Modulation characteristics of graphene-based thermal emitters

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In the last few years, thermal emission from graphene has primarily been used to probe the electronic and thermal properties of graphene-based transistors under bias.\(^1\)\(^-\)\(^6\) In monolayer devices, using graphene both exfoliated and grown by chemical vapor deposition (CVD), thermal emission is not spatially uniform and the maximum emission (“hotspot”) occurs at the point corresponding to the minimum conductivity, i.e., the charge neutrality point.\(^1\)\(^-\)\(^7\) The application of a gate bias, which changes the charge distribution, can be used to move the hotspot along the length of the devices. Recently, we have assessed the potential of using graphene-based thermal emitters to enable the applications of low-cost, intrinsically safe, portable infrared gas sensors in, for example, mine safety.\(^7\) For the currents used, which could be sustained by the devices for over 100 h, the emission from these devices peaked at a wavelength of around 4 µm and covered the characteristic absorption of many important gases, demonstrating the feasibility of developing a graphene-based mid-infrared light-emitting device. Ultimately, such devices could be more cost-effective and sustainable to manufacture than either silicon MEMS or compound semiconductor-based LED alternatives.\(^8\),\(^9\)

However, one key advantage of semiconductor LEDs is their ability to be modulated at very high frequencies, which potentially enables both faster response and the use of more sophisticated signal processing approaches. Although a measurable thermal emission was obtained from graphene devices when the drive current was modulated up to a frequency of 100 kHz, the amount of signal (corresponding to the difference between the thermal emission obtained when the current is on and off) measured at 100 kHz was approximately a factor of 100 smaller than that measured at 1 kHz.\(^7\)

In this study, we investigate the dependence of thermal emission over a range of drive current frequencies for multilayer graphene-based thermal emitters, both experimentally and by COMSOL finite element simulation.

Multilayer graphene (MLG; from Graphene Square and Graphene Supermarket), pre-transferred on 300-nm-thick SiO\(_2\) on a highly p-doped Si substrate, was used to fabricate devices. Electron beam lithography followed by reactive ion etching in an O\(_2\)/Ar plasma were used to define 600 × 500 µm\(^2\) areas of graphene. Source and drain contacts, 600 µm long and 200 µm wide, of Cr/Au (7/70 nm) were deposited on graphene by thermal evaporation, resulting in an exposed graphene area of 500 × 500 µm\(^2\). Figure 1 shows a schematic of a typical device. The uniformity and nature of the graphene were confirmed by Raman spectroscopy, which indicated that multilayer samples contained 3–6 layers of graphene. For electrical characterization and thermal emission measurements the devices were mounted on a ceramic chip holder, and placed inside a vacuum chamber with a CaF\(_2\) window for optical access. The vacuum chamber was evacuated to \(~10^{-5}\) mbar. For all measurements, a pulsed DC current with a 50% duty cycle was applied using a Keithley 6221 current source. A two-terminal current–voltage measurement was used to obtain the resistance of the device, which was typically around 1300 Ω, at room temperature, with the resistance due to the contacts and leads assumed to be negligible. The resistance is typical of those obtained using CVD graphene.\(^10\) Spatial emission measurements were performed by collecting the emitted light using a reflecting objective lens (NA = 0.28) and then focusing the light using a CaF\(_2\) lens onto a liquid-nitrogen-cooled MgCdTe detector, with a 2–12 µm response. The reflecting objective, CaF\(_2\) lens, and detector were mounted on an xy-stage and the spatial
The measured spatial variation of the thermal emission from a typical multilayer device for a peak current density of \(1.0 \times 10^7\) A cm\(^{-2}\), well below the breakdown current of graphene, at a drive frequency of 1 kHz is shown in Fig. 3(a).\(^{21}\) In contrast to that from monolayer graphene-based devices, the thermal emission from multilayer devices has a maximum intensity at the center of the emitting area, as might be expected from a conventional semimetal filament. The emission intensity as a function of position across the center of the device is shown in Fig. 3(b), where the intensities have been normalized to the peak intensity at the center of the device. Simulated normalized intensities obtained from the COMSOL model are also shown in Fig. 3(b). These were obtained by first extracting the temperature of the emitting area as a function of position across the emitting area with the current on or off. Using the extracted temperatures and assuming that the emission is that of a grey body, we calculated the measured intensity using the known spectral response of the detector. Overall, there is good agreement between the measured and simulated results, demonstrating the validity of the COMSOL model.

