Molecular Dynamics of Extreme Mass Segregation in a Rapidly Collapsing Bubble

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A molecular dynamic simulation of a mixture of light and heavy gases in a rapidly imploding sphere exhibits virtually complete segregation. The lighter gas collects at the focus of the sphere and reaches a temperature that is several orders of magnitude higher than when its concentration is 100%. Implosion parameters are chosen via a theoretical fit to an observed sonoluminescing bubble with an extreme expansion ratio (25:1) of maximum to ambient radii.

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The collapse of a gas cavity occurs when its pressure is less than the pressure at infinity of the surrounding fluid. Collapses strong enough to generate plasmas with broadband photon spectra (sonoluminescence) characteristic of thermal motion have been observed for a variety of physical systems. These include single pulsating bubbles in a strong sound field [1–3], bubbles generated by flow through a Venturi tube [4], bubbles activated by a water hammer [5,6], bubbles crushed by shock waves [7], and fluid plugs [8,9]. A key question in cavitation science is: what are the limits of energy density, charge density, mass density, and pressure which can be achieved inside of collapsing bubbles. These issues can be approached via experimental studies of sonoluminescence [10,11], hydrodynamical analysis of collapsing bubbles [12–15], and molecular dynamics (MD) simulations of the interior of a collapsing bubble [16,17]. Here, we use MD to study bubbles containing a binary mixture of light and heavy atoms. For a sufficiently rapid collapse, we find that the lighter gas segregates to the center of the bubble where it is hammered by a shock from the heavier gas so as to reach temperatures vastly higher than are achieved in the pure lighter gas.

In this Letter, we study molecular dynamics (for detailed description of our model see Refs. [17,18]) under conditions where the most extreme temperatures and densities can be expected inside an imploding bubble. Figure 1, for instance, shows the radius R as a function of time t for bubbles which exhibit the largest expansion ratio (~ 30) of maximum radius to ambient radius yet measured. For boundary conditions inspired by Fig. 1, simulations reveal that an imploding shock wave is launched when it is filled entirely with xenon gas [Fig. 2(a)]. But, for the same $R(t)$, a collapsing helium bubble does not experience an imploding shock [Fig. 2(b)] with the result that its simulated peak temperature (200 000 K) is 1000 times lower than the corresponding xenon bubble.

In the framework of hard-sphere MD, gas properties are determined by two parameters: atomic mass and cross-section. By comparing various simulations, in which masses and cross sections were interchanged, we established that the launch of a shock wave was correlated with the atomic mass, probably due to its inverse correlation with the speed of sound. As it is important to understand the conditions under which lighter gases, such as Deuterium, can be energized via an implosion, we were motivated to simulate the tradeoffs involved (e.g., higher temperature versus lower collision rate) in mixing a lighter gas (which for the purpose of this Letter is helium) with a shock wave forming gas, xenon. The main result of the Letter is shown in Fig. 3, which displays a successive series of density profiles for He and Xe inside of a rapidly collapsing bubble. According to this figure, the low atomic mass which suppresses shock formation now causes the helium to separate virtually completely from the xenon so as to form a highly concentrated shell of helium which leads the imploding xenon shock wave, and is subsequently hammered by the xenon shock wave as it approaches $r = 0$, the geometric center of the bubble and the focus of the imploding shock wave. At the moment of collapse, the region, which consists of almost 100% helium, reaches temperatures that are much higher than for a pure helium bubble and about $1/2$ those of a pure xenon bubble. Elemental concentration in collapsing bubbles was first calculated hydrodynamically by Szeri [19] who observed a $\sim 10\%$ thermodiffusion effect (also known as Soret effect) for a bubble with a smaller expansion ratio of about 10.

Molecular dynamics simulations are carried out for a bubble which has the same maximum and ambient radius as a physical bubble. The number of atoms in such a real bubble is about 8×10^8, which exceeds standard computational capabilities. To make the calculations feasible, we assume spherical symmetry and simulate gas inside a cone [18] with a vertex half angle of 0.8°, which therefore contains about 55 000 atoms. Some cases were verified for cone angles of 1.6° where the number of atoms is roughly 4 times greater. The simulations are carried out for two limiting models of molecular dynamics: (a) the hard-sphere model [20] and (b) the variable soft sphere

