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小尺度表面粗糙度与介质吸收对介质折射率测量的影响

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摘　要:将介质表面的小尺度粗糙度等效为覆盖在理想光滑表面上的多层等厚折射率渐变的薄膜,并通过特征矩阵计算多层等效膜模型的P光反射率与入射角的关系.将吸收介质的折射率虚部带入菲涅尔公式进行计算.运用COMSOL Mutiphysics软件对表面粗糙度和介质吸收进行建模和仿真计算.计算结果表明,小尺度表面粗糙度与介质吸收都会导致折射率测量产生误差.分别考虑以布儒斯特角和全反角作为折射率测量的手段,为了得到优于10^{-5}的测量准确度,测量表面粗糙介质的折射率时,采用全反角进行判定;测量具有吸收效应的介质折射率时,采用布儒斯特角进行判定.

关键词:折射率测量;表面粗糙度;布儒斯特角;全反角;COMSOL Mutiphysics应用

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Effects of Small-scale Surface Roughness and Absorption on Refractive Index Measurement by the Brewster-angle and Critical-angle Techniques

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Abstract: Small-scale roughness on surface is equivalent to a multilayer structure with same thickness and gradient refractive index. By using characteristic matrixes, the reflectivity of P waves of the effective multilayer model with different incident angle is calculated; the refractive index of the absorbing medium is a complex number in the calculation of Fresnel formula; COMSOL Mutiphysics is used to model and simulat the small-scale roughness and absorption. The calculation results show that both the small-scale roughness and absorption of the dielectrics can make errors in measurement of the refractive index. In conclusion, in order to get the precision of 10^{-5}, the refractive index of rough dielectrics should be measured by critical-angle technique, while it is better to measure absorbing dielectrics by Brewster-angle technique.

Key words: Refractive index measurement; Surface roughness; Brewster angle; Critical angle; COMSOL Mutiphysics

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0 Introduction

Fresnel's equations give the reflectance R, the transmittance T, and the ratios of the reflected and transmitted irradiances to the total incident irradiance of an ideal interface [1]. In the equations, it is found that there are two special incident angle, the Brewster angle (θ_b) and the critical angle (θ_c). If the refractive index of ambient environment (n_a) and θ_b or θ_c are known, the refractive index of the medium under tested (n_m) can be calculated[2]. Because of this, Brewster-angle and critical-angle techniques can be used in the measurement of the refractive index of solid and liquid dielectrics, thin films and any other optical materials [2-3].

The study related to Brewster angle and critical angle is a subject for many areas including material science, biology, astronomy, and forensic analysis[4-9]. Additionally, there are many articles about measuring the refractive index by Brewster-angle and critical-angle techniques. D. Luna-Moreno et al. used Brewster-angle technique to measure the pure and Er^{3+}-doped ZrO_2-SiO_2 sol-gel film in a precision higher than 99.5% and 98% in the refractive index and thickness[3]. Cristian Bahrim et al. gave a reliable and improved setups in this measurement technique, and he found it was more accurate to use only the reflectivity of the P waves to obtain the values of θ_b[2]. VN Konopsky and EV Alieva designed a periodic multilayer coating to enhance the accuracy of critical-angle refractometers[10]. R. Pawluczyk, et al [11], and any other researchers had also done the work related to those.

In this paper, it gives the influence of small-scale interfacial roughness and absorption in measuring the refractive index by the techniques, and the errors caused by roughness and absorption are compared in the two techniques.

1 Methods

Surface roughness is inevitable in practice, and it is usually described by Root Mean Square (RMS) roughness and power spectral density function. The former ignores the characteristic of surface spatial wavelength, and the latter is difficult for recurrent computation[12]. Alexander V. Tikhonravov et al. [13] proposed the concept of large and small-scale RMS roughness according to the ratio of the optical wavelength and spatial period. It was found that the effect on reflectivity caused by small-scale roughness on the surface can be equal to which caused by a thin over-layer with specific thickness and refractive index on it. Jiang Qiyuan et al. [12] continued this work and had made strict theoretical verification to prove the effectiveness of the equivalent in the case of oblique incidence. Therefore, in this paper, in order to improve the precision, the model is based on a multilayer structure.

