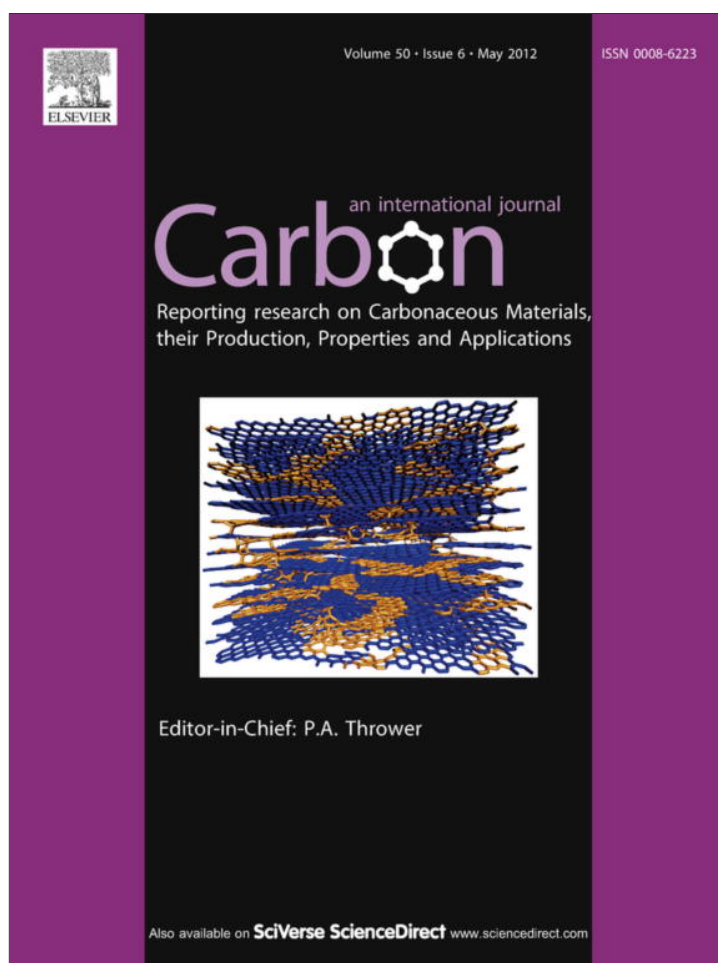


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Observation of the semiconductor–metal transition behavior in monolayer graphene

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ABSTRACT

We have observed that during temperature-dependent four-terminal resistance measurement of monolayer graphene, the resistance exhibits anomalous rising and falling behavior at different temperature regions. At lower temperature region (2–200 K) the resistance decreases gradually, but when the temperature rise further it turn to a sudden increase, and after 280 K it resumes gradual decrease. The rising and falling resistance behavior is characteristic of semiconductor or metal property. Consequently, the resistance transition follows a phase of semiconductor–metal–semiconductor. However, when a perpendicular magnetic field is applied, the resistance shows reverse transition behavior which follows a sequence of metal–semiconductor–metal. The novel transition property is attributed to the competition between the disorder of lattice defects as a short-range scattering in monolayer graphene and the Landau levels interaction. Magneto-transport measurement reveals that the excitonic gap induced by magnetic field in the monolayer graphene show an anomalous thermally activated property.

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1. Introduction

The rise of the newly-discovered two-dimensional one atomic thick carbon material or graphene promises a realm of exciting new technological applications and also interesting physics [1]. The electronic properties of monolayer graphene have been extensively studied due to its intriguing energy band structure with linear dispersion around the Dirac point [2,3]. Such electronic behavior can give rise to a number of interesting phenomena, such as the anomalous quantum hall effect [4], Klein tunneling effect [5] and Shubnikov–de-Haas (SdH) effect [6]. Owing to its exceptionally high charge mobility and crystal quality, graphene has been widely predicted to be the platform for: ultra-small field-effect transistors [2], wearable electronics [7], ultracapacitors [8], solar cells [9], biosensors [10] and future generation spintronic and

nano-electronic devices [11]. Additionally, and of no less importance, the unique chiral natures of electron in monolayer graphene show sign of relativistic electrons property [1–3]. The massless Dirac fermions properties make graphene an attention-grabbing material for fundamental studies of mesoscopic phenomena and the understanding of spin-qubits [12], with potential application in the solid-state quantum computation. The electronic properties of graphene are strongly influenced by the application of external magnetic field. The unique chiral nature of electron in monolayer graphene has a profound effect in weak localization [13] and results in unevenly spaced Landau level (LL) [14]. There are interesting physics to be explored in monolayer graphene when it is subject to the influence of magnetic field due to an extra twofold orbital degeneracy in the LL spectrum [15], which results in a fourfold-degenerate LL at the zero energy

