Haze of surface random systems: An approximate analytic approach

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Approximate analytic expressions for haze (and gloss) of Gaussian randomly rough surfaces for various types of correlation functions are derived within phase-perturbation theory. The approximations depend on the angle of incidence, polarization of the incident light, the surface roughness, σ, and the average of the power spectrum taken over a small angular interval about the specular direction. In particular it is demonstrated that haze (gloss) increase (decrease) with σ/λ as $\exp[-A(\sigma/\lambda)^2]$ and decreases (increase) with a/λ, where a is the correlation length of the surface roughness, in a way that depends on the specific form of the correlation function being considered. These approximations are compared to what can be obtained from a rigorous Monte Carlo simulation approach, and a good agreement is found over large regions of parameter space. Some experimental results for the angular distribution of the transmitted light through polymer films and their haze are presented and compared to the analytic approximations derived in this paper. A satisfactory agreement is found. In the literature haze of blown polyethylene films has been related to surface roughness. Few authors have quantified the roughness and others have pointed to the difficulty in finding the correct roughness measure.

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I. INTRODUCTION

Optical properties of polyethylene films have attracted considerable attention due to the importance in applications such as packaging. Studies of haze and gloss of films have been carried out addressing the effect of polymer structure [1–4], rheological properties [5,6], additives [4,7,8], and processing conditions [2,9–13].

The notion of haze ("cloudiness") is supposed to quantify the ratio between the *diffusely* reflected or transmitted light to the *total* light reflected (reflectance) or transmitted (transmittance). To this end, haze of a film is defined as the fraction of transmitted light that deviates from the directly transmitted beam by more than given amount (e.g., 2.5°) [14–16]. A similar definition applies for reflection. For thin films, haze is recognized to be caused mainly by scattering from surface irregularities, in contrast to bulk randomness. Previously, only a few groups have reported on the dependence of haze on surface roughness [4,12,17]. In a study of different polyethylene materials, the bulk contribution to haze of 40-μm-thick films was found to vary in the range 10–30 % of the total value [4]. Two main mechanisms for surface roughness have been identified [18]: (i) flow-induced irregularities originating from the die (extrusion haze) and (ii) protruding crystalline structure such as lamellae, stacks of lamellae or spherulites (crystallization haze).

Surface roughness is often characterized (in the engineering literature) by the root-mean-square deviation from the mean surface height, σ. Different roughness generating mechanisms, such as die irregularities and inhomogeneous distribution of additives, will influence the roughness at different length scales. Large-scale trends (compared to the wavelength) will not affect the diffuse light scattering and must be eliminated from the analyzes. Implicit difficulties in deriving a relevant measure of surface roughness may obscure its correlation with haze. Various techniques have been applied to characterize roughness. In a number of recent studies atomic force microscopy (AFM) has been applied [4,10,19–21]. Many authors, however, associate only qualitative differences in roughness, as observed by e.g., AFM, with haze values. Robust methods still have to be developed to extract the relevant roughness measure based on a sufficiently high statistics. A discussion of surfaces characterizing and relation to haze is given in [4].

Sukhadia *et al.* [22] related both crystallization haze and extrusion haze to the elastic properties measured as the recoverable shear strain of the polymer. For a low level of melt

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elasticity orientations from the die relax quickly and crystalline aggregates form on or close to the film surface. For highly elastic materials the surface roughness was attributed to melt flow effects generated at the die exit. Plotting haze versus the recoverable shear strain [22] based on a large data set of blown and cast films, results in a parabolic curve with lowest haze for materials of intermediate level of elasticity.

Studies of electromagnetic wave scattering have a long history, and the effect of surface roughness on the scattering has also been studied for many years in optics [23–35] and, more recently, for x-ray scattering [36–40]. Today it is fair to say that the main features are rather well understood, at least for one-dimensional roughness, where one recently has started to address inverse (optical) problems [41,42]. In the case of two-dimensional roughness there are still open questions to be answered, in particular for optical frequencies where the dielectric contrast (and therefore the scattering) is the most pronounced. This paper concentrates on the range of optical frequencies even if the theoretical results derived herein hold for any frequency for which the adopted approximations are expected to hold. This choice is made due to the fact that the concept of haze (and gloss) mainly is used within optics, to the best of our knowledge.

Different approximations are available for scattering of electromagnetic waves scattered by rough surfaces or bulk inhomogeneities, depending e.g., on the typical range of roughness/wavelength ratios (σ/λ), scattering angles, and electrical conductivity [14,36,37,43,44]. Vectorial or scalar formulations are used, depending on whether or not polarization is taken into account. Vectorial formulations, such as the Rayleigh-Rice perturbation theory, are considered to be more accurate for smooth surfaces (typically $\sigma \ll \lambda$) with finite conductivity and large scattering angles. However, the scalar Kirchhoff theory is claimed to be more accurate than Rayleigh-Rice for rougher surfaces, and it is easier to handle mathematically [43].

Some calculations of gloss vs surface roughness parameters have been published over the last years. Alexander-Katz and Barrera [45] calculated the angular distribution of reflected light using the scalar Kirchhoff approximation. With a Gaussian height distribution and various height correlation functions, the gloss depended on two parameters. It increased with decreasing σ and increasing correlation length. The sensitivity of gloss to correlation length depended on σ. In particular, the sensitivity was low for very low and high σ/λ values. Wang *et al.* [20] performed similar calculations. With surface parameters (from AFM) and refractive indices as input, the calculations agreed fairly well with gloss values in the range 30–70 %, measured on PE films.

Haze has been calculated as function of bulk inhomogeneities [14], but theoretical literature on haze vs surface roughness is sparse although there are related studies, e.g., on radio transmission [46]. Willmouth [14] referred to the unpublished geometrical optics calculations relating film clarity to surface roughness on a scale greater than the wavelength of light. In most cases, however, the roughness is on a finer scale, and the calculations must be based on physical optics. The scalar Kirchhoff approximation can, in principle, be applied to transmission calculations [47], but this is more difficult than the reflection case due to contributions from two surfaces, possible bulk effects, and a high probability of multiple scattering [43].

More recently Wang *et al.* [21] presented a method for calculating haze. Since the (real) surfaces were spherulitic, the combined scattering of the two surfaces could be modeled by Mie scattering theory (valid for a single sphere of any size and refractive index). Calculated haze values agreed with experimental data for films of six different materials. Furthermore, the model predicted a maximum in haze for a spherulite diameter of 800 nm ($\lambda = 550$ nm), while the clarity decreased monotonously with increasing spherulite diameter. This critical diameter, corresponding to maximum haze, decreased with increasing refractive index, but it was insensitive to the volume fraction of spheres, in the range studied. Although this model provides insight into the relationship between surface roughness and haze, it has some limitations. In particular, it cannot account for nonspherical protrusions and lateral correlation.

In this paper we study the effect of surface roughness on haze by addressing both the height distribution as well as the height-height lateral correlations. A motivation has been that problems in deriving experimental measures of surface roughness have obscured the correlation with haze [4]. Our main goal is to establish the relationship between surface structure—both roughness amplitude and surface lateral correlations—and haze. An analytical approximation to haze for Gaussian randomly rough surfaces is derived which is compared with a rigorous Monte Carlo simulation approach. Models are compared with experimental results on haze as well as with the angular distribution of the transmitted light through films.

This paper is organized as follows: the following section introduces the scattering geometry to be discussed in this work as well as the properties of the surface roughness. In Sec. III the scattering theory and notation to be used in the following discussion is presented. The main results of this work starts in Sec. IV where the definition and analytic approximate expressions for haze are presented. This approximation is compared to rigorous computer simulations in Sec. V. Finally the conclusions that can be drawn from the present work are presented in Sec. VI

II. SCATTERING GEOMETRY

The scattering geometry that we will consider in this study is depicted in Fig. 1. In the region $z > \zeta(x)$ it consists of vacuum $[\varepsilon_0(\omega)=1]$ and for $z < \zeta(x)$ of a dielectric characterized by an isotropic frequency-dependent dielectric function $\varepsilon_1(\omega)$. Here $\zeta(x)$ denotes the surface profile function. It is assumed to be a single-valued function of x that is differential as many times as is necessary. Furthermore, it constitutes a zero mean stationary Gaussian random process that is defined by

$$\langle \zeta(x) \rangle = 0, \tag{1a}$$

$$\langle \zeta(x)\zeta(x') \rangle = \sigma^2 W(|x - x'|). \tag{1b}$$

Here $W(|x|)$ denotes the (normalized) autocorrelation, or height-height correlation function and will be specified later,

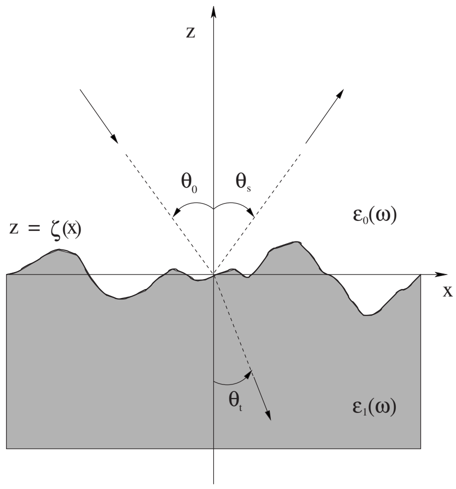

FIG. 1. The scattering geometry used in this study. The rough surface is defined by $z=\zeta(x)$. The region above the surface, $z>\zeta(x)$, is assumed to be vacuum $[\varepsilon_0(\omega)=1]$, while the medium below is a dielectric characterized by a frequency-dependent dielectric function $\varepsilon_1(\omega)$. Notice for which direction the angle of incident (θ_0), scattering (θ_s), and transmission (θ_t) are defined as being positive. An angle of transmission is only well defined if the lower medium is transparent, i.e., if Re $\varepsilon_1(\omega)>0$.

