Spatially mapping of the thermal conductivity of graphene by an opto-thermal method

Oliver Braun,†,‡ Roman Furrer,† Pascal Butti,† Kishan R. Thodkar,†,§ Ivan Schorubalko,† Ilaria Zardo,‡ Michel Calame,†,‡,¶ and Mickael L. Perrin*;†

†Empa, Swiss Federal Laboratories for Materials Science and Technology, Transport at Nanoscale Interfaces Laboratory, Überlandstrasse 129, CH-8600 Dübendorf, Switzerland
‡Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland
¶Swiss Nanoscience Institute, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland
§Current address: Department of Mechanical and Process Engineering, ETH Swiss Federal Institute of Technology, Tannenstrasse 3, CH-8092 Zürich, Switzerland

E-mail: mickael.perrin@empa.ch

Abstract

Mapping the thermal transport properties of materials at the nanoscale is of critical importance for optimizing heat flows in devices. In particular, graphene’s extraordinary high thermal conductivity values has attracted significant interest over the past decade. Several methods to determine the thermal conductivity have been developed, most of them yielding an average thermal conductivity of the sample under study, thereby disregarding the role of local variations. Here, we present a method for the spatial mapping of the thermal conductivity of suspended graphene using a combination of confocal Raman spectroscopy and finite-element calculations. Our method is
non-invasive, contact-free and does not required on-chip heaters. We demonstrate the working principle of this method by extracting the thermal conductivity map of a suspended graphene sheet that has been irradiated using He-ions. This method paves the way for spatially resolving the thermal conductivity of other types of two dimensional materials, also after incorporation into a device.

Introduction

Thermal properties of materials are of crucial importance for optimizing thermal management in nanoscale device, with as key material property the thermal conductivity\cite{1}. The thermal conductivity is typically determined by monitoring the response of the sample temperature to a local heat source, in combination with an analytical or numerical model. For instance, for bulk materials, the well-known $3\omega$ technique\cite{2,3} is used, while for nanoscale materials methods such micro-bridge thermometry\cite{4} and micro-Raman spectroscopy\cite{5} provide the thermal conductivity of the material under study. However, with these methods, only an average thermal conductivity value is obtained. For going beyond this mean material property value, approaches mapping the temperature distribution in the sample are required. A range of techniques have been developed for nanoscale thermometry, such as time-domain thermoreflectance\cite{6}, Raman spectroscopy\cite{7,8}, scanning thermal probe microscopy\cite{9-11}, polymer imprint thermal mapping\cite{12} and electron energy loss spectroscopy\cite{13}, providing means to locally map the sample temperature. Temperature mapping has recently also gained particular interest to investigate heat flows in various transport regimes. For example, a crossover from ballistic to diffusive thermal transport has been observed experimentally\cite{14} and predictions have been made for the emergence of viscous flows once heat transport enters the Poiseuille and Ziman regime\cite{15-17}.

Of particular interest are also the thermal properties of layered materials. Due to their broad range of conductivity values and their atomically thin nature, such materials are highly relevant for heat management at the nanoscale\cite{18,19}. One of the most appealing materials is
graphene, with extraordinarily high thermal conductivity values. However, extracting the thermal properties of 2D materials is challenging, in particular when suspended to reduce the influence of the substrate. For example, time-domain thermoreflectance cannot be applied to 2D materials, as the material is too thin. Scanning thermal probe microscopy, on the other hand, despite possessing nanometer resolution, is highly delicate to perform on suspended sheets of atomically thin 2D materials. Moreover, both techniques rely on the presence of on-chip heaters for pumping heat into the system. Raman spectroscopy can overcome these difficulties, as it can utilise the excitation laser to locally heat the device, while at the same time measuring the local temperature. Moreover, Raman spectroscopy can easily be performed on suspended graphene for eliminating the influence of the substrate. In fact, using Raman spectroscopy, Balandin and Ghosh et al. determined the thermal conductivity of suspended graphene to be as high as $\sim 5000 \text{ Wm}^{-1}\text{K}^{-1}$ at room temperature. Their opto-thermal method, measuring the shift of the Raman G-band upon laser irradiation for estimating the local temperature, has been extensively used in literature since. Alternatives based on the intensity ratio of Stokes to anti-Stokes Raman scattering or the Raman 2D-band have also been reported.

