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Measurement of thickness of a thin film by means of laser interference at many incident angles

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Abstract

An optical method for directly measuring the thickness of a thin transparent film has been proposed by means of multi-wave laser interference at many incident angles, and confirmed experimentally by means of equipment made on an experimental basis. Two methods are available: one can be used when an index of refraction of the film, a wavelength λ, and two successive angles of incidence at which the sinusoidal light intensity has minimum values, are known (Method I), and another can be used without an index of film refraction when three successive angles of incidence and a wavelength are known (Method II). The smallest measurable thickness is 1.43λ for Method I, and 2.5λ for Method II. The largest measurable thickness is about 100λ for both methods. The measurement error by means of numerical calculation is $\Delta h/h \fallingdotseq -1.01 \times 10^{-2}$, and that obtained experimentally with an angular resolution of incident light of 0.3° is $\Delta h/h \fallingdotseq 7 \times 10^{-2}$ for Method I. The refractive index can also be measured by means of Method II.

Keywords: Laser interference; Film thinckness; Refractive index

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1. Introduction

Transparent, protective ceramic coatings have been applied to various materials such as electronic circuits and a building's outer wall tiles. The application of such coatings is usually performed by spraying, making it difficult to apply the coat completely and uniformly, even though the thickness of the coat determines the durability of, for example, an outer wall tile.

The many methods that have been developed for measuring the thickness of a thin film or coating have been classified into two main groups: the contact method and the non-contact method. The non-contact method is more effective than the contact method, in particular in regard to all sample conditions and the real-time checking of the samples. A representative example of the non-contact method is the optical method [1], which is based mainly on light interference. The interference method can usually measure only a relative thickness by means of interference fringe. However, a method for an absolute measurement has recently been developed by means of multi-wavelength interference [2–4] and an usual interference [5]. The former methods [2–4] are based on the multi-wave interference of white light, and are time consuming and require the use of an expensive spectrometer. The last method [5] uses a super luminescent diode, whose coherent length is about 10 μm, and has been successfully applied for measuring both refractive index and thickness. However, this method can measure only a thickness between 20 μm and a few mm, because its resolution (smallest measurable thickness) was about coherent length.

In this paper, we propose a method for an absolute measurement by means of the multi-wave interference of a single wavelength. This method requires only a device for continuously changing the angle of incident light in place of a spectrometer, as described in the literature [2,4], and has a measurable range of between sub-μm and 100 μm.

2. Principle of the method

Fig. 1 illustrates the principle of measurement using the multi-wave interference of a single wavelength on a single layer, i.e., a single film in air. A portion of the incident light, L_{ir}, is reflected on the upper surface of the film, and the remainder, L_{it}, penetrates the film, while a part of L_{it} passes through the film and the remainder is reflected on to the lower surface of the film L_{itr}, which results in other transmitted light, L_{itrt}, and reflected light, L_{itrr}. Similar processes occur on both surfaces of the film, and then all the light from the lens, $L_{\mathrm{ir}}, L_{\mathrm{itrt}}, L_{\mathrm{itrrrt}}, L_{\mathrm{itrrrrrt}}, \ldots$ is focused onto a receiver. The intensity ratio of this focused light to that incident light, i.e., reflexibility R, can therefore be expressed for a Fresnel's reflection of a s-polarized light wave as follows [6]:

$$R = 2r^2\{1 - \cos(2\delta)\}/[1 + r^2\{r^2 - 2\cos(2\delta)\}], \tag{1}$$

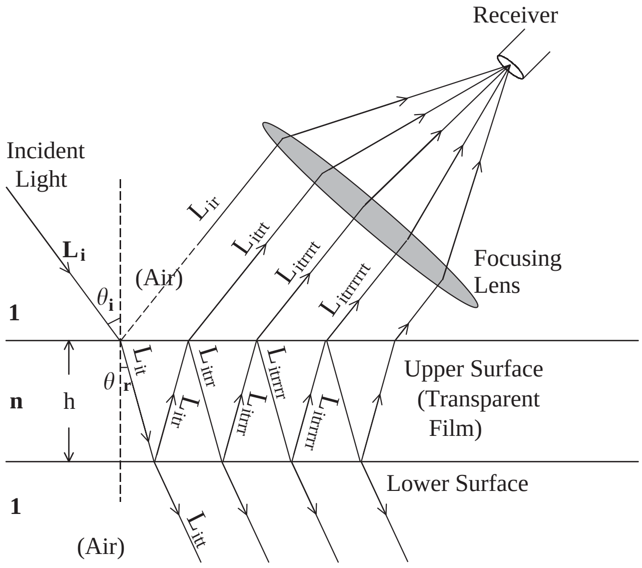

Fig. 1. Principle of multi-wave interference on a film.

