ANL/MSD/CP--85268
Conf-941144--79

Reflection of Neutrons from an Optical Grating

Alan E. Munter^{1,2}, S. Adenwalla¹, G. F. Felcher¹, X. L. Zhou³

¹Materials Science Division
Argonne National Laboratory
Argonne, IL 60439

²Department of Nuclear Engineering
103 S. Goodwin Avenue
University of Illinois
Urbana, IL 61801

³Department of Nuclear Engineering
Massachusetts Institute of Technology
24-215A, 77 Mass Avenue
Cambridge, MA 02139

RECEIVED

FEB 17 1995

OSTI

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

December, 1994

DISCLAIMER

/sm

Distribution:

- 1-2. M. J. Masek
3. B. D. Dunlap
4. G. W. Crabtree
5. J. D. Jorgensen
6. B. S. Brown
7. Editorial Office
8. Authors

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

This work is supported by the Division of Materials Sciences, Office of Basic Energy Sciences of DOE, under contract No. W-31-109-ENG-38.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ALAN E. MUNTER^{*#}, S.ADENWALLA^{*}, G.P.FELCHER^{*}, AND X.L.ZHOU[†]^{*}Argonne National Lab, MSD-223, Argonne, IL 60439[#]Department of Nuclear Engineering, 103 S. Goodwin Ave, University of Illinois, Urbana, IL 61801[†]Department of Nuclear Engineering, Massachusetts Institute of Technology, 24-215A, 77 Mass Ave, Cambridge, MA 02139

ABSTRACT

Specular and off-specular neutron intensities were measured at grazing incidence from an optical grating. The periodicity of the grating was determined from the locations of the intensity peaks in the off-specular data. An analysis of the widths of the off-specular intensity peaks showed that the effect of the coherence length of the neutron beam on the determination of a surface length scale $\sim 30 \mu\text{m}$ was negligible compared to that of the instrument resolution.

INTRODUCTION

Rough surfaces and interfaces exist in many material systems. For instance, intestinal lining is a biological membrane with a regular surface structure. The surface consists of a series of microvilli which are about $1.5 \mu\text{m}$ wide. These microvilli serve to increase the surface area of the intestinal wall for efficiently drawing nutrients out of the food. For another example, when symmetric diblock copolymers (for instance, of polystyrene and polymethylmetacrylate) self-organize in a lamellar structure with layers several hundred $\text{\AA}$ thick, an incomplete layer of the lamella forms a surface with roughness on the order of the layer thickness which is correlated over a distance of a few microns¹. Both of these examples have a laterally correlated surface structure in the micron range. The study of such irregularities of an interface may be difficult to accomplish by direct microscopy if the interface is buried deep under a surface. Neutron reflectivity is a potentially useful probe for such organic systems since slow neutrons do not cause radiation damage to organic materials, and selective deuteration provides excellent contrast between the components². However, off-specular neutron reflectivity is not well developed for characterization of such systems. Simple theoretical models to relate the roughness structure to the measured data would be particularly useful. One of the main difficulties consists of the lack of the experimental capabilities for measuring off-specular reflectivity over sufficient dynamic ranges and with sufficient resolution. In this regard, an effort has been made to compare the main features in the off-specular data and the main features in the real-space structures over large length scales. More specifically, an optical diffraction grating was selected to investigate the correspondence of the grating structures to the data structures.

MEASUREMENTS

A commercial diffraction grating was obtained for the experiment³. Its surface is $50 \times 50 \text{ mm}^2$, with 1200 lines/mm, or 8333 $\text{\AA}$ spacing from peak to peak. The grooves were printed on epoxy from a master grating surface of fused silica, and the epoxy is then coated with aluminum in order to reflect light. The grating has a blaze angle of $8^\circ 37'$ so that the surface of the grating has a sawtooth structure $\sim 1200 \text{ \AA}$ high.

Specular and off-specular intensities from this grating were measured at the POSY II reflectometer⁴ at the Intense Pulsed Neutron Source at Argonne National Lab. The POSY II

reflectometer utilizes a white beam of neutrons with wavelengths ranging from $2\text{\AA}$ to $16\text{\AA}$ emitted by a cold moderator consisting of solid methane at 20K . The scattered intensities are recorded as a function of time of flight by a one-dimensional position sensitive detector which measures both specular and off-specular neutrons within the incidence plane with an angular resolution of $\Delta\theta \sim 0.02^\circ$. At POSY II the detector accepts neutrons scattered out of the incident plane by $\Delta\varphi$ up to 0.8° , and all neutrons in this range are summed and treated as reflected neutrons within the reflection plane.

Figure 1: Geometry of the experiment

The geometry of the experiment is shown in Fig. 1. The incident beam makes an angle $\theta_i = 0.6$ and 0.45 with the grating surface for the two measurements shown in Fig. 2a and 2b respectively. The grooves on the grating subtend an angle α from the plane of incidence. As the angle α varies around the surface normal, the neutron beam sees roughness over different length scales in the forward direction. With this arrangement, the reflection is not completely in the incident plane. When the grating is rotated, the roughness is not only correlated in one dimension, it has a y component as well as x . However, in the experimental conditions we chose, φ , the angle of the scattered beam with the reflection plane is always less than 0.65° , well below the acceptance angle of the counter, and such that we may consider negligible the distortion from out-of-plane scattering.

Specular and off-specular intensities were measured at two values of α : $\alpha = 2.9^\circ$ and 1.6° as plotted in Fig. 2a and 2b respectively. In Fig. 2, the horizontal, q_x , and vertical, q_z , axes are defined as the momentum transfer in the forward direction and the perpendicular direction respectively. The range of q_x and q_z correspond to a range of $\theta_f \approx 0^\circ - 1.5^\circ$ and $\lambda \approx 2 - 16 \text{\AA}$.

