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Microscale Capillary Wave Turbulence Excited by High Frequency Vibration

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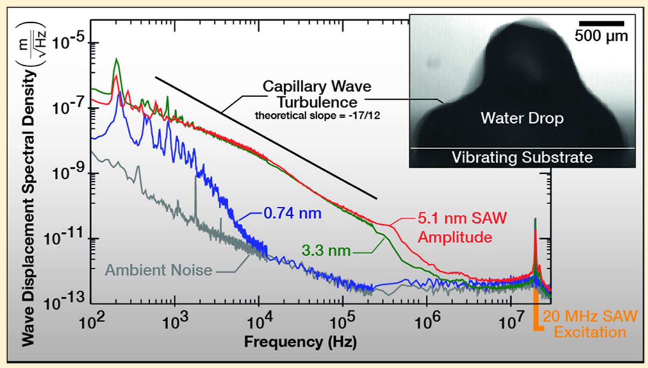
Supporting Information



ABSTRACT: Low frequency (O(10 Hz−10 kHz)) vibration excitation of capillary waves has been extensively studied for nearly two centuries. Such waves appear at the excitation frequency or at rational multiples of the excitation frequency through nonlinear coupling as a result of the finite displacement of the wave, most often at one-half the excitation frequency in so-called Faraday waves and twice this frequency in superharmonic waves. Less understood, however, are the dynamics of capillary waves driven by high-frequency vibration (>O(100 kHz)) and small interface length scales, an arrangement ideal for a broad variety of applications, from nebulizers for pulmonary drug delivery to complex nanoparticle synthesis. In the few studies conducted to date, a marked departure from the predictions of classical Faraday wave theory has been shown, with the appearance of broadband capillary wave generation from 100 Hz to the excitation frequency and beyond, without a clear explanation. We show that weak wave turbulence is the dominant mechanism in the behavior of the system, as evident from wave height frequency spectra that closely follow the Rayleigh−Jeans spectral response $\eta \approx \omega^{-17/12}$ as a consequence of a period-halving, weakly turbulent cascade that appears within a 1 mm water drop whether driven by thickness-mode or surface acoustic Rayleigh wave excitation. However, such a cascade is one-way, from low to high frequencies. The mechanism of exciting the cascade with high-frequency acoustic waves is an acoustic streaming-driven turbulent jet in the fluid bulk, driving the fundamental capillary wave resonance through the well-known coupling between bulk flow and surface waves. Unlike capillary waves, turbulent acoustic streaming can exhibit subharmonic cascades from high to low frequencies; here it appears from the excitation frequency all the way to the fundamental modes of the capillary wave at some four orders of magnitude in frequency less than the excitation frequency, enabling the capillary weakly turbulent wave cascade to form from the fundamental capillary wave upward.

■ INTRODUCTION

Deceptively simple in appearance, capillary waves generated on a fluid interface have provided nearly two centuries' worth of scientific interest, from the Faraday's crispations [1] to the development of entirely new analysis techniques in the modern era.[2] Ubiquitous in nature and man-made systems, from ocean waves to nebulizers, their importance in microfluidics comes as no surprise: capillary waves influence the bulk transport of drops and thin films driven by surface waves;[3−5] form cavitation at intense amplitudes and frequencies below a few megahertz;[6,7] alter the acoustic wave propagation within the fluid bulk;[8] affect microscale mixing[9] and particle concentration;[10] and underlie atomization, where the crests of large-magnitude capillary waves pinch off and are ejected to form a mist of fine droplets.[11−14]

The surprise is the appearance of relatively low-frequency capillary waves in these researchers' work, even when the excitation frequencies (>O(100 kHz)) are three or more orders of magnitude above the capillary waves' resonance frequencies (O(10 Hz)−10 kHz). In two centuries of work on capillary wave phenomena, researchers have considered only the use of excitation at frequencies around or below the resonance frequencies of the capillary wave,[15,16] yet throughout the new literature where high-frequency excitation is employed, capillary waves still appear and drive the consequent phenomena that are now widely used to creatively manipulate fluids. This remarkable discrepancy has only now started to be explored.[3,17,18] In the laboratory, capillary waves are traditionally generated by directly perturbing the interface using a wavemaker,[15] a vibrating rod or plate penetrating the interface,[19] a microsphere in contact with the interface,[20] creative use of electric fields,[21] or—most typically—by

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parametrically inducing the waves through a vertical vibration of the fluid container.[22−27]

A fluid surface excited by a perpendicularly oscillating support may exhibit Faraday waves vibrating at one-half the excitation frequency,[1] in recognition of Faraday's discovery of these waves in a pool of mercury excited by hand in sunlight. Classical theoretical treatment of the Faraday problem gives, along the way, a Mathieu equation in terms of the interface displacement x_m

$$\frac{d^2 x_m}{dT^2} + (p_m - 2q_m \cos(2T))x_m = 0 \quad (1)$$

where $T = ft$, p_m is the ratio of the mth discrete resonance ($m \in \{0, 1, 2,...\}$) frequency f_m to the forcing frequency f, and q_m is proportional to the forcing amplitude.[28,29] The excitation of the surface comes from an explicit time-dependent coefficient in the equation of motion: the $\cos(2T)$ term adds energy to the system. Stability analysis shows that the response frequency of the Faraday wave system depends on its representation in $p-q$ space, defining the ratio of the excitation frequency to the natural frequency and the amplitude of excitation. Figure 2 of Benjamin and Ursell[28] shows that the widely quoted $^f/_2$ response is seen only when the ratio p_m is on the order of 1 or less and the amplitude of forcing is not greater than the order of the natural wavelength. A finite-amplitude analysis of the problem places a greater restriction on the excitation frequency such that

$$\varepsilon \equiv \frac{f - f_m}{f} \ll 1 \quad (2)$$

for any resonance f_m;[29] in other words, the difference between the resonance frequency in question, f_m, and the excitation frequency, f, must be much less than the value of the excitation frequency itself.

This restriction on the excitation frequency is overlooked, even omitted from a number of articles in the literature,[15,30−32] leading to the erroneous application of Faraday wave theory when examining capillary waves in which the excitation frequency is many orders of magnitude greater than the fundamental natural frequency of the interface.[11,25,33,34] In this study, because we use extremely high excitation frequencies and therefore violate the condition presented by eq 2, Faraday wave theory as presented in the literature simply does not apply.

