Role of Defects and Wrinkles in Thermal Conductivity of Wafer-scale hBN Films

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Abstract

Thermal conductivity of large area hexagonal boron nitride films grown by metal organic vapor phase epitaxy technique is reported. In this study, both defects and wrinkles are present in 2 nm and 10 nm-thick films, while 30 nm-thick films are free from wrinkles. Raman spectroscopy, an optothermal non-contact technique, is employed to probe the temperature and laser power dependence property of the Raman active $E_{2g}^{\text{high}}$ phonon mode; which in turn is used to estimate the rise in the temperature of the films under different laser powers. As the conventional Fourier heat diffusion law cannot be directly employed analytically to estimate the thermal conductivity of these films with defects and wrinkles, finite element modeling is used instead. The effective thermal conductivity is estimated to be 87, 55, and 117 W/m.K for the 2, 10, and 30 nm-thick films, respectively. We also present a quantitative estimation of the thermal resistance by defects and wrinkles individually to the heat flow. The average heat resistance in the film is used to incorporate an overall defect structure and Voronoi cells with contact resistance at the cell-boundaries are constructed to mimic the wrinkled domains. Our
study reveals that the defects/impurities render a much higher resistance to the heat transfer in the films than wrinkles. The thickness of the films also plays a crucial role in the determination of the thermal conductivity of the films.

1. Introduction

With the increasing demand for closely packed miniaturized electronic devices, heat management for 2D materials is becoming critical in next-generation technology. The longevity and performance of devices, such as microchips, processors, and batteries strongly depend on their operating temperature. Exceptionally high thermal conductivity, about 3080-5300 W/m.K, has been measured for graphene. 1,2 The value reduces to 850-2500 W/m.K 3–6 for the graphene layer grown by chemical vapor deposition (CVD). However, the high electrical conductivity of graphene imposes constraints on its use in thermal dissipation applications in devices.7 Monolayer hexagonal boron nitride (hBN) films with relatively high thermal conductivity, 751 W/m.K, 8 and large band gap, 5 eV 9 are potentially ideal as dielectric substrates.10–12 Almost an order of magnitude enhanced carrier mobility is found for graphene devices on hBN than devices on SiO 2.13 The high thermal stability of the hBN films helps to protect air-sensitive metallic surfaces and 2D layered materials.14 Due to the unique combination of high thermal conductivity, large band gap, and notable mechanical properties (which include high flexibility and stretchability),15 thin hBN layers are extraordinary materials for a wide range of applications, from nano-electronics to the space industry.13,16,17

There have been a few studies, exploring the thermal conductivity of hBN layers.8,18 However, these studies are on small-sized exfoliated/CVD grown films, 8,18 which find limitations in practical applications. It has been reported that by using metalorganic vapor phase epitaxy (MOVPE), wafer-scale hBN films can be grown.19–21 However, the unavoidable formation of defects, grains, grain boundaries, impurities, and wrinkles in these
large-scale films$^{19-22}$ can affect their thermal conductivity. A comparative study of the resistance caused by defects/impurities and wrinkles in the heat transfer in these wafer-scale hBN films is yet to be reported in the literature.

The objective of our study is to find the effective thermal conductivity ($\kappa_{\text{eff}}$) of wafer-scale hBN films with defects/impurities and wrinkles. We further aim to find the relative resistance rendered by defects/impurities and wrinkles individually to the heat flow in these films. We employ Raman spectroscopy, a non-contact technique, to measure the temperature in the film pumped by a laser with a Gaussian beam spot. Owing to the inhomogeneity in the film, we estimate the $\kappa_{\text{eff}}$ of the films by exploiting both experimentally measured temperature of the film for known laser power, and finite element modeling (FEM) for the heat transfer in the films. One of the main advantages of the FEM is that it studies the system at a macroscopic scale which is more suitable than molecular dynamics simulation (MD) or density functional theory (DFT) to investigate inhomogeneous 2D layers. Though one obtains the material’s response at an atomic scale by MD/DFT approach, these methodologies demand a high computational cost to model a macroscopic inhomogeneous material, such as 2D films with defects, impurities, and wrinkles. FEM considers the distribution of these structural parameters over the entire area of the film to explore the effective thermal properties of the 2D layers. Our study reveals the relative importance of defects and wrinkles toward heat flow in similar 2D materials.

