
<td>1</td>
<td>$11 \pm 5$</td>
</tr>
<tr>
<td>2</td>
<td>$A \rightarrow \text{CNAF}$</td>
<td>1</td>
<td>$26 \pm 13$</td>
</tr>
<tr>
<td>8.9</td>
<td>$A \leftarrow \text{CNAF}$</td>
<td>2</td>
<td>$230 [14]$</td>
</tr>
<tr>
<td>8</td>
<td>$A_1 \rightarrow \text{CNAF}$</td>
<td>2.5</td>
<td>$7 \pm 2.5$</td>
</tr>
</tbody>
</table>

The effective growth coefficient were calculated for different facets from different crystals at different conditions. The values of the averaged effective growth coefficient for the $<110>$ facet are compared with the effective coefficient found by other authors (see table 4.5). Notice that the values for the effective growth coefficient presented in this thesis are about one order of magnitude larger than the effective melting coefficient found in the literature. This means that the bulk coefficient was negligible in our experiments and the effective growth coefficient corresponds with the intrinsic coefficient (see section 2.2.1).

The step energies of these crystals for different facets was calculated (see figure 4.15), showing an unpredicted behavior. The $<110>$ facet is the slowest at zero magnetic field but not anymore at higher magnetic fields, passing the $<211>$ facet to be the slowest one.

The step energy of the $<110>$ and $<100>$ facets decreases by an order of magnitude at $B=2$ T, while the $<211>$ remains almost unaltered. At $B=8$ T the $<110>$ and $<100>$ facets step energies have a value a bit lower than a factor of two compared with the value at zero magnetic field, while the $<211>$ facet step energy increases by roughly a factor four its value.

The ratio $\beta/\alpha d$ has been calculated for different magnetic fields (see table 4.6) in order to determine which limit is valid for the interaction between the liquid/solid interface with the crystal lattice (see section 2.2.2).

It was found by Tsepelein et al. [2] that at zero magnetic field the interface couples
Figure 4.15: Step energy of a few facets below the Néel transition at different magnetic fields. Close symbols correspond with the step energies found in this thesis while the open symbols correspond with the work done by Tsepelin et al. [2].

strongly to the crystal lattice ($\beta/\alpha d \sim 1$) while it has been found experimentally that for $^4$He $\beta/\alpha d \simeq 0.057$ [16]. It has been suggested [17] that in $^3$He the coupling of the interface to the lattice would be even weaker than in $^4$He [18] due to the larger zero-point motion of $^3$He. The results presented in this thesis show that at zero magnetic field the interface couples strongly to the crystal lattice, but at $B=0.8$ T and $B=2$ T they seem to be in the weak-coupling limit ($\beta/\alpha d \ll 1$). At $B=8$ T it seems that the interface couples again strongly to the crystal lattice.

Definitely more experiments are needed since there are too little experimental points to make a definite conclusion. The work presented in this thesis shows indications that the phase transition from the $U2D2$ phase to the $CNAF$ phase clearly affects the growth mechanisms of the crystals (figures 4.9 and 4.12).

It would be especially interesting to grow crystals slightly above and below the phase transition ($B=450$ mT) where the liquid is in the superfluid $B$-phase. Then, from the same liquid phase, crystals could be grown into the two different magneti-
cally ordered phases of the solid. As it can be seen in the work by Ni et al. [11], the magnon velocity differs almost by a factor 2 in the transition from the $U2D2$ phase to the $CNAF$ phase. If the magnon velocity turns out to be really the critical velocity (see equation 2.27), the growth rates should also differ by a factor 2 below and above the transition.

Another interesting measurement would be to grow crystals slightly below 520 mT, where the liquid is in the superfluid $B$-phase and the solid is in the $CNAF$ phase, and just above 520 mT, where the liquid is in the superfluid $A$-phase and the solid is in the $CNAF$ phase. This would also give the possibility of studying the growth mechanisms starting from different initial liquid phases and to the same solid phase.

Another possible experiment would be to grow crystals at different magnetic fields in the $CNAF$ phase and try to find out what the relation is between the step energy and the external magnetic field.

It would be also interesting to study crystals at high magnetic field and grow them from both the normal (Fermi liquid state) and superfluid phase and always below the Néel transition, since it seems to have some effect in the growth mechanisms (see figure 4.13.d). Difference in the thermal conductivity and diffusion properties in both liquid phases may affect the growth process.

Unfortunately the equilibrium shape of the crystals could not be studied in this thesis. A new electrically actuated cold valve has been built [19]. In order to investigate the (slow) approach to the equilibrium shape of the crystal it is essential that no fluctuations in the volume of $^3$He are present. These fluctuations are not caused directly because $^3$He capillary is fully plugged, but indirectly through fluctuations in the $^4$He pressure. With this valve closing off the $^4$He filling line near the experimental cell, thermal and pressure fluctuations will be avoided and the crystal can go to its equilibrium shape without any external perturbation.

| Table 4.6: $\beta/\alpha d$ calculated for different facets at different magnetic fields. |
|-----------------|-----------------|---------|---------|---------|
| $B(T)$ | Liquid $\rightarrow$ Solid | $<110>$ | $<100>$ | $<211>$ |
| 0 | $B \rightarrow U2D2$ | 0.335 | 0.151 |
| 0.8 | $A \rightarrow CNAF$ | 0.038 |
| 2 | $A \rightarrow CNAF$ | 0.026 | 0.012 | 0.024 |
| 8 | $A_1 \rightarrow CNAF$ | 0.139 | 0.059 | 0.081 |
4.8 References


Chapter 5

Improvements to the Cryostat
Abstract

After few experimental runs with the cryostat, some changes were made in order to improve the accuracy in the measurements. Explosion welded heat switches (after failure of the ones described in chapter 3), a titanium pressure gauge which works at high magnetic fields (with much higher accuracy) and an electrically actuated cold valve (for the study of the equilibrium shape of the crystals) have been designed and built up. All these new parts are described in this chapter.
Chapter 5

Improvements to the Cryostat

5.1 The Explosion Welded Heat Switches

Although the heat switches had been changed recently [1], they broke down at the diffusion welding plane after a quench of the demagnetization magnet. Both heat switches were repaired (diffusion welded again) but the joint never became as strong as before and after few cool downs they broke again.

It was then decided to fabricate new heat switches with a stronger weld between the copper and the aluminum. Three millimeters thick copper (99.99% purity) and aluminum (99.999% purity) plates were bought [2]. A sandwich of three plates (Cu-Al-Cu) was made and joined by means of explosion welding at TNO [3].

The sandwich has a thickness of 9 mm and was spark-cut to the correct shape and then bent (as shown in figure 5.1) in order to make it fit in the old heat switch holder and therefore use the same magnet for switching them from the normal to the superconductor state. Cuts in the copper link were made, so the whole assembly can work as a spring to accommodate differential thermal contraction.

The explosion welded heat switches have been mounted on the cryostat and the nuclear stage has been cooled down below 1 mK. Unfortunately, these heat switches were not characterized as well as the previous ones. Nevertheless, checking the cooling time and comparing them with the old heat switches (see section 3.4), the performance of the explosion welded heat switches is at least as good.

5.2 The Titanium Pressure Gauge

The sensitivity of the sapphire pressure gauge [4] in the experimental cell is not very high ($\sim 0.5$ pF/bar at $P = 35$ bar). Therefore it was not easy to measure accurately the applied overpressure obtained in the growth of a crystal, giving a large uncertainty in its reading. In fact, it was not sensible to apply overpressures smaller than
100 µbar because the noise was of the same order than the overpressure. Thus, a new pressure gauge with higher sensitivity was designed, constructed and tested [5].

