# The motion of the medium matters for self-assembling particles, research shows

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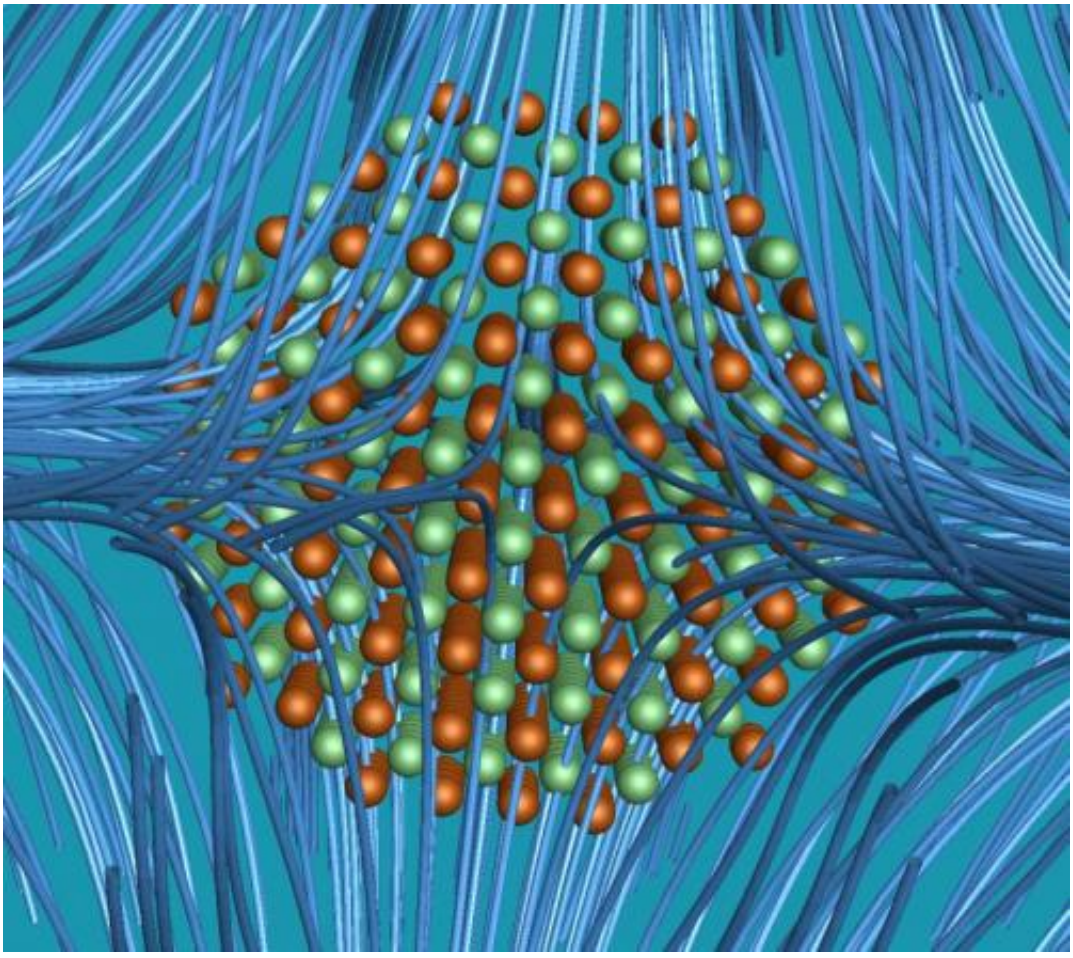


The spheres that make up the crystal follow each other in slipstreams, making some patterns more likely to form. Credit: Ian Jenkins

(Phys.org) —By attaching short sequences of single-stranded DNA to

nanoscale building blocks, researchers can design structures that can effectively build themselves. The building blocks that are meant to connect have complementary DNA sequences on their surfaces, ensuring only the correct pieces bind together as they jostle into one another while suspended in a test tube.

Now, a University of Pennsylvania team has made a discovery with implications for all such self-assembled structures.

Earlier work assumed that the liquid medium in which these DNA-coated pieces float could be treated as a placid vacuum, but the Penn team has shown that fluid dynamics play a crucial role in the kind and quality of the structures that can be made in this way.

As the DNA-coated pieces rearrange themselves and bind, they create slipstreams into which other pieces can flow. This phenomenon makes some patterns within the structures more likely to form than others.

The research was conducted by professors Talid Sinno and John Crocker, alongside graduate students Ian Jenkins, Marie Casey and James McGinley, all of the Department of Chemical and Biomolecular Engineering in Penn's School of Engineering and Applied Science.

It was published in the *Proceedings of the National Academy of Sciences*.

