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Chaotic dynamics and control in a resonance-mediated acoustic droplet vaporization model

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Abstract

Acoustic droplet vaporization (ADV) is increasingly used in biomedical research, yet whether its underlying liquid-droplet dynamics under high driving pressure can become chaotic remains unresolved. Such complex dynamics could impair predictable activation and control of imaging and therapeutic responses. Here, we report, to our knowledge, the first evidence of deterministic chaotic dynamics in a liquid-droplet model of resonance-mediated ADV and demonstrate their control by using a weak second harmonic force. An irregular phase portrait, a broadband spectrum, and a positive Lyapunov exponent indicate chaos, and the bifurcation diagram traces the route from period-doubling to chaos. Furthermore, a second harmonic force with 4% of the primary pressure amplitude is applied to reconstruct a period-eight orbit by controlling chaos. Switching the second harmonic force on and off resolves the approach to recurrence and the control of chaotic motion. These results extend ADV resonance theory from amplification to chaotic dynamics and identify chaos control with little loss of radial excursion. The liquid-state droplet model predicts that chaos control can improve the harmonic ADV activation for nonlinear acoustic applications.

Keywords: Acoustic droplet vaporization, Deterministic chaos, Bifurcation, Chaos control, Lyapunov exponent, Nonlinear acoustics

1 Introduction

Acoustic droplet vaporization (ADV) uses ultrasound to convert liquid droplets into gas bubbles [1], linking an acoustic driving force to a local imaging or therapeutic response. Applications include vascular occlusion [2], enhancement of focused-ultrasound ablation [3], oxygen scavenging [4], triggered delivery [5, 6], and phase-change contrast imaging [7–10]. Reviews describe the opportunities and dynamical complexity of these agents [11–13]. Across these applications, reliable activation requires understanding of how

the driven liquid droplet approaches conversion. The central physical question is therefore not only how strongly the droplet responds, but also whether its motion remains periodic under high driving pressure.

Resonance-based ADV theory explains harmonic amplification but leaves the aperiodic dynamics of the strongly driven liquid response unresolved. Superharmonic focusing can concentrate pressure within a droplet [14], and Lajoinie, Segers, and Versluis [15] showed that liquid-droplet resonance promotes vaporization. Neither amplification nor a bounded radial excursion

establishes that successive cycles are repeatable. If strong driving instead produces deterministic chaos, small differences in the initial liquid state could generate different pre-conversion histories. This possibility introduces a dynamical question alongside the nucleation-site, threshold, and local-mechanics questions addressed by existing measurements [16–18]. We investigate it using the nonlinear radial equation of the resonance droplet model [15], whose acoustic restoring response derives from liquid-sphere theory [19, 20].

Gas-bubble acoustics provides a precedent for chaos, not an answer for the liquid droplet before vaporization. The Rayleigh–Plesset family and its compressible extensions describe bubble motion [21–28]; experiments have observed period doubling and chaos [29–31], and numerical studies have mapped their bifurcation structures [32–37]. However, the liquid-droplet resonance model has a different restoring response and concerns a different physical state [15]. We therefore establish its dynamics by combining phase portraits, Poincaré maps, power spectra, and largest Lyapunov exponents. Together, these diagnostics test both the chaotic dynamics of the droplet motion and its sensitivity to initial state [38, 39].

A second gap of ADV control: can a weak second harmonic force restore periodic liquid motion under high driving pressure? Feedback control [40, 41], weak periodic or parametric forcing [42–45], and harmonic control of acoustic resonances [46] establish that small perturbations can reorganize nonlinear responses. Dual-frequency forcing also enables attractor selection in Duffing and Toda oscillators [47, 48]. Gas-bubble studies have examined dual-frequency thresholds, stability, and bifurcation structure [49–53], including suppression of chaos and the allocation of acoustic power [53]. However, a control possibility in the chaotic liquid-droplet ADV model has not yet been revealed. The objective is to suppress chaos at high primary forcing, rather than attenuation of the radial motion associated with resonance.

In this paper, we will investigate chaotic dynamics and its control in a resonance-mediated ADV model. To our knowledge, this is the first observation of chaotic radial motion in this model class. The bifurcation diagram will be employed to investigate the route of the ADV model from

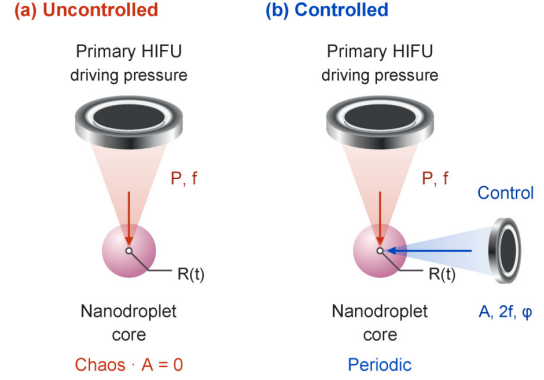


Fig. 1 Schematic of the resonance-mediated ADV model and its harmonic control, showing a liquid droplet of equilibrium radius R_0 driven by the primary pressure P at angular frequency ω together with the second control pressure A at 2ω and relative phase φ

period-doubling bifurcation to chaos. Furthermore, we will control the chaotic motion of the ADV into a period-eight orbit by using the second control pressure with 4% of the primary driving pressure. This extends ADV resonance theory from amplification to chaotic dynamics and motivates tests of stability-aware excitation for phase-change agents.

2 Resonance-based droplet vaporization model

Acoustic droplet vaporization converts a superheated liquid droplet into a gas bubble when an incident ultrasound pulse drives the droplet past its vaporization threshold. In the resonance-mediated regime treated here, the incident wave excites the radial resonance of the liquid droplet, amplifying the pressure within it and initiating the phase change at a lower incident amplitude than off-resonance excitation requires [15]. We retain the liquid-state radial dynamics that precede this conversion and ask how the driving pressure governs their regularity.

The dynamic imposed pressure (Fig. 1) is

$$p_{ac}(t) = P \sin(\omega t) + A \sin(2\omega t + \varphi) \quad (1)$$

With $u = R/R_0$, $\tau = \omega t$, $q = du/d\tau$, and $P^* = \rho_0 R_0^2 \omega^2$, we adopt the free-field droplet dynamics equation developed to describe resonance-mediated ADV initiation [15] and generalize its imposed waveform to two harmonics:

$$u' = q, \quad uq' = -\frac{3q^2}{2} + N \quad (2)$$

$$\begin{aligned} N = & -K \sin(au)(1 - 1/u + dq) \\ & -Vq/u - C(1/u - 1) \\ & -\Pi \sin \tau - \Pi_c \sin(2\tau + \varphi) \end{aligned} \quad (3)$$

Subscripts 0 and 1 denote the exterior liquid and the droplet liquid, respectively, as used in the coefficients below; primes denote derivatives with respect to τ . The coefficients are $a = \omega R_0/c_1$, $d = \omega R_0/c_0$, $K = \rho_1 c_1^2/[P^* j_1(a)]$, $V = 4(\mu_0 - \mu_1)\omega/P^*$, $C = 2\sigma/(R_0 P^*)$, $\Pi = P/P^*$, and $\Pi_c = A/P^*$, where $j_1(a) = (\sin a - a \cos a)/a^2$. Positive imposed pressure is compressive.

