

Thermal Transport in a 2D Amorphous Material

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Two-dimensional (2D) crystals proved revolutionary soon after graphene was discovered in 2004. However, 2D amorphous materials down to a single layer of atoms only became accessible in 2020, and they remain largely unexplored. In particular, the thermophysical properties of amorphous materials are of great interest upon transition from 3D to 2D. Here, we probe and simulate thermal transport in monolayer amorphous carbon (MAC). An ultralow cross-plane thermal conductivity (κ) is measured for van der Waals stacked multilayers, which is comparable to that of randomly stacked graphene despite the extra disorder in MAC. This result reveals the predominant role of the weak interlayer interactions in 2D materials. Meanwhile, an unexpectedly high in-plane κ is obtained for freestanding monolayers, which is a few times higher than what is predicted by conventional wisdom for 3D amorphous carbon with a similar sp^2 fraction. This observation is primarily attributed to the dimensionality-induced reduction of anharmonicity and the unique low-frequency out-of-plane vibrational modes in MAC. Amorphous materials at the 2D limit open up new avenues for understanding and manipulating heat at the atomic scale.

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I. INTRODUCTION

Amorphous materials such as glass have captured human imagination for thousands of years and continue to be of immense value for diverse technologies including advanced manufacturing, high-performance electronics, and thermal

barrier coatings, in light of their intriguing structures and fascinating properties [1,2]. In particular, recent years have witnessed substantial progress in terms of fabrication, characterization, and simulation. For example, the atomic structures of glassy solids (e.g., silica and high-entropy alloy) have been directly observed [3–5] ever since Zachariasen [6] proposed the continuous-random-network (CRN) model in 1932. Further, extreme mechanical and dielectric properties have been realized in amorphous carbon and boron nitride, respectively [7,8]. Moreover, *ab initio* and machine-learning-assisted calculations have markedly facilitated the mechanistic understandings of amorphous materials [9–11]. Among all the endeavors, the synthesis of two-dimensional (2D) amorphous materials [12] is noteworthy, especially amorphous carbon down to a single atomic layer, which offers unprecedented

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opportunities for achieving deeper insights, unusual properties, and novel applications [13–16]. However, apart from scarce structural, mechanical, and electrical characterizations, there remains a compelling need to explore thermal transport in amorphous materials at the 2D limit [17,18].

Heat flow in nonmetallic crystals is mainly carried by phonons, which can lead to ultrahigh thermal conductivities over $2000 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature (RT), such as in diamond [19,20], owing to the large group velocities and long lifetimes. In contrast, glasses are usually characterized by a low thermal conductivity (κ) around $1 \text{ W m}^{-1} \text{ K}^{-1}$, as limited by the random walk of vibrational energy among disordered atoms [21,22]. However, notable exceptions exist. For example, an unusually high κ_{RT} of over $20 \text{ W m}^{-1} \text{ K}^{-1}$ has recently been realized in 3D amorphous carbon with around 95% sp^3 bonding [8], which confirms a conventional wisdom that predicts higher κ for materials with more sp^3 and less sp^2 bonding [23,24]. Following this line of thought, the in-plane thermal conductivity of 2D amorphous carbon should reach a minimum due to its 100% sp^2 bonding. Furthermore, an ultralow cross-plane κ_{RT} down to around $0.04 \text{ W m}^{-1} \text{ K}^{-1}$ has recently been achieved in van der Waals (vdW) stacked 2D crystals of transition metal dichalcogenides (TMDs) with random interlayer orientations [25], which motivates us to wonder whether the additional intralayer disorder can lead to even lower cross-plane κ in stacked 2D amorphous carbon.

In this work, by systematically measuring and simulating thermal transport in 2D amorphous carbon, we attempt to address one important scientific question: How would thermal transport in amorphous materials vary upon transition from 3D to 2D? First, multiple monolayers are assembled into a set of vdW stacks for cross-plane thermal measurement using the laser pump-probe technique of frequency-domain thermoreflectance. Subsequently, for in-plane transport, the microbridge method is employed with the monolayer amorphous carbon (MAC) samples suspended across pairs of custom-fabricated heating and sensing micro-islands. We observe a cross-plane $\kappa_{\perp}$ down to $0.08 \pm 0.01 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature, which is one of the lowest values reported to date, and a remarkable in-plane $\kappa_{\parallel}$ up to $5.47 \pm 0.32 \text{ W m}^{-1} \text{ K}^{-1}$, which is a few times higher than the conventional prediction for 3D amorphous carbon. In order to understand these unusual observations, we then perform systematic molecular dynamics and lattice dynamics simulations, which highlight the role of disorder and dimensionality in both directions.

