I. SUPPLEMENTARY DISCUSSION S1 - INTERFACE DEFORMATION USING SAW

The acoustic pressure on an interface has been the focus of several publications by many prominent authors in the field of nonlinear acoustics. [1–4]. The acoustic radiation force on a surface in the path of an acoustic beam can be generally broken up into the Langevin and Rayleigh radiation pressures; the first of these refers to the time-averaged force tensor in the direction of acoustic propagation on a surface placed in the path of a beam, while the Rayleigh pressure on a surface is the combination of this and the isotropic static pressure induced. Both formulations of acoustic pressure are equally valid, though apply to different systems, depending on whether the interface the acoustic beam is confined by the interface it acts upon. In the case of an acoustic beam acting on an oil-water interface, there is no method, aside from interface movement, to transfer the isotropic pressure induced by the beam to the oil side of the interface, meaning this component of the pressure will influence the interface shape and must be taken into account. The Rayleigh pressure acting on an interface is then given by [2, 3]

$$p_r = \langle p - p_0 \rangle + \langle \rho v^2 \rangle, \tag{1}$$

where v is the instantaneous fluid particle velocity and $\langle \rho v^2 \rangle$ is simply $\langle E \rangle$, the energy density in the fluid. To a first order approximation the fluid particle velocity $v \approx v_0$, where $v_0 = (\xi\omega)$ is the substrate velocity, ξ is the surface displacement and ω is the angular frequency. If the substrate velocity is oscillating sinusoidally as in a SAW, the time average $\langle \rho v^2 \rangle$ is nonzero, resulting in a nonzero pressure term in a fluid media placed on top of the substrate. In terms of the Fox and Wallace coefficients [5], which are determined by the first and second order compressibility of a fluid, Eq. (1) can be expressed as

$$p_r = \frac{B}{2A}\langle E \rangle + \langle E \rangle, \tag{2}$$

where the first term on the right side represents the static pressure term, and the second term arises from the nonlinear interaction between the acoustic wave and the water-oil interface. To model the interaction of an acoustic beam arising from surface acoustic waves (SAW) with the water-oil interface, the static pressure terms and the force term at the interface need to be treated separately as the surface topology of the interface will be non-planar during droplet formation. Eq. (2) can then be written as

$$p_r = \frac{B}{2A}\langle E_0 \rangle + \sin\left(\phi(z) - \theta_R\right)\langle E_1 \rangle, \tag{3}$$

where $\sin(\phi(z) - \theta_R)$ is the vector normal to the interface surface and $\langle E_0 \rangle$, $\phi(z) = \tan^{-1}(\partial h/\partial z)$ the inclination angle of the interface in the $x - z$ plane, where x projects horizontally along the SAW propagation direction and z projects vertically from the substrate to the PDMS upper surface, with h being the distance between any point on the interface and the orifice and $\langle E_1 \rangle$ representing the energy density in the water bulk and at the water-oil interface. Whereas $\langle E_0 \rangle$ is simply equal to $\langle \rho v_0^2 \rangle$ [3], the energy density at the interface must take into account that the oil-water boundary is only partially reflecting, with the energy density given here as a function of the density and sound speeds in oil (ρ_o, c_o) and water (ρ_w, c_w) [2];

$$\langle E_1 \rangle = \frac{2\left[1 + \left(\frac{\rho_o c_o}{\rho_w c_w}\right)^2\right]\langle E_0 \rangle}{\left(1 + \frac{\rho_o c_o}{\rho_w c_w}\right)^2}. \tag{4}$$

At steady state the acoustic pressure at the oil-water interface is balanced by the capillary pressure $-\, p_c = p_r$. Assuming the interface shape is equivalent in both the $x - z$ and $x - y$ plane, i.e. a square orifice shape with $p_c(x, z(\tau)) \approx p_c(x, y(\tau))$, then p_c is simply twice the value in the $x - z$ plane, with

$$p_c \approx -2\gamma \frac{\partial^2 h}{\partial z^2}, \tag{5}$$

where $\gamma \approx 0.024$ N/m [6] is the value for surface tension at the oil-water interface. Combining the capillary pressure in Eq. (5), the Rayleigh pressure in Eq. (3) and the capillary pressure of an oil-water interface at rest yields the final steady state interface shape equation;

$$2\gamma \frac{\partial^2 h}{\partial z^2} = \frac{B}{2A}\langle E_0 \rangle + \sin\left(\phi(z) - \theta_R\right)\langle E_1 \rangle - \gamma \frac{1}{L}, \tag{6}$$

