Hierarchical Assembly

Among the most promising yet challenging aspects of nanoscience is the hierarchical assembly of materials. Nature has given us many elegant examples of how this can be done and has shown us the extraordinary results of accomplishing this feat. We are just now decoding snippets of how such biological systems come together and function and co-opting some of these to create what are, by comparison, still primitive assemblies. At the same time, we are developing new methods of and strategies for assembly and are trying to determine the associated rules and scales. This is no simple task, as even measuring the assembled systems is at or beyond the limits of our current abilities.

A hallmark of work in this area has been the creativity of those involved, and one has the sense that we are exploring largely uncharted territory. Rarely are we able to get atomic-scale views of the results, which would serve as the best guide to further advances. Instead, we get lower resolution glimpses with available methods or see the results of our work less directly. In this issue, several articles and features touch on this topic from different points of view.[1–14]

In our Conversation, Prof. Ned Seeman discusses structural DNA nanotechnology, a field that he has developed by working out the associations between strands of DNA, exploiting some naturally occurring features and inventing related others.[1] You will see how the creative process outstripped our ability to measure these structures and has led to long incubation times and much frustration along the way. The measurements of these systems have become more direct over time, moving from gel electrophoresis, to Förster resonance energy transfer (FRET, an optical technique that measures proximity at the few nanometers scale), to atomic force microscopy at present; this has both accelerated the advances and enabled increasing complexity of the DNA and hybrid structures assembled. Prof. Seeman points us toward the next steps in this field as well as new enabling capabilities to come.

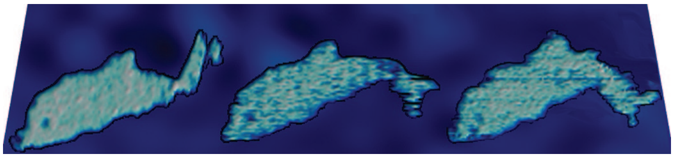

Figure 1. DNA origami of dolphin-shaped structures, as detailed in ref 2. Image courtesy of Flemming Besenbacher and Jørgen Kjems.

Such efforts have advanced sufficiently that features and properties can now be designed into these assemblies. Prof. Seeman describes a number of such advances. Also in this issue, Prof. Jørgen Kjems and co-workers have furthered the design tools for and elements of DNA origami (Figure 1), originally developed by Dr. Paul Rothemund.[2,15]

Prof. Dmitri Talapin explores competing interaction strengths and driving forces for hierarchical assembly in his Perspective.[3] He points out how these strengths scale differently for nanoparticles than they do for atoms and molecules. The result is that our intuition fails us, and we have to reconsider the interplay between these driving forces for hierarchical assembly. Often, the strengths are sufficiently close that we have not developed a predictive capability. Thus, systematic studies, such as those reported by Chen and O'Brien on binary nanoparticle superlattices,[4] or by Weller and co-workers on the preparation of highly ordered nanoparticle films,[5] will be critical to developing the basis for tuning the structures and thus properties of these hierarchically assembled materials.

Hierarchical assembly will likely be a key aspect of efficient solar cells since photoabsorbers and charge donors and acceptors must be placed not only in proximity but in optimized positions, in analogy to biological systems. In their article in our last issue, Prof. Ted Sargent and co-workers sought to capture the near-infrared portion of the solar spectrum more efficiently using molecularly coupled PbSe quantum dot arrays.[16] Several papers this month use clever techniques for the proximal placement of dyes and chromophores to sculpted semiconducting materials in order to couple light in or out.[6–8]

Prof. Steven Buratto and his group take this to the single- and few-molecule level in using the controllable properties of porous Si[17] and sensitive optical methods to probe the interactions of chromophores placed within the pores.[9] Such clever optical techniques give us glimpses into sites thus far inaccessible to imaging methods.

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In their Perspective, Yoshida and Lahann describe what "smart materials" can do, how such materials are used in nature for exquisitely complex functions, and how simple our current abilities are by comparison.[10] They suggest that one path to achieving greater function will be hierarchical assembly.

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Prof. Anna Balazs and her co-workers propose exploiting Janus nanoparticles as "artificial proteins" to open and to close pores in membranes controllably, and show through simulations that nanoparticles with hydrophilic and hydrophobic faces should be able to achieve this function.[11] The means of control could be an external force or a change in conditions.

As shown in the work of Prof. Horst Weller and his group,[5] Prof. Seeman,[1] and others in this field, the details of the assembly process have critical impact on the resulting structures and properties. Thus, comparing targeted structures to reality, developing the tools that combine ultrahigh resolution and functional measurements, and *your* creativity are going to be the keys to this burgeoning field. We will continue to explore hierarchical assembly with you, as so much remains to be done and the potential rewards for success are so great.

As we were going to press, we were delighted to learn that Editorial Advisory Board member Prof. Louis Brus and freqent contributor Dr. Sumio Iijima shared the inaugural Kavli Prize in Nanoscience for their discoveries in semiconductor quantum dots and carbon nanotubes, repsectively.[18–21] Congratulations to both and please look foward to upcoming Focus articles on their work!

Paul S. Weiss
Editor-in-Chief

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