X-ray reflection and transmission by rough surfaces

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Expressions are given for the coherent (specular) and incoherent (diffuse) reflection and transmission of x rays by rough surfaces. In particular, the results from the distorted-wave Born approximation are critically compared with those obtained by us using the Rayleigh method. It is shown that the validity of the various expressions depends on the values of the perpendicular wave vector, the root-mean-square surface roughness, and the lateral correlation length of the roughness. The conservation of intensity in the various approximations is considered.

I. INTRODUCTION

X-ray reflectometry and related techniques form a powerful tool for the investigation of samples with rough interfaces.[1] Specular reflectivity measurements can yield the magnitude of the average roughness perpendicular to an interface, and diffuse scattering experiments give information about the lateral extent of the roughness.[2,3] Moreover, the compositional depth profile can be measured. Whereas specular reflectivity provides the density profile of a sample, the elemental depth profile can be found from angle-dependent x-ray fluorescence (AD-XRF) experiments,[4] where the amount of adsorbed x rays (i.e., transmitted by the surface) is measured.

If a well-collimated x-ray beam is incident at a glancing angle on a rough sample, coherent scattering gives rise to a specularly reflected beam and a single transmitted, refracted beam, whereas incoherent scattering causes diffusely scattered reflected and transmitted radiation. Several theories exist to describe the influence of roughness on x-ray scattering.[2,3,5] One of the factors determining the applicability of a theory is the lateral correlation length of the roughness.[5,6] In this paper we will critically review existing theories relating to coherent and incoherent reflection and transmission, and will consider the conservation of intensity in the various cases. We will show that the existing theory for incoherent scattering may fail in the case of large correlation lengths, and we will propose an improved description for that case.

In all theories discussed below the sample is characterized by its (complex) refractive index, and its atomic structure is neglected, an assumption which is justified if the inverse wave-vector transfer is much larger than the interatomic distances. For simplicity's sake, the sample surface is assumed to have a Gaussian random roughness, i.e., the height deviation has a normal distribution. We will explicitly give results for the case of s-polarized radiation, although they are also valid for p polarization in very good approximation (cf. Ref. 4). Most of the conclusions are expected to be valid for neutron scattering as well.

II. COHERENT SCATTERING AT SMALL CORRELATION LENGTHS

Névot and Croce[5] used the reciprocity principle[7] to derive expressions for the specular reflectivity by starting with the solution for a smooth surface and calculating the change in the electric field due to the roughness. Considering a plane wave with wave vector $\mathbf{k}$ incident on a sample with refractive index n and a rough top surface with root-mean-square (rms) surface roughness σ, a self-consistent solution for the reflection coefficient is

$$\tilde{r}_k = r_k \exp(-2k_0 k_1 \sigma^2) , \qquad (1)$$

where $r_k = (k_0 - k_1)/(k_0 + k_1)$ is the reflection coefficient at the smooth surface, k_0 is the perpendicular component of the wave vector of the incident wave, and k_1 is the perpendicular component of the wave vector of the refracted wave. Defining the critical wave vector k_c according to $k_c^2 = |\mathbf{k}|^2(1-n^2)$, one can write $k_1 = (k_0^2 - k_c^2)^{1/2}$. In the derivation the approximation is made that just above the surface one can use the analytical continuation of the electric field just below the surface. This is a good approximation for small roughness ($k_0\sigma \ll 1$) and also if $k_0 \gg k_c$, where $k_1 \simeq k_0$.

Névot and Croce argued that their method is valid in the case of a roughness profile with a predominance of high spatial frequencies,[5] or, in our terminology, with a small correlation length. It can be shown that, if the correlation length is ξ, the above method is correct if $\xi k_0^2/|\mathbf{k}| \ll 1$.[8]

Many other authors[9–11] follow the same scheme. That is, they start with the solution for the smooth surface and make the approximations of small correlation length as well as small roughness or large k_0.

Vidal and Vincent[9] used Green's theorem to find a relation between the electric fields in the cases of smooth and rough interfaces in a multilayer. Their result implies that Eq. (1) is valid for the reflection coefficient, whereas the transmission coefficient is given by

$$\tilde{t}_k = t_k \exp[(k_0 - k_1)^2 \sigma^2 / 2] , \qquad (2)$$

where $t_k=2k_0/(k_0+k_1)$ is the transmission coefficient for a smooth surface. Expressions like Eqs. (1) and (2) apply for each interface in a mulitlayer.[9]

Sinha *et al.*[3] used the distorted-wave Born approximation (DWBA) to calculate the scattering. In this approach, the deviation of the refractive index from that of a smooth surface is considered as a perturbation. In this way, a formula is found for the reflection coefficient which up to $O(k_0^2\sigma^2)$ is the same as Eq. (1). With the same method and up to the same order, we found Eq. (2) for the transmission coefficient.[6] Pynn[10] also found that in the DWBA Eq. (1) is a self-consistent solution.[12]

Caticha[11] used a Green's-function method, equivalent to the Névot-Croce[5] formalism. He found that different results may be obtained, depending on which side of the surface the analytical continuation of the electric field is chosen. There are two possible methods which give the same result [Eq. (1)] for the reflection coefficient, whereas one of the methods gives Eq. (2) for the transmission coefficient and the other one yields

$$\tilde{t}_k'=t_k\exp[-(k_0-k_1)^2\sigma^2/2](1-\tilde{r}_k^2)/(1-r_k^2)\ .$$

Caticha suggests that the geometrical mean $(\tilde{t}_k\tilde{t}_k')^{1/2}$ be used. The expressions are the same up to $O(k_0^2\sigma^2)$ and for $k_0 \gg k_c$, but, as will be seen in Fig. 3, can differ in the neighborhood of the critical wave vector.

Other methods[13,14] also lead to the above results, at least up to $O(k_0^2\sigma^2)$. In conclusion, for this section, Eqs. (1) and (2) are expected to give a good description in the case of small correlation lengths of the roughness.

