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The appropriateness of current three dimensional refinements for multislice simulations

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ABSTRACT: A number of techniques for the refinement of the basic Multislice Approach have been proposed for the simulation of, in the main, high resolution images. Many of these have merely considered how the calculations can be made more efficient. Some have re-addressed the variety of approaches which can be made for the incorporation of three dimensional effects. Here we examine the circumstances under which these latter considerations are appropriate and assess the need or otherwise for the more sophisticated algorithms.

1. INTRODUCTION

There is a trend in many current papers involving the use of the multislice approach (Cowley and Moodie 1957) for the simulation of high resolution images, to fail to specify not only many of the parameters involved but more dangerously the specific form of the algorithm used.Until a few years ago most authors, particularly when dealing with calculations for non centro-symmetric crystals, tended to make a point of saying that "upper Laue zone effects were taken into account" by using the method described by Lynch (1971). However , despite the work of Kilaas et al. (1987), which was at face value a serious attempt to clarify what should and should not be done, a considerable amount of confusion remains.For example, in the light of current practice, it is not clear to us whether Lynch, when considering sub-unit cell thickness slices, actually attempted to include the effects of atoms out of the individual slice considered or included there only the "chopped" potential distribution of those atoms present within the slice. Certainly others discussing the problem (e.g. Wilson 1983 and Kilaas et al., 1987) appear to us to have differing impressions of the original algorithm. While this is not particularly important, one of us at least is clear that until a few years ago high resolution image simulations described as following Lynch and emanating from the Cambridge HREM group used the incorrect "chopped" form of the slice potential.

Kilaas et al. were actually mainly interested in improving the accuracy to which the amplitudes in the genuine Upper Laue Zone reflections are predicted, as excited at high **g**, so as to deal with the relatively rare phenomena associated with non cancellation of dynamical contributions from such beams near to the axis. Such effects are extremely sensitive to the precise crystal orientation (as discussed by Tu and Howie, 1978 and Steeds and Evans, 1980) and are of negligible importance in high resolution imaging at a zone axis, while perhaps being better considered by a Bloch wave approach. The easiest way to deal with the manner in which intensity is bled from the zero Laue zone to the upper Laue zones would be to include their effect by an experimentally derived absorption parameter, while doing a multislice simulation which did not include their prediction (this is relatively easy!).What is of more general concern is the way in which the treament of the vertical projection of the potential in the slices used in a simulation changes the accuracy of the predicted amplitudes in the *zero* Laue Zone reflections. Of particular interest are of course the amplitudes at the "forbidden" reflections as exemplified

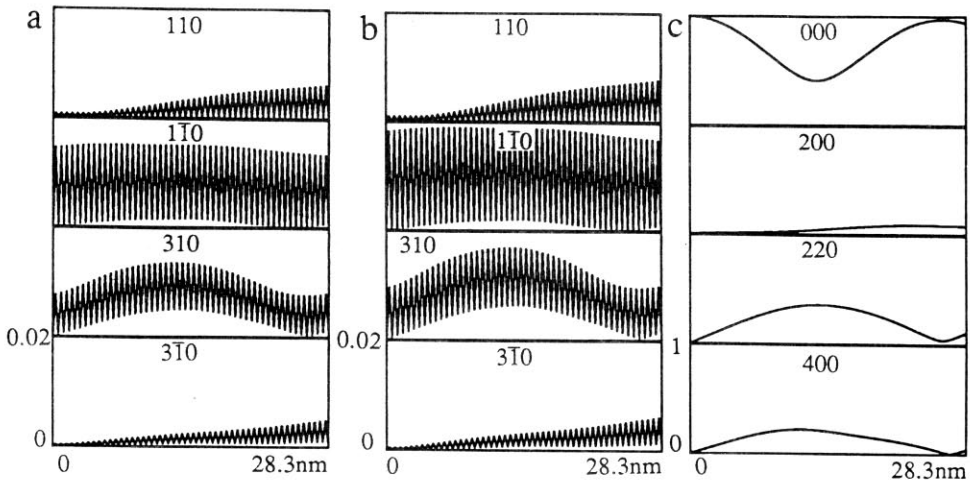

Fig. 2. Beam amplitudes for sucessive half unit cells of GaAs for "forbidden" reflections obtained using a) the "tail" model and b) the "chop" model. c) Beam amplitudes for allowed reflections for both models.

