

# METASTABLE SUPERHEAT IN NUCLEATE BOILING OF CRYOGENIC LIQUIDS

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## INTRODUCTION

The metastable superheat  $\Delta T$  attainable by a liquid above its saturation temperature is important in nucleate boiling. Aside from the boiling description, however, a knowledge of  $\Delta T$  also provides criteria for heat transport transition when bubble formation is entirely suppressed. A nonboiling liquid may be heated to the maximum metastable superheat before it disintegrates [1]. As soon as the wall excess temperature increases beyond the limiting superheat value, the system will enter the Leidenfrost regime, within which liquid is converted into the completely disordered phase when it approaches the hot walls.

The present considerations extend previous studies of peak quantities [2]. Presuming thermodynamic similitude, we compare superheat data of liquefied gases with an empirical nitrogen correlation.

## THERMODYNAMIC SIMILITUDE

The nucleate boiling superheat  $\Delta T$  has been described as a function of the heat flow density  $q$  by means of more-or-less complex equations. There exist essentially two kinds of boiling correlations: the transport equations valid for a special configuration and equations closely related to the equilibrium thermodynamics of quasi-static systems. The transport equations determine the details of the heat transport mechanism with consideration of the force field and special conditions of each particular system, but they are so complex that it is difficult to arrive at satisfactory equations which will be valid in the entire range of pressures from the triple to the critical point.

The second kind of description, on the other hand, disregards the details of the transport mechanism and relies primarily on equilibrium relations of macroscopic thermodynamics instead. A fairly satisfactory correlation of this kind [3] was previously compared with simple thermodynamic estimates [2]. The comparison suggested a simplification which avoids extended property evaluation and makes use of thermodynamic similitude. This approach is near at hand since  $\Delta T$  does not vary appreciably with  $q$  in fully established nucleate boiling. Further, in many applications it is sufficient to know only one pair of coordinates at peak conditions:  $(q_p, \Delta T_p)$ . The function  $q(\Delta T)$  then may be obtained by making use of the derivative  $\partial q / \partial \Delta T$  from a similar known configuration.

To arrive at a reduced function  $\Delta T / T_c$  expressed in terms of reduced coordinates, e.g.,  $T / T_c$ , we consider data of a frequently investigated substance representative of a group of similar liquids. Within the group of simple liquefied gases much effort has been concentrated on nitrogen. Its average superheat at peak conditions may be described by

$$\overline{\Delta T}_p / T_c = 0.10(1 - T / T_c)^{3/4} + 2.0(1 - T / T_c)^5 \quad (1)$$

The reduced superheat function can be refined as soon as more information is available on

nucleation properties. The latter are presently approximated in a crude way by specifying the mean nucleus radius [1]. Thus, the approximate relation

$$\Delta T = (2\sigma/R_0)T_s/L\rho_v \quad (2)$$

may be considered. The superheat can be small for rough surfaces, which trap large amounts of gas, while the  $\Delta T$  should be large for polished surfaces. Ultraclean systems may attain the maximum possible superheat  $\Delta T_{\max}$ . Since the nucleation properties of technical systems are neither well known, nor controllable, we may specify limits by defining a degree of metastability

$$\epsilon \equiv \Delta T_p / \Delta T_{\max} \quad (3)$$

where  $0 < \epsilon < 1$ . Alternatively, the actual superheat is given simply as a multiple of the average,  $\overline{\Delta T_p}$ . Values of  $\epsilon$  cannot be readily evaluated owing to the lack of information on state functions in the metastable range. Van der Waal's equation, however, permits crude estimates of nominal values, which come out too low if the excess pressure of the vapor cluster is neglected. For instance, at  $T/T_c = 0.6$  the following nominal  $\epsilon$  values (at constant pressure) are obtained: 0.14 at  $\Delta T_p = 0.5 \overline{\Delta T_p}$ ; 0.28 at the average superheat; and 0.42 at  $1.5 \overline{\Delta T_p}$ .

In view of the uncertainties involved, we base our comparison on the average data of (1), and account for the real superheat by specifying the multiple  $\Delta T_p / \overline{\Delta T_p}$ . Excess pressure corrections will be neglected. Reduced superheat values of various liquefied gases are expected to agree closely provided thermodynamic similitude really exists. However, complete agreement should not be anticipated since differences in molecular structure are reflected in the different  $T/T_c$  values at the triple point of various substances. Thus, an extrapolation of (1) beyond its triple point value, or about  $T/T_c \approx 0.6$ , is not recommended without experimental support.