where

$$\delta = (2\pi/\lambda)nh\cos\theta_r, \tag{2a}$$

$$r = \{\sin(\theta_r - \theta_i)\}/\{\sin(\theta_r + \theta_i)\}, \tag{2b}$$

$$n = \sin\theta_i/\sin\theta_r, \tag{2c}$$

where δ expresses the phase difference across the film, λ the wavelength of the light, n the index of refraction of the film, r the Fresnel's reflection coefficient on the surface of the film for s-polarized light, h the thickness of the film, θ_i the angle of incidence on the film and θ_r the angle of refraction. Most types of film that are practically used absorb some degree of light even if the film is transparent. In this case, the effect of absorption can be expressed by replacing a real number n with a complex number, i.e., $n = n_0(1 - ja)$, where $j = \sqrt{-1}$.

As is expected from Eqs. (1)–(2c), the reflexibility R, in other words, the light intensity received on the receiver, changes like a sin-wave with λ and θ_r, i.e., θ_i. This leads to two methods for measuring the thickness of the film. The first method is to measure the light intensity with a change of λ, which is a basis of the method described in [4]. In this case, however, a costly spectrometer is required, and the method is time consuming. The second method is to measure the light intensity with a change of θ_i, which is the basis of our method. Instead of a spectrometer, this method requires only a device for continuously changing the angle of incidence. Thus, our method is simple and inexpensive to construct.

To examine the intensity distribution with regard to θ_i, i.e., reflexibility R in Eq. (1), we calculated $\mathrm{d}R/\mathrm{d}\theta_\mathrm{i}$ as follows:

$$\mathrm{d}R/\mathrm{d}\theta_i = F(n, \cos\theta_\mathrm{r}, r, \sin\delta, \sin\theta_\mathrm{i})\sin\delta\{2(r^2+1)n\sin\delta\cos\theta_\mathrm{r} + \delta(r^2-1)\cos\theta_\mathrm{i}\cos\delta\} \quad (3)$$

and we then obtained the following relation as a solution of $\mathrm{d}R/\mathrm{d}\theta_\mathrm{i} = 0$, because the function F should be $F \neq 0$, for $r \neq 0$, $n > 1.0$, and $0 < \theta_\mathrm{i}$, $\theta_\mathrm{r} < \pi/2$, conditions which are usually valid for commonly used films.

$$\sin\delta = 0, \quad (4a)$$

or

$$2n(r^2+1)\sin\delta\cos\theta_\mathrm{r} + \delta(r^2-1)\cos\theta_\mathrm{i}\cos\delta = 0. \quad (4b)$$

Eq. (4a) is simpler to use than Eq. (4b), although both of these equations can be used to estimate the thickness h. We can therefore obtain the following relation from Eq. (4a):

$$\delta = (2\pi/\lambda)nh\cos\theta_\mathrm{r} = k\pi, \quad (k = 0, 1, 2, \ldots). \quad (5)$$

Combining Eqs. (5) and (2c), the following relation can be obtained:

$$\sin\theta_\mathrm{i} = [n^2 - \{k\lambda/(2h)\}^2]^{1/2}. \quad (6)$$

In this equation, θ_i can be determined experimentally. We can then calculate thickness h by means of Eq. (6), if other parameters, n, k and, λ are known. However, we cannot calculate the thickness because a positive integer k is also unknown. That is, two or more values for the angle of incidence, θ_i, should be measured for the calculation of h, because two unknown parameters, h and k, are included in Eq. (6). Some angles of incidence usually exist, which we denote as $\Theta_{\mathrm{i},k}$ $(k = 0, 1, 2, \ldots)$, because k has certain integer values. The range of k can be calculated as follows, since it should be $0 < n^2 - \{k\lambda/(2h)\}^2 < 1$ in Eq. (6),

$$k_\mathrm{min} = 2(n^2-1)^{1/2}h/\lambda < k < 2nh/\lambda = k_\mathrm{max}. \quad (7)$$