5.2.1 Pressure Gauge Design

In previous designs the gauge was usually made of Cu-Be. It was decided to use titanium because of its low heat capacity at low temperatures and because it is insensitive to magnetic fields. In this way also problems machining Cu-Be, which is very poisonous, were avoided. The elastic properties of titanium ($E_{Ti}=160$ GPa, $S_y, Ti=980$ MPa) are also similar to the elastic properties of Cu-Be. The Straty-Adams type design of Miura et al. [6], who were the first to use titanium as material for the membrane, has been adapted.

The aim is to improve the sensitivity at least twenty times with respect to the sapphire gauge (see the noise in $\Delta P$ in figure 4.4). The second criterium is to make
the design fit into the cryostat. There is not so much space left close to the nuclear stage. Only a gauge with dimensions smaller than 25 mm in diameter and 20 mm in height would fit in the cryostat. These two were the main considerations for the design of the pressure gauge. To determine the behavior of the gauge the model of a circular membrane fixed at the edge is considered. This gives a deflection \[ y(r, P) = \frac{3P}{16Ed^3} (r^4 - 2r^2R^2 + R^4), \] \[ y(0, P) = \frac{3P}{16Ed^3} R^4, \]

where \( R \) is the membrane radius, \( E \) is the Young’s modulus, \( d \) is the membrane thickness and \( r \) is the distance from the center of the membrane. The deflection will be bigger using the largest diameter as possible and it will be maximum for the minimum membrane thickness for which deformation is still in the elastic regime at the maximum pressure. To be sure that the membrane is below the elastic limit it was kept below about 60% of the yield stress \( S_y \) of the material. The maximum radial stress on the membrane is given by \[ S_m = \frac{3p}{16Ed^2} R^2. \]

The pressure gauge, shown in figure 5.2, consists of a silver body where 0.35 g of silver have been sintered getting a surface area of approximately 0.5 m\(^2\). This piece has also a M3 threat to attach the gauge to the nuclear stage at the cryostat. The membrane is made of titanium with a diameter of 14 mm and a thickness of 0.45 mm. The volume between the silver body and the membrane is sealed with an indium ring tightened with twelve M2 stainless steel screws. The \(^3\)He comes into this volume through a capillary placed in the silver body. The membrane ends up with a tip (1.2 mm in diameter) where a capacitor plate (10 mm in diameter) is placed. In order to electrically insulate it, a plastic cup is glued between the tip on the membrane and the capacitor plate.

The upper capacitor plate (11.8 mm in diameter) is glued with Stycast 2850FT to the holder and electrically isolated from it with a 100 \( \mu \)m Kapton ring. Both capacitor plates have been polished flat with an accuracy within 0.1 \( \mu \)m. A cover is placed on top of the holder and the whole pressure gauge is kept together with four M2 stainless steel screws. Two wires are glued with silver epoxy (EE129-4A + EE129-4B) on the capacitor plates in order to read out the capacitance. The final stage of the assembly is to glue the lower capacitor plate on the tip of the membrane. This is done as follows. After gluing the plastic cup to the lower capacitor plate with Stycast 2850FT and drying it, both capacitor plates have to be cleaned. The lower capacitor plate is placed on top of the upper one and then everything is mounted together, gluing the plastic cup to the tip with Stycast 1266, with four M2 stainless steel screws,
pressurizing the chamber with 25 bar (which corresponds with a touching pressure at liquid helium temperature of 36 bar) in such a way that the two plates are touching.

5.2.2 Results and Discussion

The pressure gauge was tested at room and liquid helium temperature. Data points were taken both going up and down in pressure, showing a linear behavior in $1/C$ vs. $P$ in the region of interest between 28 and 36 bar and no hysteresis (as can be seen in figure 5.3).

The points were fitted using a general fit

$$
\frac{1}{C} = \frac{c - bP}{a}, \quad (5.4)
$$

where $a$ depends on the effective area of the capacitor, $c$ is the gap between the two plates at $P=0$ and $b$ describes the behavior of the membrane and is dependent on the Young’s modulus. Comparing data and fits from room and liquid helium temperature it was observed that the gap between the two plates became ten microns larger when it was cooled due to thermal contraction, probably of the plastic cup. Also a change in the Young’s modulus of titanium about 30-40% was found, while only 10-15% was expected. With this design sensitivities up to 14 pF/bar at melting pressure were reached.
5.3 The Cold Valve

The equilibrium shape of the crystals could not be studied in this thesis, since there were fluctuations in the volume of the cell containing the $^3$He, via fluctuations in the $^4$He filling capillary. A new electrically actuated cold valve has been designed, constructed and tested [8]. With this valve closing off the $^4$He filling line near the experimental cell, thermal and pressure fluctuations will be avoid and the crystal can go to its equilibrium shape without any external perturbation.

It was decided to design an electrically actuated valve based on the movement of a permanent magnet. A tip placed on the moving magnet closes a small hole in a seat, making the system leak tight, by means of a coil wound around the valve. One other advantage of using this mechanism is that there is no volume change (inducing a pressure change) in the valve when closing or opening. The second criterium is to make the cold valve fit into the cryostat. Typically there is not much free space in ultra low temperature cryostats, so the design of the valve has to be as compact as possible. This means a maximum diameter of 25 mm and a length of 35 mm. These

**Figure 5.3**: Inverse of the capacitance is plotted versus the pressure at a temperature of 4K. The round grey data points were measured while going up in pressure while the square black points were taken while going down. No hysteresis is seen.
were the two main considerations in the design of the cold valve.

To estimate the behavior of the valve, the force created by a gradient in the magnetic field acting on a permanent magnet needs to be calculated. This force is given by

\[ F_z = \int_V \overrightarrow{\mu} \frac{d\overrightarrow{B}}{dz} dV, \]  

with \( \mu \) the magnetic moment of the permanent magnet given by

\[ \overrightarrow{\mu} = \int_V \overrightarrow{M} \mu_0 dV, \]  

where \( M_i \) is the magnetization, \( V \) is the volume of the permanent magnet and \( \mu_0 \) is the permeability of free space.

To create this gradient of magnetic field, two coils in the Helmholtz configuration (distance between the coils equals the diameter of a coil) were (counter) wounded. This gives the most uniform gradient. The force needed to close the valve varies between 10 and 30 N depending on the design [9, 10, 11, 12, 13]. It was assumed that the force required for closing this valve will be similar.

### 5.3.1 Cold Valve Design

A schematic drawing of the cold valve is shown in figure 5.4. The main body of the valve is made out of brass. Two grooves are machined in this part in order to wind the Helmholtz coils in them. This main body is closed with a cap where six M2.5 screws make the indium ring seal leak tight. Inside this volume the permanent magnet is placed inside a holder made out of Araldite with a needle machined out of brass in the center of it. At the end of this needle a ruby is clamped to the needle getting it fixed. This ruby will act as the tip pressing on the seat, which is glued on the cap. All the important sizes of the cold valve are given in table 5.1.

<table>
<thead>
<tr>
<th>Dimensions [mm]</th>
<th>Coils</th>
<th>Magnets</th>
<th>Seat</th>
<th>Ruby</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outer diameter</td>
<td>22</td>
<td>10.6</td>
<td>10.6</td>
<td>1</td>
</tr>
<tr>
<td>Inner diameter</td>
<td>14</td>
<td>5.2</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Thickness</td>
<td>4</td>
<td></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Heart to heart distance</td>
<td>19</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Table 5.1: Sizes of the important parts of the cold valve.*
In the following sections a more detailed description of each part of the valve will be given.

The Permanent Magnet

A material with the highest intrinsic induction possible, in order to make the force larger, is needed. The chosen material is NdFeB [14, 15, 16, 17], a sintered ceramic material. NdFeB has a remanent field of 1.17 T. Four rings were used, placed one on top of the other, giving a total length of 10 mm. A 13 mm long needle with the tip is glued into the magnet. This is the reason the shape of the magnet was chosen as a ring. In the center of this ring a needle, with a ruby clamped at the top (exploded view in figure 5.4), is glued with Stycast 2850FT. This composite permanent magnet is glued into an Araldite holder that allows the magnet to slide better inside the valve. This whole construction is kept in the right place (where the gradient of
magnetic field is linear) by two springs, one at the bottom, which lifts the magnet up, and one on top, which makes sure the valve opens again when the current is released (although the valve might also be opened by reversing the current in the coil). The spring constant has a value of 190 N/m. To close the valve, the tip needs to move 1 mm. This means that a force of 0.19 N needs to be applied for compressing the spring, which is a factor 100 lower than the force typically needed to close the valve. The force due to the spring can be neglected.

