Probing liquid surface waves, liquid properties and liquid films with light diffraction

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Abstract
Surface waves on liquids act as a dynamical phase grating for incident light. In this paper, we revisit the classical method of probing such waves (wavelengths of the order of mm) as well as inherent properties of liquids and liquid films on liquids, using optical diffraction. A combination of simulation and experiment is proposed to trace out the surface wave profiles in various situations (e.g. for one or more vertical, slightly immersed, electrically driven exciters). Subsequently, the surface tension and the spatial damping coefficient (related to viscosity) of a variety of liquids are measured carefully in order to gauge the efficiency of measuring liquid properties using this optical probe. The final set of results deal with liquid films where dispersion relations, surface and interface modes, interfacial tension and related issues are investigated in some detail, both theoretically and experimentally. On the whole, our observations and analysis seem to support the claim that this simple, low cost apparatus is capable of providing a wealth of information on liquids and liquid surface waves in a non-destructive way.

Keywords: capillary waves, optical diffraction

1. Introduction

Optical probes are usually non-destructive—they do not change the properties of the medium being probed in any significant way (unless the probe power is large). Diffraction and scattering of light, as is known over centuries, are therefore capable of providing a variety of information. In this paper, we revisit a well-known classical method of investigating liquid surface waves using light diffraction. Although this method is well studied in the context of measurements of surface tension of liquids [1], our motivation for revisiting it is primarily aimed at demonstrating its utility and capability in understanding a wider class of situations/phenomena. In particular, we investigate two aspects: surface wave profiles and liquid properties for waves on a single liquid and also for waves on liquid films.

Before we begin with the details, it is worth summarizing some of the earlier work on liquid as well as solid surface waves. Use of laser light scattering is a unique technique for a non-contact, non-invasive study of surface acoustic waves and capillary waves [2]. Technological advances directed towards diverse applications of surface acoustic waves on solid surfaces are now well established, and have resulted in important developments in the fabrication of practical devices [3–11].

However, relatively few articles deal with surface waves on liquid surfaces [12–20]. To mention a few, high frequency capillary waves at the surface of liquid gallium and the mercury liquid–vapour interface have been studied by means of quasi-elastic light-scattering spectroscopy [21]. The spatial damping coefficient of low-frequency surface waves at air–water interfaces, using a novel heterodyne light-scattering technique, has been measured and reported in [22]. Quasi-elastic light scattering has been used to study thermally excited capillary waves on free liquid surfaces over a considerably wider range of surface wavenumbers [13, 16]. Measurements
of the decay coefficient of capillary waves in liquids covered with monolayers of stearic acid, oleyl alcohol and hemicyanine are reported in [23]. Also, the measurement of the pressure–area compression isotherm in Langmuir monolayer films using the laser light diffraction from surface capillary waves is reported in [24].

In a recent article, we have shown how the effect of interfering waves on a liquid surface could be inferred from the nature of the diffraction pattern [15]. Here, we begin by proposing a combined method of simulation and experiment, sufficiently general in nature and capable of verifying the nature of the actual liquid surface wave profile. Knowing the profile, we then focus on the dispersion relation and discuss experiments to determine the surface tension and viscosity of liquids. Finally, we turn to liquid films where we try to understand the profile through various modes (surface and interface) and then go on to check the values of interfacial tension.

Section 2 describes the basic scheme of our experimental arrangement. The methodology of our experiment and typical results pertaining to the surface wave profile are discussed in section 3. In section 4, we discuss theory, observations and measurements of surface tension and viscosity for different liquids. Section 5 first discusses the theoretical platform for a surface capillary wave on liquid films. We then verify our theory with experiments. Finally, in section 6, we summarize our results with some concluding remarks.

2. Experimental details and the theoretical background

2.1. Experimental set-up

As shown in figure 1, a petri dish of diameter about 18.5 cm is filled with the experimental liquid to a depth of ∼1 cm. A metal pin with its blunt end glued vertically upright to the surface of the liquid produces a Fraunhofer diffraction pattern on a screen positioned behind the petri dish. When slightly immersed in the liquid and driven by a low-frequency sinusoidal signal generator, this exciter vibrates vertically up and down and generates the desired liquid surface waves. By varying the vertical displacement of the exciter, we can control the amplitude and frequency of the wave. The laser beam incident on the dynamical phase grating formed by the surface wave is given as

\[ \phi(x') = \frac{2\pi}{\lambda} \left( (2h \cos \theta_i) \sin \left( \omega t - \frac{K x'}{\cos \theta_i} \right) \right), \]  

where the factor of \( \cos \theta_i \) appears due to oblique incidence of the incident monochromatic light [11, 14, 15, 25]. The field strength \( E \) of the diffraction pattern can be estimated from the Fourier transform of the aperture function (in this case the surface wave produced by a single exciter). The intensity of the diffraction patterns, obtained from \( E E^* \), is given by [25, 26]

