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Luminescence of transient bubbles at elevated ambient pressures

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The light emission of transient laser-produced cavitation bubbles in water is investigated in a range of ambient pressures up to 5 bar and laser energies up to 30 mJ. At elevated pressures bubble luminescence can be increased more than two fold for bubbles created with the same laser energy, and up to almost an order of magnitude comparing bubbles of the same maximum radius. Both the conversion of large laser energies into mechanical energy of the bubble, and the conversion of mechanical energy into light are improved at higher pressure.

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I. INTRODUCTION

Gas or vapor bubbles in liquids can emit light when collapsing violently upon strong excitation. This phenomenon was known for a long time in the form of multibubble sonoluminescence (MBSL) where a great number of cavitation bubbles excited by an ultrasonic field take part in the emission. More recently, bubble luminescence has been thoroughly investigated with single, acoustically levitated bubbles (single bubble sonoluminescence, SBSL) [1,2] that feature a dynamics undisturbed by neighboring bubbles or other obstacles. A number of remarkable characteristics of SBSL could be revealed, e.g., the short duration of the light flashes in the range 60–250 ps [3], their blue spectrum with associated temperatures in excess of 10 000 K [4], or the sensitive dependence of the phenomenon on noble gas concentration [2]. However, due to the required stability of the bubble's sustained oscillations and its position, only a limited region of parameter space (spanned by rest radius R_0, acoustic driving pressure, gas concentration, temperature, chemical composition of the bubble medium, etc.) is available for scrutiny in SBSL experiments [5].

This restriction is partly relieved with the collapse of single transient cavities that give rise to single cavitation bubble luminescence (SCBL) [6]. Such bubbles can be produced by methods as different as [7] electrical discharge, the collapse of evacuated glass spheres, injection of gas into flow fields, or optic cavitation, i.e., laser-induced breakdown (LIB) in the liquid [8].

With laser nucleation bubbles can be generated in the bulk of the liquid sufficiently far from the walls of the vessel and with no mechanical obstacles, e.g., spark electrodes, near the forming cavity. Furthermore, bubble size can be easily manipulated by adjusting the laser energy. Due to the breakdown mechanism having a certain energy threshold bubbles generated in water by laser pulses of a few nanoseconds duration are, in general, considerably larger than the bubbles that can be stably trapped in SBSL experiments. The light energy of SCBL pulses increases with the maximum bubble radius, but decreases very fast when the bubble collapse becomes more aspherical [6,9]. SCBL pulse widths are in the nanosecond region and increase with the bubble size [10]. As in SBSL, a reduction of the liquid's temperature raises the luminescence yield [11]. Furthermore, the noble gas content of the water, which has significant influence on SBSL light output, has virtually no effect on SCBL of laser-generated bubbles [12]. In a sound field luminescence from laser-generated bubbles can be enhanced or reduced, depending on the frequency and on the phase of the field with respect to the instance of bubble generation [13].

In this paper we consider the influence of an elevated ambient pressure on SCBL intensity of laser-produced bubbles in a range of laser pulse energies up to 30 mJ and ambient pressures up to 5 bar. It is shown that the light output from such bubbles can be increased significantly at higher pressures. This result is in contrast with observations of SBSL where the luminescence is brighter when the ambient pressure is decreased [14].

II. EXPERIMENT SETUP

The bubbles are generated by focusing the light of a *Q*-switched Nd:YAG laser ($\lambda = 1064$ nm, pulse duration approx 8 ns, maximum pulse energy 780 mJ) in a sealed cell by means of a specially designed, aberration minimized lens system (Fig. 1). The focal spot can be observed on axis and off axis through three quartz windows of 8 mm thickness, built into the cell walls. The cell is equipped with a piston to adjust the pressure in the cell, a pressure sensor, and valves connecting it to a closed filling system. For the measurements we used clean, distilled water whose gas content could be controlled by adjusting the gas pressure in the mixing vessel of the filling system. In preliminary experiments the

