Scattering–reduction effect with overcoated rough surfaces: theory and experiment

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We show that a scattering–reduction effect is obtained by coating a rough surface with an antireflection layer. This research is a generalization of Amra's [J. Opt. Soc. Am. A **10,** 365–374 (1993)] study of smooth surfaces conducted with a first-order theory to the case of rough surfaces. We show that the differential method with the *R* matrix algorithm can be used to study scattering from multilayered rough surfaces. A comparison between numerical and experimental results is given. © 1997 Optical Society of America

Key words: Scattering, optical coatings, thin films, rough surfaces, differential method.

1. Introduction

Scattering from high-quality multilayers has been studied by numerous authors[1–3] for several decades. The objective is to understand the origin and to control the magnitude of off-specular optical losses as regards thin-film technologies (ion-assisted deposition, electron-beam deposition, ion plating, ion-beam sputtering, etc.). For this purpose, numerous experimental and theoretical tools have been developed; they have provided solutions to inverse problems that consist of characterizing material microstructures from light-scattering measurements.[4–13] Though the results might strongly depend on numerous factors such as substrate roughness, materials, and deposition techniques, the phenomena are now well understood; a good level of understanding is now reached and there are recent results (see Refs 14–16) that deal with multiscale roughness, ellipsometric measurements, localized defects as well as surface and bulk scattering. Generally, these studies are performed on low-loss ($<10^{-4}$) optical filters deposited on supersmooth substrates ($\approx$0.1 nm roughness), which largely warrant the use of first-order electromagnetic theories in this field of application for optical communication, laser mirrors, etc. Note that first-order theories present the advantage of dealing with two-dimensional overcoated surfaces. Hence they allow prediction of polarization effects outside the incidence plane.

However, there is another range of applications concerning, in particular, astronomy (for the design of light traps, solar cells, etc.) in which very rough surfaces can be overcoated with multilayers to modify their scattering pattern, which is the subject of this paper. We wondered whether it was possible to reduce the scattering pattern of a very rough surface by depositing one or more layers on it. Numerous solutions have been predicted by first-order theories, but these solutions were not valid *a priori* for rough surfaces (roughness $\approx$ wavelength). This also added to our motivation to develop a computer code based on a rigorous electromagnetic theory for scattering from rough surfaces covered with multilayers. Our method was valid for one-dimensional surfaces, and it allowed us to predict either enhancement or reduction of scattering in the multilayer design. It was based on the differential method,[17] which was improved with the use of the *R* matrix[18] (or *S* matrix) algorithm. To our knowledge, this was the first time that the differential method was used to calculate the scattering from randomly rough surfaces. Although this method was slower than the surface integral method,[19–21] its wide range of applications made it a suitable candidate for tackling this issue. Indeed, without modification, it can be applied to overhang-

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ing surfaces, to rough layers with interpenetrating surfaces, and to rough inhomogeneous films.

Section 2 describes the numerical method used and compares the results with those obtained with a surface integral method for both TE and TM polarizations. In Section 3, we compare the results given by the differential method with those given by a first-order perturbation method in the case of coated smooth surfaces. We use our model to study the extent to which the solutions offered by a first-order theory remain valid when roughness is increased. Section 4 describes the experiment. We show that good agreement is found with theory, and we demonstrate that antireflection coatings can be used to reduce scattering from rough surfaces.

2. Differential Method for Rough Surfaces

The problem of scattering from rough surfaces has been studied for both industrial and scientific purposes. Among many formalisms that have been proposed, the surface integral method is the most commonly used.[19–21] This method usually deals with homogeneous rough media whose profiles are defined by a function $h(x)$. Although this method's potentialities have been demonstrated many times, calculation of the scattering from a surface composed of rough, interpenetrating layers is needed to solve coupled integral equations. Therefore this method is not adapted when a bare rough surface is covered by more than one layer. Other methods, such as the differential method and the coupled-wave method,[17,22] permit one to calculate the diffracted pattern from any kind of surface profile.[23] However, these two methods, already used to study diffraction by gratings, have been plagued for many years by numerical instabilities, especially for the TM (p) polarization when the height of the grooves is much greater than the wavelength. Today such limitation can be overcome thanks to the R or the S matrix[24] algorithms. Now the fields of applications of the differential method can be extended to the study of scattering from rough surfaces. Moreover, the differential method is well adapted to the study of multilayered rough structures.

