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Nanosecond Near-Spinodal Homogeneous Boiling of Water Superheated by Pulsed CO₂/ Laser

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near the liquid-vapor (LV) spinode curve, where incoherent, spontaneous *homogeneous nucleation* (HN) of separate vapor bubbles merges with coherent *spinodal decomposition* (SD) of the corresponding superheated liquid correlated over a long scale [1,2]. Classical HN theory predicts metastability of the bulk superheated liquid with respect to non-interacting finite fluctuations of its local entropy S and volume V values representing bubbles of finite critical and supercritical radii ($r \geq r_{\text{cr}}$) [1,2]. At increasing degrees of superheating, the Gibbs free energy HN barrier $\Delta G(T, P, r)$ for critical bubbles and surface tension of the superheated liquid σ tend simultaneously to zero when approaching its liquid/vapor (LV) spinode curve [1], and so does r_{cr} (in practice, size-dependent $\sigma(r) \approx C_1 - C_2 r \approx C_1$ for $r \rightarrow 0$ and the numerical constants C_1 and C_2 [3], so there should be a small, but finite r_{cr} value even at the LV spinode). At the LV spinode – the ultimate stability limit of the superheated liquid with regard to infinitely small thermal and/or acoustic perturbations $\delta S, \delta V$ – a non-steady state LV transformation known as “spinodal decomposition” occurs, according to the Cahn-Hilliard model [4], as strongly-correlated large-scale fluctuations at characteristic wavelengths $\lambda \rightarrow \infty$ increasing exponentially in time (as compared to small-scale fluctuations) and setting up a large scale spatial periodic structure, while at higher temperatures – beyond the spinode – characteristic λ continuously decreases vs. increasing T to atomic or molecular dimensions in the vapor phase. However, if intense thermal fluctuations are explicitly taken into account in SD theory (*Cook, Langer* [5]), the sharp spinodal curve “washes out” because the probabilities of fluctuations for *all* λ values grow towards their asymptotic equilibrium magnitudes and, thus, HN and SD regimes merge together in the near-spinode transition region characterized by a single finite effective characteristic size of fluctuations. Despite these and other theoretical efforts [1-2,5], such combined dynamics of HN and SD in superheated liquids near their corresponding LV spinodes is not yet well understood. Recently, such initial stages of

fast LV transformation have been studied in superheated liquid argon near its LV spinode using a molecular dynamics (MD) approach [6]. In qualitative agreement with results of HN theory, these studies demonstrate shorter and shorter induction (incubation) times for nucleation of nanometer-sized density fluctuations (sub- or near-critical bubbles) in 9.5-nm MD cells vs. increasing T while approaching picosecond values at temperatures $T \approx 0.9T_{\text{crit}}$. However, since collective (correlation) and other size effects crucial for bubble nucleation in the near-spinodal region have not been included in these small-scale simulations, their important results may require experimental verification that has not been performed to date.

In this paper we report the use of contact broadband photoacoustic (PA) spectroscopy to study sub- μs near-spinodal HN of multiple steam bubbles in a μm -thick layer on a free water surface superheated by a TEA CO_2 laser above its explosive boiling threshold at heating rates $\sim 10^{10}$ K/s. Multi-MHz oscillations of acoustic pressure recorded by a state-of-the art acoustic transducer at different incident laser fluences have revealed a broad weak high-frequency spectral band and a number of strong low-frequency spectral lines interpreted, respectively, as weak random oscillations of short-living nm-sized near-critical bubbles and stimulated synchronous oscillations of μm -sized supercritical steam bubbles and sub- μs coalescence products.

In the PA experiments we used an experimental setup described elsewhere [7]. A 10.6- μm , TEA CO_2 laser beam (Lumonics 100-2, TEM_{00} , 0.1 J/pulse, the initial spike of duration $\tau_1 \approx 70$ ns (FWHM) storing about $\gamma_1 \approx 50\%$ of the pulse energy, the pulse tail of the duration $\tau_2 \approx 0.6 \mu\text{s}$, repetition rate of 1 Hz) was focused by a ZnSe spherical lens (focal distance $L = 10$ 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 cylindrical plastic container (height $H \approx 8$ mm, diameter $D \approx 14$ mm). Laser energy was varied using a number of clear polyethylene sheets (20% attenuation per piece) and measured in

