An improved differential algorithm for the critical-angle refractometer*

YE Jun-wei (叶骏伟)[1]**, **XIA Min** (夏珉)[2], **and YANG Ke-cheng** (杨克成)[2]
1. Wuhan National Laboratory for Optoelectronics, Huazhong Institute of Electro-Optics, Wuhan 430223, China
2. School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China

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Due to the limit of the pixel size of the charge-coupled device (CCD) or complementary metal oxide semiconductor (CMOS) sensor, the traditional differential algorithm has a limited measuring accuracy by determining the critical angle in integral pixel. In this paper, we present a practical algorithm based on the centroid value of the reflective ratio around the critical angle pixel to address the traditional differential algorithm problem of determining the critical angle under sub-pixel in a critical angle refractometer (CAR). When the change of refractive index (RI) of a liquid sample is beyond the sensitivity of the traditional differential algorithm, the RI of the liquid can be obtained by using the centroid value of reflectivity around the critical angle pixel. The centroid value is associated with the RI change of the liquid in sub-pixel. Demonstrated by both theoretical analyses and experimental results using saline solutions with RI that changes in sub-pixel tested through the reflective CAR, the algorithm is found to be computationally effective and robust to expand the measuring accuracy of the Abbe-type refractometer in sub-pixel.
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The reflective Abbe-type critical angle refractometer (CAR) has been widely used for refractive index (RI) measurement in the science and engineering fields for its advantages of high environment immunity, high accuracy and high stabilization[1]. Traditionally, the digital CAR is composed of a divergent light source, an optical prism, a lens and a charge-coupled device (CCD) camera. The CAR measures the RI of a liquid by measuring the reflectance of a divergent light beam from the glass-liquid interface. It is based on finding the critical angle position between the total internal reflection (TIR) region and non-TIR region, and establishing the function between the critical angle and the refractive index[2-5].

The reflective ratio curve of a digital CAR is traditionally obtained by two steps. The first step is measuring the reflective beam intensity E_{liquid} of the glass-liquid interface, and E_{liquid} is the product of the light source intensity and the reflective ratio curve. The second step is measuring the reflective beam intensity E_{air} of the glass-air interface, and E_{air} is the light source intensity[2]. Owing to the non-simultaneous measuring of E_{liquid} and E_{air}, the light source fluctuation causes a bad influence on the reflective ratio curve.

The RI information is extracted from the cutoff edge of the reflective ratio curve near the critical angle by using existing algorithms, such as the differential[2], threshold[6] and the curve-fitting algorithms[7-12], and the measuring accuracy of the CAR mainly depends on the precision of detecting the critical angle. The threshold algorithm was introduced by Bail, who suggested that the pixel point of a fixed percentage of the maximal reflective light intensity is taken as the critical-angle point[6]. The curve-fitting algorithm[7-12] utilizes the regression analysis between the experimental reflective curve and the Fresnel reflective model to obtain RI information. Most of these algorithms can only determine the critical angle point at an integral pixel. The differential algorithm is an outstanding and straight-forward algorithm for commercial CAR. Mohammadi[13] demonstrated the differential method by defining the presentational critical angle as the maximum change of the slope of the reflective ratio curve. Guo[2] applied the differential algorithm in two-reflection CAR to improve the accuracy of RI measurement and determine the critical angle at integral pixel. However, the pixel size of the CCD restricts the sensitivity of the differential algorithm. Thus, it is important to provide an algorithm to find the critical angle accurately in sub-pixel.

In this paper, we present an improved differential algorithm to measure the critical angle in sub-pixel, and also ameliorate the traditional CAR to eliminate the light source fluctuation and improve signal to noise ratio by

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** E-mail: yejunweiin717@sina.com

adding a reference light path. The improved differential algorithm combines the differential method and the centroid value of the reflective ratio near the critical angle pixel. The main difference of the proposed algorithm from the differential algorithms is that the improved differential method determines the critical angle in sub-pixel by the centroid value of the local reflective ratio. When the cutoff edge has a slight displacement in a pixel, the critical angle pixel does not change, but the centroid value around the pixel has a corresponding change. Using numerical analyses and experiments on saline solutions, we find that the centroid value around the critical angle pixel corresponds to the RI change of the liquid, when the critical angle pixel is fixed. We demonstrate that the improved differential algorithm can simply enhance the accuracy and sensitivity of the CAR, and measure the critical angle in sub-pixel.