For the default parameter values $R_0 = 3.50 \mu\text{m}$, $f = \omega/(2\pi) = 45.4 \text{ MHz}$, and $P = 78 \text{ MPa}$, the primary driving pressure leads the ADV model to produce periodic vibration. Throughout this work each forcing level is reported as the dimensionless ratio $\bar{P} = P/P_0$, where $P_0 = 78 \text{ MPa}$ is the pressure that supports the period-one solution in Fig. 2, so that the reference case is $\bar{P} = 1.000$. This choice reflects the amplitudes available in high-intensity focused ultrasound (HIFU), where ADV is driven at focal pressures of tens of megapascals [3], so that P_0 lies within the range accessible to therapeutic focused sources rather than being an artificially large forcing. The second periodic force has the amplitude of $A = 3.48 \text{ MPa}$, so $\eta = A/(P_0 \bar{P}) = 4\%$, and its relative phase is $\varphi = 25\pi/16 \approx 281^\circ$. Here η is the ratio between the second control pressure and the primary driving pressure. The material constants are the mass densities $\rho_0 = 998$ and $\rho_1 = 1630 \text{ kg m}^{-3}$, the sound speeds $c_0 = 1480$ and $c_1 = 406 \text{ m s}^{-1}$, the dynamic viscosities $\mu_0 = 1.00$ and $\mu_1 = 0.650 \text{ mPa s}$, and the interfacial tension $\sigma = 0.0100 \text{ N m}^{-1}$, with ambient pressure $1.01 \times 10^5 \text{ Pa}$. The ADV model has the initial condition of $(u, q) = (1.07, 0.0198)$. The OFF-ON-OFF calculation simulates the switch process with and without control.

We sample the droplet radial state $z_n = (u, q)$ at $\tau = 2\pi n$. The resulting Poincaré map identifies cycle-to-cycle recurrence. Radius spectra are

computed for $u = R/R_0$ using Welch averaging [54]. Each segment is mean-subtracted and multiplied by a periodic Hanning window. We average seven one-sided periodograms with 50% overlap, normalized by sampling rate and window energy, without peak normalization. For $\nu = \text{frequency}/f$, the density $S_u(\nu)$ has units $(R/R_0)^2$. A spectral peak height is a density, not an oscillation amplitude. To further quantitatively describe the dynamics of the ADV model, we integrate the variational equation and renormalize the tangent vector once per primary cycle [55]. After discarding 8000 transient cycles, let ℓ_j be the logarithmic tangent-growth increment in retained cycle j . The cumulative largest Lyapunov exponent is $\lambda(k) = k^{-1} \sum_{j=1}^k \ell_j$, in inverse primary cycles [55].

3 Existence of chaotic droplet dynamics

Primary driving pressure can produce both periodic motion and deterministic chaos in the liquid-droplet model. We establish this distinction with complementary phase portraits, Poincaré maps, frequency spectra, and largest Lyapunov exponents at four pressures, then use the bifurcation diagram to show how the ADV model evolves from period to chaos with the increase of the driving pressure.

The four driving pressure cases at $\bar{P} = 1.000, 1.026, 1.103$, and 1.128 produce distinct radial trajectories of the ADV model in the phase spaces. At $\bar{P} = 1.000$, the phase portrait forms a compact loop; the higher-pressure responses span larger, folded paths [Fig. 2(a)]. Sampling once per drive cycle isolates their Poincaré section structure in a much smaller coordinate range. Both panels use $u = R/R_0$ and $q = du/d\tau$, but the Poincaré section reveals recurrence rather than a continuously sampled enlargement of the phase portrait. Increasing driving pressure from $\bar{P} = 1.000$ to 1.026 and 1.103 changes the Poincaré section from one state to two and four states, identifying period-one, period-two, and period-four motion [Fig. 2(b)]. At $\bar{P} = 1.128$, extended bands of the chaotic attractor replace the discrete points of periodic motions at lower driving pressures. In addition, the frequency spectra make the corresponding change in harmonic components visible.

