

LETTER TO THE EDITOR

Interpreting electron micrographs of amorphous solids

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Abstract. Using the diffraction theory of random wavefunctions, we argue that recent interference electron microscopy of thin amorphous films cannot be employed to distinguish between the crystallite and random network models.

It is argued by Rudee and Howie (1972, hereafter called RH) that the fringes sometimes seen in images of thin films of amorphous Ge and Si, obtained by interference electron microscopy, imply that these materials contain, or may even consist wholly of, crystallites a few interatomic distances across. We wish to show that the important features of such micrographs would result from *any* atomic arrangement which correctly reproduced the observed diffraction pattern, provided the 'range of order' were small in comparison with the film thickness. Such possible arrangements include the random network model (Grigorovici and Manaila 1969) as well as the crystallite model, so that these are not distinguished by the type of observation under discussion.

The 'object' which is imaged by the microscope is the wavefunction $\psi_0(\mathbf{R})$ at the exit face of the specimen, \mathbf{R} being a two-dimensional position coordinate in planes parallel to the film. We may write $\psi_0(\mathbf{R})$ as a superposition of two-dimensional plane waves, with wavevectors \mathbf{K} , that is as

$$\psi_0(\mathbf{R}) = \frac{1}{2\pi} \iint d\mathbf{K} \bar{\psi}(\mathbf{K}) \exp(i\mathbf{K} \cdot \mathbf{R}) \quad (1)$$

whose 'power spectrum', defined as

$$I(\mathbf{K}) \equiv |\bar{\psi}(\mathbf{K})|^2 \quad (2)$$

gives the intensity of the diffraction pattern observed in the focal plane of the instrument. This takes the familiar form (shown schematically in figure 1) of diffuse rings centred on $\mathbf{K} = \mathbf{K}_0$, where \mathbf{K}_0 is the transverse component of the wavevector of the (approximately) plane wave of electrons incident on the specimen.

The image is formed after filtering $I(\mathbf{K})$ through an aperture, usually circular, in the focal plane. The aperture transmissivity $f(\mathbf{K})$ is unity in the aperture, zero on the screen outside. Thus the image wavefunction $\psi_1(\mathbf{R})$, neglecting magnification, lens aberrations and defocusing effects, is

$$\psi_1(\mathbf{R}) = \frac{1}{2\pi} \iint d\mathbf{K} f(\mathbf{K}) \bar{\psi}(\mathbf{K}) \exp(i\mathbf{K} \cdot \mathbf{R}) \quad (3)$$

whose power spectrum is $f(\mathbf{K})I(\mathbf{K})$.

It is therefore possible to regard the image as the interference pattern of many plane waves, containing only those periodicities which correspond to differences between K -values lying on those parts of the rings of $I(K)$ which are accepted by the focal aperture. In the interference micrographs published by RH, the aperture admits either the central spot and a portion of the first ring (position A, figure 1), or portions of the first and second rings (position B, figure 1). The shortest periodicity—corresponding to the so-called ‘lattice fringes’—arises from the large separation K_s between the central spot and first ring (A), or between the first and second ring (B). However, these fringes are affected in a fundamental way by the finite widths ΔK of the rings.

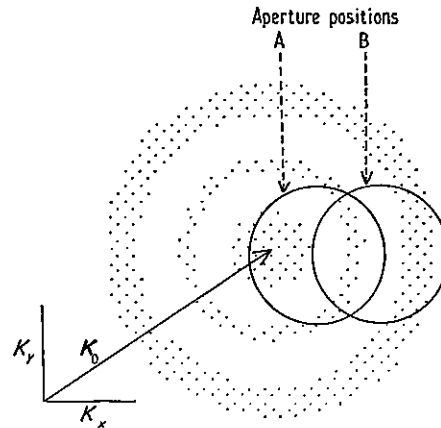


Figure 1. Diffraction rings and aperture positions in the focal plane (schematic).

The manner in which ΔK affects the fringes depends on the *phases* of the plane-wave components of the image—that is of $\psi(K)$ —which cannot be deduced from $I(K)$. Let us suppose these phases to be *random*—an assumption to be justified below. Then the image $\psi_I(R)$ is a gaussian random function of R (Rice 1944), and the two-dimensional analysis by Longuet-Higgins (1956) shows that the effect of ΔK is to divide the fringes into *groups*. Our present example has a rather complicated power spectrum, but a rough estimate of the number N of bright fringes in each group is

$$N = \frac{K_s}{\pi \Delta K \sqrt{2}} \quad (4)$$

where ΔK is, precisely, the full width of half maximum of the wider of the rings (B) or the wider of the first ring and central spot (A). On the contrary assumption, that all the plane waves have the same phase, there is only one group, containing N fringes, where N is still given by (4) (for example, a single small crystallite would give such an image).

