Surface tension of liquid ^{4}He as measured using the vibration modes of a levitated drop

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Recently, different measurements of the surface tension γ of liquid helium have given values that differ from each other by as much as 6%. In this paper we report a measurement in which we have studied a helium drop that was magnetically levitated. From measurements of the radius of the drop and the frequencies of the vibrational normal modes, the surface tension could be determined. Measurements were made over the temperature range 0.6 K to 1.6 K, and from an extrapolation, it was estimated that the value of γ at $T=0$ K is 0.375 ± 0.004 dyne cm^{-1}. The trap potential resulted in small shifts of the frequencies of the vibrational modes. We have developed a theory of these shifts and find good agreement with the experimental measurements.

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I. INTRODUCTION

The surface tension γ of liquid ^{4}He was first measured by van Urk *et al.* in 1925.[1] Since then many other measurements have been made, primarily with the goal of investigating the unusual temperature dependence of the surface tension.[2] The techniques that have been used include measurements of the rise of the liquid level in capillaries,[1,3–6] a determination of the force needed to lift a metal ring from the liquid surface,[7] and measurements of surface wave velocity.[8,9] From these measurements, the value of γ extrapolated to zero temperature ranged between 0.350 and 0.378 dyne cm^{-1}. More recently, Iino *et al.*[10] found the frequency of standing capillary waves inside an experimental cell, and from this measurement obtained the result $\gamma=0.354$ dyne cm^{-1}. This was followed by an experiment by Roche *et al.,*[11] who measured the frequency of capillary waves with wavelength in the micron range. Their result for γ was 0.375 ± 0.003 dyne cm^{-1}. They presented arguments that Iino *et al.*[10] had neglected to include a correction to allow for the effects of the meniscus at the walls of their cell. However, this was disputed in a later paper by Nakanishi and Suzuki,[12] who presented further measurements that were in agreement with the data of Iino *et al.*

Because of these differences, it is of interest to make a different measurement by an independent technique. In this paper, we report measurements of the frequency of the vibrational normal modes of magnetically levitated helium drops. From the measured frequency for drops of known size, we are able to determine the surface tension.

II. EXPERIMENTAL METHOD

In our experiment, we use helium drops that have been magnetically levitated. Helium is slightly diamagnetic and hence is repelled from regions of strong magnetic field and seeks to move toward a lower field region. In an inhomogeneous magnetic field $B(\vec{r})$, the total force on unit volume of liquid helium is

$$-\rho g\hat{k}+\frac{1}{2}\chi\nabla B^2(\vec{r}), \tag{1}$$

where $\hat{k}$ is a unit vector in the vertical direction (z direction), ρ is the density, and χ is the magnetic susceptibility. We use a static field with axial symmetry produced by a superconducting magnet. Figure 1 shows the schematic layout of the system. In order for a liquid drop to be levitated along the magnet axis, there needs to be a point at which

$$\chi B\frac{\partial B}{\partial z}=\rho g. \tag{2}$$

This condition is satisfied when $B\partial B/\partial z$ equals 20.7 T^2 cm^{-1}. The details of the levitation apparatus have been published previously.[13]

It is convenient to describe the combined effect of gravity and the magnetic field by means of a function $U(x,y,z)$, which is the potential energy of unit volume of helium at position x,y,z. If the origin of the coordinate system x,y,z is chosen to coincide with the minimum of the trap potential,

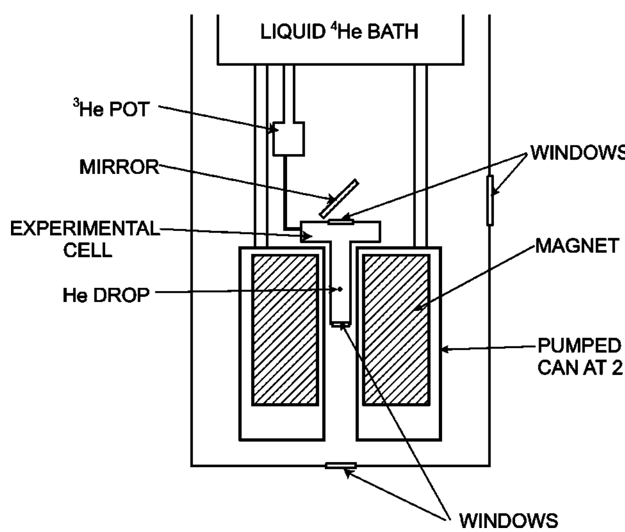

FIG. 1. The experimental apparatus showing the sample cell within the annular magnetic can. Two sets of windows on the cryostat tails and a mirror on the cell allow viewing of drops within the magnet bore. The small circle within the cell shows the approximate position of the levitated drop.

