

A Scheme for Smoothed Groove Tracking

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A new scheme that involves Fourier transformation to determine a correlation function of the first derivative of an electronic-density profile is proposed to improve the groove-tracking method. Tests of application of this scheme on simulated data demonstrate that this scheme is helpful in analysis of reflectivity data to retrieve profiles of electronic density that are not completely smooth, and operates with little or no prior information about unknown profiles. The mathematical basis of the new scheme is discussed.

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I. Introduction

Specular reflection of both x-rays and neutrons from a plane surface can yield information about the profile of the surface, i.e., its electronic structure in a perpendicular direction [1-3]. Procedures to analyse x-ray reflectivity data either depend on some models or are independent thereof [4, 5]. Because reflectivity data lack phase information and because the range of q_z over which reflectivity can be measured is limited, neither approach can yield an inherently unique profile of electronic density [6, 7]; this phase problem will be solved if various theoretical [8, 9] and experimental considerations [10-12] yield a method to determine the phase of reflectivity data. The effect of a limited range of q_z is discussed theoretically in our previous paper [13]. In practice a model-independent scheme for analysis of reflectivity data has an advantage that such a method can retrieve a profile of electronic density directly from reflectivity data with little or no prior information about an unknown profile. Therefore a model-independent method is helpful in revealing from experimental data physical phenomena of surface structures that might be neglected or undiscovered in related theories and speculations.

According to the groove-tracking method (GTM) [14-16] originated by Zhou and Chen, which works independently of a model, the density profile is first approximated with a few steps of equal width and independent height. The reflectivity for this model interface is computed, compared with experimental data, with a cost function defined in Ref. [16]; the density of each step is then independently varied to minimize the cost function. Successive approximations are made on subdividing each step and repeating the process while allowing subsequent amplitudes for narrower steps to vary. The procedure is complete when the calculated cost function attains an acceptable value and reveals a step-like profile to resemble the smooth unknown profile of the particular sample.

Improving the original GTM, which has proved applicable to many samples, we developed a smoothed groove-tracking method (SGTM) [17], by imposing a requirement that a profile of

electronic density be smooth. According to this method, the surface region of a sample is divided into thin layers; then the reflectivity is calculated more precisely by using Parratt's formula. The fitting procedure in the original GTM, which sometimes encounters problems with local minima, is also modified for mathematical reasons [13]. A combination of improved GTM and successive smoothing procedures enables a reasonable extrapolation of reflectivity data beyond $q_{z,\max}$. Hence providing more physically reasonable profiles than typically jagged and discontinuous profiles generated with the original GTM, the SGTm makes the original GTM significantly more practical especially in cases in which experimental data are measured within a limited range of q_z . In this way the SGTm provides an undiscovered but meaningful profile [18].

With this perspective in mind, the SGTm is applicable also to neutron reflectivity data and can serve to analyze reflectance with phase information after the phase problem becomes solved in the future with the help of mature techniques for measurements of the phase.

In applying SGTm to reflectivity of a sample with a non-smooth profile there remains a problem, even though GTM inherently yields a step-like retrieved profile. The disadvantage arises because precise positions of sharp edges of an unknown non-smooth profile can not be determined in advance. Here we present a new scheme based on Fourier transformation of reflectivity data divided by Fresnel reflectivity of bulk density that serves to reconstruct such an unknown profile.

II. Theory of x-ray reflectivity and a new scheme Introduction

The quantity generally measured is the reflectivity, which is obtained theoretically according to Parratt's formula [19]. The region of a surface of interest is divided into N equal slices, each of thickness Δ such that $N\Delta = d$. The depth d of the surface region must be large enough to ensure that all features of a profile of electronic density are distributed within this depth. N can be as large as duration of computation allows so that the profile of electronic density within each slice can be regarded as constant. The real profile of electronic density $\rho(z)$ is then replaced without loss with a discrete $\rho_N = [\rho_1, \rho_2, \rho_3, \dots, \rho_N]$. As a function of the free-space wave number $k_o = (2\pi/\lambda) \sin \theta$, in which θ denotes grazing angle and λ the length of the wave in free space, the theoretical reflectance of N uniform slices is given in terms of profile ρ_N through a recurrence relation [19],

$$r_i = \frac{R_{i+1} + r_{i+1} \exp(2ik_{i+1}\Delta Z_{i+1})}{1 + R_{i+1} + r_{i+1} \exp(2ik_{i+1}\Delta Z_{i+1})} \quad (1)$$

in which $\Delta Z_{i+1} = \Delta$ is the thickness of layer $i+1$, and $i=N-1, N-2, \dots, 2, 1, 0$.