σ is the root mean square (rms) of the surface roughness, and $\langle\cdot\rangle$ denotes the average over an ensemble of realizations of the surface roughness. For the later discussion one will also need the power spectrum of the surface roughness, defined as the Fourier transform of the correlation function, i.e.,

$$g(|k|) = \int dx e^{-ikx} W(|x|). \tag{2}$$

In this paper, we will mainly deal with correlation function of the exponential type, $W(x)=\exp(-|x|/a)$, where a is the so-called correlation length. For such a correlation function the power spectrum becomes

$$g(|k|) = \frac{2a}{1+k^2a^2}. \tag{3}$$

The incident wave will be assumed to be either p or s polarized, as indicated by the subscript ν on field quantities, and the plane of incidence will be the xz plane. Furthermore, the angle of incidence, scattering, and transmission, θ_0, θ_s, and θ_t respectively, are measured positive according to the convention indicated in Fig. 1.

In order to demonstrate that the assumption made above for the statistics of the surface roughness is not unrealistic, we in Fig. 2(a) present an AFM measurement of the surface topography of a polyethylene film surface. The corresponding height distribution and height-height correlation function that can be obtained on the basis of such topography measurements are can be found in Figs. 2(b) and 2(c). It is observed from these figures that the measured surface roughness, to a good approximation, is a Gaussian random process. For this particular example, an exponential correlation function is a reasonable, but not perfect, choice for the correlation function.

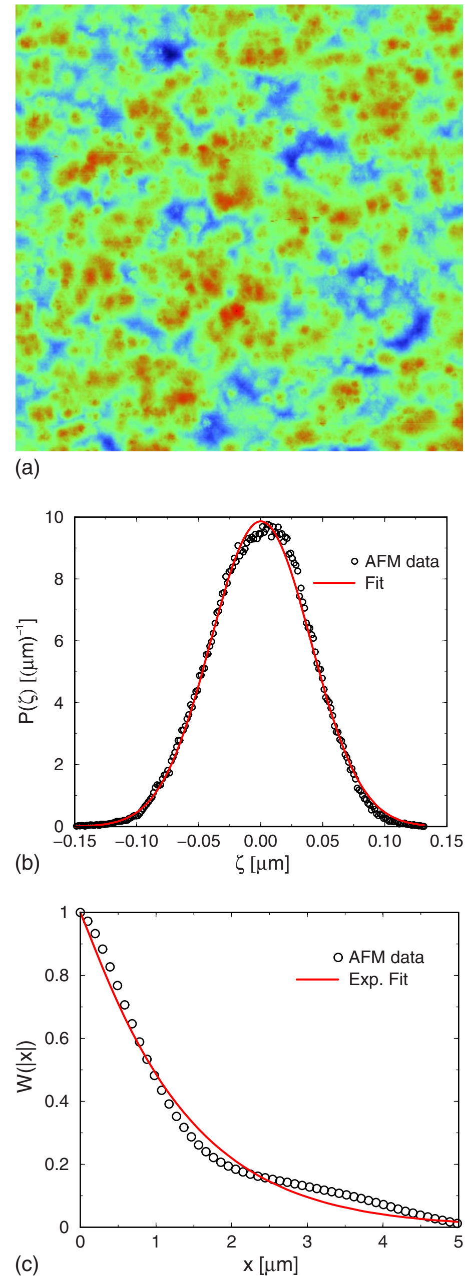

FIG. 2. (Color online) Experimental height measurements obtained from atomic force microscopy (AFM) measurements on a LLDPE film. The material was of the narrow (molecular weight distribution) metallocene grade. (a) A contour plot of the experimental height function measured over a 50×50 μm^2 quadratic area. The color code is so that red corresponds to 0.015 μm and blue to -0.015 μm. (b) The height distribution function $P(\zeta)$ calculated from the AFM data (open circles) and a Gaussian fit (solid line) corresponding to $\sigma=0.04$ μm. (c) The height-height correlation function $W(|x|)$ obtained from the AFM data (open circles) and fitted with an exponential correlation function $W(|x|)=\exp(-|x|/a)$ (solid line) corresponding to a correlation length of $a=1.3$ μm.

III. SCATTERING THEORY

In this section, elements of the scattering theory that will be useful for the discussion that will follow will be presented. A more complete and detailed presentation can be found, e.g., in Ref. [31].

A. Reflection and transmission amplitudes

Due to the one-dimensional character of the surface topography, $z=\zeta(x)$, a generic scalar field may be introduced that fully can describe, together with Maxwell's equations, the electromagnetic field. Such generic field is defined as

$$\phi_\nu(x,z,\omega) = \begin{cases} H_y(x,z|\omega), & \nu = p \\ E_y(x,z|\omega), & \nu = s, \end{cases} \tag{4}$$

where ν is a polarization index and H_y and E_y are used to denote the second component of the electric and magnetic fields. The advantage of using the field variable $\phi_\nu(x,z,\omega)$ is that the Maxwell (vector) equations satisfied by the electromagnetic field are equivalent to the scalar wave equation for $\phi_\nu(x,z,\omega)$. To be able to use a scalar equation instead of vector equations represents a great simplification of the problem. However, it should be noted that this simplification comes about because of $\zeta(x)$ being one-dimensional, and it does not hold true in general.

For the scattering system considered in this paper, the field can be written as

$$\phi_\nu(x,z,\omega) = e^{ikx-i\alpha_0(k)z} + \int_{-\infty}^{\infty} \frac{dq}{2\pi} R_\nu(q|k) e^{iqx+i\alpha_0(q)z} \tag{5a}$$

when $z > \max\, \zeta(x)$ and a plane incident wave is assumed and

$$\phi_\nu(x,z,\omega) = \int_{-\infty}^{\infty} \frac{dq}{2\pi} T_\nu(q|k) e^{iqx-i\alpha_1(q)z} \tag{5b}$$

when $z < \min\, \zeta(z)$. In writing Eqs. (5), we have introduced $R_\nu(q|k)$ and $T_\nu(q|k)$—the reflection and transmission amplitudes, the lateral momentum variable

$$q = \sqrt{\varepsilon}\frac{\omega}{c}\sin\,\theta, \tag{6}$$

where ε is the dielectric constant of the medium considered, and θ denotes the angle of incidence, reflection, or transmission depending on context. Furthermore, in Eqs. (5), we have also defined

$$\alpha_m(q) = \begin{cases} \sqrt{\varepsilon_m \dfrac{\omega^2}{c^2} - q^2}, & |q| < \sqrt{\varepsilon_m}\dfrac{\omega}{c} \\ i\sqrt{q^2 - \varepsilon_m \dfrac{\omega^2}{c^2}}, & |q| > \sqrt{\varepsilon_m}\dfrac{\omega}{c}, \end{cases} \tag{7}$$

where $m=0$ corresponds to the medium above the rough surface and $m=1$ to the dielectric medium below. Notice that when $|q| \leq \sqrt{\varepsilon_m}(\omega/c)$ (nonradiative region), it follows that $\alpha_m(q) = \sqrt{\varepsilon_m}(\omega/c)\cos\,\theta$.

B. Mean differential reflection and transmission coefficients

The mean differential reflection coefficient (DRC) and differential transmission coefficient (DTC) are two experimentally and theoretically accessible quantities frequently used to study the angular distribution of the reflected or transmitted light. We will here denote them by $\langle \partial R_\nu / \partial\theta_s \rangle$ and $\langle \partial T_\nu / \partial\theta_t \rangle$, respectively, where subscript ν, as before mentioned, is a polarization index. The mean DRC is defined as the fraction of the incident power that is scattered by the rough surface into an angular interval $d\theta_s$ about the scattering angle θ_s. The power (energy flux) crossing a plane parallel to the xy plane can be calculated from

$$P = \int dx \int dy\ \mathrm{Re}\langle \mathbf{S}_3 \rangle_t, \tag{8}$$

where $\mathbf{S} = 1/2\mathbf{E} \times \mathbf{H}^*$ is the (complex) Poyntings vector [48] for the electromagnetic field $(\mathbf{E},\mathbf{H})$ and $\langle \cdot \rangle_t$ denotes a time average. By using Eqs. (5) one finds that the incident power is given by

$$P_{inc} = \frac{L_1 L_2}{2}\frac{c^2}{\omega}\alpha_0(k), \tag{9a}$$

while the scattered power becomes

$$P_{sc} = \varepsilon_0 \frac{L_2}{2}\frac{c^2}{\omega}\int_{-\sqrt{\varepsilon_0}\omega/c}^{\sqrt{\varepsilon_0}\omega/c} \frac{dq}{2\pi}\alpha_0(k)|R_\nu(q|k)|^2 = \int_{-\pi/2}^{\pi/2} d\theta_s p_{sc}(\theta_s). \tag{9b}$$

This latter relation implicitly defines the angular-dependent scattered power $p_{sc}(\theta_s)$. Thus from the definition of the differential reflection coefficient it is realized that $\partial R_\nu / \partial\theta_s = p_{sc}(\theta_s)/P_{inc}$. However, since the surface is randomly rough, it is the *mean* differential reflection coefficient that should be of interest. Such a quantity will be given by

$$\left\langle \frac{\partial R_\nu}{\partial\theta_s} \right\rangle = \left\langle \frac{p_{sc}(\theta_s)}{P_{inc}} \right\rangle = \frac{\sqrt{\varepsilon_0}}{L_1}\frac{\omega}{2\pi c}\frac{\cos^2\,\theta_s}{\cos\,\theta_0}\langle |R_\nu(q|k)|^2 \rangle, \tag{10}$$

where we have used $\langle \cdots \rangle$ to denote the average over surface realizations. In the same way, the mean differential transmission coefficient may be defined as $\langle \partial T_\nu / \partial\theta_t \rangle = \langle p_{tr}(\theta_t)/P_{inc} \rangle$, where $p_{tr}(\theta_t)$ denotes the power transmitted into an angular interval about the angle of transmission θ_t. Instead of giving the expression for this quantity explicitly, we will instead for later convenience, introduce a generic notation for both the mean differential reflection and transmission coefficients. This generic quantity will be denoted by $\langle \partial U / \partial\theta \rangle$ so that

$$\left\langle \frac{\partial U}{\partial\theta} \right\rangle = \begin{cases} \left\langle \dfrac{\partial R_\nu}{\partial\theta_s} \right\rangle & \text{in reflection} \\ \left\langle \dfrac{\partial T_\nu}{\partial\theta_t} \right\rangle & \text{in transmission,} \end{cases} \tag{11a}$$

where θ stands for θ_s and θ_t in reflection and transmission, respectively. In general it may be written in the following form:

$$\left\langle \frac{\partial U}{\partial \theta} \right\rangle = \frac{1}{L_1} \frac{\varepsilon_m}{\sqrt{\varepsilon_0}} \frac{\omega}{2\pi c} \frac{\cos^2 \theta}{\cos \theta_0} \langle |U(q|k)|^2 \rangle. \tag{11b}$$

In Eq. (11b) $U(q|k)$ denotes the reflection or transmission amplitudes depending on context, and m takes on the values $m=0$ in reflection and $m=1$ in transmission.