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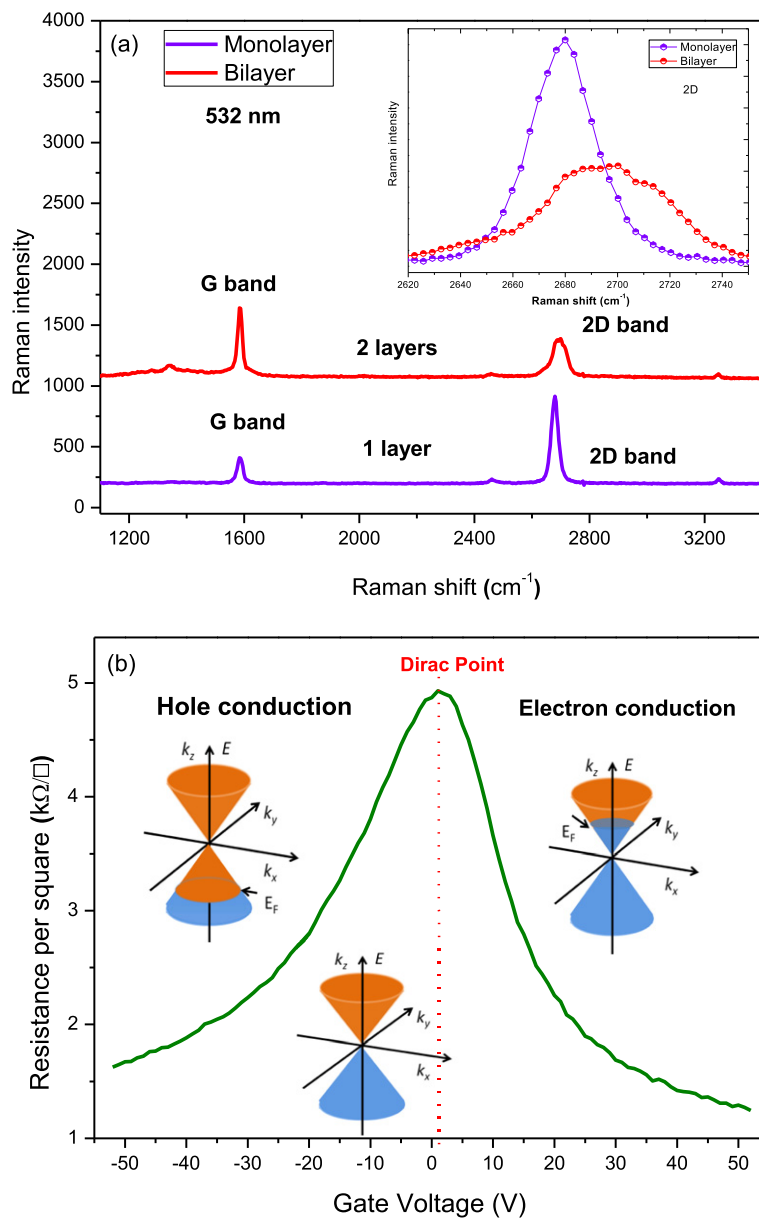


Fig. 1 – (a) Comparison of Raman spectra at 532 nm for monolayer and bilayer graphene. The position of G peak and the spectral features of 2D band confirm the number of atomic layer of the graphene. The number of graphene layer is distinguishable from the full-width half maximum of the 2D band. (b) Resistance per square as a function of back gate voltage V_{bg} . A resistance peak appears close to a gate voltage of $V_{bg} \approx 1$ V indicates that very minimum chemical doping is present in our graphene sample.

level. Hence, the enhanced interactions under high magnetic field are expected to lift fourfold zero-energy LL degeneracy in monolayer graphene [16–18]. To date, experimental and theoretical study regarding the limiting source of scattering, with and without the application of magnetic field, in monolayer graphene is a subject of intense research and debate [19]. The understanding of the nature of the disorder and how mesoscopic ripples affect the graphene transport properties are still under scrutiny.