According to the Fresnel reflection law, the reflective ratio of the light source is incremental as the incident angle θ increases in the non-TIR region and invariant as θ increases in the TIR region, as shown in Fig.1[14]. The critical angle is decided by the RI of the liquid n_{liquid}, which is between the non-TIR and TIR regions in the CAR. The angular reflective ratio curve $R(\theta, n_{\text{liquid}})$ has a single peak in the differential curve $DR(\theta, n_{\text{liquid}})$, which is expressed as

$$DR(\theta, n_{\text{liquid}}) = \frac{R(\theta, n_{\text{liquid}}) - R(\theta - \Delta\theta, n_{\text{liquid}})}{\Delta\theta} . \quad (1)$$

The peak position p is deemed as the critical angle point of the CAR when the liquid is transparent and low optical absorbing, such as the distilled water and the saline solution. The differential method establishes the function between p and n_{liquid}, and measures n_{liquid} by p of $DR(\theta, n_{\text{liquid}})$.

The CCD sensor discretizes the analog $R(\theta, n_{\text{liquid}})$ into a discontinuous curve at angle direction. The peak position p of $DR(\theta, n_{\text{liquid}})$ is an integral pixel, due to the discrete character of the CCD sensor. The accuracy Δn of the method can be written as

$$\Delta n = \frac{n_p \sin\theta_{\max} - n_p \sin\theta_{\min}}{p_{\max} - p_{\min}} = \frac{n_p \sin\theta_{\max} - n_p \sin\theta_{\min}}{l_{\text{pixel}} C} = \frac{n_{\text{range}}}{l_{\text{pixel}} C} , \quad (2)$$

where n_p is the RI value of the optical prism, $n_p\sin(\theta_{\max})$ is the upper n_{liquid} value, l_{pixel} is the pixel size, n_{range} is the measurement range, and C is a constant. TIR only occurs when the ray is at the maximal incident angle $\theta_{\max}$, and $\theta_{\max}$ corresponds to the boundary pixel $p_{\max}$ of the CCD sensor. $n_p\sin(\theta_{\min})$ is the lower n_{liquid} value, TIR occurs when the ray is at the minimal incident angle $\theta_{\min}$, and $\theta_{\min}$ corresponds to another boundary pixel $p_{\min}$ of the CCD sensor. When n_{range} is invariant, Δn depends on the pixel size of the CCD sensor. l_{pixel} is generally between 3 μm and 6 μm in a commercial CCD or complementary metal oxide semiconductor (CMOS) sensor which is restricted by the semiconductor manufacturing technique.

The differential method establishes the function between the RI value and the peak value pixel of the differential curve. The reflection of the glass-liquid interface is modulated with the RI information of the liquid and composed of a bright region and a dark region as shown in Fig.1. The critical angle pixel of the angular reflective ratio curve is approximately N in Fig.1, even if the RI of the liquid changes t ($|t|< 0.5\Delta n$). $R(\theta, n_{\text{liquid}})$ is different from $R(\theta, n_{\text{liquid}}+t)$ in the reflective ratio distribution around critical angle pixel, while the critical angle pixel of $R(\theta, n_{\text{liquid}})$ is the same as $R(\theta, n_{\text{liquid}}+t)$. The differential method is ineffective under above condition, however t can be extracted from the ratio distribution of $R(\theta_{n-1}, n_{\text{liquid}})$, $R(\theta_n, n_{\text{liquid}})$ and $R(\theta_{n+1}, n_{\text{liquid}})$. And $R(\theta_n, n_{\text{liquid}})$ is the reflective ratio in peak value pixel N, θ_{n-1} corresponds to the left pixel of N, and θ_{n+1} corresponds to the right pixel of N.

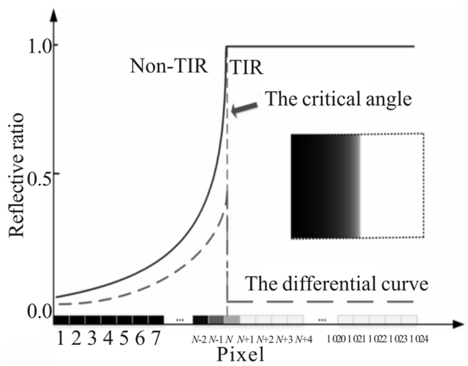


Fig.1 The bright-dark reflective image and reflective ratio curve in pixel direction