III. INCOHERENT SCATTERING AT SMALL CORRELATION LENGTHS

A formula for the differential cross section for diffuse reflection, which is applicable for small roughness ($k_0\sigma \ll 1$), is[2,15,16]

$$\frac{d\sigma(k_0\to p_0)}{d\Omega}=AP(k_0\to p_0)\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\ , \qquad (3)$$

where the incident wave vector is $\mathbf{k}=(\mathbf{k}_\parallel,k_0)$, the scattered wave vector is $\mathbf{p}=(\mathbf{p}_\parallel,p_0)$, A is the (irradiated and detected) sample area, and $\tilde{C}(\mathbf{q}_\parallel)$ is the power spectral density of the surface roughness, i.e., the two-dimensional Fourier transform of the correlation function of the roughness profile (Appendix A). The prefactor is

$$P(k_0\to p_0)=|k_c|^4|t_k|^2|t_p|^2/(16\pi^2)\ . \qquad (4)$$

For the case of larger roughness, Sinha *et al.*,[3] with the use of DWBA, derived

$$\frac{d\sigma(k_0\to p_0)}{d\Omega}=AP(k_0\to p_0)S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_1+p_1)\ , \qquad (5)$$

where

$$S(\mathbf{q}_\parallel,q)=\left|\frac{\exp(-q^2\sigma^2/2)}{q}\right|^2 \times\int d^2X\exp(i\mathbf{q}_\parallel\cdot\mathbf{X}) \times\{\exp[|q|^2C(\mathbf{X})]-1\}\ . \qquad (6)$$

With the same method [using the results from Ref. 6 and Appendix B, Eq. (B7)], for the differential cross section for diffuse transmission we find

$$\frac{d\sigma(k_0\to p_1)}{d\Omega}=AP(k_0\to p_1)S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_1-p_0)\ , \qquad (7)$$

with

$$P(k_0\to p_1)=|k_c|^4|t_k|^2|t_{\bar{p}}|^2(p_1'^2/|p_1|^2)/(16\pi^2)\ , \qquad (8)$$

where the scattered wave vector is $(\mathbf{p}_\parallel,p_1)$, $t_{\bar{p}}=2p_1/(p_1+p_0)$ and $p_1'=\mathrm{Re}(p_1)$.

There has been some discussion of whether the transmission coefficients in Eqs. (4) and (8) should not be substituted by those of Eq. (2).[10,17] Considering all arguments,[18] we conclude that the use of Eqs. (4) and (8) should be preferred, with the effect of the roughness included in the factor $|\exp(-q^2\sigma^2/2)|^2$ of Eq. (6).

A problem with Eq. (5) is that it can yield a total scattered intensity which exceeds the incident intensity. The integrated incoherently reflected intensity divided by the incident intensity is (cf. Appendix B)

$$R_i=\frac{1}{Ak_0}\int_{|\mathbf{p}_\parallel|<|\mathbf{k}|}\frac{d^2p_\parallel}{p_0}\frac{d\sigma(k_0\to p_0)}{d\Omega}\ . \qquad (9)$$

For instance, consider the case in which the scattered intensity distribution is so narrow that we can approximate $p_0\simeq k_0$. Then we find

$$R_i=|r_k|^2(k_0^2/|k_1|^2)\{\exp(8k_1''^2\sigma^2) -\exp[-4(k_1'^2-k_1''^2)\sigma^2]\}\ ,$$

where $k_1'=\mathrm{Re}(k_1)$ and $k_1''=-\mathrm{Im}(k_1)$. If σ is large, this can easily exceed 1 in the neighborhood of the critical wave vector (cf. Fig. 1). In Sec. VI we will derive formulas which fulfill the requirement of intensity conservation

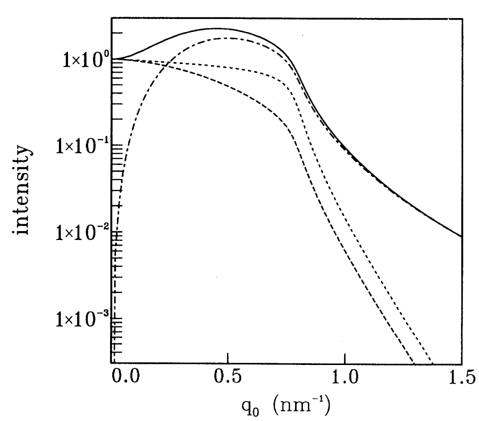

FIG. 1. Coherent and incoherent reflectivity vs wave-vector transfer, calculated in the DWBA for Cu $K\alpha_1$ radiation on gold with rms roughness $\sigma=1.5$ nm and large correlation length ξ. Long dashes: coherent reflectivity $|\tilde{r}_k|^2$ [Eq. (12)]; short dashes: only the first-order contribution to the coherent reflectivity $|\tilde{r}_k|^2$ [Eq. (1)]; dash-dotted line: integrated incoherent reflection [Eq. (5)]; solid line: sum of coherent and incoherent reflection. All intensities are relative to the incident intensity.

and do not suffer from this problem.

The formulas of Sec. VI are valid in the case of large ξ. Indeed, Eq. (5) only gives too large an intensity if both σ and ξ are large. Admittedly in the derivation it was not explicitly stated that the correlation length is small. However, it was assumed that the electric fields are continuous at the average, smooth surface. Moreover, the DWBA assumes that the electric field can be written as a perturbation series (cf. Appendix C). Both approximations are expected to fail in the case that both ξ and σ are large.

Hence we expect Eqs. (5) and (7) to be valid for general σ if $\xi k_0^2/|\mathbf{k}| \lesssim 1$. In the limit of $\xi k_0^2/|\mathbf{k}| \ll 1$, it can be shown[8] that incoherent scattering can be neglected if compared to the coherent scattering. The fact that the incoherent scattering vanishes for small ξ can also be inferred by noting that $S(\mathbf{q}\|,q)$ is proportional to ξ^2, as is found by substituting a formula like Eq. (A1) into Eq. (6) for $C(\mathbf{X})$. Then it is found that the integrated incoherent scattering [Eq. (9)] can be neglected if $\xi k_0^2/|\mathbf{k}| \ll 1$.

IV. COHERENT SCATTERING AT ARBITRARY CORRELATION LENGTHS

Recently, we showed[6] that in principle the DWBA should be pursued up to second order to calculate the coherent scattering[19] up to $O(k_0^2\sigma^2)$. We found that the second-order contribution depends on the lateral correlation length ξ of the roughness. If $\xi k_0^2/|\mathbf{k}| \ll 1$, the second-order contribution can be neglected. Hence in that case the DWBA results discussed in the preceding sections remain correct.