In this paper, we report on temperature-dependent four-terminal resistance measurement of monolayer graphene. Depending on the rising temperature regions the measured

resistance value shows anomalous rising or falling behavior, which indicates characteristic of semiconductor or metal, and the transition behavior follows a sequence of semiconductor–metal–semiconductor. However, when a perpendicular magnetic field is applied, the resistance show a reversed transition behavior, which follows the sequence of metal–semiconductor–metal. The novel transition property is attributed to the competition between the disorder of lattice defects in monolayer graphene and the Landau levels interaction. Magnetoresistance measurement reveals that the excitonic gap in the monolayer graphene show an anomalous thermally activated properties. The result implies that

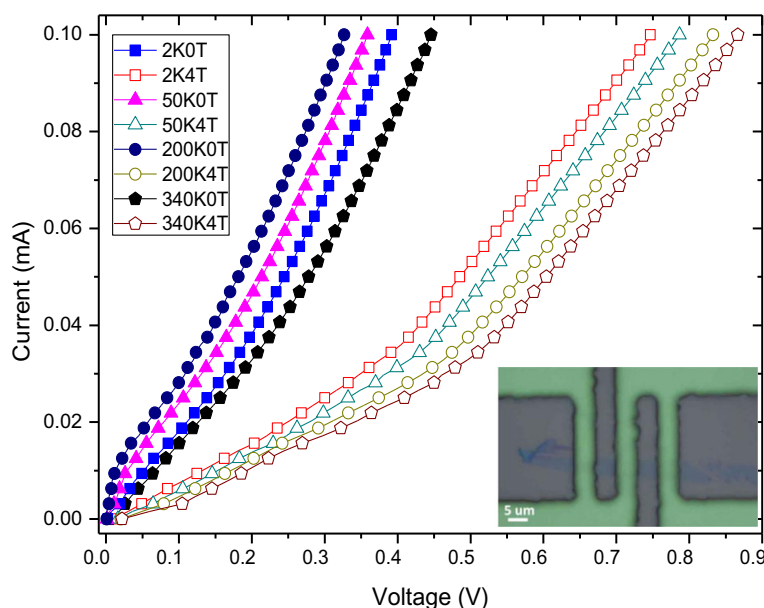


Fig. 2 – Temperature-dependent current-voltage characteristics of monolayer graphene. The measurements show the dependence of resistance on temperature and the introduction of a perpendicular magnetic field has increased the resistance. The nonlinear behavior is due to energy relaxation as slow carriers are heated more effectively under a low dc electric field. As the applied dc voltage increases the energy relaxation grows with the heating effect, and the process of carrier recombination is dominant compared with thermal generation.

magnetic field could induce high resistance and the field dependence of resistance at the Dirac point is a qualitative proof of the energy gap opening.

2. Experimental details

The monolayer graphene samples were prepared using mechanical exfoliation techniques from bulk highly oriented pyrolytic graphite (grade ZYA, SPI Supplies) and transferred onto the surface of a lightly-doped silicon substrate that was covered with a 300-nm-thick SiO₂ layer. Four Cr (10 nm)/Au (60 nm) electrical electrodes were patterned using photolithography technique and deposited via thermal evaporation at a base pressure of 10⁻⁷ mbar, with a subsequent lift off in warm acetone. A standard four-terminal configuration was used for electrical transport characterization. The transport measurements were carried out by a PPMS (Quantum Design) system. The measurements were performed in the temperature range 2–340 K and a magnetic field up to 12 T was applied. Thermal annealing was carried out at 300 °C in vacuum for 1 h to eliminate contamination and to restore clean surfaces of graphene. In situ sample cleaning by magnetic and electric field was performed to improve the measurement quality.

