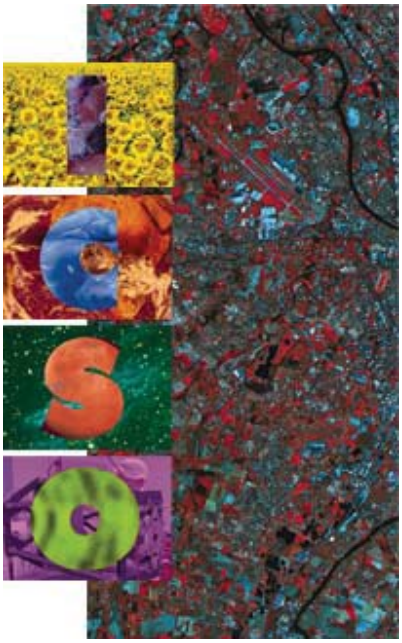
International Conference on Space Optics—ICSO 2000

Toulouse Labège, France

5–7 December 2000

Edited by George Otrio



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International Conference on Space Optics — ICSO 2000, edited by George Otrio, Proc. of SPIE Vol. 10569
105691C · © 2000 ESA and CNES · CCC code: 0277-786X/17/$18 · doi: 10.1117/12.2307934

ANGULAR SCATTERING AND ELLIPSOMETRY OF THE SCATTERED FIELD: MULTISCALE ROUGHNESS AND CONTAMINATION OF SURFACES

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RESUME - La technique de diffusion lumineuse, développée depuis plus de vingt ans au LOSCM, permet de caractériser les rugosités de surface et les hétérogénéités de volume. Des mesures réalisées avec des longueurs d'onde variant depuis l'UV jusqu'au moyen IR, complétées par des mesures par Microscopie à Force Atomique, permettent de caractériser de façon multiéchelle les composants. Cette technique a été encore améliorée grâce à son adaptation pour des mesures d'ellipsométrie sur flux diffus, qui permettent d'étudier le paramètre "déphasage polarimétrique" pour compléter les mesures classiques d'intensité. Ces mesures ont alors permis de séparer de manière non destructive les effets de diffusion de surface et de volume. Le déphasage polarimétrique s'est également avéré comme extrêmement sensible à l'état de contamination des composants.

ABSTRACT *- Light scattering is well known to be dependent on the optical properties and surface roughness or bulk inhomogeneities of components* ***[1, 2]****. Angular scattering measurements and the development of electromagnetic theories at Institut Fresnel Marseilles permit to quantify the roughness behaviour. Measurements can be performed at different wavelengths from the UV to the near IR to access to different scales of characterization. Atomic Force Microscopy is used to complete these measurements at the microscopic scale, and predict the surface behaviour in the X rays domain. All these techniques rise to a multiscale characterization of all surfaces, which reveals in most cases fractal behaviours* ***[3]****. The scatterometer has been extended and allows to perform ellipsometric measurements on scattered light in each direction of space* ***[4]****. Results can be investigated by electromagnetic theories. They permit to directly separate bulk and surface effects in the case of bare substrates and reveal the high sensitivity of the polarimetric phase difference to the presence of contaminants on surfaces, even in the case of first order contaminants, that is to say whose size is in the same order as the substrate roughness.*

1. INTRODUCTION

Light scattering is known to be dependent on the optical properties and surface roughness or bulk inhomogeneities of components **[1, 2]**. In a first part, we will show how angular scattering measurements at wavelengths varying from the UV to the IR and the development of electromagnetic theories at Institut Fresnel Marseilles permit to quantify the multiscale roughness of components. In a second step, we show how the scatterometer has been

extended to allow ellipsometric measurements of the scattered light in each direction of space [4]. Results can be investigated by electromagnetic theories and permit to directly separate bulk and surface effects in the case of bare substrates and to reveal the high sensitivity of the polarimetric phase difference to the presence of contaminants on surfaces, even in the case of first order contaminants, that is to say whose size is in the same order as the substrate roughness.