2. Experimental Details

The hBN films were grown on commercially available 2” diameter sapphire substrates by a closed couple shower head type MOVPE reactor (AIXTRON). Growth was carried out at a temperature of 1350°C using triethylboron and ammonia as the precursors. The details of the growth process are available in Ref. [20]. In this work, we used hBN films of thicknesses 2,
10, and 30 nm. The 30 nm films could be delaminated from the sapphire substrate spontaneously by bringing it in contact with de-ionized water. For the 2 and 10 nm-thick hBN films, spin-coating with PMMA followed by immersion in a 2% hydrofluoric acid bath was used for film transfer. Acetone was used to remove PMMA residue. The delaminated films were then rinsed and transferred onto (i) SiO$_2$ (2 µm) coated, and (ii) gold (200 nm)/chromium (10 nm) coated prefabricated (using photo- and electron beam lithography) circular microwells of diameter 3 µm with a well-defined boundary.

A schematic diagram of the experimental arrangement is shown in Figure 1. The optical image of one of the circular microwells with trenches, used in the experiment, is shown in the inset of Figure 1. During heating, the trenches allowed the trapped air in the hBN-covered microwell to escape rather than resulting in volume expansion. While the former set (films suspended on SiO$_2$/Si microwells) was used to study the temperature coefficient of the Raman shift of the $E_{2g}^{\text{high}}$ mode in these films, the latter set (films

Figure 1. Schematic diagram of the experimental arrangement to measure the thermal conductivity of the films (not to the scale). Inset: Optical image of the fabricated microwells with trenches.
suspended on Au-Cr/Si microwells) was used to estimate the Raman shift of the same mode by laser radiation of known power. We would like to mention that the films were also suspended on microwells of larger diameters (8, 15, and 20 μm). However, the sagging of the films over microwells of these larger diameters constrained us to use only the microwells of smaller diameters in further measurements. Atomic force microscopy (AFM) (Multimode 8, Bruker) under tapping mode operation was used for studying the surface topography of the films.

Raman measurements were performed with LabRAM HR Evolution (Horiba) system which included a confocal microscope with a 100× objective lens. The microscope objective served for both focusing the incident laser light and collecting the backscattered signal. 532 nm emission line of a diode laser was used as the excitation wavelength. A charge-coupled device (CCD) was used to record the spectrum. The laser power on the sample was varied using neutral density filters in the path of the incident laser beam. Temperature-dependent Raman spectra of hBN were measured by placing the sample inside a cooling-heating microscope sample stage (Model: THMS600, Make: Linkam Inc. UK) and using a 50×L objective lens.

3. Results and Discussion

3.1. Surface topography of the hBN films

Figure 2 presents the surface topography of the 2, 10, and 30 nm-thick hBN films transferred onto SiO₂/Si substrate. The 2 and 10 nm films exhibit wrinkles. During cool down after growth, the film experienced a compressive strain which led to wrinkling to release the energy due to the difference in lattice thermal expansion coefficient between film and substrate.²⁰,²³ The average wrinkle wavelengths for 2 and 10 nm films are estimated to be 134 and 240 nm, respectively. The delaminated 30 nm film appears to be free of wrinkles. The detailed morphology of the films has been discussed earlier in Ref. [23,24].
Figure 2. AFM images showing the surface topography of (a) 2 nm, (b) 10 nm, and (c) 30 nm-thick hBN films transferred on SiO$_2$/Si substrates.

3.2 Variation of Raman shift with temperature rise due to applied laser power

Over the last couple of decades, Raman spectroscopy has emerged as a precise contactless technique to measure the thermal conductivity of 2D materials.$^{25}$ The focussed laser beam, which is used as the excitation source in Raman measurements, locally generates heat. By quantifying the intensity ratio of the Stokes ($I_S$) and anti-Stokes ($I_{AS}$) modes from Raman measurements, one can deduce the temperature rise in the sample due to absorbed laser power.$^{26}$ From this measured parameter, one can estimate the thermal conductivity of the sample under investigation. Unfortunately, the intensity of the in-plane phonon mode of hBN at $\sim$1367 cm$^{-1}$ is very weak under non-resonant Raman spectroscopy, and therefore, it is non-trivial to record anti-Stokes Raman signals of hBN. Thus, the ratio $I_S/I_{AS}$ could not be used to estimate the temperature of the thin hBN films for the available laser powers.
Figure 3. (a)-(c) Raman spectra of $E_{2g}^{\text{high}}$ mode of 2, 10, and 30 nm-thick films suspended over SiO$_2$/Si microwells at different heating temperatures. (d) Relative change in Raman shift ($\Delta\omega$) of the $E_{2g}^{\text{high}}$ mode (from its room temperature value) as a function of change in temperature ($\Delta T$) and the corresponding linear fit to the data points. Red filled triangles, green circles and black squares correspond to the data points for the 2, 10, and 30 nm-thick films on SiO$_2$/Si substrate, respectively. Gold open triangles are for the 2 nm film on Au-Cr/Si microwells. The top axis labels the set values of the temperature.