In theoretical studies it is customary to separate $\langle \partial U / \partial \theta \rangle$ into two terms—one coherent term and one incoherent term. That this is possible can be realized from the following (trivial) rewriting:

$$\langle |U(q|k)|^2 \rangle = |\langle U(q|k) \rangle|^2 + [\langle |U(q|k)|^2 \rangle - |\langle U(q|k) \rangle|^2]. \tag{12}$$

If this expression is substituted back into Eq. (11b), the first term will give rise to the *coherent* or specular contribution to $\langle \partial U / \partial \theta \rangle$, while the last term, within the square brackets, will result in the *incoherent* or diffuse contribution to the same quantity. We will use subscripts *coh* and *incoh*, respectively, to indicate these whenever needed. This identification follows from observing that in the average $\langle U(q|k) \rangle$ one only gets contributions from those portions of $U(q|k)$ that are *in phase* from one surface realization to another.

From Eq. (11b) a quantity that defines the fraction of the incident energy that is either reflected or transmitted can be defined as

$$\mathcal{U} = \int_{-\pi/2}^{\pi/2} d\theta \left\langle \frac{\partial U}{\partial \theta} \right\rangle. \tag{13}$$

Notice, that when no absorption takes place in neither media involved, i.e., Im $\varepsilon_m=0$, one should have that the sum of this quantity in reflection and transmission should add up to one, $\mathcal{U}_s+\mathcal{U}_t=1$. This is a direct consequence of energy conservation. In practice, however, there will always be some absorption, but for many dielectric media at optical frequencies it is a reasonable approximation to neglect it.

IV. HAZE

In the optical industry two quantities—*haze* and *gloss*—are often used to quantify the visual appearance of materials [49]. Gloss, crudely speaking, is related to the amount of light being reflected (or transmitted) into angles *around* the specular direction. This quantity has previously been studied experimentally [50] for transparent plastic materials and recently also studied theoretically [45]. In this latter study, the authors investigated how gloss depends on the level of roughness and surface correlations. It was found that the incoherent (diffuse) contribution to the scattered light could contribute significantly to the gloss. Haze, on the other hand, measures the fraction of reflected (transmitted) light that is reflected (transmitted) *away* from the specular direction [15,16]. In the former case one talks of *haze in reflection* and in the latter of *haze in transmission*. Notice that haze can almost be considered as a complementary quantity to gloss. If the haze of a transparent plastic film, say, is large, then an object viewed through the film will look unsharp or blurry. It is this kind of visual effect that the haze value is supposed to quantify.

Even though gloss is important in many applications, we will in the present study concentrate on haze since this quantity has not been studied that extensively in the literature by theoretically means.

A. Definition of haze

For the purpose of this theoretical study, we will focus on a semi-infinite medium, instead of a film geometry. The reason for this choice is purely practical. Let us start by assuming that the incident light is impinged onto the planar mean surface at an angle θ_0 measured counterclockwise from the normal to the mean surface (cf. Fig. 1). In case of no surface roughness the light will be scattered (or transmitted) in accordance with Snell's law. Hence all the energy will be propagate in the directions defined by the angle

$$\Theta = \arcsin\left(\frac{\sqrt{\varepsilon_m}}{\sqrt{\varepsilon_0}} \sin \theta_0 \right), \tag{14}$$

where $m=0$ should be used in reflection (for which $\Theta=\theta_0$) and $m=1$ in transmission.

Formally, haze, $\mathcal{H}(\theta_0)$, is defined [15,16,51] as the *fraction* of the reflected (transmitted) light that is reflected (transmitted) into angles lying *outside* the angular interval (θ_-, θ_+) with

$$\theta_\pm = \Theta \pm \Delta\theta, \tag{15}$$

and where $\Delta\theta$ is an angular interval to be defined. In commercially available haze meters [15,16,51], one for this angular interval uses the value $\Delta\theta=2.5°$ [52], and for the present study this value will be adopted. Notice that the angles $\theta_\pm$ can be related to the lateral momentum variable, $q_\pm$, in accordance with Eq. (6). Furthermore, the angular interval $\Delta\theta$ is related to a corresponding momentum interval in the following way:

$$\Delta q = \frac{1}{2}(q_+ - q_-) = \sqrt{\varepsilon_m}\frac{\omega}{c}\cos \Theta \sin \Delta\theta. \tag{16}$$

Haze, as defined above, can readily be related to the mean differential reflection or transmission coefficients $\langle \partial U / \partial \theta \rangle$ (cf. Sec. III B). In terms of these quantities, haze that in general will depend on the angle of incidence θ_0 can be written in the form

$$\mathcal{H}(\theta_0) = I\left(-\frac{\pi}{2}, \theta_-\right) + I\left(\theta_+, \frac{\pi}{2}\right) \tag{17a}$$

$$= 1 - I(\theta_-, \theta_+), \tag{17b}$$

where one has introduced

$$I(\theta_a, \theta_b) = \frac{1}{\mathcal{U}} \int_{\theta_a}^{\theta_b} d\theta \left\langle \frac{\partial U}{\partial \theta} \right\rangle, \tag{17c}$$

and where $\mathcal{U}$ has been defined earlier in Eq. (13). In the transition from Eq. (17a) to Eq. (17b) it has been used that $I(-\pi/2, \pi/2)=1$ by definition. The reason for the presence of the factor $1/\mathcal{U}$ in Eq. (17c) is that haze is defined in terms of the fraction of the reflected or transmitted light, while

$\langle \partial U/\partial\theta\rangle$ is defined as the fraction of the incident power. Hence, this factor is present to ensure that the defining expression of haze has been given the correct normalization. From the definition [Eqs. (17)], it follows that haze is a dimensionless number between zero and one [53]. Furthermore, notice that an ideal scattering system, i.e., one with no surface or bulk randomness, will correspond to a haze value of $\mathcal{H}(\theta_0)=0$ for all angles of incidence since all light will be reflected or transmitted into the specular direction [54]. However, as the randomness of the scattering system is increased, and therefore the reflected or transmitted intensities as a consequence become more and more diffuse, the corresponding haze value will increase. To reach a haze of one ($\mathcal{H}=1$) is, however, rather unlikely for random systems encountered in practical situations since it will require a vanishing intensity over the angular interval (θ_-,θ_+). Surface random systems with such a property can, however, be artificially manufactured [41]. From a practical point of view, a more likely scenario for a strongly random system is probably that of a Lambertian diffuser [55], i.e., a scattering system giving raise to $\langle \partial U/\partial\theta\rangle \propto \cos\theta$ independent of angle of incidence. For such a scenario, haze at normal incidence will be $\mathcal{H}(0)=1-\sin\Delta\theta \simeq 0.956<1$ according to definition (17). For naturally occurring surfaces this is probably a more realistic upper limit of haze.

B. Naive approximation to haze

In many situations encountered in practical applications of the concept of haze, it is illuminative to have available an approximate expression to the formal definition [Eq. (17)]. Often the dependence on various parameters on that haze will depend, can be made more apparent via approximate expressions.

Before considering an analytic approximation to haze (see next subsection) we will present some approximations that are based on the concept of coherent and incoherent scattering. Such picture is often useful to bear in mind when working with the haze (and gloss) concept.

If the scattering system is not too diffuse, the main contribution to the I integral present in Eq. (17b) will come from the coherent component of the scattered or transmitted light. Furthermore, since the coherent component is nonzero outside the angular interval from θ_- to θ_+, one arrives at the following approximation to haze:

$$\mathcal{H}(\theta_0) \simeq 1-\frac{1}{\mathcal{U}}\int_{-\pi/2}^{\pi/2} d\theta \left\langle \frac{\partial U}{\partial\theta}\right\rangle_{coh}, \tag{18a}$$

based exclusively on the coherent component of the scattered or transmitted field. Thus, it is natural to refer to Eq. (18a) as a *coherent approximation to haze*. From a mathematical point of view, one may rewrite Eq. (18a) by noting that the last term is just one minus the corresponding incoherent component. By recalling Eqs. (12) and (13) one, therefore, arrives at the following equivalent *incoherent approximation to haze*:

$$\mathcal{H}(\theta_0) \simeq \frac{1}{\mathcal{U}}\int_{-\pi/2}^{\pi/2} d\theta \left\langle \frac{\partial U}{\partial\theta}\right\rangle_{incoh}. \tag{18b}$$

Later it will be shown that the coherent approximation [Eq. (18a)] only takes into account the surface roughness and *not* its correlation. The incoherent approximation [Eq. (18b)] however, will be shown to *also* depend on the surface correlation. Even though the coherent and incoherent approximations to haze are equivalent from a purely mathematical point of view, they give rise to different physical interpretations. Since Eq. (18b) includes the dependence on the surface correlation, as well as surface roughness, we will prefer this approximation over that of Eq. (18a).