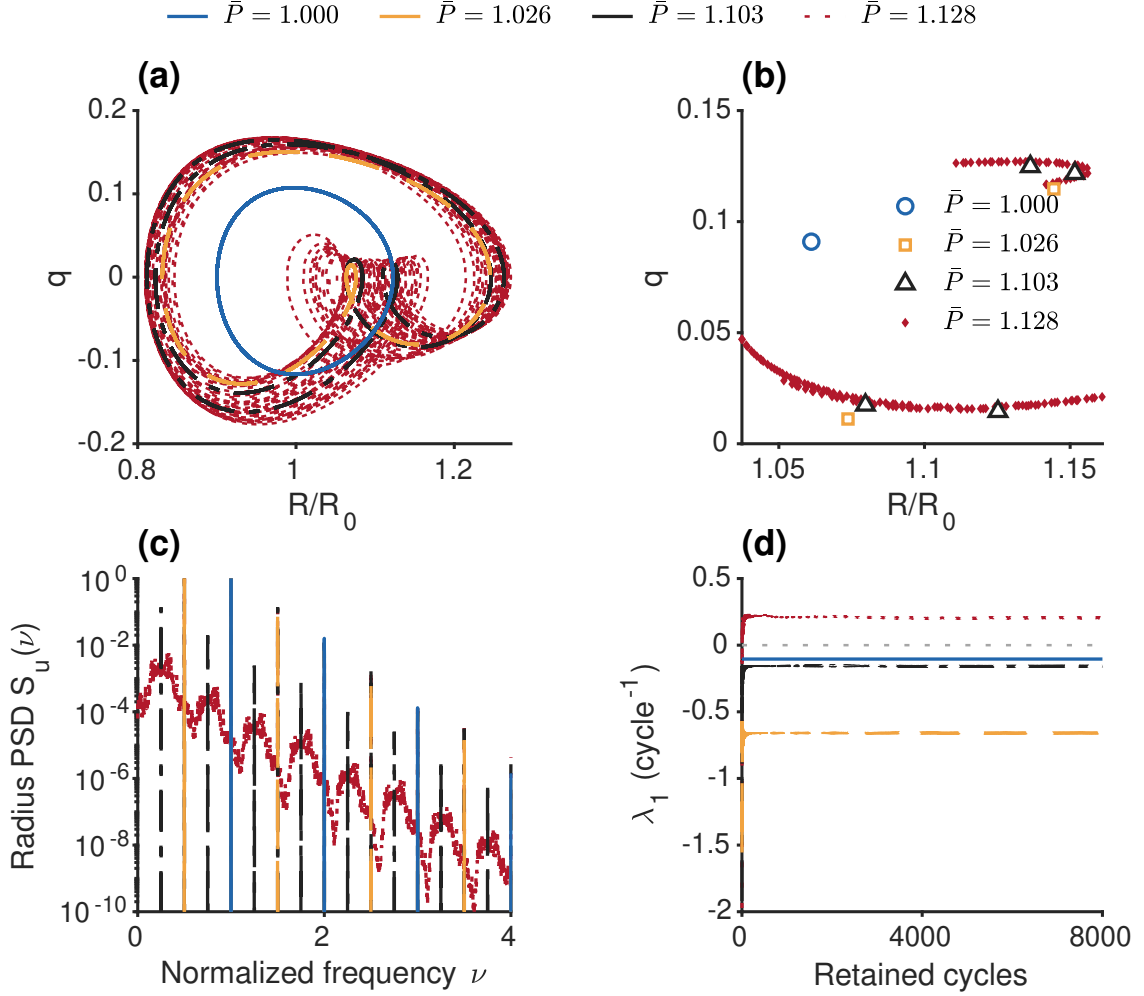


Fig. 2 Primary-only droplet dynamics at $\bar{P} = P/P_0 = 1.000, 1.026, 1.103$, and 1.128 , where $P_0 = 78$ MPa is the pressure supporting the period-one solution; $f = 45.4$ MHz, and $R_0 = 3.50$ μm . Blue, orange, black, and red traces identify the four driving pressures in increasing order. All four pressures appear in every panel. (a) Phase portraits (u, q), with $u = R/R_0$ and $q = du/d\tau$, over the final 128 cycles. (b) Poincaré sections at $\tau = 2\pi n$, containing all 8000 retained states. Circles, squares, triangles, and diamonds correspond to the four pressures. (c) One-sided radius PSD $S_u(\nu)$ per unit normalized frequency $\nu = \text{frequency}/f$, using seven mean-subtracted, periodic Hanning-windowed 1024-cycle segments with 50% overlap from the final 4096 cycles. (d) Cumulative largest Lyapunov exponent λ_1 , in inverse primary cycles, against the number of retained cycles, where the dotted horizontal guide marks zero. The estimates converge to $-0.104, -0.657, -0.155$, and 0.206 , respectively.

The periodic cases have narrow harmonic and sub-harmonic peaks, whereas the $\bar{P} = 1.128$ response has a broadband background between the frequency components [Fig. 2(c)]. With the time evolution, the largest Lyapunov exponent converges to $-0.104, -0.657, -0.155$, and 0.206 per primary cycle at $\bar{P} = 1.000, 1.026, 1.103$, and 1.128 , respectively [Fig. 2(d)]. The negative values of λ support the periodic motions at $\bar{P} = 1.000$,

1.026 , and 1.103 . Positive largest Lyapunov exponent λ confirms the chaotic vibration of the ADV model at $\bar{P} = 1.128$.

With the increase of the primary driving pressure, the bifurcation diagram shows the route from period-doubling to chaos, as shown in Fig. 3(a). Chaos occupies pressure-dependent regions interrupted by periodic windows, rather than a

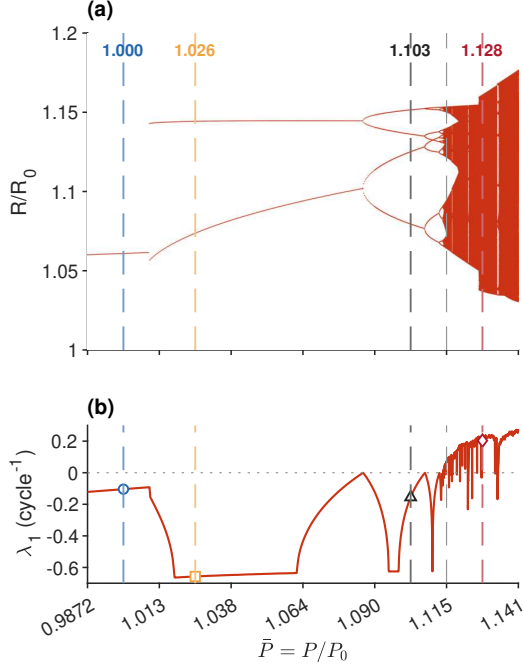


Fig. 3 The effects of primary driving pressure on droplet dynamics, where $f = 45.4$ MHz and $R_0 = 3.50$ μm , and the drive is reported as $\bar{P} = P/P_0$ with $P_0 = 78$ MPa. (a) Bifurcation diagram. (b) Largest Lyapunov exponents. Colored dashed guides identify the Fig. 2 pressures of $\bar{P} = 1.000, 1.026, 1.103$, and 1.128 in blue, orange, charcoal, and red. Gray vertical guides correspond to the chaos control case at $\bar{P} = 1.115$

single isolated operating point. At lower driving pressures, the ADV model produces discrete Poincaré map points, while at higher pressures, extended bands alternate with narrow windows [Fig. 3(a)]. Positive largest exponents distinguish chaotic regions from periodic windows [Fig. 3(b)]. Colored guides locate all four Fig. 2 cases in both panels. A finer grid over $\bar{P} = 1.090$ – 1.141 resolves the region used for the chaos control study. Each pressure starts independently from the same initial condition of the ADV model.

4 Chaos control of droplet vaporization model

Although chaos was established, we test whether the periodic ADV motion can be restored without substantially reducing radial excursion. We add a second control pressure to the ADV model.

4.1 Stabilization and its pressure dependence

The weak 4% pressure can control the chaotic dynamics of the ADV model to produce a period-eight orbit. The nearly overlapping phase portraits show the retained excursion [Fig. 4(a)], but their Poincaré sections reveal sharply different dynamic structures: an extended uncontrolled section and eight discrete controlled states [Fig. 4(b)]. The controlled spectrum has harmonic components, while the uncontrolled spectrum is broadband [Fig. 4(c)]. The largest exponent changes from $+0.0654$ to -0.00593 per primary cycle before and after control [Fig. 4(d)]. Thus weak control pressure reorganizes the chaotic motion into a periodic eight-cycle sequence without requiring a large reduction in its radial envelope.

The bifurcation diagram shows that the control pressure shifts the periodic windows and may suppress chaos. The bifurcation points of period doubling change [Figs. 5(a,b)]. The largest Lyapunov exponents before and after control have opposite signs, indicating the dynamic changes from chaos to period, while at higher pressures, positive exponent regions persist in both cases [Fig. 5(c)]. On the other hand, the periodic motion before control may induce chaos after control. The practical consequence is parameter-specific control. A harmonic control setting that regularizes one pressure need not regularize a neighboring one. Figure 3 supplies the primary-only chaotic dynamic context, and Fig. 5 identifies how the second pressure controls the dynamics.

