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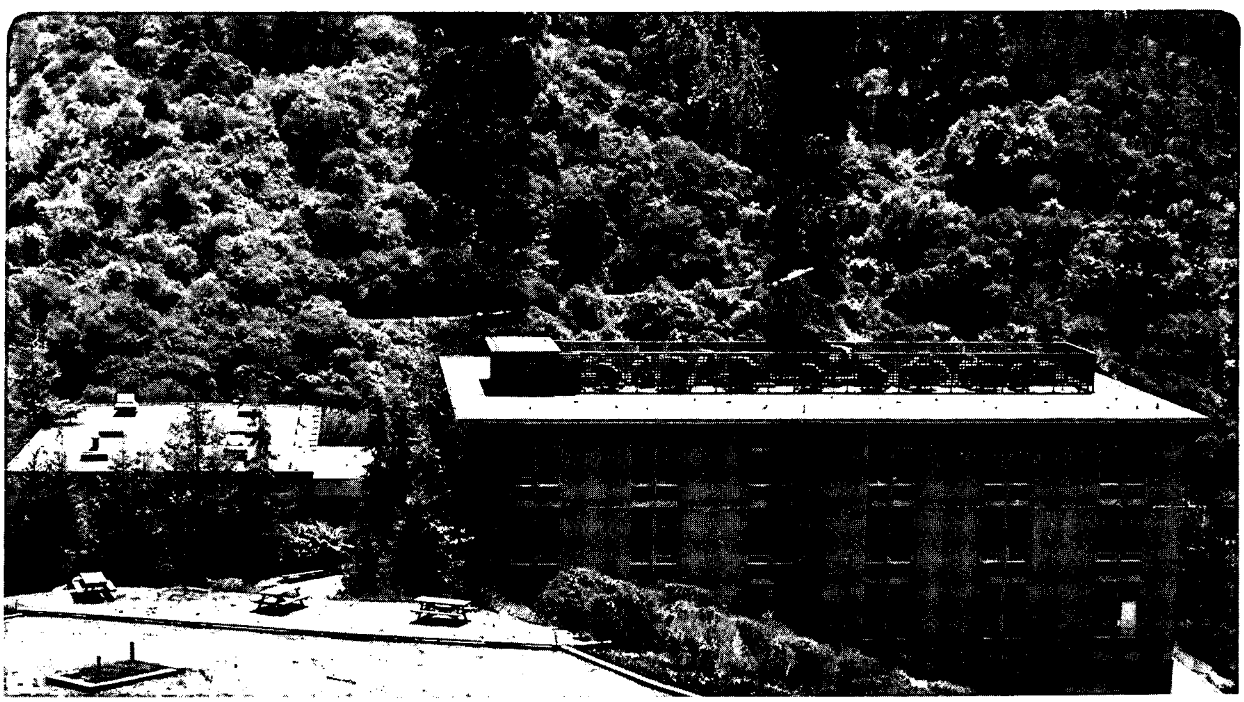

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On the Improved Phase Grating Method

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On The Improved Phase-Grating Method

Abstract

A new method for computing scattering amplitudes in High Resolution Transmission Electron Microscopy has been examined. The method, which is called the Improved Phase-Grating (IPG) method, is shown to produce reasonable results only for very small specimen thicknesses and diverges for thicknesses larger than 20 Å – 40 Å in copper [001] for accelerating voltages between 200kV – 1MV. The validity of the method is discussed and is shown to depend on electron wavelength, slice thickness,the number of reflections that are included in the calculation and the choice of specimen. It is also shown that the method does not readily allow for slice thicknesses smaller than the specimen periodicity along the incident electron beam direction.

1. Introduction

The ability of present multislice calculations to include upper Laue layer (ULL) interactions has been studied elsewhere [1,2] and it is shown [1] that the Second-Order Multislice (SOM) [3] method allows for a larger slice thickness than the conventional first-order multislice method employing fast Fourier transforms (the FFT method) while still including ULL effects. In order to include ULL effects into the SOM method and the FFT method it is necessary to use slice thicknesses smaller than the crystal periodicity (c) in the electron beam direction. The improved phase-grating method proposed by Van Dyck [4] allows for the inclusion of higher-order zones even when the slice thickness is equal to c. However, no results using this method have been published and it is not clear that the IPG method presents an alternative to existing methods. This work was undertaken in the hope of shedding light on the applicability of the method. The model system is a specimen of Cu [001], and the formulation presented in [1] is used. In order to facilitate the reading, some of that theory is repeated below.