1.1 Model description

The roughness on surfaces and interfaces might not only change the reflectance and transmittance of the medium, but also produce diffuse scattering. As divided into large-scale and small-scale roughness, the later will not cause any losses by scattering[13]. As defined in Ref. [13], roughness harmonics with spatial wavelength T_m that satisfy the inequality

$$\ln(\lambda/T_m) > \ln n_m \tag{1}$$

to be small-scale harmonics. In Eq. (1), n_m is the refractive index of the measuring medium. λ is the wavelength of the light. In order to make it more easily to be solved in mathematical, the sine surface is used to simulate its actual rough surface. As known, any complex surface can be divided into a series of sine surfaces, which just likes Fourier series. Therefore, this simulation is reasonable and effective. The only problem is that the surface seems to be smoother when it is seen from a large incidence angle, which means the value of RMS may be related with the angle. However, there is not sufficient evidence to confirm it, and the relationship between surface spatial wavelength and incident light wavelength does not change[12]. In Tikhonravov's way, the effective single-layer with a thickness of 2σ (σ denotes the value of the small-scale RMS roughness) and a refractive index n that satisfies the equation[13]

$$n^2 = (n_a^2 + n_m^2)/2 \tag{2}$$

can be equal to the small-scale roughness. However, it may not suitable in oblique incidence. Based on Ref. [12], it was proved that the oblique incidence can also be correct in the equivalent layer mode by a strict mathematical deduction of transfer matrix. Then, dividing the amplitude of the sine surface, 2σ, into N equal parts, the equivalent refractive index of dth layer, n_d, can be described as follows[12]

$$\begin{cases} n_d = n_a + (n_m - n_a) \cdot \dfrac{N}{2\pi}\left[(\arccos U - \pi)\cdot U - (\arccos V - \pi)\cdot V + \left(\sqrt{1-V^2} - \sqrt{1-U^2}\right)\right] \\ U = \dfrac{N-3d}{N} \\ V = \dfrac{N-2(d-1)}{N} \end{cases} \tag{3}$$

The steps of how to build the effective multilayer model is shown in Fig. 1

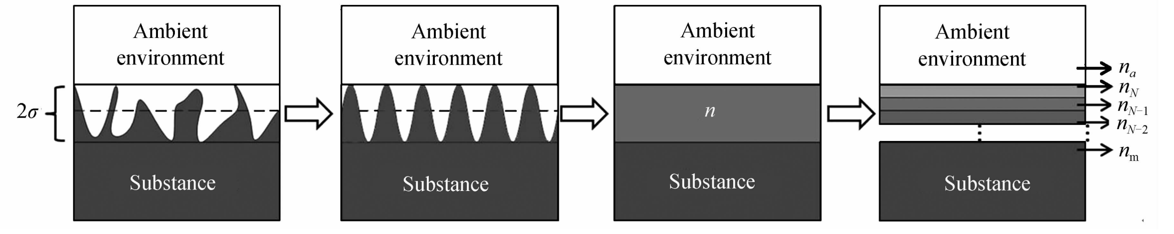


Fig. 1 Steps to build the model

1.2 Reflectivity calculation

It is difficult to calculate the reflectivity of a multilayer by Fresnel's formula directly. Thus, a calculation method should be used. The optical admittances of jth layer is given as[14]

$$\zeta_j = \begin{cases} n_j \cos\theta_j, \text{S-polarization} \\ \dfrac{n_j}{\cos\theta_j}, \text{P-polarization} \end{cases} \tag{4}$$

where θ_j denotes the transfer angle of jth layer. As proved by calculation and experimental data in Ref. [2], the difference of the incidence angle between $[R_s(\theta)]_{max}$ and $[R_P(\theta)]_{min}$ is too large to determine accurate values for the Brewster angle form the S waves component. In the article, only the reflectivity of the P waves ($R_P(\theta)$) is calculated.

