

Vibrations of a diamagnetically levitated water droplet

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We measure the frequencies of small-amplitude shape oscillations of a magnetically levitated water droplet. The droplet levitates in a magnetogravitational potential trap. The restoring forces of the trap, acting on the droplet's surface in addition to the surface tension, increase the frequency of the oscillations. We derive the eigenfrequencies of the normal mode vibrations of a spherical droplet in the trap and compare them with our experimental measurements. We also consider the effect of the shape of the potential trap on the eigenfrequencies.

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I. INTRODUCTION

If the surface of a spherical liquid drop is briefly deformed by a puff of air, for example, it vibrates, ringing at several different frequencies. The eigenfrequencies of these shape oscillations were determined by Lord Rayleigh,

$$\sigma_T = \left(\frac{Tl(l-1)(l+2)}{\rho a^3} \right)^{1/2} \text{ rad s}^{-1}, \quad (1)$$

where T is the surface tension, l is the mode number, ρ is the density, and a is the radius of the spherical drop at rest [1,2]. By measuring σ , we can determine the surface tension of the liquid.

Beaugnon *et al.* used diamagnetic levitation to measure σ of the lowest-order ($l=2$) mode of a diamagnetically levitated liquid droplet [3], and recently, we used diamagnetic levitation to investigate dynamics of a spinning water droplet [4]. A diamagnetically levitated droplet is confined within a magnetogravitational potential trap [5,6]. The trap acts as an additional cohesive force on the drop, perturbing its eigenfrequency spectrum. Beaugnon *et al.* observed the shift to higher frequency of the lowest-order $l=2$ mode and determined an expression for the increase in terms of an enhanced effective surface tension [3]. However, their result cannot be generalized to the higher-order modes. Here, we use diamagnetic levitation to measure the normal mode frequencies of a levitating drop for $l \geq 2$. We derive an expression for the eigenfrequencies of a liquid droplet confined by the magnetogravitational potential trap and compare it with our measured frequencies. Our analysis points the way to achieving accurate measurements of surface tension using this noncontact technique. We also consider the effect on the eigenfrequencies of the shape of the potential trap and consider analogies with the vibrations of a model “star.”

II. EXPERIMENTAL DETAILS

We use a vertical-bore superconducting solenoid magnet with a room temperature, 50-mm-diameter bore to levitate droplets of water with radii ~ 1 cm. The droplets levitate

approximately 80 mm above the geometric center of the solenoid, where the diamagnetic force, proportional to $B \nabla B$, is equal in magnitude to, and opposite in direction to the gravitational force on (i.e., the weight of) the droplet [5,6]. The magnetic field is $B \approx 12$ T and the vertical field gradient is $\partial B / \partial z \approx 120$ Tm⁻¹ at the levitation point.

Figure 1 shows a spatial map of the magnetogravitational potential

$$U(x, z) = gz - \frac{\chi B^2(x, z)}{2\rho\mu_0} \quad (2)$$

of a unit mass of water in the potential trap, where x and z are radial and vertical cylindrical coordinates with origin at the geometric center of the solenoid coil, $\chi = -9 \times 10^{-6}$ and $\rho = 1 \times 10^3$ kg m⁻³ are the volume magnetic susceptibility (in SI units) and density of water, respectively, and $g = 9.8$ m s⁻². The center of the levitating droplet coincides with the position of the stable levitation point L_0 at the center of the trap, as shown. (There is a second point of unstable levitation L_1 , at the saddle point in U , as shown in Fig. 1. However, this point is not useful for these experiments, and so we do not discuss it further here.) The net force on a unit mass of water, $\mathbf{\Gamma} = -\nabla U$, is zero at the levitation point. The equilibrium shape of a liquid with no surface tension follows the contours of U , as demonstrated recently, using liquid H₂ close to the critical point [7]. For water droplets with $a \sim 1$ cm, however, the surface tension dominates the magnetic and gravitational forces on the drop so that its equilibrium shape is nearly spherical.

The shape of the magnetogravitational potential trap can be altered by adjusting the current in the magnet solenoid coils. A convenient measure of the current is the magnetic field at the geometric center of the solenoid, $B_0 = B(0, 0)$. The field $B(x, z)$ is everywhere proportional to B_0 . To levitate water requires $B_0 \approx 16$ –17 T using our magnet. Figure 1 shows the $U(x, z)$ for $B_0 = 16.5$ T. The field profile $B(x, z)$ of the magnet used to construct this plot was computed by numerical integration of the Biot-Savart integral, using a thin-shell approximation for the current density in the solenoid.

We expand the potential U in a multipole expansion about L_0 :

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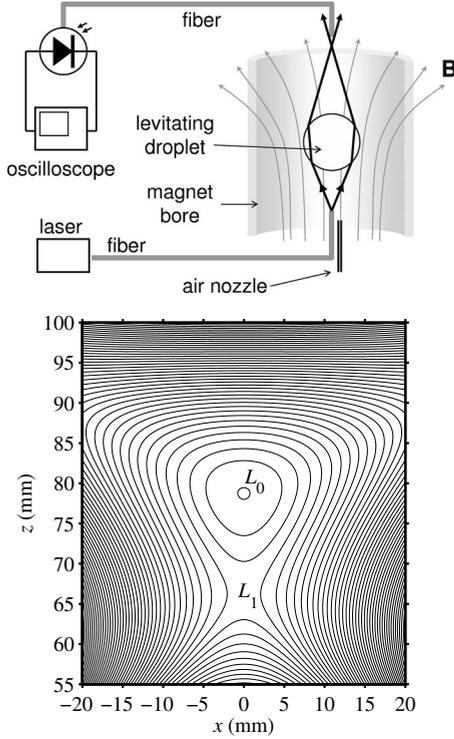


