Determining the full 3-D adsorption geometry *in-situ* of PbSe nanocrystal monolayers at liquid-air interfaces

Jaco J. Geuchies[a,b], E. Geraffy[a], Carlo van Overbeek[a], Joep L. Peters[a], Marlou R. Slot[1], Federico M. Montanarella[1], Oleg Konovalov[b], Andrei V. Petukhov[c,d], and Daniel Vanmaekelbergh[a]

[a] *Condensed Matter and Interfaces, Debye Institute for Nanomaterials Science, Utrecht University, The Netherlands*
[b] *ID10, European Synchrotron Radiation Facility (ESRF), France*
[c] *Physical and Colloidal Chemistry, Debye Institute for Nanomaterials Science, Utrecht University, The Netherlands*
[d] *Laboratory of Physical Chemistry, Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, The Netherlands*

The adsorption and self-assembly of PbSe nanocrystals (NCs) at liquid-air interfaces has led to remarkable new materials, which show atomic order and a superimposed nanoscale geometry [1]. Recent experiments have unraveled the mechanism of formation of these superlattices, governed by remarkable phase transitions [2]. Following these results, we now focus on the adsorption geometry of PbSe NC monolayers at the liquid-air interface. We combine in-situ grazing-incidence small (wide) X-ray scattering (GIS(W)AXS) combined with X-ray reflectivity (XRR) to obtain the 3-D adsorption geometry of the nanocrystal monolayer

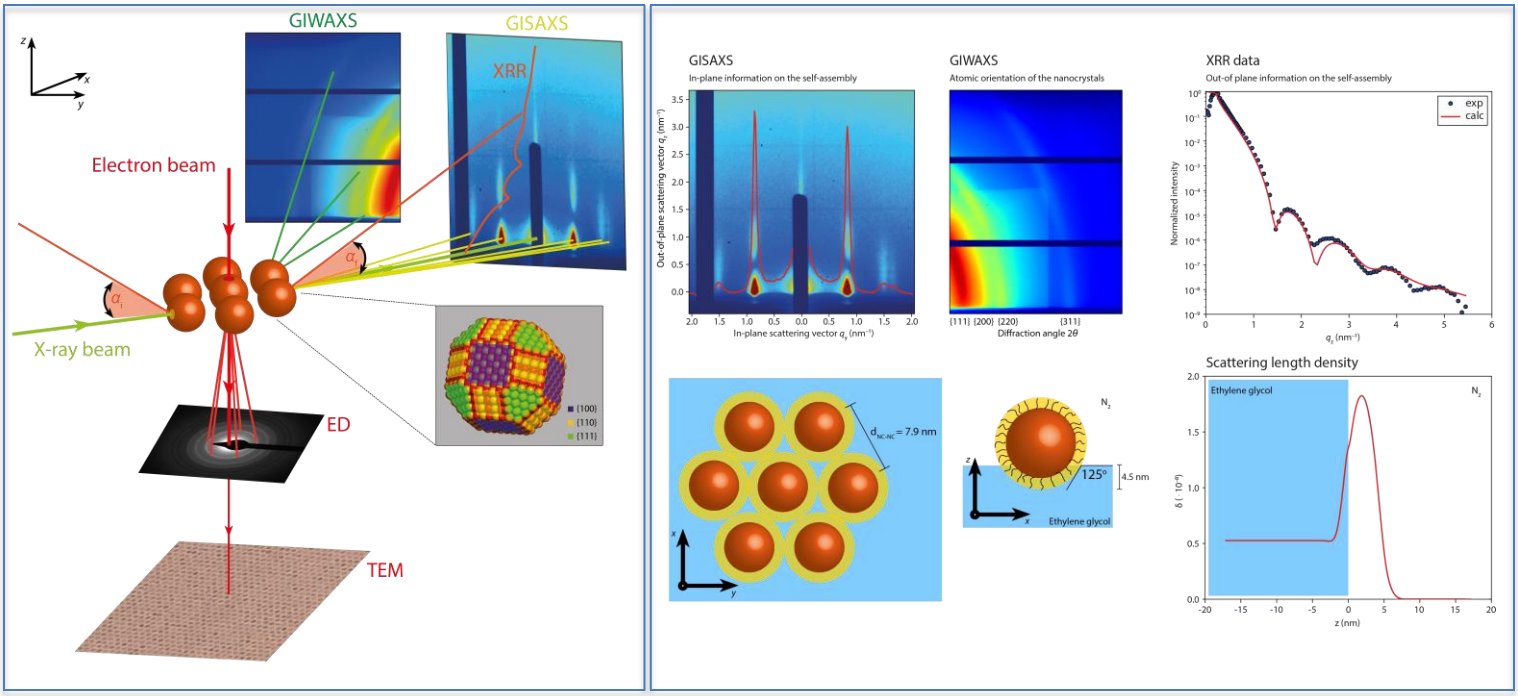

Fig. 1 Left: Experimental geometry of the experiments. The GISAXS patterns give information on the nanocrystal structure in the plane of the interface. The GIWAXS pattern gives information on the crystallographic orientation of the atomic lattice of the NCs. XRR gives the out-of plane scattering component and gives the electron density profile in the direction perpendicular to the liquid-air interface. **Right:** GISAXS/GIWAXS/XRR data and results obtained for 5.4 nm PbSe nanocrystals. Combining the data leads to a 3D in-situ model of the NCs at the liquid-air interface.

We show that all the nanocrystal monolayers attain an hexagonal symmetry in the plane of the EG-air interface from GISAXS. Moreover, we extract the electron density profile in the direction perpendicular to the liquid-air interface from the XRR measurements, which shows that all nanocrystals adsorb on the top part of this interface. The information obtained gives for the first time a full three-dimensional in-situ picture of PbSe nanocrystals adsorbed at a liquid-air interface. The adsorption geometry of the NCs in the early stages of oriented attachment are expected to have great impact on the atomically connected 2-D superlattices.

1) Boneschanscher, M. P. *et al.* *Science,* **2014** 1252642 doi: 10.1126/science.1252642.
2) Geuchies, J. J. *et al. Nat. Mater.,* **2016**, 15, 1248–1254.