**The Magnetic Field Gradient**

To create a gradient in the magnetic field two coils were counter wound around the valve body as is explained earlier. Nb-Ti wire in copper matrix with a diameter of 110 µm (including 10 µm insulation) was used to wind both coils. A total number of 800 turns per coil were wound, getting a total resistance of 250 Ω at room temperature and an inductance of 15.44 mH (at \(T=4.2\) K). The field profile along the central axis of this configuration was calculated and is shown in figure 5.5.

![Figure 5.5: Magnetic field profile along the central axis as function of position, with the origin in the middle between the two coils.](image-url)
5.3 The Cold Valve

To crosscheck the simulation the law of Biot-Savart was used. The magnetic field in the center of a circular loop of radius \( R \) is given by the following formula:

\[
B(z = 0) = \frac{\mu_0 N I}{2R},
\]

(5.7)

where \( \mu_0 \) is the permeability of free space, \( N \) is the number of turns, \( I \) is the current in the coil and \( R \) is the radius of the coil.

Using this formula with the values of the coil, \( B(0) = 5.9 \times 10^{-2} \) T/A. This compares very well with the simulated value shown in figure 5.5 at \( z = +9.5 \) mm and \( z = -9.5 \) mm.

The gradient is determined to be \( dB/dl = 6.1 \) T/m when 1 A is applied (figure 5.5). Using equations 5.5 and 5.6 and the parameters of the NdFeB magnets, the force per unit of current applied with this setup is estimated to be 4.1 N/A.

This force was also measured at room temperature. Small weights were lifted by sending current through the coils. Only very low currents in order to prevent overheating of the coil wire could be used. Since the force is linear with the current, it was possible to extrapolate obtaining a force per unit of current of 4.6 N/A, which agrees with the values estimated before.

\[\text{Figure 5.6: Scheme of the setup where the cold valve was tested.}\]
The Tip and the Seat

For the selection of tip and seat a few materials were tried out. The idea is to have a hard material as tip pressing onto a soft material chosen as seat. The seat has a 0.5 mm hole in the center, which gets closed by the tip when current is applied. Because there was good experience with ruby tip valves in the group [11], ruby was immediately chosen as the tip material. Finding a good material for the seat was more difficult.

For testing the cold valve, an experimental setup was prepared as shown in figure 5.6. The cold valve was pumped with a leak detector and filled with He gas coming from a cylinder, previously going through a cold trap. When the valve was working properly and closing, the leak rate at the outlet of the valve with the leak detector could be determined. On the other hand, when the valve was not closing, the leak detector was going out of scale.

In the above mentioned work with the ruby tip [11] a metal seal was used. Hence a seat made of phosphor-bronze was used at the beginning. With this combination (ruby and phosphor-bronze) the valve was not closing. The impedance of the outlet pumping line was changing but the leak detector placed at the outlet of the valve was going out of scale and a pressure of few millibars could be read. Several seats with the holes drilled under different angles (see figure 5.7) were tried but with similar results. All these seats were tested with currents up to 10 A. Phosphor-bronze turned out to be too hard material.

So it was decided to gold plate [18] the phosphor-bronze seat with a layer of 50 µm. Gold is a material softer than phosphor-bronze. Besides it could be expected that the gold fills in the surface roughness produced on the edge of the hole in the seat by the drilling process. It is also possible that cracking of the oxide layer on the phosphor-bronze seat, leaving pores behind through which the helium can flow, was the source of the leak. Gold is a noble metal, so no oxide layer is formed on it. Unfortunately, this configuration did not work either.

The next step was to glue a piece of Kapton foil (100 µm thick) onto the phosphor-bronze seat for making the pressing surface softer. Kapton was glued with Stycast
1266 always being very careful not to block the hole already machined in the phosphor-bronze seat. When the Stycast 1266 was dry a 0.4 mm hole was machined in the Kapton in such a way that a very smooth surface was obtained. The valve only worked once (as shown in figure 5.8), getting a leak rate in $^4$He lower than $3 \times 10^{-8}$ mbar l/s with 20 bar pressure difference across the valve.

The reason why the phosphor-bronze with Kapton glued on it only worked once might be that, even if Kapton is an elastic material, the foil was too thin and the ruby eventually was pressing on the phosphor-bronze instead of the Kapton. After this result, it was decided to use plastic seats instead of metal ones.

![Figure 5.8: $^4$He background at the outlet of the valve as function of pressure difference across the valve. A phosphor-bronze seat with 100 µm Kapton foil glued on it was used in this case. The closing current is 8 A, which corresponds with a closing force of 33 N. (Notice that the background is still going down).](image)

The first plastic material which was tried out was Vespel, a sintered Kapton powder. A seat completely machined out of Vespel (phosphor-bronze was not longer used as the seat) was made and a 0.5 mm hole was drilled in the center. In order to get a leak tight closure this hole needs to be completely symmetric and the surface around the hole very smooth. Small cracks or scratches around the hole might produce a leak in the valve. This combination of tip-seat gave a good performance (shown in figure 5.9), getting a $^4$He leak rate less than $10^{-9}$ mbar l/s with 5 bar pressure difference across the valve. The same seat could be cooled down several times, and the system was still leak tight. Reproducibility does not seem to be a problem here. Also different seats were tried getting similar results.
Figure 5.9: $^4$He background at the outlet of the valve as function of pressure difference across the valve. A Vespel seat was used. The closing current is 8 A, which corresponds with a closing force of 33 N. (Notice that the background is still going down).

Once a leak tight valve was obtained, it was decided to optimize the system trying to find a configuration which the current required to close the valve was lower than 8 A. First thing that was tried was to make the hole in the seat of a different diameter. A 0.4 mm hole was tried out, getting a close system but with a higher leak rate (see figure 5.10) than with the previous configuration. When machining a 0.6 mm hole instead of a 0.5 mm hole the valve was not even closing and the leak detector placed at the outlet of the valve was going out of scale every time. The impedance of the line was changing but the valve was never leak tight. It can be concluded that a hole of 0.5 mm is the best for the performance of this design.

Table 5.2: Elastic properties of the different plastic materials that were used as seats [2].

<table>
<thead>
<tr>
<th>Properties</th>
<th>Ultimate tensile strength</th>
<th>Young’s modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kapton</td>
<td>$2.3 \times 10^8$</td>
<td>$2.6 \times 10^9$</td>
</tr>
<tr>
<td>Vespel</td>
<td>$8.7 \times 10^7$</td>
<td>$2.4 \times 10^9$</td>
</tr>
<tr>
<td>Torlon</td>
<td>$1.2 \times 10^8$</td>
<td>$4.1 \times 10^9$</td>
</tr>
</tbody>
</table>
5.3 The Cold Valve

Figure 5.10: $^4$He leak rate at the outlet of the valve as function of pressure difference across the valve. A Vespel seat with a hole of 0.4 mm was used as the seat instead of 0.5 mm hole. The closing current is again 8 A. The leak rate is higher even at lower pressure differences. (Notice that the leak rate goes up).

Having a closing current of 8 A is not desirable for the cryostat. In order to reduce this current another combination of tip-seat was tried out. This time Torlon was used as seat. Torlon is a plastic with similar elastic properties to Vespel (see table 5.2 below).

