Photo-thermal response in dual-gated bilayer graphene

M.-H. Kim,1 J. Yan,1,2 R. J. Suess,3 T. E. Murphy,3 M. S. Fuhrer,1 and H. D. Drew1

1Center for Nanophysics and Advanced Materials, Department of Physics, University of Maryland, College Park, Maryland 20742, USA
2Physics Department, University of Massachusetts, Amherst, MA 01002, USA
3Institute for Research in Electronics and Applied Physics, University of Maryland, College Park, Maryland 20742, USA

The photovoltaic and bolometric photoresponse in gapped bilayer graphene was investigated by optical and transport measurements. A pulse coincidence technique at 1.5 μm was used to measure the response times as a function of temperature. The bolometric and photovoltaic response times were found to be identical implying that the photovoltaic response is also governed by hot electron thermal relaxation. Response times of \( \tau \sim 100 \text{ - } 20 \text{ ps} \) were found for temperatures from 3 - 100 K. Above 10 K, the relaxation time was observed to be \( \tau = 25 \pm 5 \text{ ps} \), independent of temperature within noise.

There is growing recognition that graphene has exceptional potential as a new optoelectronic material, which has led to a flurry of recent research activity and rapid advances.1-3 Graphene’s unique massless band structure gives rise to direct transitions and strong (specific) coupling to light at all wavelengths,4 and ultra-fast response (from nanosecond to femtosecond)4 room temperature operation for many applications. A photovoltaic response has been observed for visible light and we have recently observed both photovoltaic and bolometric response in bilayer graphene at THz frequencies.5 Diode-like rectification behavior is observed with contacts to dissimilar metals.1,2,3,6 However, the mechanism of the photovoltaic response has not been definitively identified. Both p-n junction physics similar to conventional semiconductor photovoltaic sensors and a thermoelectric mechanism remain viable possibilities. In a recent study we observed a hot electron bolometric response in bilayer graphene, which highlighted the outstanding thermal properties of graphene.6 Therefore, understanding the role of hot electron effects in the photoresponse of graphene is critical to the development of graphene-based optoelectronic devices such as bolometers and photovoltaic sensors.6

Excited electrons in graphene thermalize quickly on the femtosecond time scales6,8 by electron-electron scattering.10 These hot electrons transfer their thermal energy to the graphene lattice by the emission of phonons on a much longer time scale because of the weak electron-phonon interaction.10,13 The thermal relaxation of hot electrons by optical phonons3,14,15 and by acoustic phonons5,16 has received much recent attention. The processes of cooling by optical and acoustic phonons are clearly distinguishable because their thermal timescales differ by a few orders of magnitude. High pulse energy radiation produces hot electrons with energies above the optical phonon energy (\( \sim 200 \text{ meV} \)) that cool by optical phonon emission in a timescale of a few picoseconds.3,14,15 For longer times and/or lower pulse energy radiation, acoustic phonon assisted cooling is dominant with sub-nanosecond timescales.5,16

Hot electrons can be utilized for bolometric and photovoltaic photoresponse detection.3,7,17 The bolometric response makes use of the temperature dependence of the resistivity, which is significant in gapped bilayer graphene. On the other hand, the hot electrons can also give rise to a photo-thermoelectric response. Diffusion of heat and carriers to the contacts produce a thermoelectric response. A competing mechanism for photovoltaic response is charge separation by the built-in electric fields at metal-graphene junctions due to proximity doping. It remains unclear which of these two mechanisms dominates in graphene photovoltaic devices.

In this paper, we use electrical transport and optical photoresponse measurements to characterize the bolometric and photovoltaic response of a dual-gated bilayer graphene device. The temperature-dependent resistance of the device which allows a bolometric response is characterized both optically and with AC transport measurements, which together establish that the response is thermal. We found that light also generates a voltage across the sample with zero bias current. We compare this photovoltaic response with the well-understood bolometric signal in the same device as functions of dual-gate voltages and temperature. In particular pulse coincidence measurement reveals that the photo voltage displays the same temperature-dependent relaxation time as the bolometric response, demonstrating that diffusive hot carrier relaxation in graphene dominates the observed photo voltage of the device.

The bilayer graphene device we studied was fabricated by mechanical exfoliation of natural graphite on a resistive silicon wafer (200 Ωcm) which was ion implanted with boron and annealed to provide a highly conducting but transparent back-gate at cryogenic temperatures. A 300 nm thick SiO₂ layer was grown by a dry oxidation process from the silicon wafer. A micrograph of the device is shown in Fig. II(a) inset and schematic of device geometry is shown in Fig. II(b) inset. A thin Nichrome film was used as a semitransparent top-gate. Details of
allows pulse coincidence measurements with precise time delays from a few ps to 10 ns without a mechanical delay line. The absorption of 1.56 µm radiation in the graphene was estimated to be 1.2% by considering effects due to the silicon substrate and the Nichrome top gate. The graphene absorbs an average power of 0.37 nW from the pump and probe pulses and generates a temperature rise \(\Delta T\), which can be measured using the temperature dependence of \(R\).