\[ I(x') = \sum_n J_n^2 \left( \frac{4\pi h \cos \theta_i}{\lambda} \right) \delta \left( \frac{x'}{\lambda z} - \frac{n}{\Lambda \cos \theta_i} \right), \]  

where \( z \) is the horizontal distance between the location of the laser spot on the liquid surface and the screen and \( \Lambda = \frac{2\pi}{\lambda} \) is the wavelength of the surface wave. \( h \) is the amplitude of the surface wave, \( x' \) is the coordinate which measures the distance of the diffraction spots from a reference point (central spot) on the observation plane. \( J_n \) is the Bessel function of order \( n \), and \( \delta (\cdot) \) denotes the Dirac delta function. The intensity distribution on the diffraction pattern vanishes at points where \( J_n \) is zero satisfying equation (2). The quantity involving the Bessel function in this equation gives the magnitude of the intensity of diffraction spots for different orders \( n \), while the positions of the spots are given by the delta function.

When we generate capillary waves by means of a single pin exciter, the amplitude of the wave at the point of oscillation is almost the same as the amplitude of the oscillating pin in a low viscous liquid [15]. The intensities of different orders depend only on the amplitude of the capillary wave \( \phi \) if other parameters such as \( \theta_i \) and \( \lambda \) are kept constant (equation (2)). Theoretical plots of wave amplitude versus the intensity ratios, \( I_0/I_1, I_1/I_0 \) and \( I_0/I_2 \), using equation (2) (keeping \( \theta_i \) and \( \lambda \) fixed) are shown in figure 2, where \( I_0, I_1 \) and \( I_2 \) are the intensities of central order, first order and second order spots respectively. In our experiment, for a particular \( \phi \), the intensity of different order spots in the diffraction pattern are measured...
In such cases, we used the ratio $I_{\text{eye}}$. One may appreciate this when we simulate the surface wave profile using realistic dimensions and parameters. In a previous study [15], we measured the surface wave parameters (wavelength, amplitude at the point of excitation) at a given oscillation frequency (220 Hz). Here, we use similar typical values to generate theoretically the distribution of waves on the liquid surface. The equations which we use to simulate the wave profile for the cases of single and double pin excitors are (at a fixed time ‘$t’$, say $t = 0$)

$$\psi_1(X, Y) = h \exp\left(-\delta \sqrt{(X - x_1)^2 + (Y - y_1)^2}\right) \times \cos\left(\frac{2\pi}{\Lambda} \sqrt{(X - x_1)^2 + (Y - y_1)^2}\right)$$

and

$$\psi_2(X, Y) = h \exp\left(-\delta \sqrt{(X - x_1)^2 + (Y - y_1)^2}\right) \times \cos\left(\frac{2\pi}{\Lambda} \sqrt{(X - x_2)^2 + (Y - y_2)^2}\right) + h \exp\left(-\delta \sqrt{(X - x_2)^2 + (Y - y_2)^2}\right) \times \cos\left(\frac{2\pi}{\Lambda} \sqrt{(X - x_2)^2 + (Y - y_2)^2}\right),$$

respectively. Here, $(x_1, y_1)$ and $(x_2, y_2)$ are the centres of oscillations and $\delta$ is the spatial damping coefficient of the liquid. To mimic the intended experiments, we assume that each wave has the same frequency $\omega$, wavelength $\Lambda$ and amplitude $h$ (at the centre of oscillation). In our simulations, we use the typical values of $\Lambda = 2.1$ mm, $h = 1.0$ mm and $D = 8.4$ mm (separation between pin exciters) [15]. $\delta$ is chosen to be 0.235 cm$^{-1}$ (for water at 220 Hz, we obtain this value from the experiments discussed later in section 4). The corresponding surface wave profiles estimated for the two cases of interest are shown in figures 3(a) and (b), respectively. Following this recipe, one can obtain the surface wave profile for any number of oscillation sources. The simulated profiles enable a better visualization, which, in turn, can act as a guideline for their actual experimental verification.

3. The surface wave profile

3.1. Simulation of the profile

Low-frequency surface waves are not visible to the naked eye. One may appreciate this when we simulate the surface by a photo-diode detector when the laser beam is focused along a radial line on the liquid surface. The intensity ratios such as $I_0/I_1$ and $I_1/I_2$ are then evaluated. By comparing our experimentally measured ratios with those obtained from the theoretical plot (figure 2), we estimate the average value of $h$. It is to be noted that, for $I_0/1$, the value of this ratio $I_0/I_1$ becomes very small and could yield erroneous results. In such cases, we used the ratio $I_1/I_0$ for estimation of $h$ (however, we were always below the limit for which this ratio becomes very large). Again, by measuring $x'$, we determined the wavelength ($\Lambda$) of the surface capillary waves to be 2.1 mm (for more details, see [15]). For several exciters placed at different types of geometric configurations (e.g. along a line, a polygon, etc), the surface wave patterns change, and hence equation (2) must also change.

