Ion Write Microthermotics: Programing Thermal Metamaterials at the Microscale

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ABSTRACT: Considerable advances in manipulating heat flow in solids have been made through the innovation of artificial thermal structures such as thermal diodes, camouflages, and cloaks. Such thermal devices can be readily constructed only at the macroscale by mechanically assembling different materials with distinct values of thermal conductivity. Here, we extend these concepts to the microscale by demonstrating a monolithic material structure on which nearly arbitrary microscale thermal metamaterial patterns can be written and programmed. It is based on a single, suspended silicon membrane whose thermal conductivity is locally, continuously, and reversibly engineered over a wide range (between 2 and 65 W/mK) and with fine spatial resolution (10−100 nm) by focused ion irradiation. Our thermal cloak demonstration shows how ion-write microthermotics can be used as a lithography-free platform to create thermal metamaterials that control heat flow at the microscale.

KEYWORDS: Thermal metamaterial, thermotics, thermal conductivity, thermal cloak, ion irradiation

Thermal metamaterials are heterogeneous structures designed to provide qualitatively new thermal functionalities that cannot be achieved in homogeneous, natural materials. At the macroscale, theoretical advances in transformation thermotics have stimulated experimental demonstrations of various thermal devices to precisely route the heat flow in thermal cloaks, shields, camouflages, inverters, concentrators, or thermostats. These macroscopic devices utilize different materials with highly contrasting thermal conductivities (e.g., metals and polymers) that are mechanically integrated into the designed geometric pattern. One previous approach to scale these macroscopic thermal devices down to the microscale relies on drilling small holes in suspended crystalline membranes by nanopatterning with electron-beam lithography. These holes contrast with the crystalline regions with zero thermal conductivity and enable the formation of binary composite thermal structures, similar to those of macroscopic devices; we refer to this configuration as "digital", because the thermal conductivity takes two discrete, fixed values in the composite. However, this approach is insufficient in general cases, and a different "analog" configuration is needed where more than two values, or even continuous variation of local thermal conductivity, are needed. For example, in order to implement a thermal cloak, there must be at least three distinct values of thermal conductivity available to block the heat from entering the cloaked object and to route the heat around the object. However, mechanical integration of distinct materials at the microscale presents daunting technical challenges, and even if successful, mismatched thermal expansion and non-negligible thermal resistances at the interfaces between different materials would destroy the desired heat flow manipulation. Analog-like, continuously varied values of thermal conductivity could be mimicked by averaging over an area with different filling fractions of small holes, but this effective-medium approach is fundamentally incompatible with the microscopic length scales.

In this work, we develop a platform for ion-write microthermotics (IWMT), where a thin (~120 nm),
suspended, single-crystal Si membrane is locally irradiated by a tightly focused (~1 nm) He⁺ ion beam in a helium ion microscope (HIM) to create microscopic patterns. Depending on the dose of the irradiation, the local thermal conductivity ($\kappa$) of Si can be reduced by more than 1 order of magnitude from the crystalline phase value to the amorphous phase value, as recently demonstrated in nanowires at room temperature. The continuous tuneability of $\kappa$ and the monolithic nature of the composite eliminate the aforementioned issues arising from the “digital” approach. The thermal conductivity suppression is also reversible upon a modest thermal annealing in a $N_2$ environment, offering reprogrammability of the IWMT. The IWMT provides a versatile platform with which microscopic patterns of thermal metamaterials can be designed and written to reroute local heat flow and achieve desired thermal functions. For example, we show that the IWMT enables thermal cloaking of objects that are 3 orders of magnitude smaller in size than in previous demonstrations.\(^3,4\)

Figure 1a illustrates the device structure that demonstrates and characterizes the IWMT platform. The structure is based on the suspended micropad devices that were developed to measure the thermal conductivity of individual nanotubes,\(^10,11\) nanowires,\(^12,13\) and nanoribbons.\(^14\) Here, a single-crystal Si membrane bridges two micropads. The Si membrane is patterned with a thin circle (region iii, darker reflection) at a heavy dose of $10^{14}$ ions/cm$^2$; a wider circle (region ii) remaining pristine, and everywhere else (region i) at a low dose of $5 \times 10^{14}$ ions/cm$^2$. The outer boundary of region i is highlighted with a dashed circle to guide the eye. (c) SEM image of another Si membrane where a periodic pattern of pristine (region i) and heavily irradiated (region iii) is created with a period of 60 nm. (d) HRTEM images of a 30 nm thick Si layer irradiated with doses equivalent to regions i and iii in panel b, showing single crystal and amorphous Si phase, respectively.

