Particle modeling of strongly collapsing microbubbles

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Introduction

Gas or vapor bubbles in water encountered in experiments on acoustic or optic cavitation have a large range of sizes (rest radii R_0) from above 1 mm down to less than 1 μm. In particular, trapped stably oscillating bubbles emitting single bubble sonoluminescence (SBSL) light (Fig. 1) have a rest radius of a few μm, being the smaller the higher the excitation frequency is.

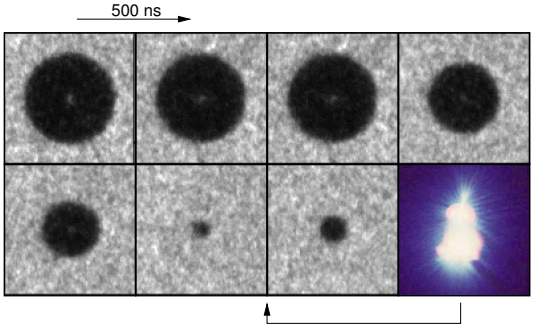

Figure 1: Sonoluminescing bubble collapsing spherically, taken with short-exposure photography. Maximum bubble radius is about 50 μm. The very fast collapse takes place between the 6th and 7th frame. Frame 8 is a long-time exposure (several h) showing the integrated SBSL light emission with higher magnification (courtesy of R. Geisler).

Such small bubbles are microfluidic objects with involved physics taking place in their interior as the bubble medium is highly compressed, heated and even ionized. Numerical calculations with continuum (CFD) codes exist that take into account heat and mass diffusion, phase change and chemical reactions [1]. A different approach to numerically study bubble collapse using direct particle simulation (molecular dynamics, MD) [2] has been pursued during the past years by our group [3] and by other researchers [4]. The main ideas and some results obtained so far are briefly summarized.

Why use MD?

A microbubble of radius $R_0 \approx 1\,\mu$m at normal conditions contains about 10^8 molecules only. With present-day desktop computers, MD calculations with several million (up to 10^7) particles are possible, though demanding. Soon, such equipment will be able to simulate the considered microbubbles realistically on a molecular scale.

Presently, a hard-sphere molecular dynamics model is used to represent the gas and vapor mixture inside a bubble during a single collapse cycle. The liquid motion is described by an ODE of Rayleigh-Plesset type. Particle gas and liquid are coupled self-consistently via pressure and velocity at the bubble wall. This phase boundary is assumed to be a sharply defined spherical (or elliptical) surface. Condensation of vapor is modeled by partial absorption of molecules with a given accomodation coefficient when they strike the wall. For details on the model, see [3, 5].

Particle simulation in fluid mechanics has to be employed when the continuum hypothesis of CFD breaks down, either for very small systems where statistical fluctuations become important, or in situations where gradients of macroscopic quantities become very steep, i.e., where the local Knudsen numer $\mathrm{Kn} = \lambda/L$ is not small compared with unity. Here, λ denotes the mean free path of the molecules and L a characteristic length scale, for example, the thickness of a shock front. Thus, in simulations of the bubble medium, the MD method may be superior to CFD approaches in the following cases: (i) with very small bubbles of submicron size; (ii) with highly excited microbubbles where strong shocks (Mach number $M \geq 2$) may occur, or where a shock singularity exists; (iii) with chemical reactions involving species of low concentration, which can be regarded as a 'rarefied gas' concerning the chemical reaction process. Furthermore, realistic ab-initio molecular dynamics simulations are also attractive for capturing the kinetics of non-equilibrium evaporation/condensation and mass transport across the fast moving phase boundary, and the detailed structure of this boundary, in particular, when surfactants are present.

Results

As an example of a microbubble collapse calculated with MD, Fig. 2 shows the spatiotemporal evolution of density of a very small argon bubble without vapor ($R_0 =$ 100 nm) under strong acoustic excitation (frequency 11

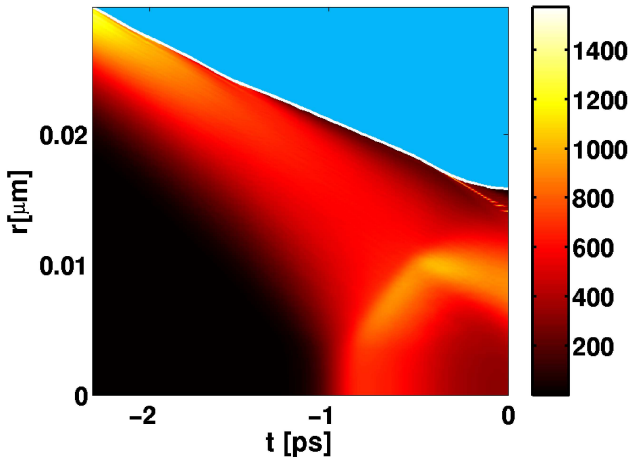

Figure 2: Spatiotemporal evolution of density (kg/m^3) within a microbubble of $R_0 = 0.1\,\mu$m, driven at 11 MHz with an acoustic pressure of 100 bar.