II. RESULTS AND DISCUSSION

A. Basic characteristics of MAC

Our 2D amorphous carbon samples were grown on fresh-cleaved mica via atmospheric pressure chemical vapor deposition (see Sec. IV). For characterizations, the monolayers were delaminated from the growth surface

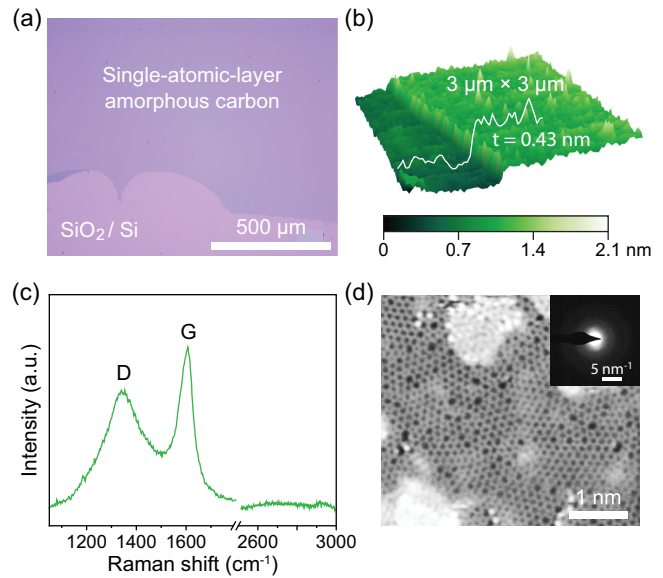


FIG. 1. Microscopic and spectroscopic characterizations of 2D amorphous carbon. (a) Optical image of single-atomic-layer amorphous carbon transferred onto a SiO_2/Si substrate. (b) Atomic force microscopy mapping of an as-grown monolayer amorphous carbon sample across the edge. The line profile shows the typical thickness. (c) Representative Raman spectrum and (d) scanning transmission electron microscopy image of monolayer amorphous carbon. The inset of panel (d) shows an electron diffraction image of a selected area.

using water intercalation and then transferred to a target substrate. In Fig. 1(a), we show a representative optical image of a continuous MAC film on a silica-coated silicon (SiO_2/Si) substrate. Using atomic force microscopy (AFM), the intrinsic monolayer thickness was determined to be about 0.4 nm directly on as-grown samples [Fig. 1(b)], which agrees well with an earlier report [15]. Values of around 0.6 nm were also observed—both by others [14] and in our study (Fig. S1 in the Supplemental Material [26]), likely due to sample handling and substrate conditions. The slightly larger thickness compared to graphene (also a single layer of carbon atoms) was previously attributed to the out-of-plane buckling due to Stone-Wales defects [14–16]. Further, Raman spectroscopy confirmed the absence of the 2D peak and yielded broad D and G peaks with an intensity ratio of around 0.8 [Fig. 1(c)], indicating an amorphous carbon network [14,15]. This finding was subsequently corroborated by selected area electron diffraction [Fig. 1(d), inset]. More directly, representative MAC samples were imaged with scanning transmission electron microscopy (STEM), which verified the disordered atomic structure containing randomly distributed pentagons, hexagons, heptagons, and octagons [Fig. 1(d)].

B. Cross-plane thermal transport

To probe cross-plane thermal transport, monolayer amorphous carbon was first assembled layer by layer into