2. EXPERIMENTAL SETUP

An angular scatterometer has been developped at L.O.S.C.M (Laboratoire d'optique des surfaces et des couches minces), now integrated in Institut Fresnel, since several years. This experimental set-up is represented on Figure 1. Measurements are usually performed in the incidence plane. The sample can move in its plane: two translation directions can be used, and one rotation. The incidence angle can be changed. In order to perform multi-scale studies, we can use several sources at different wavelengths varying from the UV (325 nm) to the IR (10.6 µm). Different detectors are used depending on the wavelength. The dynamic is 7 decades in the visible and 5 decades in the IR. The calibration of light scattering measurements are performed thanks to the use of a commercialized lambertian sample. All the motions are motorized (10 motorized axes) and controlled with a computer.

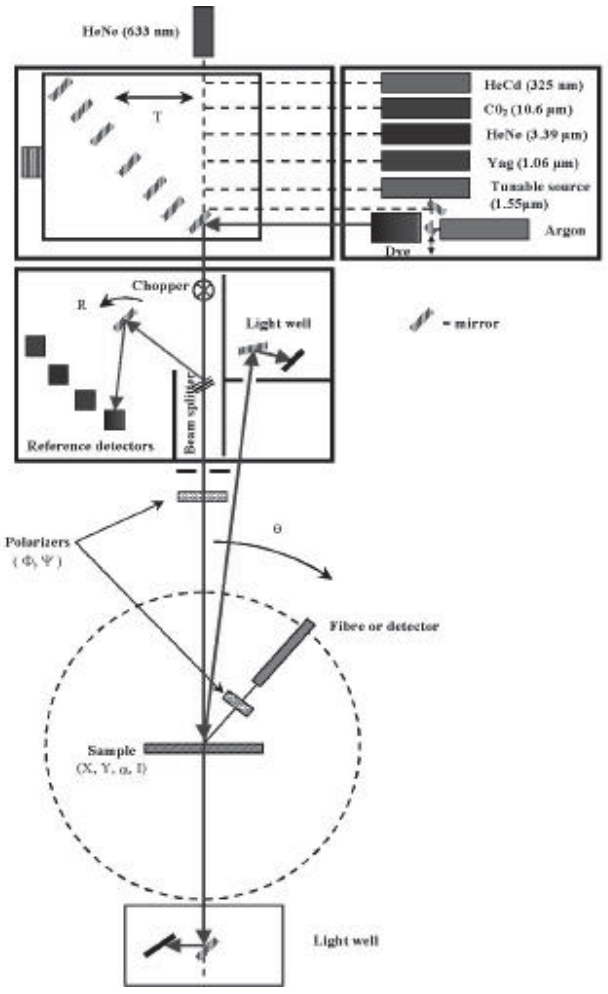


Figure 1: Experimental set-up at Institut Fresnel – Marseilles

This experimental set-up can be used in a classical configuration, as described before. Multi-scale studies have been performed, and have been completed with Atomic Force Microscopy measurements at the microscopic scale. These techniques will be presented in the next paragraph. The great accordance between both techniques proves the great calibration of our experimental set-up. Moreover, anisotropy characterizations can be performed. Polarization effects can be investigated thanks to the use of several rotating polarizers on the incident and the scattered beam. In this configuration, ellipsometry measurements of the scattered beam can be performed, as presented in paragraph 4.

3. MULTISCALE ROUGHNESS

The origin of light scattering can be the surface or the bulk of componants. Many studies have been performed to separate the effects of surface or bulk scattering [2]. In the case of a surface scattering, numerical calculations have been performed at Institut Fresnel-Marseilles. The developpement of first-order electromagnetical theories rises to an expression of I, the scattered Intensity in the direction (θ, ϕ) of the incidence plane, where θ is the scattering angle reported to the sample normal in the incidence plane, and ϕ the polar angle. We can write the *Bidirectional Reflectance Distribution Function* (BRDF) as a function of I: $I = BRDF.\cos\theta$.

