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Accuracy in the Gupta method of optical constant determination

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Abstract. The Gupta method leads to the determination of the optical constants of an absorbing material through two measurements of reflectivity. In this paper, the effect of experimental errors on both the accuracy of the determination and the conditions for the achievement of unambiguous results is studied.

1. Introduction

The determination of the optical constants of a material is of fundamental importance in a large variety of applications. In the commonly used experimental set-ups, a sample of the material under study is made to interact with a radiation beam and the unknown optical constants are deduced through the measurement of some characteristics of the transmitted and/or reflected beam [1–9].

In the case of strongly absorbing mateial, reflection measurements are obviously preferable and many methods have been envisaged in order to determine the optical constants through simple measurements of the reflectivity of the material [10–19]. Among these methods, that proposed by Gupta in 1988 is of particular interest for its simplicity [20]. From an experimental point of view, the Gupta method requires only the measurement of the reflectivity of the material in the presence and absence of a transparent overcoat film. However, as a common drawback in this kind of problem, the analytic determination of the optical constants starting from the measured reflectivities can lead to multiple solutions and the unknown optical constants can be unambiguously determined only if the wrong solutions are somehow recognizable. It has been demonstrated that, for a suitable range of the optical thickness of the overcoat film, the determination of the unknown optical constants is mathematically unambiguous and the limiting values of such a 'useful interval' of thicknesses have been found analytically [21].

In [21], the analytic treatment of the Gupta method and the subsequent evaluation of the conditions for an unambiguous determination of the optical constants are accomplished without taking into account the measurement errors. Errors in the measured reflectivities influence the application of the Gupta method in two ways. First, of course, they determine the uncertainties in the estimated values of the optical constants. Second, they can affect the ambiguity problem. A solution that is unambiguous in the absence of errors could become ambiguous if the errors exceed certain values. In this paper, we extend the analysis of [21] to determine quantitatively the effects of the measurement errors. In section 2, we briefly recall the mathematical basis of the

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Gupta method showing the existence of a 'useful interval' for an unambiguous determination of the optical constants. In section 3, we estimate the accuracy which is achievable in the Gupta method and its dependence on the experimental conditions. In section 4, the ambiguity problem is studied by taking into account the experimental accuracy and a 'practical useful interval' is determined inside the theoretical one. Finally, in section 5, the results obtained are briefly discussed and some conclusions are drawn.

2. The mathematical basis of the method

Let us denote by n and k the unknown optical constants of the material under study and by $n_1 = n - ik$ its complex refractive index. The normal incidence air–material reflectivity is given by [22]

$$R_1 = \left|\frac{1 - n_1}{1 + n_1}\right|^2 \tag{1}$$

where the refractive index of the air has been assumed equal to one and the material has been supposed sufficiently thick to completely absorb the wave reflected from the second interface.

In addition to R_1, a second measurement is needed in order to determine the two unknown quantities n and k. In the Gupta method, the material is coated with a homogeneous plane-parallel non-absorbing film of known real refractive index n_2 and thickness d and the new reflectivity, say R_2, is measured. We have [21]

$$R_2 = \left|\frac{1 - n_1 z}{1 + n_1 z^*}\right|^2 \tag{2}$$

where

$$z = \frac{2}{1 + n_2^2 + (1 - n_2^2)\cos(\gamma)} - \mathrm{i}\,\frac{(1 - n_2^2)\sin(\gamma)}{n_2[1 + n_2^2 + (1 - n_2^2)\cos(\gamma)]}. \tag{3}$$

the asterisk denotes complex conjugate and

$$\gamma = 4\pi n_2 d/\lambda. \tag{4}$$

In equation (4), λ is the wavelength of the radiation used.

Making use of the two equations (1) and (2), the unknown quantities n and k can be calculated starting from the measured reflectivities R_1 and R_2 for a given γ value. The solution of this inverse problem has been discussed in [21]. For later convenience, we reproduce the essential steps in appendix 1.

As it has been shown in [21], in the general case two pairs of solutions are obtained which we will denote by the superscripts I and II, i.e.

$$n^{\mathrm{I}}, k^{\mathrm{I}} \qquad \text{(first solution)} \tag{5}$$

and

$$n^{\mathrm{II}}, k^{\mathrm{II}} \qquad \text{(second solution).} \tag{6}$$

By inserting back into equation (1) and (2) the solutions given by equations (5) and (6), we obtain the corresponding values of the reflectivities, say R_1^{I}, R_2^{I} and R_1^{II}, R_2^{II}, respectively. These results have to be compared with the measured reflectivities R_1, R_2, so that the case of a wrong solution for the optical constants can be recognized.

It can be shown that the following relationship is always true

$$R_1^{\mathrm{I}} = R_1^{\mathrm{II}} = R_1 \tag{7}$$

while the corresponding relationship for R_2 is true only outside a certain interval. Such an interval can be called a 'useful interval', because inside it one of the two solutions does not reproduce the right reflectivity R_2 and it can then be rejected as a spurious solution. As a consequence, for γ inside the 'useful interval', the optical constants n, k can be unambiguously determined through the Gupta method. Furthermore, in [21], the extreme points (denoted by γ_1 and γ_2) of the 'useful interval' have been explicitly evaluated in the general case and their numerical values for the practical case originally considered by Gupta [20] have also been calculated.

