#### Growing <sup>4</sup>He crystals

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#### Abstract

<sup>4</sup>He crystal growth was observed using an interdigital capacitor, an external pressure sensor and an internal pressure sensor. The internal and external pressure sensors were used in the measurement of overpressurization and constant pressure regimes. These regimes are indicators of <sup>4</sup>He crystallization. The interdigital capacitor was used as a level detector to study the edge height of the <sup>4</sup>He crystal.

## Introduction

W. H. Kessom<sup>1</sup> first observed solid <sup>4</sup>He (one of the isotopes of helium) in 1926, but it wasn't until the 1980's that physicists began to study its surface properties. One of the results in the study of solid 4He's surface is the observation of stress induced surface instabilities. We are growing <sup>4</sup>He crystals in order to study stress driven surface instabilities

This paper describes the first phase of a study of stress driven surface instabilities. To study such instabilities, it is necessary to develop a reproducible method for growing crystalline <sup>4</sup>He and to have reliable measuring devices for the monitoring the properties of the crystals grown.

#### Motivations for studying solid helium

#### Solid helium approaches an ideal crystal

Solid <sup>4</sup>He approaches an ideal crystal because: it can be made chemically and isotopically pure; its bonding is due only to the Van der Waal force; its lattice structure is uniform throughout in a given phase; it can be grown with very few lattice defects and its crystal growth is fast in comparison to that of other solids.

#### <sup>4</sup>He has an unusual phase diagram

The most notable property of <sup>4</sup>He is that at low pressure it will not solidify as its temperature approaches absolute zero. This is a property unique to helium and can be seen in the following phase diagram. In the phase diagram below, solid helium is

above the line and liquid helium is below the line. The phase diagram (Figure 1) also indicates that at solid <sup>4</sup>He's boundary it will be in contact with its liquid state.

550 500 Pressure(PSI) 450 He I solid helium 400 He II 350 0.4 1.2 1.4 2.0 0.6 0.8 1.0 1.6 1.8 T(K)

Figure 1. PT phase diagram for <sup>4</sup>He at the temperature range our experiment runs at.

## Crystal growth in solid helium is essentially a mechanical process

The Clausius-Clapeyron relationship associates the slope of the PT phase diagram to the latent heat (L) divided by the product of the change in volume ( $\Delta V$ ) and temperature (T) of the material according to.

$$\frac{dp}{dT} = \frac{L}{T\Delta V}$$

where the latent heat is the heat per amount of mass added or removed during phase change. For <sup>4</sup>He at low temperatures its latent heat approaches zero. With the latent heat being almost zero the freezing pressure is independent of temperature as understood in the Clausius-Clapeyron relation. "As a result we should consider freezing to be essentially a mechanical process, rather than a thermal process as it is for ordinary materials"<sup>2</sup>

When crystals other than helium are grown in an ordinary liquid phase the latent heat is liberated at the liquid/solid interface. This release in energy creates a rise in

local temperature. The heat must be conducted away from the material or crystallization will cease. The growth of the crystal is predominantly controlled by heat conduction in the bulk phases and the kinetic processes that occur at the interface are secondary. This is not the case for helium. If a small amount of heat is generated, it is dissipated by superfluid convective flow. "The combined effect of high thermal conductivity and low latent heat is that the growth of a helium crystal is limited primarily by processes occurring at the interface, making the kinetics of growth easy to study."<sup>3</sup>

## **Experimental procedure**

**Cooling down**: To make helium crystals in the region where the change in pressure divided by the change in temperate is approximately zero we lower its temperature from 300K to 1.2K. This is done by immersing the helium in a very cold bath. The apparatus used to cool the experimental cell down to that temperature is called a dewar. The dewar in Figure 2 consists of two tubes with sealed bottoms one placed inside the other. The experimental chamber sits in the inner dewar. The dewars are basically like two glass thermos bottles, one placed inside the other. The inner dewar is connected to a large vacuum pump and is sealed to the air.



Figure 2

To cool the inner dewar and experimental cell down to 1.2K the inner dewar and the cell are precooled by filling the outer

dewar with liquid nitrogen and the inner dewar with nitrogen gas. The heat from the inner dewer and cell is then conducted outward, eventually lowering the temperature to 77K. The nitrogen gas in the inner dewar is then pumped out and replaced with liquid helium, a process that lowers the experimental apparatus temperature to 4K. The experimental cell's temperature is then lowered to 1.2K by using the evaporation of the liquid helium in the inner dewar for cooling. Specifically, the evaporation is forced by pumping away the helium vapor from the inner dewar, which allows more helium atoms to move out of the liquid state. The pumping on the liquid helium is done by slowly opening the valves connected to a vacuum pump. When the valves are open all the way and the pumping rate is maximized, the dewar attains its lowest possible temperature.