To investigate the modulation characteristics of the graphene-based emitter, the emission from the devices was simulated as well as measured as a function of drive fre-
The emission intensities were extracted from the simulated temperatures by again assuming that the emission from graphene is that of a grey body, and they are plotted as a function of frequency in Fig. 4(b). From the simulated intensities, it is clear that increasing the thermal resistance between the MLG emitter and the underlying SiO$_2$ causes an increase in the intensity, with an approximate increase of 1.5-fold above the frequency of 30 kHz. The measured values of emission at a current density of $2.0 \times 10^7$ A·cm$^{-2}$ are also plotted in Fig. 4(b), where the open triangles correspond to the emission from the MLG device. The measured intensity drops rapidly as a function of frequency, but is approximately 5-fold lower than those obtained from the simulations across the frequency range.

However, it is more instructive not to compare the measured and simulated values, but to explore the consequences of experimentally increasing the thermal resistance between the MLG emitter and the underlying substrate. The closed symbols in Fig. 4(b) therefore correspond to the measured intensity obtained from a new device in which the multilayer graphene is encapsulated above and below by multilayer hexagonal boron nitride. Hexagonal boron nitride (h-BN) makes for a good dielectric support for graphene owing to its clean, atomically smooth surface and the fact that it belongs to the same hexagonal layered family as graphene and has a similar lattice constant. In comparison with multilayer graphene on SiO$_2$, the thermal emission from MLG on multilayer h-BN increases by a factor of 2 at high frequencies ($\geq 30$ kHz). This can be explained by considering that the anisotropy of the thermal conductivity of h-BN is due to its layered crystal structure with a $c$-axis thermal conductivity of $\sim 2 \text{ Wm}^{-1}\text{K}^{-1}$. In the basal plane, the thermal conductivity has been shown to be $\sim 400 \text{ Wm}^{-1}\text{K}^{-1}$ at room temperature. As heat loss from the graphene is dominated by the vertical heat sinking path, the anisotropy of the thermal conductivity has a marked effect on the modulation characteristics of the device. Taking the thickness of the bottom multilayer h-BN to be $\sim 13$ nm and the interfacial thermal resistance per unit area to be $1.35 \times 10^{-7}$ m$^2$K$^{-1}$m$^{-1}$ for the graphene/h-BN interface and $2.2 \times 10^{-7}$ m$^2$K$^{-1}$m$^{-1}$ for the h-BN/SiO$_2$ interface, the additional thermal resistance per unit area for the h-BN device is calculated to be $1.435 \times 10^{-7}$ m$^2$K$^{-1}$m$^{-1}$ relative to graphene/SiO$_2$ devices with an interface thermal resistance of $\sim 2 \times 10^{-8}$ m$^2$K$^{-1}$m$^{-1}$ (derived from the value obtained for exfoliated samples). Increasing the thermal vertical resistance in the experiments therefore leads to an increase in the measured intensity at high frequencies, in agreement with the results of COMSOL simulations where the interface thermal resistance was increased by a similar amount. Further optimization of the device design should enable the emission intensity at higher frequencies to be further increased.

In summary the thermal modulation as a function of drive frequency for large-area CVD graphene devices was investigated experimentally and simulated by finite element method modelling in COMSOL. For devices with a multilayer graphene emitter, a measurable modulation was observed at 100 kHz, but the measured intensity was approximately 100-fold less than that measured at 1 kHz. COMSOL simulations indicate some of the complexity in designing a real device.
simulations showed that the measured intensity at high frequencies can be increased by increasing the thermal resistance between the graphene emitter and the underlying SiO₂. Measurements showed that the encapsulation of the emitting area with hexagonal boron nitride can increase the thermal resistance. This approach therefore provides a promising route to the realization of practical infrared emitters that can be used to replace expensive semiconductor LED equivalents.

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