Liquid refractometer based on immersion diffractometry

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Abstract: This study presents a laser diffractometric refractometer for measuring the refractive index of liquids. The refractive index is determined by rotating a reflection grating that is immersed in the fluid under test, and measuring the first-order Littrow diffraction angle. The Littrow angle is easily detected form the interferogram formed by the diffracted beam from the grating and the reflected beam from the liquid surface. No special cell for liquids is required. The alignment and measuring processes are simpler than those of other refractometers. The results of a feasibility experiment reveal that the accuracy of the proposed approach is about 0.003 for a refractive index of around 1.3.

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1. Introduction

Accurately determining the refractive index of liquids is important in industrial applications, such as finding fluids that are suitable for immersion photolithography [1, 2]. Various laser refractometers for liquids have been proposed. They are commonly based on either refraction [1-4] or interference [5-8]. Ordinarily, refractometers require a special container with which to fill the fluid. A prismatic cell, a rectangular cell, a 90° inverted-prism cell, and a stepped cell are the already known structures of the container. To ensure high accuracy requirements, the cell walls must be planar and parallel. Furthermore, the parameters of the glass cell must be

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known precisely. These include the refractive index of the cell, the angles of the surfaces, the separation between the walls, and the thickness of the walls.

Interferometric refractometers are sensitive to the refractive index but also to environmental disturbances. The interference signal is ambiguous regarding the direction of phase changes. Hence, a prism goniometer, a Hilger-Chance refractometer, and an Abbe refractometer are frequently applied in making routine measurements of liquids. Measuring the angle of refraction of the laser beam through the fluid-filled cell provides information on the refractive index. Conventional refractometers provide the direction of the refracted beam using a telescope and eye, in a cumbersome measuring procedure with limited accuracy. Current technology overcomes these limitations by employing a position sensitive detector [8, 9], or a photodiode detector with a pinhole or slit [2] to detect the centroid of the laser beam.

This work develops a new diffraction-based refractometer for liquids. The concept is derived from our earlier work on measuring nanoscale grating periods [9] and the experiments of Refs. [10, 11]. The proposed refractometer does not require a special cell to contain the tested liquid. The refractive index is obtained by measuring the first-order Littrow diffraction angle of a reflecting grating that is immersed in the liquid. The determination of the Littrow angle is based on the interferogram because of the interference between the diffracted beam from the grating and the reflected beam from the liquid surface. Accordingly, the alignment and measuring processes are much simpler than with other refractometers. This study describes the general principles and features of the proposed scheme, and presents the results of feasibility testing.

2. Operation principles

Figure 1 depicts a diffractometric liquid refractometer. A vessel contains the investigated liquid and a reflection grating with a known grating period *p*.

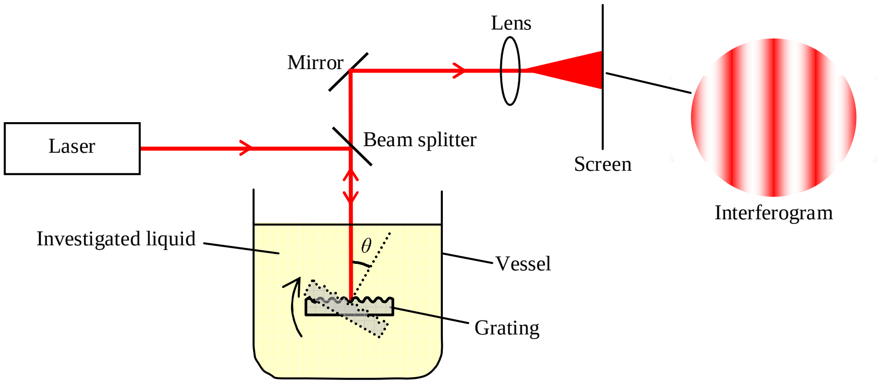

Fig. 1. Diffractometric liquid refractometer

Part of the laser beam that is reflected by a beam splitter strikes the liquid surface at normal incidence. At the air-liquid interface, a small portion of the laser beam is reflected. The rest of the laser beam enters into the liquid and is diffracted by the grating. Two light spots that are associated with the reflected and diffracted beams appear on the screen. The position of the reflected light spot can be referred to as the spatial reference position if the liquid surface is still. The immersion grating is rotated such that the diffracted beam is collinear with the reflected beam. When autocollimation occurs, the superposition of the two return beams produces a line-fringe interferogram on the screen. The diffracted beams of interest in this investigation are zero and first orders. The autocollimation conditions of zero-order and first-order diffracted lights correspond, respectively, to the grating in the normal incidence and Littrow configuration. If the difference between two autocollimation angles is

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θ, also called the first-order Littrow diffraction angle, then the refractive index of the investigated liquid n_s is related to the diffraction θ by

$$n_s = \frac{\lambda_v}{2p\sin\theta} \tag{1}$$

where λ_v is the vacuum wavelength of the laser.

