High-Frequency Nanofluidics: An Experimental Study using Nanomechanical Resonators

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Abstract

Here we apply nanomechanical resonators to the study of oscillatory fluid dynamics. A high-resonance-frequency nanomechanical resonator generates a rapidly oscillating flow in a surrounding gaseous environment; the nature of the flow is studied through the flow-resonator interaction. Over the broad frequency and pressure range explored, we observe signs of a transition from Newtonian to non-Newtonian flow at $\omega\tau \approx 1$, where $\tau$ is a properly defined fluid relaxation time. The obtained experimental data appears to be in close quantitative agreement with a theory that predicts purely elastic fluid response as $\omega\tau \rightarrow \infty$.

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The Navier-Stokes equations based upon the Newtonian approximation have been remarkably successful over the centuries in formulating solutions for relevant flow problems both in bulk and near solid walls [1]. The Newtonian approximation breaks down, however, when the particulate nature of the fluid becomes significant to the flow. The Knudsen number, $Kn = \frac{\lambda}{L}$, is one parameter, which is commonly used to settle whether the Newtonian approximation can be applied to a medium or not. Here, one compares the mean free path $\lambda$ in the medium to an *ill-defined* characteristic length $L$. A second defining parameter, especially for oscillatory flow, is the Weissenberg number, $Wi = \frac{\tau}{T}$, which compares the characteristic time scale $T$ of the flow with the relaxation time $\tau$ in the medium. As $\tau/T = \omega \tau$ is varied — for instance, by varying the flow frequency $\omega$ or the relaxation time $\tau$ — the nature of the flow changes drastically.

Recent developments in nanometer scale engineering have created a vibrant subfield of fluid dynamics called nanofluidics [2]. Most nanofluidics work is concerned with flow in nanoscale channels and remains strictly in the Newtonian regime. In contrast, emerging nanometer scale mechanical resonators [3, 4], with frequencies already extended into the microwaves [3, 6], offer an uncharted parameter space for studying nanofluidics. For a high-frequency nanomechanical resonator with resonance frequency $\omega/2\pi$, one can tune $\omega \tau$ over a wide range — in fact, possibly reaching the limits of the Newtonian approximation in a given liquid or gas. This not only allows experimental probing of a flow regime that was inaccessible by past experiments [7, 8], but also presents the unique prospect of designing nanodevices for key technological applications.

To complement the recent theoretical interest in high frequency nanofluidics [2, 9, 10, 11], we experimentally studied the interaction of high-frequency nanomechanical resonators with a gaseous environment. The gaseous environment presents an ideal fluid for these studies, where one can effectively tune $\tau$ by changing the pressure $p$. On the other hand, varying the resonator dimensions changes the mechanical resonance frequency $\omega/2\pi$. When combined, the two experimental parameters allow $\omega \tau$ to be varied over several orders of magnitude effectively.

In order to cover a broad frequency range, we fabricated silicon doubly-clamped beam resonators with varying dimensions $w \times h \times l$ displayed in Table I using standard techniques [12]. To further extend the frequency range, we employed fundamental and first harmonic modes of two commercial silicon AFM cantilevers (Table I). For the measurements, we used

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TABLE I: Device parameters, transition pressure $p(W_i = 1)$ and the approximate lower pressure limit $p_{min}$ for accurate measurements for the devices used in the study. 1$^{\text{st}}$ harmonic mode was also employed for some AFM cantilevers.

| $(w \times h \times l)$ (µm) | $\omega_0/2\pi$ (MHz) | $Q_0$ | $p(W_i = 1)$ (Torr) | $p_{min}$ (Torr) |
|-----------------------------|------------------------|-------|----------------------|------------------|
| $53 \times 2 \times 460$ (1$^{\text{st}}$ Harmonic) | 0.078 | 8321 | 1.0 | 0.05 |
| $36 \times 3.6 \times 125$ (Fundamental) | 0.31 | 8861 | 3.0 | 0.05 |
| $36 \times 3.6 \times 125$ (1$^{\text{st}}$ Harmonic) | 1.97 | 3522 | 17.5 | 0.06 |
| $0.50 \times 0.28 \times 17.1$ | 10.4 | 1840 | 110 | 1.9 |
| $0.50 \times 0.28 \times 11.2$ | 18.1 | 1530 | 200 | 2.9 |
| $0.93 \times 0.22 \times 9.9$ | 22.8 | 1335 | 176 | 0.8 |
| $0.76 \times 0.22 \times 9.9$ | 22.9 | 1200 | 216 | 1.2 |
| $0.23 \times 0.20 \times 9.6$ | 24.2 | 415 | 280 | 2.77 |
| $0.50 \times 0.28 \times 9.1$ | 27.1 | 909 | 290 | 2.6 |
| $0.32 \times 0.20 \times 7.7$ | 33.2 | 780 | 320 | 15.8 |
| $0.50 \times 0.28 \times 5.9$ | 45.7 | 1066 | 310 | 2.3 |
| $0.25 \times 0.20 \times 5.6$ | 53.2 | 571 | 400 | 1.2 |
| $0.73 \times 0.23 \times 5.6$ | 58.6 | 525 | 490 | 19.0 |
| $0.24 \times 0.20 \times 3.6$ | 102.5 | 495 | – | 11.9 |

