Surface Correlation Effects on Gloss

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ABSTRACT: A general expression for gloss within the scalar Kirchhoff's theory is derived in terms of the detector collecting angle, and two statistical parameters that characterize the surface roughness. Analytical expressions for gloss are derived for an exponential and a Gaussian correlation function, and numerical results for these and other quasi-exponential correlation functions are presented. It is shown that the incoherent contribution to gloss is significant in common polymeric surfaces. The latter implies that surface height correlations cannot be neglected in the evaluation of gloss. It is also shown that for a correlation function with a single characteristic length, gloss scales with the correlation length L_c in the same way as with the detector collecting angle. This fact can be used to determine L_c with a glossmeter, and an experimental method to achieve this is proposed. © 1998 John Wiley & Sons, Inc. J Polym Sci B: Polym Phys 36: 1321–1334, 1998
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INTRODUCTION

Gloss is an appearance attribute highly demanded by many applications such as paints and plastic goods in houseware and car industry. From a physical point of view gloss is related to the amount of light scattered around the specular direction. Usually it is associated with surface roughness because in many cases the bulk contribution is small compared to the one coming from the surface.[1,2] In this article we shall assume that this is the case.

Gloss depends not only on the material properties but also on process variables. For example, in the plastics industry, most commercial plastics are reinforced with some kind of fillers. Under elongational flow, inherent in all plastic manufacturing processes, these fillers migrate to the surface affecting its roughness.[3–7] The size distribution, mechanical properties, and index of refraction of the fillers, as well as the viscosity and index of refraction of the matrix, together with process variables such as melt and mold temperatures, injection speed, etc., define the surface properties of the sample and, therefore, its reflectance. Examples like this can be also found in other areas such as the paint industry, where one has a highly concentrated suspension of latex particles (which finally filmify) mixed with pigments and other fillers. Again, the surface tension, the index of refraction of the latex film and the fillers, the viscosity of the mixture, the particle size of the fillers, etc., will be some of the relevant material variables on which the surface properties depend. However, equally important in the formation of surface roughness are, for example, the method used in spreading the film on the substrate, together with the external variables involved in the filmification process. To develop a product with controlled gloss one must be able to relate the material and process variables to gloss, as measured by standard testing methods.

There are two kinds of gloss measurements—one, and probably the more commonly used, is a

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measure of the specular gloss, that is, the specular reflectance of a specimen relative to that of a reference standard (ASTM D 523-89).[8] The other kind measures the reflection haze and the distinctness-of-image gloss in addition to specular gloss (ASTM E430-91).[9] Distinctness-of-image gloss is the attribute that characterizes the sharpness of images of objects produced by reflection from a given surface. It is related to the relative scattering (relative to a standard) at an angle close to the specular direction ($\pm 0.3°$). On the other hand, reflection haze is given by the ratio of the scattering intensity at an angle off the specular direction to that of a reference surface. If this angle is $\pm 2°$ ($\pm 5°$), one refers to narrow-angle (wide-angle) haze. In specular gloss one measures a mixture of coherent and diffuse reflected light, as defined by the receiving aperture, while reflected haze and distinctness-of-image gloss are a measure of the sharpness of the angular pattern of diffuse scattering intensity around the specular angle. The latter test is addressed particularly to high-gloss samples. However, many applications in the plastics and paint industries lie in an intermediate region between weak and strong scattering surfaces, and specular gloss is commonly used as a monitoring measure of appearance. For these reason in this article deals with specular gloss, although the results here derived could be extended to reflection haze and the distinctness-of-image gloss.

Because gloss depends on material and process variables via the effect that these have on surface roughness, we must first establish the relation between surface roughness and gloss. The latter is the main purpose of this article.

The theoretical basis to understand the properties of the far-field of light scattered from a rough surface, in terms of the statistical properties of the surface, was established long ago in the work of Davies[10] and Beckmann.[11] Nevertheless, in most approaches,[12] including Beckmann's, the normalization of the relative reflectance corresponds to a point detector. This leads to expressions for gloss that cannot be compared to measurements made with standard glossmeters. Although inherent to the design of a glossmeter is the size of the detector apertures, we have not found in the literature explicit expressions for gloss that take into consideration the size of the detector, except in the early work of Porteus[13] on the reflectance at normal incidence for Gaussian-correlated surfaces. In this work, the detector–acceptance angle appears explicitly in the expression of the relative reflectance. However, as we shall show below, his expression corresponds to the present results in the limit of weak scattering surfaces.

One of the important inputs in the theory by Beckmann is the surface correlation function of heights. However, in spite of the growing interest in Atomic Force Microscopy studies of polymer surfaces,[14] we have not found a systematic study of correlation functions of polymeric surfaces. Most authors characterize only the RMS mean height. Even in the applied polymer literature on optical properties one finds publications[15] that relate gloss only to the RMS average height (coherent reflectance). In this article we will show that the surface correlation will have a significant effect on gloss.

Within the few articles that characterize the correlation function of polymer surfaces is that of Méndez et al.[1] for the case of poly(acrylonitrile–butadiene–styrene) (ABS), a plastic commonly used in the automotive and electrical–houseware industry. There it has been shown that, for different processing conditions, a simple Kirchhoff scalar theory with a Gaussian height distribution and an exponential height correlation function, accurately describes the angle-resolved scattering in the vicinity of the specular angle. Another example, now in the context of the paper industry, has been given by Lettieri et al.,[2] where a simple Kirchhoff scalar theory with a Gaussian height distribution together with a quasi exponential correlation function, gave an adequate description of the angle-resolved light scattering by glossy paper. In a more general context Bennett and Mattsson[16] have pointed out that in a wide variety of surfaces, the surface height correlation function is, in general, closer to an exponential one. This implies that a simple scalar Kirchhoff theory with a quasi-exponential correlation function seems to be a reasonable model in explaining the surface scattering features of many complex systems encountered in industrial applications. We shall limit here the discussion to statistical-type surfaces, that is, surfaces where roughness is statistical in nature or at least is dominant over any other systematic source of roughness.

In this article we derive a general expression for gloss, within the scalar Kirchhoff's theory with a Gaussian height distribution and show that, in the visible frequency range, the contribution of the diffuse beam is significant. Our expression takes into account the size of the aperture of the detecting system. For correlation functions with a single correlation length, L_c, we show that gloss, at a given angle, depends only on two parameters: $(\sigma/\lambda)\cos\theta_1$ and $(L_c/\lambda)(\delta\theta)_D$. Here, σ is the RMS average height, λ is the wavelength of light in air, $(\delta\theta)_D$ is the detector collecting half-angle, and θ_1 is the angle of incidence relative to the surface normal. We give explicit expressions for a Gaussian and an exponential surface correlation function. The Gaussian correlation allows us to compare our results with those of Porteus,[13] and it provides a model for a quasi single-length scale type of surface roughness. The exponential, on the other hand, corresponds more closely to statistical real surfaces as pointed out above. The power spectrum of an exponential correlation function has a longer tail, indicating the presence of higher spatial-frequencies components and, therefore, implying a multiple-scale length type of surface. In any case, the results with an exponential correlation function are also compared with those obtained with modified exponentials, as proposed by Lettieri et al.,[2] and with K-correlation functions such as suggested by Hoenders et al.[17] The latter represent a family of correlation functions that encompass Gaussians, exponentials, modified fractal-type and other nonfractal-type surfaces.