Varying the control pressure η also controls the dynamics of the ADV model. The bifurcation diagram and largest Lyapunov exponent with respect to η describe different aspects of the controlled response as shown in Fig. 6(a) and (b). Period-doubling bifurcation can be found. For sufficiently small η , chaotic bands with positive largest Lyapunov exponent appear. However, when $\eta > 0.8\%$, periodic motions with negative Lyapunov exponents can be produced by suppressing chaos. The selected $\eta = 4\%$ setting in Fig. 6 lies in the periodic window and gives the period-eight vibration in Fig. 4(b). It is therefore a demonstrated control setting and suggests the control to an ADV model or experiment that includes phase conversion.

We finally show the dynamic change of the ADV model by switching the second pressure ON

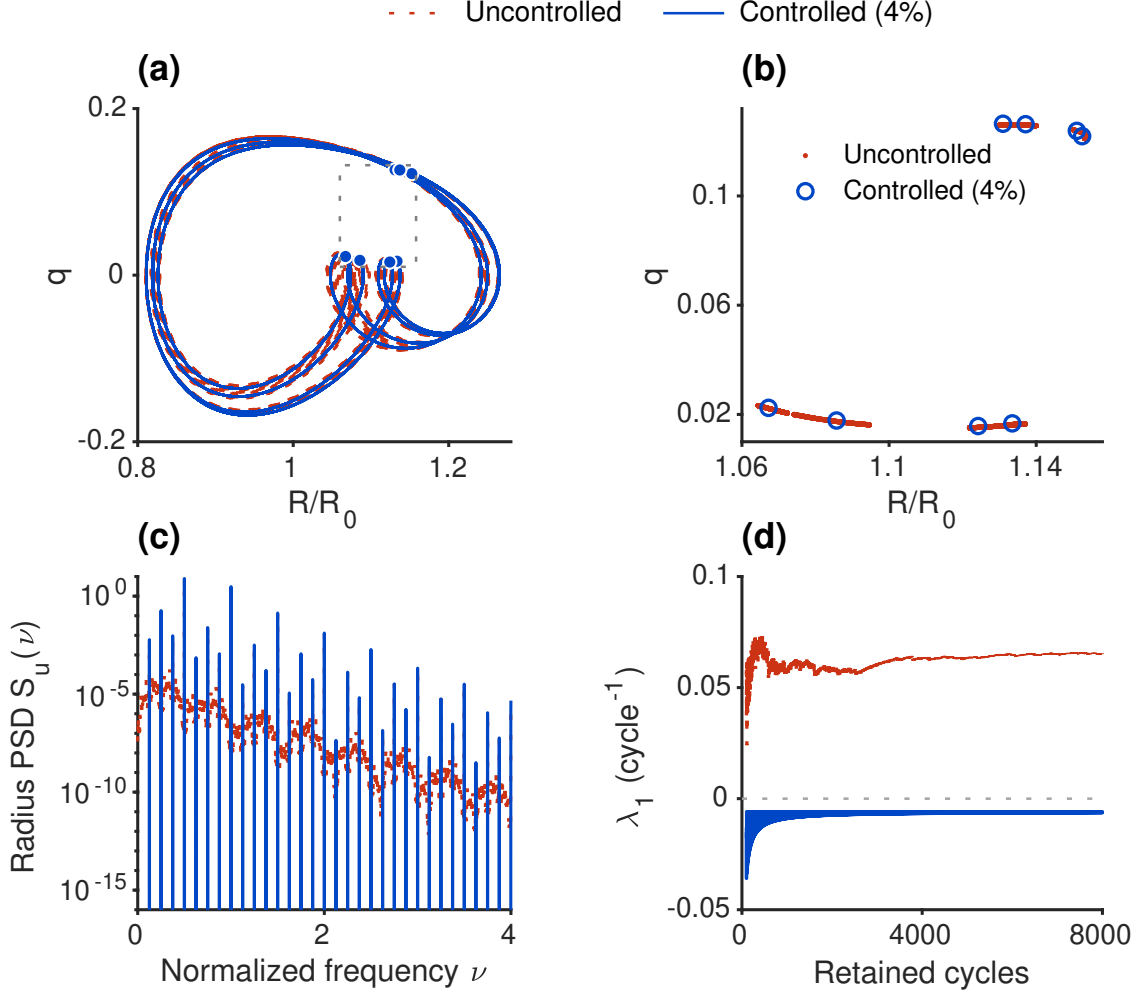


Fig. 4 Chaos control of the ADV model at $\bar{P} = 1.115$, $f = 45.4$ MHz, and $R_0 = 3.50$ μm , where the red dotted curves correspond to chaos before control and blue solid curves correspond to period after control with $\eta = 4\%$ and $\varphi = 25\pi/16 \approx 281^\circ$. (a) Phase portraits. (b) Poincaré sections. (c) One-sided radius PSD per unit $\nu = \text{frequency}/f$. (d) Largest Lyapunov exponents converging to 0.0654 (before control) and -0.00593 (after control) per primary cycle, respectively

and OFF. Figure 7 illustrates the three 8000-cycle intervals: uncontrolled OFF, controlled ON, then uncontrolled OFF. The primary drive remains at $\bar{P} = 1.115$ throughout. At cycle 8000, A is stepped from zero to $\eta = 4\%$ with $\varphi = 25\pi/16$. At cycle 16000, A returns to zero. During OFF, the stroboscopic radius occupies broad bands [Fig. 7(a)], the return error $E_8(n) = \|z_{n+8} - z_n\|_\infty$, with $z_n = (u, q)$, remains at a high value [Fig. 7(b)], and the 512-cycle tangent-growth rate is positive [Fig. 7(c)]. The return error $E_8(n)$ tests recurrence of both radius and velocity rather than radius alone.

During ON, the stroboscopic radius collapses onto the period-eight state of the controlled response. E_8 decreases over several orders of magnitude, and the tangent-growth rate becomes negative.