A. Description of the Method

We considered a plane wave that impinges on a one-dimensional surface (see Fig. 1). The randomly one-dimensional rough surface is decomposed into Q slices, each one corresponding to the interval $[y_m, y_{m+1}]$. The slices are assumed to be thin enough that we can consider that the function ε_m describing the permittivity between y_m and y_{m+1} is only x dependent. In this case $\varepsilon_m(x)$ can be written as

$$\varepsilon_m(x) = \int_{-\infty}^{+\infty} \hat{\varepsilon}_m(\sigma)\exp(2\pi i\sigma x)\mathrm{d}\sigma, \tag{1}$$

where $\hat{\varepsilon}_m(\sigma)$ is the Fourier transform of $\varepsilon_m(x)$. For another slice corresponding to an interval $[y_p, y_{p+1}]$, the corresponding permittivity is given by another

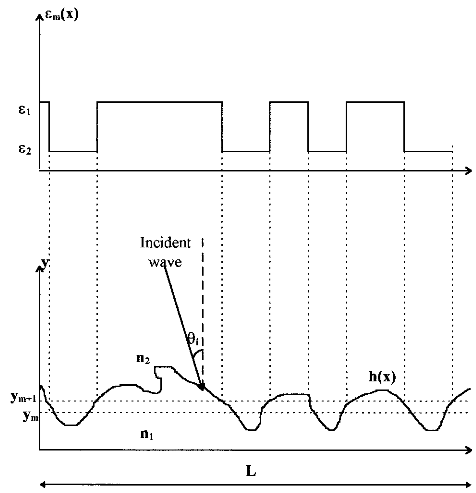

Fig. 1. Decomposition of the rough region into Q different slices.

function $\varepsilon_p(x)$. Then the permittivity variation with respect to y is contained in the different functions $\varepsilon_m(x)$ ($m \in \{1, \ldots, Q\}$).

When the surface of length L is discretized with a sampling interval $\Delta x = L/N$, Eq. (1) leads to

$$\varepsilon_m(x) \approx \sum_{n=-N/2}^{N/2-1} \hat{\varepsilon}_m{}^n \exp\left(\frac{2\pi i n}{L}\, x\right), \tag{2}$$

where $\hat{\varepsilon}_m{}^n$ are the Fourier coefficients of function $\varepsilon_m(x)$. A fast Fourier transform (FFT) algorithm permits us to compute the coefficients $\hat{\varepsilon}_m{}^n$ ($m \in \{1, \ldots, Q\}$, $n \in \{-(N/2), \ldots, (N/2) - 1\}$) for each layer. The use of a classical Runge–Kutta method in addition to the R-matrix algorithm allows integration of the set of coupled differential equations obtained.[9] The boundary conditions lead to a linear system of N equations in which the unknowns are the amplitudes of the reflected and the transmitted waves. The flux of the Poynting vector of the scattered waves through a plane parallel to the z axis is directly obtained, and then the differential reflection coefficient (DRC) can be computed. The method permits one to deal with both TE and TM polarizations; the only difference between the two cases is that the expression of the coupled differential equations to be solved. Here the spatial frequencies α_n of the diffracted waves are given by

$$\alpha_n = \alpha + (2\pi/L)n, \tag{3}$$

where

$$\alpha = \frac{2\pi}{\lambda}\, n_2 \sin\theta_i, \tag{4}$$

where λ is the wavelength of the incident wave and θ_i is the incidence angle. Hence, because of the FFT sampling $\Delta\sigma = 1/L$, the L length plays the same role as period d for gratings. The directions of the diffracted waves are given by Eq. (2). To increase the number of scattered waves considered, one has to increase L. In this case the surface has to be sampled in a higher number of points, and the time of computation increases. Because of the set of coupled differential equations to be solved, the calculation of M orders needs to sample the surface into $2.M$ points. In addition, as for gratings, to determine the number of evanescent waves that must be considered to describe the near field in the region of the grooves, one needs to test the convergence of the result by increasing M. As for multilayer gratings,[17] the differential method can be used to calculate the scattering from multilayer rough surfaces. The only difference from a single bare rough surface is that the function $\hat{\varepsilon}_m(x)$ ($m \in \{1, \ldots, Q\}$), which must be developed in Fourier series, is more complicated.

