# THE EFFECT OF INTERFACIAL DISORDER ON THE X-RAY DIFFRACTION OF SUPERLATTICES

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ABSTRACT

We have generalised our x-ray diffraction results from amorphous/crystalline multilayers, to include random interfacial disorder of a gaussian type. A general relation is obtained which can be applied to both crystalline/crystalline and crystalline/amorphous multilayers. This gaussian fluctuation or "roughness" can strongly reduce the long-range atomic order along the growth direction of the multilayer. Using classical structure factor calculations, we simulate the evolution of x-ray patterns as a function of the fluctuation amplitude, the superlattice wavelength, and the interatomic distances. Applying this model to the crystalline/crystalline case we fit the experimental Nb/Cu data, deduce a fluctuation amplitude of about 0.4 Å, and relate it to the lattice mismatch between Nb and Cu. For crystalline/amorphous systems (Pb/Ge) this amplitude can be significantly larger (2 Å).

## I. Introduction

The structural analysis of superlattices and multilayers is fundamental in the understanding of their novel physical and metallurgical properties. Especially x-ray diffraction has been used [1,2] in determining the chemical composition and modulation wavelength of these materials. However, since structural details cannot be obtained directly from an inversion of the diffraction pattern, theoretical models are introduced which are then fitted to the experimental data.

Recently, a variety of models for compositionally modulated structures have been developed [3-14] . The "step model" assumes an abrupt composition profile and uses the bulk lattice spacing distance for each material, whereas the "strain model" assumes lattice-spacing variations due to in-plane coherency strains [4]. These one-dimensional models have been successfully used in semiconducting [4] and metallic superlattices [5-8] to derive peak intensities and positions.

In more realistic models one takes into account fluctuations at the interfaces, on the thickness of the layers as well as lateral fluctuations (in the plane of the layers) [3,9-16]. The fluctuations can be continuously distributed [11-15] (for instance the distance at the interface between atoms of material A and B) or can be discretely distributed [9,10,15] (for instance the number of atoms in a layer). At high angle (large q), the latter distribution of width $c^{-1}$ equal to an atomic distance gives rise to a slight reduction in diffraction peak intensity and a disappearance of the secondary peaks [15]. In a previous study [14] we showed that a continuous distribution explains the total loss of high angle superlattice peaks in crystalline/amorphous systems.

A number of different mechanisms which cause thickness fluctuations

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include : i) imperfections in the deposition process and ii) geometric constraints at the interfaces. The difference in lattice parameter and in symmetry of the constituent planes are accomodated by distorting the layers or the interfaces. This can be achieved by the introduction of in-plane coherency strain, or by the creation of misfit dislocations. The former mechanism occurs in multilayers with a small lattice mismatch (<1%) (Nb/Al [8], Nb/Ta [18]), while the latter mechanism is present in multilayers with an important lattice mismatch (Nb/Cu [5,19], Pb/Ag [7,10], Fe/Mg [13], Mo/Ni [17], Pd/Au [11]). Furthermore, when the thickness of the layers increases, a transition from a coherent to an incoherent structure is observed in multilayers with a small lattice mismatch.

In this paper, we report on the effect of continuous fluctuations of the interface distance on the line-broadening in multilayers. Using a one dimensional kinematical diffraction model, a relation is derived which can be used for crystalline/crystalline (Nb/Cu) as well as crystalline/amorphous multilayers (Pb/Ge). The width of the high-angle diffraction peaks can be explained and important structural information is obtained.