each pulse by splitting off a part of the beam to a pyroelectric detector (Gentec ED-500) with digital readout. The front quartz window (thickness $h \approx 3$ mm) of a fast acoustic transducer (LiNbO₃ piezoelement, flat response in the 1-100 MHz range, manufactured in the Laboratory of Laser Optoacoustics at Moscow State University) served as the bottom of the water container. A LeCroy storage oscilloscope (Wavepro 940) was used to record voltage transients (delayed by the 7-8 μ s needed for the corresponding acoustic pressure transients $p(t)$ to propagate in the water volume and the quartz window) from the transducer. The relatively small laser spot on the water surface provided PA measurements in the acoustic far field resulting in differential shapes of recorded transients strongly distorted by diffraction (the diffraction parameter $(H+h)/L_D(f) \sim 1-10$, where $L_D(f) = \pi f \times \sigma_{1/e}^2 / C_l \sim 1-10$ mm is the diffraction length [8] for acoustic pulses with characteristic frequencies $f = 10-10^2$ MHz and the speeds of sound in water, $C_l \approx 1.4$ km/s [9]). Moreover, according to the values of the non-linear attenuation coefficient for water, $\alpha/f^2 \approx (2.42-2.5) \times 10^{-14}$ s²/m for $f = 7-1.9 \times 10^2$ MHz [9], multi-MHz components of the acoustic transients are not subject to significant attenuation when propagated in the 8-mm thick water volume. Acoustic reverberations in the water volume and acoustic delay line exhibiting periods of 10-11 and 2 μ s, respectively, were not present in fast Fourier transform (FFT) spectra (Origin 7, OriginLab) of the acoustic transients taken for the first μ s after the beginning of the CO₂ laser pulse.

PA studies performed at various incident laser fluences $F = 0.8-11$ J/cm² show characteristic waveforms $p(t)$ consisting of a main pulse ($t = 0-0.15$ μ s) and an oscillatory tail at $t > 0.15$ μ s (Fig.1). The main thermoacoustic (TA) pulse increases slowly for $F \leq F_B = 1.7 \pm 0.3$ J/cm², but then rises rapidly at fluences exceeding this threshold value (Fig.2) (see similar thresholds for similar TEA CO₂-laser temporal pulse shapes in [10]). Moreover, near the threshold the main acoustic pulse transforms from tripolar to bipolar (see transients 1-3 in Fig.1), where both the tri-

polar and bipolar waveforms are first time derivatives of the actual bipolar and unipolar waveforms generated during the laser spike (their FWHM equal that of the laser spike) via the TA or explosive boiling (EB) mechanisms [8,11], respectively, and slightly perturbed by surface vaporization [10] and cavitation in the superheated water. The differential effect results from diffraction of the acoustic transients in the far field [8] where data acquisition was performed, and explains slower increase of the compressive pressure amplitude p_{comp} (positive phases in Fig.1) at higher $F \approx 4\text{-}5 \text{ J/cm}^2$ after its initial rapid rise at $F \geq F_B$ by large increase of the lateral size for the explosively boiling region in the superheated interfacial water layer vs. F . Visible formation of mm-sized surface bubble and expulsion of a water jet accompanying laser ablation for $F > F_B$ strongly supports the EB character of the threshold F_B .