2. Theory

The improved phase-grating method is an approximate solution of the following modified Schrödinger equation [3]:

$$\frac{\partial\phi}{\partial z} = i\sigma V\phi + \frac{i\lambda}{4\pi}\nabla_\perp^2\phi = i\sigma V\phi + \Delta\phi \tag{2.1}$$

where

$$\sigma = \frac{2\pi m e\lambda}{h^2} \tag{2.2}$$

and

$$\nabla_\perp^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \tag{2.3}$$

and the wave function ψ has been written as a modulated plane wave of the form:

$$\psi(\mathbf{r}) = \phi(\mathbf{r})\, e^{i\mathbf{k}_0\cdot\mathbf{r}} \ . \tag{2.4}$$

V is the crystal potential in volts. If the effect of the potential is greater than that of Δ, it is appropriate to start from an exact solution in V and treat Δ as a perturbation. Van Dyck [4] suggests writing the wave function as a modulated phase grating of the form:

$$\phi(x,y,z) = \exp\left\{ i\sigma\int_o^z V(x,y,z')dz' \right\}\theta(x,y,z) \tag{2.5}$$

which, after substitution into (2.1), yields:

$$\theta(x,y,z) = \theta(x,y,o) + \frac{i\lambda}{4\pi}\int_o^z dz'\Big\{\nabla_\perp^2\theta(z') + i\sigma\Delta z[\nabla_\perp^2 V_p(z')]\theta(z')$$
$$+ 2i\sigma\Delta z\nabla_\perp V_p(z')\cdot\nabla_\perp\theta(z') + (i\sigma\Delta z)^2\,\theta(z')[\nabla_\perp V_p(z')]^2\Big\} \ . \tag{2.6}$$

The first-order perturbation result for theta becomes

$$\theta(z) \approx \theta(o) + \frac{i\lambda\Delta z}{4\pi}\Big\{\nabla_\perp^2\theta(o) + i\sigma\Delta z\theta(o)\int_o^z dz'$$
$$\Big[\frac{1}{\Delta z}\nabla_\perp^2 V_p + i\sigma\Delta z\frac{1}{\Delta z}(\nabla_\perp V_p)^2\Big] + 2i\sigma\Delta z\nabla_\perp\theta(o)\cdot\int_o^z dz'\,\frac{1}{\Delta z}\nabla_\perp V_p\Big\} \ . \tag{2.7}$$

It is shown in [1] that the integrals appearing in (2.7) can be expressed as follows:

$$\frac{1}{\Delta z}\int_{z_m}^{z_m+\Delta z} \nabla_\perp^2 V_p dz' = -(2\pi)^2 \sum_{h,k} e^{2\pi i(\frac{hx}{a}+\frac{ky}{b})}\left(\frac{h^2}{a^2}+\frac{k^2}{b^2}\right) \times \sum_{\ell} V(h,k,\ell)\frac{n}{2\pi i\ell}\left(e^{2\pi i\ell z_{mo}/c}\frac{\sin \pi\ell/n}{\pi\ell/n}-1\right) \tag{2.8}$$

$$\frac{1}{\Delta z}\int_{z_m}^{z_m+\Delta z} \frac{\partial V_p}{\partial x} dz' = 2\pi i \sum_{h,k} e^{2\pi i(\frac{hx}{a}+\frac{ky}{b})}\left(\frac{h}{a}\right)\sum_{\ell} V(h,k,\ell)\frac{n}{2\pi i\ell}\left(e^{2\pi i\ell z_{mo}/c}\frac{\sin \pi\ell/n}{\pi\ell/n}-1\right) \tag{2.9}$$

