Influence of the roughness profile on the specular reflectivity of x rays and neutrons

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For the calculation of specular x-ray and neutron reflectivity from rough surfaces two expressions exist, which are valid under different circumstances. Another expression is derived using the second-order distorted-wave Born approximation, which smoothly connects the two existing expressions. The shape of the reflectivity curve depends not only on the average roughness, but also on the lateral correlation of the roughness profile.

Specular x-ray and neutron reflectivity measurements of materials form a well-known technique for determining density profiles perpendicular to the surface.[1] For a rough surface the reflectivity depends, among other things, on the root-mean-square surface roughness σ. If compared with a smooth surface, the reflectivity is reduced and the factor by which the reflection coefficient is multiplied decreases with σ.[2-5] For this factor two forms are in use, which may give somewhat different results. The first form is the so-called static Debye-Waller (DW) factor[2,6] $\exp(-2k_0^2\sigma^2)$, where k_0 is the perpendicular component of the wave vector of the incident radiation. The second from, due to Névot and Croce (NC),[4,7] is $\exp(-2k_0k_1\sigma^2)$, where k_1 is the perpendicular component of the wave vector inside the material, after refraction. The difference between the two will be seen in Fig. 1, where in the upper curve the NC factor is used and in the lower curve the DW factor. Generally, the NC form is applied in the vicinity of the region of total reflection, whereas at large k_0 the DW form may give a better description.

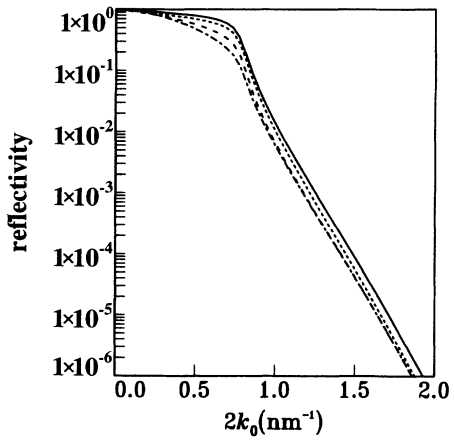

FIG. 1. Calculated specular reflectivity vs wave-vector transfer for Cu $K\alpha_1$ radiation on a gold sample with average roughness $\sigma=1.5$ nm and four different values of the lateral correlation length ξ. From top to bottom: $\xi=0$ (NC form, solid line); $\xi=100$ nm (dotted); $\xi=1000$ nm (dashed); and $\xi=\infty$ (DW form, dash-dotted).

In this paper we will present a theoretical description, which exhibits the correct behavior at all angles. It is found that the NC and DW form are limiting cases of a more general form. We will show that the exact shape depends on the lateral correlation of the roughness profile. Whereas it is known that this lateral correlation can cause nonspecular scattering (where the parallel wave vector is not conserved),[5] it is somewhat contraintuitive that it also influences the specular, where the parallel wave vector is conserved. The reason, as will be explained below, is that in specular reflectivity contributions are possible from two-step scattering *via* intermediate states which have a different parallel wave vector and hence are influenced by the lateral correlation.

As the theoretical framework for scattering from a rough surface, we will use the distorted-wave Born approximation (DWBA) as advocated by Sinha *et al.*,[5] i.e., the roughness is considered as a perturbation on the flat surface. Whereas Sinha *et al.* go to first order in the perturbation, we will show the effect of going to higher order.

We assume that the wave field considered obeys the wave equation

$$\nabla^2\phi+k^2\phi-V\phi=0 ,$$

where ϕ is the electric field parallel to the (average) surface for (*s*-polarized) electromagnetic radiation or the wave function for particles like neutrons (provided that spin plays no role); k is the length of the wave vector in vacuum and V describes the interaction with the material. For *p*-polarized electromagnetic radiation the wave equation is not exact, but is a good approximation for short wavelengths (<10 nm) (cf. Ref. 8).

