

Theory of differential and integral scattering of laser radiation by a high-precision dielectric surface

V V Azarova, V G Dmitriev, Yu N Lokhov, K N Malitskiĭ

Abstract. A theoretical study was made of the differential and integral scattering of light by high-precision optical dielectric surfaces. The technique of modified curvilinear transformation was employed to derive the expressions that correspond to the effect of irregularity slope fluctuations, which is not considered in the conventional vector theory of differential scattering. It is shown that it is possible to neglect the effect of irregularity slope to a high degree of accuracy and thereby resort to conventional vector theory to determine the roughness parameters. Expressions were derived for the total integral scattering for different ratios between the correlation length and the wavelength of the scattered radiation.

1. Introduction

Optical reflectometry is used extensively for metrological studies of rough optical surfaces (the rms roughness and the correlation length). This method involves measurements of the power of the scattered radiation normalised to the power of the incident or specularly reflected radiation: integral power P_{TIS} in the method of total integral scattering (TIS) or the angular power P_{ARS} in the method of differential scattering [angular resolved scattering (ARS)] (see, e.g. Refs [1–7]).

The interpretation of the results of measurements by the TIS and ARS methods calls for development of an adequate physico-mathematical model of light scattering by a rough optical surface. At present, the development of this model is still far from completion, and our work is an attempt to improve it.

To calculate the scattering by a rough optical surface, it is standard practice to invoke the model based on the so-called vector theory [3], which uses a curvilinear transformation of coordinates [2]. This approach permits the use of perturbation theory. The statistical properties of surface irregularities are described in this case by the function of spectral power density. In doing this, it is usual to restrict the consideration to the Gaussian or the exponential statistics to describe the surface roughness. However, the experimental data are in sat-

isfactory agreement with the vector-theory calculations only in a narrow range of scattering angles. This theory fails to explain large scattering at angles remote from the specular angle; i.e. the vector theory cannot describe the experiment over the entire range of scattering angles.

It is likely that the primary reason for the discrepancy between the vector-theory calculations and the experimental data is the complex nature of the irregularity statistics, which depends on the range of scattering by angles. In this case it is impossible to describe scattering BY using a correlation function with unique values of the rms roughness and the correlation length.

In optical reflectometry, the rms surface roughness σ at a wavelength λ is commonly measured by the TIS method, by using the expression [1]

$$P_{\text{TIS}} = \left(\frac{4\pi\sigma}{\lambda} \right)^2. \quad (1)$$

Formula (1) is widely used in the literature. However, we will show here (also, see Ref. [8]) that it is appropriate only for those surfaces for which the correlation length l satisfies the relationship $l \geq \lambda$. A different formula holds for short correlation lengths ($l \ll \lambda$) (see below). This means that the experimental data obtained by the TIS method cannot be interpreted without prior measurement of the correlation length l (e.g. by atomic-force microscopy) and determining the relation between l and λ (otherwise only some ‘effective’ rms roughness should be considered).

2. Formulation of the problem and basic equations

Consider a plane monochromatic wave

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0 \exp(-i\omega t + i\mathbf{k}_0 \mathbf{r}), \quad (2)$$

incident at an angle θ_0 from vacuum on a rough surface of a dielectric with the real permittivity $\varepsilon(\omega)$. The surface micro-profile is described by the random function $z = f(x, y)$ (the z axis is directed normal to the surface). On average, the surface is plane, $\langle f(x, y) \rangle = 0$, and the rms deviation of the profile height σ is small compared with the wavelength of the optical radiation and the correlation length:

$$\sigma \ll l, \quad \sigma \ll \lambda \cos \theta_0. \quad (3)$$

The stationary problem of light scattering by a rough surface involves solving the wave equation

$$\Delta \mathbf{E}(\mathbf{r}) + \varepsilon(\mathbf{r}) \frac{\omega^2}{c^2} \mathbf{E}(\mathbf{r}) = 0, \quad (4)$$

$$\varepsilon(\mathbf{r}) = \begin{cases} 1, & z > f(x, y), \\ \varepsilon, & z < f(x, y) \end{cases}$$

V V Azarova, V G Dmitriev, Yu N Lokhov Polyus Scientific-Research Institute (Federal State Unitary Enterprise), ul. Vvedenskogo 3, 117342 Moscow, Russia; e-mail: vgdmitr@orc.ru
K N Malitskiĭ Moscow Institute of Physics and Technology (State University), Institutskii per. 9, 141700 Dolgoprudnyi, Moscow Province, Russia

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with the boundary conditions at the surface

$$E_{n1} = \varepsilon E_{n2}, \quad E_{t1} = E_{t2} \quad (5)$$

where the subscripts 1 and 2 refer to vacuum and the dielectric, whereas the subscripts n and t correspond to the normal and tangential components of the field vector.

