Trapped Bubble Dynamics in Cryogenic Fluids

O. Baghdassarian, H. Cho[†], E. Varoquaux[*], and G. A. Williams

Department of Physics and Astronomy, University of California , Los Angeles, CA 90095, USA

Initial studies of the properties of helium bubbles acoustically trapped in liquid nitrogen at 66 K have been carried out. Laser scattering is used to monitor the dynamics of the bubble radius as a function of the acoustic driving field. In this apparatus, no sonoluminescence has yet been observed from helium bubbles in liquid nitrogen, although sonoluminescence was previously observed from xenon bubbles in ethanol cooled to 165 K. It is found that bubbles can be stably trapped at quite low acoustic drive levels, enabling the observation of new dynamical bubble behavior.

1. INTRODUCTION

The phenomenon of single-bubble sonoluminescence (SL) in liquids is still not well understood.[1] The physical mechanism that produces a 50-100 picosecond burst of photons from a collapsing gas bubble is a subject of debate. It is clear that the adiabatic compression of the gas heats it to rather high temperatures, but precisely how this translates to light emission is not known. A number of different mechanisms have been proposed, such as shock waves and plasma radiation,[2] liquid jet formation,[3] and explanations based on atomic and molecular processes.[4] Most experimental work has been in water and a few organic liquids near room temperature. These have the drawback that they are fairly complex liquids, and strongly absorb light in the ultraviolet (UV). Since most of the SL intensity is emitted in the UV,[5]the most interesting spectral region for SL is not readily accessible.

We have undertaken initial studies investigating whether single-bubble sonoluminescence can be observed in cryogenic fluids such as nitrogen, oxygen, and argon. These liquids have considerably less absorption in the UV than water, and could allow investigation of the SL spectrum over a wider

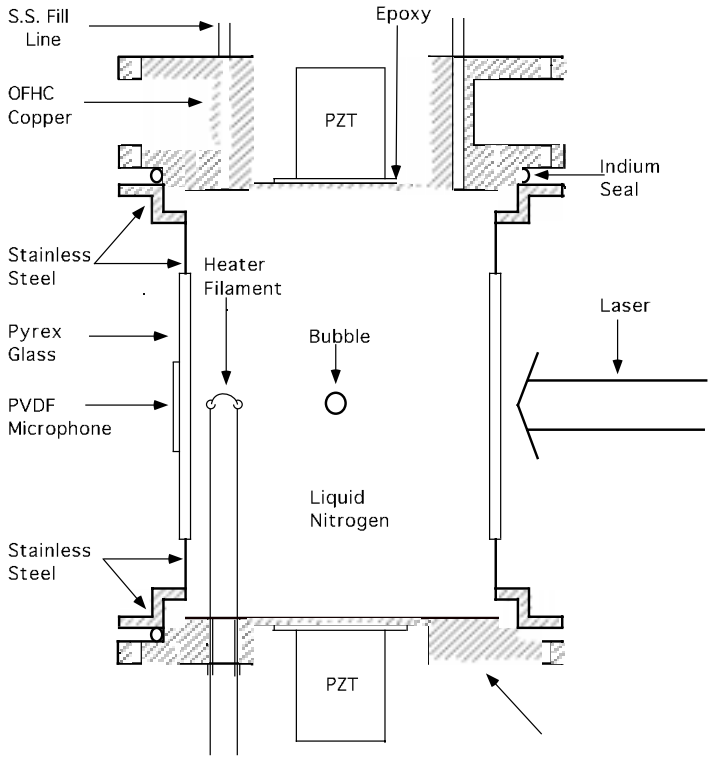

Fig. 1. Schematic of the experimental cell.

range. The lower starting temperature for the compression of the bubble does not necessarily mean that the SL will be quenched: the first measurements in our cryogenic apparatus used xenon bubbles in ethanol, from which strong SL could be observed at temperatures down to 165 K, with increasing brightness as the temperature was lowered.[6] There has also been an early report[7] of observation of weak multi-bubble SL in liquid nitrogen at 77 K, under conditions of transient cavitation.

It is known that the dynamics of the bubble motion are strongly connected with the occurence of SL.[1] Since our previous work showed that single bubbles could be successfully trapped in liquid nitrogen,[8] we have undertaken light-scattering studies to investigate the dynamics of helium bubbles in liquid nitrogen, and some preliminary results are presented here.

2. APPARATUS

A schematic of the experimental cell is shown in Figure 1. The acoustic resonator is a cylinder 3.7 cm in diameter and 10 cm in length, with a cylin-

drical Pyrex glass window 5 cm long in the middle. The cell is acoustically driven by two hollow PZT cylinders 1.9 cm in length, which are epoxied to a recess in the copper end plates where the copper thickness is 0.6 mm. The bubble-trapping mode with an antinode at the center occurs near 28.5 kHz with liquid nitrogen in the cell, as monitored with a PVDF microphone epoxied on the outside surface of the Pyrex window. The bubbles are generated by briefly pulsing current through a heater filament.

