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RESEARCH PAPERS

The Resolution Function in Diffuse X-ray Reflectivity

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Abstract

A comprehensive discussion of the resolution function in specular and diffuse X-ray reflectivity is given. This is particularly relevant due to the proliferation of this technique in the field of liquid and solid surfaces and multilayer systems. A simple quantitatively correct interpretation of the diffuse reflectivity is possible if the resolution function is separable for the two directions in the scattering plane. This can be accomplished using a symmetric resolution set-up and specific types of scans (so-called radial scans and rocking scans).

1. Introduction

Specular X-ray reflectivity is presently a widely used technique for determining the electron-density profile in the direction normal to the surface of a flat sample. To obtain the required profile, models for the specular reflectivity are calculated and compared with experimental results (Als-Nielsen, 1991). Inevitably, this leads to some uncertainty concerning the uniqueness of the model chosen. However, in fact the specularly reflected signal is only part of the total reflectivity (Sinha, Sirota, Garoff & Stanley, 1988). The diffuse signal contains not only information about the in-plane structure of the surface, but may also contribute to the quality of the model used to describe the specular reflectivity. Measurements of the diffuse reflectivity have been used so far to determine the heigh–height correlation of liquid surfaces (Sanyal, Sinha, Huang & Ocko, 1991; Tidswell, Rabedeau, Pershan & Kosowsky, 1991), solid surfaces (Savage *et al.*, 1991; Weber & Lengeler, 1992) and multilayers (Holy & Baumbach, 1994; Salditt, Metzger & Peisl, 1994; Schlatmann, Shindler & Verhoeven, 1995). Applications to soft condensed matter comprise soap films (Daillant & Bélorgey, 1992), liquid-crystal polymer films on a substrate (Geer, Shashidar, Thibodeaux & Duran, 1993; Geer & Shashidar, 1995) and free-standing smectic liquid-crystalline films (Shindler, Mol, Shalaginov & de Jeu, 1995).

As in all X-ray measurements, the measured intensity $I(\mathbf{q})$ is a convolution of the structure-factor intensity $S(\mathbf{q})$ and the experimental resolution function $R(\mathbf{q})$:

$$I(\mathbf{q}) = \int S(\mathbf{q}) R(\mathbf{q} - \mathbf{q}')\, d\mathbf{q}'. \qquad (1)$$

Typically, a model for $S(\mathbf{q})$ is available and the folding integral is calculated with the variation of free parameters in the structure factor until the best fit to the observed intensity is obtained. In practice, the three-dimensional convolution of (1) is not easy and some possibilities to avoid complications have not always fully been appreciated. The objective of this paper is to give a precise account of this resolution problem. It turns out that specific types of scans (so-called radial scans and rocking scans) in combination with a symmetric resolution set-up allow a simple quantitatively correct interpretation of the full (specular and diffuse) reflectivity.

2. The asymmetry in incoming and outgoing divergences $\Delta\alpha$ and $\Delta\beta$

We refer to Fig. 1 for the definition of the relevant angles:

$$\alpha = \theta + \omega \quad \alpha + \beta = 2\theta \quad \alpha > 0 \text{ cw} \quad \beta > 0 \text{ ccw}$$
$$\beta = \theta - \omega \quad \tfrac{1}{2}(\alpha - \beta) = \omega \quad \omega > 0 \text{ ccw}$$

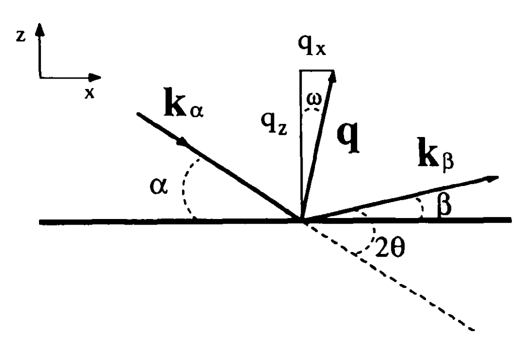


Fig. 1. Definition of the scattering geometry and the various angles.

