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ด้วยเทปกาว

Graphene Deposition onto a Si/SiO₂ Substrate Using
Micromechanical Exfoliation

นางสาวรัชชก สมพรเสน่ห์



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ได้รับทุนสนับสนุนงานวิจัยจากงบประมาณเงินรายได้ ประจำปีงบประมาณ พ.ศ. 2558

ภาควิชาฟิสิกส์ คณะวิทยาศาสตร์

สถาบันเทคโนโลยีพระจอมเกล้าเจ้าคุณทหารลาดกระบัง

Research Title: Graphene deposition onto a Si/SiO₂ substrate using micromechanical exfoliation

Researcher: Dr. Ratchanok Somphonsane

Faculty: Science **Department:** Physics

ABSTRACT

We study the technique of micromechanical exfoliation for isolating graphene and successfully prepare graphene flakes on SiO₂ substrate. We find that different types of graphite used as a source material gives different yield of graphene flakes. HOPG and natural graphite provides flakes that tend to be rather narrow and “ripped”, and that frequently are found to be close to large pieces of graphite, making it difficult for subsequent lithography processing. Kish graphite, in the other hand, provides much larger graphitic flakes that are of high quality and so it is a better choice for experimental studies. To preliminarily determine the number of graphene layers obtained from Kish graphite we use simple and reliable optical microscopy technique. With a proper choice of substrate — a 300-nm thick SiO₂ layer on top of a Si substrate — we are able to identify several potential flakes, both monolayer and bilayer graphene, that can be used for fabrication of graphene FET in the future.

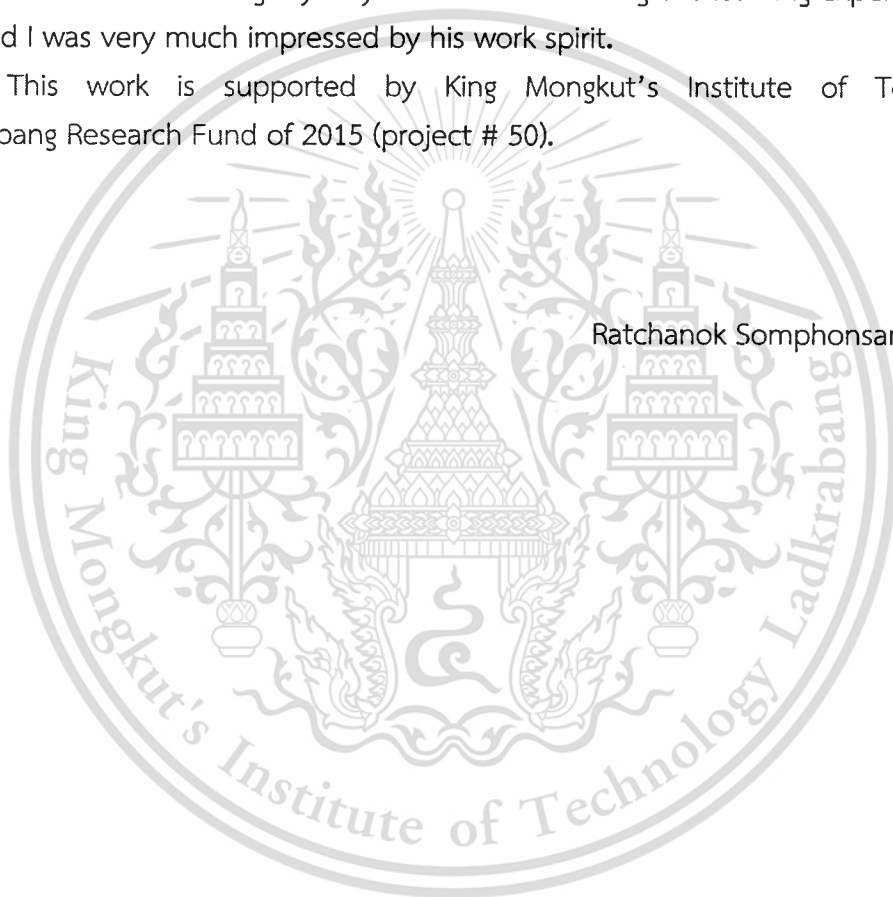
Keywords: 2D materials; graphene; exfoliation; van der Waals force; processing

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Ratchanok Somphonsane



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Chapter 1

Introduction

1.1 Introduction to the Research Problem and Its Significance

Carbon is one of the most abundant of chemical elements, and provides the basis for all organic chemistry. In the past few years, however, this element has aroused enormous interest around the world, as a potentially-new material for future electronics. More specifically, this interest has focused on the properties of a graphene – a single layer of carbon atoms, bound into a two-dimensional (2D) honey-comb lattice, and which may be thought of as being composed of benzene rings that are stripped away from their hydrogen atoms. Graphene is the fundamental component of various carbon allotropes, including three-dimensional graphite, one-dimensional carbon nanotubes and zero-dimensional fullerenes. While these allotropes have been widely studied experimentally [1-3], in some cases for as long as several decades, until recently graphene had remained the subject [4] of purely theoretical interest. In 2004, however, the successful isolation of graphene was first reported by Geim and Novoselov [5], who received the Nobel Prize in 2010 for their achievements. Their work made graphene the first truly 2D system to ever be isolated in nature. This remarkable feat was achieved in spite of the fact that it had not previously been expected that graphene could exist in the free state. Rather, it was long believed that any small, non-zero, temperature would cause the 2D crystal to lose its long-range order and break up [6, 7]. The experimental observation of an unusual quantum Hall effect in this material, reported simultaneously by the Geim and Kim groups in 2005 [8, 9], provided another noteworthy milestone in our understanding of this material. Since then, this one-atom-thick material has spurred enormous interest, in both the physics and engineering communities.

The reasons for the ever growing scientific interest in graphene are many-fold. A remarkable property of graphene is that its charge carriers behave like "massless Dirac fermions", contrary to the massive charge carriers that are found in most metallic or semiconducting systems. For this reason, carrier transport in graphene is described by the Dirac equation, instead of the conventional Schrödinger equation, allowing graphene to serve as a bridge between condensed-matter physics and quantum electrodynamics. In fact, a "counterintuitive" tunneling phenomenon of relativistic particles - also known as the Klein paradox - was observed experimentally for the first time in graphene [10, 11]. Additionally, quantum electrodynamics and the introduction of a sublattice pseudospin have led to the understanding of the half-integer quantum Hall effect in graphene [8, 9, 12, 13]. The isolation of graphene involves a relatively straightforward process, when compared to the synthesis of

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conventional semiconductors such as silicon, since all that is needed is graphite and adhesive tape to isolate graphene of high crystalline quality!

Apart from being an ideal candidate for fundamental physics research, graphene has also attracted widespread technological interest [14]. The fabrication of graphene devices is compatible with existing CMOS technologies, making it of significant interest as a platform for carbon-based nanoelectronics [15]. The first publication on graphene reported a strong electric field effect, which allows for wide, voltage-controlled, tunability of its carrier density, in the range of $0 < n < \pm 10^{14} \text{ cm}^{-2}$ [5]. This effect should serve as a fundamental component in the design of future electronic devices based on this material. According to Moore's law [16], modern silicon-based electronics is approaching the limits of miniaturization, with gate lengths on the order of 22 nm, whereas a narrow graphene strip just a few nanometers wide may be used as a transistor [17].

Graphene has several other interesting properties which further make it an excellent candidate for device applications. These include high carrier mobility, high electrical and thermal conductivities, high mechanical strength, high flexibility, almost complete transparency, impermeability to gas, strong interaction of carriers with incident photons, etc. [18-21]. Potential applications include gas sensors for detecting individual molecules, touch screens, microprocessors, conductive coatings, and transparent electrodes for solar cells. The extremely high carrier mobility in graphene ($\sim 10^4 \text{ cm}^2/\text{Vs}$ at room temperature) allows one to use it to build spin valve-, superconducting- or ballistic transistors [7], as well as ultra-high frequency and high-speed devices [12, 22, 23].

Motivated by these considerations, as a first step we have conducted preliminary study on preparation of graphene on Si/SiO₂ substrate using mechanical exfoliation technique. We hope to obtain several potential flakes, both monolayer and bilayer graphene, for using in fabrication of graphene field effect transistors (FETs) in the future.

1.2 Objectives

To prepare several potential graphene flakes (monolayer and bilayer) on Si/SiO₂ substrate, which we will need for fabrication of graphene field effect transistors (FETs), using mechanical exfoliation technique

1.3 Scope of Research

- 1.3.1 Utilizing the technique of mechanical exfoliation to prepare graphene onto Si/SiO₂ substrate

1.3.2 Optimizing parameters that affects the yield of graphene flakes, such as baking temperature and time, cleaving process, and rubbing time

1.3.3 Identifying graphitic flakes using optical microscopy technique

1.4 Method

1.4.1 Wafer preparation

- We use Si/SiO₂ as a substrate for graphene deposition
- The Si/SiO₂ wafer into pieces about 1.5 × 1.5 cm² in size
- The coordinate grid is defined on the wafer pieces by photolithography, followed by metallization
- The wafers pieces are immersed in acetone for 10 min, rinsed in isopropanol, and blown dry with nitrogen gas
- The wafers are then baked on a hot plate at 120°C for 10 min to remove any hydration layer

1.4.2 Mechanical exfoliation

- We peel a small piece of either bulk HOPG, natural graphite, or Kish graphite with adhesive tape. When we pull away the tape away, a thin graphite layer remains adhered to it
- This thin and fresh layer of graphite is then transferred to a new piece of scotch tape with a tweezer and is cleaved by folding and unfolding the tape repeatedly until graphene flakes are spread over the tape
- We then immediately press the adhesive side of the tape containing the peeled flakes on a pre-cleaned and pre-heated chip
- We gently rub the tape on the substrate using the flat end of a Teflon tweezer for about 1-2 min, before the tape is then slowly pulled away, leaving freshly cleaved graphene flakes and graphite on the substrate
- The sample is then annealed in a tube furnace under Ar(95%)/H₂(5%) gas flow at 400°C for 1 hour with a flow rate of 1.5 L/min to remove glue residues

1.4.3 Flake identification

- We use an optical microscope to scan the entire wafer at 20× magnification, looking for flakes with a greenish tint that we then study under magnifications of 50× and 100×. The flake with the lowest contrast is assigned as monolayer graphene

More details on these steps will be discussed in Chapter 3.

Chapter 2

The Properties of Graphene

In this chapter, we provide a basic introduction of graphene and discuss some of its key physical properties. We start our discussions by studying the various allotropes of carbon, graphene being one among them that exists in the two-dimensional (2D) form. We briefly touch upon the aspects of mechanical stability in graphene and the reason why it exists in nature despite the previously held belief that it is impossible to exist in its free form. Finally we take the reader through a brief history of graphene where we discuss some of the earlier works done by eminent groups across the world, prior to the major findings reported by the Manchester group in 2004.

2.1 Carbon in Two Dimensions

Carbon is not only one of the most abundant elements of the periodic table but is also the basis for all organic chemistry and the existence of life as we know it. The ability of carbon atoms to readily form complicated structures [24] has long been known as the reason for the existence of several allotropes of this element. Among the oldest known are diamond and graphite, the allotropes that exist in three-dimensional form. Carbon nanotubes (1D) [2] and fullerenes (0D) have been discovered relatively recently [25, 26] and have caught the attention of many physicists and chemists. Ironically, these forms were theoretically studied using the 2D structure of graphene as the basis for calculations. The missing 2D structure named graphene, remained elusive for a while until finally being discovered in 2004 [5, 6]. The various forms of carbon that exist in nature as of today are shown in the Figure 2.1 below.

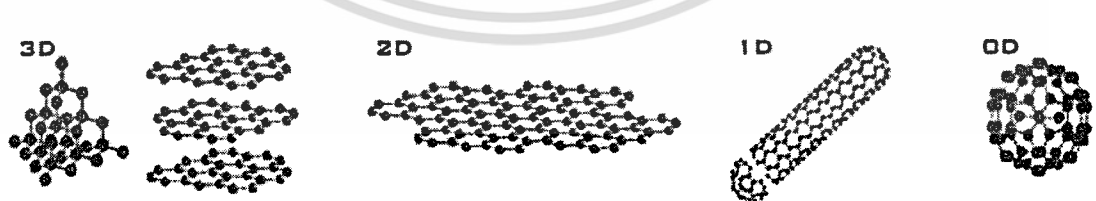


Figure 2.1: Crystal structures of the different allotropes of carbon. From left to right, three-dimensional diamond and graphite, two-dimensional graphene layer, one-dimensional carbon nanotube and zero-dimensional buckyball structure. Adopted from Ref. 13.

2.2 Two-Dimensional Stability

Graphene is known to be the first example of a 2D atomic crystal, although in principle such materials were believed to be non-existent in nature in the free-state. This was theoretically argued by Landau and Peierls nearly 70 years ago [27, 28]. According to them, crystals become thermodynamically unstable and should melt if the displacement of their atoms from their equilibrium positions exceeds a certain fraction of the interatomic spacing. For three dimensional solids these fluctuations are found to be very small and therefore allow their existence. However, the situation gets rather complicated when the dimensionality is reduced. In this limit, Landau and Peierls showed that the atomic displacements would diverge. These arguments were later supported and extended by Mermin [29], who stated that long-range order is completely absent in two dimensions and consequently dislocations are expected to appear thereby disallowing any spatial continuity over long distances. In fact, these theoretical claims were backed up by experimental observations where it was shown that thin films, in the limit of vanishing thickness, can become unstable and form segregated islands or in some cases even decompose [8, 30]. The common wisdom that 2D materials could not exist was disproved when graphene was discovered in isolated form along with other free-standing 2D crystals like boron nitride and molybdenum disulphide [5, 6]. What really took many by surprise was that, in addition to being continuous, these single-atom-thick layers of graphene exhibited high crystal quality and could be obtained on crystalline and non-crystalline substrates (see Figure 2.2(a) & 2.2(b)) [5, 8, 9], in liquid suspensions [5, 31], and also in suspended form [32]. More recent theoretical work has demonstrated that anharmonic coupling between so called bending and stretching modes can suppress the thermal fluctuations of the atoms. This in turn implies that atomically thin membranes can indeed exist in nature, however they need to be in “rippled” form so as to minimize the total free energy [33]. Such ripples (see Figure 2.2(c)) are quite commonly observed in graphene and are known to also affect the electronic properties of the material significantly [34-36].

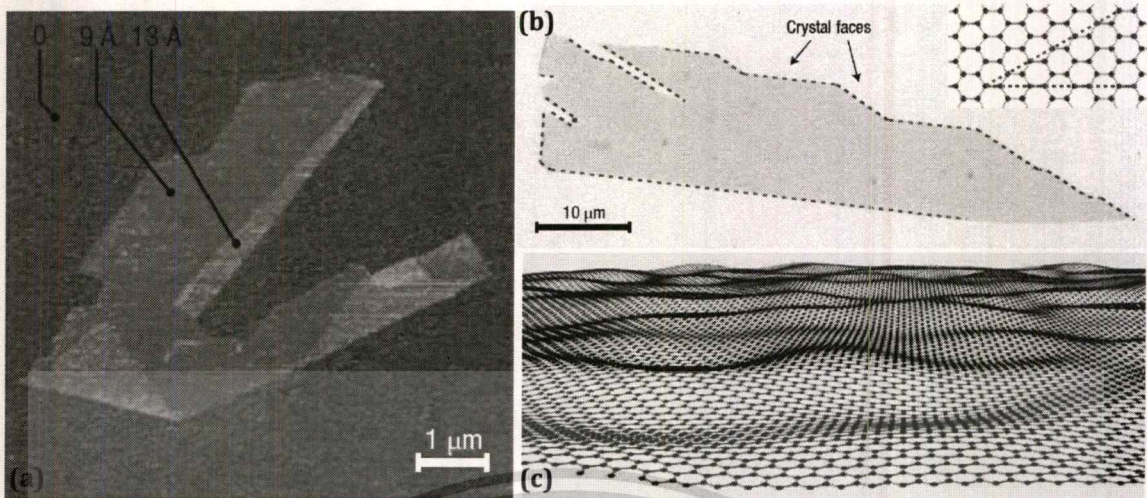


Figure 2.2: (a) Atomic force microscope (AFM) image of a single layer of graphene isolated in 2004 [13] (b) TEM image of a large area graphene sheet, (c) Graphic depicting the “rippled” structure of graphene.

2.3 A Brief History of Graphene

Although graphene has risen in popularity only in the past decade or so, following the pioneering work of the Nobel laureates Andre Geim and Kostya Novoselov, it is informative to look into the longer history of graphene research. Some of the earliest observations of this material actually date back to as far as 1859, when the British chemist Benjamin Brodie [37] discovered what he called as “graphon”, a new form of carbon which he obtained by reacting graphite with strong acids (see Figure 2.3 (a)). Today, it is known from our sophisticated knowledge of chemical science and technology that what Brodie observed more than a century ago is what we today know to be graphite oxide [38]. Following the discovery by Brodie, there were several papers describing the laminated structure of graphite oxide but the most significant contributions to this end were made in 1948 by two German physicists G. Ruess and F. Vogt, who used transmission electron microscopy (TEM) and observed flakes down to a remarkable thickness of just a few nanometers [39]. This was followed by a search for the thinnest possible graphite oxide fragments by Hanns-Peter Boehm and Ulrich Hofmann who identified some of these fragments as monolayers in 1962 [40] (see Fig. 2.3 (b)). Unfortunately, however, these remarkable discoveries received very little attention until the beginning of the twenty-first century, the main reason being that the 1962 identification relied only on “relative” TEM contrast, which in today’s scientific world would have encountered a lot of scrutiny. As a result, unambiguous identification of graphene monolayers using TEM was made only in recent years by counting the number of folding lines [41-43]

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and by understanding how the contrast depends on focusing conditions [44]. Nevertheless, Boehm and co-workers deserve due credit, for they did indeed provide the first observation of monolayer graphene. Moreover, the term “graphene” was introduced by them in 1986, by deriving it from the combination of the word “graphite” and the suffix “-ene” which refers to polycyclic aromatic hydrocarbons [12]. In 1994, they also authored the International Union of Pure and Applied Chemistry (IUPAC) report that formally defined the term graphene.

Another significant branch of pre-2004 graphene research that cannot be ignored is the work on its epitaxial growth. In the late 1970s studies of epitaxially-grown thin films had kindled huge research interest, with the growth of ultrathin graphitic films popular among these studies. These films were grown on several different substrates such as conductive metals, insulating carbides and graphite and a few of these groups even reported the successful growth of monolayer material [45] (see Figure. 2.3 (d)). Thomas Ebbeson and Hideo Hiura envisioned the idea of graphene based nanoelectronics in their 1995 paper [46]. More interesting is the fact that, prior to the efforts by the Manchester group, thin films of graphite were produced by the very same cleaving technique and their properties were studied. For instance, Heinrich Kurz’s group studied carrier dynamics in graphite obtained by cleaving with transparent tape [47] in 1990, while nanometer thickness graphite was visualized by atomic force microscopy (AFM) [46] in 1995. Scanning electron microscopy (SEM) was used to obtain images of thin graphite platelets [48] in 1999 (see Figure. 2.3 (c)), while scanning tunneling microscopy (STM) was employed to cleave and observe monolayers of graphite [49] in 2003.

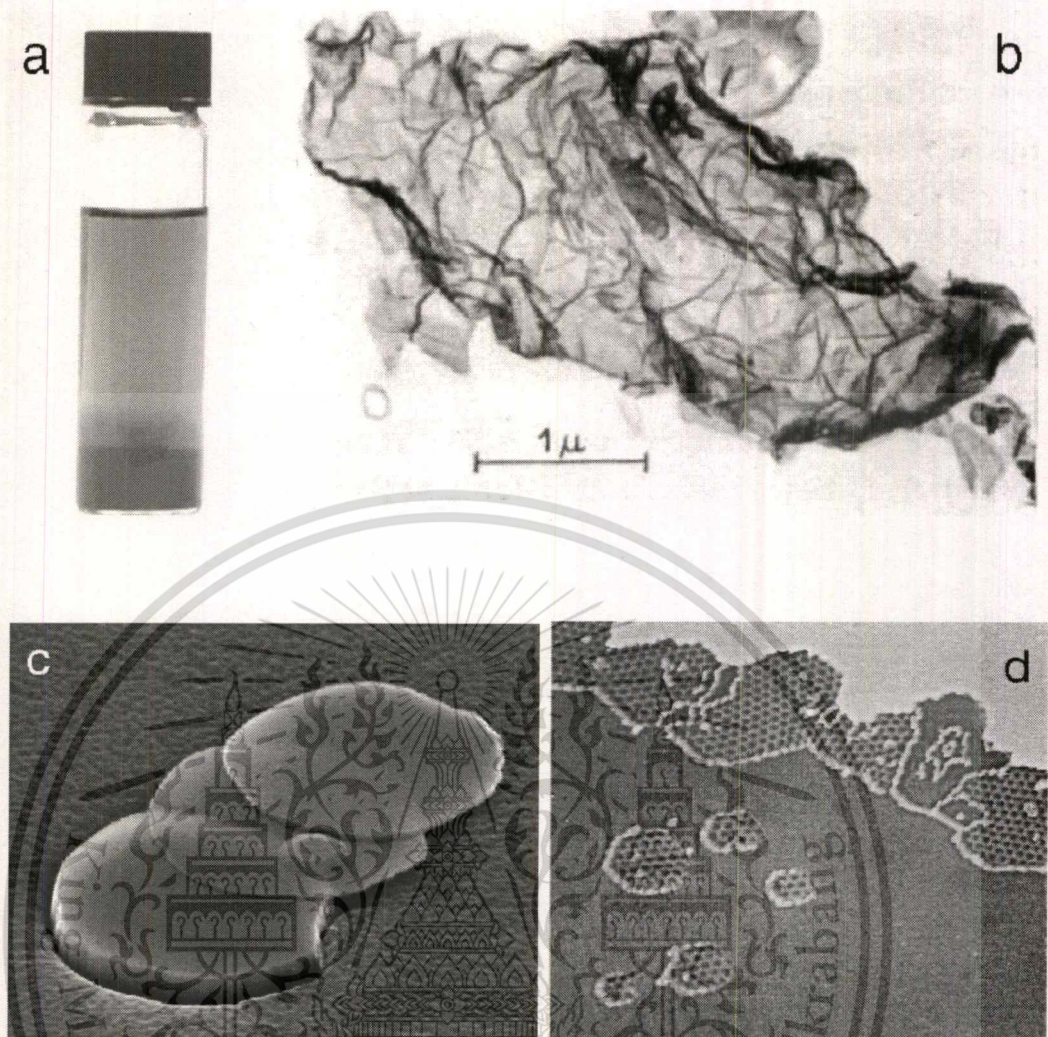


Figure 2.3: (a) Graphene as probably seen by Brodie back in 1859. Upon dissolving in water, graphite oxide separates into graphene flakes that can be found floating on the top surface. (b) TEM image of ultrathin graphitic flakes discovered by Boehm in 1962 (c) SEM image of thin graphite platelets obtained by cleavage. (d) STM image of graphene grown epitaxially on Platinum. Figures adopted from Ref. 45.

Electrical studies of these thin films were also performed. Ohashi and coworkers were able to measure an electric-field effect on 20 nm thick graphite flakes and observed resistivity changes of up to 8 percent [50]. With regards to theoretical work on graphene, the very first calculation of its band structure was carried out by Phil Wallace in 1947 [4]. The theoretical models revealed the similarities of graphene with relativistic particles and therefore graphene was seen as a condensed matter analogue of quantum electrodynamics (QED), allowing various questions in this area to be answered by studying the properties of graphene. Many of these theories found immediate use well before 2004, when carbon-nanotube research was at its peak.

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To conclude the history of graphene it is noteworthy to mention that the earlier experiments were purely observational, in that no distinguishing properties of graphene or the ultra-thin graphitic layers were reported, and hence were not sufficient to spark a graphene ‘revolution’. The interesting physics that pertains to graphene was not brought to the forefront until Geim and Novoselov’s *Science* paper was published in 2004 [5]. Research in graphene and other 2D materials began to rise exponentially at that point and is rapidly progressing even today.



Chapter 3

Methodology

Since its first demonstration by the Manchester group in 2004 [44], micromechanical cleavage – or “exfoliation” – from crystalline graphite remains the most popular approach to isolate graphene for device research. This is primarily a consequence of the high quality of the resulting graphene flakes, and the relative ease that this method affords in device fabrication. In this chapter, we describe the process of graphene deposition onto a silicon substrate by mechanical exfoliation using Scotch tape. We begin with the procedure for wafer preparation and mechanical exfoliation, before we close the chapter with flake identification process.

3.1 Wafer Preparation

We use highly *n*-doped silicon wafers, covered with a 300-nm layer of SiO₂, as the supporting substrate for graphene exfoliation. The highly doped silicon works as a back-gate electrode that allows us to apply an electric field to the finalized graphene devices. As in the case of a capacitor, the insulating SiO₂ serves as a dielectric, allowing the charge density in the graphene layer to be tuned via the back-gate voltage. The SiO₂ thickness of 300 nm is chosen in order to optimize optical contrast when inspecting graphene flakes with an optical microscope [51].

Device fabrication starts by cleaving the Si/SiO₂ wafer into pieces about 1.5 × 1.5 cm² in size using a precision diamond scribe. A coordinate grid is then defined on the wafer pieces by photolithography, followed by evaporation of 10 nm of chromium and 45 nm of gold. We prepare this grid on the substrate to allow us to determine the coordinates of subsequently-exfoliated graphene flakes, and for use as alignment markers when we make contact to the flakes. These markers are pre-designed with various symbols, on an optical mask that defines a 100 × 100 matrix. Each marker is 25 μm × 25 μm in size, while neighboring markers are spaced 200 μm from each other.

3.2 Mechanical Exfoliation

To begin with, the marker samples obtained earlier are once again cleaned with acetone and methanol in an ultrasonic bath following which they are dried in a flow of compressed nitrogen gas. The wafers are then baked on a hot plate at 120°C for 10 min to remove any hydration layer. Exfoliation of graphene onto the Si/SiO₂ substrate is then performed using adhesive tape (either 3M™ Scotch tape or Nitto™) as described extensively in the literature [52, 53]. For the purpose of this study, we

have made use of either highly oriented pyrolytic graphite (HOPG), natural graphite, or Kish graphite.

Although the exfoliation procedure cannot be standardized, it can nonetheless be illustrated by considering the case where we peel a small piece of bulk HOPG with Scotch tape. When this piece is pulled away from the tape, a thin graphite layer remains adhered to it. This thin and fresh layer of graphite is then transferred to a new piece of scotch tape with a tweezer and is cleaved by folding and unfolding the tape repeatedly. By doing so, graphene flakes are peeled off from the graphite and are spread over the tape. We normally repeat this peeling process several times until a light grey shade of graphite can be seen on the tape. This ensures minimum thickness of graphite pieces. The adhesive side of the tape containing the peeled flakes is then immediately pressed on a pre-cleaned and pre-heated chip. If the tape is not applied within a minute after pre-heating, the yield of flakes will be reduced, presumably due to accumulated moisture on the substrate. We typically obtained only one or two monolayer flakes when the substrate was hydrophobic. To maximize the adhesion of graphite, we gently rub the tape on the substrate using the flat end of a Teflon tweezer for about 1-2 min, since this increases the chance of finding usable graphene flakes. The tape is then slowly pulled away, leaving freshly cleaved graphene flakes and graphite on the substrate, as can be seen in Figure 3.1.