5 Discussion

This study adds chaotic dynamics to the linearized description of the resonance-mediated ADV model by Lajoie et al. [15]. Acoustic amplification explains how a periodic forcing can produce a

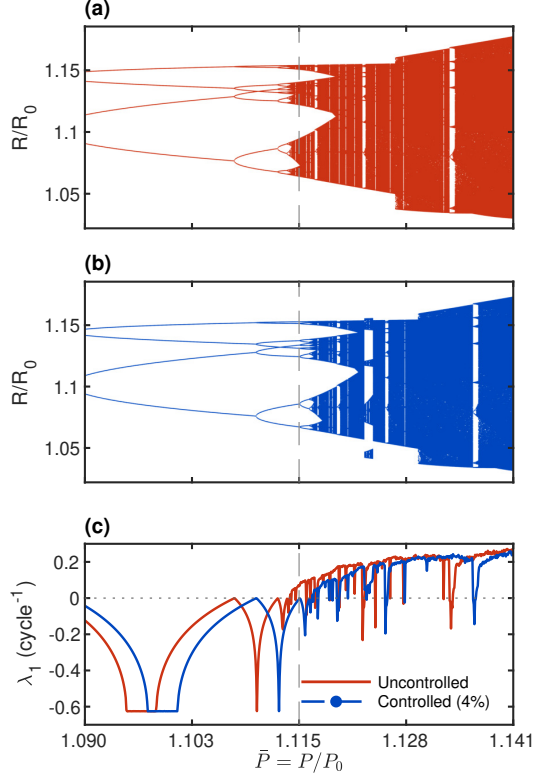


Fig. 5 Dependence of chaos control over the primary pressure $\bar{P} = 1.090\text{--}1.141$, where $f = 45.4$ MHz, $R_0 = 3.50$ μm , $\varphi = 25\pi/16 \approx 281^\circ$, and the vertical dashed line corresponds to $\bar{P} = 1.115$. (a) Bifurcation diagram of the ADV model before control. (b) Bifurcation diagram of the model after control, where the control pressure amplitude is $\eta = 4\%$. (c) Largest Lyapunov exponents before and after control

large liquid-droplet response. Our present calculations show that high driving pressure can induce period-doubling bifurcation and deterministic chaos, which may not be revealed using the linearized ADV theory. The combined diagnostics in Fig. 2 establish these nonlinear behaviors, and Fig. 3 places them among pressure-dependent periodic and chaotic windows. For acoustic excitation design, these results make stability a property to evaluate alongside the strength of the resonant response.

Furthermore, this study shows that removing the controlled pressure restores the chaotic dynamics of the ADV model, while sustaining control maintains the periodic trajectory. A weak control pressure with 4% amplitude replaces chaos

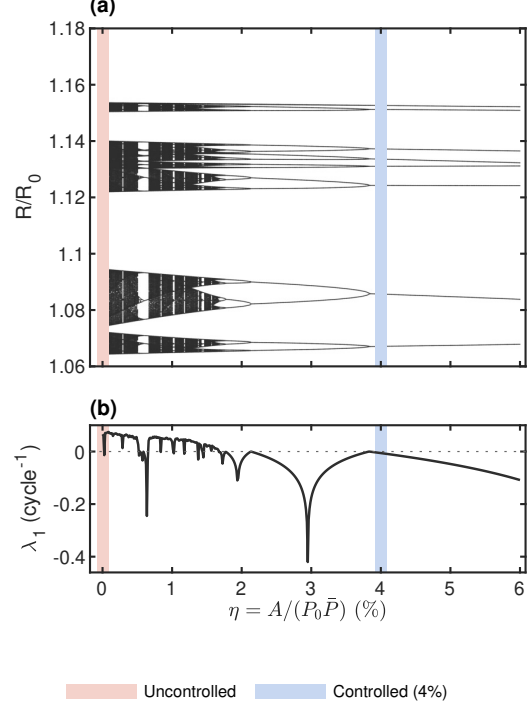


Fig. 6 Dependence of chaos control over the control pressure where $\bar{P} = 1.115$, $f = 45.4$ MHz, $R_0 = 3.50$ μm , and $\varphi = 25\pi/16 \approx 281^\circ$. (a) Bifurcation diagram from $\eta = A/(P_0\bar{P}) = 0$ to 6%. (b) Largest Lyapunov exponents from $\eta = 0$ to 6%, where red and blue stripes correspond to $\eta = 0$ and 4%, respectively

with a period-eight response (Fig. 4). The bifurcation diagram identifies the parameter dependence of that intervention (Figs. 5 and 6). Chaos control is established in other oscillators [42–45, 53]; its role in controlling the liquid-droplet resonance dynamics has not yet been found in ADV. The resulting design principle is to control nonlinear dynamics, not simply to reduce the primary drive for linearization. For ADV therapy, the application opportunity is to control the liquid dynamics that prepares an agent for conversion [1–13]. A chaotic ADV may lead to an unpredictable response even under a prescribed condition. However, chaos control with an additional controlling source offers an effective way to reduce the dynamical sensitivity and improve the predictability while retaining substantial radial motion. The testable link is from liquid-state regularity to activation timing or the fraction of droplets that convert; the present calculations establish the first element, not the conversion outcomes.

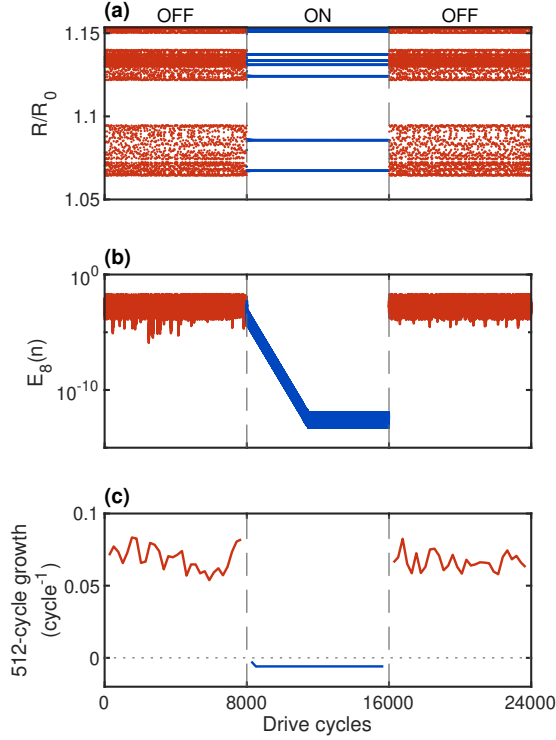


Fig. 7 OFF-ON-OFF control of the ADV model at $\bar{P} = 1.115$, $f = 45.4$ MHz, and $R_0 = 3.50$ μm , where the second control pressure changes from $\eta = 0$ to $\eta = 4\%$ at cycle 8000 and back to zero at cycle 16000. ON and OFF correspond to the controlled and uncontrolled settings. (a) Stroboscopic radius R/R_0 sampled once per drive cycle. (b) Full-state eight-cycle return error $E_8(n) = \|z_{n+8} - z_n\|_\infty$, where $z_n = (R/R_0, q)$. (c) Running 512-cycle tangent-growth rate, in inverse drive cycles, where the dotted horizontal guide marks zero and red and blue denote the uncontrolled OFF and controlled ON intervals

Nanodroplet applications make the available liquid-state interval decisive. High-speed observations of low-boiling-point nanoemulsions show that short acoustic pulses can initiate vaporization and that later cycles can change the newly formed bubbles [56]. A controlled liquid orbit is useful for activation only if the response approaches it before conversion. Figure 7 switches the auxiliary drive while retaining a liquid droplet; its final OFF interval represents renewed chaotic motion, not condensation of a vapor bubble. A direct experimental test would compare primary-only and phase-coherent control pressure exposures while measuring activation timing and the resulting droplet response. Repeated activation introduces

a further opportunity and a different physical process. Perfluorohexane nanodroplets can vaporize repeatedly after recondensation [57], motivating tests of chaos control across successive liquid-state intervals. Extending the control idea to periodic activation therefore requires phase conversion and recondensation in the model.