$$\frac{1}{\Delta z}\int_{z_m}^{z_m+\Delta z} \frac{\partial V_p}{\partial y} dz' = 2\pi i \sum_{h,k} e^{2\pi i(\frac{hx}{a}+\frac{ky}{b})}\left(\frac{k}{b}\right)\sum_{\ell} V(h,k,\ell)\frac{n}{2\pi i\ell}\left(e^{2\pi i\ell z_{mo}/c}\frac{\sin \pi\ell/n}{\pi\ell/n}-1\right) \tag{2.10}$$

$$\frac{1}{\Delta z}\int_{z_m}^{z_m+\Delta z} \left[\left(\frac{\partial V_p}{\partial x}\right)^2+\left(\frac{\partial V_p}{\partial y}\right)^2\right] dz' =$$

$$-(2\pi)^2 \sum_{\substack{h,k\\h',k'}}\left(\frac{hh'}{a^2}+\frac{kk'}{b^2}\right) e^{2\pi i(\frac{h+h'}{a}x+\frac{k+k'}{b}y)}$$

$$\times \sum_{\ell\ell'} V(h,k,\ell)V(h',k',\ell')\frac{n}{2\pi i\ell}\,\frac{n}{2\pi i\ell'}\left[e^{2\pi i(\ell+\ell')Z_{mo}/c}\,\frac{\sin\pi(\ell+\ell')/n}{\pi(\ell+\ell')/n}\right.$$

$$\left. - e^{2\pi i\ell Z_{mo}/c}\,\frac{\sin\pi\ell/n}{\pi\ell/n} - e^{2\pi i\ell' Z_{mo}/c}\,\frac{\sin \pi\ell'/n}{\pi\ell'/n} + 1\right] \tag{2.11}$$

where the slice thickness Δz is equal to c/n. Equation (2.11) simplifies in two cases :

i) the limit $\Delta z \rightarrow 0$

$$\frac{1}{\Delta z}\int_{z_m}^{z_m+\Delta z} \left[\left(\frac{\partial V_p}{\partial x}\right)^2+\left(\frac{\partial V_p}{\partial y}\right)^2\right] dz' = -\frac{(2\pi)^2}{3}\left[\left(\sum_{h,k}\frac{h}{a}V(h,k,0)e^{2\pi i(\frac{hx}{a}+\frac{ky}{b})}\right)^2 + \left(\sum_{h,k}\frac{k}{b}V(h,k,0)e^{2\pi i(\frac{hx}{a}+\frac{ky}{b})}\right)\right] \tag{2.12}$$

ii) n = 1

$$\frac{1}{c}\int_{z_m}^{z_m+c}\left(\frac{\partial V_p}{\partial x}\right)^2 dz' = \left[\frac{1}{3}+\frac{z_m}{c}\left(1+\frac{z_m}{c}\right)\right]\left[\sum_{h,k} 2\pi i\left(\frac{h}{a}\right)V(h,k,0)e^{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)}\right]^2 \tag{2.13}$$

$$+ 2\left[\sum_{h,k} 2\pi i\left(\frac{h}{a}\right)V(h,k,0)e^{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)}\right]\left[\sum_{h,k} 2\pi i\left(\frac{h}{a}\right)e^{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)}\sum_{\ell\neq 0}\left(\frac{1}{2\pi i\ell}\right)^2 V(h,k,\ell)\right]$$

$$+ \left(\frac{1}{2}+\frac{z_m}{c}\right)\left[\sum_{h,k} 2\pi i\left(\frac{h}{a}\right)V(h,k,0)e^{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)}\right]$$

$$\times\left[\sum_{h,k} 2\pi i\left(\frac{h}{a}\right)e^{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)}\sum_{\ell\neq 0}\frac{i}{2\pi\ell}V(h,k,\ell)\right]$$

$$+\left[\sum_{h,k} 2\pi i\left(\frac{h}{a}\right)e^{2\pi i\left(\frac{hx}{a}+\frac{ky}{b}\right)}\sum_{\ell\neq 0}\frac{1}{2\pi i\ell}V(h,k,\ell)\right]^2$$

The equivalent expression for the derivative with respect to y follows from (2.13).

