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Highly Spherical Nanoparticles Probe Gigahertz Viscoelastic Flows of Simple Liquids Without the No-Slip Condition

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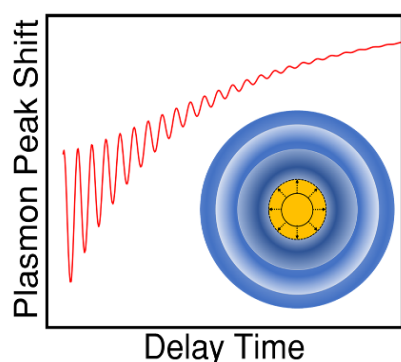
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ABSTRACT

Simple liquids are conventionally described by Newtonian fluid mechanics, based on the assumption that relaxation processes in the flow occur much faster than the rate at which the fluid is driven. Nanoscale solids, however, have characteristic mechanical response times on the picosecond scale, which are comparable to mechanical relaxation times in simple liquids; as a result, viscoelastic effects in the liquid must be considered. These effects have been observed using time-resolved optical measurements of vibrating nanoparticles, but interpretation has often been complicated by finite velocity slip at the liquid-solid interface. Here, we use highly spherical gold nanoparticles to drive flows that are theoretically modelled without use of the no-slip boundary condition at the particle surface. We obtain excellent agreement with this analytical theory that considers both the compression and shear relaxation properties of the liquid.

TOC IMAGE



Newtonian fluid mechanics is widely used to describe the flow of simple liquids such as water (1-3). The Navier-Stokes equation together with the widely accepted no-slip condition readily describes the flow of such liquids when they interact with solid objects down to the micrometer scale (4,5). However, this treatment fails to accurately describe flows generated by the vibration of nanoparticles, predicting much greater damping of the nanoparticle vibrations than is observed experimentally (6). This disagreement can be resolved by including the natural viscoelastic response of the liquid. Fundamentally, this viscoelastic response arises because the short vibrational period of the small particles is comparable to molecular relaxation times in the liquid.

The first such direct mechanical observations of the viscoelastic response of simple liquids were made using the ~ 20 GHz extensional vibrations of bipyramidal gold nanoparticles (6). Similar effects have been seen for the extensional vibrational mode of gold nanorods in poly(vinyl alcohol), a viscoelastic polymer (7). Complimentary methods using time-domain Brillouin scattering have also been developed to probe liquids in the GHz frequency regime (8) and were used to measure both shear and longitudinal viscoelastic properties of liquid glycerol (9).

In the case of the gold-bipyramid extensional vibrations, however, systematic deviations have been observed between the measured and predicted damping rates (6). This deviation is attributable to violation of the no-slip boundary condition at the particle-liquid interface (10). This assumption, that there is zero difference in tangential velocity between a solid surface and the liquid immediately adjacent to the surface, is known to break down at the nanometer scale (11,12). The precise nature of this no-slip violation is difficult to theoretically predict and quantify. There is thus a need for experiments that directly probe the viscoelastic response of simple liquids

without invoking the no-slip boundary condition at the liquid-solid interface. This would enable the unequivocal assessment of viscoelastic constitutive models for simple liquids.

This goal can be accomplished using purely radial flows, such as those generated by the breathing-mode vibrations of purely spherical or cylindrical nanoparticles (13-18). Since there is no tangential motion of the nanoparticle surface, the resulting flow is independent of the no-slip boundary condition. However, most previous measurements used nanoparticles that were only approximately spherical and highly polydisperse; the particle surface motion was therefore not purely radial. The variance in particle diameter present in typical samples meant that inhomogeneous dephasing dominated the measured signal, so that energy damping rates could not be resolved (19). Single-particle measurements overcame this inhomogeneous dephasing but were still limited by non-ideal particle geometries. For example, measurements have been made of the breathing-mode vibrations of individual gold nanorings in glycerol (20), of single quasi-spherical gold nanoparticles in optical tweezers (15), or of single gold nanowires suspended over a trench (21). However, the nanorings had flat faces; the quasi-spherical particles had irregular, faceted geometries; and the nanowires had pentagonal cross-sections. In all cases, then, accurate characterization of viscoelastic effects generated by the vibrating particles required knowledge and modeling of the precise particle geometry and involved slip at the solid-liquid interface. Moreover, these previous experiments probed either the viscous regime (15) or the elastic regime (21,22) but did not access the viscoelastic crossover between the two.