The Torlon seat was glued and the cold valve was tested. A current of 3 A, which corresponds with a value in force of 12 N, closed the valve such that the $^4$He leak rate was lower than $10^{-7}$ mbar l/s with 5 bar pressure difference across the valve were obtained (figure 5.11). This background is good enough for the purpose of the experiment. Different Torlon seats were checked out for reproducibility. Each of them was tested at least twice and all of them worked fine.

5.3.2 Assembling the Cold Valve

After gluing the needle in the center of the permanent magnet with Stycast 2850FT the magnet has to be also glued in the Araldite holder with GE varnish. The inner part of the valve body and the magnet holder need to be clean in order to allow a good movement of the magnet inside the valve. Then a spring has to be put inside the valve body to lift the magnet to the position with the most uniform gradient in the magnetic field (see figure 5.5). Another spring will be placed on the magnet opening
the valve after releasing the current. The Torlon seat is glued on the cap with Stycast 2850FT. Both seat and ruby must be very clean before closing the valve. A small piece of any material (or dust) can produce a leak due to a bad sealing. The ruby is cleaned by using lens paper on a sharp-cut wooden stick spraying a little ethanol on it. The seat can be cleaned in the ultrasonic bath with ethanol. Important for getting a leak tight valve is the form and shape of the hole in the seat. It needs to be really clean and symmetric. All tiny or loose pieces of Torlon should be removed with a wooden stick applying no force to the seat, otherwise it will deform, but if the hole is drilled properly none of these loose pieces should be visible. No knife should be used because otherwise the seat will be damaged. Once both ruby and seat are cleaned the valve can be assembled with an indium ring that seals the valve body with the cap.

Important to notice is that when the ruby is pushed for the first time towards the seat at low temperature the system usually leaks. This is due to the fact that the seat leaves some Torlon loose pieces on the ruby. This happens only the first time the valve is closed and cooled down. After the valve has been warmed up and opened and both ruby and seat have been cleaned again, the system is leak tight for the next cool downs. This can be explained by the fact that at low temperature the Torlon becomes more rigid and some of it is tweaked at the edge of the hole and becomes loose due to the force applied on it.
The needle (and therefore the ruby) will find its most favorable position in closing the hole by itself. It seems that using a guiding piece will only prevent somehow the ruby to find its optimal position.

5.3.3 Discussion

In this section the hydraulic actuated valves [9, 10, 12, 13] and the magnetically driven valve are compared, noting the advantages of the cold valve presented in this thesis.

With the design of Roach et al. [9] the leak rate is determined to be lower than $10^{12}$ molecules/sec. With the design of Bischoff et al. [10] a leak rate of $10^{13}$ molecules/sec is reached. With this design a leak rate which is lower than $10^{10}$ molecules/sec is measured, which is a factor of hundred and thousand better than the leak rate obtained by Roach et al. and Bischoff et al. respectively, although the leak rate is ten times higher than in the valve designed by Bruckner et al. [13].

Compared with the 10-30 N closing force in earlier designs, 12 N were needed for the ruby-Torlon combination. The actual closing current can be reduced by using stronger permanent magnet or winding a coil that produces a larger gradient in the magnetic field.

While the valve of Tsuda et al. [12] can be used at room temperature, the current design of the cold valve presented here only works at low temperature where the coil is superconducting. Besides, if the Torlon seat gets pressed at room temperature, it deforms too much, meaning that at low temperature it will not close.

A very important advantage of this design is that there is no dead volume in the valve when it closes. The permanent magnet slides in the valve body allowing the helium to flow around the holder. The volume of the valve is not dependent whether the valve is open or closed.

5.4 References


Chapter 6

A Search for a Supersolid Transition
Abstract

The aim of these measurements was a search for a superfluid state of solid $^4$He. The experiments consisted in a high accuracy measurement of the $^4$He melting curve with a very accurate capacitive pressure gauge. If there were a superfluid-like transition, it would be observed by a kink in the melting pressure due to the rapid change of the entropy in the system. Previous works by other authors are mentioned and compared with the measurements presented in this thesis. More accurate results on the $^4$He melting curve are shown, especially at low temperatures, where an anomaly has been seen. In the last section of this chapter, the possible candidates for this transition are discussed. This work was performed during a three months stay in the Interface Group in the Low Temperature Laboratory at Helsinki University of Technology.
Chapter 6

A Search for a Supersolid Transition

6.1 Searches for Supersolid

Theoretical studies suggest the possibility that in quantum solids (such as \(^4\)He) vacancies might exist even at zero temperature due to the large zero point motion \([1, 2]\). In \(^4\)He the zero point vacancies will behave like bosons and they are expected to Bose-condense giving superfluid-like properties to the crystal. Most experiments searching the evidence of supersolid concentrated on superfluid mass flow \([3, 4, 5]\). However, no proof of superflow was found. A clear review was presented by Meisel \textit{et al.} \([6]\).

Recent experiments with a torsional oscillator by Kim and Chan \([7, 8]\) have indicated a reduction of the rotational inertia in a cell containing solid \(^4\)He in Vycor and also in a cell containing only bulk \(^4\)He. This loss of rotational inertia was explained as a superflow of vacancies. These observations in bulk \(^4\)He were confirmed by Rittner and Reppy \([9]\) who pointed out that this effect was found only in non-uniform samples created at high temperatures and cooled down quickly. Rittner and Reppy also stated that the effect disappears once the samples are annealed \([9]\).

Van de Haar \textit{et al.} \([10]\) tried to detect the zero point vacancies measuring the melting pressure of \(^4\)He between 1.5 and 120 mK with a sensitivity of 20 \(\mu\)bar. No superfluid-like transition was found in this experiment and an upper limit for the density of vacancies in the solid of about 100 ppm was stated.

The melting pressure has contributions from different sources such as phonons, vacancies and impurities. These contributions are discussed in the following sections.

6.1.1 Contribution due to Phonons

The thermodynamic properties of solid \(^4\)He have been extensively studied in the past and are considered to be well known. These results can be used to estimate the phonon contribution to the melting pressure.
Below 0.5 K the heat capacity of the solid, $c_s$, can be described by a Debye $T^3$-term

$$c_s = \frac{12}{5} \pi^4 R \left( \frac{T}{\Theta_D} \right)^3$$

(6.1)

where $R = 8.3144 \text{ J mole}^{-1} \text{ K}^{-1}$ and $\Theta_D$ is the Debye temperature (approximately 26 K and temperature independent below 0.5 K [11]).

In the case of solid and liquid $^4\text{He}$ coexistence, the temperature dependence of the melting pressure is given by the Clausius-Clapeyron equation

$$\frac{dP_{MC}}{dT} = \frac{s_s - s_l}{v_s - v_l}$$

(6.2)

where $s_s$ and $s_l$ are the molar entropies of the solid and liquid $^4\text{He}$ respectively and $v_s - v_l$ is the difference in the molar volume of the solid and liquid $^4\text{He}$ ($v_s - v_l = -2.165 \times 10^{-6} \text{ m}^3/\text{mole}$ [12]).

The molar entropy of the solid can be derived from equation 6.1, giving an expression

$$s_s = \frac{4\pi^4 R}{5} \left( \frac{T}{\Theta_D} \right)^3$$

(6.3)

and the molar entropy of the liquid

$$s_l = \int \frac{c_l}{T} dT$$

(6.4)

where $c_l = 2 \times 10^{-2} T^3 \text{ J mole}^{-1} \text{ K}^{-4}$ is the specific heat of $^4\text{He}$ liquid at constant volume near the melting curve below 0.5 K [13].

Substituting all these expressions into equation 6.2, the following expression is obtained

$$\frac{dP_{MC}}{dT} = -0.146 \left( \frac{T}{K} \right)^3 \left( \text{bar} \right)$$

(6.5)

In other form

$$\Delta P_{MC} \equiv P_{MC}(T) - P_{MC}(0) = -0.0365 \left( \frac{T}{K} \right)^4 \text{ (bar).}$$

(6.6)
6.1.2 Thermally Activated Vacancies

The entropy of configuration $S_c$ of a perfect crystal at temperature $T$ and volume $V$ is given by

$$S_c = k_B \ln W_c$$  \hspace{1cm} (6.7)

where $k_B$ is the Boltzmann’s constant and $W_c$ is the number of distinguishable configurations for putting $N$ atoms in $N$ lattice sites.