Following vector theory [3], we pass from the orthogonal coordinate system xyz to the curvilinear system $u_1 = x$, $u_2 = y$, $u_3 = z - f(x, y)$ [2], in which the boundary conditions are now specified at the surface $u_3 = 0$. The existence of a microprofile may be treated as a perturbation for the ideal problem (i.e. the problem with an ideal plane surface, or the Fresnel problem). Vector theory uses a perturbation linear in the microprofile function $f(x, y)$ and its derivatives. In the general case, the random function $f(x, y)$ and its derivatives can be expanded in spatial Fourier harmonics, so that a rough surface can be regarded as a superposition of two-dimensional diffraction gratings. For an individual partial Fourier harmonic, vector theory is in essence a regular theory of the interaction of light with a diffraction grating (with the subsequent synthesis of the scattered partial fields over the entire ensemble of these partial gratings).

Conventional vector theory does not take into account the fluctuations of the surface slope, which change the boundary conditions and thereby affect the scattering, because the corresponding perturbations are quadratic in $f(x, y)$ and its derivatives. Furthermore, the solution of the scattering problem in vector theory is sought in the curvilinear coordinate system, which is not equivalent to the orthogonal one over the entire space. After deriving the solution in the curvilinear system, we require an inverse passage to the orthogonal coordinate system to attain a rigorous solution of the problem. Otherwise, the solution associated with a change in the boundary conditions may be lost.

Consider the modified curvilinear transformation

$$u_1 = x, \quad u_2 = y, \quad u_3 = z - f(x, y) \exp\left(-\frac{u_3^2}{2a^2}\right), \quad (6)$$

where a is the width (in z) of the domain of scattering formation. We assume below that only the boundary dipoles make a contribution to scattering, and therefore the passage to the limit $a \rightarrow 0$ is used in estimates. (In reality, different physical approaches to the interpretation of scattering sources yield different expressions for a . These issues will be considered in detail in our next paper.)

The existence of a 'cut-off factor' in one of expressions (6) has the effect that the coordinate systems will be equivalent in the domain $z \gg a$ and, hence, in the region $z \gg \lambda$ in which the scattered field resides. This modification allows us to find a more rigorous (in comparison with the vector theory) solution that is adequate for the experimental situation.

After passage to the curvilinear coordinate system (6), equation (4) with the boundary conditions (5) can be written as

$$\Delta_0 \mathbf{E}(\mathbf{U}) + \varepsilon_0 \frac{\omega^2}{c^2} \mathbf{E}(\mathbf{U}) = \hat{H}(\mathbf{U}) \mathbf{E}(\mathbf{U}), \quad (7)$$

$$\varepsilon_0 = \begin{cases} 1, & u_3 > 0, \\ \varepsilon, & u_3 < 0, \end{cases}$$

$$\Delta_0 = \frac{\partial^2}{\partial u_1^2} + \frac{\partial^2}{\partial u_2^2} + \frac{\partial^2}{\partial u_3^2},$$

where \mathbf{U} is the radius vector in the curvilinear coordinate system and $\hat{H}(\mathbf{U}) \mathbf{E}(\mathbf{U})$ are the terms that appear in passing to the curvilinear coordinate system. As shown in Appendix 1, the perturbation operator $\hat{H}(\mathbf{U})$ can be represented as a sum of two perturbation operators \hat{H}_1 and \hat{H}_2 :

$$\hat{H}(\mathbf{U}) = \hat{H}_1(\mathbf{U}) + \hat{H}_2(\mathbf{U}). \quad (8)$$

The first perturbation, linear in $f(x, y)$, corresponds to the scattering from microstructures without considering their slope. A calculation with this perturbation corresponds to a calculation within the framework of the vector scattering theory [3]. The second perturbation, which comprises the products of $f(x, y)$, its derivative, and the square of its derivative, takes into account the irregularity of slope fluctuations.

Therefore, the passage to the curvilinear coordinate system reduces the initial wave equation (4) to an inhomogeneous wave equation whose solution may be sought by the perturbation theory techniques in the small-roughness approximation.

