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Optical losses of sputtered Ta$_2$O$_5$ films

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1. Introduction.

Optical losses in dielectric thin films are composed of portions originating from the various layer volumes as well as from their interfaces. A detailed knowledge of these loss mechanisms is necessary for choosing appropriate film design, materials, and deposition techniques to achieve high quality optical coatings. Scattering losses of thin film optical coatings are mainly caused by surface and interface micro-irregularities and, hence, vector scattering theories can be used to predict both angular scattering and total integrated scattering [1, 2]. For dielectric single layers it has recently been shown that light scattering markedly depends on the cross-correlation properties of the rough interfaces [3, 4]. Moreover, the results reported there with respect to the total integrated backscattering of quarterwave and halfwave layers reveal the fundamental influence of the layer thickness chosen. Thus, scattering investigations of films with optical thickness varying in a determined manner, such as wedge-shaped films seem to be promising.

Surface and interface absorption play an important role in optical thin films. When using samples suitably prepared, both volume and interface parts of the total absorption become separable. As an example, the influence of interface absorption on the quality of thin films has been investigated in a relatively simple manner using $\lambda/2$ MgF$_2$-SiO$_2$ multilayer systems [5]. These systems are of fixed optical thickness, but consist of a different number of sublayers. For single films deposited onto glass substrates, however, a more complete set of measurements is required in order to separate different parts of absorption.

Following a method first suggested by Temple [6] the absorption of wedge-shaped single-layer films should be measured by making use of the characteristic changes of the relative power density at the air-film and film-substrate interface for quarterwave and halfwave optical thickness, respectively; i.e., not only for scattering but also for absorption measurements wedge-shaped samples are advantageous. The aim of our paper is to investigate both absorption and total integrated backscattering (TIS) of wedge-shaped Ta$_2$O$_5$ single layers. Theoretical predictions as well as the corresponding experimental results of scattering and absorption measurements are presented and compared in order to get a deeper insight into the main loss mechanisms of these layers.

2. Sample preparation.

Conventionally polished disk shaped BK 7 substrates were carefully cleaned and coated with a wedge-shaped Ta$_2$O$_5$ film by reactive dc-magnetron sputtering. The sputtering parameters were the same as described in detail in [7]. Starting at the bare substrate, films were obtained with an approximately linearly increasing geometrical thickness up to ~500 nm. The refractive index was estimated by a method described in [8] to be about 2.1 with respect
to the wavelengths used in the scattering and absorption measurements.

3. Scattering losses.

3.1 Theory. — In our investigations we will consider the optical thickness dependence of the total integrated backscattering (TIS). As following from vector scattering theory [1, 2], the TIS of a dielectric single layer on a microrough substrate is given by the expression

$$\text{TIS} = \int_0^{2\pi} \int_0^{\pi/2} \left( F_{11} \theta_{11} + F_{12} \theta_{12} + F_{22} \theta_{22} \right) \times \sin \theta \, d\theta \, d\varphi \, . \quad (1)$$

The indices 1 and 2 correspond to the substrate-film and film-air interface, respectively. $\theta$ and $\varphi$ are the polar and azimuthal angles of scattering. The factors $F_{ij}$ include only parameters of the ideal smooth layer (refractive indices, film thickness) as well as the wavelength and the angles of illumination, observation and polarization. $\theta_{ij}$ are the power spectral densities, representing the Fourier transform of roughness correlation functions $G_{ij}$ of the interfaces. If $i = j$, $G_{ij}$ is referred as autocorrelation function, else it is signified as cross-correlation function. For isotropic roughness the $G_{ij}$ are defined as

$$G_{ij}(r) = \langle \xi_i(r) \xi_j(r + \tau) \rangle \, , \quad (2)$$

where $\xi_i(r)$ symbolizes the microroughness height measured from the mean surface level of the $i$-th interface, $r$ is the value of the position vector $r$ in the plane of the surface, $\tau$ is the correlation distance, and $\langle \cdot \rangle$ denotes ensemble averaging.

Obviously, there exist two limiting cases of correlation:

1. Both interfaces are fully correlated, i.e., the substrate profile is identically reproduced by the film. Then we are given

$$G_{11} = G_{12} = G_{22} \, . \quad (3)$$

2. The interfaces are completely uncorrelated, that means the cross-correlation vanished

$$G_{12} = 0 \, , \quad (4)$$

and, for simplicity, the autocorrelation functions may be assumed to be identical, $G_{11} = G_{22}$ [9].