Recently, measurements of vibrations of gold nanoplates have accessed the viscoelastic regime (23); however, analysis of these results followed a previous theory that considered only shear relaxation processes in the liquid and did not rigorously account for bulk (compressional) relaxation processes. Qualitatively different behavior has been predicted for the viscoelastic

response of simple liquids depending on whether shear or bulk relaxation effects are activated (24-26), meaning that models that only account for a single relaxation process (27) are not expected to correctly characterize the liquid response. There has been an ambiguity in the literature regarding how to correctly account for both shear and bulk relaxation in a constitutive equation; recently, this ambiguity was clarified by proper consideration of the classical thermodynamic relationship between the mechanical and thermodynamic pressures (28). The resulting constitutive relation provides the required Newtonian (liquid-like) behavior at low frequency and elastic (solid-like) response at high frequency (29,30), and is the basis of the model used here.

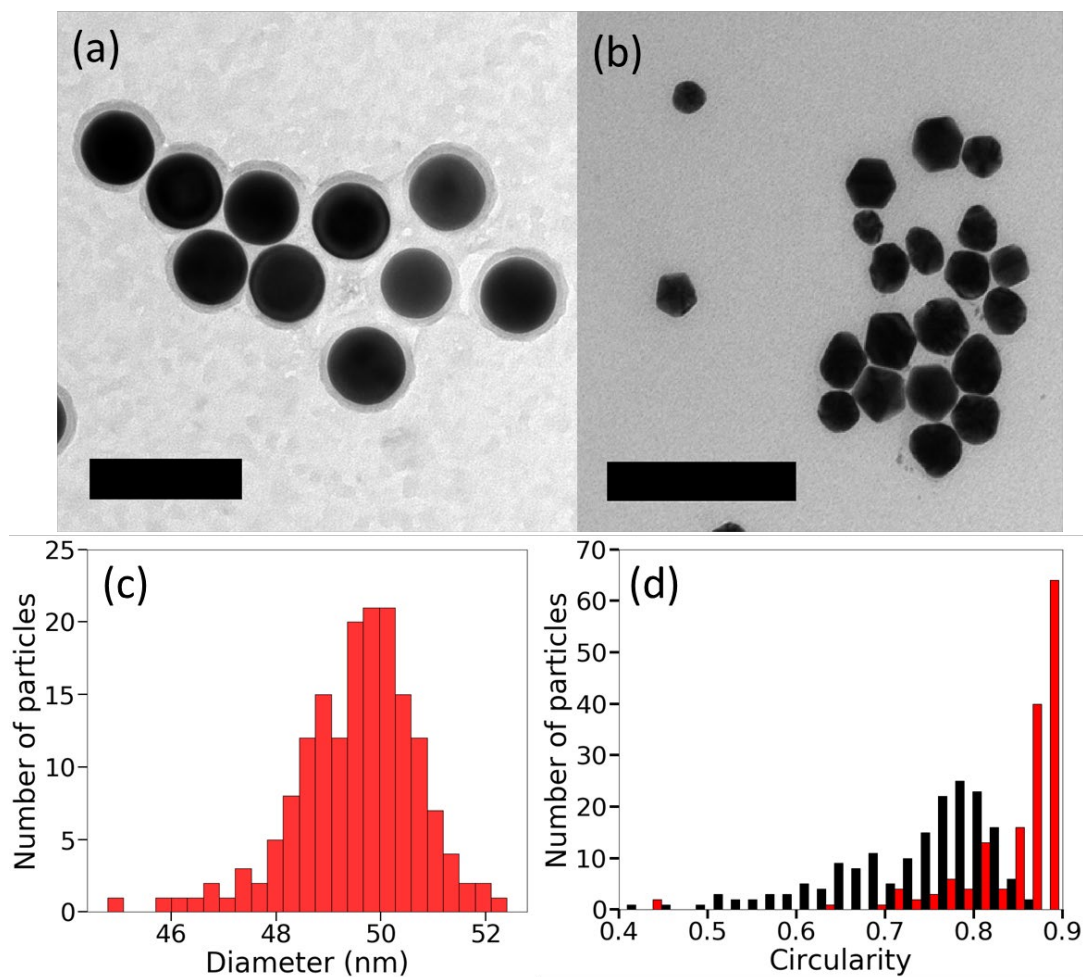


Figure 1. Comparison of highly spherical gold nanospheres used to generate compressional flow in water-glycerol mixtures to a conventional sample of gold nanospheres. (a) Transmission-electron-microscope (TEM) image of the highly spherical gold nanoparticles; scale bar equals 100 nm. (b) TEM image of a conventional sample of spherical gold nanoparticles purchased from Sigma Aldrich; scale bar equals 200 nm. (c) Histogram of measured sphere diameters for the highly spherical nanoparticles obtained from TEM images. (d) Histogram of the measured circularity from the TEM images of the highly spherical (red) and conventional (black) gold nanosphere samples.