For arbitrary ξ, the results can be written as[6]

$$\tilde{r}_k = r_k \exp\left[-2k_0k_1\sigma^2 - \frac{1}{2\pi^2}k_0k_c^2\int_{|\mathbf{p}_\parallel|<|\mathbf{k}|}\frac{d^2p_\parallel}{p_0+p_1}\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\right], \quad (10)$$

$$\tilde{t}_k = t_k \exp\left[\tfrac{1}{2}(k_0-k_1)^2\sigma^2 - \frac{1}{4\pi^2}(k_0-k_1)k_c^2 \times \int_{|\mathbf{p}_\parallel|<|\mathbf{k}|}\frac{d^2p_\parallel}{p_0+p_1}\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\right]. \quad (11)$$

In the case that $\xi k_0^2/|\mathbf{k}| \gg 1$, the integrals can be performed analytically and Eqs. (10) and (11) become

$$\tilde{r}_k = r_k \exp(-2k_0^2\sigma^2), \quad (12)$$

$$\tilde{t}_k = t_k \exp[-(k_0-k_1)^2\sigma^2/2]. \quad (13)$$

In Eq. (12), r_k is multiplied by the well-known so-called Debye-Waller or Rayleigh factor.[20] Equations (12) and (13) can also be obtained by averaging the phase factors of the reflection and transmission coefficients obtained for normally distributed surface heights.[5,6] In Sec. VI we will discuss a more thorough derivation of these equations.

V. INTENSITY CONSERVATION AT SMALL CORRELATION LENGTHS

In a scattering process we expect that the incident intensity (expressed as the number of photons per second) is equal to the sum of reflected and transmitted intensities. This equality is a manifestation of the conservation of (energy) flux through a plane parallel to the average smooth sample surface. The methods to calculate scattering from rough surfaces involve approximations. Then the sum of all scattering contributions is not necessarily equal to the incident intensity. However, the degree of intensity conservation can give a clue to the applicability of a theory.

The conservation of intensity can be checked by calculating explicitly the reflected and transmitted intensities. We will assume that an x-ray beam of unit intensity impinges on the sample. Without roughness, the reflected intensity is $R_0=|r_k|^2$, the transmitted intensity is $T_0=|t_k|^2k_1'/k_0$, and it is easily checked that $1-R_0-T_0=0$.

Next we will consider the case of scattering at rough interfaces if there is no absorption. If $k_0<k_c$, we find that, since $k_1'=0$, the transmitted intensity $T=|\tilde{t}_k|^2k_1'/k_0=0$. As was indicated above,[8] incoherent scattering can be neglected in the case of small correlation lengths. From Eq. (1) we see that in that case the coherently reflected intensity $R=1$ for $k_0<k_c$. That is, the reflectively is not affected by the roughness if the transmissivity is zero and the correlation length is small (a conclusion already made by Rayleigh for the reflection of sound[21]).

We conclude that for $k_0<k_c$ the formulas guarantee intensity conservation if there is no absorption. For $k_0>k_c$ this is not necessarily true. It is easily checked that the intensity is conserved if we follow Caticha's[11] suggestion and use $(\tilde{t}_k\tilde{t}_k')^{1/2}$ for the transmission coefficient in the case that there is no absorption.[22] If Eq. (2) is used, the intensity is only conserved up to $O(k_0^2\sigma^2)$.

In the case with absorption it is convenient to calculate the absorbed intensity explicitly. In that way, the deviation of the position of the rough surface from that of the average surface is taken into account correctly. This will give rise to a contribution from absorption in the rough interfacial layer (cf. Appendix C). Elementary considerations show that, if an x-ray beam of unit intensity strikes a sample with surface area A, the absorbed intensity is given by

$$T=\frac{2k_1'k_1''}{Ak_0}\int d^3r|\phi_{\mathbf{k}}(\mathbf{r})|^2, \quad (14)$$

where $\phi_{\mathbf{k}}(\mathbf{r})$ is the electric field due to the x rays at position $\mathbf{r}$ in the sample. We note that the absorption can be measured, e.g., in an AD-XRF experiment.[4,23]

In Appendix C the absorbed intensity in an infinitely thick sample is calculated using the DWBA. It is found that, if $\xi k_0^2/|\mathbf{k}| \ll 1$, the significant contributions to T are the absorption of the coherently transmitted beam $T_0+T_1 \simeq T_0|\exp[(k_0-k_1)^2/2]|^2$ and the absorption in the rough interfacial layer $T_4=T_0[\exp(2k_1''^2\sigma^2)-1]$.

The incoherent scattering can be neglected and, according to Eq. (1), the coherently reflected intensity is $R=R_0|\exp(-2k_0k_1\sigma^2)|^2$. With this we find $1-R-T=O(|k_0-k_1|^4\sigma^4)$. That is, there is no strict conservation of intensity. This result is not improved by pursuing the calculations of Appendix C up to higher order. The reason seems to be that the electric field was approximated by its analytical continuation, which is only true up to $O(k_0^2\sigma^2)$ in the case of small roughness and up to $O(k_c^2/k_0^2)$ in the case of large k_0.

For larger correlation lengths we find in the DWBA (Sec. IV and Appendix C) that even this is no longer true, but that $1-R-T$ can be of $O(|\mathbf{k}|\sigma^2/\xi)$.

From Sec. IV and Appendix C it is readily checked that the intensity is conserved up to $O(k_0^2\sigma^2)$ again if the correlation length is so large that $\xi k_0^2/|\mathbf{k}|\gg 1$. If both ξ and σ are large, however, the intensity in the DWBA is not conserved (cf. Sec. III). In Sec. VI we will give formulas which guarantee intensity conservation at large ξ and arbitrary σ.

VI. SCATTERING AT LARGE CORRELATION LENGTHS

In Appendix D we calculate the coherent and incoherent scattering for the case of large correlation lengths. The calculation method was used originally by Rayleigh[21] to describe the reflection of sound at a corrugated surface. For coherent scattering of x rays the method has already been used by Croce, Névot, and Pardo.[24] In that case, Eqs. (10) and (11) are obtained.

In the case of incoherent reflection, the result is

$$\frac{d\sigma(k_0\to p_0)}{d\Omega}=AP(k_0\to p_0)S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_0+p_0)\ , \tag{15}$$

with the prefactor given by Eq. (4), whereas in the case of incoherent transmission Eq. (7) is obtained.

Hence the expression for incoherent transmission obtained with the Rayleigh method is the same as in the DWBA. However, the expression for reflection differs in that Eq. (15) has $q=k_0+p_0$, whereas Eq. (5) has $q=k_1+p_1$ in the argument of $S(\mathbf{q}_\parallel,q)$. The integrated incoherently reflected intensity obtained from Eq. (15) is $R_i=|r_k|^2[1-\exp(-4k_0^2\sigma^2)]$ (see Sec. VII). This never exceeds 1, in contrast to the intensity obtained in the DWBA (Sec. III). Note that both Eqs. (5) and (15) reduce to Eq. (3) in the case of small roughness.

