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Inside a Collapsing Cavity: Sonoluminescence as a Spectroscopic Probe

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SUMMARY

Extreme temperatures and pressures are produced through acoustic cavitation: the formation, growth and collapse of bubbles in a liquid irradiated with high intensity ultrasound. Single bubbles have generally been assumed to give higher temperature conditions than bubble clouds, but confirmation from the single bubble sonoluminescence (SBSL) emission spectra have been problematic because SBSL typically produces featureless emission spectra that reveal little about the intra-cavity physical conditions or chemical processes. Here we present definitive evidence of the existence of a hot, highly energetic plasma core during SBSL. From a luminescing bubble in sulfuric acid, excited state to excited state emission lines are observed both from noble gas ions (Ar^+, Kr^+, and Xe^+) and from neutral atoms (Ne, Ar, Kr, and Xe). The excited states responsible for these emission lines range from 8.3 eV (for Xe) to 37.1 eV (for Ar^+) above the ground state. Observation of emission lines allows for identification of intra-cavity species responsible for light emission; the energy levels of the emitters indicate the plasma generated during cavitation is comprised of highly energetic atomic and ionic species. Ionization and plasma can also be created during multi-bubble sonoluminescence and also from high velocity interparticle collisions of powders in slurries, which results in mechanoluminescence at the point of impact.

INTRODUCTION

Models of spherical supersonic bubble implosion in cavitating liquids predict the generation of temperatures and densities that may be sufficient even for thermonuclear fusion [1,2]. Experimental protocols have not yet been found that provide convincing evidence of fusion, but the transient conditions during acoustic cavitation can certainly be extreme [3-5]. There is, however, a remarkable lack of observable data on the conditions created during bubble collapse. Indeed, only recently has strong evidence of plasma formation been obtained [6].

RESULTS

Shown in Figure 1 is a typical emission spectrum along with an image of the acoustic resonator containing a brightly luminescing bubble at its center (i.e., the acoustic field velocity node). The spectrum consists of lines from electronically-excited Ar atoms and a featureless continuum attributed to radiative plasma processes (e.g., bremsstrahlung, recombination, etc.). The emission lines are from electronic transitions between states within the $4p – 4s$ array, the energies of which range from 11.5 to nearly 14 eV. By measuring the relative intensities of the lines, we are able to determine the temperature of the environment from which the Ar atoms are radiating (Figure 1c).

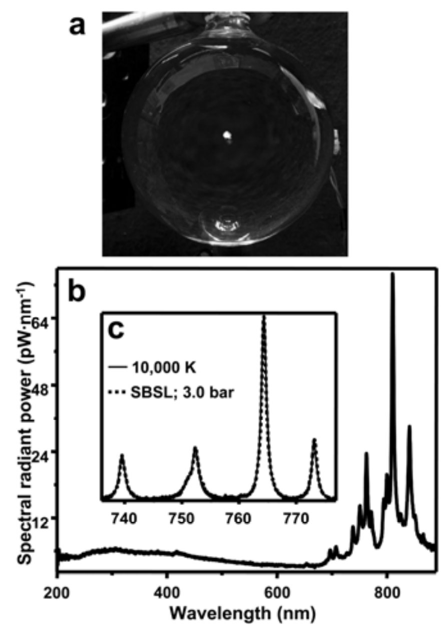

Figure 1. Single bubble sonoluminescence (SBSL) from sulfuric acid (85 wt. % H_2SO_4 containing Ar at 5% of saturation). (a) Photograph of a rapidly translating sonoluminescing bubble at the velocity node of a spherical quartz resonator. The driver piezoceramic is partially visible at the bottom of the image, while the microphone is to the right. The entire apparatus is rigidly clamped at the narrow neck of the quartz flask (top of image). **(b)** A typical SBSL emission spectrum from a bubble driven with a relatively low acoustic driving pressure, P_a. The emission lines (~700 – 900 nm) are due to electronic transitions between states within the $4p – 4s$ array of neutral Ar. **(c)** A higher resolution spectrum of SBSL Ar emission and a least-squares Lorentzian fit at a thermally-equilibrated temperature of 10,000 K.

DISCUSSION

From an analysis of the asymmetry from atomic line emission from SBSL, we have determined for the first time the plasma electron density, ion broadening parameter, and degree of ionization during single-bubble sonoluminescence and examined them as a function of acoustic driving pressure, as summarized in Table I. We find that the electron density can be controlled over four orders of magnitude and can exceed 10^{21} cm^{-3} (which is comparable to the densities produced by intense laser-induced inertial confinement fusion experiments [7]) with effective plasma temperatures ranging from 7,000 to more than 16,000 K. At the highest acoustic driving force, neutral Ar lines can no longer be used as spectroscopic reporters due to the extent of ionization and to leveling of the population of states.

Accounting for the temporal profile of the sonoluminescence pulse and the potential optical opacity of the plasma suggests the ultimate conditions generated inside the collapsing bubble may far exceed those determined from emission from the outer transparent region of the light-emitting volume. A bubble acoustically-driven into nonlinear radial oscillation can focus the diffuse energy of the sound field by many orders of magnitude [8]. The energy focusing is such that broadband light emission is observed (sonoluminescence, SL) [4] and molecular bonds are broken (sonochemistry) [9]. Measurement of the bubble dynamics of a single sonoluminescing bubble (SBSL) has shown the implosion velocity to be greater than the speed of sound with enormous acceleration near maximum collapse [10]. The bubble dynamics and the properties of the emitted light suggest the generation of extreme intracavity conditions. Indeed, recent molecular dynamics simulations predict temperatures approaching 10^8 K but lasting for only a few hundred femtoseconds [2]. The extreme conditions generated during SBSL arise from quasi-adiabatic compression of the bubble contents. One measure of the intensity of bubble implosion is the ratio of maximum to minimum bubble volume (i.e., compression ratio). The value of the compression ratio, and hence the bubble kinetic energy, increases with increasing acoustic pressure (P_a) [11]. Thus, at high P_a there is more energy available to be transferred to the bubble contents, which should ultimately produce more extreme intracavity conditions.

Table I. Summary of the SBSL Ar line profiles, antisymmetric deviations, and plasma conditions as a function of the acoustic driving pressure, P_a.

P_a (bar)	x_c (nm)	w (nm)	T_{Ar} (K)	Δ_{amp} (%)	A	N_e (cm^{-3})	α
2.7	764.26	0.78	7,000	-1.9	0.093	4×10^{17}	$3 \times 10^{-4(p)}$
3.0	764.37	0.95	10,000	-2.1	0.10	1×10^{18}	$8 \times 10^{-4(p)}$
3.3	764.43	1.21	13,000	-3.8	0.20	2×10^{19}	$0.02^{(p)}$
3.6	764.48	1.47	15,000	-6.9	0.45	5×10^{20}	$0.4^{(l)}$
3.8	764.44	1.55	16,000	-8.7	0.69	4×10^{21}	$3^{(l)}$

CONCLUSION

It will be of great interest to determine the SBSL plasma properties as a function of time during the emission flash. The implications for the ultimate conditions that may be generated during acoustic cavitation in exotic liquids are remarkable. As bubble implosion proceeds, there is experimental evidence of the formation of an optically opaque core [12-14]. The interior plasma at the core of the collapsing bubble, which is not visible by emission spectroscopy, must have conditions that exceed, perhaps dramatically, temperature and electron densities measured for the outer emitting surface shown in Table I. The plasma conditions within the optically opaque core of a collapsing bubble may be truly extraordinary.

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