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On phase change of a vapor bubble in liquid water

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Abstract

We consider a bubble of vapor and inert gas surrounded by the corresponding liquid phase. We study the behavior of the bubble due to phase change, i.e. condensation and evaporation, at the interface. Special attention is given to the effects of surface tension and heat production on the bubble dynamics as well as the propagation of acoustic elastic waves by including slight compressibility of the liquid phase. Separately we study the influence of the three phenomena heat conduction, elastic waves, and phase transition on the evolution of the bubble. The objective is to derive relations including the mass, momentum, and energy transfer between the phases. We find ordinary differential equations, in the cases of heat transfer and the emission of acoustic waves partial differential equations, that describe the bubble dynamics.

From numerical evidence we deduce that the effect of phase transition and heat transfer on the behavior of the radius of the bubble is negligible. It turns out that the elastic waves in the liquid are of greatest importance to the dynamics of the bubble radius. The phase transition has a strong influence on the evolution of the temperature, in particular at the interface. Furthermore the phase transition leads to a drastic change of the water content in the bubble, so that a rebounding bubble is only possible, if it contains in addition an inert gas.

In a forthcoming paper the equations derived are sought in order to close equations for multi-phase mixture balance laws for dispersed bubbles in liquids involving phase change. Also the model is used to make comparisons with experimental data on the oscillation of a laser induced bubble. For this case it was necessary to include the effect of an inert gas in the thermodynamic modeling of the phase transition.

1 Introduction

Mathematical modeling of fluid flow with phase change between liquid water and its vapor has many applications in science and technology, such as the study of cloud formation, of bubbles in boiling water, spray cooling in metal production or damage of ship propellers due to cavitation. For numerical computations of such flows, via mathematical equations describing the balances of mass, momentum and energy, one needs appropriate terms modeling the transfer of these physical quantities between the phases. It is a commonly found situation that one phase is dispersed in the other. Here we consider vapor bubbles containing some additional inert gas in liquid water.

The subproblem concerning the precise modeling of the evolution of a single bubble is a challenge, because various different phenomena are involved and strongly couple with each other. For example, here we meet heat conduction, elastic waves, phase transitions and diffusion. The latter occurs if the initial bubble is created by a laser beam, see e.g. Akhatov et al. [1] and especially Müller et al. [15], so that the bubble might contain inert gas, i.e. most probably hydrogen and oxygen, in the vapor due to the high temperature during the bubble creation process. We aim to reproduce as closely as possible measurements of the subsequent evolution of the bubble radius. Experimental results were obtained by

the group of Lauterborn in Göttingen, see Müller et al. [15]. We will refer to this as the Göttingen experiment in this paper. In the experiment one observes a collapsing and rebounding bubble. In fact, the bubble radius increases to a maximum, then decreases to a non-zero minimum, and hereafter the cycle is repeated, however, an apparent damping is observed. We will present numerical results using four different models in order to give an indication which terms are important. Detailed comparisons with experimental data will be reported in a forthcoming joint paper with the experimental group. A comparison of the observed evolution of a bubble with a model is a subtle problem, because the initial states of all physical quantities involved cannot be precisely measured experimentally. At this time we have to work with very incomplete experimental data.

In Akhatov et al. [1] the problem is treated numerically by a full system of coupled partial differential equations, which obviously is the correct description. However, the objective of our paper is to explore if a description by a much simpler system of equations is possible. To this end it is important to determine the influence of the various participating phenomena on the observed evolution. Thus we describe the problem by a hierarchy of model systems of increasing complexity. We start out with the model of the undamped oscillation of a bubble immersed in an incompressible liquid at constant temperature without phase transition, given by the Rayleigh-Plesset equation [12], [10]. Hereafter we take the liquid-vapor phase transition into account. Next we consider in addition heat conduction. Finally elastic waves due to the compressibility of the liquid are incorporated.

In the Göttingen experiment a laser pulse is focussed inside a vessel of water. This leads to the formation of a plasma and creation of a vapor bubble that collapses and rebounds. If a liquid-vapor phase transition is allowed, the latter effect is only possible when an inert gas is present in the bubble. A pure water vapor phase cannot persist beyond the first collapse since the vapor phase is unstable under the conditions of the experiment. Currently it is not known by measurements what constitutes the inert gas. The most likely seem to be oxygen and hydrogen. So for this paper we are assuming this to be the case. We are considering the data described in Müller et al. [15]. Earlier experimental work on this topic is described in Akhatov et al. [1, 2] and Kurz et al. [8].

The theoretical study of a spherical bubble surrounded by liquid has quite a long history. Much of the literature is an extension of the 1917 paper of Rayleigh [14] on the pressure during collapse of a spherical bubble. He mentions that his work was motivated at that time by the noise emitted by a boiling kettle and the cavitation behind propelling screws. He considered a constant outer pressure and basically integrated the equations of motion, under the assumption of complete radial symmetry, of an incompressible, inviscid liquid. The work was then extended 1949 by Plesset [12] in a study on cavitation considering the case of a non-constant *external pressure* $P(t)$ leading to a bubble which grows and then collapses. This was observed in the cavitation experiments he discussed. Let us consider the *density* ρ of the liquid and the *pressure of the liquid at the interface* $p(R)$. In his paper he derived the second order nonlinear ordinary differential equation for the time dependent bubble radius R

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 + \frac{P(t) - p(R)}{\rho} = 0 \tag{1}$$

which has become known as the *Rayleigh-Plesset equation* of bubble dynamics.

Later extensions were summarized by Lauterborn [9] and studied numerically. Next Keller and Miksis [7] were interested in the acoustics of oscillating bubbles and included the effect

of acoustic radiation by considering compressibility. Also they carried the viscosity term with *viscosity* μ through the derivation. Further their derivation involved the *speed of sound* c in the liquid and the *pressure inside the bubble* $p_b(R)$. They considered a pressure at infinity perturbed by an incoming sound wave $P(t) = p_\infty - P\sin\omega\left(t + \frac{R}{c}\right)$. Assuming that the interface velocity is equal to the liquid velocity at the interface, i.e. the phase transition is ignored, this leads to the *Keller-Miksis equation* [7, (3.9)], [13] which in our notation is

$$\begin{aligned}\ddot{R}\left(\frac{4\mu}{\rho} - R(\dot{R}-c)\right) &= \frac{1}{2}\dot{R}^3 + \dot{R}\frac{p_b(R)-p_\infty}{\rho} - c\left(\frac{3}{2}\dot{R}^2 + \frac{4\mu\dot{R}}{\rho R} + \frac{2\sigma}{\rho R} - \frac{p_b(R)-p_\infty}{\rho}\right)\\ &\quad + R\frac{\dot{p}_b(R)}{\rho} + \left(1+\frac{\dot{R}}{c}\right)\frac{cP}{\rho}\sin\omega\left(t+\frac{R}{c}\right).\end{aligned}$$

This equation is only valid for small Mach numbers. We simplify the equation by setting $\mu = 0$. Then we divide by c and use the *Laplace-Young relation* $p_b(R) = p(R) + \frac{2\sigma}{R}$ to obtain

$$R\ddot{R}\left(1-\frac{\dot{R}}{c}\right) = \frac{3}{2}\dot{R}^2\left(\frac{\dot{R}}{3c}-1\right) + \dot{R}\frac{p_b(R)-P(t)}{\rho c} - \left(\frac{P(t)-p(R)}{\rho}\right) + R\frac{\dot{p}_b(R)}{\rho c}. \qquad (2)$$

Taking the limit $c \to \infty$ reduces this equation to the Rayleigh-Plesset equation (1). Prosperetti and Lezzi [13] have generalized these equations even further using singular perturbation methods. Wu and Roberts [17] considered a version of this equation coupled to radially symmetric Euler equations in order to study sonoluminescence. In Akhatov et al. [1] a model for the laser induced bubble experiment ist introduced. Near the recollapse of the bubble the bubble radius velocity becomes very high, so the Euler equations of gas dynamics are used to simulate this part of the bubble dynamics. The paper [1] includes quite extensive citations of the relevant literature.

As a result of our considerations we obtain a hierarchy of models consisting of a system of ordinary and partial differential equations describing the evolution of the bubble with the main objective to identify the various driving forces involved.

From the numerical computations we can clearly see that the elastic waves in the liquid are of greatest importance to the evolution of the bubble radius in comparison with the experiment. Phase transition and heat conduction play no significant role for movement of the bubble radius. The phase transition has an enormous influence on the evolution of the temperature, in particular at the interface. Furthermore the phase transition leads to a drastic change of the water content in the bubble, so that a rebounding bubble is only possible, if it contains in addition an inert gas.

After this introduction we have organized the paper as follows. In Section 2 we first start with the balances of mass, momentum and internal energy for an inviscid compressible fluid in Subsection 2.2, including the jump relations that describe the conservation properties at moving interfaces. Appropriate constitutive relations for mixtures are introduced in Subsection 2.3 and complemented by the entropy principle in the bulk and in Subsection 2.4 at liquid-vapor interfaces. The entropy principle is exploited in Subsection 2.5 to introduce the interfacial mobility. In Subsection 2.6 the constitutive relations for a vapor phase containing water vapor and oxygen as well as hydrogen as inert gases are formulated. Next we derive in Subsection 2.7 a kinetic equation for the evolution of the bubble mass

under phase transition. The condensation rate may be taken from well established kinetic considerations. An important point is that the evaporation rate at the interface is derived in a thermodynamically consistent way. In Section 3 we recall the data from the laser induced bubble experiment that we are modeling.

The central part of the paper is Section 4 in which we give a hierarchy of models for radially symmetric bubbles in order to study numerically which physical effects are important to explain the experimentally observed dynamics of laser induced bubbles. Starting point is the well known second order Rayleigh-Plesset equation in Subsection 4.3. In Subsection 4.4 the mass dynamics due to phase transition is introduced to give a system of implicit first order ordinary differential equations. Then we add in Subsection 4.5 the heat conduction in the liquid. Finally a wave equation for acoustic waves is coupled to the other phenomena. The details of the derivations of these models are given in Section 5. In Section 6 we present some of our numerical computations within the setting of these models. The paper ends with a short summary of the main conclusions.

