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Directing Nanoscale Heat Flow in Nanocrystal-Based Metamaterials

Today, nanocrystals are establishing themselves as a versatile solution for the manufacture of next-generation of optoelectronic devices such as LEDs, photodetectors and photovoltaic cells. Recent work even suggests that ordered assemblies of nanoparticles, known as supercrystals or metamaterials, exhibit an even wider range of emergent properties ranging from mechanics to thermoelectricity and even catalytic functions. However, the heat generated during the use of such devices is likely to cause physical and chemical instabilities leading to the degradation of the active material and thus its performance. In collaboration with the University of Hamburg and the institutes LPS and MONARIS, members of the Physical Chemistry and Dynamics of Surfaces team at INSP showed that it is possible to control heat flux at the nano-scale in these metamaterials by playing with the shape and organization of the nanocrystals that compose them. This discovery paves the way for new thermal management methods that promise to increase the robustness and efficiency of these supercrystalline solids.

Thanks to a recent thermo-reflectance technique resolved both in space and in time, it was possible to image the heat flow propagating in supercrystals of gold nano-spheres, nano-rods and nano-bipyramids. These measurements were correlated with scanning electron microscopy images to directly visualize the impact of complex structures and defects on thermal properties at the nanoscale (Figure 1). The study of ordered and curved arrangements of nano-rods as well as grain boundaries revealed that nanoscale to microscale heat transport is anisotropic. The heat propagates more quickly in the direction parallel to the long axis of the nanoparticles and can therefore be guided progressively within the metamaterial itself.



Figure 1

Images of the samples and visualization of heat flow. (a-c) Scanning electron microscopy (SEM) images of a sample of (a) nano-spheres, (b) nano-rods with an aspect ratio of 4:1 and (c) nano-bipyramids with an aspect ratio of 3.2. (d-g) Correlative measurements superimposing a SEM image with the measured heat flux 150 nanoseconds after excitation (d) in an ordered assembly of nanorods, (e) on a grain boundary separating two supercrystals composed of rods of different orientations, (f) in a curved assembly and (g) near a fracture.

Finite element simulations and equivalent resistance models made it possible to establish how the different thermal and geometric parameters govern the emergence of the anisotropy. In this study, the research team shows that the degree of anisotropy can be experimentally controlled by changing the aspect ratio of the nanocrystals (Figure 2). An anisotropy whose magnitude exceeds the aspect ratio can even be achieved by optimizing the shape and arrangement of the nanoparticles, for example by replacing rods with bipyramids.



Figure 2

Control of anisotropy through aspect ratio. (a) Time series of images obtained by thermo-reflectance in a sample of nano-rods with an aspect ratio of 4:1. (b) Spatio-temporal evolution of the variances of the minor and major axes of the elliptical Gaussian fitted to the thermal profile in (a). (c) Ratio of thermal diffusion coefficients in directions parallel to and perpendicular to the major axis of the nanoparticles (anisotropy) as a function of the aspect ratio. The principles established in this study can be extended to other nanocrystal-based metamaterials of different shapes and compositions. These findings pave the way for new heat dissipation techniques by directly using the active material and maintaining control over the optical and electronic effects dependent on quantum confinement.

Reference

"Anisotropic Thermal Transport in Tunable Self-Assembled Nanocrystal Supercrystals" **Matias Feldman**, Charles Vernier, Rahul Nag, Juan Barrios-Capuchino, **Sébastien Royer, Hervé Cruguel, Emmanuelle Lacaze, Emmanuel Lhuillier, Danièle Fournier**, Florian Schulz, Cyrille Hamon, Hervé Portalès, **James K. Utterback. ACS Nano** 18, 34341–34352 (2024)

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