

INTERFEROMETRIC DETERMINATION OF THE REFRACTIVE INDEX IN THE INFRARED REGION

I. A CALCULATION METHOD FOR THE TRANSMITTANCE OF THE FABRY-PEROT INTERFEROMETER

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1. Introduction

Interferometry is a relatively simple and very suitable method for the determination of the real part of the complex refractive index. RAMADIER-DELBÈS was the first to propose this method for the investigation of liquids in the infrared spectrum range [1]. She used a Fabry-Perot etalon made from an optical material of high refractive index as interferometer. Her results were developed by VINCENT-GEISSE and LECOMTE [2]. A very important suggestion by KAGARISE and MAYFIELD was to make the interferometer of germanium [3]. This material has a very high reflectivity and its absorptivity is practically zero over a large spectrum range. It is a hard one and after polishing its surface is smooth and resistant.

In the present paper the optical properties of the empty Fabry-Perot etalon will be dealt with. The phenomenon of the interference occurring in the etalon has been interpreted up to now by the Airy's formula deduced for the case of a single thin plane parallel plate [4] of the form:

$$T = \frac{(1-r^2)^2}{1-2r^2 \cos \delta + r^4} \quad (1)$$

where T is the transmittancy of the non-absorbing plate, r is the reflection coefficient of the surface (ratio of reflected to incident amplitudes) that may be calculated from the refractive index n of the optical material by the following expression:

$$r^2 = \left\{ \frac{n-1}{n+1} \right\}^2 \quad (2)$$

The connection between the reflection coefficient r and reflectivity \mathfrak{R} of a surface (ratio of reflected to incident intensity) is given by $\mathfrak{R} = r^2$.

The transmission coefficient is defined as:

$$\mathfrak{T} = (1-r^2)^{1/2} \quad (3)$$

The phase difference δ involves the layer thickness D , the frequency ν , the refractive index of the layer, n , and the velocity of light in vacuum, c .

$$\delta = 4\pi n\nu \frac{D}{c} \quad (4)$$

The expression for the maximum and minimum transmittancies of the interference fringes derived from Eq. (1) are:

$$T_{\max} = \left\{ \frac{1-r^2}{1-r^2} \right\}^2 = 1 \quad (5a)$$

$$T_{\min} = \left\{ \frac{1-r^2}{1+r^2} \right\}^2. \quad (5b)$$

In reality, the Fabry-Perot etalon consists of two thick plane parallel plates and the internal reflection among the four surfaces is left unconsidered in the Airy's formula. This formula is only suitable for the calculation if the etalon is of a low-reflectivity optical material. Especially the ratio of maximum to minimum transmittancy (so called contrast factor) may closely be approximated in this case. It must be noted that according to Airy's formula, the layer thickness D in Eq. (4) corresponds to that provided by the spacer, the refractive index n , however, to that of the thick plates material. The KRS-5, Si and Ge have very high reflectivities in the infrared range, so the effect of the internal reflection can not be neglected.

In the following chapters an expression in closed form will be deduced for the transmittancy of a single thick transparent plate, and an infinite series for that one of the Fabry-Perot etalon, using two assumptions: the light beam is incident perpendicularly on the surfaces and the absorptivity of the material may be neglected.

2. The transmittance of a single thick plate

Let us consider a plane wave incident perpendicularly on the surface of a thick plate. The resultant wave (wave-front) consists of an infinite number of plane waves each having a phase difference δ with respect to the adjacent one. The phase difference δ is due to the internal reflection. The wave-front has the following form [4]

$$\psi = E_0 \vartheta^2 e^{-i(\omega t - \delta/2)} \sum_{k=0}^{+\infty} r^{2k} e^{ik\delta} \quad (6)$$

from which the transmittancy of the plate is given by multiplication with the complex conjugate of function (6)

$$T = \vartheta^4 \sum_{k=0}^{+\infty} \sum_{l=0}^{+\infty} r^{2(k+l)} e^{i(k-l)\delta}. \quad (7)$$

There is a very important difference between the effects produced by a thin and a thick plate, since in the case of a thick plate the average of the transmittancy function (7) can only be recorded by a spectrophotometer. This average is:

$$T = \vartheta^4 \sum_{k=0}^{+\infty} \sum_{l=0}^{+\infty} r^{2(k+l)} \frac{1}{\Delta\nu} \int_{\nu_1}^{\nu_2} e^{i(k-l)\delta} d\nu \quad (8)$$

where the frequency interval $\Delta\nu$ is comparable to the resolution of the instrument.