In addition, translation to nanodroplets also requires size-specific dynamics. The present radius is 3.50 μm . Because $a = \omega R_0/c_1$ couples size and frequency [15], preserving this resonance parameter at a smaller radius requires a frequency change; that change alone does not preserve the viscous and interfacial coefficients in Eqs. (2) and (3). A coated nanodroplet model must additionally include coating mechanics, nucleation, and the evolving liquid-vapor interface. The transferable result is the stability-and-settling framework, not the numerical pressure, frequency, or harmonic ratio used here. Translation to HIFU carries a further requirement specific to the control scheme. The primary amplitudes examined here are within reach of therapeutic focused sources, but chaos control also demands a second harmonic at a few percent of the primary amplitude held at a fixed relative phase throughout the pulse. Delivering that component through an aberrating and attenuating path, and maintaining its phase at the focus, is therefore the principal practical obstacle rather than the primary pressure itself. Two considerations make this pressure range worth studying rather than treating it as an edge case. First, focal amplitudes of this order are exactly where HIFU metrology is now being extended. Traceable calibration has historically stopped at a few megapascals while treatment fields reach the order of 100 MPa, a gap that has been identified as compromising metrological traceability and impeding clinical translation; interferometric realization has since been demonstrated up to 124 MPa peak-to-peak with resolution better than 0.25 MPa [58]. The dynamics reported here therefore fall inside a window that is becoming both experimentally accessible and traceably measurable, which is a precondition for translating any pressure-dependent design rule into the clinic. Second, dual-frequency forcing is an established route to chaos suppression in the closely related problem of acoustically driven gas bubbles, where it remains effective across a wide parameter range even at high acoustic pressure amplitude,

and where the allocation of power between the two components governs the outcome [53]. That a weak second harmonic organizes the liquid-droplet dynamics of ADV in the same way suggests the mechanism is not specific to a single oscillator, and it identifies the primary-to-harmonic power split as the design variable to characterize next.

Chaos control suggests a broader question for nonlinear acoustics: can a weak control tool stabilize a chaotic behavior? Nonlinear nondestructive evaluation, for example, uses harmonic and modulation signatures from imperfect contacts to detect defects [59]. Chaos control while retaining defect-sensitive signatures would extend the present design question to that setting. This prospective analogy concerns excitation design, not a demonstrated defect-detection result.

The numerical findings apply to the spherical liquid-state model. They identify chaotic motion and its control, but do not determine the liquid-state lifetime, ADV threshold, nucleation statistics, or therapeutic response. The next step is to combine size-specific phase-change models with finite-pulse measurements of activation. That comparison would determine whether the demonstrated control of liquid-state regularity can deliver the reproducibility sought in ADV applications.

6 Conclusions

We identify deterministic chaos as a feature of strongly driven liquid-droplet resonance and demonstrate its control by a weak control pressure. To our knowledge, this is the first numerical observation of chaotic dynamics in a liquid-droplet model of resonance-mediated ADV. Periodic motions give way to chaotic response with scattered Poincaré map, broadband spectrum, and positive Lyapunov exponent by sufficiently increasing driving pressure, while the ADV model shows the route from period-doubling bifurcation to chaos. Furthermore, a 4% control pressure stabilizes a chaotic response into a period-eight orbit. The bifurcation diagram locates the control windows, and continuous harmonic switching resolves the development and loss of chaos. Together, these findings extend the nonlinear ADV resonance theory beyond linear amplification: chaotic dynamics

and its control. This study provides a theoretical basis for testing chaotic excitation in phase-change droplet agents, with the application goal of improving activation consistency while retaining strong radial motion.

References

- [1] Kripfgans, O.D., Fowlkes, J.B., Miller, D.L., Eldevik, O.P., Carson, P.L.: Acoustic droplet vaporization for therapeutic and diagnostic applications. *Ultrasound Med. Biol.* 26, 1177–1189 (2000). [https://doi.org/10.1016/S0301-5629\(00\)00262-3](https://doi.org/10.1016/S0301-5629(00)00262-3)
- [2] Kripfgans, O.D., Fowlkes, J.B., Woydt, M., Eldevik, O.P., Carson, P.L.: In vivo droplet vaporization for occlusion therapy and phase aberration correction. *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 49, 726–738 (2002). <https://doi.org/10.1109/TUFFC.2002.1009331>
- [3] Zhang, M., Fabiilli, M.L., Haworth, K.J., Padilla, F., Swanson, S.D., Kripfgans, O.D., Carson, P.L., Fowlkes, J.B.: Acoustic droplet vaporization for enhancement of thermal ablation by high intensity focused ultrasound. *Acad. Radiol.* 18, 1123–1132 (2011). <https://doi.org/10.1016/j.acra.2011.04.012>
- [4] Radhakrishnan, K., Holland, C.K., Haworth, K.J.: Scavenging dissolved oxygen via acoustic droplet vaporization. *Ultrason. Sonochem.* 31, 394–403 (2016). <https://doi.org/10.1016/j.ultrasonch.2016.01.019>
- [5] Lea-Banks, H., O'Reilly, M.A., Hynynen, K.: Ultrasound-responsive droplets for therapy: A review. *J. Controlled Release* 293, 144–154 (2019). <https://doi.org/10.1016/j.jconrel.2018.11.028>
- [6] Xiao, H., Aliabouzar, M., Fabiilli, M.L.: Acoustically responsive scaffolds: Unraveling release kinetics and mechanisms for sustained, steady drug delivery. *J. Controlled Release* 374, 205–218 (2024). <https://doi.org/10.1016/j.jconrel.2024.08.001>
- [7] Sheeran, P.S., Dayton, P.A.: Phase-change contrast agents for imaging and