2 Variables, equations of balance and local entropy principle

2.1 Basic variables and constitutive quantities

As a starting point we take a spherical bubble $B = B_R(0) = \{x \in \mathbb{R}^3 | \, |x| < R(t)\}$ of vapor sitting at the origin surrounded by the liquid. Here t is time and R the time dependent radius of the bubble. However, later on we also will treat the case of a bounded planar interface between liquid and vapor. For the case of the bubble we will work in radial coordinates assuming that all physical quantities depend on time and distance from the origin. All vector fields are then assumed to be radial.

We have one vapor bubble in a closed, very large control volume. One could assume either that the control volume is subject to a given constant *outer pressure* p_0 or that the volume in the control volume remains constant in time. Note that the physics of phase transition depends considerably on whether the pressure or the volume is held constant in the control volume. In the first case the phases cannot coexist in equilibrium, whereas in the second case they may coexist. In nature the gas phase usually consists of a mixture of several constituents, for example vapor, oxygen, and nitrogen. This again changes the physics of the phase transition, allowing for fog in the pressure controlled situation found in nature.

In this paper we do not consider the case of constant volume. We only consider the case that the outer pressure p_0 of the global control volume is held constant, e.g. in a container with a moveable piston that is controlled by the outer pressure.

2.2 Equations of balance in regular and singular points

Here we exclusively consider the gas and the liquid phase as inviscid heat conducting fluids. The gas consists of water vapor, oxygen, and hydrogen. The appearance of oxygen and hydrogen is due to the fact that a bubble is created by a focussed laser beam in water. This initially leads to a high temperature so that some of the water vapor dissociates into O_2 and H_2. In the following we will only consider temperatures under which no further chemical reactions take place. Furthermore, we study short time bubble evolutions during which

hydrogen and in particular oxygen cannot leave the bubble into the liquid. Consequently the liquid consists of pure water only, thus we describe the liquid by 5 variables which are the mass density ρ, velocity $\mathbf{v}$, and temperature T. In the gas we have as variables three partial mass densities ρ_a, with $a = W, H, O$ for water, hydrogen, and oxygen. Further, there are the velocity of the mixture $\mathbf{v}$ and the temperature T. Unless stated otherwise, physical fields are assumed to depend on the time variable $t \in \mathbb{R}$ and space variable $x \in \mathbb{R}^3$.

The determination of the variables relies on the local conservation laws for mass and momentum, as well as the balance law for internal energy. In regular points of the liquid phase these are

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \operatorname{div}(\rho \mathbf{v}) &= 0 && (3) \\ \frac{\partial \rho \mathbf{v}}{\partial t} + \operatorname{div}(\rho \mathbf{v} \otimes \mathbf{v} + p\,\mathbf{1}) &= 0 && (4) \\ \frac{\partial \rho u}{\partial t} + \operatorname{div}(\rho u \mathbf{v} + \mathbf{q}) &= -p\,\mathbf{1} \cdot \operatorname{grad} \mathbf{v}\,. && (5) \end{aligned}$$

Here we use the second order tensor $\mathbf{1} = (\delta_{ij})_{1 \le i,j \le 3}$ with $\delta_{ij} = 1$ for $i = j$ and $= 0$ for $i \neq j$, the tensor product of vectors $\otimes$ giving a second order tensor. The divergence of a second order tensor is the vector of divergences of each row and the product $\cdot$ of two such tensors is the scalar obtained by double contraction.

In these equations there are further quantities which are not among the basic variables. We call them constitutive quantities, and these are the internal energy density u, the heat flux $\mathbf{q}$ and the pressure p. The constitutive quantities are related to the basic variables in a material dependent manner, which will be given in Section 2.6.

In the gas phase, the equations for momentum and energy of the mixture have the same structure as (4) and (5). However here we have to consider 3 conservation laws for the partial mass densities

$$\frac{\partial \rho_a}{\partial t} + \operatorname{div}(\rho_a \mathbf{v}_a) = 0. \qquad (6)$$

The partial mass densities are defined so that they sum up to the mass density of the mixture, and the weighted sum of partial velocities gives the (barycentric) velocity of the mixture, i.e.

$$\rho = \sum_a \rho_a, \qquad \mathbf{v} = \sum_a \frac{\rho_a}{\rho} \mathbf{v}_a \qquad \text{which implies with (6)} \qquad \frac{\partial \rho}{\partial t} + \operatorname{div}(\rho \mathbf{v}) = 0.$$

The constitutive quantities are pressure p, internal energy e, and heat flux $\mathbf{q}$, as before, and the partial velocities $\mathbf{v}_a$, but these are usually substituted by the diffusion fluxes $\mathbf{J}_a = \rho_a(\mathbf{v}_a - \mathbf{v})$.

We will use the superscripts L, V and the subscript I to specify physical quantities Ψ in the liquid, the vapor, and on the interface respectively. The combination of the subscript I with one of the superscripts denotes the one sided limit $\Psi_I^{L,V}(t, x_I) = \lim_{x^{L,V} \to x_I} \Psi(t, x)$ of the quantity in the respective bulk fluid at the interface. The superscript ν together with I denotes the scalar obtained by projection of a vector on the chosen normal vector ν on the surface. We denote vectors, except for the spatial coordinate vector of a point, and higher order tensors in boldface.

Along the singular surface I between the phases we consider the normal vector ν pointing into the liquid region. It is moving with normal speed w^ν. Across this interface we use the jump bracket $[\![\Psi]\!] = \Psi_I^L - \Psi_I^V$ for any physical quantity Ψ. We have the following relations

$$[\![\rho_a(v_a^\nu - w^\nu)]\!] = 0 \tag{7}$$

$$\rho(v^\nu - w^\nu)[\![\mathbf{v}]\!] + [\![p\,\nu]\!] = 2\sigma k_m \nu \tag{8}$$

$$\rho(v^\nu - w^\nu)[\![u + \frac{p}{\rho} + \frac{1}{2}(\mathbf{v}-\mathbf{w})^2]\!] + [\![q^\nu]\!] = 0\,. \tag{9}$$

Here the interface is exclusively equipped with *surface tension* σ. This is the term appearing on the right hand side of the momentum balance. We ignore for example tangential heat and diffusion fluxes. Additionally we assume that the interface has no mass and no inertia, and in particular we ignore the dependence of the surface tension on the concentration of the constituents of the gas and the temperature. The complete interface relations may be found in Dreyer [4, Sections 5-14] and Müller [11, Section 2.2.2, Chapter 3]. The term k_m is the *mean curvature* of the surface, with $k_m = -\frac{1}{R}$ for a sphere and $k_m = 0$ for a plane.

The assumption that there is no oxygen and hydrogen in the liquid phase implies that the equations (7) can be written as

$$v_O^\nu = v_H^\nu = w^\nu \qquad \text{and} \qquad [\![\rho_W(v_W^\nu - w^\nu)]\!] = 0\,. \tag{10}$$

In contrast to the need of partial mass balances, even for a mixture we only have to take a single energy balance, as it was given above, because there is only one temperature of the mixture. However, for a reduction of the necessary numerical data, which are needed to evaluate the energy balance, it is useful to decompose the internal energy density, the pressure, and the heat flux of the mixture into the corresponding quantities of the constituents, according to the detailed description in [11] without quadratic terms of the diffusion fluxes we have

$$\rho u = \sum_{a\in\{W,H,O\}} \rho_a u_a, \qquad p = \sum_{a\in\{W,H,O\}} p_a, \quad \text{and} \quad \mathbf{q} = \sum_{a\in\{W,H,O\}} \left(\mathbf{q}_a + \left(u_a + \frac{p_a}{\rho_a}\right)\mathbf{J}_a\right). \tag{11}$$

In terms of these quantities the balance of internal energy (5) in the gas phase now reads

$$\sum_{a\in\{W,H,O\}} \left(\rho_a \dot{u}_a + \operatorname{div}(\mathbf{q}_a + \mathbf{J}_a \frac{p_a}{\rho_a})\right) = \frac{p}{\rho}\dot{\rho}\,, \tag{12}$$

where $()^\cdot = \frac{\partial}{\partial t}() + \mathbf{v}\cdot\operatorname{grad}()$ denotes the material time derivative. Across the interface the internal energy balance (9) assumes the form

$$q_L^\nu - \sum_{a\in\{W,H,O\}} q_a^\nu = \rho^L(u^L + \frac{p^L}{\rho^L})(v_L^\nu - w^\nu) - \sum_{a\in\{W,H,O\}} \rho_a(u_a + \frac{p_a}{\rho_a})(v_a^\nu - w^\nu)\,. \tag{13}$$

2.3 General constitutive model and local entropy principle for the bulk phases of the two phase system

The general constitutive model for the system under consideration relies on two functions for the *specific Helmholtz free energy*

$$\psi = u - Ts \tag{14}$$

that is a combination of specific internal energy u and *specific entropy* s. We assume that we have in the liquid resp. the vapor phase

$$\psi^L = \widehat{\psi}^L(T, \rho^L) \qquad \text{and} \qquad \psi^V = \widehat{\psi}^V(T, \rho_W, \rho_H, \rho_O) = \widetilde{\psi}^V(T, \rho^V, X_W, X_H),$$

where the *mole fractions* are defined by $X_a = \frac{\rho_a/m_a}{\sum_b \rho_b/m_b}$. The knowledge of the specific free energy allows in combination with the local entropy principle for the bulk phases to calculate the pressure p, the specific entropy s, the specific *Gibbs free energy* g, and the chemical potentials μ_a, see Müller [11],

$$p = \rho^2 \frac{\partial \widetilde{\psi}}{\partial \rho}\,, \qquad s = -\frac{\partial \widetilde{\psi}}{\partial T}\,, \qquad g = \psi + \frac{p}{\rho}\,, \qquad \mu_a = \frac{\partial \rho \widehat{\psi}}{\partial \rho_a}\,. \tag{15}$$

Moreover the Gibbs-Duhem equation for the two phases, viz.

$$g^L = \mu_W^L\,, \qquad g^V = \frac{\sum_{a=W,H,O} \mu_a^V \rho_a}{\rho} \tag{16}$$

is likewise a consequence of the entropy principle, which also gives the entropy flux ϕ, and an inequality that controls the flow of heat

$$\phi^L = \frac{\mathbf{q}^L}{T}\,, \qquad \phi^V = \frac{\mathbf{q}^V}{T} - \frac{\sum_{a=W,H,O} \mu_a \mathbf{J}_a}{T}\,, \qquad \mathbf{q} \cdot \operatorname{grad} \frac{1}{T} \geq 0. \tag{17}$$

2.4 The local entropy principle across the liquid-vapor interface

The entropy principle holds point wise in a given body, thus we also must have an entropy principle at the interface. In the current study that principle relies on two basic assumptions: (i) There is no tangential entropy flux within the surface, (ii) the tangential velocities of the bulk phases at the interface are zero.