Furthermore, the fluid interface response cannot be fully described by a series of harmonic eigenmode resonances for finite-amplitude excitation; the response is inherently broad band in nature. The vibration response undergoes a transition from a discrete series of quantifiable, finite-amplitude eigenmode waves to a broad-band continuum of waves at a very low amplitude of excitation.[12,35] The mechanism driving this change from discrete to continuous vibrational spectra is the nonlinear, three-wave interaction between waves: after a set of traveling waves collide, the waves that arise from that interaction will extract some energy from the original set but possess different energies, frequencies, and phases from each other and that original set. These new waves go on to interact with each other, the original set, and additional waves not a part of this original collision to form other waves that likewise have new characteristics and so on in a cascade of ever-shorter-wavelength waves with reduced amplitude.[36,37]

Such a broad-band power-law spectrum[12] is a strong indicator of turbulence-like nonlinear behavior in a system.[22] Linear stability theory and similar approaches are therefore inappropriate under these circumstances, unfortunately, because they cannot accommodate the spatiotemporal exchanges in energy exhibited by these nonlinear systems. Kolmogorov[38] first proposed a Rayleigh−Jeans energy spectrum with respect to wavenumber for isotropic hydrodynamic turbulence; the concept has since been applied to wavelike systems, where it has come to be known as wave turbulence.[37,39] Unlike the general case of hydrodynamic turbulence, wave turbulence is analytically tractable because the system can be simplified to a lower order,[37] an approach unavailable in an analysis of the vorticity in hydrodynamic turbulence. Kolmogorov-like wave turbulence solutions may be found for a great variety of systems, but the nature of the solutions and the route to finding them rely upon the Hamiltonian of the system;[40] the lowest-order nonzero nonlinear part of the frequency-domain Hamiltonian determines the number of waves that participate in the interactions. Capillary wave turbulence is a three-wave system; gravity wave turbulence, for example, is instead a four-wave system.

The theory of weak wave turbulence offers three important results pertinent to our capillary wave system: first, the power-law relation between the wave amplitude η and frequency f can be found[22,37] as $\eta \approx \omega^{-17/12}$ through dimensional analysis, as illustrated in the Supporting Information. The second important result is that the inertial range in time or length scale over which this power law applies exists between an energy source (so-called pumping) and an energy sink (fluid damping). Because the cascade represents some transfer of energy from one length scale to another, a stationary solution that obeys the second law of thermodynamics requires an energy source and sink. Finally, it can be shown that the energy flux in wavenumber space must be positive. For capillary wave turbulence, this implies that energy flows in one direction and only one direction: from its source at wavenumber k_0 to higher wavenumbers k_m where the energy is damped. This result is a property of the medium and appears as a consequence of the $-^{17}/_{12}$ power law and the dispersion relation.[41] Other media support inverse cascades, where the pumping wavenumber is greater than the damping, notably Langmuir waves in plasma[42] but not capillary waves on fluids. The power-law relation between wave amplitude and frequency is readily observed: typical experiments in capillary wave turbulence employ a wave generator inside a large tank[19,43] or pistonlike support excitation;[35] however, typical excitation frequencies are less than 10 Hz whereas the capillary wave phenomenon appears at frequencies above this value.

Strong nonlinearities in a capillary wave system can give rise to rather more complex phenomena with a deviation from weak wave turbulence theory and the consequent power-law scaling. Lee et al.[40] found only recently that a renormalization of the dynamics and therefore the dispersion relationship enables the determination of the wave spectra even with strongly nonlinear interactions. They note that a major question with weak wave turbulence theory remains with regard to what happens to real wave systems if any of the assumptions required in weak wave turbulence theory are violated. A platform permitting a broad examination of the capillary wave phenomena, like the one described in this study, would be useful for this purpose.

In this work, we examine the peculiar behavior of capillary waves in a microfluidic system as excited by high-frequency acoustic waves between 500 kHz and 20 MHz. As our reference systems, we will consider the parametric vibration induced by

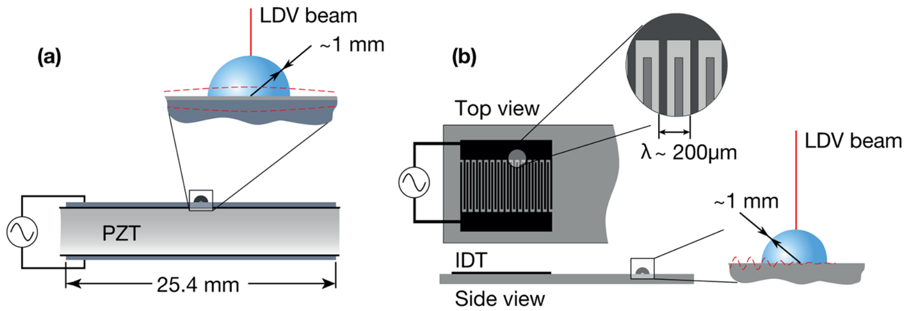

Figure 1. Schematic representation of (a) thickness-mode PZT and (b) SAW device actuation. For PZT, AC power is applied to electrodes on the upper and lower surfaces (upper surface shown in the inset), inducing a thickness-polarized vibrational mode at 500 kHz as indicated by the dashed line. The length scale of this deflection is greater than the contact length scale of the drop, $O(1\text{ mm})$, so the phase under the drop is constant. For SAW, the Rayleigh wave is generated on the piezoelectric LN substrate at 19.425 MHz; the wave damped as energy is absorbed by the fluid (schematically depicted in the inset with the dashed line).

both a thickness-mode lead zirconate titanate (PZT) transducer and a lithium niobate ($LiNbO_3$ or LN)-based surface acoustic wave (SAW) device. These two devices provide different vibrational motions and two mutually exclusive ranges of vibration frequencies—the former 1 to 1000 kHz and the latter 1 to 1000 GHz—and are representative of an entire class of devices finding application with some excitement in micro to nanofluidics.[17] Comparison with wave turbulence theory will show that turbulent capillary waves may be generated by extremely high-frequency excitation, far beyond what is typically reported in the literature, offering a new route to generating and studying these wave phenomena. Furthermore, we explain the underlying phenomena driving the formation of the capillary waves and their characteristics.