Finally, using our result that variations on L_c/λ are equivalent to changes in $(\delta\theta)_D$, we propose a method to determine the correlation length via specular gloss measurements. This will be complementary to the classical Bennett–Porteus method of determining σ from coherent reflectance measurements.[18]

KIRCHHOFF SCALAR THEORY OF GLOSS

As stated in the introduction, glossmeters that comply to the ASTM Standard D523-89[8] measure the specular reflectance of a rough surface relative to that of a smooth standard surface at a given angle. The relative specular reflectance around a solid angle Ω_1 is defined as:

$$\rho(\Omega) = \frac{1}{P_N}\int_{\Omega_1}^{\Omega_1+\Omega_D}\left(\frac{dP}{d\Omega}\right)d\Omega, \tag{1}$$

where $dP/d\Omega$ is the power scattered by the surface per solid angle, Ω_D is the solid angle defined by the receiving optics of the detector, and the subscript 1 refers to the specular direction. P_N is the total power scattered by a smooth surface, received by the same detector-collecting system, at the specular angle θ_1; that is:

$$P_N = \int_{\Omega_1}^{\Omega_1+\Omega_D}\left(\frac{dP_o}{d\Omega}\right)d\Omega, \tag{2}$$

where $dP_o/d\Omega$ is the power scattered per solid angle by a smooth surface.

To calculate $dP/d\Omega$ we use Kirchhoff scalar theory.[11,19] For that, we divide the scattered field into its coherent and incoherent components. Here we shall understand by coherent, the scattered-field component that preserves the phase relationships of the incident field. In the literature in this field,[11,19] it is common to associate the origin of incoherence in the scattered field to the random phase changes induced by the surface roughness itself rather than by the coherence properties of the light source employed. Therefore, it is usual to identify the specular field with the coherent component of the scattered field because for glossy to moderately glossy surfaces this component is dominant over the diffuse light scattered at the specular direction. The diffuse intensity is, therefore, identified with the incoherent component of the scattered light, and in this article we shall use these indistinctly.

Standard glossmeters use large receiving field apertures (≈ 2–$4°$) and illuminate a large sample area (≈ 1–$2\ \text{cm}^2$.). Their light source is an incandescent lamp that can be partially coherent by introducing a spectral filter at the source exit. For a large receiving field aperture, the diffuse intensity measured using a coherent source or a partially coherent one (centered at the same wavelength) will be the same, as long as the size of the speckles is small compared with the aperture's size, because the receiving optical system is averaging over a large number of speckles. For the illuminated sample area indicated above, the speckles will be small. Under this condition, its seems reasonable in our discussion to identify the diffuse field with the incoherent component, independently of the source used, as is common in this field.

The coherent field amplitude $\langle E_s^{(c)}\rangle$ is given by Ogilvy[19] as:

$$\langle E_s^{(c)}\rangle = \frac{|r|}{|r_o|}\,\chi(kC)E_{\text{smooth}}, \tag{3}$$

where E_{smooth} is the amplitude of the field scattered by a smooth surface, and k is the magnitude

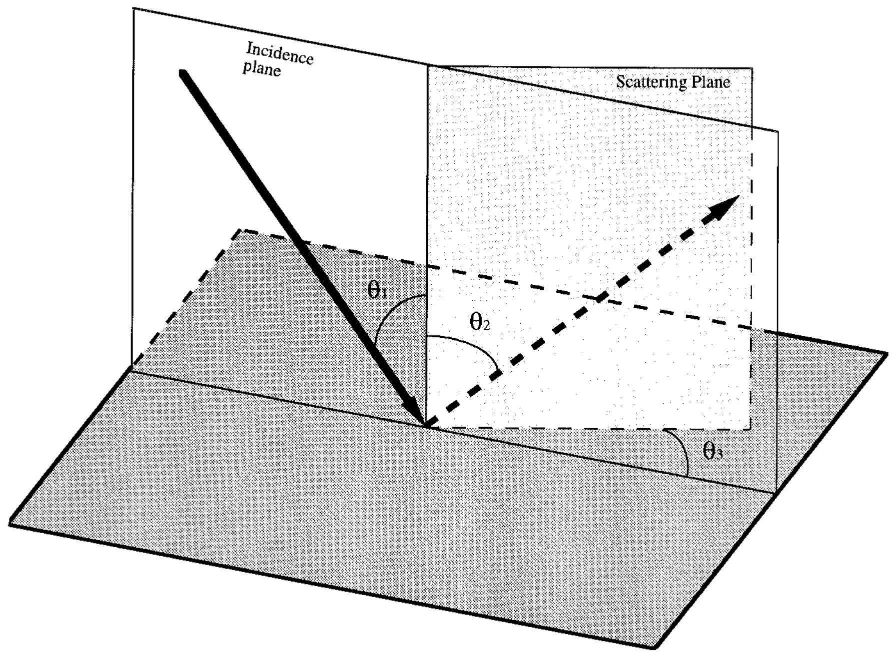

Figure 1. Scattering geometry for plane wave incidence.

of the wave vector of the incident beam. Here $|r|^2$ is the average reflectance in the specular direction of the rough surface, $|r_o|^2$ is the reflectance of the smooth surface, as given by the usual Fresnel formulas. χ is the Fourier transform of the height probability density, $p(h)$:

$$\chi(s) = \int_{-\infty}^{\infty} p(h)\exp(\text{ish})\,dh, \tag{4}$$

and s is the corresponding variable in Fourier space that represents the inverse of a surface "wavelength" and, therefore, χ provides a measure of the phase modulation of a wave at a rough surface. C is defined as:

$$C = -(\cos\theta_1 + \cos\theta_2), \tag{5}$$

Here, θ_1 and θ_2 are the incident and scattering angles relative to the surface normal, respectively, as shown in Figure 1.

Therefore, the coherent contribution to the power scattered per solid angle is:

$$\left(\frac{dP}{d\Omega}\right)^{(c)} = |\chi(kC)|^2 \frac{|r|^2}{|r_o|^2}\left\{\frac{dP_o}{d\Omega}\right\}. \tag{6}$$

For Gaussian height statistics, $\chi(kC)$ is given by:

$$|\chi(kC)|^2 = \exp(-g), \tag{7}$$

where

$$g = k^2\sigma^2(\cos\theta_1 + \cos\theta_2)^2, \tag{8}$$

and σ is the surface RMS height.

The power scattered incoherently per solid angle, in *cgs* units, is given by[19]:

$$\left(\frac{dP}{d\Omega}\right)^{(d)} = \left(\frac{c_o}{4\pi}\right)|r|^2 \frac{k^2F^2}{2\pi} A_M \times \int_0^{\infty} J_o(kR\sqrt{A^2+B^2})[\chi_2(kC, -kC, R) - |\chi(kC)|^2]R\,dR, \tag{9}$$

where A_M is the area illuminated, c_o is the velocity of light in vacuum, J_o is a Bessel function of zero order, and A, B, and F are given by:

$$A = \sin\theta_1 - \sin\theta_2\cos\theta_3 \tag{10}$$

$$B = -\sin\theta_2\sin\theta_3 \tag{11}$$

$$F = \frac{(1 + \cos\theta_1\cos\theta_2 - \sin\theta_1\sin\theta_2\cos\theta_3)}{(\cos\theta_1 + \cos\theta_2)}, \tag{12}$$

Here, θ_3 is the angle of the scattered field relative to the plane of incidence as shown in Figure 1. χ_2 is the two-dimensional characteristic function defined as:

$$\chi_2(s_1, s_2, \mathbf{R}) = \int_0^\infty \int_0^\infty p(h_1, h_2, \mathbf{R})\exp[i(s_1 h_1 + s_2 h_2)]dh_1 dh_2, \quad (13)$$

where $p(h_1, h_2, \mathbf{R})dh_1dh_2$ is the probability that two points separated by a vector $\mathbf{R}$ have heights between $h_1 + dh_1$ and $h_2 + dh_2$, respectively, and s_1 and s_2 are the corresponding variables in Fourier space.