$$k_{i+1} = \sqrt{(k_0^2 - 4\pi\rho_{i+1})} \quad (2)$$

is the wave number in layer $i+1$, $R_{i+1} = (k_i - k_{i+1})/(k_i + k_{i+1})$ is the Fresnel reflectance of the interface between layers i and $i+1$, and $r_N = (k_N - k_\infty)/(k_N + k_\infty)$ is the Fresnel reflectance of the interface between layer N and the bulk. For given ρ_N , the reflectance of the entire assembly of N layers is $r_o(k_o)$ and the reflectivity is expressed as $|r_o(k_o)|^2$

According to the Born approximation, $R_i \ll 1$, and the Fresnel reflectance is expressed as

$$R_{i+1} \approx \frac{4\pi(\rho_{i+1} - \rho_i)}{(2k_0)^2} \quad (3)$$

Therefore, Eq. (1) becomes

$$r_i \approx R_{i+1} + r_{i+1} \exp(2ik_{i+1}\Delta Z_{i+1}) \quad (4)$$

Assuming that variation of k_{i+1} in separate slices, of which the electron density varies, can be neglected, i.e., $k_{i+1} \approx k_o$ for all i , and denoting k_{i+1} as k_o , we obtain the reflectance

$$r(q_z) = \frac{4\pi}{q_z^2} \int \frac{d\rho(z)}{dz} e^{iq_z z} dz, \quad (5)$$

with $q_z = 2k_o$. Integrating Eq. (5) by parts, one can also express the reflectance as

$$r(q_z) = \frac{-4\pi i}{q_z} \int \rho(z) e^{iq_z z} dz \quad (6)$$

for convenience. Following Eq. (5), the reflectivity is given by

$$R(q_z) = \left| \frac{4\pi}{q_z^2} \int \frac{d\rho(z)}{dz} e^{iq_z z} dz \right|^2. \quad (7)$$

According to Eq. (3), the Fresnel reflectivity of the bulk density is

$$R_F(q_z) = \left| \frac{4\pi\rho_1}{q_z^2} \right|^2. \quad (8)$$

Hence,

$$\frac{R(q_z)}{R_F(q_z)} = \left| \frac{1}{\rho_1} \int_{i-1}^1 \frac{d\rho(z)}{dz} e^{iq_z z} dz \right|^2 \quad (9)$$

In the following the factor $1/\rho_1$ is neglected; hence a profile of electronic density $\rho(z)$ has units of ρ_1 for numerical simplicity; we also let $d\rho(z)/dz = f(z)$. It follows that

$$\frac{R(q_z)}{R_F(q_z)} = \left| \int_{i-1}^1 f(z) e^{iq_z z} dz \right|^2 \quad (10)$$

$$= \left(\int_{i-1}^1 f(z) e^{iq_z z} dz \right) \times \left(\int_{i-1}^1 f(z') e^{iq_z z'} dz' \right) \quad (11)$$

$$= \int_{i-1}^1 \int_{i-1}^1 f(z) f(z') e^{iq_z(z+z')} dz dz'. \quad (12)$$

Setting $z - z' = x$, $z = x + z'$ and $dz = dx$, we obtain from the above equation

$$\frac{R(q_z)}{R_F(q_z)} = \int_{i-1}^1 \int_{i-1}^1 f(x+z') f(z') e^{iq_x x} dx dz' \quad (13)$$

$$= \int_{i-1}^1 e^{iq_x x} dx \left[\int_{i-1}^1 f(x+z') f(z') dz' \right] \quad (14)$$

$$= \int_{-1}^1 P(x) e^{iq_x x} dx, \quad (15)$$

in which

$$P(x) = \int_{-1}^1 f(x+z) f(z) dz \quad (16)$$

$$= \int_{-1}^1 \frac{d\rho(z)}{dz} \frac{d\rho(x+z)}{dz} dz. \quad (17)$$

The correlation function $P(x)$ is a convolution integral of the derivative of a profile of electronic density. In practice, if the derivative of such a profile is not complex, $P(x)$ reveals the positions of sharp edges of a profile. For example, for a profile of a sample either a thin film or a wetting film, there are generally only two sharp edges of which the positions are readily obtained from the correlation function. In applying the GTM to reflectivity data of such samples, we thus divided the surface region according to information about edge positions indicated by correlation function $P(x)$.