GENERATING AND MEASURING MICROSCALE CAPILLARY WAVES

To measure the vibration of a capillary interface, we require a working fluid, an actuation device, and a means of measurement. For the fluid, a 2 μL sessile hemispherical drop of deionized water was placed on a solid substrate set to vibrate to generate capillary waves on the fluid interface as described later. A precision pipette (Eppendorf single-channel adjustable 0.1–2.5 μL pipet, North Ryde NSW Australia) mounted on a translation stage was used to deliver the drop; in between experiments a regular protocol of cleaning using acetone, ethanol, deionized water, and dry nitrogen gas was used to clean the surface. These practices allowed us to avoid variations in the drop size and contact angle from experiment to experiment; the contact angle was verified using a video microscopy system (Infinity K2/SC, Infinity Photo-Optical Company, Boulder, CO) with laboratory-written contact angle measurement software in MATLAB (Mathworks, Chatswood NSW, Australia). The drop size was allowed to vary as a parameter, although its volume was chosen to be less than the volume at which its shape significantly deforms due to gravity,[44,45] with a size less than the capillary length for gravity waves, $l = (\gamma/(\rho g))^{1/2} \approx 3$ mm for water, where γ is the surface tension and g is the acceleration due to gravity.[36] In the absence of gravity, the most relevant fluid property for capillary wave generation is the ratio of surface tension to density, which is fairly constant to an order of magnitude among most simple fluids, making the choice of working fluid somewhat arbitrary.

Two excitation devices were employed: PZT thickness-mode and LN SAW vibration devices. Both devices provide parametric excitation through the substrate but have distinctly different operating frequency ranges and motions; the former as an analog to published approaches and without vibration phase changes across the drop, albeit at a higher frequency of 500 kHz, and the latter as a specific method to examine very high frequency excitation at 20 MHz, though with an unusual excitation motion that possesses rapid spatiotemporal phase changes across the drop. The pair was chosen to assess the effects of the method of excitation on the form of generated capillary waves; the SAW devices in particular are foreseen to be useful in a broad range of micro- and nanofluidics devices that employ fluid interfaces,[17] but we will show the capillary wave phenomena that they impart are also present in the simpler PZT thickness-mode system.

The PZT transducer, illustrated in Figure 1a, possesses a thickness-mode resonance with a wavelength much larger than the contact area of the drop, akin to excitation motions typical in the literature.[23,24,26,30,36,46,47] Thickness-mode PZT transducers can be fabricated to effectively generate motion from a few kilohertz to a few megahertz. The thickness-polarized, face-electroded device used for this work (C-203, Fuji Ceramics, Tokyo, Japan, with Cr–Ag electrodes and dimensions as shown) was driven at its fundamental thickness-mode resonance at 500 kHz ± 2%; the variation is due to ambient temperature changes. Although the amplitude of piston vibration is not uniform throughout such a disc, it was nearly uniform over the area of the drop's footprint. Because the surface of the Cr–Ag electrodes possesses micrometer-scale roughness, a sessile water drop will consistently form a contact angle of about 90°.

The SAW device, illustrated in Figure 1b, generates 19.425 MHz Rayleigh waves on its surface because of the application of a sinusoidal electric signal to an interdigital transducer (IDT) patterned using photolithography on a single-crystal, piezoelectric, single-side-polished, 127.68° y-axis rotated cut, x-axis wave-propagating LN substrate. The frequency was selected with precision through the definition of the simple IDT to resonate at a wavelength of 206 μm: in the simple IDT used here, the fingers and gaps between them all have a width of one-fourth the desired wavelength. In contrast to the PZT device, the SAW is a planar traveling wave, exhibits almost no resonance frequency variation and generally operates at a much higher frequency between about 10 and 1000 MHz. The motion of the surface is elliptical and retrograde. Wave energy may be absorbed into a fluid in contact with the substrate through the phenomenon of "leaky SAW".[48] The principal features of SAW excitation are its high frequency, the fact the substrate undergoes a rapid phase change in its displacement over the propagation path of the SAW, and the presence of in-plane and transverse components of vibration that are one-quarter of a wavelength in time out of phase with each other.

Although SAW provides parametric excitation to a fluid in a manner similar to that described by Miles,[29] it fundamentally differs in that the SAW is a traveling Rayleigh wave with a wavelength so short that the phase of the wave spatially changes underneath the drop. Unlike the traditional wave-maker technique[19] and its variants,[20] under SAW excitation the fluid surface is never directly contacted by any apparatus except at the boundary. This makes SAW far more suitable for microscale devices: directly contacting the drop surface in any location would disturb the dynamics of the surface and possibly the internal fluid flow. SAW devices are capable of extremely narrow-band excitation (the bandwidth is typically <0.005% of the excitation frequency) over a range of frequencies from 1 MHz to 10 GHz, spanning 4 orders of magnitude. The particle velocity at the substrate surface as the SAW passes is typically $O(1\ \text{m/s})$, and displacements scale inversely with the SAW frequency and are generally $O(1-100\ \text{nm})$; acceleration scales with frequency and can be exceptionally large, up to $O(10^7\ \text{m/s}^2)$. If the substrate is rigorously cleaned before use, then the surface is hydrophilic; however, the contact angle was consistently 90° using our procedure of cleaning and droplet deposition.

The excitation level is characterized here by measuring the transverse component of the vibrational amplitude of the substrate. Electrical power input is sometimes selected as a basis for defining the behavior of the capillary waves; it is not appropriate here because the relationship between input power and vibrational amplitude is very nonlinear, being dependent upon the electrical properties and the electromechanical coupling of the specific device, the temperature, and how these devices are mounted and driven. Our surface accelerations are typically between 15×10^6 and $90 \times 10^6\ \text{m/s}^2$ for both devices, corresponding to 1−6 nm SAW amplitudes. Beyond 6 nm, the sessile drop tends to be transported across the substrate surface, so measurement of the capillary wave is no longer reliable. Potentially, the drop could be restrained with a well, but this may introduce a different boundary condition and additional complications in its excitation and was not considered here. However, the electrical power used to drive the drop vibration is generally less than 1 W.[12]

A laser Doppler vibrometer (LDV) (MSA-400, Polytec, Waldbronn, Germany) was used to measure the vibration velocity spectrum of the fluid surface[49] along the axis of the laser's path at its intersection with the measurement surface; the diameter of the measured area is approximately 1 μm.[49] This technique has several advantages over alternatives: high sensitivity (to $O(1\ \text{pm})$); broad measurement range of DC to 25 MHz, where direct imaging or the measurement of scattering through the surface[35] would require extremely high camera frame rates and correspondingly high-intensity illumination; and noncontact measurement, where other techniques in use require intrusive capacitive probes[19] that would distort the geometry of the drop. However, the LDV has some drawbacks worth noting. First, the LDV directly measures vibrational particle velocity along the laser path; this means that the fluid surface must be nearly perpendicular to the incident beam, restricting the area of the drop that we can measure to the central part to avoid substantial errors in measurement.[50] Second, the signal returned by the vibrometer will contain information from all of the laser energy to enter the detector; this includes not only the desired portion that has reflected from the fluid surface but also any light that has passed through the drop and reflected off the substrate. Shrinking the depth of field eliminates this issue; the fluid may also be colored to absorb any light passing through the surface, although the coloring agent will change the properties of the fluid and lower the intensity of reflected light. In practice, the first effect has minimal impact if the measurement point is carefully positioned; colored-fluid experiments demonstrate that the second effect affects only the magnitude of the measurement at the excitation frequency. Third, the limit of 25 MHz in measurement limits our ability to explore SAW excitation to the lower frequency range of such devices, precluding an examination of higher-frequency devices widely available to 1 GHz and beyond. Finally, the LDV is incapable of simultaneously measuring multiple points along the interface: it must scan from point to point, making the measurement at each point one after another. Fortunately, because turbulent capillary waves may be considered to be a nearly ergodic system,[41] temporal and spatial ensemble averaging are considered to be equivalent. To observe the stochastic properties of the system vibration, we must perform some manner of averaging on the spectrum. For this study, we ensemble average 360 spectra recorded sequentially in time at a single point and benefit from the near-ergodicity of the system in the interpretation of the results.

