THERMAL TRANSPORT

Harmony with superatoms

The thermal conductivities of superatom crystals have significant contributions from extended phonon states and show a remarkable temperature dependence due to orientational ordering.

Longji Cui, Edgar Meyhofer and Pramod Reddy

Understanding how heat is transported in nanostructured materials and devices is indispensable for the efficient conversion of heat to electricity and for thermal management in nanoscale devices[1]. Beyond their importance to future energy-conversion applications, nanostructured materials also offer unique opportunities to study the fundamental properties of phonons — quantized modes of vibration of the lattice — which are usually the dominant carriers of heat in non-metals. Intriguingly, past computational work[2] has pointed out that thermal transport in superlattices cannot be modelled accurately by just considering the phonon spectra of their individual components (Fig. 1a). Instead, the extended phonon states of the entire structure need to be considered, owing to the wave-like properties of phonons, as has been confirmed experimentally as well[3,4]. However, a clear understanding of the importance and tunability properties of such effects is still lacking because of the scarcity of good model systems. Writing in *Nature Materials*, Xavier Roy, Jonathan Malen and colleagues[5] study thermal transport in a new class of materials, called superatom crystals (SACs) — three-dimensional periodic arrays of nanoscale molecular clusters (Fig. 1b), which feature atomic-level control of materials and large unit cell sizes.

Roy and collaborators used a pump-probe optical technique[6] to measure the thermal conductivities of two classes of SACs (Fig. 1b) formed from either identical superatoms (unary crystals) or a mixture of superatoms and C_{60} (binary crystals). The measured thermal conductivity was found to correlate well with the estimated velocity of sound for each of the SACs, providing first evidence in support of the hypothesis that thermal transport in SACs has dominant contributions from extended phonon states that naturally emerge from 'harmonious' wave effects in the SAC (Fig. 1c). Furthermore, measurements of the thermal conductivities in SACs from ~120 K to 330 K reveal that they either remain temperature-independent or are very weakly decreasing with temperature for all the unary crystals. This behaviour is consistent with the expectation that the phonons associated with the collective modes of the SACs (Fig. 1c) have relatively long mean free paths, which increase further with decreasing temperature and potentially offset the expected reduction in thermal conductivity due to a decreasing heat capacity.

These observations differ markedly from past studies[7] of thermal transport in nanocrystal arrays (NCAs), despite their structural similarities with SACs. Specifically, the thermal conductivity in NCAs was found to decrease rapidly with temperature, in agreement with the expectation that the thermal conductivity should follow the same trend as the heat capacity of the system when the mean free paths of phonons are small. These differences between SACs and NCAs can be attributed to the fact that NCAs are too disordered for phonons associated with the extended phonon states of the NCAs to make appreciable contributions to thermal transport.

In contrast to the unary SACs, the thermal conductivity of binary SACs was found to have a more complicated dependence on temperature. Specifically, Roy and colleagues show that in some of the binary SACs the thermal conductivity is independent of temperature, until a threshold is reached, below which a rapid increase is observed[5]. This signals a phase transition to a highly ordered lattice, as crystalline solids show rapid increases in thermal conductivity[8] when the temperature is decreased to values below room temperature. Indeed, the authors associate the observed transition in thermal transport characteristics with a structural phase transition that suppresses the rotational modes (Fig. 1c) of the C_{60} molecules at low temperatures and introduces orientational ordering within the binary crystals, which in turn reduces the scattering of phonons associated with the extended phonon states of the SAC.

Taken together, the evidence presented by Roy and co-authors makes a compelling case that phonon states associated with the entire SAC make important contributions to thermal transport. This work highlights the suitability of SACs with tunable

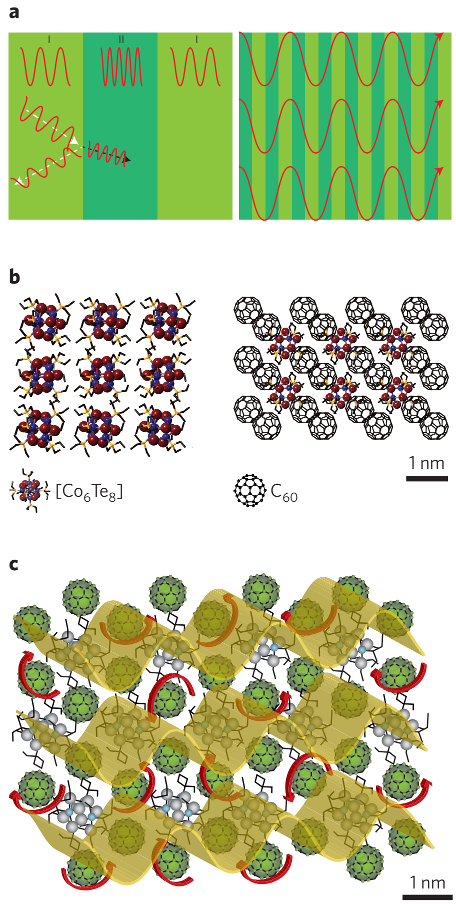

Figure 1 | Schematic description of thermal transport in nanostructured materials. **a**, Thermal transport in long- and short-period superlattices. In long-period (left) superlattices, heat transport can be modelled by considering the phonon spectra of the individual materials (labelled I and II) and the transmission probability for incident phonons at the interfaces. For superlattices with short periods (right) it is necessary to consider the phonon states associated with the entire superlattice for accurately modelling thermal transport. Red sinusoidal curves represent phonon states. **b**, Schematics of unary (left) and binary (right) SACs studied by Roy and co-workers[5]. **c**, Extended phonon states in a binary SAC, along with the rotational degrees of freedom of C_{60} molecules. Panels **b** and **c** Reproduced from ref. 5, Nature Publishing Group.

