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PHYSICAL OPTICS

A Method for Measuring the Phase of the Reflection Coefficient in the Visible Range of the Spectrum

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Abstract—A method for measuring the phase of the reflection coefficient in the optical wavelength range is proposed. The method is simple in experimental implementation and is based on measuring the energy-reflection coefficients of a sample in two media with different refractive indices. Analytical and numerical estimates show that the measurement accuracy of the phase is on the order of 1°. The possibilities of using the results of the phase measurement in practice for a more complete characterization of materials and structures under investigation are considered.

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Methods of reflection and transmission spectrophotometry are widely used to characterize the optical properties of materials and layered structures. In both cases, the intensity of light reflected from or transmitted through a sample is measured. In this case, changes in the phase of the reflected (transmitted) wave remain unknown and defy experimental determination by photometric instrumentation. Meanwhile, the phase shift of a wave upon its reflection provides additional information on the object under study and can characterize its properties, as was shown in [1], in which the results of microwave sounding of cryospheric formations were presented. Methods for measuring the phase are most developed in the microwave range [2]. In the visible range, the phase characteristics of reflection are determined by interference methods [3], which entail performing rather complicated measurements.

In this work, we consider a simple method for experimental determination of the phase of the reflection coefficient, which is based on measuring the energy-reflection and transmission coefficients of a sample under study that is placed in optically transparent media with different refractive indices.

In order to describe the interaction of light with a sample, we will use the formalism of the scattering matrix [4], which relates the electric fields of the waves on either side of the sample restricted by media "*a*" and "*b*." The scheme of the experiment is shown in Fig. 1. Here, E_a^+ and E_a^- are the amplitudes of the electric vector of the waves that travel in the positive and negative directions of the OX axis to the left of the sample (medium *a*); E_b^+ and E_b^- are the amplitudes of the similar waves to the right of the sample (medium *b*). These four amplitudes are related between each other by scattering matrix $\hat{S}$ as follows:

$$\begin{pmatrix} E_a^+ \\ E_a^- \end{pmatrix} = \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix} \begin{pmatrix} E_b^+ \\ E_b^- \end{pmatrix}. \tag{1}$$

By considering alternately the cases of the light incidence in the positive and negative directions, we arrive at the relations that explain the meaning of the elements of the scattering matrix:

$$S_{11} = \frac{1}{T}, \quad S_{12} = \frac{-\tilde{R}}{T}, \quad S_{21} = \frac{R}{T}, \quad S_{22} = \frac{T\tilde{T} - R\tilde{R}}{T}. \tag{2}$$

Here, $T = \frac{E_b^+}{E_a^+}$ and $\tilde{T} = \frac{E_a^-}{E_b^-}$ are the complex amplitude-transmission coefficients for the light that is incident on the sample from the left and from the right, respectively; $R = \frac{E_a^-}{E_a^+}$ and $\tilde{R} = \frac{E_b^+}{E_b^-}$ are the complex amplitude-reflection coefficients for the light that is incident on the sample from the left and from the right, respectively.

In order to determine complex reflection coefficient R, we consider two experimental situations that correspond to the incidence of a wave onto the structure under investigation from the left (from region *a*) and restrict ourselves to the case of normal incidence.

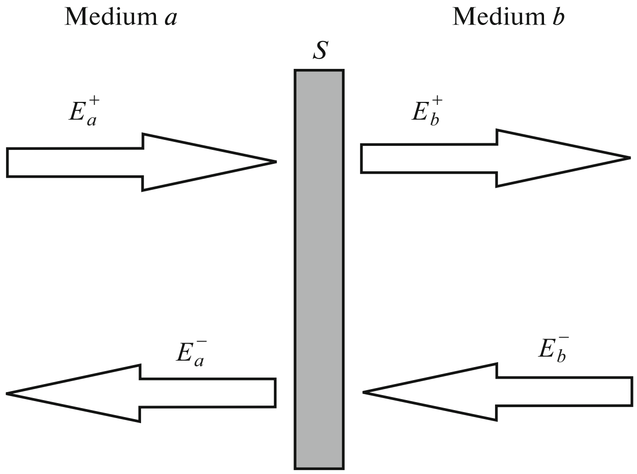

Fig. 1. Schematic image of the interaction of light with structure *S*. The superscripts "+" and "−" indicate the directions of propagation of the light wave, and the subscripts "*a*" and "*b*" indicate the medium in which it propagates.