■ VALIDATION OF CAPILLARY WAVE WEAK TURBULENCE THEORY

For deep-water capillary wave turbulence theory to apply to such a system, a number of assumptions must be validated: that the depth of the fluid is much greater than the amplitude of the waves, that gravitational forces may be neglected, that the dispersion relation approaches

$$\omega^2 = (2\pi f)^2 = \frac{\gamma}{\rho}k^3 \tag{3}$$

and that the domain of the wave system be effectively infinite.

The linearized dispersion relation for deep-water capillary-gravity waves is widely accepted to be

$$(2\pi f)^2 = gk + \frac{\gamma}{\rho}k^3 \tag{4}$$

for a flat interface. This implies a crossover frequency f^* from $(2\pi f^*)^2 = 2gk = 2\gamma/\rho k^3$ where gravity and capillary forces have equal importance; for water, this crossover frequency is approximately 10 Hz. At frequencies greater than this, capillary forces dominate and the capillary wave dispersion relation reduces to eq 3.

Equations 3 and 4 are given in the context of an infinitely flat interface. However, we show in the Supporting Information that, subject to interrelated constraints on wavenumber and wave amplitude, the relation for a curved surface typical of drops in our system is identical: for wave amplitudes of $O(10^{-9}\ \text{m})$, eq 3 holds for frequencies $f \ll O(10^7\ \text{Hz})$. We expect the linearized Lagrangian L of the system to take the form $L = a^2G(k, f)$; by minimizing the variation of the Lagrangian, we find $G(k, f) = 0$, giving the dispersion relation. Because the constant component of surface curvature does not participate in the variation, it will have no effect on G; therefore, it does not alter the dispersion relation.

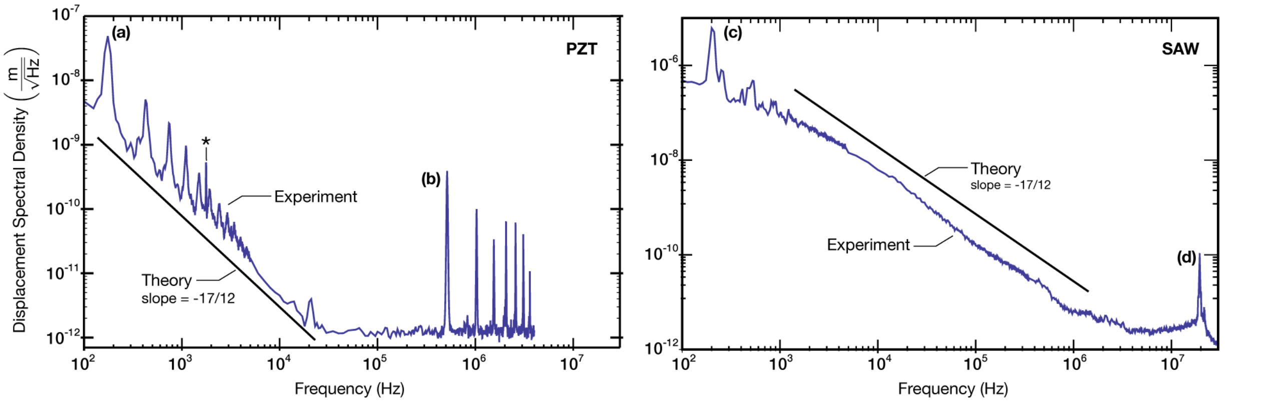

Figure 2. Typical vibrational spectra for 500 kHz thickness-mode excitation via PZT (left) and 20 MHz Rayleigh wave excitation via SAW (right). Over the frequency range of 100 Hz−2 kHz for the PZT and 1 kHz−2 MHz for the SAW, the experimentally measured spectra possess slopes that are remarkably close to $-{}^{17}/_{12}$, the value predicted by wave turbulence theory and indicated by the adjacent line. For PZT excitation, the (a) fundamental vibrational mode is the largest response of the capillary wave, though successive resonances may be seen. The response decays to below the noise floor of the LDV at about 20 kHz, about 2 pm/(Hz)$^{1/2}$, until the (b) Lorentzian response is seen for the excitation frequency of 500 kHz. Note the absence of subharmonic resonances but the presence of superharmonic resonances at $2f$, $3f$,..., $7f$; for the SAW device, the superharmonics, if present, lie beyond the upper measurement limit of the LDV, 25 MHz. The (c) fundamental mode exhibits the largest response of the system to (d) SAW excitation at (b) 20 MHz, and the response at the excitation frequency is a Lorentzian distribution.

The assumption of infinite depth, however, requires the product of wavenumber k and depth below the resting surface position h to be $kh \gg 1$. Neglecting gravity, we assume h is of the same order as the radius of the drop, $O(1$ mm$)$ in our system; through the dispersion relation, the frequency associated with the limiting wavenumber is approximately 40 Hz.

The deep-water capillary wave dispersion equation can therefore be applied to a drop of radius $O(1$ mm$)$ over the frequency range of 40 Hz $\ll f \ll 10^7$ Hz; we will show that all observed wave turbulent phenomena in our system occurs in this range.

As a final criterion for the applicability of wave turbulence theory, we must consider the effect of the pinned contact line and the finite domain. Wave turbulence theory assumes an infinite unbounded surface; however, such a domain is impractical for both numerical and experimental investigations. It has been shown that boundary effects do not dominate the response on all length scales when the system is in a disordered state.[35] However, so-called "frozen turbulence", where wave energy is concentrated at resonant points in the spectrum, may occur where the length scale of the boundary is close to the length scale of interest,[51] although it is possible for frozen turbulence to exist side-by-side with broad-band turbulence.[52] Fortunately, in our system the length scale of the boundary, $O(1$ mm$)$, is larger than the capillary wavelength at frequencies beyond 1 kHz. Furthermore, we are measuring the deflection in the center of the drop, the point farthest from the boundary possible. We expect that, as in past work,[35,51,52] the domain may be considered to be effectively infinite for the capillary waves we are examining.