Figure 1. The IWMT platform created by local He⁺ ion irradiation. (a) Schematic showing writing of local thermal conductivity in suspended single-crystal Si membrane using the He⁺ ion beam. (b) Optical image of a real thermotic device consisting of a suspended Si membrane bridging two micropads. The Si membrane is patterned with a thin circle (region iii, darker reflection) at a heavy dose of $10^{14}$ ions/cm$^2$; a broader circle (region ii) remaining pristine, and everywhere else (region i) at a low dose of $5 \times 10^{14}$ ions/cm$^2$. The outer boundary of region i is highlighted with a dashed circle to guide the eye. (c) SEM image of another Si membrane where a periodic pattern of pristine (region i) and heavily irradiated (region iii) is created with a period of 60 nm. (d) HRTEM images of a 30 nm thick Si layer irradiated with doses equivalent to regions i and iii in panel b, showing single crystal and amorphous Si phase, respectively.

Figure 2. Modulating thermal conductivity using He⁺ ion irradiation. (a) T-dependent thermal conductivity ($\kappa$) of the Si membrane after irradiation for a wide range of doses (in ions/cm$^2$). The curves are fits using a model including phonon scattering by defects, boundaries, and other phonons. (b) HRTEM images of a 30 nm thick Si layer irradiated with different doses. Insets: SAED pattern corresponding to the different doses of irradiation for 120 nm thick Si.
micropads; the irradiation dose is calculated from the beam current and irradiation time and area. The thermal conductivity measurements were performed inside a vacuum of \(<10^{-6}\) Torr to minimize the parasitic convection loss (see Methods for details). The Si membranes used for the \(\kappa\) measurements have typical widths of 2 \(\mu m\), thicknesses of 120 nm, and lengths of 20 \(\mu m\); these small widths were used to ensure accurate thermal resistance measurements for all sample irradiation levels (see Methods).

The pristine Si membrane shows a typical, nonmonotonous \(T\)-dependent \(\kappa\) with a room-temperature value of \(65 \pm 5 W/mK\), which is in good agreement with reported values in literature\(^{18}\) for size-reduced single-crystal Si with comparable thickness. We note that as these membranes are only lightly doped with a resistivity above 20 \(\Omega cm\), the electronic contribution to \(\kappa\) is negligible and the measured \(\kappa\) is completely dominated by the phonon contribution. As the irradiation dose increases from \(2 \times 10^{13}\) to \(1 \times 10^{18}\) ions/cm\(^2\), a monotonic suppression of \(\kappa(T)\) is clearly observed. For example, at the dose of \(10^{16}\) ions/cm\(^2\), the room-temperature \(\kappa\) and peak \(\kappa\) are reduced from those of the pristine Si by a factor of 30 and 33, respectively. It is worth noting that in addition to the overall suppression of \(\kappa\), the peak of \(\kappa(T)\) shifts gradually toward higher temperatures, indicating increased scattering of phonons by defects and domains newly created in the crystal. The peak vanishes in the measurement temperature range \(<300 K\) for irradiation doses exceeding \(\approx 10^{17}\) ions/cm\(^2\), corresponding to amorphization of the Si membrane as shown later.

The suppression of \(\kappa\) in Figure 2a is quantitatively consistent with that in ref 9 where \(\kappa\) of irradiated Si nanowires was measured only at room temperature. In contrast, the \(\kappa\) suppression observed over the wide temperature range and peak shifts measured in this work provide much richer information regarding the suppression mechanism. In addition, the suppression of the pristine \(\kappa\) value is less severe for our membrane (room-temperature \(\kappa = 65 W/mK\) for 120 nm film thickness) than for the previous Si nanowire geometry (room-temperature 50 W/mK for 160 nm nanowire diameter),\(^9\) which enables better heat transfer rates through the pristine region of the IWMT, an effect beneficial for heat flow control.