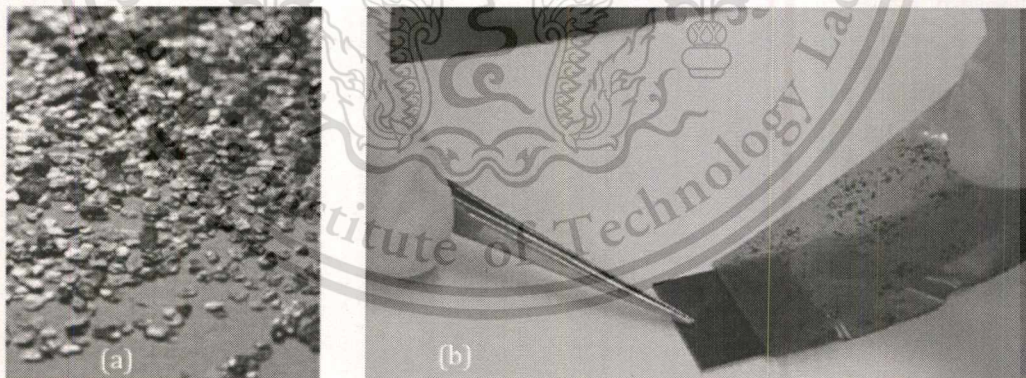


Figure 3.1: (a) Tiny pieces (~ 2 mm) of Kish graphite as purchased from the supplier.
(b) Peeling of the graphene laden blue tape after transfer.

While pulling away the tape, a competition arises between the adhesive interaction at the graphite/SiO₂ interface, and the van der Waals bonding between the graphene layers [54]. Since the adhesion between the bottommost graphene layer and the underlying SiO₂ is strong [55], it is expected to screen the van der

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Waals bonding between graphene layers adjacent to this interface. Consequently, thin graphene layers, ranging in thickness from monolayer to a few layers, can be favorably attached to the SiO₂ substrate.

A baking step is subsequently used to improve the adhesion of graphene to the substrate. Without this step, we noticed that flakes, especially large ones, sometimes detach from the substrate when dipped in acetone or during the other stages of device fabrication. Since cleaning in acetone is insufficient to remove glue residues, we also anneal the sample in a tube furnace under Ar(95%)/H₂(5%) gas flow at 400°C for 1 hour [19, 56]. The gas flow is maintained with a flow rate of 1.5 L/min throughout the heating and cooling process.

After exfoliation, we store the samples in a vacuum box if further processing is not to be performed immediately, since this allows us to avoid contamination. If kept in ambient air for a prolonged period, graphene will become contaminated with moisture and other impurities.

4.2 Flake Identification

Some of the most frequently used methods for the identification of graphene flakes are optical microscopy, Raman spectroscopy and atomic force microscopy (AFM). Of these the optical method is most popular as it is simple and reliable technique. Graphene on a proper choice of substrate can be viewed under an optical microscope, due to the different optical contrast between graphene on that substrate and the bare substrate itself. As noted already, a 300-nm thick SiO₂ layer on top of a Si substrate is most commonly used since the resulting contrast is high enough to allow monolayer graphene flakes to be observed under a conventional optical microscope (using white light without any filter) [51, 57-59]. We also make use of the 300-nm SiO₂ layer in this work, allowing us to take advantage of optical microscopy when selecting candidate flakes.

With an optical microscope we can scan over the surface of our cm² sized wafer and identify promising graphene flakes in less than an hour. Even small flakes, less than 3 μm, can be identified by this method. We start our search by scanning the entire surface at 20× magnification, looking for flakes with a greenish tint that we then study under magnifications of 50× and 100×. Monolayer graphene will appear as a light-purple shade under such higher magnification. Once potential flakes have been selected, we record images of them with a microscope-mounted camera at these different magnifications (20×, 50× and 100×).

Chapter 4

Results and Discussions

In this chapter we present the results obtained from our study on preparation of graphene on silicon substrate using mechanical exfoliation technique, starting with showing the result of the substrate after the preparation procedure. We then proceed to present our graphene flakes obtained from mechanical exfoliation technique using different types of graphite. We also comment on the influence of the type of graphite on the yield of graphene flakes in this section. Finally, we close the chapter with the discussion on how we identify suitable graphene flakes by visual inspection.

4.1 Substrate for graphene deposition

As mentioned in Chapter 3, prior to the exfoliation of graphene onto SiO_2 substrate, it is essential to have a marker grid that will aid in identification of the precise location of the flakes. In order to do this we employ a standard procedure of optical lithography in which a pre-defined set of markers printed on an optical mask is used to transfer the pattern onto the substrates. A metal stack consisting of 3 nm of chromium (Cr) and 45 nm of gold (Au) is deposited onto the substrates using an e-beam evaporator. The Cr acts as an adhesive layer for the main contact material (Au), whose thickness is optimized to provide the high visibility needed when performing subsequent processing steps in an electron microscope. An optical image of a substrate with the alignment markers is shown in Figure 4.1.

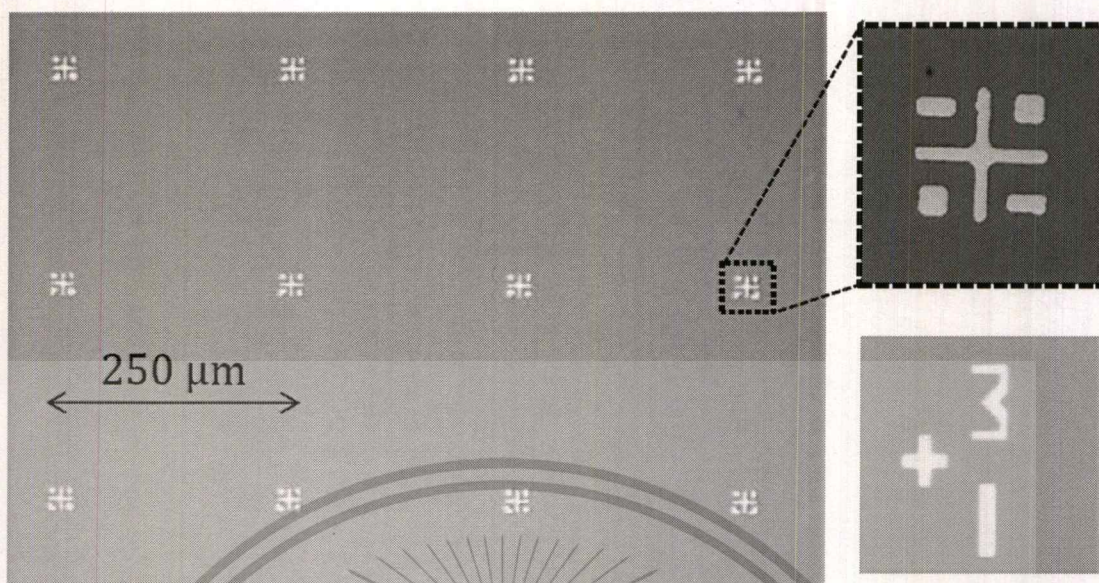


Figure 4.1: Optical image of the marker grid consisting of equally spaced markers that are each ~ 25 microns in lateral dimension. The upper inset shows a magnified view of a single marker where individual symbols distinguish each marker from the other. Alternatively, for easier searching and identification markers containing row and column numbers (as shown in the lower inset) can also be fabricated using a sharper lithography method such as e-beam. The optical mask aligner system available to us can achieve reliable resolution down to ~ 2 microns.

4.2 HOPG vs. Natural vs. Kish Graphite

While the HOPG is of very high crystalline quality [60], the flakes obtained from it tend to be rather narrow and “ripped”. A typical result of the mechanical cleavage from HOPG crystal is shown in Figure 4.2(a). Moreover, they are frequently found to be close to large pieces of graphite, which can lead to difficulties in subsequent lithographic processing. While natural graphite is known to have larger grain size and results in larger graphene flakes [60], we did not obtain results very different than those shown for HOPG. In addition, natural graphite is known to contain many defects, dislocations and chemical impurities, all of which will negatively affect the resulting transport properties. For these reasons, a better choice for experimental studies is Kish graphite, which provides much larger graphitic flakes that are of higher quality than those exfoliated from natural graphite [60]. By using Kish graphite, we not only obtained larger flakes (in the range of tens of μm , compared to a few μm produced by HOPG) but also achieved a higher yield. Figure

4.2(b) shows an example of graphene flakes obtained from Kish graphite. The large area of these flakes is noteworthy.

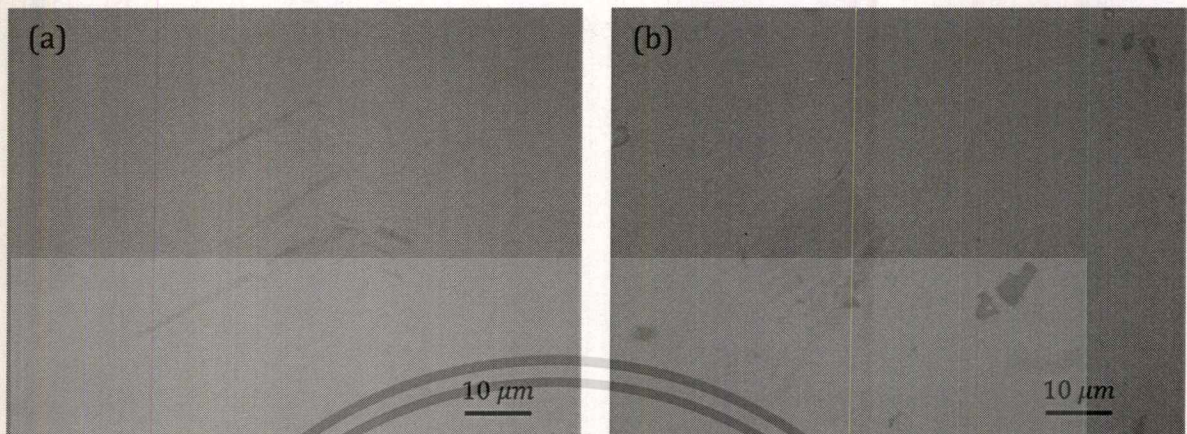


Figure 4.2: Optical images of graphene on Si/SiO₂ substrate obtained from (a) HOPG and (b) Kish graphite.

4.3 Monolayer vs. Bilayer vs. Many-layer Graphene

Graphitic flakes deposited onto the Si/SiO₂ substrate by micromechanical exfoliation are generally comprised of a mixture of monolayer and multilayer flakes that have different electronic properties. In this work we use optical microscopy, the most commonly used method, to determine the number of layers of graphitic flakes. A typical result of the exfoliation as seen under an optical microscope is shown in Figure 4.3(a), where thick graphite pieces appear as a bright white color and thinner but multilayered pieces have a strong blue color. At a low magnification (20x) such as the one shown in Figure 4.3(a), it is impossible to see any monolayer flakes. However, as we approach a higher magnification (100x) monolayer flakes can be easily detected as they appear as an almost transparent (lowest contrast) layer revealing the substrate behind it. At times it can be tricky to distinguish between monolayer and bilayer samples but this process becomes quite reliable with experience. Figure 4.3(b) shows an example of monolayer, bilayer and multilayered graphene identified by using optical microscopy. As already mentioned earlier, such distinct optical contrast is made possible by choosing the optimum oxide thickness of 300 nm [51]. However, owing to its near-transparent nature monolayer flakes can be very difficult to find if the raster scanning is not performed very slowly and vigilantly.

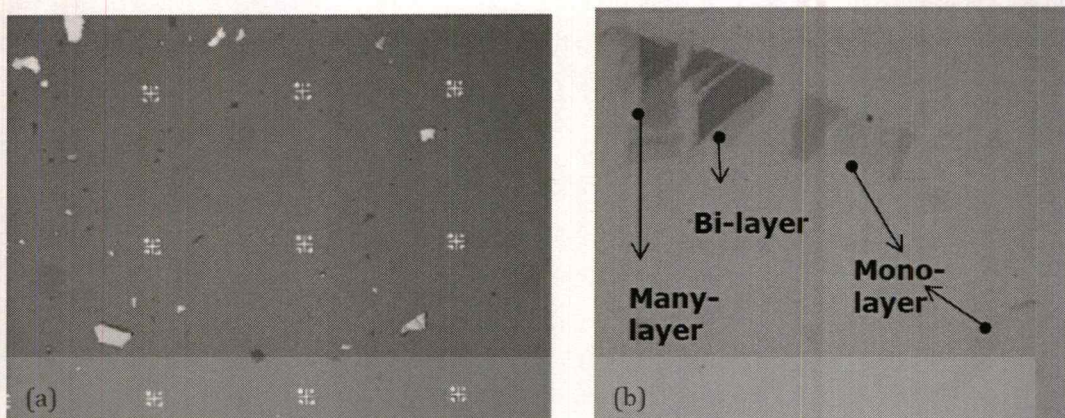
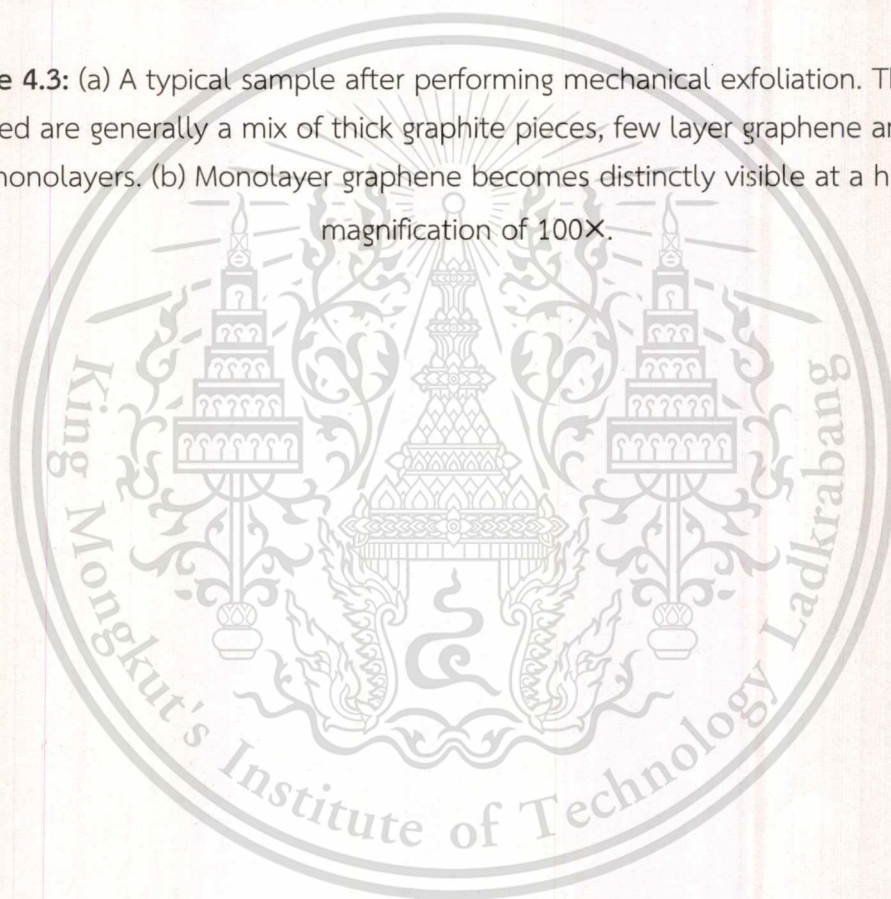


Figure 4.3: (a) A typical sample after performing mechanical exfoliation. The flakes obtained are generally a mix of thick graphite pieces, few layer graphene and hidden monolayers. (b) Monolayer graphene becomes distinctly visible at a higher magnification of 100X.



Chapter 5

Summary and Outlook

In the course of this work, we have studied the technique of micro mechanical cleavage or exfoliation from crystalline graphite and used it to prepare graphene flakes onto Si/SiO₂ substrate. This exfoliation technique is the most popular approach to isolate graphene for device research due to the high quality of the resulting graphene flakes, and the relative ease that this method affords in device fabrication. By using this technique, we were able to obtain several potential flakes, both monolayer and bilayer graphene, that can be used for fabrication of graphene FET in the future. The number of layers is determined by using a simple and reliable optical microscopy technique. The key findings of our experimental studies were as follows:

- By using exfoliation technique, we were able to prepare several graphene flakes on highly *n*-doped silicon wafers, covered with a 300-nm layer of SiO₂
- A pre-defined marker grid on the substrate is needed for identification of the precise location of the flakes
- Pre-heating the substrate at high temperature (~150 °C) helps to improve the adhesion of graphene to the substrate and hence the yield of graphene flakes
- The flakes obtained from HOPG tend to be rather narrow and “ripped”, and frequently are found to be close to large pieces of graphite, which can lead to difficulties in subsequent lithographic processing
- Although natural graphite is known to have larger grain size and results in larger graphene flakes, we did not obtain results very different than those shown for HOPG
- Kish graphite provides much larger graphitic flakes that are of high quality and so it is a better choice for experimental studies
- We can determine the number of layers of graphene flakes easily and reliably using optical microscope with a proper choice of substrate, which is a 300-nm thick SiO₂ layer on top of a Si substrate
- Using the difference in optical contrast between graphene on that substrate and the bare substrate itself, the flake with the lowest contrast is assigned as monolayer graphene

Chapter 6

Output

This grant has been utilized for the study of graphene deposition onto a Si/SiO₂ substrate using micromechanical exfoliation the result of which is included in the following publications:

- [1] G. He, K. Ghosh, U. Singiseti, H. Ramamoorthy, R. Somphonsane, G. Bohra, M. Matsunaga, A. Higuchi, N. Aoki, S. Najmaei, Y. Gong, X. Zhang, R. Vajtai, P. M. Ajayan, and J. P. Bird, "Conduction Mechanisms in CVD-Grown Monolayer MoS₂ Transistors: From Variable-Range Hopping to Velocity Saturation", *Nano Lett.* **15**, 5052 (2015)
- [2] D. K. Ferry, R. Somphonsane, H. Ramamoorthy, and J. P. Bird, "Plasmon-mediated energy relaxation in graphene" *Appl. Phys. Lett.* **107**, 262103 (2015)
- [3] D. K. Ferry, R. Somphonsane, H. Ramamoorthy, and J. P. Bird, "Energy relaxation of hot carriers in graphene via plasmon interactions" *J. Comput. Electron.* **15**, 144-153 (2016)
- [4] B. Liu, R. Akis, D. K. Ferry, G. Bohra, R. Somphonsane, H. Ramamoorthy, and J. P. Bird, "Conductance fluctuations in graphene in the presence of long-range disorder" *J. Phys.: Condens. Matter* **28**, 135302 (2016)
- [5] H. Ramamoorthy, R. Somphonsane, J. Radice, G. He, C.-P. Kwan, and J. P. Bird "Freeing" Graphene from its Substrate: Observing Intrinsic Velocity Saturation with Rapid Electrical Pulsing" *Nano Lett.* **16**, 399–403 (2016)
- [6] G. He, H. Ramamoorthy, C.-P. Kwan, Y.-H. Lee, J. Nathawat, R. Somphonsane, M. Matsunaga, A. Higuchi, N. Aoki, Y. Gong, X. Zhang, R. Vajtai, P. M. Ajayan, and J. P. Bird "Thermally-Assisted Nonvolatile Memory in Monolayer MoS₂ Transistors", *Nano Lett.* **16**, DOI: 10.1021/acs.nanolett.6b02905 (2016).
- [7] R. Somphonsane, H. Ramamoorthy, G. He, J. Nathawat, C.-P. Kwan, Y.-H. Lee, J. Fransson, and J. P. Bird "Evaluating the contribution of impurities and phonons to the resistivity of graphene by differential-conductance mapping", *Phys. Rev. B* (2016, under revision)

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Appendix

Conductance fluctuations in graphene in the presence of long-range disorder

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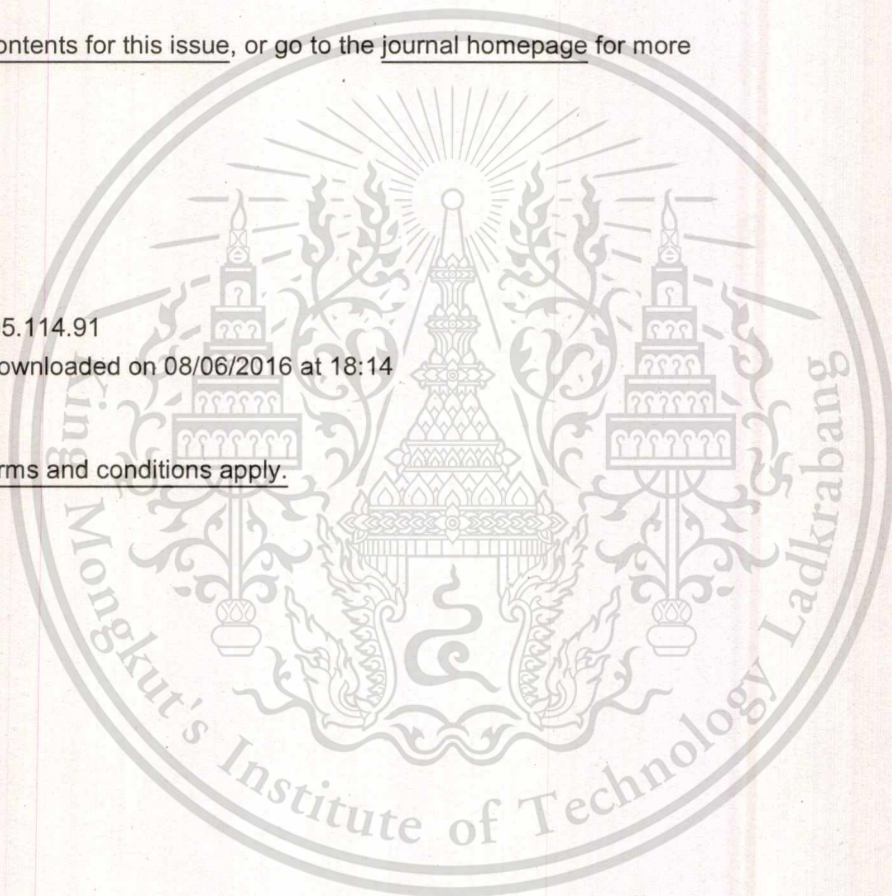
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Conductance fluctuations in graphene in the presence of long-range disorder

Bobo Liu¹, Richard Akis¹, David K Ferry¹, Girish Bohra²,
Ratchanok Somphonsane³, Harihara Ramamoorthy² and Jonathan P Bird²

¹ School of Electrical, Computer, and Energy Engineering, Arizona State University, Tempe, AZ 85287, USA

² Department of Electrical Engineering, the University at Buffalo, Buffalo, NY 14260, USA

³ Department of Physics, King Mongkut's Institute of Technology Ladkrabang, Bangkok 10520, Thailand

E-mail: ferry@asu.edu

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Abstract

The fluctuations in the conductance of graphene that arise from a long-range disorder potential induced by random impurities are investigated with an atomic tight-binding lattice. The screened impurities lead to a slow variation of the background potential and this varies the overall potential landscape as the Fermi energy or an applied magnetic field is varied. As a result, the phase interference varies randomly and leads to fluctuations in the conductance. Recently, experiments have shown that an applied magnetic field produces a remarkable reduction in the amplitude of these conductance fluctuations. We find qualitative agreement with these experiments, and it appears that the reduction in magnetic field of the fluctuations arises from a field induced smoothing of the conductance landscape.

Keywords: graphene, conductance, fluctuations, transport

(Some figures may appear in colour only in the online journal)

1. Introduction

The phenomenon of fluctuations in the conductance of a disordered metal or semiconductor at low temperature has been studied for many years [1]. In such systems, it is observed that the conductance will exhibit such fluctuations as the Fermi energy, or an applied magnetic field, is varied. It is presumed that this arises from the variations in the local potential landscape and changes in the electron wave interference as these variations are introduced. Recently, these fluctuations have been observed in monolayer and bilayer sheets of graphene [2–8]. In some of these experiments, it was found that the fluctuations observed [9] for varying magnetic field were smaller than for varying the gate voltage (energy) and decreased as the density increased [7–9]. These behaviors are contrary to the idea of ergodicity⁴ which arose from early diagrammatic theories for fluctuations in disordered materials [10], and from recent theories for graphene in the metallic regime of

high conductivity [11–13]. However, such behavior has been observed in some simulations of the fluctuations in normal semiconductors [14, 15]. It was originally suggested that this reduction of the amplitude with magnetic sweeps was due to the breaking of microscopic symmetries connected with the detailed electronic structure of the graphene itself. However, some of these symmetries could be expected to be broken by the in-plane magnetic field as well, and this was not observed [9].

Motivated by the experimental situation described above, we have undertaken simulations of quantum transport in graphene to investigate the connection of the conductance fluctuations (CF) to the nature of the disorder potential. With a very short-range atomic potential used to simulate the disorder, no decay of the fluctuation amplitude with magnetic field was observed [16, 17]. In these earlier calculations, an atomic-basis tight-binding lattice also was used to study the structure and transport in a graphene nanoribbon. The random potential was introduced via a local potential in the Anderson model [18], an approach that we have used successfully for finite difference lattices in the past [14, 15]. But, this potential appears

⁴ The term ergodic in this context would mean that the amplitude would be independent of which physical variable (energy, magnetic field, sample, etc) was varied.

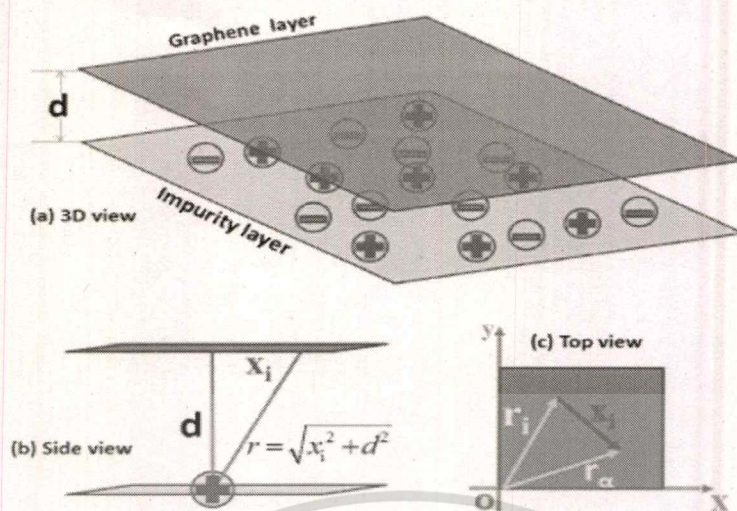


Figure 1. A typical schematic of remote charge impurity. (a) 3D view: the distance between graphene layer and impurity layer is d and the positive and negative impurity charges are randomly located within the impurity layer. (b) Side view: the distance between the impurity charge and the point where potential is calculated is r . (c) Top view: the distance between the impurity charge and the point where potential is calculated is obtained by $|r_a - r_i|$.

to be too short range for the atomic-basis lattice, a point we return to later. On the other hand, it has been pointed out that a long-range potential, appropriate to describe the potential induced by remote impurities trapped beneath the graphene sheet, is an important approach [13]. Hence, we have used here a long-range potential which derives from the impurities trapped below the graphene sheet in/on the silicon dioxide substrate. In the following, we briefly first describe our simulation approach, and then discuss how the results give agreement with available experimental data.