Using this opto-thermal method, the influence of the quality and structure of the graphene, as well as the environment have been extensively investigated. For instance, Cai et al. reported values for $\kappa$ exceeding $\sim 2500 \text{ Wm}^{-1}\text{K}^{-1}$ for suspended graphene grown by chemical vapor deposition (CVD), and Chen et al. studied the influence of the environmental on thermal conductivity of graphene. Isotopically pure $^{12}$C (0.01 % $^{13}$C) graphene has been shown to exhibit $\kappa = 4000 \text{ Wm}^{-1}\text{K}^{-1}$, a factor of two higher than $\kappa$ in graphene composed of a 1:1 mixture of $^{12}$C and $^{13}$C. Also, the influence of CVD-graphenes polycrystallinity on the thermal conductivity was studied by Lee et al. and Ma et al., revealing that smaller grain sizes drastically reduce $\kappa$ due to grain boundary scattering. Along similar lines, wrinkles, oxygen-plasma induced defects and ebeam irradiation have been shown to reduce the thermal conductivity.
In all the above-mentioned Raman spectroscopy studies, the temperature of a suspended graphene membrane was determined upon heating by a laser. The 1-dimensional heat equation assuming a cylindrical symmetric was then used to fit the experimental temperature and extract the thermal conductivity values. Despite the extensive use of the opto-thermal method in literature, this approach yield only an average thermal conductivity value, not a spatially resolved map. Moreover, the reported values vary over several orders of magnitude. This spread is attributed to several factors. First, as a circular symmetry for the heat dissipation is assumed, local variations caused by defects, folds, contaminants, etc are neglected. As a result, a single value of the thermal conductivity is assigned to the entire membrane. Second, these same local variations result in a high sensitivity of the extracted temperature, and hence of the thermal conductivity, on the exact laser spot position.

Here, we introduce an opto-thermal method that allows for two-dimensional mapping of the thermal conductivity of suspended graphene membranes. The presented method relies on a combination of scanning µ-Raman spectroscopy with finite element method (FEM) calculations. The workflow for our approach is presented in Fig.1. In the first, experimental stage, a series of two-dimensional Raman spectroscopy maps are used to construct a temperature map of the membrane upon illumination. More specifically, Raman maps are recorded for various temperatures and at a low laser power. This series of maps is used to construct a calibration map of the Raman peak shifts with temperature. Then, another Raman map is recorded at high-laser-power, which, combined with the calibration map, is used to construct a temperature map of the membrane upon illumination.

This constructed temperature map is used as input for the FEM-based fit procedure. Here, the starting point is an initial guess of the thermal conductivity. From this two-dimensional map, a temperature map upon illumination at high laser power is computed. The thermal conductivity is iteratively adjusted by comparing the target temperature to the computed one. This process is repeated until convergence is reached.
Figure 1: Experimental and Finite Element Method description. a) Schematic drawing of the suspended graphene membrane. b) Experimental workflow to obtain a temperature map upon laser illumination and computational workflow to fit the corresponding thermal conductivity map. c) assumed thermal conductivity map. d) schematic representation of the sample and the calculation mesh. Temperature profiles of the graphene membrane with the heating laser spot at three different positions 1)-3). e) Temperature profile upon laser illumination.
The computation method is depicted in Figure 1 and validated on a simulated temperature profile that is shown in section 2 of the Supporting Information. We show that a predefined thermal conductivity map can be recovered using our FEM-based fitting procedure. We then apply this method to extract the thermal conductivity of a pristine graphene membrane that is suspended over a silicon nitride frame. Finally, we demonstrate that the thermal conductivity of the graphene membrane can be tuned in a controlled way by the introduction of He$^+$-ion induced defects in the membrane.