[21] aimed at studying the mathematical ambiguity connected with the optical constant determination in the ideal limit case of absence of measurement errors. When these are taken into account, the optical constant determination is affected by a certain degree of uncertainty and this influences the width of the 'useful interval'. In the next section, we will evaluate the accuracy with which the optical constants can be calculated starting from given errors in the reflectivity measurements. Correspondingly, we will be able to determine the effective width of the 'useful interval' in any practical situation.

3. The accuracy of the method

In this section, we evaluate the uncertainties in the calculated optical constants due to the measurement errors in the reflectivities.

Let us indicate by $R_{1\mathrm{m}}$ and $R_{2\mathrm{m}}$ the measured values of the reflectivities R_1 and R_2, respectively, and by ΔR_1 and ΔR_2 the corresponding measurement errors. Starting from some reflectivity values R_1 and R_2, included inside the intervals $(R_{1\mathrm{m}} - \Delta R_1, R_{1\mathrm{m}} + \Delta R_1)$ and $(R_{2\mathrm{m}} - \Delta R_2, R_{2\mathrm{m}} + \Delta R_2)$, respectively, the corresponding optical constants n^{I} and k^{I} (or n^{II} and k^{II}) can be calculated through the well known formulae of error propagation

$$n^{\mathrm{I,II}} - n_{\mathrm{m}}^{\mathrm{I,II}} = \left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1 = R_{1m}} (R_1 - R_{1\mathrm{m}}) + \left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2 = R_{2m}} (R_2 - R_{2\mathrm{m}}) \tag{8}$$

$$k^{\mathrm{I,II}} - k_{\mathrm{m}}^{\mathrm{I,II}} = \left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1 = R_{1m}} (R_1 - R_{1\mathrm{m}}) + \left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2 = R_{2m}} (R_2 - R_{2\mathrm{m}}) \tag{9}$$

where $n_{\mathrm{m}}^{\mathrm{I}}$ and $k_{\mathrm{m}}^{\mathrm{I}}$ (or $n_{\mathrm{m}}^{\mathrm{II}}$ and $k_{\mathrm{m}}^{\mathrm{II}}$) are the optical constant values corresponding to the measured reflectivities $R_{1\mathrm{m}}$ and $R_{2\mathrm{m}}$. In equations (8) and (9), the superscripts I or II must be assumed simultaneously and the explicit expressions of the partial derivatives at the right-hand sides of equations (8) and (9) are given in appendix 2 (equations (A2.10)–(A2.13)).

It can be easily seen [23] that n^{I} and k^{I} (or n^{II} and k^{II}) vary in the plane n^{I}, k^{I} (or n^{II}, k^{II}) inside a parallelogram, centered on the point $n_{\mathrm{m}}^{\mathrm{I}}$, $k_{\mathrm{m}}^{\mathrm{I}}$ (or $n_{\mathrm{m}}^{\mathrm{II}}$, $k_{\mathrm{m}}^{\mathrm{II}}$), whose corners have the following n coordinates

$$n_{\mathrm{m}}^{\mathrm{I,II}} + \left[\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1 = R_{1\mathrm{m}}} \Delta R_1 + \left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2 = R_{2m}} \Delta R_2\right] \tag{10}$$

$$n_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}(-\Delta R_1)+\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}\Delta R_2\right] \tag{11}$$

$$n_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}(-\Delta R_1)+\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}(-\Delta R_2)\right] \tag{12}$$

$$n_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}\Delta R_1+\left(\frac{\partial n^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}(-\Delta R_2)\right] \tag{13}$$

and the following corresponding k coordinates

$$k_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}\Delta R_1+\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}\Delta R_2\right] \tag{14}$$

$$k_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}(-\Delta R_1)+\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}\Delta R_2\right] \tag{15}$$

$$k_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}(-\Delta R_1)+\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}(-\Delta R_2)\right] \tag{16}$$

$$k_{\mathrm{m}}^{\mathrm{I,II}}+\left[\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_1}\right)_{R_1=R_{1m}}\Delta R_1+\left(\frac{\partial k^{\mathrm{I,II}}}{\partial R_2}\right)_{R_2=R_{2m}}(-\Delta R_2)\right]. \tag{17}$$

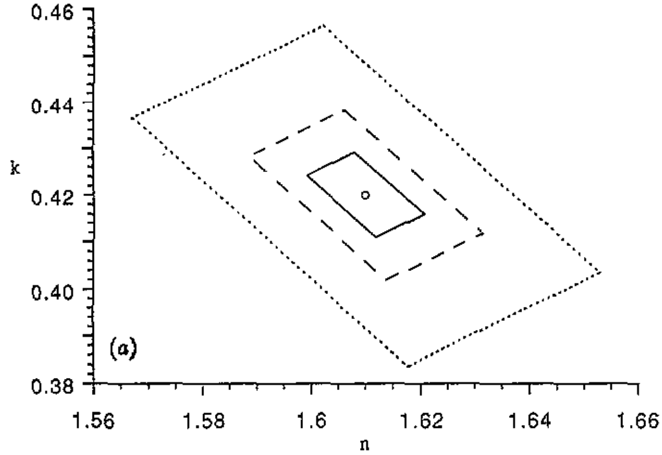
In equations (10)–(17), the superscripts I or II must be assumed simultaneously.