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then close to the potential minimum, the trap can be described by a harmonic potential,

$$U(x,y,z)=U_0+k_{xy}(x^2+y^2)+k_z z^2. \tag{3}$$

In order for the drop to have lateral stability, it is necessary to have $k_{xy}>0$. This is achieved if the magnitude of the magnetic field increases as a function of the horizontal distance (x^2+y^2) from the levitation point.

The constants k_{xy} and k_z are functions of the current in the magnet and in general are not equal. Thus, the trap does not have spherical symmetry and so a drop suspended in the trap will not be spherical. The distortion from spherical shape increases with increasing drop size. A drop with a maximum size of 1 cm in diameter can be levitated stably in our magnetic trap. We usually kept the size of our drops around 2 mm in diameter. For drops of this size, the difference between the length of the diameter of the drop along the z axis and the diameter in the xy plane is calculated to be typically 1% or less.

If, for the moment, we take the drop to be spherical, the frequencies of the vibrational modes are given by the formula derived by Rayleigh,

$$\omega_l^2=\frac{\gamma}{\rho a^3}(l-1)l(l+2), \tag{4}$$

where γ is the surface tension, ρ is the density of the liquid, and a is the radius of the drop.[14] l can be any integer equal to or larger than 2. In the derivation of Eq. (4), the drop is taken to be incompressible and inviscid. A measurement of the vibrational frequency can thus be used to determine γ provided that the radius and density are known.

The method used to excite and detect the vibrational normal modes has been described previously.[15] A voltage sufficient to cause an electrical discharge through the helium vapor was first applied to the electrodes. This discharge always left the drop with a net positive charge. The amount of charge on the drop could then be determined by using the electrodes to apply a dc field. This caused a static displacement of the drop inside the trap. From a knowledge of the trap potential, the applied electric field, and the static displacement the charge could be calculated. It was typically of the order of $10^7 e$. After the drop was charged, an ac voltage of typical magnitude 10 to 100 V was applied to the electrodes to excite one of the vibrational modes. The vibration was monitored by shining a helium-neon laser bean onto the edge of the drop and detecting the refracted light with a photodiode as shown in Fig. 2. The output of the photodiode was amplified using a lock-in amplifier with reference voltage synchronized to the driving voltage applied to the electrodes.

By sweeping the frequency of the driving ac voltage applied to the electrodes, we observed several resonance peaks, corresponding to the modes of different l. It was found that the $l=2$ mode gave a larger signal than the higher modes, and so this mode was used for the measurements. The frequency of the $l=2$ mode could be determined with an accuracy of $\pm 10^{-3}$ Hz.

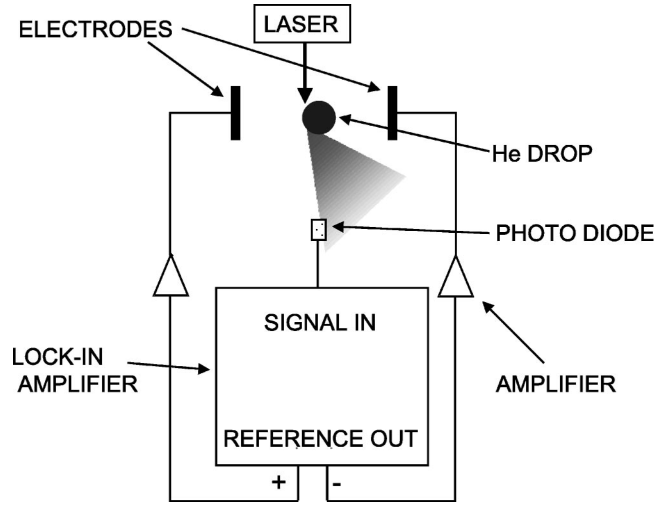

FIG. 2. Schematic diagram showing the method used to detect the vibration of a drop.

