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On the surface tension corrections in nonuniform and nonequilibrium liquid–gas systems

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Abstract

Expressions for calculating corrections to surface tension in nonuniform and nonequilibrium liquid–gas systems are obtained in terms of the thermodynamical properties of equilibrium systems in the framework of van der Waals theory of capillarity. It is shown that the pressure and temperature nonuniformities in the interface lead to significant surface tension corrections at high-rate phase transitions. The effect of thermodynamical instability and dissipation of the liquid–gas interface at intense evaporation is predicted.

Keywords: Surface tension corrections; Liquid–gas interface; Tolman length; Phase transitions

1. Introduction

The aim of current investigation is to obtain simple relations for corrections to surface tension of liquid–vapor system, caused by nonuniformities of field variables—pressure and temperature—inside the interface. The bright example of such correction is a size effect—dependence of surface tension of bubbles and droplets on curvature of interface [1]. Nonuniformity of normal component of pressure tensor inside interface of bubbles and droplets changes their surface tension. This correction is noticeable only for very small formations, but it is quite important for processes of nucleation and growth of new phase and thus more deep insight into nature of this phenomenon is helpful in understanding of physics of liquid–gas phase transitions in microscale systems. But the sign and value of this correction for surface tension of nuclei are still uncertain and theoretical, experimental and even simulation results allow different interpretations (see e.g. [2,3]).

In addition to the size effect there are some experimental evidences of influence of evaporation mass flux on surface tension [4]. Our recent simulations of nonequilibrium systems [5–7] show that presence of temperature gradient at interface at intense phase transitions affects the surface tension. Here we should distinguish the well known Marangoni effect, which is connected with temperature gradient along the interface, and this effect, which appears due to nonuniformity of temperature field across the liquid–gas interface.

Usually we consider a liquid–gas interface as a surface of zero thickness and apply boundary conditions for equations of continuous matter there, that corresponds to Gibbs approach [1]. This common technique is useful and very effective when the interface thickness is negligibly small. And for most cases it does so, because at normal conditions the interface thickness is about several nanometers. We can estimate the effective scale of temperature nonuniformity in liquid at interface as $\lambda_l \Delta\rho / q(\partial\rho/\partial T)_l$, where $\Delta\rho = \rho_l - \rho_g$, and if this effective scale becomes comparable with interface thickness we have to take into account a modification of internal structure of interfacial zone. When we consider nucleation, when the radii of critical nuclei are about tens of nanometers, or investigate microscale processes we have to examine possibility of corrections arising due to nonzero interface thickness and its internal structure. And this corrections can be significant and we have to be careful applying ordinary boundary corrections to such objects as small bubbles and droplets.

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Nomenclature

a	thermal diffusivity
$B(T)$	parameter from Eq. (7)
C_P	heat capacity
$C(T)$	parameter from Eq. (7)
d	Tolman length
$G(T)$	parameter from Eq. (7)
H_{fg}	specific heat of evaporation
j	mass flux
Ja	Jacob number
K_{T}	isothermal compressibility
L	interface thickness
l_{T}	characteristic thermal length
l_{w}	Widom's characteristic length
m	parameter from Eq. (2)
n	numerical density
P	pressure
q	heat flux
R	radius of bubble or droplet
r	radius
r_{c}	correlation radius
T	temperature
x	coordinate tangential to a surface
z	coordinate normal to a surface

Greek symbols

Γ	difference between components of pressure tensor
γ	surface tension
Δ	difference
λ	thermal conductivity
ρ	density
τ	time
Ω	great thermodynamical potential

Subscripts and superscripts

c	critical point
g	bulk gas
l	bulk liquid
s	saturation
ξ	perturbed functions

Our aim in present article is to connect macroscale and microscale approaches, basing on structure of interface, which we know from simulations and suggest some simple method of calculation of corrections for surface tension, which appear for curved interfaces, at intense phase transitions and under an action of external fields. These corrections should be expressed in the terms of thermophysical properties of bulk phases at equilibrium, because for most of the substances these properties are well investigated experimentally and theoretically and can be used in engineering calculations.

