

Evidence from simulations for orientational medium range order in fluctuation-electron-microscopy observations of a-Si

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The recent technique of fluctuation electron microscopy (FEM) [1] has been shown to be sensitive to two-, three- and four- atom correlations in disordered materials. The FEM signal is the statistical variance in the spatial image intensity distribution $V(k, Q) = \left[\frac{\langle I^2(r, k, Q) \rangle_r}{\langle I(r, k, Q) \rangle_r^2} \right] - 1$, where $I(r, k, Q)$ is the diffracted image intensity, r is the position image, k is the dark field scattering vector magnitude, $0.61/Q$ is objective aperture size, while $\langle \dots \rangle_r$ denotes a spatial average over the image. In a-Si and a-Ge, both the structure factor and $V(k)$ consistently show two broad peaks at $k_1 = 3.2 \text{ nm}^{-1}$ and $k_2 = 5.5 \text{ nm}^{-1}$. The paracrystalline model (pc-Si) of a-Si consisting of nano-sized, strained crystalline grains embedded in a continuous random network (CRN) matrix is consistent with FEM data. The magnitude of $V(k)$ varies with the paracrystallite size and volume fraction, but the first peak is *always* higher than the second as observed in hydrogenated a-Si:H and pure a-Si sputtered at low substrate temperatures. However, a-Si films deposited at substrate temperature $T > 250^\circ \text{C}$, or under ion-bombardment during growth, and for a-Ge films grown by evaporation, the second peak is higher. No explanation existed for this observation [2].

To investigate these differences, 12 realistic models of pc-Si were constructed using a modified WWW algorithm, each consisting of 1000 atoms in a cubic box. A fraction f_g ($\approx 0.1, 0.2, 0.3$ and 0.4) of the atoms were in m ($= 1, 2, \text{ or } 4$) paracrystalline grains while the remaining atoms were in a continuous random network (CRN) matrix. A CRN model ($f_g = m = 0$) was also constructed. All these models give an average bond-angle of $109^\circ \pm 10^\circ$, and a bond length of $0.235 \pm 0.01 \text{ nm}$, characteristic of the best a-Si models. Some basic results are [3]: (i) For the 12 pc-Si models $V(k)$ and $I(k)$ show two peaks, the first at k_1 and the second one at k_2 . (ii) For a given grain fraction f_g , the $V(k)$ signal decreases with m . (iii) For a given m , the $V(k)$ magnitude decreases with f_g . (v) $V(k)$ is insensitive to small strains.

We found that the second peak is higher than the first only for models containing a significant volume fraction of grains with a local preferred orientation or with significant shape anisotropy. Hence we show that FEM can detect textural and orientational ordering not seen in diffraction experiments. This conclusion is suggested to be general.

We first consider the effects of relative grain orientation. Fig. 1 shows calculated $V(k)$ curves for the CRN and 4 models of pc-Si. The unfilled (filled) circles show a model M1 (M2) with four spherical grains and $f_g = 0.43$. Model M1 has its grains randomly oriented while M2 has them all aligned along the same crystal axes. M2 has a lower overall magnitude of $V(k)$ than M1 because it has less spatial variation in diffraction; all of its grains diffract together. However, the second peak for aligned case M2

is higher than its first peak while for M1 the reverse is true. Similar models, with $f_g = 0.21$ (squares) and $f_g = 0.3$ (not shown) show no such peak height reversal. This suggests that a minimum fraction $f_{gc} \approx 0.4$ of *aligned* spherical grains is necessary for the second peak to be greater than the first. However there is no global preferred orientation implied by a larger second peak in $V(k)$. In calculating $V(k)$, each model is rotated many times with respect to the incident beam direction, so the effective sample consists of locally aligned regions, but is globally isotropic. This observation is consistent with recent observations of $V(k)$ which showed no global preferred orientation in the MRO of a-Si [2].

The shape and orientation of individual paracrystallites can also influence the relative peak heights. Fig. 2 shows $V(k)$ curves for three cylinders of perfect Si crystal with length equal to the diameter. The cylinder axes are along $[111]$, $[110]$, and $[113]$. The highest peaks $V(k)$ in each case occur at the k corresponding to the inter-planar spacings parallel to the axis, $k_{111} = 3.2 \text{ nm}^{-1}$, $k_{110} = 5.2 \text{ nm}^{-1}$, and $k_{113} = 6.1 \text{ nm}^{-1}$. The change in relative peak heights with grain shape anisotropy is evident.

References

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- [4] We acknowledge support from the National Science Foundation under grant DMR02-05858.

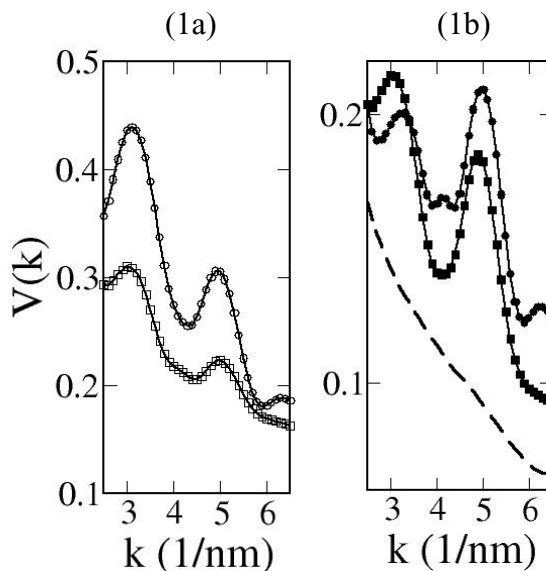


Figure 1. FEM signal $V(k)$ for four pc-Si models each with $m = 4$ grains occupying a fraction $f_g = 0.43$ (circles) or $f_g = 0.21$ (squares) of the model atoms. A CRN is shown by the dashed line in (b). Models with four grains: (a) having random relative rotations (unfilled symbols) and (b) all having the same orientation (filled symbols).

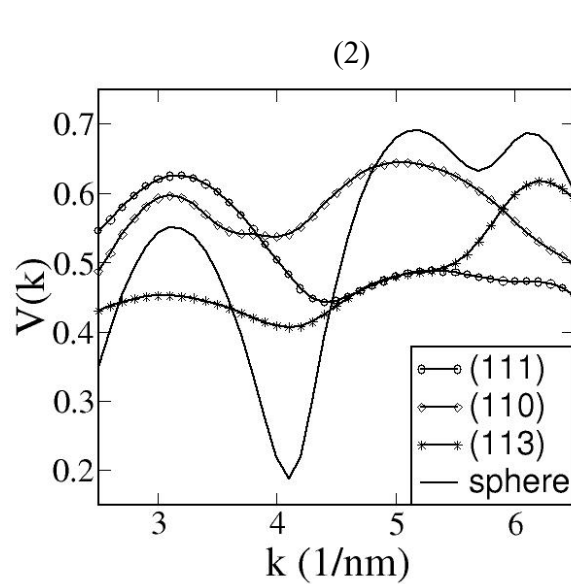


Figure 2. FEM signal $V(k)$ for grains of unstrained c-Si with no matrix. Symbols are for cylinders with their lengths equal to their diameters and cylinder axis along the indicated crystal direction. The solid line is for a sphere.