The task then becomes to derive a correlation function $P(x)$ from experimental or simulated data. Defining $R(q_z)/R_F(q_z) = F(q_z)$, we rewrite Eq. (15) as

$$F(q_z) = \int_{-1}^1 P(x) e^{iq_x x} dx, \quad (18)$$

and obtain

$$P(x) = \frac{1}{2\pi} \int_{-1}^1 F(q_z) e^{iq_z x} dq_z. \quad (19)$$

Because $F(q_z)$ is an even function,

$$P(x) = \frac{1}{2\pi} \int_{-1}^1 F(q_z) \cos(q_z x) dq_z \quad (20)$$

$$= \frac{1}{\pi} \int_0^1 F(q_z) \cos(q_z x) dq_z \quad (21)$$

which is used to obtain numerically $P(x)$ from experimental or simulated data $R(q_z)$ divided by Fresnel reflectivity $R_F(q_z)$ depending on bulk density.

III. New scheme and its application

We proceed to use reflectivity data for two simulated profiles for electronic density, which are not completely smooth, to illustrate the new scheme and to demonstrate that this scheme provides profiles faithfully reproducing the original ones. We employ simulated data so that the

correctness of the retrieved profile is verified directly against the originally simulated profiles to confirm the reliability of the new scheme.

The first simulated profile, depicted as a solid curve in Fig. 1(a), resembles that of a thin film on a substrate. The corresponding reflectivity data of this profile appear in Fig. 1(b). With an aim to derive a correlation function $P(x)$, we apply GTM to only two data points near the critical q_z for total external reflectivity to determine the bulk density ρ_1 . Then it is easy to obtain the Fresnel reflectivity of the bulk density, shown as a dashed curve in Fig. 2(a), and $R(q_z)/R_F(q_z)$, shown as a solid curve in Fig. 2(a). Employing Eq. (21), we obtain correlation function $P(x)$ shown in Fig. 2(b). Because $P(x)$ is the convolution integral of the derivative of the pertinent profile of electronic density, the inverse peak of function $P(x)$ at 79\AA reveals that the distance between two sharp edges of the profile along the perpendicular direction is about 79\AA . Therefore, instead of being divided into layers arbitrarily but inflexibly in the original GTM [15], the surface region of the sample is divided into layers of thickness $D = 79, 39.5, 19.75, 9.875, 4.938\text{\AA}$, respectively at separate stages of the refined GTM. At various stages the data points distributed within $q_{z,\max} = \pi/D$ are taken into consideration for fitting [13]. The vertical bars, shown in

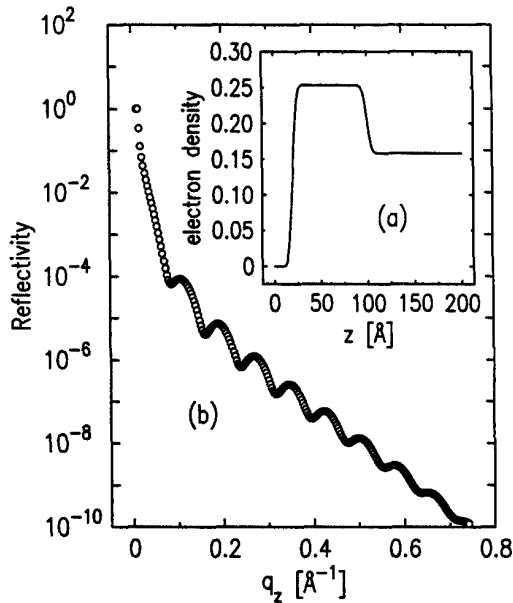


FIG. 1. (a) An original profile of electronic density, and (b) reflectivity data for this profile.