It is interesting to compare the above results with those of the Born approximation (BA),[3] which is correct if $k_0^2\gg k_c^2$. In the BA the starting point is a simple incident plane wave, and the whole sample is considered as a perturbation. In the case of coherent scattering, this yields Eq. (12) for the reflection coefficient [with $r_k\simeq k_c^2/(4k_0^2)$], whereas it gives $\tilde{t}_k\simeq t_k\simeq 1$. For the incoherent reflection and transmission the results are

$$\frac{d\sigma(k_0\to p_0)}{d\Omega}=A\frac{|k_c|^4}{16\pi^2}S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_0+p_0)\ ,$$

$$\frac{d\sigma(k_0\to p_1)}{d\Omega}=A\frac{|k_c|^4}{16\pi^2}S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_0-p_0)\ .$$

Note that in the case of reflection the roughness dependence is the same in the BA as obtained with the Rayleigh method for large correlation lengths.

VII. INTENSITY CONSERVATION AT LARGE CORRELATION LENGTHS

To check the conservation of intensity for the Rayleigh method, we will calculate the total reflection and absorption, just like in Sec. V.

The coherently reflected intensity is, according to Eq. (12), $R_0\exp(-4k_0^2\sigma^2)$. The total incoherent reflection is found from Eqs. (9) and (15), setting $p_0\simeq k_0$: $R_i=R_0[1-\exp(-4k_0^2\sigma^2)]$. The sum of the two is $R=R_0$, just as if no roughness were present (cf. Fig. 2).

Analogously to the calculation performed in Appendix C, we can calculate the various contributions to the absorption in an infinitely thick sample. Setting $p_0\simeq k_0$, we find the following. For the absorption of the coherently transmitted beam: $T_0|\exp[-(k_0-k_1)^2\sigma^2/2]|^2$. For the absorption of the incoherently transmitted beam: $T_0|\exp[-(k_0-k_1)^2\sigma^2/2]|^2[\exp(|k_0-k_1|^2\sigma^2)-1]$. For the correction due to absorption in the rough interfacial layer: $T_0[1-\exp(2k_1''^2\sigma^2)]$. The sum of the three is just T_0, again as if no roughness were present (cf. Fig. 4). In the DWBA (Appendix C) the three contributions are $T_0+T_1+T_2$, T_3, and T_4+T_5, respectively, which are the same up to $O(k_0^2\sigma^2)$.

The conclusion is that for the Rayleigh method with $\xi k_0^2/|\mathbf{k}|\gg 1$ the intensity is conserved.

VIII. EXAMPLES

To compare the result obtained with the various methods, in Figs. 1–6 we show calculations for Cu $K\alpha_1$ radiation ($|\mathbf{k}|=40.784\ \mathrm{nm}^{-1}$) incident on a gold sample with $\sigma=1.5$ nm.

Figure 1 shows the reflected intensity as a function of

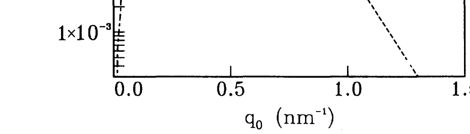

FIG. 2. Coherent and incoherent reflectivity vs wave-vector transfer, calculated with the Rayleigh method for Cu $K\alpha_1$ radiation on gold with rms roughness $\sigma=1.5$ nm and large correlation length ξ. Long dashes: coherent reflectivity $|\tilde{r}_k|^2$ [Eq. (12)]; dash-doted line: integrated incoherent reflection [Eq. (15)]; solid line: sum of coherent and incoherent reflection (or $|r_k|^2,\sigma=0$).

wave-vector transfer $q_0=2k_0$ calculated in the DWBA for the case that $\xi k_0^2/|\mathbf{k}|\gg 1$. In the first-order DWBA, the effect of ξ on the coherent reflectivity is not taken into account and the result, Eq. (1), is given by the short-dashed line. The second-order result for the coherent reflectivity [Eq. (12)], given by the long-dashed line, is smaller. The integrated incoherently reflected intensity, obtained from Eqs. (5) and (9), is also shown (dash-dotted line) as well as the sum of the latter two (solid line). Note that the reflected intensity exceeds the incident intensity. It can be concluded that this approximation fails if both σ and ξ are large.

Figure 2 shows the same, but now obtained with the Rayleigh method. The coherent reflectivity (dashed) is again given by Eq. (12). The integrated incoherently reflected intensity (dash-dotted line), obtained from Eqs. (9) and (15), does not exceed the incident intensity. The sum of the two (solid line) is the same as the reflectivity R_0 without roughness.

Figures 3 and 4 show $|\tilde{t}_k|^2$ and the other contributions to the so-called transmission factor Tk_0/k_1'. By plotting the transmission factor instead of the transmissivity T, the magnitude of the various contributions is shown more clearly.[25]

Figure 3 shows results for the case that $\xi k_0^2/|\mathbf{k}|\ll 1$. The long-dashed line shows the coherent transmission obtained from Eq. (2) (essentially equal to T_0+T_1 from Appendix C). For comparison, the result obtained with the formula $|\tilde{t}_k\tilde{t}_k'|$, suggested by Caticha,[11] is shown by the dash-dotted line. The difference between the two is only small. The short-dashed line shows the contribution due to absorption in the rough interfacial layer, T_4 from Appendix C. This is seen to be only appreciable for $k_0<k_c$. The solid line shows the total transmission, equal to the sum of the long-dashed and short-dashed contributions.

Figure 4 shows the various contributions for the case that $\xi k_0^2/|\mathbf{k}|\gg 1$. The coherent transmission is given by the long-dashed line. In this case, the correction due to absorption in the rough interfacial layer (short dashes) is negative. The dash-dotted line shows the integrated incoherent transmission. The sum of the three contributions (solid line) is just the value $|t_k|^2$ without roughness.