In this letter, we eliminate any ambiguity in particle geometry by using highly spherical particles (Ultra Uniform gold nanospheres purchased from nanoComposix). This eliminates any influence of the no-slip condition at the particle-liquid interface and thus provides an ideal model system to investigate viscoelastic effects generated in the liquid. A representative transmission electron microscope (TEM) image of the nominally 50 nm diameter nanoparticles is given in Figure 1a; for details of the sample preparation and the extinction spectrum in water see the Supporting Information. The nanosphere diameter was chosen to access both the Newtonian (viscous) regime and the high-frequency (elastic) regime for the water-glycerol mixtures and temperatures studied (22).

Figure 1c shows a histogram of the measured sphere diameters obtained from the TEM images for approximately 170 nanoparticles. The diameters were measured using ImageJ (31), yielding an average diameter of 49.5 ± 1.2 nm (errors are standard deviations). The sphericity of the particles was characterized using the circularity function in ImageJ (31), with circularity defined as $C = 4\pi(A/P^2)$, where A is the cross-sectional area of the particle in the TEM image and P is its perimeter; $C = 1$ corresponds to a perfect circle. A representative TEM image of a conventional sample of gold nanospheres purchased from Sigma Aldrich is given in Figure 1b and is used to compare the geometry to the highly spherical sample. Figure 1d shows a histogram of the measured circularity of the highly spherical (red) and the conventional (black) nanosphere samples.

Determination of the circularity is limited by the contrast of the TEM images; specifically, the thresholding procedure used to distinguish the particle from its surroundings sometimes incorporates features from the surroundings into the nanoparticle. This limited contrast may explain the upper limit of 0.9 for the circularity of all samples. The high circularity evident in the distribution means that vibration of the highly spherical sample will selectively excited radial motion in the surrounding liquid, minimizing any influence of the no-slip boundary condition. The high monodispersity of the sample minimizes inhomogeneous dephasing, enabling the characterization of homogeneous damping by the surrounding liquid (19).