a pressure-controlled optical characterization chamber connected to a high purity N$_2$ source. We actuated the out-of-plane modes of the resonators electrostatically and measured the displacements optically [12]. All measurements were performed under linear drive; moreover, the results remained independent of the rms displacement amplitudes of 1-10 nm as confirmed by Michelson interferometry. Figure [11] depicts the typical resonant response of a nanomechanical resonator as the background N$_2$ pressure in the chamber is increased. The frequency shift is due to the mass loading from the boundary layer [23], while the broadening results from the energy dissipation in the fluid. The analysis can be simplified by using a one-dimensional damped harmonic oscillator approximation [13], $\ddot{x} + \gamma \dot{x} + \omega^2 x = f/m$, where $f/m$ represents the force per unit effective mass of the resonator. The quality fac-
tor $Q$, which is a comparison of the stored energy to the dissipated energy per cycle, is related to $\gamma$ as $\gamma \approx \omega/Q$. Here, we extracted both the resonance frequency $\omega/2\pi$ and $Q$ using nonlinear least squares fits to the Lorentzian response of the resonator. In addition, for low-$Q$ (high pressure), we verified the Lorentzian fit results through fits to the real and imaginary components of the complex transmission [14]. Typical changes in $\omega$ and $Q$ of a nanomechanical resonator during a pressure sweep are shown in the inset of Fig. 1. Both $\omega$ and $Q$ approach their respective intrinsic values, $\omega_0$ and $Q_0$, at low pressure.

Before presenting further results, we must clarify the nature of the fluidic energy dissipation. The motion of the fluid with respect to the solid boundary creates a complex, position-dependent shear stress on the resonator surface. The inertial and dissipative components of the net shear force are proportional to the displacement and the velocity, respectively. For a single device, as the pressure is changed, the fluidic dissipation can be quantified by either the fluidic quality factor $Q_f$ given by $Q_f^{-1} = Q^{-1} - Q_0^{-1}$ or the fluidic dissipation $\gamma_f = \omega/Q_f$. To compare different devices with varying sizes and geometries, one needs to further realize that the fluidic dissipation is proportional to the effective surface area $S_{eff}$, while the stored energy in the resonator is proportional to the effective mass $m$ [24]. With this naive assumption, we define a normalized fluidic dissipation, $\gamma_n = \gamma_f m/S_{eff}$. The lower pressure limit $p_{min}$ for accurate $Q_f$ measurement is set by $Q_0$: as one approaches $p_{min}$, the intrinsic losses in the resonator dominate the measurement. The upper limit is 1000 Torr. Table I displays $p_{min}$ for each device along with $Q_0$.

The normalized fluidic dissipation $\gamma_n$ observed in three different resonators are presented in Fig. 2(a), (b) and (c) as a function of gas pressure. A change in the slope is noticeable for the data in (a), and (b) at approximate pressures of 1 and 300 Torr, respectively. The turn points marked in the plots correspond to $\omega \tau \approx 1$, and is discussed in detail below. For the highest frequency beam at 102.5 MHz shown in (c), the turn point falls outside the available pressure range, i.e., 1000 Torr. Molecular flow model [10], which takes into account specular collisions, fits our data only at the ideal gas limit at low pressure. Note that a multiplicative constant of 0.9 was used in all three to improve the fits. Viscous effects [15, 16] and squeeze-film effects [17, 18], commonly observed in MEMS, did not introduce significant damping for the small high-frequency devices up to atmospheric pressure.