2.1 Validity of the improved phase-grating method

Equation (2.7) represents the second term in a series expansion for theta, the first being $\theta_0 = \theta(o)$. In order to get a feeling for the error introduced by the truncation of the series it is instructive to look at the Fourier transform of (2.7). To simplify the expressions one can include only terms where $\ell = 0$ and ignore terms with $\ell \neq 0$, that is, ignore the effect of higher-order zones. Using (2.8) through (2.11) one obtains the following for the Fourier transform of theta:

$$\theta(\mathbf{g},z_m+\Delta z) = \theta(\mathbf{g},z_m) - i\pi\lambda\Delta z\{g^2\theta(\mathbf{g},z_m) \tag{2.1.1}$$

$$+ (i\sigma\Delta z)\left(\tfrac{1}{2}+\frac{z_m}{\Delta z}\right)\sum_{\mathbf{g}'} g'^2\theta(\mathbf{g}-\mathbf{g}',z_m)\, V(\mathbf{g},0)$$

$$+ (i\sigma\Delta z)^2\left[\tfrac{1}{3}+\frac{z_m}{\Delta z}\left(1+\frac{z_m}{\Delta z}\right)\right]\sum_{\mathbf{g}',\mathbf{g}''} \mathbf{g}''\cdot(\mathbf{g}'-\mathbf{g}'')\,\theta(\mathbf{g}-\mathbf{g}',z_m)$$

$$\times\, V(\mathbf{g}'',0)\, V(\mathbf{g}'-\mathbf{g}'',0)$$

$$+ 2(i\sigma\Delta z)\left(\tfrac{1}{2}+\frac{z_m}{\Delta z}\right)\sum_{\mathbf{g}'} \mathbf{g}'\cdot(\mathbf{g}-\mathbf{g}')\,\theta(\mathbf{g}-\mathbf{g}',z_m)\, V(\mathbf{g}',0)\}$$

As seen from the expression above, the convergence of the series depends on the wavelength, slice thickness, strength of the crystal potential and the maximum reciprocal wavevector (g_{max}) included in the calculation. By inserting the first-order perturbation result for theta into (2.6) it is possible to get a second-order result for theta and obtain the equivalent of (2.1.1). However, the higher-order expressions quickly become very complicated and it is difficult to obtain a useful criterion for the validity of the expansion. The part of the expansion that does not involve the potential goes as

$$\theta(g,z_m+\Delta z) = (1 - i\pi\lambda\Delta z g^2 + \frac{(i\pi\lambda\Delta z)^2}{2} g^4)\,\theta(g,z_m) \ , \tag{2.1.2}$$

and this corresponds to the expansion of the propagator that shows up in the formulation of the real space (RSP) method [5]. However, since there is no such requirement that the intensity of each $\theta(h,k)$ remains unchanged from slice to slice, only that $\sum_{h,k} |\theta(h,k)|^2 = 1$, one cannot apply the same condition as for the RSP method. Also there are terms involving the strength of the crystal potential which complicate matters. In the limit that the interaction parameter σ or the strength of the potential go to zero the criterion of validity becomes

$$K \equiv \lambda\Delta z g^2 << 1/\pi \ . \tag{2.1.3}$$

That this is not a sufficient restriction will become evident from the results in paragraph 4.

3. Procedure

The theory outlined in the previous paragraph was implemented in computer programs and run on a CDC 7600. The model system is copper in the [001] orientation and the calculation was performed for an accelerating voltage of 200 kV and 1000kV. Because of the practical problems associated with the calculation of the expression in (2.11), the only slice thickness considered was 3.6 Å, which corresponds to the specimen periodicity in the incident electron beam direction. The difficulties associated with use of a smaller slice thickness are discussed in paragraph 5.

4. Results

The results consist of a series of comparisons between the conventional (FFT) multislice method, the phase-grating method and the improved phase-grating method and are shown in Figs. 1 through 3. In Fig. 1 the accelerating voltage is 200 kV while in Figs. 2 and 3 the voltage is 1 MV. As seen from the figures the amplitudes calculated by the IPG method begin to diverge after approximately 20 – 40 Å depending on λ and g_{max}. In Figs. 1 and 2 the slice thickness is equal to c allowing the inclusion of higher-order zones into (2.11). The contribution by terms given by $\ell \neq 0$ is essential for including out-of-the-zone effects, but is small compared to the term $\ell, \ell' = 0$ and was ignored in (2.11) such that amplitudes and phases in the case of $\Delta z < c$ could be computed. Thus Fig. 3 shows amplitudes calculated for three different values of g_{max} (2.0 $Å^{-1}$, 2.8 $Å^{-1}$ and 4.0 $Å^{-1}$) while varying Δz (n) to keep the value of K constant (0.126 and 0.063). The results indicate that varying the slice thickness while keeping the wavelength and g_{max} constant has little effect.