2. Impurities and the simulation approach

One of the critical factors for fluctuations is the types of extrinsic scattering sources that can exist in graphene ribbons. Although the scattering scenario is a complicated picture with contributions from adsorbates, ripples, corrugations and remote charged impurities, etc, here we consider only the long-range Coulomb potential arising from the random distribution of charged impurities. The resulting disordered potential will be the main scattering mechanism, at least as based upon experimental evidence [19–21]. Utilizing the superposition principle, the total electric potential of all charged impurities is simply the sum of potentials due to the individual charged impurities, which can be described in a form [21]

$$V_\alpha = \sum_{i=1}^{N_i} \frac{1}{4\pi\epsilon_0\epsilon_r} \frac{e_i}{\sqrt{|r_\alpha - r_i|^2 + d^2}} \exp\left(-\xi \cdot \sqrt{|r_\alpha - r_i|^2 + d^2}\right). \quad (1)$$

Here, N_i is the number of the impurities, ϵ_0 is the vacuum permittivity, and the relative permittivity is ϵ_r . e is the elementary electric point charge which may have a different sign for different impurities. The exponential term $\exp\left(-\xi \sqrt{|r_\alpha - r_i|^2 + d^2}\right)$ represents the modification of the

potential by both the screening and the set-back of the impurity from the graphene sheet. Here, ξ is the screening coefficient [22]. The distance between the point charge and the position where potential $V(r)$ is calculated is the argument of the square root term in the exponential. We use a constant screening, which has been shown to be comparable to a density-dependent screening in terms of fluctuation amplitudes [13]. Important parameters are illustrated in figure 1.

We use an atomic-basis tight-binding model to study the transport with such a long-range potential. The atoms are treated in slices, which contain two atomic rows in order to preserve the slice symmetry [16]. The graphene nanoribbon sample used in our calculation has a width of 199 atoms and a length of 200 columns (100 slices). So the area of the sample will be around 24.3 nm (width) \times 42.4 nm (length) = 1030.32 nm² (we refer to this as the ‘normal’ size). We consider a density of impurities of $3 \times 10^{12} \text{ cm}^{-2}$. This means that the number of charged impurities in the area of the ribbon is 31. Then, these impurity charges are randomly distributed throughout the area of the graphene nanoribbon. In addition, the distance between the graphene layer and the impurity charge layer is set to $d = a_0$ and the screening coefficient is $\xi = 1/(10 \times a_0)$, corresponding to a screening length of $10 \times a_0$. Here, $a_0 = 0.142 \text{ nm}$ is distance between two adjacent carbon atoms in the nanoribbon. A typical remote charge impurity potential landscape is shown in figure 2. This potential distribution is changed for each simulation run.

This random potential provides the landscape in which the transport is computed by the above prescriptions. Because the conductance fluctuations are random, they will change with each implementation of the impurity potential. Hence, we use several samples, each of which has the same impurity density of $3 \times 10^{12} \text{ cm}^{-2}$ but a different distribution pattern for the impurities. From these impurities, the screened potential is determined and this potential is then mapped onto the atomic lattice to produce the on-site potentials, one example of which

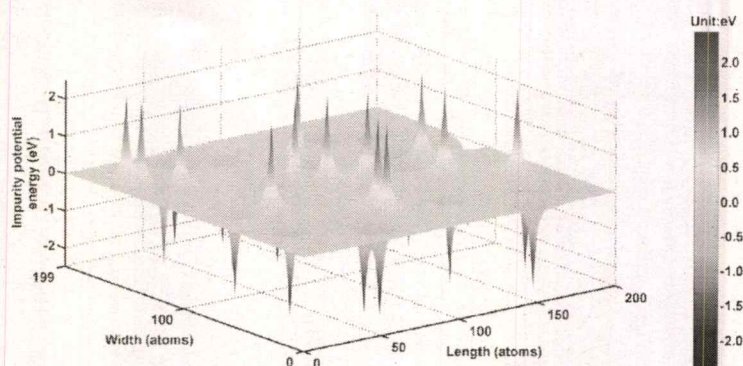


Figure 2. The potential resulting from one implementation of the remote impurity distribution. Here, the positively charged impurities are the larger quantity and the Fermi level would sit about 65 meV above the Dirac point in the absence of any gate bias.

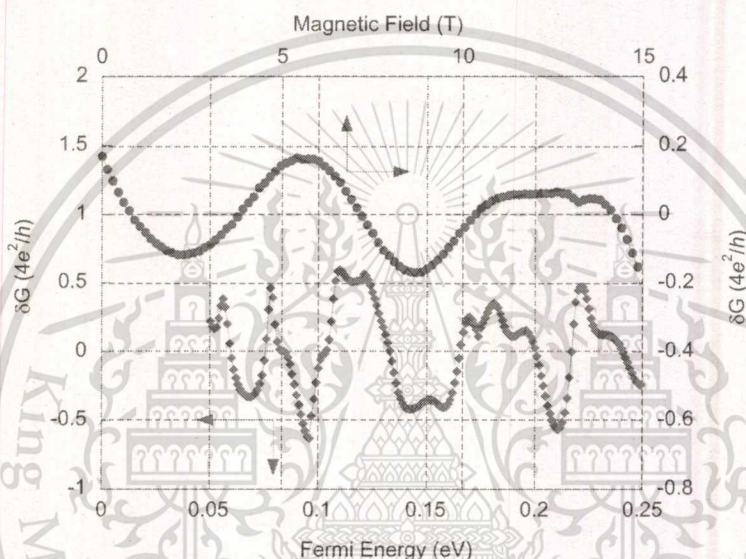


Figure 3. Typical fluctuations in the conductance observed for a magnetic field sweep (upper curve and right scale) and for a Fermi energy sweep (lower curve and left scale).

is shown in figure 2. As mentioned, the normal lattice has a width of 199 atoms and a length of 200 atomic columns, but we have considered other sizes. The position of the Fermi level, relative to the Dirac point, will depend upon the fraction of the impurities that are donors (or acceptors). Transport through this nanoribbon is computed with a stabilized variant of the scattering matrix formalism subject to magnetic field strengths applied perpendicular to the plane of the graphene sheet. The details of the computational technique have been given earlier [16, 17]. The transmission computed with this approach is then used in the multi-channel Landauer formula.

3. Results

When the conductance is computed for various values of the Fermi energy or an applied magnetic field, it is observed to have fluctuating values due to the disorder. In figure 3, we plot typical traces for the fluctuations that are observed in both a Fermi energy sweep and a magnetic field sweep (here, a background conductance has been removed to better illustrate the fluctuations themselves). Although the two curves appear to

have similar amplitudes, please note that the scale on the left, for the Fermi energy sweep, is $2.5 \times$ larger, so that the fluctuation amplitude for this latter sweep is correspondingly some $2.5 \times$ larger than that of the magnetic field sweep. In addition, the magnetic field sweep displays a rather slow variation with magnetic field; indeed, this variation is much slower than that observed in GaAs [15]. It is not exactly clear what leads to this slow variation in magnetic field, but it may be connected with the peculiar band structure and density of states in graphene, where the cyclotron energy scales as the square root of the magnetic field rather than linear in magnetic field as in GaAs [23]. However, we showed in [15] that the correlation magnetic field in GaAs was about one flux quantum through the sample. For these samples, that corresponds to a magnetic field of 3.8 T, and could explain the slow variation in magnetic field. In the main panel of figure 4, we plot the RMS amplitude (δG_{rms}) of the CF induced by the Fermi energy sweeps, undertaken in the presence of various static magnetic fields oriented normal to the graphene plane. In the inset to this figure, we also plot the CF as a function of the Fermi energy for two different values of the magnetic field. It can clearly be seen that the amplitude of the fluctuation is significantly smaller with

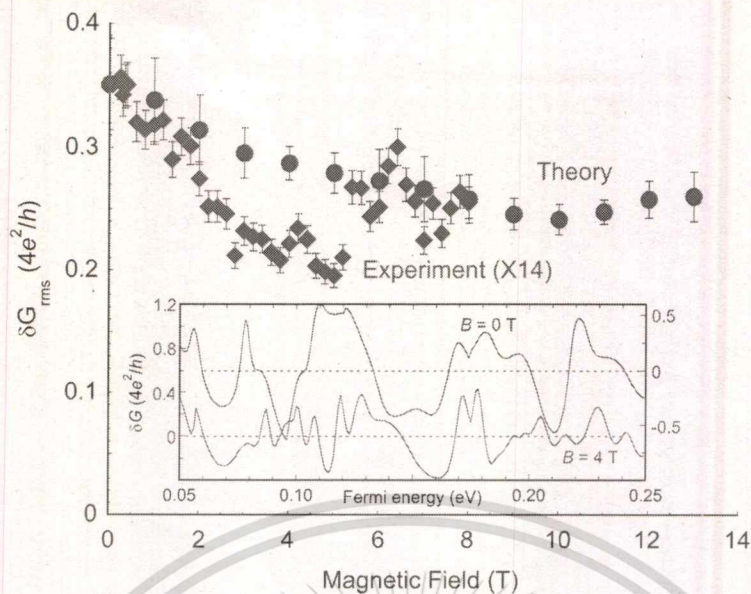


Figure 4. Amplitude of the conductance fluctuations obtained with a sweep of Fermi energy (or gate voltage in experiment) for our calculations (blue circles) and from the experiments of [9] (red diamonds). The latter has been multiplied by a factor of 14 to account for averaging over phase coherent regions in the experiment. The inset shows actual fluctuations versus Fermi energy for two different magnetic fields.

the presence of the magnetic field. Also plotted in the same figure are the experimental data from [9], which have been multiplied by a factor of 14. In the experiments, studies of the correlation function of the fluctuations observed in magnetic field sweeps were used to estimate the phase coherence length l_ϕ , which was found to be about 200 nm [9]. This distance is much smaller than the size of the sample over which the measurements were made, and smaller than the estimated thermal diffusion length. Hence, it is expected that the fluctuations observed experimentally should be considerably smaller than the theory due to statistical averaging of many phase coherent regions. The averaging length L in the measurements is about 2 μm . For the conditions discussed here, the reduction in amplitude is expected to be about $[24]\sqrt{6}(l_\phi/L)^{3/2}$, which suggests that the experimental values should be smaller than the zero-temperature calculation by a factor of 12.9. Here, we focus upon a qualitative comparison with the experiment. Hence, we have arbitrarily used a factor of 14 in figure 4 to match the experimental and theoretical data at zero magnetic field, as this gives agreement for the values of theory and experiment at this point.

Referring to the field-induced reduction in the magnitude of the computed fluctuations in figure 4, we may conclude that this is not due to any symmetry breaking process (such as that due to the breaking of time reversal symmetry or the lifting of spin or valley degeneracy), since such mechanisms are not included in our calculations. Indeed, spin degeneracy should be lifted also when the magnetic field is applied in the plane of the graphene, but no such evidence of such symmetry breaking was found in the study of [9]. So, there must be another explanation for the magnetic-field induced reduction in the amplitude of the conductance fluctuations. We speculate that it may be related to where Büttiker has shown that, under certain conditions, and in a large perpendicular magnetic field,

the latter can lead to a suppression of both elastic and inelastic back scattering [25]. As edge states begin to form in the magnetic field, most of the perceived resistance is found at the current contacts, and the forward and backward edge states are unequally occupied. Reflection from impurities cannot induce back scattering of the topologically-protected edge states, and the magnetic field also leads to a reduction in the number of channels that actually propagate through the sample. The result of these two effects should be to suppress the amplitude of the fluctuations in the conductance, and we speculate that it is this behavior that is seen in figure 4. However, we caution that the edge states are nowhere near fully formed in these samples. The fact that the experimental data falls more rapidly than our calculations suggest that the reduction of back scattering may well be more effective in the presence of additional scattering mechanisms, and when there are several phase coherent regions that lead to the averaging seen in the experimental data.

In figure 5, we plot the amplitude of the conductance fluctuations observed in experimental and simulated magnetic field sweeps as a function of the electron density. The calculations are actually done as a function of the Fermi energy, while the experiments are performed by varying the back gate voltage. We have converted both variations to one in terms of carrier density, so as to have a common parameter for comparison. Again, the experimental data has been multiplied by a factor of 14 to account for the multiple phase coherent area averaging [24]. A similar reduction in the amplitude of the fluctuations with density has been seen for both gate voltage sweeps and magnetic field sweeps [7]. It may be seen from this figure that the amplitude of the fluctuations is smaller than that in figure 4 for the Fermi energy sweeps. The difference is of the order of a factor of 3, which has also been seen in simulations for conductance fluctuations in GaAs [15]. This is

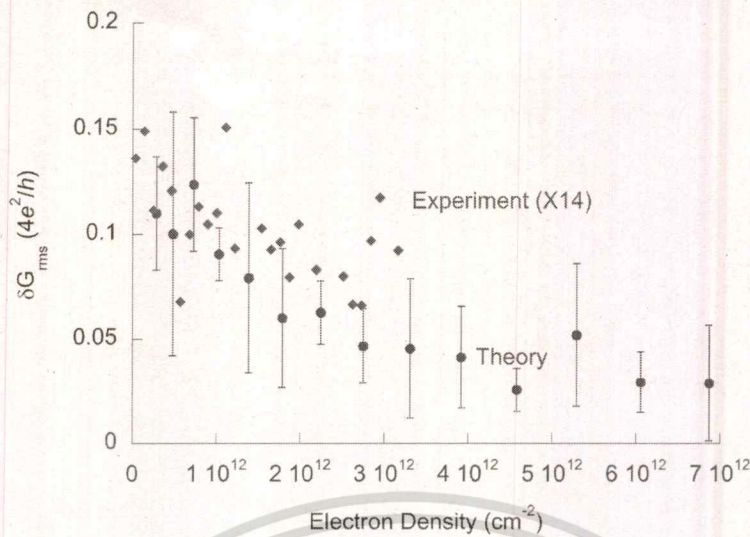


Figure 5. The average value of the conductance fluctuations that arise in a magnetic field sweep as a function of the density. The experimental data (red diamonds) has been multiplied by a factor of 14 (discussed in the text).

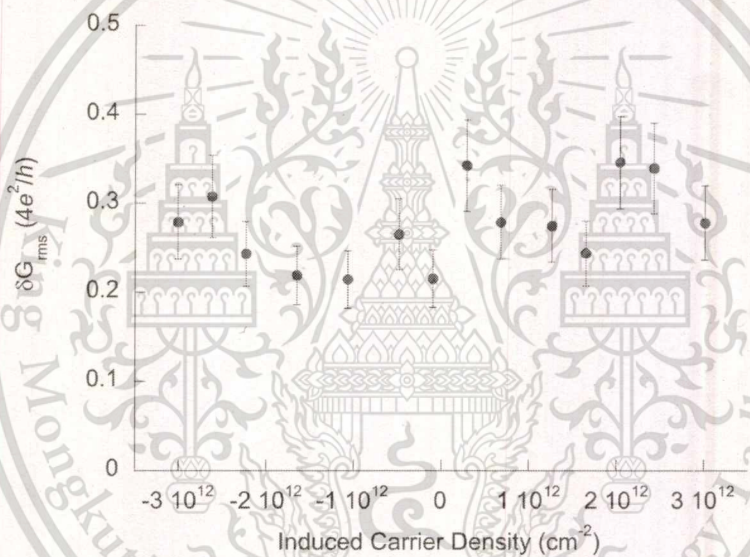


Figure 6. The conductance fluctuations observed for a fixed Fermi energy sweep on the electron side as a function of the net charge induced solely by the impurities themselves.

a clear indication that the CF do not possess any ergodic properties, as such a property would lead to equal amplitude fluctuations for different perturbations, such as sweeps in energy versus sweeps in magnetic field, or for different samples. Such a difference has also been seen in studies of $1/f$ noise in mesoscopic structures, where it is thought that this noise arises from similar interference effects as the conductance fluctuations [26]. In addition, it may be observed that the amplitude of the computed fluctuations decreases somewhat as the carrier density is raised, behavior that is captured in experiment also. This might be explained in the experiments by a decreasing phase breaking time at higher densities [27, 28], which would lead to a reduction of the phase coherence length. But, this cannot be the case in the theory, as the phase coherence length is assumed to be set by the simulation size. The observation, in both experiment and theory, of the density-dependent

variations in the fluctuation amplitude points, once again, to the non-ergodic character of these fluctuations.

We have also investigated the role played by the net charge of the impurity distribution. In figure 6, we plot the conductance-fluctuation amplitude as a function of the net impurity charge, or the induced carrier density in the graphene sheet as a result of the impurity charge. That is, when the impurities are all negatively charged, a hole concentration of $3 \times 10^{12} \text{ cm}^{-2}$ is induced in the graphene and the Fermi energy will sit below the Dirac point in energy. With only positive impurity charge, the reverse is observed. However, the Fermi energy sweeps in figure 5 are always on the electron side over the range 50 to 250 meV, or for an actual electron density that varies from roughly 2.9×10^{11} to $7.2 \times 10^{12} \text{ cm}^{-2}$. In the experiment, of course, the required gate voltage varies, and the actual carrier density depends upon the voltage at which the Dirac point is

located [9]. The background charge indicated in figure 5 is the charge that shifts the Fermi energy (in the absence of gate voltage) away from the Dirac point. The difference that arises as the net impurity charge is varied is that the total charge in the system (carriers plus impurities) will vary. While the variation is too large to draw much of a conclusion, there does seem to be a slightly higher fluctuation when the induced charge is electrons. However, in the experiment, one must face the presence of the puddle state in which both electrons and holes are present with the Fermi energy near the Dirac point [29–31]. These puddles can form self-consistently and are of the order of 5–7 nm in extent [31, 32]. In the simulations done here, there is no puddle state, as the simulation does not include the many-body effects which would lead to the formation of these puddles. So, in the presence of the puddles, it would not be clear what the role of the exact net impurity density would be, nor would it be clear what the role of the additional random potential on the conductance fluctuations would be. However, it should be pointed out that an increase of the amplitude of the fluctuation near the Dirac point is observed in some cases [7], although a reduction in the amplitude of the fluctuations near the Dirac point also has been reported [33, 34] as has a reduction in the $1/f$ noise [26]. We do not see this in the simulations, and such a increase/reduction may be a natural result of the vagaries of the puddle state and its stability.

4. Discussion

Here, we have shown that the presence of a long-range potential, arising from the charged impurities lying between graphene and its substrate, may induce fluctuations due to quantum interference in the graphene conductance. We find that this long-range potential allows the carriers to respond to the presence of a magnetic field normal to the graphene sheet, something that was not observed in the case of a very short range atomic potential [17]. Indeed, for a short-range atomic potential, we find that there is no variation in the fluctuation amplitude in the presence of a magnetic field [17], which is quite similar to the experimental case of having the magnetic field in the plane of the graphene [9]. But, with the magnetic field oriented normal to the graphene, we find that the amplitude of the conductance fluctuations varies with magnetic field and density in a qualitatively similar manner as that observed in experiments of [9], confirming the idea that the quantum interference in graphene does not exhibit the ergodic character typically assumed to describe quantum interference. One question is whether this magnetic field dependence is observed in other semiconductors, such as GaAs. We have examined this question, and find that the answer is yes (which will be discussed further elsewhere).

Overall, this study confirms the idea that the theoretical framework developed several decades ago, in which mesoscopic transport in disordered material is described as ergodic is not applicable. Indeed, an alternative analytical theory of CF in monolayer graphene, developed by Kharitonov and Efetov [35] demonstrates that the fluctuations can have different universal values for different disorder type. We have

shown in both normal materials, such as GaAs [14, 15], as well as in graphene [17], that the amplitude of the CF depends quite sensitively upon the amplitude of the random potential. This means that the fluctuations do not possess a universal property, especially as they are known to be suppressed in very high quality material. In addition, the response to sweeping the Fermi energy directly in the system, and to varying it via the magnetic field, produce dramatically different amplitudes of the fluctuations, which means that these fluctuations are not ergodic either. Similar behavior is seen in other interference situations, such as measurements of the $1/f$ noise in graphene [26]. While it may still be true that the actual physical basis for the fluctuations is a variation in the quantum interference of carrier waves in the random potential, the resulting CF are quite sensitive to the actual nature of this potential, both its amplitude and its range. Hence, one may expect that the improvement in the quality of the material under study will naturally lead to reduced amplitudes of these fluctuations. In fact, these results are all consistent with an understanding based on Anderson's much older description of disordered materials [18].

We do know that in very high quality material, no CF are observed, so apparently this material does not possess a sufficient degree of disorder. Secondly, we have shown previously that as the disorder is increased, states are gradually localized. We would expect the potential landscape to show more variations as the disorder increases, and this should then lead to larger amplitudes of CF, just as observed in our previous studies. Indeed, along with the onset of fluctuations, we observe the localization of the low-lying transverse modes in the device [15]. When the fluctuations are fully developed, at the higher values of the disorder potential, their amplitude is essentially just that expected by the turning on or off of a single transverse mode in the sample. Thus, we expect that this will lead to a maximum rms value of the conductance fluctuation as

$$\delta G_{\text{rms}} = \frac{\eta_{\text{valley}} \eta_{\text{spin}} e^2}{2\sqrt{2} h},$$

where the η are the degeneracy factors. This value actually agrees well with the earlier perturbation theory estimates [10], but is based more on empirical observations than on analytical methods. It should be remarked that the peak rms amplitude seen in graphene can be less than this, with this peak value of $0.35(4e^2/h)$ being seen only at zero magnetic field in figure 4.

Third, there is no concept of ergodicity that should be applied to CF. As the Fermi energy is varied, the number of transverse modes and the density also vary. But, the variation of the density with Fermi energy is quite different in e.g. GaAs and graphene. Similarly, the magnetic field also will cause the Fermi energy to change the number of transverse modes even when the density is constant. Yet, the energy scale for this, as given e.g. by the cyclotron energy, differs between e.g. GaAs and graphene, as discussed above. Hence, there is no reason to believe that there should be the same behavior for these different variations. Fourth, there is no *a priori* reason to expect

that the degree of disorder will be the same for different samples (assuming that they do not come from the same slice of material) from different materials. Hence, there should be no expectation that the CF will have the same amplitude in these different samples.

Finally, as the CF depend upon the nature of the disorder potential, there may well be a density dependence, as seen in figure 5 above. Whether this arises from variations in the screening of the random potential, or from another effect, this further highlights the fact that there should be no expectation of ergodic behavior in the CF. The predictions of Anderson universality is that each and every sample is unique and one should not extend conclusions about the CF seen in a particular sample to other samples and materials.

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"Freeing" Graphene from Its Substrate: Observing Intrinsic Velocity Saturation with Rapid Electrical Pulsing

H. Ramamoorthy,[†] R. Somphonsane,[‡] J. Radice,[†] G. He,[†] C.-P. Kwan,[§] and J. P. Bird^{*,†,||}

[†]Department of Electrical Engineering, University at Buffalo, the State University of New York, Buffalo, New York 14260-1900, United States

[‡]Department of Physics, King Mongkut's Institute of Technology Ladkrabang, Bangkok 10520, Thailand

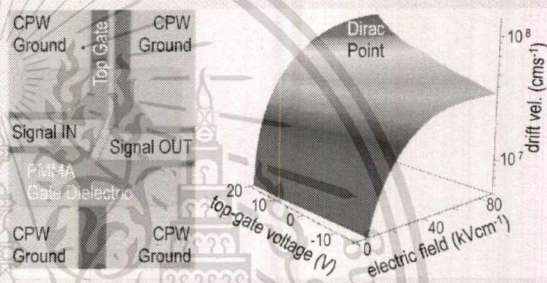
[§]Department of Physics, University at Buffalo, the State University of New York, Buffalo, New York 14260-1500, United States

^{||}Graduate School of Advanced Integration Science, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan

Supporting Information

ABSTRACT: Rapid (nanosecond-scale) electrical pulsing is used to study drift-velocity saturation in graphene field-effect devices. In these experiments, high-field pulses are utilized to drive graphene's carriers on time scales much faster than that on which energy loss to the underlying substrate can occur, thereby allowing the observation of the highest saturation velocities reported to date. In a dramatic departure from the behavior exhibited by conventional metals and semiconductors, as the electron or hole density is reduced toward the charge-neutrality point, the drift velocity is found to reach values comparable to the Fermi velocity itself. Corresponding current densities are as large as 10^9 A/cm², similar to the values reported for carbon nanotubes and for graphene-on-diamond transistors. In essence, our approach of rapid pulsing allows us to "free" graphene from the deleterious influence of its substrate, revealing a pathway to achieve the superior electrical performance promised by this material. The usefulness of this approach is not merely limited to graphene but should extend also to a broad variety of two-dimensional semiconductors.

KEYWORDS: Velocity saturation, hot carriers, graphene, transient transport, thermal effects in nanodevices



Realizing the full potential of graphene¹ as a novel material for nanoelectronics^{2,3} requires strategies to maximize its current-carrying capacity, while minimizing Joule losses to its surrounding environment. At the microscopic scale, this problem reduces to one of managing drift-velocity saturation (DVS), which arises when carriers, strongly accelerated by the electric field, rapidly lose their excess energy by spontaneously emitting optical phonons. By limiting the ultimate velocity to which carriers can be accelerated, DVS constrains the maximal currents that can be carried by any material, and must therefore be mitigated in many device scenarios. Although the large optical phonon energies (>150 meV) intrinsic to graphene promise high saturation velocities⁴⁻⁷ (v_{sat}), experiments typically yield much lower values that are degraded by unintended heating of the underlying substrate.⁸⁻¹⁶ This problem was apparent from the earliest investigations of the high-field characteristics of graphene, which revealed the velocity saturation to be consistent with a process in which the drifting carriers lose energy by exciting lower-energy (~55 meV) surface optical phonons (SOPs) in the SiO₂ substrate.¹⁷ This result may be understood quantitatively in terms of the (approximate) relation for the saturation velocity¹⁰

$$v_{\text{sat}} = \frac{2}{\pi} \frac{\omega_{\text{OP}}}{\sqrt{\pi n}} \quad (1)$$

where n is the two-dimensional (2D) carrier density. This relation shows that the value of v_{sat} is directly proportional to the energy ($\hbar\omega_{\text{OP}}$) of the specific optical phonon responsible for hot-carrier energy loss. Because the energy of the SiO₂ SOPs is much lower than that of graphene's intrinsic optical modes, energy transfer to the substrate can dominate the velocity saturation, resulting in the surprisingly low values reported for v_{sat} .⁸⁻¹⁶ In quantitative terms, using a density $n = 10^{12}$ cm⁻² as a reference value, eq 1 predicts a saturated velocity $v_{\text{sat}} \sim 10^8$ cm s⁻¹ for intrinsic graphene, while typical experiments performed with SiO₂ substrates yield values around a factor of four or five times smaller than this.

Recognizing the negative influence of the substrate on the high-field characteristics of graphene devices, one approach to overcome its effects are to eliminate it completely, making use of devices in which the graphene channel is freely suspended.¹⁸⁻²⁰ Measurements²¹ of such devices do indeed exhibit higher saturation velocities than those reported for graphene-on-SiO₂ transistors, yet still do not reach the values expected for intrinsic graphene. Alternatively, exfoliation of

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graphene onto BN substrates has been explored as a means to suppress substrate-related scattering and has been demonstrated to significantly improve low-field mobility.^{22–24} While studies of DVS have been limited in this system, they have nonetheless been found¹⁴ to exhibit low values for v_{sat} consistent with a phonon pathway with an energy of just ~ 40 meV.