(cw = clockwise; cww = counterclockwise). The wavevector transfer is given by

$$\mathbf{q} = \mathbf{k}_\beta - \mathbf{k}_\alpha,$$

with (2)

$$q = |\mathbf{q}| = 2k_0 \sin\theta,$$

and $k_0 = 2\pi/\lambda$, where λ is the X-ray wavelength. We want to consider deviations $\delta\alpha$ and $\delta\beta$ from the nominal values α and β, respectively, leading to divergence distributions $\Delta\alpha$ and $\Delta\beta$ [full width at half-maximum (FWHM)]. These will be assumed to be Gaussian distributions leading to a resolution area in the (q_x, q_z) plane (scattering plane) determined by $k_0\Delta\alpha$ and $k_0\Delta\beta$ at an angle of 2θ with each other (see Fig. 2). The y direction is orthogonal to this plane and can be treated separately. In practice, the resolution in this direction is often set coarse enough to be effectively integrated over. Also note that the half-width σ of a Gaussian at its $1/e$ value and a FWHM value like $\Delta\alpha$ are related by $\Delta\alpha = 2\sigma(2 \ln 2)^{1/2}$.

The components of $\mathbf{q}$ can be related to the angles α and β (see Fig. 1):

$$q_z = k_0(\sin\beta + \sin\alpha), \quad (3a)$$

$$q_x = k_0(\cos\beta - \cos\alpha). \quad (3b)$$

Differentiation leads to (see also Gibaud, Vignaud & Sinha, 1993)

$$\delta q_z = k_0(\cos\beta\, \delta\beta + \cos\alpha\, \delta\alpha) + \delta k_0(\sin\beta + \sin\alpha), \quad (4a)$$

$$\delta q_x = -k_0(\sin\beta\, \delta\beta - \sin\alpha\, \delta\alpha) + \delta k_0(\cos\beta - \cos\alpha), \quad (4b)$$

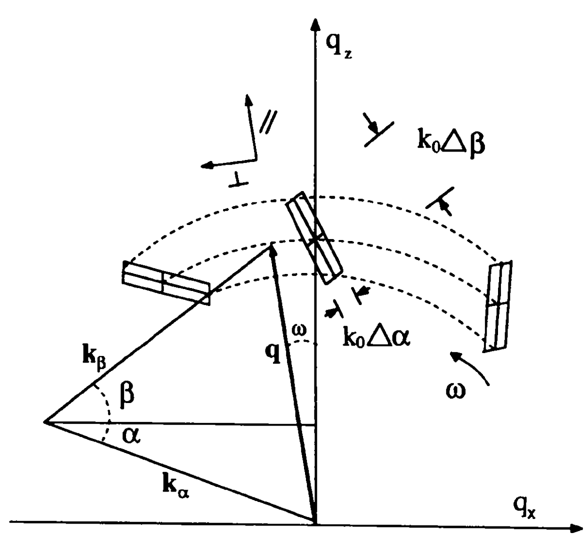


Fig. 2. Rotation of the resolution area upon variation of ω from $-\theta$ to θ (after Gibbs *et al.*, 1988). For reasons of clarity the unrealistically large value $2\theta = 60°$ has been chosen.

where from its definition δk_0 is related to the wavelength dispersion by $\delta k_0 = k_0\delta\lambda/\lambda$. For random Gaussian distributions, $\Delta\beta$ and $\Delta\alpha$, the distributions of uncertainty in q_z and q_x are also Gaussians and given by:

$$\Delta q_z^2 = k_0^2[\cos^2\beta(\Delta\beta)^2 + \cos^2\alpha(\Delta\alpha)^2] + \Delta k_0^2(\sin\beta + \sin\alpha)^2, \quad (5a)$$

$$\Delta q_x^2 = k_0^2[\sin^2\beta(\Delta\beta)^2 + \sin^2\alpha(\Delta\alpha)^2] + \Delta k_0^2(\cos\beta - \cos\alpha)^2. \quad (5b)$$