5. Discussion

It is clear from Figs. 1 through 3 that the improved phase-grating method fails to give reasonable results beyond a thickness of 20 Å – 40 Å for the combinations of wavelength, slice thickness and sampling interval that were used. As expected, the method works better for higher voltages where the wavelength gets smaller and the propagator becomes less important. Surprisingly, reducing the slice thickness does not appear to increase the accuracy of the method as (2.1.1) would indicate, although the results in Fig. 3 are slightly inaccurate since the contribution of higher-order zones were ignored in the term involving the square of the derivatives. Apart from wavelength and slice thickness, the convergence of the expansion depends also on sampling interval (g_{max}) and on the strength of the crystal potential. The dependence on sampling is clearly seen in the results, where extending the calculation further into reciprocal space causes the amplitudes of diffracted beams to diverge at a decreasing thickness. The Fourier coefficients of the potential are determined by choice of specimen and were not varied.

Copper with an atomic number of 29 represents a compromise between heavy and light elements and the results serve as a useful guide for other elements.

By neglecting the terms involving the potential it is possible to set an upper limit on the value of K for which the expansion for theta converges. This limit corresponds to $K_{max} = 1/3$, but it is clear from the calculations that when the potential is included more severe restrictions are imposed, restrictions that now also depend on the the crystal potential. However it is very difficult to find a useful expression in this case.

Because the method rapidly diverges, it might only be of academic importance to consider the extent to which upper Laue layer effects are included in the improved phase-grating method (IPG). Compared to commonly used multislice methods that rely on small slice thickness to include higher-order interactions [1], the IPG method includes higher-order effects even in the case where the slice thickness is equal to the crystal periodicity along the incident electron beam direction. However, because of the cross-terms that appear in (2.11), the method becomes impractical when n is different from 1. In the case of $n \neq 1$ the calculation of (2.11) requires a minimum of 10^9 operations (convolution over 6 indices) for 32^3 sampling points, and needs to be repeated n times. Even with the use of modern-day super computers this is hardly a small calculation.

6. Conclusion

The results show that due the limited range of validity of the improved phase-grating method it is not suitable for computation of scattering amplitudes in HRTEM. Even though the results in Fig. 3 show very little dependence on Δz, the validity of the method depends on the slice thickness, and the calculation should improve with smaller slice thicknesses. However, computational considerations prohibit the proper use of arbitrary slice thickness and prevent further investigation into thickness dependence. An upper limit of $K = \lambda \Delta z g_{max}^2 = 1/\pi$ is necessary to give convergence to the series expansion for theta, but it is clear that the sampling interval in combination with the strength of the crystal potential is more important in determining the conditions for convergence. However, an exact expression for convergence was not found.

Acknowledgements

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References

[1] Manuscript in preparation.

[2] D.F. Lynch, Acta Cryst. A27 (1971) 399.

[3] D. Van Dyck, J. Microscopy 199 (1980) 141.

[4] D. Van Dyck, J. Microscopy 132 (1983) 31.

[5] R. Kilaas and R. Gronsky, Ultramicroscopy 11 (1983) 289.

FIGURE CAPTIONS

Fig. 1. Amplitude and phase (in units of π) vs. thickness for the reflections 000, 200 and 440 in copper [001]. Calculations are performed by the conventional (FFT) multislice method (--), the phase-grating (PG) approximation (++) and the improved phase-grating (IPG) method (**). Accelerating potential is 200 kV and the crystal potential has been sampled out to 2.0 $Å^{-1}$. The slice thickness is 3.6 Å corresponding to $n = 1$.

Fig. 2. As in Fig. 1 except that the accelerating potential has been set to 1 MV.