FIG. 1. Bottom: magnetogravitational potential $U(x, z)$ of a water droplet levitated by the magnetic field $B(x, z)$ of a vertical-bore superconducting solenoid magnet (contours at $\Delta U/g=0.05$ mm intervals); x and z are radial and vertical cylindrical coordinates, respectively, with origin at the geometric center of the solenoid. The field at the center of the solenoid is $B_0=B(0,0)=16.5$ T. The field profile $B(x, z)$ used to construct the plot was computed by numerical integration of the Biot-Savart integral. The stable levitation point at a local minimum in the potential is labeled L_0 . An unstable levitation point at a saddle point in the potential is labeled L_1 . Top: schematic of the experimental setup. The magnetic field lines are shown in gray. Rays are shown as bold lines.

$$U(r, \theta) = \sum_{j \geq 0} c_j(r) P_j(\cos \theta), \quad (3)$$

where r and θ are spherical coordinates with origin at L_0 ; θ is the polar angle (i.e., $r \sin \theta = x$). Only the derivative of U normal to the droplet's surface (i.e., the radial component of the force) influences the eigenfrequencies of the normal modes,

$$\Gamma_r(r, \theta) = - \frac{\partial U(r, \theta)}{\partial r} = \sum_{j \geq 0} c'_j(r) P_j(\cos \theta), \quad (4)$$

where $c'_j = \partial c_j / \partial r$. By adjusting B_0 , we reduce the quadrupole component until it is small compared to the spherically symmetric component $c'_2 \ll c'_0$. The octopole harmonic c'_3 , which cannot be reduced this way, remains comparable to c'_0 . All other harmonics are small compared to c'_0 . We shall return to discuss these points further in Sec. V.

Figure 1, top, shows a schematic diagram of the water droplet levitating in the vertical magnet bore. The droplet at rest is close to spherical; we measure the ratio of the equatorial (horizontal) diameter to the polar (vertical) diameter to

be 1.00 ± 0.02 . An optical fiber directs light from a HeNe laser at the droplet. The drop focuses the light onto the aperture of a second fiber, which transmits the light to a photodiode outside the magnet. The emf of the photodiode is measured by a storage oscilloscope. A 1-mm-diameter nozzle directed at the center of the underside of the droplet (see Fig. 1) is connected to a rubber bulb outside the magnet by a tube. When the bulb is struck on a hard surface, the resulting pulse of air from the nozzle excites several shape oscillation modes simultaneously, with amplitude $< 0.05a$. Since the focal length of the drop depends on its shape, the intensity of the laser light falling on the photodiode oscillates as the drop vibrates. The temperature of the water was brought to 16°C in a water bath, before it was injected into the potential trap, to match the ambient temperature in the magnet bore. At this temperature, the density of the liquid is $\rho = 999 \text{ kg m}^{-3}$, its surface tension is $T = 73.3 \text{ mN m}^{-1}$ and its kinematic viscosity is $\nu = 1.11 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ [8]. The liquid was injected into the trap using a glass pipette. The volume of liquid injected was determined to better than 1% uncertainty from the difference in the weight of the pipette before and after injection. The liquid was drawn out of the bore using a paper towel, by capillary action, after the experiment. By measuring the difference in weight between the wet and dry paper, we obtained a second measurement of the droplet volume. Using this simple and accurate technique, we were able to determine that the mass loss through evaporation of the drop, during the measurement period (approximately 30–60 min), was always less than 2%. Experiments were performed both in air and in dry nitrogen gas (by filling the bore with N_2 from a pressurized gas cylinder). The air experiments were performed at $B_0 = 16.2$ T and the nitrogen experiments at 16.5 T. The reason for the different B_0 may be explained by the oxygen content of the air, which, being paramagnetic, buoys up the droplet [9,10] by an additional force $\Gamma_{\text{air}} = -\nabla U_{\text{air}}$. Here, $U_{\text{air}} = \chi_{\text{air}} B^2 / (2\mu_0 \rho)$ and $\chi_{\text{air}} = +3.7 \times 10^{-7}$ is the volume magnetic susceptibility of air (SI units). The c'_2 component of $U + U_{\text{air}}$ is minimized at $B_0 = 16.2$ T. In nitrogen, the buoyancy force is negligible compared to the diamagnetic force, and c'_2 is minimized at 16.5 T as shown in Fig. 1.

III. RESULTS

Figure 2, bottom, shows the oscillations in the photodiode emf $\mathcal{E}(t)$ developed by the light refracted through an $a = 6$ mm droplet; they decay exponentially with a time constant $\tau \sim 0.1\text{--}10$ s, dependent on a and l , due to the viscosity of the water, i.e., $\mathcal{E}(t) = \sum_l \mathcal{E}_l \exp(i\omega_l t - t/\tau_l)$. The power spectrum of these oscillations is shown in Fig. 2, center. Several peaks are evident in this spectrum, corresponding to the $l=2$ to 7 Rayleigh modes of the drop. Peak 1 is due to a small oscillation of the droplet's center of mass of about L_0 (amplitude ~ 1 mm). The frequency of this peak is independent of a . The upper panel of Fig. 2 shows the measured peak positions for different drops with radii between $a=4.5$ and 14 mm; the broken lines show the Rayleigh frequency spectrum for a water drop, according to Eq. (1). Note that the measured frequencies are slightly higher, by ~ 1 Hz, due to