As an example of reflected scattering distributions, Figs. 5 and 6 show the calculated reflected intensity around $k_0=0.3$ nm^{-1} for the case that entrance and receiving slits are used which have a divergence of 10^{-4} rad in the scattering plane and are wide open in the perpendicular direction. Figure 5 shows the intensity as a function of parallel wave-vector transfer $q_\parallel=p_\parallel-k_\parallel$ in transverse scans (in good approximation equal to rocking scans) at $k_0+p_0=0.6$ nm^{-1}. Figure 6 shows detector scans at $k_0=0.3$ nm^{-1}, where the intensity is plotted as a function of perpendicular wave-vector transfer $q_0=k_0+p_0$. In both figures the dashed and dash-dotted

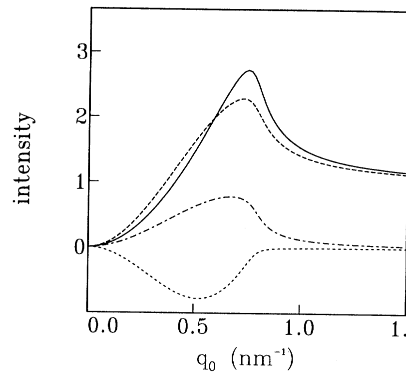

FIG. 4. Coherent and incoherent transmission factor Tk_0/k_1' vs wave-vector transfer, calculated with the Rayleigh method for Cu $K\alpha_1$ radiation in gold with rms roughness $\sigma=1.5$ nm and large correlation length ξ. Long dashes: coherent transmission $|\tilde{t}_k|^2$ [Eq. (13)]; dash-dotted line: incoherent transmission [Eq. (7)]; short dashes: correction for absorption in rough interfacial layer; solid line: total transmission (or $|t_k|^2,\sigma=0$).

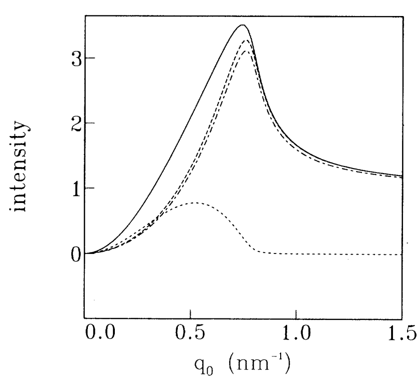

FIG. 3. Coherent and incoherent transmission factor Tk_0/k_1' vs wave-vector transfer, calculated in the DWBA for Cu $K\alpha_1$ radiation on gold with rms roughness $\sigma=1.5$ nm and small correlation length ξ. Long dashes: coherent transmission $|\tilde{t}_k|^2$ [Eq. (2)]; dash-dotted line: coherent transmission according to Caticha (Ref. 11) (see text); short dashes: absorption in rough interfacial layer; sold line: total transmission.

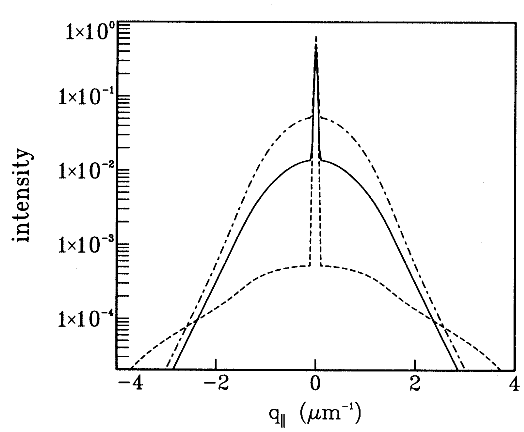

FIG. 5. Coherent and incoherent reflectivity vs parallel wave-vector transfer in a transverse scan at $q_0=0.6$ nm^{-1}, calculated for Cu $K\alpha_1$ radiation on gold with rms roughness $\sigma=1.5$ nm and two different correlation lengths ξ. Dashed line: $\xi=20$ nm (DWBA); dash-dotted line: $\xi=2$ μm (DWBA); solid line: $\xi=2$ μm (Rayleigh method).

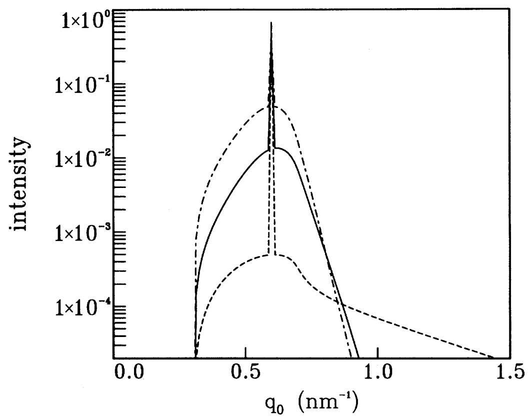

FIG. 6. Coherent and incoherent reflectivity vs perpendicular wave-vector transfer in a detector scan at $k_0=0.3\ \mathrm{nm}^{-1}$, calculated for Cu $K\alpha_1$ radiation on gold with rms roughness $\sigma=1.5$ nm and two different correlation lengths ξ. Dashed line: $\xi=20$ nm (DWBA); dash-dotted line: $\xi=2\ \mu$m (DWBA); solid line: $\xi=2\ \mu$m (Rayleigh method).

lines show the results of calculations in the DWBA for $\xi=20$ nm and 2 μm, respectively, whereas the solid line is obtained using the Rayleigh method for $\xi=2\ \mu$m. It is seen that the incoherent scattering is small for small ξ and large for large ξ. The integrated incoherent scattering R_i can be obtained by numerical integration of a detector scan. From Fig. 6 we find $R_i=0.025$ in the case that $\xi=20$ nm. In the case that $\xi=2\ \mu$m (where $\xi k_0^2/|\mathbf{k}|=4.4$), we find $R_i=1.29$ for the DWBA and $R_i=0.35$ for the Rayleigh method. This can be compared with the numbers for $\xi k_0^2/|\mathbf{k}|\gg 1$ obtained from Figs. 1 and 2: $R_i=1.59$ and 0.44, respectively. Also here we see that in the DWBA the calculated scattered intensity exceeds the incident intensity.

IX. CONCLUSION

For small lateral correlation lengths of the roughness, $\xi k_0^2/|\mathbf{k}|\ll 1$, the existing expressions for the coherent scattering, Eqs. (1) and (2), are expected to give a good description at small $k_0\sigma$ and for $k_0\gg k_c$ and they interpolate smoothly between the two regimes. For the transmission coefficient other interpolations are possible, but the difference is not dramatic (cf. Fig. 3). However, in some cases it is important to take into account the absorption in the rough interfacial layer. The incoherent scattering can be obtained from Eqs. (5) and (7).

For large correlation lengths $\xi k_0^2/|\mathbf{k}|\gg 1$, expressions (7), (12), (13), and (15), obtained with the Rayleigh method, are expected to give a good description.

For intermediate values of ξ, one can use Eqs. (10) and (11) for the coherent scattering. We propose to neglect the correction for absorption in the rough interfacial layer, since in general it is a relatively small correction, which changes sign in going from small to large ξ. For the incoherent transmission we expect that Eq. (7), which is valid at both small and large ξ, can be used. A problem is what to use for incoherent reflection. We would like to have a way to interpolate between the small-ξ behavior, Eq. (5), and the large-ξ behavior, Eq. (15). A possibility, suggested by Eq. (10), is to substitute k_1 in Eq. (5) by

$$k_1+\frac{1}{4\pi^2\sigma^2}k_c^2\int_{|\mathbf{p}_\parallel|<|\mathbf{k}|}\frac{d^2p_\parallel}{p_0+p_1}\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\ ,$$

and analogously for p_1. Comparison with experiments has to show whether this is a sensible approach.