This conclusion is also confirmed by simple energy balance analysis. EB of water occurs during the laser pulse near its LV spinode at positive pressures P ($P_0 = 1\text{atm} < P < P_{\text{cr}}$) and temperatures $T \approx (0.9\text{-}1)T_{\text{cr}} \approx (5.9\text{-}6.5) \times 10^2 \text{ K}$ [1,9] (the critical pressure and temperature of water are $P_{\text{cr}} \approx 22.4 \text{ MPa}$ and $T_{\text{cr}} \approx 647 \text{ K}$ [9], respectively) in the “thermal confinement” regime [6], when the threshold volume energy density $\varepsilon_{\text{th}} \approx (1 - R) \times F_{\text{th}} / \delta \approx 1 \times 10^3 \text{ J/cm}^3$ [1] is supplied by the incident laser fluence $F_{\text{th}} \approx 0.9 \text{ J/cm}^2$ for $R \leq 0.01$ [12] and $\delta \approx 9 \text{ }\mu\text{m}$ [13] (the reflectivity and penetration depth for a flat surface of bulk water at normal incidence and $10.6\text{-}\mu\text{m}$ laser wavelength). For increasing total laser fluence F , the onset of laser-induced EB in water at the instantaneous fluence $F(t) \approx F_{\text{th}}$ can be achieved at an earlier time t during the CO_2 -laser heating pulse and can be resolved using the nanosecond acoustic transducer. For example, for the 50:50 energy content ratio of the CO_2 -laser spike and tail there are two reference total fluence thresholds $F_1 \approx 0.9 \text{ J/cm}^2$ and $F_2 \approx 1.8 \text{ J/cm}^2$ ($F_2 \approx F_1 / \gamma_1 \approx 2F_1$), which should provide EB of water at the end of the tail and right after the spike, respectively. Therefore, at $F_1 \leq F \leq F_2$ one can see the main bipolar TA

pulse accompanied by oscillatory EB (cavitation) signal during the laser pulse tail, while at $F \geq F_2$ the EB effect and, to a minor extent, surface vaporization builds up the main unipolar EB pulse and the accompanying cavitation signal [10]. Note that, according to such energy balance analysis and the experimental data of other studies, surface vaporization of water starts at lower $F \approx 0.3 \text{ J/cm}^2$ [10], corresponding to the normal boiling temperature $T_{\text{boil}} \approx 373 \text{ K}$ at $P = P_0$ [9].

In accordance with the reference thresholds F_1 and F_2 , at $F \approx 0.8 \text{ J/cm}^2 \leq F_1$ only the main symmetric tripolar TA pulse was recorded followed for $t > 0.2 \mu\text{s}$ by a few low-amplitude oscillations at a background level (transient 1 in Fig.3). In contrast, at $F \approx 1.4 \text{ J/cm}^2 > F_1$ accompanying the main asymmetric tripolar pulse is a pronounced oscillatory tail which sets up after the laser spike at $t > 0.4 \mu\text{s}$ (transient 2 in Fig.3) and represents characteristic cavitation dynamics of steam bubbles with resonant frequencies $f \approx 10\text{-}40 \text{ MHz}$ (Fig.4b), which μs -scale dynamics has been studied in our previous work [14]. The amplitude FFT spectra in Fig.4 were obtained for the first $0.7\text{-}\mu\text{s}$ ($t = 0.2\text{-}0.9 \mu\text{s}$) slices of this and other acoustic transients for $F > F_1$. At higher $F \approx 2.1 \text{ J/cm}^2 \geq F_2$ an oscillatory tail starts at the end of the laser spike at $t \geq 0.1 \mu\text{s}$ (transient 3 in Fig.3). Similar oscillations in the range $15\text{-}30 \text{ MHz}$ were earlier detected in water at similar CO_2 -laser fluences [10], but were interpreted as oscillations of bubbles produced around micron-sized solid dust species suspended in water of industrial districts. However, since in this work no visible oscillations were observed in the acoustic transients on sub- μs or μs timescales at $F < F_1$ (Fig.3), such multi-MHz bubbles can only be related to sub- μs explosive HN in superheated interfacial water layers at $F > F_1$. Surprisingly, the spectral amplitudes of bubble modes in Fig.4 do not change significantly at higher $F < 6 \text{ J/cm}^2$ inspite of increasing axial and radial dimensions of the superheated surface water layer, while in agreement with the abovementioned F -independent onset of EB at $F(t) \approx F_{\text{th}}$ and the virtual diffraction-limited increase of p_{comp} in Fig.2 at $F = 1.7\text{-}6$

J/cm^2 at the slowly increasing actual p_{comp} amplitude. Altogether these facts demonstrate that under our experimental conditions the EB process is driven by thermodynamic (e.g., degree of superheating) rather than kinetic (e.g., heating rate) factors and exhibits a *single* threshold F_{th} for different laser fluences $F > F_{\text{th}}$, indicating EB at some superheating limit which could be the LV spinode (see below). Also, the main oscillation modes in Fig.4 show their damping during the laser tail with characteristic times of about $1 \mu\text{s}$ consistent with our previous measurements [14].

