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Boiling-up of superheated liquid argon in an acoustic field

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Abstract

The method of lifetime measurement has been used to investigate spontaneous cavitation kinetics in superheated liquid argon in weak acoustic fields. It is shown that acoustic cavitation may proceed both by the mechanism of homogeneous nucleation of the vapor phase and by way of 'build-up' of vapor bubbles generated by high-energy particles or the action of some other external factor. Acoustic cavitation thresholds are adequately described by homogeneous nucleation theory.

1. Introduction

When an ultrasonic wave passes through liquid media in periods of pressure decrease, stretches can be achieved which are sufficient for the break of continuity (cavitation) and formation of periodically evolving bubbles. As a rule, cavitation begins on completed and easily activated boiling sites in a liquid volume or on the walls of a vessel (heterogeneous nucleation). In the absence of such sites liquids withstand considerable tensile stresses. In this case the liquid cavitation strength is determined by fluctuation nucleation (homogeneous cavitation).

According to classical homogeneous nucleation theory [1–3], the average number of viable nuclei forming in a unit liquid volume in a unit time (nucleation rate J) may be presented as

$$J = \rho B \exp(-G), \tag{1}$$

where ρ is the number of molecules in a unit liquid volume, B is the kinetic factor, $G = W_*/k_\mathrm{B}T$ is the Gibbs number, the work of formation of a critical bubble W_* related to the double average thermal motion energy per degree of freedom, k_B is the Boltzmann constant, and T is temperature.

At positive pressures and temperatures close to the critical point the dynamics of growth of a near-critical-sized bubble is mainly determined by the molecule evaporation rate, and for the kinetic factor we have [4, 5]

$$B = \left[\frac{6\sigma}{\pi m(3-b)}\right]^{1/2}, \tag{2}$$

where σ is the surface tension, m is the molar mass, $b = 1 - p/p_\mathrm{s}$, p is the pressure in the liquid, and the index 's' designates a saturation line.

If the liquid is incompressible, and the vapor in the bubble is an ideal gas, then under moderate superheats, when $G \gg 1$, the work of formation of a critical bubble is

$$W_* = \frac{16\pi\sigma^3}{3(p_\mathrm{s}-p)^2(1-\rho_\mathrm{V}/\rho_\mathrm{L})^2}, \tag{3}$$

where ρ_L, ρ_V are the densities of the liquid and vapor phases on the phase-equilibrium line.

The present paper investigates the kinetics of spontaneous boiling-up of superheated liquid argon in weak acoustic fields, when the amplitude of the acoustic pressure p_m in the liquid is much lower than its static tension $p_\mathrm{s} - p_0$. The static pressure p_0 was chosen close to the boundary of the liquid cavitation strength. The acoustical actions on a superheated (stretched) liquid were used in this case not as a means of creating large stretching forces but as a means of diagnosing the nature of the liquid boiling-up (cavitation) centers. The aim of the paper consists in establishing the nucleation mechanism, homogeneous or heterogeneous, by the response of a metastable liquid to weak acoustical actions.

The cavitation strength (limiting superheat) for liquid argon in the absence of acoustic fields was investigated earlier [6–8]. Focused acoustic fields were used to investigate the boundary of cavitation strength for liquid helium in [9–11].

The paper consists of an introduction and three sections. Section 2 describes the experimental procedure and presents experimental data. Section 3 is devoted to discussion of the data obtained and their comparison with homogeneous nucleation theory. Section 4 is a conclusion.