In this case the entropy principle at the interface consists of two parts

$$[\![T]\!] = 0 \qquad \rho(v^\nu - w^\nu)[\![s]\!] + [\![\phi^\nu]\!] \geq 0\,. \tag{18}$$

The continuous temperature at the interface is denoted by T_I. In equilibrium the equality sign holds, whereas in nonequilibrium the left hand side of the inequality in (18) must be greater than zero. The axiom of continuous temperature across the interface can be given up. However, in this case the subsequent treatment of the problem becomes very involved.

2.5 Exploitation of the entropy principle

We introduce the jump of the *specific kinetic energy* $[\![e_{kin}]\!] = [\![\frac{1}{2}(\mathbf{v}-\mathbf{w})^2]\!] = [\![\frac{1}{2}(v^\nu - w^\nu)^2]\!]$. Next we multiply the entropy inequality (18) by T, subtract the result from the energy equation at the interface (9), and use (15) to obtain

$$\rho(v^\nu - w^\nu)[\![g + e_{kin}]\!] + [\![\sum_{a=W,H,O} \mu_a \mathbf{J}_a]\!] \leq 0\,. \tag{19}$$

Now we assume that oxygen and hydrogen cannot cross the interface from the bubble into the liquid, i.e. $v_O^\nu = v_H^\nu = w^\nu$. In this case the inequality (19), using $g^L = \mu_W^L$, can be reduced to

$$\rho_W(v_W^\nu - w^\nu)[\![\mu_W + e_{kin}]\!] \leq 0. \tag{20}$$

In nonequilibrium the flux $\rho_W(v_W^\nu - w^\nu)$ of mass across the interface is driven by $[\![\mu_W + e_{kin}]\!]$. For this reason that factor is called the driving force. The left hand side of (20) is of the form *flux* $\times$ *driving force*.

In thermodynamics it can be shown that in equilibrium the driving force must be zero. Thus the product assumes its maximum in equilibrium. We now assume that there is a relation between the flux and the driving force in non-equilibrium which usually is called *kinetic relation*. A consequence of the existence of the kinetic relation is that

$$[\![\mu_W + e_{kin}]\!] = 0 \qquad \Longleftrightarrow \qquad \rho_W(v_W^\nu - w^\nu) = 0\,, \tag{21}$$

see [6].

The simplest ansatz to satisfy the inequality in nonequilibrium is to assume that the two factors of the product are positively proportional to each other. We write

$$-4\pi R^2 \rho(v^\nu - w^\nu) = 4\pi R^2 B_I [\![\mu_W + e_{kin}]\!] \qquad \text{with} \qquad B_I > 0\,, \tag{22}$$

the quantity B_I is called *interfacial mobility* which must be measured or calculated from an underlying model. Such a model will be discussed in Section 2.7.

2.6 Special constitutive model for the two phase system

The vapor phase consists of the three constituents water vapor, hydrogen and oxygen. These are described by the ideal thermal and the caloric equations of state for the partial pressures and for the partial internal energies. These are for $a \in \{W, H, O\}$

$$p_a(T, \rho_a) = \rho_a \frac{kT}{m_a} \qquad \text{and} \qquad u_a(T) = z_a \frac{k}{m_a}(T - T_*) + u_a(T_*) \tag{23}$$

with

$$z_a = \begin{cases} 3/2 & \text{for a} \quad \text{monoatomic,} \\ 5/2 & '' \qquad \text{diatomic,} \\ 3 & '' \qquad \text{polyatomic} \quad \text{gas.} \end{cases}$$

The *Boltzmann constant* is denoted by k, the *molecular mass* of constituent a is m_a. We use the symbol $*$ to denote arbitrary reference values.

The constitutive laws for the partial specific entropies are also needed here

$$s_a(T, \rho_a) = z_a \frac{k}{m_a} \log \frac{T}{T_*} - \frac{k}{m_a} \log \frac{\rho_a}{\rho_a^*} + s_a(T_*, \rho_a^*)\,. \tag{24}$$

Likewise to (11) the *entropy density* ρs for the mixture is given by

$$\rho s = \sum_{a \in \{W,H,O\}} \rho_a s_a\,. \tag{25}$$

We have already assumed that the vapor mixture is inviscid but we allow heat conduction. However, due to a homogeneity assumption in the bubble we do not need the constitutive law for the heat flux $\mathbf{q}$.

The liquid phase consists only of water which is modeled as a compressible inviscid liquid. We use the following constitutive laws for the liquid pressure and energy

$$p^L = p_* + K\left(\frac{\rho}{\rho_*} - 1\right) \tag{26}$$

and

$$u^L(T,\rho) = c(T - T_*) + (p_* - K)\left(\frac{1}{\rho_*} - \frac{1}{\rho}\right) + \frac{K}{\rho_*}\ln\frac{\rho}{\rho_*} + u^L(T_*,\rho_*)\,. \tag{27}$$

The pressure is related to the density variation by a linear law, and additionally we ignore thermal expansion and the temperature dependence of the *modulus of compression* K. Thus the pressure is assumed to be independent of temperature. For the specific heat capacities we have $c_p = c_v = c$. This implies that the specific entropy of the liquid does not depend on the mass density, and we have

$$s^L(T) = c\log\frac{T}{T_*} + s^L(T_*)\,.$$

The constitutive law for the heat flux in the liquid is given by Fourier's law, i.e.

$$\mathbf{q}^L = -\kappa^L \operatorname{grad} T \qquad \text{with} \qquad \kappa^L > 0\,. \tag{28}$$

The liquid *heat conductivity* κ^L is assumed to be constant.

From the above constitutive equations for the vapor we obtain

$$\mu_a^V(T,p,X_a) = g_a(T,p) + \frac{kT}{m_a}\ln X_a\,, \tag{29}$$

where the terms have been combined, s.t. the chemical potential of constituent a can be written as the specific Gibbs free energy for the pure substance a under the total pressure of the mixture p^V plus the so called entropy of mixing. Thus $g_a(T,p)$ is defined by

$$g_a(T,p) = g_a(T_*,p_a^*) + \frac{kT}{m_a}\ln\left(\frac{p}{p_a^*}\right) + (z_a+1)\frac{k}{m_a}\left[T - T_* - T\ln\left(\frac{T}{T_*}\right)\right] - (T-T_*)s_a(T_*)\,. \tag{30}$$

Recall that the liquid consists of the single substance water. Its specific Gibbs free energy can easily be calculated and reads

$$g^L(T,p) = g^L(T_*,p_*) + \frac{K}{\rho_*}\ln\left(1 + \frac{p-p_*}{K}\right) + c\left[T - T_* - T\ln\left(\frac{T}{T_*}\right)\right] - (T-T_*)s^L(T_*)\,. \tag{31}$$

The incompressible liquid is included here as the limiting case $K \to \infty$ that gives

$$g^L(T,p) = g^L(T_*,p_*) + \frac{1}{\rho_*}(p-p_*) + c\left[T - T_* - T\ln\left(\frac{T}{T_*}\right)\right] - (T-T_*)s^L(T_*)\,. \tag{32}$$

2.7 Simple kinetic model for the evolution of a single bubble

In this section we introduce a kinetic model for the case at hand. To this end we consider a bubble with radius $R(t)$ filled with the vapor mixture from above. The bubble is immersed in the liquid, and the evolution $\dot{m}$ of the total bubble mass is given by

$$\frac{dm}{dt} = \frac{d}{dt}\int_{\Omega^V} \rho^V \, dx = -\oint_{\partial\Omega^V} \rho^V (v^\nu - w^\nu)\, dS = -4\pi R^2 \rho^V (v^\nu - w^\nu)\,, \tag{33}$$

where we have used the continuity equation (3) and Reynolds transport theorem [16]. Now we calculate $\dot{m}$ by a simple kinetic model. We start from the representation

$$\dot{m} = m_W(\gamma^E - \gamma^C) \tag{34}$$

where γ^E and γ^C are the *evaporation* respectively the *condensation rate*. The latter results from the encounters of the gas molecules with the interface of the bubble. We assume that each incoming water molecule leads to condensation, and furthermore we assume that the two other constituents remain in the bubble. In this case the kinetic theory of ideal gases gives the condensation rate by the expression

$$\gamma^C(R, T_I, p_W) = 4\pi R^2 \sqrt{\frac{kT_I}{2\pi m_W}}\frac{\rho_W}{m_W} = 4\pi R^2 \frac{p_W}{\sqrt{2\pi m_W k T_I}}\,. \tag{35}$$

The relation (34) is usually called *kinetic relation*, but it is also known as the classical Hertz-Knudsen-theory, see Bond and Struchtrup [3].

Next we calculate the evaporation rate γ^E. At first we calculate γ^E in equilibrium, and according to (21) and (33) we have here $\dot{m} = 0$. As stated in (34) this is equivalent to $\tilde{\gamma}^E = \tilde{\gamma}^C$, where $\tilde{}$ indicates an equilibrium state.