Given the geometry of our apparatus, it was not easy to make an accurate measurement of the drop diameter. The optical distance from the drop to the room temperature window was about 0.4 m. A Questar model QM-100 long working-distance microscope with an attached charge-coupled device (CCD) camera was used.[16] At a working distance of 0.4 m, a point object can be located with this system with an accuracy of ± 3.5 μm. To determine the size of a drop, the CCD image of the drop was first recorded. The scale of the image was then calibrated by recording a second image of a known length scale viewed from the same working distance. In this way, it was possible to determine the size of drop with radius around 0.1 cm to an accuracy of $\pm 0.35\%$. This leads to an uncertainty in the surface tension measurement of $\pm 1\%$. The appearance of the drop did not change when it was excited, and from this result we conclude that the amplitude of the shape oscillation was less than 10 μm.

We did not have a way to make a direct measurement of the drop temperature T_{drop}, and could only monitor the temperature T_{wall} of the cell wall. However, it is important to note that there is some bulk liquid in the bottom of the cell. This liquid must have the same temperature as the cell wall and is in thermodynamic equilibrium with the helium vapor in the cell. Hence, the pressure of the helium vapor must be $P_{\mathrm{SVP}}(T_{\mathrm{wall}})$. Because of the open cell geometry, the pressure of the helium vapor at the surface of the drop must be the same as the pressure at the cell wall. The vapor pressure at the drop surface is then[17]

$$P_{\mathrm{SVP}}(T_{\mathrm{drop}})+\frac{2\gamma}{a}\frac{v_l}{v_g}, \tag{5}$$

where v_l and v_g are the molar volumes of the liquid and gas, respectively. The pressure $2\gamma v_l/a v_g$ is very small, i.e., of the order of few dynes cm^{-2}, and as a consequence $P_{\mathrm{SVP}}(T_{\mathrm{drop}})\approx P_{\mathrm{SVP}}(T_{\mathrm{wall}})$ and so the temperature of the drop is very close to the temperature of the cell wall. However, the small difference between T_{wall} and T_{drop} does cause the drop to slowly shrink due to evaporation. This made the vibrational frequency increase slowly with time. The change of the frequency over a period of the few days that were needed in order to make a surface tension measurement did not lead to a significant error.

III. RESULTS AND ANALYSIS

Using Rayleigh's formula, the surface tension can be found from the measured frequency ω_2 of the $l=2$ vibrational mode using the equation

$$\gamma = \frac{\omega_2^2 \rho a^3}{8}. \tag{6}$$

Before presenting the results for the surface tension determined this way, we analyze a number of corrections to this relation.

(a) Charge. Rayleigh has calculated the resonant frequency for a drop that has an electrical charge Q. His result is[18]

$$\omega_l^2 = \frac{(l-1)l}{\rho a^3}\left[(l+2)\gamma - \frac{Q^2}{4\pi a^3}\right]. \tag{7}$$

For a drop with charge $10^7 e$ and radius $a \sim 1$ mm, the fractional shift in frequency of the $l=2$ mode is 6×10^{-4}. Hence, this effect can be neglected.

(b) Helium vapor. The Raleigh formula, Eq. (4), is correct for a drop in vacuum. In the presence of a second fluid of density ρ' surrounding the drop, the formula for the frequency becomes

$$\omega_l^2 = \frac{\gamma(l-1)l(l+1)(l+2)}{a^3[(l+1)\rho + l\rho']}. \tag{8}$$

Hence, the fractional frequency shift for the $l=2$ mode is

$$\frac{\delta\omega_2}{\omega_2} = \frac{\rho'}{3\rho}. \tag{9}$$

Inclusion of this effect gives a correction to the surface tension that is 0.1% at the highest temperature for which we have made measurements (1.6 K), and which decreases rapidly at lower temperatures. This correction is significantly smaller than the experimental error and we will neglect it.

(c) Viscosity. In the derivation of the Rayleigh formula, the drop is considered to be composed of a single fluid with no viscosity. The motion of superfluid helium is described by the two-fluid model. The liquid is considered to act as two interpenetrating fluids: the superfluid component that has no viscosity and density ρ_s, and the normal fluid that has viscosity η_n and density ρ_n. The sum of the densities of these two fluids equals the density of the liquid ρ. When the superfluid drop vibrates, a motion is induced in the vapor surrounding it. The dissipation in the normal fluid and the motion of the vapor result in a damping of the drop vibration, and also a shift in frequency. The theory of the damping and frequency shift is given in Ref. 19. The calculated damping rate is in good agreement with experimental measurements. We have calculated the frequency shift and find that for drops with radius in the range investigated in the present experiment, the frequency shift is less than 0.03% over the temperature range 0.7 to 1.6 K. Hence, this shift is insignificant.

