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Robust acoustic wave manipulation of bubbly liquids

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Experiments with water–air bubbly liquids when exposed to acoustic fields of frequency ~ 100 kHz and intensity below the cavitation threshold demonstrate that bubbles ~ 30 μm in diameter can be “pushed” away from acoustic sources by acoustic radiation independently from the direction of gravity. This manifests formation and propagation of acoustically induced transparency waves (waves of the bubble volume fraction). In fact, this is a collective effect of bubbles, which can be described by a mathematical model of bubble self-organization in acoustic fields that matches well with our experiments. © 2016 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4944893>]

The radiation force acting on the interface of two media with different properties has been the subject of many theoretical discussions beginning from Rayleigh^{1,2} and Brillouin.³ This is a phenomenon common for many kinds of wave motion including electromagnetism and acoustics,^{4–7} and therefore, it is interesting from a basic science perspective and of great practical importance. Particularly, the acoustic radiation force can be used to deform soft tissues in some bio-medical applications⁸ and move nanoparticles on liquid drop surfaces.²⁸ As we report in the present letter, this force can also be used to move or manipulate some media, such as bubbly liquids. Current applications are not aware on a simple way to control the bubble distribution besides the use of gravity. In the present work, we demonstrate organization of bubbly liquids in mild acoustic fields, i.e., in the absence of cavitation.

Our experimental observations of water–air bubbly liquids exposed to acoustic fields of frequency ~ 100 kHz and intensity below the cavitation threshold demonstrate that bubbles can be “pushed” away from acoustic sources independently from the direction of gravity. This forms a region of the liquid almost free from bubbles. The boundary of this region propagates towards the bulk of the bubbly liquid. This can also be interpreted as formation and propagation of acoustically induced transparency waves, or waves of the bubble volume fraction. The effect is very strong and repeatable, which suggests that an array of acoustic transducers can be used for robust manipulation with the cloud of bubbles. It is noticeable that the observed effect cannot be explained by well-known theories of a single bubble drifting in acoustic waves. This is a collective effect of bubbles, which, as we show, can be described by a simplified mathematical model of bubble organization in acoustic fields.

The behavior of bubbles in the prescribed acoustic fields as well as propagation of acoustic waves in a bubbly medium is well studied experimentally and numerically.^{9–13,24} Such studies can be referenced as cases of one-way field–particle interaction, because neither bubbles modify the field nor the field modifies the medium. Two-way field–particle interaction occurs due to nonlinearities, which cause (a) a relatively slow motion of bubbles (“drift”) driven by acoustic radiation forces and (b) a change of the bubble sizes due to rectified diffusion.^{14–19} Such slow time scale motion modifies acoustic properties of the medium and therefore affects the spatial distribution of the acoustic pressure. The phenomenon of the two-way field–particle interaction can also be classified as a bubble self-organization effect in acoustic fields or self-action of the acoustic waves. The manifestation of the two-way interaction was observed experimentally in acoustic cavitation²⁰ (“structure formation”) and modeled numerically.^{21–23}

The experimental setup for the present study includes a home-made transparent acrylic cuvette with the inner dimensions of $30 \times 30 \times 30$ mm³ and the wall thickness of 5 mm shown in Fig. 1. A piezoelectric ceramic disc transducer (PZT) (STEMiNC) with the resonance frequency of 1 MHz (we used lower frequency radial resonances of the piezo) is glued to the bottom from the outside of the cuvette. A sinusoidal voltage is applied to the piezotransducer through a waveform generator (WFG 33522A, Agilent Technologies) and a RF amplifier (AG 1012, T&C Power Conversion). A needle hydrophone (HNR-1000, Onda) and a storage oscilloscope (HRO 66Zi, LeCroy) are used to measure the pressure in the liquid filled tank. The bubble dynamics within the tank is recorded with a high-speed camera (FASTCAM SA5, Photron) with a resolution 1024×1024 pixels and at a frame rate of 2000 fps. The camera is equipped with a macro objective lens (Micro-Nikkor 60 mm f/2.8D, Nikon) that leads to a resolution of approximately 30 μm per pixel. To observe individual bubbles, the camera was attached to the long-distance

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FIG. 1. Experimental setup: (a) for observation of the transparency waves for the transducer placed on the bottom of the cell; (b) for measurements of the acoustic field in pure liquid.

microscope (K2/SC, Infinity Photo-Optical Company) resulting in an increased resolution of $\sim 3 \mu\text{m}$ per pixel.