We adopt the Fresnel problem as the zero-order approximation:

$$\Delta_0 \mathbf{E}(\mathbf{U}) + \varepsilon_0 \frac{\omega^2}{c^2} \mathbf{E}(\mathbf{U}) = 0, \quad u_3 = 0. \quad (9)$$

We will use well-developed methods of the Green function and expansion in spatial Fourier integrals to find the intensity of the scattered field in the first order of the perturbation theory (for more details, see Refs [4, 5]).

3. Results of calculations

Let $R^{(1,2)}$ be the scattering coefficients corresponding to the first and second perturbations in expression (8) and equal to the normalised intensities of the radiation scattered in a unit solid angle in the direction of an angle θ (Fig. 1). It is obvious that these coefficients describe the differential scattering. The normalisation was performed to the incident radiation intensity. Calculations give the expressions

$$R_{ik}^{(1)} = \frac{\omega^4 (\varepsilon - 1)^2}{\pi^2 c^4} g(|\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|) F_{ik}^{(1)}(\theta, \varphi, \theta_0, \varepsilon), \quad (10)$$

$$R_{ik}^{(2)} = \frac{\omega^4 (\varepsilon - 1)^2}{\pi^2 c^4} h(|\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|) F_{ik}^{(2)}(\theta, \varphi, \theta_0, \varepsilon), \quad (11)$$

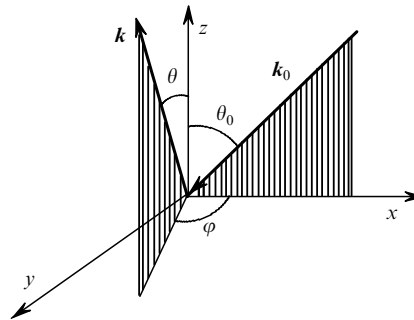


Figure 1. Schematic diagram showing the mutual arrangement of the wave vectors of the incident and scattered waves.

where the first (i) and the second (k) subscripts denote polarisations of the scattered and incident fields, respectively ($i, k = s, p$);

$$h(|\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|) = \frac{1}{4} |\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|^4 g^2(|\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|); \quad (12)$$

$$g(|\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|) = \int \exp[-i(\mathbf{k}_\perp - \mathbf{k}_{\perp 0})\mathbf{r}] \Psi(r) dS$$

is the function of spectral power density of the surface irregularities, which is a spatial two-dimensional Fourier transform of the correlation function $\Psi(r) = \Psi(|\mathbf{r}' - \mathbf{r}''|) = \langle f(\mathbf{r}') f(\mathbf{r}'') \rangle$ (the averaging is performed over the area of the scattering surface);

$$\mathbf{k}_\perp = \{k_x, k_y, 0\} = \frac{2\pi}{\lambda} \{\sin \theta \cos \varphi, \sin \theta \sin \varphi, 0\}, \quad (13)$$

$$\mathbf{k}_{\perp 0} = \{k_{x0}, 0, 0\} = \frac{2\pi}{\lambda} \{\sin \theta_0, 0, 0\}$$

are the wave-vector components of the scattered and incident waves, respectively, perpendicular to the normal to the surface. The expressions for the angular functions are given in Appendix 2.

In the derivation of relations (10) and (11), we took advantage of the fact that the cross correlator of a random function and its derivative is zero whereas the correlator of the derivative of a random function is equal to the second derivative of the correlation function (see, e.g. Ref. [7]).

The angular intensity of the field scattered by a rough surface at angles θ, φ is found as the sum of intensities (10) and (11), which correspond to the chosen polarisations of the incident and scattered fields. Expressions (10) represent the result of vector theory [3] and expressions (11) describe the additional scattering associated with the irregularity slope fluctuations.

The power of integral scattering of the radiation with the i th polarisation, i.e., of the total radiation scattered into the upper half-space, is found by integrating the corresponding expressions for the power of differential scattering over all possible scattering and azimuth angles:

$$P_{\text{TIS}}^{(i)}(\theta_0) = \int_0^{\pi/2} \sin \theta d\theta \int_0^{2\pi} [R_{si}(\theta, \varphi, \theta_0) + R_{pi}(\theta, \varphi, \theta_0)] d\varphi. \quad (14)$$