Our investigation is based on the modern van der Waals theory of capillarity [1,8]. It is clear, that not all of our considerations can be obtained within the thermodynamic approach [9] and we use the results of our molecular dynamics simulations [5–7,10] to verify our phenomenological approaches.

Surface tension is commonly defined as an excess surface free energy and can be calculated using the mechanical definition as the integral of difference between normal and tangential components of pressure tensor:

$$\gamma = \int_{z_1}^{z_g} \Gamma(z)\,\mathrm{d}z = \int_{z_1}^{z_g} (P_z(z) - P_x(z))\,\mathrm{d}z, \tag{1}$$

where x and z are the tangential and normal to the surface coordinates, and hereafter the subscripts "l" and "g" are refer to bulk liquid and gas. According to the van der Waals theory the influence of fluid inhomogeneity is described by the square of the density gradient term in free energy:

$$\gamma = \int m[\rho(z), T]\rho'(z)^2\,\mathrm{d}z \approx m\frac{(\Delta\rho)^2}{L}, \tag{2}$$

where $\rho(z)$ is the interface density profile, $m[\rho(z), T]$ is a parameter, which in common case is a weak function of density and related to a direct correlation function of a fluid [1], L is the interface thickness. Definition (2) corresponds to the minimum of the great thermodynamic potential Ω of a two-phase system and is equivalent to definition (1). This is why definitions (1) and (2) are valid not only at thermodynamic equilibrium, but also in nonequilibrium cases, e.g. during evaporation or condensation.

Since surface tension is determined by the material distribution inside the interfacial layer $\rho(z)$, any external influence affecting the density distribution should lead to a modification of surface tension. If, for any reason, pressures or temperatures of bulk phases change, the surface tension can change too. This modification of surface tension may be estimated from Eq. (2) by using the thermodynamic perturbation technique [9] and expanding the following equation:

$$\frac{\gamma_\xi}{\gamma} = \frac{L}{L_\xi}\left(\frac{\Delta\rho_\xi}{\Delta\rho}\right)^2. \tag{3}$$

The subscript ξ here denotes the perturbed functions, i.e. functions corresponding to the system in the shifted thermodynamic state. Using this general relation we can investigate not only equilibrium, but also nonequilib-

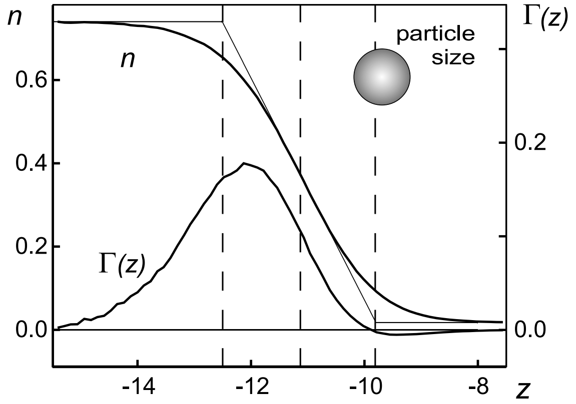

Fig. 1. Structure of interface: profiles of the numerical density n (bulk liquid boundary is $z_l = -12.5$, bulk gas boundary is $z_g = -9.8$, equimolar surface is $z_e = -11.1$) and $\Gamma_{(z)}$ obtained from MD simulations [10] for $T = 0.72T_c$, all variables reduced via Lennard–Jones potential parameters σ and ε.

rium systems and find corrections to the surface tension arising due to phase transitions.

