Transition from Boundary-Driven to Bulk-Driven Acoustic Streaming Due to Nonlinear Thermoviscous Effects at High Acoustic Energy Densities

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Microscale acoustofluidic devices are used to manipulate and control microparticles and cells. In such devices, two main forces act on the suspended particles, the acoustic radiation force and the drag force due to acoustic streaming, which is a time-averaged flow caused by the inherent nonlinearities of fluid dynamics. Recent work has clarified many subtle details pertaining to the radiation force on microparticles, including thermoviscous effects [1] and microstreaming [2]. Concurrently, similar progress has been made in the theory of acoustic streaming, especially regarding thermoviscous effects. The fundamental boundary-driven streaming caused by time-averaged forces in the oscillatory boundary-layer flow [3], and the fundamental bulk-driven streaming generated by the time-averaged dissipation of traveling waves [4], have recently been supplemented by bulk-driven baroclinic [5,6] and thermoacoustic [7,8] streaming, caused by an interplay between standing acoustic waves and steady temperature gradients. However, as noted in Refs. [7,8], the validity of the conventional perturbation approach breaks down at moderately high, but experimentally obtainable average acoustic energy densities $E_{ac}$ (1700 J/m$^3$) is reported in Ref. [9], or even lower, when combined with moderate thermal gradients above 1 K/mm. This need for an extension of the theory beyond perturbation theory is addressed in this Letter and in the jointly submitted paper [10] containing a detailed derivation of the nonperturbative model.

We introduce a nonperturbative iteration approach to investigate theoretically and numerically, the nonlinear effects appearing in a conventional acoustofluidic channel at high $E_{ac}$ in the fluid, and we validate experimentally the model predictions. For general discussions, we consider a straight microchannel with arbitrary cross section embedded in an elastic solid driven at the standing half-wave acoustic resonance by actuating part of the exterior surface. The streaming at low $E_{ac}$ is dominated by conventional boundary-driven streaming with four streaming rolls as sketched in Figs. 1(a)--1(b). For specific numerical and experimental studies, we employ the widely used rectangular channel shown in Fig. 1(d) [10–14]. We show how nonlinear effects in the form of heating by viscous dissipation from the acoustic field inside the boundary layers set up a steady temperature gradient $\nabla T_0$. This gradient drives a strong thermoacoustic streaming in the bulk, which changes the streaming flow qualitatively from...
four to two flow rolls, as sketched in Fig. 1(c), and which by thermal convection alters the temperature field.

Our nonperturbative analysis of this nonlinear phenomenon and its underlying mechanism fills a knowledge gap in nonlinear acoustics, and it provides a guidance for understanding and optimizing acoustofluidic systems running at high $E_{ac}$ such as high-intensity ultrasound focusing [15–17], acoustic streaming-based micromixers [18–21], particle manipulation devices [22–24], and high-throughput acoustophoresis devices [25–27].

Physical model.—The fluid in the microchannel in Fig. 1 is characterized by nine material parameters: density $\rho$, thermal conductivity $k_{th}$, specific heat $c_p$, dynamic and bulk viscosity $\eta$ and $\eta_b$, thermal expansion coefficient $\alpha_p$, the ratio of specific heats $\gamma = c_p/c_v$, and the isentropic and isothermal compressibility $\kappa_s$ and $\kappa_f = \gamma \kappa_s$. The elastic solid is characterized by density $\rho$, longitudinal and transverse sound speed $c_{lo}$ and $c_{lt}$, thermal conductivity $k_{th}$, thermal expansion coefficient $\alpha_{p0}$, and isothermal compressibility $\kappa_f$. For specific experimental and numerical studies, we consider the 24-mm long, silicon-glass chip used in Ref. [28] with the rectangular cross section; see Fig. 1(d). The horizontal half-wave resonance mode in the fluid (water) is excited at frequency $f_0 = 1.911$ MHz by a nanometric bottom-edge actuation displacement $u_{exc}$. The response to the acoustic actuation is governed by the conservation equation for mass, momentum, and energy in the fluid and solid. The independent fields are the pressure $p$, the velocity $v$, and the temperature $T$ in the fluid, and the displacement $u$ and $T$ in the solid. The material parameters including temperature dependencies are given in Refs. [10,13] and in Table S1 in the Supplemental Material [29].