where L is the length scale of the orifice. If the interface is pinned at the borders of the orifice, the boundary conditions can be specified by

$$h|_{z^*=0} = 0 \qquad h|_{z^*=1} = 0, \tag{7}$$

Equations (6) and (7) were solved numerically on a mesh of 1000 points using MATLAB's boundary value problem solver `bvp4c` [7] on the domain $z^* = [0, 1]$, where $z^* = z/L$ and $x^* = x/L$. The interface shape for different v_0 is given in Fig. 1 for both the Langevin (Fig. 1a) and Rayleigh (Fig. 1b) radiation pressures. Note that if the static pressure term in omitted, the interface shape tends towards θ_R.

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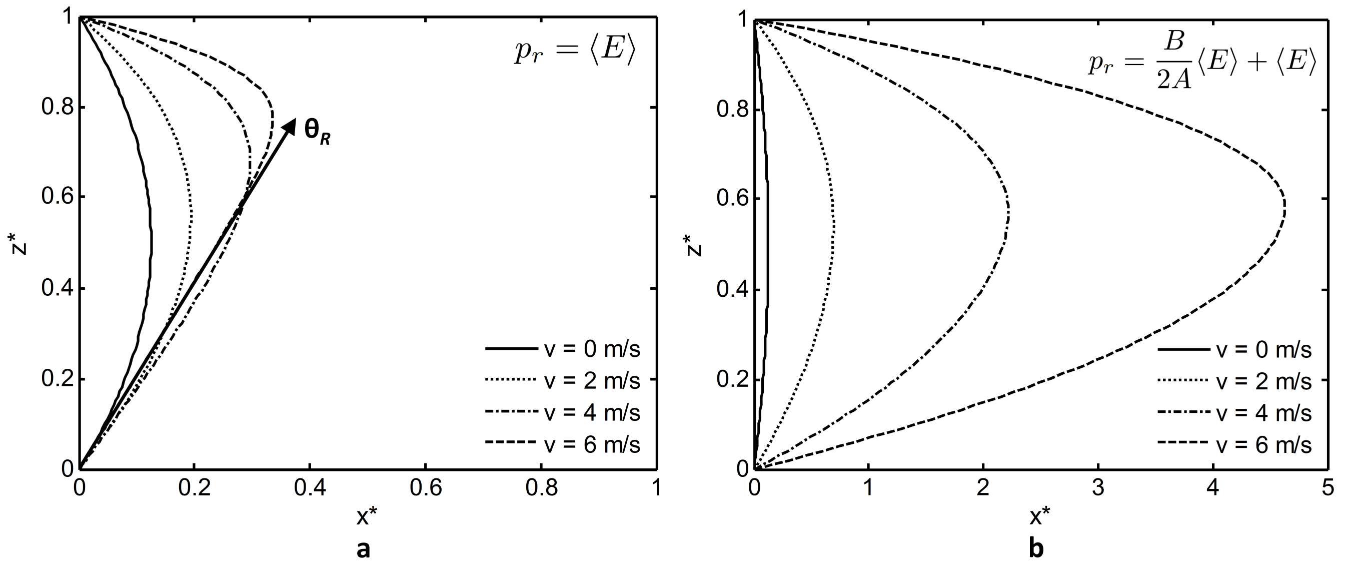

FIG. 1. A fluid-fluid interface (here oil and water) will be deformed by a time averaged second order acoustic radiation pressure, provided the two fluids have a non-zero acoustic contrast. In the case of an incident travelling wave, both the direct interfacial and static pressures pressures will determine the interface shape. (a) Considering only the pressure due to the nonlinear interaction between the interface and acoustic wave (the so-called Langevin radiation pressure), with the waves traveling upward from the SAW substrate at the Rayleigh angle θ_R, this figure shows the steady-state water-oil interface shape for different particle velocities (v = 0, 2, 4 and 6 m/s) in the fluid at a square orifice, where $v \approx v_0 = \xi\omega$, the substrate surface velocity. If only the Langevin pressure is accounted for, the leading edge of the meniscus is generally unable to exceed the line denoted by θ_R, and is unable to advance into the channel sufficiently for droplet formation. (b) Shows the same interface shapes when the static pressure component, due to the nonlinear propagation of an acoustic wave through the fluid medium (water), is taken into account.

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