Fig. 3. Amplitude vs. thickness for the reflection 200 for two sets of values of K. In the first column K = 0.126 and in the second column K = 0.063. The value of g_{max} has been set to 2.0 $Å^{-1}$, 2.8 $Å^{-1}$ and 4.0 $Å^{-1}$ in the first, second and third row respectively. The slice thickness required to keep K constant is indicated by the value of n ($\Delta z = 3.6$ Å/n).

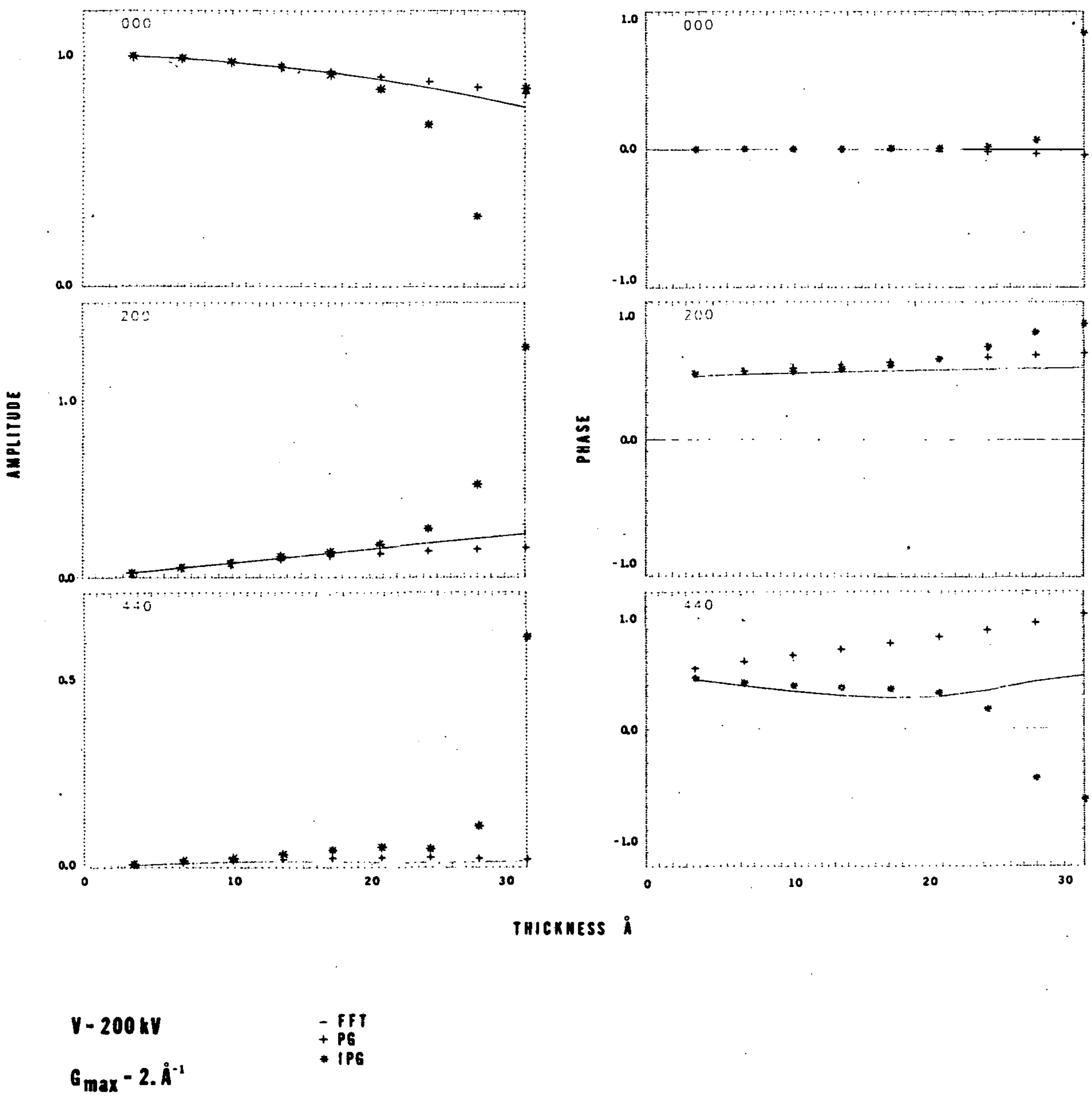

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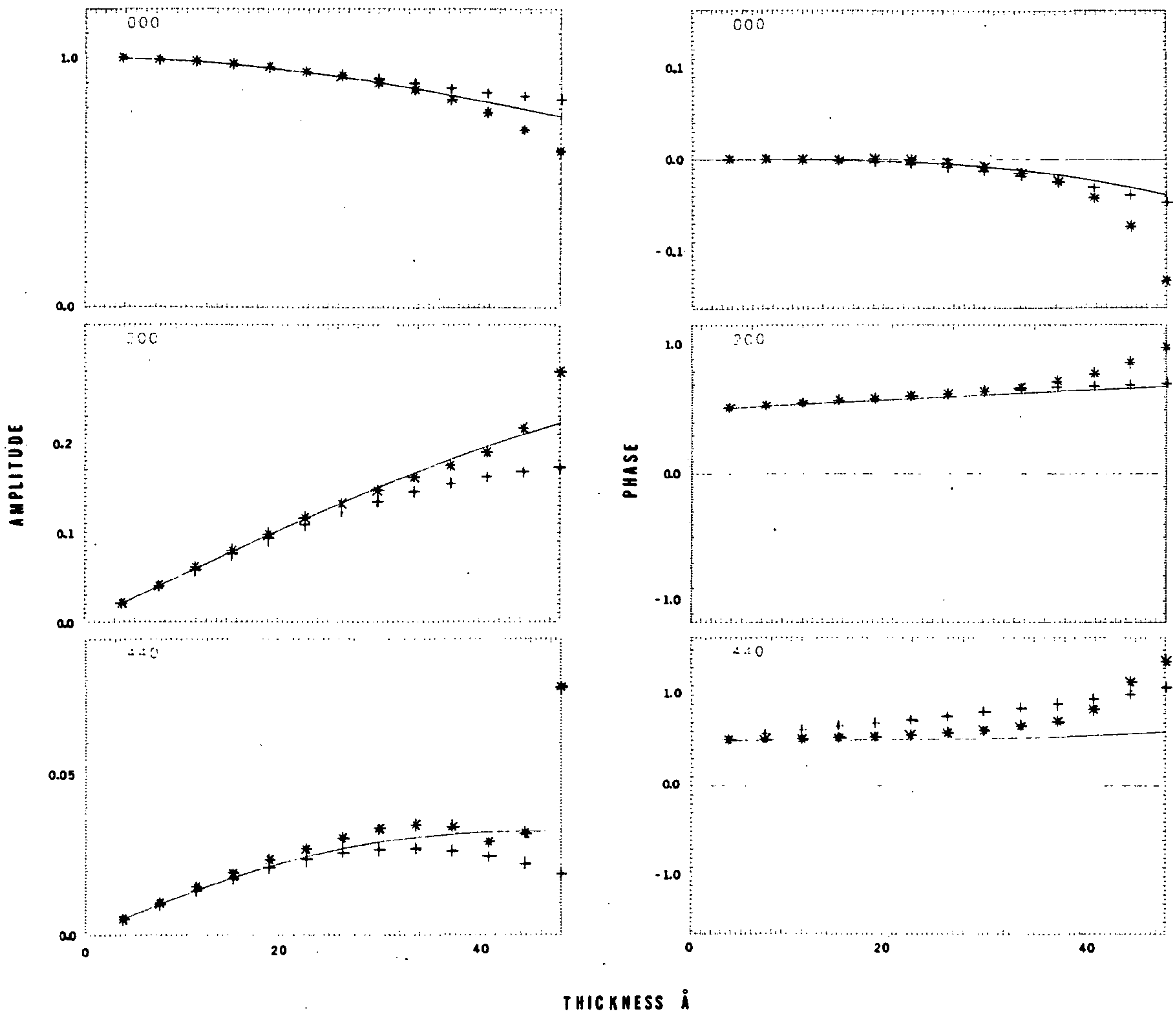
Figure 1.