**Pressurizing**: The gas handling panel used for the pressurization procedure is shown in a photograph in Figure 7and schematically in Figure 8. The process to crystallize helium is done in three steps. To begin the crystallization process, helium gas from the pressure panel is released into the cell at a fine rate, the cell is at 1.2K and very low pressure. At 1.2K and a pressure greater than 0.012 psi helium gas begins to condense in the cell. We increase the flow rate as the cell begins to fill with liquid helium until the entire cell is filled. We then increase the pressure of the cell by about 0.5 psi per second until the cell's pressure is 360 psi. In the final step we close

off the dipstick (see Figure 10) to the cell and use the pressure regulator (see Figure 9) to take the cell up to about 369 psi.

**Measuring 'He solid**: The measuring devices we used in the helium crystallization project were an external (see Figure 2) and internal pressure sensor, an interdigital capacitor (see Figure 3) and thermometers (see Figure 6) placed in the cell and bath. The internal and external pressure sensors were used to measure in the overpressurization and constant pressure regimes (see below) during crystallization. The interdigital capacitor measured the capacitance of the helium. The thermometers measured the temperature of the bath and cell.

**External pressure sensor**: Figure 2 shows the external pressure sensor model 740 made by Paroscientific Inc.



**Interdigital capacitor**: The interdigital capacitor (see Figure 4a) senses the changes of the effective dielectric constant of the material deposited in its vicinity. The capacitance increases as the density of the helium increases. The interdigital capacitor was placed in the cell at one end. When solid <sup>4</sup>He is forming, the interdigital capacitor detects growth only along the surface of the crystal. (see Figure 4b)



Figure 4a



Figure 4b

**Internal pressure sensor**: The internal pressure sensor is the copper cylinder in the top right corner seen in Figure 5 and is connected to the experimental cell by tubing. Inside the copper cylinder are two parallel conducting plates that change distance as the pressure changes inside the cell. The two plates act as a parallel plate capacitor and the capacitance between the two plates changes as the plates move closer or farther apart due to an increase or decrease in pressure.



Figure 5



#### The thermometers: Carbon

resistors are used as thermometers for measuring the helium bath and cell temperature. The resistor works as a very good thermometer because as the resistor's temperature decreases its resistivity increases exponentially. Thus, at low temperatures slight changes in temperatures cause large changes in resistance, which makes the resistors very sensitive thermometers at low temperatures. In Figure 6 the small resistor is the bath thermometer used to measure the bath temperature and the larger wire wound resistor is used as a heater. The cell thermometer is not visible in the figure.

Figure 6

**The Pressure Panel**: To crystallize helium slowly we needed to regulate the flow rate of helium gas. The pressure panel was used to regulate the flow of helium, to filter the impurities out of the <sup>4</sup>He before it went into the experimental chamber and to raise the pressure from 0-369 psi. The pressure panel consists of 4 elements: the charcoal filter, the pressure regulator, the dipstick and a series of valves that allowed for routing the gas through the different elements and to the experimental chamber.



Figure 7



Figure 8. Schematic for the pressure panel used in crystallizing helium.

The charcoal filter: The charcoal filter is used to remove condensable contaminants from the helium before it enters the experimental cell. To do this the filter is submerged into a liquid nitrogen bath. At this temperature water vapor, CO<sub>2</sub>, AR and various other gases in the <sup>4</sup>He are captured in the charcoal's matrix. In Figure 9 the charcoal filter is the small steel cylinder at the bottom center of the image. The two steel tubes entering the cylinder are in and out lines. The in line is from the helium source and the out line is to the experimental chamber.

The pressure regulator: The pressure regulator (see Figure 10b) is used for two purposes: to keep a desired pressure in the experimental chamber and to make small changes in the pressure during the crystallization of helium. It does these two activities by raising and lowering the temperature of the gas inside a container that is connected to the valve system. The container is placed inside a metal jacket that is sealed to the outside and maintained at low pressure. The pressure regulator's temperature is controlled by a heating element wrapped around the container's body and by having the container in thermal contact with the metal jacket, which is submerged in a liquid nitrogen bath. Figure 10a shows the pressure regulator submerged in a liquid nitrogen bath.





