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Measurement of the thermal conductance of the graphene/SiO₂ interface

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We have examined the interfacial thermal conductance G_K of single and multilayer graphene samples prepared on fused SiO₂ substrates by mechanical exfoliation of graphite. By using an ultrafast optical pump pulse and monitoring the transient reflectivity on the picosecond time scale, we obtained an average value of G_K of $G_K=5000$ W/cm² K for the graphene/SiO₂ interface at room temperature. We observed significant variation in G_K between individual samples, but found no systematic dependence on the thickness of the graphene layers. © 2010 American Institute of Physics. [doi:10.1063/1.3511537]

Graphene, a monolayer-thick sheet of graphite, has attracted much attention because of its unique electronic properties.^{1,2} While our understanding of the electronic states and transport in graphene has advanced dramatically,^{1,2} our knowledge of thermal transport in this material system²⁻⁴ is currently less advanced. The thermal transport properties are, however, of interest for both fundamental reasons and for applications. From the fundamental perspective, thermal transport provides insight into the properties of phonons and their interactions.⁵⁻⁷ For applications, power dissipation often limits device performance.⁸ Of particular importance is the issue of interfacial thermal transport between graphene layers and the substrate,^{9,10} which plays a critical role in current saturation of graphene devices.^{11,12}

In this paper, we report the determination of the interfacial thermal conductance G_K of graphene on a SiO₂ substrate. The measurements were performed using sudden heating of the graphene layers, exfoliated on transparent SiO₂ substrates, by a femtosecond laser pulse. The subsequent heat flow across the graphene-substrate interface, which occurs on the time scale of tens of picosecond, was monitored with a time-delayed optical probe pulse. From analysis of these data, we deduce an average room-temperature value of $G_K=5000$ W/cm² K for graphene on SiO₂. A significant variation in G_K was observed for different individual samples, with values as high as 11 000 W/cm² K having been observed. On the other hand, for graphene samples of $N=1-13$ monolayers thickness, we found no systematic variation in G_K with N .

The pump pulses for this measurement were supplied by a mode-locked Ti:sapphire laser operating at a wavelength of 800 nm and providing 100 fs pulses at an 80 MHz repetition rate. The probe pulses at a 400 nm wavelength were obtained by frequency doubling in a β -barium borate crystal. The 400 nm probe wavelength was chosen because of its enhanced sensitivity to temperature associated with optical transitions near the M-point in the graphene Brillouin zone.¹³ The pump and probe beams were focused on the graphene samples with spot sizes of $\sim 5-10$ μm . The pump-probe measurements were performed by modulating the pump laser at 1 kHz and detecting the synchronous change in intensity of the probe

reflection. The pump fluence was varied between 100 and 500 $\mu\text{J}/\text{cm}^2$, with the probe fluence kept below 10% of this value.

The graphene samples were prepared by mechanical exfoliation² of kish graphite deposited on transparent SiO₂ substrates (Chemglass Inc.) that had been thoroughly cleaned in methanol. The surface topography of the substrate was characterized by atomic-force microscopy. A roughness of 1–2 nm was observed over a typical 10×10 μm^2 area, similar to that of SiO₂ films grown thermally on Si substrates.¹⁴ The exfoliated graphene samples studied were of homogeneous layer thickness and areas of several hundreds to thousands of squares of micrometers. The sample thickness was determined by optical absorption measurements, which provide monolayer accuracy.¹⁵ All experimental measurements were carried out at room temperature under ~ 10 mTorr vacuum.

Figure 1 displays transient reflectivity data for monolayer, eight- and 13-layer graphene samples over a 200 ps time window. The decay of these transients can be fit using a biexponential form. The fast component has a time constant of $\tau_1 \sim 2$ ps; the slow component has a thickness-dependent time constant of $\tau_2 \sim 10-100$ ps, as shown in Fig. 2. The slow-decay component τ_2 exhibited no dependence on the

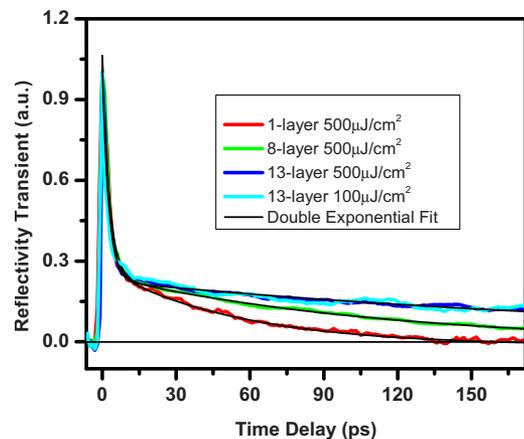


FIG. 1. (Color online) Transient reflectivity decay dynamics of monolayer, eight-, and 13-layer graphene samples, together with double-exponential fits to the data. The decay dynamics for the 13-layer graphene sample is plotted for two different pump fluences. After rescaling, identical decay dynamics are observed.