An approximation is not worth much without information about its range of validity. In order to get an idea of the accuracy of the coherent and incoherent approximations, the Lambertian diffuser will be considered once more. Such a diffuser represents in many ways a worst case scenario since there is no coherent component at all in this particular case. Due to the vanishing coherent component, one therefore, within the approximations of Eqs. (18), has that $\mathcal{H}\simeq 1$. However, above one found by using the formal definition of haze [Eqs. (17)] that $\mathcal{H}\simeq 0.956$. Hence, the error obtained by calculating the haze from the approximate expressions (18) is of the order of 4.5% for this highly diffusive case. For less diffusive surfaces the error is expected to be less.

C. Analytic approach to haze

In this subsection, an analytic expressions for haze will be derived. This will be achieved by applying the so-called phase-perturbation theory [56,57]. This approximation is reviewed in the Appendix, and it can, at least in reflection, be viewed as an extension (or correction) to the more well-known (and used) Kirchhoff approximation [24–27]. In particular, phase-perturbation theoretical results reduce in the limit of large correlation length for the surface roughness to those that can be obtained from Kirchhoff theory. In addition, phase-perturbation theory naturally can handle transmission problems in an analytic fashion, while such a generalization is not straight forward for the Kirchhoff approximation.

Within phase-perturbation theory, the mean DRC or DTC, $\langle \partial U/\partial\theta\rangle$, can be written in the following form (cf. Appendix):

$$\left\langle \frac{\partial U}{\partial\theta}\right\rangle = \frac{1}{L}\frac{\varepsilon_m}{\sqrt{\varepsilon_0}}\frac{\omega}{2\pi c}\frac{\cos^2\theta}{\cos\theta_0}|u_0(k)|^2 J(q|k), \tag{19a}$$

with $u_0(k)$ being the polarization-dependent Fresnel reflection or transmission coefficients [30] corresponding to the scattering system of a planar (nonrough) interface and

$$J(q|k) = Le^{-\sigma^2\Lambda^2(q|k)}\int_{-\infty}^{\infty} du e^{i(q-k)u} e^{\sigma^2\Lambda^2(q|k)W(u)}, \tag{19b}$$

where

$$\Lambda(q|k) = \begin{cases} \alpha_0(q)+\alpha_0(k) & \text{in reflection} \\ \alpha_1(q)-\alpha_0(k) & \text{in transmission} \end{cases} \tag{19c}$$

denotes the momentum transfer perpendicular to the mean surface. In writing Eqs. (19), we recall that σ and $W(u)$ are the surface roughness and correlation function, respectively [cf. Equations (1)], and that L denotes the length of the rough surface measured along the mean surface. Notice, that for a planar surface ($\sigma=0$), $J(q|k)$ will be proportional to $\delta(q-k)$, and hence the effect of the surface roughness in Eq. (19a) is fully contained in the $J(q|k)$ integral.

In order to derive an approximate analytic expression to haze, we start by assuming that $\sigma^2|\Lambda^2(q|k)|\ll 1$ for all values of q in the radiative region defined by $|q|\le\sqrt{\varepsilon_m}\omega/c$. Within this approximation, the J integral takes on the following form:

$$J(q|k) \simeq Le^{-\sigma^2\Lambda^2(q|k)}[2\pi\delta(q-k)+\sigma^2\Lambda^2(q|k)g(|q-k|)],$$

$$\sigma^2|\Lambda^2(q|k)|\ll 1, \tag{20}$$

where Eq. (2) has been used to relate $W(|x|)$ to the power spectrum $g(|k|)$. It should be noticed that the first term of Eq. (20) is a coherent (specular) contribution, while the last term represents the lowest order contribution from the incoherent (diffuse) field. Moreover, the coherent term depends on the parameter that defines the surface roughness only through the rms roughness, σ, while the incoherent contribution in addition shows a dependence on the correlation length a via the power spectrum $g(|k|)$.

In order to calculate haze, we need to get an expression for the (integrated) fraction of the incident power that is either reflected or transmitted into any angle, i.e., one is looking for an expression for $\mathcal{U}$ as defined by Eq. (13). The main effect of surface roughness is to alter the angular distribution of the light being reflected from or transmitted through a randomly rough surface. However, the amount of integrated reflected or transmitted light is much less sensitive to the presence of surface roughness. Notice that this is only true if we are not close to a roughness induced surface resonance such as, e.g., those due to the excitations of surface plasmon polaritons [58]. Another situation where the above assumption is known to fail is in situations where the scattered or transmitted intensity is rather low for the planar surface so that the presence of roughness might renormalize this result in a significant way. This is for instance the case for the Brewster angle phenomenon [30]. Hence, for the purpose of this study, it will be assume that $\mathcal{U}$ shows little sensitivity to surface roughness so that it can be well approximated by the planar result. With Eq. (19a) and $\sigma=0$ one therefore has

$$\mathcal{U} \simeq \frac{\alpha_m(k)}{\alpha_0(k)}|u_0(k)|^2, \tag{21}$$

where one, as before, should use $m=0$ in reflection and $m=1$ in transmission.

By substituting Eqs. (20) and (21) into Eqs. (19) an approximate result for the I integrals, Eq. (17c), used to define haze, can be obtained as

$$I(\theta_-,\theta_+) \simeq \frac{1}{L}\int_{q_-}^{q_+}\frac{dq}{2\pi}\frac{\alpha_m(q)}{\alpha_m(k)}J(q|k) \simeq e^{-\sigma^2\Lambda^2(k|k)} + \int_{q_-}^{q_+}\frac{dq}{2\pi}\sigma^2\Lambda^2(q|k)\frac{\alpha_m(q)}{\alpha_m(k)}e^{-\sigma^2\Lambda^2(q|k)}g(|q-k|), \tag{22}$$

where $q_\pm=k\pm\Delta q$. To try to obtain an estimate for the last integral of Eq. (22) will now be approached. Since the momentum interval Δq is small [cf. Eq. (16)], $\alpha_m(q)$ and $\Lambda(q|k)$ are therefore slowly varying functions of q in the interval $q_-\le q\le q_+$. The power spectrum $g(|q|)$, however, can in particular for large correlation lengths, $\Delta qa\gg 1$, become strongly q dependent over the interval of interest even for small values of Δq. Thus, the integrand of the last term of Eq. (22), accept the power spectrum $g(|p|)$, can be approximated by its value at $q=k$ (specular direction). Hence, one may write

$$I(\theta_-,\theta_+) \simeq e^{-\sigma^2\Lambda^2(k|k)}\left[1+\sigma^2\Lambda^2(k|k)\frac{G(a)\Delta q}{\pi}\right], \tag{23}$$

where

$$G(a) = \frac{1}{2\Delta q}\int_{q_-}^{q_+}dq g(|q-k|) = \frac{1}{2\Delta q}\int_{-\Delta q}^{\Delta q}dq g(|q|) \tag{24}$$

denotes the *power spectrum factor* calculated over a momentum interval of half width Δq around zero momentum transfer $q-k=0$. It is important to realize that $G(a)$ is independent of the angle of incidence but will, of course, depend on the type of power spectrum used for the surface roughness. Hence, when the type of power spectrum is known, $G(a)$ can be calculated for *all* angles of incidence, as well as for both reflection and transmission. For an exponential power spectrum, defined by Eq. (3), the power spectrum factor becomes

$$G(a) = \frac{2}{\Delta q}\arctan(\Delta qa), \tag{25}$$

and for a Gaussian power spectrum, $g(|q|)=\sqrt{\pi}a\times\exp(-q^2a^2/4)$, often used in practice, $G(a)=(\pi/\Delta q)\mathrm{erf}(\Delta qa/2)$ where $\mathrm{erf}(\cdot)$ is the error function [59]. Notice that whenever $\Delta qa\ll 1$, the power spectrum factor may be expanded with the result that $G(a)$ is directly proportional to the power spectrum at zero momentum; $G(a)\simeq g(0)$. In general, however, this is not the case.

In the spirit of phase-perturbation theory [56,57,60], one observes that the terms in square brackets in Eq. (23) are the first few (nontrivial) terms of an exponential function. Hence, one may write

$$I(\theta_-,\theta_+) \simeq \exp\left[-\sigma^2\Lambda^2(k|k)\left(1-\frac{G(a)\Delta q}{\pi}\right)\right], \tag{26}$$

and after substituting this expression back into Eq. (17b), the following approximate expression for haze has finally arrived at

$$\mathcal{H}(\theta_0) = 1 - \exp\left[-\sigma^2\Lambda^2(k|k)\left(1 - \frac{G(a)\Delta q}{\pi}\right)\right]. \tag{27}$$

Here the perpendicular momentum transfer, $\Lambda(q|k)$, has already been defined in Eq. (19c), the power spectrum factor $G(a)$ in Eq. (24), and the momentum interval Δq in Eq. (16). Approximation (27) and its numerical confirmation (to be presented later), are the main results of this study.

There are several important observations to be made from the approximate expression to haze (27): First, it should be observed that according to Eq. (27) haze can be written in terms of *two dimensionless quantities*: $\sigma\Lambda(k|k)$ and $G(a)\Delta q$. The product of the rms roughness of the surface topography and the perpendicular momentum transfer of the reflection (or transmission) process, $\sigma\Lambda(k|k)$, does depend on the "amount" of roughness *but not* on how it is being correlated. The quantity $G(a)\Delta q$, on the other hand, depends on the power spectrum, and is therefore sensitive to the type of height-height correlation function respected by the rough surface. Second, the dependence on the angle of incidence *only* enters through the momentum transfer: $\sigma\Lambda(k|k)$ (perpendicular) and Δq (lateral). Third, to apply approximation (27) in practical applications, it is the power spectrum of the surface roughness around zero momentum that is of interest since only this portion of the power spectrum enters the definition of the power spectrum factor $G(a)$. This is important to realize since the whole power spectrum, and in particular its tail, is often difficult to assess in a reliable way from direct measurements of the surface topography [26].