Note added in proof. G. Palasantzas [Phys. Rev. B **48**, 14 472 (1993)] recently proposed a correlation function very similar to Eq. (A2).

APPENDIX A: THE CORRELATION FUNCTION AND ITS POWER SPECTRAL DENSITY

The correlation function of the roughness profile of a rough surface is defined as

$$C(\mathbf{x}-\mathbf{x}')=\langle z(\mathbf{x})z(\mathbf{x}')\rangle\ ,$$

where $\langle\ \rangle$ indicates a configurational average and $z(\mathbf{x})$ denotes the height deviation of the sample surface at position $\mathbf{x}=(x,y)$ with $\langle z(\mathbf{x})\rangle=0$. For a random surface, the correlation function only depends on $|\mathbf{x}-\mathbf{x}'|$. Its two-dimensional Fourier transform

$$\tilde{C}(\mathbf{q}_\parallel)=\int d^2X\exp(i\mathbf{q}_\parallel\cdot\mathbf{X})C(\mathbf{X})$$

is the power spectral density (PSD) of the surface roughness.

A functional form for $C(\mathbf{X})$ which is often used is[3]

$$C(\mathbf{X})=\sigma^2\exp[-(|\mathbf{X}|/\xi)^{2H}]\ , \tag{A1}$$

where ξ is the lateral correlation length of the roughness, and the parameter H describes how jagged the surface is ($0<H\le 1$). For $|\mathbf{X}|\ll\xi$ the surface is a self-affine fractal. Alternative forms with the same limiting behavior were discussed by Palasantzas and Krim.[26] Following Church and Takacs,[2] we recently proposed a different form,[27] which is more in agreement with fractal growth models:[28]

$$C(\mathbf{X})=P\xi^H|\mathbf{X}|^HK_H(|\mathbf{X}|/\xi)\ , \tag{A2}$$

where K_H denotes the modified Bessel function of order H, and P is a constant which is related to σ according to $\sigma^2=P\xi^{H-1}2^{H-1}\Gamma(1+H)/H$. For the PSD this yields the simple form

$$\tilde{C}(\mathbf{q}_\parallel)=4\pi H\sigma^2\xi^2(1+|\mathbf{q}_\parallel|^2\xi^2)^{-1-H}\ .$$

In all cases, $\tilde{C}(\mathbf{q}_\parallel)$ is a bell-shaped curve centered at $\mathbf{q}_\parallel=\mathbf{0}$, with a height proportional to $\sigma^2\xi^2$ and a width proportional to ξ^{-1}.

APPENDIX B: CALCULATION OF COHERENT AND INCOHERENT SCATTERING

We assume that a plane wave with wave vector $\mathbf{k}$ is incident on a sample with surface area A. The incident electric field at position $\mathbf{r}=(\mathbf{x},z)$ can be written as

$$\phi_k(\mathbf{r})=E_0^\downarrow(k)\exp[i(\mathbf{k}_\parallel\cdot\mathbf{x}+k_0z)]\ , \tag{B1}$$

the scattered reflected field as

$$\phi_k(\mathbf{r}) = \sum_p E_0^{\uparrow}(p)\exp[i(p_{\parallel}\cdot\mathbf{x} - p_0 z)] \ , \tag{B2}$$

and the scattered transmitted field as

$$\phi_k(\mathbf{r}) = \sum_p E_1^{\downarrow}(p)\exp[i(\mathbf{p}_{\parallel}\cdot\mathbf{x} + p_1 z)] \ , \tag{B3}$$

where the subscripts 0 and 1 denote the materials above and below the surface, respectively, and the superscript arrow denotes the direction of the perpendicular wave vector.

The total outgoing flux (averaged over the sample surface) is given by $A^{-1}\int d^2x|\phi_k(\mathbf{r})|^2$, which equals $\sum_p |E_0^{\uparrow}(p)|^2$ above the surface and $\sum_p |E_1^{\downarrow}(p)|^2$ below it. We will divide this by the incident flux $|E_0^{\downarrow}(k)|^2$. For a rough surface one has to take a configurational average. In the case of reflection the coherent part of the outgoing flux divided by the incident flux is $\sum_p |\langle E_0^{\uparrow}(p)/E_0^{\downarrow}(k)\rangle|^2$ and the incoherent part is $\sum_p[\langle |E_0^{\uparrow}(p)/E_0^{\downarrow}(k)|^2\rangle - |\langle E_0^{\uparrow}(p)/E_0^{\downarrow}(k)\rangle|^2]$ (cf. Ref. 20), whereas in the case of transmission $E_1^{\downarrow}(p)$ has to be substituted for $E_0^{\uparrow}(p)$. For the coherent case only the terms with $p=k$ will survive, if the amplitudes are given by expressions like Eqs. (D3) or (D4) [or the expression for $T^{(n)}(p,k)$ from Ref. 6]. Then the coefficients for coherent reflection and transmission are given by

$$\tilde{r}_k = \langle E_0^{\uparrow}(k)/E_0^{\downarrow}(k)\rangle \ , \tag{B4}$$

$$\tilde{t}_k = \langle E_1^{\downarrow}(k)/E_0^{\downarrow}(k)\rangle \ . \tag{B5}$$

The incoherent scattering is most conveniently expressed as a differential cross section, i.e., the outgoing intensity per unit solid angle divided by the incident flux. We will calculate this assuming that the sample is so large that we can substitute $\sum_p \simeq A/(4\pi^2)\int d^2p_{\parallel}$. In the case of reflection $\int d^2p_{\parallel} = |\mathbf{k}|\int d\Omega p_0$, in the case of transmission $\int d^2p_{\parallel} = |\mathbf{k}|\int d\Omega p_1'$, where $d\Omega$ is an element of solid angle. In the case of reflection, the average outgoing flux divided by the incident flux can be written as $A/(4\pi^2)|\mathbf{k}|\int d\Omega p_0[\langle |E_0^{\uparrow}(p)/E_0^{\downarrow}(k)|^2\rangle - |\langle E_0^{\uparrow}(p)/E_0^{\downarrow}(k)\rangle|^2]$. Multiplying by the cross section $Ap_0/|\mathbf{k}|$ of the outgoing reflected beam, for the differential cross section for incoherent reflection we obtain

$$\frac{d\sigma(k_0\to p_0)}{d\Omega} = \frac{A^2}{4\pi^2}p_0^2\left[\left\langle\left|\frac{E_0^{\uparrow}(p)}{E_0^{\downarrow}(k)}\right|^2\right\rangle - \left|\left\langle\frac{E_0^{\uparrow}(p)}{E_0^{\downarrow}(k)}\right\rangle\right|^2\right] . \tag{B6}$$