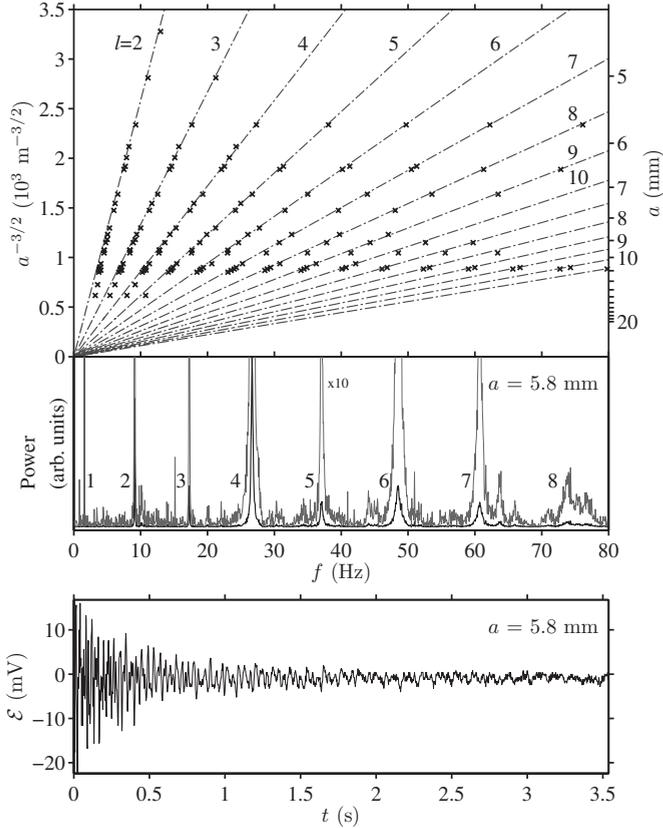


FIG. 2. Bottom: photodiode voltage \mathcal{E} measuring the shape oscillations of an $a=5.8$ mm water droplet. Center: power spectrum of the oscillations of an $a=5.8$ mm water droplet, as a function of oscillation frequency $f=\omega/2\pi$. Top: measured frequencies f of the oscillation modes of water droplets with radii between $a=4.5$ and 14 mm, up to $l=16$. The broken lines show the Rayleigh frequency spectrum for a water drop, according to Eq. (1). The measured frequencies (crosses) are slightly higher, by ~ 1 Hz, due to the effect of the magnetogravitational potential trap.

the effect of the magnetogravitational potential trap. We shall calculate this frequency shift in the following sections of the paper. Although small, it is important to be able to account for this shift if the technique is to be used to obtain accurate measurements of surface tension, for example.

An estimate of the effect of viscosity on the frequency of oscillation can be obtained by calculating the ratio of the magnitude of viscous stresses to surface tension forces, $f\nu\rho/(TD^{-1})=\text{Oh}[2l(l-1)(l+2)]^{1/2}/\pi$, where $D=2a$, $f=2\pi\omega$, and Oh is the Ohnesorge number [11]. This ratio is much smaller than unity for the droplet sizes used in these experiments. This indicates that we can neglect the influence of viscosity on the oscillation frequencies as a small effect. For example, we expect the viscosity of the water to marginally lower the frequency ω of the $l=2$ mode of an $a=5$ mm droplet by $(5 \pm 1) \times 10^{-4}\%$, but this is a small reduction compared to the frequency increase resulting from the trapping potential. (We obtained these estimates by solving numerically the Chandrasekhar equation for the eigenfrequencies of a viscous drop [12,13].) The Oh number indicates that viscosity has a significant effect on the frequency of modes $l < 20$ only for water drops smaller than $a \sim 10 \mu\text{m}$. The

shape of the peaks in the power spectrum agrees well with the Lorentzian shape expected for an exponentially decaying oscillation, with half-width at half maximum (HWHM) $\Delta\omega = 1/\tau \approx \nu a^{-2}(l-1)(2l+1)$ given by the Chandrasekhar equation [12,13]. (Since the power spectrum is the square of the magnitude of the Fourier transform of the oscillations, we compare the shape of the peak with the *square* of the Lorentzian function.)

IV. SPHERICAL POTENTIAL APPROXIMATION

We now consider the effect of a spherically symmetric magnetogravitational potential well on the eigenfrequencies of the drop. We shall discuss the effects of additional harmonics in Sec. V. Our derivation follows Lamb's derivation of the Rayleigh frequencies [2] closely, with the addition of the force Γ_r on the droplet's surface due to the gradient of the magnetogravitational potential at the surface. We write the shape of the l th mode of a drop oscillating with frequency ω , for oscillations with small amplitude ϵ , as

$$r = R(\theta, t) = a + \zeta = a + \epsilon P_l(\cos \theta) \sin \omega t, \quad (5)$$

where P_l is a Legendre polynomial of degree $l \geq 1$ ($l=1$ corresponds to an oscillation of the droplet's center of mass in the potential trap).