We will consider a homogeneous material with (complex) refractive index n. First we give the results for a flat surface separating the material (at $z<0$) from vacuum: $V=V_0$ with $V_0=0$ for $z>0$ and $V_0=k^2(1-n^2)\equiv k_c^2$ for $z<0$, where k_c is the critical wave vector. Starting with a plane wave in vacuum with wave vector $\mathbf{k}$, the solution of the wave equation is then

$$\phi_{\mathbf{k}}^{(0)}(\mathbf{r})=\psi_k(z)\exp(i\mathbf{k}_\parallel\cdot\mathbf{x}) ,$$

where $\mathbf{k}_\parallel$ and $\mathbf{x}=(x,y)$ are the projections of $\mathbf{k}$ and the position vector $\mathbf{r}$ parallel to the surface, respectively, and

(apart from a normalization constant)

$$\psi_k(z)=\exp(ik_0z)+r_k\exp(-ik_0z) \quad \text{for } z>0 ,$$

$$\psi_k(z)=t_k\exp(ik_1z) \quad \text{for } z<0 .$$

The Fresnel coefficients for reflection and transmission are, respectively, $r_k=(k_0-k_1)/(k_0+k_1)$ and $t_k=2k_0/(k_0+k_1)$, whereas $k_1=(k_0^2-k_c^2)^{1/2}$. In the following we also need the "irregular" solution (i.e., a wave starting inside the material, sometimes confusingly referred to as the "time-reversed" solution):

$$\psi_{\bar{k}}(z)=t_{\bar{k}}\exp(-ik_0z) \quad \text{for } z>0 ,$$

$$\psi_{\bar{k}}(z)=\exp(-ik_1z)+r_{\bar{k}}\exp(ik_1z) \quad \text{for } z<0 ,$$

where $r_{\bar{k}}=(k_1-k_0)/(k_0+k_1)$ and $t_{\bar{k}}=2k_1/(k_0+k_1)$.

Next we consider the influence of surface roughness. For a rough surface with at position $\mathbf{x}$, a height deviation $z(\mathbf{x})$ from the average surface, one can write

$$V=V_0+V_1(\mathbf{r}) ,$$

where $V_1(\mathbf{r})=k_c^2$ for $0\le z<z(\mathbf{x})$, $V_1(\mathbf{r})=-k_c^2$ for $z(\mathbf{x})<z\le 0$, and $V_1(\mathbf{r})=0$ elsewhere. We will treat $V_1(\mathbf{r})$ as a small perturbation on V_0. The solutions to the wave equation can be written as a perturbation series:

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

where $\phi_{\mathbf{k}}^{(0)}(\mathbf{r})$ is given above and the higher-order functions can be expressed as:[9]

$$\phi_{\mathbf{k}}^{(n)}(\mathbf{r})=\int d\mathbf{r}'G(\mathbf{r},\mathbf{r}')V_1(\mathbf{r}')\phi_{\mathbf{k}}^{(n-1)}(\mathbf{r}') .$$

It is convenient[10] to express the Green's function $G(\mathbf{r},\mathbf{r}')$ as a two-dimensional Fourier integral parallel to the surface:

$$G(\mathbf{r},\mathbf{r}')=\frac{1}{4\pi^2}\int_{|\mathbf{p}_\parallel|<k}d\mathbf{p}_\parallel\exp[i\mathbf{p}_\parallel\cdot(\mathbf{x}-\mathbf{x}')]g_p(z,z') ,$$

where $\mathbf{p}=(\mathbf{p}_\parallel,p_0)$ is a wave vector with length k. Perpendicular to the surface, the one-dimensional Green's function is[9]

$$g_p(z,z')=-\psi_p(z^<)\psi_{\bar{p}}(z^>)/W_p ,$$

where $z^<=\min(z,z')$, $z^>=\max(z,z')$, and the Wronskian $W_p\equiv\psi_{\bar{p}}d\psi_p/dz-\psi_p d\psi_{\bar{p}}/dz$.[9] Substituting ψ_p and $\psi_{\bar{p}}$ yields $W_p=2ip_0t_{\bar{p}}=2ip_1t_p$ and