Another quantity used frequently by the optical industry to quantify visual appearance of surfaces are, as mentioned earlier, gloss [49]. Gloss is, crudely speaking, a measure of how specular a surface appears in reflection or transmission. This can be measured by, e.g., the fraction of the reflected (transmitted) light that is reflected (transmitted) into in a small angular interval around the specular direction Θ. Hence, gloss is more or less complementary to haze, i.e., it can more or less be written as $1-\mathcal{H}(\theta_0)$. Hence, the last term of Eq. (27) may therefore be considered as an expression for gloss. In practice one talks of at least two types of gloss, wide angle gloss and specular gloss [49]. They are distinguished by the values of $\Delta\theta$ (and therefore Δq) used to define them. When using the expression for gloss described above, one has to substitute the correct values for $\Delta\theta$ (as well as θ_0). The expression for gloss, $1-\mathcal{H}(\theta_0)$, is probably best suited for specular gloss due to the approximation introduced in order to arrive at Eq. (23). However for large values of $\Delta\theta$ it is not expected to work too well.

Previously, Alexander-Katz and Barrera [45], while studying gloss (in reflection), found that the reduced variables for this problem were $(\sigma/\lambda)\cos\theta_0$ and $(a/\lambda)\cos\theta_0$. The results of these authors, have quite a few common features with those presented in this study. However, this is probably not so surprising since gloss and haze can be viewed as more or less complementary quantities. In particular, in reflection, we find that $\sigma\Lambda(k|k)$ indeed scales as $(\sigma/\lambda)\cos\theta_0$ [cf. Eq. (29a) below]. Hence, this agrees with the result found by Alexander-Katz and Barrera for gloss [45]. However, for the correlation function dependent quantity, we do not in general seem to agree with these authors. Only in the limit $\Delta qa \ll 1$, when $G(a)\simeq g(0)a$ as mentioned earlier, do we tend to get the same scaling behavior as reported previously by Alexander-Katz and Barrera. The reason for this "discrepancy," is related to the level of accuracy applied in the approximation of the integral of Eq. (22).

When $|\sigma\Lambda(k|k)| \ll 1$, the exponential function of Eq. (27) can be expanded with the result that

$$\mathcal{H}(\theta_0) \simeq \sigma^2\Lambda^2(k|k)\left(1 - \frac{G(a)\Delta q}{\pi}\right), \quad |\sigma\Lambda(k|k)| \ll 1. \tag{28}$$

This has the consequence that the ratio of haze in reflection $[\mathcal{H}_s(\theta_0)]$ and transmission $[\mathcal{H}_t(\theta_0)]$ will be independent of the rms roughness σ and only depend on the correlation length a (and the form of the power spectrum) as well as the parameters defining the scattering geometry.

For completeness, the expressions for haze in terms of the "defining" quantities will be explicitly given. With Eqs. (16), (19c), and (24) one has for reflection

$$\mathcal{H}_s(\theta_0) \simeq 1 - \exp\left[-16\pi^2\varepsilon_0\left(\frac{\sigma}{\lambda}\right)^2 \times\cos^2\theta_0\left\{1 - 2\sqrt{\varepsilon_0}\frac{G(a)}{\lambda}\sin\Delta\theta\cos\theta_0\right\}\right], \tag{29a}$$

while in transmission the haze can be approximated by

$$\mathcal{H}_t(\theta_0) \simeq 1 - \exp\left[-4\pi^2\varepsilon_0\left(\frac{\sigma}{\lambda}\right)^2 \times\left\{\sqrt{\frac{\varepsilon_1}{\varepsilon_0} - \sin^2\theta_0} - \cos\theta_0\right\}^2 \times\left\{1 - 2\sqrt{\varepsilon_1}\frac{G(a)}{\lambda}\sin\Delta\theta\sqrt{1 - \frac{\varepsilon_0}{\varepsilon_1}\sin^2\theta_0}\right\}\right]. \tag{29b}$$

In obtaining Eq. (29b) it has been used that for the specular direction (in transmission) $\cos\Theta_t = \sqrt{1-(\varepsilon_0/\varepsilon_1)\sin^2\theta_0}$.

Before closing this section, it should be stressed that the approximate expression to haze, Eq. (27) and related expressions, are based on phase-perturbation theory. Hence, its validity will become questionable when multiple scattering starts to contribute significantly to the scattered or transmitted fields. We stress that phase-perturbation theory is not a small amplitude perturbation theory, so it may still give reliable results for strongly rough surfaces in the large correlation length limit (i.e., small slopes). The accuracy of Eq. (27) will be investigated in Sec. V.

D. Gloss

In addition to haze, gloss is a quantity frequently used in the optical industry to quantify optical materials. As haze quantifies the fraction of the reflected or transmitted light that is directed outside a given angular interval, gloss, on the other hand, is related to the amount of light that falls inside the same interval.

Mathematically, gloss, $\mathcal{G}(\theta_0)$, is defined as the integral $I(\theta_+,\theta_-)$ of Eq. (17c) with $\theta_\pm$ adapted to the particular definition of gloss of interest. Hence, in terms of the surface parameters, gloss can be expressed by Eq. (22), or by the subsequent approximate expressions that followed it. In particular take notice of Eq. (26) that represents an approximate formula for gloss

$$\mathcal{G}(\theta_0) \simeq \exp\left[-\sigma^2\Lambda^2(k|k)\left(1-\frac{G(a)\Delta q}{\pi}\right)\right]. \qquad (30)$$

In light of Eq. (27), one, as expected, observes that $\mathcal{H}_i(\theta_0)+\mathcal{G}_i(\theta_0)=1$, where $i=s$(reflection), t(transmission) and $\theta_\pm$ are the same for both haze and gloss. This means that when the value of haze is increasing, gloss is reduced and visa versa. We stress that both quantities in the equation above refer to reflection or transmission. Mathematically there is no problem in consider gloss in transmission, however, this term is not commonly used in practical applications. [61] For the implications and validity of the above approximate expression to gloss [Eq. (30)], the interested reader is referred to a separate publication [62].

V. SIMULATION RESULTS AND DISCUSSION

A. Comparison to Monte Carlo simulations

The approximate expression for haze [Eq. (27)] is based on phase-perturbation theory and is therefore not rigorous. In order to investigate how well this analytic approximation is performing, and when this scaling form breaks down, it will be compared to what can be obtained from a rigorous computer simulation approach [31,63]. Such an approach is formally exact, since it solves the Maxwell equations numerically without applying any approximations. It will thus take into account any higher order scattering process and not just those accounted for by phase-perturbation theory.

Such exact Monte Carlo simulations can be performed by formulating the Maxwell equations as a coupled set of integral equations. This is done by taking advantage of Green's second integral identity in the plane as well as the boundary conditions satisfied by the fields and their normal derivative on the randomly rough surface. These integral equations can be converted into matrix equations and solved for the sources—the fields and their normal derivative evaluated at the surface. From the knowledge of these sources, the scattered (transmitted) field at any point above (below) the surface may be calculated and there from the mean DRC (DTC). The whole detailed procedure for doing such Monte Carlo simulations can be found in Refs. [31,63,64].

In the numerical simulation results for haze to be presented below, one will first calculate $\langle \partial U/\partial\theta\rangle$ and then, by numerical integration, calculate haze directly from Eq. (17). An example of a mean differential reflection coefficient curve, obtained by numerical simulations for a polymer material, is given in Fig. 3. In obtaining this result one assumed a Gaussian height distribution function of $\sigma/\lambda=0.058$ and an exponential correlation function of correlation length $a=1.58\lambda$. These parameters are similar to those found for the measured surface of Fig. 2. The vertical dash-dotted lines that can be seen in Fig. 3 are at an angular position $\theta_\pm=2.5°$, i.e., at the angles about the specular direction $\theta_s=\theta_0=0°$ used in the definition of haze. By numerical integration one may from this result calculate haze, and for this particular example one finds $\mathcal{H}_s(\theta_0=0°)=0.34$.

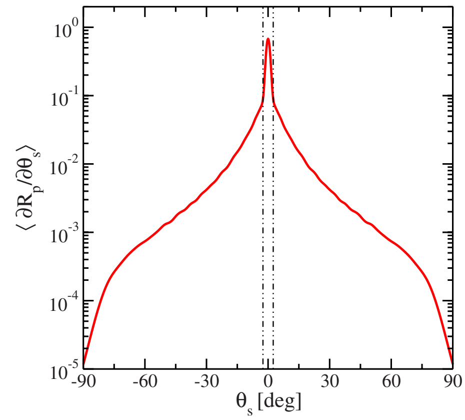

FIG. 3. (Color online) The mean differential reflection coefficient, $\langle \partial R_p/\partial\theta_s\rangle$, vs the scattering angle θ_s as obtained by rigorous Monte Carlo simulations. The incident p-polarized light of wavelength $\lambda=0.6328$ μm was incident at normal incident ($\theta_0=0°$) onto the rough dielectric surface, and for the incident wave a finite sized beam of half-width $g=6.4$ μm was used in order to reduce end effects. This surface of length $L=25.6$ μm$=40.5\lambda$ separates vacuum, above the surface, from the dielectric medium of dielectric constant $\varepsilon_1(\omega)=2.25$ below the surface. The statistical properties of the randomly rough surface was characterized by a Gaussian height distribution function of (rms) width $\sigma=0.037$ μm$=0.058\lambda$ and an exponential correlation function of correlation length $a=1$ μm $=1.58\lambda$. The surface was discretized at $N_\zeta=500$ equally distributed points, and the result was averaged over $N_\zeta=5000$ surface realizations. The vertical dash-dotted lines are at $\theta_\pm=\pm 2.5°$. Notice the specular (coherent) peak around $\theta_s=\theta_0$. The haze in reflection for this surface is according to the numerical simulations $\mathcal{H}(\theta_0)=0.33$, while the prediction of Eq. (27) is $\mathcal{H}(\theta_0)=0.34$.