3.2. Experimental results for two-source pattern

The simulated profiles are built on the assumptions made in equations (3) and (4). It is necessary to verify experimentally

![Figure 2](image1.png)

Figure 2. The theoretical plot of the wave amplitude versus intensity ratio for $\theta_1 = 77^\circ$, $\lambda = 632.8$ nm. The solid line, dashed line and dotted line stand for the ratios $I_0/I_1$, $I_1/I_0$ and $I_0/I_2$ respectively.

![Figure 3](image2.png)

Figure 3. The simulated surface capillary wave profiles for (a) a single exciter and (b) two excitors.
whether the assumptions are good enough to represent the characteristics of the surface waves. We note that the simulated two-source interference pattern (shown in figure 3(b)) exhibits hyperbolic loci for the constructive and destructive interference nodes. Using this fact as a guideline, we trace the hyperbolic loci (maxima and minima of the interfering waves) on the liquid surface by observing the changes in the corresponding diffraction patterns. Initially, when the laser beam is incident along the central line PQ (see figure 4) where constructive interference of waves occurs, the diffracted light shows, as expected [15], a central spot along with higher order ones symmetrically located on either side of the central spot (see figure 5(a)). As we shift the probe beam gradually away from the central line along a circle (note the arrow in figure 4), the number of higher order spots progressively decreases till we reach the position of the adjacent hyperbola (O₂ A), where the diffraction should ideally contain only the central order. In the experiment, however, at this position, we find the first order spots too, though with very low intensity (see figure 5(b)). This happens due to the finite spot size of the laser beam. Nevertheless, beginning with the central line on one side and gradually rotating the prism-table till we reach 180°, we observe a repetition of the patterns (shown in figures 5(a) and (b)) symmetrically at all complementary angles. We note the angular positions of all consecutive minima. These results are shown in table 1. Thus, a comparison of our experiment (figure 4) with the simulation (upper half of figure 3(b)) for identical values of parameters shows the same number (eight) of destructive interference lines at almost the same angular separation. Beyond 180° and up to 360° (i.e. while scanning the lower semicircular region in figure 4) identical patterns are observed. This confirms that equation (4) used to describe and simulate the two-source surface wave profile is, indeed, a correct representation of the observed profile. We note that our method of investigation is sufficiently general (we have indeed checked up to six exciters) and can be used for several exciters located along a line or in other geometric configurations (e.g. exciters on a triangle, quadrilateral, pentagon, hexagon, etc).

3.3. Minima distribution for oscillation sources in regular array

When multiple oscillating sources are used, different surface wave profiles can be generated by appropriately configuring their positions. It is interesting to study these interference patterns of the surface waves for multiple sources (e.g. a linear chain of oscillation sources or those placed in a closed regular geometric figure). A typical example of a simulated interference pattern for three oscillation sources is shown in figure 6. For this purpose, we choose to trace the distribution of minima lines in the simulated interference pattern in terms of the number of lines and their angular positions. It is obvious that the number of minima lines increases with the increase in the number of oscillation sources. For example, our simulation of the pattern for 2, 3, 4, 5 and 6 oscillation sources placed in a straight line with a spacing of \( D = 8.4 \) mm between adjacent two oscillators, and \( \Lambda = 2.1 \) mm, exhibits the number of hyperbolic lines respectively as 8, 16, 24, 32 and 40. To derive this behaviour theoretically, we use the interference treatment for \( n \)-oscillating sources. We consider \( n \ (\geq 2) \) sources lying in a straight line. Representing the wave generated by each oscillating source as \( h \exp(iK \cdot r_n - \omega t) \), where \( h \) is the amplitude and \( r_n \) is the position vector from the \( n \)th source to the point of observation, we find the resultant field \( \psi_P \) at a certain point P as [26]

\[
\psi_P = h \frac{\sin\left(\frac{\omega t}{2}\right)}{\sin\left(\frac{\omega t}{2}\right)} \exp(iK \cdot r_n - \omega t),
\]

where \( R = \frac{1}{2}D(n - 1)\cos(\phi) + r_1 \) is the distance from the centre of the line of oscillators to the point P and \( \beta = KD\cos(\phi) \) is the phase difference between adjacent sources. Again, \( \phi \) is the angle between \( R \) and the line joining the sources of oscillation. For minima at P, the condition is

\[
\sin\left(\frac{\omega t}{2}\right) = 0.
\]

Using the boundary condition mentioned in [15], we calculate the allowed number of minima lines and their corresponding angular positions. For \( n = 3 \), for example, the above minima condition becomes

\[
\frac{\omega t}{2} = \pm \left(\frac{\pi}{4}\right),
\]

where \( m \) is an integer but not equal to or a multiple of \( n \). If we
Figure 6. Simulated interference profile of a surface capillary wave for (a) three exciters in a straight line and (b) three exciters at the vertices of an equilateral triangle.

