Cross-Plane Thermal Conductivity of *h*-BN Thin Films Grown by Pulsed Laser Deposition

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Primary CNF Tools Used: SC4500 Odd-Hour Evaporator, P7 Profilometer, AFM-Veeco Icon

Abstract:

The distinguished properties of *h*-BN, specifically its atomically smooth surface, large critical electric field, and large electronic band gap, make it ideal for thin film microelectronics and as an ultrawide bandgap (UWBG) semiconductor. Owing to weak van der Waals interactions between layers, *h*-BN exhibits a significant degree of anisotropic thermal conductivity (κ). Exfoliation from bulk crystals is not a sustainable method for mass production of *h*-BN due to its low repeatability, low yield, poor control of sample thickness, and limitation to small areas. Thus, it is necessary to investigate the thickness-dependence of $\kappa_\perp$ for thin films grown by a practical growth method, such as pulsed laser deposition (PLD), which enables the production of reliable and large-area *h*-BN films with control of film thickness. We grew *h*-BN using PLD at 750°C and observed a non-monotonic trend of $\kappa_\perp$ as thickness increased from 30 nm to 300 nm, varying from ~ 1.47 to ~ 0.19 W/(mK). We observed a high $\kappa_\perp$ value for *h*-BN at a thickness of 30 nm, providing insights into the $\kappa_\perp$ of PLD-grown films suitable for electronics applications.

Summary of Research:

The continued scaling down of device dimensions [1] and operation in high-voltage or high-frequency regimes aggravates thermal loads, and effective heat dissipation has become one of the most critical challenges in device performance and reliability [2]. Hexagonal boron nitride (*h*-BN) is a two-dimensional (2D) material with excellent properties for the thermal management of next-generation electronics. Existing literature regarding the thermal conductivity (κ) of *h*-BN films typically focuses on the in-plane thermal conductivity ($\kappa_\parallel$) and samples produced by the exfoliation from bulk *h*-BN. The $\kappa_\parallel$ of few-layer *h*-BN nanosheets falls in the range of 100 -360 W/(mK) at room temperature [3].

Owing to weak van der Waals interactions between layers, *h*-BN has a highly anisotropic κ. The only measured data of the cross-plane thermal conductivity ($\kappa_\perp$) of *h*-BN is 0.2 to 8.1 W/(mK) between 7 nm to 585 nm, respectively, for *h*-BN flakes exfoliated at room temperature [4]. Exfoliation from bulk crystals is not a sustainable method for mass production, thus there is a need to investigate the thickness-dependent $\kappa_\perp$ for a scalable growth method, which provides reliable and large-area *h*-BN with control of film thickness.

We utilized pulsed laser deposition (PLD), which offers several benefits compared to other growth methodologies. PLD involves the deposition of thin films from a single polycrystalline target directed upon the substrate, therefore ensuring the preservation of stoichiometry throughout the deposition process. We measured $\kappa_\perp$ of *h*-BN thin films ranging from 30 nm to 300 nm implementing the optical pump-probe method, frequency-domain thermoreflectance (FDTR), depicted in Figure 1a. The samples were modeled as a three-layer system, with each layer defined by the layer thickness, volumetric heat capacity c_p, the thermal boundary conductances G1 and G2, the $\kappa_\perp$ and in-plane thermal conductivity ($\kappa_\parallel$). To maximize the coefficient of thermoreflectance of the 532 nm probe, Au was chosen as the transducer layer and the SC4500 Odd-Hour Evaporator at CNF was utilized to deposit a 100 nm layer of Au (thickness determined by the P7 profilometer).

Two curves are plotted in Figure 2a, assuming lower and higher bounds of $\kappa_\parallel$. Despite the slight increase of $\kappa_\perp$ for the isotropic case and for film thicknesses ≤ 50 nm, the $\kappa_\perp$ of the *h*-BN thin films decrease from 1.47 to 0.19 W/(mK) with an increase in film thickness from 30 nm to 300 nm. Previous experiments observed long phonon mean free path > 100 nm for $\kappa_\perp$ of exfoliated *h*-BN flakes; predictions by first-principles calculations indicated ~ 80% of the heat is transported by phonons of MFPs ranging from 3 nm to 90 nm [4].