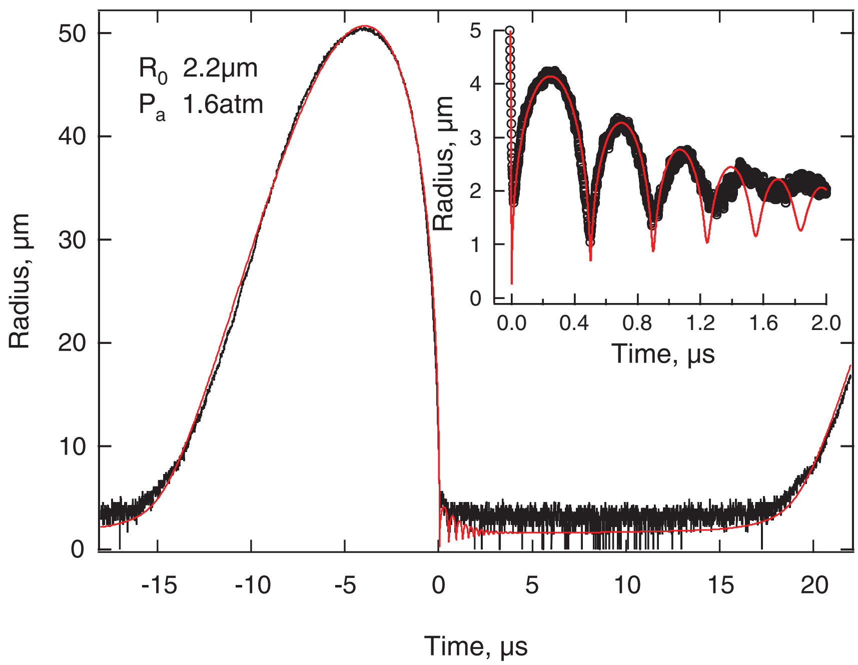

FIG. 1 (color). The figure shows experimentally achievable sonoluminescing bubbles with a large expansion ratio, which is the maximum radius divided by the ambient radius R_0 (i.e., the radius of the bubble immediately after the sound field is turned off). The expansion ratio is independent of which noble gas is used but is dependent on drive level and the concentration of gas in the surrounding water. The largest expansion ratio realized is 30:1; simulations for this Letter were carried out for a bubble with an ambient radius of 2.2 microns and an expansion ratio of 24. The expansion, collapse, and after bounces are fit by the Rayleigh-Plesset "RP" equation (see discussion in Ref. [2]). The radius versus time is not directly experimentally determined for $R < R_0$. For purpose of the simulations, $R(t)$ is extrapolated into this region via the solution to the RP equation.

"VSS" model [17,21–23]. In the VSS model, the collision cross section decreases with increasing center of mass energy. The customary VSS dependence is stronger than would be found for the Leonard-Jones 6–12 potential. The physical system lies between these limiting cases. Simulations included cooling due to ionization (with a maximum of 5 levels for xenon), and heat bath (constant temperature) boundary conditions at the wall of the bubble. Thermal conductivity of the electrons is ignored as is the vapor pressure of the surrounding fluid. The bubble radius as a function of time $R(t)$, which drives the implosion, is the solution to the Rayleigh-Plesset (RP) equation that best fits the experimental data obtained from light scattering for that portion of the acoustics cycle where $R > R_0$: the ambient radius. For radii much smaller than the ambient radius, the wall motion is obtained from this best fit, which includes the effects of viscosity and acoustic radiation damping but not the launch of outgoing shock waves [24,25] by the bubbles surface whose best fit velocity reaches 10 times the speed of sound in water. While cooling of the ions due to ionization is included (the free electrons are assumed to be created at rest), the local electric field of the plasma and recombination plays no role in our analysis. The effect of thermal radiation transport is also not included. Although these molecular dynamic simulations are endowed with the capability of describing large gradients (e.g., Fig. 3) that occur on the scale of the gas dynamic mean-free path, these results do not yet constitute a first principles theory of sonoluminescence.