Table 1. The simulated and experimental angular separations between the two successive destructive minima of the two-source interference pattern (see figures 3(b) and 4).

| Angle between the lines | A–B | B–C | C–D | D–E | E–F | F–G | G–H |
|-------------------------|-----|-----|-----|-----|-----|-----|-----|
| Simulated angular differences between the two corresponding minima | 25.5° | 18° | 16° | 15° | 18° | 25.5° |
| Experimental angular differences between the two corresponding minima | 25.1° | 17.6° | 16° | 15° | 17.5° | 25.3° |

choose $\frac{D}{\Lambda} = 4$, then $\cos(\phi)$ has only 16 allowed values, i.e. 16 hyperbolic minima lines are possible. The angular distribution of these asymptotic lines perfectly matches with the observed pattern. We have carried out this comparison successfully for higher order $n$-values up to 5. From the above calculations, we have seen that the number $(N_o)$ of asymptotic minima lines in the region above (or below) the line joining the oscillation sources is directly proportional to the ratio $\frac{D}{\Lambda}$ and also the number of the interval $(n - 1)$ between the exciters. Taking care of the ± sign of $\cos(\phi)$, we can express $N_o$ as

$$N_o = \frac{2D}{\Lambda} (n - 1)$$

which gives the same result (for different $n$ values) as obtained before. This relationship can, just by measuring the number of minima lines for given $D$ and $n$, estimate $\Lambda$ of the capillary waves.

Next, we study $n \geq 3$ oscillation sources forming a closed loop shape such as an equilateral triangle, square, pentagon and a hexagon. The number $(N_c)$ of minima lines (which are lying on either side of a symmetric axis) in this case can be similarly expressed as

$$N_c = \frac{2D}{\Lambda} \left( \frac{n}{2} \right)$$

The number of lines for the above-stated closed geometrical figures at the same values of $D$ and $\Lambda$ are 12, 16, 20 and 24 respectively. We have not been able to calculate exactly the angular distribution of the minima lines. However, using the simulation mentioned above, one can determine the interference pattern due to any number of oscillation sources placed along a line or at the vertices of polygons (regular or irregular). Such a study might be useful for generating a desired dynamic phase grating aperture for diffraction-based micro-photonic systems.

4. The dispersion relation and liquid properties using light diffraction

Knowing the surface wave profile is, of course, not enough. A crucial element of any wave phenomenon is the dispersion relation, which, obviously, contains within it quantifiers of material properties (here, liquid properties, such as surface tension and viscosity). In this section we describe how we use the Fraunhofer diffraction of laser light by the surface profile of ripples on liquids, described in section 3, to study such liquid properties. Several methods to accurately determine these two properties are now well established and available in the literature. For measurement of surface tension, the techniques widely used are the capillary rise method, drop-weight method, Jaeger’s method, Rayleigh’s method, etc [27, 28]. Likewise, for the measurement of viscosity, popular methods such as Poiseuille’s method, Stokes’ method, etc are well known and give accurate results [27, 28]. Here, we focus on a single optical set-up which can measure both surface tension and viscosity of low-viscous liquids with reasonable accuracy. Though the measurement of surface tension of liquids using the Fraunhofer diffraction of laser light by surface waves is well known in the literature [1], to the best of our knowledge, the estimation of the viscosity of a liquid using this probe and, in particular, the simple method which we have followed is certainly new. In the following, we elaborate on the above-mentioned measurements in some detail.
4.1. Surface tension

Consider the well-known dispersion relation for surface capillary wave on liquids [29–31]:

\[ \omega^2 = gK + \alpha K^4/\rho. \]  

(8)

Here, \( \omega \) and \( K \) are the angular frequency and wavevector of the surface capillary wave, respectively. \( \alpha \) and \( \rho \) are the surface tension and density of the liquid and \( g \) is the acceleration due to gravity. The above relation shows that waves at the surface of the liquid are dominated by gravity as well as surface tension. Neglecting the gravitational effect and taking the logarithm of both sides of equation (8), we get

\[ \ln \omega = \frac{3}{2} \ln K + \frac{1}{2} \ln \left( \frac{\alpha}{\rho} \right). \]  

(9)

which is an equation of a straight line for \( \ln \omega \) versus \( \ln K \) with a slope of \( \frac{3}{2} \) and y-intercept \( \frac{1}{2} \ln \left( \frac{\alpha}{\rho} \right) \), which contains the surface tension \( \alpha \).