## II. Theoretical Model

The structure factor for a multilayer consisting of M crystalline blocks of material A (lattice spacing $d_a$, scattering power $f_a$, number of planes $N_a$) and B (lattice spacing $d_b$, scattering power $d_b$, number of planes $N_b$), separated by an interface distance $a_i$, is given by:

$$F(q) = \sum_{n=0}^{N_a-1} f_a \exp(iqnd_a) + f_b \exp[iq((N_a-1)d_a+a_1)] \sum_{m=0}^{N_b-1} \exp(iqmd_b)$$
$$+ \exp[iq((N_a-1)d_a + (N_b-1)d_b+a_1+a_2)]\{ \sum_{n=0}^{N_a-1} f_a \exp(iqnd_a) +$$
$$f_b \exp[iq((N_a-1)d_a+a_3)] \sum_{m=0}^{N_b-1} \exp(iqmd_b) \} + \ldots \qquad (1)$$

Following reference [2] we assume that the interface distance is not constant but fluctuates around an average value $\hat{a}$ following a continuous gaussian distribution of width $c^{-1}$.The distribution function of every interface distance $a_i$ is given by:

$$p(a_i) = (c/\sqrt{\pi}) \exp[-c^2(a_i-\hat{a})^2] \qquad (2)$$

Integrating $F(q)F^*(q)$ over all real values $a_i$ gives the average diffracted intensity:

$$I(q) = M\,(A^2 + B^2 + 2AB \exp(-q^2/4c^2) \cos(q\Lambda/2))$$
$$+ 2 \sum_{m=1}^{M-1} (M-m)\,\{ (A^2+B^2) \exp(-2mq^2/4c^2) \cos(2mq\Lambda/2)$$
$$+ AB\,(\exp(-(2m+1)q^2/4c^2) \cos((2m+1)q\Lambda/2)$$
$$+ \exp(-(2m-1)q^2/4c^2) \cos((2m-1)q\Lambda/2) \} \qquad (3)$$

where $A = f_a\,[\sin(N_a q d_a/2)]/[\sin(q d_a/2)]$

$B = f_b\,[\sin(N_b q d_b/2)]/[\sin(q d_b/2)]$

$\Lambda = (N_a-1)d_a + (N_b-1)d_b + 2\hat{a}$

For $c^{-1} = 0$, eq. (3) reduces to the step model, while for $c^{-1} = \infty$ it reduces to the scattering of two independent blocks of material A and B without any trace of superstructure. For crystalline/amorphous systems, eq. (5) of ref. 14 is recovered when $f_b=0$.

Using eq. (3), the high-angle x-ray diffraction pattern of a crystalline/crystalline multilayer is calculated for different values of the distribution width $c^{-1}$ (see Fig. 1). The distribution width $c^{-1}$ is expressed as a percentage of the average interface distance â, conventionally taken to be $(d_a + d_b)/2$. An increase of $c^{-1}$ gives rise to a decrease of the peak intensities and an increase of the linewidth.

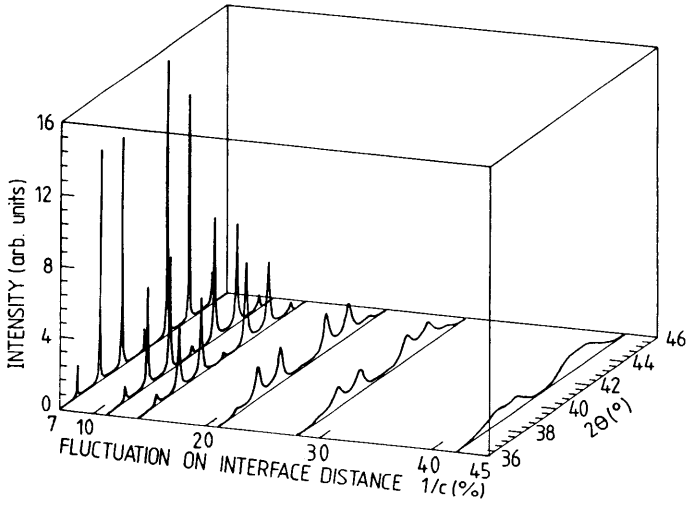


Fig. 1. Evolution of simulated high angle spectra for different values of the fluctuation amplitude $c^{-1}$, for $d_a$ = 2.33 Å, $d_b$ = 2.08 Å, $N_a = N_b = 24$, $\hat{a} = (d_a + d_b)/2$.