If a certain number of vacancies $n$ is introduced to the crystal, then the configurational entropy is increased to

$$S_c = k_B \ln \frac{(N+n)!}{N!n!}$$  \hspace{1cm} (6.8)

which can be simplified using the Stirling’s formula ($\ln N! = N \ln N$) since $N$ is of the order of Avogadro’s number and $n \ll N$, to

$$S_c = k_B n \ln \frac{N}{n}$$.  \hspace{1cm} (6.9)

The crystal is in equilibrium when the free energy, $F = U - TS$ ($U$ and $S$ are the energy and the entropy of the crystal respectively), is minimized.

The free energy can be reduced in the crystal by forming vacancies and therefore increasing the entropy. This process increases the energy and the entropy by $u_v$ (vacancy formation energy) and $s_v$ (vacancy formation entropy) respectively

$$f_v = u_v - T s_v$$. \hspace{1cm} (6.10)

The free energy of the system is given by

$$F = F_0 + n f_v - T S_c = F_0 + n f_v - T k_B n \ln \frac{N}{n}$$  \hspace{1cm} (6.11)

which becomes minimal when

$$n = N \exp \left( - \frac{f_v}{k_B T} \right)$$. \hspace{1cm} (6.12)

Substituting this value into the previous expression for the configurational entropy (equation 6.9), the following formula is obtained

$$S_c = \frac{n f_v}{T} \exp \left( - \frac{f_v}{k_B T} \right) = \frac{n f_v}{T} \exp \left( - \frac{u_v - s_v}{k_B T} \right) = \frac{n f_v}{T} \exp \left( \frac{s_v}{k_B T} \right) \exp \left( - \frac{\phi}{T} \right)$$.  \hspace{1cm} (6.13)

where $\phi$ is the vacancy formation energy (K).
This activation energy $\phi$ has been measured by various authors [14, 15] using different techniques and it has a value of about 10 K near the melting pressure.

### 6.1.3 Zero-point Vacancies

The density of thermally activated vacancies rapidly decreases with temperature and vanishes completely at $T=0$ K. However, the zero-point motion of the lattice atoms can keep the vacancy density finite even at absolute zero.

Vacancies in solid $^4$He are expected to behave like $^3$He impurities in solid $^4$He [2]. Vacancies can tunnel through the crystal very fast and they cannot be described as localized impurities (like thermally activated ones) but by a wave-function [16].

While the atom-atom tunnelling rate in helium is of the order of $10^7$ Hz, the vacancy tunnelling rate is much higher, $10^{11}$ Hz [17, 18], due to less steric hinderance. Therefore vacancies are much more mobile than $^3$He impurities. Zero-point vacancies will behave as a dilute gas of bosons.

These zero-point vacancies behave as boson and therefore, at low enough temperatures, they undergo a critical temperature $T_c$ and bose condense. Assuming the pressure-temperature relation of an ideal gas [2] and $T > T_c$, the excess pressure due to the energy of the zero-point vacancies $P_{ZPV}$ can be expressed as

$$P_{ZPV} = \frac{x_{ZPV}}{V_s}RT$$  \hspace{1cm} (6.14)

where $x_{ZPV}$ is the fraction of zero-point vacancies.

The molar entropy of solid $^4$He has to increase due to the vacancies. The molar entropy of an ideal gas is given by [19]

$$S = \frac{3}{2}R \ln T + R \ln V - S_0$$  \hspace{1cm} (6.15)

where $S_0$ is a constant.

Applying this to the vacancies, it is assumed that the entropy of the zero-point vacancies vanishes at the Bose-condensation critical temperature $T_c$, so the entropy can be written as

$$s_{ZPV} = \frac{3}{2}x_{ZPV} \ln \frac{T}{T_c},$$  \hspace{1cm} (6.16)

where $s_{ZPV}$ is the excess molar entropy of solid $^4$He due to the zero point vacancies at $T > T_c$. 
Including the excess molar entropy of solid $^4$He due to the zero point vacancies, equation 6.5 becomes

$$\frac{dP_{MC}}{dT} = \frac{3}{2} x_{ZPV} R \ln \frac{T}{T_c} \left( \frac{T}{K} \right)^3 \left( \frac{\text{bar}}{K} \right) - 0.146 \left( \frac{T}{K} \right)^3 \left( \frac{\text{bar}}{K} \right).$$

These vacancies are expected to Bose-condense at low temperatures implying a non-classical behavior of the crystal such as a non-dissipative mass current, commonly known as superflow.

## 6.2 Experimental Setup

The cryostat consists of a dilution refrigerator in combination with a copper nuclear demagnetization stage, although this last one was not used in this experiment. The experimental cell is made out of copper and it has about 20 m$^2$ of silver sinter and a volume of 13 cm$^3$ (more details about the cell can be found in [20]). The cell is mounted on the copper nuclear stage, which is pre-cooled by the dilution refrigerator to a base temperature of about 10 mK. The cryostat has an optical setup for optical studies in both $^3$He [21] and $^4$He [22]. A scheme of the vacuum can of the cryostat can be seen in figure 6.1.

The capacitive pressure gauge is made out of copper-beryllium following the standard Straty-Adams design [23] with a flexible membrane as the sensitive element. One of the electrodes is glued on it. The other electrode is glued at a fixed position. The gluing was made when both electrodes were touching (25.25 bar in the cell). At melting pressure (25.31 bar) the sensitivity of the pressure gauge is $dC/dP = 44 \text{ pF/bar}$. The capacitance was measured by a commercial Andeen-Hagerling 2500A capacitance bridge. The time and temperature stability at zero pressure is of the order of $10^{-5} \text{ pF}$ (figure 6.2) during one week and over the temperature range of 10 mK to 0.4 K.

Temperature was measured by a $^3$He melting curve thermometer thermally anchored to the experimental cell. The $^3$He pressure was converted into temperature using the Provisional Low Temperature Scale, PLTS-2000 [24]. The calibration of the $^3$He melting pressure gauge was done at 1.1 K.

## 6.3 Results and Data Analysis

A $^4$He crystal was nucleated at a temperature of 1.1 K and rapidly cooled down below 0.6 K as in the experiments by Rittner and Reppy [9]. Crystals created at such a high temperature are known to have a significant amount of dislocations [25]. Thus, crystals can grow without applying a high overpressure [22].
Chapter 6. A Search for a Supersolid Transition

Figure 6.1: Scheme of the vacuum inner can [21].

The $^4$He melting pressure was measured in the temperature range of $10 - 500$ mK. The data shows the expected $T^4$-behavior being in a good agreement with earlier results (the melting curve of $^4$He measured by Grilly [12], the curve calculated from the measurements on the density of liquid $^4$He by Hanson et al. [26], and the curves calculated from the heat capacity measurements of solid $^4$He by Gardner et al. [11] and by Castles and Adams [27]) at high temperatures, as can be seen in figure 6.3.

The analysis of the experimental data can be followed in figures 6.4 and 6.5. First of all, the high temperature data (above 150 mK) is plotted versus $T^4$ (figure 6.4.a) in order to obtain the phonon contribution. A coefficient equal to 0.03531 bar/K$^4$ was found, which is in a good agreement with the values found by Gardner et al. (0.03520 bar/K$^4$) [11] and by van de Haar et al. (0.03571 bar/K$^4$) [10]. The obtained value changed depending the temperature range used for the fit. However, it was possible to set an error less than three per cent. Therefore, the contribution due to
6.3 Results and Data Analysis

The phonons was determined to be $0.035 \pm 0.001 \text{ bar/K}^4$. This coefficient was used in the rest of the analysis.