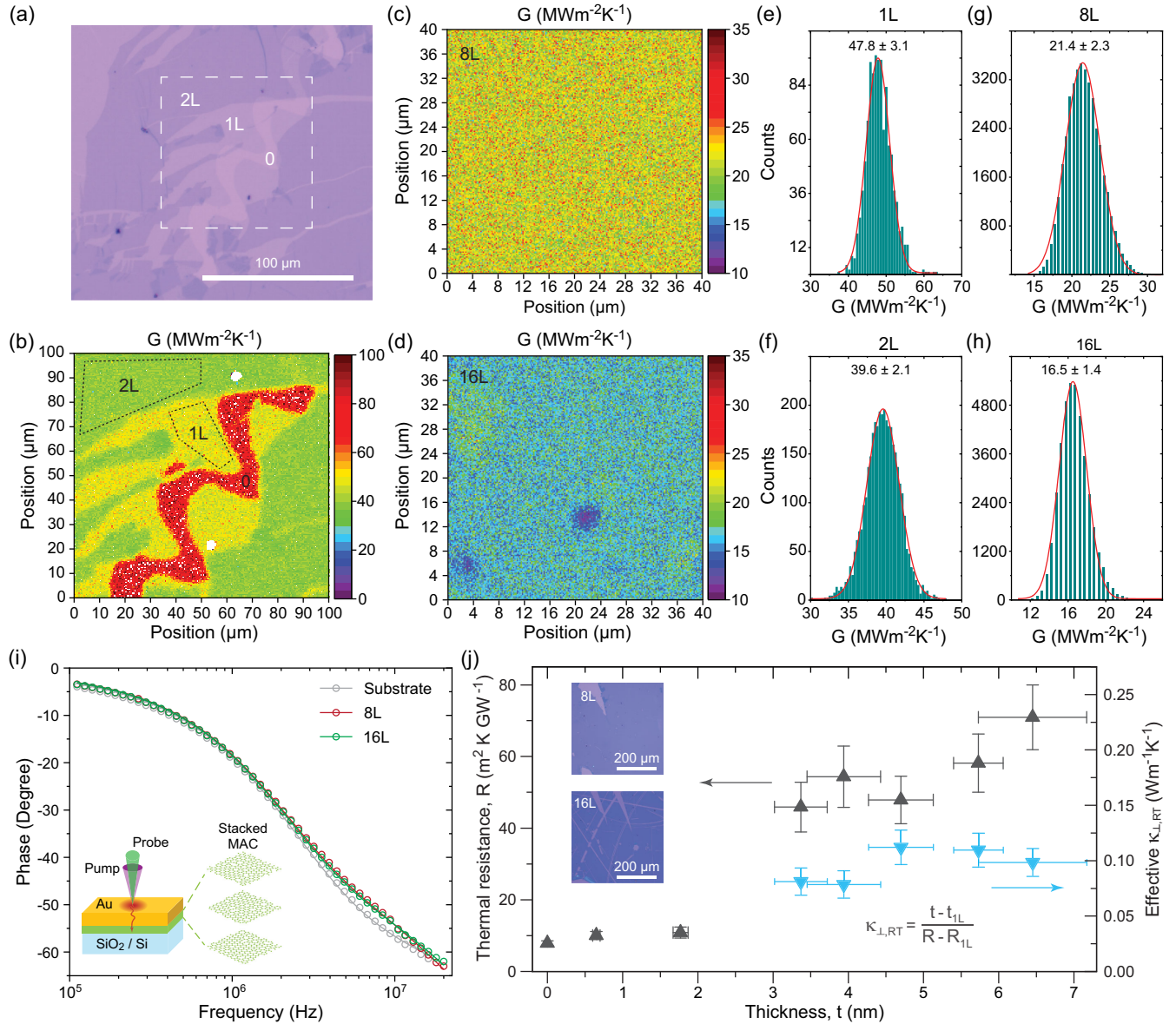


FIG. 2. Cross-plane thermal transport measurement in stacked 2D amorphous carbon. (a) Optical image showing 1L and 2L regions on a SiO₂/Si substrate (0). (b) FDTR mapping of the interfacial thermal conductance G within the white square in panel (a). (c), (d) G map of 8L and 16L MAC, respectively. (e), (f) Histograms of G for the 1L and 2L areas, respectively, marked in panel (b). (g), (h) Histograms of G for 8L and 16L MAC, respectively, obtained from panels (c) and (d). (i) FDTR signals (circles) and fittings (lines) as a function of modulation frequency measured on 8L and 16L samples and also the bare substrate. The inset shows a schematic of the experiment. (j) Thermal resistance of stacked MAC as a function of thickness for 1L, 2L, 6L, 8L, 12L, 14L, and 16L samples. Data for the substrate are also included. The effective cross-plane thermal conductivity ($\kappa_{L,RT}$) for thick samples ($\geq 6L$) is also plotted, with $t_{1L} = 0.46$ nm. The error bars include both the experimental noise and uncertainties from various input parameters for the fitting. The inset shows optical images of 8L and 16L samples.

a vdW stack on a SiO₂/Si substrate [26]. Subsequently, frequency-domain thermoreflectance (FDTR) was employed to measure the thermal conductance of the stacked MAC samples (Sec. IV), which required the evaporation of a gold (Au) nanofilm as an opto-thermal transducer [Fig. 2(i)]. Here, the sample conductance is obtained as part of the total interfacial thermal conductance (G) between the Au film and

the substrate [25,27]. First, a two-layer (2L) sample [Fig. 2(a)] was prepared by a polymer-free approach to minimize contamination. The white box marks a 100 μm × 100 μm area selected for FDTR mapping of G . The regions labeled 1L and 2L contain monolayers and bilayers, respectively [Figs. 2(a) and 2(b)]. The average G values of around 47.8 and 39.6 MW m⁻² K⁻¹ were extracted by fitting the

histograms in Figs. 2(e) and 2(f) to normal distributions, compared to around $100 \text{ MW m}^{-2} \text{ K}^{-1}$ for the bare substrate without MAC.

Subsequently, a set of eight samples including a bare substrate and stacked MAC of 1L, 2L, 6L, 8L, 12L, 14L, and 16L were prepared for further study. Because of challenges in preparing thick stacks with the polymer-free method, we switched to a polymer-assisted approach that involved mechanical contact with only the top layer to keep the internal interfaces clean (Fig. S2). In Figs. 2(c) and 2(d), we plot the G maps for the 8L and 16L samples, respectively. The corresponding histograms are shown in Figs. 2(g) and 2(h), which yield average G values around 21.4 and $16.5 \text{ MW m}^{-2} \text{ K}^{-1}$ comparable to that of multi-layer graphene [27]. In addition, we also performed single-spot measurements at random locations within the scanned area. Figure 2(i) shows the FDTR signals for three different samples, which yield G values consistent with those from G mapping (Table S2 in the Supplemental Material [26]).

To obtain the effective cross-plane thermal conductivity ($\kappa_{\perp}$) of the stacked MAC samples, in Fig. 2(j), we plot the thermal resistance ($R = 1/G$) from single-spot measurement as a function of film thickness (t). Here, t was systematically measured with AFM, yielding an average monolayer thickness of 0.46 nm for the 6L to 16L samples. For the 1L and 2L samples, higher G values were measured after annealing (Sec. IV). For the thicker samples, R increases approximately linearly with t , indicating diffusive thermal transport. By using the 1L sample as a reference, we can subtract the contributions of the Au/MAC and MAC/SiO₂ interfaces from the total interfacial thermal resistance [26]. The effective cross-plane thermal conductivity is then obtained as $\kappa_{\perp} = (t - t_{\text{IL}})/(R - R_{\text{IL}})$ and ranges from about 0.08 to $0.11 \text{ W m}^{-1} \text{ K}^{-1}$ for different samples [Fig. 2(j)]. This conductivity is nearly 2 orders of magnitude lower than the $\kappa_{\perp}$ of graphite [28,29] and many other vdW crystals, and approaches the lowest κ observed among solids [20,25,30,31].