The pressure equation at the surface of the droplet (to first order) is [2]

$$\left. \frac{\partial \phi}{\partial t} \right|_{r=a} = U(R) + \frac{p(R)}{\rho} + F(t), \quad (6)$$

where $F(t)$ is an arbitrary function of t only and ϕ is the velocity potential [2]

$$\phi(r, \theta) = -\frac{r^l}{la^{l-1}} \omega \epsilon P_l(\cos \theta) \cos \omega t. \quad (7)$$

The pressure difference across the surface resulting from the surface tension is [2]

$$p(R) = T \left(\frac{2}{a} + \frac{\zeta(l-1)(l+2)}{a^2} \right). \quad (8)$$

The magnetogravitational potential U at the surface of the drop is (to first order)

$$U(R(\theta, t)) = U(a) - \Gamma_r(a, \theta)(R - a) \quad (9a)$$

$$= U(a) - \epsilon \Gamma_r(a, \theta) P_l(\cos \theta) \sin(\omega t), \quad (9b)$$

where $\Gamma_r(a, \theta) = -c_0'(a)$ for a spherically symmetric well [see Eq. (4)]. Inserting Eqs. (7), (8), and (9b) into Eq. (6), we obtain

$$\omega^2 = \sigma_T^2 + \sigma_0^2, \quad (10)$$

where

$$\sigma_0^2 = \frac{c_0'(a)l}{a} \quad (11)$$

is the oscillation frequency (squared) of a hypothetical drop with $T=0$, held together by the magnetogravitational trap

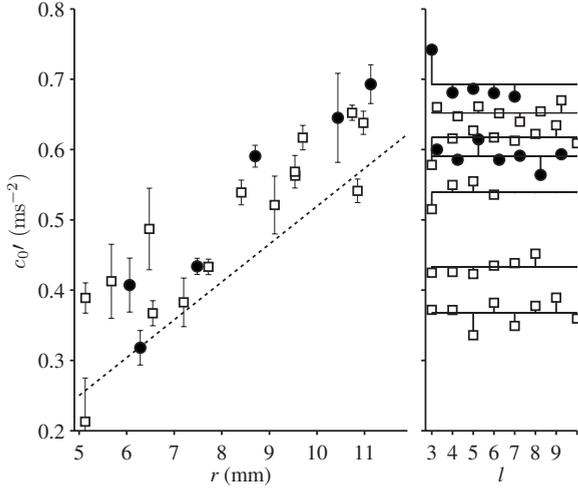


FIG. 3. Left panel: gradient of the magnetogravitational potential trap c'_0 obtained from the measured frequencies of two modes, $l=2$ and $l>2$, of a drop with radius $a=r$ at rest. Error bars show the standard error. Filled circles: data obtained in nitrogen atmosphere. Open squares: data obtained in air atmosphere. Dotted line: c'_0 computed from the solenoid geometry. Right panel: dependence of the experimentally obtained c'_0 on the mode number with $l>2$ (see text).

alone. The fact that the square of the *measured* frequency ω^2 is a simple sum of the square of the Rayleigh frequency σ_T^2 and σ_0^2 is due to the fact that l remains a good eigen-number for oscillations in a *spherically symmetric* potential well. In Sec. V, we will consider the effect of a nonspherical magnetogravitational well. In a nonspherical well, l is not a good eigen-number in general, but for small deviations from spherical, we can use perturbation theory to obtain corrections to Eq. (10).

We can obtain an experimental measurement of c'_0 by examining the difference between the measured frequencies ω of any two of the modes $l, n \geq 2$:

$$h_l \sigma_{0,n}^2 - h_n \sigma_{0,l}^2 = h_l \omega_n^2 - h_n \omega_l^2, \quad (12)$$

where $h_l = l(l-1)(l+2)$. Dividing by $(nh_l - lh_n)/a$, we obtain an experimental measurement of $c'_0(r)$ from the oscillations of a drop that has radius $a=r$ at rest:

$$c'_0(r) = r \frac{h_l \omega_n^2 - h_n \omega_l^2}{nh_l - lh_n}. \quad (13)$$

By using two measured frequencies, ω_l and ω_n , rather than a single frequency, we obtain a measurement of c'_0 independent of the surface tension.

Figure 3 (left panel) shows the values of c'_0 that we obtain from the above “two-frequency” method using the lowest-order mode $n=2$ and a second mode $l>2$. We use the $n=2$ mode since the corresponding peak in the power spectrum is always clearly resolved. We plot the mean of the values obtained for each mode $l>2$. Error bars (standard error) indicate the variation in the measured c'_0 obtained from different modes l . Filled circles and open squares show data obtained from a drop in a nitrogen atmosphere at $B_0=16.5$ T and in air at $B_0=16.2$ T, respectively. The broken line on Fig. 3

shows the value of $c'_0(r)$ computed from the potential U shown in Fig. 1 (i.e., from the geometry of the solenoid and the current B_0). This line is in reasonably good agreement with the experimentally measured values of c'_0 , although the data points fall at slightly higher values. As an additional check of our method, we plot the dependence of the measured c'_0 on l (right-hand panel). If the trap-induced frequency enhancement that we measure experimentally is accurately given by Eq. (11), then the c'_0 values that we determine by this method should exhibit no dependence on l . Although there is some scatter in the data due to experimental error, there is no clear dependence on l .

V. NONSPHERICAL POTENTIAL

In the previous section, we assumed that the potential well was spherically symmetric around L_0 . However, while we have chosen the field B_0 to minimize the quadrupole component of the trap, the octopole component remains significant, as can be seen clearly in Fig. 1. Thus it initially appears surprising that we can treat the well *as if* it were spherical, in order to calculate its effect on the vibrations of the droplet’s shape. (Had we determined an unphysical dependence of our measured c'_0 on l in the previous section, it would also have followed that the spherical well approximation was inadequate.) We now consider the effect on the eigenfrequencies of the drop of additional harmonic components c'_j of the potential trap and consider why, if the trap has a significant octopole component, the spherical well approximation is so effective at reproducing the measured frequencies of the droplet.