Raman spectroscopy is a non-destructive, reliable tool for the confirmation of the number of graphene layers and it is carried out via the 2D-band deconvolution procedure [20–22]. The Raman spectra of our graphene structure were measured at room temperature using a WITTEC CRM200 instrument at 532 nm excitation wavelength in a backscattering configuration [23,24]. Fig. 1a shows the characteristic Raman spectrum with a clearly distinguishable G peak and 2D band. The two most intense features are the 2D band and the G

peak, which are sensitive to the number of graphene layer. By comparing the position of the shapes of the 2D band and the G peaks the number of graphene layers can be precisely determined. Additionally, the number of graphene layer can be easily distinguished from the full width half maximum of the 2D band, its mode changes from a narrow and symmetric feature for monolayer to an asymmetric distribution on the high-energy side for bilayer [24]. The 2D band shown in the inset of Fig. 1a is the Raman spectra of the bilayer graphene that is red-shifted and broadened with respect to that of the monolayer graphene. In addition, the 2D peak of monolayer graphene is symmetric, while the 2D band corresponding to the few layer graphene has a complex asymmetric shape. Fig. 1b shows resistances per square as a function of back gate voltage V_{bg} . The results show a resistance peak close to a gate voltage of $V_{bg} \approx 1$ V, which indicates that the graphene sample is of high purity.

3. Results and discussion

We have carried out current (I)–voltage (V) measurement of the monolayer graphene via a four-terminal configuration. Shown in Fig. 2 is the representative I – V curve of the monolayer graphene under different magnetic field strengths and temperatures. The magnetic field is applied along the perpendicular direction of the graphene sample. An optical image of the monolayer graphene sample with the corresponding Cr/Au electrodes is shown in the inset of Fig. 2. The nonlinear behavior shown in the I – V measurement is due to energy relaxation as slow carriers are heated more effectively under low applied dc electric field. As the strength of electric field is increased the energy relaxation grows with the heating effect and the carrier

recombination process becomes more dominant compared with the thermal generation process [25]. The gradient of the I - V curve correspondings to the conductivity of the monolayer graphene. The measured gradient is increasing as the temperature increases from 2 to 200 K, which is a characteristic of semiconductor behavior. An unusual conductivity drop is observed after 200 K and the details will be given in Fig. 3 analysis. However, when the I - V measurement is subject to a perpendicular magnetic field the gradient decreases with increasing temperature. Such temperature and magnetic field dependent behavior of conductivity is a characteristic of metal. The decrease in the conductivity of the monolayer graphene with increasing magnetic field is due to the induction of an excitonic energy gap by magnetic field. In the absence of a magnetic field, the band structure of the monolayer graphene at the Dirac valley is associated with a linear dispersion relation. When a magnetic field is present, the band structure is changed to Landau level splitting [16–18]. The conductivity dependence on magnetic field suggests that the resistance ($\rho = \frac{1}{\sigma}$) R of graphene is a qualitative fingerprint of its band gap.

Fig. 3a represents the magnetoresistance measurement of the monolayer graphene as a function of applied magnetic field at temperature 2 K. The normalized magnetoresistance $R(B)/R_{B=0T, T=2K}$ exhibits non-linear behavior with plateau-like features appearing at high field region, but such features gradually disappear when the measurement temperature is increased. Krstić et al. [17] reported similar observation and they argued that those features originate from an augmented sublattice spin-splitting due to the high surface-impurity concentration of the graphene layer. However, our measurements show that such plateau-like feature is also present in bilayer graphene at temperature 2 K but it disappears when the temperature is increased. Moreover, it was completely not seen in similar magnetoresistance measurement for three, four and five layer graphene samples [26,27]. Hence, we propose that the origin of the observed plateau-like feature in the monolayer graphene is due to the ripple effect, which has introduced some anisotropic potential that may have led to a low-field lift of the sublattice degeneracy [17,28]. One of the unusual properties of massless Dirac electrons in monolayer

