Reflectivity and Roughness of Layered Synthetic Microstructures

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Abstract

The effect of uncorrelated roughness on the reflectivity of layered synthetic microstructures (LSM's) is studied. A computer code which includes uncorrelated roughness is developed allowing the subtraction of roughness from experimental measurements. This allows the use of LSM's to measure the optical constants of materials in the x-ray region of the spectrum. The method is applied to an V/C LSM to measure the resonance correction to the atomic factor, f', across the vanadium K-absorption edge.

Introduction

The understanding and control of layer roughness in Layered Synthetic Microstructures (LSM's) appears to be a key in their further development and use. Layer roughness has been treated in the visible by several authors.[1,2,3,4] Rosenbluth and Forsyth have extended this treatment into the x-ray regime.[5] Two cases can be handled with relative ease, i.e. correlated roughness and uncorrelated roughness.

Correlated roughness implies that each layer is an exact copy of the roughness of the substrate. In contrast the layer roughness between successive layers is completely random in the uncorrelated case. The rms roughness between layers may be the same for each layer, but there is no correlation from layer to layer.

We will consider the case of uncorrelated roughness which should apply near grazing incidence. At grazing incidence, even if the substrate roughness is reproduced in the vertical direction, the radiation will see uncorrelated roughness because it covers a large distance in the lateral direction between each layer. Figure 1 illustrates the path of the radiation through the structure. We have used a model of uncorrelated roughness to modify a computer code developed by Underwood[6] to give the effects of uncorrelated roughness on multilayer reflectivity.

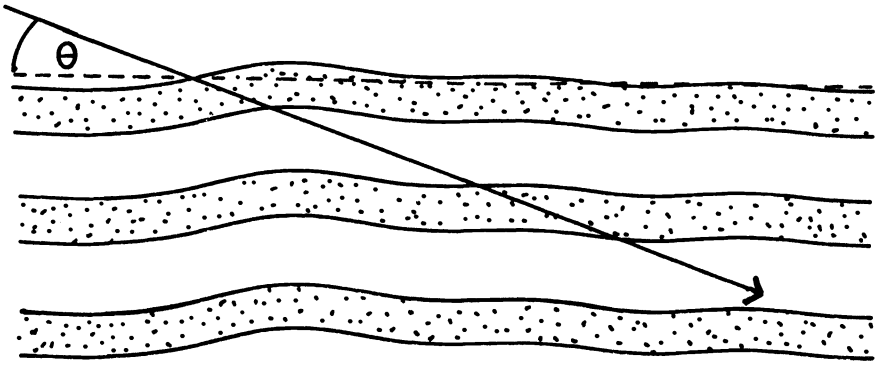


Figure 1. X-Rays incident on a rough LSM near the glancing angle. The long transverse path ensures that the radiation will encounter uncorrelated roughness patterns.

Since the heights of the x-ray reflection peaks are sensitive to the index of refraction, experimental measurements can be used to determine optical constants. We will illustrate this application of LSM's by analyzing data taken with a vanadium-carbon LSM to experimentally determine the dispersion corrections of the form factor for vanadium near the vanadium K-absorption edge.

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Reflection from Rough Layered Synthetic Microstructures

The reflectivity of an ideal LSM can be calculated using a recursion formula first derived by Parratt[7], modified and coded for a computer by Underwood and Barbee.[8]

$$R_{i-1,i} = a_{i-1}^4 \frac{R_{i,i+1} + F_{i-1,i}}{R_{i,i+1} F_{i-1,i} + 1} \tag{1}$$

$$\text{where} \quad R_{i,i+1} = a_i^2 \frac{E_i^R}{E_i} \tag{2}$$

E_i, E_i^R are the electric field values midway through the layer i. The superscript R indicates the reflected field.

$$a_i = \exp\left(-ik_1 g_i \frac{d_i}{2}\right) \tag{3}$$

$$\text{where } g_i \equiv (n_i^2 - \cos^2\theta)^{1/2} \tag{4}$$

$n_i = 1 - \delta_i - i\beta_i$, is the complex refractive index of layer i. (5)

θ is the glancing angle of incidence. k_i is the propagation constant for the layer i, and d_i is the layer thickness. For σ polarization

$$F_{i-1,i} = \frac{g_{i-1} - g_i}{g_{i-1} + g_i} \tag{6}$$

and for π polarization

$$F_{i-1,i} = \frac{\frac{g_{i-1}}{n_{i-1}^2} - \frac{g_i}{n_i^2}}{\frac{g_{i-1}}{n_{i-1}^2} + \frac{g_i}{n_i^2}} \tag{7}$$