If we assume that the surface is both isotropic and stationary and with Gaussian height statistics, then:

$$\chi_2(kC, -kC, R) = \exp(-\{g[1 - C(R)]\}), \quad (14)$$

where $C(\mathbf{R})$ is the surface correlation function defined as,

$$C(\mathbf{R}) = \frac{\langle h(\mathbf{r})h(\mathbf{r} + \mathbf{R})\rangle_s}{\sigma^2}. \quad (15)$$

Here $h(\mathbf{r})$ is the height of the surface at $\mathbf{r}$ relative to a reference nominal surface defined such that $\langle h\rangle_s = 0$, where $\langle \cdots \rangle_s$ denotes spatial averaging. Due to stationarity and the isotropic character of the surface, $C(\mathbf{R})$ will depend only on the absolute distance $\mathbf{R}$ between any two points at the surface.

Under these assumptions eq. (9) can be written as:

$$\left(\frac{dP}{d\Omega}\right)^{(d)} = \left(\frac{c_o}{4\pi}\right)|r|^2 \frac{k^2F^2}{2\pi} A_M \exp(-g) \times \int_0^\infty J_o(kR\sqrt{A^2 + B^2})[\exp(gC(R)) - 1]RdR. \quad (16)$$

For the smooth surface, the power scattered per solid angle for a rectangular illuminated section of sides $2X$ and $2Y$ is given by:

$$\frac{dP_o}{d\Omega} = \left(\frac{c_o}{4\pi}\right)\left(\frac{k^2}{16\pi^2}\right)A_M^2|r_o|^2(\cos\theta_1 + \cos\theta_2)^2 \operatorname{sinc}^2(kAX)\operatorname{sinc}^2(kBY), \quad (17)$$

where $A_M = 4XY$ and $\operatorname{sinc} x = \sin x/x$.

In most treatments[12] since Beckmann's treatise,[11] the total power scattered by the smooth surface is taken as that coming only at the specular angle, that is $\theta_3 = 0$ and $\theta_2 = \theta_1$. In that case, the sinc functions are equal to unity and P_N will be given by:

$$P_N^s = \left(\frac{c_o}{4\pi}\right)|r_o|^2\left(\frac{1}{\lambda^2}\right)A_M^2 \cos^2\theta_1\Omega_D. \quad (18)$$

The superscript "s" indicates that this normalization is equivalent to taking a very small detector collecting angle where $\Omega_D \ll \lambda^2/A_M$ and, therefore, the detector picks up only the very central part of the zero-order diffraction peak. However, because in common glossmeters $\Omega_D \gg \lambda^2/A_M$, eq. (2) implies an integration of the specularly scattered diffraction pattern, leading to the following normalization power:

$$P_N^\ell = \left(\frac{c_o}{4\pi}\right)|r_o|^2 A_M \cos\theta_1 D(\Omega_D), \quad (19)$$

where $D(\Omega_D)$ is a function that tells us how much of the diffraction pattern was collected by the detector. The extra superscript ℓ states explicitly that the case of a large detector collecting angle is being considered. If $\Omega_D \gg \lambda^2/A_M$, $D(\Omega_D)$ will be always very close to unity. The details of this normalization procedure are given in Appendix 1. We shall use P_N^ℓ as normalization power because it corresponds to the reference reflected power commonly used in specular gloss determination procedures. Notice that P_N^s scales as $A_M^2\cos^2\theta_1\Omega_D$ rather than $A_M\cos\theta_1$ as in P_N^ℓ, and consequently, all our results will differ from Beckmann's due to the normalization chosen here.

Therefore, the coherent $\rho_s^{(c)}$ and the incoherent $\rho_s^{(d)}$ contributions to the specular relative reflectance in a glossmeter are given by

$$\rho_s^{(c)} = \frac{|r|^2}{|r_o|^2}|\chi(kC)|^2, \quad (20)$$

and

$$\rho_s^{(d)} = \frac{|r|^2}{|r_o|^2}\frac{k^2}{2\pi\cos\theta_1} \times \int_{\Omega_1}^{\Omega_1+\Omega_D}\left\{\exp(-g)F^2\int_0^\infty J_o(kR\sqrt{A^2 + B^2}) \times [\exp(gC(R)) - 1]RdR\right\}d\Omega, \quad (21)$$

respectively. Gloss, $\mathcal{G}$, can then be written as:

$$\mathcal{G} = \rho_s^{(c)} + \rho_s^{(d)}. \tag{22}$$

CORRELATION LENGTH AND COLLECTING DETECTOR ANGLE EFFECTS ON GLOSS

In this section we shall develop expressions for gloss for correlation functions that are of the form $C(R/L_c)$, that is, with a single characteristic length. L_c is usually called the correlation length. In particular, we shall derive explicit expressions for an exponential and a Gaussian surface correlation function.

Because the angular range of integration is small, we can introduce the following approximations in eq. (21):

I) The factor $F^2 \exp(-g)$ can be written to first order in $\delta\theta = \theta_2 - \theta_1$ as:

$$F^2 \exp(-g) = \exp(-g_s)\cos^2 \theta_1 \times \{1+ (g_s - 1)\tan \theta_1\, \delta\theta + \cdots\}, \tag{23}$$

where

$$g_s = 16\pi^2 \frac{\sigma^2}{\lambda^2} \cos^2\theta_1. \tag{24}$$

The angle θ_3 will be of the same order as $\delta\theta$ (except at normal incidence), and it will contribute only to second order in eq. (23). We observe that for reasonably rough surfaces ($\sigma \approx 0.1\ \mu$m), and typical values for λ, θ_1, and collecting apertures, say, $\lambda \approx 0.5\ \mu$m, $\theta_1 = 20°$ and $\delta\theta_{\max} = \pi/180$, respectively, the maximum error in $F^2 \exp(-g)$, to zeroth order, is less than 3%. Therefore, the error in the full angular integral of eq. (21), by the substitution of $F^2 \exp(-g)$ to zeroth order in $\delta\theta$, will be smaller than 3%. Substitution of $gC(R)$ by $g_sC(R)$ will give an error similar to the one just discussed.

II) The argument of the Bessel function can also be expanded into powers of $\delta\theta$ and θ_3, and can be approximated by:

$$kR|\sqrt{A^2 + B^2}| \approx kR\sqrt{(\delta\theta)^2 \cos^2 \theta_1 + \theta_3^2 \sin^2 \theta_1}. \tag{25}$$

III) To the same order in $\delta\theta$, the element of solid angle can be approximated as $d\Omega \approx \sin \theta_1\, d\theta_3 d(\delta\theta)$.