Characteristic matrixes can reflect the physical parameters of the media and can be used to calculate the reflectivity. The characteristic matrix of a multilayer is defined as[14]

$$\begin{bmatrix} B \\ C \end{bmatrix} = \left\{ \prod_{i=1}^{N} \begin{bmatrix} \cos\delta_i & \dfrac{j}{\zeta_i}\sin\delta_i \\ j\zeta_i \sin\delta_i & \cos\delta_i \end{bmatrix} \right\} \begin{bmatrix} 1 \\ \zeta_m \end{bmatrix} \tag{5}$$

In the equation, δ_i denotes the thickness of the phase of ith layer, and it can be described below[14]

$$\delta_i = k d_i \cos\theta_i = \frac{2\pi}{\lambda} n_i d_i \cos\theta_i \tag{6}$$

where, n_i is the refractive index of the ith layer, and d_i is the thickness. From the Eqs. (4), (5) and (6), the reflectivity of the multilayer with ambient environment and substrate is

$$R = \tilde{r}\tilde{r}^* = \left(\frac{\zeta_a B - C}{\zeta_a B + C}\right)\left(\frac{\zeta_a B - C}{\zeta_a B + C}\right)^* \tag{7}$$

For a medium with absorption, the calculation is the same while the refractive indexes and optical admittances should be replaced as complex numbers. What should be stressed is that the calculation of reflectivity contains all the energy of the reflected light and transmitted light to the ambient environment, which means the light oscillate infinitely many times in the multilayer. It is obvious that the multilayer and the light may not meet the requirements to do so. However, this is just an effective model. In practice, the light would not oscillate on the rough surface. So this is not a real problem in this model. In this article, $\lambda = 632.8$ nm, $n_a = 1$ is set in the calculations, as the He-Ne laser is usually used in optical measurement and ambient environment is the air upon most occasions.

2 Results

2.1 Small-scale surface roughness

The results caused by small-scale surface roughness is quite different in the two techniques. Thus, they are given in the two small sections below.

2.1.1 *Brewster-angle technique*

Obviously, the effective model of the small-scale roughness would have a different incidence angle to reflectivity curve from the ideal smooth surface. Setting $n_m = 1.457$, the curves of reflectivity with different incidence angles and different RMS values are shown in Fig. 2(a).