An extra linear term was used to fit the high temperature data. This linear term can be explained as the contribution of $^3$He impurities in the $^4$He liquid. The sample is commercial $^4$He with about 0.2 ppm (or less) $^3$He impurities [28]. The experimental cell was used before in $^3$He experiments, but it is expected to be clean of $^3$He contamination since it has been carefully cleaned and used for several experiments with pure $^4$He since then. These impurities contribute to the entropy of the liquid. Comparing this high temperature linear term and equation 6.17, which can be used as well here since $^3$He impurities behave as an ideal gas with a sign change, the number of $^3$He impurities can be obtained, yielding about 1 ppm in this experiment (see figure 6.4.b). This linear term will be also present in the low temperature data since it is impossible to get rid of the $^3$He impurities.

For the analysis of the low temperature data (below 150 mK), the coefficient for the phonon contribution ($T^4$-term) and the coefficient for the contribution of $^3$He impurities in the $^4$He liquid (linear term) found in the analysis of the high temperature data were used. However, an anomaly below $T=80 \text{ mK}$ was seen (see figure 6.5.a).

**Figure** 6.2: Time and temperature stability of the capacitive pressure gauge
Figure 6.3: Previous measurements of the $^4$He melting curve compared with the work presented in this thesis.

It was investigated whether adding a quadratic term due to a glassy state that may develop at low temperatures and is caused by a distribution of tunnelling systems in the crystal [29] was the cause of such an anomaly. This contribution turned out to be negligible and therefore was not considered as the reason for the low temperature anomaly.

The low temperature data was also fitted with an extra exponential term obtaining from the fit an activation energy of 35 mK, which is far too low for being the activation energy of vacancies, which is about 10 K [14, 15]. Thus, this exponential term has no physical meaning and it was not used in further analysis.

It can be seen in figure 6.5.b that after subtracting the $T^4$-term, a straight line due to the $^3$He impurities in the $^4$He liquid there is a clear change in the slope of this line around 80 mK, showing an unexpected anomaly in the solid. The reason for this extra (roughly linear) term in the low temperature data remains still unclear.
6.3 Results and Data Analysis

Y = P₀ + P₄ * X

<table>
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<th>Parameter</th>
<th>Value</th>
<th>Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>P₀</td>
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<td>5.1319E-8</td>
</tr>
<tr>
<td>P₄</td>
<td>-0.03531</td>
<td>1.46218E-5</td>
</tr>
</tbody>
</table>

Y = P₀ + P₁ * X + P₄ * X⁴

Chi²/DoF = 1.6463E-12
R² = 0.99967

<table>
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</tr>
<tr>
<td>P₄</td>
<td>-0.03531</td>
<td>0</td>
</tr>
</tbody>
</table>

Figure 6.4: a) High temperature ⁴He melting pressure plotted versus T⁴; b) High temperature data with linear term due to impurities.
Figure 6.5: a) Low temperature data. The solid line is the expected phonon contribution. Note that below T=80 mK the melting pressure presents an almost linear anomaly; b) $^4$He melting pressure after subtracting the phonon contribution.
Up to eleven different crystals nucleated in the temperature range between 0.6 and 1.4 K have been measured, showing all of them similar behavior. No supersolid transition has been seen and there is an anomaly below $T=80$ mK. This additional extra term in the melting curve implies that the entropy of the solid would increase with lowering the temperature, which strongly contradicts the thermodynamics.

6.4 Discussion

In this section the possible explanations for the observed low temperature anomaly are discussed.

The first thing one can think of is that the observed effect is caused by the pressure gauge itself. The geometrical sizes and the dielectric constant of the non-metallic parts of the gauge could be temperature dependent.

When temperature decreases, the sizes of all parts of the pressure gauge also decrease. Therefore, the measured capacitance should decrease as well, meaning that the pressure decreases instead of the increase of melting pressure observed below 80 mK. Besides, the linear thermal expansion coefficient of copper-beryllium is $10^{-5}$ K$^{-1}$ and in the temperature range from 0 to 100 mK the contraction of the capacitor plates is $\Delta L/L \approx 10^{-10}$. The relative change in the capacitance value is of the same order of magnitude, which is much lower than the accuracy of the gauge.

The possible change of the capacitance due to a temperature dependence in the non-metallic parts of the pressure gauge should be detected at constant pressure. The pressure gauge has been cooled down at zero pressure and no change in the capacitance value was found (see figure 6.2). The possible effect is smaller than $10^{-5}$ pF, which is the accuracy of the pressure measurements. This value corresponds to a pressure of about 0.02 µbar, which is by two orders of magnitude smaller than the observed effect of the transition near 80 mK.

Another possible reason for the anomaly of the $^4$He melting curve at low temperatures could be that the temperature measured with the $^3$He melting curve thermometer does not correspond to the temperature of the $^4$He contained in the experimental cell. At low temperatures the $^4$He melting curve is almost flat (and thus not sensitive to temperature changes) while the $^3$He melting curve has still a considerable slope. Therefore, the latent heat is very small in $^4$He, meaning that it cools down faster than the $^3$He. In this case, hysteresis should be observed between cooling and warming traces. In fact, this hysteresis has been seen. The measured melting pressure obtained by warming and cooling differs by 2 µbar but this effect is smaller than the anomaly. The pressure is slightly higher during cooling compared with warming. This
is probably due to some heat capacity of the experimental cell which induces a time delay in the cooling of the $^3$He. Several sweeps have been done at different cooling and warming rates and all of them show roughly the same hysteresis. Thus it can be concluded that the $^4$He experimental cell was in good thermal equilibrium with the $^3$He melting curve thermometer.

The possible effect of the hydrostatic pressure change is not completely negligible. The crystal had a volume of approximately 200 mm$^3$. The dependence of the pressure on the equilibrium crystal height is exponentially weak. The crystal grows from the sides, although a change of 0.3 mm in the height of the crystal would correspond already to a change in pressure of 6 µbar, which is about the observed deviation. However, the reason why this effect would be only observed below 80 mK is not clear. The hydrostatic pressure difference between the crystal and the pressure gauge changes only a little due to the growth or melting. The possible deviation from the equilibrium shape due to the growth thresholds on facets depends on the density of dislocations. For a reasonable amount of dislocations per facet (few tens) the deviation results in an overpressure lower than 1 µbar [22]. Moreover, the effects of the shape and growth should be stronger at higher temperatures where the melting curve is steeper.

The observed linear term in the low temperature tail of the $^4$He melting curve could be explained by a concentration of $1\,\text{--}\,2$ ppm of $^3$He impurities in the crystal. This explanation faces also some problems since the effect should have opposite sign. The linear dependence corresponds with an excess of entropy of about $1.5 \times 10^{-6} R$ in the solid. In other words, the entropy increases when the solid undergoes the transition, which violates the classical thermodynamics.

### 6.5 References


[28] pure 4He supplied by Oy Woikoski AB, Huvudkontor, 47901 - Vuohijrvi, Finland
Summary

The main goal of the research described in this thesis is to improve the understanding of the growth and melting kinetics of $^3$He crystals, and determine how the magnetic ordering of the solid affects the growth mechanisms. Several experiments at temperatures between $0.65 \text{ mK}$ at zero magnetic field and $2.5 \text{ mK}$ at $B=8 \text{ T}$ were performed with a dilution refrigerator in combination with a copper nuclear demagnetization stage to achieve the results presented in this thesis. Images of $^3$He crystals at these temperatures were taken, so different types of facets could be followed during the growth and melting sequences.

Chapter 1 is a short introduction to low temperature physics. Some general effects that only become relevant at low temperatures are described. This chapter gives also the motivation for the work presented in this thesis, discussing briefly previous experiments on $^3$He crystals and the crystallization waves found for solid $^4$He. In this chapter a guideline through this thesis is also presented.

