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Liquid-to-vapor homogeneous nucleation in liquid nitrogen

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Experimental data on the liquid-to-vapor homogeneous-nucleation temperature, which provides a fundamental limit on the maximum attainable superheat, are presented for liquid nitrogen in the temperature range 77.4 K $< T_s <$ 124.6 K, where T_s is the saturated bath temperature of liquid nitrogen. These data were obtained from a transient-heating experiment and are relevant to the design of superconducting systems using the recently discovered high-temperature superconductors and liquid nitrogen as the coolant.

INTRODUCTION

With the recent discovery of high-temperature ($T_c > 90$ K) superconductors,[1,2] the need for the extensive use of liquid nitrogen as a coolant is close to reality. In the case of superconductors, the coolant serves two purposes: (1) it provides the required low-temperature environment for the superconducting system to operate efficiently, and (2) it prevents the superconducting system, e.g., high-field magnets or transmission lines, from quenching due to localized normal-zone creation by arresting the spread of such normal zones through efficient heat transfer into the liquid coolant. In regard to the second requirement for the coolant, it is necessary to understand better the transient heat transfer from a solid into liquid nitrogen. In particular, it is important to learn if there is any fundamental limitation to the heat-transfer capacities of liquid nitrogen imposed by the amount it can be superheated, and the extent to which the liquid can sustain metastability.

It is common knowledge that a liquid can withstand a certain amount of superheating before a transition to the vapor phase takes place. In the absence of external initiating influences, such as impurities or high-energy radiation, a homogeneous liquid can be significantly superheated relative to the saturation temperature before a sufficiently larger number of vapor nuclei induced by thermodynamic fluctuations can generate and grow spontaneously in the liquid. This process of spontaneous initiation of nucleation restricts further superheating of the liquid and is termed homogeneous nucleation. Thus, homogeneous nucleation in practice places a significant constraint on the maximum attainable superheat in a liquid. The kinetic theory of homogeneous nucleation[3,4] predicts the nucleation rate that depends exponentially on parameters related to the degree of superheating. Consequently, one obtains a relatively sharp definition of the maximum attainable superheat in a liquid. Because of the extreme sensitivity of the nucleation rate to the superheat temperature of the liquid, one adopts the more or less conventional usage of referring to a single temperature, the homogeneous-nucleation temperature T_h. Because T_h depends exclusively on the intrinsic thermophysical properties of the liquid, its determination may prove to be essential in providing a reference temperature that can be used as a design parameter for large-scale superconducting systems, such as superconducting magnets, transmission lines, etc., in which liquid nitrogen is likely to be used as a coolant.

We report here our measurements of the homogeneous-nucleation temperature in liquid nitrogen in the bath temperature range 77.4 K $< T_s <$ 124.6 K, where T_s is the saturated bath temperature. This corresponds to the pressure range $0.03 < P_s/P_c < 0.91$, where P_s and P_c are the saturated bath pressure and the critical pressure of the liquid, respectively. Previous measurements of T_h in nitrogen[5] were made by continuous isobaric heating of the liquid in sealed capillaries. Our measurements, on the other hand, are made by rapidly heating a solid wire in a bath of liquid nitrogen and by monitoring the superheating of the liquid in immediate contact with the wire. Such measurements provide both the transient heat-transfer characteristics from a solid into liquid nitrogen and also the important information on the limiting superheat of the liquid. Thus, this experimental study is directly relevant to the design of superconducting systems using the recently discovered high-temperature superconductors.

EXPERIMENT

The measurements were made in a specially constructed stainless-steel cryostat of 7-cm inner diameter, capable of withstanding pressures up to 10 MPa. The liquid-nitrogen bath temperature was determined with a platinum resistance thermometer, and the bath pressure was monitored using a pressure transducer with a digital pressure indicator.

The test section consisted of a platinum heating wire, roughly 7 cm in length and 0.102 mm in diameter, that was attached to a spring-loaded holder to keep the wire taut. The whole test section assembly was mounted vertically inside the pressure vessel. The platinum wire also served as its own thermometer to monitor the temperature of the wire (which was heated by a step-function current) and also the temperature of liquid in contact with it. Platinum was chosen because of its relatively large temperature coefficient of resistance and its high degree of linearity in its electrical resistance versus temperature over the temperature range of interest. The electronic circuitry used, and the procedure employed for the transient superheat temperature measurement employing the platinum wire heater-thermometer, are described in detail elsewhere[6,7] and will not be repeated here.