The necessary condition can be read from $(21)_1$ with $e_{kin} = 0$. At the interface it states that $\mu_W^L(T_I, p_I^L, X^L) = \mu_W^V(T_I, p^V, X_W)$. In the liquid we have $X^L = 1$. For the vapor we insert into (29) the relation $X_W = p_W/p^V$, which holds for ideal gases. Thus we obtain from (30) that $g^L(T_I, p_I^L) = g_W(T, p_W)$. Thus for given T_I and p_I^L we may solve this condition for $p_W = \tilde{p}(T_I, p_I^L)$, so that the condensation rate in equilibrium can be written as

$$\tilde{\gamma}^C = \gamma^C(R, T_I, \tilde{p}(T_I, p_I^L)) = 4\pi R^2 \frac{\tilde{p}(T_I, p_I^L)}{\sqrt{2\pi m_W k T_I}}\,. \tag{36}$$

Next we introduce a trick to represent the function $p_W = \tilde{p}(T_I, p_I^L)$ by a form that does not explicitly refer to equilibrium. To this end we introduce the saturation pressure $\bar{p}$, which denotes the equilibrium pressure at a planar interface between the vapor and the liquid phase of pure water. In such a situation the equation $g^L(T_I, p) = g_W(T_I, p)$ holds for the equilibrium pressure $\bar{p}$. Consequently for a given T_I we have $p = \bar{p}(T_I)$.

Using (30) we calculate $g_W(T_I, \tilde{p})$ and $g_W(T_I, \bar{p})$ to obtain the difference

$$g_W(T_I, \tilde{p}) - g_W(T_I, \bar{p}) = \frac{kT}{m_W}\ln\frac{\tilde{p}}{\bar{p}}\,. \tag{37}$$

From $g^L(T_I, p_I^L) = g_W(T_I, \tilde{p})$ we conclude

$$g^L(T_I, p_I^L) = g_W(T_I, \bar{p}(T_I)) + \frac{kT}{m_W}\ln\frac{\tilde{p}}{\bar{p}}\,, \tag{38}$$

thus we get

$$\tilde{p} = \bar{p} \exp\left(m_W \frac{g^L(T_I, p_I^L) - g_W(T_I, \bar{p})}{kT} \right) . \tag{39}$$

In equilibrium we have $\tilde{\gamma}^E = \tilde{\gamma}^C$, so using (39) the evaporation rate in equilibrium can be calculated according to

$$\begin{aligned} \tilde{\gamma}^E = \tilde{\gamma}^C &= 4\pi R^2 \frac{\tilde{p}}{\sqrt{2\pi m_W k T_I}} \\ &= 4\pi R^2 \frac{\bar{p}(T_I)}{\sqrt{2\pi m_W k T_I}} \exp\left(m_W \frac{g^L(T_I, p_I^L) - g_W(T_I, \bar{p}(T_I))}{kT_I} \right) . \end{aligned} \tag{40}$$

Analogously to (37) we obtain from (30)

$$g_W(T_I, \bar{p}) = g_W(T_I, p_W) - \frac{kT}{m_W} \ln \frac{p_W}{\bar{p}} . \tag{41}$$

Using this expression the equilibrium evaporation rate can be given in the following form

$$\tilde{\gamma}^E = 4\pi R^2 \frac{p_W}{\sqrt{2\pi m_W k T_I}} \exp\left(m_W \frac{g^L(T_I, p_I^L) - g_W(T_I, p_W)}{kT_I} \right) . \tag{42}$$

Finally, due to $g^L(T_I, p_L) = \mu^L(T_I, p_L, 1)$ and $g_W^V(T, p_W^V) = \mu_W^V(T, p_V, X_W)$ using (35) we obtain

$$\tilde{\gamma}^E = \gamma^C \exp\left(\frac{m_W}{kT_I} [\![\mu_W]\!] \right) . \tag{43}$$

It is important to note that the equilibrium evaporation rate is now given in terms of non-equilibrium quantities. This fact motivates to assume that the condensation rate γ^E in non-equilibrium is given by the same expression as $\tilde{\gamma}^E$. However, the presented motivation has ignored the contribution of the kinetic energy to the chemical potential. A more careful study leads to

$$\gamma^E = \gamma^C \exp\left(\frac{m_W}{kT_I} [\![\mu_W + e_{kin}]\!] \right) . \tag{44}$$

For small deviations from equilibrium the expression

$$\gamma^E = \gamma^C \left(1 + \frac{m_W}{kT_I} [\![\mu_W + e_{kin}]\!] \right) \tag{45}$$

gives a good approximation. A comparison with the phenomenological ansatz (22) identifies the mobility B_I as

$$B_I = m_W \frac{p_W}{\sqrt{2\pi m_W k T_I}} \frac{m_W}{kT_I} . \tag{46}$$

As the final result we use

$$\dot{m} = 4\pi R^2 m_W \frac{p_W}{\sqrt{2\pi m_W k T_I}} \frac{m_W}{kT_I} [\![\mu_W + e_{kin}]\!] . \tag{47}$$

Here we would like to make two remarks. (i) If the liquid were incompressible, a case that we do not consider here, the relation (44) can be derived without the assumption from above. (ii) If we were to ignore the kinetic energy the same result can also be obtained by the *principle of detailed balance*. However, that principle must be handled with care. An important counterexample is given by Dreyer and Duderstadt in [5].

3 The Göttingen laser induced bubble experiment

Our objective is to apply the developed model to a single spherical bubble that is produced by laser pulses. The bubble is created in a cuvette filled with clean distilled water. The temperature and the pressure far from the bubble are kept constant at $20°C$ and 1 bar. Everything that is seriously known from the experiment can be read of from the Figure 1 that gives the bubble radius versus the time.

Figure 1:
Evolution of bubble radius, experimental data from [15].

Apparently there is a growing bubble that reaches its maximum radius $R_{\max} = 7.469 \cdot 10^{-3}\, m$ at 70.7 μs and collapses hereafter to a minimal radius $R_{\min} = 12.467\, \mu m$ which is assumed at $140\, \mu s$. Hereafter the bubble starts to grow again, however, the new maximal radius is very much smaller than the first one, so that one may conclude that a large damping mechanism accompanies the observed process. The experimental researchers in Göttingen report that the second cycle is followed by further cycles that almost show no damping. It is also reported that presumably there is plasma in the bubble, which is created by the laser, which has completely recombined at time $t = 0$. Further observations and in particular data, that give information on the thermodynamic state of bubble and liquid, are not available.

Obviously, on this slim data basis a serious simulation of the described process is not possible without further speculations. We do not describe here speculations that are given in [15]. Instead from now on we present our point of view to formulate at least the necessary initial conditions. For simplification we start the simulation at the first maximum radius so that we need to know the thermodynamic state of the bubble-liquid system at that instant.

According to our point of view the main assumption to reach coincidence between measured and simulated data concern the permanent existence of an inert gas in the bubble. We believe that oxygen and hydrogen molecules have survived the dramatic period that

followed the bubble creation by the laser, and we properly adjust their amount at the first maximal radius of the bubble, so that the first subsequent minimal radius is met. Thus the initial data for our simulation are given by $T = 293.15\,K,\ N_W = 7.5 \cdot 10^{15},\ N_O = 2.5 \cdot 10^{13}$ implying $N_H = 2N_O,\ p_V(t=0) = 17350\ Pa$.

The temperature is assumed to be homogeneous in the bubble-liquid system at $t = 0$ and is equal to the liquid temperature far away from the bubble. Initially we take $R(0) = R_{\max}$ and $v^L = 0$.

We already mention here that agreement with the experimental data can only be achieved if the bubble-liquid system enters a single phase state for a finite period of the evolution. This happens if the interfacial temperature T_I exceeds the critical temperature. In the above model the single phase region is characterized by $\dot{m} = 0$.

4 Special cases

4.1 Introduction of special cases

The evolution of the bubble radius is accompanied by three damping mechanisms which are due to: phase transition, heat conduction and generation of waves in the liquid. In order to see their influence on the damping of the bubble radius we separately consider the three phenomena.

In this section we give only the main results for the bubble dynamics. The detailed derivation of the formulas is given in Section 5.

4.2 Spherical symmetry and homogeneous bubble

We consider exclusively a bubble-liquid system with spherical symmetry which implies that the conservation laws assume in the bubble as well as in the liquid the following form

$$\frac{\partial \rho}{\partial t} + \frac{1}{r^2}\frac{\partial (r^2 \rho v)}{\partial r} = 0 \tag{48}$$

$$\frac{\partial \rho v}{\partial t} + \frac{1}{r^2}\frac{\partial (r^2 \rho v^2)}{\partial r} + \frac{\partial p}{\partial r} = 0 \tag{49}$$

$$\frac{\partial \rho u}{\partial t} + \frac{1}{r^2}\frac{\partial (r^2 \rho u v)}{\partial r} + \frac{1}{r^2}\frac{\partial (r^2 q)}{\partial r} = -\frac{p}{r^2}\frac{\partial (r^2 v)}{\partial r}\,. \tag{50}$$

The corresponding constitutive equations will be separately inserted for the various special cases.

The interfacial conservation laws (10), (8) and (9) in spherical coordinates are with $w^\nu = \dot{R}$

$$v_I^H(t) = v_I^O(t) = \dot{R}(t) \qquad \text{and} \qquad [\![\rho_W (v_W - \dot{R})]\!] = 0, \tag{51}$$

$$\rho(v_I - \dot{R})[\![v]\!] + [\![p]\!] = -\frac{2\sigma}{R}\,, \tag{52}$$

$$-\rho(v - \dot{R})[\![u + \frac{p}{\rho} + \frac{1}{2}(v - \dot{R})^2]\!] + [\![q]\!] = 0\,. \tag{53}$$

The total mass of the spherical bubble B is given by $m(t) = 4\pi \int_0^{R(t)} r^2 \rho^V(t,r)dr$. From now on we exclusively consider a homogeneous bubble, thus we have

$$m(t) = \frac{4}{3}\pi R(t)^3 \rho^V(t) \qquad \text{implying} \qquad \dot{m} = 4\pi R^2 \dot{R}\rho^V + \frac{4}{3}\pi R^3 \dot{\rho}^V \,. \tag{54}$$

We eliminate $\dot{m}$ by means of the mass balance (33) and obtain

$$\dot{\rho}^V = -\frac{3\rho^V v_I^V}{R}\,, \tag{55}$$

so that $(54)_2$ can be written as

$$\dot{m} = 4\pi R^2 \rho^V (\dot{R} - v_I^V) = 4\pi R^2 \rho_I^L (\dot{R} - v_I^L)\,. \tag{56}$$