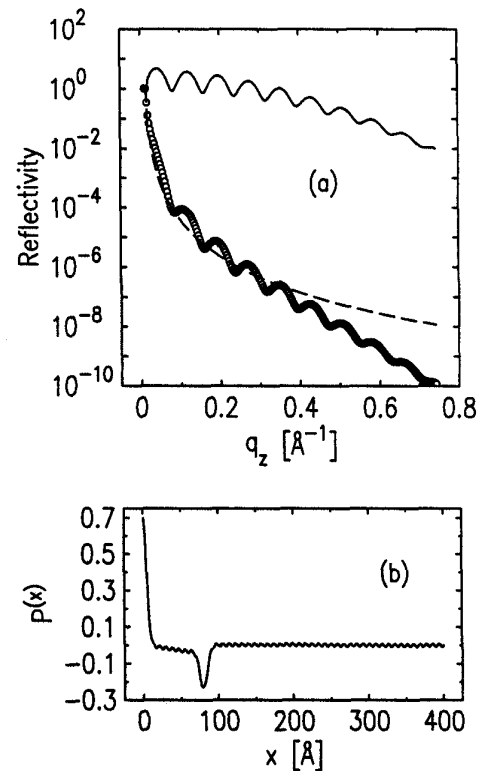


FIG. 2. (a) Reflectivity data (circles) shown in Fig. 1(b) with Fresnel reflectivity $R_F(q_z)$ (dashed curve) and $R(q_z)/R_F(q_z)$ (solid curve). (b) Function $P(x)$ obtained through Fourier transformation.

Fig. 3(a), indicate the boundary at which $q_{z,\max} = \pi/D$ for values of D . The evolution of profiles retrieved at consecutive stages of the GTM is illustrated in Fig. 3(b). If layers of thickness $D = 4.938 \text{ \AA}$ are further divided into thinner layers of thickness $D \approx 2.5 \text{ \AA}$, data points distributed within $q_{z,\max} = 1.2 \text{ \AA}^{-1}$ are needed for reasonable fitting, but such data points in this case are distributed only within the cutoff $q_z \approx 0.74 \text{ \AA}^{-1}$. In the final stage of the refined GTM only data points within $q_{z,\max} = 0.62 \text{ \AA}^{-1}$ are thus fitted to construct the resultant GTM profile with resolution D about 4.938 \AA . In order to minimize the cost function between simulated and fitted reflectivity data, the whole GTM procedure, including all the stages, is undertaken in three iterations. A step-like profile, denoted ‘‘third iteration’’ in Fig. 3(b), is eventually obtained on fitting data within $q_{z,\max} = 0.62 \text{ \AA}^{-1}$. This step-like profile is successively smoothed. While the SGTM is applied to the smoothed profile, the smoothed profile is re-divided into layers of thickness 4.245 \AA . A relation between the thickness of divided layers and the boundary $q_{z,\max}$ of data considered for fitting is satisfied, i.e., $0.74 \text{ \AA}^{-1} = \pi/4.245 \text{ \AA}$, and all simulated (or experimental) data within a cutoff $q_z = 0.74 \text{ \AA}^{-1}$ are fitted to obtain the best resolution of the retrieved profile. The computed reflectivity from the resultant SGTM profile is compared with simulated reflectivity data in Fig. 4(b). Satisfactory agreement is demonstrated between the simulated reflectivity data and the the computed reflectivity. Furthermore, the resultant SGTM profile and the original profile, shown in Fig. 4(a), are so similar that the dashed curve is scarcely distinguishable from the solid curve.

Another simulated profile, shown in an inset of Fig. 5(a), is inspired by x ray measurements on films of liquid hexane adsorbed on a rough silicon wafer [20]. The corresponding reflectivity $R(q_z)$ of this profile, the Fresnel reflectivity $R_F(q_z)$ and $R(q_z)/R_F(q_z)$ are shown in Fig. 5(b). According to Eq. (21), we obtain correlation function $P(x)$ shown in Fig. 5(a). A large maximum of $P(x)$ located at 84 \AA indicates that the distance between two sharp edges of the profile in this example is about 84 \AA . Using the refined GTM procedure, we divide the surface region into layers of thickness $D = 84, 42, 21, 10.5$ and 5.3 \AA respectively at various stages. The $q_{z,\max}$, within which data points are used for fitting, is determined as π/D for each value of D at each stage of the refined GTM and is indicated with vertical bars in Fig. 6(a). Related details of the GTM procedure are shown in Table I. The evolution of profiles, which are retrieved at successive stages

TABLE I. Data points for fitting at each stage in GTM.

stage	1	2	3	4	5
No. of Data points for fitting	5	16	39	86	179
thickness of divided layers	84 \AA	42 \AA	21 \AA	10.5 \AA	5.3 \AA
$q_{z,\max}$	0.0375 \AA^{-1}	0.075 \AA^{-1}	0.15 \AA^{-1}	0.3 \AA^{-1}	0.6 \AA^{-1}