For small angles, the contribution from Δk_0^2 can be disregarded. Using the definitions of the various angles, we can write (see also Shindler & Sutter, 1992):

$$\Delta q_z = k_0[(\Delta\alpha)^2 + (\Delta\beta)^2]^{1/2} \quad (6a)$$

$$\Delta q_x = k_0[\alpha^2(\Delta\alpha)^2 + \beta^2(\Delta\beta)^2]^{1/2} = (q/2)[(1 + \omega/\theta)^2(\Delta\alpha)^2 + (1 - \omega/\theta)^2(\Delta\beta)^2]^{1/2}. \quad (6b)$$

For $|\omega| \ll \theta$, we find $\Delta q_x = (q/2k_0)\Delta q_z = \theta\Delta q_z$, while for $\omega = \theta$ and $\omega = -\theta$ we see that Δq_x is equal to $q\Delta\alpha$ and $q\Delta\beta$, respectively. The resolution is asymmetric as long as $\Delta\beta \neq \Delta\alpha$, as illustrated in Fig. 2. In Fig. 3, the resolution area along q_z is pictured for small angles and the situations $\Delta\beta \gg \Delta\alpha$ and $\Delta\beta = \Delta\alpha$, respectively. The resolution Δq_z, that is relevant for the diffusely reflected signal is given by the *projection* of the resolution area on the q_z axis. In this situation, the results for Δq_z and Δq_x can be calculated from (6); they are summarized in Table 1 for both $\Delta\beta \gg \Delta\alpha$ and $\Delta\beta = \Delta\alpha$. The situation is different for a specular scan, which in principle is strictly confined to the q_z axis. Then, the resolution Δq_z is given by the length along q_z that is *enclosed* by the resolution area. For this idealized situation, one can calculate from Fig. 3 $\Delta q_z^{\rm spec} = 2k_0\Delta\alpha$, independent of $\Delta\beta$.

3. Transformation from $R(\delta\alpha, \delta\beta)$ to $R(\delta q_x, \delta q_z)$ and to $R(\delta q_\perp, \delta q_\parallel)$

All derivations will further be made in the small-angle approximation. Also disregarding possible variations in δk_0, one can write (4*a*, *b*) as

$$\delta q_z = k_0(\delta\beta + \delta\alpha), \quad (7a)$$

$$\delta q_x = -k_0(\beta\, \delta\beta - \alpha\, \delta\alpha). \quad (7b)$$

This can easily be solved to give

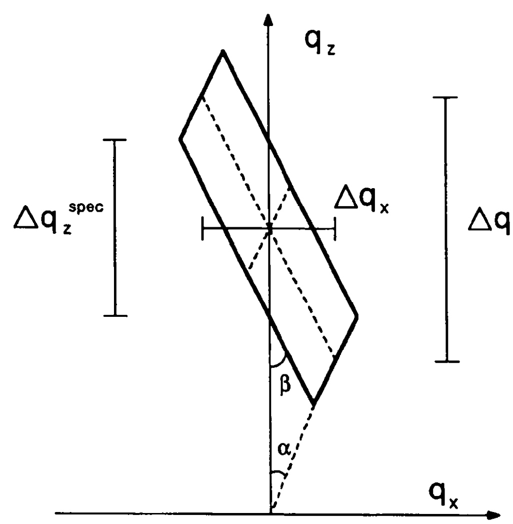
$$\delta\alpha = (\delta q_z\beta + \delta q_x)/[k_0(\alpha + \beta)] = (1/q_z)(\delta q_z\beta + \delta q_x), \quad (8a)$$

$$\delta\beta = (\delta q_z\alpha - \delta q_x)/[k_0(\alpha + \beta)] = (1/q_z)(\delta q_z\alpha - \delta q_x). \quad (8b)$$