The theory of electromagnetic scattering from rough surfaces indicates that the reflected radiation from the rough surface is equal to the reflected radiation from a smooth surface multiplied by an exponential term containing the rms roughness of the surface.[9]

$$\exp\left(-\frac{1}{2}\left(\frac{4\pi\ \sigma \sin\theta}{\lambda}\right)^2\right) \tag{8}$$

E_{i-1}^R is the field halfway through the layer so the intensity is decreased by a factor

$\exp\left(-\frac{1}{2}\left(\frac{4\pi\sigma_{i-1}}{\lambda} \sin\theta\right)^2\right)$. Thus the ideal reflectivity at that point

is $E_{i-1}^R \exp\left(\frac{1}{2}\left(\frac{4\pi\sigma_{i-1}}{\lambda} \sin\theta\right)^2\right)$.

The effect of uncorrelated roughness can be calculated by incorporating this correction into the reflectivity recursion relation given by equation (1).

$$R_{i-1,i} = a_{i-1}^4 \exp\left(-\frac{1}{2}\Sigma_{i-1}^2\right) \frac{R_{i,i+1}\exp\left(-\frac{1}{2}\Sigma_i^2\right) + F_{i-1,i}}{R_{i,i+1}\exp\left(-\frac{1}{2}\Sigma_i^2\right) F_{i-1,i} + 1} \tag{9}$$

where $$\Sigma_i = \frac{4\pi\sigma_i}{\lambda} \sin\theta$$

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If we assume that σ_i is the same for each layer (9) becomes

$$R_{i-1,i} = a_{i-1}^4 \, e^{-\frac{8\pi^2\sigma^2\sin^2\theta}{\lambda^2}} \; \frac{R_{i,i+1}e^{-\frac{8\pi^2\sigma^2\sin^2\theta}{\lambda^2}} + F_{i-1,i}}{R_{i,i+1}e^{-\frac{8\pi^2\sigma^2\sin^2\theta}{\lambda^2}} F_{i-1,i} + 1} \tag{10}$$

Calculations including roughness can be made by modifying the Underwood code as indicated by the recursion relation (9).

Figure 2 shows a calculation of reflectivity for a tungsten-carbon LSM around the first order resonance including the effect of uncorrelated roughness. The peak reflectivity is near 90%.

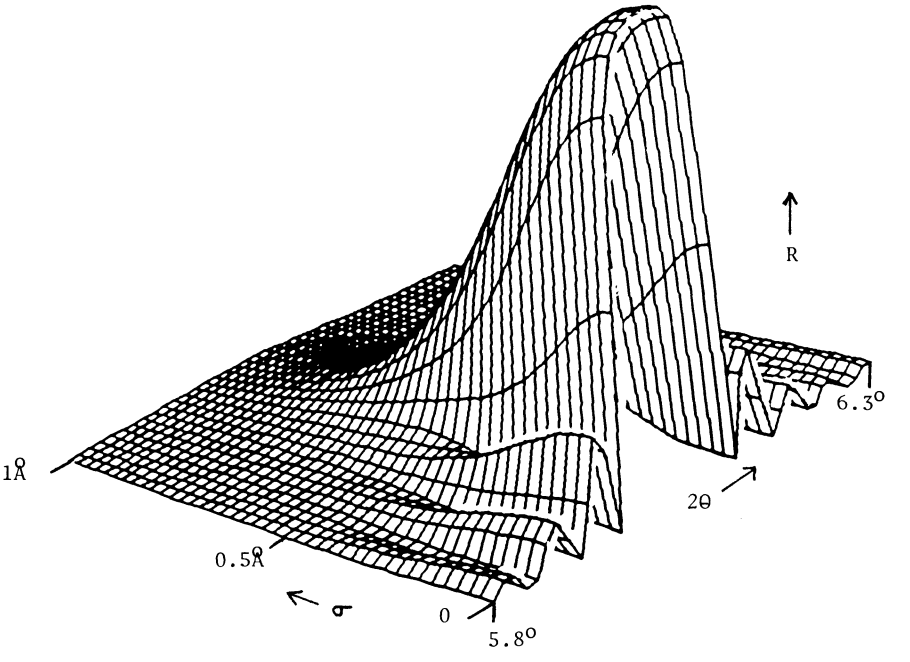


Figure 2. The calculated reflectivity of a W/C LSM. R vs. θ vs. σ.