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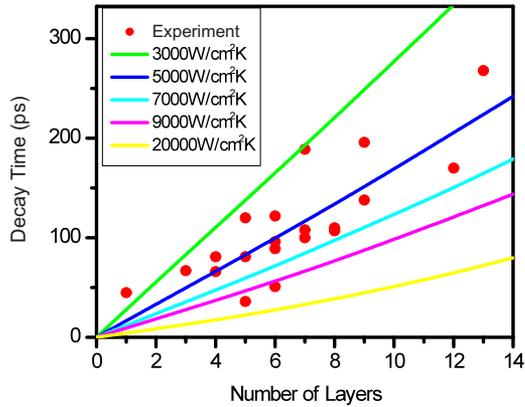


FIG. 2. (Color online) Decay time constants τ_2 for interfacial heat flow as a function of graphene layer thickness. Dots: experimental data, with multiple points corresponding to values for different samples of the given thickness. Lines: numerical simulations, as described in the text, for different values of the interfacial thermal conductance G_K .

pump fluence (Fig. 1). This indicates that we remain in the regime of weak perturbation.

For the purpose of this investigation, we are not concerned with the fast relaxation component. As discussed in earlier publications, this picosecond response is associated with the equilibration of the electronic excitations and optical phonons with other phonons in the system.^{13,16} On the time scale of the slow relaxation component, full equilibrium between the different degrees of freedom in the graphene samples should be achieved, with essentially all of the thermal energy residing in excitations of acoustic phonons. On this time scale, we can then relate the reflectivity transient to the temporal evolution of graphene temperature. Given the very slight change observed (10^{-6} – 10^{-5}) in the reflectivity, we can assume that the change in sample temperature is linear in the change in optical reflectivity.

Once the different sub-systems of the sample have reached thermal equilibrium with one another, subsequent heat dissipation can in principle be achieved by in-plane heat flow within the graphene and out-of-plane heat transfer to the substrate. Given the micron size of the laser spot, lateral heat flow will not be significant on the relevant (subnanosecond) time scale, since $\tau_p = d^2 D_p \approx (25 \mu\text{m}^2) / (2.5 \text{cm}^2/\text{s}) \approx 100 \text{ns}$, where τ_p , d , and D_p denote, respectively, the lateral heat diffusion time, the laser spot size, and the in-plane thermal

diffusivity of the sample (approximated by that of graphite¹⁷). In addition, cooling by lateral heat flow is incompatible with the observed dependence of τ_2 on the thickness of the graphene sample (Fig. 2). The observed decay in temperature thus arises from heat flow into the substrate.

Heat flow into the substrate is characterized by the interfacial (Kapitza) thermal conductance G_K . The values of G_K can be extracted from τ_2 by considering vertical heat flow within the graphene sample and the SiO_2 substrate, which we treat as diffusive, and heat flow across the interface as described by interfacial conductance G_K . The initial condition for the heat-flow problem is defined by the optical excitation. As the optical penetration length, $\alpha^{-1} \approx 15 \text{nm}$,¹⁵ is much larger than the thickness of the graphene samples, the graphene is heated homogeneously with depth. The substrate is completely transparent and is not directly heated by the laser radiation.¹⁸ The absolute temperature rise of the graphene samples is not needed for our analysis, but can be determined from the absorbed laser fluence and its total heat capacity (again approximated by that of graphite¹⁹), as appropriate for full thermal equilibrium. For the highest applied fluence of $F = 500 \mu\text{J}/\text{cm}^2$, we obtain a temperature rise of about 150 K.

We fit the experimental data using the thermal diffusivities of graphite¹⁷ (out-of-plane diffusivity of $0.017 \text{cm}^2/\text{s}$) and fused silica²⁰ ($0.009 \text{cm}^2/\text{s}$) and treating G_K as an unknown parameter. We find (Fig. 2) that the calculated decay time constants τ_2 vary nearly linearly with N . Particularly for the lower values of G_K , interfacial heat flow completely controls cooling of the graphene samples. In this interface dominated regime, τ_2 varies linearly with N , since the total amount of heat that must be transported across the interface increases linearly with the amount of materials. On the other hand, for sufficiently high values of G_K , greater substrate heating for thicker graphene samples leads to reduced heat flow across the interface. The relation between τ_2 and N then deviates from linearity, as can also be seen in Fig. 2. When cooling of the sample is completely defined by bulk heat flow, either through the sample or the substrate, we obtain $\tau_2 \propto N^2$, as expected for diffusive heat transport. Based on our experimental data (Fig. 2), we see that we are in fact essentially in the regime of *interface limited heat flow*.

