Thin Films and Coatings

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HRTEM Image Simulation of Model Structures for Fullerene-Like Nanotructures

Z. Czigany[1]

[1]Institute for Technical Physics and Materials Science in RCNS of HAS, Thin Film Physics, Budapest, Hungary

czigany.zsolt@ttk.mta.hu
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Carbon based amorphous and fullerene–like (FL) thin films have a great potential in applications due to their mechanical resiliency. However, high resolution transmission electron microscopy (HRTEM) imaging of fullerene-like structures in the solid phase is limited by the overlap of spherical nm-sized fullerene-like features as reported recently for CN_x [1] and CP_x [2] thin films. In this paper the prospects and limitations of HRTEM imaging is investigated by image simulations.

Model structures were created for CN_x and CP_x thin films using close packed clusters of nano-onions of encapsulated Goldberg polyhedra (of C_{60} C_{240} and C_{540}) and P doped randomized C_{20} fullerites [3], respectively. The 10at% P doping in the model for CP_x was implemented by application of $C_{18}P_2$ isomers [3].

HRTEM image simulations were made by the JEMS program (by P. Stadelmann) using the multislice method [4]. Conventional and C_s corrected HRTEM images were simulated with the parameters of FEI Tecnai G^2 (200 kV, equipped with ultratwin objective lens; point resolution of 0.19nm) and FEI Titan (300 kV) microscopes, respectively. Conventional HRTEM images were calculated at Scherzer defocus. For the C_s corrected HRTEM images optimized parameters were applied to minimize the point spread [5]. For 300kV acceleration voltage and $g_I \approx 15nm^{-1}$ information limit C_s=7.5µm and -4.5nm defocus were applied.

Results of conventional (non C_s corrected) HRTEM image simulations of a nano-onion ensemble in two orientations are shown in Fig.1. For small overlap of two 3 shell nano-onions faceting like artificial fringes appear at the shell coincidences which is demonstrated in a symmetric arrangement in Fig.1c. In general, if the overlap of two 3 shell nano-onions exceeds 50% the interpretation of the image is uncertain. Overlap of more than two onions results in amorphous-like appearance of the image (Fig.1d). The features in the image depend strongly on the defocus in agreement with experimental observations [1]. In C_s corrected image simulations (Fig.2 – simulated image of Fig.1b arrangement) the innermost shell is not visible even in the non-overlapping onions at the periphery of the cluster. At the overlapping central region of the cluster the structure has an amorphous-like appearance with a finer scale pattern compared to non C_s corrected images.

Simulated HRTEM images of clusters of $C_{18}P_2$ cages, containing 13, 20, 40, 60 and 90 cages for FEI Tecnai G^2 microscope at Scherzer defocus and FEI Titan microscope are compared in Figure 3. For the model clusters of close packed $C_{18}P_2$ molecules (built for FL-CP_x) the contrast of fringes is very similar for different cluster sizes and multiplicity of overlaps. An amorphous appearance dominates the image over a few nm specimen thicknesses. In the simulated C_s corrected images the slightest overlap, producing ~1.6nm specimen thickness, may provide an amorphous appearance of the structure with finer scale pattern compared to the non C_s corrected case.

The simulated HRTEM images of the above model structures are also in agreement with the experimental observation [1,2] and provide a general indication, that quite complex nanostructured materials may have amorphous-like appearance in real TEM observations – independent of the actual resolution [6].

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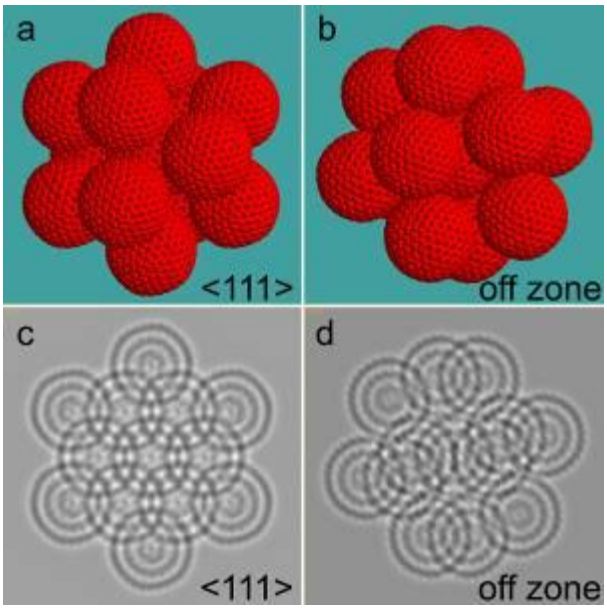

Figure 1. Close packed (fcc) clusters of 13 pieces of 3-shell nano-onions in different orientations are depicted in panels (a and b) and corresponding simulated HRTEM images calculated for FEI Tecnai G^2 microscope at Scherzer defocus (-43nm) are shown in panels (c and d). The panels are 8 nm wide. False fringes and confusion of shell structure can be recognized in the central region of the cluster for <111> and "off zone" orientations, respectively, due to multiple overlap.