Results

Experimental temperature maps

Pristine graphene exhibits distinct Raman peaks in the G-band ($\sim 1581$ cm$^{-1}$) and the 2D-band ($\sim 2680$ cm$^{-1}$). In addition, defective graphene exhibits a peak in the D-band ($\sim 1350$ cm$^{-1}$), its height is proportional to the number of defects present. Here, Raman spectroscopy is employed to obtain the lattice temperature of the suspended graphene membranes upon illumination, inferred from the peak position shift with temperature. In the following, we focus on the 2D-band due to its high sensitivity to temperature changes of around $-0.07$ cm$^{-1}$/K. Alternative approaches reported in literature rely on the G-peak due to its high linearity in peak shift versus temperature.

The membranes are prepared as follows. Graphene grown by chemical vapor deposition (CVD) on copper is transferred onto a Si$_3$N$_4$ frame using a wet transfer technique. For more details, we refer to the experimental section. The thermal anchoring of the graphene to the Si$_3$N$_4$ frame was ensured using a Ti/Au (5/40 nm) layer evaporated on top of the frame. The prepared membranes are clamped on a hot plate that is in turn fixed on the piezo stage of the Raman microscope for mapping. To ensure thermalization of the membrane, a waiting time of $\sim 45$ min is used before each Raman map is acquired.

Figure 2a) presents maps of the Lorentzian-fitted 2D-peaks acquired at temperatures $T_1$-
Figure 2: Experimental determination of temperature map. Spatially resolved mapping of laser induced temperature rise of graphene. a) Raman 2D-peak position obtained with $P_{laser} = 0.25 \text{ mW}$ at different bath temperatures $T_1$ to $T_7$. b) density plot of the temperature evolution of the 2D-peak position. Two spacial points (center and edge) are highlighted to represent the method. The inset shows a histogram of $d\omega_{2D}/dT$ of the complete membrane. c) spacial distribution of change in Raman shift per temperature change $d\omega_{2D}/dT$ obtained from linear fits to the data shown in b). d) Raman 2D-peak position obtained with $P_{laser} = 4 \text{ mW}$ at 297 K. e) temperature distribution obtained by combining the results from c) and d)
ranging between 298 K and 425 K. The Raman spectra have been acquired at low set laser power (0.25 mW) to limit any heating effects using a 532 nm excitation laser. For each pixel, the peak shift with bath temperature \( \frac{d\omega_{2D}}{dT} \) is fitted using a first-order polynomial, as shown in Fig. 2(b). The inset of the figure presents a histogram of the slopes, showing a Gaussian distribution centered around -0.07 cm\(^{-1}\)/K. The spatial distribution of the peak shifts with temperature is displayed in Fig. 2(c), showing substantial spatial variations. Once this calibration map is acquired, a Raman map of the graphene membrane is acquired at high laser power (4 mW), as shown in Fig. 2(d). The high laser power causes the graphene to locally heat up, resulting in a shift in the Raman peak position. By combining this high-power measurement with the \( \frac{d\omega_{2D}}{dT} \) map obtained at low laser power, a map of the averaged temperature within the laser spot is obtained for each laser position, as shown in Fig. 2(e). We note that local variations in the temperature upon illumination on the order of 50-100 K are observed, highlighting the importance of spatially mapping the temperature, rather than using a single laser spot position to calculate the thermal conductivity.

FEM calculations are employed for the computation of the temperature map of the system upon laser illumination for a given spatial thermal conductivity distribution. This concept is illustrated in Fig. 1(c)-e). As input, a two-dimensional map of the thermal conductivity is provided, of which an example is shown Fig. 1(c). Fig. 1(d) presents the layout of the system (not to scale). It consists of two concentric circles; the largest one represents the graphene on the support \( R_{\text{supp}} \) with a radius of 20 \( \mu \text{m} \), while the smaller one depicts the suspended graphene \( R_{\text{memb}} \), with a radius of 4 \( \mu \text{m} \). A third circle represents the laser spot \( r_0 \) and is scanned across the graphene membrane. Within the domain, the two-dimensional, steady-state, heat equation in Cartesian coordinates is solved, as defined below:

\[
\frac{\partial}{\partial x} \left( \kappa(x, y) \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( \kappa(x, y) \frac{\partial T}{\partial y} \right) = -Q - Q_{\text{laser}}(x, y) \tag{1}
\]