The solid lines in Fig. 2 are fits to a theory by Yakhot and Colosqui [9] developed from the Boltzmann Equation in the relaxation time approximation. After geometric normalization
FIG. 1: Resonance of a silicon doubly-clamped beam of width $w = 500 \text{ nm}$, thickness $h = 280 \text{ nm}$ and length $l = 11.2 \mu \text{m}$ at various $N_2$ pressures in the chamber: $p = 0.049, 5.4, 32, 100, 302$ and 942 Torr. The inset shows the extracted quality factor $Q$ and normalized resonance frequency $\omega/\omega_0$ of the same device as a function of pressure. Here, $Q_0 \approx 1530$.

FIG. 2: Normalized fluidic dissipation $\gamma_n$ as a function of pressure for (a) a cantilever with dimensions $(w \times h \times l) 53 \times 2 \times 460 \mu \text{m}$, and nanomechanical doubly-clamped beams with dimensions of (b) $230 \text{ nm} \times 200 \text{ nm} \times 9.6 \mu \text{m}$ (c) $240 \text{ nm} \times 200 \text{ nm} \times 3.6 \mu \text{m}$. Resonance frequencies are as indicated and the approximate turning points are marked with arrows. The lines are fits to molecular collision model [10] and Eq. [9] using $\tau \approx 1850/p$ [9]. The molecular collision and the Yakhot-Colosqui predictions were multiplied by 0.9 and 2.8, respectively, for all resonators. (d) Relaxation time $\tau$ as a function of pressure. The points were extracted from fluidic dissipation data sets, such as those shown in (a) and (b), of 13 resonators. The solid line is a least-mean-squares fit and indicates that $\tau \approx 1850/p$. 
FIG. 3: (a) Normalized fluidic dissipation $\gamma_n$ as a function of the resonator frequency $\omega_0$ for several resonators at four different pressures. From top to bottom, $\tau \approx 2.3, 4.6, 9.2, 18.5$ ns. The lines are fits calculated using Eq. 1. Wi $\approx$ 1 points are marked with an arrow for each pressure. (b) The scaling of $\gamma_n/\sqrt{\omega \mu \rho_f}$ for all resonators with $\omega \tau$. Each symbol corresponds to an individual resonator. All the predictions were multiplied by the same fitting factor of 2.8 (also see Fig. 2).

and imposing the no-slip boundary condition, this theory culminates in the expression

$$\begin{align*}
\gamma_n \approx & \frac{1}{(1 + \omega^2 \tau^2)^{3/4}} \sqrt{\frac{\omega \mu \rho_f}{2}} \left[(1 + \omega \tau) \\
& \cos \left(\frac{\tan^{-1} \omega \tau}{2}\right) - (1 - \omega \tau) \sin \left(\frac{\tan^{-1} \omega \tau}{2}\right) \right].
\end{align*}$$

In Eq. 1, $\gamma_n$ is expressed in terms of the viscosity $\mu$, the density $\rho_f$, the effective relaxation time $\tau$ of the fluid, and the frequency $\omega$ of the resonator. The only unavailable parameter is the relaxation time $\tau$. In order to obtain the fits, we assumed that $\tau$ satisfied the empirical form, $\tau \propto 1/p$ [8, 19]. The key prediction of the Yakhot-Colosqui [9] theory is that the turn point in $\gamma_n$ occurs when $\tau \approx 1/\omega$. Thus our experiments provided a direct and unique way to extract $\tau$ as a function of pressure $p$: Fig. 2(d) displays the experimentally extracted $\tau$ from the transition points of multiple resonators as a function of pressure. Through linear
fitting, one can obtain the expression $\tau \approx \frac{1850}{p}$ [in units of nanoseconds when $p$ is in units of Torr]. The end result of this exercise are the self-consistent fits in Fig. 2(a), (b) and (c). To improve the fits, the results emerging from Eq. 1 with the appropriate material properties and $\tau$ were multiplied by 2.8. In general, all our data sets could be fit adequately using Eq. 1 after multiplying by $2.8 \pm 0.7$.