In an analogous way, for the differential cross section for incoherent transmission we find

$$\frac{d\sigma(k_0\to p_1)}{d\Omega} = \frac{A^2}{4\pi^2}p_1'^2\left[\left\langle\left|\frac{E_1^{\downarrow}(p)}{E_0^{\downarrow}(k)}\right|^2\right\rangle - \left|\left\langle\frac{E_1^{\downarrow}(p)}{E_0^{\downarrow}(k)}\right\rangle\right|^2\right] . \tag{B7}$$

If an x-ray beam of unit intensity impinges on the sample, the scattered intensity per unit solid angle is obtained by dividing Eqs. (B6) and (B7) by $Ak_0/|\mathbf{k}|$. The total scattered intensity is calculated by integrating over $d\Omega$ (expressed above in $d^2p_{\parallel}$), yielding Eq. (9).

APPENDIX C: ABSORPTION IN THE DWBA

In this Appendix we calculate the x-ray absorption in an infinitely thick sample with a rough surface using the second-order DWBA. Up to second order, the electric field in the samples is given by[6]

$$\phi_{\mathbf{k}}(\mathbf{r}) = \phi_{\mathbf{k}}^{(0)}(\mathbf{r}) + \phi_{\mathbf{k}}^{(1)}(\mathbf{r}) + \phi_{\mathbf{k}}^{(2)}(\mathbf{r}) \ ,$$

where $\phi_{\mathbf{k}}^{(0)}(r) = t_k\exp[i(k_{\parallel}\cdot\mathbf{x} + k_1 z)]$, and $\phi_k^{(n)}(\mathbf{r})$ for $n>0$ is given by Eq. (2) of Ref. 6:

$$\phi_{\mathbf{k}}^{(n)}(\mathbf{r}) = \frac{i}{8\pi^2}\int_{|\mathbf{p}_{\parallel}|<|\mathbf{k}|}\frac{d^2p_{\parallel}}{p_1}T^{(n)}(\bar{p},k)\exp[i(\mathbf{p}_{\parallel}\cdot\mathbf{x} + p_1 z)] \ .$$

The T matrix $T^{(n)}(\bar{p},k)$ and its configurational average are given in Ref. 6, and will be substituted below.

The square of the electric field is

$$|\phi_{\mathbf{k}}(\mathbf{r})|^2 = |\phi_{\mathbf{k}}^{(0)}(\mathbf{r})|^2 + |\phi_{\mathbf{k}}^{(1)}(\mathbf{r})|^2 + |\phi_{\mathbf{k}}^{(2)}(\mathbf{r})|^2 + 2\,\mathrm{Re}[\phi_{\mathbf{k}}^{(0)*}(\mathbf{r})\phi_{\mathbf{k}}^{(1)}(\mathbf{r}) + \phi_{\mathbf{k}}^{(0)*}(\mathbf{r})\phi_{\mathbf{k}}^{(2)}(\mathbf{r}) + \phi_{\mathbf{k}}^{(1)*}(\mathbf{r})\phi_{\mathbf{k}}^{(2)}(\mathbf{r})] \ .$$

The adsorbed intensity, given by Eq. (14), can be split into two parts:

$$T = \frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{-\infty}^{0}dz\,|\phi_{\mathbf{k}}(\mathbf{r})|^2 + \frac{2k_1'k_1''}{Ak_0}\int d^2x\int_0^{z(\mathbf{x})}dz\,|\phi_k(\mathbf{r})|^2 \ .$$

In the case of Gaussian random roughness, the surface height $z(\mathbf{x})$ is normally distributed with standard deviation σ, and configurational averages can be taken. In principle, 12 terms are involved, but up to $O(k_0^2\sigma^2)$ only the following six terms are present (T_0 to T_5):

$$T_0 = \left\langle\frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{-\infty}^{0}dz\,|\phi_{\mathbf{k}}^{(0)}(\mathbf{r})|^2\right\rangle = \frac{k_1'}{k_0}|t_k|^2 \ ,$$

i.e., the transmitted intensity in the case of a smooth surface;

$$T_1 = \left\langle\frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{-\infty}^{0}dz\,2\,\mathrm{Re}[\phi_{\mathbf{k}}^{(0)*}(\mathbf{r})\phi_{\mathbf{k}}^{(1)}(\mathbf{r})]\right\rangle \simeq T_0\mathrm{Re}[(k_0-k_1)^2\sigma^2] \ ,$$

i.e., the change in transmission for small correlation lengths. Up to $O(k_0^2\sigma^2)$ we have $T_0 + T_1 = k_1'/k_0|\tilde{t}_k|^2$, with $\tilde{t}_k$ given by Eq. (2);

$$T_2=\left\langle \frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{-\infty}^{0}dz\,2\,\mathrm{Re}[\phi_{\mathbf{k}}^{(0)*}(\mathbf{r})\phi_{\mathbf{k}}^{(2)}(\mathbf{r})]\right\rangle$$

$$\simeq T_0\mathrm{Re}\left[-\frac{1}{2\pi^2}(k_0-k_1)k_c^2 \times\int_{|\mathbf{p}_\parallel|<|\mathbf{k}|}\frac{d^2p_\parallel}{p_0+p_1}\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\right] ,$$

i.e., the change in transmission for larger correlation lengths. Up to $O(k_0^2\sigma^2)$ we have $T_0+T_1+T_2=k_1'/k_0|\tilde{t}_k|^2$, with $\tilde{t}_k$ given by Eq. (11). If $\xi k_0^2/|\mathbf{k}|\ll 1$, $T_2\simeq 0$, whereas if $\xi k_0^2/|\mathbf{k}|\gg 1$, $T_2\simeq T_0\mathrm{Re}[-2(k_0-k_1)^2\sigma^2]$;

$$T_3=\left\langle \frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{-\infty}^{0}dz\,|\phi_{\mathbf{k}}^{(1)}(\mathbf{r})|^2\right\rangle$$

$$=T_0\frac{|k_c|^4}{4\pi^2k_1'}\int_{|\mathbf{p}_\parallel|<|\mathbf{k}|}\frac{p_1'd^2\mathbf{p}_\parallel}{|p_0+p_1|^2}\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel) ,$$