C. In-plane thermal transport

For in-plane thermal transport, we employed the microbridge method (Sec. IV and Supplemental Material [26]) so that freestanding monolayer amorphous carbon could be measured (Fig. 3). To this end, we custom fabricated a series of microdevices consisting of two closely spaced silicon nitride (SiN_x) islands, both of which were suspended by long and narrow SiN_x beams to achieve good thermal isolation from the substrate, and integrated with platinum serpentine resistors for precise heating and temperature sensing [Figs. 3(a) and 3(b)]. To measure the in-plane thermal conductivity, a MAC film was first transferred onto the microdevice and then cut into a well-defined rectangle bridging the heater and sensor islands (Fig. S12), as illustrated in Fig. 3(b). Subsequently, a

sinusoidal electric current (i) at a frequency f was supplied to the heater island so that its temperature could be elevated to T_h and modulated at $2f$. This process generated a heat flow Q at $2f$ through the suspended MAC sample into the sensor island at a lower temperature of T_s , which was measured resistively by applying a direct current (I). Using this modulated heating scheme, temperature variations and heat currents can be resolved with microkelvin and picowatt resolution, respectively [26].

In Figs. 3(c) and 3(d), we show representative SEM images of two microbridge devices together with the suspended MAC samples (MAC-1 and MAC-2). A total of four samples were measured (Fig. S13), which represent three independent MAC films, and a range of lengths ($2\text{--}6 \text{ }\mu\text{m}$) and widths ($7\text{--}21 \text{ }\mu\text{m}$), assuming the same thickness of 0.4 nm [Fig. 1(b), Fig. S1, and Table S3]. The in-plane thermal conductivity ($\kappa_{\parallel}$) of all four samples was obtained in the temperature range from 80 K to 450 K under high vacuum. As shown in Fig. 3(e), $\kappa_{\parallel}$ consistently increases with increasing temperature, which is one of the most prominent features of thermal transport in 3D amorphous materials, as evidenced by the literature data for tetrahedrally bonded amorphous carbon (ta-C) and hydrogenated ta-C (ta-C:H) [32,33]. At room temperature, the $\kappa_{\parallel}$ values of these single-atomic-layer amorphous carbon films are around $4.49\text{--}5.47 \text{ W m}^{-1} \text{ K}^{-1}$, which are over 2 orders of magnitude lower than that of graphene [34] but generally higher than those of 3D amorphous carbon. The small variations (around 20%) of $\kappa_{\parallel}$ across different samples may be attributed to variations in the exact atomic arrangement [18] and imperfections in sample preparation, although a quantitative and microscopic understanding is not yet available. Interestingly, the normalized $\kappa_{\parallel}$ values with respect to κ_{RT} follow a very similar trend [Fig. 3(e), inset]. Together, these observations imply that the in-plane thermal transport in MAC is dominated by the disordered structure.

D. Molecular and lattice dynamics simulations

In addition to experimental measurements, we performed systematic molecular dynamics (MD) simulations and incorporated quantum corrections (Sec. IV and Supplemental Material [26]). Atomic models of monolayer amorphous carbon were constructed using the kinetic Monte Carlo method, which began with randomly distributed carbon atoms in 2D and proceeded to lowering the system energy via probabilistic Stone-Wales transformations (Fig. S17). Comprehensive structural analyses were conducted in terms of the ring size, bond length, bond angle, and the pair correlation function (Fig. S18), yielding results consistent with our STEM observations and previous reports [14,15].

In Fig. 4(a), we plot the cross-plane and in-plane thermal conductivity for stacked MAC and freestanding monolayers, respectively, as a function of temperature. The calculated