V - 1MV

G_{max} - 2. Å^{-1}

- FFT
+ PG
* IPG

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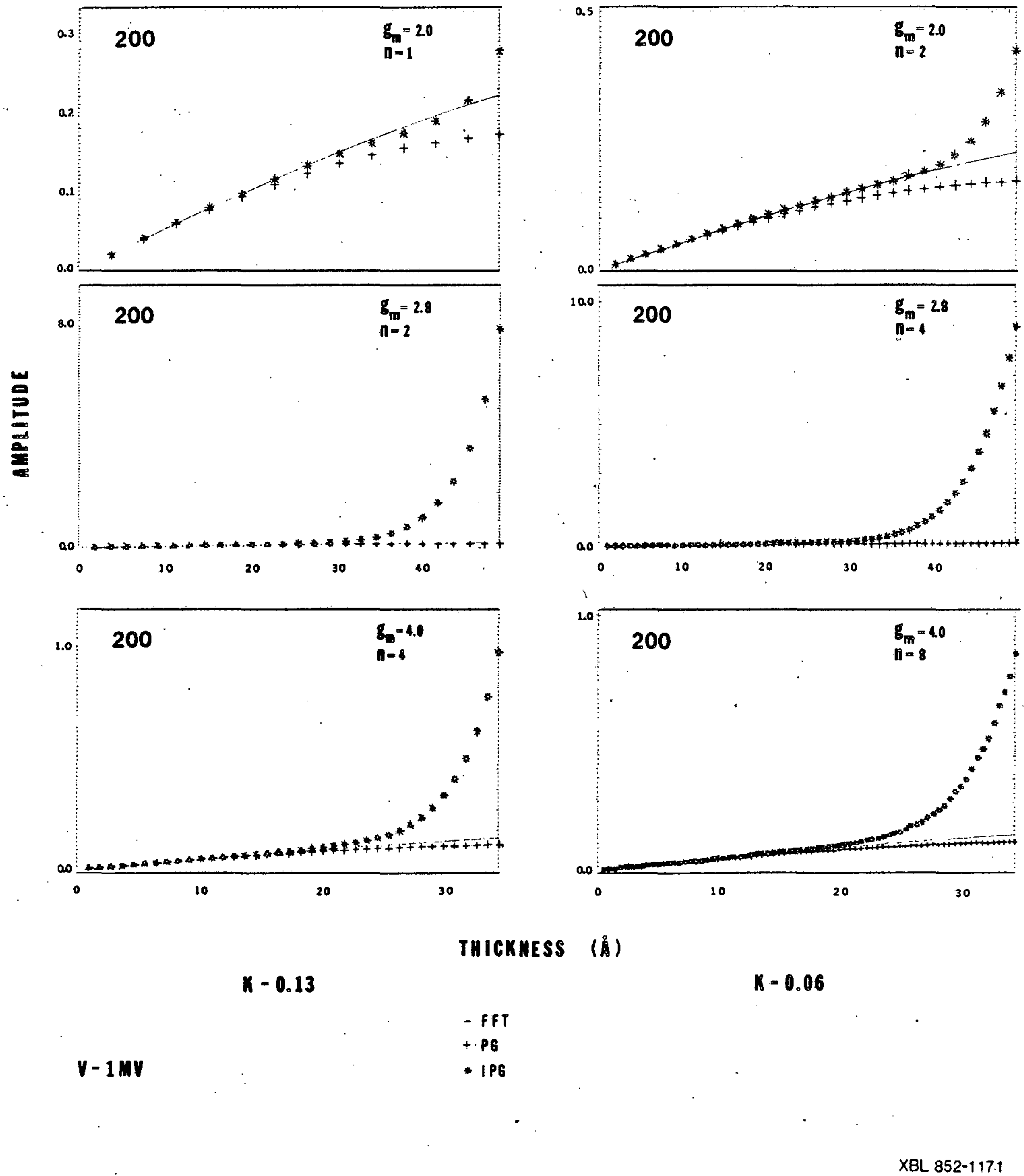
Figure 2.



200
g_m = 2.0
n = 1
200
g_m = 2.0
n = 2
200
g_m = 2.8
n = 2
200
g_m = 2.8
n = 4
200
g_m = 4.0
n = 4
200
g_m = 4.0
n = 8
AMPLITUDE
THICKNESS (Å)
K - 0.13
K - 0.06
- FFT
+ PG
* IPG
V - 1MV
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Figure 3.

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