The iterative approach.—We exploit that the acoustic fields vary much faster (~$10^{-7}$ s) than the hydrodynamic and thermal flows (~$10^{-2}$ s). We study the steady limit of the slow timescale and decompose physical fields and material parameters $Q_{phys}$ into a steady term $Q_0$ and a time-harmonic acoustic term $\text{Re}\{Q_1 e^{-i\omega t}\}$ with a steady complex-valued amplitude $Q_1$, be

$$Q_{phys}(t) = Q_0 + \text{Re}\{Q_1 e^{-i\omega t}\}. \tag{1}$$

We neglect higher harmonics with angular frequency $n\omega$, $n = 2, 3, \ldots$, and use this ansatz to separate the governing equations in one set that controls the acoustic fields, and another set that controls the steady fields. Since products of two acoustic terms $a_1 b_1$ and $b_1$ contain a steady time-averaged part $\langle a_1 b_1 \rangle = \frac{1}{2}\text{Re}\{a_1 b_1^*\}$, where the asterisk denotes complex conjugation, the acoustic terms appear as source terms $\langle a_1 b_1 \rangle$ in the governing equation of the steady fields. Conversely, the steady fields $p_0$, $v_0$, and $T_0$ determine the material parameters on which the acoustic fields depend. In the nonperturbative thermoviscous model presented here, and with more details added in Sec. II-D in Ref. [10], the combined set of equations for the coupled acoustic and steady fields are solved by a self-consistent iterative sequence until convergence is obtained. This procedure allows for studies beyond the traditional perturbative models of acoustofluidics [7,13].

Acoustofluidic systems also exhibit dynamics on two different length scales, one set by the system size and wavelength of the acoustic fields, $d \gtrsim 100$ μm, and another by the viscous and thermal boundary layer thickness of width $\delta_{s} = \sqrt{2\nu_0/\omega}$ and $\delta_{t} = \sqrt{2D_{th}/\omega}$, respectively ($\delta_s \lesssim \delta_t \lesssim 500$ nm $\ll d$). In the refined version of the iterative thermoviscous model presented in Ref. [10], we use this length-scale separation to decompose all fields $Q$ into bulk and boundary-layer fields which vary on the scales $d$ and $\delta$, respectively, $Q = Q^b + Q^\delta$. The boundary-layer fields are solved analytically and then taken into account as effective boundary conditions on the bulk fields $Q^b$. This so-called effective boundary-layer model avoids the computationally costly resolution of the thin boundary layer, and is in practice a necessity to enable simulations in three dimensions (3D). Since in this Letter, we only perform simulations in two dimensions (2D), we do not decompose $Q$ spatially, but instead use a so-called full model, in which the governing equations are solved numerically by resolving the boundary layers.

Acoustic and stationary fields.—The thermoviscous model is derived in detail in Ref. [10], but is briefly summarized here. The fluid stress is written $\tau = -\nabla p + \tau$, with $\tau = \eta_0 ( \nabla v + (\nabla v)^T ) + \eta_0 \gamma (\nabla \cdot v) I$ being the viscous part. Inserting $v = v_0 + v_1$ and $\eta = \eta_0 + \eta_1$ gives $\tau = (\tau_0 + \tau_{ac}) + \tau_1$, where the steady part $\tau_0 + \tau_{ac}$ contains terms $\eta_0 \nabla v_0$ and $\langle \eta_1 \nabla v_1 \rangle$, respectively, and the acoustic part $\tau_1$ contains terms $\eta_1 \nabla v_1$. The governing equations for the acoustic temperature $T_1$, pressure $p_1$, and velocity $v_1$ in the fluid as well as temperature $T_1^s$ and displacement $u_1$ in the solid, become

$$-i\omega T_1^0 + i\alpha p_0 (\gamma - 1) \frac{k_{th}}{\alpha_{p0}} p_1 = D_{0}^{th} \nabla^2 T_1^0, \tag{2a}$$

$$-i\omega v_1 = -\nabla p_1 + \nabla \cdot \tau_1, \tag{2b}$$

$$-i\omega T_1^0 - i\alpha \frac{\gamma - 1}{\alpha_{p0}} \nabla \cdot u_1 = D_{0}^{th} \nabla^2 T_1^0, \tag{2c}$$

$$-i\omega^2 p_0 u_1 = -\frac{\alpha_{p0}}{k_{th}} \nabla T_1^0 + (c_0^2 - c_u^2) \nabla (\nabla \cdot u_1) + c_u^2 \nabla^2 u_1. \tag{2e}$$