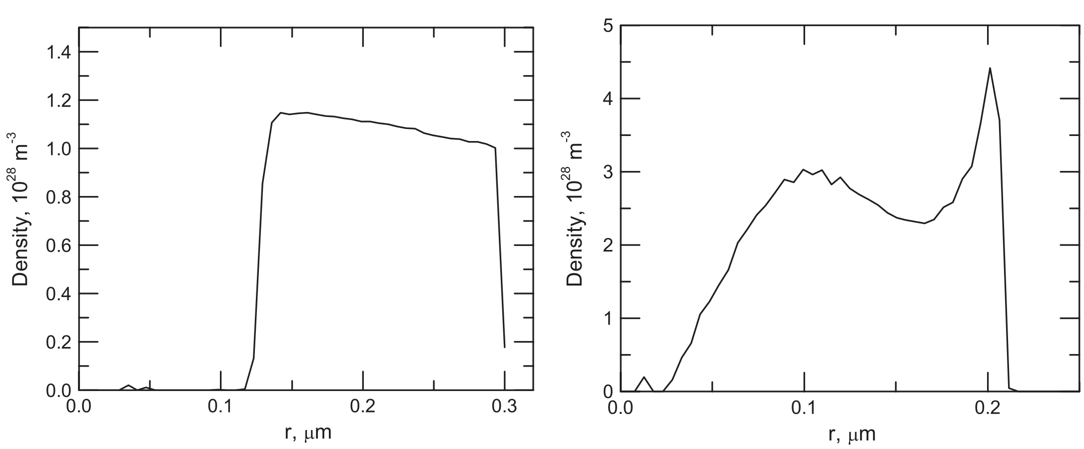

FIG. 2. Density profiles of pure xenon (a) and helium (b) bubbles at an intermediate time between R_0 and the minimum radius. This simulation is carried out for the hard-sphere model.

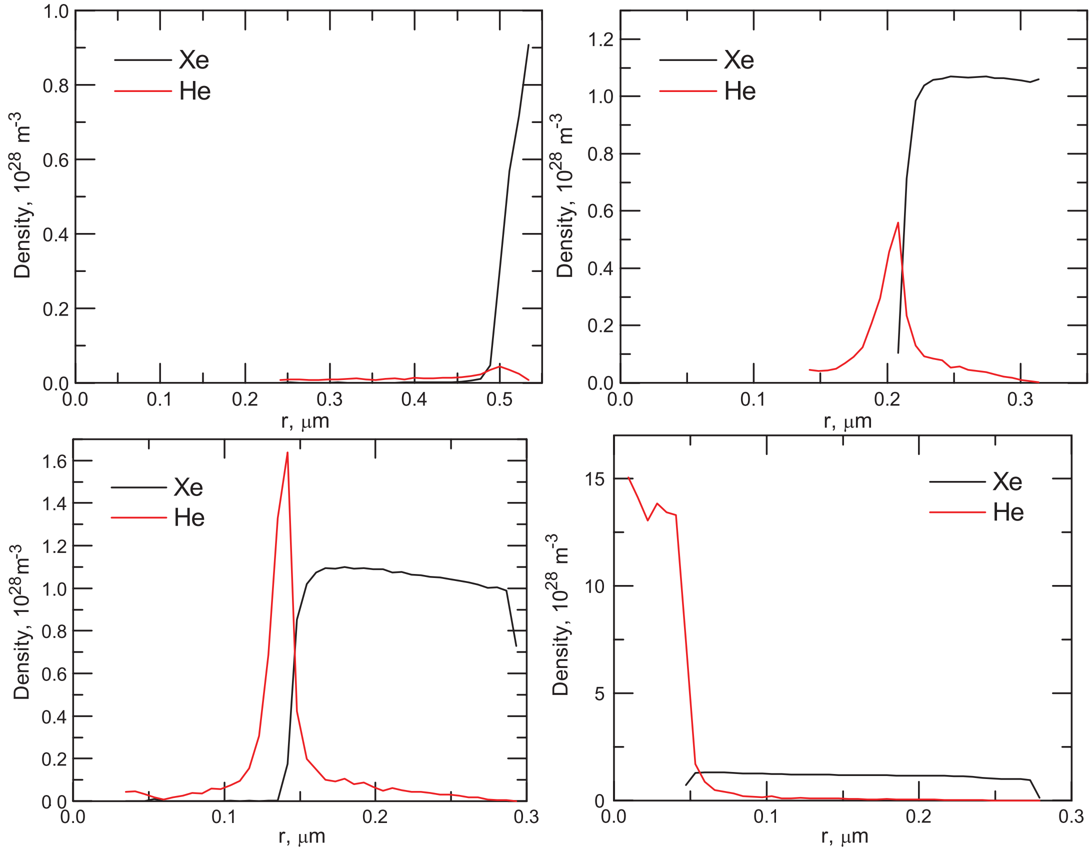

FIG. 3 (color). Density profile of helium and xenon inside of a collapsing bubble at four successive moments between R_0 and the minimum radius. This calculation was carried out for the hard-sphere model of molecular dynamics.

