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## Direct Gas-Phase Formation of Complex Core-Shell and Three-Layer Mn-Bi Nanoparticles †

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### 1 High-resolution TEM Images

The images in this section are intended to show the difference between core-shell and three layer morphology as seen in high resolution images. To claim a separate layer is present within the particle using high-resolution image becomes difficult because the core region is amorphous. An amorphous region can easily be misconstrued for an off- zone axis particle or can be due to focusing conditions. We have looked into several particles and the common feature of all the three layer particles is a core, which has no lattice fringes. In case of a core-shell particle, the core always has lattice fringes. Given below are two Figures 1 and 2, which show these characteristic features.

### 2 Quantitative Analysis of STEM Image Contrast

Figure 3, presented in the main text, is a grayscale image with pixel intensities ranging from 0 (dark)-255(bright). Supplementary figure 3 (a,b) show the original image and the intensity distribution of it. It is evident from Figure 3b that there is considerable amount of variation in the gray area of the image. This variation is shown within the image by selectively choosing the grayscale intensity range. Figure 3(c-e) show the areas corresponding to gray scale intensity range 50-100, 200-220, and >230. Figure 3c and 3d correspond to the shell and the regions marked with arrow within the particles, respectively. Figure 3e shows the brightest parts of the particles. These intensity maps, without enhancing image attributes such as brightness, contrast, gamma, etc., clearly

show variation in intensity at different parts of the particles. The results are combined into a false color map (supplementary figure 4b) and presented along with the original grayscale map for comparison (supplementary Figure 4a). As the contrast in a HAADF-STEM image comes primarily from atomic number Z, elemental mapping were further performed on these images.

