Critical Overpressure for Nucleation of $^4\text{He}$ Crystals in Aerogel

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Abstract. In an aerogel contained in a flat glass tube, we measured critical overpressures at which the very first $^4\text{He}$ crystal appeared. Crystallization and melting were repeated 50 times at a constant temperature in a variable volume cell and the nucleation probability as a function of overpressure was obtained. An anisotropic S shape characteristic of the nucleation process was successfully observed.

1. Introduction

Although two states coexist in an equilibrium condition, metastability of matter is often observed in the first order phase transition due to the interfacial energy between two phases. By sweeping a thermodynamic parameter, a new stable state nucleates in the original state away from the equilibrium condition when the energy barrier becomes small enough. In ordinary classical processes, the nucleation results from the thermal activation at high temperatures. At low temperatures, thermal activation ceases and a quantum tunneling process may play a role in the nucleation. Condensed phases of He have been a good playground for the study of both quantum and thermal nucleation[1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13]. Here we report the metastability and critical overpressure of $^4\text{He}$ crystals in an aerogel. The melting pressure of $^4\text{He}$ is known to shift to a higher pressures in porous materials but the nucleation has not been studied in details.

2. Experimental

Usually $^4\text{He}$ crystals are grown by a blocked capillary method in porous materials and their melting pressure is found to shift to higher pressure[14]. By this method it is not possible to investigate the nucleation process in detail because the crystallization pressure or temperature are mostly determined by the initial pressure of liquid $^4\text{He}$ in the cell. We adopted a Pomeranchuk-type variable-volume cell in order to investigate the nucleation process at various temperatures[15, 16]. The sample cell consisted of two chambers, a high-pressure (A) and a low-pressure (B) chamber as shown in Fig. 1b in Ref. [16]. Part of both chambers was made of bellows which were connected by a rigid rod. We were able to control the volume of chamber A by the liquid in chamber B thanks to the larger cross section of the bellows of chamber B. Pressure of chamber A was measured by a capacitive pressure gauge whose diaphragm was made of BeCu and constituted a part of the chamber wall. Pressure in chamber B was measured by a
pressure transducer at room temperature. Chamber B was pressurized at a fixed rate adjusted by a mass flow controller. The cryostat and chamber A had two optical windows aligned in a straight line. We illuminated through the back window by a parallel light (Xenon lamp, USHIO UXL-151DO) and recorded the images through the front window by a high-quality CCD camera (KEYENCE VW-100C) at room temperature.

We used an aerogel with a porosity of 98% grown directly in a thin glass tube. The tube was 10 mm in height, 8 mm in width and 1 mm in depth; the bottom was glued to a glass plate and only the upper surface was open. Aerogel was placed in chamber A and crystals were grown there at a constant temperature. $^4$He was condensed in chamber A above the critical temperature to avoid damage in the aerogel by the liquid-gas interface. Chamber A was pressurized to the melting pressure, and bulk crystals and superfluid liquid coexisted in it. At this stage, only the superfluid occupied the aerogel and overpressure would be needed to grow crystals in the aerogel. Thereafter, chamber B was continuously pressurized at a fixed rate and temperature, and the bellows of chamber A shrank. The bulk crystals outside the aerogel grew in chamber A and eventually occupied the outside. The pressure in chamber A began to increase above the bulk melting pressure and resulted in the nucleation of the first crystals in the aerogel; we recorded the critical overpressure $\delta P_c$ in the chamber at which that first occurred. After the crystals filled the aerogel, we depressurized chamber B until the crystals in the aerogel were completely melted. This sequence was repeated 50 times at 1.0 K to see whether we could observe the metastability of the superfluid liquid in the aerogel and the distribution of $\delta P_c$ characterizing the nucleation process.