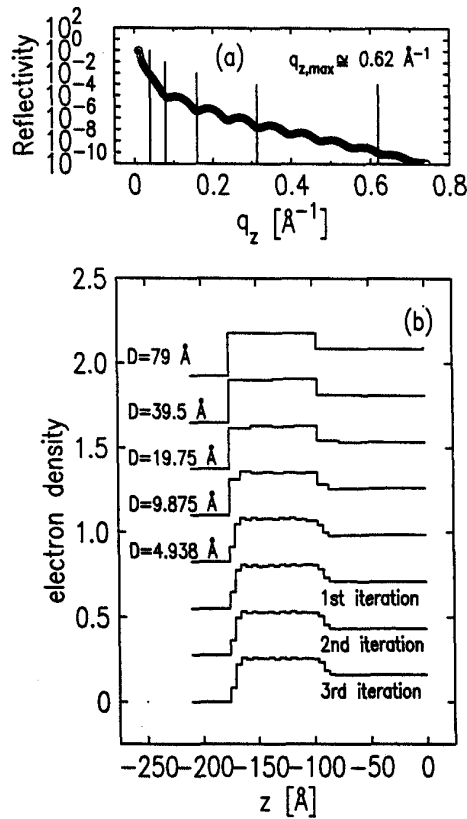


FIG. 3. (a) Vertical bars indicate the boundary within which reflectivity data points, shown in Fig. 1(b), are taken into consideration in fitting at various stages of the refined GTM. (b) Profiles obtained at various stages of GTM with thickness of divided layers $D = 79 \text{\AA}$, 39.5\AA , 19.75\AA , 9.875\AA , and 4.938\AA . With profiles obtained after the first, second iteration and third iterations of GTM.

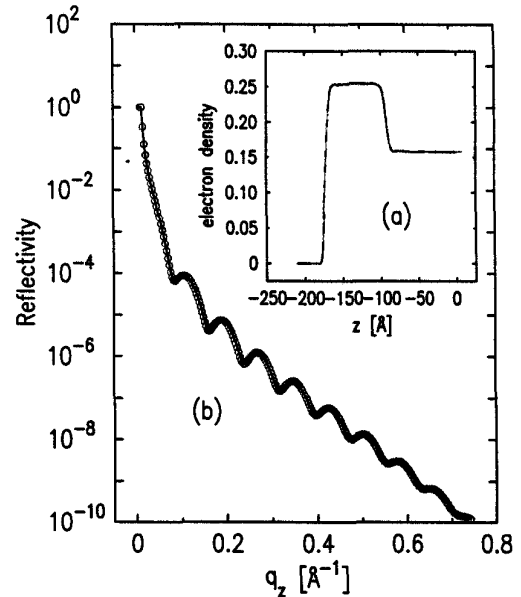


FIG. 4. (a) The resultant profile (dashed curve), retrieved from SGTM, compared with the original profile (solid curve). (b) Reflectivity data, shown in Fig. 1(b), compared with fitted reflectivity of the resultant SGTM profile in (a).

of the GTM and refined on operating the following GTM iteration, is illustrated in Fig. 6(b). In this case, we iterate the GTM procedure ten times to minimize the cost function between simulated and fitted reflectivity data. The resultant profile, denoted as “tenth iteration” in Fig. 6(b), is constructed, and, still being step-like, is smoothed. The SGTM is used to retrieve the resultant profile, shown in Fig. 7(a), while the smoothed profile is re-divided into layers of thickness $\pi/(0.79 \text{\AA}^{-1})$.