By making use of these error parallelograms, the accuracy patterns shown in figures 1 to 3 can be constructed. In all the figures, the numerical values of Gupta's original example are used. Although the degree of accuracy represented by a given parallelogram is somewhat arbitrary [23], it is evident from the figures that the accuracy decreases when the measurement errors increase (figure 1(*a*)) and that, for a given reflectivity error, less accurate parallelograms arise from γ values near the border of the 'useful interval' (figure 1(*b*)). Furthermore, it can be noted from figures 2 and 3 that, when γ approaches π, the two solutions given by equations (5) and (6) tend to coincide and the corresponding parallelograms partially overlap. As a consequence, a γ interval exists inside which the propagation of the measurement errors makes the two solutions for the optical constants indistinguishable. Figures 2 and 3 refer to different per cent errors in R_1 and R_2: these amount to 2% in figure 2 and to 4% in figure 3. The comparison between the two figures shows that the 'indistinguishableness interval' enlarges in correspondence to greater per cent errors in the measured reflectivities.

In the next section, we will consider the effects of the measurement errors on the ambiguity problem.

4. The ambiguity problem

Starting from the pairs of solutions n^{I}, k^{I} and n^{II}, k^{II}, we can trace back to the corresponding reflectivities R_1^{I}, R_2^{I} and R_1^{II}, R_2^{II}. As already mentioned in section 2, in the absence of measurement errors, a 'useful interval' of γ values exists for which $R_2 = R_2^{\mathrm{II}} \neq R_2^{\mathrm{I}}$ or $R_2 = R_2^{\mathrm{I}} \neq R_2^{\mathrm{II}}$, so that the true optical constants can be unambiguously determined. In [21], the extreme values γ_1 and γ_2 of this interval have been analytically



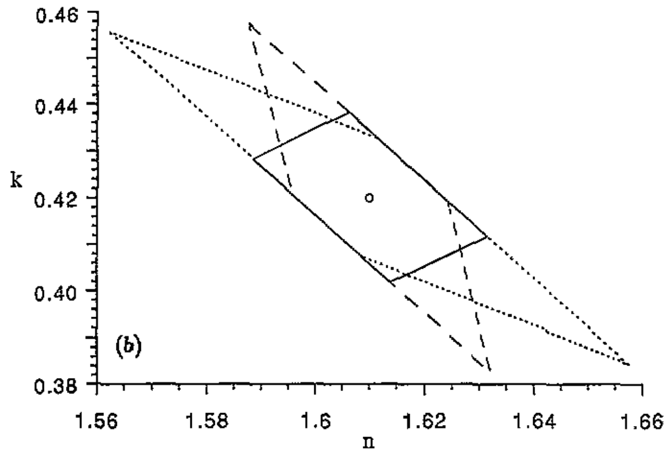


Figure 1. (*a*) Error parallelograms centred on the right solution for the optical constants n and k, in correspondence to several values of the per cent errors in the reflectivities R_1 and R_2 (full line, $\Delta R_1/R_1 = \Delta R_2/R_2 = 1\%$; broken line, $\Delta R_1/R_1 = \Delta R_2/R_2 = 2\%$; dotted line, $\Delta R_1/R_1 = \Delta R_2/R_2 = 4\%$). The phase shift γ undergone by the reflected radiation for a round trip through the coating material corresponds to the centre of the 'useful interval' ($\gamma = 4.6$). The true values of the optical constants are $n = 1.61$ and $k = 0.42$; the real refractive index of the coating film is $n_2 = 1.95$. (*b*) Error parallelograms centred on the right solution for the optical constants n and k, in correspondence to γ values corresponding to the centre of the 'useful interval' (full line, $\gamma = 4.6$) or to its borders (broken line, $\gamma = 2.7$; dotted line, $\gamma = 5.9$). The per cent errors in the reflectivities R_1 and R_2 are $\Delta R_1/R_1 = \Delta R_2/R_2 = 2\%$. The true values of the optical constants are $n = 1.61$ and $k = 0.42$; the real refractive index of the coating film is $n_2 = 1.95$.

calculated to hold

$$\gamma_1 = 2\tan^{-1}\left(\frac{2n_2k(P+R)}{(1+n_2^2)(M+R)}\right) \tag{18}$$

and

$$\gamma_2 = 2\tan^{-1}\left(\frac{2n_2k(P-R)}{(1+n_2^2)(M-R)}\right) + 2\pi \tag{19}$$