Thus, if two successive angles of incidence, $\Theta_{\mathrm{i},k}$ and $\Theta_{\mathrm{i},k+1}$, can be obtained experimentally, the thickness h can be calculated as follows:

$$h = (\lambda/2)\{(n^2 - \sin^2\Theta_{\mathrm{i},k+1})^{1/2} + (n^2 - \sin^2\Theta_{\mathrm{i},k})^{1/2}\}/(\sin^2\Theta_{\mathrm{i},k} - \sin^2\Theta_{\mathrm{i},k+1}). \quad (8)$$

Further, if three successive angles of incidence, $\Theta_{\mathrm{i},k-1}$, $\Theta_{\mathrm{i},k}$ and $\Theta_{\mathrm{i},k+1}$ can be obtained experimentally, the thickness h can be calculated as follows:

$$h = (\lambda/\sqrt{2})/(2\sin^2\Theta_{\mathrm{i},k} - \sin^2\Theta_{\mathrm{i},k-1} - \sin^2\Theta_{\mathrm{i},k+1})^{1/2}. \quad (9)$$

Thus, the thickness can be calculated when only the value of λ is known, and then Eq. (9) will be very useful for measuring the thickness of the film.

3. Experimental results

A preliminary experiment was carried out by means of a numerical calculation. Fig. 2 shows the results for Eqs. (1), (4a), and (4b) for $h = 5\,\mu m$, $\lambda = 0.65\,\mu m$ and $n = 1.4(1 - j3 \times 10^{-3})$, respectively. As is shown, the light intensity received on a silicon photodiode R varies with θ in the manner of sin-wave. Further, Eq. (4a) shows the angle of incidence θ where the light intensity has the minimum value, i.e., Θ_i, and Eq. (4b) corresponds to the maximum value.

A device for continuously changing the angle of incidence is required for this method to be used practically, as described above. Photo 1 shows an experimental setup, and Fig. 3 an optical arrangement. The angle of incidence on the film, θ_i, was

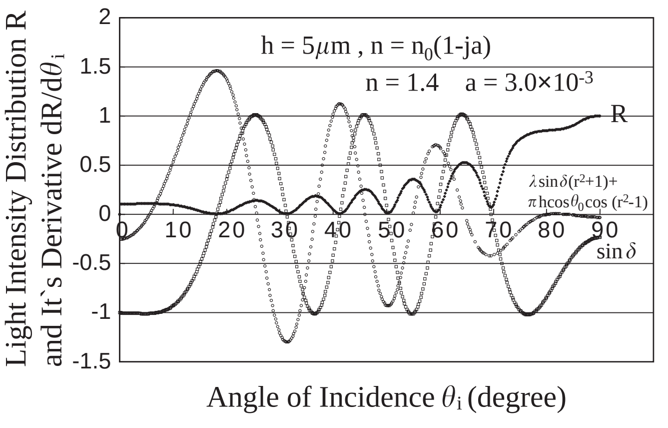

Fig. 2. Numerical calculations for light intensity R given by Eq. (1), and its differentiation, $dR/d\theta$, given by (4.1) and (4.2), where, $h = 5\,\mu m$, $n = 1.4$ and $\lambda = 0.65\,\mu m$.

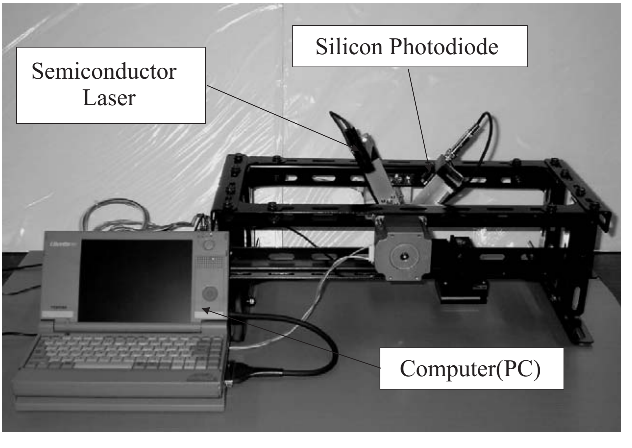

Photo 1. Experimental setup.

