Observation of supercooling of superfluid ^{4}He during crystallization

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A supercooling of superfluid helium-4 during crystallization has been observed experimentally. The average lifetime of the metastable liquid increases with decreasing temperature. The transition from a cubic phase to a hexagonal phase sharply reduces the lifetime.

Studies of the crystallization kinetics in helium are of particular interest since this phase transition exists all the way down to absolute zero. With decreasing temperature, the probability (W) for the appearance of a critical nucleation center as a result of thermal fluctuations falls off exponentially: $W \to 0$ as $T \to 0$ (Ref. 1). According to the theoretical predictions of Kagan and Lifshits,[2] at $T = 0$ the crystallization should occur as a result of the appearance of a supercritical nucleation center due to tunneling. This approach was subsequently developed in several theoretical papers.[3–5] In the present letter we report the first experimental results on the supercooling of helium at fairly high temperatures, 1.25 – 1.75 K.

Metastable liquid helium is produced through a monotonic cooling of a container in a vacuum jacket in a bath of ^{3}He. The pressure is determined (within a measurement error $\sim 2 \times 10^{-4}$ atm) from the buckling of one of the container walls, 0.55-mm thick. A copper cooling duct passes through the opposite stainless-steel wall and is soldered in place. A resistance thermometer is mounted on this duct. The temperature is determined within ~ 5 mK. The pressures and temperatures are measured, and the heater is controlled, by an IBM PC microcomputer, which is connectd through an AKK-83 controller[6] to a CAMAC crate. The measurement cycle begins with the establishment of the temperature of the cold point (the ^{3}He bath) at a level ~ 20 mK below the crystallization temperature T_m. The temperature of the heater container is raised to a level ~ 20 mK above T_m; then the heating is stopped, and the container begins to cool monotonically at a rate of $T = 10^{-3}$–10^{-4} K/s. When a nucleating center reaches the critical size, it begins to grow very rapidly; the process is accompanied by a decrease in the pressure in the container. On the recording, the event is seen as a jump (Fig. 1). The magnitude of this jump, Δp, is determined within 7×10^{-4} atm. The sample is then melted, and the measurement cycle is repeated. The total number of jumps detected at each temperature point ranged from ~ 100 to 690. The difference between the chemical potentials of the liquid helium before and after the crystallization, $\delta\mu$, is found from the pressure jump: $\delta\mu = k(T)(V_l - V_s)\Delta p$, where V_l and V_s are the volumes per atom in the liquid and solid phases, respectively, and $k(T)$ is a correction for the heat of crystallization [$k(1.75) = 1.47$; $k(1.53) = 1.09$; $k \to 1$ as $T \to 0$]. The rate of change, $\dot{\delta\mu}$, is calculated from the measured rates of change of the pressure and the temperature; the scatter in the values was $\sim 30\%$.

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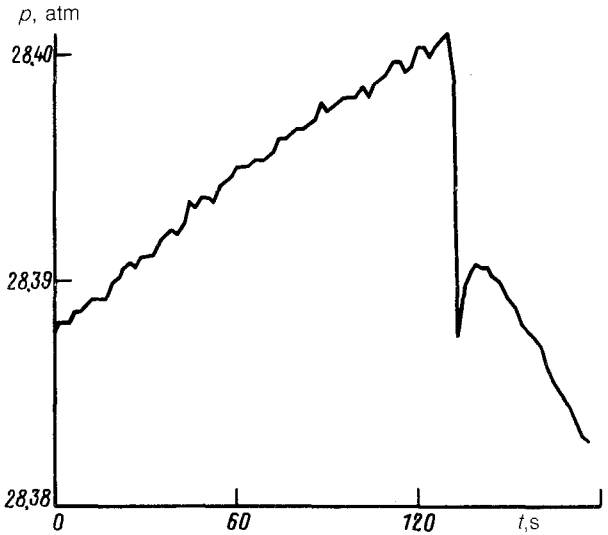

FIG. 1. Example of a recording of a pressure jump upon the formation of a nucleation center of the solid phase (bcc).