It is clear that in expression (8) only those members differ from zero for which indices k equal l . The result is an infinite geometrical series, the first member of which is $(1 - r^2)^2$ according to Eq. (3), the quotient equals r^4 . From the series the following simple formula may be deduced:

$$T = \frac{1 - \mathfrak{R}}{1 + \mathfrak{R}}. \quad (9)$$

Note that the reflectivity \mathfrak{R} of a surface is not equivalent to the reflectivity of a plate, R arising from the internal reflection. This latter quantity may be calculated from the transmittancy by the trivial relationship $R + T = 1$.

The transmittancies of some optical materials calculated by Eq. (9) and (2) are in good agreement with the measured data (Table I).

The refractive index of KRS-5 published in the literature concerns a material consisting of 50% TII + 50% TIBr.

Table I

	λ [μ]	n_{20}	\mathfrak{R}	T calc.	T exp.
NaCl	3.00	1.5243 ^[5]	0.0431	0.917	0.915
KRS-5	3.00	2.3876 ^[6]	0.1678	0.713	0.725
Ge	10.00	4.00 ^[7]	0.3600	0.471	0.470

3. The transmittance of the Fabry-Perot etalon

Let us consider a plane wave ψ_0 which is incident perpendicularly on the first surface of a Fabry-Perot etalon consisting of two plates with the same thickness H . The plane wave ψ_0 is divided into two parts on the surface. One

of them, $r\psi_0$ is reflected and the other, $\vartheta\psi_0$ is transmitted. This latter one is separated into an infinite number of waves characterized by their optical paths in the etalon. These individual plane waves may be obtained in the following form:

$$\psi_{pq} = E_0 c_{pq} \exp \{ -i [\omega t - \delta_{pq}] \} \quad (10)$$

where

$$\delta_{pq} = \frac{\omega}{c} \{ (2p+2)nH + (2q+1)D \} \quad (11)$$

in which $(c/\omega) \delta_{pq}$ is the optical path.

An optical path characterized by p and q may be realized by one or more different ways. In the following these ways will be called paths. Both the number of internal reflections (ϱ) and the transmissions through the surfaces (τ) may be different in the case of paths corresponding to a given pair p, q . The coefficient c_{pq} in relation (10) has the form of

$$c_{pq} = \left\{ \sum_{\tau} \sum_{\varrho} A_{\varrho\tau}^{pq} \vartheta^{2\tau} r^{2\varrho} \right\}^{1/2} \quad (12)$$

where $A_{\varrho\tau}^{pq}$ is a weight factor equivalent to the number of the paths having common ϱ and τ . It is evident that the sum of the $A_{\varrho\tau}^{pq}$ gives the number of the paths in an investigated optical path.

According to the above statement, on the last surface the wave-front is a superposition of the individual plane wave (10)

$$\psi = E_0 e^{-i\omega t} \sum_{q=0}^{\infty} \sum_{p=0}^{\infty} c_{pq} e^{i\delta_{pq}}. \quad (13)$$

The transmittancy of the etalon is:

$$T = \sum_{\substack{q=0 \\ q'=0}}^{+\infty} \sum_{\substack{p=0 \\ p'=0}}^{+\infty} c_{p'q'} c_{pq} \exp i \{ \delta_{pq} - \delta_{p'q'} \}. \quad (14)$$