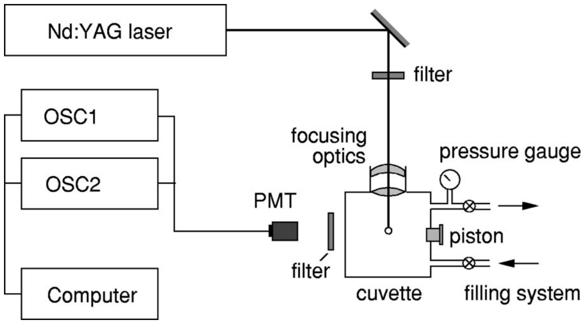

FIG. 1. Experimental arrangement for the investigation of light emission from laser-generated bubbles at controlled ambient pressure and gas content of the liquid.

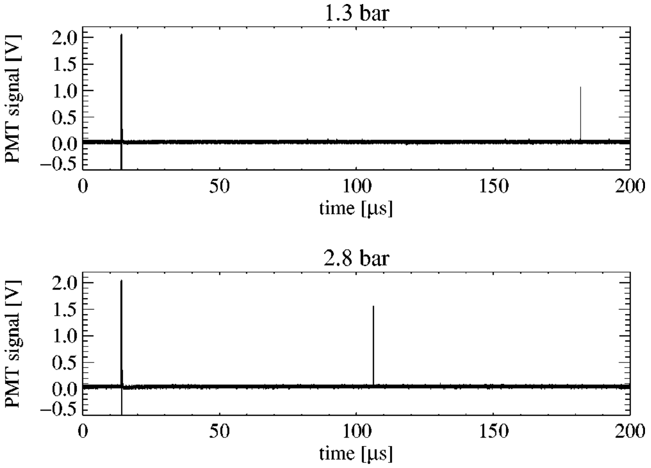

FIG. 2. PMT signal at indicated ambient pressures for $E_L = 11.2$ mJ with the LIB pulse (first spike) and SCBL pulse (second spike).

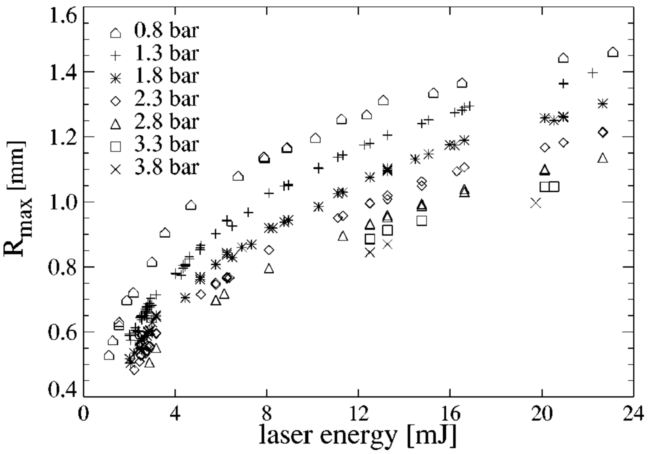

FIG. 3. Maximum bubble radius versus laser energy for degassed water at the indicated ambient pressures.

gas concentration of the liquid appeared to have no significant effect on SCBL strength [12,15]. Therefore we did not systematically vary the gas content but used water either saturated with air, completely degassed, or saturated with argon. The temperature of the water was kept fairly constant at 24 °C during the experiments.

To improve pulse stability the laser was operated at a relatively high power (~400 mJ/pulse) with a repetition rate of 0.5 Hz. Infrared filters were used to attenuate the laser light to the desired energy before it entered the focusing optics. The light emitted perpendicularly to the laser beam axis was detected by a photomultiplier tube (PMT, Hamamatsu R5600U-06), after passing through a 10% gray filter to reduce detector overload.