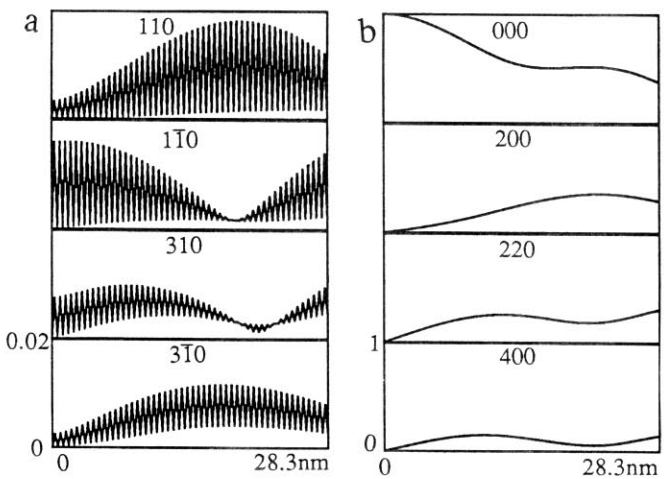

Fig. 3. Beam amplitudes for sucessive half unit cells of AlAs using the "chop" model. a) Forbidden reflections and b) allowed reflections.

successive half unit cell slices of GaAs and AlAs and the differences in magnitudes of $f_{III}+f_V$ and of $f_{III}-f_V$ for these two systems enable useful comparisons to be made between their beam amplitudes.Their behaviour slice by slice can be used to compare the two calculation methods and to help to understand the relative importance of dynamical and other slice thickness dependent effects.

Fig.1a shows the [001] projected arrangement of a half III-V unit cell and fig.1b the associated diffraction pattern. The beam amplitudes for characteristic S_F and D_F reflections for successive half unit cell thicknesses are shown in fig. 2a for the "tail"model and fig. 2b for the "chop" model. Using the conventional structure factors for Ga and As the overlapping tail between successive 0.28 nm slices was estimated to be 10%.We can first note that there are finite values of the amplitudes for the full unit cell thicknesses which build slowly for D_F and oscillate for S_F with the behaviour of the allowed beams, reaching in the latter case values as high as 0.006 for *both* models. We have found that this was not caused by "beam leakage" from the calculation and note that the effect could only be minimised by using considerably thinner slices. The zero amplitudes of the forbidden reflections should of course be maintained for

by the {110} and {310} beams for a III-V structure examined in the [001] direction. This is the same class of reflection that was used by Cherns (1974) to image in dark field the effect of the presence of fractions of unit cells in (111) foils of Au: a reflection such as 1/3(2, $\bar{4}$, 2) is of oscillating strength as the foil thickness is increased, becoming essentially zero as every third close packed plane is added.

The behaviour of this type of "forbidden" reflection is of increasing topicality in the context of the plan view examination of multilayers of either III-V materials or of Si and Ge with small individual layer thicknesses which might be fractions of a unit cell. Vincent et al. (e.g.1987) have considered the way the extra Laue zone reflections are strengthened in convergent beam patterns as a function of the form of the multilayer present for such a system and even imaged multilayer foils using these upper Laue Zone reflections. As a pilot study for the potential characterisation of, for example, the layer strains in such multilayers either by high resolution imaging or, probably more realistically, by using the Zero Laue zone "forbidden" reflections individually, we have investigated the way different multislice calculation methods predict the beam amplitudes for GaAs and AlAs in the "forbidden" as well as in the allowed reflections for an [001] beam direction. We describe here the implications of our results in relation to the degree of accuracy which would be realistically required in multislice simulations used for these types of problem.

2. SIMULATIONS FOR [001] ORIENTED GaAs AND AlAs

There are in principle many ways of dealing with the scattering effect of a vertically periodic but finite object which differ from one another to varying degrees. Here we neglect the range of refinements, due to a number of authors, largely associated with computational technique, as have been discussed by, for example, Kilaas et al. (1987). A normal multislice program assumes that each slice is embedded in an infinite lattice. Thus we might attempt to take account of the external surface by treating the scattering differently for the first few layers. Ideally this would appear to be sensible if we were dealing with an extremely thin object, paricularly if it was vertically polar and contained atoms of very high atomic number. We should remember that any differences between the results of such a calculation and those obtained by one of the more usual approaches would depend on the accuracy to which the low angle part of the atomic scattering factors was known. It would also neglect the effects associated, in Bloch wave terminology, with the distance over which the current distribution becomes stabilised. The next most realistic approach is to determine the transmission function of each slice either by completing a full three dimensional Fourier transform or equivalently in real space by including the effect of atoms in neighbouring slices using the form of the four Gaussians normally used to describe their individual scattering amplitudes (Wilson 1983).Related methods for completing the summation of the "projected" potential for slices of sub unit cell thickness have been considered by for example Self et al.(1983). A still simpler approach for slices of sub unit cell thickness, but based on the same principles as those described above, is to use an error function analysis of the vertical form of the atomic scattering potential as a whole including in each slice the relevant fractions due to atoms in neighbouring slices. Such a method would become dangerously inaccurate only when the slice thickness was made rather less than an atomic 'radius' and it should be noted that this might be necessary for very strong scatterers such as gold. It is this "tail" method which we compare here with the clearly incorrect "chop" method of neglecting the effects of atoms outside the slice altogether. The asymmetry of