$$I(\theta,\phi) = C(\theta,\phi)\gamma(\theta,\phi) \tag{3.1}$$

The optical factor $C(\theta,\phi)$ is calcultated with the electromagnetical theory. It doesn't depend on the surface profile. The term $\gamma(\theta,\phi)$ is for the roughness spectrum of the surface. It is defined as:

$$\gamma(\vec{\sigma}) = \frac{4\pi^2}{S}\left|\hat{h}(\vec{\sigma})\right|^2 \tag{3.2}$$

The roughness δ, or root mean squared, is calculated with the roughness spectrum:

$$\delta^2 = \int_{\vec{\sigma}} \gamma(\vec{\sigma})\, d\vec{\sigma} \tag{3.3}$$

In this expression, $\vec{\sigma} = 2\pi n/\lambda \sin\theta(\cos\phi, \sin\phi)$ is the spatial pulsation of the propagated waves.

The roughness can be written as a function of the scattering angle: $\sigma = 2\pi/\lambda \sin\theta$.

Let's notice that the roughness depends on the experimental conditions, particularly on the wavelength. The frequency bandwidth that can be reached during experiments is given by:

$$B(\lambda, \theta_{min}) = \left[2\pi\frac{\sin\theta_{min}}{\lambda}; 2\pi\frac{1}{\lambda}\right] \tag{3.4}$$

The use of different laser sources on the experimental set-up permits to increase this frequency bandwidth.
Several experimental results have proved that the roughness spectra obtained by light scattering measurements at different wavelengths varying from the UV to the IR overlap perfectly at the intersection of these frequency bandwidths.

In order to increase this frequency domain, light scattering measurements can be completed with Atomic Force Microscopy Measurements. This technique rises to a spatial

sampling of the measured surfaces. In this case, the roughness spectrum can be calculated from a discrete profile:

$$\gamma_{pq} = \gamma\left(p\Delta\sigma_X, q\Delta\sigma_Y\right) = \left(\frac{2\pi}{L}\right)^2 \left|\hat{h}\left(p\Delta\sigma_X, q\Delta\sigma_Y\right)\right|^2 \tag{3.5}$$

where $\Delta\sigma_X = \Delta\sigma_Y$ is the sampling spectral step. In adequation with the Shannon theorem, we choose:

$$\Delta\sigma_X = \Delta\sigma_Y = \frac{\pi}{N\Delta x} = \frac{\pi}{L}. \tag{3.6}$$

The calculation of this roughness spectrum is performed thanks to the use of a discrete Fourier transform. It is obtained in a limited frequency bandwidth : $\sigma_{min} = 2\pi/L$ and $\sigma_{max} = \pi/\Delta x = \pi N/L$.

The adjustement of the sampling interval during Atomic Force Microscopy measurements permits to reach a very large domain of spatial frequencies, and to complete light scattering measurements, as shown in figure 2. In this case we can talk about multiscale characterization of these surfaces.

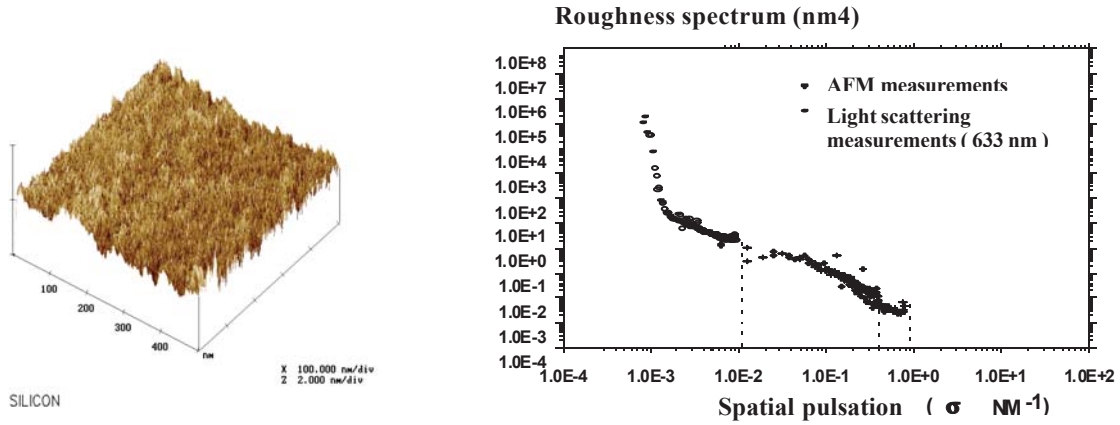


Figure 2: Roughness spectra obtained thanks to light scattering measurements and AFM measurements in the case of a supersmooth silicon substrate.

