Supporting Information to

Ultrasound Modulated Droplet Lasers

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Number of pages: 9
Number of figures: 3
Number of videos: 1
Number of equations: 15

Theoretical Estimation of Droplet Deformation

Here we provide a quantitative estimation of acoustic radiation force in the focused ultrasound (US) field and the corresponding deformation of a droplet based on its surface tension. We make the following assumptions: (1) The droplet is isotropic and compressible; (2) The radius of the sphere, r (25 μm), is much smaller than the acoustic wavelength, λ_a (3 mm at 500 kHz); (3) The surrounding fluid is non-viscous; (4) The ultrasound field is axial symmetric; (5) $kR \gg 1$, where k is the wavenumber of ultrasound and R is the radius of the sound source. Following the derivation in the literature, the acoustic radiation force on the droplet along the axial can be expressed as[1]:

$$\boldsymbol{F_a} = \left(\frac{\boldsymbol{A_\alpha + 4B_\alpha}}{\boldsymbol{2c}}\right)\frac{\boldsymbol{\partial I}}{\boldsymbol{\partial z}} \tag{S1}$$

where

$$\boldsymbol{A_\alpha} = \frac{\boldsymbol{12\pi r^3}}{(\boldsymbol{1+\delta_0^2})^3\left\{\frac{1}{[9\nu(1-\Omega^2)\delta^2]} - \frac{\nu}{[3(1-\Omega^2)(1+2\nu)]}\right\}} \tag{S2}$$

$$\boldsymbol{B_\alpha} = \frac{\frac{4}{3}\boldsymbol{\pi r^3(\nu - 1)}}{\boldsymbol{1+2\nu}} \tag{S3}$$

$$\boldsymbol{\delta_0} = \frac{(\boldsymbol{kr})^3\left(\boldsymbol{\nu} - \frac{1}{\delta^2}\right)}{[\boldsymbol{3\nu(1-\Omega^2)}]} \tag{S4}$$

$$\boldsymbol{\Omega} = \frac{\boldsymbol{w}}{\boldsymbol{w_0}} = \frac{\boldsymbol{w}}{\sqrt{\boldsymbol{3\nu}}\boldsymbol{c\delta/r}} \tag{S5}$$

$$\boldsymbol{I} = \frac{|\boldsymbol{p}|^2}{\boldsymbol{2\rho_0 c}} \tag{S6}$$

I is the sound intensity. z is the axial direction of the ultrasound field. c is the sound speed in the surrounding solution. c_s is the sound speed in the droplet. w is the angular frequency. w_0 is the angular resonance frequency of droplet, $\delta = \frac{c_s}{c}$ is the ratio of the sound speeds. $k = \frac{w}{c}$ is the wavenumber. ρ_s is the density of the droplet. ρ is the density of the surrounding solution. $\nu = \frac{\rho_s}{\rho_0}$ is the ratio of the densities. p is the ultrasound pressure.

In Eq. S1, $\left(\frac{A_\alpha + 4B_\alpha}{2c}\right)$ is determined by the material and size of the droplet. $\frac{\partial I}{\partial z}$ can be estimated experimentally. We can measure the pressure difference of a small step Δz and calculate ΔI so that $\frac{\partial I}{\partial z}$ can be estimated with $\frac{\Delta I}{\Delta z}$. Inserting the numerical value for each parameter (US frequency: 500 kHz): r =25 μm, c =1450 m/s, $\nu = 0.93$, δ = 0.9, $w = 2\pi \times 500000/\text{s}$, we have:

$$F_a = 5.74 \times 10^{-18}\frac{\partial I}{\partial z} \tag{S7}$$

Then we analyze the droplet deformation with a surface tension dominated model (Figure S1a)[2]. When the acoustic radiation force, F_a, is balanced by the restoring surface tension force, F_{st}, which can be expressed as:

$$F_{st} = -2\pi a\gamma + \pi a^2\gamma\left(\frac{1}{a} + \frac{a}{b^2}\right) \tag{S8}$$

where a is the equatorial radius, b is the vertical conjugate radius, and γ is the surface tension coefficient. At small deformation, Eq. S8 can be linearized as:

$$F_{st} = 3\pi\gamma\Delta b \tag{S9}$$

The force balancing equation can be expressed as:

$$F_a - F_{st} = 0 \tag{S10}$$

By combining Eqs. S7 - S9 while keeping the volume constant, i.e., $\frac{4}{3}\pi r^3 = \frac{4}{3}\pi a^2 b$ and inserting $\gamma = 0.005\text{N/m}$, we get the deformation ratio, $\Delta b/b$, as a function of F_a:

$$\frac{\Delta b}{b} = 4.88 \times 10^{-12}\frac{\partial I}{\partial z} \tag{S11}$$