Figure 2 plots the refractive index n_s as a function of the diffraction angle θ for four different wavelength-to-period ratios λ_v/p. In the range of refractive indices from 1.0 to 2.0, a larger λ_v/p corresponds to higher measurement sensitivity. In this study, λ_v/p is around 0.76.

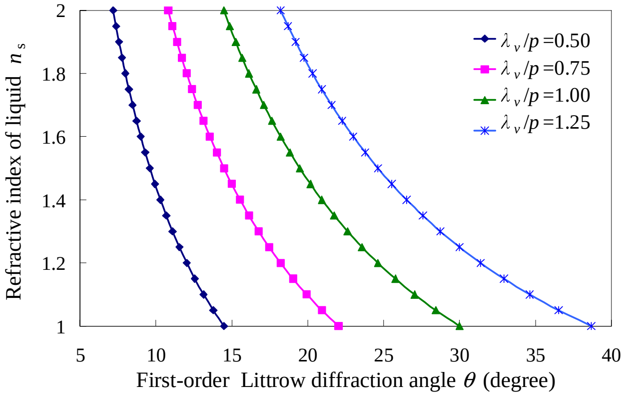

Fig. 2. Relationships between the refractive index of a liquid n_s and the diffraction angle θ corresponding to different wavelength-to-period ratios λ_v/p.

3. Experiments

Figure 3 shows the experimental configuration that was used to demonstrate the feasibility of the proposed method. The configuration is similar to that in Fig. 1 except that the grating and vessel are rotated simultaneously.

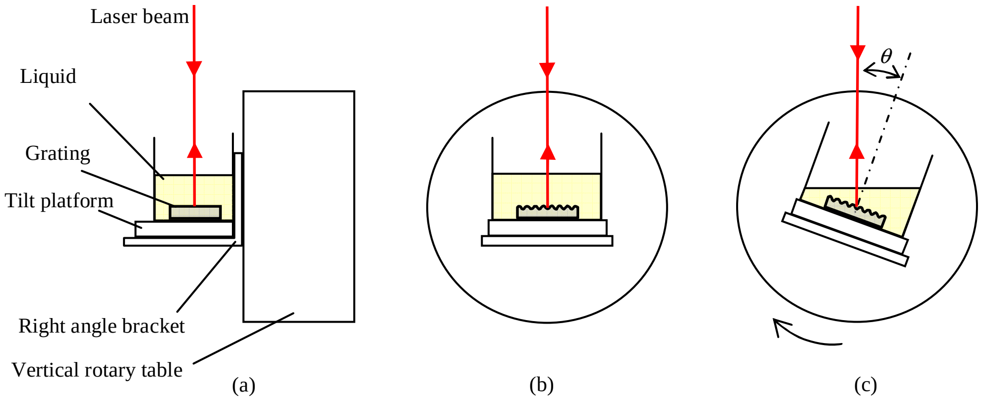

Fig. 3. Experimental configuration used to measure the refractive index of a liquid. (a) Side view (b) Front view (c) Grating in Littrow configuration.

A holographic grating with 1200 groves/mm is attached to the bottom of an ordinary vessel. The vessel is fixed on a tilt platform. A right angle bracket is adopted to connect the tilt platform to a vertical rotary table. The angular resolution and repeatability of the rotary table are 0.0025° and 0.02°, respectively. The light source used in this experiment is a 633 nm He-Ne laser. The fluid to be measured is poured into the vessel. First, the optical components

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are arranged such that the laser beam strikes the liquid surface at normal incidence. Next, the tilt platform is adjusted until the zero order diffracted beam from the grating and the reflected beam from the air-liquid interface are collinear. This requirement is met when the number of the line fringes in the interferogram, formed by the diffracted and reflected beams, is less than one. The grating surface is now perpendicular to the laser beam as presented in Fig. 3(b). The angle position of the rotary table is set to zero. Finally, the table is rotated to find the next angular position at which the interferogram again appears. At that position, the grating is in the Littrow configuration, as displayed in Fig. 3(c), and the angle position of the table is equal to the first-order Littrow diffraction angle θ.