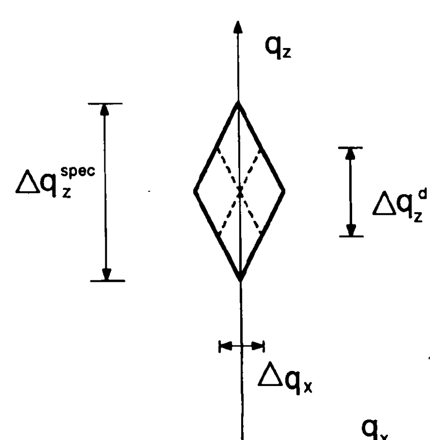
First, we want to calculate the resolution function $R(q_x, q_z)$ starting from the known distribution of α and β, or from

$$R(\delta\alpha, \delta\beta) = \exp(-\delta\alpha^2/\Delta\alpha^2)\exp(-\delta\beta^2/\Delta\beta^2). \quad (9)$$

This expression is separable in α and β because α and β are statistically independent, even though the distributions $k\Delta\alpha$ and $k\Delta\beta$ are in general not orthogonal. However, a transformation of these one-dimensional Gaussians will give a resolution function that is in general not separable in q_x and q_z. Substituting (8) in (9), we



(*a*)



(*b*)

Fig. 3. Details of the resolution area at $\omega = 0$ for (*a*) $\Delta\beta \gg \Delta\alpha$ and (*b*) $\Delta\beta = \Delta\alpha$.

Table 1. *Values for the resolution in various situations*

		$\Delta\beta \gg \Delta\alpha$	$\Delta\beta = \Delta\alpha$
	Δq_z^{diff}	$k_0\Delta\beta$	$2^{1/2}k_0\Delta\alpha$
At $\omega \simeq 0$	Δq_x	$q\Delta\beta/2$	$q\Delta\alpha/2^{1/2}$
At $\omega/\theta = 1$	Δq_x	$q\Delta\alpha$	$q\Delta\alpha$
At $\omega/\theta = -1$	Δq_x	$q\Delta\beta$	$q\Delta\alpha$

obtain an expression that makes this explicit.

$$\begin{aligned} R(\delta q_x, \delta q_z) &= \exp[-(\delta q_z\beta + \delta q_x)^2/(q_z\Delta\alpha)^2] \\ &\quad \times \exp[-(\delta q_z\alpha - \delta q_x)^2/(q_z\Delta\beta)^2] \\ &= \exp\{[-(\delta q_z)^2(\alpha^2\Delta\alpha^2 + \beta^2\Delta\beta^2) \\ &\quad - (\delta q_x)^2(\Delta\alpha^2 + \Delta\beta^2) \\ &\quad - 2\delta q_z\delta q_x(\beta\Delta\beta^2 - \alpha\Delta\alpha^2)]/(q_z\Delta\alpha\Delta\beta)^2\}. \end{aligned} \quad (10)$$

Now, if we consider the situation $\Delta\alpha = \Delta\beta$, (10) can be written as

$$\begin{aligned} R(\delta q_x, \delta q_z) &= \exp\{[-(\delta q_z)^2/(\Delta q_z)^2][(\alpha^2 + \beta^2)/(2\alpha^2)\} \\ &\quad \times \exp\{[-(\delta q_x)^2/(\Delta q_x)^2][(\alpha^2 + \beta^2)/(2\alpha^2)]\} \\ &\quad \times \exp\{[-\delta q_z\delta q_x/(\Delta q_z\Delta q_x)] \\ &\quad \times [(\beta - \alpha)(\alpha^2 + \beta^2)^{1/2}/(2^{1/2}\alpha^2)]\}. \end{aligned} \quad (11)$$