Introducing these approximations into eq. (21) and making the following change of variables:

$$\alpha = kL_c \cos \theta_1 \delta\theta, \tag{26}$$

and

$$\beta = kL_c \sin \theta_1 \theta_3, \tag{27}$$

the expression for $\rho_s^{(d)}$ can be written as:

$$\rho_s^{(d)} = \frac{2}{\pi} \frac{|r|^2}{\pi |r_o|^2} \exp(-g_s) \times \int_0^{y_D} \int_0^{y_D \cos\theta_1} \left\{ \int_0^\infty J_o(\sqrt{\alpha^2 + \beta^2}x) \times [\exp(g_sC(x)) - 1]x dx \right\} d\alpha d\beta, \tag{28}$$

where

$$y_D = kL_c|(\delta\theta)_D|.$$

In this expression, we have taken into account that the detector collecting solid angle is defined entirely by the collecting optics and, therefore, Ω_D should be independent of the specular angle. We have taken the integration limits over $\delta\theta$ and θ_3 such that this is satisfied. That is, the integration range of $\delta\theta$ is $0 \leq \delta\theta \leq (\delta\theta)_D$, where $(\delta\theta)_D$ is the detector collecting half-angle, while the corresponding integration limits for θ_3 are $0 \leq \theta_3 \leq (\delta\theta)_D/\sin \theta_1$. This means that the collecting optics aperture is approximated by a spherical square sector.

We can observe that for a given angle of incidence the incoherent contribution to gloss depends only on two parameters, g_s and y_D. This implies that $\rho_s^{(d)}$ varies with L_c in the same way as with the detector collecting angle. Equation (28) is valid for all correlation functions with a single characteristic length.

To find an explicit expression for gloss in terms of g_s and y_D, we must know the functional form of the correlation function. In what follows we shall discuss the analytical solutions for two types of correlation functions, an exponential and a Gaussian one. Substituting an exponential correlation function $C(R) = \exp(-R/L_c)$ in eq. (28) and performing a series expansion of the exponential inside the integral, we get:

$$\rho^{(d)}{}_s = \frac{2}{\pi}\frac{|r|^2}{|r_o|^2}\exp(-g_s) \times \sum_{n=1}^{\infty}\int_0^{y_D}\int_0^{y_D\cos\theta_1}\left\{\int_0^{\infty} J_o(\sqrt{\alpha^2+\beta^2}x) \times\left[\frac{g_s^n\exp(-nx)}{n!}\right]xdx\right\}d\alpha d\beta. \quad (29)$$

Integrating the above expression and using:

$$\int_0^{\infty} J_o(yx)\exp(-nx)x\,dx = \frac{n}{(n^2+y^2)^{3/2}}, \quad (30)$$

we arrive at:

$$\rho^{(d)}_{s(\exp)} = \frac{2}{\pi}\frac{|r|^2}{|r_o|^2}\exp(-g_s)\sum_{n=1}^{\infty}\left[\frac{g_s^n}{n!}\right] \times\arctan\left\{\frac{y_D^2\cos\theta_1}{n\sqrt{n^2+y_D^2(1+\cos^2\theta_1)}}\right\}. \quad (31)$$

We have included an additional subscript (exp) to indicate explicitly that this expression is only valid for the case of an exponential correlation function. If we add to eq. (31), the coherent specular relative reflectance, we finally get a simple expression for gloss:

$$\mathcal{G}_{(\exp)} = \frac{|r|^2}{|r_o|^2}\exp(-g_s)\left\{1+\frac{2}{\pi}\sum_{n=1}^{\infty}\left[\frac{g_s^n}{n!}\right] \times\arctan\left\{\frac{y_D^2\cos\theta_1}{n\sqrt{n^2+y_D^2(1+\cos^2\theta_1)}}\right\}\right\}. \quad (32)$$

This formula applies for all angles, within the range of validity of Kirchhoff's approximation, except in the neighborhood of normal incidence, because in this case θ_3 is not small and will vary between 0 and 2π. Therefore, the normal-incidence case has to be treated separately. However, in this case, $\theta_1 = 0$ and $\beta = 0$; thus, the integrand of eq. (29) will only depend on $\delta\theta$. Additionally, in this limit, $d\Omega$ can be approximated by $d\Omega \approx \delta\theta d(\delta\theta)d\theta_3$. Following the same steps as above we get:

$$\mathcal{G}_{(\exp)}(\theta_1=0) = \frac{|r|^2}{|r_o|^2}\exp(-g_s)\left\{\exp(g_s) - \sum_{n=1}^{\infty}\frac{g_s^n}{(n-1)!(n^2+y_D^2)^{1/2}}\right\}. \quad (33)$$

For a Gaussian correlation function $C(R) = \exp(-[R/L_c]^2)$ we follow the same steps as in the previous case, and by using

$$\int_0^{\infty} J_o(yx)\exp(-nx^2)x\,dx = \frac{1}{2n}\exp\left(-\frac{y^2}{4n}\right), \quad (34)$$

we find

$$\rho^{(d)}_{s(\mathrm{Gauss})} = \frac{|r|^2}{|r_o|^2}\exp(-g_s) \times\sum_{n=1}^{\infty}\frac{g_s^n}{n!}\operatorname{erf}\left(\frac{y_D\cos\theta_1}{2\sqrt{n}}\right)\operatorname{erf}\left(\frac{y_D}{2\sqrt{n}}\right), \quad (35)$$

where erf(x) is the error function. We have included an additional subscript (Gauss) to indicate explicitly that this expression is only valid in the case of a Gaussian correlation function.

Adding the coherent contribution to the relative specular reflectance, we get that gloss is given by:

$$\mathcal{G}_{(\mathrm{Gauss})} = \frac{|r|^2}{|r_o|^2}\exp(-g_s)\left\{1+\sum_{n=1}^{\infty}\frac{g_s^n}{n!} \times\operatorname{erf}\left(\frac{y_D\cos\theta_1}{2\sqrt{n}}\right)\operatorname{erf}\left(\frac{y_D}{2\sqrt{n}}\right)\right\}. \quad (36)$$

Analogously, at normal incidence this expression is not valid. Again, calculating this case separately and following the same steps as above we get,

$$\mathcal{G}_{(\mathrm{Gauss})}(\theta_1=0) = \frac{|r|^2}{|r_o|^2}\exp(-g_s) \times\left\{1-\sum_{n=1}^{\infty}\frac{g_s^n}{n!}\left(1-\exp\left(-\frac{y_D^2}{4n}\right)\right)\right\}. \quad (37)$$

This expression and the one given by Porteus[13] only coincide to first order in g_s, that is, in the limit of weak scattering surfaces.