The transient heat-transfer characteristics in any liquid depend strongly on the heating rate of the solid and the pressure the liquid is subjected to. For sufficiently high heating rates, on a millisecond time scale or faster, neither natural convection nor heterogeneous nucleation have time to develop, and the superheating of the liquid adjacent to the solid heating surface takes place solely due to thermal conduction prior to the onset of homogeneous nucleation in the superheated liquid layer. Because of the short times involved, heterogeneous nucleation from the heating-surface irregularities, such as grain boundaries, ledges, cracks, and scratches does not have time to develop. We had previously employed this transient-heating technique to determine the homogeneous nucleation temperature of liquid helium I.[8,9]

RESULTS

Typical sets of superheat temperature ΔT, versus time curves at two representative bath pressures, are shown in Figs. 1(a) and 1(b). In Fig. 1(a) note the kinks in the temperature rise curves occurring at a constant superheat temperature ΔT_h. Curves *b* and *c* illustrate the transition in the characteristics of the transient superheating curves from rounded temperature overshoots to the ones that continually rise toward stable film boiling. As the bath pressure is increased, this transition moves to shorter time scales, and approximately above 0.96 MPa, only temperature overshoots characterized by sharp peaks are observed, as shown in Fig. 1(b). It is worth pointing out that the behavior of the high-pressure ΔT-versus-time curves [Fig. 1(b)] is almost identical to that observed in liquid helium.[8,9]

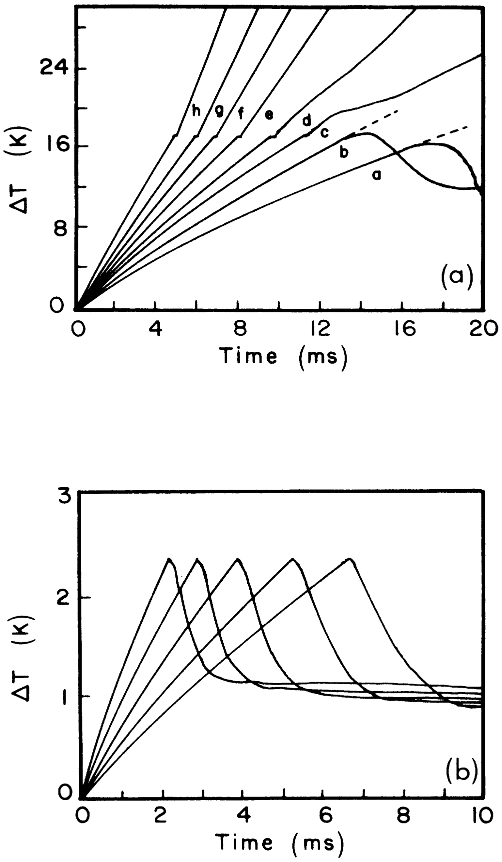

FIG. 1. Superheat temperature ΔT vs time. Transient superheat temperature curves are shown as a function of increasing applied power to the platinum wire at two bath pressures: (a) 0.55 MPa and (b) 2.2 MPa. Note that the kinks in the temperature rise in (a) and the sharp peaks in (b) all occur at constant superheat temperatures.

We interpret the sharp peak in the temperature overshoots [Fig. 1(b)] and the kinks in the temperature rise curves [Fig. 1(a)], both occurring at constant superheat, as indications of a sudden onset of homogeneous nucleation in the superheated liquid nitrogen layer in contact with the platinum heating wire. As previously mentioned, the superheating of the liquid is achieved entirely by heat transfer into the liquid by thermal conduction from the heating wire, because the time scales involved are much shorter than the delay times involved in the onset of both heterogeneous nucleation and convective heat transfer. This conclusion can be inferred from the excellent agreement between the observed temperature rise and the temperature rise predicted,[10] assuming that the heat transfer is purely due to thermal conduction, as illustrated by the dashed lines superimposed upon curves *a* and *b* in Fig. 1(a). Therefore, during the superheating process, a large amount of thermal energy is accumulated in the superheated liquid layer. When T_h is reached, a large number of critical vapor nuclei ($\sim$50 Å in radius) are formed spontaneously in the bulk of the superheated liquid layer. Their subsequent explosive growth is promoted by the high vaporization rate associated with such high superheat.

The behavior of the wire temperature at the onset of homogeneous nucleation is governed by a combination of two factors, namely, rate of heat removal by vaporization, and the thermal insulation around the wire provided by the growing vapor film. At low pressures, this leads to stable film boiling. At high pressure, both the liquid superheat [see Eq. (2)] and the bubble break-off diameter[11,12] are small. These two factors help reduce the chances of a vapor-film formation because before a vapor film can form the vapor bubbles break off and leave the wire surface. Consequently, we see a sharp peak in the temperature overshoot indicating homogeneous nucleation with a subsequent cooling of the wire down to steady-state nucleate boiling.