■ RESULTS

The LDV provides displacement spectral density distributions with respect to frequency from 100 Hz to 25 MHz, as illustrated in typical response spectra provided in Figure 2 for both the thickness-mode PZT device and the Rayleigh-wave-generating SAW device. As previously observed,[8,12,49] note the presence of a capillary wave at the excitation frequency, which is remarkable given the very high frequency of the excitation in either case. This cannot be the response of the substrate because the focal depth is sufficient only to measure the fluid interface, the amplitude of vibration of the substrate is far lower than what is measured here, and the width of the response peak at the excitation frequency is far more narrow for the substrate: the response of the substrate absent the drop is identical to the noise floor except for a narrow peak at 500 kHz for the PZT and 20 MHz for the SAW device to a displacement spectral density amplitude on the order of 10^{-11} m/(Hz)$^{1/2}$. As remarkable as the absence of a capillary wave at one-half the excitation frequency is, there is no Faraday wave. Finally, note the presence of the capillary wave response at (Hertz to kiloHertz order) frequencies far lower than the excitation frequency.

The capillary waves do appear to behave as a system driven by weak wave turbulence. The majority of the spectra is dominated by a response cascade that follows a power-law relationship $\eta \approx f^{-1.420}$, which is closely correlated to the capillary wave turbulence theory-derived $-{}^{17}/_{12} \approx -1.417$ slope (R^2 = 0.990 and 0.998 for the PZT and SAW devices, respectively), providing strong evidence of capillary weak wave turbulence in the system for two rather entirely different modes of fluid interface excitation.

The progression of resonance responses from the fundamental resonance frequency at about 200 Hz upward appears according to the well-known[3,18,45] Lamb model of elastic resonance of a spherical capillary surface[53]

$$f_m = \sqrt{\frac{(m+1)(m+2)(m+4)\gamma}{3\pi\rho L^3}} \tag{5}$$

providing the frequencies f_m = {247, 479, 742, 1030, 1350, 1700,...} Hz that correlate well with the experimentally measured fundamental and higher-order resonances driven by the PZT device, f_m = {176, 432, 735, 1140, 1470, 1940,...} Hz (R^2 = 0.999), and the SAW device, f_m = {259, 506, 668, 862, 1220, 1688,...} Hz (R^2 = 0.990): there appears to be no need to resort to more thorough analysis provided elsewhere.[54] Note that there is nothing in these calculations that involve the excitation frequency. Furthermore, there is a strong correlation (R^2 = 0.961) between the experimentally and theoretically

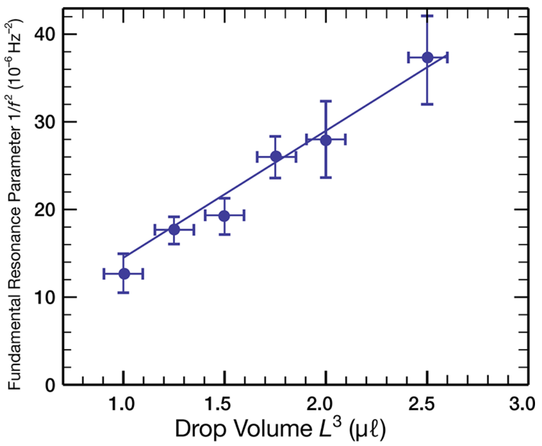

Figure 3. Driven by SAW at 20 MHz, the fundamental resonance frequency of the drop scales with the cube of its length according to eq 5 with good correlation (R^2 = 0.961); the line through the data represents the theoretical prediction. The error bars are twice the standard error; the horizontal error is due to pipetting because there were no observable evaporation effects for the short duration of each experiment. The data taken at each drop volume appear to be normally distributed (n = 5).

determined fundamental resonance frequencies as the drop size is varied with the SAW device as illustrated in Figure 3. The exceptionally narrow peak indicated with an asterisk in Figure 2 is a specious single data point appearing as a result of the encoder card's analog-to-digital conversion at 2^{14} = 16 384 Hz; its absence in the SAW-driven response is simply due to the stronger response driven by this system that acts to hide it under those conditions. Notably, there are no subharmonic cascades in this system from the excitation frequency at 500 kHz. These results, taken together, indicate that the system does not exhibit Faraday waves[31] but, more importantly, that it is behaving as a weak wave turbulent system.

For PZT excitation there are superharmonic resonances from the excitation frequency of 500 kHz upward in a rational cascade $\{f, 2f, 3f,...,7f\}$, and perhaps beyond if not for encountering the upper limit of the LDV's measurement range at this resolution. The upper limit of the LDV's measurement range prevents us from determining whether there are similar superharmonic resonances in the SAW-driven system.

The ultimate source of energy in this system is the parametric excitation from either the SAW or PZT device; indeed, the excitation frequency is strongly indicated by Lorentzian distributed drop surface vibration responses (Figure 2b,d; Lorentzian response distributions are characteristic of a second-order linear system with viscous damping). However, the strongest response whatever the excitation method appears at the drop's fundamental resonance frequency (Figure 2a,c). The drop visibly vibrates in either case at its fundamental resonance under most conditions. The Faraday wave mechanism is clearly not the cause: the excitation of the fundamental drop resonance represents a downshift from the excitation frequency of 3 orders of magnitude for the PZT device and 5 orders of magnitude for the SAW device, far beyond the usual $^f/_2$ treatment resulting from the standard Mathieu equation-based result of Faraday wave analysis. Furthermore, the capillary wave resonance frequencies clearly do not depend on the frequency of excitation, at odds with Faraday wave theory, which is not surprising given the violation of a fundamental precept of the theory in eq 2. However, the capillary wave resonances that appear at 200 Hz in Figure 2 are a consequence of the physics of the capillary surface itself, as made clear by Figure 3.

We have shown here that the capillary waves behave under weak wave turbulence theory: their amplitude exhibits a Rayleigh–Jeans distribution of $\eta \approx f^{-17/12}$ over a broad frequency range. We will show now that this is actually a cascade from the fundamental capillary wave resonance frequency upward in a one-way response. It is tempting to assume that energy is transported from the high-frequency excitation to eventually give rise to the fundamental resonance of the drop through a subharmonic cascade (akin to Lauterborn and Cramer[55]). However, the direction of energy flux for capillary wave weak turbulent spectra is always toward higher frequencies: for capillary waves, the energy source for the cascade must be at a lower frequency than the cascade itself, as discussed above.