To verify the accuracy of our approximations, we calculated $\rho_s^{(d)}$, for an exponential correlation function, by direct numerical integration of eq. (21) and compared the results with the corresponding values obtained by using eq. (31). In the

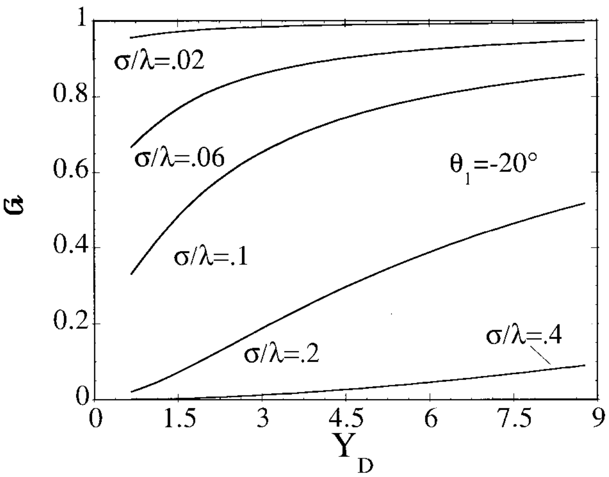

Figure 2. Gloss vs. y_D at an incidence angle of 20° for an exponential correlation function for different values of σ/λ. $(\delta\theta)_D = 1°$.

interval of σ/λ and y_D where gloss is sensitive to $y_D(0.02 \leq \sigma/\lambda \leq 0.2$ and $0.6 \leq y_D \leq 6)$, the maximum difference found was less than 1.5%. In conclusion, the series representation for gloss given in eqs. (32), (33), (36), and (37) provide a simple way to calculate gloss for a Gaussian and an exponential correlation function and which can be accomplished by hand calculators.

RESULTS AND DISCUSSION

In this section we discuss the consequences of the expressions for gloss derived above. In Figures 2 and 3 we plot gloss as a function of y_D, for several

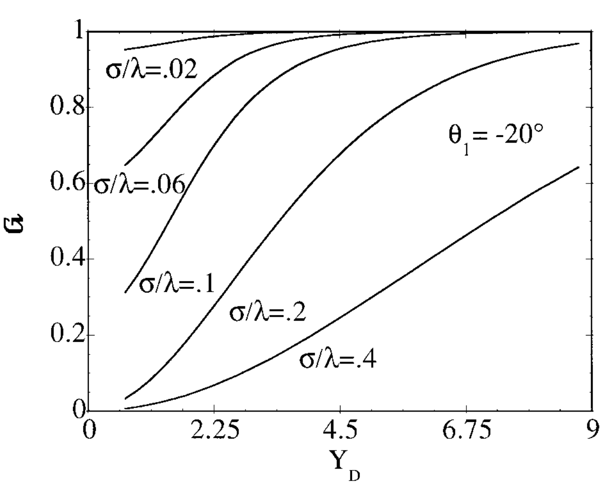

Figure 3. Gloss vs. y_D at an incidence angle of 20° for a Gaussian correlation function for different values of σ/λ. $(\delta\theta)_D = 1°$.

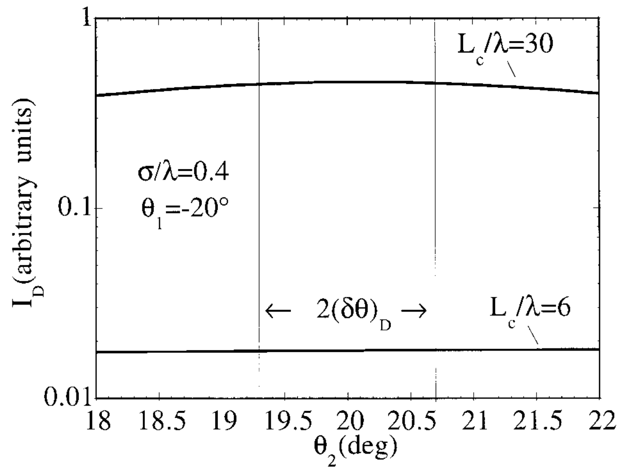

Figure 4. Correlation length effects on the diffuse scattering intensity (I_D) for an exponential correlation function and Gaussian distribution of heights. $\sigma/\lambda = 0.4$, $\theta_1 = 20°$, $\theta_3 = 0$.

values of σ/λ, for both an exponential and a Gaussian correlation functions, respectively. As expected, for very small values of σ/λ, gloss should be highly insensitive to y_D, because the coherent component will be dominant and the incoherent contribution will represent a small percentage of the total specularly reflected light. In this limit, eqs. (32) and (36) reduce to the Bennett and Porteus[18] expression given by

$$\mathcal{G} \approx \frac{|r|^2}{|r_o|^2} \exp(-g_s). \tag{38}$$

In Figure 2 we also observe that for very large σ/λ, again gloss is insensitive to changes in y_D. However, the reason for this is different from the previous case, because in this limit the incoherent contribution to the relative reflectance is fully dominant. To understand this, we show in Figure 4 the scattering pattern of the incoherent field for an exponential correlation function and a large σ/λ ($g_s \approx 22$). One can see that the light scattered diffusely is almost angle independent within the angular interval defined by the detector receiving system. In Figure 5 we show two mathematically generated one-dimensional random surface profiles with an exponential correlation function and a Gaussian distribution of heights, for the same parameters used in Figure 4. All random surfaces profiles were generated using the spectral method.[20] Because L_c roughly corresponds to the average lateral size of the surface's protuberances, the surface profile with $L_c/\lambda = 30$ and σ/λ

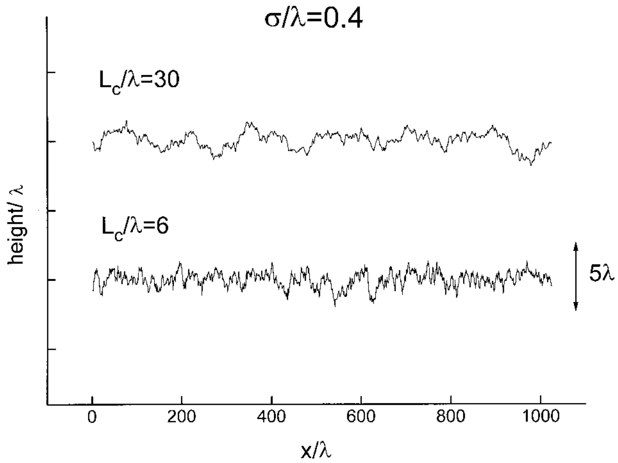

Figure 5. Mathematically generated random surface profiles for a Gaussian distribution of heights and an exponential height correlation function for $\sigma/\lambda = 0.4$.

$= 0.4$ looks less "wiggly" than for $L_c/\lambda = 6$. As one can see from Figure 5, the average slope ($\approx \sigma/L_c$) of the protuberances is large enough to give rise to an almost isotropic diffuse field, as shown in Figure 4. In this case, the integral within brackets in eq. (21) is independent of Ω, and the angular integration is done readily. This leads to a simple approximation for large g_s, namely:

$$\mathcal{G} \cong \frac{|r|^2}{|r_o|^2} \frac{y_D^2}{2} \cos\theta_1 \left(\frac{\Omega_D}{\pi(\delta\theta)_D^2} \right) \exp(-g_s) \times K(g_s) (g_s \gg 1), \quad (39)$$

where $K(g_s)$ is given by:

$$K(g_s)_{\text{exp}} = \int_0^\infty [\exp(g_s \exp(-x)) - 1] x \, dx, \quad (40)$$

for an exponential correlation function, and by:

$$K(g_s)_{\text{Gauss}} = \int_0^\infty [\exp(g_s \exp(-x^2)) - 1] x \, dx, \quad (41)$$

for a Gaussian correlation function. The factor $\Omega_D/(\pi(\delta\theta)_D^2)$ will be close to unity, depending on the geometry of the collecting aperture. For a circular aperture its value is unity.