Note that, even in this case of a symmetric resolution, there is a cross-term in $\delta q_z\delta q_x$ proportional to $(\beta - \alpha)$ that causes a changing 'tilt' of the resolution area during the course of a scan varying ω. Similarly, if we substitute $\alpha = \beta$ (but with $\Delta\alpha \neq \Delta\beta$), there is a cross-term proportional $(\Delta\beta - \Delta\alpha)$. A separable resolution function of the desired form,

$$R(\delta q_x, \delta q_z) = \exp[-(\delta q_z)^2/(\Delta q_z)^2]\exp[-(\delta q_x)^2/(\Delta q_x)^2], \quad (12)$$

can only be obtained if we take both $\Delta\alpha = \Delta\beta$ (symmetric resolution) and $\alpha = \beta$, *i.e.* for the specular reflectivity only. As a consequence of these transformations, for the diffuse reflectivity (where necessarily $\alpha \neq \beta$), there is always a coupling between δq_x and δq_z as given by (11). This will complicate any analysis of the scattered intensity, as a full two-dimensional convolution process is required to obtain data that can be interpreted quantitatively.

The problem described above can be considerably simplified if we consider in the *xz* plane apart from (q_x, q_z) also $(q_\perp, q_\parallel)$; see Fig. 1. For the small angles under consideration, $q_\parallel = q_z = |\mathbf{q}|$ and $q_\perp = q_x$. However, similar simple relations do not hold for the resolution function. As $\mathbf{q}$ and the resolution area rotate together with ω, the directions parallel and perpendicular to $\mathbf{q}$ always make a constant angle with the principal axes of the resolution area. Consequently, neither $\Delta q_{\parallel}$ nor $\Delta q_{\perp}$ varies as a function of ω, in contrast to Δq_z and Δq_x. Hence, to calculate $R(\delta q_{\perp}, \delta q_{\parallel})$ we can choose the situation $\omega = 0$ (corresponding to $\alpha = \beta$):

$$R(\delta q_{\perp}, \delta q_{\parallel})|_{\text{all }\omega} = R(\delta q_x, \delta q_z)|_{\omega=0}. \quad (13)$$

If we now set again $\Delta\beta = \Delta\alpha$, then $R(\delta q_{\perp}, \delta q_{\parallel})$ is fully separable in $q_{\perp}$ and $q_{\parallel}$:

$$R(\delta q_{\perp}, \delta q_{\parallel}) = \exp[-(\delta q_{\parallel})^2/(\Delta q_{\parallel})^2] \exp[-(\delta q_{\perp})^2/(\Delta q_{\perp})^2]. \quad (14)$$

This separable resolution function applies if scans are made parallel to $\mathbf{q}$ (radial scan: θ, 2θ scan at constant offset of ω) and perpendicular to $\mathbf{q}$ (rocking scan with only ω changed and $\alpha + \beta$ kept constant), respectively.

4. Discussion

For the specular reflectivity, the resolution Δq_z^{spec} is constant only when a δ function along the z axis is considered. In that situation, Δq_z^{spec} is independent of the value of $\Delta\beta$ and thus the same for the two situations depicted in Fig. 3. In real experiments, the observed integrated intensity does depend on Δq_x, which is much smaller for $\Delta\beta = \Delta\alpha$ then for $\Delta\beta \gg \Delta\alpha$ (compare Figs. 3*a* and *b*). This means that part of the diffusely reflected intensity is lumped together with the specular one. Although this problem is in principle well known and has been analysed in some detail (Sinha *et al.*, 1988; Daillant & Bélorgey, 1992), its relevance is not always appreciated. Fortunately, the diffuse reflectivity at small angles often shows a q_z dependence similar to that of the specular one. Then, there is at least no qualitative dependence of the measured reflected intensity on the choice of Δq_x. However, if a quantitative result is desired, especially at large values of q_z this effect should be carefully taken into account.