The momentum transfer is related to the wavelength and angle of the reflected neutrons as follows:

$$\begin{aligned} q_x &= \frac{2\pi}{\lambda} (\cos\theta_f - \cos\theta_i) \\ q_z &= \frac{2\pi}{\lambda} (\sin\theta_f + \sin\theta_i) \end{aligned} \quad (1)$$

Figure 2a

Figure 2b

Intensity contours of the neutrons scattered from an optical grating for different grating spacings. The contour plots show the natural log of the intensities with levels at integer values of log intensity.

ANALYSIS

The off-specular data for both measurements clearly show localized intensity peaks separated in the q -space. Since the sample has a coherent and periodic surface structure, the positions of the off-specular peaks can be treated under conventional diffraction theory. The peak positions are given by

$$\theta_n = \cos^{-1} \left(\cos \theta_i + \frac{n\lambda}{d_x} \right) \quad (2)$$

where n is the order of the diffracted peak.

Fig 2a shows solid lines which represent separations of $3.75 \times 10^{-5} \text{Å}^{-1}$ between peaks, and Fig 2b, separations of $2.1 \times 10^{-5} \text{Å}^{-1}$. These can be directly related to the periodicity of the surface by Eq. 2 as

$$d_x = \frac{2\pi}{\Delta q_x} \quad (3)$$

to yield periods of 16.7 microns from Fig. 2a, and 30 microns from Fig 2b. The angle of rotation around the surface normal, α , can also be found by $\alpha = \sin^{-1}(8333 \text{Å}/d_x)$.

When very long length scales like this are measured in the plane of the surface, there is the question of whether or not the neutron beam is coherent over the whole distance or not and what relationship the coherence length has with the resolution function of the instrument⁵. It is evident from the contour plots in Fig. 2 that the off-specular, $n \neq 0$, peaks are broader than the specular, $n=0$, ones are. At least three things can influence this broadening. First, if the beam is incident on the sample at an angle θ_1 with an inherent width of Δx_1 , the width of the outgoing beam, neglecting any broadening properties the surface may have, is

$$\Delta x_2 = \frac{\Delta x_1 \sin \theta_2}{\sin \theta_1} \quad (4)$$

From this equation one can see that for outgoing angles greater than the incoming angle (negative order diffraction peaks) the beam width will be slightly wider than the incident beam width. Secondly, since $\Delta q_x = -\sin(\theta_f) \Delta \theta_f 2\pi/\lambda$, as θ_f increases the width in q_x also increases giving an additional appearance of broadening when the data is plotted in reciprocal space as is

done in Fig 2. A third contribution to peak broadening could be the coherence length of the neutron beam. The upper bound for the longitudinal coherence length of a neutron beam at a pulsed source is the distance traveled by the neutrons in the width of one pulse. This distance is on the order of a few centimeters for a moderator at 20 K and a pulse length of a hundred μ s. The transverse coherence length is limited due to the fact that the neutron beam is divergent and not a perfect plane wave. The actual longitudinal coherence length will be less than this due to the fact that the neutrons do not have uniform wave fronts like a laser. MRS

When the widths of the off-specular peaks are measured and compared to that of the specular, the broadening that is observed can be completely accounted for by the first two geometrical and resolution effects. If the incoherence of the neutron beam is having any effect on peak broadening, it can not be determined at this resolution. When calculations are performed under conventional diffraction theory for the width of the respective orders it is apparent that we can safely assume that the neutron beam has a longitudinal coherence of at least 60 μ m and probably much greater distances as well at our present resolution.

The determination of the periodicity of an optical grating from off-specular intensity was demonstrated. An assessment was made of the effect of the neutron beam coherence length on the surface spatial resolution by analyzing the widths of off-specular intensity peaks. A more detailed analysis of the off-specular intensity data for more detailed roughness information is in progress. From such an analysis, the relationship between the height and shape of surface roughness and the off-specular intensity data will be determined. The influence of the detector resolution function upon the inverse problem of surface structure determination from off-specular data will be assessed.

Although the structure of an optical grating used in this paper is simple, it has served to illustrate the simple relationship between the off-specular data structure and the real-space properties. For more general rough systems, a practical theoretical model will be needed to relate the off-specular data to the real-space roughness structure so that the diffuse data can be analyzed. Owing to the intrinsic complexity of diffuse scattering, the development of such a theoretical model will be a significant technical challenge, and its solution will constitute a major breakthrough in the development of the diffuse reflectivity technique for rough surface/interface characterization.

A.E.M. acknowledges support from Argonne National Lab, Department of Educational Programs and the University of Illinois Research Board. Work at Argonne was supported by the U.S. Department of Energy, Basic Energy Sciences, under Contract No. W-31-109-ENG-38. Work by XLZ was supported by the Massachusetts Institute of Technology Sloan Funds.

References

1. G.P. Felcher, R.J. Goyette, S. Anastasiadis, T.P. Russell, M. Foster, and F. Bates, Phys. Rev. B **50**, 9565 (1994).
2. G.P. Felcher, Neutron News **5**, 18 (1994).
3. Edmund Scientific catalog number N41,034
4. A. Karim, B.H. Arendt, R. Goyette, Y.Y. Huang, R. Kleb, and G.P. Felcher, Physica B **173**, 17 (1991).
5. S.K. Sinha, in Surface X-Ray and Neutron Scattering, edited by H. Zabel and I.K. Robinson (Springer Verlag, Berlin, 1991), p. 1.