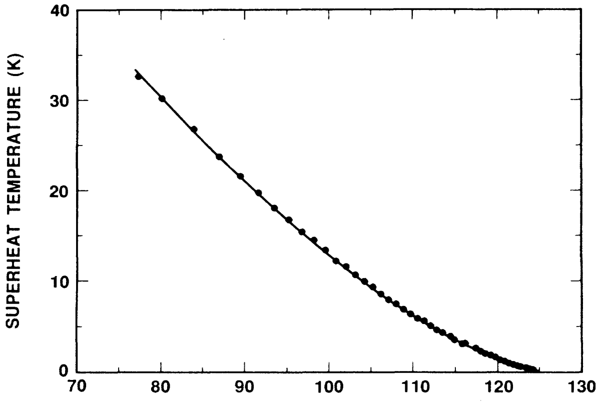

FIG. 2. Superheat temperature ΔT_h as a function of bath temperature. The experimental superheat temperature ΔT_h is plotted (solid circles) as a function of the bath temperature. The solid line represents the theoretical predictions of the homogeneous nucleation temperature derived from Eq. (1) for a nucleation rate of $J=10^6$ critical nuclei/cm^{3}s.

In order to verify that the observed constant superheat temperatures, ΔT_h ($\Delta T_h = T_h - T_s$), are independent of the wire material, we have repeated the measurements at atmospheric pressure with wires of different dimensions and materials (Constantan and Manganin). We have also repeated the measurements for both vertical and horizontal wires. In all cases, the same value of the constant superheat ΔT_h was observed within ±0.3 K independent of wire size, composition, and orientation. This is consistent with our identification of the sharp temperature changes with the homogeneous nucleation temperature of the liquid.

Finally, in Fig. 2, we show the experimental data on superheat temperature, ΔT_h, obtained from the transient superheating technique described above, as a function of the bath temperature. For comparison, we also show in Fig. 2 (solid line) the theoretical estimates for ΔT_h based on the predictions from the classical nucleation theory. We employ the form recently given by Blander and Katz[13] for the homogeneous nucleation rate as

$$J = N\left(\frac{2\gamma}{\pi m B}\right)^{\frac{1}{2}} \exp\left(\frac{-16\pi\gamma^3}{3\kappa T(P_e - P_L)^2\delta^2}\right) , \tag{1}$$

where the notation employed is that of Ref. 13 and the thermophysical properties of liquid nitrogen were taken from Jacobsen *et al.*[14] The solid line in Fig. 2 corresponds to a nucleation rate of $J=10^6$ nuclei/cm^{3}s. This nucleation rate is based on the heating rates employed in this study.

It should be mentioned that this rate is not the same for the entire pressure range studied, but the error introduced in assuming a single value of J is entirely negligible because of the exponential nature of the nucleation rate. The excellent agreement between the experimental data and the theoretical estimates of ΔT_h supports our interpretation that the kinks and the sharp peaks occurring at constant superheat temperature are indeed the result of a sudden onset of homogeneous nucleation in the superheated liquid in contact with the heater-wire surface.

The experimental data on ΔT_h can be described well in terms of the saturated bath temperature by a single power law of the form

$$\Delta T_h = A\left(1 - \frac{T_s}{T_c}\right)^a , \tag{2}$$

where $A=136.19$ K, $a=1.496$, and T_c ($T_c=126.193$ K) is the critical temperature of nitrogen. Equation (2) fits the experimental data with a standard deviation of 0.11 K. Similarly, in terms of the reduced bath pressure, P_s/P_c, the data can be expressed as

$$\left(1 - \frac{T_h}{T_c}\right) = B\left(1 - \frac{P_s}{P_c}\right)^b , \tag{3}$$

where $B=0.1329$, $b=0.9395$, and P_c ($P_c=3.3978$ MPa) is the critical pressure of nitrogen.

Finally, from the transient heat transfer studies presented here, we suggest that film boiling, which may occur in superconducting systems following a sudden temperature rise, can be prevented if the nitrogen bath is pressurized above 0.96 MPa. In such a situation, the onset of homogeneous nucleation can result in improved heat transfer and possibly in stabilizing the system. Moreover, the homogeneous nucleation temperature data can be used as a fundamental design parameter. The effect of coolant flow on the transient heat transfer characteristics should also be examined to determine under what flow conditions a transition to film boiling can be prevented.

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