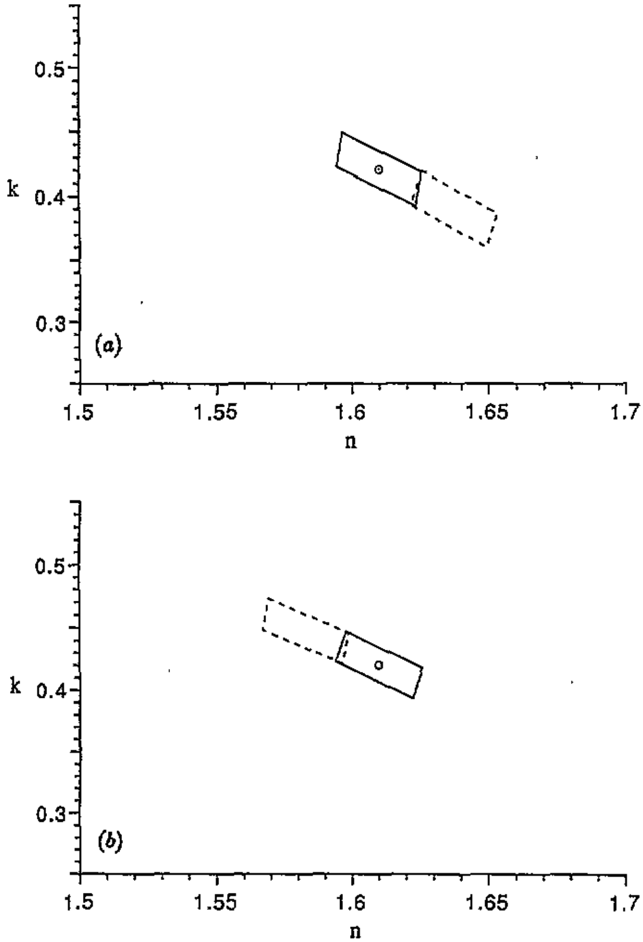


Figure 2. Error parallelograms centred on the right (full line) and wrong (broken line) solutions for the optical constants n and k, in correspondence to (a) $\gamma = 3.09$ and (b) $\gamma = 3.19$. The true values of the optical constants are $n = 1.61$ and $k = 0.42$; the real refractive index of the coating film is $n_2 = 1.95$. The percent errors in the reflectivities R_1 and R_2 amount to 2%.

where

$$P = 1 + k^2 + n^2 \tag{20}$$

$$M = 1 + k^2 - n^2 \tag{21}$$

$$R = [(1 + k^2 + n^2)^2 - 4n^2]^{1/2}. \tag{22}$$

In the presence of the measurement errors ΔR_1 and ΔR_2, the optical constants can be calculated with the uncertainties determined by the error parallelograms constructed in the preceding section.

As a consequence, also the quantities R_2^{I} and R_2^{II} are affected by some uncertainties, say ΔR_2^{I} and ΔR_2^{II}, respectively. These uncertainties can be easily evaluated as

$$\Delta R_2^{\mathrm{I,II}} = \left|\frac{\partial R_2^{\mathrm{I,II}}}{\partial n^{\mathrm{I,II}}}\frac{\partial n^{\mathrm{I,II}}}{\partial R_1} + \frac{\partial R_2^{\mathrm{I,II}}}{\partial k^{\mathrm{I,II}}}\frac{\partial k^{\mathrm{I,II}}}{\partial R_1}\right| \Delta R_1 + \left|\frac{\partial R_2^{\mathrm{I,II}}}{\partial n^{\mathrm{I,II}}}\frac{\partial n^{\mathrm{I,II}}}{\partial R_2} + \frac{\partial R_2^{\mathrm{I,II}}}{\partial k^{\mathrm{I,II}}}\frac{\partial k^{\mathrm{I,II}}}{\partial R_2}\right| \Delta R_2 \tag{23}$$

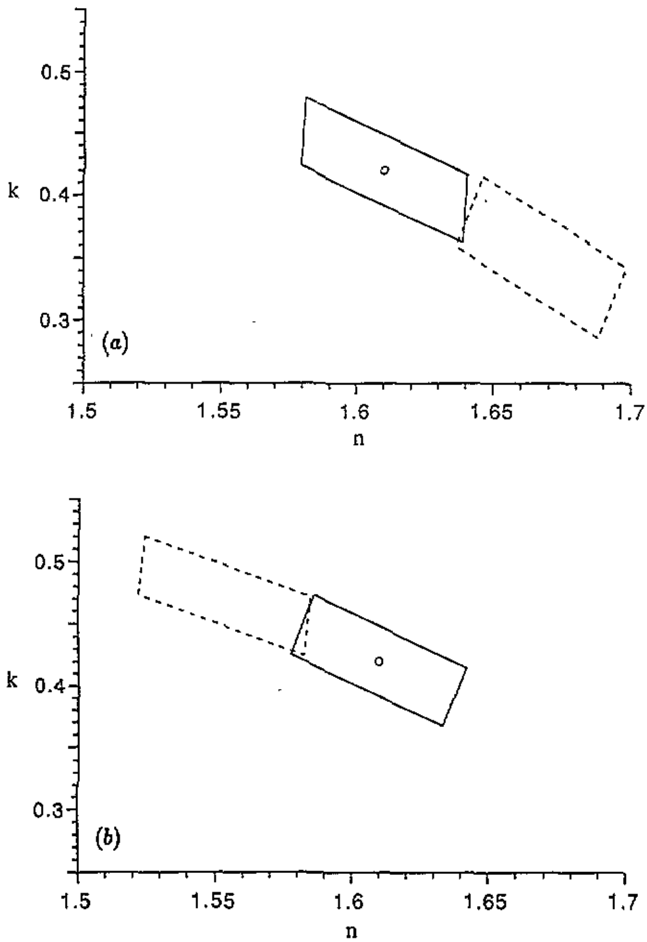


Figure 3. Error parallelograms centred on the right (full line) and wrong (broken line) solutions for the optical constants n and k, in correspondence to (a) $\gamma = 3.03$ and (b) $\gamma = 3.24$. The true values of the optical constants are $n = 1.61$ and $k = 0.42$; the real refractive index of the coating film is $n_2 = 1.95$. The per cent errors in the reflectivites R_1 and R_2 amount to 4%.

where

$$\frac{\partial R_2^{\mathrm{I,II}}}{\partial n^{\mathrm{I,II}}} = \frac{4XZ^2[(n^{\mathrm{I,II}})^2 - (k^{\mathrm{I,II}})^2] - 4X[1 + 2Yk^{\mathrm{I,II}}]}{\{1 + Z^2[(n^{\mathrm{I,II}})^2 + (k^{\mathrm{I,II}})^2] + 2Xn^{\mathrm{I,II}} + 2Yk^{\mathrm{I,II}}\}^2} \tag{24}$$

$$\frac{\partial R_2^{\mathrm{I,II}}}{\partial k^{\mathrm{I,II}}} = \frac{8Xn^{\mathrm{I,II}}(Z^2k^{\mathrm{I,II}} + Y)}{\{1 + Z^2[(n^{\mathrm{I,II}})^2 + (k^{\mathrm{I,II}})^2] + 2Xn^{\mathrm{I,II}} + 2Yk^{\mathrm{I,II}}\}^2}. \tag{25}$$

In equations (23)–(25), the superscripts I and II must be assumed simultaneously.