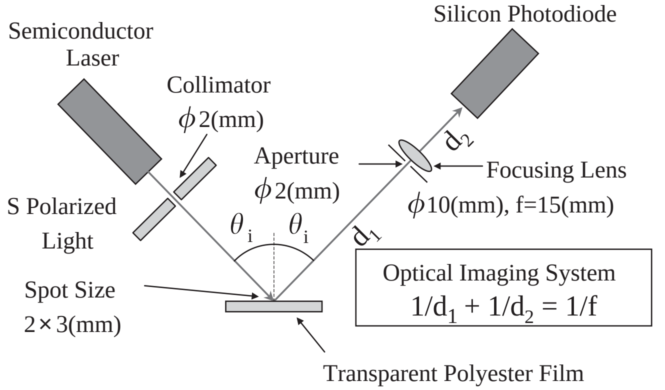

Fig. 3. Optical arrangement of the experimental setup.

varied from 7° to 83° by means of a stepping motor with an angular resolution of 0.3°. The illuminated spot area is focused onto the silicon photodiode by means of the optical arrangement with $f = 15$ mm, $d_1 = 55$ mm and $d_2 = 21$ mm. A semiconductor laser having a wavelength of 0.65 μm was used as a light source, and a silicon photodiode was used as a receiver. The laser is usually stable enough over a long period and thus does not affect the fluctuation in h since a measurement can be done within a minute. A laser light was collimated in both the source and receiver sides to obtain a collimated light. Although the diameter of a collimated beam is about 2 mm as shown in Fig. 3, the actual spot size on the film surface varies with the incident angle. This variation, however, changes only the measured area, and does not affect the accuracy of the measurement since the incident angle, where the reflexibility has the minimum value, is hardly affected by this variation. This is one of the most advantageous features of this method.

Fig. 4 shows an example of the measured light intensity for a transparent polyester film with a refraction index of 1.4, and a mean thickness of about 20 μm which was measured as the mean value of a 100-sheet stack by means of micro-meter. Table 1 shows the thickness calculated by means of each of two successive values of $\Theta_{i,k}$ and $\Theta_{i,k+1}$ and Eq. (8). Thus, the thickness measured by this method agrees quite closely with the mean value obtained by means of a micrometer. A slight fluctuation in the value of h in Table 1 is due primarily to: (1) a quantization error of incident angle Θ_i whose angle of resolution is 0.3°, which is discussed in Section 4.2 and (2) a scattered light on the film surface, whose intensity depends on the optical arrangement as well as the surface roughness of the film. However, the intensity of light scattered on the film is usually small enough to compare with that of reflected light since the film surface is sufficiently specular, and thus the fluctuation in the value of h due to the scatted light is negligibly small. The thickness can also be calculated by means of three successive values of $\Theta_{i,k-1}$, $\Theta_{i,k}$, and $\Theta_{i,k+1}$ and Eq. (9). This will be discussed below.

In Fig. 4, no minimum of R can reach 0. This results from the light absorption in the film as mentioned above. Fig. 5 shows some examples of this effect calculated numerically for $h = 5$, 10 μm and $n = 1.4(1 - ja)$, where $a = 1.0 \times 10^{-4}$ and $3.0 \times$

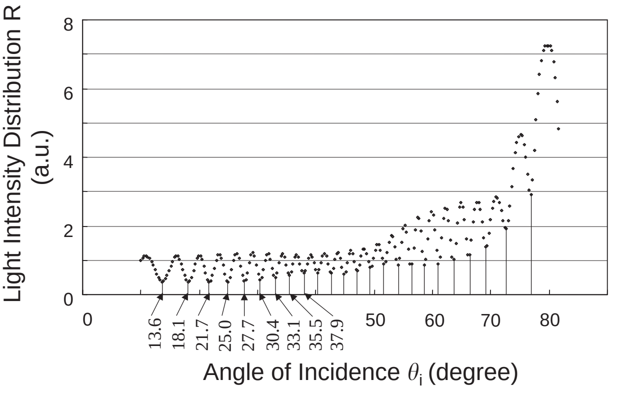

Fig. 4. Light intensity distribution, R, with regard to the angle of incidence, θ_i, for a transparent polyester film with $n = 1.4$ and $h \fallingdotseq 20\,\mu m$.