More complex, but noisy acoustic transients exhibiting higher-resolution FFT spectra more rich in higher f were obtained at $F \approx 2.1 \text{ J}/\text{cm}^2$ (Fig.5) using thinner water layers ($H \approx 1\text{-}1.5 \text{ mm}$), since, in this case, the diffraction effect for the multi-MHz acoustic waves is considerably weaker. In particular, one finds in Fig.5 that the bubble oscillation mode at $f \approx 32 \text{ MHz}$ has much shorter lifetime ($\approx 150 \text{ ns}$ according to its FWHM parameter $\Gamma \approx 6 \text{ MHz}$) than the other modes in Fig.4, while another mode at $f \approx 63 \text{ MHz}$ exhibits slightly longer lifetime $\approx 250 \text{ ns}$ ($\Gamma \approx 4 \text{ MHz}$ in Fig.5). Potentially, the latter mode could be the second harmonic of the former one with its lifetime longer because of higher thermal agitation. Supercritical steam bubbles representing both of these modes may be the precursors of the larger bubbles coalesced on a ns timescale [15] from the smaller ones and oscillating at lower $f < 30 \text{ MHz}$ as shown in Figs.4 and 5.

Furthermore, FFT spectra in Fig.5 show also a weak diffuse band at $f \approx 70\text{-}250 \text{ MHz}$ with the central frequency $f_{\text{C}} \approx 1.6 \times 10^2 \text{ MHz}$ and FWHM $\Gamma \approx 1 \times 10^2 \text{ MHz}$, which has the average amplitude at the level of RF noise from the CO_2 -laser gas discharge. We believe that this band may represent a multitude of transient near-critical steam bubbles (rather than non-oscillating periodic spinodal structures) which are precursors of the 32- and 63-MHz supercritical bubbles and grow very slowly and randomly for $(dr/dt) = 0$ and $(d^2r/dt^2) = 0$ at $r = r_{\text{cr}}$ [16] driven presumably by a random force $\Psi(T, P, r, t)$ resulting from thermal fluctuations. Indeed, the Gibbs free energy

$\Delta G(T, P, r)$ for HN of bubbles has a maximum at $r \approx r_{\text{cr}}$ and the thermodynamic driving force for bubble growth $(\partial \Delta G / \partial r)_{T, P, r_{\text{cr}}} \approx 0$, defining the critical bubble radius as $r_{\text{cr}} = 2\sigma(P, T) / [P_S(T) - P_0]$ [1,2] where $P_S(T)$ is the saturated vapor pressure at T . Such critical and near-critical bubbles have a chance to make a few random oscillations near $r \approx r_{\text{cr}}$ under the influence of $\Psi(T, P, r, t)$ during their slow evolution in size and frequency domains in the near-critical region, emitting a number of short and low-amplitude acoustic wave packets with random frequencies and phases at strongly broadened linewidths, as seen in Fig.5 from the close correspondence of their f_C and Γ . The number of near-critical bubbles per unit volume and periods of their near-critical oscillations (random acts of growth and shrinkage) are strong (exponential) functions of T [1,2]. Therefore, one can expect to observe such random oscillations of tiny near-critical bubbles in FFT spectra in the form of a very weak and broad band with amplitudes and central frequencies rapidly changing versus F , which seems to be the case in this work.