In Figs. 4 rigorous numerical simulation results for haze (open symbols) are presented versus various parameters of the scattering geometry in both reflection and transmission. Here an exponential correlation function $W(|x|)=\exp(-|x|/a)$ that is a reasonable fit to measured data for some polymer films (see Figs. 2) has been assumed. Furthermore, the light of wavelength $\lambda=0.6328$ μm was incident normally onto the mean surface [Figs. 4(a) and 4(b)], and for the dielectric constant of the media involved, $\varepsilon_0=1$ and $\varepsilon_1=2.25$, were used. For all simulations, the results were averaged over at least $N_\zeta=500$ surface realizations. The vertical dashed-dotted lines present in Figs. 4(a) and 4(b) correspond to the roughness parameters used in obtaining the results of Fig. 3. The solid lines in Figs. 4 are the predictions of Eq. (27)—the approximate expression to haze. In particular, Fig. 4(a) presents the dependence of haze vs σ/λ for a fixed value of the correlation length $a/\lambda=1.58$ and the angle incidence was $\theta_0=0°$. This value for a/λ corresponds to the vertical dashed-dotted line in Fig. 4(b). From Fig. 4(a) it is observed that the analytic approximations (solid lines) performs im-

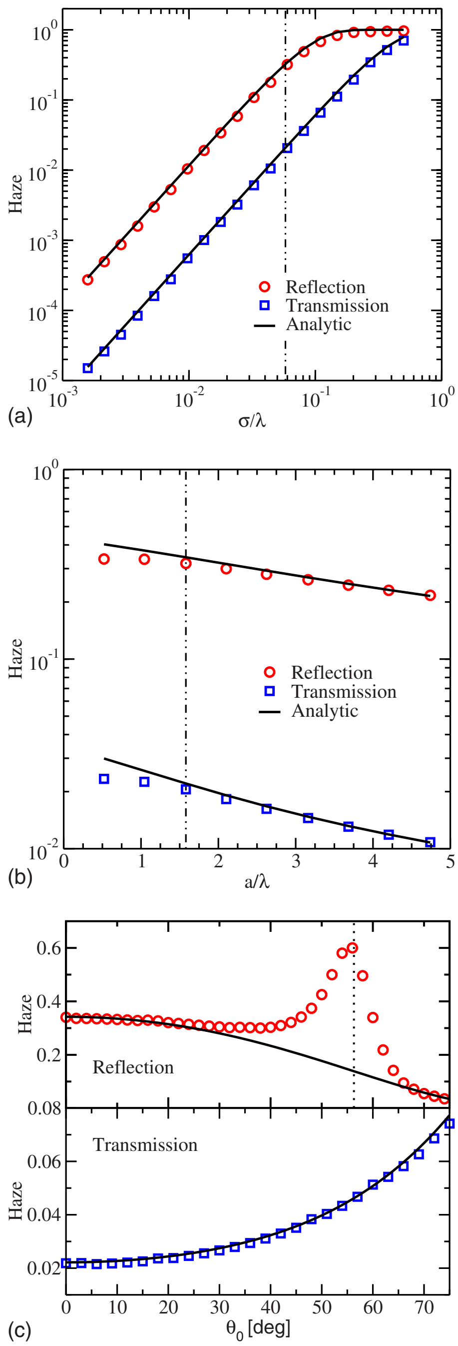

FIG. 4. (Color online) Haze, $\mathcal{H}$, as a function of (a) the surface roughness σ/λ for $a/\lambda=1.58$ at normal incidence, (b) the correlation length a/λ for $\sigma/\lambda=0.058$ at normal incidence, and (c) the angle of incidence θ_0 for $\sigma/\lambda=0.058$ and $a/\lambda=1.58$. For all figures the wavelength of the p-polarized incident light was $\lambda=0.6328$ μm. The open symbols are results of rigorous Monte Carlo simulations, while the solid lines are the predictions of Eq. (27). The dashed-dotted line in the upper panel (reflection) of (c) corresponds to the position of the Brewster angle $\theta_0=\theta_B$ determined by $\tan^2\theta_B=\varepsilon_1/\varepsilon_0$.

pressing well, also for the roughest surfaces considered. It should be noted that both the $(\sigma/\lambda)^2$ increase in haze for low levels of roughness [cf. Eq. (28)] as well as the "bend-off" or saturation that takes place for the haze for strong roughness seem to be correctly predicted by Eq. (27). In this latter case, however, the numerical simulation results saturated around $\mathcal{H}\simeq 0.95-0.96$, while the theoretical curves approach the value of one. Hence, for strongly rough surfaces one has a relative error of about 5% as discussed in an earlier section.

In Fig. 4(b) the dependence of haze vs correlation length a/λ for $\sigma/\lambda=0.058$ (vertical dashed-dotted line) and $\theta_0=0°$ is depicted. The agreement, also in this cases, is rather satisfactory. However, from this figure there is an indication that at smaller correlation lengths, the agreement becomes less good. This is caused by phase-perturbation theory not being a good approximation in the small correlation length limit (that corresponds to large local slopes σ/a).

Finally in Figs. 4(c), how the angle of incidence influence haze is studied. Only positive angles of incidence are being considered since the scattering geometry is so that there is a symmetry with respect to a change in sign in θ_0. As was done to obtain the results of Figs. 4(a) and 4(b), one has also here fixed the roughness and correlation length to the values used in obtaining the results of Fig. 3, i.e., $\sigma/\lambda=0.058$ and $a/\lambda=1.58$. In the case of transmission [lower panel of Fig. 4(c)], the agreement between the Monte Carlo simulation result and the analytic approximation is of a good quality for all angle of incidence considered. In reflection [upper panel of Fig. 4(c)], the agreement is of a good quality only for the smallest scattering angles. However, as the angle of incidence approaches roughly $\theta_0=55°$, from below or above, the disagreement between the simulation and approximate result (for reflection) becomes pronounced. The reason for this discrepancy is the so-called Brewster angle phenomenon [30]. This phenomenon express itself for the planar geometry in p polarization by the reflection coefficient being exactly zero at the Brewster angle θ_B defined by $\tan^2\theta_B=\varepsilon_1/\varepsilon_0$. However, as roughness is introduced into the system, the reflectivity of the (rough) surface will not be zero any more, not even at the Brewster angle, but will instead go through a minimum for an angle of incidence close to θ_B (the "quasi"-Brewster angle phenomenon). This has the consequence that haze (in reflection and for p polarization) will go through a corresponding maximum for the same angle of incidence. So in the region about the Brewster angle, the presence of surface roughness will strongly renormalize the corresponding planar geometry result with the consequence that the integrated reflected energy is not any more well approximated by Eq. (21). By taking into account roughness in the estimation of the total integrated scattered intensity, one will most likely be able to also predict the behavior of haze for such angles of incidence with more confidence. However, the penalty for doing so is that the resulting expressions become much more complicated and must be evaluated numerically. Since this is not the aim of the present study, we will not follow up this line of actions here, but only keep in mind that the simple approximation [Eq. (27)] breaks down around the Brewster angle. Notice that there is no Brewster angle phenomenon in transmission as shown explicitly in the lower panel of Fig. 4(c) nor is there any such phenomenon in reflection (or transmission) for s-polarized incident light. Hence, for s polarization, the approximate expression for haze [Eq. (27)] should apply for all angles of incidence, something that has been confirmed by numerical simulations (results not shown).