Having identified how energy loss from graphene's hot carriers strongly suppresses its saturated velocity, a question that arises concerns whether suitable schemes can be implemented to alleviate this problem? The answer to this question is suggested by recent studies of transient transport in graphene, which show that heating of the substrate does not develop instantaneously, but rather takes place over many tens of nanoseconds.^{12,25,26} This result can be attributed to the large thermal mismatch between channel and substrate and indicates that pulsed investigations of graphene, performed on nanosecond time scales, should allow its intrinsic nonequilibrium characteristics to be revealed.¹¹ While there have previously been several different reports of transient transport in graphene,^{11,25–28} none of them have explored the behavior in this regime of rapid pulsing. In this Letter, we overcome this problem by using rapid (nanosecond) pulses, allowing us to probe the true hot-carrier dynamics of graphene before its performance is degraded through its coupling to the substrate. In this way, we are able to achieve saturation velocities consistent with those expected for intrinsic material, and higher than those reported for suspended graphene²¹ and for devices realized on boron-nitride substrates.¹⁴ In a dramatic departure from the behavior exhibited by conventional metals and semiconductors in which the drift-induced shift of the Fermi surface is typically small, as the Dirac point is approached from either the conduction or valence bands we find that v_{sat} reaches values comparable to the Fermi velocity ($\sim 10^8$ cm s⁻¹) itself. Corresponding current densities ($\sim 10^9$ A/cm²) are similar to those found in carbon nanotubes²⁹ and in graphene-on-diamond transistors.³⁰ In essence, our approach of rapid pulsing allows us to "free" graphene from the influence of its substrate and to therefore reveal a pathway to achieve the superior electrical performance promised by this material. Beyond graphene, we expect that this approach should also be amenable to investigations of DVS in many other two-dimensional materials.

Rapid pulsing of graphene was performed at room temperature in an impedance-matched setup (Supporting Information) in which electrical contact to graphene flakes, exfoliated on SiO₂, was provided by on-chip coplanar waveguides (CPWs, see Figure 1a). The carrier density was varied by an aligned top-gate (TG), isolated from the graphene by 120 nm of hardened PMMA (insets to Figure 1a,b). Pulses of various amplitude (v_p^{in}) and duration were applied to the input CPW, and the resulting current was measured by connecting the output waveguide to the 50- Ω input of a sampling oscilloscope. For sufficiently small pulse amplitudes, the transient resistance (R) determined at different gate voltages (V_{TG}) agreed well with that obtained by small-signal lockin measurement (Figure 1b). Five devices (D1–D5) were investigated here with D2–D5 being monolayer while D1 was realized from bilayer graphene (see the Supporting Information for the dimensions of the devices and their room-temperature mobility.)

The transient electrical characteristics of graphene can be strongly influenced by extrinsic factors, most notably by charge

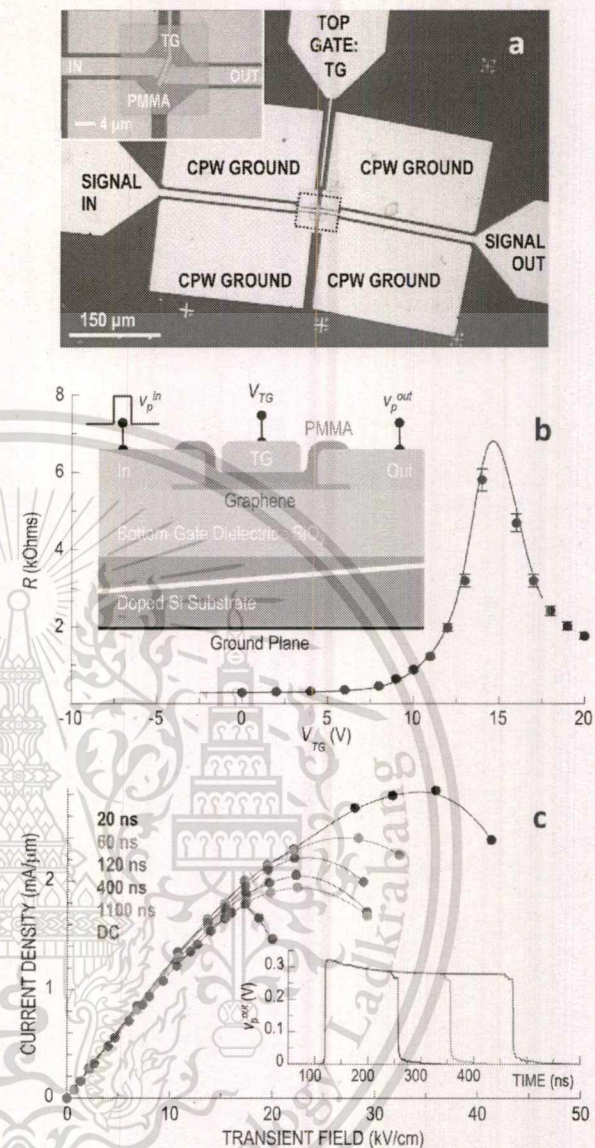


Figure 1. (a) The main panel is an optical micrograph of one of the graphene transistors (Device D5) studied here, showing the CPW structure used to make contact to the graphene flake. The area enclosed by the dotted line is shown in expanded form in the inset, in which the hardened PMMA layer has been colorized to highlight the top-gate geometry. (b) Comparison of the Dirac curve measured for Device D1 at room temperature by small-signal lockin detection (solid line) and low-amplitude transient pulsing ($v_p^{\text{in}} = 100$ mV, corresponding to a field strength $F \sim 0.6$ kVcm⁻¹; duration = 4 ns; filled symbols). The inset is a cross-sectional schematic indicating the essential components of the top-gated devices. (c) The main panel shows the current–voltage characteristic determined at $V_{\text{TG}} = 0$ and room temperature for Device D1 for pulses of varying duration (indicated) and for DC biasing. The inset plots three long pulses of varying duration ($v_p^{\text{in}} = 3.05$ V, corresponding to $F \sim 34$ kVcm⁻¹) for Device D4, exhibiting the influence of charge trapping and substrate heating.

injection into the dielectric layers of the device and by thermal conduction to its substrate.^{25–28,31} Under such conditions, it is easy to reach misleading conclusions regarding the physical

origins of current saturation, as demonstrated by the results of Figure 1c. Here we plot current–voltage characteristics of one of our devices for transient pulses of varying duration and amplitude. (Throughout this Letter, quoted current values, and the drift velocities inferred from them, correspond to those determined at the end of the current pulse just before its falling edge.) At fields (F) below ~ 15 kV/cm, the transient current is independent of pulse duration and is also consistent with that obtained by DC biasing. At higher fields, however, the current decreases systematically with increasing pulse length and tends toward the DC result. Present in each data set is an apparent region of negative-differential conductance (NDC), whose onset shifts to higher field for shorter pulses. This behavior does not represent an intrinsic NDC, however, but rather results from the extrinsic influence of trapping and heating.²⁶ This may be seen by referring to the inset of Figure 1c, where we show measurements of pulses up to several hundred nanoseconds long. A monotonic decay of v_p^{out} is seen on this time scale, which has previously been identified as arising from the combined influence of charge trapping and substrate heating.^{25–28,31} It is this decay that is responsible for the observed NDC and in order to avoid its influence it is necessary to pulse graphene on time scales much faster than those considered in Figure 1c.

An example of our rapid pulsing of graphene is presented in Figure 2a, the contour of which represents the output transients (v_p^{out}) measured in response to the application of short (4 ns) pulses of varying amplitude (v_p^{in}). The contour nicely reproduces the form of the input pulses, as shown in the insets to Figure 2a where we directly compare the input and output signals. The faithful manner in which the input transient is reproduced at the output demonstrates that parasitic circuit elements exert little influence on these measurements, and that trapping and substrate heating are both suppressed. In Figure 2b–d, we plot the current–(drain) voltage characteristics inferred from experiments of this type. Figure 2b plots gate voltages on the hole side of the Dirac curve (whose charge-neutrality point is at $V_{\text{TG}} \sim 16.5$ V), while Figure 2d shows similar data for electrons. Clear current saturation is observed for both carrier types with no evidence of NDC, despite the fact that the peak field strength in the measurements is almost twice that in Figure 1c.

From data such as those of Figure 2, and a knowledge of the carrier density (Supporting Information), the field-velocity characteristics of graphene are determined. Representative results are plotted in Figure 3a,b, where we see clear saturation of the drift velocity (v_d) for field strengths exceeding ~ 30 kV/cm. Consistent with the form of eq 1, v_{sat} increases as the carrier density is reduced from either the hole or electron sides. In the vicinity of the Dirac point, however, the drift velocity does not saturate (Figure 3a, inset), but rather shows a “kink-like” feature. The kink is also apparent in the current–voltage characteristics (see the data for $V_{\text{TG}} = 10$ and 15 V in Figure 2b and for $V_{\text{TG}} = 20$ V in Figure 2d) and has previously been attributed to the role of minority-carrier injection from the drain.^{8,13,32}

The results obtained from our rapid-pulsing studies exhibit a number of important differences with those reported in prior (DC or quasi-DC) investigations. First, the current densities that we achieve (~ 4 mA/ μm , see Figure 2b) are much larger than reported previously. Second, we note that velocity saturation has previously been parametrized^{10,11,16,33} by appealing to Thornber’s equation³⁴

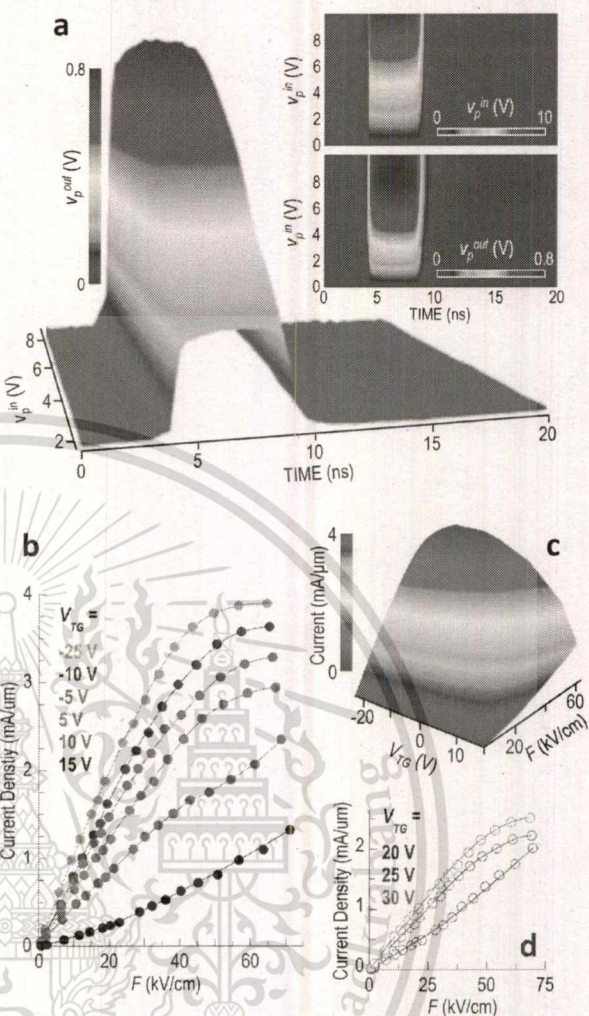


Figure 2. (a) Output pulses of duration 4 ns, measured in Device D2 in response to the application of input pulses of varying amplitude. The top-gate voltage is fixed at $V_{\text{TG}} = -10$ V. The inset plots the form of the input and output pulses as two-dimensional color contours to allow a comparison of their forms. (b) Transient current–voltage characteristics determined using 4 ns pulses such as those in panel a. Data are for holes in Device D3 and the different top-gate voltages are indicated. (c) Data in panel b replotted as a color contour. (d) Transient current–voltage characteristics determined using 4 ns pulses such as those in panel a. Data are for electrons in Device D3 at the indicated top-gate voltages. Solid lines in panels b and d are interpolation curves.

$$v_d = \left[\left(\frac{1}{\mu F} \right)^\beta + \left(\frac{1}{v_{\text{sat}}} \right)^\beta \right]^{-1/\beta} \quad (2)$$

where μ is the low-field mobility, F is the electric field responsible for drift, and β is a phenomenological parameter. In work based on DC (and quasi-DC) methods, the field dependence of v_d has been found to be well fitted^{10,11} with $\beta \approx 2$. As we show in Figure 3c, however, our data are also well described by eq 2 but only by taking $\beta = 5$. This larger value translates into a more rapid initial increase of current with electric field and to the onset of saturation at larger v_d (see Figure 3c). Overall, the implication of these observations is that

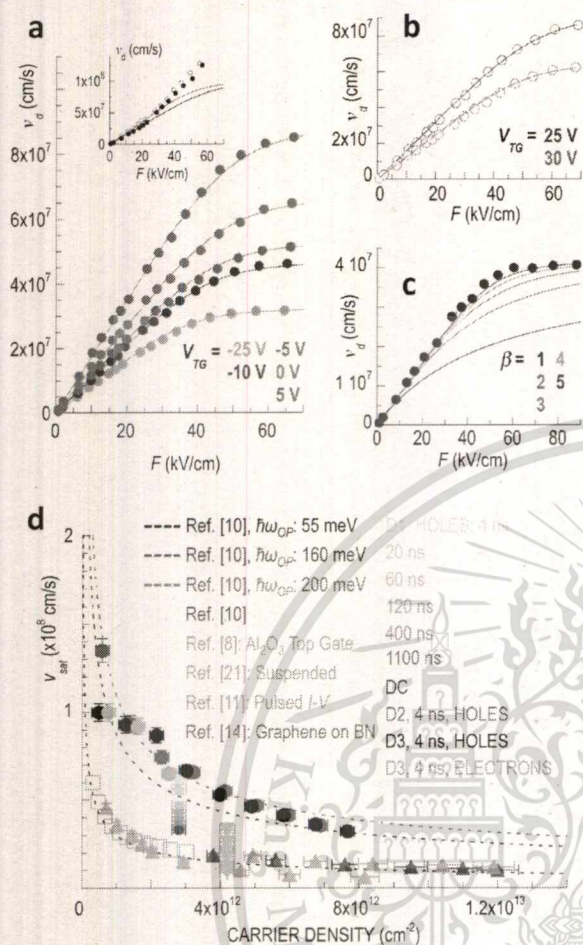


Figure 3. (a) Transient velocity-field characteristic determined for holes in Device D3 by 4 ns pulsing. Solid lines through the data are fits to eq 2 using the experimentally observed v_{sat} and a fit parameter $\beta = 5$. The inset plots corresponding data for the same device near its charge-neutrality point, where the current-voltage characteristics show the “kink” behavior arising from minority-carrier injection from the drain. Solid lines are fits to eq 2, obtained by assuming $\beta = 5$ and varying v_{sat} to obtain a best fit to the lower-field data. Filled symbols: $V_{TG} = 15$ V. Open symbols: $V_{TG} = 20$ V. (b) Similar plot as in panel a except for electrons in Device D3. (c) Fit of the velocity-field characteristic to eq 2, using the same values of v_{sat} but different β values. Filled symbols were obtained for Device D2 for $V_{TG} = -10$ V. (d) Variation of saturated drift velocity with carrier density. The different symbols are identified in the inset to the plot. Dashed lines correspond to the predictions of eq 1, using different characteristic phonon energies (also indicated in the inset).

relative to DC investigations our strategy of rapid pulsing allows us to more effectively drive graphene’s carriers by avoiding the onset of trapping and heating.

The most important result of our study is presented in Figure 3d, which shows the density-dependent variation of v_{sat} determined in three of our devices. Data obtained by rapid pulsing for both electrons and holes clearly follow the variation expected from eq 1, for graphene’s intrinsic optical-phonons ($\hbar\omega_{OP} = 160\text{--}200$ meV). As the density is reduced below 10^{12} cm $^{-2}$, v_{sat} approaches the Fermi velocity of graphene ($v_F \sim 10^8$ cm/s), providing a dramatic difference with the behavior exhibited by typical metals. (To determine v_{sat} in this low-

density limit, we restrict our fitting in terms of eq 2 to the field regime below the kink, see the inset to Figure 3a). These results should be contrasted with the data obtained from prior studies^{8,10,11,14,15} (data plotted as filled triangles), performed under DC, or quasi-DC, conditions and which clearly follow the variation expected for scattering from the lower-energy ($\hbar\omega_{OP} = 55$ meV) SOPs of SiO $_2$. In fact, our values for v_{sat} even exceed those obtained for freely suspended graphene²¹ (inverted triangles) and for graphene isolated on a BN substrate¹⁴ (open squares). On the basis of these observations we conclude that rapid pulsing of graphene can indeed be used to probe its intrinsic DVS, free of the negative impact of current-induced Joule heating of the substrate.

Some further points should be made regarding the importance of using very short pulses to obtain the intrinsic saturation velocities demonstrated in Figure 3d. In the inset to Figure 1c, the decaying portions of the different transient curves overlap very closely with one another, which leads one to consider whether it should be possible to infer the intrinsic velocity saturation from these curves also, simply by using the maximum value of the current just after the rising edge? While this would be reasonable if our measurements were performed in a “single-shot” scheme, it must be noted that our transient curves are instead captured by repetitive sampling. Consequently the entire duration of the transient pulse, including that obtained at short times, may reflect processes occurring on much longer time scales. These include substrate heating and charge trapping, the latter of which is known²⁶ to yield long recovery times. This point is discussed in more detail in the Supporting Information, where we provide an example to demonstrate the significantly increased saturation velocities that can be achieved by using short-pulse studies. As a further, more technical, point we note that such pulsing allows us to apply much higher fields to the devices (compare the maximum field strength in Figure 1c to that in Figures 2b–d and 3a–c) without irreversibly degrading their performance or even destroying them as the local substrate temperature is driven well-beyond ambient.

As a final comment on the results of Figure 3d, we point to the data plotted as the solid blue circles in this figure. These data show how the inferred drift velocity is steadily degraded as the measurement pulse is made longer; on increasing the duration from 4 to 20 ns, the drift velocity already drops by some 10%, while for a pulse length of around a microsecond the decrease is as much as 50% (these data are plotted more clearly in the Supporting Information). This figure again emphasizes the need to probe transport on very short (nanosecond) time scales when studying graphene’s intrinsic characteristics. A useful avenue to pursue in the future, to study the dynamics of thermal decoupling from the substrate in more detail, would be to fix the pulse duration at its shortest value and to steadily increase the pulse-repetition frequency up to a value (125 MHz) approaching the pulse length. Unfortunately, this was not possible with our experimental setup in which the maximum repetition frequency of the pulse generator was 1 MHz. Up to this frequency, at least we found the short-pulse characteristics to be independent of the repetition rate, as we describe in further detail in the Supporting Information. Despite being unable to extend our measurements to higher repetition frequencies, for the first time we emphasize that the main value of this work lies in its demonstration of the use of short well-separated pulses to reveal the intrinsic velocity saturation of graphene.

While previous theoretical work^{6,35,36} has explored the influence of different substrates on high-field DC-transport in graphene, our work demonstrates how rapid pulsing may be used to effectively eliminate any such influence. In this way, we achieve significantly higher saturated velocities than those reported previously, despite the poorer low-field mobility of our devices (Supporting Information) when compared to suspended material²¹ or to graphene on BN.¹⁴ The lessons learned from our study are not only relevant to graphene but rather point to a broader strategy in which rapid pulsing may be used to manage heat dissipation in different 2D semiconductors.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.5b04003.

The high-frequency measurement setup used to perform the pulsed studies; basic low-field characterization of device mobility; calculations of the drift velocity accounting for quantum capacitance, residual charge, and thermal carriers; and a discussion of the influence of pulse duration on the velocity-saturation characteristics.

(PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: jbird@buffalo.edu. Phone: +1 (716) 645-1015.

Author Contributions

All authors contributed equally to this work.

Notes

The authors declare no competing financial interest.

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Plasmon-mediated energy relaxation in graphene

D. K. Ferry,¹ R. Somphonsane,² H. Ramamoorthy,³ and J. P. Bird³

¹School of Electrical, Computer, and Energy Engineering, Arizona State University, Tempe, Arizona 85287-5706, USA

²Department of Physics, King Mongkut's Institute of Technology, Ladkrabang, Bangkok 10520, Thailand

³Department of Electrical Engineering, University at Buffalo, the State University of New York, Buffalo, New York 14260-1500, USA

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Energy relaxation of hot carriers in graphene is studied at low temperatures, where the loss rate may differ significantly from that predicted for electron-phonon interactions. We show here that plasmons, important in the relaxation of energetic carriers in bulk semiconductors, can also provide a pathway for energy relaxation in transport experiments in graphene. We obtain a total loss rate to plasmons that results in energy relaxation times whose dependence on temperature and density closely matches that found experimentally. © 2015 AIP Publishing LLC.
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The study of “hot” carriers, driven out of equilibrium by an electric field, has long been pursued, since the relaxation properties of these carriers are important for semiconductor-device performance.¹ With the emergence of graphene as a promising electronic material,^{2–4} there has been significant experimental^{5–18} and theoretical^{19–24} interest in the dynamics of its hot carriers. One aspect of graphene is the high energy of its optical phonons, which should render these modes ineffective at mediating cooling below room temperature.²⁵ Similarly, momentum-conservation constraints dictate that cooling via acoustic phonons is quasi-elastic, and therefore also inefficient. Consequently, the role of other scattering processes has drawn increasing attention, with particular interest having focused on the implications of impurity-assisted “supercollisions.”²³

Commonly in studies of hot-carrier cooling, one compares the variations of some transport feature (e.g., mesoscopic fluctuations, localization-related magneto-resistance, and other quantum corrections), as a function of lattice temperature (T_L) and electric field, to infer hot-carrier temperature (T_e). We study the dependence of this parameter on electric field and T_L yields information on the scattering processes responsible for cooling, with this assignment achieved by introducing the hot-carrier energy-loss rate (P_e). Defined as the power dissipated per electron, this rate usually varies as²⁶

$$P_e(T_e, T_L) = A(T_e^p - T_L^p), \quad (1)$$

with A and p characteristic parameters reflective of the specific relaxation process. Acoustic-phonon scattering in pristine graphene, for example, is predicted to yield $p=4$,^{19,20} while $p=3$ is expected for supercollisions.²³ While both power laws have been reported in separate experiments,^{10,14–17} other work has demonstrated an exponent of $p=3$ in a regime where supercollisions are clearly not expected to be effective.^{13,18} This suggests that some other mechanism may be responsible for cooling, and in this letter, we suggest that this may be provided by plasmon-mediated^{27,28} scattering.

The relaxation of carrier energy through plasmon-induced losses has been studied for transport in metals^{29,30} and semiconductors^{31–37} (the single-particle carrier-carrier scattering, of course, does not relax either the net energy or the net momentum). Plasmons themselves have also been studied in carbon nanotubes³⁸ and graphene,³⁹ where the dielectric function has been calculated.⁴⁰ The plasmons of interest here arise from the free carriers in graphene, and their influence on cooling is determined by starting from the standard expression for the energy-loss rate^{1,25,41}

$$P_e \equiv \left\langle \frac{dE}{dt} \right\rangle = \sum_{k,q,\omega} \{ f(E_k)[1 - f(E_k - \hbar\omega)] \hbar\omega W_{k \rightarrow k-q} - f(E_k)[1 - f(E_k + \hbar\omega)] \hbar\omega W_{k \rightarrow k+q} \}, \quad (2)$$

where the symbols E and k represent the carrier energy and momentum, respectively, while $\hbar\omega$ and $\hbar q$ are the energy and momentum exchanged in each collision, here that of the plasmon modes, and $f(E)$ is the carrier distribution function. The scattering element W describes the plasmon-mediated interaction, the form of which we take from treatments of the low-temperature dephasing time (although with some differences). In (2), the energy-relaxation process depends on the difference in emission and absorption terms, whereas the phase-breaking time depends upon the sum of these terms. Each of these two terms is expressed as

$$W_{\pm} = \frac{2\pi}{\hbar} N \text{Im} \left\{ \frac{V(q)}{\epsilon(q, \omega)} \right\} \delta \left(\omega - \frac{E_k - E_{k \pm q}}{\hbar} \right), \quad (3)$$

where N is either the Bose Einstein distribution, in the absorption term, or 1 plus this distribution, in the emission term. The scattering function arises from the imaginary part of the screened Coulomb potential $V(q) = e/2\epsilon_s q$, where e is the electron charge and ϵ_s is the permittivity of graphene.^{28,42} Here, the screening is done by the full frequency and momentum dependent dielectric function. As may be expected, the dielectric function is dominated by the plasmon pole, but in using the inverse of the dielectric function in (3), we

incorporate the collision broadening of this resonance. This is given as $\text{Im}(V/\varepsilon) = \omega e^2 / \varepsilon_s \chi_2 q^2 \tau$, where $\chi_2 = n_s e^2 / 2m^* \varepsilon_s = n_s e^2 v_F / 2\hbar k_F \varepsilon_s$, n_s is the sheet density of carriers, v_F is the Fermi velocity, k_F is the Fermi momentum, and τ is the scattering time. In two dimensions, the plasmon frequency involves the momentum as $\omega_p^2 = n_s e^2 q / 2m^* \varepsilon_s$, where n_s is the sheet density and m^* is the electron or hole effective mass ($=\hbar k / v_F$). The broadening arises from the total scattering rates, which yield a scattering time τ . A key step in the development of the energy loss rate lies in the fact that (3) may be used to rewrite (2) as

$$P_e = 2\pi \sum_q \int \frac{\omega}{2\pi} d\omega \text{Im} \left\{ \frac{V(q)}{\varepsilon(q, \omega)} \right\} \delta_\omega \Phi(\hbar\omega), \quad (4)$$

where δ_ω represents the energy-conserving delta function and the various distribution functions (both Fermi-Dirac and Bose-Einstein) are pulled into the expression

$$\Phi(x) = \sum_k \{ [N(x, T) + 1] f(E) [1 - f(E - x)] - N(x, T) f(E) [1 - f(E + x)] \}, \quad (5)$$

where x will ultimately involve the plasmon frequency and the electron distribution function is at the carrier temperature T_e . Carrying out the various integrations (see supplementary material⁴³) yields the energy-loss rate

$$P_e \equiv \left\langle \frac{dE}{dt} \right\rangle = \frac{2.4 k_B^3}{2\pi \hbar^2 v_F^2 \tau} (T_e^3 - T_L^3), \quad (6)$$

where v_F is the Fermi velocity. Comparison with the form of (1) indicates two important features of this expression. First, the plasmon mechanism yields an exponent of $p=3$, similar to that obtained for supercollisions. Second, the prefactor (A) in the energy-loss rate of (6) is governed by a system-dependent scattering time τ , which describes the broadening, that can be determined from the density and temperature dependent mobility of the device under test. In most cases at low temperature, this is dominated by the impurity scattering.²⁵

An important parameter related to the energy-loss rate is the energy-relaxation time (τ_e), which may be obtained from (6) by factoring the cubic terms in T to give a definition of τ_e from the theoretical parameters⁴⁰

$$P_e = \frac{k_B(T_e - T_L)}{\tau_e}. \quad (7)$$

In Fig. 1, we compare the variation of $\tau_e(T_e)$, obtained in the experiment of Ref. 13, with that predicted by (6) for the base temperature of 1.8 K, as used in the experiments. Theoretical values of τ_e are determined by taking the experimentally-determined energy-loss rate for the given density and T_e and introducing this into (6) and (7) to compute τ_e . From this figure, we see that the plasmon-based model reproduces not only the low-temperature magnitude of τ_e , for both electrons and holes, but that it also captures its quantitative dependence on temperature.