Since critical bubbles are “bottleneck species” in the HN kinetics exhibiting the minimal possible growth rates [1,2,16], their oscillation frequencies $f \sim f_C$ characterize a bubble nucleation frequency $f_{\text{nuc}}(P, T, r_{\text{cr}})$, the parameter used to define at a given P, T a steady-state HN rate $J(P, T) = N_0(P, T) \times B \times \exp[-\Delta G(P, T, r_{\text{cr}}) / kT]$, where $N_0(P, T) \sim 10^{22} \text{ cm}^{-3}$ is the density of molecules in a liquid and $B \sim 10^{10-12} \text{ Hz}$ is the kinetic pre-factor [1,2]. The product $B \times \exp[-\Delta G(P, T, r_{\text{cr}}) / kT] = f_{\text{nuc}}(P, T, r_{\text{cr}})$ and, as a result, for EB conditions assumed in this work $-T^* \approx 0.9T_{\text{cr}} \approx 5.9 \times 10^2 \text{ K}$, $P_S(T^*) \approx 8.6 \times 10^6 \text{ Pa}$ [9] and $f_{\text{nuc}}(P_S(T^*), T^*, r_{\text{cr}}) \approx f_C \approx 1.6 \times 10^2 \text{ MHz}$ – one finds $\Delta G(P_S(T^*), T^*, r_{\text{cr}}) \approx (4-8) \times 10^{-20} \text{ J}$. Simultaneously, one can write for $\Delta G(P, T, r_{\text{cr}}) = \frac{1}{3} \times (4\pi r_{\text{cr}}^2) \times \sigma(P, T) = (2\pi r_{\text{cr}}^3) / 3 \times [P_S(T) - P_0]$ [1] which gives for the known $\Delta G(P_S(T^*), T^*, r_{\text{cr}})$ and $[P_S(T^*) - P_0]$ the estimate $r_{\text{cr}} \approx 1.5-2 \text{ nm}$ consistent with $r_{\text{cr}} \sim 1-3 \text{ nm}$ in Refs. [2,6]. Moreover, for the known r_{cr} and $\Delta G(P_S(T^*), T^*, r_{\text{cr}})$ one can estimate $\sigma(P_S(T^*), T^*) = 3 \times \Delta G(P_S(T^*), T^*, r_{\text{cr}}) / (4\pi r_{\text{cr}}^2) \approx (4-5) \times 10^{-3} \text{ N/m}$,

which corresponds, according to data for $\sigma(P_S, T_S)$ on the water binode curve [9], to the near-spinodal EB temperature $T^* \approx 6.2 \times 10^2 \text{ K} \approx 0.95T_{\text{cr}}$ in good agreement with the energy balance analysis above and typical spinodal temperatures $T_{\text{spin}}(P>0) > 0.92T_{\text{cr}}$ for real liquids [1].

When these critical steam bubbles grow to supercritical dimensions blown up by $P_S(0.95T_{\text{cr}})$, their maximum diameter D_{max} for maximal $f \approx 32 \text{ MHz}$ can approach $D_{\text{max}}(32 \text{ MHz}) \approx 6 \text{ }\mu\text{m} \leq \delta_{\text{wat}} \approx 9 \text{ }\mu\text{m}$ [13] calculated using the Rayleigh's formula [16] written as $D_{\text{max}} \approx 1.1 \times [P_S(0.9T_{\text{cr}}) \times 1/\rho(0.9T_{\text{cr}})]^{1/2}/f$ and values of the water density $\rho(6.2 \times 10^2 \text{ K}) \approx 0.5 \times 10^3 \text{ kg/m}^3$ [1] and $P_S(6.2 \times 10^2 \text{ K}) \approx 1.5 \times 10^7 \text{ Pa}$ [9]. This result shows single-layer packaging of the supercritical bubbles in the superheated interfacial water layers and explains why acoustic waves emitted by the bubbles can be readily recorded in the normal direction in the acoustic farfield without significant attenuation by other bubbles or loss of coherence of their oscillations in the recorded acoustic signals. As a result, the subsequent nanosecond coalescence of these micron-sized bubbles presumably occurs along the surface of the superheated water layers.

In conclusion, we have demonstrated that transient micron-sized supercritical steam bubbles nucleate and coalesce on a nanosecond timescale in a thin surface layer of free bulk water superheated by the TEA CO_2 laser. We have also revealed some indications of random oscillations of nanometer-sized near-critical steam bubbles growing toward supercritical sizes, and estimated their dimensions, nucleation frequency and work of formation, as well as temperature and surface tension of the superheated water at the explosive boiling threshold. The extracted bubble nucleation parameters and energy balance analysis indicate explosive boiling of the superheated water near its liquid/vapor spinode curve. These results provide new experimental facts about near-spinodal explosive boiling of laser-superheated liquids in the thermal confinement regime.