We first discuss how close to spherical it is possible to make the potential trap; that is, how small the coefficients c'_j , $j>0$ can be made. Lorin and Mailfert have considered the relationship between the coil geometry (the distribution of the current density) and the shape of the potential trap [14]. Although we cannot alter the coil geometry, we can alter the coefficients c'_j by adjusting the current flowing in the solenoid. The magnitude of the quadrupole component of the radial force $|c'_2|$ is smallest (for our magnet) at $B_0=16.2$ T in air, and at $B_0=16.5$ T in nitrogen gas; at this field, $|c'_2| \ll |c'_0|$ and only weakly dependent on r . Increasing B_0 beyond this value gives a positive c'_2 , increasing with r . Decreasing B_0 below this value gives a negative c'_2 , becoming more negative with increasing r . The gradient of the octopole component c'_3 , however, is of the same order as c'_0 and increases as r^2 . This component is a feature of the magnetogravitational potential trap generated by a solenoid. It can be reduced slightly, by increasing B_0 , but only at the expense of significantly increasing $|c'_2|$. Note that the dipole component of the trap c_1 is necessarily zero (hence $c'_1=0$ too) since the net vertical force on the droplet F_z must be zero for levitation. Since F_z is proportional to the difference between the mean surface potentials of its upper and lower hemispheres, i.e.,

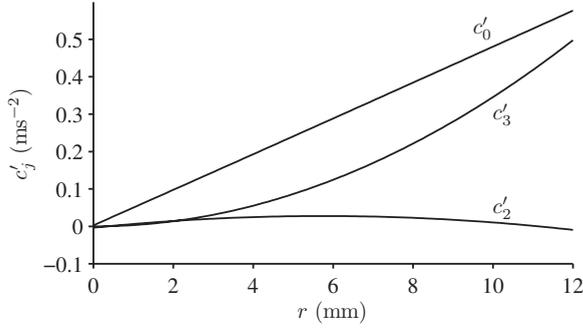


FIG. 4. Values of the $j=0$ (spherically symmetric) and $j=2,3$ (quadrupole and octopole) coefficients c'_j in a multipole expansion of the potential well gradient, Eq. (4), at $B_0=16.5$ T (in nitrogen atmosphere). Note, $c'_1=0$ (see text). The values were determined from the magnetogravitational potential [Eq. (2)] shown in Fig. 1. The c'_0 line is the same as that shown by the dotted line on Fig. 3.

$$\begin{aligned} F_z &\propto \int_0^1 |w| U(a,w) dw - \int_{-1}^0 |w| U(a,w) dw \\ &\propto \sum_j c_j \int_{-1}^1 w P_j(w) dw \\ &\propto c_1, \end{aligned}$$

it follows that $c_1=0$ where $w=\cos \theta (=P_1)$. The harmonic components $j>3$, are small compared to both c'_0 and c'_3 , in our system. Figure 4 shows the values of the $j=0$ and $j=2,3$ (quadrupole and octopole) coefficients c'_j at $B_0=16.5$ T (for nitrogen atmosphere) computed from the solenoid geometry. The spheres in Fig. 5 show the computed magnitude of $\Gamma=-\nabla U$ at the surface of three droplets, radius $a=5.0, 7.5$, and 10.0 mm. Radiating lines indicate the direction and magnitude of Γ , which is directed toward the interior of the droplet. The variation in $|\Gamma|$ over the surface of the droplet is due to the octopole component of the potential trap.

We now consider the effect on the droplet's eigenfrequencies of adding a harmonic component c'_j , $j \geq 2$ to the potential trap. The analysis proceeds as above, however, we must now include higher-order harmonics in the eigenfunction of the shape oscillation,

$$r = R(\theta, t) = a + \epsilon \sin \omega t \sum_{l \geq 1} b_l P_l(\cos \theta), \quad (14)$$

since l is not, in general, a good eigen-number in a non-spherical potential. In principle, we should decompose the shape into spherical harmonics Y_l^m since the degeneracy in m

is also lifted in a nonspherical potential (see Fig. 6). However, our method of inducing shape oscillations in the droplet tends to excite only the axisymmetric shapes (i.e., with $m=0$) since the air jet is aligned along the solenoid axis. For this reason, we derive here the frequencies of the $m=0$ oscillations only [Eq. (14)], which are sufficient to interpret the experimental results. We summarize the treatment of the general case $|m| \leq l$ in the Appendix.

The velocity potential is

$$\phi(r, \theta) = -\epsilon \omega \cos \omega t \sum_{l \geq 1} b_l r^l l^{-1} a^{-l+1} P_l(\cos \theta). \quad (15)$$

The magnetogravitational potential U at the surface of the drop is [see Eq. (9a)]

$$U(R) = U(a) - \epsilon \Gamma_r(a, \theta) \sin(\omega t) \sum_{l \geq 1} b_l P_l(\cos \theta), \quad (16)$$

where $-\Gamma_r(a, \theta) = c'_0(a) + c'_j(a) P_j(\cos \theta)$ in this case [see Eq. (4)]. Inserting Eqs. (14)–(16) into Eq. (6) and equating the time-varying terms, we obtain

$$a \omega^2 \sum_{l \geq 1} \frac{b_l P_l}{l} = \sum_{l \geq 1} \left[(c'_0(a) + c'_j(a) P_j) + \frac{T}{\rho a^2} (l-1)(l+2) \right] b_l P_l. \quad (17)$$