(d) Effect of the trap. The potential of the trap also has an effect on the frequency of the vibrational normal modes. This theory is developed in detail in the Appendix. The result is that the five-fold-degenerate $l=2$ mode is split into three frequencies, two of which are doubly degenerate. Explicitly,

$$\omega_{20} = \sqrt{\frac{8\gamma}{\rho a^3}}(1+2A+B), \tag{10}$$

$$\omega_{21} = \omega_{2\bar{1}} = \sqrt{\frac{8\gamma}{\rho a^3}}(1+A+B), \tag{11}$$

and

$$\omega_{22} = \omega_{2\bar{2}} = \sqrt{\frac{8\gamma}{\rho a^3}}(1-2A+B), \tag{12}$$

where

$$A = \frac{19a^3}{168\gamma}(k_z - k_{xy}), \tag{13}$$

$$B = \frac{a^3}{12\gamma}(k_z + 2k_{xy}), \tag{14}$$

and k_z and k_{xy} are the coefficients entering into the trap potential, Eq. (3).

In principle, we are able to calculate the potential due to the trap from the magnet current and the known geometry of the magnet windings. As a test of this calculation, we first found the minimum current needed to levitate a helium drop, i.e., the current required to produce a field for which the condition equation (2) is satisfied at some point. The calculation gave the result 114.6 A, whereas the experimental result was 112.9 A, i.e., 1.5% lower. We then proceeded to calculate the values of the spring constants k_{xy} and k_z as a function of the magnet current. The results are shown in Fig. 3. Since we know that k_z is zero for a current of 112.9 A, we have applied a correction of 1.5% to the current to make this happen. We then find that at a current of 113.5 A, $k_z = k_{xy}$, and so the trap is spherical.

Data showing the splitting of the $l=2$ mode are presented in Fig. 4. In this example, the frequency difference between the lowest and the highest mode is 1.3%. To investigate the effect of the trap on the vibrational modes, we have made measurements of the three resonant frequencies for a series of magnet currents and the results are shown in Fig. 5. To make a comparison with theory, we have taken the mode frequency for the spherical trap as an adjustable parameter and then used Eqs. (10)–(12) to calculate the splitting as a function of magnet current. It can be seen that the agreement between theory and experiment is very good.

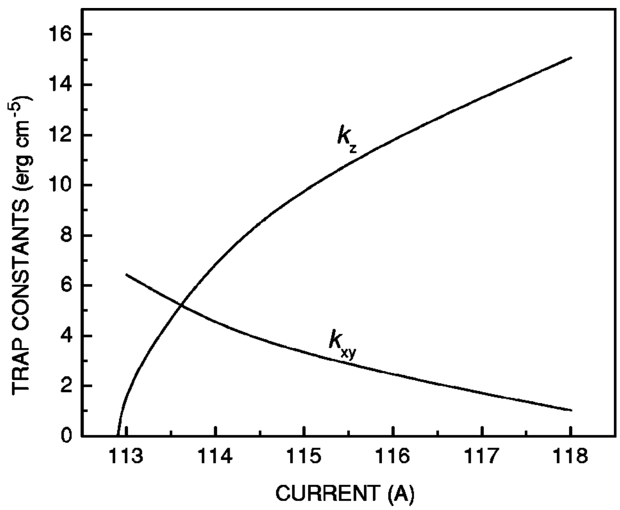

FIG. 3. The parameters k_z and k_{xy} for the trap potential as a function of the magnet current. The calculation of these parameters is described in the text.

To make surface tension measurements, we decided to work with the magnet current that gave no mode splitting (113.5 A). For this current, the effect of the trap was to raise the frequency by an amount

$$\frac{\delta\omega}{\omega} = \frac{a^3 k_z}{4\gamma}. \tag{15}$$

For a drop with radius 1 mm, this correction was $\approx 0.3\%$, and so amounted to a correction of 0.6% to the surface tension. Measurements were made on drops of radius 0.103, 0.119 and 0.132 cm. Figure 6 shows the results of surface tension measurements made between 0.6 and 1.6 K, along with the results of Zinov'eva and Bolarev,[5] Iino *et al.*,[10] and Roche *et al.*[11] It can be seen that within the experimental uncertainty of $\pm 1\%$, the results are in agreement with the data of Zinov'eva and Bolarev and of Roche *et al.*, and do