where \( T(x, y) \) is the temperature distribution, \( \kappa(x, y) \) the thermal conductivity, \( Q \) the heat losses via convection and/or the support, and \( Q_{\text{laser}}(x, y) \) the heating due to the laser.
As boundary condition, a fixed temperature on the outer boundary of the support is used, set to the bath temperature. For graphene on the support, the following heat losses are used to account for the interaction with the support and convection:

\[ Q_{\text{support}} = -g \frac{T - T_0}{t_{gr}} - h_{\text{conv}} \frac{T - T_0}{t_{gr}} \]  

(2)

where \( T_0 \) [K] is the bath temperature, \( t_{gr} \) [m] the thickness of graphene (0.334 nm), \( h_{\text{conv}} \) \([\text{Wm}^{-1}\text{K}^{-1}]\) the convection coefficient of air, and \( g \) \([\text{Wm}^{-1}\text{K}^{-1}]\) is the interface thermal conductivity between the support and the graphene. For suspended graphene, only convection is present:

\[ Q_{\text{suspended}} = -2h_{\text{conv}} \frac{T - T_0}{t_{gr}} \]  

(3)

where the factor of 2 accounts for the convection occurring at both sides of the membrane.

The laser is modeled as a Gaussian distribution, introducing a heat source equal to:

\[ Q_{\text{laser}}(x, y) = \frac{Q_{\text{abs}}}{2\pi r_0^2 t_{gr}} e^{-\frac{(x-x_0)^2}{2r_0^2} - \frac{(y-y_0)^2}{2r_0^2}} \]  

(4)

where \( Q_{\text{abs}} \) [W] is the absorbed laser power, \( r_0 \) [m] the laser spot size, \( x_0 \) [m] and \( y_0 \) [m] the laser position. All calculation parameters can be found in table S1 of the supporting information. Furthermore, a homogeneous absorption, defined by the fine structure constant \( \alpha = e^2/\hbar c \) giving \( \pi \alpha \approx 2.3 \% \), is initially assumed for the validation of the model.

While scanning the laser across the membrane, the full temperature distribution is calculated for each laser spot position on the membrane (three examples are provided in Fig. 1d). For each of these temperature distributions, the average temperature within the laser spot is calculated, from which all values are combined to obtain a two-dimensional map of the graphene temperature upon illumination. This induced-temperature map is presented in Fig. 1e) and is the temperature map that is obtained experimentally upon illumination of the sample with high laser power.
Experimentally, however, the situation is reversed. From a series of Raman maps, we construct the induced temperature maps (see Fig. 1b), while the corresponding thermal conductivity maps are unknown. To obtain the thermal conductivity map, an iterative minimization procedure is employed. Its starting point is an initial (typically uniform) guess of the thermal conductivity. In each iteration of the process, the corresponding induced temperature map is compared to the experimental temperature map, after which the thermal conductivity is adjusted pixel-wise according to the temperature difference. This process is repeated until the mean error across the membrane drops below the set threshold value \( i.e., \) the procedure is converged. We first validate our numerical method on a simulated system with a known thermal conductivity map, as shown in supporting information Fig. 2.