Similarly, the governing equations for the steady temperature $T_1^s$, pressure $p_0$, and streaming velocity $v_0$ in the fluid, as well as the temperature $T_1^s$ in the solid, are

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\[ 0 = -\nabla \cdot (\rho_0 \nu_0) + \dot{\rho}_\text{ac} \]  
\[ 0 = -\nabla p_0 + \nabla \cdot \tau_0 - \nabla \cdot (\rho_0 \nu_0) + \tilde{f}_\text{ac} \]  
\[ 0 = \nabla \cdot [k_0 \nabla T_0] - c_p \rho_0 \nu_0 \cdot \nabla T_0 + \rho_0 \nu_0 + P \]  
\[ 0 = \nabla \cdot (k_0 \nabla T_0) + P_{\text{ac}} + P. \]

with the acoustic-product source terms given by

\[ \dot{\rho}_\text{ac} = -\nabla \cdot \langle \rho_1 \nu_1 \rangle, \]  
\[ \tilde{f}_\text{ac} = \nabla \cdot [-\rho_0 \nu_1 + \tau_\text{ac}], \]  
\[ P_{\text{ac}} = \nabla \cdot (k_0 \nabla T_0), \]  
\[ P_{\text{ac}} = \nabla \cdot [(k_0 \nabla T_0) - \langle \rho_1 \nu_1 \rangle + \nu_1 \cdot \tau_1] - \rho_0 c_p \rho_0 \langle \nu_1 \cdot \nabla T_0 \rangle. \]

Here, \( P_{\text{ac}} \) and \( P \) are the power density supplied internally by the acoustic fields and externally by given sources, respectively, and we have neglected small terms \( |\rho_1 \nu_1| \ll |\rho_0 \nu_0| \) and \( |\eta_1 \nu_0| \ll |\eta_0 \nu_1| \), which holds for typical acoustic-fluidic devices. Moreover, since the thermal expansion of the solid due to the gradients in \( T_0 \) is minute, we have assumed \( u_0 = 0 \). The boundary conditions are continuous velocity and stress fields across the oscillating fluid-solid interface for both steady (including the Stokes drift) and acoustic fields, as detailed in Ref. [10]. A no-stress condition is applied on the exterior surface, except at the actuation region, where a displacement condition \( u_1 = u_{\text{exc}} \) is applied to \( u_1 \).

We note that in the bulk, the acoustic body force takes the form \( f_{\text{ac}} = f_{\text{ac}} = \nabla (\nabla \cdot \rho_0) \). The gradient term is absorbed in the pressure gradient \( -\nabla p_0 \), leaving the second-order field \( f_{\text{ac}} \) of the acoustic body force, which drives thermoacoustic streaming [7,8] in the bulk of the fluid [10].

\[ f_{\text{ac}} = -\frac{1}{4} |\nu_1|^2 \nabla p_0 + \left[ 1 - \frac{2a_0 (\gamma - 1)}{\beta + 1} \frac{\Gamma_\omega}{c_0^2} \langle \nu_1 \rho_1 \rangle \right] \nabla \mathbf{p}_0 \]  
\[ -\frac{1}{4} |\nu_1|^2 \nabla \kappa_1 + 2a_0 \eta_0 (\gamma - 1) a_0 \frac{\alpha}{c_0^2} \langle \nu_1 \cdot \nabla \nu_1 \rangle \]  
\[ \approx -\frac{1}{4} \left[ |\nu_1|^2 \frac{\partial \kappa_1}{\partial T} \right]_{T_0} + |\nu_1|^2 \left[ \frac{\partial \rho}{\partial T} \right]_{T_0} \nabla T_0. \]  

Here, the last expression is valid for sufficiently large temperature gradients \( \nabla T_0 \) in the bulk. In the usual perturbative limit, the first-order fields \( p_1, \nu_1 \), and \( u_1 \) depend linearly on the actuation amplitude \( u_{\text{exc}} \). As time-averaged products of these fields are sources for the time-averaged second-order fields \( T_2 \) and \( v_2 \), the spatial patterns of the latter are independent of \( u_{\text{exc}} \), whereas their amplitudes scale as \( u_{\text{exc}}^2 \), and \( \nabla T_0 \) all scale as \( E_{\text{ac}}^2 \). Hence, \( f_{\text{ac}} \) leads to nonperturbative effects in the thermoviscous streaming \( v_2 \), such as an \( E_{\text{ac}} \)-dependent spatial pattern and an amplitude nonlinear in \( E_{\text{ac}} \). These effects will dominate over the usual perturbative response at sufficiently high values of \( E_{\text{ac}} \), as shown experimentally and numerically below.