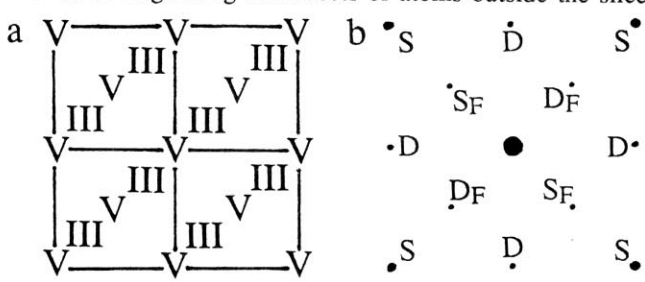

Fig. 1. a) Half of four III–V semiconductor unit cells in [001] projection and b) their diffraction pattern. S represents a sum reflection ($f_{III}+f_V$), D a difference reflection ($f_{III}-f_V$) and subscript F indicates a forbidden reflection for the whole cell.

integer unit cell thicknesses except to the very small extent to which there is genuinely dynamical contrast within a half unit cell thickness. By comparison, there is a negligible effect of using half unit cell thicknesses (on either model) on the allowed beams (see fig.2c) at integer thicknesses. It is thus amusing that, to the extent that any change would be visible in a *simulation,* the effect of the low order incorrectly simulated reflections would make either "improved" model give a poorer representation of an image than a simulation using full unit cell slice thicknesses.

The "forbidden" beams associated with the full foil thickness containing an extra half unit cell have amplitudes of up to 0.015. It is thus possible that the asymmetry associated with the S_F and D_F behaviours would make a real interference image show asymmetry, in regions containing an extra half unit cell, for a total thickness of up to about 10 nm. By contrast with the full unit cell the dynamical effects of the changes in total thickness on these beams is real. However, if observed experimentally, such an effect would almost certainly be attributed to beam tilt. It should be easier to see the predicted asymmetry effects in a simple comparison of a S_F and D_F dark field pair of images, and it is in relation to this type of experiment that the use of the "tail" model would give rather improved predictions of the expected image behaviour than the "chop" model. Even so it is questionable whether the predicted differences could be quantified given uncertainties in contributions to these beams of other origins (Boothroyd and Stobbs 1989). Figs 3a and 3b are equivalent calculations for AlAs to those in figs 2b and 2c for GaAs. We can now compare the changed effects of the differences between $f_{III}+f_V$ and $f_{III}-f_V$ and synergistically of cumulative dynamical scattering on the half integer thickness S_F.and D_F amplitudes. Now, for example, the asymmetries between the behaviours of these beams is both less marked and depends on thickness differently than for GaAs since the 200 beams build in amplitude more quickly.

3. CONCLUSIONS

We had two reasons for considering the above problem. The first was to see if we could use relatively simple approaches, by comparison with those involving the determination of slice transmission functions using full three dimensional Fourier transforms, to assess to a realistic degree the importance or otherwise of disallowed reflections on interference images. For the example considered it seems that it would probably be more accurate to refrain from using any of the "improved" models. On the other hand it would be interesting to compare the relative behaviour of S_F and D_F beams individually for these systems. For this, when we remember the degree to which comparison with experiment is realistic given that the effects of inelastic scattering and contamination are generally not included, a simple approach to the incorporation of the tails of the scattering potentials of the atoms above or below a given slice would be adequate. Our second reason for examining these effects was to decide how best to model plan view multilayers of, for example, successive periods of half a unit cell of AlAs followed by a full unit cell of GaAs. The effects of the non integer part of each wavelength of such a structure will clearly (if only from the above calculations) be to a degree cumulative. For such a configuration the reflections which are forbidden for the individual unit cells can now become allowed.It would seem that it will, in general, be adequate to use a "chop" model in the prediction of the beam amplitudes for the extended unit cell.

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