The most striking observation of Ref. 13 was of a strong variation of τ_e as a function of carrier density. This behavior

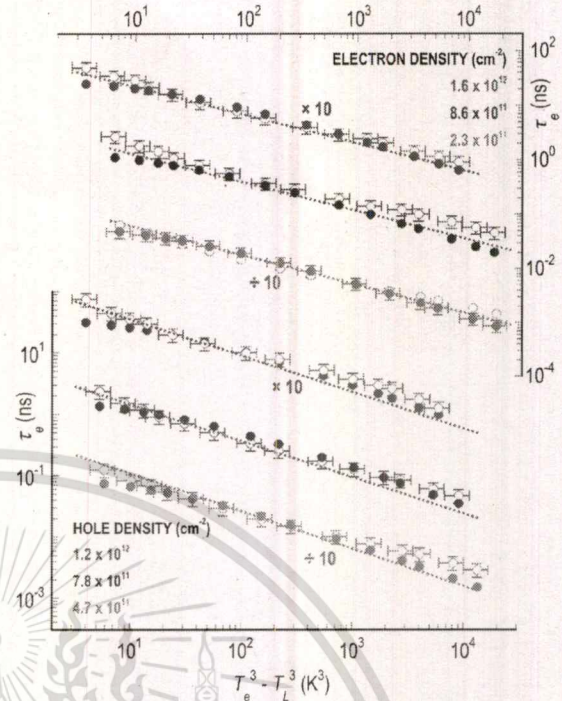


FIG. 1. The energy relaxation time for several values of the electron and hole density. Experimental values from Ref. 13 are plotted as open symbols while theoretical values obtained from (6) and (7) are the closed symbols. Error bars are shown for the experimental data only. Dotted lines through the theoretical data are guides to the eye that indicate a linear variation of power input with T^3 . In each case, the red and green curves have been offset by a factor of 10 for clarity as indicated.

is reproduced in Fig. 2, along with the results of our calculations. The main panel compares the behavior observed at a number of different electron temperatures, and again shows close, if not perfect, agreement between experiment and theory. In the inset to the figure, we plot the variation of τ_e as a function of electron density, for three different measurement currents. As with the data in the main panel, the theory reproduces the experiment well.

The plasmon-loss mechanism rearranges the energy distribution function, but this energy remains in the electron gas as a whole. Under the excitation current (or electric field), the distribution function is shifted in momentum space to reflect the current, and it spreads as the electron temperature rises. The shift can also distort the distribution to produce an elongated extension in the direction of the field or current. The ultimate limit of this elongation is a delta function in the field direction.¹ Between the shift in momentum space, and the elongation, the distribution function has more carriers in the forward direction than the backward direction. The plasmon loss takes carriers from the streaming forward direction and moves them to the backward direction, in the symmetric terms of the distribution function, on the average. This scattering process relaxes the streaming distribution, but the ultimate result still remains that another process must pass the energy to the lattice. This can be due to acoustic-phonon scattering,¹⁹⁻²⁴ or even supercollisions.²³ However, it is the plasmon interaction that dominates what is seen experimentally, as it is this process which moves the current carrying particles out of the forward extension of the distribution

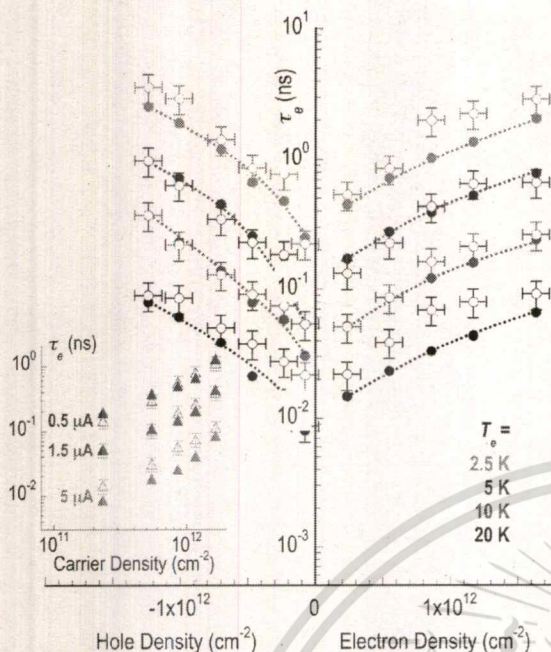


FIG. 2. The energy relaxation time as a function of the carrier density with the electron temperature as a parameter. Experimental values from Ref. 13 are plotted as open symbols while theoretical values obtained from (6) and (7) are the closed symbols. Dotted lines provide a guide though the theoretical data (filled symbols). At the lower left, we plot the variation of the relaxation time with density for a few values of the excitation current at $T_L = 4.2$ K.

function. The other processes which ultimately move the energy to the lattice are thus masked, and this would be even more true with the supercollision process as it has the same temperature dependence (i.e., $p = 3$) as that found here.

In conclusion, a good quantitative agreement between the experimental data of Ref. 13 and the theoretical results obtained here suggest that the plasmon interaction dominates hot-carrier energy relaxation in graphene at low temperatures. Since the role of this mechanism in governing energy loss does not appear to have been appreciated thus far, it suggests that its relevance may be far wider than simply the experimental study analyzed here.

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⁴³See Supplementary material at <http://dx.doi.org/10.1063/1.4938760> for the process of getting to (6) from (4).

Energy relaxation of hot carriers in graphene via plasmon interactions

D. K. Ferry¹ · R. Somphonsane² · H. Ramamoorthy³ · J. P. Bird³

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Abstract Energy relaxation of hot carriers in graphene is studied theoretically and experimentally at low temperatures, where the loss rate may differ significantly from that predicted for electron–phonon interactions. We show here that plasmons, important in the relaxation of energetic carriers in bulk semiconductors, can also provide a pathway for energy relaxation in transport experiments in graphene. Reflecting the linear nature of graphene’s bands, we obtain a total loss rate to plasmons that is independent of carrier density. This results in energy relaxation times whose dependence on temperature and density closely matches that reported experimentally.

Keywords Graphene · Energy relaxation · Plasmons · Carrier heating

1 Introduction

The study of “hot” carriers in semiconductors has been pursued for many decades (See, e.g., [1]). By hot carriers, we refer to those carriers driven out of equilibrium by an applied electric field, or equivalently by passing a large current through the device. Since the relaxation of these carriers is important for electronic-device performance, it is important

to understand just how this relaxation process occurs. When an electric field is applied to a semiconductor, or a current is passed through it, the distribution function is dramatically affected. While spherically symmetric in momentum space at thermal equilibrium (or cylindrically symmetric in the case of a two-dimensional semiconductor such as graphene), under non-equilibrium conditions this function splits into what can be considered as two separate parts. The first of these remains largely symmetric, while the second, the so-called “streaming”, term is responsible for the current [2–4]. Electrical transport measurements typically probe *only* the streaming term, while telling us nothing about the symmetric one, behavior that should be contrasted with optical measurements. In the latter, the small photon momentum does not create the streaming term, and *only* information about the symmetric term can be obtained. Here, we want to treat plasmon-mediated energy relaxation, which is different since the interaction is between carriers in the streaming part of the distribution and those in the symmetric part. That is, the plasmon interaction transfers net energy *from* the streaming terms, thus serving to relax that part of the distribution that is measured by transport. It does this by primarily (near) back-scattering (for materials like graphene, exact back scattering is chirally forbidden). In a purely symmetrical distribution, we would not expect this process to be important, so that it is not likely to be significant in an optical experiment.

With the emergence of graphene as a promising material for future nanoelectronics [5–7], there has been significant experimental [8–21] and theoretical [22–27] interest in the processes governing its hot carrier dynamics. One aspect of graphene is the very high energy of its optical phonons, which ensures that these are ineffective at cooling electrons and holes except at higher temperatures, near or above room temperature [28]. At lower temperatures than this, the major cooling pathway has been thought to involve the

✉ D. K. Ferry
ferry@asu.edu

¹ School of Electrical, Computer, and Energy Engineering, Arizona State University, Tempe, AZ 85287-5706, USA

² Department of Physics, King Mongkut’s Institute of Technology, Ladkrabang, Bangkok 10520, Thailand

³ Department of Electrical Engineering, University at Buffalo, The State University of New York, Buffalo, NY 14260-1500, USA

interaction of hot carriers with acoustic phonons. Due to momentum-conservation constraints, however, this process is quasi-elastic, and so inefficient at mediating energy transfer. Consequently, the role of other phonon processes has drawn increasing attention, with significant interest having focused on the implications of impurity-assisted “supercollisions” [26]. Experiments utilizing noise thermometry [15, 20], coupled microwave and dc heating [29], and p-n junction photocurrent spectroscopy [18, 19], have established the role of this mechanism at elevated carrier/phonon temperatures. Nonetheless, lower-temperature studies of mesoscopic devices [16, 21] suggest that this mechanism does not provide a complete picture of hot-carrier cooling.

In evaluating the heating of carriers induced in a transport measurement, it is convenient to express the results in a form (See e.g., [30])

$$P_e(T_e, T_L) = A (T_e^p - T_L^p), \quad (1)$$

where P_e is the power input per carrier (the energy loss rate) and A and p are parameters determined by the specific mechanism of energy relaxation. Scattering of hot carriers by acoustic phonons in pristine graphene, for example, is predicted to be characterized by an exponent $p = 4$ [22, 23], while $p = 3$ is expected for the aforementioned supercollisions [26]. The situation with regards to the latter mechanism is unclear, however, since some experiments [16, 21] have also yielded $p = 3$ while clearly being performed in the regime in which supercollisions are not expected to dominate. Motivated by this, we therefore explore the possibility of cooling in graphene by a very-different process, namely that arising from plasmon-mediated scattering of hot carriers. Here, we demonstrate that such an exponent ($p = 3$), in fact, also is consistent with the role of plasmon-mediated relaxation. By deriving an expression for the energy-relaxation time (τ_e), the time scale governing the loss of energy from hot carriers in the system, we demonstrate here that this interaction can provide an important relaxation pathway in graphene. To demonstrate the validity of this approach, we show that the relaxation time predicted by this model reproduces well the temperature- and density-dependent variations of this parameter found in a recent experiment [16]. Most importantly, this agreement is achieved *without the use of any adjustable parameters*. Thus, this highlights how the weak electron–phonon coupling in graphene opens up the possibility of hot-carrier dissipation via novel mechanisms, beyond those typically considered in more conventional materials.

2 Plasmons in graphene

Scattering of carriers by the screened potential of other electrons and from the collective plasmon modes are both part of

the total electron–electron (or hole–hole) interaction among the free carriers. Starting from the non-interacting electron gas, the inter-carrier Coulomb interaction is usually treated as a perturbation, but is problematic due to the long range of the Coulomb potential. The coupled dielectric function then has two important poles, one at zero frequency and one at the plasmon frequency. These correspond to the single-particle scattering potential and the plasmon mode, respectively. Generally, one introduces a screening wave vector to the Coulomb potential, which separates the long-range and the short-range parts. Then, the potential can be split into a short-range part, where the scattering wave vector is larger than this screening vector, and this corresponds to the free-particle Coulomb scattering. The long-range part, on the other hand, is responsible for the interactions with the collective plasmon mode. These collective modes are bosons, so that the scattering rate can be found in quite normal ways. The relaxation of carrier energy through plasmon-induced losses has been studied for transport in metals [31, 32] and semiconductors [33–39] for some time. Plasmons themselves have also been studied in carbon nanotubes [40] and graphene [41], where the dielectric function has been calculated [42]. It has also been suggested that the plasmon interaction can be important for phase breaking in two dimensional systems [43–45].

To be sure, there has been a lively debate in the literature over whether or not the electrons can actually interact with the plasmons. It has been stated in some papers that this interaction cannot occur in graphene [46–48], since graphene is *qualitatively different from a parabolic band semiconductor*. It has been known for quite some time that the plasmons can be scattered by the single-particle interactions, a process known as Landau damping. If this scattering is large, then the plasmon modes are heavily damped and one could say that they don’t really exist. However, it has been conclusively shown that the plasmon dispersion curve (energy versus wave vector) in graphene remains outside the region of single particle electron–electron interactions [42]. However, the dispersion curve does enter the region of electron–hole scattering. In spite of this, away from the Dirac point, one does not have strong electron–hole interactions as one or the other of these particles is absent. Moreover, in a recent review of many-body interactions in graphene [49], it was found that a double feature is present in the decay rate given by the imaginary part of the self-energy. One of these was a peak at positive energies, signaling an onset of plasmon emission. In another context, plasmonics is a new area of interest in graphene. Here, high frequency waves are used to excite the plasmons. In a recent paper, the authors computed the dielectric function and plasmons in graphene, and find that [50] “...scattering between plasmons and electrons is possible and will lead to damping of the plasmons.” This is plasmon absorption by the electrons. Of course, one then expects that the reverse will be true and this scattering will lead to emis-

sion of plasmons by energetic electrons. So, it appears that the plasmons are well defined excitations, even in graphene, and can be involved in the relaxation of hot carriers.

The scattering potential for the plasmons $V(q)$ remains the Coulomb potential, and this is combined with the momentum- and frequency-dependent dielectric function in the dynamic screening approach [51,52]. In general, this approach arises from the Lindhard RPA dielectric function, and the dynamic screening expression can be written as

$$\epsilon(q, \omega) = \epsilon_s \left[1 + \frac{\omega_p^2}{Dq^2/\tau - (\omega - i/\tau)^2} \right], \quad (2)$$

where the square of the plasmon frequency in two dimensions is given as

$$\omega_p^2 = \frac{n_s e^2 q}{2m^* \epsilon_s} = \chi_2 q. \quad (3)$$

In the dielectric response, one normally just keeps the plasmon-pole approximation, which means keeping only the frequency term in the denominator of (2). However, we require the imaginary part of the inverse dielectric function, which means keeping at least the leading term in the scattering time. Then, we may write the scattering term as

$$\text{Im} \left\{ \frac{V}{\epsilon} \right\} \simeq \frac{\omega e^2}{\epsilon_s \chi_2 q^2 \tau}, \quad (4)$$

with $\chi_2 = n_s e^2 / 2m^* \epsilon_s = n_s e^2 v_F / 2\hbar k_F \epsilon_s$ (ϵ_s is the permittivity of the graphene), and τ is the total scattering time in the material. We will use these results in the next section for the energy relaxation rate.

3 The energy relaxation rate

As we remarked above, the role of the electric field is to distort the distribution function to account for the hot carriers. We illustrate this in Fig. 1, where we compare the equilibrium distribution function (in two dimensions) and the streaming distribution function that results in the hot carriers. The electric field causes a portion of the distribution to stream along the field direction, and this accounts for the current that flows through the sample. The electric field provides a forcing function which is balanced by the relaxation of energy back to those states that have been emptied by the formation of the streaming part of the distribution. We indicate this for plasmons by the arrow that flows from the streaming part to the back of the distribution. Of course, the inverse process, the absorption of the plasmons, is also possible, and the two processes have to balance in equilibrium. The net energy loss by the carriers depends upon the difference between the

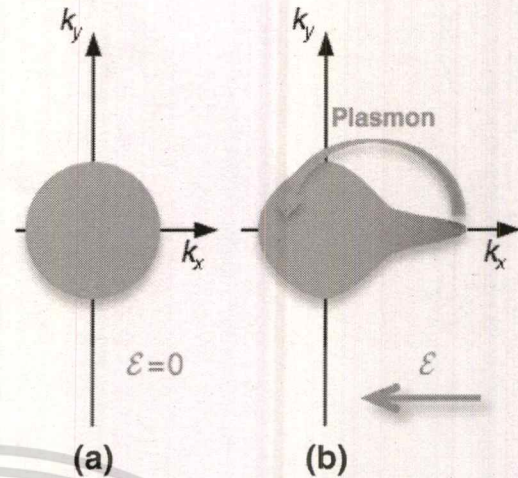


Fig. 1 a A diagrammatic depiction of the equilibrium distribution function in two dimensions. b A similar depiction of the formation of the streaming portion of the distribution, and the role of plasmons in the energy relaxation

emission rate and the absorption rate. The general approach we use to describe this overall process is quite straightforward. We begin with a standard equation for the energy loss rate, which can be written as [1], (See e.g., [30]), [53]

$$\left\langle \frac{dE}{dt} \right\rangle = \sum_{k,q,\omega} \left\{ f(E_k) [1 - f(E_k - \hbar\omega_q)] \hbar\omega_q W_{k \rightarrow k-q} - f(E_k) [1 - f(E_k + \hbar\omega_q)] \hbar\omega_q W_{k \rightarrow k+q} \right\}. \quad (5)$$

Here, W represents the scattering function. For the scattering function we take the standard approach to the phase-breaking time in mesoscopic materials at low temperature, in which the scattering function arises from the imaginary part of the screened Coulomb potential (See, e.g., [54]), [43]. There is a difference, however. It may be noted in (5) that the energy relaxation process depends upon the *difference* in the emission and absorption terms, as described above, whereas the phase-breaking time depends upon the *sum* of these two terms. Each of these two terms is expressed as

$$W_{\pm} = \frac{2\pi}{\hbar} N \text{Im} \left\{ \frac{V(q)}{\epsilon(q, \omega)} \right\} \delta \left(\omega - \frac{E_k - E_{k \pm q}}{\hbar} \right), \quad (6)$$

where the plus (minus) sign refers to the plasmon emission (absorption). The N here is either the Bose Einstein distribution, in the absorption term, or 1 plus this distribution, in the emission term. Note that we require the imaginary part of the inverse dielectric function, which is given by (4) above. In (5), there is a delta function which conserves the energy, while the momentum is also assumed to be conserved in writing this expression. There is still the phase space argument

of the delta functions to be considered, and this is discussed in Appendix 1.

Using these preliminaries, and the results of (6), we can now rewrite (5) as

$$P = 2\pi \sum_{\mathbf{q}} \int \frac{\omega d\omega}{2\pi} \text{Im} \left\{ \frac{V(\mathbf{q})}{\epsilon(\mathbf{q}, \omega)} \right\} \delta_{\omega} \Phi(\hbar\omega) \quad (7)$$

where δ_{ω} represents the energy conserving delta function, and the various distribution functions (both Fermi–Dirac and Bose–Einstein) are pulled into the expression

$$\Phi(x) = \sum_{\mathbf{k}} \{ [N(x, T) + 1] f(E) [1 - f(E - x)] - N(x, T) f(E) [1 - f(E + x)] \} \quad (8)$$

and the distribution function is at the carrier temperature T_e . Price [55] has shown how the integral over the distribution functions can be carried out, and how the electron temperature in the emission term finally appears in the Bose–Einstein distribution. Using the properties of the linear bands in graphene, his results applied to the last integral can be rewritten as

$$\Phi(x) = \frac{x E_F}{\pi(\hbar v_F)^2} [N(x, T_e) - N(x, T_L)] \quad (9)$$

In (7), the potential and inverse dielectric function are given by (4).

We are now ready to evaluate the integrals in (7). Using the delta functions and the other properties given above and in Appendix 1, we can carry out these integrals in a straightforward manner. This leads to two terms, as in (9), each of which can be written in terms of an arbitrary temperature as

$$F(T_1) = \frac{e^2 E_F k_B^3 T_1^3}{2\pi \epsilon_s \hbar^4 v_F^2 \chi_{2T}} \int_0^{u_{\max}} \frac{u^2 du}{e^u - 1} \quad (10)$$

where $u = \hbar v_F q / k_B T_1$ and $u_{\max} = 2\hbar v_F k_F / k_B T_1$ represents an approximate upper limit. In general, this upper limit is density and temperature dependent but is large, and the integral approaches a limiting value near 2.4 as is shown in Appendix 2.

Using the above results, and those of Appendix 2 for the integral, the final power loss is given as

$$\left\langle \frac{dE}{dt} \right\rangle = \frac{2.4 k_B^3 (T_e^3 - T_L^3)}{2\pi \hbar^2 v_F^2 \tau} \quad (11)$$

An interesting point is that, with the unique band structure of graphene, the explicit density dependence has dropped out of this final power loss, but will return when we normalize it to the power loss per electron. The second important point

to note is that the loss rate in this expression is characterized by the exponent $p = 3$, consistent with the results of recent experiments [16,21].

In (10) and (11), we see the scattering time τ is the only sample dependent parameter remaining in the energy loss rate. In general, we can use the measured mobility to infer a value only for the momentum relaxation time, which is longer than τ by a numerical factor that arises from the averaging over the scattering angle. The momentum relaxation time is weighted in favor of large angle scattering (subject to chirality constraints), while there is considerably more small angle scattering with the Coulomb interaction. Since there are other scattering processes than just the Coulomb scattering from impurities, it is difficult to assign an exact value that arises from this weighted average. We have assumed the dominance of impurity scattering for the mobility, and this allows us to compute an estimate of the actual scattering time from the momentum relaxation time. In Appendix 3, we plot

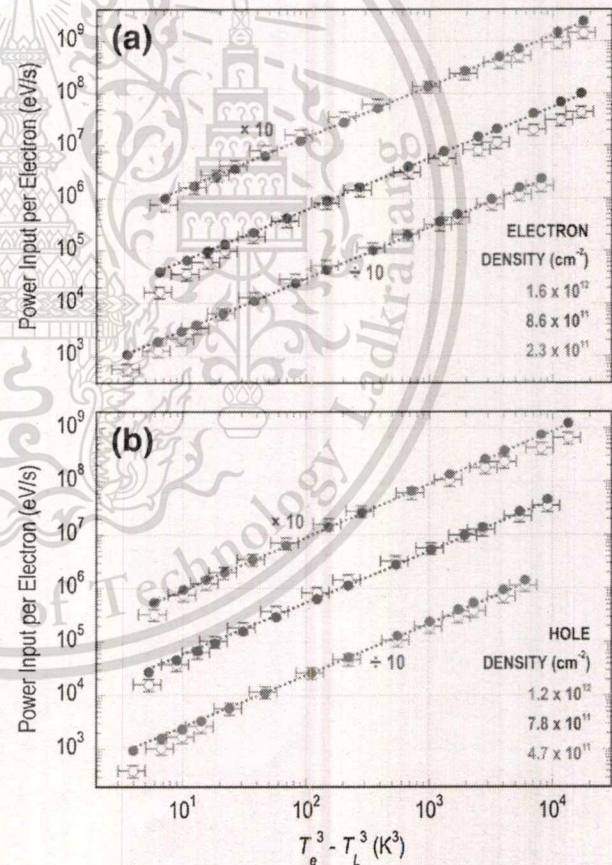
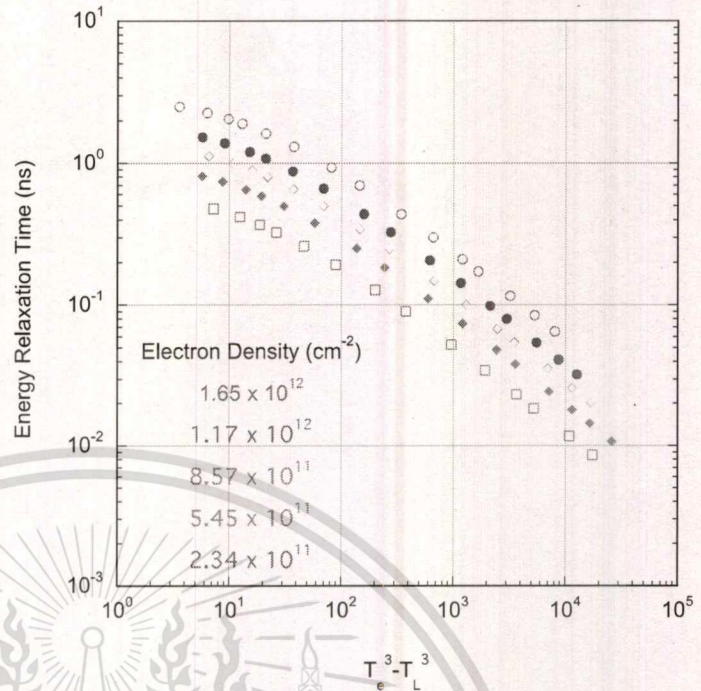


Fig. 2 The energy loss rate per carrier for **a** Electrons and **b** holes. In each case, the *solid symbols* are the theory (11) and the *open symbols* are the data from Ref. [16]. The *color* corresponds to a particular density and *error bars* are shown for the experimental data only. *Dotted lines* through the theoretical data are guides to the eye that indicate a linear variation of power input with T^3 (Color figure online)

Fig. 3 Variation of the energy relaxation time with temperature for several values of the electron density



the mobility that is measured in the samples as a function of temperature for various densities.

We are now ready to show an experimental test of the theory developed here, and this is a direct comparison of the main results of the present work with those of Ref. [16]. There, we showed an energy-loss rate described by an exponent p close to 3, while the energy-relaxation time was found to decrease significantly as the Dirac point was approached from either the conduction or valence band. In Fig. 2, we compare the computed energy-loss rate per carrier to the experimental values determined for different electron and hole densities in monolayer graphene. It may be observed that there is some curvature in the experimental data (open symbols) that is not matched by the theory. This could arise from the variation of the Bose integral, or from a temperature variation of the scattering time, which has not been considered here. In general, though, the agreement is quantitatively good, which is remarkable as there are essentially no adjustable parameters other than the fit to the scattering rate.

4 The energy relaxation time

Another important parameter for hot carrier cooling is the energy relaxation time. Normally, this is obtained from (11) by factoring the cubic terms to give a definition of the energy relaxation time from the theoretical parameters [53]

$$P_e = \frac{k_B (T_e - T_L)}{\tau_e}, \tag{12}$$

where P_e is the power loss per electron. It is τ_e that actually describes the time over which the energy is taken out of the streaming term (Fig. 1). In Fig. 3, we compare the temperature-dependent variation of τ_e found from the present theory. To obtain the theoretical values, the electron temperature that is found from the excitation current at a particular density [16] is used to determine the power lost, and this is introduced into (12) to compute τ_e . From this figure, we see that the plasmon-based model reproduces not only the low-temperature magnitude of τ_e , but that it also captures its quantitative dependence on temperature.

The most striking observation of Ref. [16] was of a strong variation of τ_e as a function of carrier density. This behavior is reproduced in Fig. 4, along with the results of our calculations. The main panel compares the behavior observed at a number of different electron temperatures, and again shows close, if not perfect, agreement between experiment and theory. One may note that there is an asymmetry between the electron and hole sides of the plot, the origin of which is a similar asymmetry in the gate-voltage dependent conductance measured in experiment [16]. On the electron side, it appears that there is a soft saturation setting in above $\sim 10^{12} \text{ cm}^{-2}$, which is not apparent on the hole side. Whether this has significance is not clear at this time. The electron temperature is extracted from the experimental data, and we note that the sample was chemically-doped such that a positive gate bias was necessary to bring the Fermi energy to the Dirac point. It is possible then that this negative charge could have induced some asymmetry into the results. In the inset to Fig. 4, we plot the variation of τ_e as a function of electron density, for

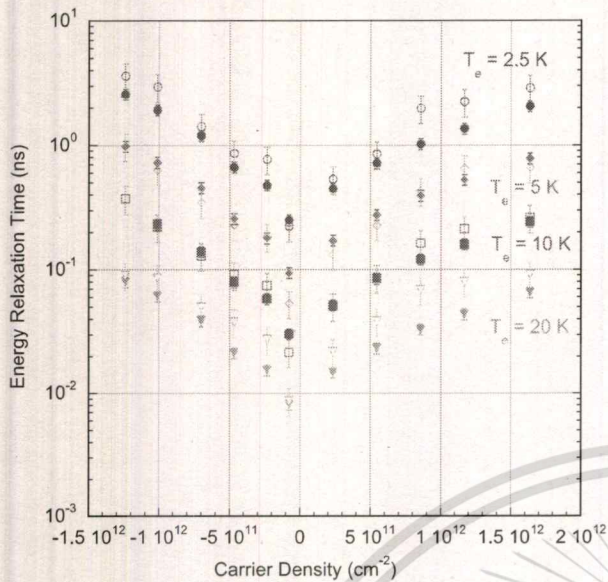


Fig. 4 Comparison of the calculated energy relaxation time with that measured in Ref. [16] for several values of the electron temperature. The *open symbols* are experimental data while the *solid symbols* are the theoretical values

three different measurement currents. As with the data in the main panel, we see that the theory reproduces the experiment well in this figure also.

