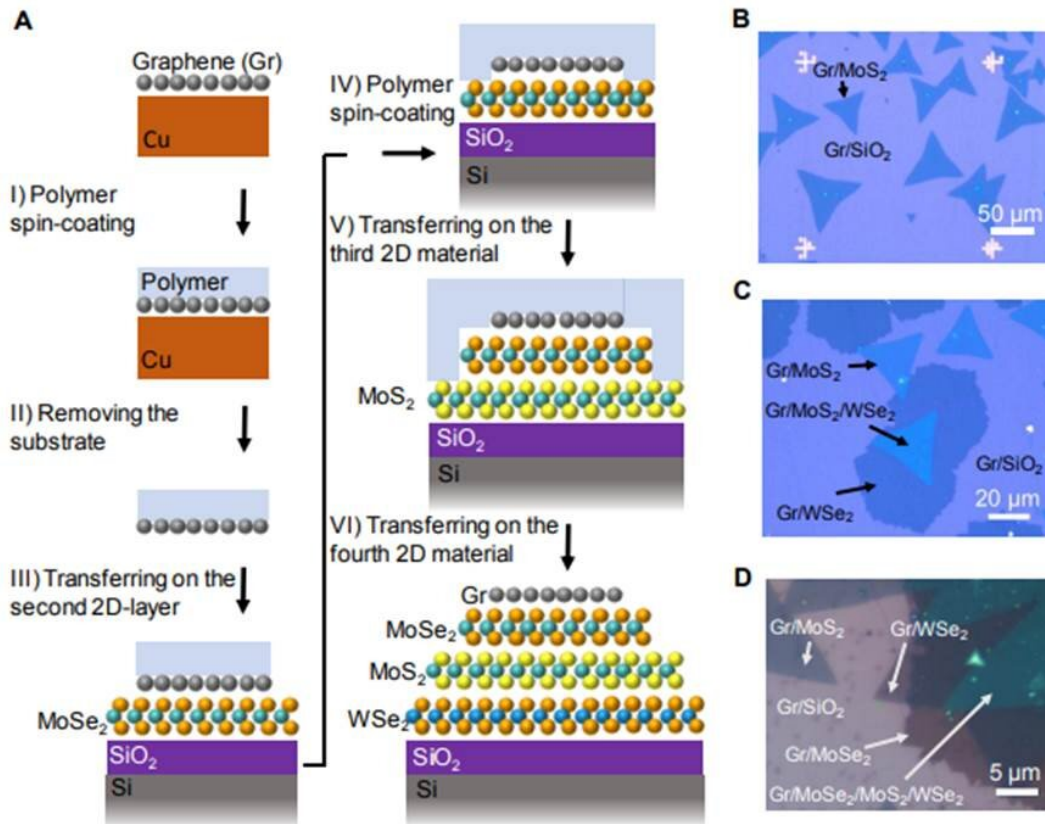


Ultrahigh thermal isolation across heterogeneously layered two-dimensional materials

August 22 2019, by Thamarasee Jeewandara



Transfer process and optical images. (A) Schematic of the multiple-transfer process flow to fabricate a new metamaterial Gr/MoSe₂/MoS₂/WSe₂ (graphene/molybdenum diselenide/molybdenum disulfide/tungsten diselenide) heterostructure on SiO₂/Si (silicon dioxide/silicon) substrate. Optical images of (B) Gr/MoS₂, (C) Gr/MoS₂/WSe₂, and (D) Gr/MoSe₂/MoS₂/WSe₂ on SiO₂/Si