Because eq. (39) is only valid in the case $g_s \gg 1$, we can replace eqs. (40) and (41) by its asymptotic limits, that is:

$$K(g_s)_{\text{exp}} \underset{g_s \gg 1}{\rightarrow} \frac{\exp(g_s)}{g_s^2}$$

and

$$K(g_s)_{\text{Gauss}} \underset{g_s \gg 1}{\rightarrow} \frac{\exp(g_s)}{2g_s}$$

Therefore, in this limit the expression for gloss will be reduced to:

$$\mathcal{G}_{(\text{exp})} \cong \frac{|r|^2}{|r_o|^2} \frac{y_D^2}{2g_s^2} \cos\theta_1 \left(\frac{\Omega_D}{\pi(\delta\theta)_D^2} \right) \quad (g_s \gg 1), \quad (42)$$

and

$$\mathcal{G}_{(\text{Gauss})} \cong \frac{|r|^2}{|r_o|^2} \frac{y_D^2}{4g_s} \cos\theta_1 \left(\frac{\Omega_D}{\pi(\delta\theta)_D^2} \right). \quad (g_s \gg 1). \quad (43)$$

Although for $g_s \gg 1$, $\mathcal{G} \approx (y_D)^2$, the proportionality factor is very small, and this is what limits the sensitivity of gloss on y_D, we will have to reach unrealistic high values of y_D to start detecting significant changes in gloss. The values of g_s for attaining these asymptotic expressions will depend on the correlation function; for instance, in the exponential case, this limit will be reached sooner because it tends to give flatter scattering patterns in the specular interval than those generated by a Gaussian correlation function. However, this limit is not the most interesting one because the surface is fully matte, whatever small changes we might register by varying y_D within experimental limits.

In many applications, surfaces are neither extremely glossy nor fully matte; they are fairly glossy with moderate values of g_s (≈ 1). It is in this intermediate region where gloss has the strongest sensitivity to y_D. To illustrate the difference in roughness in this region compared with the surface profiles shown in Figure 5, we present in Figure 6 two mathematically generated one-dimensional random surface profiles with a Gaussian distribution of heights and an exponential surface correlation function, for $\sigma/\lambda = 0.114$ and two different values of L_c/λ. As it is evident from Figure 6 the surfaces are much smoother than the one shown previously. Again, we see in this figure that the average slope of the surface

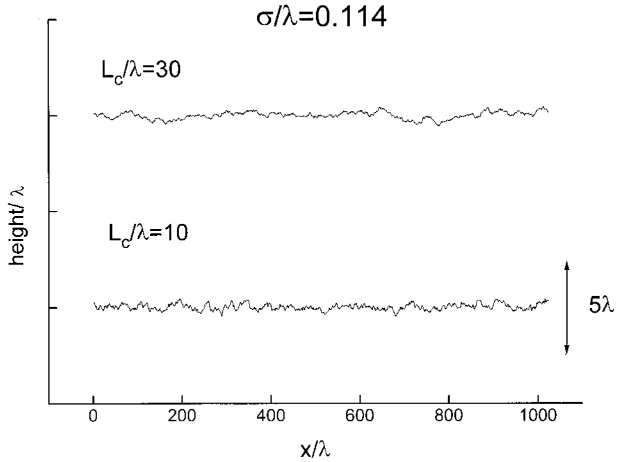

Figure 6. Mathematically generated random surface profiles for a Gaussian distribution of heights and an exponential height correlation function for $\sigma/\lambda = 0.114$.

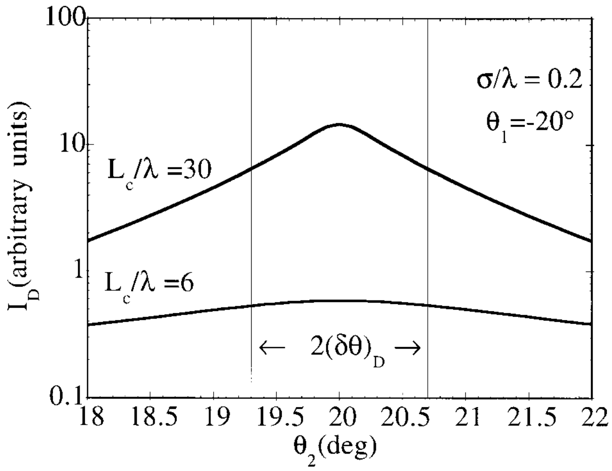

Figure 8. Correlation length effects on the diffuse scattering intensity (I_D) for an exponential correlation function and Gaussian distribution of heights. $\sigma/\lambda = 0.2$, $\theta_1 = 20°$, $\theta_3 = 0$.

protuberances decreases as the correlation length increases, and one would expect that the diffuse field will be more concentrated around the specular angle. This, in fact, is the case, as shown in Figure 7 where the diffuse intensity is plotted as a function of θ_2, for the same values of σ/λ and L_c/λ as in Figure 6. Figure 8 shows the same correlation effects on the diffuse scattering pattern for $\sigma/\lambda = 0.2$. Therefore, we see that for a moderate value of σ/λ, as we increase L_c/λ, the diffuse scattering tends to concentrate more within the detector's acceptance angle, and in consequence, gloss is increased by the contribution of the incoherently scattered field. This is illustrated in Figures 9 and 10, where we plot the incoherent contribution to gloss, $\mathcal{G}_i$, and gloss as a function of σ/λ for different values of L_c/λ, for both an exponential and a Gaussian correlation functions, respectively. As one can see, the incoherent fraction of gloss can be quite substantial for reasonably glossy surfaces ($\mathcal{G} \approx 75\%$). For example, in Figure 9 for a moderate value of $\sigma/\lambda = 0.1$, at $\theta_1 = -20°$ with $\delta\theta = 0.017$ rad and $L_c/\lambda = 40$, the incoherent fraction will be 66.4%. Even for very glossy surfaces ($\mathcal{G} \approx 90\%$), as long as L_c/λ is large, Figure 9 shows that the incoherent contribution can be quite important; for instance, for $\sigma/\lambda = 0.06$, leaving the other parameters the same, gloss is 89.9%,

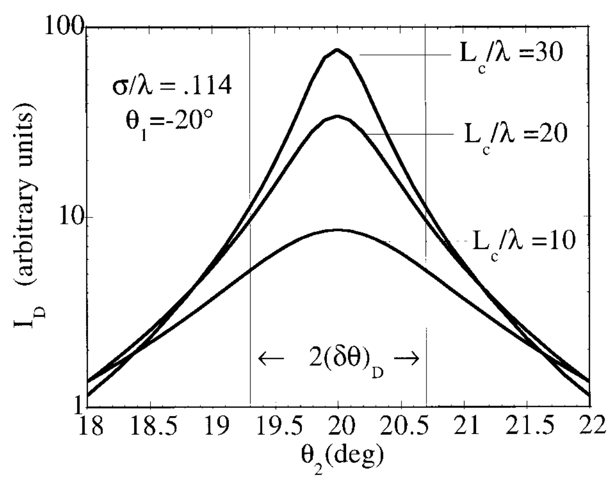

Figure 7. Correlation length effects on the diffuse scattering intensity (I_D) for an exponential correlation function and Gaussian distribution of heights. $\sigma/\lambda = 0.114$, $\theta_1 = 20°$, $\theta_3 = 0$.