These measurements revealed a particular behavior of most of our substrates. They present a linear variation in logarithmic coordinates:

$$\bar{\gamma}(\sigma) = A/\sigma^{\alpha} \tag{3.7}$$

This kind of variation can be connected with self-affine, or fractal properties of the surfaces. It permits to put in evidence the oportunity of a multiscale polishing of surfaces.

Indeed, the expression (3.3) permits to calculate the roughness δ and its variations, and the scattering power $u = \delta / \lambda$ of surfaces as a function of the exponent α. We obtain the following expressions and properties:

$$\frac{d\delta}{d\lambda} = \left(\frac{\alpha}{2} - 1\right)\frac{\delta(\lambda)}{\lambda} \quad \text{if} \quad \alpha > 2 \text{ then } \delta \text{ increases with } \lambda \tag{3.8}$$

$$u = \frac{\delta}{\lambda} \Rightarrow \frac{du}{u} = \left(\frac{\alpha}{4} - 1\right)\frac{d\lambda}{\lambda} \quad \text{if} \quad \alpha > 4 \text{ then } u \text{ increases with } \lambda \tag{3.9}$$

The adjustment of the parameter α, by working on the polishing technology of surfaces, could make appear surfaces whose roughness or whose scattering power u is constant with the wavelength, that we could qualify of "multiscale surfaces".

4. DETECTION OF CONTAMINANTS THANKS TO ELLIPSOMETRY OF THE SCATTERED FIELD

Angle resolved ellipsometry of the scattered field has been presented as an original technique developed at Institute Fresnel to complete light scattering intensity measurements **[4]**. The experimental results can be investigated via electromagnetic theories of surface and bulk scattering. We have presented calculations and measurements **[5]** performed on substrates and multilayer stacks. They prove that this technique is a powerful tool to directly separate surface and bulk effects of bare substrates. In other cases, we show that this technique rises to the identification of first and second-order contaminants on coatings.

The first-order electromagnetic theory developped at Institut Fresnel predict surface or bulk effects in light scattering measurements. This theory permits to calculate the amplitude $A_{s \text{ or } p}$ of the scattered field from a multilayer, where the subscript is for s or p polarization:

$$A_{s \text{ or } p} = \sum_i C_i(s \text{ or } p)\hat{h}_i \quad \text{in the case of surface scattering} \tag{4.1}$$

$$A_{s \text{ or } p} = \sum_i C'_i(s \text{ or } p)\hat{p}_i \quad \text{in the case of bulk scattering} \tag{4.2}$$

where $C_i(s \text{ or } p)$ and $C'_i(s \text{ or } p)$ are optogeometrical coefficients depending on the multilayer design and on illumination and observation conditions. Irregularity profiles have spectra $\hat{h}_i$ and $\hat{p}_i$, respectively for the roughness interface in the case of surface scattering, and for index heterogeneity in the case of bulk scattering.
The amplitudes A are complex numbers which can be written as:

$$A_s = \sqrt{N_s}\exp(\delta_s)E_s \tag{4.3}$$

$$A_p = \sqrt{N_p}\exp(\delta_p)E_p \tag{4.4}$$

where A_s, respectively A_p, represents the s or p-polarised scattered field that merges in air in the incident plane, at direction θ from the sample normal, as the result of an incident polarized field $E_s(s-\text{polarized})$, respectively $E_p(p\text{-polarized})$. The polarimetric phase, or "phase term", is defined as **[4,5]**

$$\delta = \delta_s - \delta_p = \text{Arg}(A_s\overline{A_p}) \tag{4.5}$$

In the case of a bare substrate, we can show **[4,5]** that the variations of the polarimetric phase do not depend on the surface profile or bulk inhomogeneity, but on the origin of scattering. **Figure 4** shows these results of calculations and measurements in the case of bulk or surface scattering substrates.