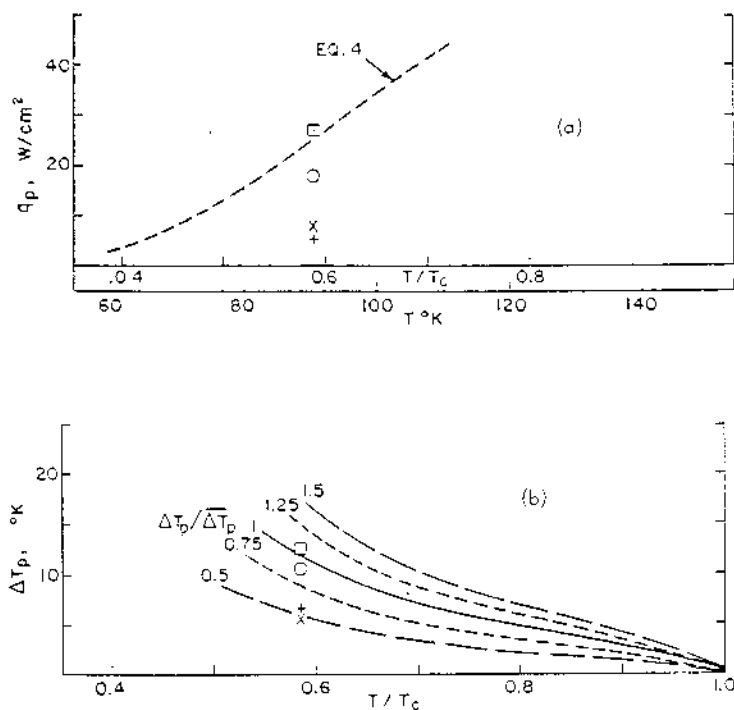


Fig. 1. Oxygen peak data. (a) peak flux; (b) metastable liquid superheat, where curves are based on (1).  $\square$  Bochirol, Bonjour, and Weil [8],  $\circ$  Weil [6],  $\times$  Haselden and Peters [1],  $+$  Mikhail, from [2].

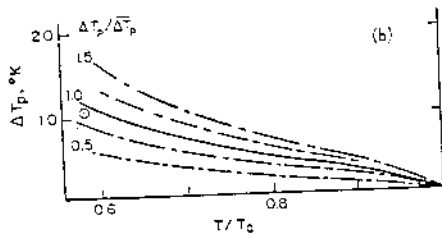
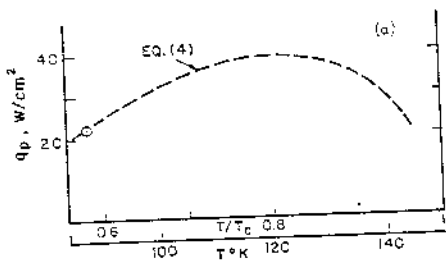


Fig. 2. Argon peak data: (a) peak flux; (b) metastable liquid superheat, where curves are based on (1). ○ Bochirrol, Bonjour, and Weil [3].

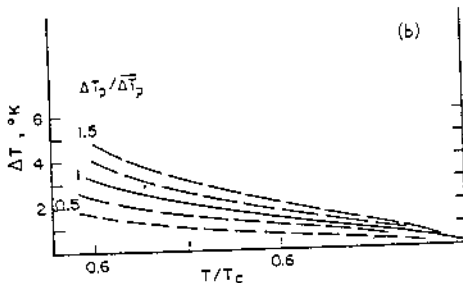
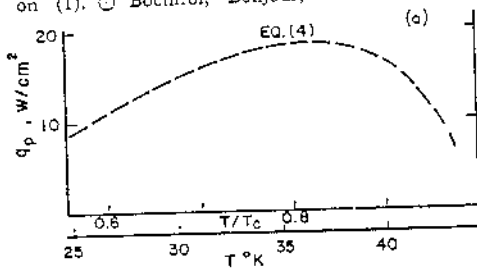


Fig. 4. Neon peak quantities: (a) peak flux; (b) metastable liquid superheat, where curves are based on (1). No experimental data available.