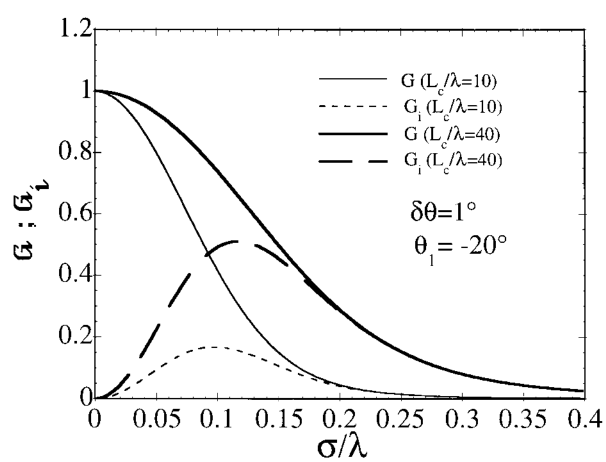

Figure 9. Gloss and the incoherent contribution to gloss vs. σ/λ for two different L_c/λs for an exponential correlation function. $\theta_1 = -20°$ and $(\delta\theta)_D = 1°$.

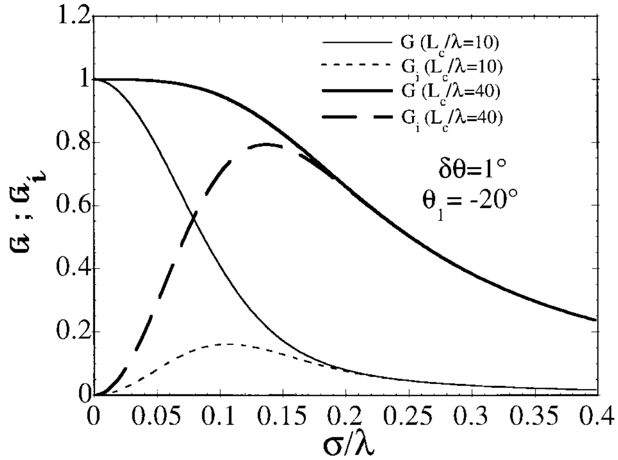

Figure 10. Gloss and the incoherent contribution to gloss vs. σ/λ for two different L_c/λ's for a Gaussian correlation function. $\theta_1 = -20°$ and $(\delta\theta)_D = 1°$.

32.7% of which is incoherent in origin. For a smaller correlation length this contribution will be reduced because the proportion of the diffusely scattered beam that reaches the detector will be smaller. As an example of the latter, for $L_c/\lambda = 10$ and $\sigma/\lambda = 0.1$ with the same values of the other parameters as above, the diffuse contribution to gloss represents $\approx$ 40.2% of the total, which is still quite large. The proportion of the incoherent contribution can be enhanced by increasing the receiving aperture, because the detector will register a larger portion of the diffuse field. In conclusion, for moderately glossy surfaces, gloss, as usually measured, can be strongly incoherent in nature.

From the introductory discussion it is clear that, in many applications, an exponential correlation function will be a close guess for the real correlation function. The two examples given, of a molded filled plastic and a coated paper, correspond to two extreme cases—one highly glossy and the other fairly matte—where the surface topography has a completely different origin. In both cases the correlation function that best fitted the experimental angle-resolved light scattering was an exponential or close to an exponential, and the correlation length was not larger than 15 μm. To study the effects on gloss produced by deviations from an exponential correlation function, we shall present numerical results of gloss, as a function of y_D, for two models of quasi-exponential correlation functions. The first one, given by Lettieri et al.,[2] consists on modified exponentials of the form $C(R) = \exp(-|R/L_c|^{\alpha})$ where α is an exponent between 1 and 2. Here, we shall calculate gloss for $\alpha = 1.15$ and 1.35, which were the values that best fitted their light-scattering experiments. These correspond to positive deviations from an exponential, that is, that the slope at $R = 0$ is lower than that of an exponential.

The other is the model discussed by Hoenders et al.[17] of a fluctuating facet-scattering surface. The family of correlation functions proposed by these authors is of the form:

$$C_{\nu}(R) = \frac{(p_{\nu}R/L_c)^{\nu}}{2^{\nu-1}\Gamma(\nu)} K_{\nu}(p_{\nu}R/L_c)$$

where K_{ν} is the modified Bessel function of order ν, Γ is the Gamma function and p_{ν} is a scalar whose value is such that for $R = L_c$, $C_{\nu}(L_c) = e^{-1}$. For $\nu = \frac{1}{2}$ the correlation function corresponds exactly to an exponential while in the limit $\nu \to \infty$, this tends to a Gaussian correlation function. To represent a quasi-exponential with this family of functions, we will choose $\nu = 0.7$ and 0.3 as examples of positive and negative deviations from an exponential. Figure 11 shows the correlation functions used in our calculations and how they compare with an exponential. Because the correlation function for $\nu = 0.7$ almost coincides with the modified exponential for $\alpha = 1.15$, we shall only show the results for one of these, the K-correlation function with $\nu = 0.7$. Because, in the case of quasi-exponentials considered here, it is not possible to derive a simple series representation for gloss, the calculations for these were done by performing a numerical integration of the triple integral that appears in eq. (28). The latter was done

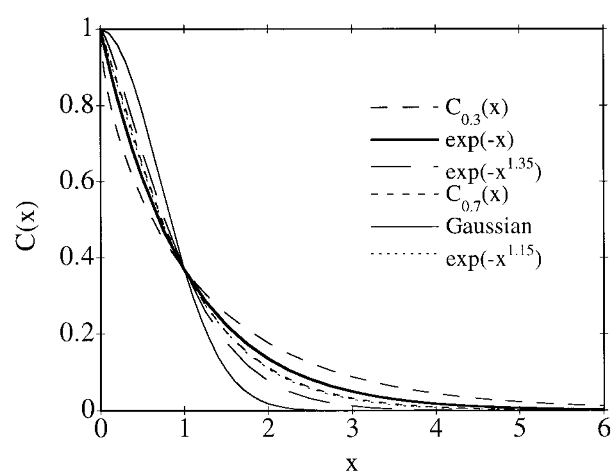

Figure 11. Quasi-exponential correlation functions compared with an exponential and a Gaussian.

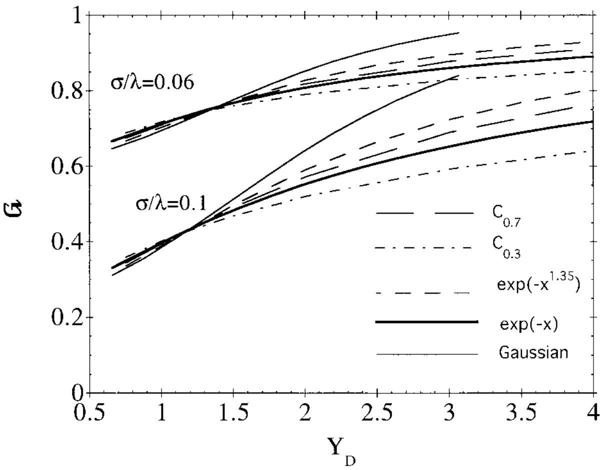

Figure 12. Gloss vs. Y_D at an incidence angle of 20° for various correlation functions for two values of σ/λ. $(\delta\theta)_D = 1°$.

by means of Mathematica[21] using a multidimensional method and keeping an absolute accuracy of the multiple-integration algorithm of at least three significant figures. In Figure 12 we plot the corresponding curves for gloss as a function of y_D and we compared them to those of an exponential and a Gaussian, for two values of σ/λ. As one can see, for a given σ, all the gloss curves intersect within a very narrow interval of y_Ds. For y_Ds between 0.6 and the zone of intersection, the gloss is rather insensitive to the correlation function. On the contrary, for large y_Ds, this difference is quite pronounced. However, for a typical glossmeter receiving aperture at $\theta_1 = 20°$ of $(\delta\theta)_D = 0.9°$, and a correlation length of 15 μm, for a wavelength of 0.5 μm, $y_D \approx 3$. For this value, the quasi exponential gloss curves differ from the exponential by at most 11%.