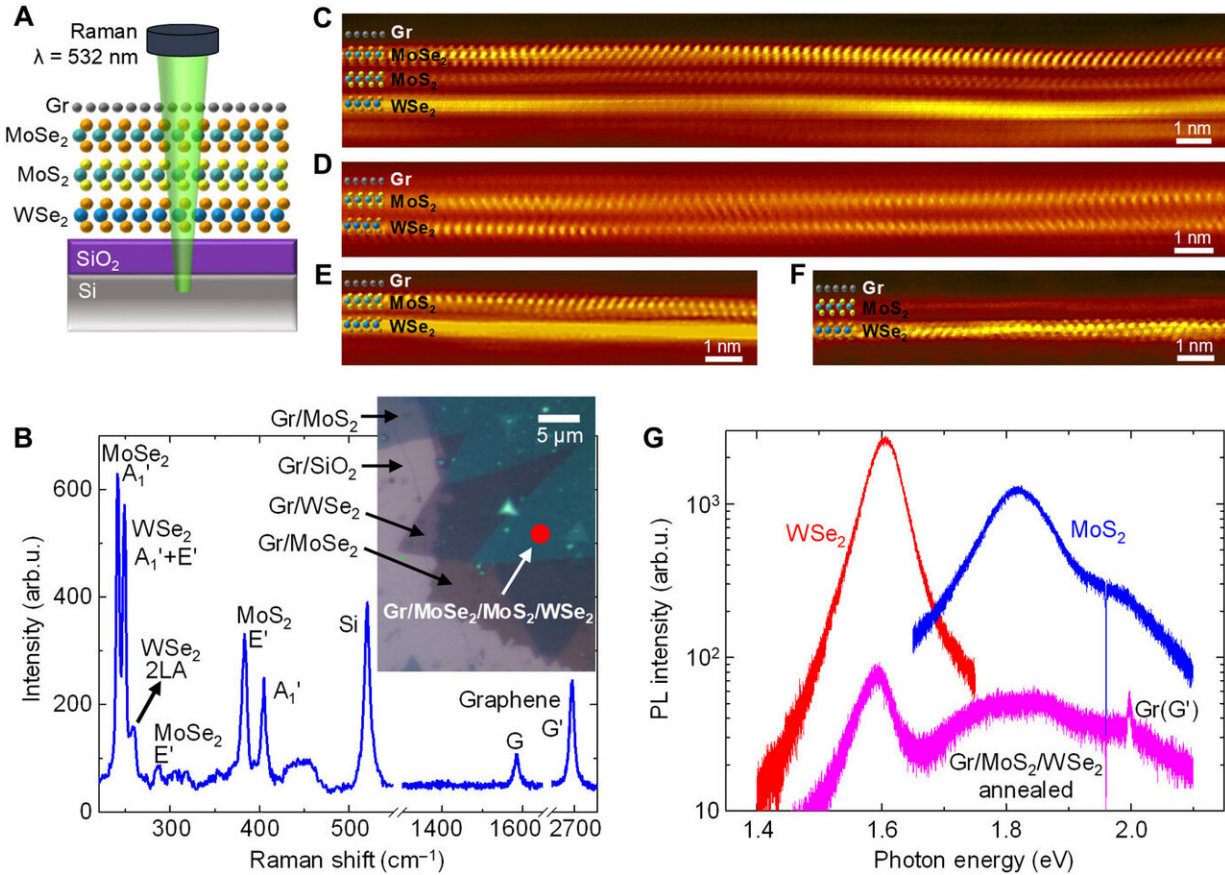
substrates. Credit: Science Advances, doi: 10.1126/sciadv.aax1325

Heterogeneous nanomaterials can now facilitate advanced electronics and [photonics](#) applications, but such progress is challenging for thermal applications due to the comparatively shorter wavelengths of [heat carriers \(known as phonons\)](#). In a new study, now published on *Science Advances*, Sam Vaziri and co-workers at Theiss Research and the departments of Electrical Engineering, Materials Science and Engineering at the National Institute of Standards and Technology (NIST), and the Precourt Institute of Energy at Stanford University, Stanford California, demonstrated unusually high thermal isolation across ultra-thin heterostructures.

They achieved this by layering atomically thin, two-dimensional (2-D) materials to form artificial stacks of monolayer graphene (Gr), molybdenum disulfide (MoS_2) and tungsten diselenide (WSe_2), with thermal resistance greater than silicon dioxide (SiO_2). Alongside effective thermal conductivity lower than air at room temperature. Using [Raman thermometry](#), the scientists simultaneously identified the thermal resistance between any 2-D monolayers in the stack to form thermal [metamaterials](#) as examples in the emerging field of phononics. Vaziri et al. propose applications of the metamaterials in ultrathin thermal insulation, thermal energy harvesting and to route heat within ultracompact geometries.

Advanced electronic and photonic devices such as [high-electron mobility transistors](#), [quantum cascade lasers](#) and [photonic bandgap crystals](#) take advantage of the [fermionic nature](#) of charge carriers during voltage gating or confinement. Then they make use of long photon wavelengths during their interference. Nevertheless, thermal nanoengineering and the [emerging field of phononics](#) only offer a few examples, despite the

existing demand for [heat management applications](#). This discrepancy results from the [short wavelengths](#) of heat-carrying vibrations in solids, where the [bosonic nature](#) of phonons can also contribute to the challenge of actively controlling heat transport in solids where it cannot be voltage-gated like charge carriers.