The cell is suspended in a vari-temp optical cryostat using pumped liquid nitrogen as a coolant. High-purity nitrogen gas (5N) is condensed into the cell through two independent 3 mm diameter filling lines after being passed through a nitrogen trap, and the liquid is condensed well up into the lines to maintain the cell pressure at about 900 mm Hg. The cell temperature is regulated using two identical heaters on the top and bottom copper plates. The plates are also thermally connected with a copper wire. The helium gas is then added at a selected partial pressure, and a small diaphragm pump is used to establish a pressure head across the two filling lines to circulate the helium gas into the liquid. This is a slow process, requiring at least 24 hours of circulation before the dissolved gas reaches an apparent equilibrium and the diaphragm pump can be switched off.

The bubble oscillations induced by the acoustic drive are monitored by scattering a 100 mW argon-ion laser beam from the bubble surface (Mie scattering). The light coming off at angles around 90° is focused onto a photomultiplier tube, and the resulting signal is amplified and averaged (for up to 1024 traces) using a fast digital oscilloscope. Since the averaged Mie scattering varies as the square of the bubble radius, the square root of the signal is taken, after first subtracting the background scattering. To test the system we initially filled the cell with water and 150 mm of air. The light scattering measurements reproduced the known bubble dynamics of this system,[1] and above a threshold drive level SL was easily detected on the photomultiplier. The bubbles trapped in liquid nitrogen are generally not as stably trapped as the bubbles in water, particularly at high acoustic amplitudes. They jitter about the trapping point, causing fluctuations in the magnitude of the received signal.

3. OBSERVATIONS

Figure 2 shows the bubble radius (normalized to one at the peak) for a time encompassing the acoustic period of 35 μsec; the different graphs show the progression as the relative acoustic amplitude in the cell (proportional to the microphone signal) is increased. On the rarefaction part of the acoustic drive the bubble radius expands and gas flows in, and then on

the compression part the radius rapidly drops. This is followed by several "afterbounces" at the natural frequency of the bubble, which is determined by the compressibility of the gas. This general behavior is well known for gas bubbles in water.[1] However, we observe several features that are quite different from the water-bubble oscillations. One feature readily apparent at low drives is the very large amplitude of the afterbounces, which has not been seen in water. We find that for high helium concentrations we can trap bubbles at quite low acoustic drive levels, apparently much lower than can be achieved with bubbles in water. In a brief run with 133 mm of helium we found that the drive could be reduced to the point where only quasi-linear oscillations of the bubble at the drive frequency could be observed, with no afterbounces at all.

Another difference between the curves in Fig. 2 and those for the water bubbles is that at higher drives the collapse of the bubble after the expansion appears to be considerably slower in the nitrogen case. The slope is measurably finite, a bubble surface velocity that can be estimated at a few m/s, compared to the almost sonic velocities seen for the water bubbles.[1] This lack of rapid compression may be the reason that we have not yet observed SL from the bubbles in nitrogen.

One major difference between water and nitrogen is the vapor pressure in the bubble. At 66.5 K the vapor pressure of nitrogen is 170 mm Hg, compared to about 20 mm in water at room temperature. Combined with the lower temperature this means that the vapor density in the bubble is more than an order of magnitude larger than in the water bubbles, and this may impact the bubble dynamics. The evaporation-condensation process at the bubble surface introduces dissipation and soaks up some of the energy input from the acoustic drive. The diffusive motion of helium in and out of the bubble could be hindered by the vapor, possibly slowing down the dynamics. We can visually observe signs of dissipation by backlighting the bubble, where a convective "halo" can be seen around the bubble, indicating that the liquid in its vicinity is being heated. This effect was very prominent at the lowest helium concentrations we tried, around 0.2 mm partial pressure (we could not produce bubbles in pure liquid nitrogen at 900 mm static pressure). For these bubbles (which could only be trapped at relatively high drive levels) the halo was very distinct, and was also accompanied by a tiny rising plume of heated liquid. At higher helium pressures the extent of the halo decreased considerably, becoming barely observable.

