1850
doi:10.1017/S1431927610059428
Microsc. Microanal. 16 (Suppl 2), 2010

Quantitative Analysis of Electron Tomograms using Auto- and Cross-Correlations

H. Friedrich*, C. J. Gommes**, P.E. de Jongh*, K. P. de Jong*

* Inorganic Chemistry and Catalysis, Utrecht University, Sorbonnelaan 16, 3584 Utrecht;
HF present address: Materials & Interface Chemistry and Soft Matter CryoTEM Research Unit, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands, h.friedrich@tue.nl
** Department of Chemical Engineering, University of Liège, Allée du 6 août 3, 4000 Liège, Belgium

In the last decade the range of materials studied by electron tomography (ET) has extended dramatically. The recent interest, in particular by the physical and chemical sciences, is strongly connected to advances in shaping and patterning materials on the nanoscale. The ability of ET to give detailed insight into the three dimensional morphology of complex nanostructures has considerably contributed to the understanding of their physico-chemical properties. While in many instances a qualitative description of structural characteristics, e.g. pore connectivity in zeolites or distribution of active components in supported catalysts, are sufficient more frequently a detailed quantification on the nanoscale, e.g. pore size distributions, and particle size distributions and particle-particle distances is required. [1] In this paper the image analysis of electron tomograms based on auto- and cross-correlations (CC) for a quantitative description of ordered mesoporous silica and nanocrystal (NC) superlattices are discussed.

SBA-15 ordered mesoporous silica was analyzed in terms of irregular cylinders with a circular cross section (CC template), the centers and radii of which are variable along the pores (Fig. 1a). This enables the quantitative characterization of pore roughness in terms of the statistical distribution of the pore diameters (Fig. 1b), and of the local deviation between the positions of the pore centers and the closest hexagonal lattice point (Fig. 1c). Variations of the mesopore centers and radii combined result in pore wall corrugation with amplitude of 1-2 nm and correlation length of 4-5 nm along the pore axis. The amplitude of corrugation agreed well with the value of the microporous corona obtained from SAXS measurements. [2] In case that the signal-to-noise ratio of electron tomograms is too low for a reliable segmentation-based image analysis, the three-dimensional grey-tone auto-correlation function can be analyzed in terms of the same morphological model of the material. [3]

For the NC superlattices an analysis by CC with 3D synthetic templates of varying size was carried out. Templates were defined as spherical dark regions (of value -1), surrounded by a bright shell (of value +1). This template model corresponds well with the appearance of the NC in the tomogram exposing dark cores and a bright rim. The bright rim results from underfocus condition during acquisition well as the reconstruction point spread function. CC provided the NC coordinates while NC core diameters were obtained from the radial intensity profiles. This enabled us to characterize thousands of NC in each tomogram containing less than 5 % false positives for high-contrast PbSe and Au NCs and around 10% false positives for the low-contrast CdSe NCs. Subsequently, positional order, but also local symmetries as revealed by local bondorder parameters were exploited to find individual crystallites. Based on the underlying superlattice, and besides characterizing point and plane defects, deviations from translational symmetry resulting in strain fields were observed (Fig. 2). The modular and versatile approach demonstrated on binary NC superlattices [4] can be easily extended to the first truly ternary structure [5] that we recently imaged by ET.

We have highlighted the versatility of auto- and cross-correlations for the quantitative 3D analysis of nanostructured materials which will be key for a more rational design of this important class of materials in the future.

[1] H. Friedrich, P.E. de Jongh, A.J. Verkleij, K.P. de Jong, *Chem. Rev.* 109 (2009) 1619.
[2] C.J. Gommes, H. Friedrich, M. Wolters, P. de Jongh, K.P. de Jong, *Chem. Mat.* 21 (2009) 1311.
[3] C.J. Gommes, H. Friedrich, P.E. de Jongh, K.P. de Jong, *Acta Mater.*, 58, (2010) 770.
[4] H. Friedrich, et al., Nano Lett. 9 (2009) 2719.
[5] W.H. Evers, H. Friedrich, L. Filion, M. Dijkstra, D. Vanmaekelbergh, *Angew. Chem. Int. Ed.* 48 (2009) 9655.