The plasmon loss mechanism rearranges the energy distribution function, but this energy remains in the electron gas as a whole. Under the excitation current (or electric field), the distribution function is shifted in momentum space to reflect the current, and it spreads as the electron temperature rises. The shift can also distort the distribution to produce an elongated extension in the direction of the field or current. The ultimate limit of this elongation is a delta function in the field direction [4]. Between the shift in momentum space, and the elongation, the distribution function has more carriers in the forward direction than the backward direction. The plasmon loss takes carriers from the streaming forward direction and moves them to the backward direction, in the symmetric terms of the distribution function, on the average. This relaxes the streaming distribution, but the ultimate result still remains that another process must pass the energy to the lattice. This can be due to the acoustic phonon process [22–27] or even the super-collision process [26]. However, it is the plasmon interaction that dominates what is seen experimentally as it is this process which moves the current carrying particles out of the forward extension of the distribution function. The other processes which ultimately move the energy to the lattice are thus masked, and this would be even more true with the supercollision process as it has the same temperature dependence (i.e., $p = 3$) as that found here.

In conclusion, the good quantitative agreement between the experimental data of Ref. [16] and the theoretical results obtained here suggests that the plasmon interaction dominates hot-carrier energy relaxation in graphene at low temperatures. Since the role of this mechanism in governing energy loss does not appear to have been appreciated thus far, it suggests that its relevance may be far wider than simply the experimental study analyzed here.

Appendix 1

We note that the emission and absorption of plasmons by hot carriers is not the normal Fermi liquid behavior. If the Fermi sea is completely full at zero Kelvin, then there can be no emission at the Fermi level since the lower energy states are full. We need a spreading of the distribution function at an elevated temperature arising from the hot electron effect, induced by the electric field as shown in Fig. 1. The phase space interaction, illustrating conservation of both energy and momentum can be sketched as in Fig. 5, for both emission and absorption. Here, we show two circles which are the constant energy rings for the states $E(\mathbf{k})$ and $E(\mathbf{k} \pm \mathbf{q})$. These rings are separated by the plasmon energy, which is q dependent. The red ring is the Fermi circle for the initial state, and the blue ring is the Fermi circle for the final state. The initial and final momenta are, of course, connected vectorially by the momentum of the plasmon, so that momentum conservation, as well as energy conservation, is assured in the interaction. Panel (a) represents the situation for plasmon emission, while (b) represents the situation for plasmon absorption.

We begin by writing the energy conservation for the emission process in terms of the magnitudes of the various vectors as

$$\hbar v_F (|\mathbf{k}| - |\mathbf{k} - \mathbf{q}|) = \hbar \omega_p = \hbar \sqrt{\chi 2q} \quad (13)$$

where the plasma frequency has been given in the main text by (3). Here, we have introduced the proper dynamic mass for graphene in the last term. We can now expand the terms in the first equation to get

$$k - \sqrt{k^2 + q^2 + 2kq \cos \vartheta} = \frac{\sqrt{\chi 2q}}{v_F} \quad (14)$$

It is important to note here that the vector properties of Fig. 5a mean that $\cos \vartheta < 0$, or that the angle is in the second or third quadrants. We can now rearrange the terms and square the result to get

$$k^2 + \frac{\chi 2q}{v_F^2} - 2k \frac{\sqrt{\chi 2q}}{v_F} = k^2 + q^2 + 2kq \cos \vartheta, \quad (15)$$

Fig. 5 Diagrams for the conservation of momentum and energy in the plasmon interaction for **a** emission and **b** absorption

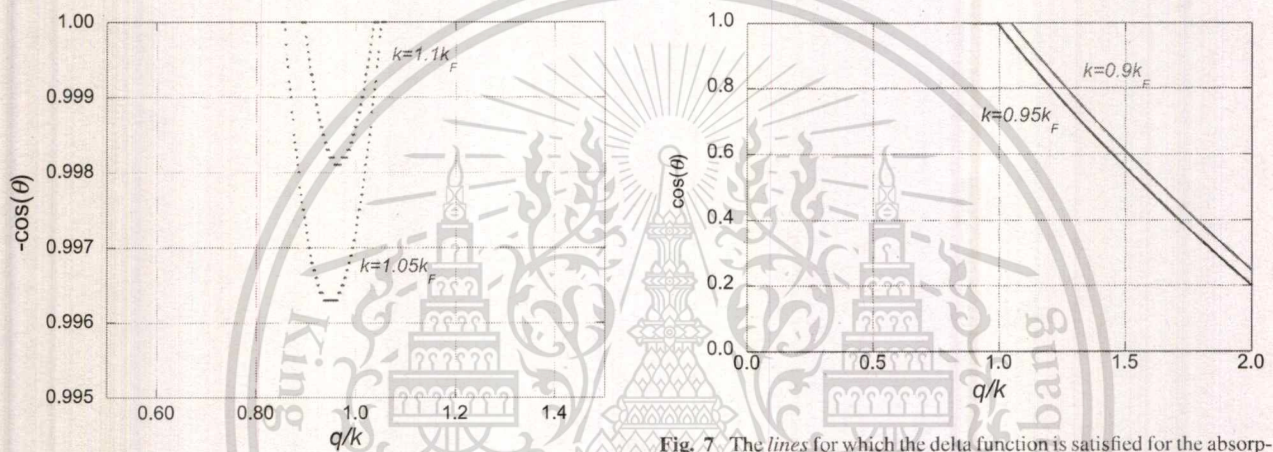
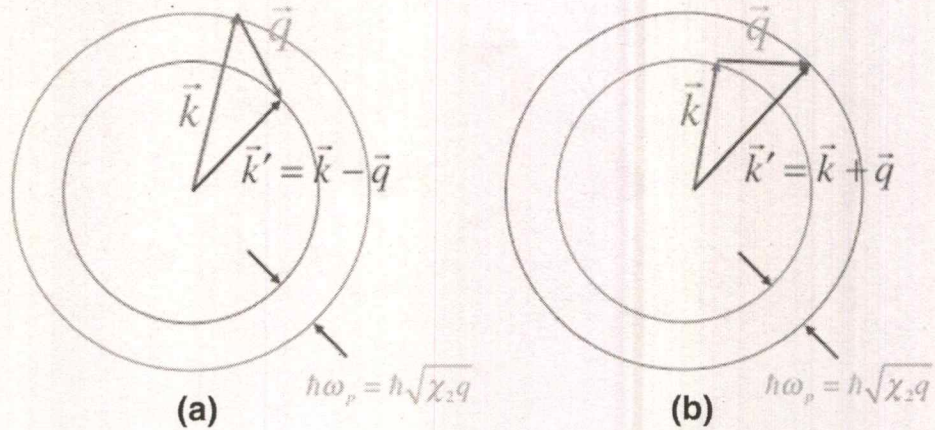


Fig. 6 The lines for which the delta function is satisfied for the emission of plasmons at two different values of energy of the carriers, as given by (16)

Fig. 7 The lines for which the delta function is satisfied for the absorption of plasmons at two different values of energy of the carriers, as given by (20)

or

$$\cos \vartheta = -\frac{q^2 - \frac{\chi_2 q}{v_F} + 2k \frac{\sqrt{\chi_2 q}}{v_F}}{2kq}$$

$$= -\left[\frac{q}{2k} - \frac{\chi_2}{2kv_F^2} + \frac{1}{v_F} \sqrt{\frac{\chi_2}{q}} \right]. \tag{16}$$

It is important to note here that the vector properties of Fig. 5b mean that $\cos \vartheta > 0$, or that the angle is in the first or fourth quadrants. We can now rearrange the terms and square the result to get

$$k^2 + \frac{\chi_2 q}{v_F^2} + 2k \frac{\sqrt{\chi_2 q}}{v_F} = k^2 + q^2 + 2kq \cos \vartheta, \tag{19}$$

or

$$\cos \vartheta = \frac{-q^2 + \frac{\chi_2 q}{v_F^2} + 2k \frac{\sqrt{\chi_2 q}}{v_F}}{2kq}$$

$$= \left[-\frac{q}{2k} + \frac{\chi_2}{2kv_F^2} + \frac{1}{v_F} \sqrt{\frac{\chi_2}{q}} \right]. \tag{20}$$

Thus, we require the term in square brackets to be positive. The solutions are shown in Fig. 6.

Now, we turn to the absorption process and write the energy conservation in terms of the magnitudes of the various vectors as

$$\hbar v_F (|\mathbf{k} + \mathbf{q}| - |\mathbf{k}|) = \hbar \omega_p = \hbar \sqrt{\chi_2 q}. \tag{17}$$

We can now expand the terms in the first equation to get

$$\sqrt{k^2 + q^2 + 2kq \cos \vartheta} - k = \frac{\sqrt{\chi_2 q}}{v_F}. \tag{18}$$

Thus, we require the term in square brackets to be positive (Fig. 7).

It is clear that there are solutions to the above equations which allow for both emission and absorption of plasmons.

In the emission case, the scattering lies in a very small range of angles whose “cone” is $6^\circ\text{--}10^\circ$ for the curves shown. This is around the back-scattered direction and has a momentum almost equal to that of the initial state. Hence, this is almost back scattering, but not quite (we note that pure back-scattering in graphene is forbidden in the equilibrium case by chirality). For absorption, there is a very large range of angles for the forward process, but again it occurs for rather large values of the momentum. In both emission and absorption, the scattering involves rather large values of the plasmon energy. There is almost no density dependence in the range of $10^{11} - 10^{12} \text{ cm}^{-2}$, but one would expect the interaction to get stronger nearer to the Dirac point, although the plasmon may be damped strongly in this case by the interband single particle interactions discussed above.

Appendix 2

The integral in (10) may be written as

$$I = \int_0^{u_{\max}} \frac{u^2 du}{e^u - 1} \tag{21}$$

where

$$u = \frac{\hbar v_F q}{k_B T}, \quad u_{\max} = \frac{2\hbar v_F k_F}{k_B T} \tag{22}$$

The first term arises from the energy conserving delta function that appears in the integration over the frequency. The second term arises from the fact that the maximum value of q that can occur is approximately the span of the Fermi circle in two dimensions, which translates into a maximum value for u . Generally, this maximum is twice the ratio of the Fermi energy to the thermal energy, and can be quite large. In Fig. 8, we plot both the integrand and the value of the integral as a function of u or u_{\max} , as appropriate. It may be seen that the peak of the argument occurs at a relatively small value of u , due to the dominance of the exponential in the denominator. The integral approaches its limiting value fairly rapidly. The smallest value of the limit occurs for low density and high temperature. In the experiments [16], the lowest value of density is about $2 \times 10^{11} \text{ cm}^{-2}$ while the highest carrier temperature is about 40 K. These numbers lead to a minimum value of u_{\max} of about 2.4 where the integral has already reached more than half its final value. So, except for low densities at the highest input power per electron, the integral can reasonably be assumed to be the limiting value of approximately 2.4.

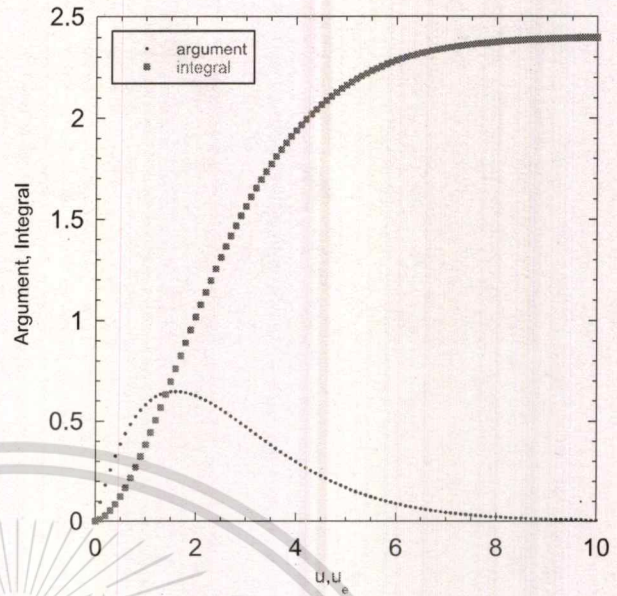


Fig. 8 The argument and value of the integral (21) as a function of the argument or limit

Appendix 3

The mobility that is measured in the samples as a function of temperature for various densities is shown in Fig. 9. To get to the needed scattering time, one must understand how the various scattering rates are averaged over the scattering angle (Sec. e.g., [56]). With several scattering mechanisms active in graphene, this is actually rather difficult to unfold. We have considered impurity scattering to be dominant. The actual scattering rate differs from the momentum relaxation

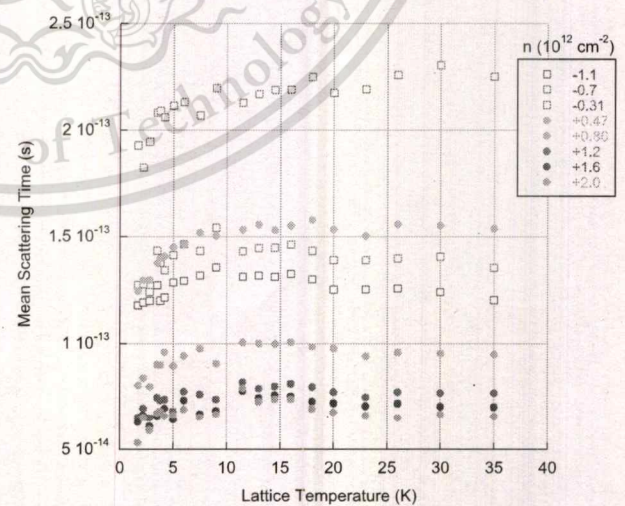


Fig. 9 The momentum relaxation time as a function of temperature for several values of the carrier density

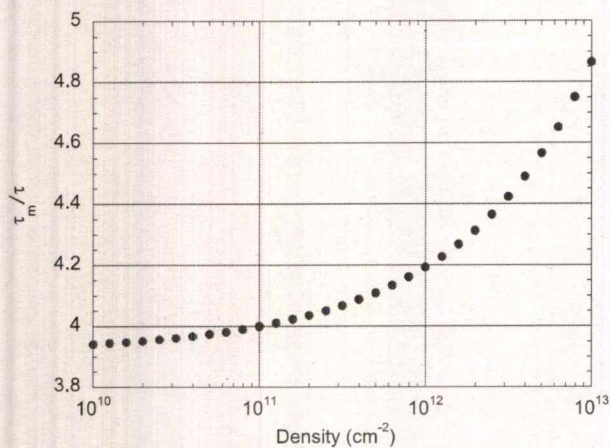


Fig. 10 The ratio of the momentum relaxation time to the actual scattering time for ionized impurity scattering in graphene

time determined from the measured mobility. The actual scattering rate from ionized impurity scattering in graphene can be written as [57]

$$\frac{1}{\tau_{imp}} = \frac{N_{imp}}{4\pi\hbar E_k} \left(\frac{Ze^2}{\epsilon_s}\right)^2 \int_0^\pi \frac{[1 + \cos(2\vartheta)] e^{-4k \sin(\vartheta) d_s} d\vartheta}{[\sin(\vartheta) + \frac{q_s}{2k}]^2} \quad (23)$$

where $e^2 k_F / 2\pi \epsilon_s$ is the screening wave vector and 2θ is the angle between the incoming and scattered wave vectors—the elastic scattering occurs with $q = 2k\cos(\theta/2)$, and the angle has been doubled in the equation. In the momentum relaxation time, there is an additional factor of $[1 - \cos(2\theta)]$ in the numerator (See, e.g., [58]). So, the ratio of the momentum relaxation time to the actual scattering time is the ratio of the integrals with, and without, this additional term. This ratio is shown in Fig. 10, with the scattering rates all evaluated at the Fermi surface, appropriate for low temperatures. In graphene, the mobility in suspended graphene is quite high, and can be of the order of several times $10^5 \text{ cm}^2/\text{s}$ [59]. The mobility in the experiments discussed in Ref. [16], and used in the manuscript, are much lower and so are assumed to be dominated by impurity scattering. Hence, the ratio in Fig. 6 was used to convert the momentum relaxation time into the actual scattering time used in the computation of the energy loss rate per electron.

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Conduction Mechanisms in CVD-Grown Monolayer MoS₂ Transistors: From Variable-Range Hopping to Velocity Saturation

G. He,[†] K. Ghosh,[†] U. Singiseti,[†] H. Ramamoorthy,[†] R. Somphonsane,[‡] G. Bohra,[†] M. Matsunaga,[§] A. Higuchi,[§] N. Aoki,[§] S. Najmaei,^{||} Y. Gong,^{||} X. Zhang,^{||} R. Vajtai,^{||} P. M. Ajayan,^{||} and J. P. Bird^{†,*}

[†]Department of Electrical Engineering, University at Buffalo, The State University of New York, Buffalo, New York 14260-1900, United States

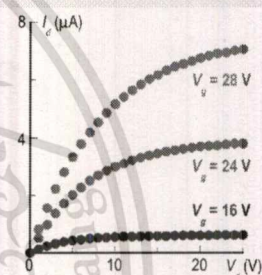
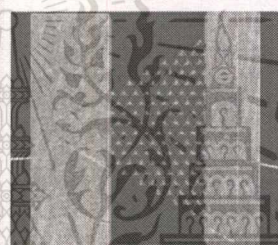
[‡]Department of Physics, King Mongkut's Institute of Technology Ladkrabang, Bangkok 10520, Thailand

[§]Graduate School of Advanced Integration Science, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan

^{||}Department of Materials Science and NanoEngineering, Rice University, Houston, Texas 77005, United States

Supporting Information

ABSTRACT: We fabricate transistors from chemical vapor deposition-grown monolayer MoS₂ crystals and demonstrate excellent current saturation at large drain voltages (V_d). The low-field characteristics of these devices indicate that the electron mobility is likely limited by scattering from charged impurities. The current–voltage characteristics exhibit variable range hopping at low V_d and evidence of velocity saturation at higher V_d . This work confirms the excellent potential of MoS₂ as a possible channel-replacement material and highlights the role of multiple transport phenomena in governing its transistor action.



KEYWORDS: Molybdenum disulfide, transition metal dichalcogenides, 2D transistors, high-field transport, velocity saturation

Transition-metal dichalcogenides have recently emerged as a new class of electronic materials, which, much like graphene, may be isolated in a two-dimensional (2D) form to implement a variety of novel electronic devices.¹ Among the materials that comprise this group, molybdenum disulfide (MoS₂) is one whose characteristics appear particularly promising for such applications. In marked contrast to graphene, MoS₂ exhibits a large energy gap (1.3–1.8 eV), the direct nature of which in the monolayer form makes it attractive for not only transistor implementations but also optoelectronics. Efficient transistor action has been demonstrated for devices based on multilayer and monolayer MoS₂,^{1–23} and these devices are characterized by large (>10⁷) room-temperature ON-OFF ratios. They are also predicted to exhibit excellent subthreshold swing and benefit from immunity to short-channel effects.²⁴ Although the large effective mass ($m^* = 0.45m_0$) of the MoS₂ electrons means that they exhibit much lower mobility than the massless Fermions of graphene, this is not expected to be problematic for many applications involving transistor switching. In one pioneering study, MoS₂ transistors were used to implement small-scale integrated circuits,²⁵ providing essential Boolean functionality. Other work has demonstrated their application as sensitive photodetectors^{26,27} capable of operating in the visible and UV ranges. Additionally, and in common with graphene, the ability to exploit valley polarization as a state variable in (monolayer) MoS₂ may open up the development of new “valleytronic” devices.^{28–30}

In the vast majority of studies reported in the literature to date, MoS₂ transistors have been fabricated by mechanically exfoliating 2D material onto an insulating substrate from crystalline molybdenite.^{2–9,13–18,20,22,23,25,26} Although this has the benefit of allowing rapid prototyping of individual devices, chemical routes to synthesis^{10–12,19,21,31–33} offer the prospect of both high material yield and uniformity. Chemical vapor deposition (CVD), involving the sulfurization of molybdenum films deposited on Si/SiO₂ substrates, is one such promising approach.^{11,12,19,31,32} Depending on the initial film thickness a variety of final structures can be realized, including monolayer crystals with areas of many tens of square microns. These structures may then easily be subjected to in situ device fabrication or may be released into solution for transfer to other substrates.¹⁹ Structural characterization (via Raman spectroscopy and atomic-force microscopy) of the CVD-grown crystals confirms their monolayer,^{11,12} highly crystalline³³ character, making this approach particularly attractive for transistor realization. Recently, there have been some reports^{11,12,19} of the electrical characteristics of such devices, suggesting that they are able to compete with or even exceed the performance offered by their exfoliated counterparts. In ref 21, the authors presented a detailed study of the influence of thermal annealing

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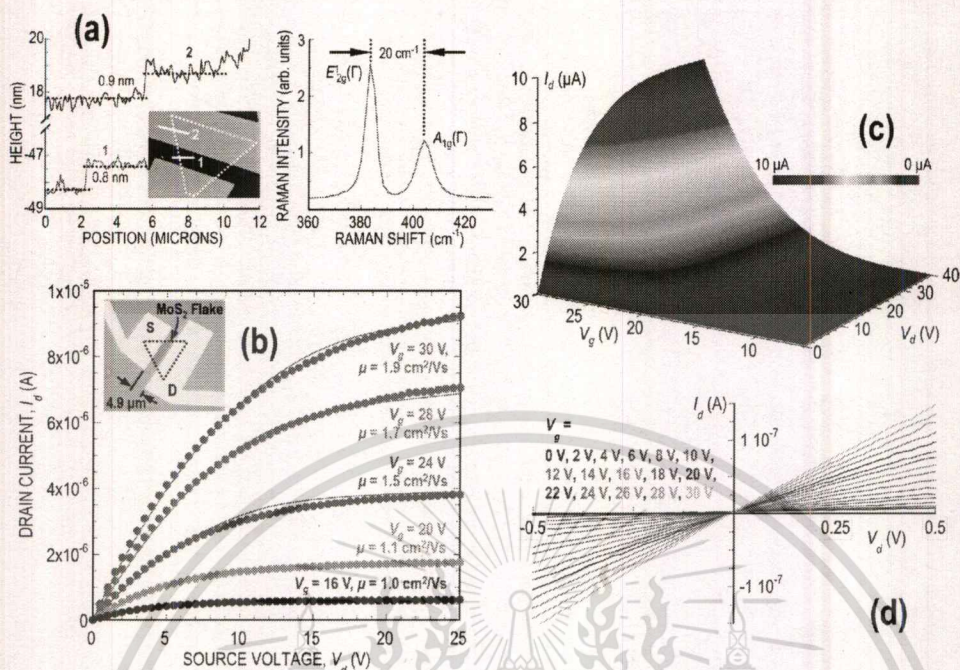


Figure 1. (a) Shown on the left are line scans recorded along the two white solid lines in the AFM image shown in the inset. The image is of MS2-13, and the outline of the MoS₂ crystal is indicated by the white dotted line. The figure on the right is a Raman spectrum obtained using a (514.5 nm wavelength, 1 μm spot size) laser to illuminate the channel of MS2-13. (b) Transistor curves measured at room temperature for transistor MS2-13 (shown in the false-color optical micrograph in the inset). Filled symbols are experimental data points, and solid lines represent the results of simulations (see main text for details). (c) Color contour plot showing the full evolution of $I_d(V_d, V_g)$ for device MS2-13. (d) Expanded view of the transistor curves around $V_d = 0$, showing the linearity of the current over the full range of V_g .

on the transistor characteristics and showed that this allowed low-temperature (10 K) mobility values as large as $500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ to be obtained. Moreover, the authors were able to demonstrate evidence of a metal–insulator transition as a function of gate voltage, suggestive of the high electronic quality of the CVD-grown crystals.

In this Letter, we present the results of studies of the electrical properties of transistors implemented in CVD-grown, monolayer MoS₂. We demonstrate, for the first time, excellent current saturation in such devices and explore the role of different transport mechanisms in governing the dependence of their drain current (I_d) on the drain (V_d) and gate (V_g) voltages. For drain voltages in the linear regime, we observe clear signatures of variable-range hopping (VRH) in the temperature (T)-dependent conduction, reminiscent of prior studies of exfoliated MoS₂.^{3,13} The VRH weakens with an increase in the gate voltage, presumably as electrons in the channel are able to more effectively screen out the impurity-related background potential. This behavior is accompanied at higher fields by the emergence of velocity saturation. The need to introduce the latter phenomenon arises from the strongly gate-voltage dependent character of the channel mobility (μ), and the saturation velocity ($v_{\text{sat}} = 3 \times 10^6 \text{ cm s}^{-1}$) used to fit the experiment is found to closely match that reported in recent studies of multilayer MoS₂.³⁴ Simulations of the electrostatics within the device confirm that the velocity saturation arises from the presence of a strongly peaked electric field (E_y , where y is the direction along the channel length), which develops in the channel near the drain contact under current saturation.