To further correlate the microstructural evolution of the material with the reduction in \(\kappa\), we performed systematic high-resolution transmission electron microscopy (HRTEM) and selected area electron diffraction (SAED) analysis on the Si membranes after irradiation with different doses as shown in Figure 2b. For the pristine sample, the analysis confirms that the membrane is perfectly single crystalline with the (100) plane parallel to the membrane plane. The membrane remains single-crystalline after the irradiation with doses up to \(10^{16}\) ions/cm\(^2\). Beyond this dose, an amorphous phase starts to form inside the crystalline Si matrix, which becomes significant after \(10^{17}\) ions/cm\(^2\) irradiation. At \(\approx 10^{18}\) ions/cm\(^2\), the membrane is completely amorphized, as seen from the corresponding SAED pattern where the diffraction spots are lost and only diffusive rings are seen. This gradual evolution from point defects to amorphization of Si is consistent with previous demonstrations of ion-irradiation driven amorphization in Si.\(^{19,20}\)

This analysis indicates that the suppression of \(\kappa\) is dominated by phonon scattering from point defects and amorphous domains in the low and high dose irradiation regimes, respectively. The measured \(\kappa(T)\) was fitted by adopting a conventional model\(^{24}\) that incorporates Umklapp, defects, and boundary scatterings using the Matthiessen’s rule and the Born–von Karman dispersion for acoustic phonons. The impurity scattering is the only free parameter adjusted in the fitting, and the obtained impurity scattering rate increases monotonically with the irradiation dose, as shown in the Supporting Information. The data demonstrate the focused ion irradiation to be a powerful tool for engineering local thermal conductivity of materials from the single-crystal phase continuously to the amorphous level. The spatial resolution is \(\approx 100 nm\) for the suppression of \(\kappa\) by a factor of 30, or even below \(\approx 10 nm\) for \(\kappa\) suppression by a factor of 2 (see Supporting Information for spatial resolution discussion). Below we will also show that such engineering of \(\kappa\) is largely reversible.

The ability of the IWMT platform to route heat flux using these fine features enables the construction of a range of microscale thermal devices. A thermal shield routes heat fluxes around a small object via exquisite design of local thermal conductivity tensors, shielding the object from the external heat flux. A thermal cloak is a special type of shield that hides the object from far-field thermal detection, which requires rerouting the heat flow around the object while also mandating that the external heat flow is unchanged by the addition of the cloak and object.\(^{2,3}\)

Thermal shield and cloak structures have been experimentally demonstrated at the macroscale (e.g., centimeters) using materials of contrasting thermal conductivity.\(^{3,4}\) However, these macroscopic devices are too large to control heat flows at the microscale. Our IWMT approach provides a monolithic platform, on which the thermal shield and cloak can be readily constructed. Proving the function of these devices, however, requires spatial mapping of temperature over the entire device area, which is challenging at the microscale. We employ thermoreflectance imaging (TRI)\(^{22,24}\) to map the temperature, which offers higher spatial resolution and temperature sensitivity than infrared thermography.

TRI is a noncontact optical technique for measurements of temperature variation on a surface by taking into account the change of optical reflectivity from the surface at different temperatures.\(^{5,26}\) The relative change in reflectivity (\(\Delta R\)) from the mean optical reflectivity (\(R\)) of a sample is related to the change in temperature (\(\Delta T\)) of the surface using the thermoreflectance coefficient (\(C_R\)) as \(\Delta R/R = C_R\Delta T\).\(^{27,30}\) Because \(C_R\) depends on the material, its surface roughness, and the wavelength of light used for measurement, it needs to be well calibrated for each sample prior to actual measurements.\(^{27,28}\) The illumination wavelength chosen to maximize\(C_R\) is typically in the range of 400–800 nm, and is suitable for submicron imaging. All TRI in this work is performed using blue light (\(\approx 470 nm\)), which is recommended for Si.\(^{30}\) The thermal cloaks are microscopic suspended membranes which make it impossible to directly calibrate them using steady-state temperature cycling, typically used in determining \(C_R\) of substrate-supported devices. For calibration of the suspended thermal cloaks, we use a transient calibration method, TransientCAL, which has been previously used in \(C_R\) estimation of nanoscale features.\(^{31}\) First, temperature cycling is used to determine a reference \(C_R\) of a substrate supported material that is also present in the suspended heater pads. The thermal cloak is uniformly heated and cooled and transient thermal images of the suspended assembly (thermal cloak and the heater pads) are taken. Using the transient calibration method, these images and the reference \(C_R\) is used for
obtaining a $C_{tr}$ map of the thermal cloak. This is then used to extract the temperature map of the suspended thermal cloak (see Methods for measurement conditions and setup and the Supporting Information for details).