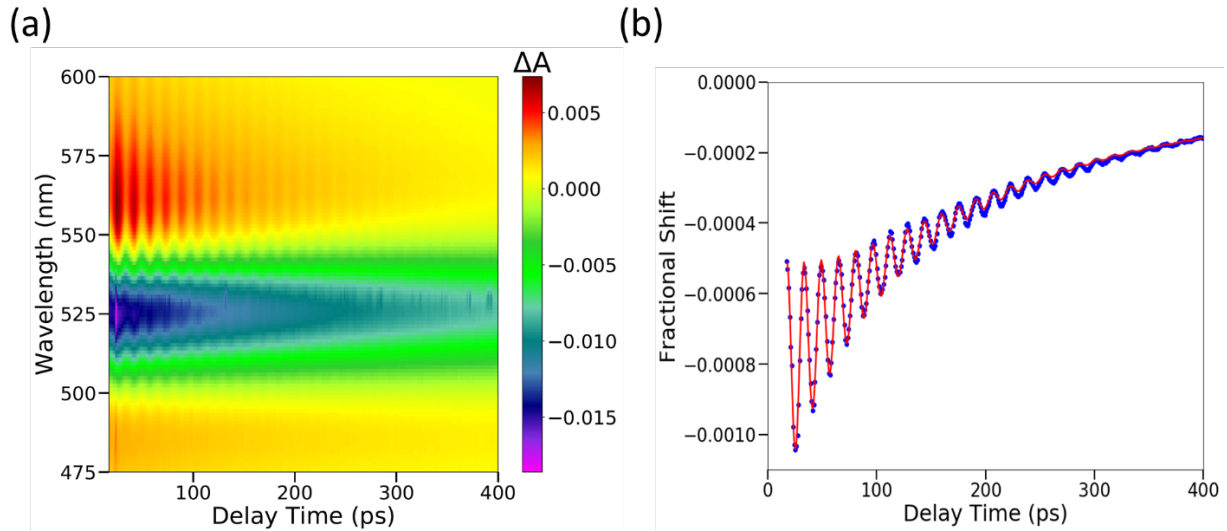


Figure 2. Breathing-mode vibrations of gold nanospheres for 60% mass-fraction glycerol at 20°C. (a) Extinction change, ΔA , after excitation by an initial pump laser pulse tuned to the plasmon resonance of the nanosphere sample as a function of probe pulse wavelength and delay time between the arrival of the pump and probe pulse. (b) Fractional shift corresponding to the change in the peak plasmon resonance frequency normalized by the linewidth, blue dots, obtained from fitting the data in (a). The red line is a fit to the corresponding fractional shifts using a model of a damped oscillation imposed on a decaying background; for details see the Supporting Information.

The frequency and decay rate for vibrations of these particles in glycerol-water mixtures with different compositions and at different temperatures are measured using transient absorption spectroscopy (13,32). Representative transient-absorption data for vibrating nanospheres are shown in Figure 2a. Our data analysis follows that of our previous work (6,33); a representative fit of the fractional shift, corresponding to the change in the peak plasmon resonance frequency normalized by the linewidth, to a model of a damped oscillator on top of a decaying background is shown in Figure 2b. The frequency and decay rate for the vibrations are obtained from these fits, and the dimensionless quality factor is determined to quantify the degree of damping; for details, see the Supporting Information. The experimental vibrational frequencies and quality factors determined from the transient absorption fitting are shown in Figure 3.

