

ESRF	Experiment title:Two dimensional semiconductor structures with nanoscale honeycomb structuring	Experiment number : HC890
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Report:

The goal of the experiment was to investigate lattice coherence in self-assembled superlattices of PbSe nanocrystals.

Recently, solution grown, 5nm diameter PbSe nanocrystals have been shown to be able to rapidly self- organise into large-scale twodimensional superlattices.[1] Drop-casting a dispersion of nanocrystals onto another, immiscible liquid under the right conditions leads to the rapid formation of tunable superlattices. Temperature, concentration and surface ligands can be tuned to produce linear aggregates, square lattices (figure 1a) or honeycomb-like structures (figure 1b). Upon assembly, the nanocrystals seem to form atomically coherent structures, meaning they essentially fuse into single crystals. The goal of this experiment was to determine the crystallinity of such lattices and investigate the range of atomic positional correlations. For this, use was made of nanofocused X-ray diffraction at beamline ID01.

Method:

The goals of the experiment was to find the range of atomic positional correlations through measurements of the width of the in-plane components of Bragg peaks and possibly their fine structure. To this end, lattices were made on Si 001 single crystal substrates for

measurements in Bragg geometry, as well as on Si_3N_4 membranes for Laue geometry measurements.

In order to get information on the lattice coherence within the monolayers, peaks with as large as possible in-plane components were measured. This prevented influences of long-range lattice deformations on the peak width.



Figure 1: Honeycomb structures of self-assembled PbSe nanoparticles. Scale bars in the left and right image are 60nm and 10 nm, respectively.

Although intensity was too low to obtain peak fine structures, widths of several reflections could be measured and used to determine atomic coherence using the Scherrer formula. For example, for the 111 specular reflection of the honeycomb structures, the peak width gave a lattice coherence of 4.8 nm, a slight underestimate of the particle size of 5.8 nm. In contrast with that, the in-plane width of the 002 reflection at the same sample position gave a 25.4 nm lattice coherence, indicating that coherence is preserved over at least 5 particles everywhere in the whole lattice captured in the 600x380 nm² beam. This value seems to correspond the the size of differently shaded regions in figure 1b, which is a TEM micrograph of the same synthesis. Different shades in TEM measurements correspond to different lattice orientations. Nanoparticle orientation therefore seems to gradually change over several particle diameters, confirming our observations at ID-01.

The extent of lattice coherence is quite impressive. First of all because the Scherrer formula tends to underestimate the extent of a crystal and the real coherence therefore entends over 25.4 nm. Second, the fact that nanoparticles can fuse into single crystals that are many times larger than their own size in a synthesis that can be completed in less than a minute opens the possibility to create tuneable exotic structures in single crystal thin films.

The predicted electronic properties of fully fused, long-range ordered nanocrystals include a graphene-like band gap, making these structures very interesting. Our studies contribute significantly to the knowledge that indeed long-range single crystals are present in these structures.

References:

[1] Evers, W., et al., Nano Letters, 13, 2317, (2013)