The PMT signal was recorded by two oscilloscopes (Tektronix TDS 220 and TDA 784A) to achieve sufficient resolution on the different time scales involved. The first oscilloscope was used to acquire both the plasma and luminescence signal. The second, faster oscilloscope (TDA 784A) acquired the luminescence pulse with high temporal resolution. After each successful shot, the memories of both oscilloscopes were transferred to the controlling computer and stored in a database for subsequent analysis. For each data point at least 20 shots were recorded.

Figure 2 shows typical recordings for two values of ambient pressure. The first pulse is generated by the LIB plasma, the second pulse by SCBL. The undershoot of the first pulse is due to clipping of the electrical signal. The time T_{12} between the two pulses gives, to good accuracy, the collapse time of the bubble, $T_c = T_{12}/2$, which was used to calculate the maximum bubble radius by means of Rayleigh's formula for an empty cavity [16]

$$R_{\max} = 1.09\sqrt{\frac{p - p_v}{\rho}}\,T_c\,, \tag{1}$$

where $\rho = 997.3$ kg/m^3 is the density of water and $p_v = 0.0298$ bar is the vapor pressure at 24 °C. Figure 2 exemplifies that, at the higher value of pressure, the time T_{12} between the two pulses is reduced significantly, and the SCBL flash is brighter.

To facilitate comparison of the data with previously published results (e.g., [2]) the energy of the SCBL light pulses is given in terms of photon numbers. They are calculated as follows. We assume isotropic emission and neglect any reflection or absorption in the path of the light. A lower bound on the optical pulse energy is obtained by integrating the electrical signal over the duration of the pulse, using the gain and the maximum spectral sensitivity (at $\lambda_0 \approx 400$ nm) of the PMT, and taking into account the solid angle subtended by the PMT entrance area. This energy is then represented by an *equivalent* number of photons, N_{ph} having the wavelength λ_0. Note that since the maximum sensitivity of the PMT was used and the spectrum of the luminescence light was not measured these photon numbers have to be taken as lower bounds on the true *equivalent* numbers.

III. RESULTS

In the experiment bubbles were generated with different laser pulse energies at ambient pressures adjusted to increasingly higher values. The resulting maximum radii of the bubbles, $R_{\max}$, are given in Fig. 3 as a function of the laser energy for a selection of ambient pressure values p. As expected, $R_{\max}$ increases with the laser energy, but decreases with the pressure p.

In Fig. 4 the photon number N_{ph} is plotted as a function of the maximum radius at different ambient pressures p. It is observed that SCBL becomes brighter with increasing static pressure when bubbles of equal maximum radius are compared. For example, the increase of the number of photons amounts to almost an order of magnitude for bubbles with $R_{\max} \approx 1.3$ mm. This behavior is plausible since more energy is stored in bubbles of the same maximum size at higher pressures. The presented data imply that these bubbles remain sufficiently stable upon collapse to also yield higher SCBL energies.

In Fig. 5 the number of photons is presented as a function of the laser pulse energy for different values of ambient pres-

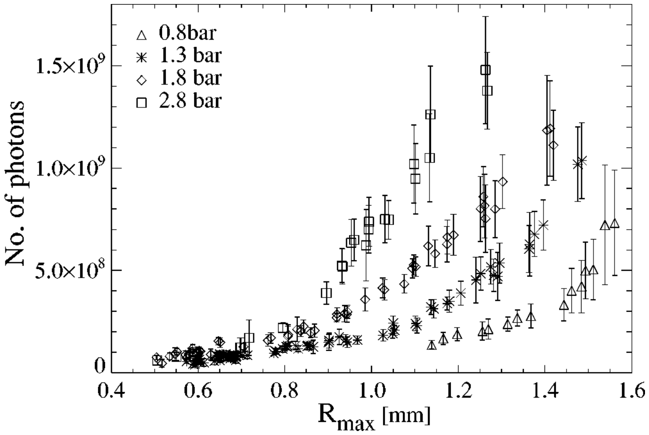

FIG. 4. Number of photons versus maximum bubble radius for degassed water at the indicated ambient pressures.