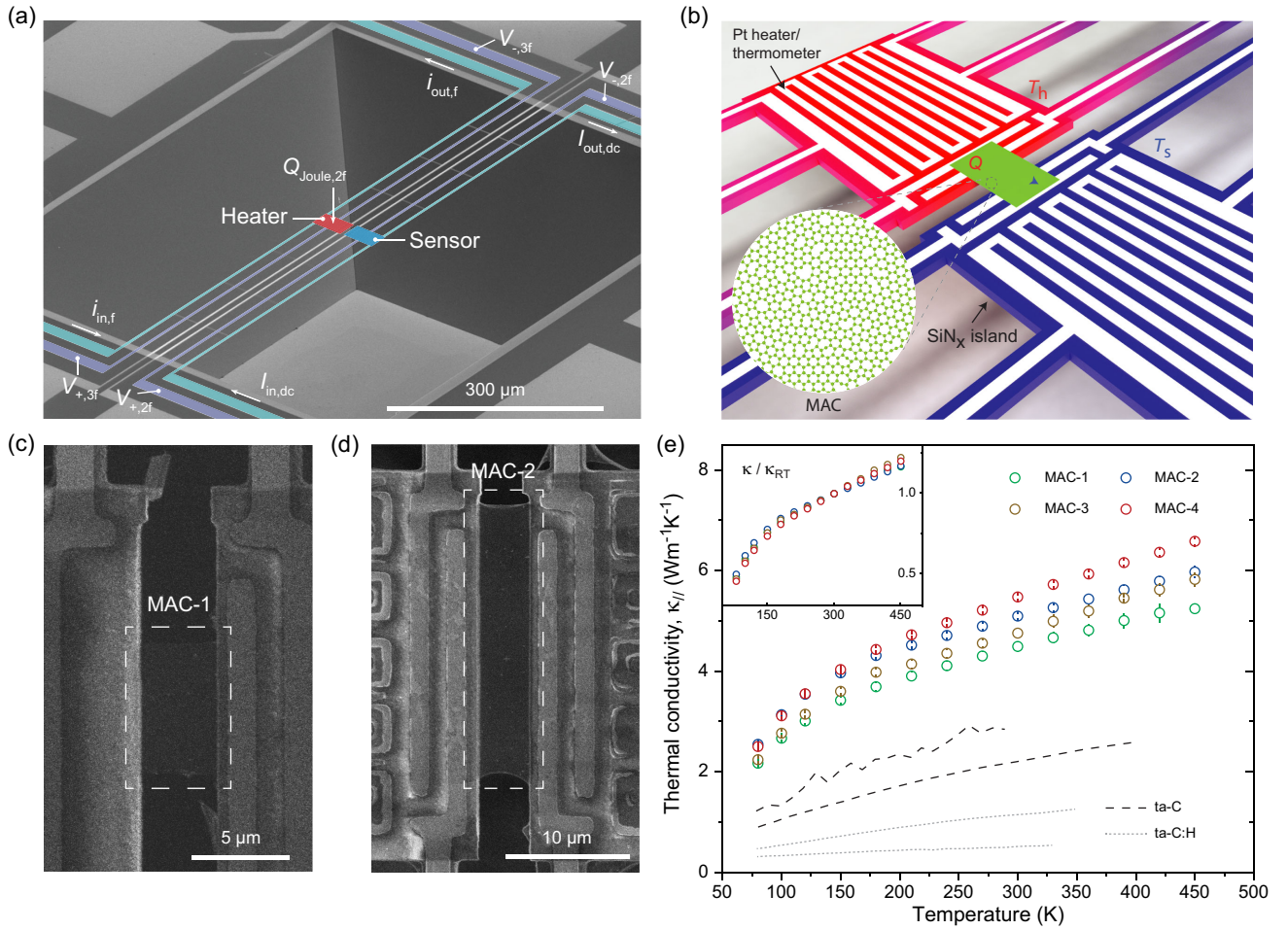


FIG. 3. In-plane thermal transport measurement of freestanding 2D amorphous carbon. (a) False-colored SEM image of a suspended microbridge device with the key electrical and thermal signals labeled. (b) Schematic of a MAC sample bridged on a microbridge device. The inset shows the simulated atomic structure. (c), (d) SEM images of microbridge devices with suspended MAC samples. (e) Measured in-plane thermal conductivities of four MAC samples (1 and 2 are from the same batch) as a function of temperature, together with literature data for thin films of 3D amorphous carbon [32,33]. The inset shows the normalized thermal conductivity of MAC with respect to the room-temperature values. The error bars represent uncertainties in the thermal conductance measurement.

$\kappa_{\perp,RT}$ of stacked MAC is around $0.04 \text{ W m}^{-1} \text{ K}^{-1}$, lower than the experimental values of around $0.08 \text{ W m}^{-1} \text{ K}^{-1}$. The calculated $\kappa_{||,RT}$ increases slightly with model dimensions and ranges from 5.87 to $7.35 \text{ W m}^{-1} \text{ K}^{-1}$. Despite some quantitative discrepancies, there is an overall agreement between the simulations and experiments (Fig. S16). Moreover, upon normalization, all the quantum-corrected and measured $\kappa_{||,RT}$ overlap well in terms of the temperature dependence. These results suggest that our simulations have captured the essential mechanisms of thermal transport in MAC. For comparison, randomly stacked graphene and 3D amorphous carbon dominated by sp^2 bonding (about 94%) were also simulated (Fig. S19). Both feature a thermal conductivity that increases with temperature, as is characteristic of amorphous materials. The calculated $\kappa_{\perp}$ of the former closely follows that of stacked MAC [Fig. 4(a)], while the predicted κ of the latter ($3.20 \text{ W m}^{-1} \text{ K}^{-1}$) is notably lower than the $\kappa_{||}$ of freestanding MAC.

In addition to MD simulations, we further calculated the $\kappa_{||}$ of MAC using lattice dynamics (LD). To this end, both the classical harmonic Allen-Feldman theory [21] and the state-of-the-art quasi-harmonic Green-Kubo (QHGK) [9] approach were employed (Sec. IV), which inherently captured the wavelike tunneling of vibrational energy that is crucial to amorphous systems [10]. A major distinction is that the former only considers the effect of disorder while the latter also incorporates the influence of anharmonicity [9,10,21]. At around 300 K and above, the $\kappa_{||}$ values from these two LD methods agree well with the measured and MD-simulated results [Fig. 4(a)]. However, at lower temperatures, both notably underestimate $\kappa_{||}$. Potential influences of the model size and quantum correction are discussed in Sec. IV.