Expression (14) may be transformed into the following form:

$$T = \sum_{\xi=0}^{+\infty} \sum_{\zeta=0}^{+\infty} c_{\xi\zeta} \cos \frac{2\omega}{c} \{ \xi nH + \zeta D \} \quad (15)$$

where

$$\zeta = |p - p'| \quad (16a)$$

$$\xi = |q - q'| \tag{16b}$$

and

$$c_{\zeta\xi} = \frac{1}{2} \sum_{p=\zeta}^{+\infty} \sum_{q=\xi}^{+\infty} c_{(p-\zeta)(q-\xi)} c_{pq} \quad \text{if } \zeta, \xi \neq 0 \tag{16c}$$

$$c_{00} = \sum_{p=0}^{+\infty} \sum_{q=0}^{+\infty} c_{pq}^2. \tag{16d}$$

By analogy to Chapter 2, it is valid to the relationship (15) that in the series those and only those members are averaged by the spectrophotometer which contain the thickness H . Using a simple trigonometrical transformation, the averages are:

$$\frac{1}{\Delta\nu} \cos \frac{2\omega}{c} \xi D \int_{\nu_1}^{\nu_2} \cos \frac{2\omega}{c} \zeta n H d\nu \begin{cases} \nearrow \cos \frac{2\omega}{c} \xi D & \text{for } \zeta = 0 \\ \searrow 0 & \text{for } \zeta \neq 0 \end{cases} \tag{17a}$$

$$\frac{1}{\Delta\nu} \sin \frac{2\omega}{c} \xi D \int_{\nu_1}^{\nu_2} \sin \frac{2\omega}{c} \zeta n H d\nu = 0 \tag{17b}$$

where frequency interval $\Delta\nu$ is comparable to the resolution of the instrument. In this interval the first slowly changing factor may be considered constant.

Using the above results, the expression of transmittancy is:

$$T = \sum_{\xi=0}^{+\infty} c_{\xi} \cos \frac{2\omega}{c} \xi D \tag{18}$$

neglecting the unnecessary index ζ .

If the distance D is very large, the relationship (18) must be averaged further to yield:

$$T = c_0. \tag{19}$$

The maximum and the minimum of the transmittancy is given by the following expression:

$$T_{\max} = c_0 + \sum_{\xi=1}^{+\infty} c_{\xi} \quad \text{for} \quad \frac{2\omega}{c} \xi D = 2\pi N \tag{20a}$$

$$T_{\min} = c_0 - \sum_{\xi=1}^{+\infty} c_{\xi} \quad \text{for} \quad \frac{2\omega}{c} \xi D = 2\pi \left(N + \frac{1}{2} \right) \tag{20b}$$

where N is the order of interference.

The coefficients $A_{\sigma\tau}^{pq}$ may not be given by a closed or a recursion formula. In the following chapter a relatively simple method is presented for determining the coefficients $A_{\sigma\tau}^{pq}$ based on the graph theory.

4. A method for determining the paths in a given optical path

Let us consider the four surfaces of the etalon as four vertices of a directed multiple-graph (digraph) numbered from 1 to 4 in the direction of the light travel.

The following conditions are to be met: Only adjacent vertices can be connected directly by the edges of the digraph. All edges of length D must be between vertices 2 and 3. Odd number of edges must be put between vertices 1 and 2, and 3 and 4, respectively. The direction of the edges must be chosen so that by one more edge start from a vertex in the direction of the light travel than arrive to the same vertex from the opposite direction. Naturally, vertex 4 is an exception. Uniting vertices 1 and 4 creates a pseudo-symmetric digraph, where the outdegree and indegree of any vertex is identical.

For a given pair p, q , $p + 1$ different graphs may be sketched. Our problem is to determine the possible distinguishable paths interpreted on these digraphs such a way that in the building of a path each edge is used once and only once. The number of internal reflection (ϱ) and transmission (τ) belonging to these paths must also be determined.

These edges will be denoted by two numbers representing the starting and the arriving vertex, respectively. The possible paths may be built up from the edges in the following manner. The first edge is always one of the 12, the last one is one of the 34. The other edges must be placed between the above two ones in such an order that the first number of the edge is identical with the second one of the previous edge. From a given set of edges, in general more than one different paths can be constructed by applying a systematic transposition.

In the case of a path built up in this way, the number of internal reflection (ϱ) is equivalent to the possible number of pairs formed from the adjacent edges of opposite direction, the number of transmission (τ) to the number of pairs formed from the adjacent edges having identical direction, plus 2 corresponding to the transmission through the first and last surfaces. For a given pair p, q the sum of ϱ and τ is constant.