The dependence of the photoresponse with pulse time delay for different bias currents is shown in Fig. 2(a) under conditions that the device is gated to its charge neutral point. At zero bias current the signal is purely photovoltaic. For non-zero bias there is also a bolometric signal given by \(V_b = I_d c \Delta R\) which was reported earlier. We find that the total photoresponse can be described as \(V_{pv}(I_{dc}) = V_b(I_{dc}) + V_{pr}\), allowing a separation of the photovoltaic and bolometric contributions. Therefore the bolometric response can be obtained from \(V_b(I_{dc}) = V_{pr}(I_{dc}) - V_{pr}(0)\). It is seen in the figure that this bolometric response is dominant except near \(I_{dc} = 0\).
where the response is purely photovoltaic.

These pulse time delay data allow a measurement of the response time $\tau$ of the two components of the photoresponse. For long pulse delay times, $t_d$, average probe-pulse induced photo voltage, $V_{pr}$, does not change with respect to the time delay $t_d$. When the delay is short ($t_d < \tau$), however, the magnitude of $V_{pr}$ is reduced due to the nonlinear radiation power dependence of the response so that the photo voltage $V_{pr}(t_d)$ displays a peak or dip at $t_d = 0$. The magnitude of this peak or dip increases with the non-linear power dependence of $V_{pr}$.

Figure 2(b) shows $V_{pv}$ and $V_b(I_{dc})$ normalized to the response in the absence of the pump pulse for several different $I_{dc}$. All of the normalized $V_b$ for different $I_{dc}$ collapse to one curve because the small Joule heating $I_b^2 R$ does not significantly raise the electron temperature above the lattice temperature. Both bolometric and photovoltaic responses produce a narrow dip with a sub-nanosecond width at zero time delay when the pump and probe pulses are coincident. Surprisingly, the widths of both bolometric and photovoltaic dips are seen to be the same to within the experimental error. The time constants determined by the half widths at half maximum of the dips are $0.12 \pm 0.01 \text{ ns}$. This demonstrates that both $V_{pv}$ and $V_b(I_{dc})$ have the same response time and since the bolometric response is clearly thermal this implies that the photovoltaic response is also thermal. Similar results are observed at $V_{tg} = 0 \text{ V}$ and $V_{bg} = 30 \text{ V}$ for temperatures below $10 \text{ K}$.

To gain further insight into the nature of the photo voltage, we measured its gate voltage dependence. Figure 3 shows back gate voltage dependence of photo voltage at $T = 15 \text{ K}$ with $V_{tg} = 0$ and $I_{dc} = 0$. As can be seen from the data in Fig. 1(a), the top gate does not gate the entire device. To obtain uniform gating we control only the back gate voltage with zero top gate voltage. Figures 3(a) and (b) display the photovoltaic response below and above the maximum $V_{pv}$ observed at around $V_{bg} = 25 \text{ V}$. The peak or dip structure is associated with the sign of $V_{pv}$, and its depth or height depends on the nonlinear power dependence of $V_{pv}$. Both sign and power nonlinearity depend on back gate voltage. For example, at $V_{bg} = 15.5 \text{ V}$ the response $V_{pv}(t_d)$ is independent of $t_d$ indicating that $V_{pv}$ is linear with radiation power. As the power nonlinearity of $V_{pv}$ grows above or below $V_{bg} = 15.5 \text{ V}$, the dip or peak of $V_{pv}$ appears and grows. Remarkably, however, all of the pump-probe data have the same $\tau = 25 \pm 5 \text{ ps}$. The gate-independent time constant shows that the photovoltaic response is thermal at all gate voltages not only at maximum $R$ with respect to $V_{bg}$ as shown in Fig. 2(b) where it could be directly compared with the bolometric response. This observation demonstrates that the photovoltaic response time in bilayer graphene is the intrinsic thermal time constant of hot electron energy relaxation.

We also measured the temperature dependence of the response time in our graphene device. Figure 4 exhibits $\tau$ obtained from the photovoltaic response in the temperature range $3 \text{ K} - 87 \text{ K}$ at several different dual-gate voltages. The response time for different gate voltages coincides within error as shown in Fig. 3. The time constant decreases from $\sim 80 \text{ ps}$ at $3 \text{ K}$ to $\sim 20 \text{ ps}$ at $80 \text{ K}$. Above $T \sim 10 \text{ K}$, $\tau$ is seen to be temperature independent to within experimental error.