The wave-turbulent cascade in our system is therefore not the mechanism by which the absorbed acoustic energy is shifted in frequency to the fundamental resonance: it cannot be because the excitation frequency is far higher than that of the entire capillary wave cascade. In fact, the cascade forms as a consequence of vibration at the fundamental resonance. It is apparent that the mechanism responsible for the downshift does not interfere with the capillary wave response at frequencies between the excitation and the fundamental mode because the surface exhibits the theoretical stochastic wave-turbulent response.

So the question turns from why the response appears as it does to why it is excited in the first place. Focusing now upon the use of SAW excitation alone, we show that a mechanism permitting a subharmonic cascade from the high excitation frequency to the low capillary wave frequencies is the formation of turbulence in the acoustic streaming induced by the SAW within the fluid bulk.

The streaming Reynolds number[17] as a consequence of SAW excitation of the water drop is first examined,

$$Re_{st} = \frac{\rho \hat{u} L}{\mu} = \frac{\rho(u_{hyd} + u_{vib})L}{\mu} = \frac{\rho[u_{hyd} + (2\pi f x_{vib})]L}{\mu} \approx \frac{(10^3)[(10^{-1}) + (2\pi 10^7 10^{-9})](10^{-3})}{(10^{-3})} \approx 10^3 \quad (6)$$

where ρ, L, μ, and $\hat{u} \equiv u_{hyd} + u_{vib}$ are the fluid density, drop size, fluid dynamic viscosity, and Lagrangian fluid velocity, respectively, with the last term being defined as a summation of the hydrodynamic fluid velocity in the drop, as observed to be on the order of $u_{hyd} \approx 0.1$ m/s, and the vibrational particle velocity as the acoustic wave passes through the fluid, u_{vib}, being dependent upon the amplitude of the vibration of the substrate. The streaming Reynolds number for PZT excitation is similar because the Lagrangian fluid velocity is the same though the excitation frequency is 2 orders of magnitude lower.

A streaming Reynolds number of $Re_{st} \approx 10^3$ is well above the critical values of $\sim 10^2$ reported in the literature (and nicely explained in the classic article by Thompson et al.[56]) predicting a transition to subharmonic, period-doubling cascades of turbulent fluid flow[55] that result in chaotic flow behavior. It is important to distinguish these results from those typical of the hydrodynamic Reynolds number where there a transition to turbulence appears, for example, at 10^5 in "typical" pipe flow.[57]

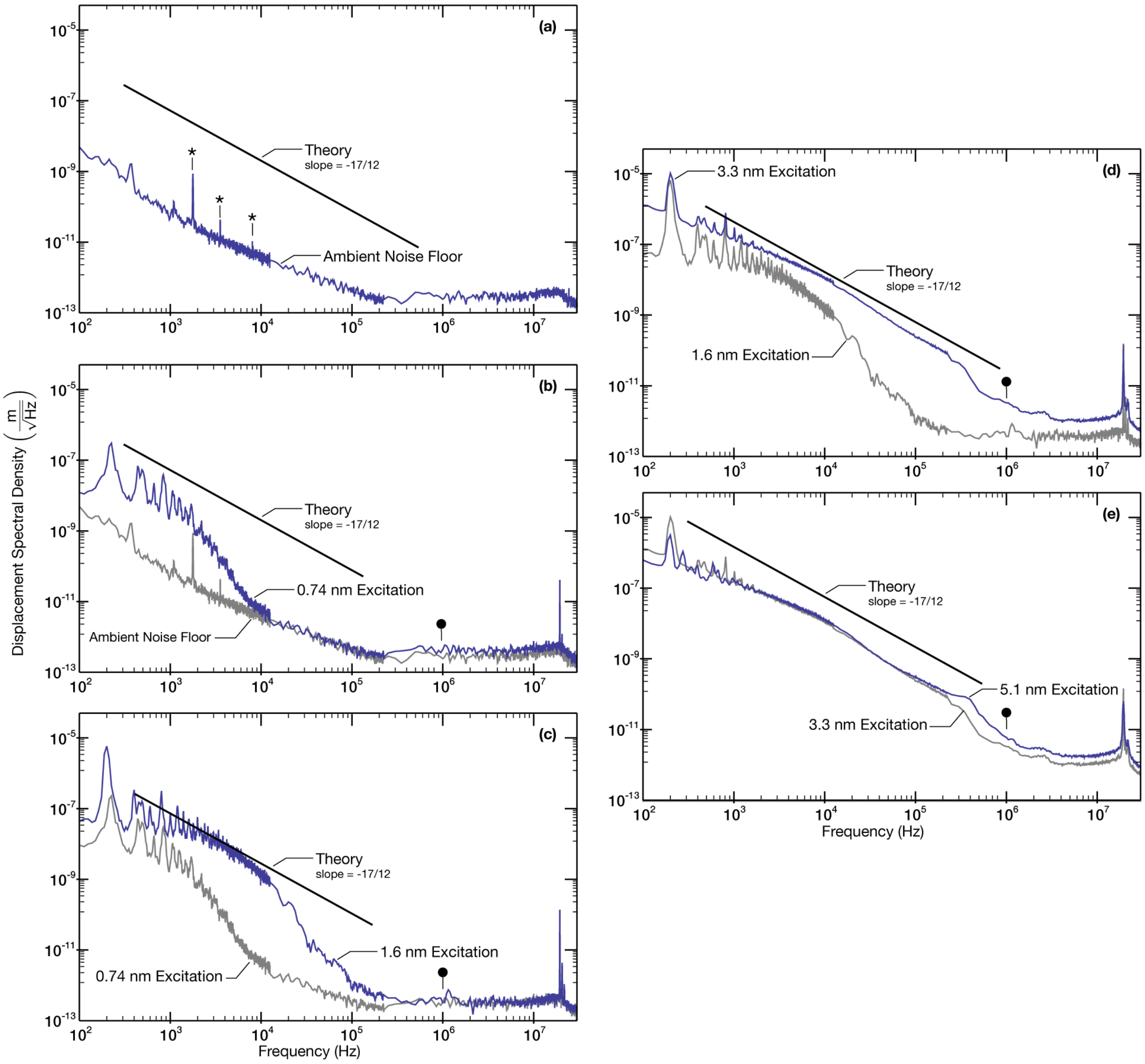

Figure 4. Evolution of the spectrum with increasing SAW excitation amplitude: (a) ambient noise floor without explicit vibration excitation, (b) 0.74 nm by comparison to ambient, (c) 1.6 nm excitation compared to 0.74 nm excitation, (d) 3.3 nm excitation compared to 1.6 nm, and (e) 5.1 versus 3.3 nm excitation.