Monolayer MoS₂ transistors were fabricated using electron-beam lithography and metal lift-off to form Cr/Au (5 nm/50

nm) source-drain contacts to CVD-grown single MoS₂ crystals synthesized on commercial Si/SiO₂ wafers. The heavily doped Si served as the back-gate of the transistor, whereas the 280 nm thick SiO₂ functioned as the gate dielectric and was also used to provide optical contrast when observing the MoS₂ crystals under an optical microscope. (In the ultimate applications, this design could be replaced with one consisting of a top gate formed on a thin layer of high- k dielectric, significantly lowering the working gate-voltage range while also enhancing the mobility through “engineering” of the dielectric environment.²) The CVD growth was performed by first dispersing MoO₃ nanoribbons onto the Si/SiO₂ substrate and then sulfurizing them in a furnace.³² This process resulted in the formation of triangular-shaped single crystals, $13 \pm 2.5 \mu\text{m}$ on a side, whose monolayer character was confirmed by both Raman studies and atomic-force microscopy. This is indicated in Figure 1(a), in which the measured thickness of the MoS₂ crystal and the separation of the E_{2g}(Γ) and A_{1g}(Γ) Raman peaks are both consistent with earlier reports for CVD grown monolayer MoS₂.^{11,21}

The electrical characteristics of 13 different transistors were investigated for this study and were all found to exhibit fairly similar characteristics (see Table 1 of the Supporting Information, where we also define the notation used to identify the different transistors). For this reason, in our discussions here we focus on providing the results of a comprehensive study of a single device (MS2-13), which we have chosen due to its representative character. This device is shown as an inset in Figure 1(b), where the source (S) and drain (D) contacts are indicated. After fabrication, transistors were wirebonded into standard DIP packages and mounted in a cryostat, allowing

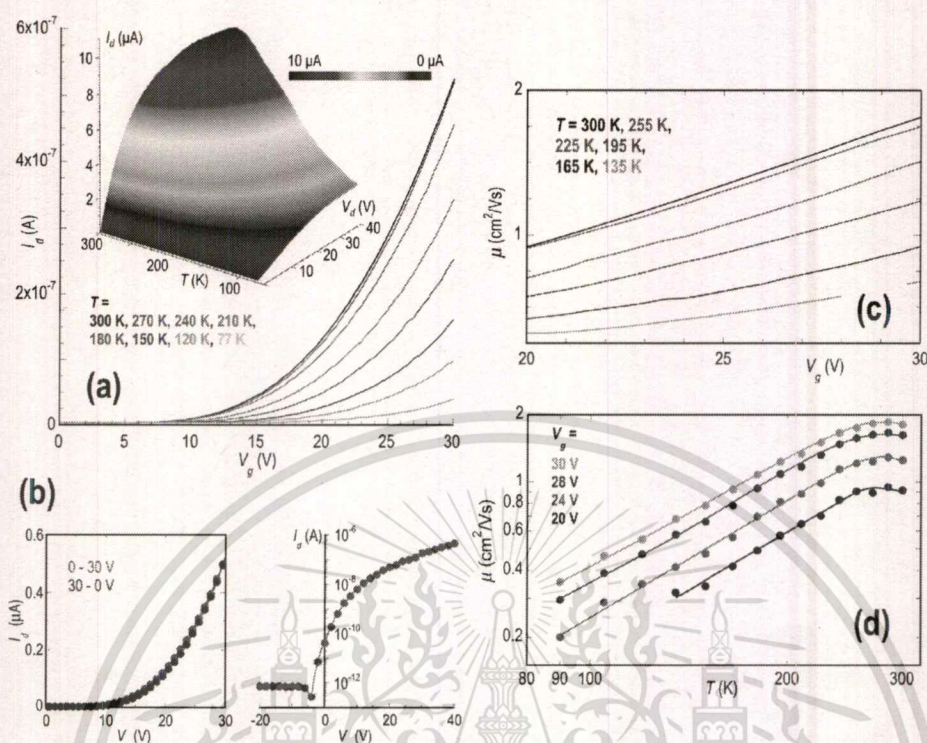


Figure 2. (a) Transfer characteristics of device MS2-13 ($V_d = 0.5$ V), measured at various temperatures from 77 to 300 K. The inset plots the variation of $I_d(V_g)$ as a function of temperature from 77 to 300 K. The gate voltage ($V_g = 30$ V) was held fixed for these measurements. (b) The left panel shows the transfer curve of MS2-13 measured at 300 K in both sweep directions (red and blue data). In the right panel, we use a semilog plot to reveal an on-off ratio at 300 K of ~ 6 orders of magnitude. The measurement was made with $V_d = 1$ V using a Keithley 4200 with a high resolution preamplifier. (c) Variation of mobility as a function of V_g , inferred (using eq 2) from the experimental data of (a) at different temperatures. (d) Variation of mobility as a function of temperature, inferred from the results of (c) at various gate voltages. The solid lines are trend lines that form a guide to the eye.

their electrical characteristics to be measured from 77 to 300 K with a Keithley 2400 source meter. The measurements reported here were obtained with the samples maintained under vacuum, in the dark, and in the absence of any annealing.^{15,21} We have performed both in situ (under vacuum, 110 °C, 10^{-5} mbar, 20 h) and ex situ (also under vacuum, 120 °C, 10^{-5} mbar, 20 h) annealing of our devices, but in contrast to earlier reports,^{15,21} do not find such a significant improvement in performance. The key features of transistor action (including the well-defined current saturation that we show below) are preserved after annealing, and current levels are increased by $\sim 50\%$, but this improvement is much less dramatic than that achieved in refs 15 and 21. The reasons for this are unclear at present and require further investigation.

In Figure 1(b) and (c), we show measurements of the transistor curves of device MS2-13 at room temperature. Figure 1(b) shows the characteristics measured at several discrete gate voltages, along with corresponding theoretical fits (solid lines), whose details are provided further below. Figure 1(c) is a color contour that expresses the evolution of I_d as a continuous function of V_d and V_g . Both figures reveal classic transistor action with excellent saturation at high drain voltages, something that has not been demonstrated for CVD-grown MoS₂ devices (for which previous works have focused primarily on analyzing the transfer characteristics^{11,12,15,19,21}). In Figure 1(d), we confirm the linear character of the current for small V_d and for a wide range of V_g . Typically, such linearity in two-

terminal measurements is taken to indicate that good ohmic contact has been achieved to the MoS₂ channel.²¹

In the main panel of Figure 2(a), we present measurements of the transfer (I_d - V_g) characteristic of transistor MS2-13 at a series of temperatures from 77 to 300 K. As indicated in Figure 2(b), the transfer curve showed little hysteresis as a function of sweep direction (left panel), and yielded a room-temperature on-off ratio of ~ 6 orders of magnitude (right panel). Regardless of the temperature in Figure 2(a), it is noted that none of the curves show the linear variation $I_d(V_g)$ expected from a simple square-law model³⁵

$$I_d = \frac{W\mu}{L} \frac{\epsilon_{ox}\epsilon_0}{x_o} \left[(V_g - V_T)V_d - \frac{V_d^2}{2} \right] \quad (1)$$

Here, V_T is the threshold voltage, L is the channel length, x_o is the oxide thickness, and ϵ_{ox} is its dielectric constant. W is the channel width, which in this work is taken as the average width of the MoS₂ crystal at the source and drain ends. μ is an effective channel mobility and, when this parameter is independent of gate voltage, its value can be determined from the linear relation between the drain current and the gate voltage [i.e., $\mu = (dI_d/dV_g)(x_o/(\epsilon_{ox}\epsilon_0))(L/WV_d)$]. The nonlinearity of the curves in Figure 2(a) indicates that the mobility in these devices is gate-voltage dependent however, requiring us to determine μ by solving the following expression (for details see the Supporting Information).

$$\mu + (V_g - V_T) \frac{d\mu}{dV_g} = \frac{L}{W} \frac{x_0}{\epsilon_{ox}\epsilon_0} \frac{1}{V_d} \frac{dI_d}{dV_g} \quad (2)$$

In Figure 2(c), we plot the variation of $\mu(V_g)$ determined from the experimental transfer curves at various temperatures using eq 2. The mobility clearly increases in an almost linear manner with increasing V_g and is also systematically larger at higher temperatures. This latter point can be seen explicitly in Figure 2(d), where we plot the variation of $\mu(T)$ at four different values of V_g . The behavior here is similar to that found previously for exfoliated MoS₂¹⁷ and is consistent with the role of charged-impurity scattering.^{1,5} The temperature dependence of the mobility is reflected as a strong variation of the transistor curves, as we indicate in the inset to Figure 2(a). This color contour plots the I_d - V_d characteristic of transistor MS2-13 at fixed gate voltage and at a series of different temperatures. As the mobility increases with temperature, the overall current level exhibits significant growth. At every temperature, however, we observe clear signatures of the linear- and saturated-current regimes, indicating that the transistor action is robust.

Turning to the issue of the conduction mechanism in the MoS₂ transistors, this is strongly dependent upon both the drain and gate voltages. For small V_d , we observe clear signatures of 2D VRH in the resistance^{3,13}

$$R \propto R_0 \exp\left[\left(\frac{T_0}{T}\right)^{1/3}\right] \quad (3)$$

where R_0 and T_0 are appropriate fit parameters. In Figure 3(a), we show the variation of the resistance (R) as a function $T^{-1/3}$

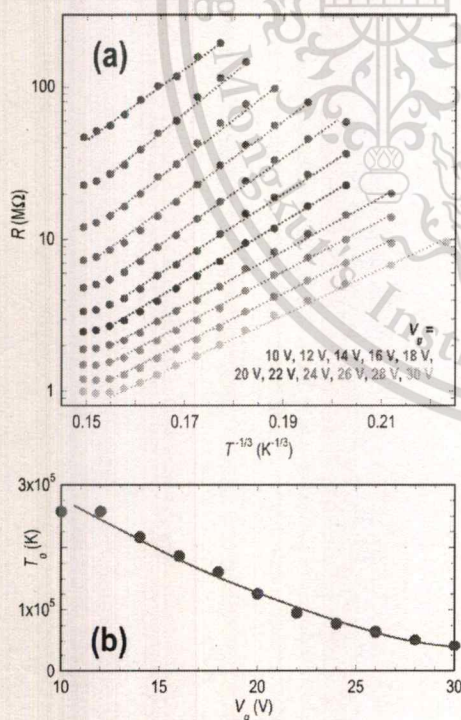


Figure 3. (a) Temperature-dependent variation in the resistance of device MS2-13 plotted to reveal the connection to VRH. A source voltage of $V_d = 1$ V was used for these measurements. (b) Gate-voltage-dependent variation of the VRH parameter T_0 (see eq 3).

(on a semilog plot) and obtain good agreement with the VRH form for a wide range of gate voltages. The resistance values plotted here were determined at $V_d = 1$ V, well within the linear regime of transistor operation. In Figure 3(b), we plot the variation of T_0 , inferred from the data of Figure 3(a), as a function of the gate voltage. Clearly, there is a trend for T_0 to decrease with an increase in V_g , indicating that the role of the VRH is correspondingly reduced. We have already noted that the increase in mobility that we observe upon increasing temperature is indicative of the role of strong charged-impurity scattering. The related observation of VRH suggests that these charged impurities, which are presumably associated with the substrate,³ also give rise to the disordered potential landscape needed for hopping to dominate. As the carrier concentration is increased by raising the gate voltage, screening of the impurity potential should then suppress the role of the hopping. MoS₂, like graphene, is known to be characterized by inefficient screening, particularly when prepared in few-layer form.^{36–39} Estimates for the screening length vary, dependent upon the number of layers, but are typically in the range of several nanometers.^{36–39} Screening of the impurity potential will therefore be incomplete in the devices, but we nonetheless expect that it should improve as the carrier concentration increases.⁴⁰ From our SILVACO simulations described in further detail below, we find that an increase in the gate voltage from 16 to 30 V should raise the carrier concentration in the MoS₂ from 0.4×10^{12} to 2.2×10^{12} cm⁻². According to recent theoretical work,⁴⁰ the increased density should lead to improved screening, and it is this phenomenon that we suggest is responsible for the modest improvements in mobility achieved in Figure 2(c).

For a more quantitative understanding of transistor action, we have simulated the performance of the devices using the SILVACO Atlas package.⁴¹ Details of the calculations are provided in the Supporting Information, along with a full list of the material parameters used. The essential idea, however, is that the monolayer MoS₂ crystal is treated as a 0.5 nm thick film, and a classical drift-diffusion model is used to simulate the drain current. The calculations explicitly consider the triangular shape of the MoS₂, and use micrographs of the actual device to determine the orientation of the crystal relative to the contacts. To account for the influence of velocity saturation, a field-dependent mobility ($\mu(E_y)$) is used, such that

$$\mu(E_y) = \mu_0 \left[\frac{1}{1 + (\mu_0 E_y / v_{sat})^\beta} \right]^{1/\beta} \quad (4)$$

where E_y is the electric field along the direction of current flow. Because the conduction in these devices is dominated by electrons, we take the parameter $\beta = 2$.⁴² The low-field mobility (μ_0) in eq 4 is then determined by fitting the drain current to match that obtained in the linear region of the transistor curves.

Using the approach described above, we are able to successfully reproduce the experimental transistor curves of Figure 1(b). In doing so, we infer low-field mobility values in good agreement with those determined from the experimental data. This is illustrated in the inset to Figure 4(b), where we plot as discrete points the mobility values used in our simulations and compare them with those determined experimentally at room temperature. The agreement between the simulation and experiment is good, being within the error bars associated with the former. (The error bars reflect

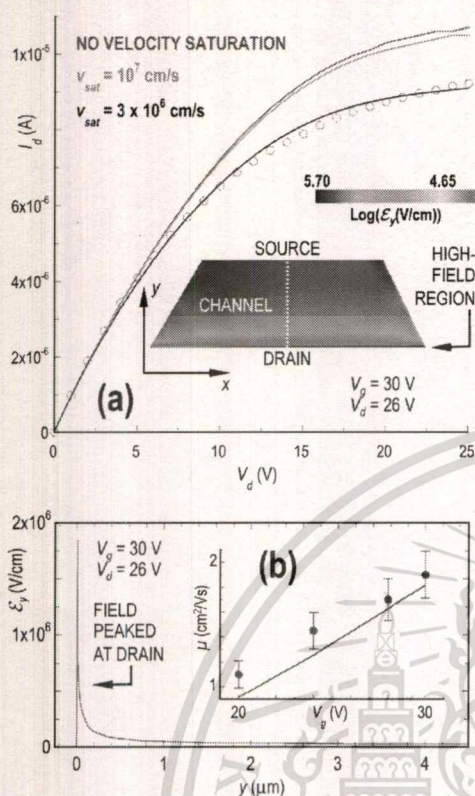


Figure 4. (a) Simulated fits to the drain current ($V_g = 30$ V), using different values for v_{sat} (indicated). A saturated velocity of 3×10^6 cm s^{-1} is found to best fit the experimental data (open symbols). The inset plots the calculated electric field strength (E_y) as a function of position within the channel (for $V_g = 30$ V, $V_d = 26$ V). The field strength is plotted on a log scale here (see color bar) and is strongly peaked at the drain end. (b) Variation of E_y as a function of position along the center line of the channel, indicated by the white dotted line. $y = 0$ corresponds to the drain end of the device. $V_g = 30$ V; $V_d = 26$ V. (inset) Comparison of experimentally determined mobility at 300 K with that used in the simulations to match the transistor curves. Device MS2-13. Solid line: experimental data. Filled symbols: simulated mobility values.

primarily the variation that arises when computing the mobility using the width of the triangular channel at its source and drain ends. The data points themselves were obtained by using an averaged value for this width.)

An important feature revealed by our simulations is that current saturation at large V_d results from an interplay between channel pinch-off at the drain end and velocity saturation of electrons in the channel. At low gate voltages, where the mobility is small, the current saturation results primarily from pinch-off, whereas the converse is true at higher gate voltages. Referring to the transistor curves of Figure 1, for example, for gate voltages lower than 25 V, our simulations show that the drain current is generally consistent with a simple square-law model without the need for any velocity saturation. For larger gate voltages, however, agreement with the experiment is only possible by properly accounting for velocity saturation. This can be seen in Figure 4(a), where we show how the choice of different values for v_{sat} influences the computed current. The experimental data are plotted as open symbols in the figure, and it is clear that, with no velocity saturation (green line) or with

an assumed $v_{\text{sat}} = 10^7$ cm s^{-1} (blue line), the simulated current is significantly larger than the experimental current. With $v_{\text{sat}} = 3 \times 10^6$ cm s^{-1} , a value suggested in previous work on multilayer MoS_2 transistors,³⁴ however, the agreement is very good. In the inset to Figure 4(a), we plot a 2D surface contour of the channel field (E_y) using a logarithmic scale. The figure reveals that the velocity saturation is due to the strongly peaked nature of this field near the drain, a feature that can also be seen in Figure 4(b). Here, we plot the variation of E_y as a function of position (y) along the center line indicated by the white dotted line in the color contour of Figure 4(a). The field at the drain end approaches 2×10^6 V cm^{-1} , from which value and the mobility of 1.9 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$, we obtain an estimate for the saturated velocity of 3.8×10^6 cm s^{-1} . This is consistent with our discussion above regarding the influence of the velocity saturation on the drain current in Figure 4(a).

The mobility values determined here ($\sim 1\text{--}10$ $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$, see Table 1 in the Supporting Information) are, in many cases, much lower than those reported elsewhere. Putting aside the fact that the estimates provided in many of these studies have been called into question by others,⁴³ there are three primary issues that are likely at play here. First, our measurements are performed on unannealed devices, whereas thermal annealing has been demonstrated to provide improvements in the mobility.^{15,21} Second, and perhaps more importantly, our work here shows that the mobility is strongly gate-voltage dependent, such that further increasing the gate voltage beyond the threshold can be expected to result in larger mobility values. In ref 21, for example, room-temperature mobility as large as $50\text{--}60$ $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ was reported but was achieved by using back-gate voltages approaching 100 V. Finally, our two-terminal measurements may be influenced by contact resistance, which would result in an underestimate of the mobility. Although contact resistance was not included in the simulations shown in Figure 4, we have performed calculations in which the source/drain contact resistance was increased up to 1 $\text{k}\Omega$ (inclusion of larger values than this introduces problems with convergence) and did not observe any significant change in drain current. This can be attributed to the much lower conductance of the FET channel itself.

It is informative to compare the results of our study with those found in similar investigations of multilayer MoS_2 transistors. We have seen here that the current saturation in our devices arises from a combination of usual electrostatics (i.e., pinch-off at the drain) supplemented by velocity saturation that becomes important as the channel mobility improves. Current saturation has also been reported for multilayer devices^{5,7,18,20,34} and has similarly been discussed in terms of long-channel pinch-off⁵ and velocity saturation.³⁴ However, the much higher mobility exhibited by multilayer MoS_2 ³⁷ results in saturated current values that may be >2 orders of magnitude larger^{5,20,34} than those reported here. In this sense, it is clear that multilayer devices are more attractive than monolayer ones for applications requiring larger drive currents. Nonetheless, in spite of its lower mobility, monolayer MoS_2 benefits from a larger gap (1.8 eV) than its multilayer counterpart (1.2 eV), offering the potential of a lower off current. Additionally, and in contrast to multilayer MoS_2 , its direct band structure makes monolayer material attractive for various light-emitting devices. In this sense, both monolayer and multilayer devices possess their own advantageous characteristics, making them suitable for very distinct applications.

In conclusion, we have fabricated transistors from CVD-grown monolayer MoS₂ crystals and have demonstrated well-behaved operation in these devices. Excellent current saturation is observed at large drain voltages, and an analysis of the low-field transport indicates that the associated mobility is likely limited by scattering from charged impurities. The dominant conduction mechanisms are strongly dependent upon both the drain and gate biases with clear evidence of VRH at small V_d. With increases in both V_g and V_d, however, a satisfactory description of the characteristics can only be achieved by invoking the role of high-field velocity saturation. The velocity saturation was shown to arise from the presence of a strongly peaked electric field near the drain end of the channel, and the value of v_{sat} inferred from our studies was shown to be consistent with reports for multilayer MoS₂ transistors. This work therefore confirms the excellent potential of MoS₂ as a possible channel-replacement material and highlights the role of multiple transport phenomena in governing its transistor action.

■ ASSOCIATED CONTENT

● Supporting Information

Information on the various devices studied, along with details of the simulation parameters, and a description of the mobility determination from the transfer characteristics. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.5b01159.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: jbird@buffalo.edu. Phone: +1 (716) 645-1015.

Author Contributions

G.H. fabricated the MoS₂ transistors, and G.H., M.M., A.H., and N.A. measured their temperature-dependent electrical characteristics. G.H., R.S., H.R., G.B., S.N., R.V., and J.P.B. collaborated on the design of the experiments. K.G. performed the computational modeling of transistor performance, working in collaboration with U.S., G.H., and J.P.B. CVD growth of MoS₂ crystals was performed by S.N. in collaboration with R.V. and P.M.A. Y.G. and X.Z. performed AFM and Raman microscopy of the fabricated devices.

Notes

The authors declare no competing financial interest.

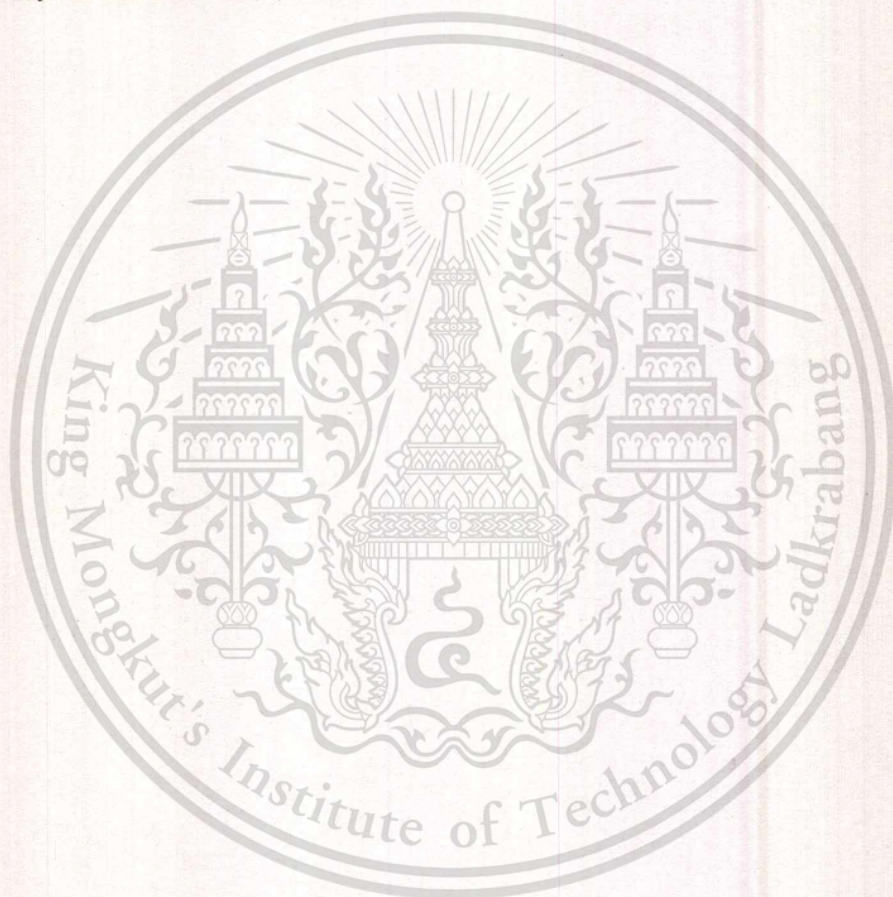
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Reversing hot-carrier energy-relaxation in graphene with a magnetic field

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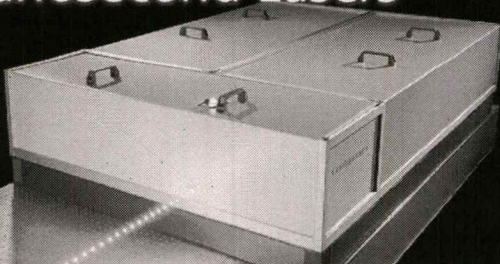
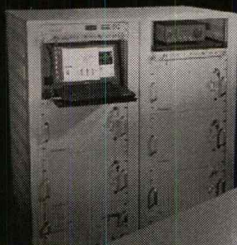
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Reversing hot-carrier energy-relaxation in graphene with a magnetic field

H. Ramamoorthy,¹ R. Somphonsane,² G. He,¹ D. K. Ferry,^{3,4} Y. Ochiai (落合勇一),⁴
 N. Aoki (青木伸之),⁴ and J. P. Bird^{1,4}

¹Department of Electrical Engineering, University at Buffalo, Buffalo, New York 14260-1900, USA

²Department of Physics, King Mongkut's Institute of Technology Ladkrabang, Bangkok 10520, Thailand

³School of Electrical, Computer, and Energy Engineering, and Center for Solid State Electronics Research, Arizona State University, Tempe, Arizona 85287-5706, USA

⁴Graduate School of Advanced Integration Science, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan

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We investigate the influence of a perpendicular magnetic field on hot-carrier energy relaxation in bilayer graphene. Working in the regime of incipient Landau quantization, we find that the magnetic field influences the relaxation in a very different manner, dependent upon the position of the Fermi level relative to the Dirac point. While for carrier densities $>10^{12} \text{ cm}^{-2}$ relaxation is slowed by the magnetic field, as the density of free carriers approaches zero it instead becomes quicker. We discuss this behavior in terms of the emergence of the zero-energy Landau level, and the role of charge puddling in graphene. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4878535>]

Over the past decade, graphene has emerged as the leading two-dimensional material, due to its broad potential for application to a range of electronic technologies.¹ As our understanding of its fundamental transport properties has matured,² a problem that has attracted growing interest concerns the mechanisms for hot-carrier energy loss in this material. Driving ongoing theoretical^{3–11} and experimental^{12–34} interest in this problem are its implications for the development of active devices. In graphene bolometers,^{23,25,34} for example, it is desirable to engineer the environment to strongly decouple nonequilibrium carriers from phonons. Similarly, in photonic devices such as solar cells³⁵ and lasers³⁶ energy transfer from excited carriers to phonons will degrade the optical output. From such examples alone, the need to quantitatively understand the various energy-loss pathways in graphene becomes apparent.

When carriers in graphene are driven out of equilibrium, their excess energy is redistributed by various mechanisms. The fastest of these occurs on a few tens of femtoseconds, as carrier-carrier scattering establishes an effective carrier temperature (T_e).³⁷ At the opposite end of the spectrum, energy exchange between carriers and phonons occurs on the energy-relaxation time (τ_e), which may be as long as nanoseconds at 4.2 K.^{23,28,31} The details of phonon-mediated cooling are predicted to differ significantly for pristine^{3,4} and disordered^{7,9} graphene, with the role of impurity-mediated “supercollisions” being emphasized for the latter.^{7,9,28,29,32} Elsewhere, we have found surprising behavior in which energy relaxation quickens significantly as the Fermi level is swept towards the Dirac point (DP) from either band. This behavior is inconsistent with existing theories^{3,4,7,9} for phonon-cooling pathways, and suggests that the mechanisms for energy relaxation may differ, near to and away from the DP.

A useful means for probing energy relaxation involves the application of a perpendicular magnetic field (B), which modifies the phase space for phonon scattering by inducing Landau quantization.² In recent work on graphene, Plochocka *et al.*¹⁶

showed that the onset of this quantization is accompanied by a slowing of carrier thermalization. Baker *et al.*,^{21,27} in contrast, used resistance thermometry to investigate electron-phonon cooling, and found this, also, to be slowed in the magnetic field. In none of these studies, however, was the carrier density varied continuously, so that the results obtained provided only a “snapshot” of the cooling at a few fixed energies far from the DP. In this Letter, however, we present a systematic study of the energy and magnetic-field dependence of τ_e in graphene. For Fermi energies away from the DP, our results reveal a slowing of energy relaxation with increasing magnetic field, reminiscent of the aforementioned studies.^{16,21,27} As the Fermi level approaches the DP, however, a distinct reversal of this behavior occurs, with the energy-relaxation rate increasing at higher magnetic fields. This observation appears to support the idea that a very different process dominates energy relaxation near the DP. We discuss these results in terms of the influence of the magnetic field on the graphene density of states, and on the charge-puddling phenomenon^{38–40} that is known to lead to strong incompressibility near the DP.