In our experiments, we have generated surface capillary wave on the liquid surface with a single exciter. As we discussed before, the expected surface wave profile (shown in figure 3(a)) acts as a reflection phase grating for the incident laser light, and the intensity distribution satisfies equation (2). If we take the separation between the central order and the first order \((n = 1)\) diffraction spots to be \( d \) (see figure 7), then from the delta function in equation (2), we get the expression for the wavenumber \( K \) as

\[ K = \frac{2\pi d}{\lambda e^\theta}, \]  

(10)

where \( \theta = \frac{\pi}{2} - \theta_0 \) is the grazing angle of incidence of the laser beam. For a small grazing angle, we can approximate \( K \) as

\[ K = \frac{2\pi ld}{\lambda e^2}, \]  

(11)

where \( l \) is the perpendicular height of the central order spot from the liquid surface level (see figure 7). As \( \omega \) changes, the distance between the bright spots of the diffraction pattern also changes. Sets of diffraction patterns are obtained on the screen for different values of \( \omega \). For each set, we have traced the positions of spots of different orders on the screen.

For Fraunhofer diffraction, values of \( z \) and \( l \) in equation (11) are quite large (342 cm and 87 cm, respectively, in our experiment). Thus, the systematic error introduced in measuring these lengths with a metre scale (with a least count of 1.0 mm) is quite small. On the other hand, a careful and accurate measurement of \( d \) (of the order of a few mm) is crucial for evaluating \( K \). From the characteristic feature of the delta function in equation (2), we find that the diffraction spots due to different orders are equidistant for a particular frequency of the capillary wave. Thus, to reduce the systematic error in measuring \( d \), we have measured the separation between the positive and the negative fourth-order (fifth-order) spots. The values of \( d \) are obtained through averaging these data. For each frequency \( \omega \), we have repeated the experiment at least four times. Hence, using equation (11), we estimate the value of \( K \) for a particular \( \omega \). In addition, it is reasonable to assume that the frequency \( \omega \) of the capillary wave is the same as that of the exciter (driven by a function generator) [15]. Thus, the values of \( \omega \) are found directly from the frequency readings of the function generator driving the exciter. Following these measurements, we plot a ln–ln graph for \( K \) versus \( \omega \) and evaluate the slope and y-intercept of this straight line. The slope of the line \((\frac{3}{2})\) should verify the power law predicted by the dispersion relation while the y-intercept should give the surface tension.

Our experimental results on three different liquids (water, 1-hexanol and kerosene—a mixture of petroleum hydrocarbons) are shown in figure 8. The nonlinear least-squares fit (shown by lines in figure 8) to the data with equation (9) verifies the 1.5 exponent of the dispersion relation within experimental uncertainty (1.5 ± 0.03). Using the known value of \( \rho \) for each liquid, we estimate the value of \( \alpha \) from the measured y-intercept of the straight lines. It is to be noted that the maximum percentage error in measuring \( \alpha \) is limited to 6–8% (for different liquids) by our measurement procedure. We have tabulated the measured values of the surface tension for different liquids in table 2. These results match quite well with the standard values of the surface tension for the corresponding liquids [32].

4.2. Viscosity

Using the same experimental set-up, we now move on towards estimating the viscosity of a given liquid. We mentioned before that for capillary waves generated by means of a single pin exciter, the amplitude of the wave at the point of oscillation is almost the same as the amplitude of the oscillating pin in a low viscous liquid [15]. However, if we imagine a radial straight line from the source of excitation (on the liquid surface), then along this line the amplitude of the surface wave is gradually damped due to the viscosity of the liquid (see figure 7). The
We have seen in section 2 that the intensities of the diffraction on the liquid surface from the point of excitation (laser beam is focused along a radial line at different distances)

Table 2. The experimental and standard values of surface tension ($\alpha$) and viscosity ($\eta$) of different liquids at room temperature (25°C). For standard values of $\alpha$ and $\eta$, see [32].