Our main assumption is that the interface thickness L is proportional to a density fluctuation correlation radius in bulk liquid r_c^l. Indeed, far from the critical point the vapor density is significantly lower than the density of liquid and almost all the mass of interface layer is concentrated near the liquid boundary. Fig. 1 based on the results of our molecular dynamics simulations illustrates this. At the interface, density smoothly changes from bulk liquid to bulk gas and most of the contribution to the surface tension is due to the liquid part of the interfacial layer. In the vicinity of the critical point the difference between phases disappears and the correlation radii of liquid and gas become close and the interface thickness can be estimated to be proportional $2r_c^l$.

The correlation length itself scales as follows:

$$r_c^l \sim \rho_l\sqrt{K_T^l}, \tag{4}$$

where $K_T^l = \rho_l^{-1}(\partial\rho/\partial P)_T^l$ is the isothermal compressibility of liquid.

Combining (3) and (4) we get:

$$\frac{\gamma_\xi}{\gamma} = \frac{r_c^l}{r_{c\xi}^l}\left(\frac{\Delta\rho_\xi}{\Delta\rho}\right)^2 = \frac{\rho_l}{\rho_{l\xi}}\left(\frac{\Delta\rho_\xi}{\Delta\rho}\right)^2\left(\frac{K_T^l}{K_{T\xi}^l}\right)^{1/2}. \tag{5}$$

Using Eq. (5) we can obtain corrections to surface tension through the equilibrium thermodynamic properties of coexisting phases with planar interface [9].

For the purposes of calculations interface thickness can be expressed explicitly within our approach assuming the density profile in the form of hyperbolic tangent [1]:

$$L = 24 l_w\left(\frac{\rho_l}{\Delta\rho}\right)^2, \tag{6}$$

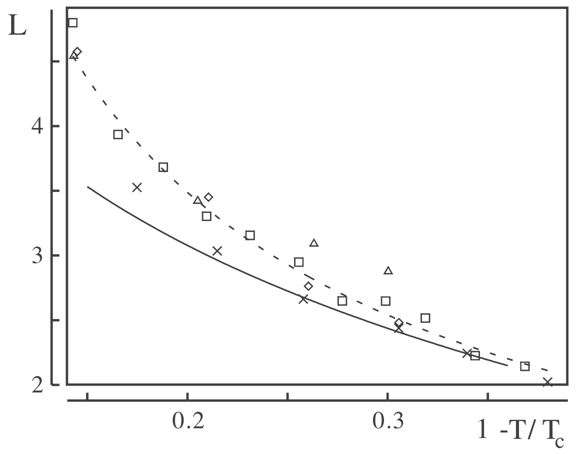

Fig. 2. Interface thickness: comparison of our MD results [10] (dots are simulation results and dashed line is the fitting line) and calculations for argon with the use of (6); (◇) ϕ_{tr}, $r_{cut} = 2.5$; (□) ϕ_{spl}, $r_{cut} = 2.5$; (×) ϕ_{tr}, $r_{cut} = 3.5$; (△) ϕ_{tr}, $r_{cut} = 5.5$.

where $l_w = \gamma K_T^l$ is Widom's characteristic length. We compared the results for interface thickness from MD simulations and calculations for argon by (6) and found good agreement at $1 - T/T_c > 0.15$ (see Fig. 2).

2. The first order correction to the surface tension

When some perturbations occur in a thermodynamic system, i.e. a pressure change, temperature drop or something else, the modification of a surface tension may be estimated with the first nonvanishing terms of the expansion series of the thermodynamic functions in Eq. (5) at the point of two-phase equilibrium with planar interface $(P_g = P_l = P_s(T),\ T_g = T_l = T_s(P),\ \gamma = \gamma(T_s))$. Indeed, such technique is valid only for small values of perturbation parameters $\partial P = P_\xi - P_s,\ \partial T = T_\xi - T_s$, which are the changes of physical parameters at the interface. For simplicity we will consider from now on the case of low gas density $\rho_l \gg \rho_g$ and neglect terms containing ρ_g/ρ_l, which lead to corrections of higher order. This assumption is valid far from the critical point. One can obtain the following equation for the surface tension correction:

$$\frac{\gamma_\xi}{\gamma} = 1 + \frac{3}{2}\frac{l_w}{\gamma}[(1 - B(T))\Delta P - (1 + G(T))C(T)\Delta T], \tag{7}$$

where $\Delta P = P_\xi^l - P_s$, $\Delta T = T_\xi^l - T_s$

$$B(T) = \frac{\rho}{3}\left(\frac{\partial^2\rho}{\partial P^2}\right)_T\left(\frac{\partial P}{\partial\rho}\right)_T^2 < 0,$$

$$G(T) = -\frac{\rho}{3}\left(\frac{\partial^2\rho}{\partial P\partial T}\right)\left(\frac{\partial P}{\partial\rho}\right)_T\left(\frac{\partial T}{\partial\rho}\right)_P > 0, \tag{8}$$

$$C(T) = \left(\frac{\partial P}{\partial T}\right)_\rho > 0.$$

Table 1
Estimation of parameters from Eq. (7) for water

P_s (MPa)	T_s (K)	$1-B(T)$	$1+G(T)$	l_w (nm)	l_T (nm)	$-d$ (nm)
0.1	372.78	2.7	2.0	0.3	122	0.12
1	453.03	2.8	2.2	0.32	14.2	0.14
2	485.52	3.0	2.4	0.35	4.8	0.15
4	523.48	2.8	2.6	0.39	2.2	0.17
6	548.71	3.2	2.8	0.42	1.4	0.19
8	568.13	3.1	2.8	0.46	0.96	0.21
10	584.11	3.5	3.0	0.49	0.76	0.23
15	615.27	2.8	3.7	0.57	0.38	0.28

The properties of water and values of thermodynamic derivatives are taken from Ref. [11].

All derivatives are calculated for liquid at phase boundary. The results of calculations of parameters from (7) for water are presented in Table 1. Analyses show that up to $P \approx 0.8P_c$ within the estimation error $B(T)$ and $G(T)$ are weak functions of temperature and pressure, the results of calculations of $B(T)$ and $G(T)$ for *para*-hydrogen with the use of NBS data [14] are close to the results for water [9]. Thus for $T < 0.8T_c$ we have $1-B(T) \approx 1+G(T)$ and in engineering calculations it is possible to use simple semi-empirical approximation for the surface tension correction:

$$\frac{\gamma_\xi}{\gamma} = 1 + (3\cdots4)\frac{l_w}{\gamma}[\Delta P - C(T)\Delta T]. \tag{9}$$

At high pressures and temperatures it is necessary to take into account terms containing the ρ_g/ρ_l ratio. Close to the critical point the corrections should be calculated from (5) using the scaling equation of state. In the following we will discuss how to apply the surface tension correction (7) to a case of curved interface (calculation of the Tolman length) and to high-rate phase transitions.

3. Curvature effect on surface tension

The dependence of surface tension on the curvature of phase boundary (the size effect) becomes appreciable for liquid drops or vapor bubbles with radii R of units and tens of nanometers. The classic Tolman expression (under condition that $R \gg L \sim r_c$) is:

$$\frac{\gamma(R)}{\gamma} = 1 \pm \frac{2d}{R}, \tag{10}$$

where the sign "+" is for bubbles and "−" is for droplets (schematic boundary conditions see at Fig. 3). The value and sign of the Tolman parameter d cannot be defined within rigorous thermodynamic approach and they are still a matter of scientific investigations and discussions. It becomes possible to estimate d in the van der Waals approach by using Eq. (7). Under conditions of thermodynamic equilibrium when $\Delta T = 0$, the difference between pressure in bulk liquid and saturation pressure is determined by Kelvin equation:

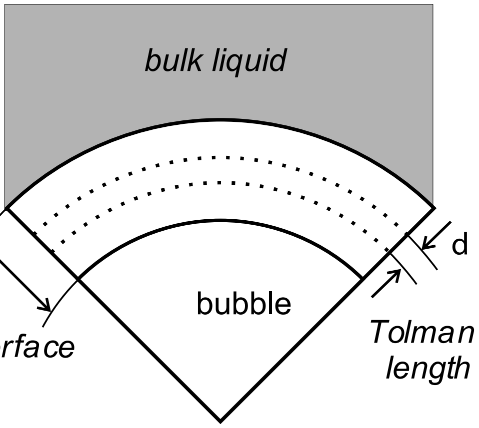

Fig. 3. Schematic view of curved liquid–vapor boundary.

$$\Delta P_i = \pm\frac{2\gamma}{R}\frac{\rho_i}{\Delta\rho}, \tag{11}$$

where "+" is for drops and "−" for bubbles, and subscripts "*i*" denotes one of the bulk phases: gas or liquid. Substituting (11) to (7) and assuming $\rho_g/\rho_l \ll 1$ we get equation for the Tolman length:

$$d = -\tfrac{3}{2}l_w(1-B(T)). \tag{12}$$

Because $B(T) < 0$ the Tolman parameter turns out to be negative for all temperatures and surface tension is always smaller for bubbles and decreases with an increase of curvature of surface R^{-1}. This is in agreement with the results of many experiments (see e.g. [12,13]). The values of d for water are presented in Table 1 and the surface tension for critical bubbles in liquid argon is shown in Fig. 4.

For superheated liquids near the spinodal critical, nucleus sizes R^* are small and the surface tension correction can significantly reduce the nucleation work. This can noticeably lower the temperature of limiting superheat $T_{\lim}$ at given pressure and nucleation rate. The shift of $T_{\lim}$ due to the size effect can be estimated with the use of Eq. (12). The results of calculations for argon [9] are in agreement with results of experiments [15].

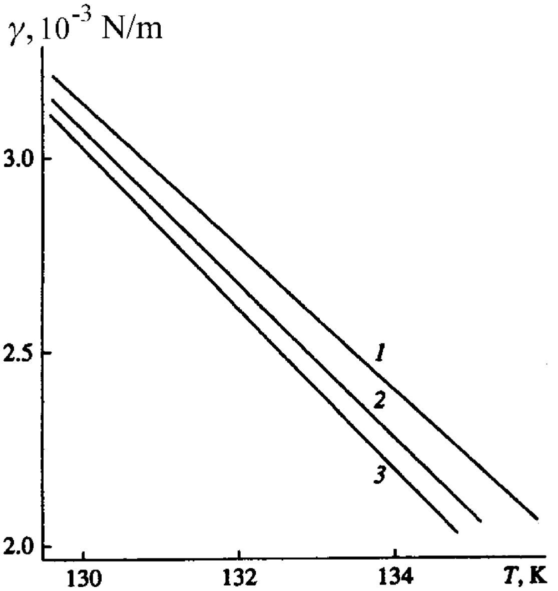

Fig. 4. Surface tension of argon for: (1) planar interface; (2) critical bubble at $P = 0.1$ MPa; (3) critical bubble at $P = 1$ MPa.

4. Surface tension correction at high-rate evaporation

At evaporation heat is conducted to interface through bulk liquid and carried away with gas flow. Boundary conditions compared with the results of MD simulations are presented on Fig. 5. At high rate evaporation, there appears a temperature jump at the interface due to its nonzero thickness and high temperature gradient in bulk liquid. Thus the thermodynamic state of liquid near the interface is shifted and we can use the difference $\Delta_T = T_l - T_s(P_s)$, where T_l is the temperature of bulk liquid at dividing surface, for calculation in (7). On the other hand, far from the critical point density of bulk gas is too low and we can assume $T_g = T_s$, because even if temperature of gas and saturation temperature differ their difference will give a correction of higher order and we do not need to take it into account.