We followed an indirect approach to estimate the temperature rise in the films under laser radiation. First, we calibrated the Raman shift of the given hBN films for various applied temperatures keeping laser power fixed at 0.20 mW. It is to be noted that the laser power was kept very low to record the spectra, so that, it resulted in negligible additional heating of the samples. To find the temperature coefficient of Raman shift, temperature-dependent Raman spectra of three wafer samples suspended over SiO$_2$/Si microwells were recorded. Figure 3 (a)-(c) display the temperature dependent Raman spectra of 2, 10, and 30 nm hBN films, respectively, suspended over SiO$_2$/Si microwells. Each spectrum was fitted with a Lorentzian function, with the intensity, width, and peak position as the free fitting parameters. The relative Raman shifts ($\Delta\omega=\omega(T)-\omega(297 \text{ K})$, where $\omega(T)$ and $\omega (297 \text{ K})$ are the measured Raman shift at temperature $T$ and at 297 K, respectively) with temperature for
the three different samples are summarized in Figure 3(d), showing a clear downward shift with increasing temperature for all samples.

Various factors are responsible for the evolution of the Raman shift of the suspended 2D films with temperature. For large scale hBN films, the dominant roles are played by the thermal contraction of the lattice and the increase in anharmonicity with the rise in temperature. Additionally, the contact regions of films with the side wall of the microwells could give rise to strain due to thermal expansion coefficient (TEC) mismatch. Figure 3(d) also includes the variation of $\Delta \omega$ with temperature for the 2 nm film suspended on Au-Cr/Si microwell. In this case, it is expected that the TEC mismatch between hBN and the substrate is more for Au-Cr/Si than for SiO_{2}/Si.\textsuperscript{27} In spite of this, the data points for both sets of samples (open and closed triangles) nearly overlap as shown in Figure 3(d). It suggests that the contact regions of the film with the substrate play an insignificant role on $\Delta \omega$. Details of the thermal effect on the Raman shift of suspended hBN films have already been discussed by us in Ref. \textsuperscript{[24]}. The first order temperature coefficient of Raman shift ($\chi$) could be estimated by assuming a linear relation\textsuperscript{8} i.e., $\Delta \omega = \chi \Delta T + c$, where $c$ is the y-intercept. From the linear fit to the data points, the values of $\chi$ ($=d(\Delta \omega)/d(\Delta T)$) are estimated to be $-0.02\pm0.001$, $-0.010\pm0.002$, and $-0.010\pm0.002$ cm\textsuperscript{-1}/K for the 2, 10, and 30 nm-thick suspended films, respectively.

Next, the incident laser beam was used as the heat source on the suspended films. The beam was focused at the center of the film on the microwell. The rise in temperature due to the laser power was investigated by recording Raman spectra of the films suspended over Au-Cr/Si microwells. The excitation power dependent Raman spectra of the films are shown in Figure 4 (a)-(c). The power level indicated next to each spectrum is the measured (applied) power on the sample. The change in Raman shift due to applied laser powers is shown in Figure 4(d). The Raman mode downshifts for all the cases, which indicates
Figure 4. (a)-(c) Raman $E_{2g}^{high}$ mode of the 2, 10, and 30 nm-thick hBN films suspended over Au-Cr/Si microwells at different laser powers. (d) Change in $E_{2g}^{high}$ mode frequency as a function of laser power.

an increased local temperature with increasing laser power. Here the change in Raman shift due to laser power relates to the capability of thermal conduction of hBN films towards the Au-Cr/Si heat sink. By using the thermal coefficient of Raman shift ($\chi$), we estimate the temperature ($d(\Delta T)+297$) of the film $d(\Delta T) = \frac{d(\Delta \omega)}{dP} \cdot \frac{d(T)}{d(\Delta \omega)} dP$ as a function of applied laser power and plot them in Figure 5 as red triangles, green circles, and black squares for the 2, 10, and 30 nm-thick films, respectively.