We first design a thermal cloak with two concentric rings (hence called “bilayer” design). Three distinct regions with different $\kappa$ are created in the system as shown in the top panel of Figure 3a; $\kappa$ values of regions i, ii, and iii are designed to be 65, 40, and 2 W/m·K at room temperature, respectively. Finite-element simulations (Figure 3a, second through fourth panels) show that such a device effectively shields the thermal flux around the central circle when it is thermally biased with a nearly uniform temperature distribution in the shielded region (see Methods). The numerical heat flux map $|q_x(x,y)|$ (Figure 3a, fourth panel) obtained from Fourier’s law confirms that the heat flux in the shielded region is very small. The larger heat flux in the pristine region of the cloak shows that the heat is efficiently routed around the shielded region, compensating for the additional thermal resistance of the low-conductivity region iii. As discussed in the Methods, the simulations predict that the total heat flow is changed by less than 1% through the addition of the cloak, and that the heat flow is only very weakly dependent on the thermal conductivity of the cloaked object (i.e., the innermost zone ii in Figure 1a).

An experimental bilayer thermal cloak is written on the Si membrane with the ion beam (Figure 3b) following the design pattern, and regions i, ii, and iii are irradiated with a dose of 0 (pristine), $5 \times 10^{14}$, and $1 \times 10^{18}$ ions/cm$^2$, respectively. The temperature distribution of the system under thermal bias, mapped by the TRI technique (second and third panels in Figure 3b), shows that indeed the temperature in the middle circular region appears as a plateau with nearly isothermal distribution. Because the pristine region (region i) of the cloak has a higher thermal conductivity than the cloaked object, this indicates that the heat is mostly routed around the central region. The heavily irradiated region (region iii) prevents the heat from leaking into the central region, as is required for thermal cloaking.

Figure 3. Thermal cloaks demonstrated with the IWMT platform. (a) Top to bottom: design of a bilayer thermal cloak. The concentric i, ii, and iii regions were irradiated with 0 (pristine), $5 \times 10^{14}$, and $1 \times 10^{18}$ ions/cm$^2$, and have $\kappa$ of 65, 40, and 2 W/m·K at room temperature, respectively; simulated temperature distribution $T(x,y)$ of the design where the right and left edges are set at 330 and 300 K, respectively; FEM-simulated temperature profile of the device along the straight lines indicated in the panel above; local heat flux map $|q_x(x,y)|$, obtained from Fourier’s law $q_x = -\kappa \frac{dT}{dx}$. (b) Top to bottom: optical image of the bilayer thermal cloak realized on the IWMT platform following the design in a (the black dot is a dust particle contamination); temperature distribution of the device mapped by the TRI technique; measured temperature profile of the device along the straight lines indicated in the panel above; local heat flux map $|q_x(x,y)|$ obtained from Fourier’s law. (c) Top to bottom: optical image of a quadlayer thermal cloak realized on the IWMT platform following the design in the Supporting Information; temperature distribution of the device mapped by the TRI technique; measured temperature profile of the device along the straight lines indicated in the panel above; local heat flux map $|q_x(x,y)|$ obtained from Fourier’s law. The three regions of darkest, intermediate, and lightest contrast in the optical image were irradiated at 0, $2 \times 10^{15}$, and $1 \times 10^{18}$ ions/cm$^2$ and have $\kappa$ of 65, 30, and 2 W/m·K, respectively. The results in panels b and c show that the IWMT platform enables heat flux routing around the central region, as required for thermal cloaking.
thermal cloaking. The last panel of Figure 3b shows the experimental heat flux map \( q_y(x,y) \) obtained from the experimental temperature profile in the second panel of Figure 3b (see the Supporting Information for more details). This heat flux map demonstrates the thermal cloaking effect predicted by the simulations in Figure 3a. Lastly, it can be noted that the TRI temperature profiles and heat fluxes in Figure 3 display asymmetries that are not observed in the idealized thermal cloak simulations. The causes of this behavior are not fully understood, but could arise due to slight asymmetries in the device introduced during fabrication or due to nonuniform thermal tilts created during operation (see Supporting Information).

The IWMT platform can also be used to implement thermal cloak designs which have finer structural features than the simplest bilayer cloak. For example, Figure 3c shows a second device with a “quadlayer” design, in which two circular heat barriers instead of just one (as used in the first device) are fabricated. This quadlayer device is designed with three distinct values of \( \kappa \) of 60, 30, and 2 W/m K, which were realized on the IWMT platform by irradiation at 0, 2 \( \times 10^{15} \), and \( 10^{18} \) ions/cm\(^2\), respectively. TRI mapping in Figure 3c shows an enhanced uniformity of the temperature distribution in the inner circle compared to the bilayer design.

