Comment on "Submicrocrystallites and Orientational Proximity Effect"

In a recent Letter,[1] Ourmazd, Bean, and Phillips have reported a new effect which they named "orientational proximity effect," on the basis of the interpretation of high-resolution electron microscopy (HREM) images of an *a*-Si/*c*-Si interface. Their claims were twofold: (1) the existence of submicrocrystals (1.5 to 2.0 nm diameter) in amorphous Si, and (2) the preferred orientation of these submicrocrystals near the *a*-Si/*c*-Si boundary. We have found that in each case the evidence supporting the effect is not convincing.

Regarding point (1), the evidence was that small patches of fringes appeared in the HREM images of amorphous thin films. However, it is known that such features may arise in images of amorphous films purely by chance[2] and may be influenced by a number of factors including misalignment of the microscope, effects of the transfer function of the objective lens, and limitations imposed by the objective aperture.[3,4] We have simulated[5] HREM images of white-noise phase objects in which local patches of fringes also occur. Statistical comparisons have been made with use of an autocorrelation analysis technique developed by Fan and Cowley.[6] The local ordering over distances of 1.5 nm in images of *a*-Si film with thickness 2 nm is not significantly different from that for images derived from purely random white noise, under the conditions of a JEM-200CX electron microscope whose resolution (0.27 nm) should be comparable with that of the Phillips 420ST used by Ourmazd, Bean, and Phillips. This result does not exclude the possibility that microcrystallites exist for *a*-Si, but it does cast doubt on the reliability of conclusions based on HREM images made with this sort of resolution.

On point (2), we have simulated the effect of limited resolution on imaging of the *a*-Si/*c*-Si interface. Figure 1(a) is a composite image of a perfect *c*-Si image in [110] orientation interfaced with a computer-generated image of white noise. Figure 1(b) shows the effect of the transfer function when the spectrum is multiplied by an aperture function of size 3.7 nm^{-1} so that information beyond the (200) reflection is cut off, limiting resolution to 2.7 Å. Although the crystal part is essentially the same, the amorphous part has changed drastically. There is a very clear impression of a continuation of the lattice fringes of the crystal part into the amorphous part, and also an impression of preferred orientation in the detail of the amorphous image. This is because an aperture or the transfer function can eliminate the information concerning the termination of the crystal lattice which is contained in streaks in the diffraction pattern running through the strong diffraction spots. The impression is given that the patches of fringes well beyond the interface show some alignment as a result of this artifact. (Fringes on top of the figure are due to periodic continuation and are not part of our argument.) The appearance of alignment is comparable with that shown in the HREM images of Ourmazd, Bean, and Phillips.

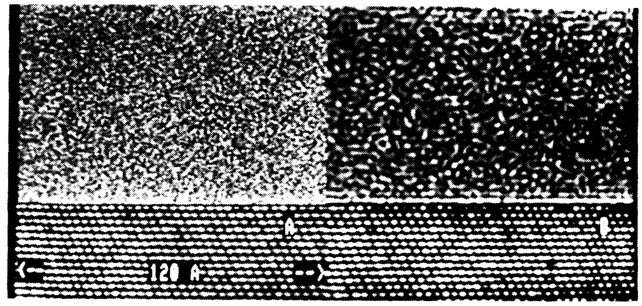

FIG. 1. Simulated images of a random white-noise object interfaced with *c*-Si in [110] orientation: (a) with no aperture limitation; (b) with aperture limitation 3.7 nm^{-1}.

In summary, our simulations suggest that recent interpretations of HREM images in terms of microcrystalline regions and proximity effect must be questioned. While it is not to be concluded that microcrystals and proximity effects do not exist, it is suggested that the evidence from HREM observations to date must be regarded as inconclusive.

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Note added.—We anticipate that images of *a*-Si obtained with better resolution (e.g., 1.6 Å with the JEM 4000EX) will show even clearer patches of fringes since our autocorrelation analysis has shown clear differences from random white-noise simulations in this case, but the question as to whether such fringes indicate crystallinity is not resolved, and awaits further analysis.

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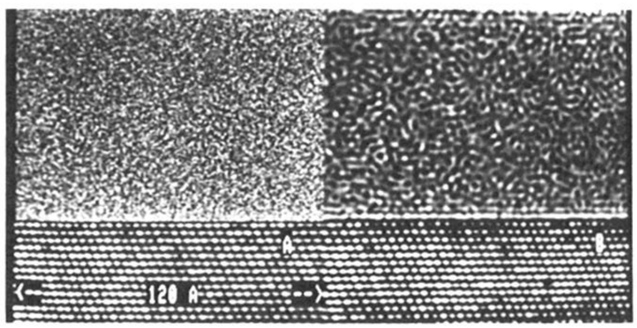

FIG. 1. Simulated images of a random white-noise object interfaced with c-Si in [110] orientation: (a) with no aperture limitation; (b) with aperture limitation 3.7 nm^{-1}.