## III. Discussion

It is well known that the coherence length, the distance over which the x-rays are coherently scattered, can be calculated from the full width at half maximum (FWHM) of an experimental diffraction line using the Scherrer equation :

$$\xi = 0.9\ \lambda_x\ /\ (\ \mathrm{FWHM}(2\theta)\ \cos(\theta_B)\ ) \qquad (4)$$

with $\lambda_x$ the x-ray wavelength, and $\theta_B$ the Bragg angle of the diffraction peak.

In the case of crystalline/amorphous multilayers (Pb/Ge), it is assumed that the crystalline regions are separated from each other by "non-scattering" layers and the coherence length varies from the finite size length of one crystalline layer (Pb) to the total multilayer thickness.

To date, all diffraction experiments on crystalline/amorphous multilayers exhibit only one broad peak at high angle. In a previous paper [14] it was shown that small fluctuations (2 Å) of the thickness $\bar{a}$ of the amorphous layer can explain this fact (Fig. 2). This value is close to the nearest-neighbour distance in a-Ge (2.5 Å). Unfortunately this model only

gives a lower limit for the fluctuation amplitude. An even better determination can be obtained if an amorphous material is layered between large d-spacing material. The reason for this is because the coherence between atoms spaced at 5 Å for instance is less affected by a fluctuation amplitude of 2 Å than it is from atoms spaced with a distance of say 2.87 Å (Fig. 3).

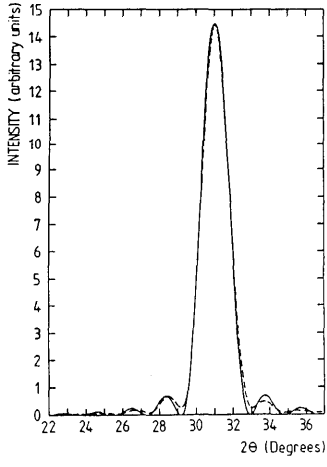


Fig. 2. Experimentally measured x-ray spectrum (dashed line) for a Pb (49 Å) / Ge (59 Å) multilayer fitted with eq. (5) of ref. 14 with $c^{-1} = 0.04\ \bar{a}$ (solid line).

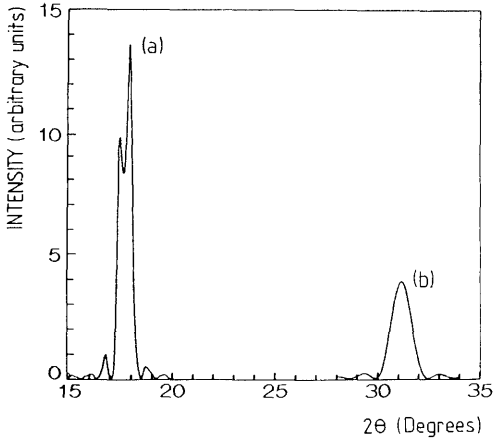


Fig. 3. Simulation of high angle spectra with eq. (5) of ref. 14 for (a) d = 5 Å, $c^{-1} = 0.07\ \bar{a}$, N = 25, $\bar{a}$ = 28 Å, M = 13, and (b) d = 2.87 Å (Pb), $c^{-1} = 0.07\ \bar{a}$, N = 25, $\bar{a}$ = 28 Å, M = 13.

For crystalline/crystalline superlattices (Nb/Cu), the coherence length observed at high angles, is much larger than the finite-size thickness of each crystalline block.

In order to further quantify our model we introduce the concept of the "number of coherent scattering modulation wavelengths", $\eta$ .This number is obtained by normalizing the coherence length by the modulation wavelength ($\eta = \xi / \Lambda$). We studied the dependence of $\eta$ on the modulation wavelength and on the fluctuation amplitude. A doubling of $\Lambda$, for a constant $c^{-1}$, simply doubles the coherence length, and keeps $\eta$ constant. This is plausible as the amount of disorder at the interface remains constant. Increasing the fluctuation amplitude, for a constant $\Lambda$, lowers the coherence length and reduces $\eta$. Figure 4 shows that for zero fluctuation amplitude, $\eta$ approaches the value of the step model while for an infinite fluctuation amplitude $\eta$ tends toward 0.5 (if $N_a d_a = N_b d_b$). This figure can directly be used to analyse experimental x-ray data : extracting the coherence length from experimental spectra gives the corresponding fluctuation amplitude.