Combining Eqs. S6 and S11, we have:

$$\frac{\Delta b}{b} = 3.25 \times 10^{-18} p\frac{\partial p}{\partial z} \tag{S12}$$

Here p and $\frac{\partial p}{\partial z}$ can be experimentally measured (i.e., 500 kHz US, p = 50 kPa, $\frac{\partial p}{\partial z} = 2.4 \times 10^{10}\ Pa/m.$). Finally, the deformation ratio for an oil droplet of 50 µm in diameter immersed in water under the 500 kHz US wave is plotted in Figure S1b. Similarly, the deformation ratio for 2.5 MHz and 10 MHz US wave can also be calculated and plotted in Figure S1b. Based on Eq. S11, 500 kHz US at 30 kPa and 50 kPa can generate a radiation force of 1.65 nN and 4.59 nN, respectively, and the corresponding deformation ratio of 0.14% and 0.39%, respectively. Through the experimental results, we can also estimate the deformation by examining the whispering gallery mode spectral shift. For a whispering gallery mode, the resonant wavelength, λ, can be expressed as:

$$nl = N\lambda \tag{S13}$$

where n is the refractive index of oil, l is the perimeter of a vertical cross-section orbit, N is the mode constant. The resonant wavelength shift of a droplet laser is proportional to the perimeter change in a whispering gallery mode orbit.

$$\Delta\lambda = \lambda\frac{\Delta l}{l} \tag{S14}$$

The perimeter change due to the deformation of vertical cross-section orbit can be calculated as:

$$\Delta l = l - l_0 = 2\pi r - \pi[3(a+b) - \sqrt{(3a+b)(a+3b)} \tag{S15}$$

During lasing enhancement with US, we did not observe any resonant peak shift at 50 kPa. Therefore, the peak shift, $\Delta\lambda$, should be below the spectrometer resolution (0.1 nm). Combining Eqs. S12 - S13 and using $n = 1.46$, $r = 25$ μm, and $\lambda = 540$ nm, we can estimate the upper limit for the corresponding deformation ratio based on the following two assumptions:

(1) conservation of volume for the 3-D model: $\frac{4\pi}{3}r^3 = \frac{4\pi}{3}a^2b$,

$$\frac{\Delta b}{b} < 0.1\%$$

(2) conservation of cross-section area for the 2-D model: $\pi r^2 = \pi a^2 b$,

$$\frac{\Delta b}{b} < 1.5\%$$

The actual deformation ratio should be between these two ideal estimations.

In Figure S1c, we also calculate the ultrasound pressure needed to achieve a certain droplet deformation ratio as a function of the droplet radius. Here we use 0.14% as the radius deformation ratio to reach the critical deformation, which is obtained from the 500 kHz calculations and measurements above.

Fluorescence Intensity of Droplets under Ultrasound

To confirm that US has no effect on the efficiency of dye molecules and pump energy distribution, in Fig. S2 we compare the fluorescence intensity of a droplet (50 μm in diameter) with a strong US pressure (50 kPa at 500 kHz) and without US (0 kPa) when the droplet was pumped at an energy density level of 10 μJ/mm^2 (below its lasing threshold). No significant difference was observed.

Lasing Intensity of Polystyrene Beads under Ultrasound

The effect of US on the lasing properties of a polystyrene bead (10 μm in diameter) doped with FITC fluorescent dye was investigated to confirm that the US enhancement of lasing is due to the deformation. The lasing intensity of the polystyrene laser bead remained unchanged after exposing to continuous US (50 kPa at 500 kHz). Compared to an oil droplet, the polystyrene bead with Young's modulus of 3 GPa is much more difficult to deform under the same US pressure (50 kPa at 500 kHz). Consequently, no lasing enhancement was observed when US was applied to the polystyrene bead.

Temporal Modulation of Droplet Lasers

Here we provide a short video acquired by a CCD to demonstrate the US modulated droplet laser. A series of 4 Hz US bursts (500 kHz, 50 kPa, 60 ms burst duration) were applied to the droplet laser that was pumped at an energy density of 40 μJ/mm^2 with a 20 Hz repetition rate. It can be seen from the video that the droplet laser emitted higher intensity at 4 Hz in response to the US burst modulation.