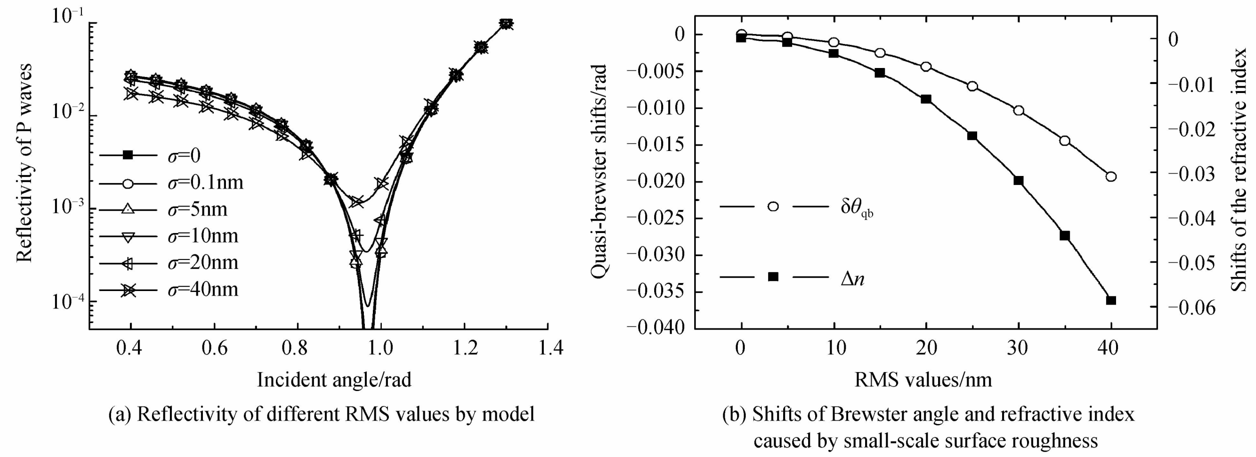


Fig. 2 Results of small-scale roughness by model: Brewster-angle technique

Here the result of $\sigma = 0$ is calculated from Fresnel′s formula directly. In other words, it is the reflectivity curve of an ideal smooth surface. The curve of $\sigma=0.1$ nm is hardly to be distinguished from it in the figure with the largest distinction of 10^{-7} in the reflectivity values near the incidence angle of 0. Hence, the equivalent single layer with a thickness of 0.2 nm is precise enough in the model. The other results with a larger σ are divided into a multilayer with the thickness of single layer of 0.2nm. For example, if $\sigma=5$ nm, the N will be 50. Due to limited space, only a few curves with different σ are listed.

From Fig. 2(a), it is found that a rough surface with a larger RMS value will have a higher reflectivity near Brewster angle, but a lower one away from it. Obviously, this could result in deviations of the measurement results. However, what makes more senses is that the curves have the minimum point at different angles. Usually, the minimum point are regarded as the Brewster angle in measurements, so this will induce errors in measuring the refractive index. Additionally, the angle having minimum reflectivity decreases as the σ becomes larger, which means that the errors will become larger when the RMS value increases.

As mentioned above, in Brewster-angle technique, the angle of the minimum reflectivity of P waves is regarded as the Brewster angle and the surface roughness will make errors in calculations of the refractive index. Naming this inaccurate Brewster angle as quasi-Brewster angle (θ_{qb}). The quasi-Brewster shifts, $\delta\theta_{qb}$ is defined as the formula below[15]

$$\delta\theta_{qb}=\theta_{qb}-\theta_b \tag{8}$$

Then, the errors of the refractive index, namely, the shifts of the refractive index, is described as

$$\Delta n=n_{qb}-n_m \tag{9}$$

The detailed formulas of $\delta\theta_{qb}$ and Δn are too complex to be described. Therefore, the approximation calculated by Matlab with a high precision of 10^{-6} is given in Fig. 2(b). In fact, Fig. 2(b) is another form of Fig. 2(a), it gives the shifts of angle having minimum reflectivity and shifts of the refractive index caused by the former. When $\sigma=40$ nm, the distinction of the refractive index is obviously larger than 0.055. Since refractive index reading errors will be important when they become larger than the refractive index precision of modern instruments, about 10^{-5}[16], the errors may not be ignored.

2.1.2 *Critical-angle technique*

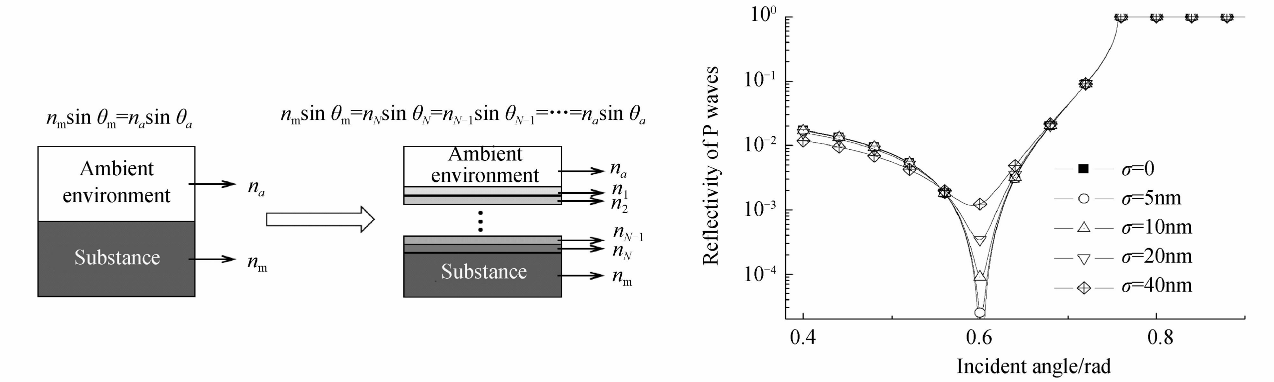
Different from Brewster-angle technique, light incidents from the medium to the ambient environment in critical-angle technique. As shown in Fig. 3(a), when the surface is ideal flat, the angles and the refractive indexes should meet the Fresnel formula as below.