For all experiments, deionized water is used (prepared from an ultrapure water purification system Milli-Q Advantage A10, EMD Millipore). The air bubbles in water with a mean radius of $16 \mu\text{m}$ and a volume fraction of 0.3%–0.5% are generated using a homemade Venturi tube placed in a separate 500 ml reservoir. A typical bubble size distribution obtained using image processing is shown in Fig. 2. The bubble contours were detected and fitted by ellipses. The eccentricity is used to distinguish between single and multiple bubbles. The volume fraction was estimated using models of a bubble in a spherical and in a cubic cell. To avoid fast coalescence of the bubbles, a salt (NaCl, concentration $c = 67 \text{ g/l}$) was added to water.

The turned on sound field results in the initially homogeneous bubbly liquid to quickly change with a remarkable redistribution of the bubble density. Initially, a reduction of the bubble density appears at the bottom of the tank near the transducer. A thin layer separates this region with the high density of bubbles (front) from the homogeneous bubbly liquid in the upper part of the tank. Further, the front was moving up to the top level of the liquid (see Fig. 3(a)) (Multimedia view) and 3(c)) (Multimedia view)). Figures 3(b) (Multimedia view) and 3(d) (Multimedia view) show the front motion for different transducer locations. The shape of the front can be explained by the fact that the amplitude

FIG. 2. Bubble radius distribution measured in the experiments.

of the field near the center of the transducer is stronger than at its periphery. The front velocity rises at the increasing waveform generator signal amplitude. Besides overall vertical motion upward, some bubbles move down picking up the neighboring bubbles, which leads to a cluster formation. The clusters move up and merge with the bubble front.

In theory, a gas bubble can be considered as a nonlinear oscillator. Being placed in a small amplitude time harmonic acoustic field of a circular frequency ω and a wavelength λ , a spherical gas bubble of radius $a \ll \lambda$ experiences small radial oscillations near the period-averaged radius a_0

$$\begin{aligned} a &= a_0 \left(1 + \varepsilon \text{Re} \left\{ -\Lambda(a_0) A e^{-i\omega t} \right\} + \dots \right), \\ p &= p_* (1 + \varepsilon \text{Re} \{ A e^{-i\omega t} \}), \\ \Lambda(a_0) &= \frac{a_0^2}{a_r^2 - a_0^2 - i\eta}, \quad a_r^2 = \frac{3\gamma p_*}{\omega^2 \rho_l}, \end{aligned} \quad (1)$$

where p and p_* are the total and static pressures, ρ_l is the liquid density, Λ is the bubble response function, a_r is the resonance radius, while γ and $\eta > 0$ are the effective adiabatic exponent and the dumping coefficient, which includes effects of heat transfer in gas, liquid viscosity, and acoustic radiation.^{10,11} The dimensionless phasor of the acoustic field here is presented in the form εA to have $|A| \sim 1$ and constant ε characterizing the magnitude of the field, which is convenient for estimations of the magnitude of nonlinear effects of sound.

In case of $a \ll \lambda$, the acoustic field in a bubbly liquid can be modeled using the multiple scattering approach.²⁵ The total field in this case is a sum of the incident and scattered fields, $A = A^{in} + A^{sc}$, where the field scattered by N bubbles of radii a_{0j} located at $\mathbf{r}_{0j}$, $j = 1, \dots, N$ is a sum of monopoles due to bubble volume oscillations, Eq. (1)

$$\begin{aligned} A^{sc}(\mathbf{r}) &= \sum_{j=1}^N s_j A(\mathbf{r}_{0j}) \frac{e^{ik_l |\mathbf{r} - \mathbf{r}_{0j}|}}{4\pi |\mathbf{r} - \mathbf{r}_{0j}|}, \quad \mathbf{r} \neq \mathbf{r}_{0j}, \\ s_j &= \frac{4\pi \rho_l \omega^2 a_{0j}^3}{p_*} \Lambda(a_{0j}), \end{aligned} \quad (2)$$