The product $P_l P_j$ appearing on the right-hand side (RHS) of this equation can be expanded as a sum of Legendre polynomials [15], which, for our purposes, is most conveniently written $P_l P_j = \sum_{p=|j-l|}^{j+l} Q(l, j, [j+l-p]/2) P_p$, in which [15]

$$Q(l, j, s) = \frac{A(l-s)A(s)A(j-s)}{A(j+l-s)} \binom{2j+2l-4s+1}{2j+2l-2s+1} \quad (18)$$

for integer s and we define $Q(l, j, s)=0$ for half-integer s ; $A(n)=1 \times 3 \times 5 \times \dots \times (2n-1)/n!$. Equating the coefficients of P_l , we obtain

$$\begin{aligned} \omega^2 b_l &= \frac{c'_0(a)l}{a} b_l + \frac{T}{\rho a^3} l(l-1)(l+2) b_l \\ &+ \frac{c'_j(a)l}{a} \sum_{p=|j-l|}^{j+l} b_p Q(p, j, [j+p-l]/2). \end{aligned} \quad (19)$$

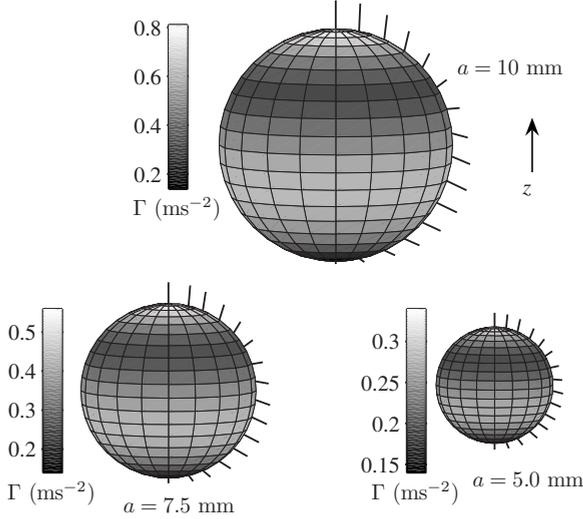


FIG. 5. The three spheres show the computed magnitude of the vector $\Gamma = -\nabla U$ on the surface of three droplets, radius $a = 5.0, 7.5$, and 10.0 mm at $B_0 = 16.5$ T (in nitrogen atmosphere); the length and direction of the radiating lines show the direction and magnitude of the (inward pointing) Γ at the surface; Γ was determined from the magnetogravitational potential [Eq. (2)] shown in Fig. 1. The variation in $|\Gamma|$ over the surface of the droplet is due to the octopole component of the potential trap.

This equation has the form of an eigenvalue problem $\omega^2 b_l = H_{l\lambda} b_\lambda = (H_{l\lambda}^{(0)} + V_{l\lambda}) b_\lambda$ (using the summation convention, $l \geq 1$), where $H^{(0)}$, representing the first two terms on the RHS of Eq. (19) is a diagonal matrix and V , representing the third term on the RHS is not, in general, diagonal. Treating V as a perturbation, the first-order correction $(\sigma_j^2)^{(1)} = b_l^{(0)} V_{l\lambda} b_\lambda^{(0)}$ due to a harmonic component j can be computed analytically to obtain the eigenfrequencies $\omega^2 = \sigma_T^2 + \sigma_0^2 + (\sigma_j^2)^{(1)} + \dots$. For a quadrupole harmonic $j=2$ we obtain

$$(\sigma_j^2)^{(1)} = \frac{c_2'(a)}{a} \frac{l^2(l+1)}{(2l+3)(2l-1)}. \quad (20)$$

For an octopole harmonic $j=3$ and all odd j , $(\sigma_j^2)^{(1)} = 0$. This explains why our spherical-well approximation works so well in predicting the eigenfrequency spectrum of the droplet: we have minimized c_2' by careful adjustment of B_0 and we expect, from the above analysis, that the effect of the octopole harmonic c_3' (which is comparable to the spherically symmetric component c_0') on the measured eigenfrequencies in our experiment, to be minimal. Figure 6 shows the eigenfrequencies determined by using a computer to solve the eigenvalue problem numerically (i.e., beyond first order). The eigenfrequencies of an $a = 7.5$ mm droplet, for various c_2' and c_3' , are shown (thick lines), along with the first-order result Eq. (20) for comparison (thin lines). In addition, we plot the eigenfrequencies of modes with $m \neq 0$ (broken lines); the calculation of the frequency of these modes is outlined in the Appendix. The lower plot of Fig. 6 shows that the effect of the octopole harmonic c_3' on the measured eigenfrequencies is minimal for all m for $|c_3'| \leq 1$. For a quadrupole harmonic c_2' , the eigenfrequencies depend strongly on m , as shown in the upper plot of Fig. 6. However, since we