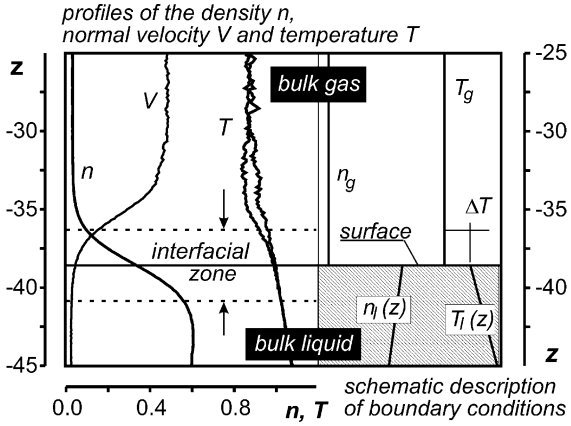

Fig. 5. Boundary conditions at the liquid–vapor interface at evaporation. At the left (from MD simulations [7]): the profiles of the numerical density n, normal velocity V and kinetic temperatures T (the Knudsen layer is well seen); at the right: the schematic description of thermodynamic boundary condition for the system.

We assume that in our case the reference thermodynamic state is bulk liquid at the interface. Indeed, it is well known, that at evaporation from a flat interface the pressures in bulk liquid and gas differ. The pressure jump due to recoil Mesherskii forces is given by [16]:

$$P_l - P_g = j^2 \frac{\Delta\rho}{\rho_l \rho_g}, \tag{13}$$

where j is the interfacial mass flux. But this forces are due to the acceleration of the evaporating gas and according to condition $j = \rho(z)v_z = \text{const.}$, the increase in velocity is connected with density. Therefore the pressure jump occurs in the gas part of interfacial layer, as clearly shown from the molecular dynamics simulations (see Fig. 5). Since the pressure decreases outside the interface, it does not affect surface tension due to the low gas density. Hence at calculation of first order correction we may set $\Delta P = 0$.

As a result from the Eq. (7) we obtain:

$$\frac{\gamma_{\text{evap}}}{\gamma} = 1 - \frac{3}{2}\frac{l_w}{\gamma}[1 + G(T)]C(T)\Delta T, \tag{14}$$

where $\Delta T = qL/\lambda$ is the temperature drop inside the interface zone, and q the heat flux at the interface, and λ the liquid thermal conductivity. The surface tension correction can be expressed in the following form:

$$\frac{\gamma_{\text{evap}}}{\gamma} = 1 - \frac{q}{q^*}, \quad q^* = \frac{\lambda\gamma}{36[1 + G(T)]C(T)l_w^2}. \tag{15}$$

One can see that evaporation lowers the surface tension. The results of our molecular dynamics simulations confirm this effect [6,7]. The characteristic heat flux q^* for water varies from 4×10^9 W/m^2 at the normal boiling point to 2.6×10^8 W/m^2 at $P = 10$ MPa (Fig. 6), and corresponds to conditions when the liquid–gas interface becomes unstable due to a strong decrease of the surface tension. Far from the critical point the decrease of the surface tension at evaporation becomes significant for heat fluxes about $q \sim 10^6$–10^8 W/m^2. Such values of q can be reached at evaporation of liquids under an action of high power radiation, in experiments with electric wire explosion, during explosive boiling of strongly superheated liquids, during atomization of liquid fuels in combustors of rocket engines, etc.

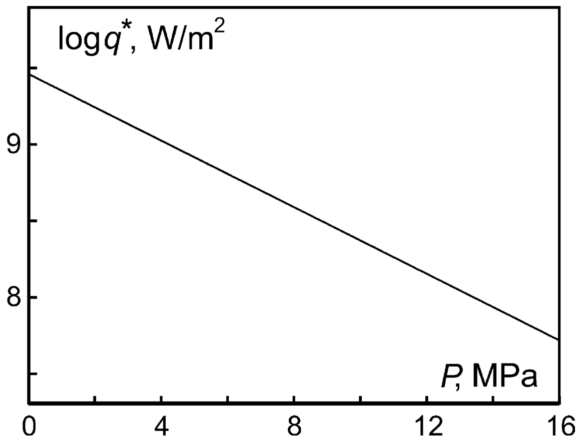

Fig. 6. The characteristic heat flux for water.