where s_j is the scattering coefficient and k_l is the wavenumber in the liquid without bubbles. Since A^{in} satisfies the Helmholtz equation with the wavenumber k_l , the total field satisfies equation

$$\nabla^2 A + k^2 A = 0, \quad k^2 = k_l^2 + k_b^2, \quad (3)$$

$$k_b^2 = \sum_{j=1}^N s_j \delta(|\mathbf{r} - \mathbf{r}_{0j}|), \quad (4)$$

where $\delta(x)$ is Dirac's delta-function. Ensemble averaging²⁵ shows that for an elementary volume, the effective wave number k for the medium has the same form as in Eq. (3), where k_b^2 is the sum of contributions of all bubbles in that volume. This is consistent with the continuum (spatial averaging) approach,^{10,11} which for diluted monodisperse systems provides the well-known relation

$$k_b^2 = s_0 n_0, \quad \alpha_0 = \frac{4}{3} \pi a_0^3 n_0, \quad (5)$$

FIG. 3. Propagation of the bubble front after switching on the acoustic field. The effect is shown for transducer locations at the bottom (a) at 89 kHz; (b) at 209.2 kHz, on the top; (c) at 85 kHz, and on the two sides of the experimental cell; (d) at 88.8 kHz. (Multimedia view) [URL: <http://dx.doi.org/10.1063/1.4944893.1>] [URL: <http://dx.doi.org/10.1063/1.4944893.2>] [URL: <http://dx.doi.org/10.1063/1.4944893.3>] [URL: <http://dx.doi.org/10.1063/1.4944893.4>]

where s_0 , $\alpha_0(\mathbf{r}, t)$, and $n_0(\mathbf{r}, t)$ are the scattering coefficient, the volume fraction, and the number density of bubbles of radius a_0 . For bubbles moving with the average velocity $\mathbf{v}_0$, we have conservation equation for n_0

$$\frac{\partial n_0}{\partial t} + \nabla \cdot (n_0 \mathbf{v}_0) = 0. \quad (6)$$

Note that in the above equations, period-averaged bubble sizes and positions can change in a time scale slower than ω^{-1} . The former effect is known as “rectified diffusion,”¹⁴ whose characteristic times²⁶ are much larger than the characteristic times of the latter phenomenon.

The momentum conservation equation governing the drift of a bubble of zero mass in an acoustic field can be written as

$$\frac{2}{3} \pi \rho_l \frac{d\langle a^3 \mathbf{v}_b \rangle}{dt} = -\frac{4}{3} \pi \langle a^3 (\nabla p + \rho_l \mathbf{g}) \rangle - 12 \pi k_\mu \mu_l \langle a \mathbf{v}_b \rangle. \quad (7)$$

Here $\mathbf{v}_b$ is the bubble velocity and $\langle \rangle$ denotes averaging over the period of oscillations. In the left hand side we have the added mass force, while the right hand side presents the Bjerknes, gravity, and viscous drag forces ($\mathbf{g}$ is the gravity acceleration and k_μ is the drag coefficient). For small volumetric (see Eq. (1)) and translational oscillations of the j -th bubble about constant radius a_{0j} and location $\mathbf{r}_{0j}$, $j = 1, \dots, N$, this equation takes the form

$$\begin{aligned} \frac{d\mathbf{v}_{0j}}{dt} &= \frac{3p_* \varepsilon^2}{\rho_l} \text{Re} \left\{ \Lambda(a_{0j}) A \nabla \bar{A} \right\} - 2\mathbf{g} - \frac{18k_\mu \mu_l}{\rho_l a_{0j}^2} \mathbf{v}_{0j}, \\ \frac{d\mathbf{r}_{0j}}{dt} &= \mathbf{v}_{0j}. \end{aligned} \quad (8)$$

Equations (3), (4), and (8) form a closed system describing two-way particle–field interaction for a discrete system of bubbles in acoustic field.