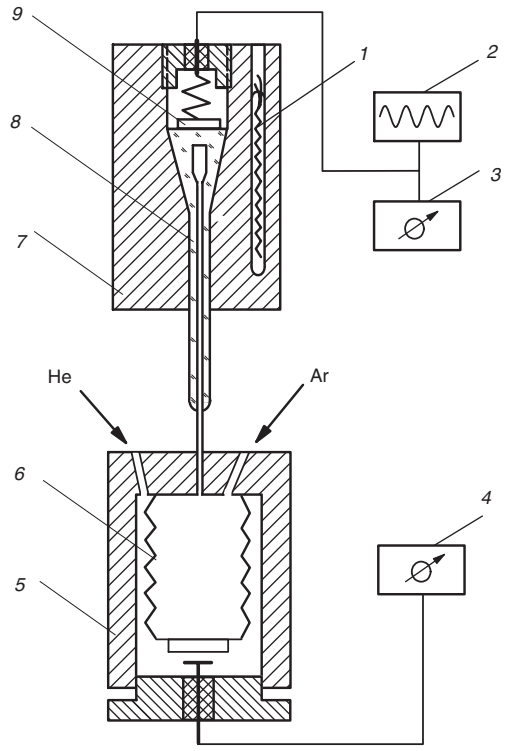

Figure 1. Schematic diagram of the experimental setup: 1—platinum resistance thermometer, 2—generator, 3—voltmeter, 4—chronometer, 5—bellows-element chamber, 6—bellows element, 7—thermostatting block, 8—glass acoustic cell, 9—piezoceramic radiator.

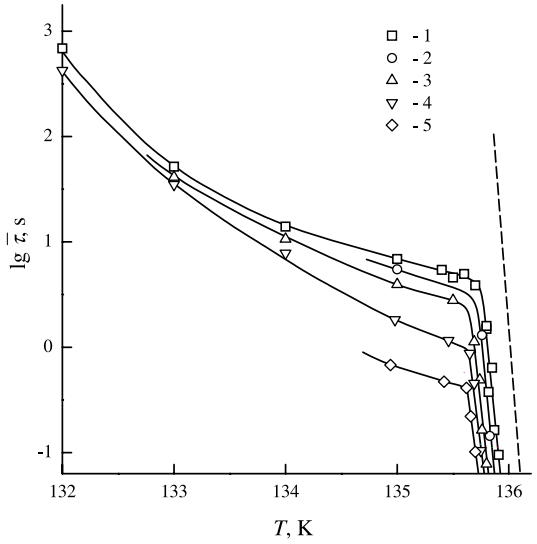

Figure 2. Temperature dependence of the mean lifetime of stretched liquid argon ($p_0 = 1.5$ MPa): 1—under natural conditions ($U_m = 0$ V), 2— $U_m = 10$ V, 3—13 V, 4—15 V, 5–18 V. The dashed line shows calculation by homogeneous nucleation theory ($p = p_0$).

2. Experiment

The method of lifetime measurement has been used to study the kinetics of spontaneous boiling-up of liquid argon in weak acoustic fields [3]. The experimental setup and the procedure of conducting experiments on the measurement of lifetimes of superheated cryogenic liquids are described in detail in [12, 13]. Here we shall point out the peculiarities of this procedure connected with measurements in acoustic fields.

The liquid under investigation was enclosed in a glass acoustic cell (labeled 8 in figure 1) of volume $V \simeq 80$ mm^3. The cell with the liquid was thermostated (± 0.005 K) in a copper block (7) at temperature T. Acoustic vibrations of frequency $f = 0.69$ MHz in the liquid were created by a piezoceramic radiator (9) situated on the outside of the cell. The cell is connected with a pressure chamber (5), whose temperature in the course of an experiment was maintained close to the temperature of normal boiling of the liquid under investigation. The pressure in the system was created by compressed helium and passed to the liquid through a metal bellows element (6). Entry into a metastable region was realized by a rapid decrease of pressure on the liquid from the initial value of $p_r(T) > p_s(T)$ to the value of $p_0(T)$. To decrease the introduced correction for the nonisothermality of the expansion process, the pressure was lowered in two steps: first to the intermediate value of p_1 ($p_s - p_1 \simeq 0.4$–0.5 MPa) with a time lag of $\sim$20 s, and then to $p = p_0$. Simultaneously with the final depressurization an exciting electric signal with a voltage amplitude U_m was delivered to the piezoradiator from a sine wave generator (2). The time τ from the moment of depressurization to the liquid boiling-up was measured in experiment by a chronometer (4). After boiling-up the liquid, the initial pressure p_r was created in the cell, and the measurement cycle was repeated. A total of $n = 40$–60 values of τ was fixed at given T, p_0, and U_m, by which the mean lifetime $\bar{\tau} = \sum_i \tau_i / n$ was determined. The temperature in experiment was measured by a platinum resistance thermometer (1) with an error of ± 0.02 K, pressure ± 0.005 MPa, and time ± 0.01 s. The electric signal amplitude on the piezoradiator was measured by a voltmeter (3) with an error ± 0.5 V and varied from 0 to 30 V. Argon of 99.998% purity was investigated in the experiments.

