Computer Program for Determining Optical Constants of a Film on an Opaque Substrate

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FOREWORD

The development and continued improvement of target-acquisition and missile-guidance systems for Fleet use are areas of special interest to the Navy. In support of these interests, the Naval Weapons Center (NWC) is conducting research projects in the fields of infrared and ultraviolet detection, solid state physics, and optics. To provide better optical target-acquisition and guidance systems, improvements are required either in systems sensitivity or in operating wavelength range. NWC's research efforts have been directed toward the production of improved detectors and optical elements.

One of the principal efforts in the optics field has been the study of corrosion films on metal surfaces. Since silver has the highest reflectance of any metal in the visible and infrared spectrum, the tarnish layer that forms on silver surfaces is of special concern. During studies conducted at NWC to investigate the growth of silver sulfide tarnish films on silver, it became necessary to obtain the optical constants of the tarnish films. However, since the films could not be removed from the substrate, conventional methods used to determine these constants were not applicable. Consequently, a computer program was developed to determine the optical constants of films on opaque substrates by using input data obtained from normal incidence reflectance measurements on two films of different thicknesses. This computer program is described in the included reprint of an article published in Applied Optics, Vol. 8, No. 11 (November 1969), pp. 2366-68.

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Technical Director
Computer Program for Determining Optical Constants of a Film on an Opaque Substrate

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In the course of studying the growth of silver sulfide tarnish films on silver,\textsuperscript{1,2,3} we needed to know their optical constants, \(n, k\). The films were under 100-Å thick and could not be removed from the silver, so that we were unable to use the method of determining \(n\) and \(k\) from the measured reflectance \(R\), transmittance \(T\), and film thickness \(t\).\textsuperscript{4} We thus decided to try to obtain the optical constants from normal incidence reflectance measurements on two silver sulfide films of different thicknesses on silver where the sulfide thicknesses were measured ellipsometrically.\textsuperscript{1} This involved an addition to our multilayer film computer program which will be described in this letter.

The basic multilayer film program\textsuperscript{5} calculates reflectance, transmittance, and phase change on reflection of a substrate covered with one or more films of known thickness and optical constants, when the optical constants of the substrate are also supplied. This program was previously modified to obtain \(n\) and \(k\) for a film given the refractive index of the (transparent) substrate, film thickness, normal incidence front surface reflectance, and transmittance of the film on the substrate.\textsuperscript{6} The program described in this letter is independent of the \(R, T, t\) modification, although it uses some of the same logic and iterative techniques.

The addition to the multilayer program which is required for the new calculational method is shown in the flow diagram in Fig. 1. The reflectance \(R_s\) of the opaque substrate is first calculated using assumed optical constants for the substrate material. For silver, the solution is not very sensitive to the values chosen.

Trial values of \(n\) and \(k\) for the film are then taken along with the film thickness \(t\) and the reflectance \(R_f\) is calculated for the film on the opaque substrate. \(R_s\) is subtracted from \(R_f\) and a quantity called \(\Delta H_f\) is obtained. A second quantity, \(\Delta H_o\), for a film of thickness \(t\) is obtained in an analogous manner. These \(\Delta H_f\)'s should be directly comparable with the measured quantities \(\Delta H_s\) and \(\Delta H_o\) obtained by subtracting the measured reflectance of a film-covered opaque substrate from the reflectance of the film-free substrate.