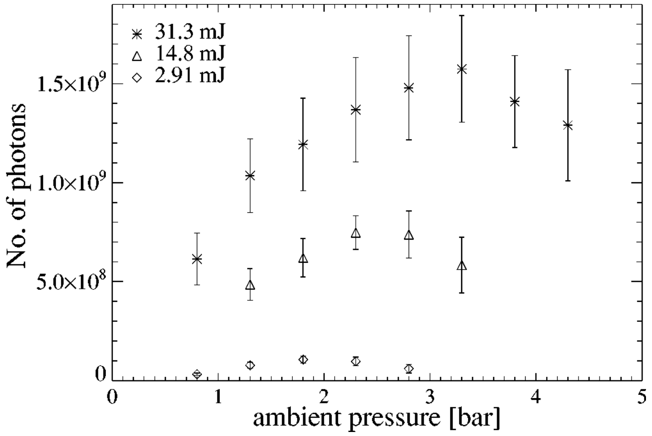

FIG. 6. Luminescence versus ambient pressure for three different laser energies.

sure. The plot demonstrates that for fixed laser energy the number of photons increases when the pressure p is raised from atmospheric pressure to moderately large values. The dependence on p is, however, not monotonic over a larger pressure range. This observation is shown in Fig. 6 where the photon number is given versus pressure p for three selected laser energies E_L. The light output attains a maximum at an optimum pressure p_{opt} that shifts upwards for larger E_L. The SCBL light is brightest at large input energy, e.g., for $E_L = 31.3$ mJ the photon number rises by a factor of ≈ 2.5 at $p_0 \approx 3.3$ bar, compared to the number pertinent to approximately atmospheric pressure. Along with the decline of luminescene above p_{opt} it is observed that the bubbles tend to become unstable and split up upon collapse. Then, in some cases, two consecutive light pulses can be detected that are closely spaced (with separation $\leqslant 20$ ns), and sometimes overlap strongly.

Table I presents the pulse widths and number of photons corresponding to the data of Fig. 6, obtained by deconvolution of the recorded SCBL signals with the impulse response of the instrument. The pulses broaden and get brighter with increasing laser energy E_L. For constant E_L the widths do not depend appreciably on the ambient pressure when $p < p_{\mathrm{opt}}$. Above p_{opt}, the average pulse widths were calculated for the signals that feature a single emission peak only. These pulses consistently have a larger width and smaller average energy.

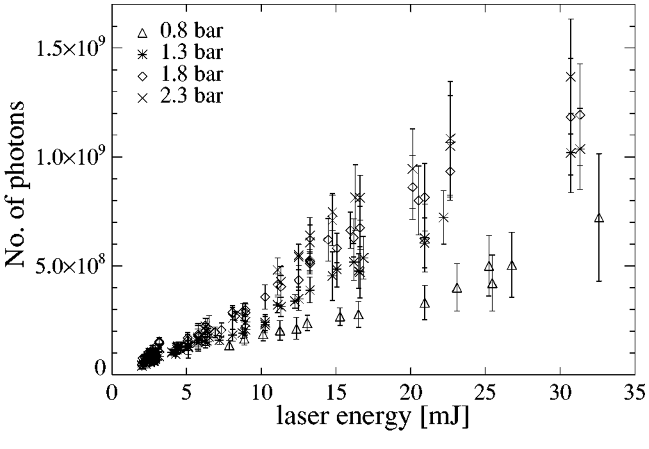

FIG. 5. Number of photons emitted in an SCBL pulse versus energy of the laser pulse generating the bubble for degassed water at the indicated ambient pressures.