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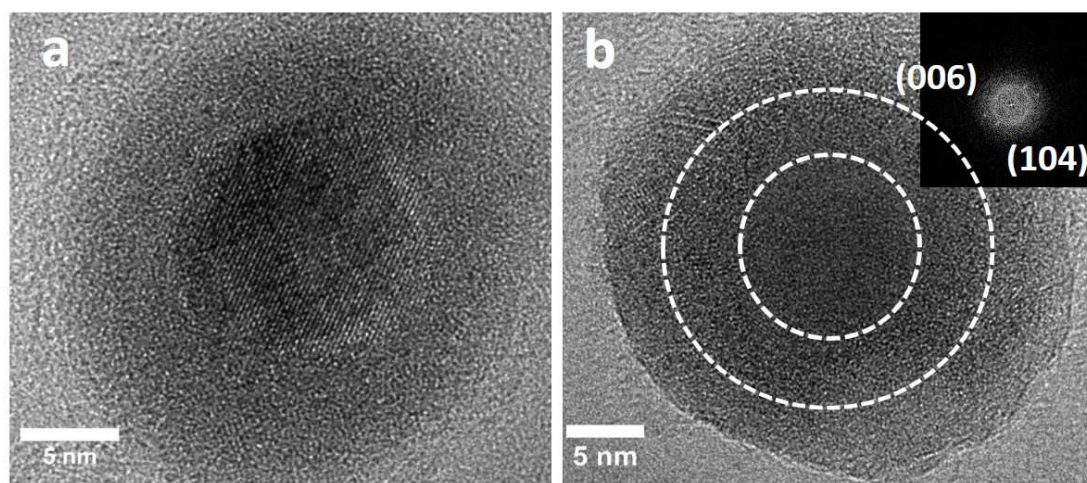
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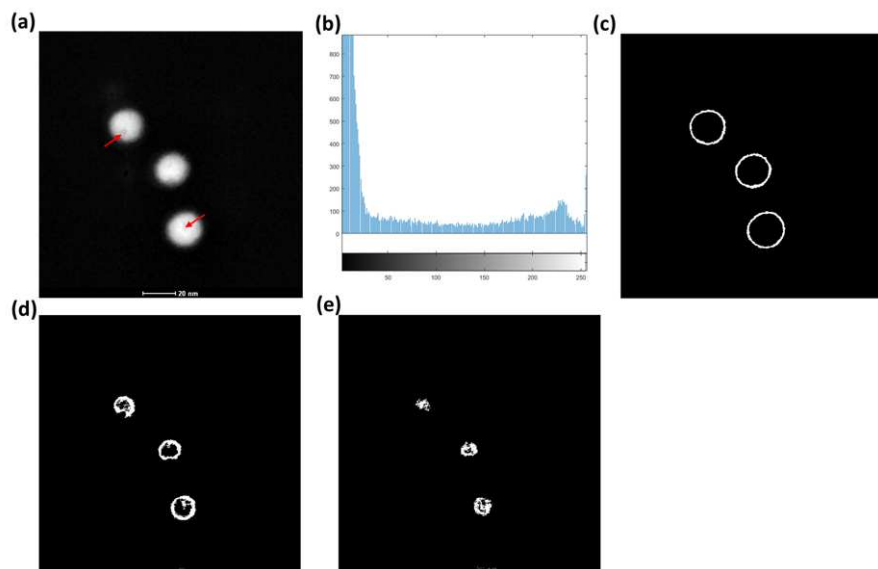
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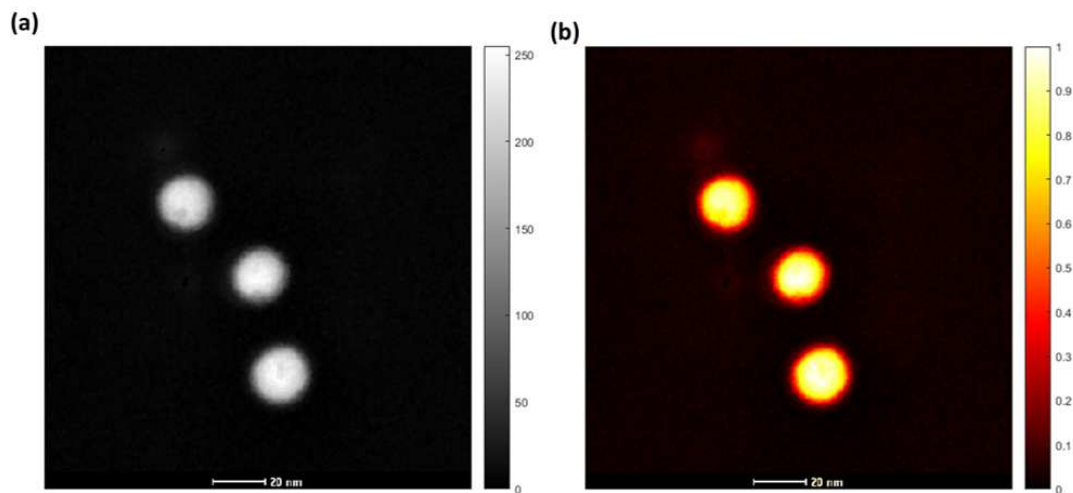
**Fig. 1** (a-c) The high-resolution images of a three layer nanoparticle taken at three different defocus conditions. The images in a, b, and c are taken in underfocus, Scherzer defocus, and overfocus conditions. In all three conditions, the core of the particle shows no lattice fringes, while the annular region show lattice fringes. The position of the core shifts a small amount as the focusing conditions are changed.



**Fig. 2** (a) The high-resolution image of a core-shell nanoparticle. The core shows clear lattice fringes. (b) The three layer particle has an amorphous core and the layer surrounding the core has faint lattice fringes. (Inset) The fast Fourier transformation of the annular region show that the spots belong to a crystalline Bi phase.



**Fig. 3** (a) HAADF-STEM image of Mn-Bi nanoparticles. The shells and the regions marked with arrow show different contrast than rest of the areas of the particles. (b) The frequency distribution of pixel intensities of figure (a). The pixel intensity range from 0 (dark)-255(bright). (c-e) The map of selected grayscale contrasts with pixel intensity range of 50-100, 200-220, and >230 are shown. The white regions in image c and d correspond to shell and areas marked with arrows in (a), respectively. The white regions in figure(e) correspond to the brightest parts of figure (a).



**Fig. 4** (a,b) The original grayscale image (figure 3a) along with the false color image of the same show the core-shell nature of the Mn-Bi nanoparticles.