Other results may appear on these experimental results. The first observation is the offset of the phase term at the origin. The other effect is the presence of oscillations. These phenomena are observed on other experimental curves, as shown in **figure 5** and **figure 6**.

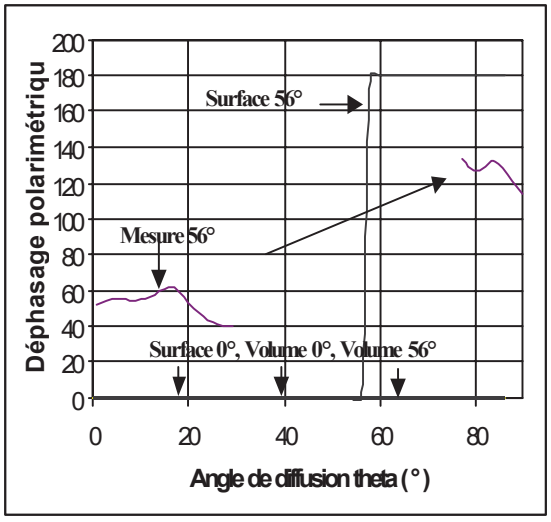


Figure 4-a : *The case of a glass substrate. The rapid variation of the phase term in the vicinity of $\theta = 56°$ for an incidence angle $i = 56°$ reveals surface scattering.*

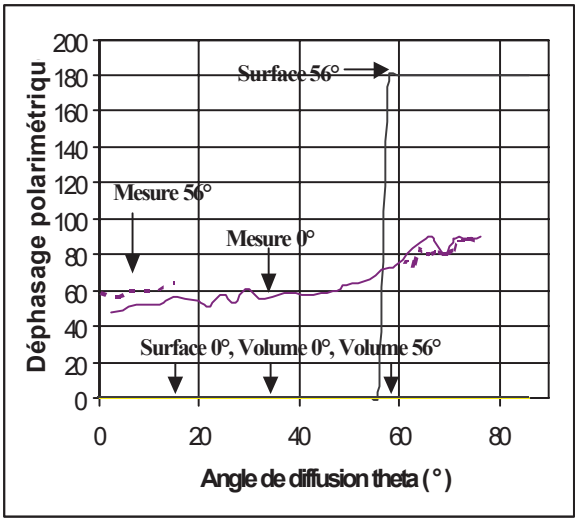


Figure 4-b*: The case of a MgF2 substrate. The low variation of the phase term, whatever the incidence angle, reveals bulk scattering.*

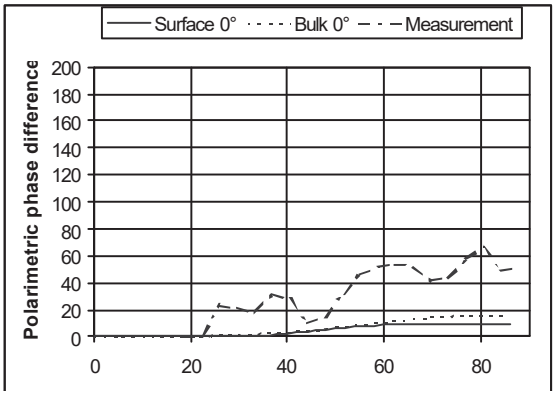
Figure 4: Ellipsometry measurements compared with numerical calculations in the case of bare substrates.

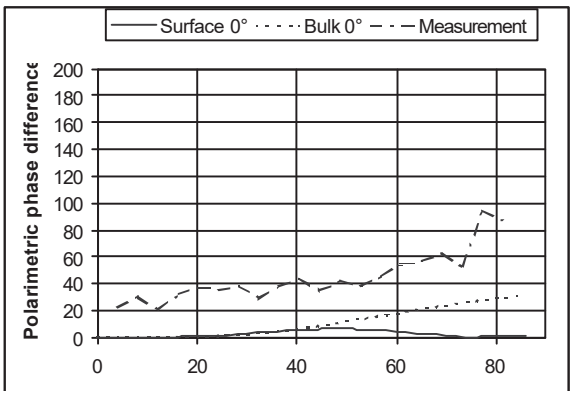
Figure 5: Measurement of the polarimetric phase difference in the case of the specific 2H2B2H structure deposited on a glass substrate, compared with the calculation curves presented in figure 8. The incidence angle is equal to 0°.