Now $(56)_2$ is used to calculate the liquid velocity at the interface

$$v_I^L = \dot{R} - \frac{\dot{m}}{4\pi R^2 \rho_I^L}\,. \tag{57}$$

The assumption of a homogeneous gas phase means, that the processes of diffusion and heat conduction are much faster than the adjustment to mechanical equilibrium. In the homogeneous case from $(23)_1$ and $(25)_1$ we obtain

$$p_I^V(t) = \frac{3N(t)kT}{4\pi R(t)^3}\,, \tag{58}$$

where the quantity $N(t) = N_W(t) + N_H + N_O$ gives the total number of particles in the bubble. With

$$m(t) = N_W(t)m_W + N_H m_H + N_O m_O \tag{59}$$

we have

$$\dot{m} = m_W \dot{N}\,. \tag{60}$$

4.3 Case 1: The undamped oscillation of a bubble immersed in an incompressible liquid at constant temperature without phase transition

In this case the evolution law for the bubble radius can be reduced to a single ODE that reads

$$\ddot{R} + \frac{3\dot{R}^2}{2R} + \frac{1}{\rho^L R}\left(p_0 - \frac{3NkT}{4\pi R^3} + \frac{2\sigma}{R}\right) = 0\,, \tag{61}$$

which is known as the Rayleigh-Plesset equation. The total number of particles in the bubble N is constant, because in this case the number of water molecules in the bubble does not change. The equation describes an undamped periodic oscillation around the equilibrium radius R_{eq} with $p_0 + \frac{2\sigma}{R_{eq}} = \frac{3NkT}{4\pi R_{eq}^3}$. With the initial conditions $R(0) = R_0 > 0$ and $\dot{R}(0) = \dot{R}_0$ integration of (61) leads to two solutions for the interfacial velocity

$$\dot{R} = \pm\frac{1}{R^{3/2}}\sqrt{\dot{R}_0^2 R_0^3 - \frac{1}{\rho^L}\left(\frac{2}{3}p_0(R^3 - R_0^3) - \frac{3NkT}{2\pi}\ln\frac{R}{R_0} + 2\sigma(R^2 - R_0^2)\right)}\,, \tag{62}$$

which describe a closed curve, see Subsection 5.1 and Figure in Section 6.

4.4 Case 2: The oscillation of a bubble immersed in an incompressible liquid at constant temperature with damping due to phase transition

Next we take into account phase transition, that means $\dot{N} \neq 0$. This case can be described by three ordinary first order differential equations

$$\dot{N} = \frac{4\pi R^2 p_W}{\sqrt{2\pi m_W kT}} \frac{m_W}{kT} \left[\frac{1}{\rho^L}(p_I^L - \bar{p}) - \frac{kT}{m_W} \ln \frac{p_W}{\bar{p}} + \left(\frac{1}{(\rho^L)^2} - \frac{1}{(\rho^V)^2} \right) \left(\frac{m_W \dot{N}}{4\pi R^2} \right)^2 \right] \quad (63)$$

$$\dot{R} = \frac{F}{R^2} + \frac{m_W \dot{N}}{4\pi R^2 \rho^L} \quad (64)$$

$$\dot{F} = \frac{F^2}{2R^3} - \frac{R}{\rho^L}(p_0 - p_I^L)\,, \quad (65)$$

where the explicit representations of p^W, p_I^L, ρ^V are given in Subsection 5.2. The resulting system describes a damped oscillation.

The system is implicit with respect to $\dot{N}$. We are interested in solutions that lie in the domain of positive R and N_W, i.e. $(R, N_W, F) \in]0,\infty[\times]0,\infty[\times \mathbb{R}$. The system is solvable for the initial conditions $R(0) = R_0$, $\dot{R}(0) = \dot{R}_0$ and $N(0) = N_0$.

Case 1 is included here by replacing equation (63) with $\dot{N} = 0$.

4.5 Case 3: The oscillation of a bubble immersed in an incompressible liquid with damping due to phase transition and heat conduction

In contrast to the previous cases the temperature field is unknown and controlled by heat conduction. Therefore we now have to consider the energy balance equations.

The resulting ODE system is almost the same as before but coupled to the energy balance equation in the liquid and to the corresponding interfacial boundary condition

$$\begin{aligned} \dot{N} = \frac{4\pi R^2 p_W}{\sqrt{2\pi m_W kT_I}} \frac{m_W}{kT_I} \Bigg[& \frac{1}{\rho^L(T_0)}(p_I^L - \bar{p}(T_0)) - \frac{kT_I}{m_W} \ln \frac{p_W}{\bar{p}(T_0)} \\ & + \left(\frac{1}{(\rho^L(T_0))^2} - \frac{1}{(\rho^V)^2} \right) \left(\frac{m_W \dot{N}}{4\pi R^2} \right)^2 \\ & + (c^L - c_W)(T_I - T_0 - T_I \ln \frac{T_I}{T_0}) - (s^L - s_W)(T_I - T_0) \Bigg] \end{aligned} \quad (66)$$

$$\dot{R} = \frac{F}{R^2} + \frac{m_W \dot{N}}{4\pi R^2 \rho^L(T_0)} \quad (67)$$

$$\dot{F} = \frac{F^2}{2R^3} - \frac{R}{\rho^L(T_0)}(p_0 - p_I^L) \quad (68)$$

$$\left. \frac{\partial T}{\partial r} \right|_{r=R} = \frac{k\dot{T}}{4\pi R^2 \kappa^L} \sum_{a \in \{W,H,O\}} N_a z_a + \frac{\dot{R}p}{\kappa^L} - \frac{kT\dot{N}}{4\pi R^2 \kappa^L} + \frac{\lambda m_W \dot{N}}{4\pi R^2 \kappa^L} \quad (69)$$

$$\frac{\partial T}{\partial t} = a^L \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r} \right) - \frac{F}{r^2}\frac{\partial T}{\partial r}\,. \quad (70)$$

In addition to $R(0) = R_0$, $\dot{R}(0) = \dot{R}_0$ and $N(0) = N_0$ we need for the temperature the

initial condition $T^L(0,r) = T_0^L(r)$ and at the outer boundary $r = R_a$ we choose $\frac{\partial T}{\partial r} = 0$ for R_a sufficiently large.

4.6 Case 4: The oscillation of a bubble immersed in a weakly compressible liquid with damping due to phase transition, heat conduction and acoustic waves

In the previous cases the liquid was assumed to be incompressible. This assumption is too restrictive for the experiment at hand. Thus now we take weak compressibility of the liquid into account. The necessary modifications lead to the following system

$$\begin{aligned}
\dot{N} &= \frac{4\pi R^2 p_W}{\sqrt{2\pi m_W k T_I}} \frac{m_W}{kT_I} \left[\frac{K}{\rho^L(T_0)} \ln\left(1 + \frac{p_I^L - \bar{p}(T_0)}{K}\right) - \frac{kT_I}{m_W} \ln \frac{p_W}{\bar{p}(T_0)} \right. \\
&\quad + \left(\frac{1}{(\rho^L(T_0))^2} - \frac{1}{(\rho^V)^2} \right) \left(\frac{m_W \dot{N}}{4\pi R^2} \right)^2 \\
&\quad \left. + (c^L - c_W)(T_I - T_0 - T_I \ln \frac{T_I}{T_0}) - (s^L - s_W)(T_I - T_0) \right] \qquad (71) \\
\dot{R} &= \frac{\phi(2R_a - R - c_0 t) - \phi(R - c_0 t)}{R^2} + \frac{\phi'(2R_a - R - c_0 t) + \phi'(R - c_0 t)}{R} \\
&\quad + \frac{m_W \dot{N}}{4\pi R^2 \rho_L(T_0)} \qquad (72) \\
\frac{p_I^L - p_0}{\rho^L(T_0) c_0} R &= \phi'(R - c_0 t) - \phi'(2R_a - R - c_0 t) \qquad (73) \\
\left. \frac{\partial T}{\partial r} \right|_{r=R} &= \frac{k\dot{T}}{4\pi R^2 \kappa^L} \sum_{a \in \{W,H,O\}} N_a z_a + \frac{\dot{R} p}{\kappa^L} - \frac{kT\dot{N}}{4\pi R^2 \kappa^L} + \frac{\lambda m_W \dot{N}}{4\pi R^2 \kappa^L} \qquad (74) \\
\frac{\partial T}{\partial t} &= a^L \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right) - v^L \frac{\partial T}{\partial r} \,. \qquad (75)
\end{aligned}$$

The newly introduced function ϕ describes in- and outgoing waves.

5 Detailed derivations

5.1 Case 1: A bubble immersed in an incompressible liquid at constant temperature without phase transition

In the isothermal case the temperature T is given and considered to be constant. Therefore we do not need the energy balance equation, we merely have to solve the mass and momentum balance equations. The liquid is assumed to be incompressible, i.e. we require that the liquid mass density ρ^L does not depend on pressure. Then mass conservation of the liquid (48) simplifies to

$$\frac{\partial (r^2 v^L)}{\partial r} = 0 \qquad (76)$$

and leads to the liquid velocity

$$v^L(t,r) = \frac{F(t)}{r^2} \qquad (77)$$

with the time dependent function F. For an incompressible liquid the momentum balance equation (49) reduces to

$$\frac{\partial v^L}{\partial t} + \frac{\partial}{\partial r}\left(\frac{(v^L)^2}{2}\right) = -\frac{1}{\rho^L}\frac{\partial p^L}{\partial r}\,. \tag{78}$$

Using (77) we replace v^L in the time derivative and obtain

$$\frac{\dot F}{r^2} + \frac{\partial}{\partial r}\left(\frac{(v^L)^2}{2}\right) = -\frac{1}{\rho^L}\frac{\partial p^L}{\partial r}\,.$$

We integrate the equation over $[R(t),\infty[$ assuming the velocity v^L to vanish at infinity and here the pressure p becomes the outer pressure. The result is

$$\dot F = \frac{F^2}{2R^3} - \frac{R}{\rho^L}(p_0 - p_I^L)\,. \tag{79}$$

Without phase transition the mass balance $(51)_2$ at the interface I simplifies to

$$v_I^L(t) = v_I^V(t) = \dot R(t)\,. \tag{80}$$

This implies

$$v_I^L(t) = v^L(t,R(t)) = \frac{F(t)}{R(t)^2} = \dot R(t) \qquad \text{i.e.} \qquad F = \dot R R^2\,. \tag{81}$$