**Experimental method.**—The experiments were performed on the glass-silicon chip of Fig. 1(d) glued to a piezoelectric transducer, the same as in Ref. [28]. The system was driven at the resonance frequency 1.97 MHz using input powers \( P_{\text{in}} = 6.1, 86.8, \) and \( 182.5 \) mW, resulting in the energy density \( E_{\text{ac}} = 27.2 \pm 1.1, 388.7 \pm 15.9, \) and \( 817.3 \pm 33.5 \) J/m\(^3\). When less than \( 140 \) J/m\(^3\), \( E_{\text{ac}} \) is determined from the focusing of 4.9-\( \mu \)m-diameter polystyrene particles at 140 fps using confocal microparticle image velocimetry (μPIV) [9]. When greater than \( 140 \) J/m\(^3\), \( E_{\text{ac}} \) is estimated using the proportionality \( E_{\text{ac}} \propto P_{\text{in}} \). To measure \( E_{\text{ac}} \) accurately, the confocal μPIV technique only captures the particle motion near the focal plane (channel midheight), excluding particles near the top and bottom walls influenced by hydrodynamic and acoustic particle-wall interactions [11]. The acoustic streaming for each \( E_{\text{ac}} \) was measured at 10 to 60 fps by tracking 0.5-\( \mu \)m-diameter particles using a defocusing-based 3D particle tracking technique [32–34]. To avoid the resonance frequency shift due to the temperature rise of the transducer under moderate (86.8 mW) and high (182.5 mW) \( P_{\text{in}} \), each measurement was run for only 2 s and repeated 100 times to improve the statistics, resulting in 7800–12 000 recorded frames for each driving condition. For further experimental details see Sec. S2 in the Supplemental Material [29].

**Numerical method.**—The numerical simulation of the model is carried out using the commercial finite-element software COMSOL Multiphysics [35] as described in Ref. [7] for the perturbative case, but extended with the iterative procedure described above to handle the nonperturbative case; see details in Ref. [10]. The only free parameter is the displacement amplitude \( u_{\text{exc}} \), which is fixed to obtain the measured value of \( E_{\text{ac}} \). A sample script is provided in Sec. S3 in the Supplemental Material [29].

**Results and discussion.**—The simulation and experimental results in Fig. 2 show how the streaming \( \nu_0 \) and temperature \( T_0 \) in a standard acoustofluidic device undergo a clear qualitative transition as \( E_{\text{ac}} \) increases. In the perturbative regime at low \( E_{\text{ac}} \lesssim 30 \) J/m\(^3\) shown in Fig. 2(a), \( \nu_0 \) is dominated by the boundary-driven streaming sketched in Fig. 1(b) that scales linearly with \( E_{\text{ac}} \) and exhibits the usual four flow rolls. Because of friction in the viscous boundary layers, heat is generated both at the top and bottom of the channel. At the bottom, this heat is removed less efficiently by the lower heat conductivity of glass, and a steady temperature gradient \( \nabla T_0 \) is established, which
together with Eq. (4b) explains the temperature \( T_0 \) seen in Fig. 2(f): The acoustic body force \( f_{ac} \) points toward the high temperature at the top, and it is strongest at the pressure antinodes at the sides \[7,8\]. Consequently, \( f_{ac} \) pushes liquid from the sides up toward the top center, which induces a backflow down along vertical center axis. The resulting streaming pattern consists of two flow rolls, one in each side of the channel. This bulk-driven nonperturbative pattern is seen in Fig. 2(d) at the high \( E_{ac} \approx 5300 \) J/m\(^3\), where \( v_0 \) is completely dominated by the thermoacoustic streaming. The transition from boundary-driven streaming at low \( E_{ac} \) to bulk-driven streaming at high \( E_{ac} \) is studied qualitatively in Figs. 2(a)–2(d) and quantitatively in Fig. 2(e). During the transition in Figs. 2(b)–2(c), the two bottom streaming rolls expand, and the two top rolls shrink, at \( E_{ac} \approx 380 \) and 800 J/m\(^3\), respectively. The bottom rolls expand, because they rotate the same way as the two thermoacoustic streaming rolls.

