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Parametric generation of multimegahertz acoustic oscillations in laser-generated multibubble system in bulk water

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Using a nanosecond CO₂ laser for explosive surface boiling of bulk water, oscillatory acoustic transients from steam bubbles were recorded using a contact photoacoustic technique. Multiple well-resolved, high-amplitude multimegahertz spectral features reflecting parametric interactions between oscillations of cavitating steam bubbles were revealed in the fast Fourier transformation spectra of these transients. A potential parametric generation mechanism for these oscillation modes of steam bubbles is discussed. © 2006 American Institute of Physics. [DOI: 10.1063/1.2206094]

Mechanical (cavitation damage),¹ optical (sonoluminescence),² chemical (sonochemistry³), and even nuclear sonofusion⁴ effects of single- and multibubble systems have been extensively studied during recent decades. Because of the highly nonlinear character of bubble dynamics,⁵ most of these studies have been focused on the much simpler single-bubble systems to study their cavitation dynamics—growth, collapse, and rebound⁶ accompanied by sonoluminescence,⁷ or interactions with sound or ultrasound⁸ using visible (multimicron or larger) single bubbles tracked by means of various optical and acoustic techniques.⁹ However, many realistic systems contain multiple gas or vapor bubbles and their cavitation dynamics may exhibit collective behavior due to interbubble interactions^{9,10} strongly enhancing the mechanical, optical, and chemical effects of a single bubble.¹⁰ Dynamics of multiple interacting bubbles were the subject of numerous theoretical studies,^{5,10} but are not yet well understood. Moreover, experimental studies of multibubble systems are very laborious or even not possible by means of common optical techniques,⁹ while more informative acoustic techniques require use of state-of-the-art acoustic transducers. Therefore, an experimental insight into multibubble dynamics provided by unique experimental tools is very important for continuous progress in this research field. In this letter we report using a contact broadband photoacoustic (PA) technique to study parametric generation of multimegahertz acoustic oscillations in a system of multiple cavitating steam bubbles produced on a free surface of bulk water by nanosecond CO₂ laser heating above its explosive boiling threshold.

A 10.6 μm, transversely excited atmospheric (TEA) CO₂ laser beam [TEM₀₀, 100 mJ/pulse, 100 ns full width at half maximum (FWHM) first peak with a 0.7 μs tail, 1 Hz repetition rate] was focused by a ZnSe spherical lens ($f=10$ cm, Gaussian focal spot radius $\sigma_{1/e} \approx 0.2$ mm) at normal incidence onto a free surface of bulk de-ionized water in a container made of a plastic tube of a height $H \approx 8$ mm. Laser energy was varied using a number of clear polyethylene plastic sheets (20% attenuation per piece) and was measured in each pulse by splitting off a part of the beam to a pyroelectric detector with digital readout (Gentec ED-500). The front 5 mm thick protective quartz window of a fast

acoustic transducer (LiNbO₃ piezoelement, nearly flat response in the 1–100 MHz range, manufactured in the Laboratory of Laser Photoacoustics at Moscow State University) served as the bottom of the container. The relatively small laser spot on the water surface provided PA measurements in the acoustic far field that were distorted by diffraction [the diffraction parameter $H/L_D \sim 1$, where $L_D = \pi f \sigma_{1/e}^2 / C_l$ is the diffraction length¹¹ for acoustic pulses with characteristic frequencies $f=1-10$ MHz and the speed of sound in water $C_l \approx 1.4$ km/s¹² (Ref. 12)], resulting in differential shapes of recorded transients.¹¹ A LeCroy storage oscilloscope (Wavepro 940) was used to record the acoustic transients (delayed by 8 μs needed for sound to propagate in water and the protective window) from the transducer. PA measurements were performed in the laser fluence range $F=0.8-11$ J/cm² and, in good agreement with literature data,¹³ the near-critical explosive boiling threshold of water was found to be equal to $F_B=1.7$ J/cm² where visible expulsion of water jet starts to occur.

A typical PA transient, representing a temporal profile of acoustic pressure $P(t)$ under explosive boiling conditions in water at $F=3.5$ J/cm² $> F_B$, is shown in Fig. 1. In the time frame $t=8-8.2$ μs this transient exhibits a main bipolar pulse with a FWHM parameter equal to that of the main peak

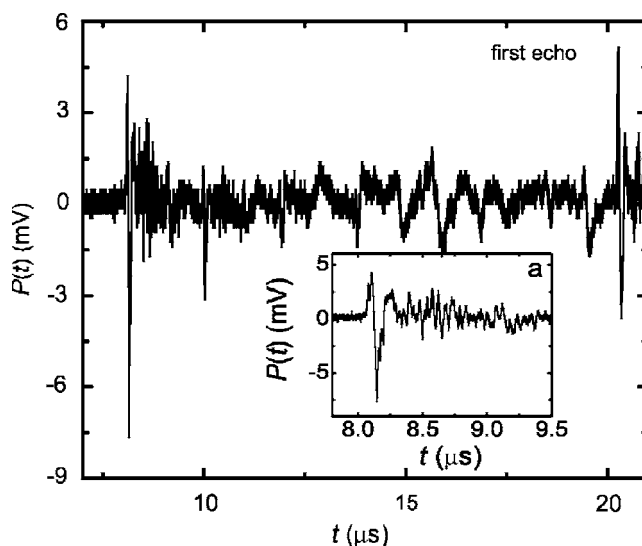


FIG. 1. Acoustic wave form recorded in water at $F \approx 3.5$ J/cm². Inset shows the same wave form on the time scale of 8.0–9.5 μs.