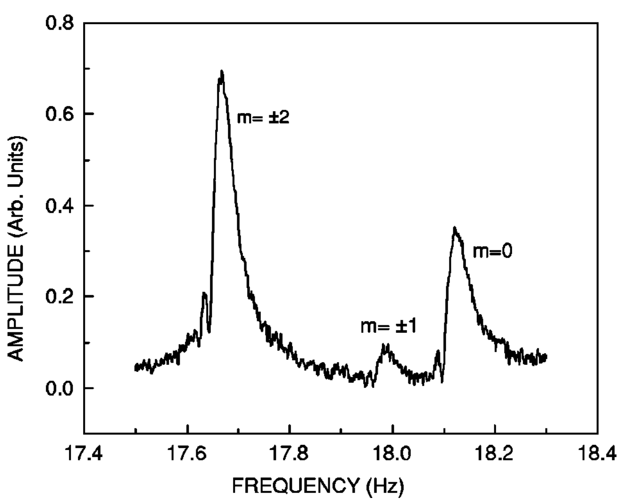

FIG. 4. Vibrational spectrum for the $l=2$ modes of a drop of radius 0.117 cm at a temperature of 1.4 K. The magnet current was 117 A. The three modes are labeled by the azimuthal quantum number m.

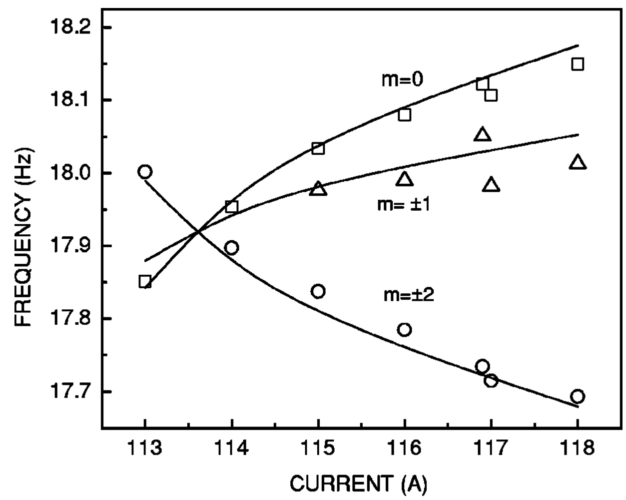

FIG. 5. Frequency of the $l=2$ modes of a drop of radius 0.117 cm as a function of magnet current at 1.4 K. The three different modes are denoted by the triangles, squares, and circles. The solid lines are the results of calculations described in the text.

not agree with the results of Iino *et al*. If we combine the value for γ that we have obtained at the lower end of our measurement range with the temperature dependence of γ found by Roche *et al*. at lower T, we can estimate that the value of γ at $T=0$ K is 0.375 ± 0.004 dyne cm^{-1}.

IV. SUMMARY

By means of measurements of the frequency of the $l=2$ vibrational modes of levitated drops, we have measured the surface tension of superfluid ^{4}He over the temperature range 0.6 K to 1.6 K. From these measurements we estimate that the value of the surface tension at zero temperature is 0.375 ± 0.004 dyne cm^{-1}. This result is consistent with the

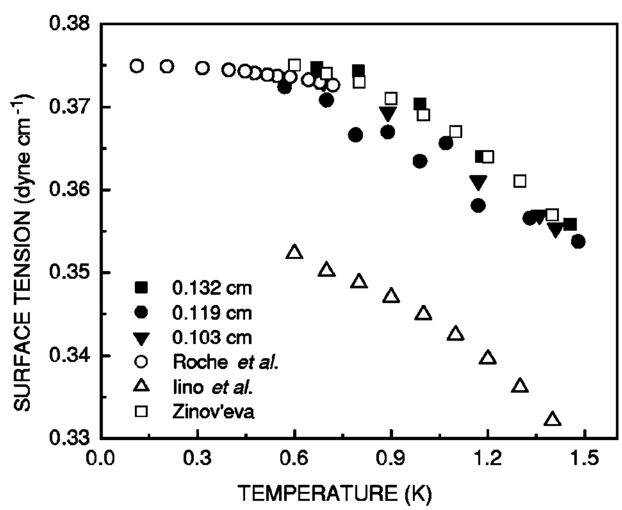

FIG. 6. Measured surface tension of liquid helium-4 as a function of temperature. The solid triangles, circles, and squares are the results from the present measurements on drops of radius 0.103, 0.119, and 0.132 cm, respectively. The results of Roche *et al*. (Ref. 11) Iino *et al*. (Ref. 10) and Zinov'eva and Bolarev (Ref. 5) are also shown.

earlier measurement of Roche *et al.*[11] We have also observed a splitting of the $l=2$ vibrational modes that arises from the anisotropy of the trap potential. The measured splitting of the modes is in good agreement with theory.