This is of no significance for the description of the method under consideration, but allows us to simplify the mathematical treatment.

Situation 1. The sample is in air and the media to the left and to the right of the sample are the same, with their refractive indices being unity. In this case, the elements of scattering matrix $\hat{S}$ are given by relations (2), and the quantities to be measured are the energy reflection and transmission coefficients, $\mathcal{R} = |R|^2$ and $\mathcal{T} = |T|^2$, respectively.

Situation 2. The refractive index of medium *a* (e.g., liquid) is *n*, while medium *b* is air. In this case, scattering matrix $\hat{S}_1$ can be found if matrix $\hat{S}$ is sequentially multiplied from the left by the matrix of a virtual air layer of a zero thickness and by matrix $\hat{I}_{n1}$ of the interface between the media with refractive indices *n* and 1. The matrix of the virtual layer is unity because of the zero thickness of the layer and does not affect the final result. However, its inclusion into the system makes it possible to relate matrices $\hat{S}$ and $\hat{S}_1$ with each other, since initial matrix $\hat{S}$ is determined with respect to the ambient medium with the refractive index equal to unity. As a result, we obtain

$$\hat{S}_1 = \hat{I}_{n1}\hat{E}\hat{S} = \frac{1}{nt}\begin{pmatrix} 1 & -r \\ -r & 1 \end{pmatrix}\hat{S}. \quad (3)$$

Here, *r* and *t* are the Fresnel reflection and transmission coefficients for the interface between air and the medium with refractive index *n*. At a normal incidence, these coefficients are expressed by the formulas

$$r = \frac{n-1}{n+1}, \quad (4)$$

$$t = \frac{2}{n+1}. \quad (5)$$

By calculating the elements of the first column of matrix $\hat{S}_1$, we obtain the complex amplitude reflection and transmission coefficients of the system,

$$R_1 = \frac{R-r}{1-rR}, \quad (6)$$

$$T_1 = \frac{ntT}{1-rR}. \quad (7)$$

The energy reflection and transmission coefficients that are measured in this experiment are expressed by the formulas

$$\mathcal{R}_1 = \left|\frac{R-r}{1-rR}\right|^2, \quad (8)$$

$$\mathcal{T}_1 = \frac{1}{n}|T_1|^2 = nt^2\left|\frac{T}{1-rR}\right|^2. \quad (9)$$

Experimental situations are also possible in which the refractive index of medium *b* is *n*, while that of medium *a* is either *n* or 1. In these cases, the scattering matrices are found as a result of multiplying initial matrix $\hat{S}$ by the matrices of the interfaces: $\hat{S}_2 = \hat{I}_{n1}\hat{S}\hat{I}_{1n}$ and $\hat{S}_3 = \hat{S}\hat{I}_{1n}$. In either case, the expressions for the amplitude reflection and transmission coefficients, $R_{2,3}$ and $T_{2,3}$, which are similar to (6) and (7), apart from variables *R* and *T*, will also contain variables $\tilde{R}$ and $\tilde{T}$. This increases the number of unknowns in equations; therefore, we will not consider such experimental situations, although they can give additional information on the structure.

In order to calculate the phase of the complex reflection coefficient, we decompose *R* into real and imaginary parts, $R = R' + iR''$, and substitute this decomposition into Eq. (8). After simple algebraic transformations and taking into account that $(R')^2 + (R'')^2 = \mathcal{R}$, we obtain the following expressions for the real and imaginary parts of the complex reflection coefficient in terms of experimentally measured quantities $\mathcal{R}$ and $\mathcal{R}_1$, and Fresnel coefficient *r*:

$$R' = \frac{\mathcal{R}_1(1+r^2\mathcal{R}) - \mathcal{R} - r^2}{2r(\mathcal{R}_1 - 1)}, \quad (10)$$

$$R'' = \pm\sqrt{\mathcal{R} - (R')^2}. \quad (11)$$

Phase φ of the complex reflection coefficient is found from the relation

$$\cos\varphi = R'/\sqrt{\mathcal{R}}. \quad (12)$$

Unfortunately, the phase in this case can be determined only with an accuracy of up to an interval that is multiple of π because of the uncertainty in expression (11).

In practice, the ambiguity in the phase can be eliminated if its interval is known at least at one point of the spectrum. For example, if a strong absorption in layers of a structure in some range of the spectrum is observed, while the reflection takes place only from the outer layer, the complex reflection coefficient will be close to the Fresnel coefficient, for which the phase interval is known. If there are no sharp phase jumps in the spectral range under consideration, then, by going along the spectrum, it is possible to track the change in the phase interval and, thus, to eliminate the ambiguity in the phase.