In many filled polymers the surface height correlation extends roughly to the protuberances within themselves, that is, it corresponds to the self correlation of the protuberances. In this case, the correlation length represents an average of the lateral extension of the protuberances. The average slope of the rough profile will be determined by the size of the fillers, melt viscosity, as well as other processing parameters such as the injection rate and mold temperature.[4–7] This means that the correlation length will be related to a combination of such parameters. Therefore, a measurement of L_c could give a deeper insight in the effect of such variables in, for example, the elongational induced stresses at surface.

The fact that the collecting detector angle scales as the correlation length can be used to determine L_c, for moderately glossy surfaces, by a very simple method, which can be incorporated in commercial glossmeters. There are two situations—one when we know the surface correlation function, and the other when this is not the case. In the first case, if we know σ, using gloss-y_D plots, such as the ones shown in Figures 2 and 3, we first look for the y_D that gives the same value of the experimentally measured gloss. From this value, $(y_D)_T$, and the experimental detector collecting half-angle, $[(\delta\theta)_D]_E$, we can calculate L_c as:

$$L_c = \frac{(y_D)_T \cdot}{k((\delta\theta)_D)_E}\,. \tag{44}$$

In practice, σ can be measured independently, for instance, incorporating an extra infrared light source at a fixed angle in the design of the glossmeter and using the classical method by Bennett an Porteus.[18]

In the case that we do not know the height correlation function of the surface, for a measured σ, we can vary the detector aperture until we reach the theoretical value for gloss at the intersection between an exponential and a Gaussian gloss curve. Because, for other correlation functions the intersection with the exponential will occur almost at the same y_D, we can determine L_c with high precision, independently of the statistical nature of the surface, from the value of the experimental receiving aperture and that of y_D at the theoretical intersection. If we neglect for the moment the error propagation introduced by the measurement of σ, for $\sigma/\lambda = 0.1$ the maximum error committed by this procedure is less than 1%, and for $\sigma/\lambda = 0.06$ is of 3.4%. The latter corresponds to the generating correlation function of $C_{0.3}$, that is, a negative deviation from an exponential. For the positive deviations calculated, the error is much smaller.

Additionally we could generate a curve of gloss vs. $(\delta\theta)_D$ by varying $(\delta\theta)_D$. To construct a gloss vs. y_D curve we just have to multiply $(\delta\theta)_D$ by the factor

$$\left.\frac{y_D}{(\delta\theta)_D}\right|$$

at the intersection. The experimental gloss curve constructed by such a procedure will give us information about the type of correlation function associated to the surface roughness.

If these modifications and calculations are incorporated into commercial glossmeters, one could obtain reasonable estimates of the main statistical parameters of the surface and serve as quantitative developing tool.

CONCLUSIONS

In this article we derived a general expression for gloss within the scalar Kirchhoff's theory where the effects of correlation length L_c, the detector collecting half-angle $(\delta\theta)_D$, and the statistical nature of the surface are taken into account. It was shown that regardless of the correlation function used, gloss at a given angle depends exclusively on two parameters, $(\sigma/\lambda)\cos\theta_1$ and $(L_c/\lambda)(\delta\theta)_D$. A corollary of the last statement is that gloss scales with the correlation length in the same way as with the detector collecting angle, for all correlation functions of the form $C(R/L_c)$. Analytic expressions in the form of series expansions for gloss were derived for an exponential and a Gaussian correlation function in terms of these parameters. Calculations with these show that the incoherent contribution to gloss is significant in common polymeric surfaces. The latter implies that surface height correlations cannot be neglected in the evaluation of gloss. It was shown that Porteus expression, for specular reflectance for a Gaussian correlation function facet model, corresponds to the present result only in the limit of weak scattering surfaces. The results for the exponential and Gaussian correlation function were compared with the ones obtained by direct numerical integration for quasi-exponential correlation functions. It was found that, for a given σ, all gloss vs. y_D curves intersect almost at the same point. The equivalence between L_c/λ and $(\delta\theta)_D$ can be used to determine the correlation length, and an experimental method by means of a glossmeter was proposed for this purpose, independent of the statistical nature of the surface.

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APPENDIX 1

In this appendix we calculate the normalization power P_N, which appeared in the definition of gloss and was defined as:

$$P_N = \int_{\Omega_1}^{\Omega_1+\Omega_D} \left(\frac{dP_o}{d\Omega}\right) d\Omega. \quad (2)$$

For a rectangular illuminated section of sides $2X$ and $2Y$ and an incident electric field of magnitude one, the power scattered per solid angle by a smooth surface is:

$$\frac{dP_o}{d\Omega} = \left(\frac{c_o}{4\pi}\right)\left(\frac{k^2}{16\pi^2}\right)A_M^2|r_o|^2(\cos\theta_1 + \cos\theta_2)^2 \times \operatorname{sinc}^2(kAX)\operatorname{sinc}^2(kBY), \quad (A1)$$

where $A_M = 4XY$, $|r_o|^2$, k, λ, A, and B have been previously defined.

The total power scattered within the solid angle subtended by the detector can be written as:

$$P_N = \left(\frac{c_o}{4\pi}\right)\left(\frac{k^2}{16\pi^2}\right)16\cos^2\theta_1 A_M^2|r_o|^2 \times \int_0^{(\delta\theta)_D/\sin\theta_1}\int_0^{(\delta\theta)_D} \operatorname{sinc}^2(kX\cos\theta_1\delta\theta)\operatorname{sinc}^2 \times (kY\sin\theta_1\theta_3)\sin\theta_1 d(\delta\theta)d\theta_3, \quad (A2)$$

where A, B, and $d\Omega$ were substituted by their first-order approximations in $\delta\theta = \theta_2 - \theta_1$.

The limits of integration are chosen such that the detector's collecting solid angle is constant and independent of the specular angle. However, for a large detector collecting angle, $\Omega_D \gg 1/k^2XY$, we can replace the upper integration limits by infinity because the linear dimensions of the main diffraction peak will be of the order of $1/kX$ (or $1/kY$). Therefore, replacing the upper integration limits by infinity, eq. (A2) reduces to eq. (19) with $D(\Omega_D) = 1$.

On the other hand, in the opposite limit, when the detector's angular aperture is so small that $\Omega_D \ll 1/k^2XY$, it will collect only the very central part of the zero-order diffraction peak. Therefore, we have to evaluate eq. (A2) at $\delta\theta = 0$ and $\theta_3 = 0$, in which case the integral will be equal to $\Omega_D/4$ and eq. (A2) will reduce to eq. (18).

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