The uncertainties ΔR_2^{I} and ΔR_2^{II} reduce the effective width of the 'useful interval' for an unambiguous optical constant determination. This is shown pictorially in figure 4, with reference to the same practical example originally considered by Gupta [20] and then studied in [21]. In this figure, the reflectivity calculated starting from the wrong solution (that is R_2^{I} for $2.6 < \gamma < \pi$ and R_2^{II} for $\pi < \gamma < 5.9$) is compared with the

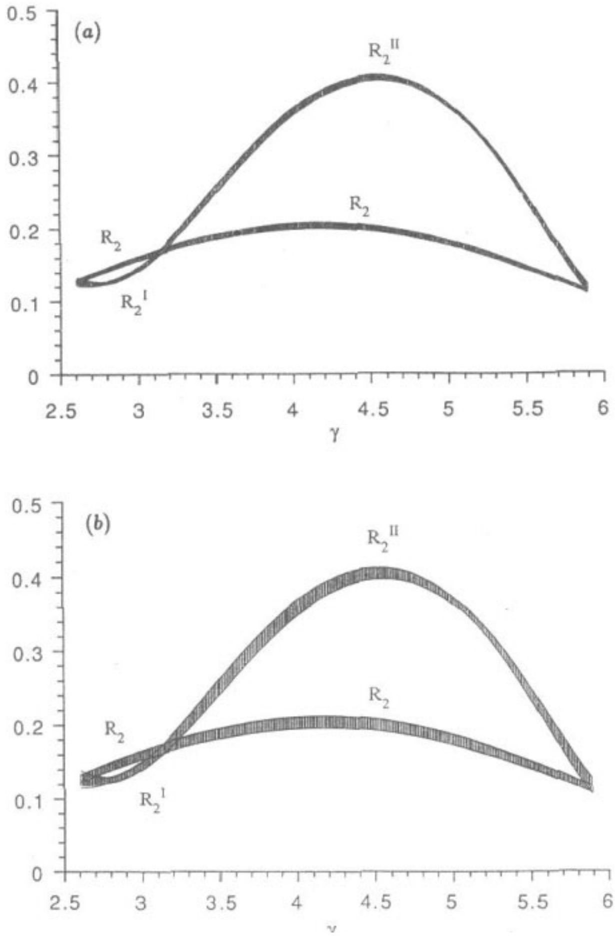


Figure 4. Comparison between the reflectivity calculated starting from the wrong solution (that is R_2^{I} for $2.6 < \gamma < \pi$ and R_2^{II} for $\pi < \gamma < 5.9$) together with its uncertainty and the measured reflectivity together with its uncertainty. The true values of the optical constants are $n = 1.61$ and $k = 0.42$; the real refractive index of the coating film is $n_2 = 1.95$. All the reflectivities are drawn as a function of the phase shift γ undergone by the reflected radiation in correspondence to a round trip through the coating material. The per cent errors in the reflectivities R_1 and R_2 are assumed equal to (*a*) 2% and (*b*) 4%. As a consequence of the measurement errors, the 'useful interval' narrows down to (*a*) $2.66 < \gamma < 5.87$ and (*b*) $2.71 < \gamma < 5.83$, while the 'indistinguishableness interval' enlarges up to (*a*) $3.09 < \gamma < 3.19$ and (*b*) $3.03 < \gamma < 3.24$.

measured reflectivity R_2. Both the reflectivities are drawn together with the uncertainty arising from the measurement errors. These amount to $\Delta R_1/R_1 = \Delta R_2/R_2 = 2\%$ in figure 4(*a*) and to $\Delta R_1/R_1 = \Delta R_2/R_2 = 4\%$ in figure 4(*b*). Due to the measurement errors, the wrong solution can be recognized and then rejected for an interval of γ values whose width decreases when $\Delta R_1/R_1$ and $\Delta R_2/R_2$ increase. This means that the 'useful interval' of γ values allowing an unambiguous determination of the optical constants gets narrower when the measurement errors increase. At the same time, the reflectivities are also indistinguishable for a γ interval around π, whose amplitude increases with the measurement errors. This effect is connected with the superposition of the error parallelograms pointed out in figures 2 and 3.