The velocity field in the liquid domain is given by

$$v^L(t,r) = \dot R(t)\left(\frac{R(t)}{r}\right)^2\,. \tag{82}$$

Using (81) in (79) we end up with

$$\ddot R R + \frac{3}{2}\dot R^2 = -\frac{1}{\rho^L}(p_0 - p_I^L)\,. \tag{83}$$

The momentum balance (52) at the interface can be written as

$$p_I^L(t) = p_I^V(t) - \frac{2\sigma}{R(t)} = \frac{3NkT}{4\pi R(t)^3} - \frac{2\sigma}{R(t)}\,. \tag{84}$$

We replace p_I^L in (83) and obtain the oscillation equation

$$\ddot R R^3 + \frac{3R^2\dot R^2}{2} + \frac{1}{\rho^L}\left(p_0 R^2 - \frac{3NkT}{4\pi R} + 2\sigma R\right) = 0\,. \tag{85}$$

This ordinary second order differential equation describes an oscillation around the stationary radius R_{stat}, that solves the equation $p_0R^3 + 2\sigma R^2 = \frac{3NkT}{4\pi}$. The left hand side of this equation is monotone increasing in R and it is zero for $R = 0$. The right hand side of the equation is positive. Thus there is a only one real positive solution R_{stat}.

Obviously we have

$$\left(\ddot R R^3 + \frac{3R^2\dot R^2}{2}\right)\dot R = \frac{1}{2}(\dot R^2 R^3)^{\cdot}\,, \tag{86}$$

and by integration of (85) we derive

$$\frac{1}{2}(\dot{R}^2R^3 - \dot{R}_0^2R_0^3) + \frac{1}{\rho^L}\left(\frac{1}{3}p_0(R^3 - R_0^3) - \frac{3NkT}{4\pi}\ln\frac{R}{R_0} + \sigma(R^2 - R_0^2)\right) = 0\,, \tag{87}$$

where the integration constant is determined by the initial condition. This is a quadratic equation in $\dot{R}$ with two solutions (63), which only depend on R and the initial conditions R_0, $\dot{R}_0$. Together these solutions describe a closed curve. We conclude, that (85) describes an undamped oscillation.

5.2 Case 2: A bubble immersed in an incompressible liquid at constant temperature with phase transition

In the case of phase transition we have to make some modifications in the interfacial mass and momentum balance. We already derived the formula $v_I^L = F/R^2$ in Subsection 5.1. Using the mass balance equation (57) at the interface and $\dot{m} = m_W\dot{N}$ we obtain

$$F = R^2\dot{R} - \frac{m_W\dot{N}}{4\pi\rho^L}\,. \tag{88}$$

The momentum balance equation (52) at the interface gives

$$p_I^L = \frac{3NkT}{4\pi R^3} - \frac{2\sigma}{R} - \left(\frac{1}{\rho^L} - \frac{1}{\rho^V}\right)\left(\frac{m_W\dot{N}}{4\pi R^2}\right)^2. \tag{89}$$

For convenience we define the number of particles of the inert gas $N_i := N_H + N_O$ and the corresponding mass $m_i := N_H \cdot m_H + N_O \cdot m_O$. This notation allows to apply all the formulas to any arbitrary inert gas. Then the density of the gas phase assumes the following form

$$\rho^V = \frac{3(m_W \cdot (N - N_i) + m_i)}{4\pi R^3}\,. \tag{90}$$

We replace p_I^L in (79) and obtain

$$\dot{F} = \frac{F^2}{2R^3} - \frac{R}{\rho^L}\left(p_0 - \frac{3NkT}{4\pi R^3} + \frac{2\sigma}{R} + \left(\frac{1}{\rho^L} - \frac{4\pi R^3}{3(m_W \cdot (N - N_i) + m_i)}\right)\left(\frac{m_W\dot{N}}{4\pi R^2}\right)^2\right). \tag{91}$$

The evolution of the water particles in the bubble is described by (47). Here we replace $\dot{m} = m_W\dot{N}$, choose $T^* = T$ and calculate $[\![\mu_W + e_{kin}]\!]$ using (29), (30), (32) and (51). The partial pressure p_W is given by

$$p_W = \frac{3(N - N_i)kT}{4\pi R^3}\,.$$

Finally we have to replace the liquid pressure at the interface p_I^L and the vapor density ρ^V as in the previous subsection, using (89) and (90). We thus obtain

$$\begin{aligned}\dot{N} = {} & \frac{3(N - N_i)m_W}{R\sqrt{2\pi m_W kT}} \\ & \left(\frac{1}{\rho^L}\left(\frac{3NkT}{4\pi R^3} - \frac{2\sigma}{R} - \left(\frac{1}{\rho^L} - \frac{4\pi R^3}{3(m_W \cdot (N - N_i) + m_i)}\right)\left(\frac{m_W\dot{N}}{4\pi R^2}\right)^2 - \bar{p}\right)\right. \\ & \left. - \frac{kT}{m_W}\ln\left(\frac{3(N - N_i)kT}{4\pi R^3\bar{p}}\right) + \frac{1}{2}\left(\left(\frac{1}{\rho^L}\right)^2 - \left(\frac{4\pi R^3}{3(m_W \cdot (N - N_i) + m_i)}\right)^2\right)\left(\frac{m_W\dot{N}}{4\pi R^2}\right)^2\right),\end{aligned}$$

which can be simplified to

$$\dot{N} = \frac{3(N-N_i)m_W}{R\sqrt{2\pi m_W kT}} \left(\frac{1}{\rho^L} \left(\frac{3NkT}{4\pi R^3} - \frac{2\sigma}{R} - \bar{p} \right) - \frac{kT}{m_W} \ln \left(\frac{3(N-N_i)kT}{4\pi R^3 \bar{p}} \right) - \frac{1}{2} \left(\frac{1}{\rho^L} - \frac{4\pi R^3}{3(m_W \cdot (N-N_i) + m_i)} \right)^2 \left(\frac{m_W \dot{N}}{4\pi R^2} \right)^2 \right) . \quad (92)$$

This equation together with (88) and (91) gives the resulting system in the considered case.

5.3 Case 3: A bubble immersed in an incompressible liquid with phase transition and heat conduction

Now we take heat conduction into account, so that the temperature field is determined by the energy balance equations of the phases and at the interface.

The energy balance of the phases can be written in the general form

$$\rho \left(\frac{\partial u}{\partial t} + \mathbf{v} \cdot \operatorname{grad} u \right) + \operatorname{div} \mathbf{q} = \frac{p}{\rho} \left(\frac{\partial \rho}{\partial t} + \mathbf{v} \operatorname{grad} \rho \right) . \quad (93)$$

In the liquid phase this can be reduced, with temperature independent compressibility K and using (14), $(15)_1$ as well as $(15)_2$, to

$$\rho^L c^L \left(\frac{\partial T^L}{\partial t} + \mathbf{v}^L \operatorname{grad} T^L \right) + \operatorname{div} \mathbf{q}^L = 0 ,$$

which holds for the incompressible as for the compressible case. Here $\mathbf{q}^L$ is determined by Fourier's law (28) and with the thermal conductivity $a^L = \kappa^L/(\rho^L c^L)$ the heat conduction equation reads

$$\frac{\partial T^L}{\partial t} + v^L \frac{\partial T^L}{\partial r} = a^L \left(\frac{\partial^2 T^L}{\partial r^2} + \frac{2}{r} \frac{\partial T^L}{\partial r} \right) . \quad (94)$$

In the gas phase we also start from (93) and replace the internal energy respectively the pressure by the constitutive laws (23) and (25) and after some rearrangements of terms we obtain

$$\sum_{a \in \{W,H,O\}} \rho_a \left(\frac{\partial u_a}{\partial t} + \mathbf{v}_a \operatorname{grad} u_a \right) + \operatorname{div} \sum_{a \in \{W,H,O\}} (\mathbf{q}_a + (\mathbf{v}_a - \mathbf{v}) p_a) = \frac{p}{\rho} \left(\frac{\partial \rho}{\partial t} + \mathbf{v} \cdot \operatorname{grad} \rho \right) . \quad (95)$$

We use $(23)_2$ to substitute $\frac{\partial u_a}{\partial t} + \mathbf{v}_a \cdot \operatorname{grad} u_a$, integrate (95) and get

$$\sum_{a \in \{W,H,O\}} q_a^I = -\frac{k\dot{T}}{4\pi R^2} \sum_{a \in \{W,H,O\}} N_a z_a - \dot{R} p + \frac{kT\dot{N}}{4\pi R^2} . \quad (96)$$

From the balance of internal energy at the interface we obtain

$$q_I^L - \sum_{a \in \{W,H,O\}} q_a^I = -\frac{\lambda m_W \dot{N}}{4\pi R^2} . \quad (97)$$

We use Fouriers law (28) on the liquid side of the interface to get

$$\frac{\partial T}{\partial r} = \frac{k\dot{T}}{4\pi R^2 \kappa} \sum_{a\in\{W,H,O\}} N_a z_a + \frac{\dot{R}p}{\kappa} - \frac{kT\dot{N}}{4\pi R^2\kappa} + \frac{\lambda m_W \dot{N}}{4\pi R^2 \kappa}\,. \tag{98}$$

The equations (88) and (91) remain the same as before, when we ignored heat conduction. In the equation (92), which describes the mass transfer across the interface, the temperature dependent function $\mu(T) = (c^L - c_{p_W})(T - T_0 - T\ln\frac{T}{T_0}) - (s^L - s_W)(T - T_0)$ does not drop out any longer here, and we set therein the reference temperature $T^* = T_0$. This leads to

$$\begin{aligned}\dot{N} \;=\; & \frac{3(N-N_f)m_W}{R\sqrt{2\pi m_W kT}}\left(\frac{1}{\rho^L}\left(\frac{3NkT}{4\pi R^3} - \frac{2\sigma}{R} - \bar{p}\right) - \frac{kT}{m_W}\ln\left(\frac{3(N-N_f)kT}{4\pi R^3\bar{p}}\right)\right. \\ & -\frac{1}{2}\left(\frac{1}{\rho^L} - \frac{4\pi R^3}{3(m_W\cdot(N-N_f)+m_f)}\right)^2\left(\frac{m_W\dot{N}}{4\pi R^2}\right)^2 \\ & \left.+(c^L - c_{p_W})(T - T_0 - T\ln\frac{T}{T_0}) - (s^L - s_W)(T-T_0)\right)\,.\end{aligned} \tag{99}$$