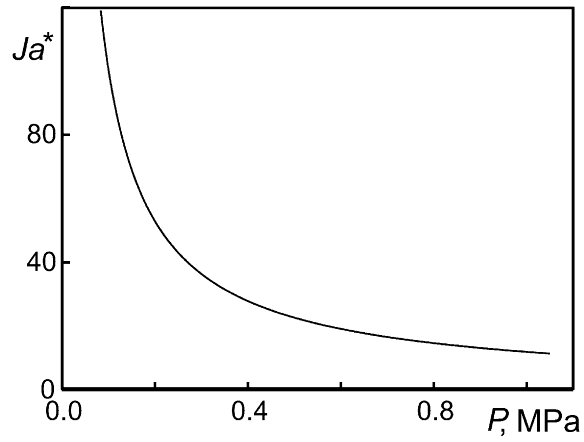

Fig. 8. The critical Jacob number for water.

5. Bubble growth in a superheated liquid

Now let consider a bubble growing in a superheated liquid (Fig. 7). In this case the reference point is the pressure in bulk liquid P_l and the saturation temperature for this pressure $T_s(P_l)$. At constant pressure and high Jacob numbers ($Ja \gg 1$) the temperature gradient at the interface of a bubble of radius R is equal to:

$$\left(\frac{\partial T}{\partial r}\right)_R = \rho_l \frac{H_{fg}}{\lambda_l}\frac{dR}{d\tau}, \tag{16}$$

$Ja = \rho_l C_{Pl}(T_l - T_s)/H_{fg}\rho_g$, $T_l - T_s$ is the liquid superheat, H_{fg} is the latent heat of evaporation. The time dependence of bubble radius $R(\tau)$ can be estimated by the Plesset–Zwick approximation [17]:

$$R(\tau) = \left(\frac{12}{\pi}\right)^{1/2} Ja(a_l\tau)^{1/2}, \tag{17}$$

where $a = \lambda/\rho C_P$ is the thermal diffusivity. Combining Eqs. (16) and (17) we obtain:

$$\left(\frac{\partial T}{\partial r}\right)_R = \frac{H_{fg}\rho_g}{C_{Pl}\rho_l}\frac{6}{\pi}Ja^2\frac{1}{R}. \tag{18}$$

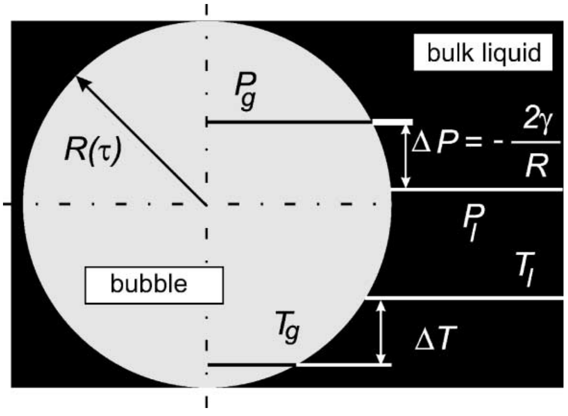

Fig. 7. Boundary conditions at the liquid–vapor interface of growing bubble. The heat-dependent model of growth for $Ja \gg 1$.

Thus, with the aid of (18) and (6) we can express the temperature drop ΔT inside the bubble interface. Substituting ΔT into (14) we get:

$$\frac{\gamma_{evap}}{\gamma} = 1 - \frac{216}{\pi}(1 + G(T))\frac{l_w}{l_T}Ja^2\frac{l_w}{R},$$
$$l_T = \frac{\gamma C_{Pl}\rho_l}{C(T)H_{fg}\rho_g}, \tag{19}$$

where l_T is the characteristic thermal length similar to the Widom's length l_w. For the critical nucleus whose radius is given by:

$$R^* = \frac{2\gamma T_s(P)}{H_{fg}\rho_g(T_l - T_s(P))}, \tag{20}$$

one obtains the following formula valid at high Jacob numbers and low pressures:

$$\frac{\gamma(R^*)}{\gamma} = 1 - \left(\frac{Ja}{Ja^*}\right)^3,$$
$$Ja^* = \left(\frac{108}{\pi}(1 + G(T))\left(\frac{l_w}{l_T}\right)^2\frac{H_{fg}\rho_g}{T_s(P)C(T)}\right)^{-1/3}. \tag{21}$$