The experimental \( q_y(x,y) \) maps for both the bilayer and quadlayer cloaks show that the heat flux magnitude is significantly larger in the pristine region of the cloak than in the central cloaked object, indicating efficient thermal cloaking. The numerical predictions for the quadlayer temperature profile and heat flux are shown in Figure S6. For the bilayer (quadlayer) cloak, the mean experimental heat flux within the cloaked region \( \langle q_y \rangle = 1.6 \times 10^7 \text{ Wm}^{-2} \) \( (0.9 \times 10^7 \text{ Wm}^{-2}) \). These values represent factor of \( \sim 4 \) \( (\sim 5) \) reductions compared to the predicted values in the absence of a thermal cloak, about \( 6 \times 10^7 \text{ Wm}^{-2} \) \( (4.5 \times 10^7 \text{ Wm}^{-2}) \). In Figure S7, we also show that the temperature gradient in the cloaked region of the quadlayer cloak is insensitive to the total heat flux imposed at the edge of the cloak, as expected.

Both the bilayer and quadlayer devices utilize the finely structured local thermal conductivity of the IWMT to bend heat flow around the central object. These designs necessitate the continuous control of thermal conductivity and monolithic integration of different constituents as achieved in the IWMT. Obviously, the IWMT can be configured to construct other types of thermal devices as well. In the Supporting Information, a classical thermal rectifier based on temperature-dependent thermal conductivities is also demonstrated in the IWMT at 200 K, where the thermal conductance differs by 2.4% when the heat flow direction is reversed under a temperature bias of 30 K.

Lastly, we show that the suppression of \( \kappa \) in the IWMT is also reversible and rewritable. As shown in Figure 4a, the room-temperature \( \kappa \) is suppressed, relative to the original \( \kappa_{\text{pristine}} \approx 65 \) to 55 W/m K \( (0.85 \kappa_{\text{pristine}}) \) by irradiation of \( 10^{14} \) ions/cm\(^2\). A moderate anneal of the device in an \( N_2 \) gas at 300 °C restores the suppressed \( \kappa \) to 62 W/m K \( (0.95 \kappa_{\text{pristine}}) \). For irradiation at higher doses, the suppression is greater whereas the recovery of \( \kappa \) is less. For example, after irradiation using \( 10^{15} \) ions/cm\(^2\) (Figure 4b), \( \kappa \) is suppressed to 33 W/m K \( (0.51 \kappa_{\text{pristine}}) \) and annealing restores it to \( \kappa = 60 \) W/m K \( (0.92 \kappa_{\text{pristine}}) \). Importantly, after the initial annealing, the system appears to be locked into two stable states, such that subsequent treatments of irradiation and annealing under the same condition always set and reset \( \kappa \) between the low \( (33 \) W/m K) and high \( (60 \) W/m K) values. Therefore, akin to nonvolatile resistive random-access memory, the initial irradiation serves as a forming process, and the subsequent annealing and irradiation can repeatedly erase and encode the thermal conductivity distribution in the membrane.

In this work, a versatile platform is developed to reversibly write microscale thermal metamaterials. Using the platform, we experimentally demonstrate thermal cloaking at the microscale for the first time. The capability of controlling microscale heat flow using this platform opens opportunities to explore novel ideas for microscopic thermal management. As the spatial resolution of the ion writing is much smaller than that of the phonon mean free path (MFP) in crystalline Si \( (\sim 50\% \) of the heat carried by phonons with MFPs of at least \( 1 \) \( \mu \)m at room temperature, and longer for lower temperatures\(^ {34} \), sub-MFP-sized devices can also be written onto the IWMT platform, which are potentially able to ballistically route thermal phonon transport for exploring exotic thermal effects beyond the Fourier law\(^ {35,15} \). The gradual and controlled transition from the crystalline to amorphous phase in the IWMT also offers an ideal material system with which to test novel thermal conduction physics, such as the transition from thermal phonons to heat-carrying local vibration modes (or the so-called propagons and locons, respectively)\(^ {36} \). Using other ultrathin materials such as graphene\(^ {37} \) as the membrane, an even wider range of modulation of thermal conductivity could be achieved, which may enable improved performance or new functions. In this sense, the ion-write microthermotics demonstrated here has the potential to serve as a versatile platform with which to control heat flow at the microscale, akin to what nanofluidics does for fluids.