$$n_m\sin\theta_m=n_a\sin\theta_a \tag{10}$$

While θ_m is larger enough and the value of $\sin\theta_a$ should be bigger than 1 to satisfy this formula, the total reflection will happen. Similarly, in a multilayer structure, or in other words, when the surface is rough, angles and the refractive indexes of every adjacent layers should also satisfy this formula. In this case, Eq. (10) can be written as

$$n_{\mathrm{m}}\sin\theta_{\mathrm{m}}=n_N\sin\theta_N=n_{N-1}\sin\theta_{N-1}=\cdots=n_{\mathrm{a}}\sin\theta_{\mathrm{a}} \tag{11}$$

Angles and the refractive indexes of the first and the last layer have the same mathematic relation as in a flat surface, which means that if incident angle θ_{m} is big enough to cause total reflection on a perfect surface, it can also do this on a multilayer structure which is used to represent the small-scale roughness, as a consequence of the recursive formula. Thus, the critical angle will not change in a rough surface and roughness makes no errors in the refractive index measurement in the effective multilayer mode. Fig. 3(b) gives the result of reflectivity with different incidence angles and different RMS values by mode. It is obvious form the figure, the critical angle are the same whatever the RMS value is. This result accords with the Eq. (11) very well. Therefore, small-scale surface roughness have no effects on critical-angle technique in this computing method.

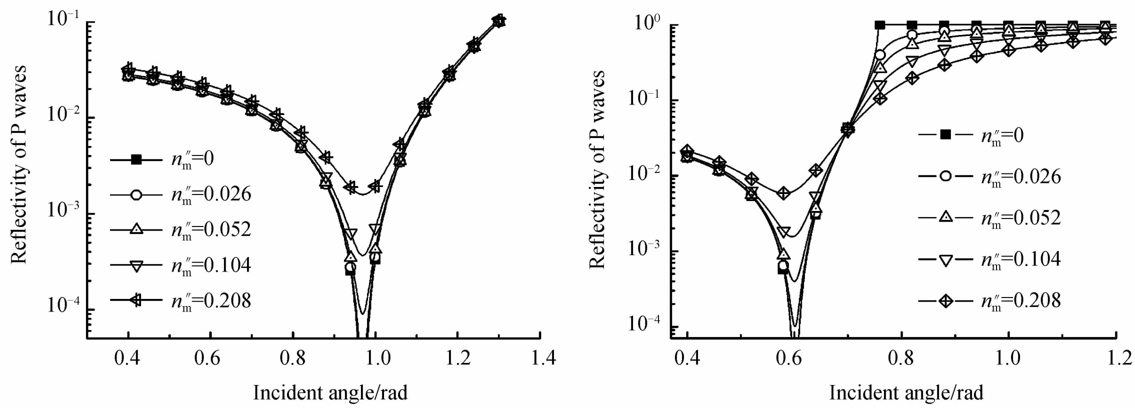


(a) Snell's law in a multilayer structure (b) Reflectivity of different RMS values by model

Fig. 3 Results of small-scale roughness by model: critical-angle technique

2.2 Absorption

G. H. Meeten[16] had already done some work about absorption. In Brewster-angle technique, it seems that effects caused by absorption has some similar features as those caused by small-scale roughness. It is clear in Fig. 4(a), absorption also makes the reflectivity increase near Brewster angle. But different from roughness, reflectivity also increases away from Brewster angle, not decreases.