The observed increase of luminescence energy with pressure, below the threshold p_{opt}, can be separated into two contributions. First, the mechanical energy stored in the bubble at maximum expansion,

$$E_m = \frac{4}{3}\pi R_{\max}^3 (p - p_v), \tag{2}$$

depends on the laser energy, as shown in Fig. 7. For bubbles of less energy the conversion factor between E_L and E_m appears to be constant, independent of the ambient pressure. However, the linear relation ceases to be valid for large bubbles created with high laser energies at low pressures. For

TABLE I. SCBL pulse characteristics (τ, FWHM pulse width, and N_{ph}, number of photons emitted) for different laser pulse energies E_L and ambient pressures p.

E_L (mJ)	p (bar)	τ (ns)	$N_{ph}/10^8$
2.91	0.8	6.67±0.23	0.33±0.06
	1.3	6.53±0.31	0.91±0.21
	1.8	6.62±0.23	1.07±0.18
	2.3	7.6±0.83	0.85±0.13
14.8	1.3	10.4±1.0	4.85±0.81
	1.8	9.6±0.76	6.19±1.0
	2.3	10.0±0.5	7.46±0.85
	2.8	10.4±1.0	7.37±0.87
	3.3	12.1±2.4	5.82±1.4
31.1	1.3	12.7±1.4	10.4±2.0
	1.8	12.5±1.4	11.7±2.7
	2.3	12.7±1.4	13.8±2.8
	2.8	12.7±1.2	14.6±3.3
	3.3	12.6±0.9	15.6±2.3

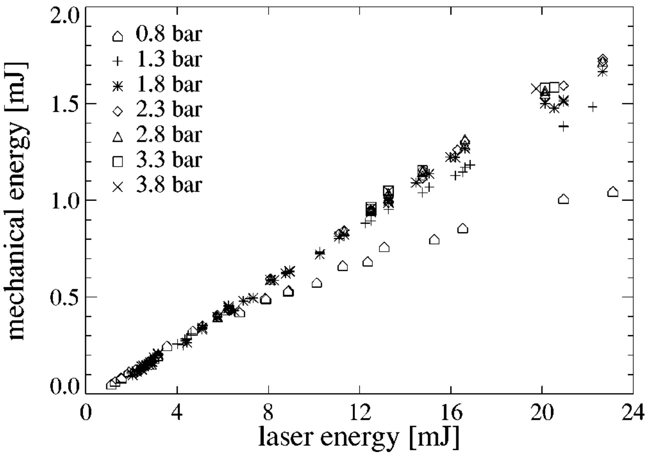

FIG. 7. Mechanical bubble energy versus laser energy for degassed water at different ambient pressures.

example, at $p = 1.3$ bar the deviation starts at $E_L \approx 13$ mJ, corresponding to a maximum bubble radius of $R_{max} = 1.17$ mm (collapse time $T_c = 94.6$ μs). For large E_L the conversion efficiency improves when the ambient pressure is increased, approaching the value encountered with smaller bubbles. As the second contribution, when comparing bubbles of equal mechanical energy, a weak dependence of SCBL output on the ambient pressure can be detected. This feature is demonstrated in Fig. 8.

IV. DISCUSSION

Two counteracting effects have to be considered to interpret our results: energy conversion efficiency and geometric bubble stability. It is well known that a key factor for energy focusing by bubbles is their shape stability during an oscillation cycle. In the case of SBSL the onset of surface instabilities (Rayleigh-Taylor like or parametric) limits the parameter range of maintained bubble luminescence [17]. As shown by molecular dynamics simulations of the collapse of SBSL bubbles [18] the energy focusing within a bubble appears to be surprisingly robust when only small shape pertubations are present. A large shape asymmetry, however, strongly suppresses light emission, as has been demonstrated experimentally for laser generated bubbles collapsing near solid boundaries [6].