**Thermal conductivity map based on fitted experimental temperature map**

The induced temperature map obtained in Fig. 2 is used as input for the iterative procedure introduced in Fig. 1 to obtain the thermal conductivity map. Besides this calibration procedure knowledge of the absorbed laser power is essential. Due to the full suspension of the graphene, measuring the transmitted laser power can be done by placing a powermeter below the sample while scanning over the graphene membrane. By subtracting the powermeter maps obtained on a graphene membrane from the one obtained on an empty hole in the SiN frame and using \( 1 - T = A + R \), where \( T \) represents the transmitted, \( A \) the absorbed and \( R \) the reflected part of the light while assuming the reflected part being neglectable due to the low optical crosssection a map of the absorbed laserpower absorption is obtained (see Fig. S1). It is observed that the absorption map shows variation over the graphene map indicating that residues, folds, and defects can do have an influence on the locally absorbed laserpower and hence heating up of the graphene membrane nonhomogenous. In particular, at the edges of the suspended graphene, the determination of the absorbed laser power is experimentally challenging. For this reason, the thermal conductivity maps are fitted using
a uniform absorption of 2.7 % for the suspended graphene and double that value (5.4 %) for the supported graphene. All the model parameters are summarized in Table S1 of the supporting information. Importantly, in the model, a diffraction-limited spot size of 300 nm is assumed. Moreover, the thermal conductivity of the graphene on the supporting part, the thermal coupling to the substrate, as well as the convection parameter are taken from literature. Finally, we note that our model has difficulties modelling the transition from the suspended graphene to the support graphene once the laser spot is in the vicinity of the edge. Several reasons may cause this: 1. reflections of the excitation laser at the edges of the support may lead to an increase in the deposited laserpower 2. quenching of the Raman scattered light on the substrate may lead to an overestimation of the local temperature as the 2D peak of the suspended graphene is more pronounced than that of the supported graphene. To circumvent this issue, the thermal conductivity of the first 0.5 \( \mu \text{m} \) of the membranes away the support are not fitted and kept at a fixed value.

First, the thermal conductivity is fitted for 100 iterations, after which the absorption is fitted for the same number of cycles. More details about this procedure can be found in section 2 of the supporting information. For numerical stability reasons, we put a lower value on the thermal conductivity at 100 Wm\(^{-1}\)K\(^{-1}\).

Fig. 3a presents the experimental temperature map, as obtained in Fig. 2e, alongside the fitted temperature map in Fig. 3b. The two maps closely resemble each other. The corresponding thermal conductivity map is presented in Fig. 3c. We find thermal conductivity values ranging from 500 to 2000 Wm\(^{-1}\)K\(^{-1}\) and an averaged value of 1007 \( \pm 450 \) Wm\(^{-1}\)K\(^{-1}\) in agreement with values reported previously as well as by other methods. In Fig. 3c), we present a histogram of the fitted thermal conductivity map for increasing bath temperatures. The bar plots show that for increasing temperature, a gradual decrease in thermal conductivity is observed. This behavior follows the trend observed by others.
Thermal conductivity of defect engineered graphene

As a final demonstration of the capability of the presented method, we study the thermal conductivity of a graphene membrane that is exposed to He\textsuperscript{+}-ions using focused ion beam lithography. As shown previously, He\textsuperscript{+}-ions can be used to induce, in a controlled fashion, defects in suspended graphene membranes and other two-dimensional materials.\textsuperscript{34,35}

Figure 4a) presents the exposure pattern as well as the used irradiation doses. The membrane is divided into four quadrants, with the He\textsuperscript{+}-ion irradiation steadily increasing in the counter-clockwise direction, starting in the lower left with no He\textsuperscript{+}-ion dose. Manually selected representative Raman spectra of each quadrant are presented in Fig.S2, exhibiting all the characteristic graphene peaks. The selection of the representative Raman spectra can also be done by an advanced clustering approach to avoid any human bias as described elsewhere.\textsuperscript{36} Upon an increase of the He\textsuperscript{+}-ion dose, the D-band intensity steadily increases. From the intensity ratio of the D and D’ band I(D)/I(D’), the type of defect can be deduced.\textsuperscript{37}
Figure 4: Thermal conductivity of defect engineered graphene. a) Schematic image of a suspended graphene membrane. The areas where the membrane was exposed to He$^+$-ions and their corresponding dose is indicated with different colors. b) experimentally observed temperature map. c) map of thermal conductivity used in the model to calculate d) fitted temperature map. e) Histogram of the thermal conductivity for various defect densities.
We find an intensity ratio of ∼15 on the defect engineered graphene. This value is comparable to the reported intensity ratio of ∼13 for sp³ type of defects. We employ the same procedure as presented in Fig. 3 for the extraction of the temperature. Figure 4b) presents the induced temperature map upon a 4 mW laser illumination. In this plot, the four quadrants are visible, with the lowest temperatures recorded in the (unexposed) lower left section of the membrane, and the highest one in the upper left (most exposed). This temperature map is used as input for the iterative FEM-based fitting procedure, resulting in the fitted thermal conductivity map shown in Fig. 4c) and the fitted temperature map in Fig. 4d). Fig. 4e) presents a histogram of the thermal conductivity of each of the four quadrants. A steady decrease in average conductivity is observed, from ∼1000 Wm⁻¹K⁻¹ for the no He⁺-ion irradiation, and ∼100 Wm⁻¹K⁻¹ for the highest He⁺-ion dose. This decrease in thermal conductivity with increasing defect density is in agreement with previous reports.

