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The Effect of Surface Roughness on the Phase of Neutrons Specularly Reflected at Grazing Incidence

by

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The effect of surface roughness on the specular intensity of a reflected neutron beam can be calculated using the distorted wave Born approximation. For Gaussian roughness the result obtained is the familiar expression of Nevot and Croce. When the distribution of surface heights, z , is proportional to $\cosh^{-2}(z/2d)$, the calculated reflectance is $\sinh[\pi d(k_0 - k_1)] / \sinh[\pi d(k_0 + k_1)]$. Here k_0 and k_1 are the components of neutron wavevectors perpendicular to the interface, evaluated outside and inside the reflecting medium, and d is a measure of the amplitude of the roughness. This result for the reflectance differs by a phase factor from that obtained in an exact calculation for an interface with the corresponding graded refractive index. The phase difference between the two expressions is small, scaling as $\sim d^3$, and usually has little effect on measured reflectivities of multilayered structures. Ways in which the phase could be determined are discussed.

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It is now widely accepted that the formulae derived by Nevot and Croce (NC) [1] describe the effect of surface roughness on the specular reflectivity of surfaces whose elevation, $h(x,y)$, above a plane has a Gaussian distribution when averaged over all points (x,y) of the surface. It was shown recently by one of us [2] that the NC result follows from the distorted wave Born approximation (DWBA) when the Fresnel wavefunctions are used in this perturbation theory. Thus, the NC result may be regarded as a perturbative estimate of the effect of roughness on specular reflectivity, valid only in limits where the amplitude of the roughness is small or where the refractive index contrast between the media separated by a rough interface is small.

The DWBA can be applied easily to distributions of $h(x,y)$ which are not Gaussian. In particular, for a distribution of the form $\cosh^{-2}(h/2d)$, the specular reflectance, R_r , is found to be:

$$R_r = \sinh[\pi d(k_1 - k_2)] / \sinh[\pi d(k_1 + k_2)] \quad (1)$$

where k_1 is the component of the neutron or x-ray wavevector perpendicular to the surface outside the reflecting medium and k_2 is the corresponding component within the medium. Evidently R_r tends to the usual Fresnel result as the width, d , of the rough interface tends to zero.

Figure 1: A comparison of graded-index interfaces generated by a Gaussian (dotted line) and $\cosh^{-2}(z)$ distribution of surface heights.

The $\cosh^{-2}$ height distribution corresponds to a graded interface for which the scattering length density, $\rho(z)$, varies perpendicular to the surface as

$$2 \rho(z) = (\rho_1 + \rho_2) - (\rho_1 - \rho_2) \tanh z/2d \quad (2)$$

The reflectance for a graded interface with this functional dependence on z can be calculated in closed form [3]. The result is

$$R_g = \exp[i\phi] R_r \quad (3)$$

where R_r is the function defined in equation (1) and the phase, ϕ , may be expressed as a quotient of gamma functions with complex arguments:

$$e^{i\phi} = - \frac{\Gamma(2iy_1) \Gamma(-i[y_1 + y_2]) \Gamma(-i[y_1 - y_2])}{\Gamma(-2iy_1) \Gamma(i[y_1 + y_2]) \Gamma(i[y_1 - y_2])} \quad (4)$$

where $y_1 = k_1 d$ and $y_2 = k_2 d$

According to Lekner [3], equation (4) may be simplified to yield

$$\phi = \frac{1}{2} \sum_{n=1}^{\infty} \arctan \left\{ \frac{2y_1}{n} \frac{y_1^2 - y_2^2}{n^2 + 3y_1^2 + y_2^2} \right\} \quad (5)$$

a result which shows that ϕ is third order in the interface thickness, d , and first order in the refractive index contrast between media 1 and 2.

It is gratifying that a perturbation calculation of the reflectance of a rough interface and an exact calculation for the corresponding graded interface give results which differ only by a term whose order is beyond those retained in the perturbation theory. Furthermore, since the difference between the two results is a phase factor, the predicted reflectivities (i.e. the squared moduli of the reflectances) of a rough surface with a $\cosh^{-2}$ height distribution and a corresponding graded-index interface are the same. Our view is that this agreement lends credence to results obtained when the DWBA is applied to rough interfaces.

