Creation and Annihilation of $^4$He Negative Crystals

T. Takahashi, R. Nomura, and Y. Okuda
Department of Physics, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro, Tokyo 152-8551, Japan
E-mail: takahashi.t.aw@m.titech.ac.jp, nomura@ap.titech.ac.jp

Abstract. When a relatively large $^4$He crystal sticking on a sample cell wall fell slowly along the wall with cooling, a number of small superfluid droplets or negative crystals spontaneously emerged in the host crystal. The negative crystals rose in the host crystal due to their buoyancy, and finally merged into bulk superfluid surrounding the host crystal. The rising velocity of the negative crystals was almost constant regardless of the host crystal’s falling motion, even when the host stayed on the bottom of the cell.

1. Introduction

Liquid or vapor inclusions are sometimes left in a host crystal and called negative crystals. Negative crystals are isolated from the outer environment and are suitable for studying the interfacial properties of crystals. Negative crystals are known to exist in $^4$He quantum crystals, whose interface is highly mobile[1, 2], and rise in a host crystal due to their buoyancy[3, 4]. Here, we report the spontaneous formation of negative crystals when a host crystal falls along a sample cell wall as it cools.

2. Experimental and results

We performed an experiment in a $^3$He evaporation cooling cryostat designed for a reduced gravity experiment of $^4$He crystals on a small jet plane. In the cryostat, we can produce an environment of less than 10% of the earth gravity $g_E$ for a period of 20 s during the top region of the parabolic flight. Here, we report the observation of bcc $^4$He crystals coexisting with the superfluid liquid in the cryostat on the ground at 1 $g_E$. A schematic of the cryostat is shown in Fig. 1 of Ref. [5]. A small GM refrigerator (Model RDK-101D, Sumitomo Heavy Industries) provided a 4 K stage with a cooling power of 0.1 W at 4 K. We installed a 1 K pot with a volume of 2 ℓ into which we directly transferred liquid $^4$He. Liquid helium in the 1 K pot lasted for about 6 h with pumping by a 1800 ℓ/h scroll pump. A $^3$He pot with a volume of 150 mℓ was also installed. The lowest temperature of 650 mK was able to be maintained for 4 hours by pumping of the $^3$He pot with another 10000 ℓ/h scroll pump.

The cryostat has optical windows for in situ observation of $^4$He crystals from room temperature. The windows are aligned in a straight line and infrared filters were installed on the thermal shields at 70 K and 4 K. We illuminated the $^4$He crystals from the back window by a parallel light and observed them from the front. The sample cell has two optical windows 26 mm in diameter with a flat bottom as illustrated in Fig. 4 of Ref. [6]. The cell body is made of stainless steel and the top flange is made of Cu. The top flange is thermally anchored to the $^3$He pot and is the coldest part.

Published under licence by IOP Publishing Ltd
Figure 1. Falling motion of $^4$He bcc crystals in superfluid liquid along the sample cell wall. Diameter of the circular sample cell is 26 mm. Cooling rate was about 1.5 mK/s. Time is indicated in each frame. As the crystals fell, a number of small superfluid inclusions or negative crystals spontaneously appeared and grew in the host crystals. The small square area at $t = 9$ s is expanded in Fig. 2 to show the details of the negative crystals.

We initially pressurized the liquid in the cell to about 29 bar at 2 K and stopped condensing the liquid. Thereafter, we started pumping the $^3$He pot slowly to cool down the system. With the slow cooling of the sample cell, one or a few $^4$He crystals nucleated on the top of the cell at around 1.72 K. They continued to grow in the coldest area as it cooled. These were in the bcc phase in this temperature range on the melting curve[1]. When they grew to a size of about 10 mm in diameter, they began to fall along the cell wall as shown in Fig. 1. In all the following figures, we set the origin of the time $t = 0$ s in the first frame of Fig. 1. At $t = 0$ s, the temperature was 1.65 K and cooling rate was about 1.5 mK/s. The right crystal dropped quickly by about 4 mm just after $t = 6$ s along the wall, but in other time periods it fell slowly. The falling process differed in crystals: some directly dropped to the bottom and others fell slowly along the wall.

Although the formation mechanism of negative crystals is not well understood, we frequently observed the following. As $^4$He crystals fell, a number of small superfluid droplets or negative crystals spontaneously nucleated and grew in the host crystals. To see the spontaneous emergence of the negative crystals, the small square area shown in Fig. 1 at $t = 9.0$ s has been expanded in Fig. 2. Initially, no sign of the negative crystal was recognized in the host crystal. But then several negative crystals with a spherical shape nucleated and grew to about 1 mm in diameter within 20 to 40 s depending on the temperature during the slow cooling.