| Liquids   | $\alpha$ (dyn cm$^{-1}$) (experimental) | $\alpha$ (dyn cm$^{-1}$) (standard) | $\delta$ (cgs units) | $\delta$ (cgs units) (experimental) | $\eta$ (cp) (experimental) | $\eta$ (cp) (standard) |
|-----------|----------------------------------------|-------------------------------------|----------------------|-------------------------------------|--------------------------|------------------------|
| Water     | 75 ± 5.3                                | 72                                  | 0.235 ± 0.026        | (0.92 ± 0.1) at 220 Hz              | 0.92 ± 0.08 at 260 Hz    | 0.89                   |
|           |                                        |                                     | (0.304 ± 0.031)      | (0.87 ± 0.09) at 300 Hz             |                          |                        |
| Kerosene  | 26.3 ± 3.7                              | 28                                  | 0.947 ± 0.04         | (1.44 ± 0.06) at 220 Hz             | 1.43 ± 0.07 at 260 Hz    | 1.40                   |
|           |                                        |                                     | 1.11 ± 0.05          |                                      |                          |                        |
| 1-Hexanol | 28.5 ± 3.4                              | 26                                  | 2.75 ± 0.17          | (4.27 ± 0.26) at 220 Hz             |                          | 4.58                   |

Figure 9. The experimental plot of the distance ($x$) versus surface capillary wave amplitude ($h$). The solid line, dashed line and dotted line are for the liquids water, kerosene and 1-hexanol, respectively.

The wavevector for a wave of a fixed real frequency $\omega$ has a small imaginary part, which contributes to the spatial damping. Substituting $K = K_0 + i\delta$ where $\delta \ll K_0$ in the well-known Navier–Stokes equation for waves at a liquid–air interface, one gets [22, 33]

$$\delta = \frac{4\eta\omega}{3\alpha},$$

where $\eta$ is the viscosity of the liquid. Experimentally, we have obtained the values of $\alpha$ and $\delta$ for the above-mentioned liquids. The estimated values of viscosity of different liquids, using equation (12), are shown in table 2. The experimental plots in figure 9 are for frequency 220 Hz. We have also checked our results for two other frequencies (260 Hz and 300 Hz) and have obtained similar results (see table 2).

5. Liquid films on liquids: theory and experiments

Till now we have been exclusively concerned with waves on the surface of a single liquid, or, more precisely, waves at the liquid–air interface. A more complicated scenario arises when there exists a film of a different liquid on top of a given liquid (the two liquids are immiscible). In such a situation, the dispersion relation changes drastically, which, in turn, changes the surface wave profile. We now investigate the novelties that arise through studies with our optical probe.

5.1. Background theoretical framework

The dispersion relation in equation (8) for capillary wave propagation on liquid surfaces is valid only when the depth of the experimental liquid is large. For lower values of the depth of the liquid (denoted by, say, $y$), the relation becomes [30]

$$\omega^2 = \frac{\alpha K^3}{\rho} \tanh(Ky),$$

where, here too, we neglect a term due to gravity. Typically, if we choose $y = 1$ cm and a range of $K$ from 20 to 80 cm$^{-1}$ (as used in our earlier experiment), $\tanh(Ky)$ is nearly unity, and the dispersion relation reduces to the one given by equation (8). The interesting problem we focus on now is that of wave propagation on a liquid film spread out on another liquid. If the thickness of the film is very low, then not only the surface tension but also the interfacial tension across the liquid–film boundary plays a role in the propagation mechanism of the waves. We first work out a general dispersion relation where the surface and interfacial tensions as well as the film thickness appear explicitly in our mathematical expressions.

Figure 10 shows the symbols used to represent various parameters in the following discussion. We denote the density of the lower liquid by $\rho$ while $\alpha$ is the interfacial tension at the liquid–liquid interface. The same quantities for the film–air interface are represented with primes, i.e. $\rho'$ and $\alpha'$ respectively. The equilibrium plane of separation (interface) between the liquids is at $y = 0$. The liquid below the $y = 0$ plane extends up to a value $y = -H$ with $H$ being very large. The film is located above $y = 0$ with a thickness $y' \ll H$. Under these assumptions, the general expressions for the surface capillary waves in region I: $-H \leq y \leq 0$ and in region II: $0 \leq y \leq y'$ are given by [30]

$$\psi = A e^{Kx} \cos(Kx - \omega t),$$
and

\[
\psi' = [B e^{-K_y} + C e^{K_y}] \cos(Kx - \omega t),
\]
respectively. Here \(A, B\), and \(C\) are arbitrary constants. The above two waves must obey the following boundary conditions arising from the continuity of the velocity component \(v_y\) and that of the pressure at the surface of separation of the two liquids, i.e.,

\[
\frac{\partial \psi}{\partial y} = \frac{\partial \psi'}{\partial y},
\]

\[
g \frac{\partial \psi}{\partial y} + \rho \frac{\partial^2 \psi}{\partial y^2} = \alpha \left( \frac{\partial^2 \psi}{\partial x^2} \right) + g \rho' \left( \frac{\partial \psi'}{\partial y} \right),
\]

\[
\rho' \frac{\partial \psi'}{\partial y} + \rho \frac{\partial^2 \psi'}{\partial y^2} - \alpha' \frac{\partial}{\partial y} \left( \frac{\partial^2 \psi'}{\partial x^2} \right) = 0.
\]