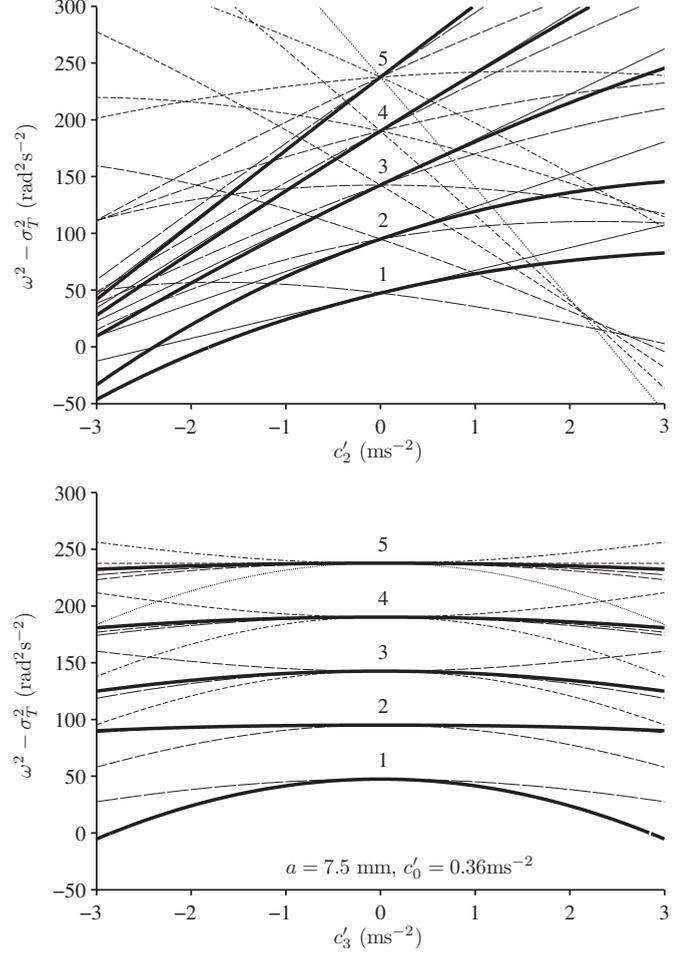


FIG. 6. Top: effect on the eigenfrequencies of an $a = 7.5$ mm water droplet of adding a quadrupole harmonic c_2' to the shape of the magnetogravitational potential well ($c_0' = 0.36$ m s $^{-2}$, all other $c_j' = 0$). The thick black lines show the eigenfrequencies (squared) of the $m=0$ modes [i.e., the eigenvalues ω^2 of Eq. (19)] corresponding to $l=1-5$; the eigenvalues of Eq. (19) were computed using MATLAB. (Note that we have plotted $\omega^2 - \sigma_T^2$ for clarity). The thin lines show the first-order approximation [Eq. (20)] departing from the numerically computed frequencies at $|c_2'| \gtrsim 1$. Long-dash, medium-dash, short-dash, dot-dash, and dotted lines show the frequencies of the $|m|=1-5$ modes, respectively (the treatment of the $m \neq 0$ modes is summarized in the Appendix). Bottom: effect on the eigenfrequencies of an $a = 7.5$ mm water droplet of adding a harmonic c_3' to the shape of the well ($c_0' = 0.36$ m s $^{-2}$, all other $c_j' = 0$).

have reduced $|c_2'|$ to smaller than ≈ 0.05 as described above and since our method of exciting the oscillations tends to excite only the $m=0$ modes, this does not alter our explanation of why the spherical-well approximation works so effectively.

VI. DISCUSSION AND CONCLUSION

Although the forces at the droplet's surface are dominated by the surface tension, we can clearly observe the influence of the magnetogravitational potential trap on the eigenfrequencies. We have demonstrated that the effect of the trap on

the eigenfrequencies closely approximates that of a spherically symmetric potential well. Although the potential well has a significant octopole component in addition to the spherically symmetric component, we have calculated that the effect of the octopole component on the eigenfrequencies is small and verified this experimentally. The small difference between the values of c'_0 calculated from the solenoid geometry and the measurement of c'_0 from the measured droplet oscillations may be due to a small error in the calculated value: the calculation of the field profile $B(x,z)$, performed by numerical integration of the Biot-Savart equation, is based on a thin-shell approximation of the current density in the solenoid coils. We note that the discrepancy cannot be explained by the octopole component of the trap since this only marginally reduces the eigenfrequencies (Fig. 6), which would only *reduce* the measured value of c'_0 slightly. Experiments have shown that nonlinear effects become significant for oscillation amplitudes greater than approximately $0.1a$ [16]. In our experiments, however, the oscillation amplitude is smaller than this, and we do not observe these nonlinear effects.

We have shown how to minimize the quadrupole harmonic of the potential trap by adjusting the solenoid current. This allowed us to use the relatively simple formula Eq. (11) to calculate the effect of the trap on the eigenfrequencies. This opens up the possibility of using diamagnetic levitation to accurately determine the surface tension of diamagnetic liquids (for example, water, and many water-based and organic solutions): subtracting the contribution of the trap to the eigenfrequencies reveals the Rayleigh-frequency spectrum, from which the surface tension can be obtained directly. A contactless measurement technique has many advantages, for example, the ability to measure highly reactive liquids and to achieve significant supercooling of the liquid, as demonstrated in experiments on electromagnetically levitated liquid metals [17]. Acoustic levitation and the oscillating drop method can be used to measure surface properties of small drops (radius <2 mm) of water and organic liquids (e.g., [18,19]). Suspension of small drops in air flow is also possible [20]. However, since the equilibrium droplet shape is distorted significantly from spherical in both of these techniques, it is necessary to make accurate measurements of the equilibrium shape in order to correct for the distortion, which introduces additional significant experimental uncertainty [18–21]. Drops can be bounced on a solid surface or a vibrating liquid bath, the drop and surface being separated by a thin layer of air [22,23]. In this case, the eigenfrequency spectrum deviates from the Rayleigh spectrum due to the periodic forcing by the oscillating surface [24,25]. An expression, similar to Rayleigh's, for the eigenfrequencies of vibrating drops in continuous point contact with a surface has been determined [25] but there remains some uncertainty over the spectrum of bouncing drops [25]. Although free fall can be used to obtain the Rayleigh frequencies directly (for example, in a drop tower [26]), we have shown how diamagnetic levitation could offer an alternative accurate method of measuring these frequencies; the levitated droplets are near spherical at rest and the eigenfrequency spectrum is very close to Rayleigh's, as shown in Fig. 2. The relatively small shift to higher frequency due to the potential trap can be