Table 1
Thickness calculated by means of measured Θ_i and Eq. (8)

Θ_k	h (μm)
13.6	21.64
18.1	21.96
21.7	20.83
25.0	23.03
27.7	21.34
30.4	20.00
33.1	21.36
35.5	20.51
37.9	19.82
40.3	19.29
42.7	21.61
44.8	18.65
47.2	21.15
49.3	18.47
51.7	18.56
54.1	18.79
56.5	21.88
58.6	19.66
61.0	Ave.20.48

10^{-3}. The effect of light absorption increases as the thickness of the film increases because the optical path length in the film increases.

4. Discussion

4.1. Resolution of this method

As was shown in Eqs. (8) and (9), the thickness can be determined experimentally when two successive values of $\Theta_{i,\,k-1}$ and $\Theta_{i,\,k}$ are obtained for Eq. (8), and when

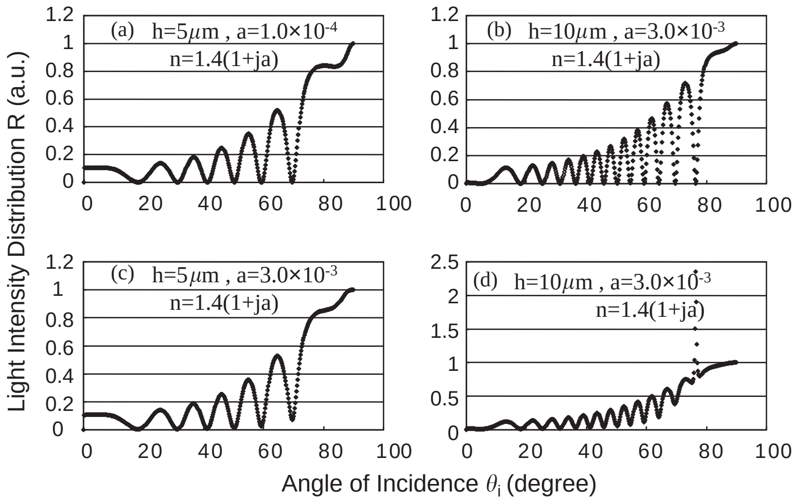

Fig. 5. Calculated light intensity for $h = 5$, 10 μm, and $n = 1.4(1 - ja)$. (a) and (b) for $a = 1.0 \times 10^{-4}$, and (c) and (d) for $a = 3.0 \times 10^{-3}$.

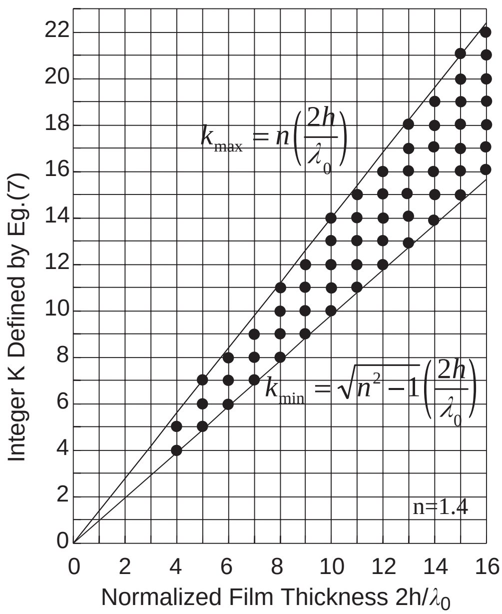

Fig. 6. Range of k in Eq. (7) for $n = 1.4$.