**Methods. Device Fabrication.** Fabrication of the suspended microdevices started with 6 inch silicon-on-insulator wafers (SOI) (vendor, SOITEC). The device layer of the SOI was lightly p-doped and single-crystalline with a resistivity of...
20–30 Ω cm, hole concentration of $\sim 10^{15}$ cm$^{-3}$, (100) crystal orientation, and thickness of 200 ± 5 nm (thinned down to 120 nm in the final device). The buried silicon oxide layer and handle substrate were 200 ± 10 nm and 625 ± 10 μm thick, respectively. The silicon layer was patterned to define and form the silicon membrane below and between the two pads using photolithography (ASML S500/300 DUV stepper) and a deep reactive ion etching process (SPTS ICP-SR deep reactive ion etcher). A 300 nm thick, Si-rich, low-stress SiNx layer was deposited by low-pressure chemical vapor deposition, followed by metallization of 2 nm Cr and 50 nm Pt for the electrodes using sequentially the stepper photolithography, sputtering, and lift-off. Afterward, the SiNx layer was patterned followed by reactive ion etching to expose the silicon membrane channel bridging the two pads. The wafer was cut into small chips of 9.2 x 9.2 mm in size, each containing 72 microdevices with silicon membrane channel of various lengths. After defining the width of the silicon membrane channel using electron beam lithography (Crestec CABL-9000) and ICP etching (Oxford PlasmaLab 150 inductively coupled plasma etcher) using hydrogen bromide, the devices were finally released from the substrate by backside photolithography and selective deep reactive ion etching.

**Irradiation.** The suspended Si membrane was irradiated by He$^+$ ions using a helium ion microscope (HIM, Zeiss ORION NanoFab). The incident ions were generated at the atomically sharp gas field-ionization source using a helium gas pressure of $\sim 2 \times 10^{-6}$ Torr. The acceleration potential and beam spot size were set to 25 kV and $\sim 1$ nm diameter (10 μm aperture), respectively. The irradiation doses were controlled by varying the beam current, dwell time, and scan spacing. A 2 pA beam current, 1 μs dwell time, and 0.25 nm scan spacing were used for most of the irradiation processes, except for the highest doses (10$^{17}$ and 10$^{18}$ ions/cm$^2$), which used a 10 nm spacing and 40 pA beam current (70 μm aperture). All patterns were designed and written using the NanoPatterning and Visualization Engine (NPVE) program from Fibics, Inc.

**Thermal Conductivity Measurements.** Thermal conductance ($G$) of the membrane was measured using the suspended micropad devices placed in a cryostat with a vacuum chamber (pressure <10$^{-6}$ Torr). Serpentine Pt electrodes predeposited on the two symmetric, suspended SiN$_x$ pads act as microheater and thermometer for the thermal measurements. $G$ was measured using the expression $G = (Q \times \Delta T_\text{r}) / \left[(\Delta T_h)^2 - (\Delta T_c)^2\right]$, where $Q$ is the Joule heating power of the Pt microheater, and $\Delta T_h$ and $\Delta T_c$ are the temperature change of the hot and cold SiN$_x$ pads, respectively.

When a dc current (0–15 μA) flowed into the Pt microheater on the heating pad, $\Delta T_h$ and $\Delta T_c$ were simultaneously recorded by lock-in amplification of the Pt heater/thermometer resistance to an additional ac current (500 nA) applied to the microheater. The temperature coefficient of resistance (TCR) of the Pt heater/thermometer was precalibrated by quadratic fitting to its resistance as a function of global temperature prior to each measurement. The monolithic silicon device fabrication method ensures that there are no thermal contact resistances at the device-pad interface. The analysis of the measurement above assumes that the heating and sensing pads are isothermal; as discussed in the Supporting Information, this isothermal-pad analysis is valid in this scenario because the sample’s thermal resistance (i.e., 1/$G$) is always at least an order of magnitude larger than the spreading thermal resistance due to conduction near the pad-device interface. The thermal conductivity measured in this study has a <5% error, considering the errors of electrical measurements, as well as measurements of the sample dimensions.

**TEM Characterization of Irradiated Silicon Membrane.** HRTEM data was taken in an FEI Titan at 300 keV from a test Si membrane sample. The sample was irradiated to the same dose as the devices but was thinner ($\sim 30$ nm, as opposed to 120 nm as in the devices) to allow for HRTEM imaging. SAED data was taken in a Zeiss Libra TEM at 200 keV. Single devices with irradiated Si regions were mounted into an appropriate substrate using a low-temperature curing epoxy (200 °C). Using the pristine crystalline region of each film, the sample was tilted until the electron beam direction coincided with the Si (100) direction. Then, diffraction data was acquired for each radiation dose.