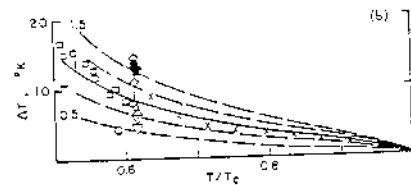
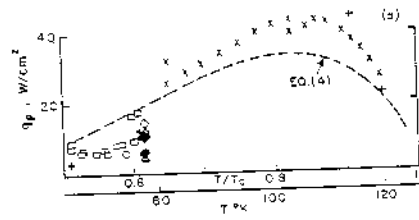


Fig. 3. Nitrogen peak data: (a) peak flux; (b) metastable liquid superheat, where curves are based on (1). × Roubeau [2], △ Weil [2], ○ Mulford and Nigon from [3], □ Frederking [12], ◇ Merte and Clark [13], ◆ Ruzicka [13], △ Haseleden and Peters [7], ▽ Mikhail from [8], ● Flynn, Draper, and Roos [12].

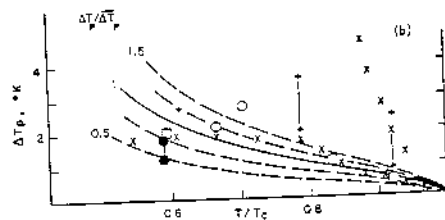
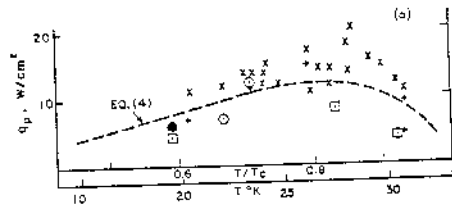


Fig. 5. Hydrogen peak data: (a) peak flux; (b) metastable liquid superheat where curves are based on (1). × Roubeau [2], △ Weil and Lacaze [14], ● Mulford and Nigon [13], □ Glass, DeHaan, Piccone, and Cost [15], ○ Walters [16].

### COMPARISON WITH EXPERIMENTAL DATA

The experimental data plotted in Figs. 1 through 6 represent the highest values reported and show the broad superheat range typical of nucleate boiling, which is often due to different nucleation conditions. For comparison, the heat flow density (the peak flux of natural convection pool boiling on a horizontal plate as given by Kutateladze)

$$q_p = 0.16L(\rho_v)^{1/2}g(\rho_L - \rho_v)^{1/4} \quad (4)$$

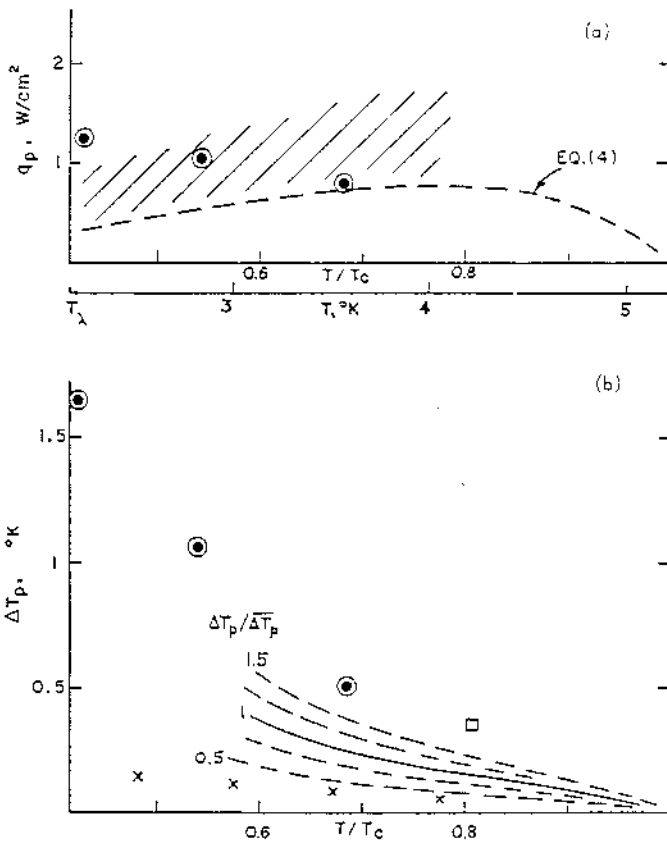


Fig. 6. Helium peak data: (a) peak flux; (b) metastable liquid superheat, where curves are based on (1).  $\odot$  Karagounis [13],  $\square$  Meissner [13],  $\times$  Reeber [20],  $\text{///}$  Frederking [14].