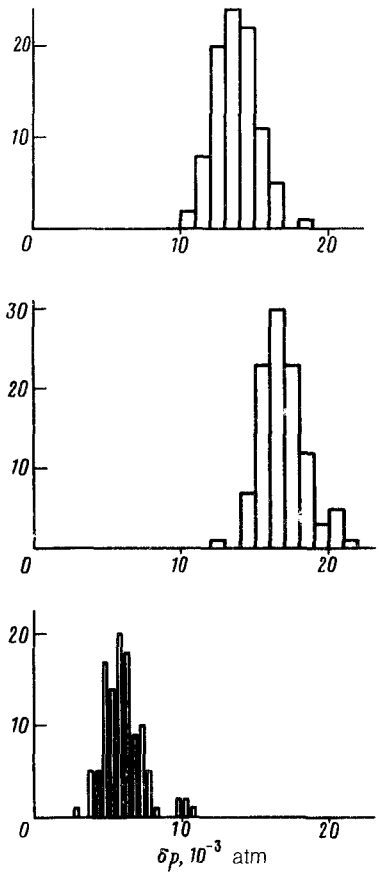

FIG. 2. Distributions of the number of events in which critical nucleation centers are produced versus the supercooling. From top to bottom: bcc, $T = 1.73$ K, $\dot{T} = 6.1 \times 10^{-4}$ K/s, 93 events; bcc, $T = 1.53$ K, $\dot{T} = 4.3 \times 10^{-4}$ K/s, 105 events; hcp, $T = 1.4$ K, $\dot{T} = 2.6 \times 10^{-4}$ K/s, 110 events.

Figure 2 shows histograms of the distribution of the sizes of the jumps in the coordinates $\delta p = \delta\mu/(V_l - V_s)$.

The number (g) of jumps detected per unit time at the time t is related to the probability for the appearance of a critical nucleating center in the container, W, by

$$g(t) = N_0 W[\delta\mu(t)] \exp\left[-\int_0^t W[\delta\mu_l(t')]dt'\right],$$

where N_0 is the total number of events. Inverting this expression, and making use of the linear relationship between $\delta\mu$ and t, we find

$$W(\delta\mu) = \dot{\delta}\mu\, g / \left[\int_{\delta\mu}^{\infty} g(\delta\mu')\, d(\delta\mu')\right] .$$

Since the half-widths of the histograms (Fig. 2) are approximately equal to the mean-square error of the measurements, however, this analysis procedure results in a large error in the functional dependence $W(\delta\mu)$. Accordingly, Fig. 3 shows the reciprocal of the lifetime $\langle t \rangle$ of the metastable state, averaged over all events. It is proportional to the probability for the appearance of a critical nucleation center at the average value $\langle\delta\mu\rangle$. For the bcc phase we have $\langle\delta\mu\rangle(V_l - V_s) = 0.014$–$0.017$ atm; for the hcp phase the corresponding values are 0.055–0.06 atm. It can be seen from this figure that a lowering of the temperature results in an increase in the average lifetime of the metastable liquid in both phases. At the transition from the cubic phase to the hexagonal phase we observe a jump in the rate of nucleation; in the low-temperature phase the rate is higher even at a supercooling smaller by a factor of three.

Similar experiments were carried out with nonsuperfluid helium at pressures of 30.5–36 atm. Pressure jumps of the type shown in Fig. 1 were not detected. Some

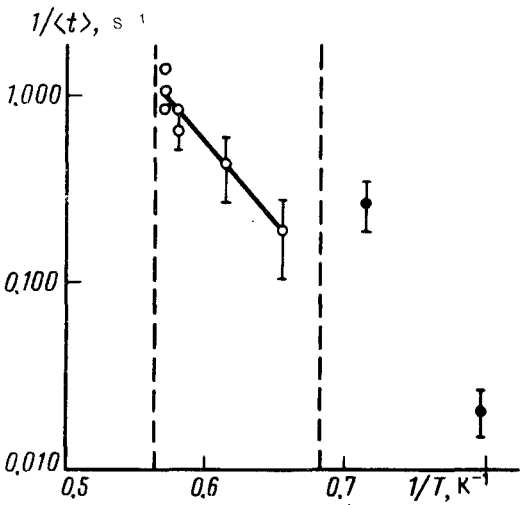

FIG. 3. Temperature dependence of the reciprocal lifetime of the metastable state. The vertical dashed lines show the bcc phase. The straight line drawn in the cubic phase corresponds to an activation energy ~25 K.

possible reasons are a temperature gradient in the container, a short lifetime of the metastable liquid, and a low kinetic coefficient of the crystal growth.

We can draw some qualitative conclusions from these results. The decrease in the rate of nucleation with the temperature in each of the phases appears to imply that at $T = 1.25$–1.75 K we are observing a classical, thermally activated appearance of a critical nucleation center. The jump at the transition to the hexagonal phase is probably due to a change in surface tension. It is difficult to make a quantitative comparison of these results with theoretical predictions, for several reasons. For example, such parameters of the theory as the surface tensions of the solid phases and the wetting angle at the surface of the container are not known accurately. The wetting angle is particularly important since the nucleation center usually appears at the wall, according to experiments (Ref. 7, for example). The presence of charges in the liquid, formed as a result of the natural radioactivity of the container walls, also reduces the potential barrier for the appearance of a critical nucleation center.

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