It must be noted that these paths are not identical to the so called Euler's paths belonging to the same digraph [8], since several Euler's paths are indistinguishable in our edge notation system corresponding to our optical problem.

Consider an example for our method: Let $2p + 2 = 6$ and $2q + 1 = 3$. Three different graphs may be sketched (Fig. 1). The following possible paths may be derived from these digraphs using our theorem:

$$\begin{array}{ll}
 a) \{12, 23, 32, 23, 34, 43, 34, 43, 34\} & \tau = 4 \quad \varrho = 6 \\
 \{12, 23, 34, 43, 32, 23, 34, 43, 34\} & \tau = 6 \quad \varrho = 4 \\
 \{12, 23, 34, 43, 34, 43, 32, 23, 34\} & \tau = 6 \quad \varrho = 4
 \end{array}$$

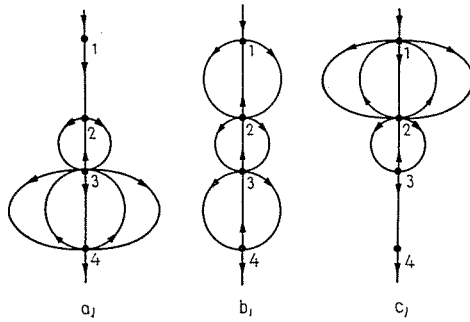


Fig. 1

b)	{12, 21, 12, 23, 32, 23, 34, 43, 34}	$\tau = 4$	$\rho = 6$
	{12, 23, 32, 21, 12, 24, 34, 43, 34}	$\tau = 6$	$\rho = 4$
	{12, 23, 34, 43, 32, 21, 12, 23, 34}	$\tau = 8$	$\rho = 2$
c)	{12, 21, 12, 21, 12, 23, 32, 23, 34}	$\tau = 4$	$\rho = 6$
	{12, 21, 12, 23, 32, 21, 12, 23, 34}	$\tau = 6$	$\rho = 4$
	{12, 23, 32, 21, 12, 21, 12, 23, 34}	$\tau = 6$	$\rho = 4$

The pictures of the internal reflections may be seen in Fig. 2.

Some constants $A_{\sigma\tau}^{pq}$ of significant weight applied in the calculations are presented in Tables 11a, b, c, d and e.

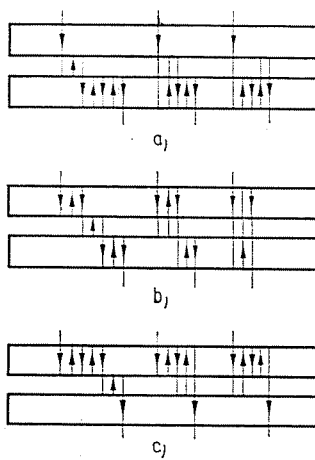


Fig. 2

Table IIa
 $2q + 1 = 1$

$\tau + \varrho$	ϱ $2p + 2$	0	2	4	6	8
4	2	1	0	0	0	0
6	4	0	2	0	0	0
8	6	0	0	3	0	0
10	8	0	0	0	4	0
12	10	0	0	0	0	5

Table IIb
 $2q + 1 = 3$

$\tau + \rho$	ϱ $2p \times 2$	2	4	6	8	10	12
6	2	1	0	0	0	0	0
8	4	2	2	0	0	0	0
10	6	1	6	3	0	0	0
12	8	0	4	11	4	0	0
14	10	0	0	9	18	5	0
16	12	0	0	0	20	20	6

Table IIc
 $2q + 1 = 5$

$\tau + \varrho$	ϱ $2p + 2$	4	6	8	10
8	2	1	0	0	0
10	4	4	2	0	0
12	6	6	12	3	0
14	8	3	17	22	4

Table IId
 $2q + 1 = 7$

$\tau + \varrho$	ϱ $2p + 2$	6	8	10
10	2	1	0	0
12	4	6	2	0
14	6	13	17	3

Table IIe
 $2q + 1 = 9$

$\tau + q$	q		
	$2p + 2$	8	10
12	4	1	0
14	6	8	2

5. Experimental

The transmittancy of the Fabry-Perot etalon made of NaCl, KRS—5 and Ge, respectively, were investigated. The NaCl and KRS—5 plates were usual cell windows (produced by C. Zeiss, Jena). The Ge plates made from 40 Ω cm, n-type polycrystalline germanium were 40 \times 22 \times 6 mm and had a high polish. These ones were obtained from Tungsram Inc. Budapest.