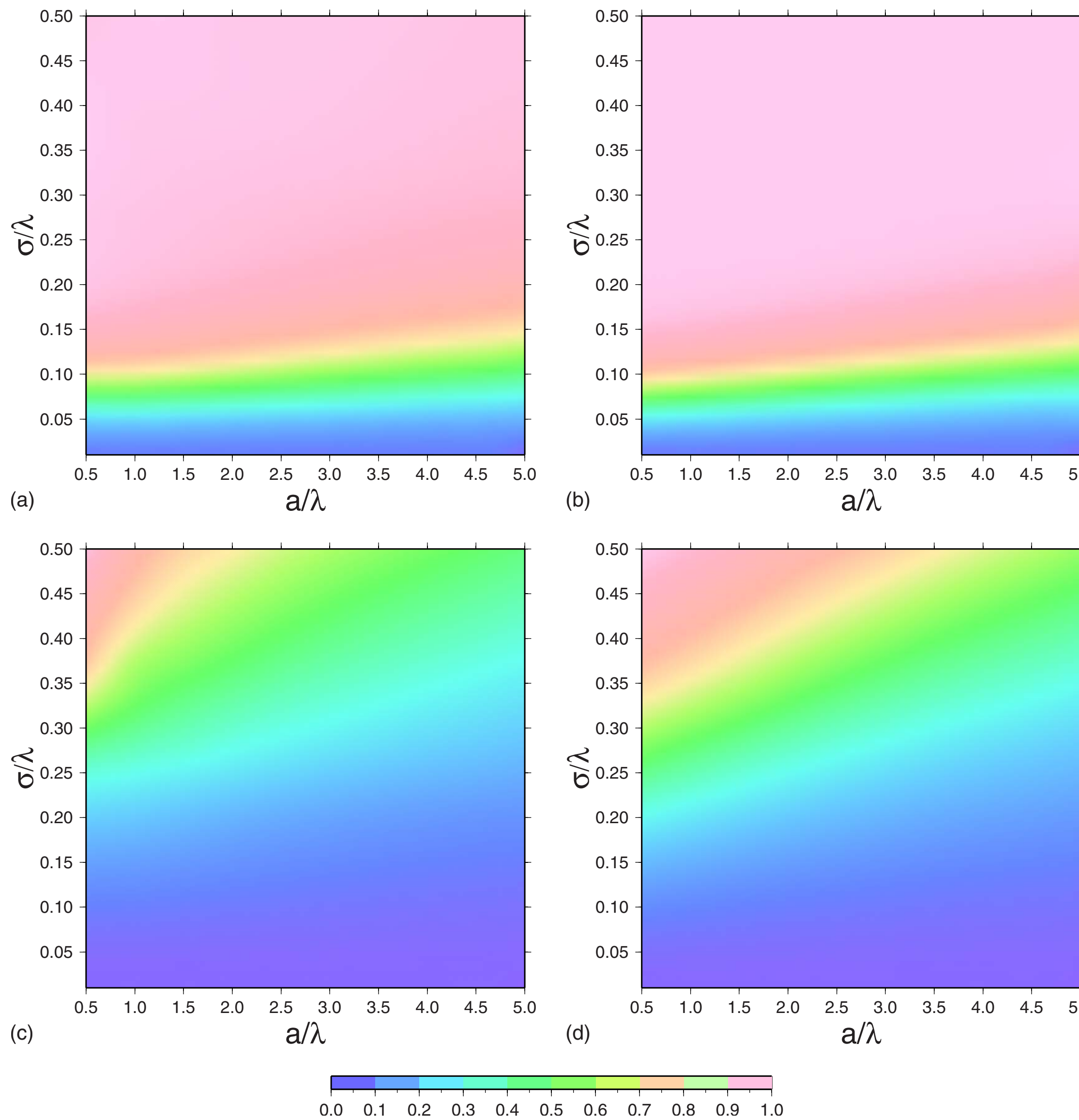

FIG. 5. (Color online) Contour plots of [(a) and (c)] the rigorous Monte Carlo simulation results and [(b) and (d)] the analytic approximation (27) for haze obtained in [(a) and (b)] reflection and [(c) and (d)] transmission for light of wavelength $\lambda=0.6328$ μm incident normally ($\theta_0=0°$) onto the rough surface. The dielectric media that was separated from vacuum by a rough interface was characterized by the dielectric constant $\varepsilon_1=2.25$. All results were averaged over at least $N_\zeta=500$ surface realizations. The random surfaces were all characterized by a Gaussian height distribution function of standard deviation σ and an exponential height-height correlation function of correlation length a. Overall the agreement between the analytic and rigorous simulation results is satisfactory over large regions of parameter space.

In Figs. 4(a) and 4(b), one or more of the parameters that characterize the surface roughness were fixed to constant values. It is, however, important to get a more complete picture of the quality of our analytic expression to haze. This can be achieved by allowing both σ/λ and a/λ to vary freely (within certain limits). Figures 5 depict contour plots for the variations in haze vs both of the two above mentioned parameters in reflection and transmission. The figures in the left column [Figs. 5(a) and 5(c)] show contour plots of haze as obtained by rigorous Monte Carlo simulations. In the right column, i.e., in Figs. 5(b) and 5(d), the corresponding plots obtained from the analytic haze (approximate) expression [Eq. (27)] are presented. By in Figs. 5 comparing the numerical simulation results to those of the corresponding analytic predictions, one can conclude that the quality of the analytic expression (27) is remarkably good over large regions of parameter space. This is particularly the situation when considering reflection. As a general trend, it seems fair to say that approximation (27) performs the best for large correlation lengths and smallest rms roughness. This is in particular the case for haze in transmission for which the approximation is poorer than in reflection. These findings fit the picture that phase-perturbation theory can be looked upon as a generalization of the more familiar Kirchhoff approximation that is known to work the best in the large a/λ limit [24,26]. Moreover, notice that when the large correlation length limit is taken, for fixed rms roughness, the small slope limit is approached since the average slope of the surface is proportional to σ/a.

So far, a semi-infinite dielectric transparent medium bounded to vacuum by a randomly rough interface has been considered. From a practical point of view, it will be rather interesting to also investigate how haze depends on the surface parameters for, say, a film geometry. For such a case, the derivation of an approximate expression for haze, analogous to those presented in Sec. IV for a semi-infinite medium, is lengthy but can be performed. However, such expressions will not be presented here. Instead we would like to add that rigorous numerical simulations for a film geometry have been preformed for normal incidence. The results of such simulations show that the characteristic dependence of haze on the parameters σ/λ and a/λ originally found for a semi-infinite randomly rough surface also seems to hold true for the film geometry.

B. Comparison to experimental results

So far in this paper we have mainly considered one-dimensional surfaces. However, naturally occurring surfaces, as well as man-made surfaces generated by, say, an industrial process, are usually two dimensional. Neither is it not uncommon that their statistical properties are not well described by simple mathematical distribution and correlation functions of the form often assumed in theoretical studies. It is therefore an open question if the approximate result for haze [Eq. (27)] obtained for one *single* randomly rough one-dimensional surface has any relevance for the more complicated scattering systems encounter in practical applications. In the most general case our approximate expression is obviously not suitable. One may still hope, however, that the general behavior of haze found in Eq. (27) could be taken over to higher-dimensional and more complicated scattering geometries. In particular, for weakly rough isotropic surfaces at normal incidence there are hopes that a one-dimensional approach might work reasonably well. This is so since under such circumstances the (two-dimensional) mean differential reflection and transmission coefficients are rotational symmetric, i.e., no ϕ dependence. Consequently, a one-dimensional mean differential or transmission coefficient might be enough to catch the main angular dependence of the scattering up to cross-polarization effects. The purpose of this subsection is to look into these questions and to compare experimental measurements with what can be obtained from a one-dimensional computer simulation approach of the type applied previously in this paper.

To investigate this further, we will consider a melt blown film of linear low-density polyethylene (LLDPE). The film and LLDPE type was referred to as *narrow molecular weight distribution metallocene* and described in detail in Ref. [4]. The haze of the film was measured by a spherical haze meter (diffusion system, type M57) according to the standard [15], and (in transmission, at normal incidence) one obtained for this material a haze of 18.9%. Light scattering at the surface, as well as in the bulk, contributed to this haze value. The bulk contribution was estimated by measuring the haze of films coated by glycerol on both sides. Glycerol has a refractive index of 1.47, which matches closely the refractive index of the blown films (approximately 1.5). Under this condition the haze was significantly reduced (to 5% for the film mentioned above, leaving, crudely speaking, about 14% haze resulting from the surface roughness). The films that were experimentally studied in Ref. [4] (with that of Fig. 2 being among them) gave a haze in the range 12–30 %. When embedded in glycerol, the corresponding values were reduced to 3–6 %. The film with the highest haze was measured to be 31.9% and reduced to about 1/10 of this value when submerged in glycerol (3%). This indicates that the contribution to haze from surface roughness dominates over the contribution from the bulk. This is consistent with the assumption made in the theory section of this paper (cf. Sec. III).

The surface topography of the above mentioned polymer film was measured and served as the basis for the characterization put forward in Fig. 2. The solid lines in Figs. 2(b) and 2(c) represent a Gaussian [Fig. 2(b)] and an exponential fit [Fig. 2(c)] to the height distribution and height-height correlation function, respectively. These functions are characterized by a root-mean-square roughness of $\sigma=0.04$ μm and a correlation length of $a=1.3$ μm. Notice that the functional fits performed in Figs. 2(b) and 2(c) are of reasonable quality and in particular for the height distribution function. In the one-dimensional Monte Carlo simulation results to be presented below, one has used the functions represented by the solid lines in Figs. 2(b) and 2(c) as a basis for generating the underlying ensemble of surface realizations.

In Fig. 6 the logarithm of the experimentally obtained angular intensity distribution, $\log I(\theta,\phi)$, of light being transmitted through the polymer film system described above is depicted. In obtaining these results, a HeNe-laser at wavelength $\lambda=0.6328$ μm was used as a source for the unpolarized normal incident light ($\theta_0=0°$). As expected, a strong specular transmission peak as well as a large dynamical range in intensity (almost 7 orders of magnitude) are observed. Moreover, a weak anisotropy in the angular distribution of the transmitted light can be observed in Fig. 6 as represented by the horizontal line of enhanced intensity. This anisotropy is caused by the polymers being partially oriented along the direction of the flow in the production of the polymer film. In Fig. 6 this direction corresponds to the vertical. Except from the weak anisotropy the angular distribution of the transmitted light is rather isotropic.

In Fig. 7, a horizontal cut through the center of Fig. 6 is represented by open symbols. The solid line in this same figure represents the rigorous (one-dimensional) Monte Carlo simulation results for the mean differential transmission coefficient. In obtaining this latter result a film geometry of mean thickness $d=40$ μm was used where the uncorrelated upper and lower rough (one-dimensional) interfaces were described by the parameters that were derived from the measured surface topography of the film (see Figs. 2). In particular, these surfaces were characterized by the functions represented by the solid lines of Figs. 2(b) and 2(c) (see also the caption of Fig. 7 for the surface parameters). From Fig. 7 it is observed that there is a quite reasonable agreement

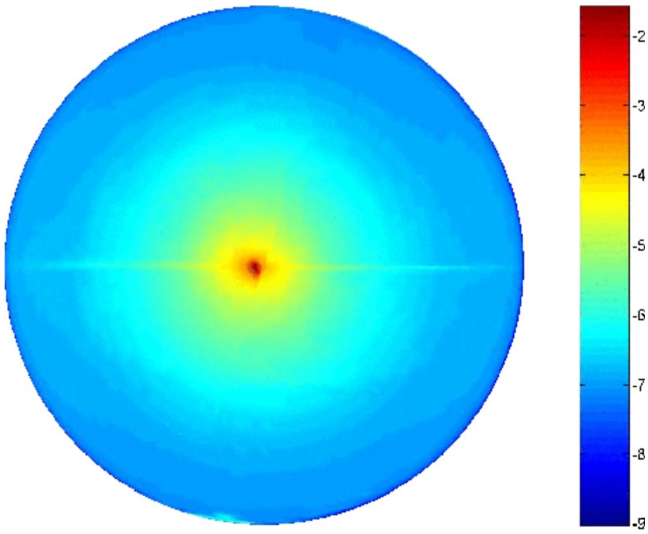

FIG. 6. (Color online) Experimental results for the angular distribution of the light being transmitted through a melt blown LLDPE film of the type described and partly characterized in Fig. 2. The (mean) thickness of the film was $d=50$ μm, and the wavelength of the light being incident normally onto the top mean surface was $\lambda=0.6328$ μm. It is the logarithm of the transmitted intensity in arbitrary units that is presented. The measurements were conducted with a spectrophotogoniometer built by SINTEF. The weak anisotropy seen in the transmitted intensity is caused by the polymers being preferentially oriented in the flow direction (vertical direction in the figure).