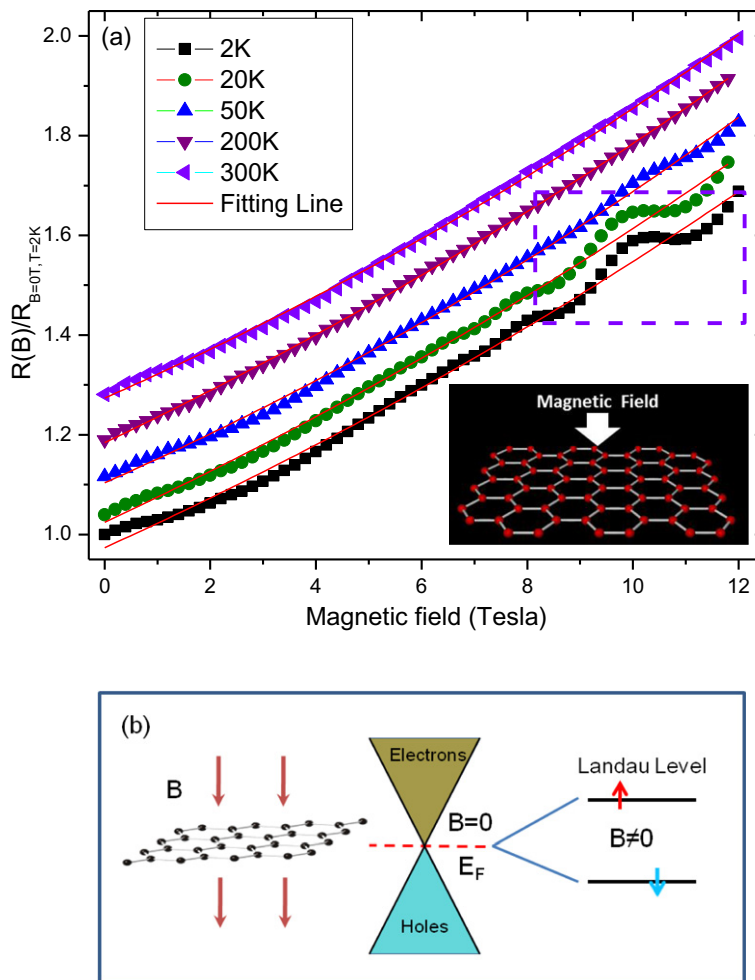


Fig. 3 – (a) Magnetoresistance per square measurements of monolayer graphene at different temperature. A higher measured resistance with increasing perpendicular magnetic field strongly supports that magnetic field can induce an excitonic gap in the monolayer graphene. The dependence of resistance on the magnetic field can be explained by the splitting of Landau level (LL). Resistance plateau-like features are observed for measurement at high magnetic field (8–12 T) but the feature gradually disappears when the temperature is increased (>50 K). The line fits following the relationship \sqrt{B} . (b) Illustration of the monolayer graphene bandgap and Landau level splitting under the influence of a magnetic field.