The Determination of the Resonance Correction to the Scattering Factor

The atomic scattering factor is given by[10]

$$f = Z + f' + if'' \tag{11}$$

where f′ is called the resonance correction, and f′′ the absorption correction to the atomic scattering factor. The real and imaginary parts of the index of refraction, δ and β respectively, can be written:

$$\delta = \frac{r_e \lambda^2 N}{2\pi} (Z + f') \tag{12}$$

$$\beta = \frac{r_e \lambda^2 N}{2\pi} f'' \tag{13}$$

Inclusion of refraction effects gives a modified Bragg equation.

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$$m\lambda = 2d \sin\theta \left(1 - \frac{\delta}{\sin^2\theta} \right) \qquad (14)$$

The calculated reflection intensity depends on f′ through δ, f′′ through β, d, the layer pair spacing, σ, the roughness parameter and γ, the ratio of the thickness of material 1 to d. γ enters the calculation because the LSM δ is given by

$$\delta = \delta_1\gamma + \delta_2(1 - \gamma)$$

(Subscripts 1 and 2 refer to the two LSM materials.) This implies that one of these parameters can be measured experimentally if the other four parameters are known and the calculated value is compared with an experimental value of reflected intensity. In particular the resonance correction to the atomic scattering factor, f′, can be measured (even near the absorption-edge) for one of the LSM materials. The parameters d and δ are determined by experimentally fitting them to equation (14). The parameters γ and σ and then determined far from the absorption edge so that accurate values for the relevant optical constants can be calculated. δ, β, and the number density are calculated for each of the materials in the LSM. Now γ and σ are used as variable parameters in the reflectivity code. These two parameters are adjusted to fit the amplitudes of the resonant peaks to as many orders as are statistically significant. γ and σ modulate the reflected peak amplitudes in different ways. γ modulates the resonant peaks much like frequency or phase modulation effects resonant signals in radio frequency, i.e. any order can be attenuated or even eliminated depending on the value of the modulating parameter. σ on the other hand has a greater effect on the higher order peaks.

It is also necessary to determine f′′, in the region of interest by some independent method. Once the absorption measurement has been made and d, γ, σ have been determined, the resonance correction can be used as a variable parameter in the code to fit the measured amplitudes at the resonant peaks for each energy.

Experimental Measurement of f′ for Vanadium Near the K-Absorption Edge

A Vanadium-Carbon LSM was used to illustrate this measurement method. The sample (80-177) used had twenty-one 45 Å thick vanadium layers separated by 100 Å carbon layers deposited onto a (100) orientation Si single crystal substrate. The reflectivity measurements taken at the SSRL Synchrotron and reported earlier by Barbee[11] were used to determine f′. Figure 3 is a schematic diagram of the Vanadium-Carbon sample (80-177). Figure 4 is a typical experimental measurement at 5.3Kev.

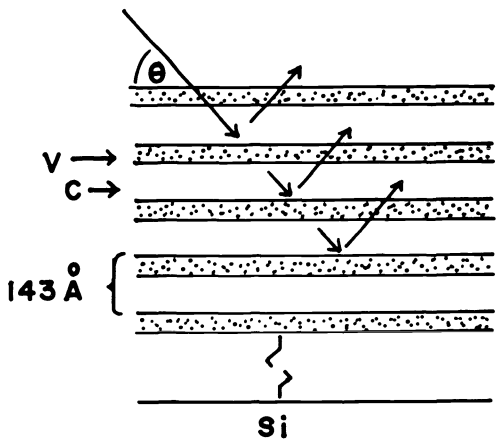


Figure 3. Schematic diagram of the V/C LSM sample (80-177).

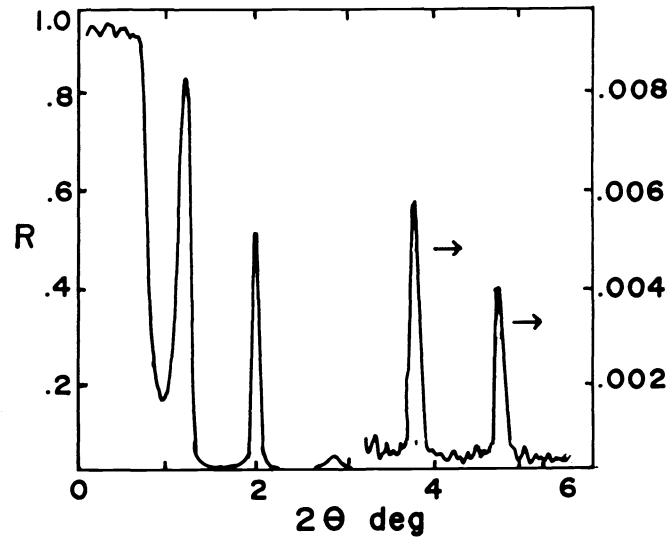


Figure 4. An experimental run at E = 5.3 Kev for sample (80-177).