Two spectrometers, a Zeiss UR—20 instrument equipped with KBr, NaCl and LiF prisms and a Hilger H900 one employing a NaCl prism and gratings combination were used to observe the interference fringes.

6. Results and discussion

In the first step the transmittancy of the etalon of great layer thickness was investigated. Data obtained in experiments and calculated by our theory, respectively, may be found in Table IIIa.

Table IIIa

	λ [μ]	T cal.	T exp.
NaCl	3.00	0.847	0.840
KRS-5	3.00	0.569	0.560
Ge	10.00	0.302	0.290

There is a very good agreement.

The shape of the interference fringes was approximated by the following finite series:

$$T_{NaCl} = 0.8470 + 0.0198 \cos \alpha + 0.0009 \cos 2\alpha \quad (21a)$$

$$T_{KRS-5} = 0.5380 + 0.0645 \cos \alpha + 0.0136 \cos 2\alpha + 0.0041 \cos 3\alpha \quad (21b)$$

$$T_{Ge} = 0.3020 + 0.0741 \cos \alpha + 0.0323 \cos 2\alpha + 0.0141 \cos 3\alpha + \\ + 0.0082 \cos 4\alpha + 0.0054 \cos 5\alpha \quad (21c)$$

where

$$\alpha = \frac{2\omega}{c} D.$$

The constants of the series were calculated by the expressions (12), (16) and the data of Table II*a, b, c, d, e*. The fourth and fifth constants in the series (21*c*) were determined by extrapolation using an empirical relationship of the form:

$$\frac{1}{c_{\xi}} = a\xi^2 + b. \quad (22)$$

The maximum and minimum transmittancies of interference fringes may be seen in Table III*b*. Unfortunately, our KRS-5 plates, with a refractive index corresponding to that of the composition 50% TlI and 50% TlBr, have not the surfaces required for good interference effects. The refractive index of the plates with adequate surfaces, however, was unknown. Therefore, the reflectivity of the plates was calculated from the transmittancy of the single plate using the expression (9). Naturally, this fact increases the error of the calculation.

Table III*b*

	λ [μ]	T_{\max} cal.	T_{\max} exp.	T_{\min} cal.	T_{\min} exp.
NaCl	3.00	0.368	0.860	0.826	0.825
KRS-4	2.00	0.620	0.640	0.456	0.425
Ge	10.00	0.436	0.440	0.168	0.170

Table III*c* contains the contrast factors T_{\max}/T_{\min} calculated by the Airy's formula, our theory and the measured data, respectively.

Table III*c*

	λ [μ]	T_{\max}/T_{\min} Airy	T_{\max}/T_{\min} cal.	T_{\max}/T_{\min} exp.
NaCl	3.00	1.19	1.05	1.04
KRS-5	2.00	2.08	1.36	1.51
Ge	10.00	4.52	2.60	2.59

Despite the fact that the convergence of the light beam of the spectrophotometer was neglected in our model, what is more, both the flatness of the surfaces and the plane parallelity of the layer may not be perfect, the

agreement between the experimental and calculated data is very good. The results according to our theory are more satisfactory than those from the Airy's formula which gives e.g. $T_{\max} = 1.000$, $T_{\min} = 0.228$ for Ge. The error may be reduced by taking into account more coefficients $A_{gr}^{e\tau}$ than are found in Table IIa, b, c, d, e.

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Summary

A new calculation method is presented for the determination of the transmittancy of a single thick plate and the Fabry-Perot interferometer. For the single thick plate a short, closed formula and for the Fabry-Perot etalon an infinite series were deduced. The determination of the constants of the series is based on the graph theory. The transmittancy was investigated in the case of three optical materials, NaCl, KRS-5 and Ge, respectively. The agreement between the experimental and calculated data is very good.

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