The major portion of the new program consists of changing \(n\) and \(k\) for the films in such a way that \(\Delta H_f - \Delta H_o = \Delta H_p\). Since there are two measured quantities, \(\Delta H_s\) and \(\Delta H_o\), and two unknowns, \(n\) and \(k\) for the films, in principle a unique solution should exist if both films have the same optical constants. (In practice there are experimental difficulties which prevent
unique solutions from being obtained; these will be discussed later.) The detailed manner in which the search routine proceeds can be summarized as follows. From the trial values of \( a \) and \( k \) for the films, a range of values of \( n \) is selected centering on the trial \( n \) value. With \( k \) held fixed, \( \triangle R_0 \) is calculated for all the \( n \) values selected, and the \( n \) saved which gives a \( \triangle R_0 \), closest to \( \triangle R_0 \) (i.e., smallest \( \triangle D R_0 \) — see Fig. 1). Using this \( n \), the trial value of \( k \) is now varied over a selected range and the \( n \) and \( k \) giving the smallest \( \triangle D R_0 \) is retained. Using this new pair of \( n \) and \( k \) values, the program shifts to the second film of thickness \( h \) and repeats the process, obtaining a different pair of \( n \) and \( k \) values which yield the smallest \( \triangle D R_0 \). The preceding process is called cycle I. Cycle II consists of a repeat of cycle I starting with the just-determined \( n \) and \( k \), only letting the allowed ranges of values of \( n \) and \( k \) be exactly half the allowed ranges for cycle I. Cycle III is a repeat of cycle II with the allowed ranges again cut in half. If the allowed ranges of \( n \)'s and \( k \)'s are properly chosen, the program should converge on a single pair of values which satisfy the conditions \( \triangle R_0 = \triangle R_0 \), and \( \triangle R_0 = \triangle R_0 \) (i.e., \( \triangle D R_0 = \triangle D R_0 = 0 \)). However, if the range of \( n \)'s is too large, \( \triangle R_0 \) can be made equal to \( \triangle R_0 \), by simply changing \( n \) (\( k \) not required to change). In this situation, when going to \( \triangle R_0 \), \( n \) may change to a different value, still without requiring \( k \) to change. In cycles II and III, the major portions of the changes may again be made by \( n \), so that there appear to be two discrete values of \( n \) corresponding to film thicknesses \( h \) and \( h \). If this situation occurs, the allowed range of \( n \) should be made smaller so that \( k \) will also have to change. If discrete solutions for \( n \) are found for the two films even though \( k \) is allowed to change, the two films may actually have different optical constants.

If \( \triangle R_0 \) and \( \triangle R_0 \) are exact and the two films have the same optical constants, the calculated \( n \) and \( k \) values will be unique. However, if the \( \triangle R_0 \)'s contain experimental measuring errors, a series of solutions for \( n \) and \( k \) will be obtained where the range of values will depend on the percentage uncertainty in the \( \triangle R_0 \)'s. As an example, consider the following problem. For two films of silver sulfide on silver (thicknesses 22.5 \( \text{Å} \) and 38 \( \text{Å} \), respectively) we have measured values of \( \triangle R_0 \) and \( \triangle R_0 \) which we will use as inputs. [For the purpose of this problem, the so-called mea-

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**Fig. 1.** Flow diagram for addition to multi-layer film program which determines optical constants of a film on an opaque substrate.

**Fig. 2.** Range of \( n \) and \( k \) values which give \( \triangle R_0 \)'s within 0.001 of the input values for films of thicknesses 22.5 \( \text{Å} \) and 38 \( \text{Å} \), respectively. The short-dashed \( n \) curve goes with the short-dashed \( k \) curve and the long-dashed \( n \) and \( k \) curves similarly go together.
...sure quantities were actually calculated from the thin-film optical constants in Ref 4. After choosing a set of trial values of $n$ and $k$ for the silver sulfide films and using the computer program, we obtain the values of $n$ and $k$ shown as the solid lines in Fig. 2. However, our measured values, $\Delta n_1$ and $\Delta n_2$, are not exact, but each contains a measuring uncertainty of $\pm 0.001$. To find the upper limit of $n$ values which can be a solution, we choose a value of $n$ larger than that on the solid line and restrict it to a very narrow range of values. We then use the program to find a single $k$ value which gives the smallest $\Delta n_1$ and $\Delta n_2$. If these differences are less than $0.001$ in absolute value, we have a solution. However, we want the $n$ value where both $\Delta n_1$ and $\Delta n_2$ are close to $0.001$, so a larger $n$ is chosen until one is found where they are. If either $\Delta n_1$ or $\Delta n_2$ is larger than $0.001$, we have chosen our initial $n$ too large and must reduce it. We can similarly find the lower limit of $n$ values, using $n$'s smaller than the numbers on the solid curve. Results of these calculations are shown as the shaded area in Fig. 2. In this figure the short-dashed $n$ curve and the short-dashed $k$ curve are solutions for which the $\Delta n$'s are exactly $\pm 0.001$ different from the original values; the long-dashed $n$ and $k$ curves are also solutions satisfying this requirement. Although these curves are strictly representative only of the particular example of silver sulfide on...
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