three successive values of $\Theta_{i,k-1}$, $\Theta_{i,k}$ and $\Theta_{i,k+1}$ are obtained for Eq. (9). This restricts the smallest measurable thickness of the film, i.e., the resolution of this method. The range of k, in other words, the number of $\Theta_{i,k}$, can be calculated by means of Eq. (7), and is shown in Fig. 6 for $n = 1.4$ as an example. Thus, the range of

k increases with thickness; in other words, $2h/\lambda$ increases for a given n. As is shown in this figure, two successive numbers of k are 3 and 4, and the smallest measurable thickness $h_{\min}$ for Eq. (8) can, therefore, be obtained by

$$h_{\min} = 4/1.4(\lambda/2) = 2.86(\lambda/2). \tag{10a}$$

In contrast, three successive numbers of k are 5, 6 and 7, and the smallest measurable thickness $h'_{\min}$ for Eq. (9) can, therefore, be obtained by

$$h'_{\min} = 7/1.4(\lambda/2) = 5.0(\lambda/2). \tag{10b}$$

Thus, the smallest measurable thickness, $h'_{\min}$, for Eq. (9) becomes larger than that, $h_{\min}$, for Eq. (8).

The largest measurable thickness $h_{\max}$ can be determined by an angular resolution for θ, i.e., an angular increment in each data acquisition, $\Delta\theta$, since the thickness can be obtained by means of measured $\Theta_{i,k}$. If $\Delta\theta$ is 0.3° as in this study, we can find at least two values of $\Theta_{i,k}$ within about 1.5° experimentally. In this case, the largest measurable thickness can be calculated as $h_{\max} \fallingdotseq 65\,\mu\text{m}$ by substituting $\Theta_{i,k} = 16.5°$, $\Theta_{i,k+1} = 15°$, $n = 1.4$, and $\lambda = 0.65\,\mu\text{m}$ in Eq. (8).

4.2. Measurement error

A measurement error for the film thickness, Δh, depends on the measurement error of $\Theta_{i,k}$, $\Delta\Theta_{i,k}$, and an increment Δh due to $\Delta\Theta_{i,k}$ can be calculated theoretically by means of Eq. (8) as follows:

$$\begin{aligned}\Delta h/h = &\\ &-\Delta\Theta_{i,k}[\{\sin\Theta_{i,k}\cos\Theta_{i,k}/(n^2-\sin^2\Theta_{i,k})^{1/2}\\ &+\sin\Theta_{i,k+1}\cos\Theta_{i,k+1}/(n^2-\sin^2\Theta_{i,k+1})^{1/2}\}/\{(n^2-\sin^2\Theta_{i,k})^{1/2}\\ &+(n^2-\sin^2\Theta_{i,k+1})^{1/2}\}\\ &+2\{\sin\Theta_{i,k}\cos\Theta_{i,k}-\sin\Theta_{i,k+1}\cos\Theta_{i,k+1}\}/(\sin^2\Theta_{i,k}-\sin^2\Theta_{i,k+1})].\end{aligned} \tag{11}$$

The maximum values of $\Delta\Theta_{i,k}$ and $\Delta\Theta_{i,k+1}$ can be considered as $\Delta\theta$ ($=0.3° = 5.23\times10^{-3}$ in this experiment), i.e., $\Delta\Theta_{i,k} = \Delta\Theta_{i,k+1} = \Delta\theta$. For an example, we can then obtain $\Delta h/h = -1.\ 923\times\Delta\Theta_{i,k} = -1.01\times10^{-2}$ for $\Theta_{i,k} = 3.13°$, and $\Theta_{i,k+1} = 18.1°$ as shown in Fig. 2.

In the experiment, the film thickness was measured as shown in Table 1. Thus, the measurement error was found to be about $\Delta h/h \fallingdotseq 0.07$ with a few marked exceptions. This error will result from the surface reflection condition. That is, an irradiated area increases as the angle of incidence θ increases, and a light from a different area will be received on the silicon photodiode.

When we use Eq. (9) to determine the thickness, it was calculated as $h = 14.29\,\mu\text{m}$ by means of $\Theta_{i,k+1} = 13.6°$, $\Theta_{i,k} = 18.1°$ and $\Theta_{i,k-1} = 21.7°$ which are the results shown in Fig. 4. The error was rather large. This is due to the fact that a square of the denominator on the right side in Eq. (9), i.e., $(2\sin^2\Theta_{i,k} - \sin^2\Theta_{i,k-1} - \sin^2\Theta_{i,k+1})$, has such a small value as 2.641×10^{-4} for $h = 20\,\mu\text{m}$

and $\lambda = 0.65\,\mu m$. This means that the values of $\Theta_{i,\,k+1}$, $\Theta_{i,\,k}$, $\Theta_{i,\,k-1}$ should be measured rather accurately, which is difficult to achieve by means of the experiment.

