Electronic Coupling in Two-Dimensional Arrays of PbSe Nanocrystals.

Peter Liljeroth[a], Karin Overgaag[a], Bruno Grandidier[b], Stephen G. Hickey[c], and Daniël Vanmaekelbergh[a]

[a] *Condensed Matter and Interfaces, Debye Institute, University of Utrecht, PO Box 80000, 3508TA Utrecht, the Netherlands.*

[b] *Institut d'Electronique, de Microélectronique et de Nanotechnologie, Département ISEN, 41 bd Vauban, 59046 Lille Cedex, France.*

[c] *Institute of Physical Chemistry, University of Hamburg, Grindelallee 117, D-20146 Hamburg, Germany*.

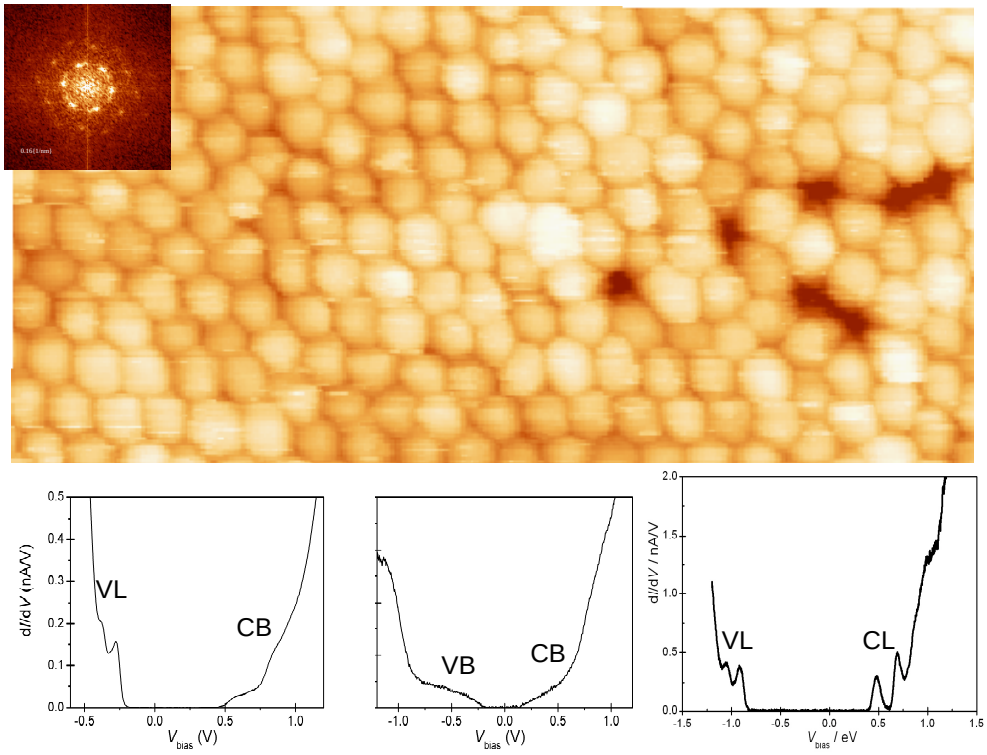
Two-dimensional arrays of semiconductor nanocrystals can be fabricated by simple drop-casting; if the quantum dots are sufficiently monodisperse, arrays with local translational order extending over hundreds of nanometres can be obtained. While electronic coupling in metallic nanocrystal superlattices has been demonstrated, it remains elusive in arrays of semiconductor quantum dots. The lower density of states at the bandedges and disorder effects could be important for this. For instance, the overlap between neighbouring quantum dot orbitals depends also on the rotational ordering of the crystals, i.e. relative orientation of the facets. It is thus possible that, even in nanocrystal superlattices possessing long-range translational order, strong electronic coupling between the particles can only prevail within limited regions of the array.

Scanning tunnelling microscopy and spectroscopy can be used to simultaneously probe the topography and the local density of states. Furthermore, tunnelling spectroscopy is carried out in the absence of selection rules, which can hamper the interpretation of optical absorbance spectra. We have carried out measurements on two-dimensional arrays of PbSe nanocrystals deposited on Au and HOPG substrates with UHV STM at low temperatures. We compare the tunneling spectra of single, individual nanocrystals with those acquired above nanocrystals in the array. While individual nanocrystals feature discrete energy levels, the density of states of PbSe nanocrystals in the array is step-like typical of a two-dimensional semiconductor. We observe that the conduction levels couple more strongly than the valence levels. This can be rationalized in terms of the tunnelling barrier height between the nanocrystals: higher lying conduction levels experience a lower effective barrier height.

Figure: Electronic coupling in an array of PbSe quantum dots probed by STM-STS.

Top: STM picture of a two-dimensional array of PbSe quantum dots after slight vacuum annaeling at 125 C. The STM picture reflects different origins of disorder in the array, due to (a) size dispersion of the NC building blocks (b) orientational disorder of faceted nanocrystals (c) “Crystallographic” defects such as a NC vacancy, an insertion of an NC line in the array.

Below: Density of states obtained with scanning tunneling spectroscopy of a PbSe quantum dots in the array featuring band-selective orbital coupling of the conduction levels (CV) (left), and coupling of the Valence Levels and Conduction Levels into VB and CB. The density of states of a single isolated quantum dot is shown at the bottom right.



References

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