3.3 Estimation of the effective thermal conductivity ($\kappa_{eff}$) of the films

For a homogenous film, the temperature distribution $T(\vec{r})$ across the film due to a heat source $Q(\vec{r})$ is governed by the steady state heat diffusion equation

$$\kappa \nabla^2 T(\vec{r}) + Q(\vec{r}) = 0,$$

where $\kappa$ is the thermal conductivity of the material. For a Gaussian laser beam as the isotropic heat source and its incidence at the centre $|\vec{r}_0|$ of the suspended part of the film,

$$Q(\vec{r}) = \frac{P}{\pi \alpha a^2} \exp \left( -\frac{|\vec{r} - \vec{r}_0|^2}{a^2} \right)$$

(2)
Here, \( a \) is the radius of the laser spot and \( P \) is the absorbed laser power. \( d \) is the thickness of the film. Introducing a dimensionless variable \( \tilde{\rho} = \frac{\tilde{r}}{a} \) the solution of Eqn. 1 for a homogeneous film and the isotropic heat source is

\[
T(0) - T(\rho) = \frac{\alpha}{2} \left( \ln \rho + \frac{\gamma}{2} \right),
\]

(3)

with \( \alpha = P/\pi\kappa d \) and \( \gamma \) is the Euler’s constant. \( T(0) \) is the temperature of the film at the center of the beam. In our experiments (for example, as shown in Figure 1), the edge of the suspended film can be considered to be at ambient temperature, i.e. \( T(R/a)=297 \) K, where \( R \) is the radius of the microwell. Note that the parameter \( P \) in Eqn. 2 is not the same as the applied laser power \( P_a \) (in Figure 4), as only part of the incident radiation is absorbed by the film. Furthermore, some of the absorbed power may be lost by air convection and radiation. For a homogeneous film, one can estimate the loss due to these factors analytically.\(^5^8\) By measuring \( T(0) \) (from the Raman shift) with respect to the same at ambient temperature at the edge, one can estimate \( \kappa.\(^2^6\) However, for a film with grain boundaries, defects, impurities, or wrinkles,\(^2^8\) an additional damping term must be included in the LHS of Eqn. 1. Nonetheless, in this way the role of defects/impurities and wrinkles cannot be decoupled. Furthermore, it is non-trivial to estimate the heat loss by the radiation for these inhomogeneous films.

Thus, instead of estimating the thermal conductivity of the film using the above analytical formalism, we determine the same numerically using the finite element approach. This method describes a system at a macroscopic scale and finds approximate solutions of partial differential equations numerically. The solutions are based on subdividing a large system into smaller parts and linearizing the equations within each of the small parts. The heat propagation in hBN films is simulated using the heat transfer module in COMSOL Multiphysics (version 4.4) software package.\(^2^9\) Using this module, temperature and heat across the film can be mapped by considering the characteristics of the films and the given heat generating source.
To mimic the experimental geometry of the suspended film in the simulation, we define a circular boundary of radius $R$ for the film. The hBN film of known thickness and specific heat is subjected to a Gaussian laser beam of radius $a$ at the center. The heat distribution due to the given laser power takes place within the circular boundary, beyond which room temperature is maintained. The model assumes an ideal thermal contact between the film and the region beyond the boundary (since we used Au-Cr/Si as the heat sink in the experiment). In the simulation, we use the values of $R$ and $a$ as 1.5 $\mu$m and 0.5 $\mu$m, respectively, corresponding to experimental values. UV-visible spectroscopy was carried out to find the absorption of radiation at 532 nm by the films. The absorbance values for the 2, 10, and 30 nm films are 0.23%, 1.15%, and 3.4%, respectively. In the simulation, the applied laser power ($P_a$) is scaled by the above absorbance values for the films to obtain $P$. Heat loss to the air, which is the sum of radiative and convective components, is also included as

$$Q_{air} = h(T - T_{amb}) + \varepsilon\sigma(T^4 - T_{amb}^4)$$

where, $h$ and $\varepsilon$ are heat transfer coefficient (3475 W/m$^2$.K) and surface emissivity (0.8) for hBN, respectively, $\sigma$ is the Stefan-Boltzmann constant and $T_{amb}$ is the ambient temperature (297 K).\(^{18}\)