All measurements of the expectation time for the boiling-up of superheated liquid argon were conducted at a static pressure $p_0 = 1.5$ MPa. The results of measuring $\bar{\tau}$ on a semilogarithmic scale are presented in figure 2. In the absence of periodic pressure pulsations at $\bar{\tau} = 0.3$ s the liquid argon superheat $\Delta T = T_n - T_s$ was 11.88 K, where $T_n = 135.86$ K, and the cavitation strength $\Delta p = p_s(T_n) - p_0$ at the superheat temperature was 1.15 MPa. The switching of an ultrasonic field leads to a decrease in the liquid cavitation strength. The superheat temperature decreases by approximately 0.25 K at a voltage amplitude on the piezoradiator of 20 V.

The character of the temperature dependence of the mean lifetime for superheated liquid argon in an acoustic field is qualitatively similar to such a dependence in its absence. On all isobars $\bar{\tau}(T)$ there is a section of an abrupt decrease in $\bar{\tau}$ on the attainment of a certain temperature (attainable superheat boundary). The position and the slope of this section in the absence of acoustic vibrations are in good agreement with predictions of homogeneous nucleation theory (figure 2, the dashed line). The attainable superheat temperature T_n was calculated by equations (1)–(3) with the use of experimental

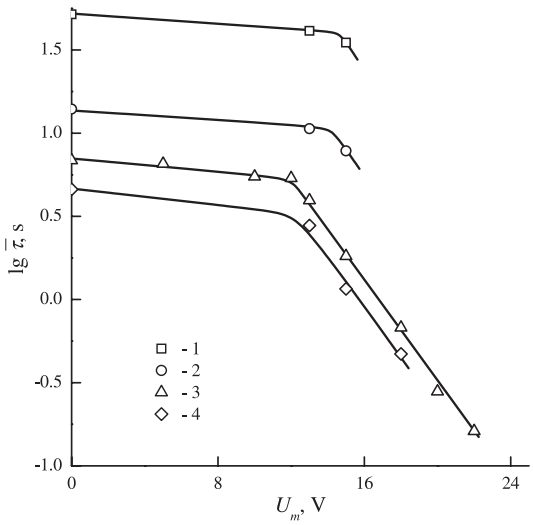

Figure 3. Dependence of the mean lifetime of superheated argon on the voltage amplitude of the piezoradiator at temperatures: 1—$T = 133.0$ K, 2—134.0 K, 3—135.0 K, 4—135.5 K.

data on thermophysical properties of argon [14–16]. It was assumed that the surface tension of critical bubbles, whose characteristic radius was approximately 4 nm, did not depend on their size and coincided with the surface tension at the flat interface (capillary approximation) [16].

A considerable deviation of the experimental curve from the theoretical line at large times ($U_m = 0$) indicates that boiling-up here is initiated by the action of external factors. Such factors initiating boiling-up may be the natural radiation background [3], and also the gas concentrated in defects of the inner surface of the measuring cell [17]. The effect of γ-radiation on the mean lifetime of superheated liquid argon was investigated in [6]. An increase in the intensity of ionizing radiation leads to a decrease in the expectation time for boiling-up of a superheated liquid. In this case the character of the dependence $\bar{\tau}(T)$ close to the boundary of spontaneous boiling-up of liquid argon in a field of γ-radiation is qualitatively similar to such a dependence in an acoustic field (figure 2).