Now consider aperture position A (figure 1): ΔK can be measured for the first ring, using the densitometer trace in the paper by Shevchik (1972), and, if we assume that the apparently large diameter of the central spot is an artefact arising from halation, inelastic scattering and beam convergence, N can be evaluated. We obtain the value 2.3. In fact the micrograph for case A (see RH) consists of groups of about three fringes, in fair agreement with this simple theory. The apparent tendency of the fringes to lie along two principal directions in a ‘tweed’ pattern is not explained by our arguments (which ignore directional effects), or by the different considerations advanced by RH.

Turning now to aperture position B, the calculated value of N (which involves ΔK for the broader second ring) is 1.4. But if the groups contain only 1.4 waves each, the resulting image should not show 'lattice fringes', and in fact no periodic structures can be discerned in the corresponding micrograph (see RH). We emphasize the approximate nature of our argument: more significance attaches to the difference between N -values than to the values themselves.

Thus we can account for the micrographs without considering the detailed atomic arrangement in the specimen, simply by examining the aperture-filtered diffraction pattern, provided the wave $\psi_{\text{I}}(\mathbf{R})$ (and hence $\psi_0(\mathbf{R})$) is a gaussian random function. Is this assumption justified? To answer this question we must consider the beam-specimen interaction; if this is such that the exit wave is the sum of a large number of independent random contributions, then $\psi_0(\mathbf{R})$ is a gaussian random function (Rice 1944). If the Born (or 'kinematic') approximation holds, each atom scatters once to the exit point \mathbf{R} . But the atomic positions are not independent: they may be arranged in tetrahedral units (random network model), or alternatively as crystallites. The average number of such independent units which scatter *obliquely* to \mathbf{R} can easily be shown to be less than unity for the film thickness concerned (200 Å) (the units contributing in this way must lie between two pairs of cones whose apices lie at \mathbf{R} , enclosing the angles of the diffraction rings). However, the number of units scattering *forward* to \mathbf{R} is simply the film thickness divided by the mean linear extent of a unit, and this is large on either model. Since these forward-scattered waves (which must not be confused with the unscattered wave) are independent, $\psi_0(\mathbf{R})$ is gaussian random, and our theory is applicable. (The theory does not apply to films consisting of 'platelet' crystallites whose size is comparable with the film thickness; in such cases it is of course perfectly correct to interpret regions crossed by fringes as the images of individual crystallites.)

These conclusions about the statistical nature of the image are reinforced by the fact that the Born approximation is rather poor for 100 kV electrons in 200 Å films, so that the number of independent contributions to $\psi_0(\mathbf{R})$ is increased by multiple scattering, between individual atoms in a random network, or between crystallites. An improvement on the Born approximation, which includes some 'dynamical' effects, is the phase-grating approximation (Cowley and Moodie 1962); this involves only the atoms lying on a ray through \mathbf{R} , thus demonstrating in a wider class of cases that it is the forward-scattered waves which contribute most strongly at \mathbf{R} . Multiple scattering also brings into question the conclusion of RH that if the film includes crystallites these have the wurtzite structure. Their calculations are based on the assumption that the observed $I(\mathbf{K})$ arises from single scattering, which cannot be correct in this case (similar considerations apply to attempts to fit $I(\mathbf{K})$ by a random network structure).

In these experiments where the wave is a random function, interference images give no information about the structure which cannot be obtained more simply from the diffraction pattern $I(\mathbf{K})$. When either the Born or phase-grating approximation is valid, $I(\mathbf{K})$ depends only on the radial distribution function $g(r)$ in the film. In more 'dynamical' situations, involving more multiple scattering, $I(\mathbf{K})$ depends on higher-order correlations in addition to $g(r)$, but the theory has not yet been fully worked out.

Chaudhari *et al* (1972) consider single scattering from a random cluster of atoms; they compute the diffraction pattern $I(\mathbf{K})$, but not the image $\psi_{\text{p}}(\mathbf{R})$, so that their results do not bear directly on the question of interpreting fringes. In any case, it should not be forgotten that, according to our arguments, the image of a random cluster will show periodic fringes only if the cluster is large enough for the diffraction pattern $I(\mathbf{K})$ to have a clearly defined ring structure.

Our conclusion is that recent electron micrographs cannot be employed to distinguish between either the random network or the crystallite model of amorphous films.

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References

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