For a specific geometry of the suspended film, the rise in temperature for a given heat flux determines the effective thermal conductivity ($\kappa_{eff}$) of the film. The presence or absence of wrinkles, defects, and impurities govern the heat flow in the film and changes the value of $T(0)$. Thus, the value of $T(0)$ can be used to estimate $\kappa_{eff}$. In other words, a homogenous film with a higher value of $T(0)$ effectively models the film in which heat flow finds higher resistance. It is to be recalled that by Raman measurements in back-scattering geometry we obtain an average value of the temperature over the area of the spot size, as shown by the data
Figure 5. Estimation of effective thermal conductivity of the films. Measured $T_m$ for different applied laser powers for films of different film thicknesses (data points). The solid lines are simulated for $T_m$ with the appropriate values of $\kappa_{\text{eff}}$ that fits the data points for each film thickness. The dashed line plots the expected change of $T_m$ with applied laser power for bulk hBN.

points in Figure 5. Thus, in the simulation, too, we use the mean temperature ($T_m$) within the spot size of the laser beam of radius $a$. Using FEM, we simulate the temperature ($T_m$) of the films of different thicknesses for different values of $\kappa_{\text{eff}}$. The experimental data points could be best matched (solid lines in Figure 5) for the value of $\kappa_{\text{eff}}$ as 87, 55, 117 W/m.K for the 2, 10, and 30 nm films, respectively. The dashed line in Figure 5 plots the variation of $T_m$ with applied laser power for a $\kappa_{\text{eff}}$ value of 420 W/m.K of bulk hBN. The values of $\kappa_{\text{eff}}$ are significantly less in the films compared to bulk hBN due to the presence of defects/impurities and wrinkles. For a homogeneous hBN film, one also expects a reduction in $\kappa_{\text{eff}}$ with increasing film thickness. However, we find that the value of $\kappa_{\text{eff}}$ does not change monotonically with the thickness of these films. As shown in Figure 2, unlike the 2 and 10 nm films, the 30 nm film is free of wrinkles. Nonetheless, defects/impurities are present in all
samples. Furthermore, the density of wrinkled domains is more for the 2 nm film than the 10 nm film (See Figure 2). The non-monotonic trend in $\kappa_{\text{eff}}$ with thickness indicates a complex role of wrinkles and defects/impurities in the films, which we discuss below.

### 3.4 Thermal resistance by defects and impurities in hBN films

In Figure 5 it is interesting to note that the presence of defects/impurities reduces the value of $\kappa_{\text{eff}}$ of the 30 nm film by nearly 70%. A mean thermal resistance ($R_D$) is introduced in the simulation to model the resistance to the heat flow caused by defects in the 30 nm film. The expected thermal conductivity of the homogenous bulk film is considered as 420 W/m.K.\(^{30}\) The film was exposed to a Gaussian laser beam of different laser powers, as above. The contour plot of the simulated temperature $T_m$ for different laser power and $R_D$ is shown in Figure 6(a). The dashed lines are isothermal lines for the measured values of $T_m$ for the 30 nm-thick film at different laser powers. For the value of $R_D = 14$ nK.m\(^2\)/W, the simulated variation of $T_m$ with applied power matches the experimentally obtained results (see diamond symbols).

### 3.5 Thermal resistance due to wrinkles in hBN films

It is to be recalled that in addition to defects/impurities, wrinkles are present in the 2 and 10 nm films. However, the density of wrinkles is significantly different in these two films. To model the 2 and 10 nm films, a discrete set of points (seeds) are randomly distributed across a plane with a predefined dimension (4.1 $\mu$m $\times$ 3.2 $\mu$m). Based on the distributed points, Voronoi cells are constructed (refer to Figure 6 (b) and (c)). The Voronoi cell has the property that any point X in the plane within the cell R(Y) is closer to seed point Y than any other seed point. The average Voronoi cell size is kept close to the measured
Figure 6. (a) Contour plots of $T_m$ for different laser powers and $R_D$ for the 30 nm-thick film. The dashed lines are isothermal plots for the experimental values of $T_m$. The diamond symbols indicate the values of $R_D$ on the isothermal lines for the applied laser powers used in the experiment. Models of wrinkled hBN films for FEM simulation: (b) average domain size of 150 nm, and (c) average domain size of 260 nm. (d) and (e) Contour plots of $T_m$ for different laser powers and $R_W$ for the 2 and 10 nm-thick films, respectively. The dashed lines are isothermal plots for the experimental values of $T_m$. The circles and triangle symbols indicate the values of $R_W$ on the isothermal lines for the applied laser powers used in the experiment. (f)-(h) Surface temperature distribution of the 2, 10, and 30 nm suspended film under laser heating.