**Thermoreflectance Imaging.** The thermoreflectance coefficient ($C_\text{tr}$) was calibrated for the suspended materials following the established TransientCAL method presented in ref 31 followed by their steady-state thermal characterization. We used the TRI measurement setup (Microsanj NT-210B) in which an LED through a microscope objective illuminated the device under test (DUT) as it was biased, and a phase-locked CCD camera captured the reflectance signals from the surface of the DUT. A blue-light LED (wavelength $\sim 470$ nm), which is suited to silicon devices, was used throughout the experiment with a 100X, 0.6 NA (numerical aperture) microscope objective. We determined the $C_\text{tr}$ of a substrate supported platinum (Pt) contact pad using steady-state measurements by hot–cold temperature cycling. A 20 μs biasing pulse (at 2% duty cycle, total period 1 ms) was applied to both heaters and transient thermal images of the suspended assembly (Si membrane and heater pads) cooling to ambient temperature were captured up to 670 μs. These were used with the TransientCAL technique and a reference $C_\text{tr}$ to obtain the calibration $C_{\text{tr}}$ map of the membrane. We extracted the $C_{\text{tr}}$ of a SiN$_x$ region on the (suspended) heater using the Pt $C_{\text{tr}}$ and used this as the reference $C_{\text{tr}}$ with the transient images to obtain the $C_{\text{tr}}$ map by the TransientCAL technique.

**HRTEM Characterization of Irradiated Silicon Membrane.** A rectangular area present on the (suspended) heater pad, was more suitable as the reference region than the narrow Pt serpentine region. After the calibration procedure, a 20 μs bias pulse was used to heat one side of the Si membrane to obtain thermoreflectance signals at the steady state operating temperature. These were used with the $C_{\text{tr}}$ map to extract the heat flow across the device and obtain the final temperature map on the surface of the Si membrane.

The TRI measurements were done near room temperature. Convective heat loss from the device is estimated to be $\sim 10^{-6}$ W, which is much lower than the heat flux flowing in the Si membrane (about $10^{-4}$ W). Similarly, the temperature of the device was kept no more than $\sim 350$ K, and radiative heat loss is estimated to be less than $10^{-8}$ W.

**Concept and Simulation of Thermal Cloak.** Two-dimensional (2D) thermal cloaks hide thermal information about a cloaked object from far-field measurements in the 2D plane. To illustrate thermal cloaking, consider one-dimensional heat flow through a medium of thermal conductivity $\kappa_m$. If an object with a thermal conductivity $\kappa_o \neq \kappa_m$ is embedded in the medium, the steady-state temperature profile and heat flux vectors are modified. The goal of the thermal cloak is to restore the original 1D temperature profile and heat flows outside of the cloaked region, such that thermal measurements do not
reveal the presence of the cloaked object. Thermal cloaks achieve this restores by routing the heat around the object without changing the magnitude of the total heat flow. Because the heat does not flow through the object, the cloaked object is ideally isothermal and the total heat flow is insensitive to \( \kappa_w \).

Building on the designs of previously developed macroscopic thermal cloaks, we designed microscale thermal cloaks with bilayer and quadlayer designs. We consider cylindrical thermal cloaks patterned into a background material that is lightly irradiated to achieve an intermediate conductivity \( \kappa_m \) (region ii in Figure 2). Thin layers of heavily irradiated Si with a low thermal conductivity \( \kappa_l \) (region iii) encircling the object provide insulation to prevent heat flow toward the cloaked object, whereas thicker cylindrical regions of pristine Si with a high thermal conductivity \( \kappa_p \) (region i) route the heat around the object. The rectangular background region has a length \( L \) along the direction of global heat flow \( x \), width \( w \) in the orthogonal in-plane direction \( y \), and thickness \( t \). We perform 2D steady-state finite-element method (FEM) COMSOL simulations to find the temperature profiles and total heat flows through cloaked objects. We applied constant temperature boundary conditions at \( x = 0 \) and \( x = L \) and adiabatic boundary conditions at \( y = 0 \) and \( y = w \). We ensure mesh convergence by calculating the total heat flow in the \( x \)-direction and verifying that the total heat flow is converged to <0.01%.