Figure 2: Phase ϕ determined from equation (4) for an air/nickel interface plotted as a function of k for roughness parameters, d , of 10 Å (dotted curve) and 20 Å.

Using a tool such as *Mathematica* it is straightforward to evaluate equation (4). Figure 2 shows the result for an air-nickel interface for two values of the interface thickness, d . The variation of the phase with k_1 is easy to understand physically. For values of k_1 less than the critical value ($k_c = 0.011 \text{ Å}^{-1}$ for nickel), the apparent mean position of the interface

varies strongly with the energy of an incident neutron or x-ray because the radiation only samples a portion of the graded interface in which the potential energy is less than or comparable to the particle energy. Thus, the range of values of z sampled depends on the value of k_1 .

One may ask whether the phase given in equation (4) should be associated with the reflectance of a rough interface as well as with a graded-index interface. Our physical intuition is that it should. Supposing for a moment that this is true, one is led to enquire how the phase may be measured, and what effect it will have on results deduced from measurements of reflectivity.

The only way to observe a phase factor due to interfacial roughness is to make a measurement in which radiation reflected by the rough interface interferes with that reflected by a neighbouring interface. The reflectance of a film of thickness t deposited on a substrate is given by:

$$R_f = \frac{R_{01} + R_{12} e^{ik_1 t}}{1 + R_{01} R_{12} e^{ik_1 t}} \quad (6)$$

where R_{01} is the reflectance of the air/film interface and R_{12} is the reflectance of the film/substrate interface. Interference minima appear in the reflectivity of the film/substrate system when the numerator in equation (6) attains minimal values. Thus, whichever interface of the film is rough, any phase introduced by surface roughness will alter the apparent film thickness deduced from the position of an interference minimum in the reflectivity. In most cases, however, this effect will not be pronounced because the magnitude of the phase introduced by the roughness (c.f. Figure 2) is expected to be small. The most significant effect is obtained for thin films because the value of $k_1 t$ at the first observable interference minimum is small in this case. Since the phase due to roughness is expected to displace successive interference minima by smaller and smaller amounts, it should be possible to determine the phase. In practice, it is probably easier, as figure 3 shows, to choose a material which naturally forms a rough surface [4] and to overlay it with a material like titanium, whose negative coherent scattering length increases the values of both y_1 and $y_1^2 - y_2^2$ in the numerator of equation (5).

Figure 3: Logarithm of reflectivity plotted against k for a 350 Å layer of titanium deposited on a rough SiO_x surface with a roughness parameter, d , of 40 Å. The first two interference minima are shown. The dotted curve is the reflectivity calculated ignoring ϕ while the solid curve includes the phase calculated from equation (4).

One might hope that a more sensitive experiment for the observation of the phase change caused by roughness could be devised by making use of a multilayer device. However, it turns out that the phase introduced by the first interface in a bilayer is to a large extent cancelled by that which results from the second interface. For this reason, the wavevectors of Bragg peaks obtained with a bilayer stack are insensitive to surface roughness if each of the interfaces is rough.

In this note we have tacitly assumed that the effect of surface roughness on the phase of the specular reflectance of an interface is identical to the effect of a corresponding uniform gradation of the scattering length density. This point should be investigated experimentally. If our assumption is correct, it may be necessary to account for this phase in the analysis of some reflectivity data. However, the numerical estimates we have made

to date indicate that for most problems the phase is unlikely to significantly effect the majority of conclusions drawn from measurements of specular reflectivity.

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References

- [1] L. Nevot and P Croce, Rev. de Phys. Appl., 15, 761 (1980)
- [2] R. Pynn, Phys. Rev (to be published)
- [3] J. Lekner, (1987). *Theory of the Reflection of Electromagnetic and Particle Waves*, Martinez Nijof Publishers, Dordrecht, Holland.
- [4] H. A. van Sprang and R. G. Aartsen, J. Appl. Phys., 56, 251 (1984)

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