Figure 6: Measurement of the polarimetric phase difference in the case of a 8H Ta_2O_5 layer deposited (Ion Platting) on a glass substrate, compared with the calculation curves. The incidence angle is equal to 0°.

The first theoretical calculations where performed in the case of a perfect replication of all interfaces during deposition. But the replication may be not perfect, what we can write as:

$$\hat{h}_{j-1} = \hat{h}_j + \hat{g}_j \qquad (4.6)$$





where $\hat{h}_i$ is the Fourier transform of the interface i, and $\hat{g}_i$ is the roughness brought by the layer i, as described in **Figure 7**.

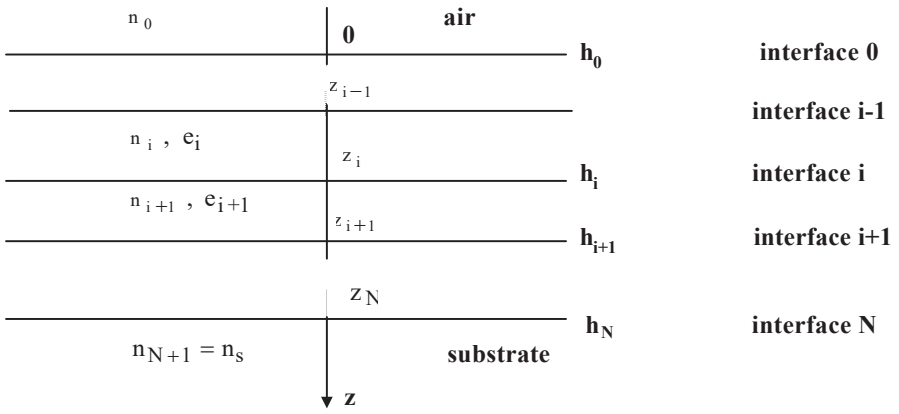


Figure 7: Definition of the opto-geometrical parameters of a multilayer. n_i and e_i are respectively for the index and the thickness of the layer number i.

In the case of N layers, and if $\hat{h} = \hat{h}_N$ is the substrate profile, we can write:

$$\hat{h}_i = \hat{h} + \sum_{p=i+1}^{N} \hat{g}_p \qquad (4.7)$$

Or $\hat{h}_i = \hat{h} + \hat{g}_i$ where $\hat{g}_i = \sum_{p=i+1}^{N} \hat{g}_p$ (4.8)

We can calculate the phase term δ with the formula (4.5). In the case of the perfect replication, we can write that $\hat{h}_i = \hat{h} \ \forall i$, and δ = Arg L_0 with:

$$L_0 = \sum_{i,j} C_i(s)\overline{C}_j(p)\hat{h}\overline{\hat{h}} \qquad (4.9)$$

When the replication is not perfect, δ = Arg L with:

$$L = \left(\sum C_i(s)\hat{h} + \sum C_i(s)\hat{g}_i \right)\left(\sum \overline{C}_i(p)\overline{\hat{h}} + \sum \overline{C}_i(p)\overline{\hat{g}}_i \right) \qquad (4.10)$$

We can deduce that:

$$\frac{L}{L_0} = \left(1 + \frac{\sum C_i(s)\hat{g}_i}{\sum C_i(s)\hat{h}} \right)\left(1 + \frac{\sum \overline{C}_i(p)\overline{\hat{g}}_i}{\sum \overline{C}_i(p)\overline{\hat{h}}} \right) \qquad (4.11)$$

This result shows that the partial correlation creates a ripple in the phase term. The ripple accurates even if the the $\hat{g}_i$ factors are in the order of the substrate profile $\hat{h}$.