Chapter 2 gives an introduction to the properties of both liquid and solid $^3$He. The phase diagram at melting pressure in the presence of magnetic field is discussed. This is followed by the theory of the crystal growth in the presence of screw dislocations. The weak and the strong coupling between the interface and the crystal lattice is also presented. This chapter ends with a discussion about the equilibrium shape of the crystals.

Chapter 3 introduces the experimental setup. The cryostat consisted of a dilution refrigerator built by Prof. Giorgio Frossati 25 years ago in combination with a nuclear demagnetization stage. After many years of operation, the performance of the cryostat had degraded, so a complete check-up as well as new parts were needed for the success of the experiments. An optical fiber going from the experimental room to the vacuum inner can of the cryostat has been mounted, and all the lenses, cold mirrors and filters have been changed, so images of the $^3$He crystals can be taken. The quality of these images has improved very much since these changes were made. The heat switches connecting thermally the mixing chamber of the dilution refrigerator and the copper demagnetization stage were partially broken, so new heat switches were constructed. They consist of a copper-aluminium-copper sandwich diffusion welded...
together. A bulky piece of aluminium was used as the superconducting material instead of the standard tin foils. At the end of the chapter, the experimental cell is also presented. It is a Pomeranchuk cell which can hold a pressure difference between the two volumes up to 23 bar. In the inner $^3$He volume there are a sapphire pressure gauge to monitor the $^3$He pressure, a vibrating wire and a speer resistor used as thermometers and a heater used to nucleate the $^3$He crystals. The experimental cell also had very pure quartz optical windows, so we had optical access to the cell during the growth and melting experiments.

Chapter 4 presents the experimental method used for the growth of $^3$He crystals and the analyzing techniques followed for the understanding of these measurements. A computer program that reconstructs the 2D images of the crystals into a 3D wireframe has been developed. The effective growth coefficients of different types of facets have been measured for $^3$He crystals at different magnetic fields ($B=0$ T, $B=0.8$ T, $B=2$ T and $B=8$ T). The values obtained for the crystals at zero magnetic field agree with previous values that can be found in the literature. However, when the magnetic field is increased and the solid magnetically orders in the CNAF phase, the effective growth coefficient of the $<110>$ facet increases by almost an order of magnitude. This was completely unexpected since the $<110>$ facet has the largest interplanar distance and it was expected to be the slowest facet also in the presence of a magnetic field. It was interpreted that the interface and the crystal lattice seem to be in the weak-coupling limit when the magnetic field is increased, while they couple strongly at zero magnetic field.

Chapter 5 shows the new improvements made on the cryostat after the realization of the experiments described in chapter 4. New heat switches providing better thermal contact between the mixing chamber and the nuclear stage were built since the previous ones broke down at the diffusion welding plane after a quench of the demagnetization magnet. They are basically the same design as the ones described in chapter 3, but with the difference that this time, the copper-aluminium-copper sandwich was explosion welded, becoming a more rigid device. The sapphire pressure gauge inside the body of the experimental cell was not sensitive enough for the purposes of these studies and therefore, a more sensitive titanium pressure gauge was designed and constructed with 30 times more sensitivity than the sapphire one. Finally, an electrically actuated cold valve has been built for closing off the $^4$He filling line near the experimental cell, so thermal and pressure fluctuations will be avoided and the crystal will go to its equilibrium shape without any external perturbation.

Chapter 6 describes the experiments carried out in the Interface Group in the Low Temperature Laboratory in the Helsinki University of Technology during a three months stay. The experiment concerns a presently hot topic: the possible observation of superfluidity in solid $^4$He. Interest in the problem of supersolid was renewed with the observation by Kim and Chan in 2004 of the reduction in the rotational inertia of a cell containing solid $^4$He below about 0.2 K. The authors named this
phenomenon “nonclassical rotational inertia” and interpreted it in terms of superfluidity of the solid, possibly caused by Bose-Einstein condensation of vacancies. In this chapter, direct high-precision measurements of the melting pressure of $^4$He in the range from 10 to 400 mK are presented. No indication of a supersolid transition was found. For $80 < T < 400$ mK the temperature behavior of the melting pressure is fully determined by the phonons giving a $T^4$-dependence. For $T < 80$ mK a deviation is seen which is not consistent with a transition in the solid since the excess entropy in the solid decreases with increasing temperature. An upper limit for the number of zero-point vacancies (1 ppm) has been set after the realization of these experiments.
Samenvatting

Het belangrijkste doel van het onderzoek dat in dit proefschrift wordt beschreven is het vergroten van het begrip van groei- en smaltprocessen van $^3$He kristallen. Daarbij wordt in het bijzonder de invloed van magnetische ordering op de groeiprocessen onderzocht. De metingen zijn verricht met een mengkoelmachine in combinatie met een koperen kerndemagnetizatietrap, bij temperaturen tussen 0.65 mK zonder magnetisch veld en 2.5 mK bij $B=8$ T. Bij deze temperaturen zijn afbeeldingen van $^3$He kristallen gemaakt, zodat de verschillende facettypes tijdens groeien en smelten kunnen worden bekeken.

Hoofdstuk 1 geeft een korte inleiding tot enkele aspecten van lage temperatuurfysica. Sommige fysische effecten die slechts bij lage temperaturen relevant zijn worden beschreven. Dit hoofdstuk geeft ook de motivatie voor het werk dat in dit proefschrift wordt voorgesteld en bespreekt kort vorige experimenten aan $^3$He kristallen en de kristallisatiegolven die in vast $^4$He zijn gevonden. Tot slot biedt het een overzicht van dit proefschrift.

Hoofdstuk 2 geeft een inleiding tot de eigenschappen van zowel vloeibaar als vast $^3$He. Het fasediagram bij smeltdruk in aanwezigheid van een magnetisch veld wordt besproken. Dit wordt gevolgd door de theorie van kristalgroei in aanwezigheid van schroefdislocaties. De zwakke en sterke koppeling tussen het grensvlak en het kristalrooster wordt ook besproken. Dit hoofdstuk eindigt met een bespreking van de evenwichtsvorm van kristallen.

Hoofdstuk 3 introduceert de experimentele opstelling. De cryostaat bestaat uit een mengkoelmachine in combinatie met een kerndemagnetizatietrap. Na vele jaren van gebruik funtioneerde de cryostaat niet meer optimaal, zodat een volledige check-up evenals nieuwe onderdelen vereist waren voor de hier bescheven experimenten. Er is een optische fiber aangebracht van de meetruimte naar de binnenste vacuüm kamer van de cryostaat. Vervolgens zijn alle lenzen, koude spiegels en filters veranderd, zodat afbeeldingen van de $^3$He kristallen konden worden gemaakt. De kwaliteit van de beelden is sterk verbeterd nadat deze veranderingen waren aangebracht. De warmte schakelaars die de mengkoelmachine en de kerndemagnetizatietrap thermisch verbinden, waren gedeeltelijk gebroken, zodat nieuwe werden geconstrueerd. Zij
Samenvatting

bestaan uit een koper-aluminium-koper verbinding die met een diffusie-las proces aan elkaar gesmeed zijn. In plaats van de standaard tin folies werd een massief aluminium blokje gebruikt als supergeleidend materiaal. Aan het eind van het hoofdstuk wordt de experimentele cel nader toegelicht. Het is een Pomeranchuk cel die een drukverschil tussen de twee volumes tot 23 bar kan weerstaan. In het $^3$He volume bevindt zich een drukmeter van saffier om de $^3$He druk te meten, evenals een trildraad en een koolstof-weerstand die als thermometers worden gebruikt, en een stookdraadje om nucleatie van vast $^3$He op een vaste plaats te initiëren. De experimentele cel heeft vensters van zeer zuiver kwarts, zodat de kristallen, en dus de groei- en smeltprocessen, bekeken worden.