$$\phi_{\mathbf{k}}^{(n)}(\mathbf{r})=\frac{i}{8\pi^2}\int_{|\mathbf{p}_\parallel|<k}\frac{d\mathbf{p}_\parallel}{p_0}T^{(n)}(p,k)\exp[i(\mathbf{p}_\parallel\cdot\mathbf{x}-p_0z)] \quad \text{for } z>0 , \quad (1)$$

$$\phi_{\mathbf{k}}^{(n)}(\mathbf{r})=\frac{i}{8\pi^2}\int_{|\mathbf{p}_\parallel|<k}\frac{d\mathbf{p}_\parallel}{p_1}T^{(n)}(\bar{p},k)\exp[i(\mathbf{p}_\parallel\cdot\mathbf{x}+p_1z)] \quad \text{for } z<0 , \quad (2)$$

where the so-called T matrix is

$$T^{(n)}(p,k)=\int d\mathbf{x}\exp(-i\mathbf{p}_\parallel\cdot\mathbf{x})\times\int_0^{z(\mathbf{x})}dz\,\psi_p(z)V_1(\mathbf{r})\phi_{\mathbf{k}}^{(n-1)}(\mathbf{r}) .$$

Strictly speaking, expressions (1) and (2) are not valid inside the rough surface layer, but it can be shown that they still are valid up to $O(k_0z)$, which will be sufficient to calculate the T matrix up to $O(k_0^2\sigma^2)$. As usual, all higher-order T matrix elements can be expressed in the first-order elements, for instance,

$$T^{(2)}(k',k)=\frac{i}{8\pi^2}\int_{|\mathbf{p}_\parallel|<k}\frac{d\mathbf{p}_\parallel}{p_0t_{\bar{p}}}T^{(1)}(k',\bar{p})T^{(1)}(p,k) .$$

Expressions (1) and (2) describe waves scattered in all directions. Up to $n=1$, the differential cross section for diffuse reflection is

$$\frac{d\sigma(p\leftarrow k)}{d\Omega}=\frac{|T^{(1)}(p,k)|^2}{16\pi^2} .$$

However, we want to calculate the specular reflectivity. To do this, we have to insert, in Eq. (1),

$$\delta_{k_x,p_x}\delta_{k_y,p_y}=(2\pi/L_x)\delta(k_x-p_x)(2\pi/L_y)\delta(k_y-p_y) ,$$

where L_x and L_y are the linear dimensions of the sample (which tend to infinity), and we have to take a configurational average (indicated by $\langle\ \rangle$). Writing the specular reflection coefficient from a rough surface as

$$\tilde{r}_k=r_k^{(0)}+r_k^{(1)}+r_k^{(2)}+\cdots ,$$

we find $r_k^{(0)}=r_k$ and

$$r_k^{(n)}=i\langle T^{(n)}(k,k)\rangle/(2L_xL_yk_0) \quad \text{for } n>0 .$$

To calculate the T matrix, we will approximate the unperturbed functions by $\psi_k(z)\simeq t_k\exp(ik_1z)$, also if $z>0$. This is reasonable for large k_0 up to $O(k_c^2/k_0^2)$, and also for small k_0 to calculate the T matrix up to $O(k_0^2\sigma^2)$. Then we find

$$T^{(1)}(p,k)\simeq k_c^2t_pt_kF(\mathbf{p}_\parallel-\mathbf{k}_\parallel,p_1+k_1) ,$$

where

$$F(\mathbf{q}_\parallel,q)\equiv -i/q\int d\mathbf{x}\exp(i\mathbf{q}_\parallel\cdot\mathbf{x})\{\exp[iqz(\mathbf{x})]-1\}$$

and

$$T^{(2)}(k,k)\simeq\frac{i}{4\pi^2}k_c^4t_k^2\int_{|\mathbf{p}_\parallel|<k}\frac{d\mathbf{p}_\parallel}{p_0+p_1}F(\mathbf{p}_\parallel-\mathbf{k}_\parallel,k_1-p_0)\times F(\mathbf{p}_\parallel-\mathbf{k}_\parallel,p_1+k_1) .$$