For the V/C LSM $\delta = \delta_v \times \gamma + \delta_c \times (1 - \gamma)$. (15)

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δ_v for vanadium and δ_c for carbon are computed using a code developed by Cromer and Liberman.[12,13] The initial d and γ are estimated from fabrication data as 145.3Å for d and γ = .3.[11] d and δ are now determined by fitting these parameters to equation (14) by a least squares minimization technique.[14,15] The 2nd-order through 5th-order peaks on 12 runs were used in the minimization procedure. This process was also used to get a better value for the offset angle, θ_o. The results are summarized in Table I.

TABLE I.

Values of layer thickness, d; average delta, δ; and 2θ offset determined by least squares minimization technique.

Run number	X ray (keV)	d (in A)	δ ($\times 10^5$)	2θ offset (degrees)
VC177.007	5.300	142.77	2.130	.00875
VC177.014	5.444	143.37	1.906	.0230
VC177.008	5.455	142.90	1.926	.00899
VC177.009	5.465	143.07	1.875	.00941
VC177.015	5.469	143.44	1.750	.0231
VC177.016	5.472	143.54	1.769	.0232
VC177.010	5.475	143.21	1.820	.00962
VC177.017	5.478	143.09	1.785	.0234
VC177.011	5.485	143.24	1.854	.00965
VC177.012	5.495	144.26	1.847	.00900
VC177.013	5.650	143.83	1.808	.0108
VC177.020	6.000	143.29	1.539	.00866

γ and σ were determined by fitting them to the ratios of the 2nd-order through 5th-order intensity peaks to the 1st-order peak intensity. The optimization converged to σ = 1.7Å and γ = .295.

The absorption correction, f'' was determined across the vanadium K-absorption edge by using a thin slab of vanadium in an EXAFS run. The resonance correction, f', can now be determined by calculating the ratio I_2/I_1 as a function of f' for each value of energy across the absorption edge. The experimental value of f' is found by matching the measured ratio with the computed curve. Figure 5 is a theoretical curve of I_2/I_1 vs. f' for E = 5.495 KeV. Table II summarizes the experimental values of f' across the vanadium absorption edge. Figure 6 compares the experimental values of f' against energy with a theoretical curve computed with the Cromer-Liberman code. The discrepancy between the two curves can be explained by the fact that the code uses single atom wavefunctions. The actual wavefunction is altered by the neighboring atoms.

TABLE II.

Results.

I_{2/I_1} for the different runs and the respective f' found by table V and I_{2/I_1}.

X-Ray energy (± 1 eV)	I_{2/I_1}	f'
5.444 keV	.513 ± .011	-2.76 ± .31
5.455 keV	.429 ± .009	-4.90 ± .22
5.465 keV	.319 ± .007	-6.64 ± .19
5.469 keV	.315 ± .007	-6.41 ± .21
5.472 keV	.293 ± .007	-6.74 ± .21
5.475 keV	.254 ± .006	-8.12 ± .24
5.478 keV	.252 ± .006	-7.86 ± .28
5.485 keV	.276 ± .007	-6.88 ± .30
5.495 keV	.297 ± .007	-5.96 ± .30

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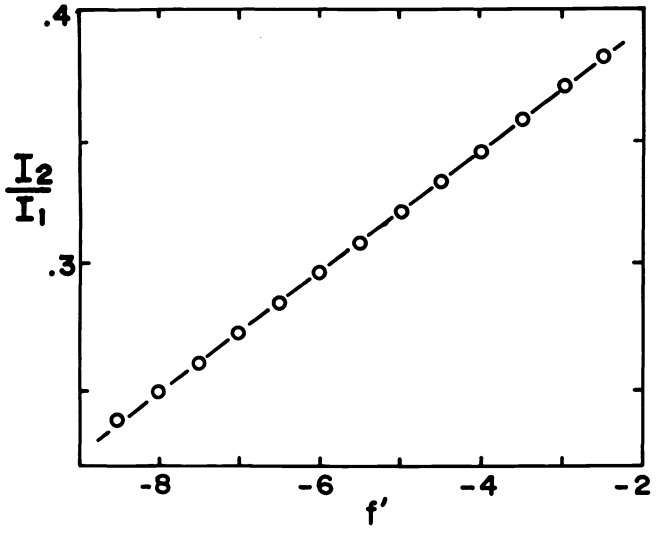


Figure 5. A computed curve of I_2/I_1 vs. f′ for E = 5.495 Kev.