ACKNOWLEDGMENTS

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APPENDIX

The theory of the vibrational normal modes of a spherical drop in the absence of an external potential is well understood.[20] Here we consider the effect on the normal modes of a weak external potential with axial symmetry. A related problem has been discussed by Cummings and Blackburn[21] (CB) and we are able to use many parts of their calculation. They considered the vibrations of an inviscid metallic drop levitated by an alternating magnetic field. The applied field generated currents close to the surface of the drop, and the interaction of the magnet field with these currents resulted in a pressure acting on the surface of the drop. Using a form of perturbation theory, CB calculated the change in the shape of the drop and the shift in the frequency of the vibrational modes. The problem of interest in the present paper differs in an essential way from the problem considered by CB. In our problem we are concerned with a trap that exerts a body force on every part of the liquid drop, whereas in the work of CB the drop is confined solely by forces applied at the surface.

1. Description of the surface

Following CB, we write the distance to the surface of the drop in the direction θ,ϕ as

$$r_\sigma(\theta,\phi,t)=[1+R(\theta,\phi)+\zeta(\theta,\phi,t)], \tag{A1}$$

where a is the radius of the drop in the absence of any fields or gravity, $R(\theta,\phi)$ describes the static distortion of the shape of the drop, and $\zeta(\theta,\phi,t)$ represents the shape oscillation. Formally, the shape of the drop at time t can be considered to be the solution of the equation

$$\sigma(r,\theta,\phi,t)=0, \tag{A2}$$

where

$$\sigma(r,\theta,\phi,t)\equiv r-r_\sigma(\theta,\phi,t). \tag{A3}$$

Points outside the drop have $\sigma>0$. The total derivative of σ at the surface is

$$\frac{d\sigma}{dt}=\frac{\partial\sigma}{\partial t}+\vec{u}\cdot\vec{\nabla}\sigma=0, \tag{A4}$$

where $\vec{u}$ is the velocity of the fluid. From this it follows that

$$\frac{\partial^2\sigma}{\partial t^2}+\frac{\partial\vec{u}}{\partial t}\cdot\vec{\nabla}\sigma=0, \tag{A5}$$

if time-dependent product terms of order u^2 are neglected.

2. Motion of the liquid

The equation of motion of an inviscid incompressible fluid is

$$\rho\frac{\partial\vec{u}}{\partial t}=-\vec{\nabla}P+\vec{F}, \tag{A6}$$

where P is the pressure and $\vec{F}$ is the externally applied force per unit volume of liquid. This assumes that the velocity is small so that the difference between the simple and convective derivative can be neglected. We write

$$\vec{F}=-\vec{\nabla}U, \tag{A7}$$

where the trap potential U is

$$U=\rho g z-\frac{1}{2}\chi B^2. \tag{A8}$$

The equation of motion becomes

$$\rho\frac{\partial\vec{u}}{\partial t}=-\vec{\nabla}P-\vec{\nabla}U. \tag{A9}$$

P can be divided into the sum of a constant P_s and a time-dependent part P_t. Then from Eq. (A9) we must have

$$P_s=-U+P_0, \tag{A10}$$

where P_0 is a constant. Also

$$\frac{\partial\vec{u}}{\partial t}=-\frac{1}{\rho}\vec{\nabla}P_t. \tag{A11}$$

Since $\vec{\nabla}\cdot\vec{u}=0$ for an incompressible fluid, we have $\nabla^2P_t=0$. Therefore we can write

$$P_t=\sum_{lm}\left(\frac{r}{a}\right)^l Y_{lm}(\theta,\phi)\alpha_{lm}(t), \tag{A12}$$

where $\alpha_{lm}(t)$ are some coefficients. The acceleration of the surface is from Eq. (A5):

$$\frac{\partial^2\sigma}{\partial t^2}=\frac{1}{\rho}\vec{\nabla}P_t\cdot\vec{\nabla}\sigma. \tag{A13}$$

Following CB, this can be expressed as

$$\rho\frac{\partial^2\sigma}{\partial t^2}=\frac{\partial P_t}{\partial r}-\frac{1}{r^2}(\hat{L}P_t)\cdot(\hat{L}\sigma), \tag{A14}$$

where the operator $\hat{L}$ is defined as

$$\hat{L}\equiv i\left(\frac{\hat{\theta}}{\sin\theta}\frac{\partial}{\partial\phi}-\hat{\phi}\frac{\partial}{\partial\theta}\right). \tag{A15}$$