B. Validation of the Numerical Results

In this paper, all the surfaces have been generated following the method and using the notations described in Ref. 20. To validate our computer code, we have considered a single rough surface with Gaussian statistics with given values of rms height δ, correlation length a, and length L at wavelength λ. All the computations have been performed on an IBM 604 workstation. We have computed the DRC's obtained with the differential method for normal incidence, with $M = 256$ and $Q = 150$. We have compared the results obtained with the differential method with those given by a surface integral method[20] for both TE and TM polarizations. Figure 2 shows that a very good agreement between the two methods has been found. Because of the limitations of computer memory size, the lengths of the surfaces considered in Fig. 2 are only 30 and 20 μm for TE and TM polarizations, respectively. This problem is typical of the computer codes based on rigorous electromagnetic theories. For this reason, owing to the speckle, the angular pattern of scattering exhibits several peaks. To simulate a case corresponding to a large lighted surface, one has to perform the computation for several surface samples with the same statistics. The scattering from a large surface is obtained when the DRC's of all the samples are averaged.[20] The computation time is approximately twice that of the surface integral method in the conditions of Fig. 2. This result shows that the differential method can be used to study scattering from rough surfaces.

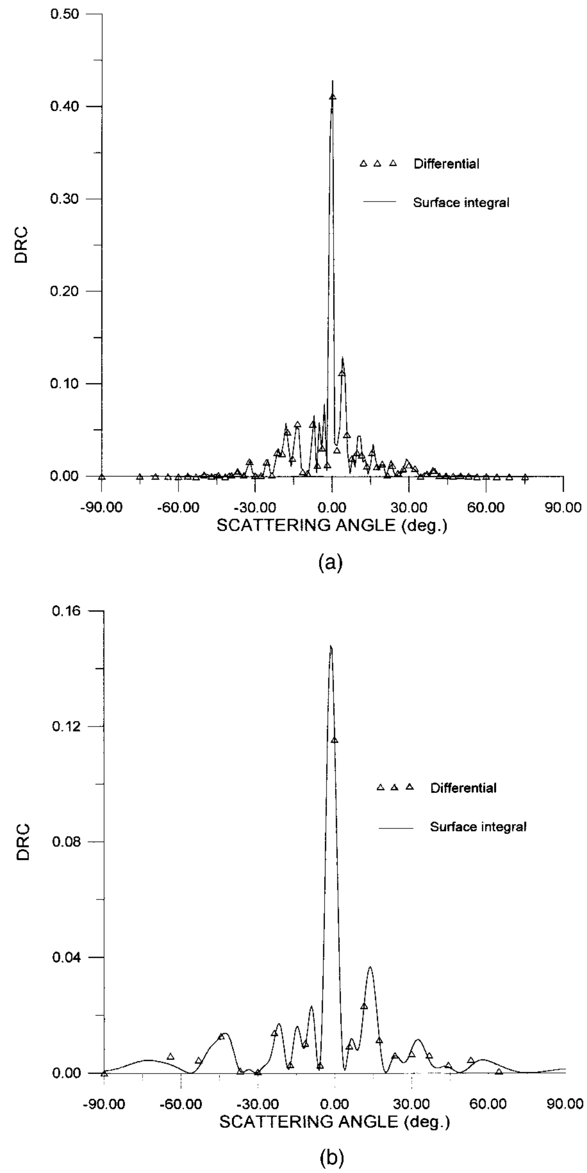

Fig. 2. Comparison between differential and integral surface models: $n_1 = 1.52$, $n_2 = 1$, and $\lambda = 1$ μm. Normal incidence: (a) TE polarization $\delta = 0.1$ μm, $a = 1$ μm, and $L = 30$ μm; (b) TM polarization $\delta = 0.1$ μm, $a = 0.3$ μm, and $L = 20$ μm.

3. Analysis of Solutions Given by First-Order Theories

Now we come to multilayer coatings. Many solutions given by a first-order theory have been proposed to reduce scattering from smooth surfaces. Thanks to the differential method, it is possible to extend this study to the case of surfaces whose roughness is of the same order of the wavelength.

A. Case of Smooth Surfaces

In this subsection, we are first concerned with smooth surfaces (for example, $\delta < \lambda/50$). It was previously shown[25] that an antiscattering effect could be obtained after deposition of a single layer on a rough surface. With the parameters corresponding to a typical case in the field of optical thin films ($\lambda = 0.633$ μm, the refractive index of the substrates $n = 1.52$, $\delta = 0.003$ μm, and $a = 2$ μm), it has been shown that a quarter-wavelength antireflection layer of refractive index $n = 1.3$ allows reduction of the amount of scattered light. This result is valid provided that the ratio $r = \delta_0/\delta_1 \in [0, 2]$ in which δ_0 and δ_1 are the rms values of the upper surface (coated surface) and of the lower surface (bare surface), respectively.[25] Moreover, it has been demonstrated that the value $r = 0.66$ leads to an antiscattering effect (zero scattering) given by an analytical result. This effect was predicted by a first-order theory[10] and confirmed by numerous experiments on supersmooth substrates.[6] In the first step, we verified these results with our rigorous method. To this end we have verified that the conclusions given by the