Finally we couple the system (88), (91), (99) with the energy balance equation (94) and (98). In (94) we substitute $v^L(t,r)$ by (77) and get

$$\frac{\partial T^L}{\partial t} + \frac{F}{r^2}\frac{\partial T^L}{\partial r} = a^L\left(\frac{\partial^2 T^L}{\partial r^2} + \frac{2}{r}\frac{\partial T^L}{\partial r}\right)\,. \tag{100}$$

5.4 Case 4: A bubble immersed in a weakly compressible liquid with phase transition and heat conduction

The model from the previous subsection leads to a damping of the bubble oscillations that is not sufficiently strong to describe the experimental data. A further damping effect is related to the propagation of waves in the liquid, which are induced by the motion of the bubble. In this case we must skip the assumption of an incompressible liquid. For this reason we now consider a weak compressible liquid and in order to avoid extreme difficulties we linearize the mass conservation law and the momentum balance, which then read

$$\frac{\partial \rho}{\partial t} + \rho_0\frac{\partial v}{\partial r} = 0 \tag{101}$$

$$\rho_0\frac{\partial v}{\partial t} + \frac{\partial p}{\partial r} = 0\,. \tag{102}$$

With the definition $v := \frac{\partial f}{\partial r}$ and $\frac{\partial p}{\partial \rho} = c_0^2$ we can rewrite these equations in the following form

$$\frac{1}{c_0^2}\frac{\partial p}{\partial t} + \rho_0\Delta f = 0 \tag{103}$$

$$\frac{\partial}{\partial r}\left(\rho_0\frac{\partial f}{\partial t} + p\right) = 0\,. \tag{104}$$

Integration of (104) leads to

$$p - p_0 = -\rho_0\frac{\partial f}{\partial t}\,. \tag{105}$$

Thus for the potential f we obtain the wave equation

$$\frac{1}{c_0^2}\frac{\partial^2 f}{\partial t^2} = \Delta f\,. \tag{106}$$

For radial symmetry its general solution is known to be

$$f(t,r) = \frac{\phi(r-c_0t)}{r} + \frac{\varphi(r+c_0t)}{r}\,, \tag{107}$$

where ϕ and φ describe outgoing respectively ingoing waves. We consider $p = p_0$ at the outer boundary $r = R_a$. This leads to the reflection condition $\varphi(r+c_0t) = -\phi(r+c_0t-2(R_a-r)) = -\phi(2R_a-r-c_0t)$. We calculate

$$p(t,r) = p_0 + \rho_0 c_0 \frac{\phi'(r-c_0t)}{r} - \rho_0 c_0 \frac{\phi'(2R_a-r-c_0t)}{r} \qquad \text{and} \tag{108}$$

$$v(t,r) = -\frac{\phi(r-c_0t)}{r^2} + \frac{\phi'(r-c_0t)}{r} + \frac{\phi(2R_a-r-c_0t)}{r^2} + \frac{\phi'(2R_a-r-c_0t)}{r}\,. \tag{109}$$

Using the balance of mass at the interface we end up with

$$\frac{\phi(2R_a-R-c_0t)-\phi(R-c_0t)}{R^2} + \frac{\phi'(2R_a-R-c_0t)+\phi'(R-c_0t)}{R} = \dot R - \frac{m_W\dot N}{4\pi R^2\rho_0} \tag{110}$$

$$\text{and} \qquad p_I^L = p_0 + \rho_0 c_0 \frac{\phi'(R-c_0t)-\phi'(2R_a-R-c_0t)}{R}\,. \tag{111}$$

Unlike (88) and (91) we now get two differential equations including the unknown function ϕ which depends on time explicitly and on the unknown time dependent bubble radius R.

The consideration of weak compressibility leads to a small modification in the derivation of the equation to describe the mass transfer. In contrast to the previous cases in Subsection 5.2 and 5.3 we have to use (31) instead of (32) to calculate $[\![\mu_W + e_{kin}]\!]$. Equation (99) modifies to

$$\begin{aligned}\dot N = \frac{3(N-N_f)m_W}{R\sqrt{2\pi m_W kT}}\Bigg(&\frac{K}{\rho^L}\ln\Bigg(1+\Bigg(\frac{3NkT}{4\pi R^3}-\frac{2\sigma}{R}-\Big(\frac{1}{\rho^L}-\frac{1}{\rho^V}\Big)\Big(\frac{m_W\dot N}{4\pi R^2}\Big)^2-\bar p\Bigg)/K\Bigg)\\ &-\frac{kT}{m_W}\ln\Bigg(\frac{3(N-N_f)kT}{4\pi R^3\bar p}\Bigg)-\frac{1}{2}\Bigg(\frac{1}{\rho^L}-\frac{4\pi R^3}{3(m_W\cdot(N-N_f)+m_f)}\Bigg)^2\Big(\frac{m_W\dot N}{4\pi R^2}\Big)^2\\ &+(c^L-cp_W)(T-T_0-T\ln\frac{T}{T_0})-(s^L-s_W)(T-T_0)\Bigg)\,.\end{aligned} \tag{112}$$

The now more complicated resulting system consists of the ordinary differential equation (112), the delay equations (110) and (111) which are coupled to the partial differential equation

$$\frac{\partial T^L}{\partial t} + v^L\frac{\partial T^L}{\partial r} = a^L\left(\frac{\partial^2 T^L}{\partial r^2} + \frac{2}{r}\frac{\partial T^L}{\partial r}\right)\,. \tag{113}$$

and to the corresponding interfacial boundary condition (98). The liquid velocity v^L in (113) can be calculated with (109).

6 Numerical results

In this section we will discuss the four described cases on the basis of the numerical computations for the corresponding systems of equations.

For all computations we choose the external pressure $p_0 = 101300Pa$, initial radius $R_0 = 0.00075m$, initial interface velocity $\dot{R} = 0$, initial particle numbers $N_O = 2.5 \cdot 10^{13}$, $N_H = 5 \cdot 10^{13}$, $N_W = 7.5 \cdot 10^{15}$, and initial temperature $T = 293.15K$.

The cases 1 and 2 are concerned with isothermal evolution, i.e. the temperature does not change. However the given initial value determines the reference values for those quantities that are listed in the table below. The composition of the gas in the bubble plays no role in the dynamics of the first case. The sum of the three particle numbers is used in order to determine the initial pressure.

Case 1: A bubble immersed in an incompressible liquid at constant temperature without phase transition.

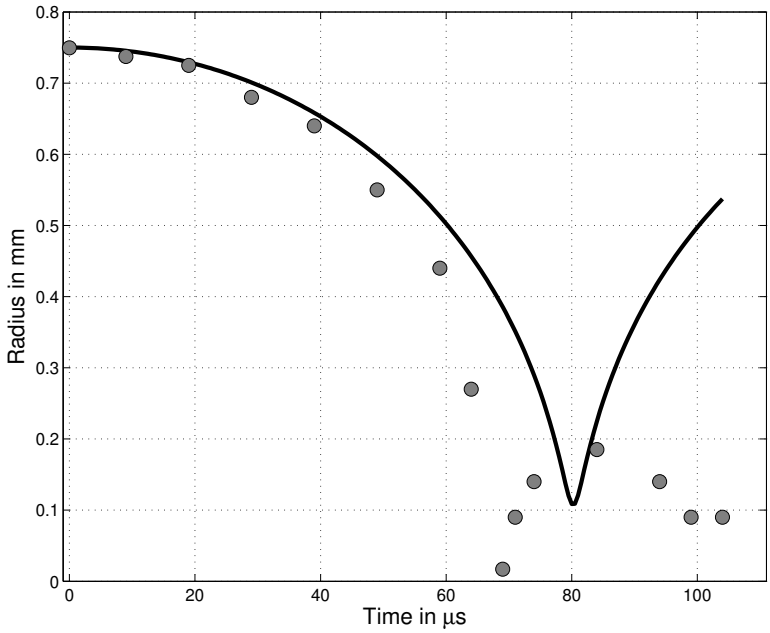

Figure 2:
Calculated bubble radius according to Case 1. Dots: Experimental data, solid line: Computation.

The numerical solution of the second order ordinary differential equation is obtained by solving the corresponding first order system with one of the second order Runge Kutta methods.

As was shown above, the bubble radius oscillates without damping in this case. This result is compared in Figure 2 with experimental data. We observe that computational oscillation period is longer than the experimental one.

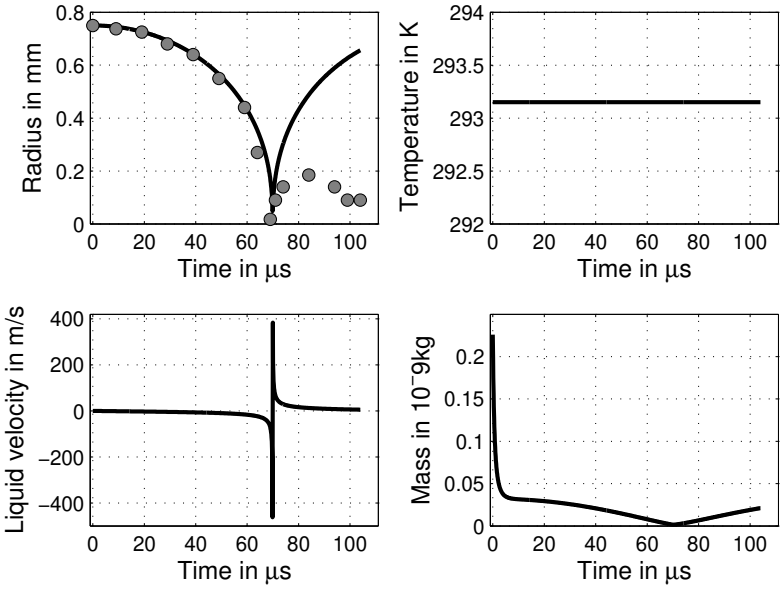

Figure 3:
Calculated bubble radius, bubble mass, liquid velocity and temperature at the interface according to Case 2. Dots: Experimental data, solid line: Computation.