Then

$$-\rho a\frac{\partial^2\zeta}{\partial t^2}=\sum_{lm}\frac{l}{a}\left(\frac{r}{a}\right)^{l-1}Y_{lm}\alpha_{lm}+\frac{a}{r^2}\sum_{lm}\left(\frac{r}{a}\right)^{l}[\hat{L}(Y_{lm}\alpha_{lm})\cdot\hat{L}(R+\zeta)]. \tag{A16}$$

In this equation $r=a(1+R+\zeta)$. If we drop terms of the order of R^2 and second-order terms in ζ or α_{lm}, we obtain

$$-\rho a\frac{\partial^2\zeta}{\partial t^2}=\sum_{lm}\frac{l}{a}[1+(l-1)R]Y_{lm}\alpha_{lm}+\frac{1}{a}\sum_{lm}\hat{L}R\cdot\hat{L}(Y_{lm}\alpha_{lm}). \tag{A17}$$

Note that this is an equation in θ,ϕ. We can convert it to a matrix equation by multiplying by Y^*_{LM} and integrating. This gives

$$-\rho a^2\left\langle LM\left|\frac{\partial^2\zeta}{\partial t^2}\right.\right\rangle=\sum_{lm}[l\langle LM|lm\rangle+l(l-1)\langle LM|R|lm\rangle+\langle LM|(\hat{L}R)\cdot\hat{L}|lm\rangle]\alpha_{lm}. \tag{A18}$$

This can be written as

$$-\rho a^2\frac{\partial^2}{\partial t^2}\langle LM|\zeta\rangle=\sum_{lm}A_{LM,lm}\alpha_{lm}, \tag{A19}$$

where the matrix $A_{LM,lm}$ has elements

$$A_{LM,lm}=l\langle LM|lm\rangle+l(l-1)\langle LM|R|lm\rangle+\langle LM|(\hat{L}R)\cdot\hat{L}|lm\rangle. \tag{A20}$$

From Eq. (2.56) of CB, we have the identity

$$\langle LM|(\hat{L}R)\cdot\hat{L}|lm\rangle=\frac{1}{2}[L(L+1)-l(l+1)]\langle LM|R|lm\rangle-\frac{1}{2}\langle LM|\hat{L}^2R|lm\rangle. \tag{A21}$$

Therefore

$$A_{LM,lm}=\frac{1}{2}[L(L+1)+l(l-3)]\langle LM|R|lm\rangle-\frac{1}{2}\langle LM|\hat{L}^2R|lm\rangle+l\langle LM|lm\rangle. \tag{A22}$$

3. Static distortion of the surface of the drop

At the surface of the drop the pressure must be balanced by the surface tension force. Thus

$$P(\text{surface})=\gamma\vec{\nabla}\cdot\hat{n}, \tag{A23}$$

From CB, the curvature of the surface of the drop is

$$\vec{\nabla}\cdot\hat{n}=\frac{1}{a}[2+(\hat{L}^2-2)R+(\hat{L}^2-2)\zeta+4\zeta R-2\zeta\hat{L}^2R-2R\hat{L}^2\zeta]. \tag{A24}$$

We now consider a trap of the form of Eq. (3), i.e.,

$$U=U_0+[U_{00}Y_{00}+U_{20}Y_{20}]\left(\frac{r}{a}\right)^2. \tag{A25}$$

Then the condition (A23) becomes

$$-[U_{00}Y_{00}+U_{20}Y_{20}]\left(\frac{r}{a}\right)^2+P_0+\sum_{lm}\alpha_{lm}(t)Y_{lm}(\theta,\phi)\left(\frac{r}{a}\right)^l=\frac{\gamma}{a}[2+(\hat{L}^2-2)R+(\hat{L}^2-2)\zeta+4\zeta R-2\zeta\hat{L}^2R-2R\hat{L}^2\zeta]. \tag{A26}$$

If we consider just the time-independent terms, this gives

$$-[U_{00}Y_{00}+U_{20}Y_{20}]\left(\frac{r}{a}\right)^2+P_0=\frac{\gamma}{a}[2+(\hat{L}^2-2)R]. \tag{A27}$$

Let $R(\theta,\phi)=\Sigma_{lm}\langle lm|R\rangle Y_{lm}(\theta,\phi)$. Then

$$-[U_{00}Y_{00}+U_{20}Y_{20}]\left(\frac{r}{a}\right)^2+P_0=2\frac{\gamma}{a}+\frac{\gamma}{a}\sum_{lm}(l-1)(l+2)\times\langle lm|R\rangle Y_{lm}. \tag{A28}$$

Then to first order in the trap potential

$$P_0=\frac{2\gamma}{a}+U_{00}Y_{00} \tag{A29}$$

and

$$\langle 20|R\rangle=-\frac{a}{4\gamma}U_{20}. \tag{A30}$$

To lowest order in the trap potential, all the other $\langle lm|R\rangle$ amplitudes are zero.