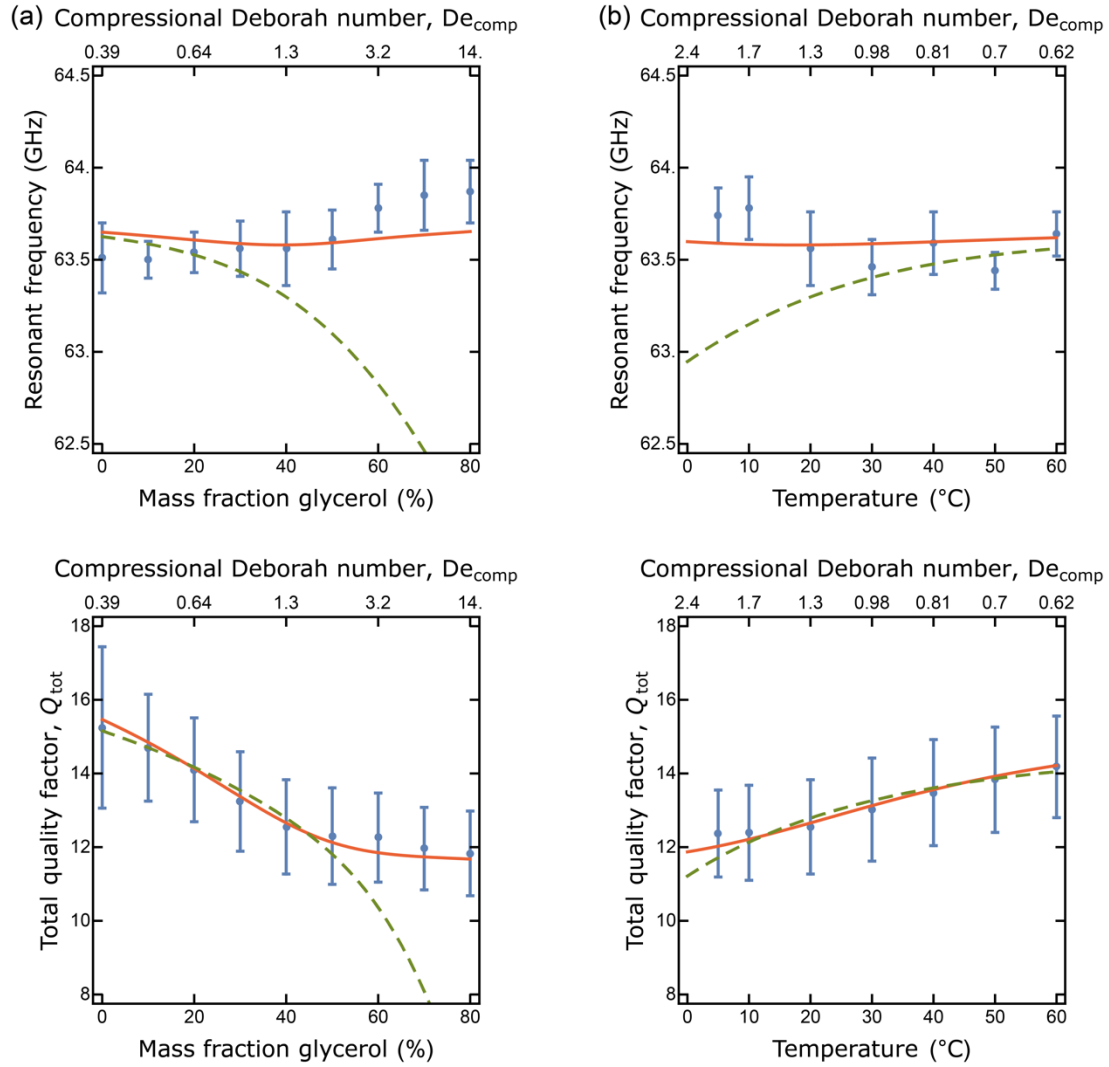


Figure 3. Frequency and quality factor for vibrations of highly spherical gold nanoparticles in water-glycerol mixtures. Blue points are experimental data (error bars are 95% confidence intervals), red solid lines are viscoelastic theory, and green dashed lines are Newtonian theory. The top row shows vibrational frequencies and the bottom row gives total quality factor. The compressional Deborah number is given on the upper horizontal axes; the relationship between the shear and compressional Deborah numbers is given in Supporting Information Figure S5. (a) Mass fraction glycerol varied while holding temperature constant at 20°C. (b) Temperature varied while holding mass fraction glycerol constant at 40%.

Figure 3a shows the frequency and quality factor as a function of the mass fraction of glycerol with the liquid held at a constant temperature of 20°C. Intuitively, Newtonian fluid mechanics predicts that an increase in viscosity leads to a corresponding increase in damping, or

a monotonic decrease in the quality factor. This damping should also lead to a monotonic decrease in the frequency of the damped oscillator due to enhanced added mass in the liquid. By contrast, as the mass fraction of glycerol is increased, the experimental quality factors and the experimental frequencies reach plateaus. These plateaus are signatures of viscoelastic effects: at high glycerol concentrations, the elastic response of the liquid contributes strongly to the stiffening of the nanoparticle-liquid system, so the vibrational frequency increases relative to its purely viscous counterpart (6). Some energy is stored in the liquid, rather than being only dissipated through viscous effects, leading to a reduction in the amount of damping. The crossover from the purely viscous to the viscoelastic regime is also seen in Figure 3b, where the quality factor and frequency are shown as a function of temperature with the glycerol concentration of the mixture now held constant (at 40% mass-fraction glycerol). Here, as temperature increases, viscosity decreases, but there is no corresponding increase in the resonant frequency as would be the case for a Newtonian fluid; instead, the frequency remains nearly constant.

To provide a quantitative description of the viscoelastic effects, we follow the recently developed theoretical framework of Ref. 29. The nanospheres exhibit small-amplitude vibrations, which enables linearization of the fluid's equations of motion:

$$\frac{\partial(\kappa p)}{\partial t} + \nabla \cdot \mathbf{v} = 0, \quad \rho_0 \frac{\partial \mathbf{v}}{\partial t} = \nabla \cdot \mathbf{T},$$

where p is the fluid pressure, \mathbf{v} its velocity, ρ_0 its equilibrium density, \mathbf{T} its Cauchy stress tensor and κ its compressibility, i.e., $\kappa = 1/K$ where K is the complex compressional modulus. Because the nanosphere vibrations are periodic, we specify all variables to have the exponential time dependence, $e^{-i\omega t}$ (e.g., $p = \bar{p}e^{-i\omega t}$), where ω is the angular frequency of vibration. Henceforth, we work in Fourier space and the overbar notation is omitted in the remainder of the analysis.