The dependence of the mean lifetime of superheated argon on the amplitude of the voltage on the piezoradiator is shown in figure 3. On the dependence of $\lg \bar{\tau}$ on U_m one can distinguish two sections: the section where $\lg \bar{\tau}$ is practically independent of U_m, and the section with a linear dependence $\lg \bar{\tau}(U_m)$. With decreasing temperature the amplitude of the voltage at which the character of the dependence $\lg \bar{\tau}(U_m)$ changes increases weakly.

The appearance of sections with a constant value of $\lg \bar{\tau}(U_m)$ on the curves $\lg \bar{\tau}(U_m)$ can be connected in the absence of a direct acoustic contact between the piezoradiator and the liquid. The absorption of an acoustic signal in the cell wall leads to the 'delay effect'.

3. Comparison with homogeneous nucleation theory

Calculation of the liquid cavitation strength in an acoustic field by homogeneous nucleation theory presupposes a knowledge of the amplitude of the sound pressure p_m. An experimental determination of this quantity is hampered by the small volume of the cavitating liquid. To evaluate p_m, we used an approach based on the statistical character of boiling-up of a metastable liquid.

We shall assign the periodic pressure pulsations in the form

$$p(t) = p_0 + p_m \cos \omega t, \tag{4}$$

with $p_m \ll (p_s - p_0)$, and $\omega \ll 1/\tau_n = 10^9$ Hz, where τ_n is the nonstationarity period (lag time). The latter inequality guarantees stationarity of the bubble size distribution in an acoustic field. Under continuous changes of pressure the stationary nucleation rate J changes as well. The nucleation rate logarithm may be presented in the form of the following series

$$\ln J(p) = \ln J(p_0) + \frac{\partial \ln J(p_0)}{\partial p}(p - p_0) + \cdots. \tag{5}$$

We shall restrict ourselves to the first two terms of the expansion (5). Owing to the weak dependence of the pre-exponential factor in (1) on thermodynamic state parameters for the coefficient with the linear expansion term we have [18]

$$\frac{\partial \ln J(p_0)}{\partial p} = -G_p \simeq -\left(\frac{\partial G}{\partial p}\right)_T = -\frac{1}{2}\frac{\upsilon_*}{k_B T}\left(1 - \frac{\rho_V}{\rho_L}\right), \tag{6}$$

where υ_* is the volume of a critical bubble.

Homogeneous nucleation is a statistical process. Both the number of forming nuclei and the moments of their appearance are of random probabilistic character. The condition that in the volume V of a superheated liquid in a certain mean expectation time $\bar{\tau}$ there appears one viable nucleus is written as

$$V \int_0^{\bar{\tau}} J[T, p(t)]\, \mathrm{d}t = 1. \tag{7}$$

Substitution of (1), (5), and (6) into (7) gives

$$J(p_0)V \int_0^{\bar{\tau}} \exp[G_p(p_0)\, p_m \cos \omega t]\, \mathrm{d}t = J(p_0)V\bar{\tau} I_0[G_p(p_0)\, p_m] = 1. \tag{8}$$

Here $I_0(x)$ is the Bessel function of an imaginary argument [19].

From (8) we have

$$\bar{\tau} = \frac{\bar{\tau}_0}{I_0(G_p p_m)}, \tag{9}$$

where $\bar{\tau}_0 = [J(p_0)V]^{-1}$ is the mean lifetime of a superheated (stretched) liquid in the absence of an acoustic field. At small values of $G_p p_m$ ($G_p p_m < 3$) formula (9) may be presented as [20]

$$\bar{\tau} = \bar{\tau}_0 \cdot \frac{G_p p_m}{\mathrm{sh}(G_p p_m)}. \tag{10}$$

Formulae (9) and (10) make it possible to evaluate the amplitude of the sound pressure p_m by experimental data on the mean lifetime of a superheated liquid in an acoustic field and in its absence. The value of G_p may be found from