Discussion

A limitation of the presented method is the time consumption of the temperature calibration, reducing it’s use in high-throughput applications. As the laser power is low, acquiring the two-dimensional Raman map at each temperature requires several hours. This long acquisition time can also lead to a drift in the sample position during the measurement. To reduce this drift, a proper clamping of the sample and a good thermalization of the sample with the environment is crucial. Furthermore, as changing the bath temperature leads to shifts of the sample position, the Raman maps acquired at various temperatures need to be aligned one versus the other.

A second limitation is the fixed value for the absorption of 2.7 % that is used for the first 100 cycles of the fitting procedure, after which the absorption is fitted as well. The accuracy of the model may be improved by experimentally determining the absorption at the various bath temperatures. Ideally, the absorption would be measured by simultaneously monitoring
the transmitted, and reflected laser power while scanning across the sample. We stress that simultaneously measuring both components is crucial, as contaminations and residues on the membrane may scatter the laser light, leading to a reduction in the transmitted light, but not to an increase in absorption. However, such a measurement is challenging, and technically unfeasible in our current setup.

Despite the previously mentioned limitations, our method is well suited for studying the thermal properties of two-dimensional materials. It can also be used for few-layer Van der Waals materials. Furthermore, the method is expendable from two to three dimensions, allowing for modelling of more complex device geometries, including, for instance, stacks of 2D-materials, or the presence of contact electrodes of finite thickness.

As such, it could be used for assessing the material quality of upon device integration. Also, as the individual two-dimensional materials in a stacked geometry each have a distinct Raman signature, one could also investigate the subsurface thermal properties of materials, such as, for instance, graphene embedded in a thin hexagonal boron nitride layer. Alternatively, when the material under study is on substrate or thick enough, other means of determining the temperature map may be used, like time-domain thermoreflectance, for reduced measurement time and improve throughput. The measured temperature map can then be used as input for the iterative FEM model we employ for extracting a map of the thermal conductivity.
Conclusion

We have introduced method for spatially mapping the thermal conductivity of single and few layer two-dimensional materials using a combination of Raman spectroscopy and finite element calculations. We applied the method to obtain the thermal conductivity of a pristine and He-ion patterned suspended graphene membrane. For the unpatterned membrane, large variations of the extracted thermal conductivity are observed, and attributed to local irregularities such as contamination, defects, or folds. These findings highlight the importance of spatial mapping of the thermal conductivity, in contrast to measurement approaches that yield a thermal conductivity averaged across the entire sample. On the patterned membrane, we demonstrate controlled engineering of the thermal conductivity by He-ion irradiation. As Raman spectroscopy is widely used in the two-dimensional materials community, our method is ideally suited for studying the thermal properties of other layered materials. Moreover, the working principle of the FEM method can easily be extended to more complex geometries, or interfaces with alternative measurement techniques for providing a temperature map of the material of interest upon heating.
Experimental

Preparation of the SiN membrane

Two types of Si/Si₃N₄-membranes were used. First, commercially available Si/Si₃N₄-membranes (Norcada Inc., Edmonton, Canada, NORCADA Low Stress SiNx Membrane NX5200D) were patterned with arrays of holes of various diameters using Gallium-FIB (Strata, FEI), see supporting information. Second, silicon nitrite frames are fabricated using dry and wet etch processes as described elsewhere. Further a Ti/Au (5/40 nm) layer is deposited using an electron beam evaporator.