To calculate the configurational average, we assume that $z(\mathbf{x})$ is a Gaussian random variable with standard deviation σ. We find

$$\langle F(\mathbf{q}_\parallel,q)\rangle=-iL_xL_y\delta_{\mathbf{q}_\parallel,0}[\exp(-q^2\sigma^2/2)-1]/q$$

and up to $O(k_0^2\sigma^2)$:

$$\langle F(\mathbf{q}_\parallel,q)F(\mathbf{q}_\parallel',q')\rangle=L_xL_y\delta_{\mathbf{q}_\parallel,\mathbf{q}_\parallel'}C(\mathbf{q}_\parallel)$$

with

$$C(\mathbf{q}_\parallel)\equiv\int d\mathbf{X}\exp(i\mathbf{q}_\parallel\cdot\mathbf{X})C(\mathbf{X}) ,$$

where $C(\mathbf{x}-\mathbf{x}')\equiv\langle z(\mathbf{x})z(\mathbf{x}')\rangle$ is the correlation function of the roughness profile.

Note that the diffuse scattering cross section $d\sigma/d\Omega$ is [up to $O(k_0^2\sigma^2)$] proportional to $C(\mathbf{q}_\parallel)$.[5] The second-order *specular* reflectivity can be considered as a sum (over all $\mathbf{p}$) of (virtual) diffuse scattering contributions from $\mathbf{k}$ to $\mathbf{p}$ to $\mathbf{k}$. It is well known[5] that if the perpendicular component of either the incoming (k) or the scattered (p) wave vector equals $|k_c|$, peaks are seen in the diffuse scattering, often called anomalous scattering, Yoneda wings[11] or angel's wings. Since contributions from close to $|k_c|$ are also important in $r_k^{(2)}$, we may call these contributions "virtual angel's wings."

The first-order contribution to the reflection coefficient [up to $O(k_0^2\sigma^2)$] is easily found as $r_k^{(1)}=-2r_k k_0 k_1\sigma^2$ (derived in a more complicated way by Sinha *et al.*[5]).

The second-order contribution is

$$r_k^{(2)}=-\frac{1}{2\pi^2}r_k k_c^2 k_0\int_{|\mathbf{p}_\parallel|<k}\frac{d\mathbf{p}_\parallel}{p_0+p_1}C(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\ .$$

To calculate this, we have to know the roughness correlation function $C(\mathbf{X})$. Following Sinha *et al.*[5] we will assume that it has the form

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

where the parameter H ($0<H\leq 1$), describing how jagged the surface is, is connected to its fractal dimension $D=3-H$,[12] and the lateral correlation length ξ acts as a cutoff length for the (self-affine)[12] fractal behavior of the surface. Note that we implicitly assumed that ξ is smaller than the coherence length of the radiation parallel to the surface. To calculate $r_k^{(2)}$, first $C(\mathbf{q}_\parallel)$ has to be calculated (which can be done analytically if $H=1$ or $\frac{1}{2}$) and next the integral over $\mathbf{p}_\parallel$ has to be performed.

First we will consider two limiting cases, depending on the value of $(k-|\mathbf{k}_\parallel|)\xi/2\simeq k_0^2\xi/k$. If this value is $\gg 1$, $C(\mathbf{p}_\parallel-\mathbf{k}_\parallel)$ approaches a δ function centered at $\mathbf{k}_\parallel$ and we find $r_k^{(2)}\simeq -2r_k k_0(k_0-k_1)\sigma^2$, yielding

$$\tilde{r}_k\simeq r_k^{(0)}+r_k^{(1)}+r_k^{(2)}=r_k(1-2k_0^2\sigma^2)\ .$$

For $k_0\gg k_c$ the second-order correction amounts to $r_k^{(2)}\simeq -r_k k_c^2\sigma^2$. Hence, in the case of large k_0 this contribution is appreciable especially for heavy elements. If, on the other hand, $(k-|\mathbf{k}_\parallel|)\xi$ is small, then $r_k^{(2)}/r_k^{(1)}=O(k_0\sqrt{\xi/k}\,)$. Hence, if $k_0\sqrt{\xi/k}\ll 1$, then $r_k^{(2)}$ can be neglected.