- pnas.1312171111
- therapy. *Curr. Pharm. Des.* 18, 2152–2165 (2012). <https://doi.org/10.2174/138161212800099883>
- [8] Sheeran, P.S., Streeter, J.E., Mullin, L.B., Matsunaga, T.O., Dayton, P.A.: Toward ultrasound molecular imaging with phase-change contrast agents: An in vitro proof of principle. *Ultrasound Med. Biol.* 39, 893–902 (2013). <https://doi.org/10.1016/j.ultrasmedbio.2012.11.017>
- [9] Rojas, J.D., Dayton, P.A.: Vaporization detection imaging: A technique for imaging low-boiling-point phase-change contrast agents with a high depth of penetration and contrast-to-tissue ratio. *Ultrasound Med. Biol.* 45, 192–207 (2019). <https://doi.org/10.1016/j.ultrasmedbio.2018.08.017>
- [10] Durham, P.G., Dayton, P.A.: Applications of sub-micron low-boiling point phase change contrast agents for ultrasound imaging and therapy. *Curr. Opin. Colloid Interface Sci.* 56, 101498 (2021). <https://doi.org/10.1016/j.cocis.2021.101498>
- [11] Lin, C.-Y., Pitt, W.G.: Acoustic droplet vaporization in biology and medicine. *BioMed Res. Int.* 2013, 404361 (2013). <https://doi.org/10.1155/2013/404361>
- [12] Kee, A.L.Y., Teo, B.M.: Biomedical applications of acoustically responsive phase shift nanodroplets: Current status and future directions. *Ultrason. Sonochem.* 56, 37–45 (2019). <https://doi.org/10.1016/j.ultrasonch.2019.03.024>
- [13] Aliabouzar, M., Kripfgans, O.D., Fowlkes, J.B., Fabiilli, M.L.: Bubble nucleation and dynamics in acoustic droplet vaporization: A review of concepts, applications, and new directions. *Z. Med. Phys.* 33, 387–406 (2023). <https://doi.org/10.1016/j.zemedi.2023.01.004>
- [14] Shpak, O., Verweij, M., Vos, H.J., de Jong, N., Lohse, D., Versluis, M.: Acoustic droplet vaporization is initiated by superharmonic focusing. *Proc. Natl. Acad. Sci. U.S.A.* 111, 1697–1702 (2014). <https://doi.org/10.1073/pnas.1312171111>
- [15] Lajoinie, G., Segers, T., Versluis, M.: High-frequency acoustic droplet vaporization is initiated by resonance. *Phys. Rev. Lett.* 126, 034501 (2021). <https://doi.org/10.1103/PhysRevLett.126.034501>
- [16] Li, D.S., Kripfgans, O.D., Fabiilli, M.L., Fowlkes, J.B., Bull, J.L.: Initial nucleation site formation due to acoustic droplet vaporization. *Appl. Phys. Lett.* 104, 063703 (2014). <https://doi.org/10.1063/1.4864110>
- [17] Giesecke, T., Hynynen, K.: Ultrasound-mediated cavitation thresholds of liquid perfluorocarbon droplets in vitro. *Ultrasound Med. Biol.* 29, 1359–1365 (2003). [https://doi.org/10.1016/S0301-5629\(03\)00980-3](https://doi.org/10.1016/S0301-5629(03)00980-3)
- [18] Aliabouzar, M., Abeid, B.A., Kripfgans, O.D., Fowlkes, J.B., Estrada, J.B., Fabiilli, M.L.: Real-time spatiotemporal characterization of mechanics and sonoporation of acoustic droplet vaporization in acoustically responsive scaffolds. *Appl. Phys. Lett.* 123, 114101 (2023). <https://doi.org/10.1063/5.0159661>
- [19] Anderson, V.C.: Sound scattering from a fluid sphere. *J. Acoust. Soc. Am.* 22, 426–431 (1950). <https://doi.org/10.1121/1.1906621>
- [20] Feuillade, C., Clay, C.S.: Anderson (1950) revisited. *J. Acoust. Soc. Am.* 106, 553–564 (1999). <https://doi.org/10.1121/1.427025>
- [21] Lord Rayleigh: On the pressure developed in a liquid during the collapse of a spherical cavity. *Philos. Mag.* 34, 94–98 (1917). <https://doi.org/10.1080/14786440808635681>
- [22] Plesset, M.S., Prosperetti, A.: Bubble dynamics and cavitation. *Annu. Rev. Fluid Mech.* 9, 145–185 (1977). <https://doi.org/10.1146/annurev.fl.09.010177.001045>
- [23] Prosperetti, A.: Nonlinear oscillations of gas bubbles in liquids: Steady-state solutions. *J. Acoust. Soc. Am.* 56, 878–885 (1974). <https://doi.org/10.1121/1.1903341>

- [24] Keller, J.B., Miksis, M.: Bubble oscillations of large amplitude. *J. Acoust. Soc. Am.* 68, 628–633 (1980). <https://doi.org/10.1121/1.384720>
- [25] Prosperetti, A., Lezzi, A.: Bubble dynamics in a compressible liquid. Part 1. First-order theory. *J. Fluid Mech.* 168, 457–478 (1986). <https://doi.org/10.1017/S0022112086000460>
- [26] Prosperetti, A., Crum, L.A., Commander, K.W.: Nonlinear bubble dynamics. *J. Acoust. Soc. Am.* 83, 502–514 (1988). <https://doi.org/10.1121/1.396145>
- [27] Leighton, T.G.: *The Acoustic Bubble*. Academic Press, London (1994). <https://doi.org/10.1016/B978-0-12-441920-9.X5001-9>
- [28] Lauterborn, W., Kurz, T.: Physics of bubble oscillations. *Rep. Prog. Phys.* 73, 106501 (2010). <https://doi.org/10.1088/0034-4885/73/10/106501>
- [29] Lauterborn, W., Cramer, E.: Subharmonic route to chaos observed in acoustics. *Phys. Rev. Lett.* 47, 1445–1448 (1981). <https://doi.org/10.1103/PhysRevLett.47.1445>
- [30] Lauterborn, W., Koch, A.: Holographic observation of period-doubled and chaotic bubble oscillations in acoustic cavitation. *Phys. Rev. A* 35, 1974–1976 (1987). <https://doi.org/10.1103/PhysRevA.35.1974>
- [31] Lauterborn, W., Parlitz, U.: Methods of chaos physics and their application to acoustics. *J. Acoust. Soc. Am.* 84, 1975–1993 (1988). <https://doi.org/10.1121/1.397042>
- [32] Parlitz, U., Englisch, V., Scheffczyk, C., Lauterborn, W.: Bifurcation structure of bubble oscillators. *J. Acoust. Soc. Am.* 88, 1061–1077 (1990). <https://doi.org/10.1121/1.399855>
- [33] Lauterborn, W., Holzfuss, J.: Acoustic chaos. *Int. J. Bifurcation Chaos* 1, 13–26 (1991). <https://doi.org/10.1142/S0218127491000038>
- [34] Ohl, C.-D., Kurz, T., Geisler, R., Lindau, O., Lauterborn, W.: Bubble dynamics, shock waves and sonoluminescence. *Philos. Trans. R. Soc. London A* 357, 269–294 (1999). <https://doi.org/10.1098/rsta.1999.0327>
- [35] Hilgenfeldt, S., Brenner, M.P., Grossmann, S., Lohse, D.: Analysis of Rayleigh–Plesset dynamics for sonoluminescing bubbles. *J. Fluid Mech.* 365, 171–204 (1998). <https://doi.org/10.1017/S0022112098001207>
- [36] Behnia, S., Jafari, A., Soltanpoor, W., Jahanbakhsh, O.: Nonlinear transitions of a spherical cavitation bubble. *Chaos Solitons Fractals* 41, 818–828 (2009). <https://doi.org/10.1016/j.chaos.2008.04.011>
- [37] Carroll, J.M., Calvisi, M.L., Lauderbaugh, L.K.: Dynamical analysis of the nonlinear response of ultrasound contrast agent microbubbles. *J. Acoust. Soc. Am.* 133, 2641–2649 (2013). <https://doi.org/10.1121/1.4796128>
- [38] Jiang, J.J., Zhang, Y., Stern, J.: Modeling of chaotic vibrations in symmetric vocal folds. *J. Acoust. Soc. Am.* 110, 2120–2128 (2001). <https://doi.org/10.1121/1.1395596>
- [39] Zhang, Y., Jiang, J.J.: Chaotic vibrations of a vocal fold model with a unilateral polyp. *J. Acoust. Soc. Am.* 115, 1266–1269 (2004). <https://doi.org/10.1121/1.1648974>
- [40] Ott, E., Grebogi, C., Yorke, J.A.: Controlling chaos. *Phys. Rev. Lett.* 64, 1196–1199 (1990). <https://doi.org/10.1103/PhysRevLett.64.1196>; erratum, *Phys. Rev. Lett.* 64, 2837 (1990). <https://doi.org/10.1103/PhysRevLett.64.2837>
- [41] Pyragas, K.: Continuous control of chaos by self-controlling feedback. *Phys. Lett. A* 170, 421–428 (1992). [https://doi.org/10.1016/0375-9601\(92\)90745-8](https://doi.org/10.1016/0375-9601(92)90745-8)
- [42] Braiman, Y., Goldhirsch, I.: Taming chaotic dynamics with weak periodic perturbations. *Phys. Rev. Lett.* 66, 2545–2548 (1991). <https://doi.org/10.1103/PhysRevLett.66.2545>
- [43] Lima, R., Pettini, M.: Suppression of chaos by resonant parametric perturbations. *Phys.*