The thermal relaxation rate is given by the ratio of the electronic heat capacity $C$ to thermal conductance $G$. The thermal conductance was obtained using transport measurements as described in Ref. 5. For $T < 8 \text{ K}$, the transport measurements give $G = 0.5 \times (T/5)^3 \text{nW/K}$ which is in reasonable agreement with the value estimated for cooling by acoustic phonons. 12 A crossover of the thermal conductance from $T^3$ to linear $T$ is predicted for $T > T_{BG}$, where $T_{BG}$ is the Bloch-Grüneisen temperature given by $k_B T_{BG} \approx 2h v_s k_F$. 13 Assuming a sound velocity $v_s = 2.6 \times 10^4 \text{ m/s}$ 21 and a disorder-induced charge density of $n_{rms} \sim 10^{12} \text{ cm}^{-2}$ 22, we find $T_{BG} \sim 70 \text{ K}$. Although our sample is nominally charge
neutral at \( R_{\text{max}} \), it is widely accepted that disorder creates electron-hole puddles \[22\] and thus \( T_{BG} \) is non-zero at all gate voltages. Transport measurements show that the Bloch-Gr"uneisen regime behavior occurs for \( T < 0.2T_{BG} \sim 14 \) K. \[20\] The behavior of \( G \) and \( C \) above \( T \sim 0.2T_{BG} \) may be complicated by disorder induced supercollision cooling \[16\], \[23\] and/or the non-parabolic band structure of gapped bilayer graphene \[4\] which leads to small Fermi energies. We measured \( G = 0.91 \times T^{1.04} \) nW/K for \( T > 8 \) K, which is reasonable in view of these considerations.

On the other hand, diffusion cooling of hot electrons also gives a linear \( T \) dependent thermal conductance. Diffusion cooling provides a thermal conductance \( k = \Delta T/R_g \), by the Wiedermann-Franz law, where \( \Lambda = 24.4 \) nWΩ/K\(^2\) is the Lorentz number, and \( R_g \) is bilayer graphene resistance. At the peak resistance for our sample, \( k = 3.4 \times 10^{-12} \times T^{1.0} \) W/K, which is two orders of magnitude smaller than the electron-phonon conductance. We conclude that acoustic phonon mediated cooling of hot electrons is dominant in our samples.

We note, however, that the thermoelectric and photovoltaic signals are a consequence of diffusion. For asymmetric contacts the thermal diffusion and charge flow at the two contacts differs leading to a net potential difference. The diffusion length \( \xi = (k/G)^{1/2} \) is estimated to be \( 0.5 \) µm at 10 K which is much smaller than the sample size of 5 µm so that the sample temperature rise and response time is dominated by the thermal conductance to the lattice which greatly reduces the thermoelectric signals in these large area, low conductance samples. The thermoelectric effect produces an electric field \( e = S \nabla T \), where \( S \) is the Seebeck coefficient and \( \nabla T \) is the gradient of the temperature. The heat conductance to the lattice diminishes \( \nabla T \) at the contact and therefore the thermoelectric field by a factor of \( 2\xi/L \) compared with pure diffusion.

At low temperatures (\( k_B T < \mu \), where \( \mu \) is the local Fermi energy in the graphene and \( k_B \) is the Boltzmann constant) the electronic specific heat is \( C = \alpha T \), where \( \alpha = (\pi^2/3)\nu(E_F)k_B^2 \), where \( \nu(E_F) \) is the density of states for bilayer graphene. In the parabolic band approximation of (ungapped) graphene \( \nu(E_F) \approx \gamma_1/(\pi h^2 v_F^2) \) where the interlayer coupling \( \gamma_1 = 390 \) meV \[24\], \( v_F = 1 \times 10^6 \) m/s is the monolayer graphene Fermi velocity. For our sample area of 25 \( \mu m^2 \), this gives \( \alpha = 2.6 \times 10^{-20} \) J/K\(^2\). Thus the thermal response time of our bilayer sample can be estimated \( \tau = C/G \approx 29 \) ps independent of temperature for \( T > 8 \) K which is in reasonable agreement with the measured \( \tau \) shown in Fig. 4.

In summary, we have reported photovoltaic response time measurements on gapped bilayer graphene. The devices show both bolometric and photovoltaic responses, which were separated by their bias current dependence. The identical response time constants observed for the bolometric and photovoltaic responses as a function of gate voltages and temperature implies that both effects are governed by the same intrinsic hot electron-phonon thermal relaxation process. The observed response times of 10 - 100 ps indicates that hot electron relaxation occurs through acoustic phonon emission. These observations support the growing realization that graphene appears to have great promise for fast sensitive photo detectors over a wide spectral range.

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FIG. 4. Temperature dependence of thermal response time. Thermal time constant \( \tau \) of the photovoltaic response vs. temperature measured by the pulse coincidence technique for several different dual-gate voltage settings.
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