4. Results and discussions

The grating period p and wavelength λ_v must be known to determine the refractive index of liquid n_s from the measured Littrow angle θ. The grating period is 833.36 nm which was measured using the same apparatus but without liquid in the vessel. The measurement procedures are described in our previous work [9]. The laser wavelength is 632.9909 nm, checked by a wavelength meter with a relative accuracy of 1×10^{-7}. The temperature of the tested liquid was measured by a thermometer and was confirmed to within 25±0.5 ºC. Each refractive index measurement takes about two minutes since the liquid in the vessel needs time to become stabilized. Table 1 lists the measured refractive indices of four different liquids. Comparing the measured refractive indices n_s with the reference values n_{ref} yields a minimum deviation of 0.0005 for water and a maximum deviation of 0.0025 for ethanol.

Table 1. Measured refractive indices of four different liquids

Liquid	θ(degree)	n_s	n_{ref} [3, 8]
Methanol	16.6113	1.3285	1.32684
Water	16.5684	1.3318	1.33128
Ethanol	16.2008	1.3612	1.35867
Microscope immersion oil (Nikon 50 Type A)	14.5300	1.5138	1.51410[a]

[a] Refractive index for λ_v = 587.6 nm at 23 ºC

The uncertainty of the refractive index measurement made by the proposed method primarily depends on the errors in the laser wavelength λ_v, the grating period p, and the measured Littrow angle θ. The effects of these error sources are separately discussed below.

4.1 Instability of laser wavelength

The sources of error in the wavelength are the relative accuracy of the wavelength meter and the variation in laser wavelength. The latter dominates since the light source is a multimode He-Ne laser. The worst wavelength variation is expected to be 1×10^{-6}. The error in the refractive index measurement δn_s caused by the wavelength instability $\delta\lambda_v/\lambda_v$ can be derived by differentiating Eq. (1):

$$\delta n_s = n_s \cdot \frac{\delta\lambda_v}{\lambda_v} \tag{2}$$

According to above equation, $|\delta n_s|$ is 1.3×10^{-6} for n_s=1.3.

4.2 Uncertainty in grating period

The relationship between the error in the measured refractive index δn_s and the relative uncertainty in the grating period $\delta p/p$ is given by

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$$\delta n_s = -n_s \cdot \frac{\delta p}{p} \tag{3}$$

The grating period was determined using the same apparatus without a liquid in the vessel. The measurement equation is similar to Eq. (1) and is given by

$$p = \frac{\lambda_v}{2n_a \sin\theta} \tag{4}$$

where n_a is the refractive index of air. The uncertainty of the measured grating period is governed mainly by the accuracy of the rotary table. Differentiating Eq. (4) with respect to angle θ gives

$$\frac{\delta p}{p} = -\left[\left(\frac{2n_a p}{\lambda_v}\right)^2 - 1\right]^{\frac{1}{2}} \cdot \delta\theta \tag{5}$$

Substituting n_a=1.000268, λ_v/p=0.76, and $\delta\theta=(2\times0.02^2)^{1/2}\approx0.028°$ into Eq. (5) yields $|\delta p/p|=1.2\times10^{-3}$. Therefore, according to Eq. (3), $|\delta n_s|$ is 1.6×10^{-3} for n_s=1.3.

4.3 Error in Littrow angle

The error in the measured refractive index δn_s is related to the error in the measured Littrow angle $\delta\theta$, which is given by

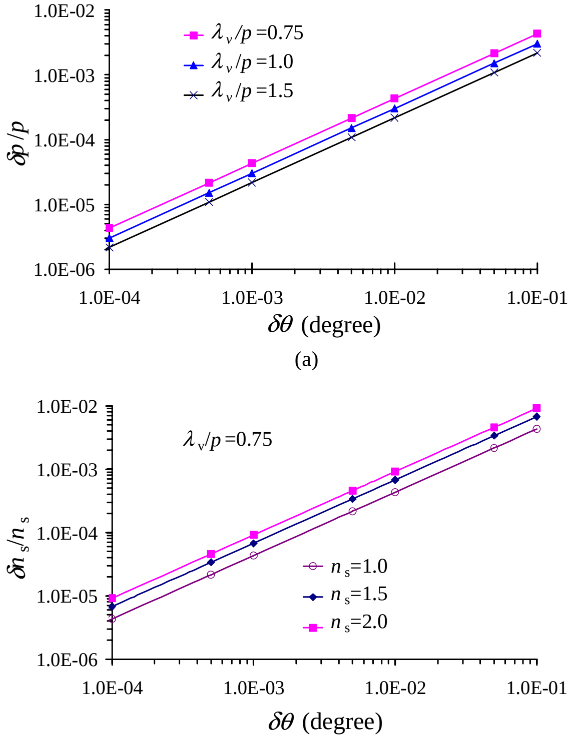
$$\delta n_s = -n_s \cdot \left[\left(\frac{2n_s p}{\lambda_v}\right)^2 - 1\right]^{\frac{1}{2}} \cdot \delta\theta \tag{6}$$