4. Small oscillations

Now consider the time-dependent terms in Eq. (A26). We keep terms that are first order in ζ or α_{lm}, and up to first order in R:

$$-2[U_{00}Y_{00}+U_{20}Y_{20}]\zeta+\sum_{lm}\alpha_{lm}Y_{lm}(1+lR)=\frac{\gamma}{a}[(\hat{L}^2-2)\zeta+4\zeta R-2\zeta(\hat{L}^2R)-2R(\hat{L}^2\zeta)]. \tag{A31}$$

We multiply by Y^*_{LM} and integrate to obtain

$$
\begin{aligned}
&-2U_{00}Y_{00}\langle LM|\zeta\rangle-\sum_{lm}2U_{20}C_{lm20LM}\langle lm|\zeta\rangle\\
&\quad+\sum_{lm}\alpha_{lm}(\langle LM|lm\rangle+l\langle LM|R|lm\rangle)\\
&=\frac{\gamma}{a}(L+2)(L-1)\langle LM|\zeta\rangle+\frac{\gamma}{a}\sum_{lm}[4\langle LM|R|lm\rangle\\
&\quad-2\langle LM|\hat{L}^2R|lm\rangle-2l(l+1)\langle LM|R|lm\rangle]\langle lm|\zeta\rangle,
\end{aligned}
\tag{A32}
$$

where

$$C_{lml'm'LM}\equiv\int Y_{lm}Y_{l'm'}Y^*_{LM}d\cos\theta\, d\phi. \tag{A33}$$

Equations (A19) and (A32) enable the frequencies to be found. To simplify the discussion, we now consider specifically the corrections to the frequency for the five modes with $l=2$ that are degenerate in the absence of the trap potential. Since we are considering a trap with axial symmetry, in the presence of the trap each of these modes will still be characterized by a definite value of m. We then let

$$\zeta=\zeta_0Y_{2m}\exp(-i\omega_{2m}t), \tag{A34}$$

$$\alpha_{lm}(t)=\alpha_0Y_{2m}\exp(-i\omega_{2m}t), \tag{A35}$$

and solve Eqs. (A19) and (A32) to find the mode frequencies correct to lowest order in the parameters U_{00} and U_{20} to obtain

$$\omega_{2m}=\sqrt{\frac{8\gamma}{\rho a^3}}\left(1+\frac{19aU_{20}}{16\gamma}C_{2m202m}+\frac{aU_{00}}{16\sqrt{\pi}\gamma}\right). \tag{A36}$$

Using the values of C_{2m202m} we obtain:

$$\omega_{20}=\sqrt{\frac{8\gamma}{\rho a^3}}\left[1+\frac{19}{112}\sqrt{\frac{5}{\pi}}\frac{aU_{20}}{\gamma}+\frac{1}{8\sqrt{\pi}}\frac{aU_{00}}{\gamma}\right], \tag{A37}$$

$$\omega_{21}=\omega_{2\bar{1}}=\sqrt{\frac{8\gamma}{\rho a^3}}\left[1+\frac{19}{224}\sqrt{\frac{5}{\pi}}\frac{aU_{20}}{\gamma}+\frac{1}{8\sqrt{\pi}}\frac{aU_{00}}{\gamma}\right], \tag{A38}$$

$$\omega_{22}=\omega_{2\bar{2}}=\sqrt{\frac{8\gamma}{\rho a^3}}\left[1-\frac{19}{112}\sqrt{\frac{5}{\pi}}\frac{aU_{20}}{\gamma}+\frac{1}{8\sqrt{\pi}}\frac{aU_{00}}{\gamma}\right]. \tag{A39}$$

The trap potential can also be expressed as in Eq. (3), i.e., as

$$U=U_0+k_zz^2+k_{xy}(x^2+y^2). \tag{A40}$$

We then finally arrive at the results given in Eqs. (10)–(14) of the text.

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