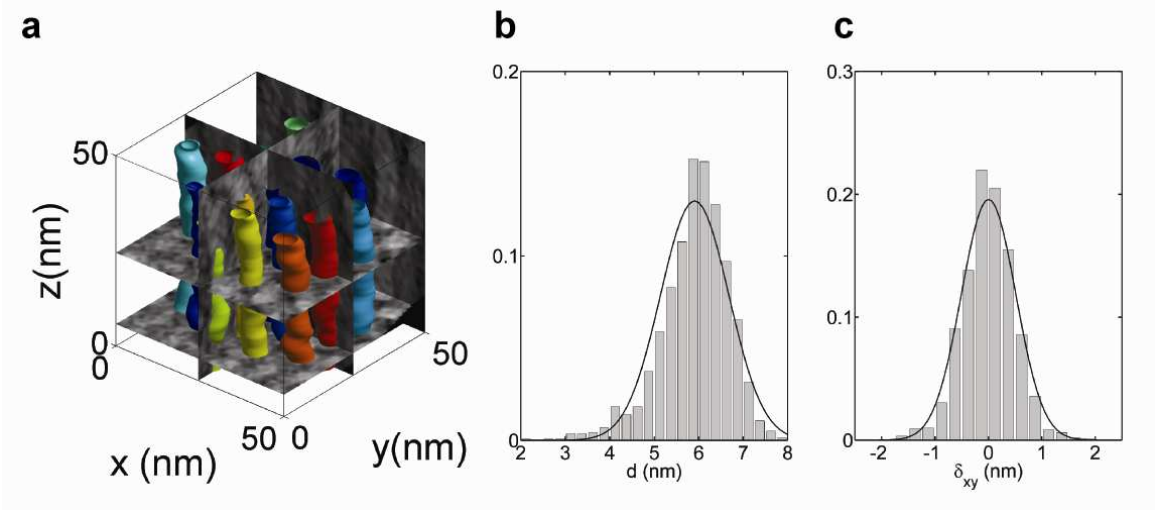

FIG. 1. Example of electron tomogram of SBA-15 with segmented mesopores (a), together with the corresponding distribution of pores diameters (b) and of the deviation between pores centers and the closest hexagonal lattice (c).

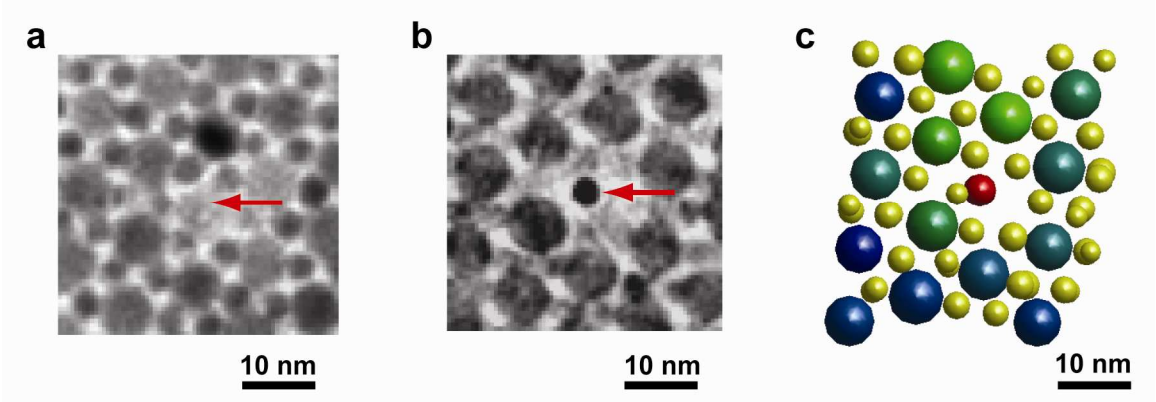

FIG. 2. Three dimensional analysis of nanocrystal (NC) superlattice composed of 7.1 nm PbSe NCs and 3.7 nm Au NCs. While in the TEM projection at zero tilt (a) the substitution, indicated by red arrow, is almost invisible, a numerical cross-section through the superlattice (b) clearly reveals the defect. Although, a small Au NC (black) has replaced a much larger PbSe NC (dark gray) the lattice appears stretched around the defect. A quantitative analysis (c) shows PbSe NCs color coded relative to their displacement (blue= on lattice; green= ~2.5 nm off lattice), Au NCs in yellow, and the substitution in red.

https://doi.org/10.1017/S1431927610059428 Published online by Cambridge University Press