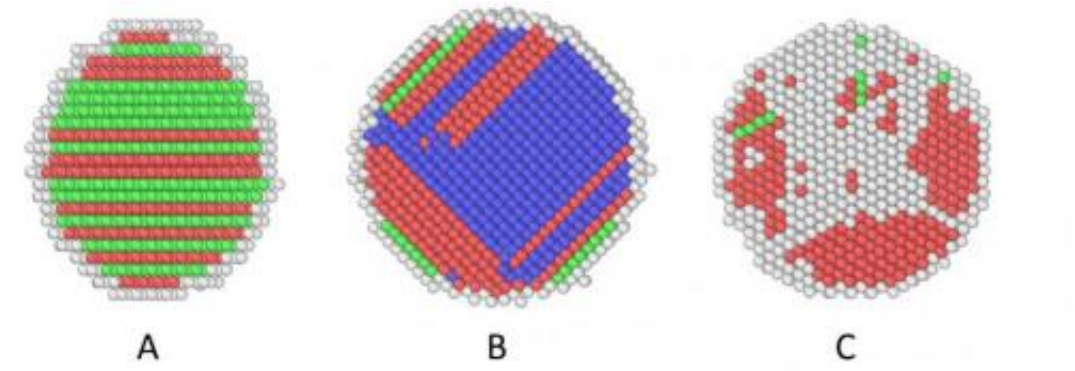
The Penn team's discovery started with an unusual observation about one of their previous studies, which dealt with a reconfigurable crystalline structure the team had made using DNA-coated plastic spheres, each 400 nanometers wide. These structures initially assemble into floppy crystals with square-shaped patterns, but, in a process similar to heat-treating steel, their patterns can be coaxed into more stable, triangular configurations.

Surprisingly, the structures they were making in the lab were better than the ones their computer simulations predicted would result. The simulated crystals were full of defects, places where the crystalline pattern of the spheres was disrupted, but the experimentally grown crystals were all perfectly aligned.

While these perfect crystals were a positive sign that the technique could be scaled up to build different kinds of structures, the fact that their simulations were evidently flawed indicated a major hurdle.

"What you see in an experiment," Sinno said, "is usually a dirtier version of what you see in simulation. We need to understand why these simulation tools aren't working if we're going to build useful things with this technology, and this result was evidence that we don't fully understand this system yet. It's not just a simulation detail that was missing; there's a fundamental physical mechanism that we're not including."

By process of elimination, the missing physical mechanism turned out to be hydrodynamic effects, essentially, the interplay between the particles and the fluid in which they are suspended while growing. The simulation of a system's hydrodynamics is critical when the fluid is flowing, such as how rocks are shaped by a rushing river, but has been considered irrelevant when the fluid is still, as it was in the researchers' experiments. While the particles' jostling perturbs the medium, the system remains in equilibrium, suggesting the overall effect is negligible.




The researchers' simulations produced crystals with random defects. Here, different colors represent different crystal patterns. Credit: University of Pennsylvania

"The conventional wisdom," Crocker said, "was that you don't need to consider hydrodynamic effects in these systems. Adding them to simulations is computationally expensive, and there are various kinds of proofs that these effects don't change the energy of the system. From there you can make a leap to saying, 'I don't need to worry about them at all.'"

Particle systems like ones made by these self-assembling DNA-coated spheres typically rearrange themselves until they reach the lowest energy state. An unusual feature of the researchers' system is that there are thousands of final configurations—most containing defects—that are just as energetically favorable as the perfect one they produced in the experiment.

"It's like you're in a room with a thousand doors," Crocker said. "Each of those doors takes you to a different structure, only one of which is the copper-gold pattern crystal we actually get. Without the hydrodynamics, the simulation is equally likely to send you through any one of those doors."

The researchers' breakthrough came when they realized that while hydrodynamic effects would not make any one final configuration more energy-favorable than another, the different ways particles would need to rearrange themselves to get to those states were not all equally easy. Critically, it is easier for a particle to make a certain rearrangement if it's following in the wake of another particle making the same moves.

"It's like slipstreaming," Crocker said. "The way the particles move together, it's like they're a school of fish."

"How you go determines what you get," Sinno said. "There are certain paths that have a lot more slipstreaming than others, and the paths that have a lot correspond to the final configurations we observed in the experiment."

The researchers believe that this finding will lay the foundation for future work with these DNA-coated building blocks, but the principle discovered in their study will likely hold up in other situations where microscopic particles are suspended in a liquid medium.

"If slipstreaming is important here, it's likely to be important in other particle assemblies," Sinno said. It's not just about these DNA-linked particles; it's about any system where you have particles at this size scale. To really understand what you get, you need to include the hydrodynamics."

**More information:** Hydrodynamics selects the pathway for displacive transformations in DNA-linked colloidal crystallites, www.pnas.org/content/111/13/4803.abstract

Provided by University of Pennsylvania