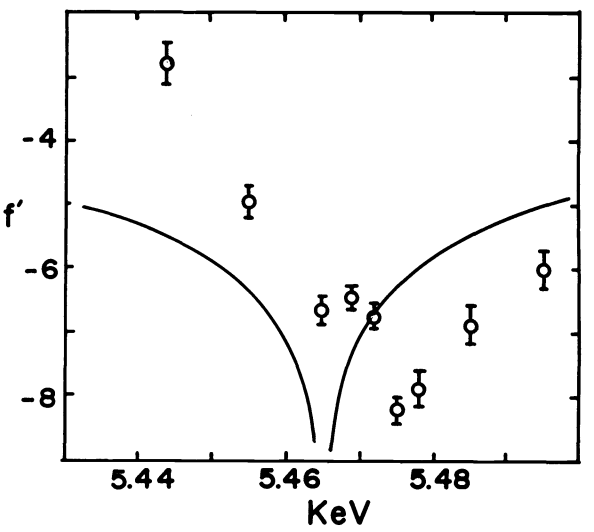


Figure 6. A comparison between experimental values of f′ vs. energy and a theoretical curve computed with the Cromer-Liberman code.

Conclusions

This work demonstrates that a simple model of uncorrelated roughness in LSM's can predict the effect of roughness on reflectivity as evidenced by the consistency of the optical constant measurements. It also appears that LSM's may make possible precise measurements of optical constants over a broad range of wavelengths. In particular, we have obtained f′ for vanadium to approximately ± 0.5 with the prospect of improvement to the ± 0.05 level attained by the Bonse-Hart method.[16] What is more, it appears that optical constants can be obtained even where absorbance is high because LSM's can be made with extremely thin layers of the absorbing material.

Acknowledgements

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References

1. Eastman, J.M., Physics of Thin Films, 10, 167 (1978).
2. Verhaeghe, M.F., Optica Acta, 17, 843 (1970).
3. Carniglia, C.K. Opt. Eng., 18, 104 (1979).
4. Elson, J.M., et al, in Applied Optics and Optical Engineering, edited by R.R. Shannon and J.C. Wynat (Acad. Press, N.Y., 1979), Vol. VII, pp. 198,199.
5. Rosenbluth, A. and Forsyth, J.M., Proceedings of AIP Topical Conference on Low Energy X-Ray Diagnostics, AIP Conference Proceedings No. 75, pg. 280 (1981).
6. Underwood, J.H. and Barbee, T.W., Jr., Proceedings of AIP Topical Conference on Low Energy X-Ray Diagnostics, AIP Conference Proceedings No. 75, pg. 170 (1981).
7. Parratt, L.G., Phys. Rev., 95, 359 (1954).
8. Underwood, J.H. and Barbee, T.W., Jr., J. Applied Optics, 20, 3027 (1981).
9. Beckmann, P. and Spizzichino, A., The Scattering of Electromagnetic Waves from Rough Surfaces. (Pergamon, New York, 1963).
10. James, R.W. The Optical Principles of the Diffraction of X-Rays, (G. Bell and Sons, Ltd., London, 1958).
11. Barbee, T.W., Jr., Proceedings of AIP Topical Conference on Low Energy X-Ray Diagnostics, AIP Conference Proceedings No. 75, pg. 131 (1981).
12. Cromer, D.T. and Liberman, D., Acta Cryst., A37, 267 (1981).
13. Cromer, D.T. and Liberman, D., Los Alamos Scientific Laboratory Report. LA-4403 (1970).
14. Davidon, W.C., Argonne National Laboratory Report No. ANL 590 (Rev.), 1959 (unpublished).
15. Fletcher, R. and Powell, M.J.D., Comp. Journal, 6, 163 (1963).
16. Cusatis, C. and Hart, M., Proc. R. Soc. Lond. A, 354, 291 (1977).

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