The values of l_w and l_T for water are given in Table 1, and Ja^* on Fig. 8.

6. The effect of dissipation of interface at intense evaporation

Equations (15) and (21) show that evaporation at high heat fluxes leads to a significant decrease in surface tension. For water, Eq. (15) predicts that surface tension disappears for temperature difference inside the interface $\Delta T \approx 40$ K and for the growing bubble the superheat of liquid estimated by Eq. (21) is $T_l - T_s \approx 30$ K. But Eqs. (15) and (21) are obtained under assumption that the correction is much smaller than the value of surface tension and hence these temperature differences give only qualitative estimation. At the same time, these equations predict new effect, that under the action of intense evaporation the interface may disappear at temperatures lower than the critical temperature. We can estimate the critical heat fluxes by using following considerations.

Inside a liquid–gas interface at temperatures far from the critical T_c, the gradient of density correlation fluctuation radius across the interface is less than unity $|\Delta r_{c\perp}| \lesssim 1$. Indeed, when density correlations inside a substance vanish rapidly the continuity of a fluid becomes broken and the system decomposes into separate parts. Thus the condition:

$$|\nabla r_{c\perp}(q)| \sim 1, \tag{22}$$

corresponds to the heat fluxes for which interface disappears due to strong evaporation and surface tension vanishes. Using the scaling equations for the correlation radius it is possible to obtain the critical heat flux:

$$q^{**} \sim \frac{\lambda T_c}{r_0 v} |t|^{v+1}, \tag{23}$$

where r_0 and v are the critical amplitude and critical exponent for the density correlation radius r_c, $|t| = |1 - T/T_c|$. For water at $|t| > 10^{-1}$ the critical heat flux is about $q^{**} \sim 10^{10}$–10^{11} W/m^2, that corresponds to the temperature drop inside the interface in about $\Delta T^* \sim 10$–100 K that is in the agreement with predictions of Eqs. (15) and (21).

The effect of interface dissipation and disappearance of surface tension at $T < T_c$ and $q \sim q^{**}$ assigns the thermodynamical limit of liquid–gas interface existence at conditions of intense evaporation. Apparently it can not be observed experimentally for bubbles and droplets with the sizes more than critical because even for heat fluxes $q < q^{**}$ the interface boundary becomes unstable due to hydrodynamic instabilities, first of all due to Landau and barocapillary instabilities [18]. Lowering of the surface tension due to heat flux at phase transition helps these instabilities to appear. On the other hand this effect can play important role at nuclei formation in superheated liquids.

7. Conclusion

According to the results of our investigation we can conclude, that nonuniformities of temperature and pressure, arising due to intense phase transitions or on curved interfaces lead to modification of properties of the interface and to modification of the surface tension. For the bubbles both for equilibrium and growing in superheated liquid the surface tension decreases, and also it decreases at intense evaporation from planar interface.

We obtained the simple thermodynamic equations, which permit to calculate the surface tension corrections with the aid of the properties of equilibrium bulk phases with planar interface. The proposed technique is useful for calculations for pure substances and on the base of Cahn–Hilliard theory can be extended to solutions.

Our results show that at high-rate phase transitions the interface can become unstable due to sudden decrease of the surface tension. There exist critical values of the heat flux q^{**} and temperature difference inside the interface ΔT^* which determine absolute thermodynamical limit of liquid–gas interface existence at evaporation.

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