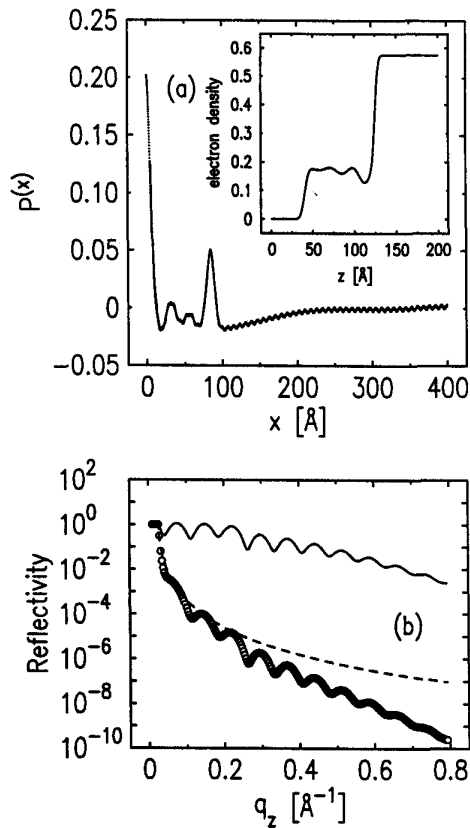


FIG. 5. (a) The function $P(x)$ obtained according to Eq. (21). The inset shows the original profile. (b) Simulated reflectivity data for the original profile in the inset of (a) along with Fresnel reflectivity $R_F(q_z)$ (dashed curve) and $R(q_z)/R_F(q_z)$ (solid curve).

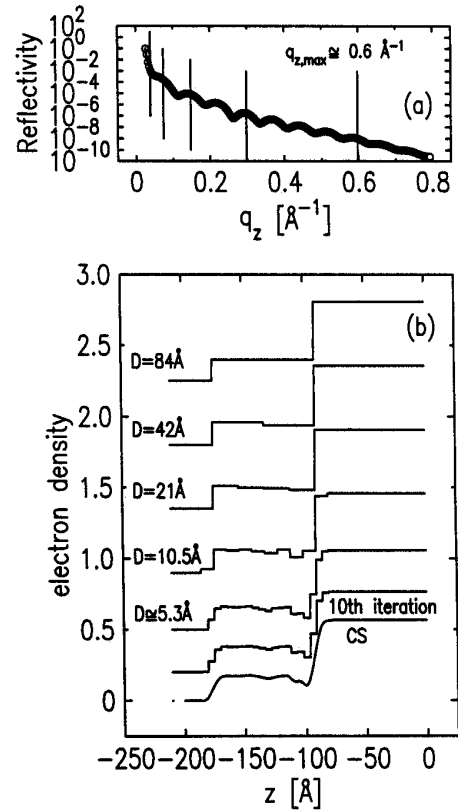


FIG. 6. (a) Vertical bars indicate a boundary within which data points are taken into consideration for fitting at various stages of GTM. (b) Profiles of electronic density, obtained at various stages of the GTM, with thickness of divided layers in a range 84 - 5.3 Å. The profile obtained after GTM is iterated ten times, denoted as "tenth iteration", and the smoothed profile. "CS" refers to a smoothing operation based on cubic-spline interpolation.

The retrieved profile reveals faithfully all features of the wetting-film region, whereas these are generally unknown in realistic cases. The reflectivity is fitted satisfactorily. Because we do not generate enough data points near the critical q_z to define precisely the critical value of q_z , the bulk density, related to this critical value, can not be determined precisely and differs slightly from that of the original profile. The minute discrepancy arises as there is a small bump in the retrieved profile appearing at -80 Å (see Fig. 7(a)). For this reason minor deviations of fitted

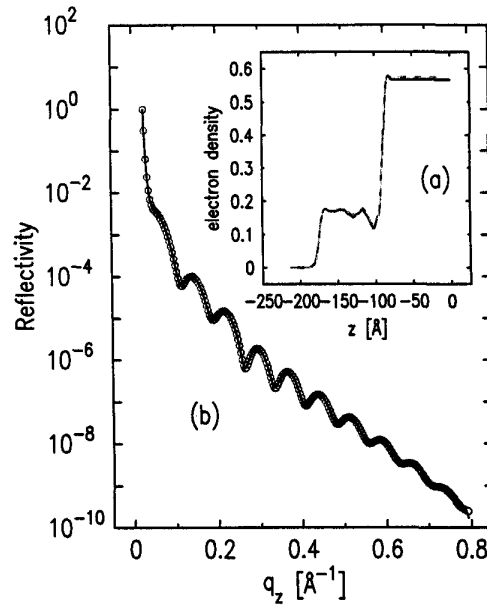


FIG. 7. (a) The resultant profile (dashed curve), obtained from SGTM, compared with the original profile (solid curve). (b) Simulated reflectivity data compared with fitted reflectivity of the resultant SGTM profile in (a).

reflectivity spectra from simulated data are discernible beyond $q_z \approx 0.7 \text{ \AA}^{-1}$ in Fig. 7(b). In an experiment, if good data near the critical q_z can be obtained, the density of data points in this region becomes sufficiently increased to determine precisely the accurate value of the critical q_z and then to determine precisely the value of the bulk density. In cases in which data near the critical q_z are poor, the bulk (substrate) density should be determined in advance. The electron density of a substrate is generally known before preparation of the wetting samples.