The temporal scale separation in the present model can be justified as follows. The length scale for the bubble position $\sim |k|^{-1}$ determines the characteristic time of the bubble drift, t_{s*} (e.g., between the pressure nodes and antinodes)

$$t_{s*} \sim \sqrt{\frac{|k|^{-1}}{|d\mathbf{v}_0/dt|}} \sim \frac{1}{|k| \varepsilon} \sqrt{\frac{\rho_l}{p_*}} \sim \frac{C_*}{\omega \varepsilon} \sqrt{\frac{\rho_l}{p_*}}, \quad (9)$$

where C_* is the characteristic sound speed, $|\Lambda| \sim |A| \sim 1$, $|\nabla A| \sim |k|$, and the gravity and viscous forces are neglected. Note that in bubbly liquids typically $C_* \gg (p_*/\rho_l)^{1/2}$, which means that even for waves of moderate amplitude, $\varepsilon \sim 1$, we have $t_{s*} \gg \omega^{-1}$.

Liquid viscosity increases this time. If the characteristic time t_{s*} is much larger than the viscous relaxation time τ_μ , then the acceleration term in Eq. (8) can be neglected, and the velocity can be determined

$$\mathbf{v}_{0j} = \tau_{\mu j} \left(\frac{3p_* \varepsilon^2}{\rho_l} \text{Re} \left\{ \Lambda(a_{0j}) A \nabla \bar{A} \right\} - 2\mathbf{g} \right), \quad \tau_{\mu j} = \frac{\rho_l a_{0j}^2}{18k_\mu \mu_l}. \quad (10)$$

Thus, in the absence of gravity, the characteristic velocity and time are

$$v_{0*} \sim \frac{\varepsilon^2 p_* \omega \tau_\mu}{\rho_l C_*}, \quad t_{s*} \sim \frac{1}{|k| v_{0*}} \sim \frac{\rho_l C_*^2}{\varepsilon^2 p_* \omega^2 \tau_\mu}. \quad (11)$$

In a one-dimensional model characterized by the spatial coordinate z , we place the origin of the reference frame at the acoustic source location and assume that initially uniform monodisperse bubbly liquid occupies semi-space $z > 0$. Let $A_f = A(z_f)$ be the phasor of the acoustic field at some point $z_f > 0$. Due to attenuation, the acoustic field consists only of the outgoing wave

FIG. 4. Computed bubble volume fraction (thick solid lines) and the acoustic pressure amplitude (thin dashed lines) $|P_a| = \epsilon p_* |A|$ at different instances of time ($\alpha_0 = 0.004$; the driving field of frequency 209.2 kHz and amplitude 700 kPa).

$$A = A_f e^{ik(z-z_f)}, \quad z \geq z_f, \quad \text{Im}\{k\} > 0, \quad \text{Re}\{k\} > 0. \quad (12)$$

This shows that the sign of the Bjerknes force acting on the bubbles at $z = z_f$ is always positive, independently on the bubble size. Indeed, we have from Eqs. (3), (5), and (6)

$$\Lambda = \frac{p_*}{3\alpha_0 \rho_l \omega^2} (k^2 - k_l^2). \quad (13)$$

With this relation, the Bjerknes force can be expressed as

$$\begin{aligned} \frac{F_B}{m_0} &= \frac{3p_* \epsilon^2}{\rho_l} \text{Re} \left\{ \Lambda A \frac{\partial \bar{A}}{\partial z} \Big|_{z=z_f} \right\} = \frac{3p_* \epsilon^2}{\rho_l} |A_f|^2 \text{Re} \{-ik\Lambda\} \\ &= \frac{1}{\alpha_0} \left(\frac{\epsilon p_* |A_f|}{\rho_l \omega} \right)^2 (|k|^2 + k_l^2) \text{Im}\{k\} > 0, \quad m_0 = \frac{2}{3} \pi \rho_l a_0^3. \end{aligned} \quad (14)$$

This contrasts with the sign of the Bjerknes force for a single bubble in a standing acoustic wave where bubbles of radius

$a_0 < a_r$ move towards the pressure antinodes, while bubbles of radius $a_0 > a_r$ move towards the nodes.