In the general case, the maximum displacement (towards the right) $\Delta\gamma_1$ of the left limit γ_1 of the 'useful interval' and the maximum displacement (towards the left) $\Delta\gamma_2$ of the right limit γ_2 of the same interval can be analytically evaluated to hold, respectively

$$\Delta\gamma_1 = \left|\frac{\partial\gamma_1}{\partial n^{\mathrm{II}}}\right| \Delta n^{\mathrm{II}} + \left|\frac{\partial\gamma_1}{\partial k^{\mathrm{II}}}\right| \Delta k^{\mathrm{II}} \tag{26}$$

$$\Delta\gamma_2 = \left|\frac{\partial\gamma_2}{\partial n^{\mathrm{I}}}\right| \Delta n^{\mathrm{I}} + \left|\frac{\partial\gamma_2}{\partial k^{\mathrm{I}}}\right| \Delta k^{\mathrm{I}} \tag{27}$$

where

$$\frac{\partial\gamma_1}{\partial n^{\mathrm{II}}} = 8n_2k^{\mathrm{II}}(1+n_2^2) \times \frac{n^{\mathrm{II}}(M^{\mathrm{II}}+P^{\mathrm{II}}+2R^{\mathrm{II}}) + (n^{\mathrm{II}}P^{\mathrm{II}}/R^{\mathrm{II}})(M^{\mathrm{II}}-P^{\mathrm{II}}) + (2n^{\mathrm{II}}/R^{\mathrm{II}})(P^{\mathrm{II}}-M^{\mathrm{II}})}{4n_2^2(k^{\mathrm{II}})^2(P^{\mathrm{II}}+R^{\mathrm{II}})^2 + (1+n_2^2)^2(M^{\mathrm{II}}+R^{\mathrm{II}})^2} \tag{28}$$

$$\frac{\partial\gamma_1}{\partial k^{\mathrm{II}}} = 4n_2(1+n_2^2)(P^{\mathrm{II}}+R^{\mathrm{II}})\frac{M^{\mathrm{II}}+R^{\mathrm{II}}+[2(k^{\mathrm{II}})^2/R^{\mathrm{II}}](M^{\mathrm{II}}-P^{\mathrm{II}})}{4n_2^2(k^{\mathrm{II}})^2(P^{\mathrm{II}}+R^{\mathrm{II}})^2 + (1+n_2^2)^2(M^{\mathrm{II}}+R^{\mathrm{II}})^2} \tag{29}$$

$$\frac{\partial\gamma_2}{\partial n^{\mathrm{I}}} = 8n_2k^{\mathrm{I}}(1+n_2^2)\frac{n^{\mathrm{I}}(M^{\mathrm{I}}+P^{I}-2R^{\mathrm{I}}) + (n^{\mathrm{I}}P^{\mathrm{I}}/R^{\mathrm{I}})(P^{\mathrm{I}}-M^{\mathrm{I}}) + (2n^{\mathrm{I}}/R^{\mathrm{I}})(M^{\mathrm{I}}-P^{\mathrm{I}})}{4n_2^2(k^{\mathrm{I}})^2(P^{\mathrm{I}}-R^{\mathrm{I}})^2 + (1+n_2^2)^2(M^{\mathrm{I}}-R^{\mathrm{I}})^2}. \tag{30}$$

$$\frac{\partial\gamma_2}{\partial k^{\mathrm{I}}} = 4n_2(1+n_2^2)(P^{\mathrm{I}}-R^{\mathrm{I}})\frac{M^{\mathrm{I}}-R^{\mathrm{I}}+[2(k^{\mathrm{I}})^2/R^{\mathrm{I}}](P^{\mathrm{I}}-M^{\mathrm{I}})}{4n_2^2(k^{\mathrm{I}})^2(P^{\mathrm{I}}-R^{\mathrm{I}})^2 + (1+n_2^2)^2(M^{\mathrm{I}}-R^{\mathrm{I}})^2}. \tag{31}$$

In equations (28)–(31), P^{I}, M^{I}, R^{I} and P^{II}, M^{II}, R^{II} are the expressions in equations (20), (21) and (22) calculated in correspondence to the solutions given by equations (5) and (6), respectively.

5. Concluding remarks

In this paper, the accuracy of the method originally proposed by Gupta for the determination of the optical constants of an absorbing material has been studied. The method is based on two measurements of the reflectivity of the material in the presence and absence of a transparent overcoat film. The unknown optical constants can be unambiguously determined provided that the optical thickness of the overcoat film is within a 'useful interval' of thicknesses. In the preceding sections, the experimental errors in the measured reflectivities have been demonstrated to influence both the accuracy with which the optical constants are determined and the amplitude of the 'useful interval' of thicknesses allowing the achievement of unambiguous results. In particular, the accuracy of the optical constant determination has been evaluated analytically through the propagation of the measurement errors and represented graphically through the construction of error parallelograms. The results show a decrease in the accuracy when the experimental errors increase and specify the optical thicknesses of the overcoat film in correspondence to which larger accuracies are obtained for given reflectivity errors. Furthermore, an optical thickness interval is also determined in correspondence to which the error parallelograms overlap and the two solutions obtained for the optical constants are indistinguishable. As expected, the amplitude of this interval turns out to be an increasing function of the measurement errors.

Finally, the limits of the 'practical useful interval', i.e. the 'useful interval' of thicknesses allowing an unambiguous determination of the optical constants in the presence of measurement errors, have been determined analytically. With reference to the numerical example originally proposed by Gupta, the limits of the 'practical useful interval' have also been determined through a graphical method. The results obtained emphasize the narrowing of the 'useful interval' when the experimental errors in the measured reflectivities increase and the rise inside it of an 'indistinguishableness interval', in correspondence to which the two solutions for the optical constants coincide (within the limits of the error uncertainty). The amplitude of this interval is shown to be an increasing function of the measurement errors.