If the structure is semitransparent and the transmission differs from zero, then, instead of relation (8), we can use Eq. (9) and obtain the formula for R' that is alternative to Eq. (10),

$$R' = \frac{(1 + r^2\mathcal{R})\mathcal{T}_1 - nt^2\mathcal{T}}{2r\mathcal{T}_1}. \quad (13)$$

Mathematically, the two formulas (10) and (13) yield equivalent expressions for R'; however, for structures with a small transmission, expression (13) should be used with care because of a considerable influence of errors. Clearly, measured quantities $\mathcal{R}$, $\mathcal{R}_1$, $\mathcal{T}$, and $\mathcal{T}_1$ are not independent of each other, and there is an interplay between them. By equating the right-hand sides of (10) and (13), we obtain an invariant relationship for the reflection and transmission coefficients,

$$\frac{1 - \mathcal{R}}{\mathcal{T}} = \frac{1 - \mathcal{R}_1}{\mathcal{T}_1}. \quad (14)$$

The physical meaning of the obtained expression is that the ratio between the energy that penetrated into the structure $(1 - \mathcal{R})$ and the energy that emerged from it $(\mathcal{T})$ is a constant quantity, which does not depend on the refractive index of a medium from which the light is incident. This invariant can be used as a criterion for estimating the absolute measurement accuracy.

Let us consider the accuracy of proposed phase measurements. An estimate can be made from an analysis of the obtained relations. By differentiating (12), we obtain

$$-\sin\varphi\delta\varphi = \delta R'/\sqrt{\mathcal{R}} - R'\delta\mathcal{R}/\mathcal{R}. \quad (15)$$

The second term in the right-hand side of (15) does not exceed the value of the relative measurement error of the reflection coefficient, which is 10^{-2}–10^{-3} for modern equipment. The expression for variation δR in the first term of (15) can be found from (10). As a result, we obtain

$$\delta R'/\sqrt{\mathcal{R}} = \frac{\delta\mathcal{R}(\mathcal{R}_1 - 1) + \delta\mathcal{R}_1(\mathcal{R} - 1)(1 - r^2)}{2r\sqrt{\mathcal{R}}(\mathcal{R}_1 - 1)^2}. \quad (16)$$

Let us consider limiting cases. At $\mathcal{R} \to 0$, we find from Eq. (8) that $\mathcal{R}_1 \to r^2$. As a result, expression (16) increases infinitely as $1/\sqrt{\mathcal{R}}$. The second limiting case takes place when $\mathcal{R} \to 1$. Here, the value of $\mathcal{R}_1$ also tends to 1, while expression (16) as a whole increases infinitely again because of multiplier $\mathcal{R}_1 - 1$ in the denominator. From the viewpoint of accuracy, an optimal value of $\mathcal{R}$ should lie in the interval [0, 1]. By substituting $\mathcal{R} \sim \mathcal{R}_1 \sim 0.5$ into (16), we find that the typical measurement error for values of $\varphi = 90°$ and $n = 1.5$ is $\delta\varphi \sim 1°$. Figure 2 shows calculated dependences $\delta\varphi(\varphi)$ for some values of $\mathcal{R}$. The error was calculated as a difference between the specified value of φ and it value determined by formula (12) taking into account (10) or (13). In this case, the deviations of different signs, $\delta\mathcal{R} = 0.005\mathcal{R}$, $\delta\mathcal{R}_1 = -0.005\mathcal{R}_1$, $\delta\mathcal{T} = 0.005\mathcal{T}$, and $\mathcal{T}_1 = -0.005\mathcal{T}_1$, were introduced into the calculated values of coefficients $\mathcal{R}$, $\mathcal{R}_1$, $\mathcal{T}$, and $\mathcal{T}_1$ in (10) and (13). The refractive index of the external medium for curves *1*–*6* was taken to be $n = 1.5$. It is seen from the calculations that, because the quantity to be measured is $\cos\varphi$ rather than angle φ, errors $\delta\varphi$ increase near $\varphi = 0$ and $\varphi = 180°$. In the range of values of φ that are rather far from these boundaries, the measurement accuracy of the phase is about 1° in both cases: upon measuring the reflection coefficient in the second medium and upon measuring the transmission coefficient. The accuracy can be somewhat improved if one optimizes the refractive index of the immersion medium. In the figure, curve 7 was calculated for $n = 2$ and $\mathcal{R} = 0.3$; in this case, the error decreased roughly twofold compared to calculations at $n = 1.5$.