The correlation function of irregularities is assumed to be Gaussian:

$$\Psi_G(r) = \sigma^2 \exp\left(-\frac{r^2}{l^2}\right), \quad (15)$$

$$g_G(\mathbf{k}_\perp - \mathbf{k}_{\perp 0}) = \pi \sigma^2 l^2 \exp\left(-\frac{|\mathbf{k}_\perp - \mathbf{k}_{\perp 0}|^2 l^2}{4}\right). \quad (16)$$

For the two limiting ratios of the correlation length to the wavelength, we obtain the following expressions for the power of the total integral scattering normalised to the intensity of radiation incident normally on the surface:

$$P_{\text{TIS}} \approx \left(\frac{4\pi\sigma}{\lambda}\right)^2 \left(\frac{1 - \sqrt{\varepsilon}}{1 + \sqrt{\varepsilon}}\right)^2, \quad l \geq \lambda, \quad (17)$$

$$P_{\text{TIS}} \approx \frac{1}{6} \left(\frac{4\pi\sigma}{\lambda}\right)^2 \left(\frac{\pi l}{\lambda}\right)^2 (1 - \sqrt{\varepsilon})^2, \quad l \ll \lambda. \quad (18)$$

One can see that expression (17) commonly used in the literature is valid only for a certain relation between the correlation length and the wavelength.

4. Discussion

A calculation made by using expressions (10) and (11), which describe the differential scattering related to the effect of fluctuations of irregularity height and slope, suggests that the irregularity height fluctuations exert a dominant effect on the scattering. This case is adequately described by conventional vector theory [3]. The error arising from neglecting the irregularity slope fluctuations is small. For the s-polarised radiation, the error is maximum for small angles of incidence and large scattering angles and does not exceed 10^{-4} . For p-polarised radiation, the error is maximum for angles of incidence $\sim 50^\circ$ and large scattering angles and does not exceed 10^{-3} . As an example Fig. 2 shows the theoretical differential pp-scattering indicatrices, which correspond to expressions (10) and (11), for the surface of amorphous silica.

The primary implication is as follows. The vector scattering theory, which disregards the effect of irregularity slope fluctuations, can be used to evaluate very precisely the roughness parameters of optical surfaces for $l \ll \lambda$.

Because the expressions for the total integral scattering for different ratios of l to λ are different, the rms roughness σ cannot be determined reliably without a preliminary measurement of the correlation length by other techniques, e.g., by atomic-force microscopy. In other words, the measurement of σ by the method of integral scattering of optical radiation yields a value averaged over the spatial structures with the lateral dimension exceeding λ . For instance, for a total integral scattering of 10^{-5} ($\lambda = 0.63 \mu\text{m}$) by a rough quartz surface with $l = 0.1 \mu\text{m}$, the rms roughness calculated by using formula (17) would be $\sim 10 \text{ \AA}$, whereas the real rms roughness calculated by formula (18) is $\sim 20 \text{ \AA}$.

The exact expression for the power of integral scattering (14) can be calculated by numerically employing relations (10) and (11) for differential scattering. The results of calculations for a quartz substrate are presented in Fig. 3, which shows the relative error in determining the rms roughness for different l by formulas (17) and (18). One can see, in particular, that

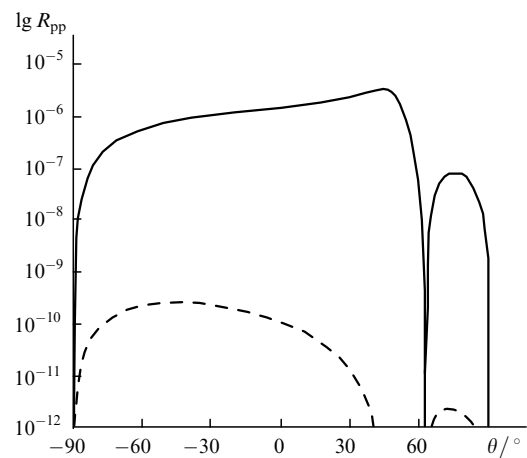


Figure 2. Indicatrices of differential scattering calculated by formulas (10) (solid line) and (11) (dashed line), for a polished quartz substrate for $\varepsilon = 2.12$, $\lambda = 0.63 \mu\text{m}$, $\sigma = 10 \text{ \AA}$, $l = 1.0 \mu\text{m}$, angle of incidence of 50° , and the Lorentzian statistics.