In Fig. 3(a) we present a histogram of the values of the interfacial thermal conductance G_K for graphene samples of

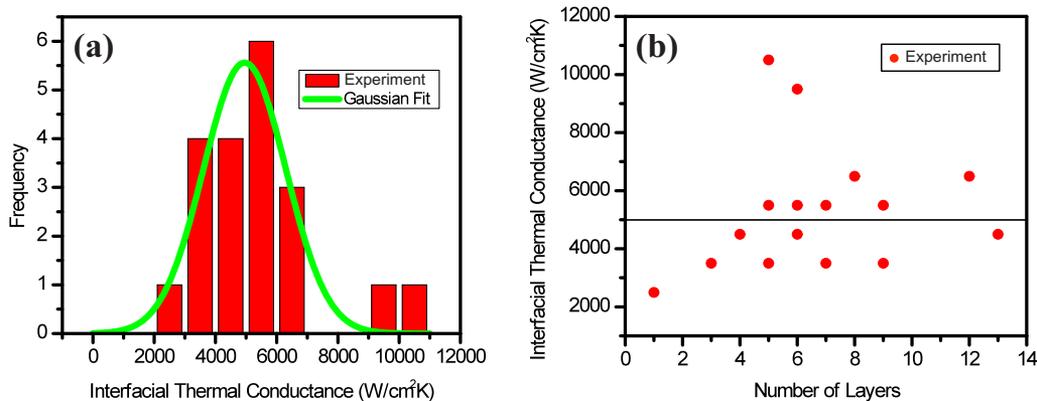


FIG. 3. (Color online) (a) Histogram of the values of the interfacial thermal conductance G_K extracted from the data in Fig. 2. Green curve: Gaussian fit to data, yielding a thermal conductance of $G_K = 5000 \pm 1300 \text{W}/\text{cm}^2 \text{K}$. (b) Measured G_K as a function of graphene layer thickness. No correlation with thickness is seen. The black horizontal line represents the average value.

all layer thicknesses. From a Gaussian fit, we obtain an average $\langle G_K \rangle = 5000 \text{ W/cm}^2 \text{ K}$ (compatible with that deduced in Ref. 9), with a standard deviation of $1300 \text{ W/cm}^2 \text{ K}$. Figure 3(b) displays the individual values of G_K from our measurements as a function of layer thickness N . Within our experimental accuracy, no correlation is seen.

The values of G_K obtained for the graphene/SiO₂ interface are comparable to those measured for single walled carbon nanotubes in solution²¹ and on SiO₂ substrates²². The values are also similar to those reported for different metal-insulator interfaces, which typically lie between 3,000 and 11,000 W/cm²K.^{23,24} While the graphene/SiO₂ interface displays good thermal transport properties, the measured values for G_K vary between 2,000 W/cm²K and 11,000 W/cm²K. This large dispersion presumably reflects the relatively poorly defined nature of interface produced by the mechanical deposition process. Some of the samples may have better surface contact with the substrate than others, yielding accordingly better interfacial heat flow. The highest value of the interfacial conductance obtained in this measurement was $G_K = 11,000 \text{ W/cm}^2 \text{ K}$. The intrinsic value for the graphene/SiO₂ interface could be still higher. The measurement technique presented in this letter can readily be extended to probe other interfaces with graphene. Investigations of graphene interfaces with different substrates over a range of temperatures should help to elucidate which phonons are responsible for the dominant transfer of energy at the interface. Careful calibration of the temperature rise for a given absorbed laser fluence could also serve to identify possible enhanced energy flow during the initial non-equilibrium response of the graphene to femtosecond laser excitation. In this regime conditions may be similar to those for high-field charge transport where energy transfer by coupling of electronic excitations directly to substrate optical phonons should be considered.¹⁰

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¹⁸In the initial (<1 ps) period after excitation by the pump pulse, the graphene is in a nonequilibrium state characterized by an elevated electronic temperature. This may result in more rapid energy transfer to the substrate through channels such as coupling of the graphene electrons to substrate polar phonons (Ref. 10). However, the absence of any fluence dependence of the measured thermal behavior suggests that this process does not affect the subsequent evolution of the equilibrium interfacial heat flow.

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