The fluid compressibility, κ , is frequency dependent for glycerol-water mixtures (9). However, for small amplitude vibrations, p exhibits a linear response to κ and hence the required governing equations are

$$-i\omega\kappa p + \nabla \cdot \mathbf{v} = 0, \quad -i\omega\rho_0 \mathbf{v} = \nabla \cdot \mathbf{T}. \quad (1)$$

The fluid is assumed to be a compressible viscoelastic Maxwell fluid so that the constitutive model for the Cauchy stress is (29,30):

$$\mathbf{T} = -p\mathbf{I} + \mathbf{S}_{\text{sh}} + \mathbf{S}_{\text{comp}}$$

where the shear and compressional components of the deviatoric stress tensor are, respectively,

$$\mathbf{S}_{\text{sh}} = \frac{2\mu}{1 - i\text{De}_{\text{sh}}} \left(\mathbf{D} - \frac{\text{tr}(\mathbf{D})}{3} \mathbf{I} \right),$$

$$\mathbf{S}_{\text{comp}} = \frac{\mu_{\text{B}}}{1 - i\text{De}_{\text{comp}}} \text{tr}(\mathbf{D}) \mathbf{I}.$$

Here, $\mathbf{D} = (\nabla \mathbf{v} + \nabla \mathbf{v}^T)/2$ is the rate-of-strain tensor; $\text{De}_{\text{sh}} = \omega\lambda_{\text{sh}}$ and $\text{De}_{\text{comp}} = \omega\lambda_{\text{comp}}$ are the shear and compressional Deborah numbers, where λ_{sh} and λ_{comp} are the shear and compressional relaxation times, respectively; and μ and μ_{B} are the shear and bulk viscosities of the fluid, respectively. The Deborah numbers quantify the relative importance of Newtonian (low Deborah number) to elastic (high Deborah number) behavior in the fluid.

The sphere's vibrations are governed by the unsteady Navier's equation:

$$-\rho_s \omega^2 \mathbf{u} = \frac{E}{2(1 + \sigma)} \left(\nabla^2 \mathbf{u} + \frac{1}{1 - 2\sigma} \nabla(\nabla \cdot \mathbf{u}) \right), \quad (2)$$

where ρ_s is the particle's density, \mathbf{u} its displacement, E its Young's modulus, and σ its Poisson's ratio. Applying continuity of stress at the particle surface, a solution is found to eqs 1 and 2 for pure radial displacement. This leads to the following eigenequation for the sphere's vibrational frequency:

$$\frac{E}{1+\sigma} \left(\frac{1-\sigma}{1-2\sigma} \frac{j_0(\tau)}{j_1(\tau)} \tau - 2 \right) = \mu \omega \left(\frac{\beta}{1-i\alpha} + \frac{4i}{1-i\text{De}_{\text{sh}}} \right), \quad (3)$$

where $j_n(x)$ is the spherical Bessel function of the first kind of order n ,

$$\tau = \omega R \sqrt{\frac{\rho_s(1+\sigma)(1-2\sigma)}{E(1-\sigma)}},$$

is the dimensionless frequency, $\beta = R^2 \omega \rho_0 / \mu$ is the acoustic Reynolds number (where R is the nanosphere's radius), and

$$\alpha = \sqrt{\frac{\beta \zeta^2 (1 - i\text{De}_{\text{sh}})}{\beta (1 - i\text{De}_{\text{sh}}) - i \zeta^2 \left(\frac{4}{3} + \frac{\mu_B}{\mu} \frac{1 - i\text{De}_{\text{sh}}}{1 - i\text{De}_{\text{comp}}} \right)}}$$

where $\zeta = \omega R \sqrt{\rho_0 \kappa}$ is a dimensionless wavenumber. Solving for the fundamental solution to eq 3 gives the complex eigenfrequency for the breathing mode of the nanosphere, from which the angular resonant frequency and quality factor are, respectively,

$$\omega_f = \sqrt{\omega_r + \omega_i}, \quad Q_{\text{fluid}} = -\omega_f / 2\omega_i, \quad (4a, b)$$

where ω_r and ω_i are the real and imaginary parts of ω , respectively.