It is interesting to compare our results with the existing expressions. In the case of large ξ ("predominance of low spatial frequencies" in the terminology of Croce and Névot[3]), one can consider reflection from a plane at $z(\mathbf{x})$, yielding a phase factor $\exp[2ik_0 z(\mathbf{x})]$ in the reflection coefficient. After averaging over all values of $z(\mathbf{x})$ (assumed to be distributed as a Gaussian random variable), one finds $\tilde{r}_k=r_k\exp(-2k_0^2\sigma^2)$, i.e., the DW form. In the case of small ξ ("predominance of high spatial frequencies"), the NC form $\tilde{r}_k=r_k\exp(-2k_0k_1\sigma^2)$ can be obtained from various approaches.[4,7,13,14] The same expression is also found using the Born approximation for a transition layer with thickness 2σ.) It is seen that, up to $O(k_0^2\sigma^2)$, these two expressions are equivalent to those found by us.

We propose to generalize our results as follows:

$$\tilde{r}_k\simeq r_k\exp\left[-2k_0k_1\sigma^2-\frac{1}{2\pi^2}k_0k_c^2\int_{|\mathbf{p}_\parallel|<k}\frac{d\mathbf{p}_\parallel}{p_0+p_1}C(\mathbf{p}_\parallel-\mathbf{k}_\parallel)\right]\ . \tag{3}$$

This expression has the right limiting values for both small and large $k_0^2\xi/k$ and interpolates smoothly in between. Although our results are valid for general ξ and small $k_0\sigma$, the interpolation is not necessarily correct for larger $k_0\sigma$. However, it is correct if both $k_0^2\sigma^2\ll 1$ and $k_0^2\xi/k\gg 1$, i.e., if $\xi\gg k\sigma^2$. It is also correct if $k_0^2\xi/k\ll 1$ for all k_0, i.e., if $k\xi\ll 1$, in which case the second term can be neglected. Hence, Eq. (3) is a manageable expression which we expect to give a good overall description for the specular reflectivity from a large variety of rough surfaces.

Now we turn to the calculation for arbitrary $k_0^2\xi/k$. The two-dimensional integral over $\mathbf{p}_\parallel$ in Eq. (3) can be transformed into pole coordinates. The angular integral can be performed both for $H=1$ and for $\frac{1}{2}$, yielding a modified Bessel function of the order zero and a complete elliptic integral of the second kind, respectively. Then one is left with a one-dimensional integral, which can be evaluated numerically.

As an example, in Fig. 1 calculations are shown for the reflectivity $|\tilde{r}_k|^2$ of Cu $K\alpha_1$ radiation ($k=40.784\ \text{nm}^{-1}$) on a gold sample ($n=1-4.644\times10^{-5}-i4.922\times10^{-6}$) with $\sigma=1.5$ nm and various values of ξ. The calculations shown are for $H=\frac{1}{2}$, but we did not find significant differences if $H=1$ was used. As is seen, the reflectivity is lower for $\xi=\infty$ than for $\xi=0$. The difference can be very large in the total-reflection region. In the case of large k_0, the difference becomes a constant factor of $\exp(2k_c^2\sigma^2)$, amounting to about 2. As anticipated, for finite values of ξ our theory interpolates between the two extremes, with the small-ξ behavior at small k_0 and the large-ξ behavior at large k_0, whereas the crossover occurs at $k_0^2\xi/k\simeq 1$.