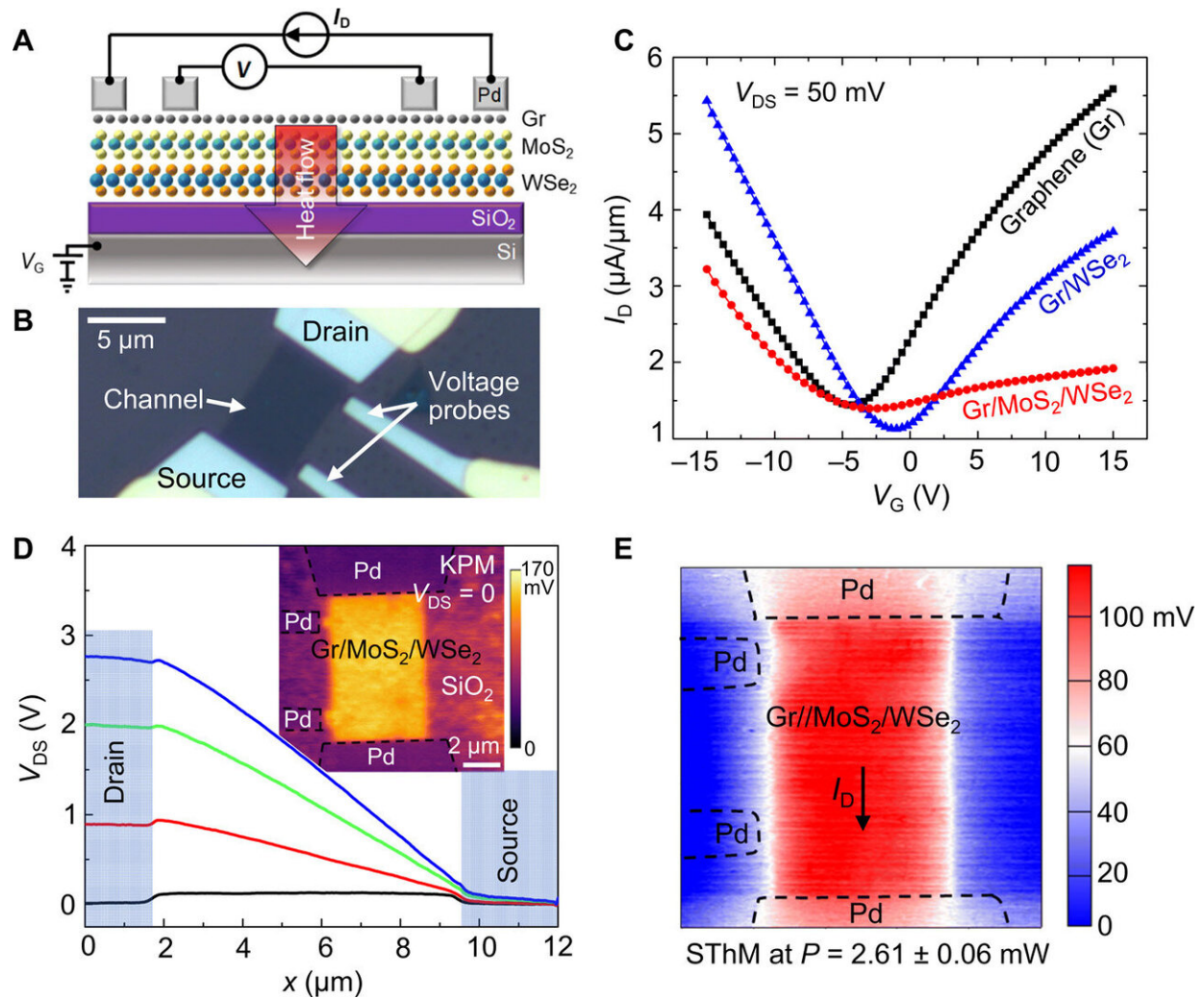
Optical and STEM characterization of vdW heterostructures. (A) Cross-section schematic of Gr/MoSe₂/MoS₂/WSe₂ sandwich on SiO₂/Si substrate, with the incident Raman laser. (B) Raman spectrum of such a heterostructure at the spot indicated by the red dot in the inset optical image. Raman signatures of all materials in the stack are obtained simultaneously. The graphene Raman spectrum is flattened to exclude the MoS₂ photoluminescence (PL) effect. arb.u., arbitrary units. (C to F) STEM cross-sectional images of four-layer (C) and three-layer (D to F) heterostructures on SiO₂. In (D), MoSe₂ and WSe₂ are