These results are illustrated in **Figures 8** and **9** where we present calculations in the case of a 2 nm rough substrate coated with a thin film of different amplitude roughnesses varying from 0.1 nm to 1000nm.

Let us notice that in the case where the amplitude of the layer becomes very high by comparison with the amplitude of the substrate, the behaviour is the same as a single interface behaviour, and doesn't depends on the profile. In this case, the phase curve doesn't present any ripple.

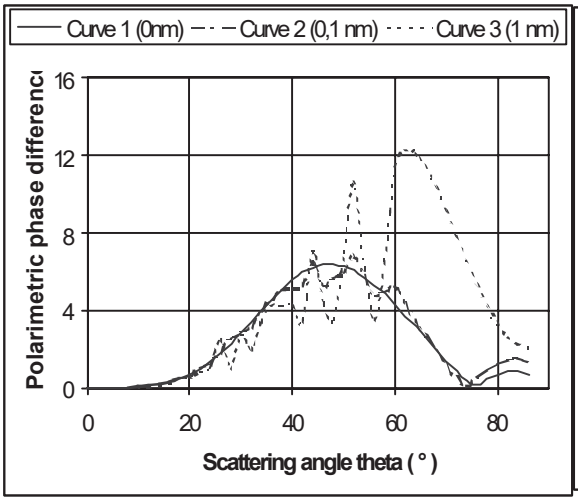


Figure 8: Calculation of polarimetric phase difference in the case of a 8H high index layer (Ta_2O_5 Ion Plating) deposited on a 2 nm rough glass substrate. The layer brought roughness is 0 nm (curve 1), 0.1 nm (curve 2) and 1 nm (curve 3).

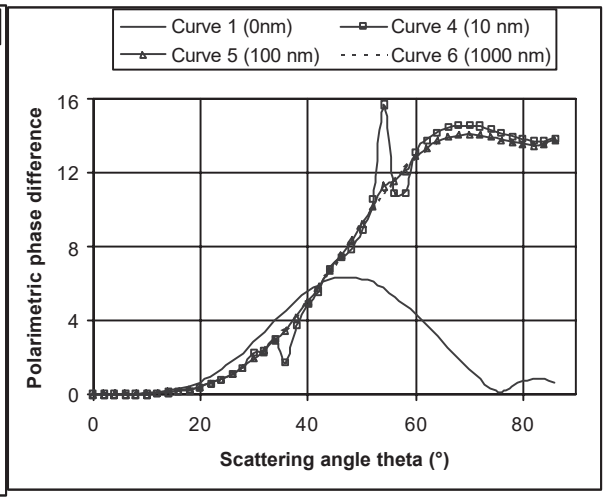


Figure 9: Calculation of polarimetric phase difference in the case of a 8H high index layer (Ta_2O_5 Ion Platting) deposited on a 2 nm rough glass substrate. The layer brought roughness is 0 nm (curve 1), 10 nm (curve 4), 100 nm (curve 5) and 1000 nm (curve 6).

These results give a solution to detect contamination effects on optical surfaces.

The first effect which can be detected is a depolarization effect. In fact, all our investigations are based on first-order electromagnetic theories that predict no cross-polarized scattering in the incidence plane. However many curves show a slight disagreement between theory and experiment. For example, **Figure 4** reveals a bulk behaviour of the MgF2 substrate, because of a slow variation of the phase term in the vicinity if θ equals to i. But this phase term is never perfectly equal to zero, particularly at low angles. This kind of behaviour may appear in other cases, in the presence of multilayer stacks. For example, **Figure 6** is plotted for a thin film layer 8H deposited on a glass. It reveals strong anomalies at low angles, since the phase term is not equal to zero.

In these cases, it is necessary to consider a small amount of light depolarization in the scattering process. This phenomenon can be measured but is not predicted with first order theory in the incidence plane. A phenomenological approach has been performed and presented in a previous paper [4]. It was shown that a depolarization of 10^{-4} causes the phase term to vary from 0° to 25°.