E. Role of disorder and dimensionality

Carbon forms a rich variety of allotropes and almost limitless compounds [41]. In Figs. 4(a) and 4(b), we

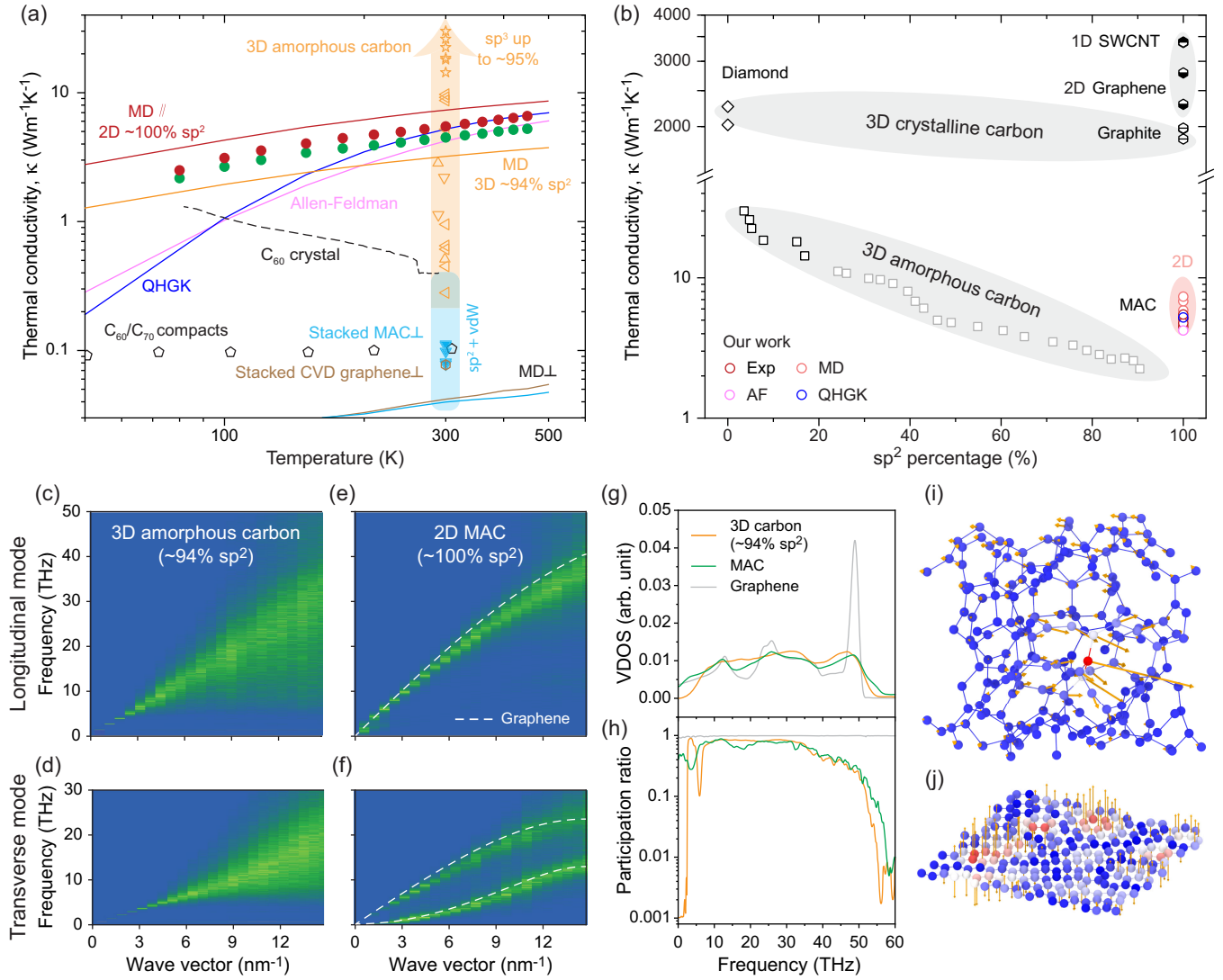


FIG. 4. Atomistic simulations of thermal transport in 2D amorphous carbon. (a) Calculated thermal conductivities of (stacked) MAC plotted against the experimental values, together with literature data for a selection of carbon allotropes with low κ , including fullerene [35,36], 3D amorphous carbon [8,23,32,33], and stacked polycrystalline graphene [27]. The thermal conductivities of 3D amorphous carbon with 94% sp^2 bonding and randomly stacked graphene are also calculated and presented for comparison. (b) Thermal conductivity of crystalline and amorphous carbon allotropes, including diamond [19,37], graphite [28,29,38], graphene [34,39], single-walled carbon nanotube [40], and 3D amorphous carbon [8,24], as a function of sp^2 percentage. (c)–(f) Dispersion relations of the longitudinal and transverse vibrational modes for 3D amorphous carbon and MAC along the Γ -M direction. The phonon dispersion for graphene is calculated using lattice dynamics and plotted for comparison. (g), (h) Vibrational density of states and participation ratio, respectively, for 3D amorphous carbon, MAC, and graphene. (i), (j) Visualization of representative eigenvectors for 3D amorphous carbon (2.34 THz) and MAC (2.39 THz), respectively.

summarize previously reported κ of representative carbon materials from 0D to 3D covering both crystals and glasses. These materials include diamond [19,37], graphite [28,29,38], graphene [34,39], single-walled carbon nanotube (SWCNT) [40], fullerene [35,36], 3D amorphous carbon [8,23,24,32,33], and stacked graphene [27]. Intriguingly, the κ_{RT} values span 5 orders of magnitude from more than 10^3 to around $10^{-2} \text{ W m}^{-1} \text{ K}^{-1}$, which is basically the full range for thermal transport in solids. In

particular, even amorphous carbon alone can have κ_{RT} values varying by a few hundredfold [see the vertical shades at room temperature in Fig. 4(a)]. Such characteristics reflect the great breadth and depth of the underlying physical mechanisms and bring us back to the scientific question that motivated this work, namely, the role of disorder combined with dimensionality.