The method under consideration can also be extended to the case of oblique incidence of light. It is only necessary that angles of incidence φ_0 and φ_1 in media with refractive indices n_0 and n_1 be related by Snell's law, $n_0\sin\varphi_0 = n_1\sin\varphi_1$, in order for relation (3) to remain valid. A similar method for measuring the phase of the reflection coefficients was previously proposed for ellipsometric measurements [5], with the only difference that, in the cited work, the experimentally measured quantities were the ratios of the complex reflection coefficients for two different polarizations, whereas, in the present study, only the amplitudes of the reflection coefficients are measured.

Let us consider ways of implementing measurements in a denser medium in practice.

For these purposes, it is most convenient to use a plane-parallel plate of a transparent material, e.g., glass (Fig. 3a). The optical contact between the plate and the sample is ensured by an immersion liquid with a refractive index close to the refractive index of the plate. For example, for a plate made of the K8 glass (crown glass), the refractive index of which at a wavelength of $\lambda = 587.56$ nm is $n = 1.5164$ [6], glycerol ($n = 1.47$ [7]) can be used as an immersion liquid. For this difference between their refractive indices, the Fresnel losses at the glass–glycerol interface will be about

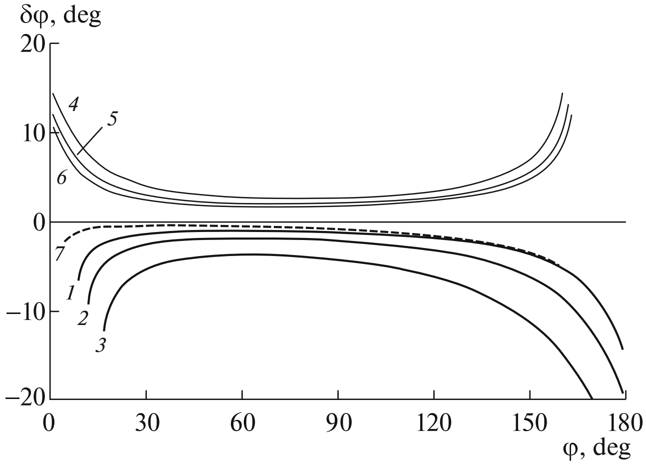

Fig. 2. Determination errors δφ of the phase of the reflection coefficient in relation to the value of phase φ calculated for the measurement errors of the reflection and transmission coefficients equal to ±0.005. The values of the reflection coefficient are as follows: $\mathcal{R}$ = (curves *1*, *4*, *7*) 0.3, (*2*, *5*) 0.5, and (*3*, *6*) 0.7. The phase was calculated by formula (10) (curves *1–3*, *7*) and formula (13) (curves *4–6*). The refractive index of the immersion medium for curves *1–6* was $n = 1.5$, while that for curve *7* was $n = 2$.

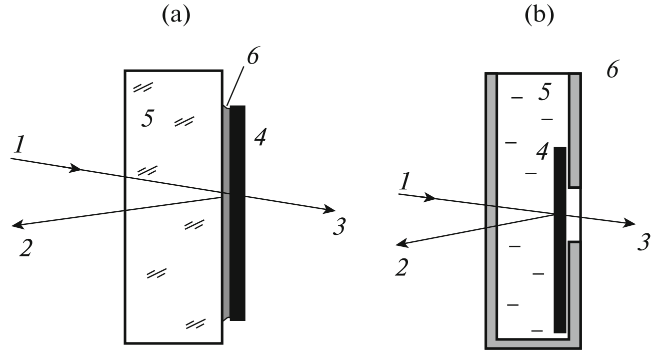

Fig. 3. Devices for performing measurements when light is incident on a sample from a denser medium. (a) A plane-parallel plate: (*1*, *2*, and *3*) incident, reflected, and transmitted rays, respectively; (*4*) sample; (*5*) transparent plate; and (*6*) immersion liquid. (b) A photometric cell: (*1*, *2*, and *3*) incident, reflected, and transmitted rays, respectively; (*4*) sample; (*5*) immersion liquid; and (*6*) cell walls made of a transparent material.

0.02%, which is an order of magnitude smaller than the instrumental errors.