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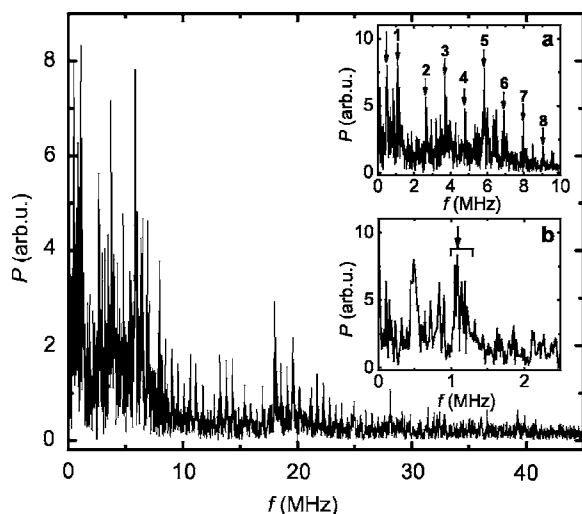


FIG. 2. Amplitude FFT spectrum of the acoustic wave form taken in the time interval of 8–20 μ s. Insets exhibit fragments of the spectrum in the frequency ranges of 0–10 (a) and 0–2.5 (b) MHz. Short arrows and numbers in the inset (a) indicate spectral lines at f_i , while the long arrow shows the position of the subharmonic mode at f_0 . The arrow in the inset (b) shows the set of steam bubble oscillations at $f_{1,k}$.

of the laser pulse. This pulse is generated in a nonlinear thermoacoustic generation regime via the “thermal nonlinearity” mechanism,¹¹ corresponding to near-critical conditions for superheated water. The actual wave form with a predominant compression phase (positive phase in the inset of Fig. 1) characteristic of explosive boiling¹³ transforms to a bipolar one with predominant tensile phase (negative phase in the inset of Fig. 3) due to the diffraction effect in the acoustic far field¹¹ where data acquisition was performed. Accompanying the intense bipolar pulse in Fig. 1 are lower-amplitude oscillations which occur at different frequencies f decreasing as a function of time: $f \approx 15$ –30 MHz at $t = 8.2$ –9.4 μ s, $f \approx 5$ –15 MHz at $t = 9.5$ –11 μ s, and for $t > 11$ μ s there are very pronounced 1 and 2 MHz oscillations.¹⁴ At later times, the first echo in the water layer can be seen at $t \approx 20$ μ s, while the next two appear near 32 and 44 μ s, respectively. Note that none of the main bipolar, oscillatory, and echo signals was observed without the laser pulse heating the water surface or with the acoustic transducer not in contact with the water container.

The pressure oscillations on the time scale $t = 8.2$ –20 μ s can be interpreted as cavitation dynamics of steam bubbles including the characteristic stages of bubble growth [positive $P(t)$], shrinkage and collapse [negative

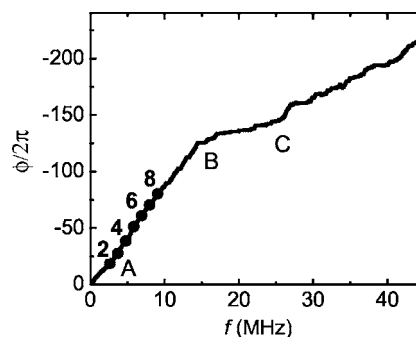


FIG. 3. Phase FFT spectrum of the acoustic wave form taken in the time interval of 8–20 μ s. Regions A–B and B–C show positions of “idler” and “pump” modes, respectively. The idler modes given in Table I are shown by the numbered large dark circles.

$P(t)$], and rebound [next positive stage of $P(t)$].^{1,6} These bubbles are formed, following CO₂-laser ablation of the top water surface layer in the form of a vapor-droplet mixture¹⁵ under explosive boiling conditions, in the underlying layer that is not superheated enough for ablative removal, rather than at the periphery of the laser spot where similar sub-threshold energy deposition into water is provided for smaller $F < F_B$. Indeed, formation of such bubbles on this time scale in the center of the laser spot for $F < F_B$ has not been observed in our experiments.

