

SHORT NOTE

LINEAR IMAGING AND DIFFRACTION OF AN AMORPHOUS FILM

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It is experimentally established that one can use linear imaging theory with implied kinematical diffraction theory to understand the high resolution image of an amorphous material of thickness 10–20 nm. It is also established that an equivalent thickness of a crystalline material cannot be understood by these models, but instead requires non-linear imaging theory and dynamical diffraction. It is shown here that an amorphous material is a special case where the non-linear and dynamical diffraction solutions reduce to simpler linear and kinematical forms.

1. Introduction

It is experimentally well established that one can use linear imaging theory, primarily the weak-phase object approach (which also assumes weak kinematical scattering), to understand the image of a thin amorphous film, for instance amorphous carbon. It is also established that neither linear imaging theory nor kinematical scattering theory can be applied to explain the image of an equally thick crystalline material. This leads to an apparent discrepancy which is clearly confusing at the very least to students of high resolution electron microscopy, and furthermore leads to some conceptual traps; for instance, if Scherzer defocus is “best” for imaging an amorphous film, one would suspect that it is also ‘best’ for imaging a crystalline material.

The intent of this paper is to clarify this apparent contradiction. It is shown that for the particular case of an amorphous material the full dynamical diffraction solutions convert to an essentially kinematical form and that a non-linear imaging analysis similarly converts to a linear form. Thus an amorphous material is an understandable exception to the general rule.

2. Theory

For simplicity we will consider separately the diffraction and imaging questions, although it is the combination of the two which leads to the simplifications.

2.1. Diffraction from an amorphous material

Our intention here is to show that for an amorphous material most of the second- and higher-order scattering is much smaller than it is for a crystalline material, and that the scattering can be well described by a kinematical model. Here we will use a scattering series approach to the general diffraction problem. Considering Schrödinger’s equation

$$\{\nabla^2 + (8\pi^2me/h^2)[E + V(\mathbf{r})]\} \psi(\mathbf{r}) = 0, \quad (1)$$

we will look for a solution of form

$$\psi(\mathbf{r}) = \int \phi(\mathbf{k}, z) \exp(2\pi i \mathbf{k} \cdot \mathbf{r}) d\mathbf{k}, \quad (2)$$

where the incoming beam is along the z direction and the different values of \mathbf{k} correspond to wave-vectors on the Ewald sphere. Substituting $\psi(\mathbf{r})$ in

the form of eq. (2) into eq. (1) and neglecting the second derivative of ϕ with respect to z as small, we obtain after integrating over the x, y plane the standard equation

$$\begin{aligned} d\phi(\mathbf{k}, z)/dz &= -(4\pi ime/h^2 k_z) \\ &\times \int_{-\infty}^{\infty} \exp[2\pi i(\mathbf{k}' - \mathbf{k}) \cdot \mathbf{z}] \\ &\times \tilde{V}(\mathbf{k} - \mathbf{k}') \phi(\mathbf{k}', z) d\mathbf{k}', \quad (3) \end{aligned}$$

where $\tilde{V}(\mathbf{k})$ is the Fourier transform of $V(\mathbf{r})$ and we are considering that the crystal is a slab in the range $0 < z < t$. In standard texts on diffraction theory, eq. (3) is solved as a set of coupled differential equations. An alternative method is to use a series expansion as in scattering theory. With the (standard) approximation that the term k_z in front of the integral can be taken as a constant for all \mathbf{k} values of interest, we can solve in the form

$$\phi(\mathbf{k}, z) = \sum_n \phi_n(\mathbf{k}, z), \quad (4)$$

where

$$\begin{aligned} \phi_n(\mathbf{k}, z) &= -(4\pi ime/h^2 k_z) \\ &\times \int_0^z \int_{-\infty}^{\infty} \exp[2\pi i(\mathbf{k}' - \mathbf{k}) \cdot \mathbf{z}'] \\ &\times \tilde{V}(\mathbf{k} - \mathbf{k}') \phi_{n-1}(\mathbf{k}', z') d\mathbf{k}' dz', \quad (5) \end{aligned}$$

and $\phi_0(\mathbf{k}, z)$ is a delta function which is non-zero when \mathbf{k} is the incident beam wavevector and independent of z . The first-order solution is the kinematical solution, whilst higher-order terms are multiple scattering contributions. What we will now show is that for an amorphous specimen these higher-order terms are anomalously small. Let us consider the second-order term, which when written out in full is

$$\begin{aligned} \phi_2(\mathbf{k}, z) &= -(4\pi me/h^2 k_z)^2 \\ &\times \int_0^z dz' \int_0^{z'} dz'' \int_{-\infty}^{\infty} \exp\{2\pi i[(\mathbf{k}' - \mathbf{k}) \\ &\cdot \mathbf{z}' + (\mathbf{k}_0 - \mathbf{k}') \cdot \mathbf{z}'']\} \\ &\times \tilde{V}(\mathbf{k}_0 - \mathbf{k}') \tilde{V}(\mathbf{k} - \mathbf{k}') d\mathbf{k}', \quad (6) \end{aligned}$$