i.e., the absorbed diffusely scattered radiation. If $\xi k_0^2/|\mathbf{k}|\ll 1$, $T_3\simeq 0$, whereas if $\xi k_0^2/|\mathbf{k}|\gg 1$, $T_3\simeq T_0|k_0-k_1|^2\sigma^2$;

$$T_4=\left\langle \frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{0}^{z(\mathbf{x})}dz\,|\phi_{\mathbf{k}}^{(0)}(\mathbf{r})|^2\right\rangle$$

$$=T_0[\exp(2k_1''^2\sigma^2)-1]\simeq T_0 2k_1''^2\sigma^2 ,$$

i.e., the absorption in the rough interfacial layer if the transmitted beam were not affected by the roughness; and

$$T_5=\left\langle \frac{2k_1'k_1''}{Ak_0}\int d^2x\int_{0}^{z(\mathbf{x})}dz\,2\,\mathrm{Re}[\phi_{\mathbf{k}}^{(0)*}(\mathbf{r})\phi_{\mathbf{k}}^{(1)}(\mathbf{r})]\right\rangle$$

$$\simeq T_0\mathrm{Re}\left[\frac{ik_1''k_c^2}{\pi^2}\int_{|p_\parallel|<|\mathbf{k}|}\frac{d^2p_\parallel}{p_0+p_1}\tilde{C}(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\right] ,$$

i.e., the correction to T_4 due to the change in transmission caused by the roughness. If $\xi k_0^2/|\mathbf{k}|\ll 1$, $T_5\simeq 0$, whereas if $\xi k_0^2/|\mathbf{k}|\gg 1$, $T_5\simeq T_0(-4k_1''^2\sigma^2)$.

APPENDIX D: THE RAYLEIGH METHOD

Here we will use a method devised by Rayleigh[21] to calculate the scattering at a corrugated surface. We will assume that at each position $\mathbf{x}$ along the surface with height deviation $z(\mathbf{x})$, the electric field and its normal derivative are continuous. We will suppose that ξ is so large that the local surface slope can be neglected.

Writing the incident, reflected, and transmitted fields as Eqs. (B1), (B2), and (B3), the continuity of the field and its normal derivative at position $\mathbf{x}$ yield

$$E_0^{\downarrow}(k)\exp\{i[\mathbf{k}_\parallel\cdot\mathbf{x}+k_0z(\mathbf{x})]\} + \sum_p E_0^{\uparrow}(p)\exp\{i[p_\parallel\cdot\mathbf{x}-p_0z(\mathbf{x})]\} = \sum_p E_1^{\downarrow}(p)\exp\{i[\mathbf{p}_\parallel\cdot\mathbf{x}+p_1z(\mathbf{x})]\} , \tag{D1}$$

$$k_0E_0^{\downarrow}(k)\exp\{i[\mathbf{k}_\parallel\cdot\mathbf{x}+k_0z(\mathbf{x})]\} - \sum_p p_0E_0^{\uparrow}(p)\exp\{i[\mathbf{p}_\parallel\cdot\mathbf{x}-p_0z(\mathbf{x})]\} = \sum_p p_1E_1^{\downarrow}(p)\exp\{i[\mathbf{p}_\parallel\cdot\mathbf{x}+p_1z(\mathbf{x})]\} . \tag{D2}$$

These form a series of coupled equations which cannot be solved exactly for the general case. However, if ξ is large, we can make some further approximations.

We can consider ξ as a measure of the largest period present in the roughness profile. If radiation is diffracted from a grating with period ξ, first-order diffraction peaks occur at wave vectors $\mathbf{p}$ with $|\mathbf{p}_\parallel|=|\mathbf{k}_\parallel|\pm 2\pi/\xi$, from which we find $p_0\simeq k_0[1\pm 4\pi|\mathbf{k}|/(\xi k_0^2)]^{1/2}$ and an analogous expression for p_1. We anticipate that higher-order diffraction peaks will only have a small intensity. Indeed, the final result, Eq. (15), will correspond to a single narrow peak for large ξ.

Then, if we make the approximations $p_0\simeq k_0$ and $p_1\simeq k_1$, Eqs. (D1) and (D2) remain correct up to $O[|\mathbf{k}|/(\xi k_0^2)]$. Multiplying Eq. (D1) by k_1 and subtracting Eq. (D2), we obtain

$$\sum_p E_0^{\uparrow}(p)\exp[i(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\cdot x]=r_kE_0^{\downarrow}(k)\exp(2ik_0z(\mathbf{x})) .$$

Since this can be considered as a Fourier series, it follows that

$$\frac{E_0^{\uparrow}(p)}{E_0^{\downarrow}(k)}=\frac{r_k}{A}\int d^2x\,\exp[-i(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\cdot\mathbf{x}]\exp(2ik_0z(\mathbf{x})) \tag{D3}$$

In the same way, after multiplying Eq. (D1) by k_0, adding Eq. (D2), and Fourier transformation, we obtain

$$\frac{E_1^{\downarrow}(p)}{E_0^{\downarrow}(k)}=\frac{t_k}{A}\int d^2x\,\exp[-i(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\cdot\mathbf{x}] \times\exp[i(k_0-k_1)z(\mathbf{x})] . \tag{D4}$$

Now we can use the formulas of Appendix B to calculate the coherent and incoherent scattering. Using Eqs. (B4) and (B5), we obtain Eqs. (12) and (13) for the coherent scattering.

Using Eqs. (B6) and (B7), for the incoherent reflection and transmission we find

$$\frac{d\sigma(k_0\rightarrow p_0)}{d\Omega}=\frac{A}{\pi^2}|k_c|^4\frac{k_0^2p_0^2}{|k_0+k_1|^4}S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,2k_0)\ ,$$

$$\frac{d\sigma(k_0\rightarrow p_1)}{d\Omega}=\frac{A}{\pi^2}|k_c|^4\frac{k_0^2p_1'^2}{|k_0+k_1|^4}S(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_1-k_0)\ ,$$

with $S(\mathbf{q}_\parallel,q)$ given by Eq. (6). At first sight, these equations do not fulfill the reciprocity principle,[7,20] i.e., symmetry in the interchange of $\mathbf{p}$ and $\mathbf{k}$. However, since we assumed that $p_0\simeq k_0$ and $p_1\simeq k_1$, the scattering cross sections are given with the same accuracy, i.e., up to $O[|\mathbf{k}|/(\xi k_0^2)]$, by the symmetric formulas Eqs. (15) and (7).

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