Synthesis and Transfer of Graphene

The single-layer graphene samples were synthesized using a Cu metal catalyst by chemical vapor deposition (CVD). A 25 µm thick Cu-foil was cleaned with acetic acid for 20 min and rinsed with DI-water and ethanol. The Cu-foil was then heated up to 1000 °C inside a quartz tube under Ar atmosphere for 3 h, and the graphene was then grown with flowing gas mixtures of Ar:H₂:CH₄ = 200:20:0.1 (sccm) for 60 min. After synthesizing the graphene, the polymethyl-methacrylate (PMMA) was coated on the graphene at 3000 RPM for 30 s. Using reactive ion etching (Ar/O₂-Plasma, 60 s) the graphene on the back-side of the PMMA/graphene/metal catalyst was removed. The metal catalyst was then etched by floating on a 0.1 M ammonium persulfate solution overnight. After rinsing the PMMA/graphene with DI-water, the PMMA/graphene was transferred onto the target substrate and baked at 110 °C for 30 min with an intermediate step at 80 °C for 10 min, increasing the adhesion between the graphene and target substrate. The PMMA was removed with acetone, IPA followed by DI-water. Besides graphene grown by us, also commercially available graphene (Easy Transfer, Graphenea and graphene grown and transferred by Applied Nanolayers) was used.
Raman setup and Spectra analysis

Raman spectra were acquired with a WITec Alpha 300 R confocal Raman microscope in backscattering geometry, equipped with 100x (NA=0.9) and 50x (NA=0.55, long working distance) objective lenses. The backscattered light was coupled to a 300 mm lens-based spectrometer with gratings of 600 g/mm or 1800 g/mm equipped with a thermoelectrically cooled CCD. The excitation laser with a wavelength of 532 nm from a diode laser was used for all Raman measurements. The laser power was set using WITec TruePower. A calibration curve of measured laserpowers at different positions in the setup can be found in the supporting information. The 2D-peak properties were extracted from the full-spectrum mapping results by fitting a single Lorentz after linear background subtraction.

Temperature calibration

Measurements were carried out under ambient conditions by placing the sample on a small hotplate. The sample was fixed using a screw to avoid sliding around and ensure good thermal anchoring. Before the measurements, a thermalization time of 45 min has waited.

Laser power absorption

Measurements were performed under ambient conditions by placing a powermeter (Thorlabs, PM100D, equipped with a photodiode) below the sample. See supporting information for details.

FEM calculations

The FEM calculations were performed using Comsol Multiphysics 5.5. The system was remeshed for each laser spot position with an ‘extremely fine’ mesh. The Gaussian laser spot size was cut-off at a radius of 2.5 µm. To improve convergence, the thermal conductivity map is adjusted as follows.
\[ \Delta \kappa = \text{sign}(T_{\text{calc}} - T_{\text{exp}}) \cdot 10^{\beta \log_{10}(|T_{\text{calc}} - T_{\text{exp}}|)} \]  

(5)

All values outside the mask are set to zero.

He\(^{+}\)-ion irradiation

For the irradiation of freestanding graphene membranes, we used a He ion microscope (Orion, Zeiss) equipped with a pattern generator (Elphy MultiBeam, Raith) operated at 30 keV using a probe current of \(\sim 0.5\) pA at a chamber pressure of \(\sim 7 \times 10^{-5}\) mbar. The ion dose was controlled by the exposure dwell time of each pixel ranging from 0.3 ms to 1.5 ms.

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Author contributions statement

O.B., I.S., M.C., and M.P. conceived and designed the experiments. K.T. developed the graphene growth recipe and transfer process. R.F. performed the graphene growth. O.B and I.S. prepared the SiN frame and performed the defect engineering using focused-ion beam. O.B. performed the Raman measurements. O.B., M.P., M.C., and I.Z. did the Raman
spectroscopy analysis. P.B. developed the finite-element model to calculate the temperature distribution for a single laser spot position. M.P. extended the model to construct the temperature map upon illumination by the Raman laser and developed the procedure to fit the thermal conductivity. M.P. performed all finite-element calculations. O.B., M.P., and M.C. wrote the manuscript. All authors discussed the results and implications and commented on the manuscript.

Competing interests

The authors declare no competing financial interests.

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Graphical TOC Entry