First, in the cross-plane direction, the ultralow thermal conductivity of stacked MAC originates from the synergistic

contributions of the cross-plane structural disorder and the weak interlayer vdW interaction characteristic of 2D materials, as demonstrated by simulations separating the two factors (Fig. S23). The comparable $\kappa_{\perp,RT}$ of stacked MAC and graphene is counterintuitive considering the extra in-plane disorder of the former; however, an intuitive physical picture can be proposed. Briefly, both stacks feature very small interlayer shear moduli since the cross-plane disorder leads to random, incommensurate interfaces that keep adjacent layers out of registry [42]. This feature substantially reduces the energy barriers for interlayer sliding and underlies the structural superlubricity observed in graphite, which in turn softens the transverse vibrational modes and dramatically suppresses cross-plane thermal transport [25,29,43]. In addition, the longitudinal modes are strongly affected by the cross-plane disorder, which is shared by stacked MAC and graphene. Together, the similarities in both the transverse and longitudinal modes (Fig. S20) explain the similar cross-plane conductivities.

Next, we consider the role of dimensionality for in-plane transport. It is known that dimensionality can impact a plethora of phenomena such as wave localization and hydrodynamic flow [44]. In addition, substantial enhancement of thermal conductivity with reducing dimensionality has been reported [45,46]. In Fig. 4(b), we illustrate dimensional crossover in the κ of both crystalline and amorphous carbon as the atomic coordination transforms from pure sp^3 to sp^2 . Notably, we observe, for the first time, that the $\kappa_{\parallel}$ values of 2D MAC (around 100% sp^2) are discernibly higher than those of 3D amorphous carbon with a similarly high percentage of sp^2 bonding [Fig. 4(b)]. This finding is especially unusual considering the traditional expectation that for 3D amorphous carbon, more sp^2 means lower thermal conductivity, which has been attributed to smaller volumetric heat capacity and stronger scattering [8,24,47,48].

To gain deeper physical insights, we calculated the dispersion relations for the vibrational modes in both 3D amorphous carbon and MAC [Figs. 4(c)–4(f) and Fig. S21] [22]. For 3D amorphous carbon with 94% sp^2 bonding, the dispersions for both the longitudinal and transverse modes are almost entirely diffuse [Figs. 4(c) and 4(d)]. This broadening computed from MD arises from both disorder and anharmonicity, which enhances scattering and impedes thermal transport [18]. In comparison, despite the similar disorder and sp^2 bonding, the dispersions for MAC clearly feature well-defined branches similar to those of graphene even at relatively high frequencies, for example, up to around 20 THz for the longitudinal modes [Figs. 4(e) and 4(f)]. This feature suggests that low-frequency propagating waves contribute dominantly to the unusual high $\kappa_{\parallel}$ of MAC. Additionally, the similar $\kappa_{\parallel}$ values from Allen-Feldman and QHGK calculations [Fig. 4(a)] also indicate that the effect of anharmonicity is relatively weak in MAC at around room temperature. The crucial role of the low-frequency

modes in MAC is further corroborated by their notably higher vibrational density of states (VDOS) and participation ratio, as shown, respectively, in Figs. 4(g) and 4(h). Ultimately, these distinct characteristics can be traced back to the 2D nature of MAC, which significantly promotes out-of-plane vibrations (Fig. S24). As an example, we compare representative eigenvectors for 3D amorphous carbon and MAC as visualized in Figs. 4(i) and 4(j), respectively. For the former, a localized mode at 2.34 THz is identified, as characterized by an overall random distribution of the vibrational directions and only a few atoms with large amplitudes. In contrast, a propagating mode at 2.39 THz is shown for MAC, as manifested by the consistent out-of-plane atomic vibrations together with more evenly distributed amplitudes.

III. CONCLUSIONS

In summary, we have grown single-atomic-layer 2D amorphous carbon films and revealed thermal transport properties distinct from 3D amorphous carbon, both along the in-plane and cross-plane directions. The cross-plane thermal conductivity is expectedly ultralow but surprisingly similar to that of randomly stacked graphene despite the extra disorder in MAC. Meanwhile, the in-plane thermal conductivity turns out to be unexpectedly high, which goes against the conventional wisdom for 3D amorphous carbon. These remarkable observations arising from the unique atomic structures and bonding anisotropy in (stacked) MAC highlight 2D amorphous materials as a rich yet uncharted territory for pushing the limit of thermal transport. As an example, a recent theoretical study of MAC has explored the consequences of hyperuniformity [18]. In the future, it would be of interest to experimentally correlate thermal transport with detailed topological and structural characteristics, and to look into the possibility of directly observing the impact of localization. Finally, we expect the unusual thermal properties of 2D MAC combined with its chemical stability, mechanical strength, and electrical tunability to be uniquely beneficial to various thermal management and energy harvesting applications.

IV. METHODS

A. Growth of 2D amorphous carbon

Two-dimensional (2D) amorphous carbon films were grown in a dual-zone horizontal tube furnace using atmospheric pressure chemical vapor deposition with aromatic compound precursors. Centimeter-sized pieces of fresh-cleaved fluorophlogopite mica were used as the substrates. Prior to film growth, the tube was evacuated and purged with argon (Ar) gas to eliminate residual air. Throughout the growth process, the quartz tube was immersed in a flow of mixed Ar and hydrogen (H_2 , 9.1 vol%). The substrates were heated up to 800 °C together with the precursors. A 30-minute growth time was used. After growth, the quartz

tube was taken out of the furnace and rapidly cooled to room temperature.