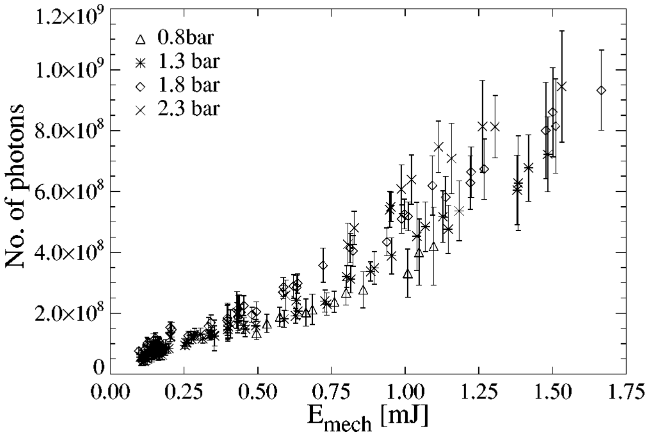

FIG. 8. Light emission versus mechanical bubble energy for different ambient pressures.

In the present experiment the elongated, elliptical or cone-like form of the laser plasma induces a small but detectable shape deformation of the bubble at maximum expansion [19] that cannot be avoided. Under certain conditions, this deformation is amplified sufficiently during the collapse to yield, for example, a dumb-bell-shaped bubble, breaking up into two smaller bubbles collapsing separately [19]. The observed decline in light emission above p_{opt}, with the occasional occurence of two emission peaks, is attributed to this collapse instability.

In MBSL experiments, a pressure dependence of luminescence featuring an emission maximum similar to our results has been observed [20]. A few explanations have been suggested for this phenomenon, considering spherical, single-bubble dynamics, or variation of the number of cavitation nuclei in the liquid. Clearly, the situation is more involved in MBSL than it is in the present experiment due to the presence of many different sized, acoustically excited bubbles that probably interact strongly. Thus, laser-generated bubbles as investigated here provide a convenient experimental model to study certain aspects of the multibubble system.

The ambient pressure dependence of SBSL was investigated in Ref. [14]. A slight decrease of static pressure below the atmospheric pressure results in an increase of light emission (at fixed acoustic excitation strength). However, the influence of ambient pressure on bubble activity in SBSL is more subtle than in our experiment. In SBSL, a stable diffusive equilibrium has to be obtained [21] that is influenced by even slight changes in the bubble dynamics, effected, e.g., by alteration of the ambient pressure. Thus, in addition to the fact that bubble energies are quite different for the two experiments, the findings of Ref. [14] and the present results are not directly comparable.

Our results imply that for the smaller bubbles created at higher pressures the energy conversion from laser energy to mechanical energy, and from mechanical energy to light energy are more efficient than for bubbles created at atmospheric pressure. The dependence shown in Fig. 6, which we consider to be our main result, may be explained qualitatively as follows. Because of the low compressibility of the liquid a change of ambient pressure will not affect the number of water molecules in the focal volume. Thus we can assume that the fraction of energy used to evaporate water during optical breakdown depends on the focal spot size and the laser energy but not on p. Furthermore, there is no evidence that ambient pressure has a significant effect on the relative energy emitted by the initial outgoing shock wave [22]. Merely considering the energy balance of the process we propose that because at higher pressure the bubble size is smaller and the expansion cycle is shorter, and thus surface area and interaction time are diminished, energy loss by thermal conduction and mass flow by nonequlibrium evaporation/condensation at the bubble wall are reduced, bringing about a higher maximum temperature in the bubble.

The present experimental evidence does not permit to assess the change of the maximum temperature attained in the bubble, and of the number of molecules taking part in the emission, with pressure. Further experiments, as well as numerical simulations of the bubble collapse including mass and heat transfer across the bubble wall, surface stability, and also chemical reactions in the bubble, are required to explain the reported results in more detail.

V. CONCLUSION

It has been shown that SCBL light emission can be increased substantially by using high-power laser pulses at elevated pressures, having a peak at a size-dependent optimum pressure p_{opt} beyond which the bubbles tend to become unstable and split. Further enhancement of bubble luminescence seems possible at still higher pressures, utilizing aberration-minimized focusing optics with large numerical aperture to create highly spherical bubbles.

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