The incident beam generates TIR on the glass-air interface, and the reflectance can be regarded as the background of the CAR. The reflective ratio curve of a liquid in the CAR is obtained by the reflective intensity curve and background N, the critical angle pixel of the reflective ratio curve, is obtained by differential method in the following situations.

$$\begin{cases} DR(N, n_{\text{liquid}}) = R(N+1, n_{\text{liquid}}) - R(N, n_{\text{liquid}}) \\ DR(N-1, n_{\text{liquid}}) = R(N, n_{\text{liquid}}) - R(N-1, n_{\text{liquid}}) \\ DR(N-2, n_{\text{liquid}}) = R(N-1, n_{\text{liquid}}) - R(N-2, n_{\text{liquid}}) \\ DR(N-1, n_{\text{liquid}}) > DR(N, n_{\text{liquid}}) \\ DR(N-1, n_{\text{liquid}}) > DR(N-2, n_{\text{liquid}}) \end{cases} , \quad (3)$$

where $R(N, n_{\text{liquid}})$ is the reflective ratio of N pixel, $R(N-1, n_{\text{liquid}})$ is the reflective ratio of $N-1$ pixel, $R(N-2, n_{\text{liquid}})$ is the reflective ratio of $N-2$ pixel, and $R(N+1, n_{\text{liquid}})$ is the reflective ratio of $N+1$ pixel. The real position of the critical angle is the boundary line between the dark region and the bright region in CCD camera chip as the blue vertical line in Fig.2 and has three possibilities in above conditions. In the first case, the boundary line is just between $N-1$ pixel and N pixel, as shown in Fig.2(a). In the second case, the boundary line is between the middle of $N-1$ pixel and left the boundary of N pixel, as shown in Fig.2(b). In the third case, the boundary line is between the middle of N pixel and left the boundary of $N+1$ pixel, as shown in Fig.2(c).

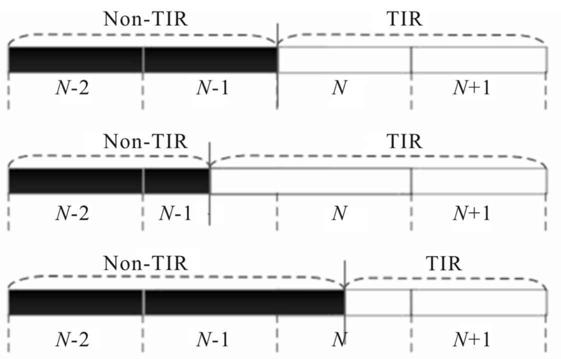


Fig.2 The reflective ratio curve near the critical angle pixel

As is followed, a numerical analysis is carried out to investigate the differential curve around the critical angle, when n_{liquid} has a slight change. We calculate a set of differential curves with n_{liquid} around 1.336 0 by the Fresnel reflective ratio equation, when the measurement range is in [1.330, 1.340]. In Fig.3, seven differential curves are shown with n_{liquid} from $1.336\,0-3\times10^{-6}$ to $1.336\,0+3\times10^{-6}$. Fig.3 shows that as the increasing of the pixel, the differential value is significantly increased when the pixel is less than 426 and sharply decreased when the pixel is more than 426. To highlight the characteristic of the differential curve, only the pixel near the critical angle pixel 426 is displayed in Fig.3. Although the differential curves have the same peak value pixel of 426, the value distribution near the critical angle pixel 426 differs considerably. As t changes from -3×10^{-6} to 3×10^{-6}, the differential value grows large until reaching a maximal value (critical angle line as Fig.2(a)), then becomes small. This demonstrates that the distribution of the differential value round the critical angle pixel determines on the deviation value between the actual critical angle position and the critical angle pixel. The distribution can potentially achieve sub-pixel determination for the critical angle in the CAR.

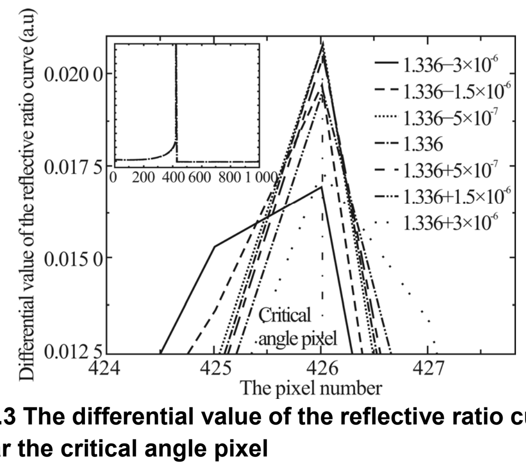


Fig.3 The differential value of the reflective ratio curve near the critical angle pixel