where we are taking \mathbf{k}_0 as the wavevector of the incident plane wave. For an amorphous material we can take $\tilde{V}(\mathbf{k})$ to have essentially constant absolute value and completely random phases, i.e. $V(\mathbf{r})$ is simply noise. Two features of $\tilde{V}(\mathbf{k})$ can now be used to show that the second-order terms are anomalously small. First if we ignore the phase variations and take $\tilde{V}(\mathbf{k}) = C$ as constant,

$$\begin{aligned} \phi_2(\mathbf{k}, z) &= -(4\pi meC/h^2 k_z)^2 \\ &\times \int_0^z dz' \int_0^{z'} dz'' \int_{-\infty}^{\infty} \exp\{2\pi i[(\mathbf{k}' - \mathbf{k}) \cdot \mathbf{z}' \\ &+ (\mathbf{k}_0 - \mathbf{k}') \cdot \mathbf{z}'']\} \phi_0(\mathbf{k}_0, z'') d\mathbf{k}' \quad (7) \\ &\sim -(4\pi meC/h^2 k_z) z \delta(\mathbf{k} - \mathbf{k}_0) \phi_0(\mathbf{k}_0, z). \quad (8) \end{aligned}$$

This asymptotic relationship holds well for quite small values of z , for instance 10 nm, even without the integration over \mathbf{k}' , and is the same in spirit as the arguments used to derive the Golden Rule for inelastic scattering. As such it depends upon having a flat, continuous spectrum of values of $\tilde{V}(\mathbf{k})$ which an amorphous material has and a crystal has not. (It is not necessary to have a completely flat $\tilde{V}(\mathbf{k})$, just a slowly varying high density of values.) This argument is reinforced when we include the random phases (it then becomes essentially the "Random Phase" method of deriving inelastic scattering intensities) since we will have additional destructive interference. It is informative to note that eq. (8) indicates that the amorphous specimen is acting as essentially a classical scatterer; the only surviving second-order term corresponds to the attenuation of the incident beam. Similar arguments can be applied to higher-order scattering terms with the same general conclusions; the kinematical result is therefore very strong.

It follows, therefore, that to a far better approximation than for a crystalline material, it is quite valid to consider an amorphous sample as a purely kinematical scatterer.

2.2. Imaging theory

We now consider the second-order imaging of an amorphous specimen. Let us first consider (for

comparison) a simple crystalline specimen for which the wavefunction after the specimen $\psi(\mathbf{r})$ is a simple sum of diffracted beams, i.e.

$$\psi(\mathbf{r}) = \sum_{\mathbf{g}} \phi_{\mathbf{g}} \exp(2\pi i \mathbf{g} \cdot \mathbf{r}). \quad (9)$$

To obtain an upper bound to the second-order contribution in the final image we allow all the second-order contrast transfer terms to be unity. The second-order contribution of frequency \mathbf{g} , $I_{\mathbf{g}}$ is then

$$I_{\mathbf{g}} = \sum_{\mathbf{h}} \phi_{\mathbf{g}-\mathbf{h}}^* \phi_{\mathbf{h}}. \quad (10)$$

This sum can be fairly significant, particularly for a large unit cell material if there are no large variations in the phases. For instance, if each of the $\phi_{\mathbf{g}}$ is equal to 0.05 and we have ten possible combinations, the fractional second-order contribution to the image will be 0.025. This can be compared to a typical image contrast of 0.1. It is accurate to say that the second-order terms are constructively interfering.

Now consider the case of an amorphous specimen. Here the $\phi_{\mathbf{g}}$ coefficients will have random phases (since $\tilde{V}(\mathbf{k})$ has random phases) and therefore destructively rather than constructively interfere. To show this qualitatively we can write the wave after the specimen as

$$\psi(\mathbf{r}) = 1 - ian(\mathbf{r}), \quad (11)$$

where $n(\mathbf{r})$ is a noise function which can take random values between plus and minus one. Setting the second-order transfer function again to

unity, the second-order contribution to the final image ($S(\mathbf{r})$) is

$$S(\mathbf{r}) = \alpha^2 n(\mathbf{r})^2 \quad (12)$$

$$\cong 0.5\alpha^2(1 - n(\mathbf{r})), \quad (13)$$

where we are allowing phase randomization to permit changing from $n(\mathbf{r})^2$ to $n(\mathbf{r})$. Compared to the crystalline case, if $n(\mathbf{r})$ is 0.05 then the upper bound to the second-order terms will be only 0.00025, which is negligible. This can be shown very simply by numerical integration of the full non-linear imaging theory for simulated carbon films; these are not included here since the widespread use of linear contrast transfer functions and optical diffraction patterns *experimentally* makes a stronger case than any numerical calculation can.

3. Conclusions

An amorphous specimen acts anomalously as a kinematical scatterer which can be understood using linear weak phase object imaging theory not because it is inherently a kinematical or a weak phase object, but instead because the dynamical diffraction and second-order imaging terms are anomalously small.

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