Concerning these cases, from equation (1) the TIS was calculated for a wedge-shaped Ta$_2$O$_5$ film ($n = 2.1$) on a BK 7 substrate ($n_s = 1.52$) assuming a Gaussian correlation function with a 400 nm correlation length. It should be emphasized that changing the correlation length from 400 nm up to several micrometers do not affect the results noticeably. Further, rms roughness was eliminated from the consideration relating the film scattering (TIS) to that of the bare substrate (TISO).

Figure 1 shows the TIS-ratio as a function of the optical thickness $nd$. Both perfect and vanishing cross-correlation cause the scattering loss to change periodically, exhibiting extreme values at $nd = p\lambda/4$ ($\lambda = 633$ nm is the illumination wavelength, $p$ is an integer). However, in the case of full correlation maxima appear for $nd$ equals odd multiples of $\lambda/4$ and, vice versa, minima appear at even multiples of $\lambda/4$. Just, the opposite behaviour is yielded for zero cross-correlation. This general behaviour described above is typical for high index films and independent of the special parameters. On the other hand, the certain value of TIS/TISO at the quarter-wave and halfwave positions is strongly determined by the refractive indices of the layer and the substrate. With the parameters mentioned above for $nd = (2p - 1)\lambda/4$ we are yielded roughly 6 in the correlated and 3 in the uncorrelated case. The corresponding values for $nd = p\lambda/2$ are 1 and 9, respectively.

![Fig. 1. — Calculated TIS/TISO-ratio as a function of optical thickness $nd$ for a layer of refractive index 2.1 on BK 7 substrate. Full line: fully correlated interface roughness; upper dashed line: uncorrelated interface roughness. The lower dashed line markes the TISO-level.](image-url)
Fig. 2. — Apparatus used for TIS measurements. (1) He-Ne laser source; (2) chopper; (3) sample; (4) sample shifting system; (5) Coblentz sphere; (6) scattered light detector; (7) lock-in amplifier; (8) specularly reflected light detector; (9) x-y-recorder.

The measurements were performed at nearly normal incidence of the laser beam while shifting the sample perpendicularly to the beam in order to change the effective optical thickness. The current optical thickness was determined from simultaneous measurement of the specularly reflected light.

In figure 3 a typical result of TIS measured on wedge-shaped Ta$_2$O$_5$ films is shown. The scattering level depends on the film thickness in a similar manner as calculated theoretically for fully correlated interface roughness: the TIS exhibits a pronounced periodicity with maxima at $nd = (2p-1)\lambda/4$ and minima at $nd = p\lambda/2$. However, the TIS/TIS$_0$-values noted in table I with respect to the extreme levels reveal that they are somewhat different from those predicted in the case of full correlation. On the one hand, the TIS/TIS$_0$-ratio experimentally observed is lower at the quarterwave positions, on the other hand it exceeds the calculated ratio at the halfwave points. Hence, the interface roughness is to be characterized as being predominantly but not fully correlated.

Finally, let us remember the assumption of negligible film volume scattering, implicitly involved in the theoretical treatment in section 3.1. We will ask for whether the experiment confirms this assumption or not. Obviously, if volume scattering contributed substantially to the scattering, a continuously increasing TIS level with increasing film thickness, superimposed on the periodically changing level would have been expected [11]. As following from figure 3 this is not to be observed. Note that the effect of slightly increasing TIS at $nd = (2p-1)\lambda/4$ for increasing $p$ while not exhibiting this behaviour at $nd = p\lambda/2$ results from the theory of interface scattering as can be seen in figure 1.

### 4. Absorption losses.

#### 4.1 Calculation of volume and interface absorption.

To investigate the absorption of single-layer films deposited onto glass substrates we have to distinguish between four regions where optical absorption originates: the air-film interface ($af$), the volume of the film ($f$), the film-substrate interface ($fs$), and the bulk of the substrate ($s$). Hence, the measured total absorption $A$ of the sample investigated consists of an air-film as well as a film-substrate interface term and volume absorption of the film as well as bulk absorption of the substrate [6]:

$$A = A_{af} + A_{f} + A_{fs} + A_{s}$$

$$= p_{af} a_{af} + \bar{a}_{f} d_f + p_{fs} a_{fs} + \bar{a}_{s} d_s$$

where $a_{af}$ and $a_{fs}$ represent the specific absorption at the air-film and film-substrate interface defined as the ratio of power absorbed at the interfaces to light power at the interface; $\bar{a}_{f}$ and $\bar{a}_{s}$ are the spatially averaged film and substrate absorption coefficients, respectively, and $d_f$ and $d_s$ are the geometrical thicknesses of the film and the substrate. Finally, $p_{af}$ and $p_{fs}$ denote the relative light power densities at
the corresponding film interfaces, and \( p_f \) and \( p_s \) are the relative light power densities within the corresponding media defined as spatially averaged over film and substrate volume, respectively [7].