Video Link

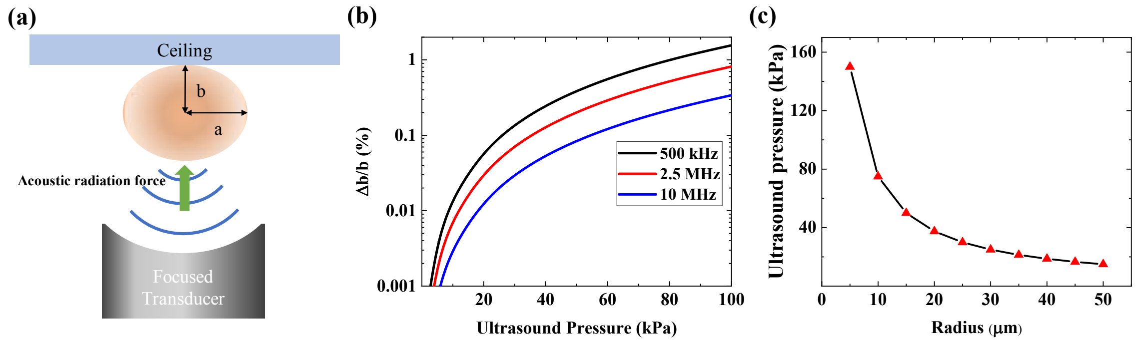

Figure S1. (a) Schematic of the deformation model. (b) Theoretical calculation of the radius deformation ratio of an oil droplet as a function of the ultrasound pressure. (c) Theoretical calculation of ultrasound pressure needed to achieve a certain radius deformation ratio (0.14%) as a function of the droplet radius.

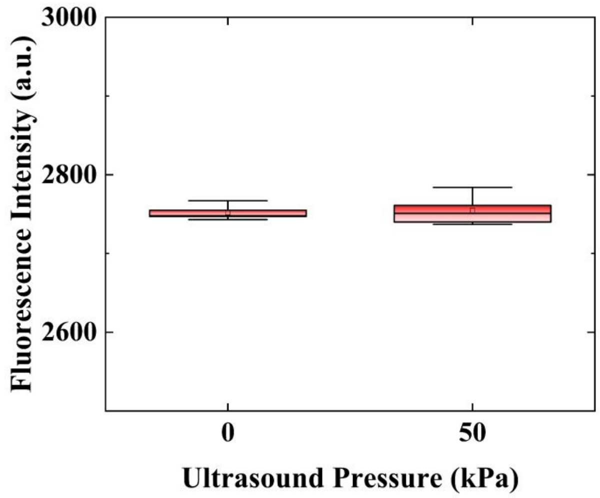

Figure S2. Fluorescence intensity of an oil droplet (50 µm in diameter) doped with BODIPY without and with applying ultrasound pressure (50 kPa at 500 kHz). All data were obtained under the same pump energy density of 10 µJ/mm^2 with a repetition rate of 20 Hz. Error bars were obtained with 5 measurements.

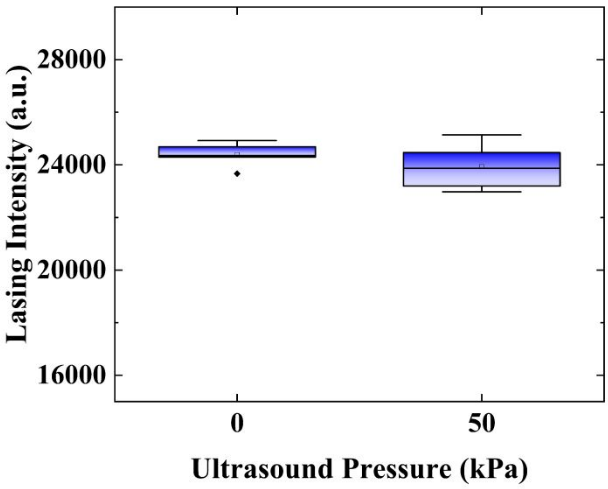

Figure S3. Lasing intensity of a polystyrene bead doped with FITC without and with applying ultrasound pressure (500 kHz, 50 kPa). All data were obtained under the same pump energy density of 40 μJ/mm^2 with a repetition rate of 20 Hz. Error bars were obtained with 5 measurements.

Reference:

(1) Wu, J.; Du, G., Acoustic radiation force on a small compressible sphere in a focused beam. *J. Acoust. Soc. Am.* **1990,** *87*, 997-1003.
(2) Møller, P. C. F.; Oddershede, L. B., Quantification of droplet deformation by electromagnetic trapping. *EPL* **2009,** *88*.