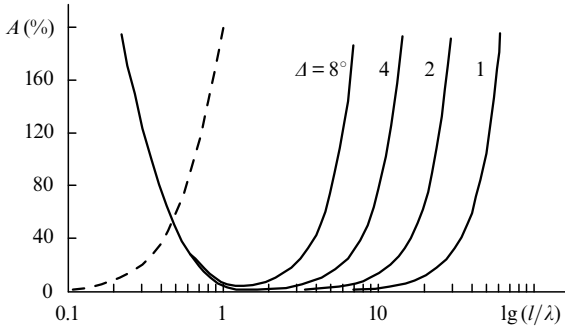


Figure 3. Relative error A of calculating the rms roughness by using the limiting formulas (17) (solid lines) and (18) (dashed line) versus l/λ for different angular widths Δ of the entrance opening (the position of the dashed line is independent of Δ).

the range where formula (17) yields accurate results is limited by the condition $l > \lambda$. For $l < \lambda/2\pi$, expression (18) can be used, and in the other cases the numerical integration should be used.

5. Conclusions

We have developed the theory of differential and integral scattering of optical radiation by rough dielectric surfaces in the case of small (in comparison with λ) irregularities. It was shown that the use of a modified curvilinear transformation allows us simultaneously to include the effect of the fluctuations of both irregularity height and slope on the differential (angular-resolved) scattering. The dominant mechanism governing the differential scattering by optical surfaces is the irregularity height fluctuation, whose effect is described by conventional vector theory. The neglect of the effect of irregular slope fluctuations results in an error that does not exceed 10^{-4} for the s-polarised and 10^{-3} for the p-polarised radiation.

Different expressions for the power of total integral scattering were derived in our work for two different ratios of correlation length to wavelength. This difference does not permit us to obtain reliable values of the rms roughness without prior determination of the correlation length by other techniques, e.g., by atomic-force microscopy. The theory elaborated in this work permits a more adequate interpretation of experimental results in the metrology of high-precision optical surfaces.

Appendix 1

Wave equation in curvilinear coordinates

The curvilinear transformation has the form:

$$\begin{cases} u_1 = x, \\ u_2 = y, \\ u_3 = z - f(x, y) \exp(-u_3^2/2a^2). \end{cases} \quad (\text{A1.1})$$

The metric tensor of the curvilinear coordinate system is

$$g_{ik} = \begin{pmatrix} 1 + \varphi^2 f_{u_1}^{\prime 2} & f_{u_1}' f_{u_2}' \varphi^2 & \frac{\varphi f_{u_1}'}{\psi} \\ f_{u_1}' f_{u_2}' \varphi^2 & 1 + \varphi^2 f_{u_2}^{\prime 2} & \frac{\varphi f_{u_2}'}{\psi} \\ \frac{\varphi f_{u_1}'}{\psi} & \frac{\varphi f_{u_2}'}{\psi} & \frac{1}{\psi^2} \end{pmatrix}. \quad (\text{A1.2})$$

Here, the subscript in the derivatives denotes the variable with respect to which the partial derivative is taken, and the following notations are adopted:

$$\varphi = \exp\left(-\frac{u_3^2}{2a^2}\right), \quad \psi = \left[1 - \left(\frac{u_3}{a^2}\right) f \varphi\right]^{-1}, \quad (\text{A1.3})$$

the Jacobian of the transformation (A1.1) being

$$g^{1/2} = |g_{ik}|^{1/2} = \psi^{-1}. \quad (\text{A1.4})$$

The Laplacian in the new coordinate system has the form

$$\Delta = g^{-1/2} \frac{\partial}{\partial u_i} \left(g^{ik} g^{1/2} \frac{\partial}{\partial u_k} \right), \quad (\text{A1.5})$$

where g^{ik} is the fundamental tensor of the curvilinear coordinate system (A1.1) related to the metric tensor by the expression

$$g^{il} g_{lk} = \delta_{ik}; \quad (\text{A1.6})$$

where δ_{ik} is the Kronecker delta. The Laplacian can be represented as

$$\Delta = \Delta_0 - \hat{H}, \quad (\text{A1.7})$$

where

$$\Delta_0 = \frac{\partial^2}{\partial u_1^2} + \frac{\partial^2}{\partial u_2^2} + \frac{\partial^2}{\partial u_3^2}. \quad (\text{A1.8})$$

