

Managing Hierarchical Supramolecular Organization with Holographic Tweezers

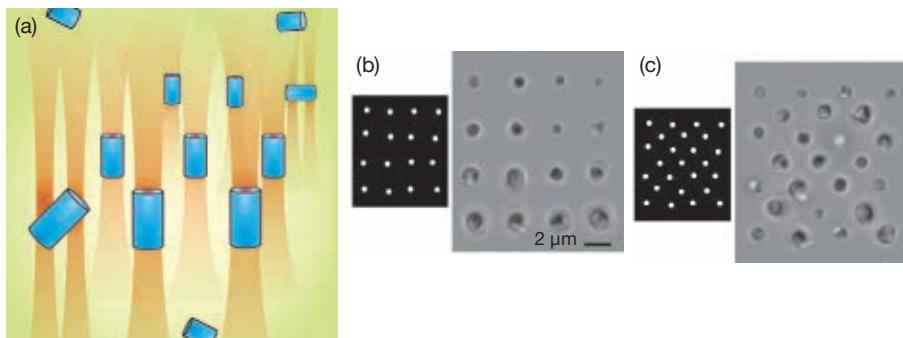
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Hierarchical supramolecular organization may be key to designing novel, functional organic or inorganic materials with tailored properties that exploit the strong relationship between molecular arrangements and resulting macroscopic properties.¹ In particular, the hierarchical organization of pre-ordered structures is one of the most promising approaches to bridging different ordering scales—from the molecular to the macroscopic.

Microporous molecular sieves such as zeolites have proven to be ideal host materials to accommodate a wide range of guest molecules² and thereby realize a first level of organization. The challenge is to create ordered assemblies of the host material after they are loaded with guest species, thus creating hierarchical supramolecular organization. There are several examples of relatively simple, large-scale organization of host materials with established chemical methods. So far, however, it has been almost impossible to achieve a higher degree of fine control on the level of the single host particles.

Where chemistry reaches its limit, optics takes over. Optical tweezers are ideal tools for a moderate number of particles; they can trap, orient and guide particles, particularly when holographic optical tweezers (HOT) are implemented.³ Creating optical landscapes to trap a larger number of particles in 3-D in a defined and preferably reconfigurable way is still a challenge, especially for objects with nonspherical symmetry.

Earlier this year, we showed that it is possible to achieve a high degree of control on elongated microscopic objects like rod-shaped bacteria by means of tailored light fields in HOT.⁴ Transferring this approach to zeolite-based host-guest materials—which also have a nonspherical shape—we can create almost arbitrary configurations.⁵ With HOT, any single host in an assembly can be controlled independently from all others, solely by



(a) Optically induced organization of multiple zeolite *L* host particles. The setup is based on a Nikon Ti Eclipse microscope where holographic tweezers are implemented by means of a 2.5-W Nd:YVO₄ laser ($\lambda=1,064$ nm), which illuminates a high-definition phase-only spatial light modulator. After the light field is structured, it is focused through the microscope objective ($M=100\times$, NA=1.49) and thus creates the desired configuration of multiple optical traps at different transversal and axial positions. (b) Sixteen polydisperse zeolite *L* particles with diameter and length of roughly 1 μm are 3-D optically trapped in a rectangular lattice configuration and sorted by size. (c) The complexity of organization is increased by adding additional zeolite *L* particles at the geometrically relevant positions, resulting in a centered rectangular lattice. Black insets show the configuration of the optical traps (white spots).

optical means and in strong contrast to the contemporary, ensemble-based methods. As the sample is observed through an inverted, optical fluorescence microscope during operation, all manipulations can be done truly interactively.

Any host particle with desired properties can be selected separately from a reservoir and precisely translated to any position in the microscopic sample, thereby allowing hundreds of hosts to be held simultaneously. The most powerful advantage of our optical approach, however, is that it allows the possibility of rotating elongated host particles into any orientation with highest precision

by means of optimized light fields that create multiple traps with different relative intensities. This formidable level of control allows for the realization of tailored microstructures with a widely tunable degree of organization.

The hierarchical supramolecular organization achieved by our approach is not limited to static assemblies.⁵ The micro-structures can be modified and re-arranged in real time. On the other hand, permanent structures can be prepared by established methods to fixate the assembly after creation. HOT is thus an optimal choice if complex assemblies are investigated or very versatile rapid-prototyping is required. ▲



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