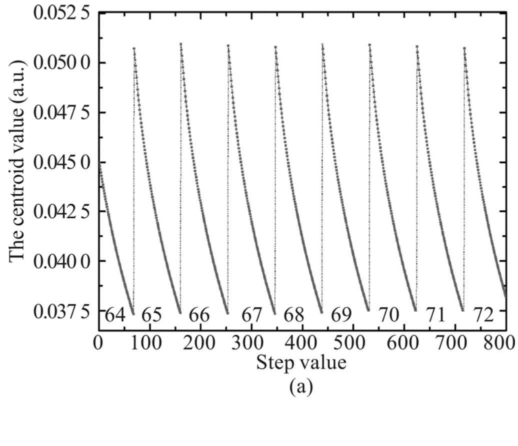
The peak value pixel by the differential method determines on the critical angle pixel at integral pixel, and the reflective ratio around the peak value pixel determines on the relative position between the real critical angle position and the left boundary of the peak value pixel. In order to express the characteristic distribution of the reflective ratio around the critical angle pixel mathematically, we define the centroid value $C(n_{\text{liqiud}})$ of the local reflective ratio near the critical angle pixel as the small change t. The centroid value $C(n_{\text{liqiud}})$ can be written as

$$C(n_{\text{liquid}})=\frac{2R(N+1,n_{\text{liquid}})+R(N,n_{\text{liquid}})-R(N-1,n_{\text{liquid}})-2R(N-2,n_{\text{liquid}})}{R(N+1,n_{\text{liquid}})+R(N,n_{\text{liquid}})+R(N-1,n_{\text{liquid}})+R(N-2,n_{\text{liquid}})},\tag{4}$$

where N is the critical angle pixel, $R(N, n_{\text{liquid}})$ is the reflective ratio of the critical angle pixel, $R(N-1, n_{\text{liquid}})$ is the reflective ratio of $N-1$ pixel, $R(N-2, n_{\text{liquid}})$ is the reflective ratio of N−2 pixel, and $R(N+1, n_{\text{liquid}})$ is the reflective ratio of $N+1$ pixel.

Another numerical experiment is carried out to analyze the relationship between the small change t and $C(n_{\text{liquid}})$ with n_{liquid} from 1.333 0 to 1.333 20 and t step 5×10^{-7}, when the measurement range is [1.330 0, 1.375 0].

Fig.4 shows the numerical calculation results for the centroid value curves. As the step number increases, the centroid value changes regularly. When the peak value pixel is stationary such as 68 in Fig.4(a), the centroid value is monotonically increased as the small RI changes. And the variation law of the centroid value is the same as the small RI change in different peak value pixel. Fig.4(b) shows that each of the centroid value corresponds to a single small RI change. The positive proportion of the small RI change and the centroid value is obtained by applying the polynomial fitting algorithm. And the fitting function between the centroid value and the small RI change enhances the sensitivity and accuracy of the differential method and makes the critical angle edge detection reaching sub-pixel. According to the simulation above, the precision of improved differential algorithm is limited to the number of calibration points and complexity degree of calibration curve. Effectively, the self-noise of CCD affects the precision of algorithm as well which can be reduced by multiple average.



(a)

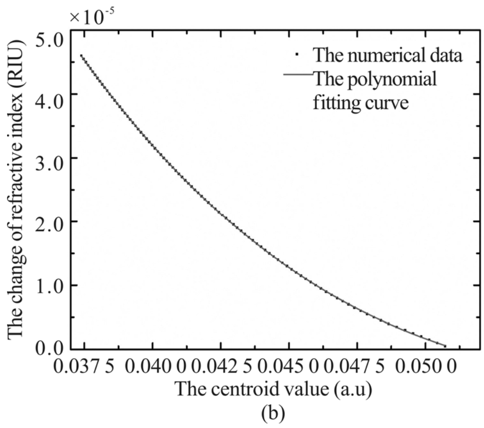


Fig.4 (a) The centroid value versus the refractive index around different critical angle pixels; (b) The change of the refractive index value versus the centroid value around a critical angle pixel