(a) Brewster-angle technique (b) Crtical-angle technique

Fig. 4 Results of absorption by different models

Additionally, there is a more significant difference between them. The reflection coefficient of an absorptive medium at Brewster angle can be described as[17]

$$r_{\mathrm{p}}=\frac{\mathrm{i}n''_{\mathrm{m}}\cos\theta}{\mathrm{i}n''_{\mathrm{m}}\cos\theta+2n_{\mathrm{a}}\sin\theta}=\frac{\mathrm{i}n''_{\mathrm{m}}}{\mathrm{i}n''_{\mathrm{m}}+2n'_{\mathrm{m}}} \tag{13}$$

where n_{m} is a complex number and $n_{\mathrm{m}}=n'_{\mathrm{m}}+\mathrm{i}n''_{\mathrm{m}}$. The equation has the minimum value at Brewster angle, but not zero, unless the n''_{m} is zero. That means the absorption would not induce the quasi-Brewster shifts, if the angle of the minimum reflectivity of P waves was regarded as the Brewster angle. And what it influences is just the values of the reflectivity. It is interesting to find that G H Meeten[16] thought that absorption would make errors in Brewster angle. As a matter of fact, his conclusion is also correct. Because the errors come from the numerical solution of an approximate formula given by Humphreys-Owen[18], as the value of $R_P(\theta_b)$ is not zero. However, if the refractive index is obtained in an experiment way and the angle of minimum reflectivity is treated as Brewster angle, the errors caused by absorption can be completely avoided. In Fig. 4(a) the calculation result accords with the Eq. (13), which means the deduction above is correct.

In critical-angle technique, if the medium is absorptive, there will not have total reflections, shown in Fig. 4(b). Thus, the critical angle cannot be determined by the reflectivity. For the medium without absorption, the reflectance gradient $dR/d\theta$ changes discontinuously at critical angle, but continuously in an absorptive one[16]. Hence, the critical angle is replaced by a quasi-critical angle θ_{qc} at a point of inflexion, where $d^2R/d^2\theta=0$ and the reflectance gradient $dR/d\theta$ is a maximum[16]. This quasi-critical angle could make big errors in the measurement. The errors are approximately linear in n''_m for n''_m up to about 10^{-3}. When bigger than it, the errors are so large that θ_{qc} would not be used as a criterion[16].

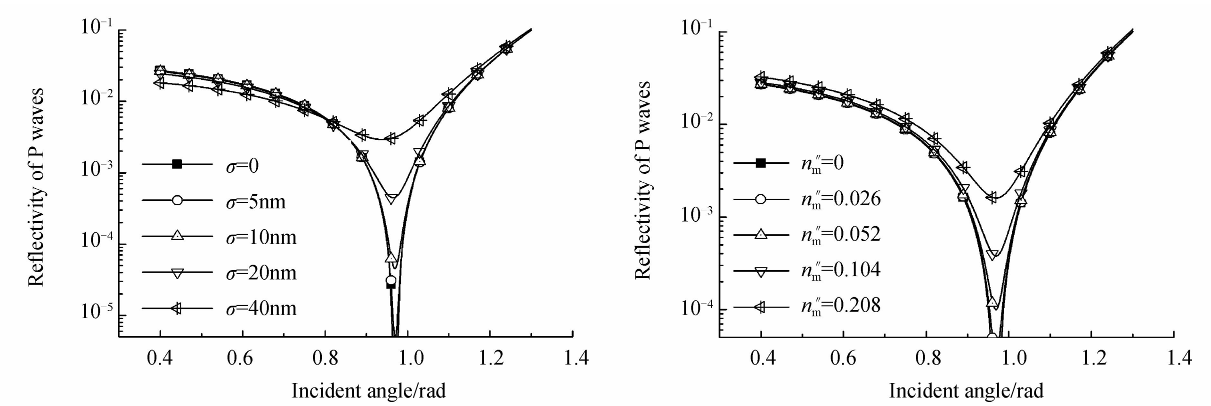
3 Simulation proving

In order to prove the validity of the model, COMSOL Multiphysics, a commercial program that uses Finite Element Method (FEM)[19], is used as a simulation method to acquire the results. This program contains a number of physical simulation modules, our study is based on the Radio Frequency (RF) module. Here, the model of a rough surface is given as an example that shows the process.

In the program, two cuboids and a parametric surface are used to build the shape of the model in a period. To coincide with the effective multilayer model, the parametric surface is set as a sine surface. Above the sine surface is ambient environment, below is the medium under tested. In all the simulations, T_m is set at 100nm, which satisfies the Eq. (1).