It should be noted that, in principle, every kind of experimental error influences the accuracy with which the optical constants can be determined and then produces a narrowing of the useful interval we discussed in this paper. For instance, instead of referring, as we did, to the experimental errors in the measured reflectivities, we could refer to the uncertainty about the knowledge of the thickness and/or of the index of refraction of the transparent overcoating layer. However it is evident that the ambiguity problem is affected in the same way by any source of error so that the conclusions we draw in this paper can be referred to any kind of experimental error.

Appendix 1

Let us introduce the following notations for the modulus and the argument of the complex refractive index $n_1 = n - ik$

$$n_1 = N\exp(i\phi) \qquad N = (n^2 + k^2)^{1/2} \qquad \phi = \tan^{-1}(-k/n). \quad \text{(A1.1)}$$

Furthermore, let us indicate by X and Y the real and imaginary parts, respectively, of the complex number z as defined by equation (3), i.e. let us put

$$z = X - iY \qquad X = \frac{2}{1 + n_2^2 + (1 - n_2^2)\cos\gamma}$$
$$Y = \frac{(1 - n_2^2)\sin\gamma}{n_2[1 + n_2^2 + (1 - n_2^2)\cos\gamma]} \quad \text{(A1.2)}$$

and let us introduce the following notation for its modulus and argument

$$z = Z\exp(-i\theta) \qquad z = (X^2 + Y^2)^{1/2} \qquad \theta = \tan^{-1}(Y/X). \quad \text{(A1.3)}$$

By making use of equations (A1.1) and (A1.3), equations (1) and (2) can be alternatively written as

$$R_1 = \frac{1 + N^2 - 2N\cos\phi}{1 + N^2 + 2N\cos\phi} \quad \text{(A1.4)}$$

and

$$R_2 = \frac{1 + N^2Z^2 - 2NZ\cos(\phi - \theta)}{1 + N^2Z^2 + 2NZ\cos(\phi + \theta)} \tag{A1.5}$$

respectively.

Starting from equations (A1.4) and (A1.5), the following equations are obtained for the modulus, the real part and the imaginary part, respectively, of the unknown complex refractive index n_1

$$N^2(h_2Z^2 - h_1X) + (h_2 - h_1X) = -h_2Y[N^2(4 - 2h_1^2) - h_1^2N^4 - h_1^2]^{1/2}. \tag{A1.6}$$

$$2N\cos\phi = h_1(1 + N^2) \tag{A1.7}$$

$$2N\sin\phi = -[4N^2 - h_1^2(1 + N^2)^2]^{1/2}. \tag{A1.8}$$

where we put

$$h_1 = (1 - R_1)/(1 + R_1) \tag{A1.9}$$

$$h_2 = (1 - R_2)/(1 + R_2). \tag{A1.10}$$

By squaring both sides of equation (A1.8), the following equation of the second degree for N^2 is obtained

$$aN^4 + 2bN^2 + c = 0 \tag{A1.11}$$

where

$$a = (h_2Z^2 - h_1X)^2 + h_1^2h_2^2Y^2 \tag{A1.12}$$

$$b = (h_2 - h_1X)(h_2Z^2 - h_1X) - (2 - h_1^2)h_2^2Y^2 \tag{A1.13}$$

$$c = (h_2 - h_1X)^2 + h_1^2h_2^2Y^2. \tag{A1.14}$$

The two solutions of equation (A1.11) will be denoted by the superscripts I and II and the same will be done for the corresponding optical constants. From equation (A1.11), we have

$$(N^2)^{\mathrm{I}} = (1/a)[-b + (b^2 - ac)^{1/2}] \tag{A1.15}$$

$$(N^2)^{\mathrm{II}} = (1/a)[-b - (b^2 - ac)^{1/2}]. \tag{A1.16}$$

and from equations (A1.7) and (A1.8), we have

$$(n)^{\mathrm{I,II}} = \tfrac{1}{2}h_1(1 + (N^2)^{\mathrm{I,II}}) \tag{A1.17}$$

and

$$(k)^{\mathrm{I,II}} = \tfrac{1}{2}\{4(N^2)^{\mathrm{I,II}} - h_1^2[1 + (N^2)^{\mathrm{I,II}}]^2\}^{1/2} \tag{A1.18}$$

respectively. In equations (A1.17) and (A1.18), the superscripts I or II must be assumed simultaneously.

Appendix 2

In this appendix, we calculate the explicit expressions of some partial derivatives that are utilized in the paper.

From equation (A1.9) and (A1.10), we obtain

$$\frac{\mathrm{d}h_j}{\mathrm{d}R_j} = \frac{-2}{(1+R_j)^2} \qquad \text{for } j = 1, 2. \tag{A2.1}$$

From equations (A1.12), (A1.13) and (A1.14), we obtain respectively

$$\frac{\partial a}{\partial R_1} = [-2X(h_2Z^2 - h_1X) + 2h_1h_2^2Y^2]\frac{\mathrm{d}h_1}{\mathrm{d}R_1} \tag{A2.2}$$

$$\frac{\partial a}{\partial R_2} = [2Z^2(h_2Z^2 - h_1X) + 2h_1^2h_2Y^2]\frac{\mathrm{d}h_2}{\mathrm{d}R_2} \tag{A2.3}$$

$$\frac{\partial b}{\partial R_1} = \{X[2h_1X - h_2(1+Z^2)] + 2h_1h_2^2Y^2\}\frac{\mathrm{d}h_1}{\mathrm{d}R_1} \tag{A2.4}$$

$$\frac{\partial b}{\partial R_2} = [2h_2Z^2 - h_1X(1+Z^2) - 2h_2(2-h_1^2)Y^2]\frac{\mathrm{d}h_2}{\mathrm{d}R_2} \tag{A2.5}$$

$$\frac{\partial c}{\partial R_1} = [-2X(h_2 - h_1X) + 2h_1h_2^2Y^2]\frac{\mathrm{d}h_1}{\mathrm{d}R_1} \tag{A2.6}$$

$$\frac{\partial c}{\partial R_2} = [2(h_2 - h_1X) + 2h_1^2h_2Y^2]\frac{\mathrm{d}h_2}{\mathrm{d}R_2}. \tag{A2.7}$$