Performing a photoacoustic absorption measurement we obtain a measuring signal caused by optical absorption. For optical and thermally thin layers onto non-absorbing substrates the photoacoustic signal is shown to be proportional to the optical absorption, i.e., proportional to the sum of the interface and volume absorption components of a single layer [7]. To separate the individual \( \alpha_f \), \( a_{ft} \), and \( a_{sf} \) wedge-shaped films have been used. In dependence on the light beam position on the layer, and, hence, on the actual film thickness, the relative power densities at both interfaces and within the film volume are all changing in a characteristic pattern, ranging from a quarterwave to a halfwave optical thickness, labelled by the indices (1) and (2), respectively. Thus, the film volume as well as interface parameters become calculable, see references [6] and [7].

4.2 EXPERIMENTAL PROCEDURE AND RESULTS. — The measurements were performed at a wavelength of \( \lambda = 515 \) nm in an experimental set-up schematically shown in figure 4 and described in detail elsewhere [12]. The optical absorption was detected by a photoacoustic gas cell-microphone set-up consisting of an argon-ion laser as light source (1), rotating disk beam chopper (2), PAA cell with measuring condensor microphone (3) and lock-in amplifier (4). A sensitivity of about 0.1 V per W absorbed power at 200 Hz was achieved connected with a noise equivalent of power of about 0.5 \( \mu \)W. In this manner accurate measurements of both amplitude and phase of weak signals were accomplished.

PAA measurements on wedge-shaped \( \text{Ta}_2\text{O}_5 \) films were carried out in \( \lambda /4 \) steps. In figure 5 the result is shown for the same sample which has been viewed in section 3.2. Obviously, the absorption \( A \) measured increases with increasing optical thickness. The averaged volume absorption coefficient \( \bar{\alpha}_f \) and the calculated specific interface parameters \( a_{ft} \) and \( a_{ft} \) are presented in table II. By comparing these values with those obtained in our preceding measurements in [7] it is evident that the volume absorption coefficient \( \bar{\alpha}_f \) and the air-film absorption \( a_{ft} \) are nearly unchanged, whereas the film-substrate interface absorption \( a_{sf} \) differs remarkably; i.e., interface absorption strongly diminishes. Moreover, the total interface absorption is lowered of about one order of magnitude. This result is explained by taking into consideration the procedure of substrate cleaning prior to the coating process. Concerning the sample presented in this paper, a special cleaning method connected with detailed inspection and extreme careful handling was applied to the substrate, whereas only a routine procedure was performed in [7]. As it has been known for a long time, the substrate condition can act as a crucial factor in thin film preparation.

5. Summarizing remarks.

For sputtered \( \text{Ta}_2\text{O}_5 \) films the scattering losses measured at \( \lambda = 633 \) nm as well as the absorption losses at \( \lambda = 515 \) nm were found to be in the range...
of some $10^{-5}$ to $\sim 10^{-4}$, considerably depending on the actual film thickness. From a comparison of the experimentally obtained results with those given by the theoretical predictions we concluded that the scattering arose from interface roughness rather than from volume effects, and a high cross-correlation between the rough interfaces was deduced.

In contrast to these findings a great part of the absorption losses originate from the volume of the film. Additionally, a contribution to the total absorption loss is due to the film-air interface, whereas a nearly zero contribution from the substrate-film interface was discovered. The pronounced distinction concerning the predominant origin of scattering and absorption losses, respectively, may be explained as follows: scattering is induced by the interfaces via the geometrical effect of microroughness, while interface absorption mainly arises from contaminations, critically depending on the substrate cleaning procedure, and therefore, zero substrate-film interface absorption corresponds to a very clean substrate surface.

Finally, the volume absorption obtained is explained by a slight non-stoichiometry, as discussed for magnetron sputtered tantalum oxide films in [13].

References