The negative crystals rose in the host crystal at a velocity $v$ with respect to the sample cell frame proportional to their size as observed in Ref. [3]. In Fig. 3, $v$ is shown as a function of the radius $R$ for three negative crystals. The same symbol represents data of the same negative crystal and $v$ became higher as the negative crystals grew. It was concluded in the previous experiment that the rise is induced by the melting of the host crystal in the upper part of the negative crystal and by the crystallization in the lower part owing to the gravitational driving force proportional to $R[3, 7]$. In the rising negative crystal, a downward superflow occurs due to the mass conservation within it. The rising velocity is described by the crystallization rate $K$ as,

$$v = K \frac{\Delta \rho}{\rho_c} g E R.$$  (1)
Figure 2. Expanded images of the square area in Fig. 1 at $t = 9$ s. Time is indicated in each frame and the length scale of 1 mm is given in the last frame. Initially no sign of the negative crystal was recognized at $t = 9$ s but then three negative crystals nucleated and grew in the host crystal, growing to about 1 mm in diameter in about 20 s.

Figure 3. Rising velocity of negative crystals as a function of the radius. Three symbols represent the data for three different negative crystals. Since they grew as they rose, $v$ can be obtained as a function of the radius. The upper and lower lines are the calculation of Eq. (1) at 1.49 and 1.63 K.

Here, $\Delta \rho$ and $\rho_c$ are the density difference between solid and liquid and the solid density. The two lines in Fig. 3 are the calculation of Eq. (1) at 1.63 and 1.49 K and roughly agreed with the observed $v$.

This rising of the negative crystals was independent of the host crystal’s falling motion: the rising velocity of the negative crystals was almost unchanged when the host fell slowly or stayed on the bottom of the cell. The negative crystals fell with the host only when the host dropped quickly. This indicates that the slow falling of the host crystal is the result of the crystallization in its lower part and the melting in its upper part. In this case, the falling velocity is also expected to be given by Eq. (1). The observed velocity was 0.23 mm/s at around $t = 9$ s which roughly agrees with the calculation of Eq. (1), about 0.3 mm/s.

The negative crystals eventually reached the upper surface of the host crystal, merged into the outer superfluid liquid and were annihilated as shown in Fig. 4. The first frame of Fig. 4 is the image through the whole window at $t = 35$ s and the following figures are an expansion of the square area. Just after the merging, there remained a hollow on the surface of the host crystal and this then relaxed to a flat surface due to gravity.

If the lattice is completely rigid in a crystal, it is impossible to have negative crystals. There must be a mass transport mechanism to grow negative crystals to compensate the density difference between the solid and the liquid. As the host crystal falls, the hydrostatic pressure increases. This increase stabilizes the crystal and does not favor the emergence of negative crystals. As cooling takes place slowly along the melting curve during the growth, the inner part of the crystal is formed at higher temperature and under higher melting pressure. The
Figure 4. Annihilation of the negative crystals. The first frame is the image through the whole window at $t = 35$ s. The square area in this frame is expanded in the following frames. Time is indicated in each frame and the length scale of 1 mm is given. The negative crystals reached the upper surface of the host crystal and merged into the outer bulk liquid. The dip on the surface slowly relaxed and became flat.

The inner part has greater density and higher temperature than the outer part. These density and temperature gradients in the host crystal may drive the formation of the negative crystals. We need to continue measurements such as the temperature dependence in order to confirm these speculations.

3. Summary
Negative crystals were observed from their emergence to annihilation. They spontaneously nucleated in the host crystal falling along the cell wall with cooling. The driving force and the mass transport mechanisms are not clear at present.

4. Acknowledgements
This study was supported in part by the GCOE at Tokyo Tech. ”Nanoscience and Quantum Physics Project”, a Grant-in-Aid for Scientific Research (Nos. 21340095 and 17071004) from MEXT of Japan and by a ”Ground-based Research Announcement for Space Utilization” promoted by the Japan Space Forum.

References
[1] S. Balibar, H. Alles and A. Ya. Parshin, Rev. Mod. Phys. 77 317, (2005).
[2] Y. Okuda and R. Nomura, J. Phys. Soc. Jpn. 77, 111009 (2008).
[3] K. Yoneyama, R. Nomura and Y. Okuda, Phys. Rev. E 70, 021606 (2004). Erratum, Phys. Rev. E 73, 049901 (2006).
[4] Y. Okuda, S. Yamazaki, T. Yoshida, H. Fujii, and K. Matsumoto, J. Low Temp. Phys. 113, 775 (1998).
[5] T. Takahashi, M. Suzuki, R. Nomura and Y. Okuda, J Low Temp Phys 162, 733 (2011).
[6] T. Takahashi, M. Suzuki, R. Nomura and Y. Okuda, K. Kamiya, T. Numazawa and P. Shirron, Microgravity Sci. and Technol. in press.
[7] M. Mukai and M. Uwaha, J. Phys. Soc. Jpn. 75, 054601 (2006).