has been entered in Figs. 1a through 6a. Since only a few experimental  $q_p$  results are available for this configuration, other data (horizontal cylinder, vertical plate) have been included in the comparison.

A few additional effects, in particular the power level at which  $\Delta T$  has been observed and an apparent  $\Delta T$  increase associated with a broad  $q(\Delta T)$  maximum in the mixed boiling-transition regime must be considered in regard to the metastable superheat. At low power levels, a small derivative  $\partial q / \partial \Delta T$  might be encountered, indicating that nucleate boiling had not yet been fully established. Accordingly,  $\Delta T$  will be smaller than the peak value. At a pronounced  $q(\Delta T)$  maximum, on the other hand, a high  $\Delta T$  value appears at the peak. This apparent superheat value belongs to the transition regime and could be eliminated by using an electrically heated specimen with low heat capacity. To obtain  $\Delta T$  for correct nucleate boiling from thick heaters, however, we should subtract the influence of local vapor patches. In the figures these effects have been marked in some cases by plotting the entire  $\Delta T$  range from full nucleate boiling to the maximum. Since details of the investigations can be found in the original references, only a few remarks will be noted here for  $N_2$ ,  $H_2$ , and He.

Nitrogen (Fig. 3) has been studied by many investigators, particularly at 1 atm ( $T/T_c \approx 0.61$ ). The high atmospheric superheat data represent mixed boiling at a broad  $q(\Delta T)$  maximum, whereas the low values are more likely actual nucleate boiling results. At subatmospheric pressures nucleate boiling has been found to be poorly developed on thin wires [14].

We note, of course, that a reduced superheat of  $1.5 \overline{\Delta T_p}/T_c$  is not easily attainable and might be close to the metastability limit  $\epsilon = 1$ .

Hydrogen investigations are beset with some additional difficulties, caused by the low boiling point of the liquid, its small heat of vaporization, and ortho- and para-states of the molecule. Accordingly, the scatter of data is large. Although most of the results lie within the range to be expected from  $N_2$  by applying similitude, a second range of high  $\Delta T$  values, above  $1.5 \overline{\Delta T_p}/T_c$ , has also been reported [3,9]. Though this effect is as yet unexplained, it seems likely that the high superheat cannot be sustained by a pure liquid during strong nucleate boiling.

Liquid helium can be regarded as an ordinary liquid; however, its physical properties are modified somewhat compared to other liquids. The superheat data of Karagounis [13] correspond to reduced values larger than  $1.5 \overline{\Delta T_p}/T_c$ , whereas results from a vertical heater [20] obtained at a very low power level, are small, as to be expected. In spite of the low temperature, there are no drastic deviations from thermodynamic similitude in the range  $0.6 < T/T_c < 1$ , where equation (1) permits reasonable superheat estimates.

### SUMMARY

The metastable superheat encountered in fully established nucleate boiling of liquefied gases at 1 atm covers a temperature range from less than 1°K to about 10°K, i.e., more than one order of magnitude. For this group of cryogenic substances agreement between experiment and thermodynamic prediction might be considered fair, in view of the uncertainties in nucleation conditions. Therefore, we conclude that thermodynamic similitude can be used to obtain fast superheat estimates in nucleate boiling.

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### NOTATION

- $g$  = acceleration due to gravity
- $L$  = heat of vaporization
- $q$  = heat flow density
- $R_0$  = nucleus radius
- $T$  = temperature, ( $T_s$  saturation value)
- $\Delta T$  = liquid excess temperature above saturation
- $\epsilon$  = degree of metastability
- $\rho$  = density
- $\sigma$  = surface tension

#### Subscripts

- $c$  = quantity at the critical point
- $l$  = liquid
- max = maximum
- $p$  = peak quantity
- $v$  = vapor

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