In any case, the error becomes small at small values of $\Theta_{i,\,k}$, where the angular interval between the two or three successive values are larger than those at larger values of $\Theta_{i,\,k}$, and then those values can be determined in more detail.

Thus, Eq. (9) can be used even when a film's refractive index, n, is unknown. This, however, requires three successive highly accurate $\Theta_{i,\,k}$ values. In contrast, Eq. (8) can be used only when n is known, but it requires only two successive values of $\Theta_{i,\,k}$, and the film thickness can be measured with a reasonable accuracy.

4.3. Application for measuring a refractive index

The method described in this paper can also be applied for the determination of refractive index as well as thickness, as follows. As expressed in Eq. (9), the thickness can be obtained experimentally even when the refractive index is unknown. We can, therefore, calculate a refractive index by introducing this thickness, H into Eq. (8). That is, the refractive index n can be calculated as follows:

$$n = (1/2)[(2H/\lambda)^2(\sin^2\Theta_{i,\,k} - \sin^2\Theta_{i,\,k+1})^2 + 2(\sin^2\Theta_{i,\,k} + \sin^2\Theta_{i,\,k+1}) + \{\lambda/(2H)^2\}]^{1/2}. \tag{12}$$

This will be useful since the refractive index of a film is usually unknown.

4.4. Proposal for a real-time measurement

About 2 min were required to obtain the results shown in Fig. 4 and Table 1 by means of the experimental setup seen in Photo 1 since the sampling frequency of data acquisition was limited to about 2 Hz in order to suppress vibration at the time of data acquisition. Thus, this experimental setup cannot be used in a production plants such as those devoted to making polyester and polyethylene films, which are usually produced at a speed of a few m/s. For such a purpose, a system employing many

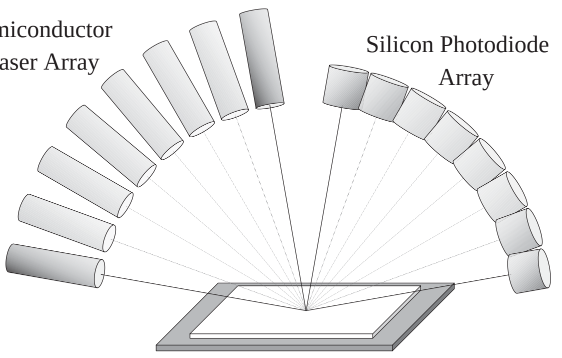

Fig. 7. Proposed system for a real-time measurement.

semiconductor lasers and corresponding receivers, each of which has a slightly different angle of incidence and a corresponding reflection angle, is proposed in Fig. 7. In this system, each datum can be obtained on each receiver by means of an electronic switch, and high-speed data acquisition can be obtained because the entire system is fixed and the data acquisition can be performed without vibration. In this case, a correction is required for each level of light intensity on each silicon photodiode. This can be done easily by adjusting the output of each laser.

5. Conclusion

The optical method for directly measuring the thickness of a thin transparent film has been proposed by means of multi-wave laser interference for many incident angles, and was confirmed experimentally by means of equipment made on an experimental basis. Two methods are available: one can be used when an index of refraction of the film, a wavelength λ, and two successive angles of incidence where the sinusoidal light intensity has minimum values are known (Method I), while the other can be used without an index of film refraction when three successive angles of incidence and a wavelength are known (Method II). The following characteristics mainly of Method I can be determined based on light intensity distribution sampled with an angular resolution of incident angles of 0.3°. Refractive index as well as thickness can be measured by means of Method II. The smallest measurable thickness is 1.43λ for Method I and 2.5λ for Method II, and the largest measurable thickness is about 100λ for both methods. The measurement error by means of numerical calculation is $\Delta h/h \risingdotseq -1.01 \times 10^{-2}$, and that obtained experimentally is $\Delta h/h \risingdotseq 7 \times 10^{-2}$ for Method I.

The system in this experiment is available only in off-line measurement; about 2 min are required for one measurement. We are now developing a system available for real-time measurement.

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