The acoustic streaming can be shown to have become turbulent via another route. Lighthill[58] shows quite eloquently how turbulent Stuart streaming appears, with convective acceleration becoming significant, at only a few microwatts at 1 MHz. When a very conservative estimate is taken for our system assuming that turbulent Stuart streaming appears when the acoustic streaming power, P, is above a critical value $P_{\text{crit}} = 1$ mW, the power of the acoustic beam propagating into the fluid drop is $P c\rho u_{\text{vib}}^2 = c\rho f^2\eta^2$, where c is the speed of the acoustic wave in the fluid, resulting in $\eta_{\text{crit}} = (1/f)(P_{\text{crit}}/(\rho c))^{1/2} = 0.61$ nm by solving for η_{crit} and substituting in typical values for water. This shows that for our SAW system we have turbulent acoustic streaming in all cases, with the 0.74 nm amplitude case near the critical amplitude, if we assume throughout that the amplitude of the acoustic wave is of the same order of magnitude as the amplitude of the SAW the drop resides on, an assumption that we have found to be valid elsewhere.[59]

The matter of turbulent acoustic streaming is important because it is key to explaining how very high frequency acoustic excitation can result in such low-frequency acoustic capillary waves, especially when there is no known mechanism to drive a subharmonic cascade in the capillary waves themselves. Unlike the capillary wave phenomena, a large-scale subharmonic cascade is possible in a turbulent jet,[55,56,58] and this appears to underlie the driving of low-frequency capillary waves by the very high frequency boundary vibration. The SAW-driven generation of turbulent acoustic streaming in improving mixing in drops with geometries similar to those in this study has been shown to appear over a wide range of viscosities and powers,[9] indicating how the bulk of the entire drop becomes turbulent through acoustic streaming.

Other potential mechanisms of subharmonic coupling including interactions in the viscous boundary layer, bubble formation and collapse or interactions in and around these bubbles, and other instability mechanisms, especially the Kelvin–Helmholtz instability.[60] All of these are unlikely: the first would require all of the energy of the capillary wave cascade to be transmitted through an exceedingly small part of

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the fluid drop's volume because the viscous boundary layer has a thickness only on the order of 10 nm. The second potential mechanism has a similar volume constraint in addition to the fact that it is known that bubbles are not formed with SAW excitation,[12] as opposed to what happens at much lower frequencies of excitation.[6,7] Kelvin−Helmholtz instabilities appear only upon sufficient flow speed at the fluid interface; the critical flow speed for our system would be $u_{crit} = (2(\gamma g \rho)^{1/2})^{1/2} = 7.2$ m/s, where γ is the air−water surface tension and g is the gravitational acceleration.[60] Even presuming that the critical flow speed did not apply here, perhaps because of the presence of turbulence, suggests that the wavelength of the least-stable wave due to the acoustic streaming flow is over 10 cm, well beyond the scale of our drop, suggesting that this form of instability is not responsible for the capillary waves we see.

However, the observation that the bulk flow and capillary wave are coupled through shear is well known in the literature, as found by Longuet−Higgins[61] and shown convincingly by Trinh and Wang;[62] though the typical treatment of the phenomenon is the generation of the bulk flow by the capillary waves, here we see evidence of the process occurring in reverse in a manner still consistent with their studies. In the absence of any detailed analysis of the formation of capillary waves due to bulk flow, if we take the liberty of considering the classical solution of Stokes' drift[61] due to capillary waves, $u = \eta^2 \omega k e^{-2kz}$ where z is the distance into the flow of the deep bulk (using a shallow treatment prevents solution here) and note that the Stokes' drift in this context is also the time-averaged fluid flow in the bulk u induced by the turbulent acoustic jet, then we can determine what order the velocity of the turbulent jet would have to be to induce the fundamental capillary waves that we observe. With the relationship as written, $u \approx \eta^2 \omega k = \omega^{-11/6} k$ if we note the capillary weak wave turbulence relation $\eta \approx \omega^{-17/12}$. Because the fundamental vibrational mode is at about 250 Hz and its wavelength is about 1 mm, $u \approx 10$ mm/s. If instead we chose to note that the typical amplitude of the fundamental vibrational mode is on the order of 100 μm, again $u \approx 10$ mm/s. Either way, the turbulent acoustic jet is driving the flow in the drop at these velocities and beyond and the turbulence of the acoustic streaming is present to drive the formation of the capillary wave at these frequencies, so via this simplistic treatment it appears entirely possible to drive capillary waves of the form that we see with the acoustic streaming we know to be present in the drop. Certainly more appropriate and detailed analysis, properly incorporating the nonlinearities in lieu of reference to classical theory, would provide a more detailed picture of the mechanism responsible for the capillary waves.

We turn now to an examination of the dynamics of the drop through its vibrational spectrum at different excitation levels. Figure 4a indicates the noise floor of the LDV, in other words, the minimum displacement spectral density as a function of frequency, ambient noise, thermal noise, analog-to-digital conversion limitations, and the shot noise of the measurement system. These culminate in a constant velocity, $^1/_f$, noise over the range of 10^4 to 10^5 Hz resulting from the limitations of the analog−digital encoder hardware in the LDV, whereas beyond 10^5 Hz the noise floor is defined by a constant value of about 0.5 $/(\mathrm{Hz})^{1/2}$ due to shot noise at the photodiode of the LDV. The asterisks refer to specific peaks present as a result of intrinsic encoder difficulties (as with all such encoders) at 2^{13}, 2^{14}, and 2^{15} Hz that are not true resonance peaks. The noise floor coincidentally matches the theoretical slope from 300 to 2000 Hz but does not match elsewhere.

The capillary wave response transforms from a discrete to a continuous response as the SAW amplitude is increased. Upon application of SAW to the drop at 20 MHz, the response at that same frequency appears as a narrow peak in Figure 4b, which grows as the excitation amplitude is increased to 1.6 nm but remains relatively constant as the excitation amplitude is increased further to 5.1 nm: the energy is moving elsewhere in the spectrum. As the excitation level increases, the response of the drop at low frequencies begins to saturate (c); energy then enters the wave turbulent cascade (d). At a high excitation level (e), the turbulent cascade is fully established over an inertial range spanning at least 10^3−10^6 Hz. Another indicator of the turbulent response is the smoothed broad-band response at the highest excitation levels (d, e) from the discrete resonances that appear at relatively lower excitation amplitudes (b, c), except for the ever present and substantial fundamental resonance of the drop at around 200 Hz.

