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Assembling nanocrystal superstructures

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Publication date

2019

Document Version

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Citation for published version (APA):

Marino, E. (2019). *Assembling nanocrystal superstructures*.

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Outlook

Not all those who wander are lost.

— J.R.R. Tolkien

As conclusive chapter, we provide a brief personal outlook on the matters of nanoscience investigated in the body of this thesis. We place the advances presented in this thesis in perspective with other breakthroughs in nanocrystal synthesis and surface chemistry. Finally, we speculate about the future endeavors of nanoscience.

In this thesis, we have investigated novel approaches to drive the assembly of nanocrystals into solids with well-defined and reproducible morphologies. In multiple instances, we have shown how morphology defines the properties of the nanocrystal solid, demonstrating the importance of improving our understanding of nanocrystal interactions and assembly. Yet, even the most careful assembly approaches can prove to be ineffective when not in synergy with advances in nanocrystal synthesis and surface chemistry.

Semiconductor nanocrystals are often described as *artificial atoms*; however, the degree of energetic disorder present in nanocrystals intrinsically distinguishes them from their atomic counterparts. This energetic disorder derives from heterogeneity of nanocrystals in size, shape, and surface coverage across the ensemble, acting as a barrier against charge transport in nanocrystal solids. Therefore, to allow for the development of efficient channels for charge transport, there is a strong push towards atomically-defined nanocrystals.

The last 20 years of research resulted in the decrease of nanocrystal polydispersity from $\sim 5\%$ to $\sim 3\%$. [4, 29] An open question is how monodisperse do nanocrystals need to be to allow miniband formation. Nevertheless, the recent synthesis of colloidal nanoplatelets with atomically defined thicknesses, [125] accompanied by the rationalization of their formation mechanism, [136] suggest that it is only a matter of time until we can prepare truly monodisperse nanocrystals. Even then, controlling the nanocrystal surface chemistry will be just as key to the realization of nanocrystal solids with well-defined transport properties.

The ideal surface ligands grant nanocrystals colloidal stability in the dispersing solvent, while being conductive enough to allow the electronic wavefunction to leak to the neighboring nanocrystals in a solid, and passivating surface traps present at the surface. With these requirements in mind, the most promising approach might be using the recently synthesized molecular solders, inorganic polymers matching the composition of the nanocrystal core. [32] Here, the development of more accessible synthetic routes for these inorganic polymers may encourage their use. Yet, there might be interesting opportunities for organic conjugated molecules and polymers, [208] harvesting on the body of knowledge accumulated in the last 40 years of research on organic photovoltaics. [209] Finally, thanks to the atomically defined cores and ligand binding sites, the field of atomic clusters may suggest novel approaches in the reduction of nanocrystal disorder. [210]

Beyond these technical, yet paramount, considerations, new physics awaits. We envision properties of nanocrystal solids defined entirely by the choice of constituent nanocrystals so that each nanocrystal may be responsible for the interaction of the overall solid with a specific external stimulus. We imagine the position-dependent composition of nanocrystal solids to allow for novel effects that are more than the bare sum of the parts, controlling and coordinating orthogonal properties of the nanocrystal-

tal solid. The multidimensional parameter space spanned by nanocrystal composition, size, shape, surface coverage, ligand chemistry, and still morphology of the nanocrystal solid, provides a number of degrees of freedom that are simply unavailable in atomic solids; this feature should be exploited as a strength of nanocrystal solids, and this multi-dimensional phase space should be scanned by experiments and simulations, to identify regions where novel effects can be expected.