Evidently, the first two conditions apply at \(y = 0\) while the third one is valid at the top surface at \(y = y'\). Under the boundary conditions above, we neglect the effect of gravity in all calculations below. Using equations (14), (15) in equations (16), (17), we obtain

\[
A = C - B
\]

and

\[
C = B \frac{\alpha K^3 - (\rho + \rho')\omega^2}{\alpha K^3 - (\rho - \rho')\omega^2}.
\]

Substituting equations (19), (20) in equation (18) with \(y = y'\), we construct a quadratic equation for \(\omega^2\) given by

\[
\omega^4 \left[ 1 + r \tanh(Ky') \right] - \omega^2 \left[ \omega'^2 \tanh(Ky') + r \omega^2 + \omega'^2 \right] + \omega^2 \omega'^2 \tanh(Ky') = 0,
\]

where we have used the notation \(\frac{\omega'}{\rho} = r, \frac{\omega K}{\rho} = \omega'^2\) and \(\frac{\omega K}{\rho} = \omega'^2\) for simplicity. The two roots of equation (21) are the new dispersion relations for our given problem. The two roots are

\[
\omega^2 = P \pm \sqrt{Q},
\]

where

\[
P = \frac{\rho K^2 + \omega'^2 \tanh(Ky') + \omega'^2}{2Q}, \quad Q = 1 + r \tanh(Ky') \quad \text{and} \quad G = P^2 - 4Q \omega^2 \omega'^2 \tanh(Ky')).
\]

Consider two limiting cases of the above dispersion relation. Let us first assume \(y' \to \infty\). Then \(\omega^2 \to \omega'^2\) and \(\omega^2 \to \omega'^2 \frac{\alpha K}{\rho} \). Thus, when \(y'\) is very large, the \(\omega_s\) mode corresponds to the waves propagating on the upper liquid surface (thus called the surface mode). On the other hand, the \(\omega_s\) mode involves the interfacial tension \([30]\) as well as the density of the lower liquid and may be termed an interface mode. In such a situation \((y' \to \infty)\), by reflecting the probe laser beam off the surface wave profile on the liquid film, we can obtain information (through the diffraction pattern) on wave propagation and properties of the liquid film. The existence of the other mode can be confirmed (for \(y' \to \infty\)) only if we perform experiments in transmitted light, though it is not clear how such information may be obtained. In contrast, assuming \(y' \to 0\) (very thin film), we get \(\omega^2 \to (\omega^2 + \omega'^2) = \frac{\rho K^2}{\rho} \) and \(\omega^2 \to 0\). Thus, the \(\omega_s\) mode now depends on properties of both the liquids \((\alpha', \rho)\) and the interface \((\alpha)\).

Furthermore, we can obtain the constants \(A, B, C\) and hence \(\psi\) and \(\psi'\) by assuming a value for the amplitude imparted initially at the top surface. In this way, one may obtain the amplitudes of \(\psi\) and \(\psi'\) for both the modes \(\omega_s\) and \(\omega_s\), respectively. In particular, we have noted (not demonstrated here) that the nature of variation of the amplitudes of \(\psi\) (with the film height \(0 < y' < h\)) for the \(\omega_s\) mode is quite different from that for the \(\omega_s\) mode. In the former case \((\alpha_s)\), the value of the amplitude drops to a much smaller value at the interface compared to the drop in the amplitude for the \(\omega_s\) mode. However, it is not possible for us to check this fact through our experiments. Hence, we refrain from discussing this aspect further in this paper.

5.2. Experiments and results

In our experiments, we have tried to figure out which of the two different modes (discussed in the previous section) that arise due to the presence of films of kerosene, \(n, n\)-dimethyl aniline or \(o\)-xylene spread on the surface of water is actually present. The choice of these liquids is due to the fact that these are immiscible with water, have density lower than water, are low viscous and have a higher boiling point. In these experiments with films, a petri dish of area \(\sim 63 \text{ cm}^2\) is filled with water to a depth of nearly \(1 \text{ cm}\) and the liquid film is formed by adding a measured quantity of the second liquid in drops from a micro-pipette. The thickness of the film is estimated from the known volume of the liquid and the surface area of the petridish. Uniformity of the films on the water surface has been checked optically. In order to avoid any decrease in thickness due to evaporation, we have taken observations within \(5 \text{ min}\) of the formation of the uniformly spread film. The measurements are of the same type as those mentioned while studying surface tension (discussed in the earlier section) except that, in the present case, we do the experiments for varying film thicknesses. Here, we show our experimental results only for \(o\)-xylene films of different thicknesses on water. For films of the other liquids mentioned above, on water, we have observed similar behaviour.