The reflection at the air–glass interface is more considerable and should be taken into account in calculations. When the light passes through this interface twice, the relationship between the reflection coefficient in the medium, $\mathcal{R}_1$, and its measured value is given by the following expression:

$$\mathcal{R}_1 = \left(\frac{(n+1)^2}{4n}\right)^2 \mathcal{R}_{1\text{meas}}. \quad (17)$$

Upon measuring the transmission coefficient, the light passes through the air–glass interface only once, and the correction for Fresnel losses is expressed in this case by the formula

$$\mathcal{T}_1 = \frac{(n+1)^2}{4n} \mathcal{T}_{1\text{meas}}. \quad (18)$$

Equations (17) and (18) do not take into account multiple reflections between the input face of the plate and the sample. They can be eliminated by tilting the sample from the normal by a small angle. If the thickness of the plate is sufficiently large, the reflected parasitic rays will prove to be outside the optical path. If the tilt angle of the sample does not exceed a few degrees, expressions (17) and (18) will remain practically unchanged.

Alternatively to the plane-parallel plate, measurements can also be performed using a standard photometric cell filled with an immersion liquid, as is shown in Fig. 3b. In this case, the refractive index of the cell wall through which the incident and reflected rays pass should be close to the refractive index of the liquid, in order to obviate additional complications in calculations. For measuring the transmission coefficients, the photometric cell should have an output hole. The sample is pressed against the cell wall, ensuring its hermeticity, its working surface faces the liquid, while the second surface is exposed to air. Corrections to the measured reflection and transmission coefficients are given by the same formulas (17) and (18).

Let us dwell on the possibilities of practical applications of results of the proposed method. Upon development and fabrication of optical devices, the measurement of the phase of the reflection coefficient may be of independent interest, e.g., upon fabrication of kinoform elements, and to characterize the parameters of phase-shifting elements or mirrors. However, the knowledge of the phase can also be used to study layered structures in order to extract additional information about their parameters. For example, from the spectrum of the light reflection coefficient of a bulk sample, it is impossible to simultaneously determine both optical parameters, refractive index n and absorption index k of the material. Indeed, the expression for $\mathcal{R}$ in the case of a normal incidence has the form

$$\mathcal{R} = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}, \quad (19)$$

and, to determine the optical constants, additional information is required, for which the equation for the phase can be used. After transformations, the expression for the final solution for the optical constants can be written in the complex form as

$$n - ik = \frac{1 + R}{1 - R}, \quad (20)$$

where the complex reflection coefficient $R = \sqrt{\mathcal{R}}e^{i\varphi}$ is expressed via its experimentally measured modulus and phase.

Optical constants n and k of a thin semitransparent layer deposited on a substrate can be measured in a similar way, provided that the optical constants of the substrate, as well as thickness d of this layer, are known. The complex reflection coefficients of such a structure are expressed by well-known equations [4], which can be solved with respect to n and k numerically. The requirement for the known layer thickness is not crucial if the spectral range of a material under study contains a transparency interval. In this case, it is first necessary to calculate the thickness by solving the obtained equation with respect to n and d, and then to calculate optical constants n and k over the entire spectral range.

Further development of this idea makes it possible, in principle, to profile optically inhomogeneous layers by sequential and thickness-controlled etching of an inhomogeneous layer with subsequent measurement of the reflection spectra and the phase of the reflection coefficient. This profiling cannot be performed based solely on the spectra of the energy-reflection coefficients, since the corresponding relations for the scattering matrix of the inhomogeneous structure under consideration have a complex form, and the loss of information on the phase makes the solution of the inverse problem impossible. However, if the phase of the reflection coefficient is measured at each etching step, then the parameters of the layers can be determined in the order that is reverse to the order in which the experiment is performed (i.e., doing sequential calculations from the substrate to the surface). At each iteration step, the parameters of the jth layer are determined and scattering matrix $\hat{S}_j$ of the j-layered structure, which serves as a substrate for determining the parameters of the $(j + 1)$th layer, is restored. We note that we have successfully applied this algorithm previously in ellipsometric measurements [8].

Thus, we proposed a method for measuring the phase of the reflection coefficient for an arbitrary reflecting structure. This method can be realized most simply in the optical range of the spectrum. For this purpose, one can use any spectrophotometer with the function of measuring the reflection coefficient, which is equipped with a device for performing immersion measurements. The thus-obtained information on the phase of the reflection coefficient qualitatively extends the functional capabilities of spectrophotometric measurements.

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