The fluidic dissipation in individual resonators shown in Fig. 2 suggests that there, indeed, is a transition at $\omega \tau \approx 1$, obtained by tuning $\tau$. Further support for this transition comes from extended measurements in the frequency parameter space. Figure 3(a) shows $\gamma_n$ from different resonators spanning a huge frequency range. Here, $\gamma_n$ is plotted against the resonator frequency $\omega_0$ at four different pressures, i.e., four different $\tau$. This comparison between different devices with different sizes is possible only after normalization of the dissipation by $S_{ef}/m$ [25]. The solid lines in Fig. 3(a) are fits to Eq. 1 using $\tau \approx \frac{1850}{p}$. The points marked by arrows correspond to $\omega \tau \approx 1$. Again, we have multiplied all the fits by 2.8 as in Fig. 2. This multiplicative constant probably arises from adapting the theoretical expressions [9] for an infinite plate oscillating in-plane to the finite and rectangular resonators oscillating out-of-plane. Our surface-to-volume normalization does not give the absolute dissipation, while it appears to be useful for comparing different devices. The elucidation of finite size effects in complex geometries is the subject of our ongoing computational research. Figure 3(b) shows all the data, $\gamma_n/\sqrt{\omega_0 \mu \rho_f}$, from all devices collapsed onto a single curve, plotted against over four decades of the dimensionless parameter $Wi = \omega \tau$. Each symbol type in Fig. 3(b) corresponds to a separate resonator; the $Wi$ is obtained by multiplying the resonator frequency by the corresponding $\tau$ from $\tau \approx \frac{1850}{p}$. The solid curve is obtained from Eq. 1.

Also apparent in Fig. 1 is a small decrease in the resonance frequency as the pressure is increased. The observed decrease is primarily due to the mass of the fluid $m_f$ that is being displaced in-phase with the resonator: $\Delta \omega/\omega_0 \approx \frac{m_f}{2m}$ [4]. Both the geometry and the frequency of the moving surface is expected to play a role in determining $m_f$ and thus $\Delta \omega$. In an effort to rule out the geometry effects, we studied the frequency shift in four beams of identical widths ($w = 500$ nm) and thicknesses ($h = 280$ nm) but varying lengths and resonance frequencies as a function of pressure (Fig. 4). Here, the plotted $\Delta \omega/\omega_0$ corresponds to the approximate fluid mass per unit beam length. The arrows mark $Wi \approx 1$ for each beam and the dashed lines represent best line fits corresponding to $\Delta \omega \propto p^{1/3}$. The molecular flow
FIG. 4: Normalized frequency $\frac{\Delta \omega}{\omega_0}$ for beams of identical widths $w = 500$ nm and thicknesses $h = 280$ nm, but varying lengths and resonance frequencies. The pressure for $Wi \approx 1$ is marked with an arrow. Dashed lines are least-mean-squares fits to $\Delta \omega \propto p^{1/3}$.

The transition observed in our experiments can be interpreted in most general terms as follows. The simple linear relation between stress and rate-of-strain in a Newtonian fluid breaks down at high frequencies. The Boltzmannian theory developed by Yakhot and Colosqui suggests that this result is independent of the nature of the fluid. This transition at $Wi = \omega \tau \approx 1$ was described as a “viscoelastic to elastic” transition owing to the fact that the waves generated in the fluid by the resonator motion do not decay as $\omega \to \infty$. There also appears to be some universality with respect to device geometry: both in cantilevers and doubly-clamped beams, the same naive geometric normalization resulted in a consistent analysis.

There is a relentless effort to develop nanomechanical resonators operating in gaseous and liquid environments. Our results should impact the design of next-generation nanomechanical resonators. Figure 3 suggests that fluidic dissipation saturates at high
frequencies. Take, for instance, two doubly-clamped beam resonators with identical widths and thicknesses but different lengths, i.e., identical $\frac{S_{eff}}{m} \approx \frac{1}{w} + \frac{1}{h}$ but different frequencies such that $\omega_1 < \omega_2$. If $\omega_1 < \omega_2 < 1/\tau$, the ratio of the quality factors of the two resonators in fluid becomes $\frac{Q_{2f}}{Q_{1f}} \sim \sqrt{\frac{\omega_2}{\omega_1}}$. On the other hand, if $\omega_1 < 1/\tau < \omega_2$, then $\frac{Q_{2f}}{Q_{1f}} \sim \omega_2 \sqrt{\frac{\tau}{\omega_1}}$. Finally, for $1/\tau < \omega_1 < \omega_2$, $\frac{Q_{2f}}{Q_{1f}} \sim \omega_2 \omega_1$. Thus, shorter, higher-frequency resonator will always be more resilient in a given fluid but the degree of resilience depends upon the fluid $\tau$. Yet, for two devices with identical frequencies, the smaller one with the larger $\frac{S_{eff}}{m}$ will have the lower $Q_f$. For the case where both $\frac{S_{eff}}{m}$ and device frequency increase, the nature of the scaling determines the end result. Finally, the surface roughness, especially for very small devices, is expected to have an important role in nanofluidics of nanomechanical resonators [22].

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