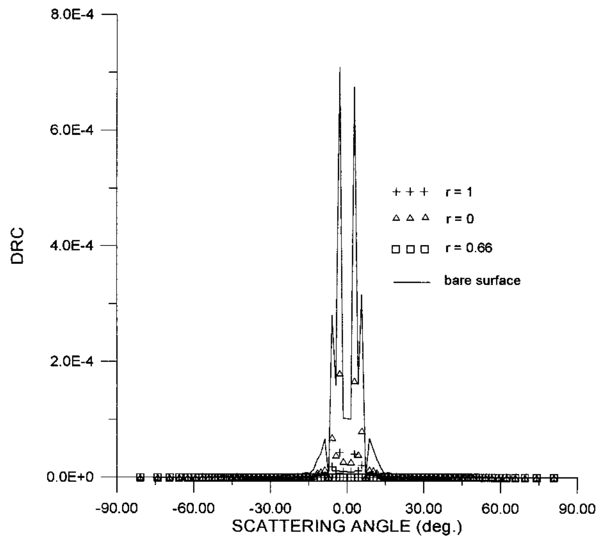

Fig. 3. Scattering-reduction and antiscattering effects obtained when a smooth surface is coated. $L = 25.32$ μm and $\lambda = 0.633$ μm. Computation with the differential method. The specular reflection is not represented. The coating is a single layer of Cryolite [Na_3AlF_6, refractive index $n = 1.3$, and thickness $e = \lambda/(4.n)$]; $n_1 = 1.52$ and $n_2 = 1$. Normal incidence.

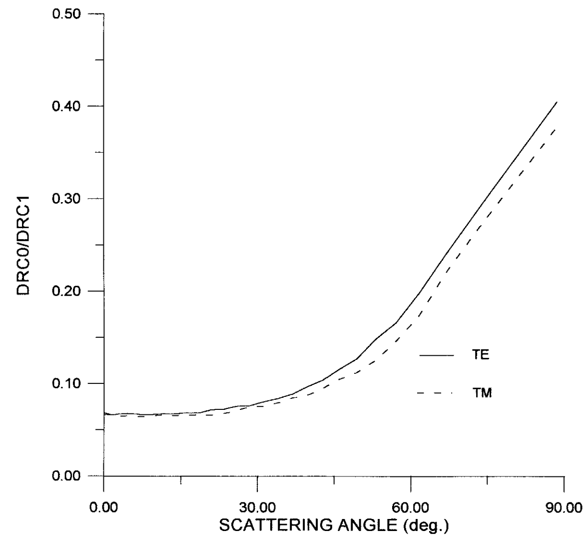

Fig. 4. Scattering-reduction effect obtained when a rough surface is coated. Numerical result.

method described in Ref. 25 are confirmed by the results obtained with the differential method described in Section 2. In Fig. 3 we have plotted the angular pattern for a smooth surface before and after the deposition of a single layer with different values of r, and we did not represent the specular reflection. As explained in Subsection 2.B, the small dimensions of the surface considered lead to the speckle observed in the angular pattern of the scattered light. The average of the angular patterns of a high number of surfaces (corresponding to the case of a large lighted surface) leads to a continuum of scattering. The DRC in Fig. 3 must be compared with the DRC of Fig. 2. One can see that for the smooth surface, the amount of off-specular light is much lower than for the rough surface considered in Subsection 2.B. These results confirm the predictions given by the first-order theory, and they show the potentiality of the differential method.

B. Case of Rough Surfaces

We then concerned ourselves with the validity of scattering reduction when the surface is very rough and when parameter δ is out of the range of validity of the first-order vector theory. We have considered a surface with Gaussian statistics with $\delta = 1$ μm, $a = 3$ μm, and $L = 30$ μm, covered with a single antireflection layer, at $\lambda = 0.633$ μm. The two surfaces (the substrate and the coating) are assumed to be similar. The refractive index n_1 of the substrate is assumed to be $n_1 = 1.52$, which is a typical value for standard glass. The refractive index of the coating is that of Cryolite: $n = 1.3$. Figure 4 shows the ratio DRC0/DRC1 as a function of the scattering angle, where DRC0 and DRC1 are the DRC's of the coated surface and the bare surface, respectively. To simulate a case corresponding to a large lighted surface,[20] we performed the computation for 100 surface samples with the same statistics. The values of DRC0 and DRC1 were then averaged over the total number of samples. The results shown in Fig. 4 show that the scattering-reduction effect still occurs for rough surfaces. Numerical experiments on various rough surfaces with other statistics have shown the same effect. The parasitic light can then be reduced by coating rough surfaces; the scattering-reduction effect obtained with smooth surfaces can also be achieved in the case of rough surfaces. However, Fig. 5 shows that in the case of rough surfaces, in contrast to the result described in Subsection 3.A (see Fig. 3), no antiscattering effect is obtained with $\delta_0/\delta_1 = 0.66$. This result clearly justifies the use of a rigorous method to calcu-