This work examines the interference pattern, formed from the diffracted and reflected beams, to determine the Littrow angle. The number of the line fringes in the interferogram is less than one when the Littrow condition is fulfilled. Thus, the angular sensitivity equals a quarter wavelength divided by the beam diameter. The diameter of the laser beam is around 1 mm so the angular error that arises from the interferometric method is approximately 0.01°. Another angular error is associated with the rotary table. The angular repeatability of the rotary table is 0.02°. Consequently, the combined angular uncertainty is the root-sum-square of these angle errors, $\delta\theta=(2\times0.01^2+2\times0.02^2)^{1/2}\approx0.032°$. Substituting $\delta\theta$=0.032° and λ_v/p=0.76 into Eq. (6) yields $|\delta n_s|=2.4\times10^{-3}$ for n_s=1.3.

Table 2. Sources of error in the measurement of refractive index by the proposed method

Error sources	Symbols	Magnitude	Errors for n_s= 1.3
Wavelength	$\delta\lambda_v/\lambda_v$	1.0×10^{-6}	1.3×10^{-6}
Grating period	$\delta p/p$	1.2×10^{-3}	1.6×10^{-3}
Littrow angle	$\delta\theta$	0.032°	2.4×10^{-3}
Combined error = $[(1.3\times10^{-6})^2+(1.6\times10^{-3})^2+(2.4\times10^{-3})^2]^{1/2} \approx 0.003$			

Table 2 summarizes the sources of error in the proposed method. The dominant errors are in the grating period and the Littrow angle. The former is governed mainly by the rotary table, and the latter is partially subject to the rotary table as well. Figures 4(a) and 4(b) show the effect of $\delta\theta$ on $\delta p/p$ and $\delta n_s/n_s$, respectively. Employing a rotary table with high accuracy can greatly reduce the angular error. If a rotary table with an accuracy of 0.001° is used, then the angular error is $(2\times0.001^2)^{1/2}\approx0.0014°$. According to Eq. (3) and Fig. 4(a), the angular error leads to $|\delta n_s/n_s|=|\delta p/p|\approx6\times10^{-5}$ for $\lambda_v/p=0.75$. Both the rotary table and the interferometric method contribute to the error in the Littrow angle. Expanding the diameter of the laser beam also increases the accuracy of the Littrow angle detection. If the laser beam is expanded to a diameter of 10 mm, then the combined angular error is $(2\times0.001^2+2\times0.001^2)^{1/2}\approx0.002°$. Referring to Fig. 4(b), $|\delta n_s/n_s|\approx1.3\times10^{-4}$ for $n_s=1.5$. Accordingly, the combined relative error of the refractive index measurement is $[(6\times10^{-5})^2+(1.3\times10^{-4})^2]^{1/2}\approx1.4\times10^{-4}$, and the refractive index can be accurately determined to the fourth decimal place.



(a)

(b)

Fig. 4. Graphs of (a) $\delta p/p$ versus $\delta\theta$ and (b) $\delta n_s/n_s$ versus $\delta\theta$.

5. Conclusion

This work demonstrates the feasibility of using a laser refractometer based on immersion diffractometry for measuring the refractive index of liquids. Unlike other methods, the proposed method does not involve a special cell. Moreover, alignment and measuring processes are simple. A reflecting grating is immersed in the fluid. The grating is rotated, and interferometric method is utilized to find the Littrow angle. The refractive index is determined from the measured Littrow angle. Preliminary results indicate that the accuracy of the proposed method is about 0.003 for a refractive index of around 1.3. The accuracy can be as high as 0.0001 by expanding the diameter of the laser beam and using a rotary table with high accuracy.

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