IV. Conclusion

A new scheme for smoothed groove tracking is applicable to reflectivity data for samples for which profiles of electronic density are not completely smooth, whereas previous methods serve for reconstruction of only smooth profiles; in this way applications of SGTM are expanded. This scheme consists of a preliminary process, based on Fourier transformation, a refined GTM and the SGTM. In outline of the method, the surface region of the sample in question is divided into layers according to information on positions of sharp edges of the probed profile, which is obtained from the preliminary process. Then a refined GTM is used to retrieve efficiently the step-like profile of electronic density revealing basic features of sharp edges of the probed profile. Finally the SGTM is employed to reconstruct the resultant profile resembling the true profile and exhibiting details of minute features of the profile. The SGTM complemented with the preliminary process is applicable to simulated reflectivity data of samples as a thin film or a wetting film with little or no prior information.

A relation between thickness D of divided layers and $q_{z,\text{max}}$ over which data points are taken into consideration for fitting is $q_{z,\text{max}} = \pi/D$. This rule must be rigorously satisfied whether in the refined GTM with the preliminary process or in the SGTM. To violate the rule might cause a serious problem of a local minimum or make fitting futile.

In realistic cases measurement of reflectivity data near the critical q_z , at which total external reflectance occurs, can be difficult, so that it becomes impossible to determine the bulk density based on experimental data, but the electronic density of a bulk or substrate is typically well known. Any accessory information, which is necessary and reliable for preparation of a sample or is provided by related theoretical work, is helpful in implementing efficiently the new scheme and in avoiding non-uniqueness of results. In this work using the new scheme with the refined GTM, we obtain quickly a step-like profile that resembles the real profile and serves as an estimated profile for the successive SGTm. A profile for the probed sample obtained by any other method dependent or independent of a model can serve as that estimated profile for the SGTm. During the iterative procedure of the SGTm, an estimated profile becomes further refined to approach the real profile in question as the computed reflectivity agrees satisfactorily with experimental or simulated data.

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References

- [1] S. Dietrich and A. Haase, *Phys. Reports* **260**, 1 (1995).
- [2] Proceedings of the 15th international conference on surface xray and neutron scattering, *Physica* **B248** (1998).
- [3] P. Mikulik and T. Baumbach, *Phys. Rev.* **B59**, 7632 (1999).
- [4] R. Lipperheide, *et al.*, *Physica* **B221**, 514 (1996).
- [5] N. F. Berk and C. F. Majkrzak, *Phys. Rev.* **B51**, 11296 (1995).
- [6] G. Reiss and R. Lipperheide, *Phys. Rev.* **B53**, 8157 (1996).
- [7] P. S. Pershan, *Phys. Rev.* **E50**, 2369 (1994).
- [8] V.-O. De Haan, *et al.*, *Physica* **B221**, 524 (1996).
- [9] V.-O. De Haan, *et al.*, *Phys. Rev.* **B52**, 10831 (1995).
- [10] M. V. Klivanov and P. E. Sacks, *J. Math. Phys.* **33**, 3813 (1992).
- [11] W. J. Clinton, *Phys. Rev.* **B48**, 1 (1993).
- [12] D. S. Sivia, W. A. Hamilton and G. S. Smith, *Physica* **B173**, 121 (1991).
- [13] C. H. Chou, *Physica* **B233**, 130 (1997).
- [14] X.-L. Zhou and S. H. Chen, *Phys. Rev.* **E147**, 121 (1993).
- [15] X.-L. Zhou and S. H. Chen, *Phys. Reports* **257**, p. 310-322 (1995).
- [16] X.-L. Zhou and S. H. Chen, *Phys. Rev.* **E47**, 3174 (1993).
- [17] C. H. Chou, *et al.*, *Phys. Rev.* **E55**, 7212 (1997).
- [18] C. H. Chou, *Solid State Comm.*, Vol **108**, No. 9, 687 (1998).
- [19] G. Parratt, *Phys Rev.* **95**, 359 (1954).
- [20] A. K. Doerr, *et al.*, *Physica* **B248**, 263 (1998).