From equations (A1.15) and (A1.16), we obtain respectively

$$\frac{\partial(N^2)^{\mathrm{II}}}{\partial R_j} = \{[-ac + 2b^2 + 2b(b^2-ac)^{1/2}](\partial a/\partial R_j) + [-2ab - 2a(b^2-ac)^{1/2}] \times (\partial b/\partial R_j) - a^2(\partial c/\partial R_j)\}[2a^2(b^2-ac)^{1/2}]^{-1} \tag{A2.8}$$

$$\frac{\partial(N^2)^{\mathrm{II}}}{\partial R_j} = \{[-ac + 2b^2 + 2b(b^2-ac)^{1/2}](\partial a/\partial R_j) + [-2ab - 2a(b^2-ac)^{1/2}] \times (\partial b/\partial R_j) + a^2(\partial c/\partial R_j)][2a^2(b^2-ac)^{1/2}]^{-1} \tag{A2.9}$$

where $j = 1, 2$.

Finally, from equations (A1.17) and (A1.18), we obtain respectively

$$\frac{\partial n^{\mathrm{I,II}}}{\partial R_1} = \frac{\partial h_1}{\partial R_1}\frac{1+(N^2)^{\mathrm{I,II}}}{2} + \frac{h_1}{2}\frac{\partial(N^2)^{\mathrm{I,II}}}{\partial R_1} \tag{A2.10}$$

$$\frac{\partial n^{\mathrm{I,II}}}{\partial R_2} = \frac{h_1}{2}\frac{\partial(N^2)^{\mathrm{I,II}}}{\partial R_2} \tag{A2.11}$$

$$\frac{\partial k^{\mathrm{I,II}}}{\partial R_1} = \frac{\{2 - h_1^2[1+(N^2)^{\mathrm{I,II}}]\}(\partial(N^2)^{\mathrm{I,II}}/\partial R_1) - h_1[1+(N^2)^{\mathrm{I,II}}]^2\,\partial h_1/\partial R_1}{2\{4(N^2)^{\mathrm{I,II}} - h_1^2[1+(N^2)^{\mathrm{I,II}}]^2\}^{1/2}} \tag{A2.12}$$

$$\frac{\partial k^{\mathrm{I,II}}}{\partial R_2} = \frac{\{2 - h_1^2[1 + (N^2)^{\mathrm{I,II}}]\}[\partial (N^2)^{\mathrm{I,II}}/\partial R_2]}{2[4(N^2)^{\mathrm{I,II}} - h_1^2[1 + (N^2)^{\mathrm{I,II}}]^2\}^{1/2}}. \tag{A2.13}$$

In equations from (A2.10)–A2.13), the superscripts I or II must be assumed simultaneously.

References

[1] Bennett J M and Booty M J 1966 *Appl. Opt.* **5** 41–3
[2] Abelès F and Thèye M L 1966 *Surf. Sci.* **5** 325–31
[3] Nilsson P O 1968 *Appl. Opt.* **7** 435–42
[4] Hjortsberg A 1981 *Appl. Opt.* **20** 1254–63
[5] Case W E 1983 *Appl. Opt.* **22** 1832–6
[6] Dobrowolski J A, Ho F C and Waldorf A 1983 *Appl. Opt.* **22** 3191–200
[7] McPhedran R C, McKenzie D R, Botten L C and Netterfield R P 1984 *Appl. Opt.* **23** 1197–205
[8] Leupacher W and Penzkofer A 1984 *Appl. Opt.* **23** 1154–8
[9] Paulick T C 1986 *Appl. Opt.* **25** 562–4
[10] Engelsrath A and Loewenstein E V 1966 *Appl. Opt.* **5** 565–7
[11] Kolb D M 1972 *J. Opt. Soc. Am.* **62** 599–600
[12] Hunderi O, 1972 *Appl. Opt.* **11** 1572–8
[13] Hunter W R 1974 *J. Opt. Soc. Am.* **64** 429–33
[14] Querry M R and Holland W E 1974 *Appl. Opt.* **13** 595–8
[15] Beauchamp W T and Rancourt J D 1980 *Appl. Opt.* **18** 3239–44
[16] Azzam R M A 1982 *J. Opt. Soc. Am.* **72** 1439–40
[17] Singleton E B and Shirkey C T 1983 *Appl. Opt.* **22** 185–9
[18] Nagendra C L and Thutupalli G K M 1983 *Appl. Opt.* **22** 587–91
[19] Lu Y and Penzkofer A 1986 *Appl. Opt.* **25** 221–5
[20] Gupta M C 1988 *Appl. Opt.* **27** 954–956
[21] Gori F and Guattari G 1991 *Appl. Opt.* **30** 36–41
[22] Born M and Wolf M 1980 *Principles of Optics* (New York: Pergamon)
[23] Ward L 1988 *The Optical Constants of Bulk Materials and Films* (Bristol: Hilger)