Figure 10b

Figure 9

Figure 10a

8



Figure 11

**The dipstick**: The dipstick is used to increase and decrease pressure in the experimental cell. To change the pressure the dipstick is raised or lowered into a helium bath. The dipstick is essentially a metal tube filled with charcoal that is connected to a helium supply. To fill the dipstick it is lowered into the helium bath and as the helium is cooled it condenses. The helium will continue to condense and as it condenses it will draw helium in from the supply source until thermodynamic equilibrium is attained. At this point the connection to the source is closed off and the connection to the experiment is opened up. The dipstick is then raised slowly out of the helium bath so that as it warms up the helium evaporates and pressure begins to rise. In Figure 11 the dipstick is about to be submerged in the liquid helium bath.

# **Preliminary results**

Figure 12 shows strip chart recordings as a function of time of data taken during a crystal growth event. The total time period is approximately 200 seconds. The top graph is a measurement of a change in capacitance of the interdigital capacitor over a period of time. The second graph depicts a change in the pressure over the entire fill line measured by the external pressure sensor. The experimental cell's change in pressure is measured by the internal pressure sensor and is shown in the third graph. The last two graphs show the changes in the bath resistor and cell resistor readings. Capacitance and pressure were measured over time; and it is the magnitude and direction of these changes over time rather than absolute values that were used as an indication of crystal growth. Overpressurization and subsequent drop in pressure due to crystal nucleation (see second and third graph in Figure 12), a change in capacitance due to the increasing density of the material (see first graph in Figure 12), and constant pressure during crystal growth were the changes of properties used to confirm the presence of a crystal.



**Pressure during crystal growth**: The difference between the pressure needed to initiate crystallization (overpressurization) and the equilibrium melting pressure (Figure1) varies in magnitude from run to run of the experiment. In general, the attainment of the equilibrium pressure, alone is insufficient to indicate an initiation of crystal growth. The second tool for detecting the presence of a crystal is a prolonged state of constant pressure during the growth process. By definition, equilibrium crystallization requires that the chemical potential of the solid state be equal at all times to the chemical potential of the liquid state. The chemical potential of the gas phase is directly related to its pressure. The second and third graphs in Figure 12 show constant pressure over a long period of time after the initial overpressurization. Thus, the observation of constant pressure in both pressure sensors indicates crystal growth is occurring.

**Overpressurization**: Crystal nodules may appear and dissolve as a result of random fluctuations in pressure above the (equilibrium) melting pressure. When super fluid <sup>4</sup>He is overpressurized; at some point one of the many crystal nodules begins to grow. On reaching a certain critical volume, it becomes energetically more favorable for the crystal to continue growing at the equilibrium temperature and somewhat lower equilibrium pressure. Thus, once a nucleation seed is "born", the

pressure of the system may fall back down to the melting pressure. The overpressurization is a hysteretic process from solid to liquid associated with a first order transition. These observations explain why crystal nucleation occurs at a pressure higher than indicated by the equilibrium PT diagram for a given melting temperature. Figure 13 shows the abrupt change that takes place after nucleation has occurred. Note that the pressure drop occurs in a time shorter than the data acquisition time (1sec.).



Measurements were taken during three periods of the crystallization process: condensation, pressurization and crystallization. The measurements made during condensation and pressurization stages were used to calibrate the measurements made during crystal growth.

The pressure measurement taken during crystal growth followed expected predictions of overpressurization followed by growth at constant pressure. However, the time dependence of the interdigital capacitor readings was very different from what was expected. We expected a constant increase in capacitance; instead, the measurement shows a stepped increase, then a decrease, then a stepped increase in capacitance (Figure 14). After several different runs we came to the conclusion that a wire connecting the capacitor to the readout equipment was moving and affecting the capacitance output. The wire configuration was changed and as shown in Figure 15 the capacitance during crystallization followed expected patterns.



**Changes in capacitance**: Figure 16 shows the output of a lock-in amplifier, which is proportional to the changes of the capacitance of an interdigital capacitor (IDC). The interdigital capacitor is in contact with the helium in the experimental cell. As the height of the solid helium in contact with the IDC increases, the capacitance measured by the interdigital capacitor increases. Helium's solid phase is denser than its liquid. When the solid layer is in contact with the interdigital capacitor the capacitance increases. The decrease in output voltage indicates melting of the crystal.



## **Future work**

The interdigital capacitor will play a crucial role in future measurements of stress driven instability because it is possible to extract crystal height from its capacitance output. At present during condensation the interdigital capacitor exhibits an unexplained drop in capacitance. Future work will address this issue.

## References

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2 H. J. Maris, and A. F. Andreev, "The surface of crystalline helium-4", Physics Today, Feb.25 (1987).

3 ibid.

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