This transition is studied quantitatively in Fig. 2(e) by plotting measured (3 points) and simulated (156 points) values of the spatial average \( \langle v_0 \rangle \) of the magnitude \( v_0 = |v_0| \) of the streaming velocity and the vertical distance \( \Delta_y \) (white bar) is the height where \( v_0 \) is maximal. The error bars on experimental \( \langle v_0 \rangle \) and \( E_{ac} \) are within the square markers \[29\]. The round markers (⊙) represent the simulations shown in panels (d) and (f)–(i). (f)–(i) Color plot of simulated temperature increase \( T_0 \) from 0 (blue) to \( T_0^{\max} \) at four \( E_{ac} \). \( \Delta_T \) (white bar) is the height where \( T_0 = \frac{1}{2} T_0^{\max} \). (j) Plots of simulated \( T_0^{\max} \) and \( \Delta_T \) vs \( E_{ac} \) showing a transition from diffusion-dominated to convection-dominated heat transport.

As \( v_0 \) increases, the heat convection \(-c_p \rho_0 v_0 \cdot \nabla T_0^0\) in Eq. (3c) affects the temperature field even more strongly, as seen in Figs. 2(f)–2(i) for \( E_{ac} = 380, 800, 5300, \) and 12 600 J/m\(^3\), and it becomes important when the Péclet number \( |v_0|H/D^{th} \) exceeds unity for \( |v_0| \geq 1 \) mm/s, consistent with Figs. 2(f)–2(j). Qualitatively, we see that for \( E_{ac} \approx 800 \) J/m\(^3\), the two flow rolls pull the temperature profile down along the vertical center axis. We quantify this effect by the maximum temperature \( T_0^{\max} \) and the vertical
distance $\Delta_T$ along the center axis from the bottom edge to the point where $T_0 = \frac{1}{4} T_{0 \text{max}}$. The thermoacoustic streaming increases the heat transport from the fluid-glass interface to the silicon wafer; thus $T_{0 \text{max}}$ increases less steeply than the perturbative result, $T_{0 \text{max}} \propto E_{ac}$, as seen in the log-log plot (blue) of $T_{0 \text{max}}$ vs $E_{ac}$ for $E_{ac} \gtrsim 5000$ J/m$^3$ in Fig. 2(j). A stronger signal is seen in the log-lin plot (dark red), where the perturbative result $\Delta_T = \text{const}$ only holds for $E_{ac} \lesssim 400$ J/m$^3$, after which point $\Delta_T$ decreases with increasing $E_{ac}$.

**Conclusion.**—In this Letter we have shown numerically and experimentally that the acoustic streaming in a standard microscale acoustofluidic device is changed qualitatively for moderately high acoustic energy densities $E_{ac} \gtrsim 400$ J/m$^3$. We have explained this effect by a nonperturbative model, in which a transition from boundary-driven to bulk-driven acoustic streaming occurs, as the acoustic body force $f_{ac}$ begins to dominate the streaming at increased $E_{ac}$ due to the internal heating generated in the viscous boundary layers. We have shown good qualitative and quantitative agreement between our model predictions and experimental data. The iterative model can easily be extended to materials other than silicon, glass, and water, such as we have done in the perturbative model with iodixanol solutions [36], rubber (polydimethylsiloxane) [37,38], aluminum [38], and polymer (PMMA) [39], and with oil in the iterative model itself [10].

$E_{ac} \gtrsim 400$ J/m$^3$ can easily be obtained in standard acoustofluidic devices, for which $E_{ac} \approx 10$–50 J/m$^3 \times (U_{pp}/(1 \text{ V}))^2$ has been reported, $U_{pp}$ being the voltage applied to the transducer [12,40–42], and higher $E_{ac}$ could be obtained by optimized actuation schemes [9,39,43,44]. The physical understanding of how such acoustofluidic devices behave at high $E_{ac}$ is important for the continued development of high-throughput devices of particular relevance in biotech and clinical applications.

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