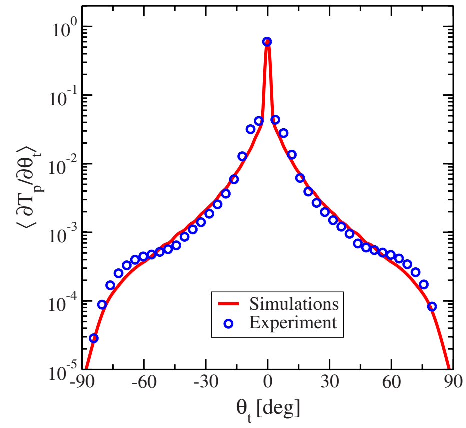

FIG. 7. (Color online) The mean differential transmission coefficient $\langle \partial T_p / \partial \theta_t \rangle$ vs angle of transmission θ_t for light of wavelength $\lambda=0.6328$ μm impinging normally ($\theta_0=0°$) onto a $d=40$-μm-thick LLDPE film ($\varepsilon_1=2.25$). The experimental results (open circles) corresponds to a cut through Fig. 6. Due to the use of arbitrary units in the experiment, the amplitude of the measurements was adjusted to fit that of the simulation results. In obtaining the simulation results (solid line), the surface parameters represented by the solid lines in Figs. 2 were used, i.e., the parameters used were $\sigma=0.04$ μm and $a=1.3$ μm for the exponentially correlated rough surface. In order to replicate the unpolarized incident light used in the experiment, the simulation results were averaged over the s- and p-polarized results. The thickness of the film was also here $d=40$ μm.

between the measured and simulated angular distribution of the transmitted light. It is at the largest angles of transmission that the discrepancy starts to emerge, while the angular distribution in the central part that represents the main part of the transmitted energy seems to be well accounted for by the simulation result. For the largest transmission angles the Monte Carlo result seems to underestimate the transmitted power. This situation is in fact not unexpected; In the experimental measurements scattering from the bulk is also present, while such effects has not been taken into consideration in the approach used to produce the solid line of Fig. 7. It is, in fact, well known that bulk scattering tends to enhance the transmitted power into large angles of transmission [65].

As mentioned above, one by direct measurements found the haze in transmission for normal incidence to be $\mathcal{H}_{2D}(0°)\approx 0.189$ for the experimental sample (with both surface and bulk randomness). From the Monte Carlo simulation results one on the other had found a haze of $\mathcal{H}_{1D}(0°)\approx 0.054$. However, in order to be able to compare this result with the experimentally available value $[\mathcal{H}_{2D}(0°)]$, one has to introduce a (multiplicative) constant that accounts for the difference between the one- and two-dimensional geometries (assuming normal incident light). Doing so results in a haze of about 14–15 %, a result that compares favorably to the experimental haze value resulting from surface randomness. Hence, one has demonstrated that the one dimensional Monte Carlo approach can be applied to predict reasonably well the haze of an isotropic experimental sample at normal incidence. This is indeed an encouraging results; but further work is needed, however, to determine the full region of applicability of the Monte Carlo approach.

VI. CONCLUSIONS

In conclusion, we have studied the dependence of haze and gloss with the parameters that normally are used to characterize randomly rough surfaces—the rms roughness σ and the height-height correlation length a. Based on phase-perturbation theory, we have derived analytic expressions that represent approximations to haze and gloss for a one-dimensional Gaussian rough surface. It is demonstrated that haze (gloss) increases (decreases) with σ/λ as $\exp[-A(\sigma/\lambda)^2]$ and decreases (increases) with a/λ in a way that depends on the specific form of the correlation function being considered. This latter dependence enters into the expression for haze and gloss as the average of the power spectrum taken over a small interval, Δq, around zero momentum transfer. In the limit $\Delta qa \ll 1$ one obtains that the haze and gloss depend linearly on the correlation length.

The range of validity for these approximate expressions to haze and gloss put forward in this paper were assessed by rigorous numerical Monte Carlo simulations. They were found to agree remarkably well over large regions of parameter space. Furthermore, some experimental results for the angular distribution of the light being transmitted through a polymer film was presented. It was found to fit reasonably well with the prediction from the Monte Carlo simulations, and consequently the predicted value of haze was found to agree relatively well.

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APPENDIX: PHASE-PERTURBATION THEORY

In this appendix we will describe the so-called phase-perturbation theory, as well as deriving some analytical expressions, on which the main text relays. What has become known as the phase-perturbation theory, was originally developed by Shen and Maradudin [56] for nonpenetrable media. The method was later extended to one-dimensional randomly rough penetrable media by Sánchez-Gil *et al.* [60]. The method has also been formulated in an explicit reciprocal way [57]. In phase-perturbation theory it is the phase of the field that is determined perturbatively [29], and it has proven well suited for reflectivity studies [60,66]. One of the interesting features of this perturbative method is that in the large limit of a/λ, with a being the surface correlation length and λ as the wavelength of the incident light, it reduces to the more well-known Kirchhoff approximation [24–27] for a nonpenetrable medium. Furthermore, as a becomes comparable to λ, phase-perturbation theory represents a correction to the Kirchhoff result. An additional practical advantage of phase-perturbation theory is that it retains its analytic form also for penetrable or absorbing media, something that is not the case for the Kirchhoff approximation.

Let us start our discussion of phase-perturbation theory and what can be derived from it by letting $U(q|k)$ collectively denote either the reflection or transmission amplitudes, $R_\nu(q|k)$ or $T_\nu(q|k)$, introduced in Sec. III B. By taking into account the boundary condition at the rough surface and assuming that there is no down-going (up-going) scattered (transmitted) waves even close to the rough interface [67] a single integral equation for $U(q|k)$ can be derived [23,31] (the reduced Rayleigh equation). Based on this integral equation one can derive the following expression for amplitude:

$$U(q|k) = u_0(k)\int_{-\infty}^{\infty} dx e^{-i(q-k)x} e^{i\Lambda(q|k)\zeta(x)}, \qquad \text{(A1a)}$$

where $u_0(k)$ is the Fresnel coefficient for the corresponding planar geometry. In writing this expression we have introduced

$$\Lambda(q|k) = \begin{cases} \alpha_0(q) + \alpha_0(k) & \text{in reflection} \\ \alpha_1(q) - \alpha_0(k) & \text{in transmission,} \end{cases} \qquad \text{(A1b)}$$

It should be noticed that if we were dealing with a more complicated scattering geometry then the one depicted in Fig. 1, say a film geometry, the amplitude could still be expression in form (A1a) if only one of the interfaces were randomly rough. In this case $\Lambda(q|k)$ will take on another form then the one given above.

In order to make contact with observable we will need an expression for $|U(q|k)|^2$. In fact, more precisely it is the average over this quantity that we should be interested in since the surface is randomly rough. With Eq. (A1a), we find

$$\langle |U(q|k)|^2\rangle = |u_0(k)|^2 \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dx' e^{-i(q-k)(x-x')} \times \langle e^{i\Lambda(q|k)[\zeta(x)-\zeta(x')]}\rangle, \qquad \text{(A2)}$$

where the average, denoted by $\langle \cdots \rangle$, is assumed to be taken over an ensemble of surface realizations of the surface profile function $\zeta(x)$. Furthermore, we have here assumed that $\Lambda(q|k)$ is real (or close to being real), as it will be automatically in reflection.

With the change in variable $x' = x + u$ the above equation becomes

$$\langle |U(q|k)|^2\rangle = |u_0(k)|^2 \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} du e^{i(q-k)u} \langle e^{i\Lambda(q|k)\Delta\zeta(u)}\rangle, \qquad \text{(A3)}$$

with $\Delta\zeta(u) = \zeta(x) - \zeta(x+u)$. If $\zeta(x)$ is a *stationary* random process, then the average in Eq. (A3) will be independent of x. Therefore, the x integration in the same equation will give the contribution L that is the length along the x direction of the rough surface. Furthermore, if in addition to being stationary ζ also is a Gaussian random process, as we assume here according to Sec. II, then $\Delta\zeta(u)$ will also be a Gaussian random variable. Hence, the average in Eq. (A3) can be done analytically with the result that

$$\langle |U(q|k)|^2\rangle = L|u_0(k)|^2 \int_{-\infty}^{\infty} du e^{i(q-k)u} e^{-\sigma^2\Lambda^2(q|k)[1-W(u)]} = |u_0(k)|^2 J(q|k), \qquad \text{(A4a)}$$

where

$$J(q|k) = L e^{-\sigma^2\Lambda^2(q|k)} \int_{-\infty}^{\infty} du e^{i(q-k)u} e^{\sigma^2\Lambda^2(q|k)W(u)}, \qquad \text{(A4b)}$$

with $W(u)$ being the surface correlation function as defined in Sec. II.

With Eqs. (11b), one for the mean DRC or DTC, collectively denoted $\langle \partial U/\partial\theta\rangle$, finally obtains

$$\left\langle \frac{\partial U}{\partial \theta}\right\rangle = \frac{1}{L}\frac{\varepsilon_m}{\sqrt{\varepsilon_0}}\frac{\omega}{2\pi c}\frac{\cos^2\theta}{\cos\theta_0}|u_0(k)|^2 J(q|k). \qquad \text{(A5)}$$

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[53] Haze is often also indicated by using a "percentage notation" so that a haze of 1 corresponds to a haze of 100%.
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