As a consequence, this technique is very sensitive to all second-order effects, or contaminants that change the polarization of light. It completes techniques which measure

crossed polarizations effects correlated with depolarization effects. These effects can be attributed to the presence of contaminants.

The second effect is relative to the detection of "first-order contaminants". In fact, the precedent remarks can be applied to detect the presence of contaminants on optical coatings, which rises to depolarization effects. But contaminants can also be first-order scatterers with low heights or index variations. The problem is thefore more complex since no depolarization occurs in the incidence plane. In this case, the contaminants are strongly analogous to classical roughness and inhomogeneity, and cannot be detected a priori by intensity measurements.

A solution to detect this kind of contaminants can be found in the partial interface correlation due to the presence of contaminants. The behaviour of the polarimetric phase difference can be simulated in this case. Such an interface can be simulated, as seen in **Figure 10**, and used to simulate this effect of contaminants. The result of calculations is presented in **Figure 11**, compared with the result obtained in the case of a perfect correlation, for the deposition of a single 8H layer on a 2 nm rough glass substrate. The result is so characterized by a strong ripple of the phase term centered near the curve calculated for a perfect correlation. The sensitivity is large since we can detect some percent on the decorrelation factors. We hope that the technique is nearly unique to discriminate first-order contaminants.

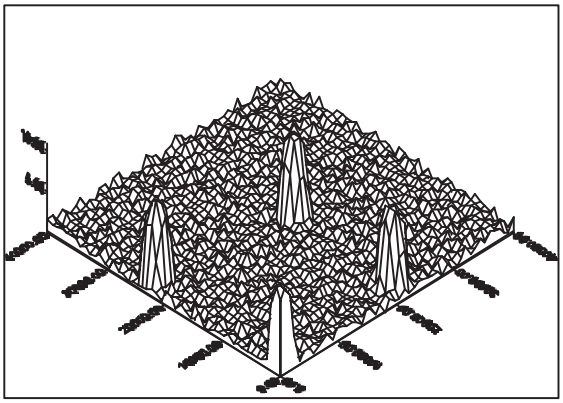


Figure 10: Simulation of a 2 nm rough interface with 4 defects (diameter 4µm, 10 nm high)

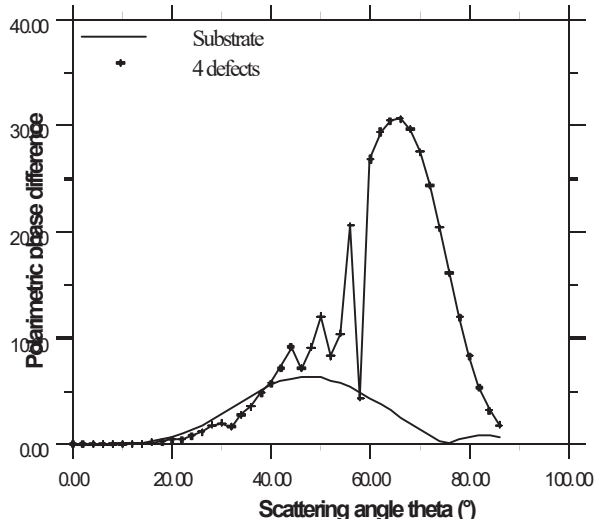


Figure 11: Simulation of the polarimetric phase difference in the case of a 8H Ta_2O_5 (Ion Platting) on a glass substrate, compared with the calculation curves. The incidence angle is equal to 0°.

5. CONCLUSION

Light scattering is a characterization technique which is well known at Institut Fresnel – Marseilles. The experimental setup is performent and has been used in the case of numerous round-robin tables. We have shown that angular scattering measurements at wavelengths varying from the UV to the IR and the development of electromagnetic theories permit to quantify the multiscale roughness of components. This technique has been extended to allow ellipsometric measurements on scattered light in each direction of space. Results can be investigated by electromagnetic theories and permit to directly separate bulk and surface effects in the case of bare substrates and to reveal the high sensitivity of the polarimetric phase difference to the presence of contaminants on surfaces, even in the case of first order contaminants, that is to say whose size is in the same order as the substrate roughness.

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