![Figure 1](image-url)

**Figure 1.** Crystallization process of $^4$He in aerogel in the high temperature creep region. Profiles of the crystal-superfluid interface were drawn at 0, 100, 200, 300, 450 and 600 s. The interface appeared from the lower part of the aerogel in (b), moved smoothly and eventually filled the aerogel. Similar movie was recorded during every pressure sweep to identify the time at which the first crystal appeared in the aerogel.
Figure 2. Typical pressure sweeps in chamber A to grow crystals in aerogel at 1.0 K. Origins of time are arbitrary. Open circles are the overpressure measured by the capacitive pressure gauge. The filled circles are the critical overpressure $\delta P_c$ at which the first crystal appeared in the aerogel. $\delta P_c$ was different in each sweep and had a rather broad distribution.

Figure 3. Cumulative distribution of critical overpressure for $^4$He crystal nucleation in aerogel at 1.0 K. The vertical axis is $\Sigma = \frac{N_0 - N}{N_0}$, $N$ is the number of nucleation events which occurred at an overpressure less than $\delta P$, and $N_0 = 50$ is the total number of the pressure sweeps. An anisotropic S shape characteristic of the nucleation process was observed.

3. Results
Profiles of the crystal-superfluid interface are drawn in Fig. 1. The interface first appeared in the lower part and moved smoothly. This growth mode is the creep growth observed in the high temperature region in the classification in our previous publications[15, 16]. At lower temperature the interface moves quite differently: the crystal grows via avalanche. In this report we focus on the high temperature nucleation. The time of nucleation at which the interface first appeared in the aerogel can easily be determined. The first crystal always appeared at the same position in the aerogel.

In Fig. 2, three pressure sweeps during the crystal growth in aerogel are shown as examples at 1.0 K. $\delta P$ is the overpressure measured from the bulk melting pressure at 1.0 K. The filled circles are the moment of the nucleation determined from the crystal growth movie and indicate the $\delta P_c$ in each sweep. $\delta P_c$ always had a different value in each pressure sweep and was distributed. Metastability of the superfluid liquid in the aerogel is clearly observable. The melting began at about the half of $\delta P_c$ during the depressurization. The equilibrium pressure in the aerogel may be between the two but we have no way to obtain the equilibrium pressure in the present experiment.
By repeating the pressure sweeps 50 times, cumulative distribution of $\delta P_c$ was obtained as in Fig. 3. Here, the vertical axis is $\Sigma = \frac{N_0 - N}{N_0}$; $N$ is the number of nucleation events whose $\delta P_c$ is less than $\delta P_c$, and $N_0 = 50$ is the total number of pressure sweeps. Typical value of the critical overpressure was $\delta P_c \approx 0.4$ bar at $\Sigma = \frac{1}{2}$, and the minimum and maximum values were 0.32 and 0.48 bar, respectively. $\delta P_c$ spread over 40% of the value at $\Sigma = \frac{1}{2}$ and was broadly distributed. In the $^4$He crystal nucleation without aerogel by Ruutu et al. [3], it was distributed about 14% of the mean value of the critical pressure. Without aerogel, the absolute value of $\delta P_c$ is naturally much smaller, on the order of a few mbar, than in aerogel. Our finding of the broad distribution of $\delta P_c$ might be a specific feature of the crystal nucleation of $^4$He in aerogel. Since the first nucleation always occurred at the same position, the broad distribution is not just a reflection of the inhomogeneity of aerogel. One of the other possible reasons for the broad distribution is that the pressure was measured through solid. It would be informative to know the temperature dependence of $\delta P_c$ since the non-uniform stresses that the crystal can support depends on temperature.

4. Summary

Critical overpressure for $^4$He crystal nucleation was measured in aerogel using a variable volume cell. It had different values in each pressure sweep and the metastability of superfluid liquid in aerogel was clearly seen. Cumulative distribution of the critical overpressure was obtained by 50 repetitions of nucleation, and an anisotropic S shape characteristic of the nucleation process was observed. The breadth of the distribution was rather large compared with that without aerogel.

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