Fig.5 shows a schematic diagram of the improved CAR. The optical system comprises a point light source, an equilateral prism, a lens and a CCD camera. A point light source, centered at 532.0 nm with a spectral width of 10.0 nm, is established by making the beam of a royal green light-emitting diode (LED) transmitting through 532.0 nm narrow band filter and coupling into a single mode fiber with a core diameter of 9.0 μm. The numerical aperture of the single-mode fiber is 0.12, and the divergence angle of the point light source is 13.780°. The optical prism with the inclined angle of 64.0°, made of K9 glass (1.51950 RIU at 532.0 nm), is the key part of the CAR. A divergent fiber-coupled royal green LED source is incident on the interface. A thermometer is used to keep the temperature of our liquid samples near 25.0±0.5 °C. The camera implements a progressive scan monochrome CCD with an image size of 1 024 pixel× 768 pixel and a pixel size of 4.65 μm×4.65 μm.

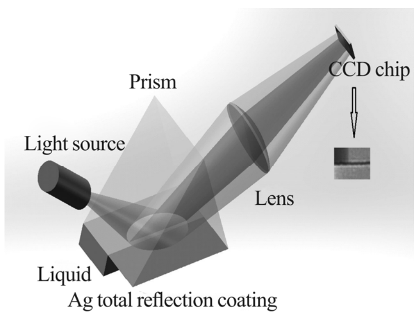


Fig.5 The schematic diagram of the reflective CAR

The interface is divided into the measuring part and the reference part. The measuring part is a glass-liquid interface, the light rays with incident angles smaller than the critical angle cause partial reflection and transmission into the liquid sample, whereas others cause TIR when the refractive index is less than the maximal value based on Snell's law in Fig.6(b). The reflection is the product of the light source intensity distribution and the reflective ratio distribution. The reference part is a glass-Ag coating. TIR of the entire incident beam will occur, and the reflection is the background of the light source intensity distribution in Fig.6(a).

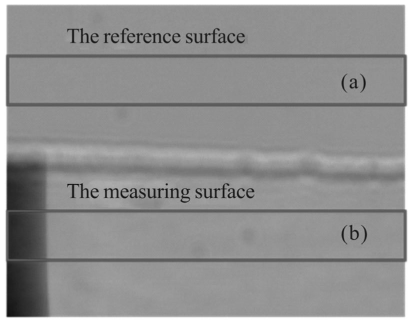


Fig.6 The reflective facula of the reflective CAR

In our experimental setup, seven saline solutions with concentration varying from 20.0 mg/L to 152.0 mg/L are made by the electron balance (accuracy 0.1 mg) and a high precision volumetric flask at 25.0 °C. We take images of different samples using the reflective CAR shown in Fig.5, and derive the reflective ratio curve and the critical angle pixel. As the concentration of the above solutions is less than the accuracy of the traditional differential algorithm, the critical angle pixels of the reflective ratio curves are the same. The traditional differential algorithm cannot distinguish the difference of these saline solutions. We derive the centroid value of the reflective ratio curve around the critical angle pixel using Eq.(3).

Fig.7 shows that the centroid value of the reflective ratio curve around the critical angle pixel varies with the concentration of the saline solution. The calibration curve of the concentration is presented as a function of the centroid value by two-order polynomial fitting with the correlation coefficient of 0.998 5. It also demonstrates that the centroid value around the critical angle pixel by our improved differential algorithm can get measure the critical angle in sub-pixel.

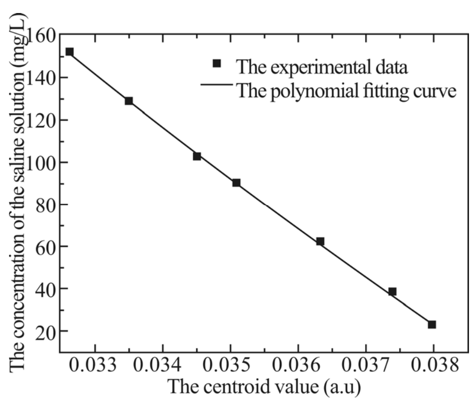


Fig.7 The concentration of the saline solution versus the centroid value

The improved differential algorithm is presented to enhance the accuracy of the refractive index of CAR. This algorithm based on the centroid value of the reflective ratio around the critical angle pixel can solve the problem of determining the critical angle in integral pixel. This algorithm is a potential supplementary of the traditional differential algorithm, and has a significant advantage for improving the measuring accuracy of the commercial CAR. This algorithm can be facilely applied without any additional cost in the digital CAR to measure the refractive index of all liquids in the natural world.

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