Figure 4b−e illustrates this by a comparison of the displacement response due to a given level of excitation to the next lowest level of excitation; Figure 4c, for example, shows that the response to 1.6 nm excitation is very similar to the 0.74 nm response between 100 and 1000 Hz but is very different between 1000 Hz and 50 kHz. A low-frequency vibration appears at even the lowest excitation level that we could sustainably operate, 0.74 nm, and this response propagates upward in frequency to fill in the turbulent cascade from 200 Hz to eventually 1 MHz as the excitation is increased to 5.1 nm. The discrete response of the system and absence of a complete cascade at 0.74 nm indicates that we are likely on the lower bound of excitation of the turbulent acoustic streaming and consequently the formation of the capillary waves (predicted to occur at 0.61 nm above); future work could consider this transition in far more detail. Curiously, there is little difference between excitations of 3.3 and 5.1 nm in Figure 4e other than the appearance of a "hump" in the higher-amplitude response that appears to be correlated to the appearance of atomization phenomena, an aspect for consideration in a separate work. It is important to remember the effect of operating with such high excitation frequencies with regard to the incredible accelerations imparted to the drop: though the displacements are only 0.74, 1.6, 3.3, and 5.1 nm, the respective accelerations are 11×10^6, 23×10^6, 50×10^6, and 76×10^6 m/s^2 for these four levels of excitation.

We should expect the inertial range to end with wave energy being rapidly damped beyond some threshold wavenumber, as shown in other studies.[43] In such a case, the cascade will steepen in descent away from the theoretical gradient. We see this behavior in Figure 4b from 10^3 to 10^4 Hz, in Figure 4c from 10^4 to 10^5 Hz, and in Figure 4d from 5×10^5 to 10^6 Hz. The presence of the noise floor of the LDV precludes our ability to measure this phenomena beyond about 1 MHz. The vibrometer's claimed displacement resolution at megahertz-order frequencies is less than 0.1 pm/Hz$^{1/2}$; our measurements show that the spectrum becomes constant at a noise floor of $O(0.1 \text{ pm/Hz}^{1/2})$ at $O(10^7 \text{ Hz})$. An alternate perspective is provided by calculating the Kolmogorov viscous time scale in water, $\tau = (\nu/\varepsilon)^{1/2} \approx O(1 \text{ MHz})$; 1 MHz has been indicated with a solid circle in Figure 4b−e, nominally representing the end of the capillary wave cascade, which appears to be consistent with the experimentally measured results.

The evolution of the low-frequency resonance with increasing excitation level is also apparent in Figure 4. The harmonic series of low-frequency resonances is quite typical of narrow-band pumping:[22,41] the pumping energy is localized around a specific frequency or characteristic length, in contrast to wide-band pumping where the pumping is spread over a range of frequencies, an extreme example being the excitation of drops by white noise.[63] Here, the energy is initially concentrated at the frequency of pumping; interactions between two waves generated by the pumping lead to energy transfer to the next harmonic in the series, and similar processes lead to the creation of ever shorter and wider harmonic peaks that gradually merge with the background level of the spectrum at higher frequencies. All of the spectra, regardless of the excitation amplitude, exhibit zero-frequency side bands, exponential decay, and power-law behavior, phenomena that according to Xia and Punzmann[35] imply the presence of turbulence throughout. Turbulence is already present from low excitation levels; increasing the amplitude of excitation acts to distribute the power to higher frequencies.

The low-frequency interactions are governed by the same weakly nonlinear mechanics as the cascade at higher frequencies. However, the effects of eigenmode resonances of the drop surface and initial energy concentration at the fundamental vibrational mode combine to limit the interactions between waves, constrained by the three-wave resonance conditions $f_1 = f_2 + f_3$ and $\mathbf{k}_1 = \mathbf{k}_2 + \mathbf{k}_3$, to a set of discrete harmonics. We observe the harmonic series, steadily decreasing in magnitude with higher frequency, superimposed upon a broad-band level decreasing at a slower rate than the harmonic peaks. This is evident in Figure 4b,c; as the excitation amplitude increases beyond 1.6 nm, the spacing between adjacent resonances becomes narrower in the frequency domain.

CONCLUSIONS

We have described a method to parametrically generate and measure capillary wave phenomena on the microscale using laser Doppler vibrometry, megahertz-order SAW actuation, and kilohertz-order thickness-mode actuation, inducing fluid flows that exhibit strongly inertial behavior. Capillary waves appear at the frequency of excitation as a linear response to the excitation but also, and most remarkably, appear at much lower frequencies, from the fundamental resonance frequency upward as a discrete modal response at low excitation amplitudes that gives way to a continuous broad-band response.

The turbulent Rayleigh–Jeans power-law cascade and pumping characteristics in our system behave according to weak wave turbulence theory, despite the fact that the excitation frequency is substantially higher than the capillary wave response frequencies, regardless of whether the system is driven through thickness vibration or surface acoustic wave vibration and despite the exceptionally large accelerations induced in the system. Notably, strongly nonlinear phenomena that might have renormalized the expected Zakharov–Kolmogorov turbulence spectrum[40] were not seen, a surprise considering the extreme accelerations induced by the SAW. The key aspects of weak turbulence theory that this system was observed to conform to include the following: (1) The capillary wave amplitude is $\eta \approx f^{-17/12}$ between the energy source frequency and the energy sink frequency. (2) The energy flux in wavenumber space must be positive: energy flows from a source at a low k_0 to a high k_m; $k_m > k_0$ is required and therefore the capillary wave energy must flow from low frequencies to high frequencies.

Finally, no Faraday wave response was detected in this system, a consequence of the excitation frequency being far greater than the observed capillary wave frequencies, which is atypical in many experiments studying capillary wave phenomena but a feature of many new acoustically driven microscale devices under consideration.

Through the one-way nature of capillary wave weak turbulence with energy flow from low to high frequencies, we may infer that the fundamental capillary wave resonance appears to be directly driven by the parametric excitation at very high frequency through acoustic streaming-driven chaotic flow in the fluid bulk. The acoustic streaming was found to be turbulent because of the presence of convective acceleration when the induced vibrational amplitude for the SAW was greater than 0.61 nm over the entire measurement range of this study. Though alternative routes of coupling to form capillary waves from the incident acoustic wave were considered, the most likely candidate remains the turbulent acoustic streaming-inducing bulk flow within the drop that drives the generation of the capillary waves through shear. In any case, the turbulence in the jet can support energy flow from high excitation frequencies to the low frequencies of the capillary wave phenomena, and it is this mechanism that drives the subsequent formation of the low- to high-frequency capillary wave cascade.

ASSOCIATED CONTENT

Supporting Information

Capillary wave weak turbulence and the scaling of wave energy. Dispersion relations on a curved surface and on a flat surface. Derivation for dispersion on a spherical surface. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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