From a design perspective, it is desirable to obtain large thermal conductivity contrast to enable efficient cloaking of large cloaked regions. For a given range of finite conductivity ratios and a certain cloaked object size, multiple cloak designs (e.g., bilayer, quadlayer, composite, or transformationalthermic) can potentially be implemented; the design details and underlying theory of such cloaks have been previously discussed in the literature. To compare the performance of the two cloak options selected for the IWMT demonstrations, we introduce several cloaking metrics. First, we want the total heat flow \( Q \) from the hot edge at \( T_h \) to the cold edge at \( T_c \) with the cloaked object present to be identical to the total heat flow through a homogeneous material with conductivity \( \kappa_m \). Therefore, a perfect cloak would have \( Q^* = \frac{Q_L}{\kappa_m(T_h - T_c)} = 1 \).

Second, we want \( Q \) to be insensitive to the value of the object thermal conductivity \( \kappa_o \), demonstrating that the cloak is effective for many objects. One way to quantify this sensitivity is the dimensionless derivative \( \delta = \frac{L}{w t (T_h - T_c)} \frac{\partial Q}{\partial \kappa_o} \) which should be small compared to unity. Lastly, we want the dimensionless temperature gradient at the center of the object \( g = \frac{L}{(T_h - T_c) \delta t} \) to be smaller than unity, indicating that the cloak is effectively rerouting the heat around the object. We note that using \( \kappa_l \approx \kappa_m \) provides the most challenging cloaking test: if \( \kappa_l \ll \kappa_o \) then \( \delta \) is always much less than 1 (even for a poor cloak) because the object is a “thermal short circuit” with no appreciable temperature gradients. Similarly, if \( \kappa_l \gg \kappa_o \) then \( \delta \) is always much less than 1 (even for a poor cloak) because the object is a “thermal open circuit” and all of the heat flows around the cloaked object.

For our final designs demonstrated in Figure 3, we selected the geometric parameters to optimize these cloaking metrics while ensuring that the object size was large enough to enable accurate TRI measurements and that the highly irradiated section was as small as possible. The limit on the highly irradiated thickness arises because the TRI signal displays artifacts due to irradiation-induced surface roughness. We found that our bilayer cloak design in Figure 3a with \( \kappa_o = \kappa_m = 40 \text{ W/m-K} \) provides simulated cloaking values of \( Q^* = 0.996, \delta = 0.07, \) and \( g = 0.50 \). In particular, the small value of \( \delta \) indicates that the measured \( Q \) is quite insensitive to the object’s thermal conductivity, because in the absence of a thermal cloak this geometry has \( \delta = 0.24 \). The simulations of the quadlayer cloak shown in Figure 3c with \( \kappa_o = \kappa_m = 30 \text{ W/m-K} \) displays similar cloaking capabilities (\( Q^* = 1.07, \delta = 0.07, \) and \( g = 0.53 \)) as the bilayer cloak in Figure 2c. Lastly, we note that our simulations neglect the interfacial thermal boundary resistance \( R_i \) between the regions of different irradiation levels because \( R_i \) is expected to be many orders of magnitude smaller than the thermal resistances due to bulk conduction. For example, molecular dynamics simulations of the crystalline/amorphous silicon interface found that \( R_i = 0.2 \times 10^{-9} \text{ K m^2 W}^{-1} \), a thermal resistance which is smaller than the thermal resistance due to conduction through 1 nm of amorphous silicon. This insensitivity to \( R_i \) is another advantage of the IWMT platform compared to macroscopically assembled thermotric devices.

**ASSOCIATED CONTENT**

Supporting Information
The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.9b00984.

Thermal conductivity modeling, SRIM simulation of ion irradiation, thermoreflectance (TRI) imaging operation, thermoreflectance coefficient (\( C_{tr} \)), calibration, and temperature mapping. TRI measurement uncertainty discussion, verifying validity of thermal conductivity measurement, demonstration of thermal rectification with IWMT, experimental heat flux mapping in the thermal cloaks, quadlayer thermal cloak simulations, negligible heat flux in the cloaked region at different levels of total heat flux (PDF)

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**Author Contributions**

H.S.C., R.P., and G.W. contributed equally to this work. J.W. conceived this project and H.S.C. and W.L. fabricated the suspended microdevices. H.S.C. and F.I.A. performed the HIM irradiation. H.S.C. measured the thermal conductivity. M.S. and A.M. performed HRTEM and SAED analysis. R.P. and J.B. performed the TRI experiments. G.W. and C.D. performed the FEM simulation and thermal conductivity modeling. H.S.C., R.P., G.W., and J.W. analyzed the data and drafted the paper. All authors discussed the results and contributed to writing the manuscript.

**Author Contributions**

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Notes
The authors declare no competing financial interest.

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