approximately aligned along the 1H [100] zone axis, and in (E and F), the layers are misaligned by $\sim 21^\circ$ with respect to the 1H [100] zone axis. The monolayer graphene on top of each heterostructure is hard to discern due to the much lower atomic number of the carbon atoms. (G) PL spectra of monolayer MoS₂, monolayer WSe₂, and a Gr/MoS₂/WSe₂ heterostructure after annealing. The PL is strongly quenched in the heterostructure due to intimate interlayer coupling. Credit: Science Advances, doi: 10.1126/sciadv.aax1325

Physicists had previously attempted to manipulate the thermal properties of solids using [non-laminate films](#) and [superlattices](#) to reduce thermal conductivity below the constituent materials to ultimately achieve thermal manipulation via structural disorder and high interface density to introduce additional thermal resistance. They found unusually [low thermal conductivity](#) in nanoengineered [silicon and germanium nanowires](#) due to strong phonon-boundary scattering and achieved large thermal conductivities in isotopically pure materials such as [diamond](#), [graphene](#) and [boron arsenide](#) via reduced phonon scattering.

Two dimensional (2-D) materials have thus [enabled a new frontier](#) with sub-nanometer thin, single monolayers to control device behaviors at atomic length scales. Existing examples include new [tunneling field effect transistors](#) and [ultrathin photovoltaics](#) with high efficiency. In the present work, Vaziri et al. used van der Waals (vdW) assembly of atomically thin 2-D layers to achieve unusually high thermal resistance across heterostructures. They showed a thermal resistance equivalent to 300 nm thick SiO₂ across sub-2-nm-thin vdW heterostructures with clean, residue-free interfaces. By layering heterogeneous 2-D monolayers with diverse atomic densities and vibrational modes the research team demonstrated the potential to tailor thermal properties at the atomic scale; on the order of the phonon wavelength. The structural basis of the new phononic metamaterials with unusual properties is not commonly found in nature. The present work represents unique

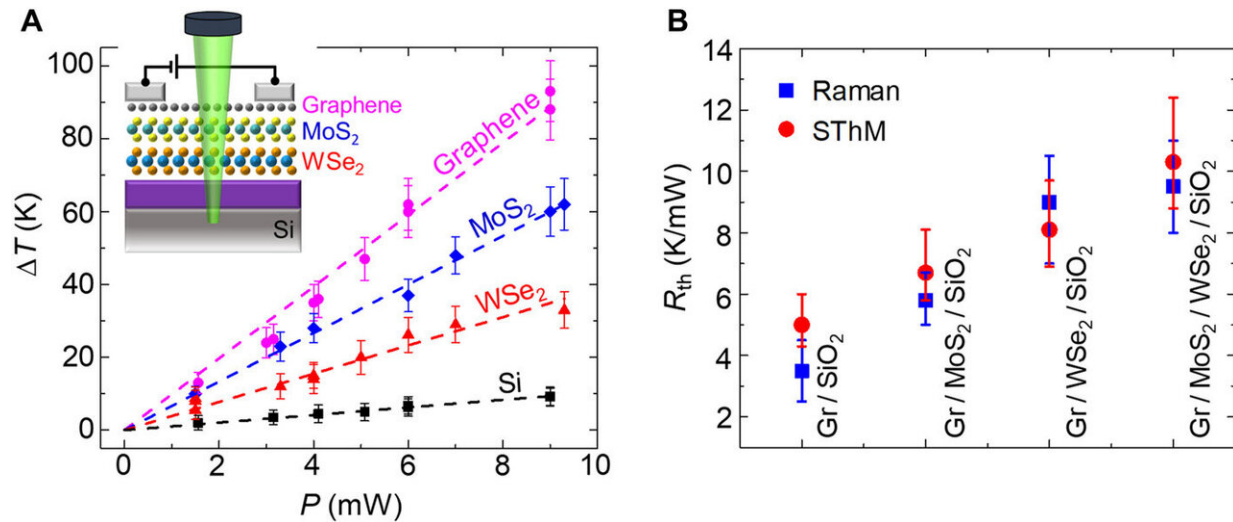
applications of 2-D materials and their weak vdW interactions for assembly to block or guide the flow of heat.