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superatom interactions for studying the emergence of extended phonon states and their impact on thermal transport. Superatom crystals featuring large electric and magnetic responses may also offer unique opportunities for achieving the long-standing goal of actively tuning the thermal conductivity through the use of external fields. Furthermore, given that the room-temperature thermal conductivity of SACs is rather low (~0.5 W m^{-1} K^{-1}), they may be suitable for new efficient thermoelectric materials if their electrical and thermoelectric properties can be engineered through suitably chosen superatom clusters and ligands. Finally, SACs may also hold potential for engineering the dielectric properties of materials and using them for control of near-field thermal radiation[9,10]. ❐

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METASURFACE LENS

Shrinking the camera size

A miniaturized camera has been developed by integrating a planar metasurface lens doublet with a CMOS image sensor. The metasurface lens doublet corrects the monochromatic aberration and thus delivers nearly diffraction-limited image quality over a wide field of view.

Cheng Sun

An ideal refractive lens bends light rays into a single focal point to produce a perfect replica of the object. In reality, intrinsic geometric and chromatic aberrations result in refractive lenses that are far from perfect. The recent emergence of metasurfaces offers radically new capabilities in controlling and manipulating the propagation of light[1,2]. Metasurfaces are composed of planar arrays of nanoscatterers that can lead to full control of the local amplitude, phase, polarization and linear/angular momentum of light at the subwavelength scale. Now, in *Nature Communications*, Andrei Faraon and colleagues report a wide-angle metasurface imaging lens doublet, corrected for monochromatic aberration[3].

The genesis of classical optics dates back to the eighth century BC, in an ancient Egyptian reference describing "simple glass meniscal lenses"[4]. Since then, crafting the surface of a piece of transparent material to manipulate the light has remained the primary strategy for building a refractive imaging lens. Precise cutting, grinding and polishing are necessary steps for the fabrication of optical components. More complex devices, such as camera lenses and microscope objectives, are subsequently assembled together with tight alignment tolerances, and are inevitably complex and expensive.

Ultra-thin 2D metasurfaces consisting of spatially varying nanoscatterers offer a possible substitute for bulk optical elements. These metasurfaces have already inspired a wide range of photonic applications, such as light bending and focusing, wave plates, vortex beam generation and holograms[5]. In particular, ultra-thin, aberration-free meta-lenses can produce something close to diffraction-limited focusing[1,2,6,7], with their efficiencies having recently improved[8–10].

Faraon and collaborators[3] have fabricated a metasurface lens doublet that consists of two polarization-independent phase plates, on the top and bottom surfaces of a 1-mm-thick transparent fused silica substrate (Fig. 1a). Each metasurface consists of a hexagonal array of 600-nm-tall amorphous silicon nanoposts with spatially

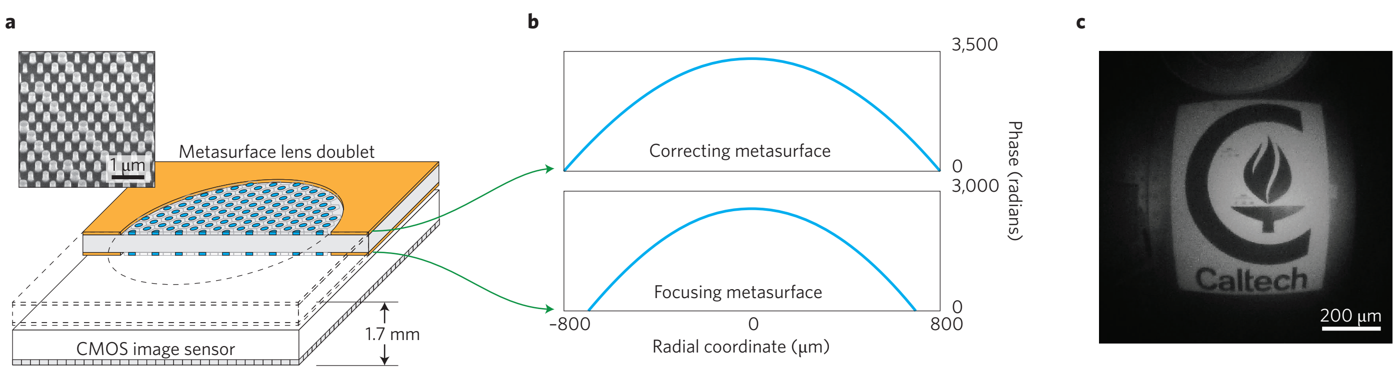

Figure 1 | Imaging with a miniaturized camera containing a monochromatically corrected planar metasurface lens. **a**, Schematic illustration of the miniaturized camera, which consists of a metasurface lens doublet integrated on the surface of a CMOS image sensor, with total thickness of 1.7 mm. The planar metasurface lens doublet consists of a correcting metasurface and a focusing metasurface, fabricated on both sides of a transparent substrate. The inset shows a magnified view of the metasurface lens, consisting of a 2D array of nanoposts with spatially varying dimensions. **b**, Phase profile of the correcting and focusing metasurfaces. **c**, Image recorded using the miniaturized camera. Adapted from ref. 3, Nature Publishing Group.

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