There is also evidence of anomalous dissipation in the curves of Fig.2. There is an "onset" point near relative microphone level 0.711 where the afterbounces begin to decay very rapidly at the higher drive levels. The decay rate becomes even faster than those we observed for the afterbounces

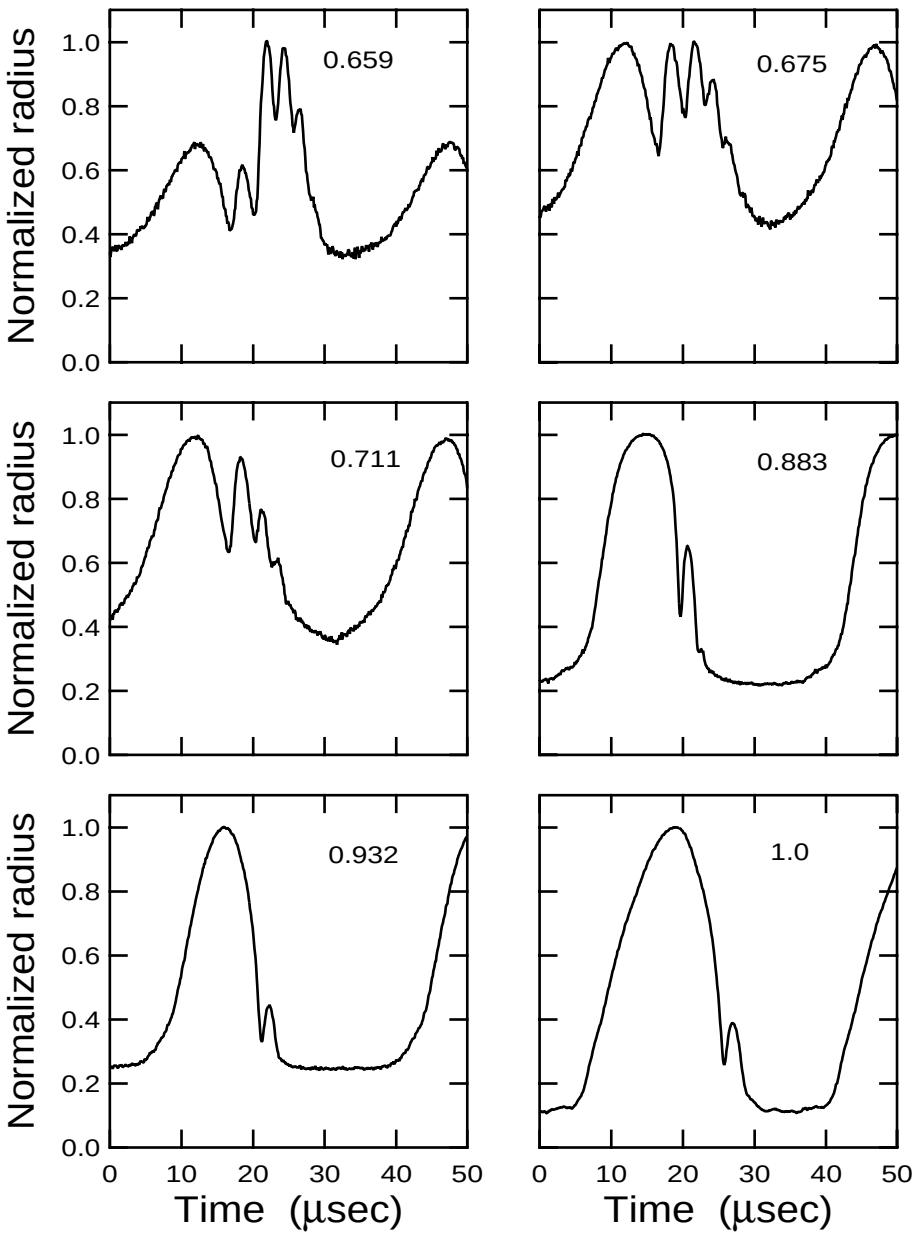

Fig. 2. Helium bubble radius (normalized) versus time, in liquid nitrogen at 66.5 K. The number in each graph denotes the relative acoustic amplitude, normalized to the last graph. The helium partial pressure is 38 mm Hg.

of water bubbles in our cell, in spite of the fact that the viscosity of the liquid nitrogen is 3-4 times smaller than that of water.

We plan to try fitting our data to the Rayleigh-Plesset equation for the bubble motion.[9] The theory has been extended recently[10] to include evaporation-condensation effects, and our system may provide a good test of the model. If successful, this should allow us to calibrate the bubble size and the acoustic pressure amplitude. In further work, the measurements will be extended to higher helium concentrations and different temperatures and pressures, as well as to neon gas and to mixtures with oxygen, argon, and xenon gas (all of which dissolve appreciably in the nitrogen). We will also test other cryogenic liquids such as oxygen and argon. Liquid oxygen should allow an interesting comparison to the liquid nitrogen, because of its much lower vapor pressure, and it may be a better candidate for the observation of single-bubble sonoluminescence.

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*Present address: Laboratoire de Physique des Solides, Université Paris-Sud, 91405 - Orsay - France.

† Present address: Physics Dept., Univ. of Chicago, Chicago IL 60637.

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