More detailed information about the origin and dynamics of steam bubbles was obtained by performing a fast Fourier transformation (FFT) of the wave form in the time interval $t = 8.2$ –20 μ s, where the signal is not perturbed by the acoustic echo in the water layer. An amplitude FFT spectrum shows a number of distinct spectral features in the frequency range $f \approx 0$ –30 MHz (Fig. 2), while their initial *negative* phases ϕ_i are presented in the corresponding conjugate FFT phase spectrum (Fig. 3). The most pronounced spectral features represented by a set of frequencies $\{f_i, i = 1, 2, 3, \dots\}$ are separated by frequency intervals of about $f_1 \approx 1.07$ MHz for $f_i \approx if_1$ [see Table I and the inset (a) of Fig. 2], while for other features frequency intervals are not so regular or easily classified. Although the abovementioned set of frequencies $\{f_i\}$ appears to be a number of higher harmonics of the vibration at f_1 , the corresponding set of phases $\{\phi_i\}$ exhibits a pronounced negative linear chirp at $f_i \leq 15$ MHz for $\phi_i \approx i\phi_1$ (Fig. 3, region A–B) and very close ($\pm 5\%$) or shifted by $2\pi m$ ($m = 1, 2, 3, \dots$) values in the range of 15–30 MHz (Fig. 3, region B–C). This fact may indicate that bubble oscillations are starting at higher frequencies

TABLE I. Experimental parameters of “idler” and “signal” oscillation modes of steam bubbles.

No. of peak	Frequency f_j (MHz)	$\Delta f_j = f_j - f_{j-1}$ (MHz)	Normalized phase $\varphi_j = \phi_j / 2\pi$	$\Delta \varphi_j = \varphi_j - \varphi_{j-1}$
0	0.5	...	-4.2	...
1	1.08	0.58	-9.1	-4.9
2	2.62	1.54	-18.8	-9.7
3	3.69	1.07	-27.7	-8.9
4	4.76	1.07	-38.5	-10.8
5	5.83	1.06	-51.1	-12.8
6	6.91	1.08	-60.8	-9.8
7	7.98	1.07	-70.4	-9.8
8	9.05	1.07	-80.2	-9.8

$f_i \approx 15\text{--}30$ MHz and then eventually are converting to slower and slower oscillations. Therefore, one can interpret oscillations in the frequency range of 1–15 MHz (region A-B in Fig. 3) as bubble oscillation modes excited at later times ($t > 8.2 \mu\text{s}$) via a parametric generation mechanism $f_i \rightarrow f_{i-1} + f_1$ with the 15–30 MHz modes as “pump” oscillations, where frequencies f_{i-1} and f_1 correspond to resulting “idler” and “signal” oscillations,¹⁶ respectively.

Regarding the origin of these pump oscillations, from the FWHM of the tensile pulse (the negative phase of the intense first bipolar pulse in Fig. 1 at $t = 8.0\text{--}8.2 \mu\text{s}$) of about $0.03 \mu\text{s}$ (or even shorter if the far-field diffraction effect increasing its duration and amplitude is accounted for), one can deduce broadband ($f \leq 30$ MHz) excitation of steam bubbles with specific frequencies in the range of 15–30 MHz coherently “blown up” by this tensile pulse. Maximum bubble size R_{max} estimated using the Rayleigh relationship⁹ in the form $fR_{\text{max}} \approx 0.5(P/\rho)^{1/2}$, for $f \sim 10$ MHz, the water density $\rho \approx 1 \times 10^3 \text{ kg/m}^3$ (Ref. 12) and characteristic saturated vapor pressure at a temperature of bubble formation, $P \sim 1$ bar,¹³ is on the order of $1 \mu\text{m}$, being lower than the penetration depth of the $10.6 \mu\text{m}$ laser radiation in water, $\delta \approx 11.5 \mu\text{m}$.¹⁷

Another interesting feature in Fig. 2 is a spectral line at $f_0 \approx 0.5$ MHz which serves as another “signal” oscillation [see less pronounced intermediate spectral features in the inset (a) of Fig. 2]. There has been some controversy^{18,19} about an excitation mechanism for $f_{\text{dr}}/2$ and other subharmonics appearing at different rational frequencies $(m/n)f_{\text{dr}}$, where $m, n = 1, 2, 3, \dots$, and f_{dr} is the multikilohertz frequency of the external piezoelectric sound source driving such oscillations.¹⁸ Although the line at $f_0 \approx 0.5$ MHz appears also to be a subharmonic for the signal oscillation at f_1 for $f_0 \approx f_1/2$ and $\phi_0 \approx \phi_1/2$ (Figs. 2 and 3 and Table I), in accordance with our experimental results it can be considered as a product of parametric generation from higher frequencies via the general scheme $f_i \rightarrow f_i' + f_0$. Actually, both these lines at f_0 and f_1 correspond to sets of a few separate lines $\{f_{0,i}\}$ and $\{f_{1,k}\}$, respectively, clearly demonstrated for $f_{1,k}$ oscillations in the inset (b) in Fig. 2. Accounting for participation of these oscillations in the parametric generation process resulting in slower and slower oscillations of steam bubbles due to their energy dissipation, one may consider the oscillations as carriers of the dissipation process.¹⁴

In conclusion, in these photoacoustic studies of a multi-bubble system in laser-superheated bulk water we have revealed a number of characteristic steam bubble oscillations which can be interpreted as pump, signal and idler acoustic oscillations related to each other via parametric interactions in steam bubbles.

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