obtained using a simple spherical-well approximation [Eq. (11)]. Diamagnetic levitation can levitate cm-size drops (up to ≈ 3 cm diameter in our magnet) [3,4,27] enabling the droplet volume to be measured easily and precisely. Currently, there is interest in the temperature dependence of the surface tension of supercooled water [28]. The lowest-temperature achieved so far, which used a contact technique, is 245 K [29]; nucleation sites on the container walls trigger freezing before colder temperatures can be reached. We propose that diamagnetic levitation and the oscillating drop technique could be used to obtain measurements of surface tension at temperatures less than 245 K.

It is interesting to consider an analog between the oscillations of the levitating droplet and that of an object in a gravitational field. We can consider the magnetogravitational force on a unit mass of water, $\Gamma = -\nabla U$, as being an effective gravitational field acting on the liquid. Reid obtained an expression for the eigenfrequencies of a gravitating body composed of a solid spherical core radius R_1 and density ρ_1 covered by an inviscid liquid "mantle" of radius R_2 , density ρ_2 [30] (see also Ref. [31]):

$$\sigma^2 = \frac{4}{3} \pi G \bar{\rho} l(l+1) \frac{1 - \eta^{2l+1}}{1 + l(1 + \eta^{2l+1})} \left(1 - \frac{3}{2l+1} \frac{\rho_2}{\bar{\rho}} \right), \quad (21)$$

where $\eta = R_1/R_2$, $\bar{\rho} = \eta^3 \rho_1 + (1 - \eta^3) \rho_2$ is the mean density, and G is the gravitational constant. Interestingly, Reid's expression becomes equivalent to Eq. (11) if we let $\rho_2 \rightarrow 0$ and $R_1 \rightarrow 0$ since this allows the surface effective gravity $\Gamma(a)$ to be equated with a fictitious point mass $m = (4/3) \pi a^3 \bar{\rho} = \Gamma(a) a^2 / G \sim 1 \times 10^8$ kg (approximately the mass of 1 mm³ of nuclear-density material). In this sense, we can view our magnetically levitated droplet as a "toy model" of an incompressible gravitating body (possessing surface tension), composed of a small-diameter massive core (the fictitious mass) and a low density outer region (the water). Note that this result differs from that of a *uniform-density* self-gravitating body, $\sigma_G = [(8/3) \pi G \rho l(l-1)/(2l+1)]^{1/2}$ [32]. Bastrukov recently obtained a similar spectrum for a self-gravitating liquid droplet with radially varying density distribution $\rho(r < a) \propto 1/r$, having a singular density at the center [33].

By adjusting the current in the magnet (i.e., B_0) we can change the quadrupole (c'_2) component of the trap. Although we have sought to minimize this component in this paper, it would be interesting to investigate the effect of this quadrupole component on the oscillation frequencies experimentally and compare it with the result we derive above. Unlike the odd harmonics, the even order harmonics have a significant effect on the eigenfrequencies to first order. Using the analogy with a model "star," adding a quadrupole component to the effective gravity Γ is equivalent to deforming the shape of the core mass from spherical to oblate, perhaps due to rotation, for example.

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APPENDIX

We consider the effect of a trap component c'_j on an arbitrary shape oscillation

$$r = R(\theta, \Phi, t) = a + \epsilon \sin \omega t \sum_{l \geq 1} \sum_{m=-l}^l b_l^m Y_l^m(\theta, \Phi). \quad (\text{A1})$$

The corresponding velocity potential is

$$\phi(r, \theta, \Phi) = -\epsilon \omega \cos \omega t \sum_{l \geq 1} \sum_{m=-l}^l b_l^m r^l l^{-1} a^{-l+1} Y_l^m(\theta, \Phi). \quad (\text{A2})$$

Inserting Eqs. (A1) and (A2) into Eq. (6) and equating the time-varying terms, we obtain

$$\begin{aligned} a \omega^2 \sum_{l \geq 1} \sum_{m=-l}^l \frac{b_l^m Y_l^m}{l} \\ = \sum_{l \geq 1} \sum_{m=-l}^l \left[(c'_0 + c'_j P_j) + \frac{T}{\rho a^2} (l-1)(l+2) \right] b_l^m Y_l^m. \end{aligned} \quad (\text{A3})$$

The product $Y_l^m P_j = N_l^m e^{im\Phi} P_l^m P_j^0$ can be expanded as $N_l^m e^{im\Phi} \sum_{p=|j-l|}^{j+l} Q(0, j; m, l; p) P_p^m$, where the Q in this case are Gaunt coefficients, as defined in Ref. [34], P_l^m are associated Legendre functions, and N_l^m are the corresponding normalization factors. Equating the coefficients of Y_l^m , we obtain $2l+1$ eigenvalue problems, i.e., one for each m , similar to Eq. (19).

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