Hence, at short times after the acoustic field is turned on, all bubbles start to move away from the source, which forms a “shock wave” of the bubble volume fraction. There are no bubbles behind the front—it is the region of pure liquid, so

$$A = B_1 \sin k_l z + B_2 \cos k_l z, \quad 0 \leq z \leq z_f(t), \quad (15)$$

where integration constants $B_1(t)$ and $B_2(t)$ should be determined from the boundary and matching conditions. This describes a standing wave, which forms when $v_f \ll C_l$. The initial velocity of the front $v_f = dz_f/dt$ can be estimated from Eqs. (10) and (14), where neglecting the gravity and denoting acoustic pressure $P_A = \epsilon p_* |A_0|$, we obtain

$$v_f(0) = \frac{\tau_\mu}{\alpha_0} \left(\frac{P_A}{\rho_l \omega} \right)^2 (|k|^2 + k_l^2) \text{Im}\{k\}. \quad (16)$$

We developed and tested a version of the particle-in-cell (PIC) algorithm. The effect of collisions was neglected, while corrections for the added mass and drag coefficients for finite Reynolds numbers and volume fractions were introduced. Also, the effect of small but finite bubble volume fraction was taken into account in the bubble response function and in the dispersion relation. At each moment of time, $\alpha(z)$ and $k_b(z)$ obtained via the particle-to-grid interpolation were used in the second order finite difference solver to find $A(z)$ for Eq. (3) with boundary conditions on the transducer ($z = 0$) and free surface ($z = H$) (for the transducer located at the bottom of the tank)

$$\frac{\partial A}{\partial z} \Big|_{z=0} = k_l, \quad A|_{z=H} = 0. \quad (17)$$

Then, the grid-to-particle interpolation of $\alpha(z)$ and $A(z)$ was used to compute the dynamics of each bubble.

FIG. 5. Comparison of the theory (thin dashed lines) and the experiment (thick solid lines).

To compare computations and experiments, we used the measured initial bubble volume fraction α_0 , the bubble size distribution shown in Fig. 2, and the amplitude of the acoustic field in the liquid without bubbles, from which the parameter ε can be found. Particularly, we estimated $\varepsilon = 2.98$ for 89 kHz and $\varepsilon = 7.00$ for 209.2 kHz.

It is shown in Fig. 4 that the region behind the wavefront is almost free of bubbles, and we have here a standing wave in pure liquid. In the region occupied by the bubbly liquid, the amplitude of the wave attenuates rapidly in space. One can also see some smaller amplitude short pressure waves just behind the wavefront. These waves are due to a relatively small amount of subresonant satellite bubbles, which form an acoustically transparent medium with a substantially smaller sound speed than in pure liquid. Figure 5 shows that the experimentally observed effect is quantitatively captured by the model. The discrepancy can be explained by a number of model assumptions (e.g., neglecting 3D effects and the value for the bubble drag coefficient used in simulations) as well as by the errors in experimentally measured parameters used in simulations (such as the initial volume fraction and bubble size distribution).

The present theory relates the appearance of the transparency wave to the strong dissipation of acoustic waves in bubbly liquids. There are three basic mechanisms providing non-zero $\text{Im}\{k\}$. First, the dissipative mechanisms related to single bubble dynamics (the thermal dissipation in the gas and the viscous dissipation in the liquid). Second, in the case when the above effects are neglected, the super-resonant bubbles ($a_0 > a_r$) cause $k^2 < 0$, $\text{Im}\{k\} > 0$ for some range of sizes.¹¹ Third, even when the mechanisms of the first kind are neglected, the polydispersity causes the Landau damping.²⁷ In the absence of these mechanisms, the effect should not exist.

It is noteworthy that a good agreement of the theory and experiment is obtained for strong acoustic fields, despite the theory neglects the nonlinearity of bubble oscillations. The explanation comes from the structure of the wave. Indeed, the presence of a few single bubbles behind the wave front, whose behavior may not be properly described by the linear theory, should not affect the driving acoustic field significantly. The bubbles in the mixture at some distance from the front also do not “feel” the acoustic field at all. Finally, the bubbles close to the front are placed either in a substantially reduced acoustic field (due to its strong attenuation) or/and in substantially constrained conditions due to relatively high volume concentrations, preventing their high amplitude

oscillations. So, in the entire domain, the effect of the bubble dynamics nonlinearity may not be very strong, while more detailed studies are required.

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