- Rev. A 41, 726–733 (1990). <https://doi.org/10.1103/PhysRevA.41.726>
- [44] Hsu, R.-R., Su, H.-T., Chern, J.-L., Chen, C.-C.: Conditions to control chaotic dynamics by weak periodic perturbation. *Phys. Rev. Lett.* 78, 2936–2939 (1997). <https://doi.org/10.1103/PhysRevLett.78.2936>
- [45] Boccaletti, S., Grebogi, C., Lai, Y.-C., Mancini, H., Maza, D.: The control of chaos: Theory and applications. *Phys. Rep.* 329, 103–197 (2000). [https://doi.org/10.1016/S0370-1573\(99\)00096-4](https://doi.org/10.1016/S0370-1573(99)00096-4)
- [46] Fyrrillas, M., Szeri, A.J.: Control of ultra- and subharmonic resonances. *J. Nonlinear Sci.* 8, 131–159 (1998). <https://doi.org/10.1007/s003329900046>
- [47] Hegedűs, F., Krähling, P., Aron, M., Lauterborn, W., Mettin, R., Parlitz, U.: Feedforward attractor targeting for non-linear oscillators using a dual-frequency driving technique. *Chaos* 30, 073123 (2020). <https://doi.org/10.1063/5.0005424>
- [48] Krähling, P., Steyer, J., Parlitz, U., Hegedűs, F.: Attractor selection in nonlinear oscillators by temporary dual-frequency driving. *Nonlinear Dyn.* 111, 19209–19224 (2023). <https://doi.org/10.1007/s11071-023-08855-3>
- [49] Hegedűs, F., Klapcsik, K., Lauterborn, W., Parlitz, U., Mettin, R.: GPU accelerated study of a dual-frequency driven single bubble in a 6-dimensional parameter space: The active cavitation threshold. *Ultrason. Sonochem.* 67, 105067 (2020). <https://doi.org/10.1016/j.ultsonch.2020.105067>
- [50] Klapcsik, K.: GPU accelerated numerical investigation of the spherical stability of an acoustic cavitation bubble excited by dual-frequency. *Ultrason. Sonochem.* 77, 105684 (2021). <https://doi.org/10.1016/j.ultsonch.2021.105684>
- [51] Klapcsik, K., Varga, R., Hegedűs, F.: Bi-parametric topology of subharmonics of an asymmetric bubble oscillator at high dissipation rate. *Nonlinear Dyn.* 94, 2373–2389 (2018). <https://doi.org/10.1007/s11071-018-4497-2>
- [52] Varga, R., Hegedűs, F.: Classification of the bifurcation structure of a periodically driven gas bubble. *Nonlinear Dyn.* 86, 1239–1248 (2016). <https://doi.org/10.1007/s11071-016-2960-5>
- [53] Zhang, Y., Zhang, Y.: Chaotic oscillations of gas bubbles under dual-frequency acoustic excitation. *Ultrason. Sonochem.* 40, 151–157 (2018). <https://doi.org/10.1016/j.ultsonch.2017.03.058>
- [54] Welch, P.: The use of fast Fourier transform for the estimation of power spectra: A method based on time averaging over short, modified periodograms. *IEEE Trans. Audio Electroacoust.* 15, 70–73 (1967). <https://doi.org/10.1109/TAU.1967.1161901>
- [55] Skokos, Ch.: The Lyapunov characteristic exponents and their computation. In: Souchay, J., Dvorak, R. (eds.) *Dynamics of Small Solar System Bodies and Exoplanets. Lecture Notes in Physics*, vol. 790, pp. 63–135. Springer, Berlin (2010). https://doi.org/10.1007/978-3-642-04458-8_2
- [56] Sheeran, P.S., Matsunaga, T.O., Dayton, P.A.: Phase-transition thresholds and vaporization phenomena for ultrasound phase-change nanoemulsions assessed via high speed optical microscopy. *Phys. Med. Biol.* 58, 4513–4534 (2013). <https://doi.org/10.1088/0031-9155/58/13/4513>
- [57] Van Namen, A., Jandhyala, S., Jordan, T., Luke, G.P.: Repeated acoustic vaporization of perfluorohexane nanodroplets for contrast-enhanced ultrasound imaging. *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 68, 3497–3506 (2021). <https://doi.org/10.1109/TUFFC.2021.3093828>
- [58] Wang, K., Xing, G., Song, D., Wilkens, V., Yang, P.: High-intensity focused ultrasound sound pressure realization up to 100 MPa based on a differential high bandwidth homodyne laser interferometer. *Opt. Express* 34, 15110 (2026). <https://doi.org/10.1364/OE.2026.15110>

- [59] Donskoy, D., Sutin, A., Ekimov, A.: Nonlinear acoustic interaction on contact interfaces and its use for nondestructive testing. *NDT & E Int.* 34, 231–238 (2001). [https://doi.org/10.1016/S0963-8695\(00\)00063-3](https://doi.org/10.1016/S0963-8695(00)00063-3)

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Data availability. The data supporting the findings of this study are not publicly available but are available from the authors upon reasonable request.