Figure 5 shows the fluctuation amplitude as a function of the modulation wavelength, for Nb/Cu multilayers [5]. The observed fluctuation amplitude is much smaller than in the Pb/Ge case. For small modulation wavelengths the observed value is of the order of the lattice mismatch between Nb and Cu (0.4 Å).

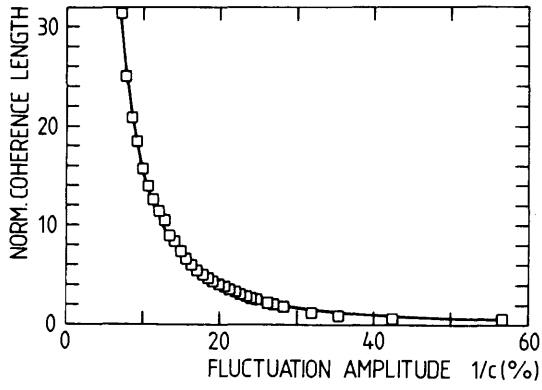


Fig. 4. The number of coherent scattering modulation wavelengths η as a function of fluctuation amplitude using eq. (3) for Nb/Cu multilayers.

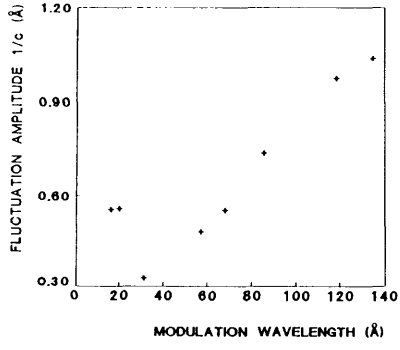


Fig. 5. Fluctuation amplitude $c^{-1}$ versus modulation wavelength for Nb/Cu multilayers.

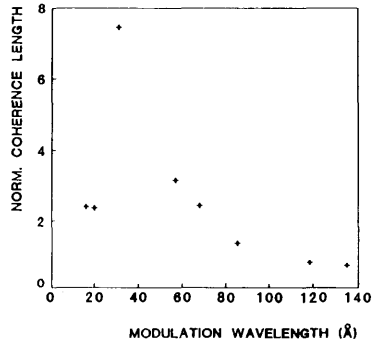


Fig. 6. Normalised coherence length versus modulation wavelength for the same samples as in Fig. 5.

Fig.6 shows the dependence of the normalized coherence length on the modulation wavelength, and indicates that the increase of the fluctuation amplitude as Λ increases is <u>not</u> an artefact of our model. This increase is a common feature in most superlattices as will be shown elsewhere [20]

Hilliard [21] calculated the energy needed to create a misfit dislocation at the interface, as well as the energy needed for epitaxial rearrangement. He found that the energy for a dislocation is inversely dependent on the thickness of the layers, whereas the energy for an epitaxial rearrangement is independent of the layer thickness. The actual configuration at the interface is determined by the balance of these energies. The increase of the fluctuation amplitude as Λ increases can thus be explained by stating that the amount of dislocations increases with Λ.

As there are many causes [16] for line-broadening it is difficult to unambiguously conclude that most of the disorder observed in diffraction patterns from multilayers is due to the above described mechanism. However the close agreement between the values of the lattice mismatch and the observed fluctuation amplitude strongly supports this model.

## IV Conclusion

We developed a model which accounts for the line broadening in crystalline/amorphous and crystalline/crystalline multilayers by assuming that all disorder is due to fluctuations of the interface distance. The value of the fluctuation amplitude is 2 Å for crystalline/amorphous while for crystalline/crystalline multilayers it is much smaller and of the order of the lattice mismatch (0.4 Å).

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