For completeness we should mention that in a similar way the transmission coefficient can also be found. We have $t_k^{(0)}=t_k$ and, using Eq. (2), $t_k^{(n)}=i\langle T^{(n)}(\bar{k},k)\rangle/(2L_xL_yk_1)$ for $n>0$. From this we obtain:

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

For $k_0^2\xi/k \ll 1$ the second term between brackets vanishes and the result $\tilde{t}_k \simeq t_k \exp[(k_0-k_1)^2\sigma^2/2]$ is the same as that found by Vidal and Vincent[13] (or using Born's approximation for a transition layer with thickness 2σ). For $k_0^2\xi/k \gg 1$ the second term approaches $-(k_0-k_1)^2\sigma^2$, yielding $\tilde{t}_k \simeq t_k \exp[-(k_0-k_1)^2\sigma^2/2]$. The same result is found by averaging the phase factor $\exp[i(k_0-k_1)z(\mathbf{x})]$ obtained for the transmission coefficient of a plane at $z(\mathbf{x})$. Note that here the effect is less drastic than for the reflectivity, since $(k_0-k_1)^2\sigma^2 \simeq k_c^2\sigma^2/(2k_0^2)$ vanishes for large k_0.

In this report we only discuss reflection at materials with a single surface. In layered materials, the reflectivity can be found by using a Fresnel expression at each interface.[15] For small $k_0^2\xi/k$, the roughness can be taken into account by multiplying the reflection coefficient at each interface by the appropriate NC factor.[8,13,14] For larger $k_0^2\xi/k$ the situation is more complicated. Often there will be correlation between the roughness of different interfaces. For conformal roughness, where all interfaces have the same roughness σ, the total reflected field can be multiplied by a DW factor $\exp(-2k_0^2\sigma^2)$.[6] Even if this correlation is absent, the phase relation between the radiation scattered at the different interfaces means that the influence of roughness in a multilayer cannot be described in a simple way.[6] Work is in progress to quantify this effect with the method outlined above.

In conclusion, we have presented a theory for the specular reflection of x rays and neutrons from rough surfaces, which predicts a shape that depends on the lateral correlation of the roughness profile. To permit a comparison with experiments, some problems have to be overcome. In the first place, one has to subtract properly the diffuse scattering, which may be larger than the specular for large σ and ξ. Furthermore, real materials often contain a thin surface layer with a density different from the bulk, e.g., due to oxidation. In that case, more parameters are needed to describe the sample and, as was indicted above, for such a layered material the calculation can be complicated. However, it will be clear that in any case the effects described in this paper should be considered for accurate surface roughness determination from x-ray or neutron reflectivity measurements.

[1] *Surface X-Ray and Neutron Scattering*, edited by H. Zabel and I. K. Robinson (Springer-Verlag, Berlin, 1992).
[2] P. Beckman and A. Spizzichino, *The Scattering of Electromagnetic Waves from Rough Surfaces* (Pergamon, Oxford, 1963).
[3] P. Croce and L. Névot, Rev. Phys. Appl. **11**, 113 (1976).
[4] L. Névot and P. Croce, Rev. Phys. Appl. **15**, 761 (1980).
[5] S. K. Sinha, E. B. Sirota, S. Garoff, and H. B. Stanley, Phys. Rev. B **38**, 2297 (1988).
[6] J. M. Eastman, Phys. Thin Films **10**, 167 (1978).
[7] R. Pynn, Phys. Rev. B **45**, 602 (1992).
[8] D. K. G. de Boer, Phys. Rev. B **44**, 498 (1991).
[9] L. S. Rodberg and R. M. Thaler, *Introduction to the Quantum Theory of Scattering* (Academic, New York, 1967).
[10] D. L. Mills, A. A. Marudin, and E. Burstein, Ann. Phys. (N.Y.) **56**, 504 (1970); E. Evans and D. L. Mills, Phys. Rev. B **5**, 4126 (1972); A. A. Maradudin and D. L. Mills, *ibid.* **11**, 1392 (1975); P. Mazur and D. L. Mills, *ibid.* **26**, 5175 (1982).
[11] Y. Yoneda, Phys. Rev. **113**, 2010 (1963).
[12] B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, New York, 1982).
[13] B. Vidal and P. Vincent, Appl. Opt. **23**, 1794 (1984).
[14] F. Stanglmeier, B. Lengeler, W. Weber, H. Göbel, and M. Schuster, Acta. Crystallogr. A **48**, 626 (1992).
[15] L. G. Parratt, Phys. Rev. **95**, 359 (1954).