MHz, acoustic pressure 100 bar). Here, simulation particles correspond to real atoms. A strong compression wave detaches from the wall, approaches the center and gives a high maximum temperature of about $4 \cdot 10^5$ K. Fast particles being reflected or passing through the center collide with the incoming wave tail. This simulation does not include energy consumption by vapor dissociation.

MD modelling per se includes linear and nonlinear (steep gradient) diffusion of mass, momentum and energy in the bubble medium. Heat and mass diffusion across the bubble wall have to be implemented by supplementary rules. Besides simple particle reflection, a wall collision rule with thermal accomodation and including a thermal boundary layer in the liquid has been realized. The thermal accomodation coefficient can be adjusted from a purely adiabatic to a purely isothermal condition. For a typical argon SBSL bubble it was found that isothermal conditions can lead to stronger collapse and higher peak temperatures than an adiabatic wall, mainly because the expanding bubble attains a larger maximum radius.

Noble gases and mixtures thereof have been chosen as the bubble medium, because (i) chemical reactions can be dispensed with and (ii) stable SBSL bubbles are mainly composed of noble gases effusing out of the liquid. With noble gas mixtures the Soret effect is observed to be effective within one collapse cycle, leading to mixture segregation. The lighter particles accumulate at the center, affecting shock formation and SBSL light emission.

When water vapor (having rotational degrees of freedom) is included, with evaporation and condensation at the wall but no dissociation, large amounts of vapor can be trapped in the bubble during expansion and the following contraction. The bubble expands to a slightly larger radius than without vapor, and collapses more strongly. However, the peak temperature decreases because the vapor molecules accumulate at the center, giving additional mass and higher heat capacity there. Shock formation is enhanced in the presence of water vapor.

Chemical reactions, in particular the endothermic dissociation of water vapor, introduce significant energy consumption during collapse (Fig. 3). When this feature is turned on in the MD simulations, large amounts of OH

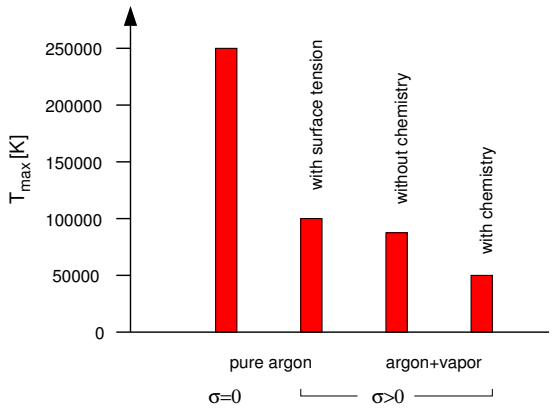

Figure 3: Effect of surface tension σ and of energy consuming mechanisms on maximum temperature achieved in a typical SBSL bubble.

are created at the bubble center and in outgoing waves, lowering the maximum temperature. Also, shock formation is counteracted by this mechanism. Fig. 3 summarizes the effects of surface tension σ, vapor and chemistry on the peak temperature calculated for a typical SBSL bubble.

One advantage of the MD method is its ability to model fully three-dimensional flow. Up to now, this feature was exploited in the investigation of shock focusing stability and the collapse dynamics and sonoluminescence light emission of elliptical bubbles. For example, Fig. 4 shows the spatially resolved, integrated light emission of an elliptically oblate bubble ($\epsilon = 0.9$) which has a pronounced elongated shape. Similar features should obtain for the asymmetric collapse of bubbles near a wall that will be investigated by MD in the near future.

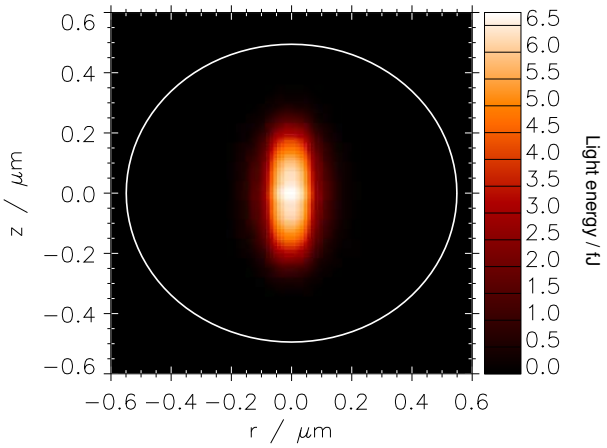

Figure 4: Total integrated light emission of an elliptical bubble ($\epsilon = 0.9$) with reflective boundary conditions, assuming a simple Bremsstrahlung model.

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