Electrical and scanning probe characterization. (A) Cross-sectional schematic of the test structure showing the four-probe configuration. Electrical current flows in the graphene top layer, and heat dissipates across layers, into the substrate. (B) Optical image of a four-probe test structure. Devices are back-gated by the Si substrate through 100-nm SiO₂. (C) Measured transfer characteristics of three test structure stacks, Gr/MoS₂/WSe₂, Gr/WSe₂, and Gr-only control devices in vacuum ($\sim 10^{-5}$ torr). All measurements display the ambipolar property of the top graphene channel. (D) KPM of an uncapped Gr/MoS₂/WSe₂ heterostructure

device. The graph displays the surface potential along the channel (averaged across the channel width) at different bias conditions. The small potential jump near the Pd electrodes represents the relative work function difference (~120 mV). The KPM maps reveal no other heterogeneities in the surface potential, confirming the spatially uniform quality of these devices. The inset shows the zero-bias KPM map. (E) SThM thermal map of Gr/MoS₂/WSe₂ heterostructure, here capped with 15-nm Al₂O₃, revealing homogeneous heating across the channel. This confirms the uniformity of the thermal interlayer coupling in the stacks. The device dimensions are the same as in the (D) inset. Credit: Science Advances, doi: 10.1126/sciadv.aax1325.

The research team obtained a cross-section of a four-layered heterostructure with graphene (Gr) on MoSe₂ (molybdenum diselenide), MoS₂ (molybdenum disulfide) and WSe₂ (tungsten diselenide) on a SiO₂/Si substrate. Using a Raman laser, they simultaneously probed the individual layers in the stack with single-layer accuracy. The research team separately grew the 2-D monolayer materials using [chemical vapor deposition](#) and transferred them to avoid polymer and other residues. To confirm the microstructural, thermal and electrical properties of the heterostructures, Vaziri et al. used extensive materials characterization techniques, including [scanning transmission electron microscopy](#) (STEM), [photoluminescence \(PL\) spectroscopy](#), [Kelvin probe microscopy](#) (KLM) and [scanning thermal microscopy](#) (SThM) alongside [Raman spectroscopy](#) and [thermometry](#). Using the techniques, they revealed the signature of every 2-D material monolayer in the stack and that of the Si substrate. Using multiple STEM images, the research team revealed atomically intimate vdW gaps without contaminants, allowing them to observe the total thickness of the heterostructures. They then confirmed interlayer coupling across large surface areas using PL spectroscopy.

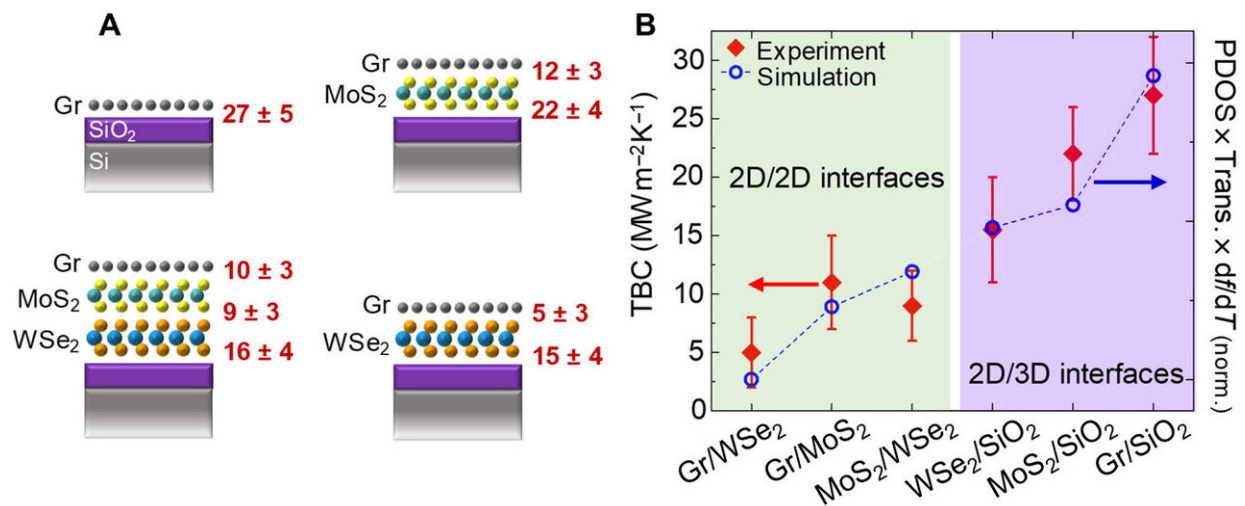


Thermal resistance of the heterostructures. (A) Measured temperature rise ΔT versus electrical input power for each individual layer in a Gr/MoS₂/WSe₂ heterostructure, including the Si substrate, shown in the inset. Graphene (pink circles), MoS₂ (blue diamonds), WSe₂ (red triangles), and Si (black squares). All measurements are carried out at VG