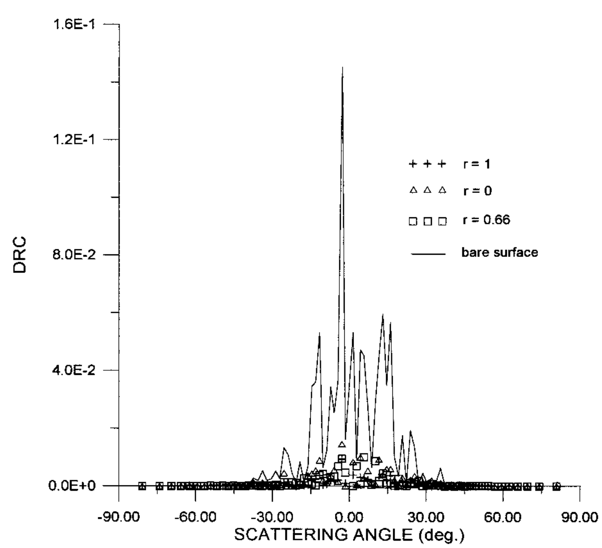

Fig. 5. Angular pattern of the bare surface and of the coated surface for different values of r. Case of a rough surface: $\delta = 0.2$ μm, $a = 0.3$ μm, and $L = 30$ μm. TE polarization.

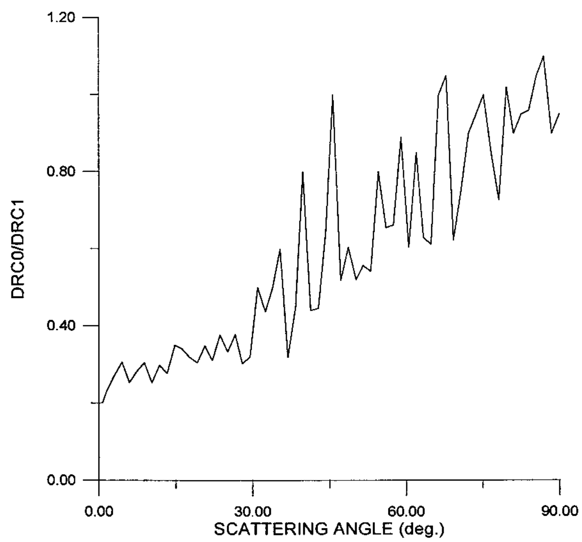

Fig. 6. Scattering-reduction effect on a rough surface. Experimental results. The wavelength of operation is $\lambda = 0.633$ μm. Normal incidence. Spot size ≈ 6 mm^2.

late the scattering from surfaces whose roughness (in terms of rms) is of the same order of the wavelength.

4. Application to Experiment

Scattering from a two-dimensional rough surface ($n_1 = 1.52 + 10^{-3}$ j) was measured[12] at $\lambda = 0.633$ μm before and after the deposition of a single, quarter-wavelength Cryolite layer. Experimentally we have ensured that the surfaces, both bare and coated, exhibit only diffuse reflectance or scattering. The characterization of the surface by an atomic force microscope has shown that $\delta = 1$ μm and $a = 3$ μm. The experimental results observed in Fig. 6 tend to agree with our predictions, although our computer code is one dimensional. The experimental results confirm the numerical predictions and show that a scattering-reduction effect is obtained for a single layer.

5. Conclusion

We have shown that the differential method can be used to study scattering from rough surfaces. The results with this method show a perfect agreement with those yelded by the surface integral method. The differential method is advantageous in that it is well suited to the case of multilayered structures. Moreover, the differential method of computation presented here permits one to deal with inhomogeneous media that are difficult to treat with other more conventional methods. The study of this topic is beyond the scope of this paper and is the subject of a future paper. The results given by the differential method have been successfully compared with those given by the first-order vector theory in the case of smooth surfaces. The effects of coatings deposited on very rough surfaces have been determined both theoretically and experimentally. In particular, it has been shown that a scattering-reduction effect is obtained when an antireflection coating is deposited on a rough surface. The results presented here will help extend research to the study of multilayer coatings deposited on rough surfaces.

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