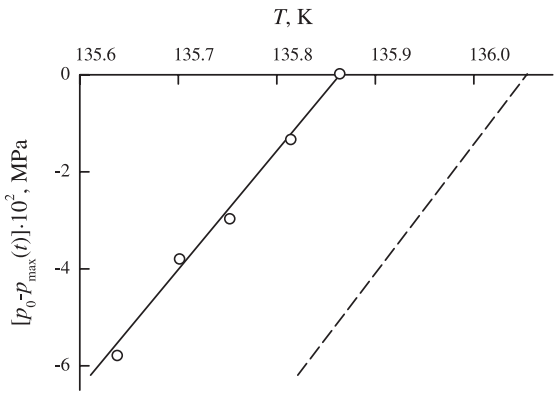

Figure 4. The change of cavitation strength $p_0 - p_{\max}(t)$ of liquid argon in an acoustic field as a function of temperature ($J = 10^7$ s^{-1} m^{-3}). The dashed line shows calculation by homogeneous nucleation theory.

homogeneous nucleation theory or obtained by experiment from the half-width of the distribution of liquid boiling-up events in pressure with its continuous lowering [12, 13]. In our calculations we assumed $G_p = 130\ 1$ MPa^{-1}.

The temperature dependence of the change of cavitation strength $p_0 - p_{\max}(t)$ of liquid argon at a nucleation rate $J \simeq 10^7$ s^{-1} m^{-3} is presented in figure 4. Shown therein are the results of calculating the dependence of $p_0 - p_{\max}(t)$ on T by homogeneous nucleation theory (equations (1)–(3), capillary approximation). To a first approximation these dependences are linear. There is good agreement in the slopes of the curves, which indirectly confirms the legitimacy of the assumptions made. The discrepancy in experimental and theoretical values of $p_0 - p_{\max}(t)$ may be attributed to the neglect in homogeneous nucleation theory of the dependence of the surface tension of vapor-phase nuclei on the curvature of the separating surface. As shown in [12], for simple liquids at positive pressures and temperatures close to the critical point the surface tension of vapor bubbles at the boundary of a liquid spontaneous boiling-up is 5–7% less than at a flat interface.

More complicated is the mechanism of acoustic cavitation in a region of liquid initiated boiling-up. Here a considerable decrease in the mean lifetime of a stretched liquid in an acoustic field cannot be explained only by decreasing effective pressure in the liquid. Experiments of the measurement of superheated liquid lifetimes under natural conditions [3, 12, 13] testify that the time $\bar{\tau}_{\text{in}}$ from which the bends of experimental curves begin has a very weak pressure dependence. In liquid argon the value of $\bar{\tau}_{\text{in}}$ changes from 2 to 5 s at a pressure increase from 0.2 to 15 MPa [6]. The constancy of $\bar{\tau}_{\text{in}}$ is one of the arguments in favor of the radiation background nature of the bends of experimental curves. The action of the radiation background that initiates boiling-up manifests itself with a certain, approximately constant frequency, which does not depend on the pressure in the liquid.

An abrupt decrease in the value of $\bar{\tau}_{\text{in}}$ from 5 to 0.4 s when a voltage of amplitude 18 V is supplied to the piezoradiator ($p_m \simeq 0.05$ MPa) may be caused both by the increase with time owing to a rectified heat transfer [21] of the average sizes of bubbles generated by high-energy particles and gas occlusions, and by an increase in the frequency of appearance of viable nuclei in the liquid.

4. Conclusion

Experimental investigations of spontaneous boiling-up kinetics of stretched (superheated) liquids in weak acoustic fields testify that depending on the state variables (p, T) cavitation may proceed both as a result of homogeneous nucleation and by way of activation of the evaporation centers currently available in the liquid. Spontaneous acoustic cavitation thresholds are adequately described by classical homogeneous nucleation theory. The mechanism of activation of completed and easily activated boiling sites by weak acoustic fields calls for a special consideration.

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