The expression for the perturbation operator in its complete form is rather cumbersome. We restrict our consideration to the perturbation terms linear and quadratic in $f(x, y)$ and its derivatives. It is assumed that the xz plane is the plane of incidence, with the consequence that the zero-order approximation field E_0 is independent of $u_2 = y$. We use the following expansion of the function ψ :

$$\psi \sim 1 + \frac{u_3}{a^2} f \varphi. \quad (\text{A1.9})$$

In the calculation of scattering, the perturbation operator \hat{H} is integrated with respect to u_3 . Rearranging the perturbation with integration by parts with respect to u_3 and taking advantage of the slowness of the variation of the zero-order approximation field, we obtain expressions which are omitted here owing to their awkwardness. In these expressions, the perturbation comprises terms which either do or do not contain the factor u_3/a^2 . Integrating the former terms gives expressions that are independent of a , and integrating the latter terms gives expressions that depend on a . For this reason, in view of the passage to the limit $a \rightarrow 0$, the perturbation terms not containing the factor u_3/a^2 can be disregarded. (This passage to the limit corresponds to the scattering sources concentrated at the interface.) Since the correlator of a random function and its derivative $\langle f f' \rangle = 0$, the corresponding terms may be omitted. Then the perturbation operator is expressed as

$$\hat{H} = \hat{H}_1 + \hat{H}_2, \quad (\text{A1.10})$$

$$\hat{H}_1 = -2 \frac{u_3}{a^2} \varphi f \frac{\partial^2}{\partial u_3^2},$$

$$\hat{H}_2 = \frac{u_3}{a^2} \varphi^2 (2 f_{u_1}^{\prime 2} + 2 f_{u_2}^{\prime 2} + f_{u_1}'' f + f_{u_2}'' f) \frac{\partial}{\partial u_3}.$$

Therefore we have obtained the perturbation associated with the representation of the wave equation in the curvilinear coordinate system, correct to the quadratic terms.

Appendix 2

Expressions for the angular functions

The expressions for the angular functions that appear in the relation for the scattering coefficients (10) and (11) have the form

$$F_{ss}^{(1)} = \frac{\cos \theta_0 \cos^2 \theta \cos^2 \varphi}{[\cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2 [\cos \theta + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2},$$

$$F_{sp}^{(1)} = \frac{\cos \theta_0 \cos^2 \theta \sin^2 \varphi (\varepsilon - \sin^2 \theta_0)}{[\varepsilon \cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2 [\cos \theta + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2},$$

(A2.1)

$$F_{ps}^{(1)} = \frac{\cos \theta_0 \cos^2 \theta \sin^2 \varphi (\varepsilon - \sin^2 \theta_0)}{[\cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2 [\varepsilon \cos \theta + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2},$$

$$F_{pp}^{(1)} = \frac{\cos \theta_0 \cos^2 \theta}{[\varepsilon \cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2}$$

$$\times \frac{[\cos \varphi (\varepsilon - \sin^2 \theta_0)^{1/2} (\varepsilon - \sin^2 \theta)^{1/2} - \varepsilon \sin^2 \theta_0 \sin \theta]^2}{[\varepsilon \cos \theta + (\varepsilon - \sin^2 \theta)^{1/2}]^2},$$

$$F_{ss}^{(2)} = \frac{\cos \theta_0 \cos^2 \theta \cos^2 \varphi}{(\varepsilon - \sin^2 \theta_0)}$$

$$\times \frac{1}{[\cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2 [\cos \theta + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2},$$

$$F_{sp}^{(2)} = \frac{\cos \theta_0 \cos^2 \theta \sin^2 \varphi (\varepsilon - \sin^2 \theta)}{[\varepsilon \cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2 [\cos \theta + (\varepsilon - \sin^2 \theta)^{1/2}]^2},$$

$$F_{ps}^{(2)} = \frac{\cos \theta_0 \cos^2 \theta \sin^2 \varphi}{(\varepsilon - \sin^2 \theta_0)} \quad (A2.2)$$

$$\times \frac{1}{[\cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2 [\varepsilon \cos \theta + (\varepsilon - \sin^2 \theta)^{1/2}]^2},$$

$$F_{pp}^{(2)} = \frac{\cos \theta_0 \cos^2 \theta}{[\varepsilon \cos \theta_0 + (\varepsilon - \sin^2 \theta_0)^{1/2}]^2}$$

$$\times \frac{[\cos \varphi \sin \theta_0 (\varepsilon - \sin^2 \theta)^{1/2} - \sin \theta (\varepsilon - \sin^2 \theta_0)^{1/2}]^2}{[\varepsilon \cos \theta + (\varepsilon - \sin^2 \theta)^{1/2}]^2}.$$

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