To measure heat flow perpendicular to the atomic planes of the heterostructure, Vaziri et al. patterned the stacks in the shape of four-probe electrical devices. They used electrical heating to accurately [quantify the input power](#) and confirmed the current conduction and heating on the top graphene layer to be orders of magnitude greater than for MoS₂ and WSe₂. To demonstrate the surface temperature uniformity of these devices they used KPM and SThM surface characterization methods and then quantified the temperature of each individual layer using Raman spectroscopy. As the graphene heat power (P) ramped up in the system, the temperature of each layer increased in a Gr/MoS₂/WSe₂ heterostructure setup. Due to uniform heating the researchers easily analyzed the thermal resistances from bottom to top. The excellent agreement between the two thermometry methods of Raman and SThM validated the values obtained in the setup.

The scientists analyzed the thermal boundary resistance (TBR) between the layers responsible for the very large thermal resistance perpendicular to the

heterostructures. The thermal boundary conductance (TBC) measurements in the study were a first for atomically intimate interfaces between 2-D/2-D monolayers and formed the first reported TBC between WSe₂ and SiO₂ monolayers. They showed that TBCs obtained for Gr/SiO₂ and MoSe₂/SiO₂ interfaces [agreed with previous studies](#), while TBC of the monolayer WSe₂/SiO₂ interface [was comparatively lower](#), which was not unexpected due to the comparatively fewer [flexural phonon modes](#) available for transmission in the monolayer. According to the results, TBC for a 2-D/2-D interface was lower than the TBC with a 3-D SiO₂ substrate. The lowest TBC recorded in the work belonged to Gr/WSe₂ and the research team explained the observations using the [Landauer formula](#). The research team obtained the phonon transmission at the interface using the acoustic mismatch model (AMM) as the [ratio of mass density of the two materials](#). The researchers captured TBC trends using a simple model of heat flow across the interfaces developed in the study.



Summary of TBC (thermal boundary conductance) trends. (A) Schematic of all TBCs measured (in MW m⁻² K⁻¹) across heterostructures consisting of, clockwise from top left, graphene (Gr), Gr/MoS₂, Gr/WSe₂, and Gr/MoS₂/WSe₂, all on SiO₂/Si substrates. (B) Measured TBC values of 2D/2D and 2D/3D (with SiO₂) interfaces (red diamonds, left axis) and the calculated product of phonon density of states (PDOS), phonon transmission, and df/dT (blue circles, right axis). Calculated values are normalized to the minimum achieved for Gr/WSe₂ (see table S2). The dashed line between the simulation symbols is a guide to the eye. Lower TBC is noted at interfaces between 2D/2D materials and those between materials with larger mismatch in mass density. Three devices were

measured for each structure, at two or more distinct positions of the Raman laser. No significant TBC variation is seen between samples with different layer (mis)alignment, within the experimental uncertainty. All values are at room temperature. Credit: Science Advances, doi: 10.1126/sciadv.aax1325

In this way, Sam Vaziri and co-workers gained knowledge to realize atomically tailored thermal interfaces and demonstrated their potential to engineer extremely thermally insulating metamaterials. The newly engineered metamaterials demonstrated properties unprecedented in nature. The heterostructures provide an example in the emerging fields of phononics to manipulate the thermal properties of solids at length scales comparable to phonon wavelengths. The 2-D layered materials offer promising, ultralight and compact heat shields to direct heat away from hotspots in electronics. The research team envision translating the metamaterials to improve the efficacy of [thermoelectric energy harvesters](#) and thermally active devices such as [phase-change memories](#) in the future.

More information: Sam Vaziri et al. Ultrahigh thermal isolation across heterogeneously layered two-dimensional materials, *Science Advances* (2019). DOI: [10.1126/sciadv.aax1325](https://doi.org/10.1126/sciadv.aax1325)

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