Hoofdstuk 4 bespreekt de experimentele aanpak die voor de groei van $^3$He kristallen werd gebruikt en behandelt de analyse van de meetresultaten. Er is een computer-programma ontwikkeld dat de 2D-projecties van de kristallen terugrekent naar de 3D vorm. De effectieve groeicoëfficiënten van verschillende facettypes voor $^3$He kristallen zijn bij verschillende magnetische velden gemeten ($B=0$ T, $B=0.8$ T, $B=2$ T en $B=8$ T). De waarden gevonden bij $B=0$ T komen overeen met de waarden uit de literatuur. Wanneer het magnetische veld wordt verhoogd en vast $^3$He magnetisch in de $CNAF$-fase komt, neemt de effectieve groei coëfficiënt van de $<110>$ facetten met bijna een orde van grootte toe. Dit was volledig onverwacht, aangezien het $<110>$ facet de grootste interplanaire afstand/staphoogte heeft en verwacht was dat het het langzaamste facet zou blijven, zelfs in aanwezigheid van een magnetisch veld. Dit kan een aanwijzing zijn dat het grensvlak en het kristalrooster zwakker gekoppeld lijken te zijn wanneer het magnetische veld toeneemt, terwijl zij bij $B=0$ T sterk koppelen.

Hoofdstuk 5 toont de verbeteringen na de experimenten uit hoofdstuk 4. Er zijn nieuwe warmte schakelaars gemaakt, die mechanisch sterker zijn. Het ontwerp van deze thermische schakelaar is hetzelfde als dat in hoofdstuk 3, maar dit keer zijn de koper-aluminium-koper lagen verbonden met de techniek van explosie-lassen, wat leidt tot een steviger constructie. De gevoeligheid van de saffieren drukmeter in de cel bleek niet goed genoeg voor de studies in hoofdstuk 4 en daarom werd een titanium drukmeter ontworpen met een factor 30 grotere gevoeligheid dan de vorige. Ook is een elektrisch aangedreven kraan gebouwd om de $^4$He vullijn dichtbij de experimentele cel af te sluiten. Daarmee worden temperatuurvariaties en drukschommelingen vermeden en kan het kristal zonder externe storing zijn evenwichtsvorm aannemen.

Hoofdstuk 6 beschrijft de experimenten die in de Interface Group in het Low Temperature Laboratory van de Helsinki University of Technology zijn uitgevoerd, waaraan ik heb bijgedragen tijdens een verblijf van drie maanden. Het experiment betreft een actueel onderwerp namelijk de mogelijke observatie van supersoliditeit in vast $^4$He. De interesse voor het probleem van supersoliditeit werd in 2004 nieuw leven ingeblazen met de observatie door Kim en Chan van de vermindering van het traagheidsmoment van een cel die vast $^4$He benedien ongeveer 0.2 K bevatte. De auteurs noemden dit fenomeen non-classical rotational inertia en interpreteerden het
in termen van superfluïditeit van het kristal, die misschien wordt veroorzaakt door Bose-Einstein condensatie van de gaten in het rooster. In dit proefschift worden zeer precieze metingen van de smeltduik van $^4$He tussen 10 en 400 mK gepresenteerd. Van 80 tot 400 mK werd geen afwijking waargenomen van het verwachte $T^4$-gedrag t.g.v. van de fononen. Echter, beneden 80 mK werd een onverwachte afwijking van het $T^4$-gedrag gezien, die echter niet aan een superfluïde overgang toegeschreven kan worden, aangezien de entropie in de vaste stof afneemt bij stijgende temperatuur. Wel kon uit onze experimenten een bovengrens bepaald worden voor het aantal roostergaten bij het absolute nulpunt (1 ppm).
Curriculum Vitae

Juan Bueno was born in Santander, Spain, in 1979. He studied physics at the Faculty of Science of the University of Cantabria. From April to October 2002 he worked as an Erasmus student in the Low Temperature Division, Technical Faculty of Physics, University of Twente (The Netherlands) under the supervision of Dr. Ir. Jaap Flokstra and Dr. Martin Podt designing and fabricating Superconducting Quantum Interference Devices (SQUIDs) used by the MiniGRAIL Project, the first spherical gravitational wave detector. After that he got his Master Degree in Physics, Specialization in Solid State Physics.

From November to December 2002 he worked in the Gravitational Wave Research Group in the Laboratori Nazionali di Frascati (LNF), Instituto Nazionale di Fisica Nucleare (INFN), Italy, under the supervision of Prof. E. Coccia, helping with the assembly of the transducers for Nautilus and Explorer gravitational wave antennas.

In January 2003 he started his PhD project at the Kamerlingh Onnes Laboratory of the University of Leiden, The Netherlands, under the supervision of Dr. Reyer Jochemsen. His research concerned the study of the growth and melting kinetics of $^3$He under the influence of an external magnetic field and resulted in the work presented in this thesis. Part of his research was carried out at the Low Temperature Laboratory of the Technical University of Helsinki, Finland, under the supervision of Dr. Harry Alles, where he helped in an experiment searching for supersolid $^4$He. During his stay at Leiden University he was also supervising several bachelor and master degree students.

He also wrote a proposal titled “Feasibility Study on SQUID MRI at Low Magnetic Fields”, which was selected among the best three in the ’Win de toekomst’ competition (November 2006) organised by FOM (Fundamenteel Onderzoek der Materie).
Participation to scientific events

November 2006  Physics Preview Lectures, 
Leiden, The Netherlands. *(oral presentation on invitation)*

April 2006  Quantum Phenomena at Low Temperatures, 
ULTI users meeting, Lammi, Finland. *(poster presentation)*

December 2005  Dutch Condensed Matter Physics meeting, 
Veldhoven, The Netherlands. *(oral presentation)*

November 2005  Physics Preview Lectures, 
Leiden, The Netherlands. *(oral presentation on invitation)*

August 2005  Ultra Low Temperature Conference, 
Gainnesville, Florida, USA. *(oral presentation on invitation)*

August 2005  24th International Conference on Low Temperature Physics, 
Orlando, Florida, USA. *(two posters and oral presentation)*

January 2005  Dutch Statistical Physics meeting, 
Lunteren, The Netherlands. *(oral presentation)*

July 2004  International Symposium on Quantum Fluids and Solids, 
Trento, Italy. *(poster presentation)*

January 2004  Dutch Statistical Physics meeting, 
Lunteren, The Netherlands. *(poster presentation)*

August 2003  International Symposium on Quantum Fluids and Solids, 
Albuquerque, New Mexico, USA.

January 2003  Dutch Statistical Physics meeting, 
Lunteren, The Netherlands. *(poster presentation)*
List of Publications

1. Growth of $^3$He Crystals at Different Magnetic Fields
   J. Bueno, R. Blaauwgeers, A.Y. Parshin, and R. Jochemsen
   in preparation

2. Melting Curve of $^4$He: No Sign of the Supersolid Transition down to 10 mK

3. A Magnetically Driven Cold Valve for $^4$He
   J. Bueno, R. Blaauwgeers, R. Partapsing, I. Taminiau, and R. Jochemsen

4. A Small and Very Sensitive Titanium Pressure Gauge for $^3$He Melting Pressure Measurements in Magnetic Field
   J. Bueno, R. Blaauwgeers, R. Partapsing, I. Taminiau, and R. Jochemsen

5. Analysis of $^3$He Crystal Images Using a Computer-Generated Wire-Frame
   D.M.T. Dekker, R. Wagner, R. Blaauwgeers, J. Bueno, and R. Jochemsen
   J. Low Temp. Phys. 139, 509 (2005)

6. Improved Optical Setup to Measure Growth and Melting of Solid $^3$He Below 1 mK in Magnetic Fields up to 9 Tesla
   R. Blaauwgeers, J. Bueno, D. Kleinmans, S. Leerink and R. Jochemsen
Acknowledgements

The work presented in this thesis would have never been possible without the contribution and constant support of many people.

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