Case 2: A bubble immersed in an incompressible liquid at constant temperature with phase transition. Again a second order Runge Kutta method is used for the system of three ordinary first order equations, including a standard step size control. Due to stability restrictions on the numerical scheme this is needed in the vicinity of the minimal radius.

The incorporation of the phase transition leads to a damping, that however is too small to describe the experimental observations. Moreover the comparison of this case with the experimental data in Figure 3. The computational oscillation period is smaller now than in Case 1.

Case 3: A bubble immersed in an incompressible liquid with phase transition and heat conduction. The non-isothermal evolution is described by a system of three first order ordinary differential equations that are coupled now to the partial differential equation for the evolution of the temperature. The latter equation is solved numerically by the method of lines. The resulting large system of ordinary differential equations is again solved with a second order Runge Kutta method. For the spatial discretization finite differences of second order are used.

In this case we observe an enormous variation of the temperature. However its influence on the evolution of the bubble radius can be ignored, in particular there is only a small additional damping in comparison to Case 2, see Figure 4 and Figure 3 for comparison.

The temperature variation has large impact on the phase transition. At the minimal radius there is a large increase of temperature that leads to a change of the gas-liquid phase transition. In fact, we observe here evaporation leading for some time to a reduction

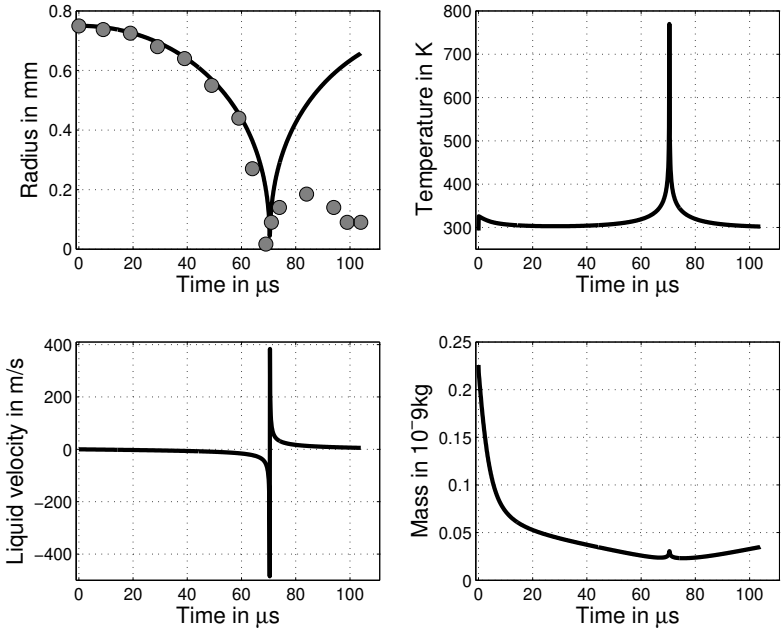

Figure 4:
Calculated bubble radius, bubble mass, liquid velocity and temperature at the interface according to Case 3. Dots: Experimental data, solid line: Computation.

of the released latent heat. During that period where the temperature exceeds the critical temperature we turn off the phase transition, by setting $\dot{N} = 0$. Note, that in addition to the initial data from above we start here with $v = 0$ at $t = 0$ in the whole liquid domain.

Case 4: A bubble immersed in a weakly compressible liquid with phase transition and heat conduction. Recall that in the cases 1 - 3 the momentum balance is reduced to an ordinary differential equation. This is not possible anymore if the compressibility of the liquid is taken into account. In order to reduce numerical complexity we restrict here to the case where the momentum balance becomes a linear wave equation, that is now coupled to a modified version of the equations of Case 3. The wave equation is solved by means of in- and outgoing waves and the resulting system of two delay equations is likewise solved with a second order Runge-Kutta method. Note, that we meet a free boundary here, and in Case 3, i.e. at constant cell number we have a variable spatial mesh size.

Figure 5 reveals a sufficiently strong damping now, so that we can observe good agreement with the experimental data. On the other hand the oscillation period is slightly smaller than in the experiment.

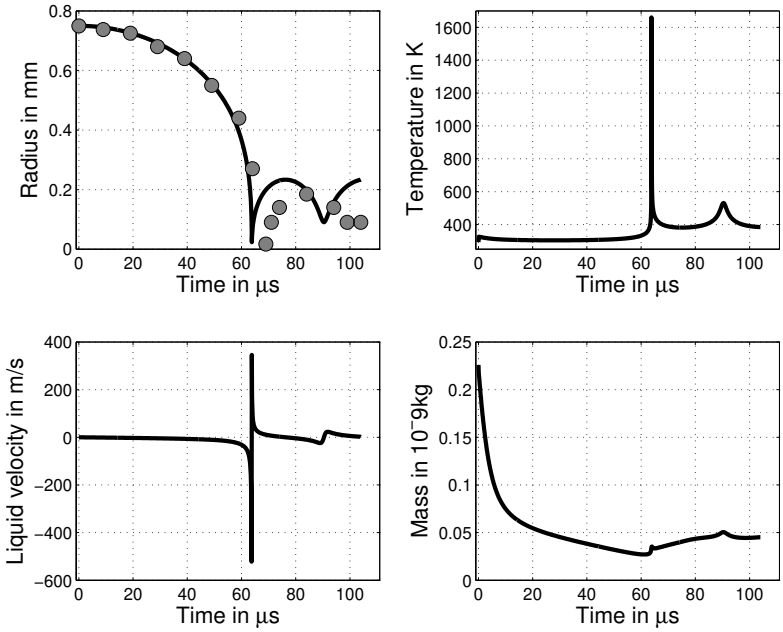

Figure 5:
Calculated bubble radius, bubble mass, liquid velocity and temperature at the interface according to Case 4. Dots: Experimental data, solid line: Computation.

$\sigma = 0.07274 N/m$	surface tension
$\bar{p} = 2330 Pa$	saturation pressure
$\rho = 998.2 kg/m^3$	liquid density
$c_p^L = 4183 J/kg/K$	specific heat capacity of liquid water at constant pressure
$c_{pW} = 1882 J/kg/K$	specific heat capacity of water vapor at constant pressure
$s^L = 296 J/kg/K$	specific entropy of liquid water
$s_W = 8665 J/kg/K$	specific entropy of water vapor
$r = 2453300 J/kg$	specific heat of evaporation
$\lambda = 0.5984 W/m/K$	heat conductivity
$c = 1482 m/s$	speed of sound in liquid water
$MH = 2 \cdot 1.0079 g/mol$	molecular weight of hydrogen
$MO = 2 \cdot 15.9994 g/mol$	molecular weight of oxygen

7 Conclusions

The main conclusions of this study concern the evolution of a laser induced bubble as it is described in Section 3, and can be summarized in five statements.

- Isothermal treatment of liquid-vapor phase transitions is not appropriate if the two phase system is exposed to atmospheric pressure.

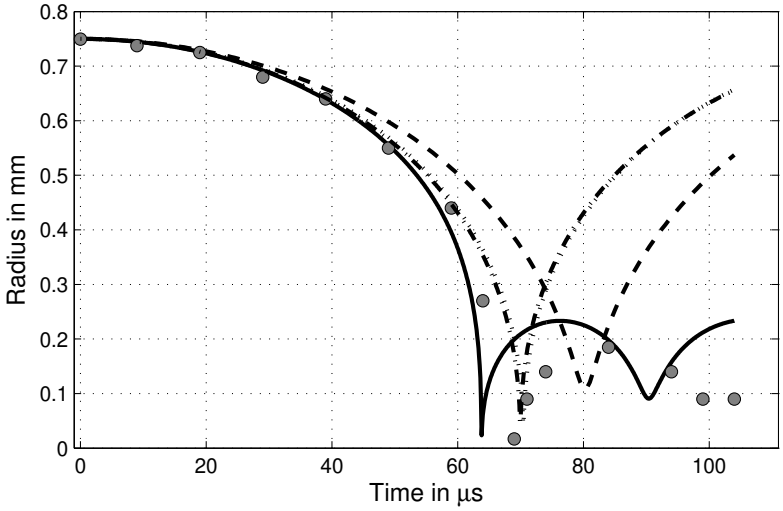

Figure 6:
Calculated bubble radii according to Cases 1 - 4. Dots: Experimental data, dashed line: Case 1, dash-dotted line: Case 2, dotted line: Case 3, solid line: Case 4. (Remark: Case 2 and Case 3 are nearly the same.)

- The modeling of the liquid as an incompressible body ignores the crucial effect that controls the damping of the bubble oscillation.
- The exclusive description of the evolution of the bubble radius, as it is done in the framework of Keller-Miksis type approximation like (2), where the phase transition is ignored, is possible. However, if the obtained data are used we calculate pressures and temperatures in the bubble as well as in the liquid, one observes implausible values for those quantities. This is related to the fact, that the Keller-Miksis approximation relies on a small Mach number expansion, that becomes unrealistic in the region of minimal bubble radius.
- If a liquid-vapor phase transition is allowed, the rebound of the bubble is only possible when an inert gas is present in the bubble. A pure water vapor phase cannot persist beyond the first collapse since the vapor phase is unstable under the conditions of the experiment.
- Surprisingly, the non-isothermal treatment including phase transition has no large impact on the evolution of the bubble radius but on the other variables of the thermodynamic states of the bubble-liquid system. However, the bubble radius is the only quantity, that currently can easily be measured.

Finally we have collected the evolutions of the bubble radius corresponding to the four considered cases in a common plot, see Figure 6.

In a further study, which is in progress, we will use the models developed in this paper in the derivation of two-phase mixture conservation laws with phase transition from averaging procedures.

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