- [1] V.P. Skripov, E.N. Sinitsyn, P.A. Pavlov, G.V. Ermakov, G.N. Muratov, N.V. Bulanov, and V.G. Baidakov, *Thermophysical Properties of Liquids in the Metastable State* (Gordon and Breach, New York, 1988).
- [2] P.G. Debenedetti, *Metastable Liquids: Concepts and Principles* (Princeton University Press, Princeton, 1996).
- [3] R.C. Tolman, J. Chem. Phys. **17**, 333 (1949).
- [4] J.W. Cahn and J.E. Hillard, J. Chem. Phys. **31**, 688 (1959).
- [5] H.E. Cook, Acta Met. **18**, 297 (1970); J.S. Langer, Ann. Phys. **65**, 53 (1971).
- [6] E. Leveugle, D.S. Ivanov, and L.V. Zhigilei, Appl. Phys. A **79**, 1643 (2004).
- [7] S.I. Kudryashov, K. Lyon, and S.D. Allen, Proc. SPIE **6086**, 60861Y (2006).
- [8] V.E. Gusev and A.A. Karabutov, *Laser optoacoustics* (AIP, New York, 1993).
- [9] I.S. Grigor'ev and E.Z. Meilikhov, *Fizicheskie Velichini* (Energoatomizdat, Moscow, 1991).
- [10] F.V. Bunkin, A.A. Kolomensky, V.G. Mikhailevich, S.M. Nikiforov, and A.M. Rodin, Sov. Phys. Acoust. **32**, 21 (1986); A.F. Vitshas, L.M. Dorozhkin, V.S. Doroshenko, V.V. Korneev, L.P. Menakhin, and A.P. Terentiev, Sov. Phys. Acoust. **34**, 43 (1988).
- [11] S.I. Kudryashov, PhD thesis, Moscow State University, 1999.
- [12] E.D. Palik (ed.), *Handbook of Optical Constants of Solids* (Academic Press, Orlando, 1991).
- [13] R.K. Shori, A.A. Walston, O.M. Stafsudd, D. Fried, and J.T. Walsh, Jr., IEEE J. Sel. Top. Quant. Electron. **7**, 959 (2001).
- [14] S.I. Kudryashov, K. Lyon, and S.D. Allen, Phys. Rev. E (2006, accepted for publication).
- [15] K.F. MacDonald, V.A. Fedotov, S. Pochon, B.F. Soares, N.I. Zheludev, C. Guignard, A. Mihaescu, and P. Besnard, Phys. Rev. E **68**, 027301 (2003).
- [16] S. van Stralen and R. Cole, *Boiling Phenomena: Physicochemical and Engineering Fundamentals and Applications* (Hemisphere Publishing Corp., Washington, 1979).

FIG.1. Fragments ($t = -0.2-0.5 \mu\text{s}$) of acoustic waveforms recorded in water at $F \approx 1.4 \text{ J/cm}^2$ ($< F_B$ (1), $1.7 \text{ J/cm}^2 \leq F_B$ (2) and $2.4 \text{ J/cm}^2 > F_B$ (3). The patterned region shows evolution of the main acoustic signal from the tripolar TA to the bipolar EB one at F close to $F_B \approx F_2$.

FIG.2. Compressive pressure p_{comp} vs. F . The arrow shows the explosive boiling threshold F_B of water.

FIG.3. Oscillatory tails ($t > 0.1 \mu\text{s}$) of acoustic waveforms at $F \approx 0.8 \text{ J/cm}^2 \leq F_1$ (1), $F_1 \leq 1.4 \leq F_2$ (2) and $2.1 \geq F_2$ (3) J/cm^2 .

FIG.4. Amplitude FFT spectra for the time interval $t = 0.2-0.9 \mu\text{s}$ of acoustic waveforms at different F (J/cm^2): $a - 1.4$, $b - 1.7$, $c - 3.5$ and $d - 6.5$.

FIG.5. Acoustic waveform at $F \approx 2.1 \text{ J/cm}^2$ (a) and amplitude FFT spectra of its slices over the intervals $t = 0-0.3 \mu\text{s}$ (b), $0.3-0.5 \mu\text{s}$ (c) and $0.5-0.7 \mu\text{s}$ (d) showing transient abundances of different steam bubbles. The arrows show positions of 32- and 63-MHz modes and the horizontal bracket shows position of the diffuse band (see text for details).

fig.1

fig.2

fig.3

fig.4

fig.5