To find out the mode which is present on the film surface for a liquid film of \(o\)-xylene on water, we first plot the experimental \(\omega \sim K\) behaviour with the film thickness as a parameter. The theoretically obtained \(\omega \sim K\) curves for the different modes \(\omega_s, \omega_s\) and \(\omega_s\) (as a limiting case of the \(\omega_s\) mode) are then placed on the same graph for a comparison. Here, we use standard values, \(\alpha = 37.2 \text{ dyn cm}^{-1}\).
Figure 11. The theoretical plot of the frequency ($\omega$) versus wavevector ($K$) for different modes in water-o-xylene liquid–liquid film at film thickness (a) 0.158 cm, (b) 0.016 cm and (c) 0.032 cm. The solid line is for the $\omega_+$ mode, the dashed line is for the $\omega_-$ mode and the dotted line is for the $\omega_0$ mode. The experimental data points are shown by the filled circles.

Figure 12. Nonlinear curve fitting of the experimental data for water-o-xylene film of thickness 0.016 cm with $\omega_+$ and $\omega_-$ modes.

$\alpha' = 29.8 \text{ dyn cm}^{-1}$, $\rho' = 0.88 \text{ g cm}^{-3}$ and $\rho = 1 \text{ g cm}^{-3}$ in equation (22) [32]. It is evident that for a film of thickness as low as 0.016 cm, the measured data correspond to the $\omega_+$ mode (figure 11(b)), while for films of thickness typically 0.158 cm, the experimental data match well with the $\omega_-$ mode (figure 11(a)). However, any intermediate thickness between 0.016 and 0.158 cm yields data within the $\omega_0$ and $\omega_+$ modes (figure 11(c)). This clearly shows that the $\omega_0$ mode always dominates over the $\omega_-$ mode. For thicker films, the $\omega_0$ mode coincides with the $\omega_+$ mode, whereas for thinner films the mode is still $\omega_+$, though it is modified by the presence of the interfacial tension and the density of the lower liquid, and is not equal to $\omega_0$.

The above conclusions have been further verified by analysing the experimental data for the estimation of the surface and interfacial tensions. We have fitted our experimental data of water-o-xylene film of thickness 0.016 cm with the $\omega_+$ and $\omega_-$ modes in equation (22) by nonlinear curve fitting (keeping surface tension and interfacial tension as varying parameters). The $\omega_+$ mode fits with the experimental data and estimates the values of surface tension ($\alpha'$) of o-xylene as (29.5 ± 3.6) dyn cm$^{-1}$ and the values of interfacial tension ($\alpha$) as (33.2 ± 8.8) dyn cm$^{-1}$ (solid line in figure 12). These values are very close to the standard values available in the literature [34]. On the other hand, the $\omega_-$ mode fails to fit the experimental data well (dashed line in figure 12); moreover, it yields unphysical values of surface and interfacial tensions. Experimental limitations, in our present set-up, did not allow us to make films of thickness lower than the values quoted above.

6. Concluding remarks

In this paper, we describe a simple optics-based experimental technique devised with the necessary background theoretical formulation to study the characteristics of surface capillary waves on liquids. Firstly, we visualize the profile of the surface waves using simulations with realistic values for the parameters involved. The locations of the nodes and antinodes which appear in the simulated profile are used to match the profile with actual experiments. We also claim that this combined method of simulation and experimentation can be used for other geometric configurations of oscillating sources on liquid surfaces. Subsequently, the two key liquid properties, surface tension and viscosity, which appear in the dispersion relation and play an integral role in the propagation of such capillary waves on a liquid surface are measured optically using Fraunhofer diffraction of laser light. We note, in particular, the novelty and simplicity in the measurement of viscosity and the fact that our measurement tools are non-destructive, but by no means less precise. The results of our experiments with several liquids, when compared to the respective values known from other sources and literature, summarily establish the efficacy and accuracy of our approach. We anticipate, in future, the introduction of technological sophistications in our set-up, which might lead to an ‘optical surfacetensometer cum viscometer’. The advantage of having a single set-up for measuring both these properties need not be further emphasized. Finally, we investigate, both experimentally and theoretically, an interesting aspect of capillary wave characteristics involved with a liquid film placed on the surface of an immiscible liquid. The dispersion relation we frame to study this case not only explains the features of the surface and interfacial modes, but also provides an estimate of the interfacial tension across the film–liquid boundary. In summary, our experimental and theoretical results seem to demonstrate the capability of our set-up in carrying out studies pertaining to capillary wave profile, liquid properties and waves on liquid films on liquids. Despite its limitations, the wide variety of information which can be obtained from such a simple set-up makes it a tool worth improving upon in future investigations.

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