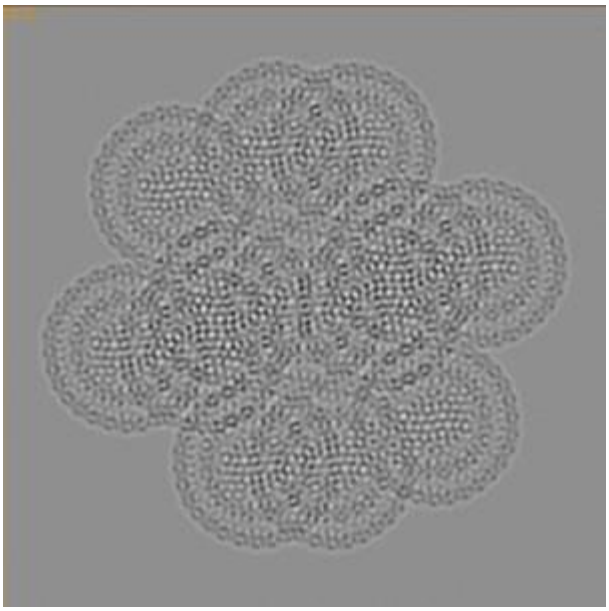

Figure 2. Simulated HRTEM image of a close packed (fcc) clusters of 13 pieces of 3-shell nano-onions in off zone orientation (Fig. 1b) calculated for FEI Titan microscope at C_s=7.5μm and Δf=-4.5nm (least confusion). The panel is 8 nm wide. The inner onion shells are not visible and the image has an amorphous appearance even in the non overlapping areas.

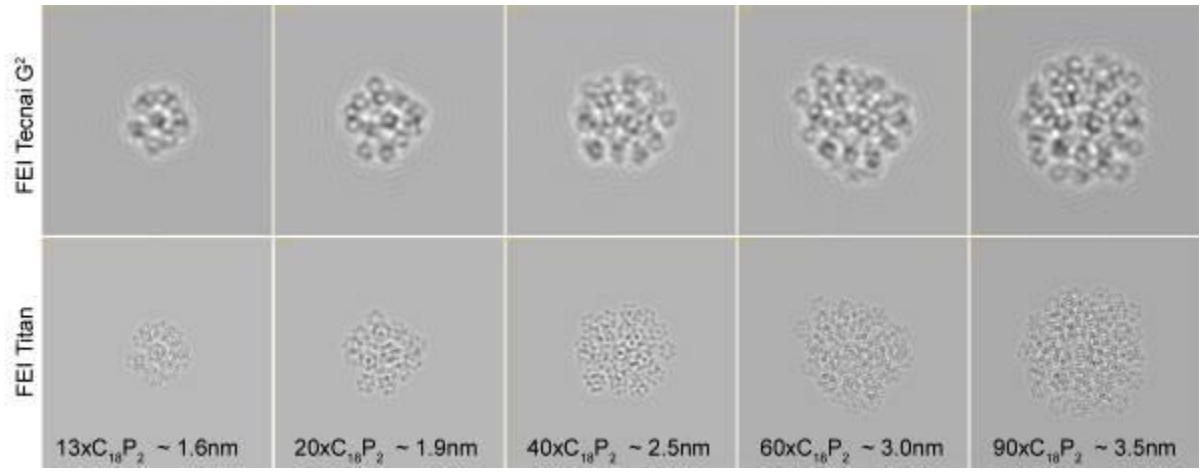

Figure 3. Simulated HRTEM images of $C_{18}P_2$ clusters containing 13, 20, 40, 60 and 90 cages for FEI Tecnai G^2 microscope at Scherzer defocus (upper line of images) and FEI Titan microscope (lower line of images). The panels are 5.5 nm wide. The corresponding cluster diameters are given in each panel. In all images of clusters for FEI Tecnai G^2 microscope small circles (resembling the projection of an individual C_{20} sized cage) can be recognized both at the periphery of the clusters and at the centre, where cage overlap is obvious. The image for C_s corrected (FEI Titan microscope) has very similar appearance having at finer scale reminding of an amorphous appearance.