Figure 4 shows peak temperature as a function of time for various mixtures of He and Xe for the VSS and hard-sphere cases. Note that the hard-sphere temperatures are much higher and display sharper peaks. Both the VSS model and the hard-sphere model yield helium mole fractions above 99% for the central region of the hot spot. For a 10% overall helium concentration, the size of this fully segregated region is about 50 nm. Helium segregation is unaffected by artificially changing its atomic radius so that it is the same as xenon, while changing the mass of the

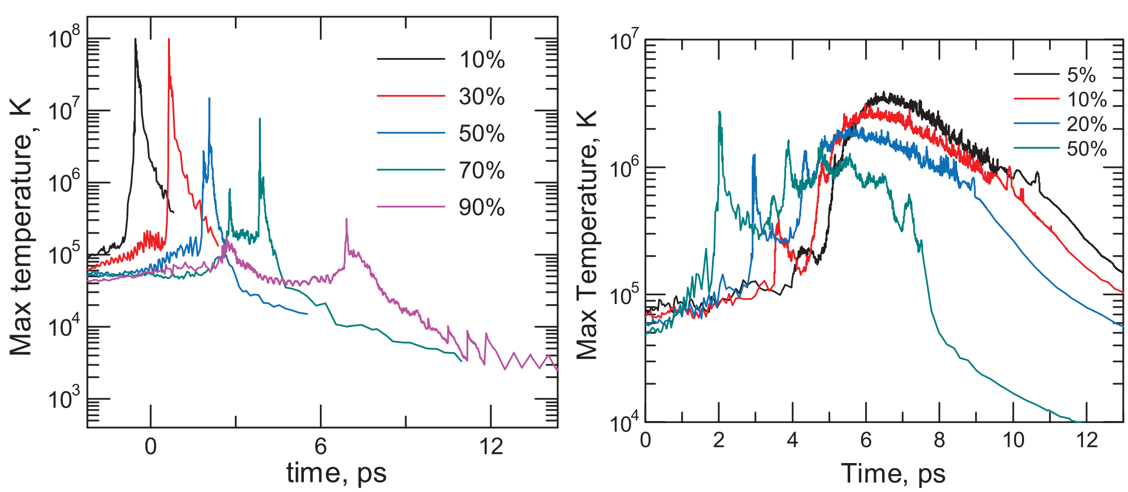

FIG. 4 (color). Maximum temperature vs time (after collapse) near hot spot for hard sphere and VSS MD simulations of various mixtures of helium and xenon inside of a strongly supersonically collapsing piston. The legend shows the percentage of He in the mixture.

helium for fixed smaller radius eliminates segregation entirely. We thus conclude that segregation is due to the smaller mass and therefore higher velocity of helium atoms for a given temperature. The strongly supersonic collapse is also important to the observation of virtually complete segregation as bubbles with expansion ratios of about 10 display only small changes in relative concentration [19].

If a physical system containing Deuterium instead of helium reached the densities and temperatures characteristic of the simulations discussed here, then the thermonuclear fusion rate [26] would yield 6×10^{-5} neutrons per collapse or 2 n/s at a repetition rate of 30 kHz. This rate uses the formulas

$$N = n^2 \langle \sigma v \rangle R_{\mathrm{hot}}^3 \Delta t_{\mathrm{hot}}$$
$$\langle \sigma v \rangle = 4 \times 10^{-12} \tilde{T}^{-2/3} \exp(-20 \tilde{T}^{-1/3}\ \mathrm{cm}^3/\mathrm{s})$$
$$\tilde{T} = T/1.16 \times 10^7\ \mathrm{K}$$

with input values for the temperature $T = 10^7$ K for a duration $\Delta t_{\mathrm{hot}} = 0.25$ ps for a hot spot of radius $R_{\mathrm{hot}} =$ 20 nm.

In conclusion, attempts to enhance the simulated temperature of a light gas inside of a rapidly collapsing spherical piston by mixing it with a heavy gas results in a win-win phenomenon. Virtually complete elemental segregation means that the mutual collision rate of the lighter gas is not diminished due to dilution by the heavier gas. Furthermore, the heavy gas still develops a shock front that hammers the lighter gas so as to reach much higher temperatures that are close to those realized in the pure gas of heavier atoms. Remarkably, the region of highest temperature sits entirely within the lighter gas. Figure 3 demonstrates the structure of the singularity which appears in a strongly shocked fluid mixture. Hydrodynamic calculations of strongly shocked fluid mixtures should be formulated so as to be able to capture this structure.

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