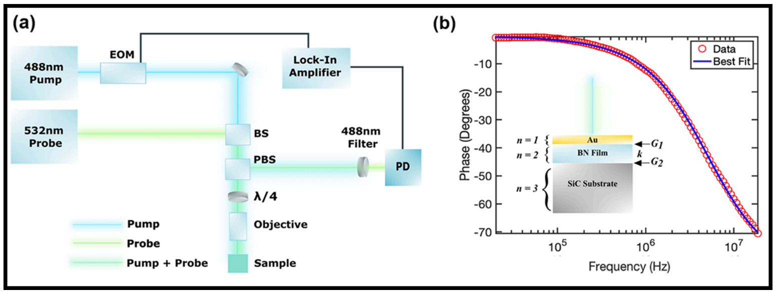

Figure 1: FDTR experimental setup and fitted data. (a) Simplified schematic of the FDTR system. (b) Phase vs. frequency data obtained from FDTR measurements for the 50 nm epitaxial h-*BN layer on bulk SiC substrate.*

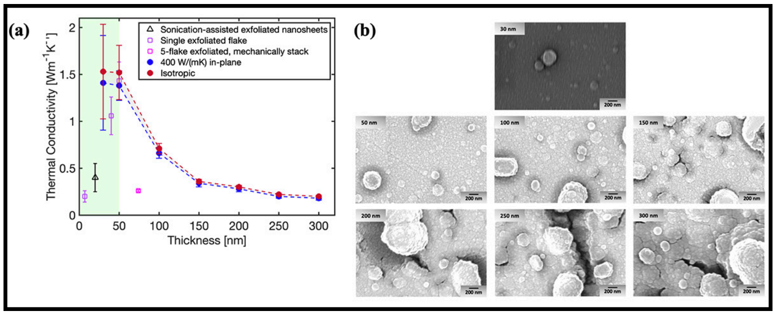

Figure 2: Thickness-dependent thermal conductivity and SEM of h-*BN films. (a) As the thickness of the thin film increases there is a decrease in sample quality, thus, reducing* $\kappa_\perp$ *(b) Crack propagation is evident as the thickness increases leading to an increase in phonon defect scattering.*

If the film quality of PLD-grown h-BN was consistent across the thickness range, we should expect the $\kappa_\perp$ increases, at least up to 100 nm, with thickness due to the size effect. However, our findings indicate the opposite trend, implying that as the thickness of the film increases, there is a significant decrease in the film quality.

To investigate the quality of the thin films, a commercial AFM system was utilized. An increase in root-mean-square (RMS) from 3.30 nm to 17.40 nm for the *h*-BN films with a thickness from 30 nm to 300 nm was determined. The increase in RMS presents increasing surface roughness, which introduces stronger phonon scattering, thereby reducing $\kappa_\perp$.

Lastly, the Zeiss Gemini 500 Scanning Electron Microscope (SEM) was utilized to capture high-resolution images of the sample surfaces, as shown in Figure 2b. As thickness increases, voids/cracks start to form and are evident in the thicker *h*-BN films, which leads to strong phonon-defect scattering, thereby reducing $\kappa_\perp$.

In conclusion, among two-dimensional materials (2D), hexagonal boron nitride (*h*-BN) stands out as a highly promising candidate for use in both micro and nanoscale devices, given its exceptional mechanical, electrical, and thermal properties. The PLD-grown *h*-BN at 30 nm showed a larger $\kappa_\perp$ value than that of the single exfoliated flake at 7 nm and 40 nm [4], and a larger value that of the sonicated-assisted exfoliated nanosheets at 20 nm [5]. These thinner samples provide the high $\kappa_\perp$, suitable for integrated circuits and high-power electrics, which are in need of heat dissipation while maintaining a compact and lightweight design.

References:

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