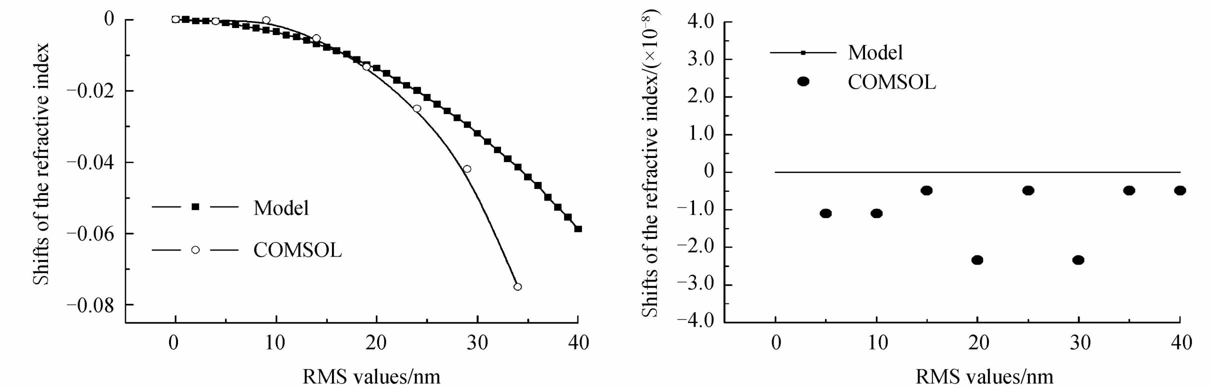
3.1 Brewster-angle technique

The results with different σ and n''_m are given in the Fig. 5(a) and (b) respectively. Curves in Fig. 5(a) and (b) prove that both roughness and absorption would improve reflectivity near Brewster angle, but different from roughness, absorption would not cause the quasi-Brewster shifts. These have a good match with results of the model. The quasi-Brewster shifts are compared by the model and simulation in Fig. 5 (c). Because of the large amount of calculation in COMSOL, only a few points of RMS values are shown in the figure. From the Fig. 5(c), it is clear that the two methods fit better in small RMS values than in large ones. One possible reason for this is that the larger roughness may cause scattering, although it has small spatial wavelength.



(a) Reflectivity of different RMS values with the Brewster-angle technique

(b) Reflectivity of different complex refractive index values with the Brewster-angle technique



(c) Comparison of the shifts of refractive index between model and simulation with the Brewster-angle technique

(d) Comparison of the shifts of refractive index between model and simulation: critical-angle technique

Fig. 5 Results given by Comsol in simulation

3.2 Critical-angle technique

The results given by COMSOL Multiphysics are given in Fig. 5(d). As absorption had been studied by G. H. Meeten[16], only surface roughness is considered. In the simulation, the step length and mesh size can influence the precision. Thus, the promiscuously and tiny shifts (less than 10^{-7}) of the refractive index in the simulation may be regarded as a result of calculation errors. Namely, the small-scale surface roughness hardly cause errors in the refractive index measurements by critical-angle technique.

4 Conclusion

In this article, the results are given in theory. Because the precision is so high that it needs special experimental environment and instruments which are almost unavailable for us. But the results have been proved by simulation and it is meaningful in optical measurement where high precision is needed, since refractive index reading errors is important when they become larger than the refractive index precision of modern instruments, about 10^{-5}[10].

In summary, the effects caused by small-scale surface roughness and absorption have some similarities in Brewster-angle technique. They can both improve reflectivity near Brewster angle. And this can make errors of the refractive index in numerical methods. However, if angles of the minimum reflectivity of P waves are regarded as the Brewster angles in practical experiments, the influence of absorption can be avoided. On the contrary, small-scale surface roughness hardly induces measuring errors in critical-angle technique, but absorption can make significantly shifts of the refractive index. Therefore, it is the best way to measure the refractive index of rough dielectrics by Brewster-angle technique, and absorptive dielectrics by critical-angle technique. These results should be meaningful in many aspects about optical applications, as they reveal the effects of small-scale surface roughness and absorption on Brewster-angle and critical-angle techniques.

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