There are typically 160 and 100 numbers of domains of 2 nm and 10 nm modeled films.

Quite a few articles in the literature have discussed the role of wrinkles in determining the thermal conductivity of graphene.\textsuperscript{28,31,32} Non-equilibrium molecular dynamics simulation\textsuperscript{31,32} suggests that the strong localization of phonon across the wrinkled domain wall reduces the thermal conductivity of the film. In view of this, in addition to the 14 nK. m$^2$/W value used for $R_D$ to represent the effect of defects/impurities, resistance ($R_w$) as contact elements of domain boundaries is also introduced. In our model, we assume that defects/impurities and
wrinkles render independent resistive processes. As before, a circular boundary of radius $R$ of the modeled film is subjected to the radiation of a Gaussian laser beam of radius $a = 0.5 \mu m$ at the center of the film. Keeping other constraints the same as described above, Figure 6 (d)-(e) present the simulated contour plots of $T_m$ for different applied laser powers and $R_w$ using a false color scale for the 2 and 10 nm films. The isothermal lines in Figure 6 (d) and (e) are shown by dashed curves for the experimentally measured temperatures $T_m$ of the films obtained in Figure 5. The circle and triangle symbols in panels (d) and (e) on the isothermal lines mark the value of $R_w$ for particular values of $P_a$ used in the experiments. The corresponding values of $R_w$ are 0.7 and 3 nK.m$^2$/W, respectively, for 2 and 10 nm films, respectively. Since the room temperature Raman linewidths are nearly the same in both samples, we assume that the level of defects/impurities is also very similar. High-resolution cross-sectional transmission electron microscopy images reveal that in these films, the individual basal planes across the domain wall are maintained. The resistance per unit thickness due to the wrinkled domain boundaries is $\sim 0.35 (0.3)$ nK. m/W for the 10 nm (2 nm)-thick film. In the above analysis, we find that the resistance to the heat flow caused by defects/impurities is much higher than that caused by wrinkles.

Thermal conductivity at the wrinkled domain boundaries strongly depends on the wavelength of phonons. Perturbed phonons at the domain boundaries do not contribute to the total conductivity of the film. The Umklapp scattering via out-of-plane acoustic phonon reduces the thermal conductivity even for homogenous multilayer films. In addition, due to softer phonons (than C-C covalent bonds in graphene), the intrinsic phonon-phonon scattering plays a dominant role in hBN films and drastically reduces their effective thermal conductivity. The effect of the thickness (i.e. the increase in the number of layers) in reducing the thermal conductivity is intrinsic for few layer 2D materials. Fourier law demands that the thermal conductivity to decrease monotonously with the thickness of the
film till it reaches the bulk value. We demonstrate that such monotonic behavior fails for inhomogeneous films with defects/impurities and wrinkles. The present study shows that defects/impurities, wrinkles, and thickness in hBN films can be exploited to modulate the phonon density of states and hence, to use the system efficiently for thermal management and thermoelectric devices.

4. Summary
We carry out a combined experimental and modeling approach to study the effective thermal conductivity of wafer-scale hBN films. Defects/impurities and wrinkles in these large scale wafers reduce the thermal conductivity from their values in homogeneous counterparts drastically. Raman spectroscopy, a non-contact technique, is used for measuring the temperature of the film for different laser powers. The measurements are performed on 2, 10, and 30 nm-thick suspended hBN films. Defects/impurities are present in all three films. Due to the inhomogeneity of the films, instead of solving the Fourier diffusion law analytically, we apply FEM to estimate the effective thermal conductivity of the films by including wrinkles and defects/impurities in the models. To model the wrinkles, Voronoi cells are constructed following the characteristic morphology of the measured wrinkled domains in the 2 and 10 nm films. The combined experimental and simulated observations suggest that the resistance to heat flow caused by defects/impurities is much higher than that caused by wrinkles.

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