

Relationship between atomistic arrangement and internal nanocrystal superstructure ordering within PbS-organic mesocrystals

L. Liebscher^a, W. Carrillo-Cabrera^b, E. Rosseeva^b, P. Simon^b, S. G. Hickey^a, A. Eychmüller^a

^a Physical Chemistry, TU Dresden, Bergstraße 66b, 01062 Dresden, Germany
L.Liebscher@chemie.tu-dresden.de

^b Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany

Nanoscale materials show behavior which is intermediate between the properties of single molecules or atoms and the properties of bulk materials. Therefore by controlling the dimensions of materials in the nanometer size regime a number of material properties which demonstrate size dependence can be tuned. For example semiconductor nanocrystals (NC) show size-dependent optical-properties, magnetic NCs show size-dependent magnetic-properties (superparamagnetism), including transitions from paramagnetism to ferromagnetism and noble-metal NCs possess size-tunable plasmonic resonances.^[1] Through the development of new optimized synthesis strategies highly monodisperse NCs can be produced, which have potential applications in optical, magnetic or electronic devices.

2D and 3D arrangements of NCs combine the properties of individual NCs with new features that derive from the unique collective properties of arrays of ordered particles. These particle arrangements have further promising applications as light emitting devices, photodetectors, thermoelectric heat-to-electricity converters and solar cells^[2]. Because of the potential locked within these new material sets the field of interest is shifting away from the synthesis of the nanoparticles as building blocks to the assembly of these small units into hierarchical structures.

In this contribution we present a novel method, using the gas phase diffusion technique (Figure 1), for the formation of hierarchal architectures (mesocrystals, superstructures) with spherical lead chalcogenide NCs as building blocks. The mesocrystal dimensions are increased by up to six times (300µm) using this new destabilization technique as compared to the technique employed by Podsiadlo *et al*^[1]. After thinning the mesocrystals using FIB, the inner crystallographic arrangement of the building blocks and the 3D arrangement of the NCs could be visualized. TEM analyses (Figure 2) have shown the relationship of the superstructure crystal arrangement to the crystal structure of the individual NCs and their facets and simulations provide insights into the superlattice packing. Electron holography studies demonstrate that the NCs in the superstructure arrangement are of the same size and morphology as before the destabilization process.

References

- [1] P. Podsiadlo *et al.*, Nano Lett., Article ASAP, DOI: 10.1021/nl103587u
 [2] S. M. Rupich *et al.*, JACS **132** (2010) 289.

Figures

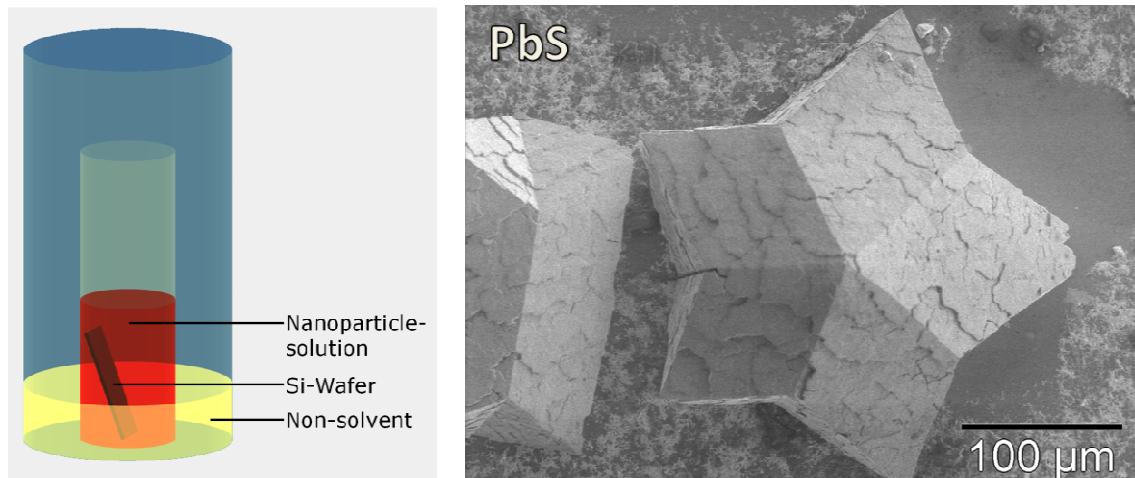


Figure 1: Gas phase diffusion technique and PbS-mesocrystal.

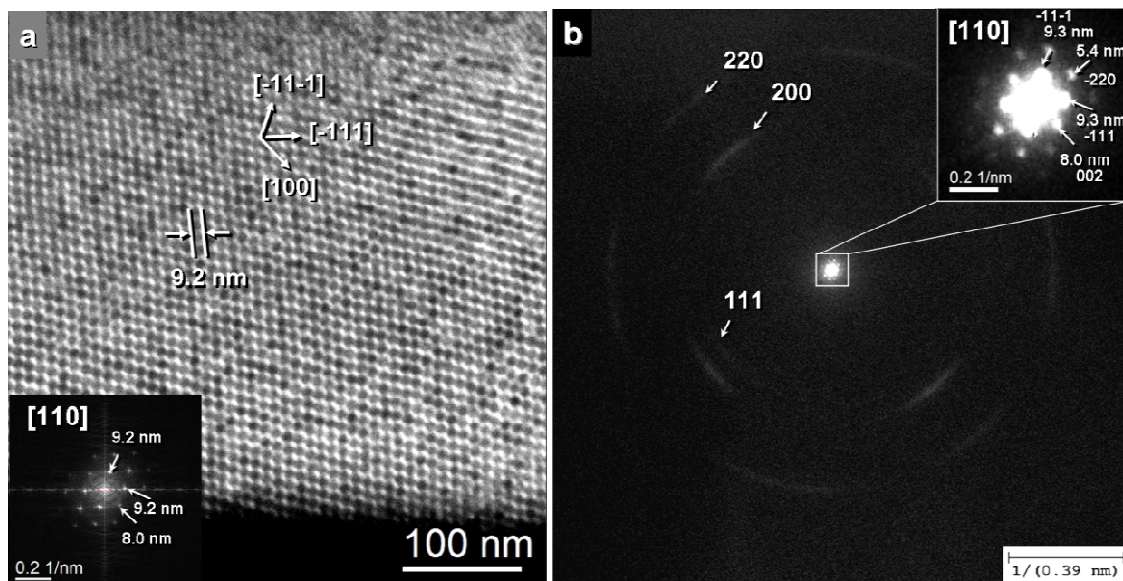


Figure 2: (a) Overview TEM image of FIB-cut with corresponding fast Fourier transform (FFT) inset, showing the superstructure ordering of the nanoparticles. (b) Electron diffraction from the same area visualizing relationship between superstructure (inset - zoomed small angle area) and PbS (galena) nanocrystals reflections