We use independently measured and tabulated shear and compressional relaxation times, compressional moduli, and bulk viscosity of glycerol-water mixtures from ref 9 and tabulated shear viscosity and density of glycerol-water mixtures from ref 34. We also choose literature values (29) for the material properties of gold at room temperature: $\rho_s = 19,320 \text{ kg/m}^3$, $E = 79 \text{ GPa}$, and $\sigma = 0.44$. (Adjusting the value of Poisson's ratio to optimize the agreement between theory and experiment results in $\sigma = 0.427$, which agrees well with the quoted literature value of $\sigma = 0.44$.) While the viscoelastic response of the liquids may exhibit a spectrum of relaxation times, the single relaxation time Maxwell model is used for consistency with tabulated data (34). The nanoparticles are single crystalline gold (35), which may break the symmetry of the radial

breathing mode. The comparison between theory and measurement reported below suggests that the effect of such a nonideality is small.

Figure 3 shows the theoretically calculated values of the vibrational frequency for different mass fractions of glycerol and different temperatures. Both theory and experiment show little variation in vibrational frequency over the range of water-glycerol mixtures and temperatures studied; this is not consistent with a Newtonian model, which predicts a monotonic and significant decrease in the frequency as viscosity increases.

The quality factor, being a dimensionless measure of fluid damping, provides a critical test of the viscoelastic model. Using the above-determined material properties, the fluid quality factor, Q_{fluid} , is calculated from eq 4b. The total quality factor, Q_{tot} , is then given by

$$\frac{1}{Q_{\text{tot}}} = \frac{1}{Q_0} + \frac{1}{Q_{\text{fluid}}}, \quad (5)$$

where Q_0 is the (constant) quality factor including both intrinsic damping and inhomogeneous dephasing of the nanospheres. A non-linear least-squares fit of eq 5 to the measured quality factors gives $Q_0 = 23.7$, consistent with reported values for previous measurements on gold nanoparticles (6,13,15-17,21). As shown in Figure 3, this single fitting parameter provides excellent quantitative agreement between the theoretical quality factors and the entire set of experimental quality factors.

Increasing the glycerol concentration, or decreasing the temperature, causes an increase in the molecular relaxation time of the liquid, which in turn increases the Deborah number. As Figure 3 demonstrates, the viscoelastic model (and experiment) diverge from the Newtonian model when the compressional Deborah number, $De_{\text{comp}} \approx 1$. That is, viscoelastic effects arise when the molecular relaxation times become comparable to the period of the breathing-mode vibrations. We note that the ratio of shear to compressional Deborah numbers varies between 0.7 and 1 for these experiments (see Supporting Information Figure S5).

In summary, this work demonstrates that viscoelastic effects in the liquid, i.e., molecular relaxation, strongly affect the breathing mode vibrations of gold nanospheres at sufficiently high glycerol concentrations or at sufficiently low temperatures. Excellent agreement is found between the compressible viscoelastic Maxwell model and all measurements of the nanosphere vibrations in glycerol-water mixtures with different compositions and with different temperatures. Use of highly spherical nanoparticles inherently eliminates the no-slip boundary condition, thus providing a direct probe of viscoelastic effects in simple liquids. The results show that, in general, a complete description of nanoscale-driven flows requires consideration of both shear and compressional relaxation effects in the liquid.

The results also show that nanoscale mechanical resonators in simple liquids can preserve higher quality factors than would be predicted by Newtonian fluid mechanics, because the elastic properties of the liquids are activated at the gigahertz resonance frequencies of these resonators (36-38). This, in turn, has the potential to extend mass sensing using the vibrations of nanoscale objects (36,37) to liquid environments (39). That is, nanostructures exploit the inherent viscoelasticity of simple liquids to overcome the strong viscous damping of small scale resonators that ultimately limits the sensitivity of such nanomechanical sensors (5,40,41). Indeed, the viscoelasticity of simple liquids will impact the mechanical dynamics of any nanoscale structure moving in a liquid, from nanomotors (42,43) to proteins (44,45).

ASSOCIATED CONTENT

Supporting Information

Supporting Information contains details of nanoparticle sample preparation, additional TEM images of the highly spherical and conventional gold nanospheres, experimental methods, and plots of the ratio of shear to compression Deborah numbers. The Supporting Information is made available free of charge [insert link].

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Author Contributions

M.P. and J.E.S. supervised the project and conceived the measurements. B.U. performed the experiment and analyzed the transient-absorption data. J.F.C., M.M. and J.E.S. developed and implemented the theory. All authors analyzed the results and contributed to writing of the manuscript.

Notes

The authors declare no competing financial interest.

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