Our study was performed on two bilayer graphene devices, realized by exfoliating Kish graphite onto a doped Si substrate with a 300-nm SiO₂ cap layer.^{31,41,42} Layer identification was performed through a combination of optical microscopy and Raman imaging, and the use of bilayer graphene was primarily driven by the relative ease with which manageable pieces of this material could be isolated. Nonetheless, there is a natural benefit to using this system for studies of this type, since the magnetic-field induced changes in the density of states near the DP are more pronounced than for monolayer graphene.² One of the devices studied (referred to hereafter as BG1) was device B1 of Ref. 31, while in Fig. 1 we show a micrograph of the second device (which we will refer to as BG2) and indicate its four-terminal measurement geometry. Since consistent results were obtained for both BG1 and BG2, below we focus on our more comprehensive study of energy relaxation in BG2. This flake was approximately 8- μm long and 2–3- μm wide,

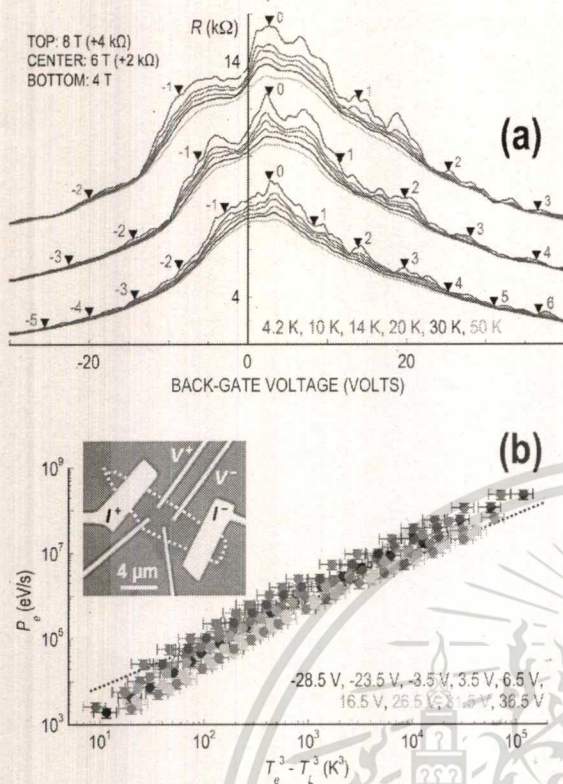


FIG. 1. (a) CF in device BG2. Measurements are at various T_L (indicated) and for $I = 100$ nA. Data at different fields are shifted vertically by the amounts indicated. Triangles identify gate voltages corresponding to filling of an integer number of Landau levels. The Landau index (N_L) is also indicated for each symbol, with negative values corresponding to filling by holes. (b) P_e vs. $T_e^3 - T_L^3$ for device BG2 at different V_g (indicated) and at zero field. The dotted line has power-law slope of one and $T_L = 4.2$ K. Inset: optical micrograph of device BG2. The graphene flake is denoted by the white dotted line and the four-terminal measurement geometry is indicated.

and the voltage probes indicated in Fig. 1 were $2\text{-}\mu\text{m}$ apart. Carrier density (n) was varied by means of the voltage (V_g) applied to the conductive Si substrate and Hall measurements could be used to reliably determine its value down to a level of $\sim 4 \times 10^{11} \text{ cm}^{-2}$.³¹ Electron (hole) mobility at 4.2 K was determined to be 4340 (3620) cm^2/Vs over a wide range of density, with a diffusion constant that ranged from 80 to 210 $\text{cm}^2 \text{ s}^{-1}$. Conductance of the graphene devices was measured by low-frequency (13 Hz) lock-in detection, using an AC current (I , whose RMS value was varied from 0.1 to 20 μA) to vary the carrier temperature relative to that of the lattice (T_L).

The hot-carrier thermometry that we perform here has been described previously in Ref. 31; it involves comparing measurements of the conductance fluctuations (CF), generated by varying the Fermi energy (V_g) in the material,^{41,42} at a series of lattice temperatures (at fixed I) and measurement currents (at fixed T_L). The CF are demonstrated in Fig. 1(a), where we plot the resistance (R) of device BG2 as a function of V_g , at various T_L (4.2–50 K) and three different magnetic fields (4-, 6-, and 8-T). The DP is located at $V_g \sim 5$ V, indicating the presence of a small amount of chemical doping. Nonetheless, we observe reproducible CF that are suppressed with increasing T_L due to increased decoherence.^{41,42} For each magnetic field, we have indicated with a set of markers

the gate voltages corresponding to complete filling of an integer number ($N_L = nh/4eB$, with the sheet density n provided by a parallel-plate capacitor expression) of Landau levels. While there is some correlation between these voltages and the presence of significant structure in the resistance, the correlation is not perfect. From this, we conclude that we are in the regime where the Landau quantization is incipient.

A key parameter in discussions of hot-carrier relaxation is the energy-loss rate (P_e), the power dissipated per carrier in the material ($P_e = I^2 R/nLW$, with R , L , and W the resistance, length, and width of the conductor, respectively). Typically, this rate is expressed in the form⁴³

$$P_e = A(T_e^p - T_L^p), \quad (1)$$

where the constants A and p depend upon the specific scattering mechanisms responsible for energy relaxation. In graphene it has been suggested that p should take values of 4 and 3, respectively, for electron-phonon scattering in pristine material,⁴ and for impurity-mediated supercollisions.⁹ In previous work, performed at zero magnetic field, we found³¹ that the loss rate in both bilayer and monolayer graphene is well described by $p = 3$, for wide variations of temperature and density. This behavior is reproduced in Fig. 1(b), where we plot P_e vs. $(T_e^3 - T_L^3)$ for device BG2. While these data are again consistent with $p = 3$, as noted already in Ref. 31 it is unlikely that such behavior is due to supercollisions. These are only expected to dominate⁹ when the Bloch-Grüneisen temperature (T_{BG}) is smaller than T_L , while our experiments are performed in the opposite regime where $T_{BG} > T_L$. ($T_{BG} = 2\hbar s k_F/k_B \sim 54n^{1/2}$ K, where $s = 2.1 \times 10^4 \text{ ms}^{-1}$ is the sound velocity in graphene, k_F is the Fermi radius, and n is the electron or hole density, in units of 10^{12} cm^{-2} . For the lowest density of around $4 \times 10^{11} \text{ cm}^{-2}$ that we are able to reliably infer from Hall measurements, $T_{BG} \sim 34$ K.) In this sense, it appears that the behavior of P_e in Fig. 1(b) represents a more general signature of hot-carrier relaxation in graphene, a conclusion that is consistent with the work of Ref. 30 where an exponent of $p = 3$ was also found outside of the supercollision limit.

In Fig. 2(a), we demonstrate how hot-carrier relaxation is influenced by the magnetic field, plotting the energy-loss rate for electrons and holes at $B = 8$ T (where the negative values denote the hole density). For both types of carrier, and at all gate voltages, the observed variations are consistent with the form of Eq. (1), with an exponent $p = 3$. In contrast to Fig. 1(b), however, the data for the different gate voltages do not collapse onto a common curve but rather exhibit a significant spread in values (implying significant differences in the prefactor A). As we reveal in Fig. 2(b), this much broader dispersion of loss rates arises from a significant change in the nature of carrier heating when the magnetic field is applied. In this figure, we plot the density-dependent variation of T_e at several different magnetic fields (and for fixed total electrical power, $I^2 R = 20$ nW). At low magnetic fields, in the range of 0–2 T, the heating is most effective near the DP, where T_e reaches a local maximum. Higher magnetic fields induce a dramatic reversal of this behavior, however, with T_e now developing a minimum near

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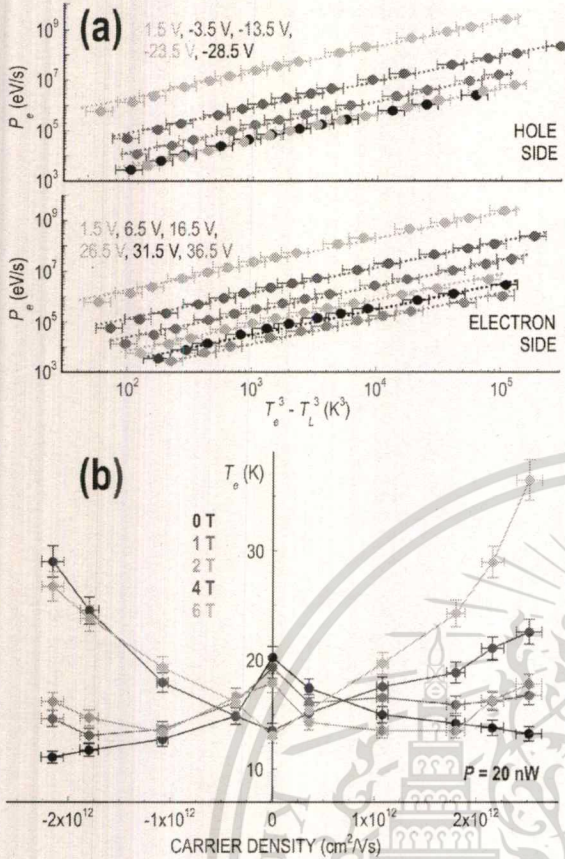


FIG. 2. (a) P_e vs. $T_e^3 - T_L^3$ for device BG2 at different V_g (indicated) at 8 T. Dotted lines through the data have power-law slope of 1 and $T_L = 4.2$ K. Top (bottom) panel: hole (electron) side of DP. (b) Variation of the effective carrier temperature, $T_e(n, B)$, obtained for fixed total electric power of 20 nW.

the DP. At the same time, T_e also exhibits a much stronger variation with density at these magnetic fields, and it is this characteristic that is responsible for the wide spread in energy-loss rates apparent in Fig. 2(a).

By imposing the requirement of energy balance, knowledge of the carrier temperature allows us to infer the energy-relaxation time⁴⁴

$$P_e = \frac{k_B(T_e - T_L)}{\tau_e} \quad (2)$$

In Fig. 3(a), we plot the density-dependent variation of τ_e at magnetic fields in the range of 0–6 T. At each magnetic field, τ_e decreases by at least an order of magnitude as the Fermi level approaches the DP from either band, consistent with our earlier work at zero field.³¹ The important aspect revealed here, however, is a strong influence of the magnetic field on heating, dependent upon the position of the Fermi level relative to the DP. For densities away from the DP ($|n| > \sim 10^{12} \text{ cm}^{-2}$), we find that τ_e increases with magnetic field, implying that relaxation is correspondingly slowed. This behavior is reversed at lower densities, however, where τ_e instead decreases with increasing field (data enclosed by the dotted line in Fig. 3(a)). This crossover is further demonstrated in Fig. 3(b), where we plot the variation of τ_e as a function of B for several gate voltages. For the data obtained

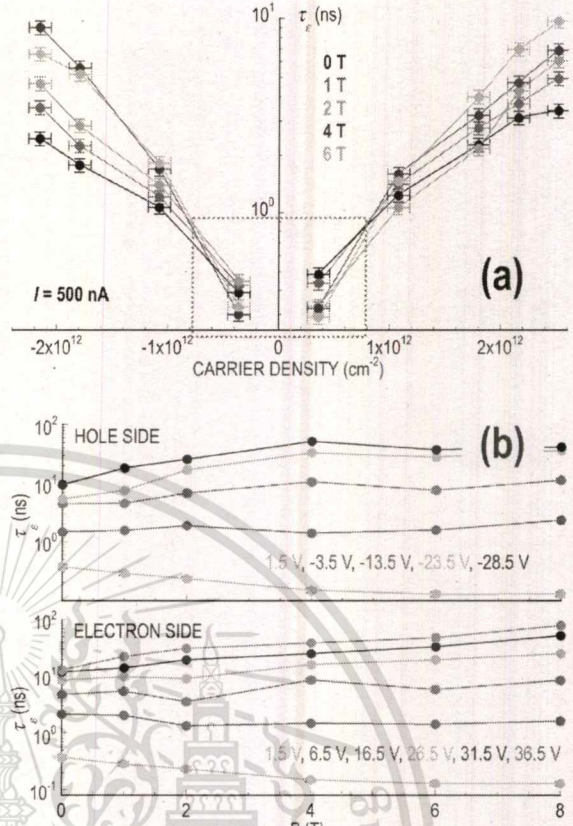


FIG. 3. (a) Variation of τ_e with density for device BG2 at fields in the range of 0–6 T (indicated, measurement current = 500 nA). (b) Variation of τ_e with B at various V_g . The upper (lower) panel is for holes (electrons) and $I = 100$ nA.

closest to the DP (gray symbols), τ_e decreases by a factor of three when the magnetic field is increased from 0- to 8-T. At the highest densities, on the other hand, it increases by as much as a factor of five for the same variation in field strength.

Previously, there have been several reports on the influence of a magnetic field on hot-carrier relaxation in graphene.^{16,21,27} The common conclusion that followed from these studies, which were performed at fixed Fermi energies away from the DP, is that relaxation is suppressed at higher magnetic fields, due to the emergence of the graphene Landau-level structure. While we do not realize well-resolved Landau levels in our work, the emergence of structure in the CF of Fig. 1(a) nonetheless suggests the presence of incipient Landau quantization. Consequently, the slowing of cooling that we observe at densities away from the DP appears to be consistent with the results of these earlier studies.

By varying carrier density via the gate voltage, we have found that energy relaxation undergoes a dramatic reversal near the DP, where the application of a magnetic field now leads to more-efficient cooling. This behavior, also, may be related to the emergence of the Landau spectrum, most notably to the development of the zero-energy level² that is unique to monolayer and bilayer graphene. Magneto-capacitance measurements provide a powerful tool to probe the structure of this level,^{45–47} and reveal two competing

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effects as the magnetic field is increased. The first of these is a growth in the density of states at the DP, while the second is a simultaneous reduction of this density on the electron and hole flanks.⁴⁵ By providing an enhanced phase space for electron-phonon coupling near the DP, it seems reasonable that the magnetic field could therefore enhance the relaxation rate.

While these general arguments concerning Landau quantization provide an attractive framework for the interpretation of our experiment, it must be emphasized that the quantization is only emergent in our experiments and that other scenarios should also be considered. Most notably, in our prior work at zero magnetic field,³¹ we noted that the short relaxation times inferred near the DP occur in a regime where the effects of charge puddling^{38–40} are expected to be important. The puddle state is characterized by strong thermodynamic incompressibility, implying that the carriers become “frozen” into a microscopic configuration that is resistant to excitation.³¹ Recent experiments in which scanning-probe techniques were used to investigate the microscopic structure of the puddling⁴⁰ have shown that the localization associated with the puddles is significantly enhanced by a magnetic field. It is therefore also possible that the unusual heating behavior we observe near the DP is related to the incompressible character of the puddling, and to its enhancement in a magnetic field.

Finally, before concluding, we comment on the possible role of screening of the carrier-phonon interaction, and the dependence of this on magnetic field and density. Starting at zero field, while screening should be effective at higher densities it should weaken as the Fermi level is swept towards the DP. This in turn should lead to stronger scattering, and so faster relaxation, reminiscent of our experiment. A problem with this picture emerges when a magnetic field is applied, however, since screening at wavelengths longer than the Landau-orbit size will be suppressed. In this case, the main effect of increasing density should be to enhance scattering via the unscreened interaction, due to the associated increase of the electron or hole density of states. This would imply faster energy relaxation at higher densities, behavior opposite to that found here. While we cannot completely rule out the role of screening in our observations, the experimental behavior therefore seems to suggest a deeper rationale for the nature of the cooling process.

In conclusion, we have investigated the influence of incipient Landau quantization on hot-carrier relaxation in bilayer graphene. The magnetic field influences the relaxation in a very different manner, dependent upon the position of the Fermi level relative to the Dirac point. For electron or hole densities greater than roughly 10^{12} cm^{-2} , relaxation is slowed by the magnetic field. As the Fermi level approaches the DP, however, a reversal in this behavior occurs, with the relaxation quickening at higher fields. We have discussed how this behavior may be related to the emergence of the zero-energy Landau level, and to the peculiar nature of the graphene puddle state in a magnetic field. While we have focused on the influence of a magnetic field on relaxation in bilayer graphene, in previous work at zero field³¹ we found the details of the relaxation to be very similar for both monolayer- and bilayer-graphene.³¹ From this, we concluded that

the density-dependent variation of τ_e should not be a consequence of the linear bands, but rather more likely arises from the vanishing of the density of states near the DP. From this perspective, we expect the magnetic field to influence relaxation in monolayer graphene in a similar manner to that found here, although this requires confirmation through proper experiment. In spite of this, our observations nonetheless provide an important perspective on our emerging understanding of hot-carrier relaxation mechanisms in graphene.

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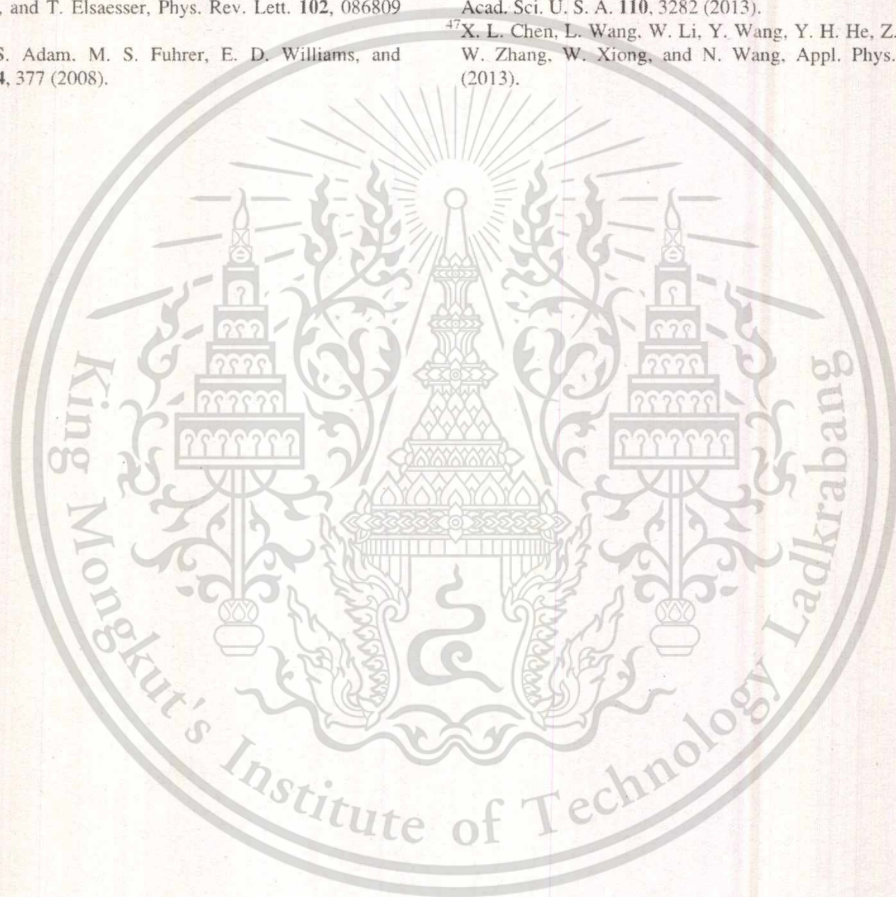
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แหล่งทุน: แหล่งเงินรายได้

ชื่อโครงการ: ศึกษาการขนส่งพลาสมาหรือไอในการสังเคราะห์กราฟีนบนซิลิกอนซัลไฟด์ด้วยวิธีการลอกด้วยเทปกา

ชื่อหัวหน้าโครงการ: ดร. รัชชก สมพรเสน่ห์

ว/ด/ป	รายการ	เลขที่อ้างอิง	รายการรับ - จ่าย		รายรับ	รายจ่าย	งบบุคลากร	งบดำเนินงาน			งบลงทุน	รวม
			รับ	จ่าย				คงเหลือ	ดอกเบี้ย	ค่าจ้างชั่วคราว		
	งบประมาณที่ได้รับอนุมัติ (ตามแผน)		50,000.00									
19/11/57	จำนวนเงินที่ได้รับ		50,000.00									
	หัก ค่าใช้จ่าย			51,478.46								
	งบประมาณคงเหลือ		50,000.00		1,478.46	0.00						
	รายละเอียดค่าใช้จ่าย											
13/7/57	BNC Tee Adapter Female-Male-Female	PSIN701012										4,918.80
13/7/57	BNC-Male to BNC Male Adapter	PSIN701012										4,158.40
13/7/57	S20 dBm P1dB, 500 MHz to 3 GHz	PSIN701016										9,437.13
13/7/57	Double Binding Post to 50 Ohm BNC	PSIN701014										9,313.25
13/7/57	10 MHz to 6 GHz SMA Bias Tee Rated To	PSIN701007										9,717.86
13/7/57	50 Ohm 0.5 Watts Nickel Plated Brass	PSIN701011										8,990.40
13/7/57	SMA-Male to SMA Female Bulkhead	PSIN701005										4,942.62
	รวม											51,478.46

ชื่อหัวหน้าโครงการ

วันที่10 สิงหาคม 2558.....

Curriculum Vitae

Name: Dr. Ratchanok Somphonsane

1. Personal Data

Female Male

Date of Birth: 4/26/1982

Affiliated University/Institute : King Mongkut's Institute of Technology Ladkrabang
Department of Physics, Faculty of Science
Chalongkrung Rd. Ladkrabang
Bangkok 10520 THAILAND

Academic Position: Lecturer

E-mail: rsomphonsane@gmail.com

Telephone: 091-706-2494

2. Education

Degree	Major	University	Year Completed
Ph.D.	Physics	The State University of New York, at Buffalo	2014
Grad. Dip.	Teaching Profession	Srinakharinwirot University	2005
B.Sc. (1 st class honors)	Physics	Prince of Songkla University	2004

3. Research Experience

2013-2014 Postdoctoral fellow, The State University of New York, at Buffalo, New York, USA

2009-2013 Research assistant, The State University of New York, at Buffalo New York, USA

2012 Visiting research fellow, Rice University, Texas, USA

Skill/Expertise: Nanoelectronics, Graphene nanoelectronics, Nanofabrication, Electrical measurement, Low-temperature measurement

4. Major Prize / Award / Honor

Year	Award	Institute
2015	Best Senior Poster Award of EDISON'19	Edison'19 Conference
2011	Best Poster Award	SEAS, The State University of New York, at Buffalo, USA
2011	Best Poster Award	EE Department, The State University of New York, at Buffalo, USA

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5. Scholarships/Fellowships

Year	Scholarships/Fellowships	Institute
2016-2017	Grant for new researcher	Ministry of Science and Technology, Thailand
2016-2017	Grant for new researcher	King Mongkut's Institute of Technology Ladkrabang
2015-2017	Grant for new researcher	Thailand research funding
2015-2017	Grant for new researcher	King Mongkut's Institute of Technology Ladkrabang
2015-2016	Grant for new researcher	King Mongkut's Institute of Technology Ladkrabang
2007-2012	Scholarship for Ph.D. study	Ministry of Science and Technology, Thailand
2004-2005	Scholarship for Graduate study in Teaching profession	The Institute for the Promotion of Teaching Science and Technology, Thailand
2000-2004	Scholarship for undergraduate study	The Institute for the Promotion of Teaching Science and Technology, Thailand

6. Publications

1. R. Somphonsane, H. Ramamoorthy, G. He, J. Nathawat, C.-P. Kwan, Y.-H. Lee, J. Fransson, and J. P. Bird "Evaluating the contribution of impurities and phonons to the resistivity of graphene by differential-conductance mapping" *Phys. Rev. B* (2016, under revision)
2. G. He, H. Ramamoorthy, C.-P. Kwan, Y.-H. Lee, J. Nathawat, R. Somphonsane, M. Matsunaga, A. Higuchi, N. Aoki, Y. Gong, X. Zhang, R. Vajtai, P. M. Ajayan, and J. P. Bird "Thermally-Assisted Nonvolatile Memory in Monolayer MoS_2 Transistors", *Nano Lett.* **16**, DOI: 10.1021/acs.nanolett.6b02905 (2016)
3. H. Ramamoorthy, R. Somphonsane, J. Radice, G. He, C.-P. Kwan, and J. P. Bird "Freeing" Graphene from its Substrate: Observing Intrinsic Velocity Saturation with Rapid Electrical Pulsing" *Nano Lett.* **16**, 399–403 (2016)
4. B. Liu, R. Akis, D. K. Ferry, G. Bohra, R. Somphonsane, H. Ramamoorthy, and J. P. Bird, "Conductance fluctuations in graphene in the presence of long-range disorder" *J. Phys.: Condens. Matter* **28**, 135302 (2016)
5. D. K. Ferry, R. Somphonsane, H. Ramamoorthy, and J. P. Bird, "Energy relaxation of hot carriers in graphene via plasmon interactions" *J. Comput. Electron.* **15**, 144-153 (2016)
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7. G. He, K. Ghosh, U. Singiseti, H. Ramamoorthy, R. Somphonsane, G. Bohra, M. Matsunaga, A. Higuchi, N. Aoki, S. Najmaei, Y. Gong, X. Zhang, R. Vajtai, P. M. Ajayan, and J. P. Bird, "Conduction Mechanisms in CVD-Grown Monolayer MoS₂ Transistors: From Variable-Range Hopping to Velocity Saturation" Nano Lett. 15, 5052 (2015)
8. H. Ramamoorthy, R. Somphonsane, G. He, D. K. Ferry, Y. Ochiai, N. Aoki, and J. P. Bird, "Reversing hot-carrier energy-relaxation in graphene with a magnetic field", Appl. Phys. Lett. 104, 193115 (2014)
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10. G. Bohra, R Somphonsane, N. Aoki , Y. Ochiai, R. Akis, D. K. Ferry, and J. P. Bird, "Nonergodicity and microscopic symmetry breaking of the conductance fluctuations in disordered mesoscopic graphene", Phys. Rev. B 86, 161405(R) (2012)
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7. Presentations

Oral Presentations

1. Gave a talk on "Relaxation Dynamics of Hot Carriers in Graphene Revealed by Current-Induced Heating Measurements" at NanoThailand 2014 (The 4th Thailand International Nanotechnology Conference), at Phatumthani, Thailand on Nov 26-28, 2014
2. Gave a talk on "Reversing Hot-Carrier Energy Relaxation with a Magnetic field" at IEEE NMDC 2014 (IEEE Nanotechnology Materials and Devices Conference) at Sicily, Italy on Oct 12-15, 2014
3. Gave a talk on on "Hot-Carrier Relaxation in Graphene and Its Dependence on a Magnetic Field" at GRPR 2014 (The 6th International Conference on Recent Progress in Graphene Research) at Taipei, Taiwan on Sep 21-25, 2014

Poster Presentations

1. R. Somphonsane, H. Ramamoorthy, and J. P. Bird, "Phonon-induced hot-carrier scattering in graphene: Insight from differential conductance" at: 9th International Conference on Electron Dynamics in Semiconductors, Optoelectronics and Nanostructures (Edison' 19), Salamanca, Spain (2015)

2. H. Ramamoorthy, R. Somphonsane, G. He, D. K. Ferry, N. Aoki, Y. Ochiai and J. P. Bird, “*Hot-Carrier Relaxation in Graphene in a Magnetic Field*” at: 21st International Conference on “High Magnetic Fields in Semiconductor Physics”(HMF-21), Panama City Beach, Florida (2014)
3. R. Somphonsane, H. Ramamoorthy, D. K. Ferry, and J. P. Bird, “*Magnetic-field dependence of hot-carrier relaxation in graphene*”, Poster presentation at: The International Symposium on Advanced Nanodevices and Nanotechnology (ISANN), Kauai, Hawaii (2013)
4. H. Ramamoorthy, R. Somphonsane, and J. P. Bird, “*Current-induced cleaning of graphene and graphene-metal contacts*” The International Symposium on Advanced Nanodevices and Nanotechnology (ISANN), Kauai, Hawaii (2013)
5. R. Somphonsane, H. Ramamoorthy, G. Bohra, G. He, D. K. Ferry, Y. Ochiai, N. Aoki, and J. P. Bird, “*Hot-carrier energy relaxation near the Dirac point of graphene*”, International Workshop on Recent Progress in Nonequilibrium Quantum Many-Body Theory, Buffalo, NY (2013)
6. R. Somphonsane, H. Ramamoorthy, G. Bohra, G. He, D. K. Ferry, Y. Ochiai, N. Aoki, and J. P. Bird, “*Energy relaxation in mesoscopic graphene*”, 3rd International Symposium on Terahertz Nanoscience (TeraNano III), Honolulu, Hawaii (2012)