B. Microscopic characterizations

Systematic microscopic characterizations of amorphous carbon films were performed using a variety of techniques, with representative data presented in Figs. 1–3 and Figs. S1, S3, S12, and S13. The optical images were taken with Olympus BX53. The thickness of MAC was measured using AFM (MFP-3D Infinity from Asylum Research and Multimode 8 from Bruker). The Raman spectra of MAC were measured using LabRAM HR Evolution, HORIBA Scientific. The scanning electron microscopy (SEM) images were obtained by Helios G4 UX, while selected area electron diffraction was performed with Talos F200X, both from Thermo Fisher Scientific. The STEM image was acquired with NION HERMES-100.

C. Frequency-domain thermoreflectance

The cross-plane thermal transport was measured by the widely used technique of FDTR. Our FDTR system is based on two continuous-wave (CW) lasers [19,49–51]. Briefly, a 405-nm pump beam was modulated to periodically heat the sample surface, and a 532-nm laser probed the surface temperature change at the modulation frequency based on variation of the reflectance. The effective laser spot size (root-mean-square average of the pump and probe beam radii) was 3.3 μm with a 10 $\times$ objective. A balanced photodetector was used together with a lock-in amplifier to enhance the signal-to-noise ratio. The phase signal was fitted to a Fourier conduction model to obtain the thermal properties of the sample. A basic assumption in the data analysis is that we treat the stacked MAC sample only as part of the total thermal resistance between the Au transducer and the SiO_2 substrate, which also includes resistances from the Au/MAC and MAC/ SiO_2 interfaces. These three parts are in series, so the contributions of the two interfaces can approximately be subtracted by using the 1L sample as a reference. Both FDTR mapping and single-spot measurement were performed, with six and 50 sampling frequencies, respectively.

D. Suspended microbridge

The in-plane thermal transport was measured using the suspended microbridge method [52,53]. To this end, we first custom designed and fabricated a series of microbridge devices at Peking University. The devices consist of two silicon nitride (SiN_x) islands that are suspended and thermally isolated from the substrate by long and narrow SiN_x beams and also integrated with thin film serpentine platinum resistors [Figs. 3(a) and 3(b)] for precise heating and temperature sensing. In a typical experiment, one of the SiN_x islands is used as the heater, while the other one serves as the sensor. The sample acts as a bridge for heat flow

between the heater and the sensor, the thermal conductivity of which can be obtained as $\kappa = G_{\text{MAC}}(L/A) = [Q/(T_h - T_s)](L/A)$. Here, G_{MAC} is the sample thermal conductance. The temperatures of the two islands (T_h and T_s , respectively) are readily monitored with the platinum resistors. The heat current through the sample (Q) is measured based on T_s and the beam conductance G_b (typically around 330 nW K^{-1}) of the sensor island. The sample length L and cross-sectional area A are obtained using SEM and AFM.

E. Construction of atomic model

The atomic structure of MAC (Supplemental Note 11 and Fig. S17) was generated by the kinetic Monte Carlo (kMC) method [54]. Models with 60% hexagonal rings were chosen as representatives for subsequent structural characterizations and thermal simulations. The distributions of ring size, bond length, and bond angle, together with the pair correlation function of the MAC models (Fig. S18) were analyzed through the Atomes software [55]. For stacked MAC, 16 different MAC structures with a common lateral size were first generated by kMC. Then, stacked MAC containing 8L, 10L, 12L, 14L, and 16L were formed by vertically assembling these monolayers [26]. Further, randomly stacked graphene with the same number of layers [26] was created by employing an algorithm that was previously used to generate twisted bilayer graphene [25,56]. The models of 3D amorphous carbon [26] were generated through a melt-quench method [24]. All atomic structures were visualized through the OVITO software [57].

F. Atomistic simulations of thermal transport

Simulations of thermal transport in freestanding and stacked MAC, stacked graphene, and 3D amorphous carbon (Supplemental Note 14 and Fig. S19) were performed with the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package using nonequilibrium molecular dynamics [58,59]. For freestanding MAC and 3D amorphous carbon, the optimized Tersoff potential was used to describe the interatomic interactions [60,61]. For cross-plane thermal transport in stacked MAC and graphene, the interlayer interaction was described by the Lenard-Jones potential. The VDOS was obtained by Fourier transforming the velocity autocorrelation function. The participation ratio could then be calculated based on the local VDOS [62]. To obtain the dispersion relations, the MD trajectories of the simulated structures were used to analyze the longitudinal and transverse current correlation function through the DYNASOR code [63]. We also calculated the phonon dispersion of graphene using lattice dynamics via the PHONOPY package [64], yielding results consistent with MD simulations. Representative eigenvectors of 3D amorphous carbon and MAC were obtained using PHONOPY.

For the in-plane thermal conductivity of MAC, both the harmonic Allen-Feldman [21] and the quasi-harmonic Green-Kubo (QHGK) [9] calculations were conducted

using the κ ALDO software [65] on a $10\text{ nm} \times 10\text{ nm}$ model. In the Allen-Feldman approach, the broadening was set to 5 times the average spacing of the vibrational mode frequencies [66,67]. Both methods underestimate the thermal conductivity at low temperatures compared to the measured and MD-simulated results, which may be attributed to limitations in the model dimensions [68,69] and to the way in which the quantum effects are incorporated.

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DATA AVAILABILITY

The data supporting this study's findings are available within the article.

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