To obtain results for the diffuse X-ray reflectivity that can be interpreted quantitatively, a resolution function that is separable in the components of $\mathbf{q}$ is highly desirable. This can be accomplished by setting $\Delta\beta = \Delta\alpha$ (symmetric resolution) and making radial scans and rocking scans, so that the directions parallel and perpendicular to $\mathbf{q}$ are probed and (14) applies. While for the small angles under consideratioin $q_{\parallel} = q_z = |\mathbf{q}|$ and $q_{\perp} = q_x$, it is found that $(\Delta q_x, \Delta q_z)$ differs from $(\Delta q_{\perp}, \Delta q_{\parallel})$. As an illustration of this choice, we consider a typical structure factor for surfaces (Sinha *et al.*, 1988) that is also relevant for layered systems:

$$S(\mathbf{q}) \propto \int \mathrm{d}x\, \mathrm{d}y \exp[q_z^2 C(x, y, q_z)] \exp[-i(q_x x + q_y y)], \quad (15)$$

where $C(x, y, q_z)$ describes either the height–height correlation of the surface or the displacement–displacement correlation of a layered system. As mentioned before, the y direction perpendicular to the scattering plane can usually be integrated out. To interpret the total (specularly and diffusely) reflected intensity in general, a two-dimensional convolution with the resolution function would be necessary, with all the complications described above. However, owing to its separability, a two-dimensional convolution with $(\Delta q_{\parallel}, \Delta q_{\perp})$ can be performed as a one-dimensional convolution (denoted $\otimes$) along $q_{\parallel}$, and a real-space cut-off of $1/\Delta q_{\perp}$ owing to the structure-factor integration along x. This leads to (Shindler *et al.*, 1995)

$$I(\mathbf{q}) \propto \{[\Delta q_{\perp}/(2\pi)^{1/2}] \int \mathrm{d}x \exp(-\tfrac{1}{2}x^2 \Delta q_{\perp}^2) \times \exp[q_z^2 C(x, q_z)] \exp(-iq_x x)\} \otimes \exp(-\tfrac{1}{2}q_{\parallel}^2/\Delta q_{\parallel}^2). \quad (16)$$

Note that the directions $(\parallel, \perp)$ can be identified with z, x in the integration coordinates, but not in $(\Delta q_{\parallel}, \Delta q_{\perp})$. With the method described, a complicated separate normalization of the specular and diffuse component – which is artificial anyhow – can be avoided.

At synchrotron sources and high resolution, $\Delta\alpha$ can be quite small; we have explored typically $\Delta\omega = 35$ µrad at ID10 at ESRF (Grenoble). At a detector distance of 1 m, the condition $\Delta\alpha = \Delta\beta$ then leads to a small but still practical slit width of 35 µm. This is the price to be paid for the convenience of not solving the full two-dimensional resolution problem. In such a high-resolution set-up, however, often $\Delta\omega_{\text{mos}}$ due to the mosaic distribution of the sample might be comparable or even larger than $\Delta\alpha = \Delta\beta$ as determined by the instrument. This is equivalent to the addition to $\Delta q_{\perp}$ of an extra sweep in ω equal to $\Delta\omega_{\text{mos}}$ (see Fig. 2). Again with the assumption of independent Gaussian distributions, this leads to $\Delta q_{\perp}^2(\text{tot}) = \Delta q_{\perp}^2 + q^2 \Delta\omega_{\text{mos}}^2$, while $\Delta q_{\parallel}$ is not influenced. Problems will arise if $\Delta\omega_{\text{mos}}$ is not constant during the experiment, for example because of a variation of the footprint of the X-ray beam on the sample. In that situation, it is preferable to choose a coarser resolution where the influence of $\Delta\omega_{\text{mos}}$ is eliminated.

5. Conclusion

We have shown that a simple quantitatively correct interpretation of the diffuse reflectivity is possible if the resolution function is chosen to be separable for two directions in the scattering plane. This can be accomplished by setting a symmetric resolution set-up and making radial scans and rocking scans, parallel and perpendicular to the wave-vector transfer $\mathbf{q}$, respectively.

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