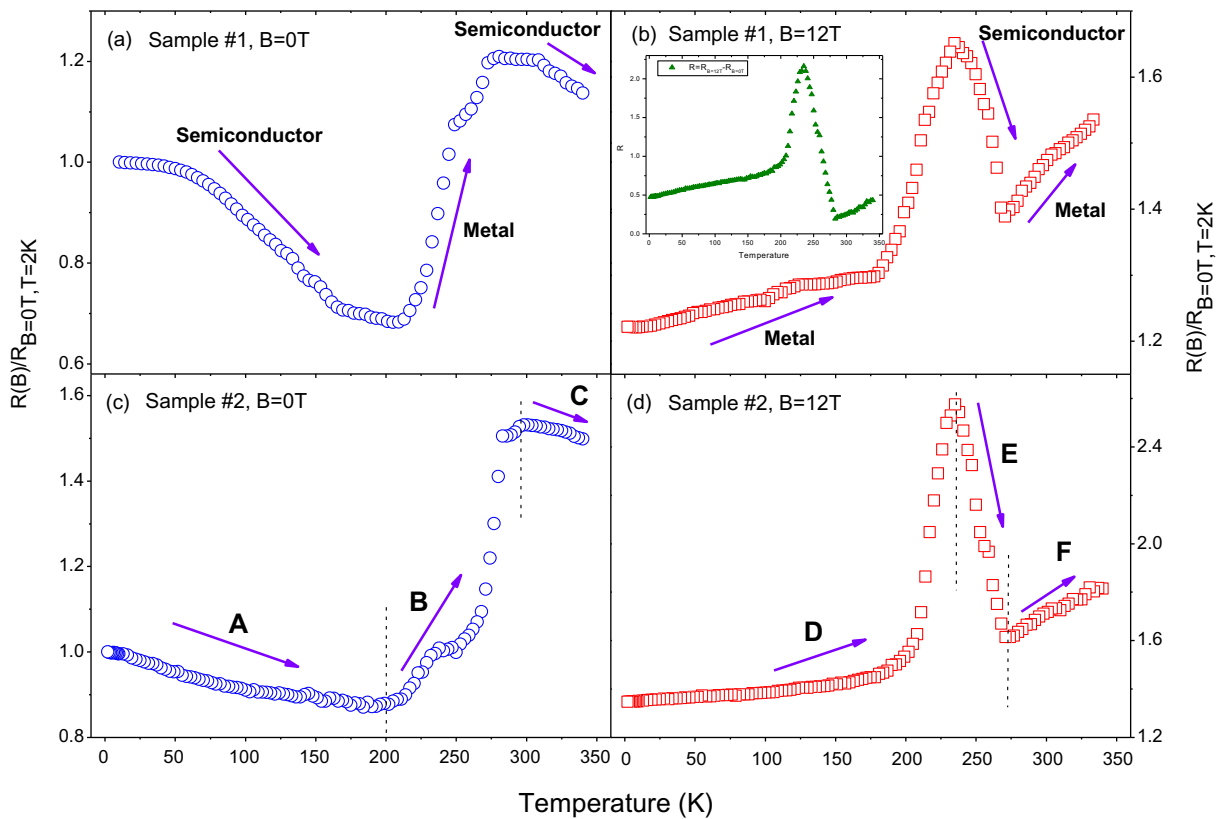


Fig. 4 – Temperature-dependent resistance measurements of monolayer graphene (sample #1 and sample #2) under the conditions with and without an applied magnetic field. The results reveal that as temperature increases from 2 to 340 K, the monolayer graphene undergoes a semiconductor–metal–semiconductor transition. However, when a perpendicular magnetic field ($B = 12\text{ T}$) is applied, the transition becomes metal–semiconductor–metal. The opposite transition sequence of the biased and unbiased magnetic field condition is attributed to the competition between the disorder of lattice defects as a short-range scattering in the monolayer graphene and the interaction Landau levels splitting. Inset shows the relative resistance change $\Delta R = R_{B=12T} - R_{B=0T}$ of the monolayer graphene as a function of temperature.

graphene is \sqrt{B} that it has intrinsic Zeeman energy that is accurately one half of the cyclotron energy in magnetic field [15]. Such property has caused Landau-level (LL) splitting and its energy spectrum comprises of fourfold degeneracy at zero-energy and fourfold degeneracy at non zero-energy levels [15,28]. This potentially indicates sublattice symmetry breaking and gap formation due to many-body correction in this LL [28–30]. Fig. 3b illustrates the bandgap of the monolayer graphene and Landau level splitting when it is under the influence of a magnetic field. When Coulomb interaction is considered, the interaction between electron–hole pairs has formed an excitonic condensation that can give rise to an energy gap at zero energy level [15]. In our magnetoresistance measurements, the non linear behavior is proportional to but it cannot fully account for the plateau-like feature. Hence, these considerations only provide qualitative explanation for the nonlinear magnetoresistance $R(B)/R_{B=0T, T=2K}$ relationship with the magnetic field.

Among the main scattering mechanisms that characterize the electronic transport property in monolayer graphene are Coulomb scattering [31,32], short range scattering [33], and graphene surface phonon scattering [34,35]. The temperature dependence of resistance in monolayer graphene is predominantly determined by short-range and Coulomb scatterings,

which are caused by lattice defects and substrate charged impurities in graphene sample, respectively. Shown in Fig. 4 is the temperature-dependent resistance measurement of the monolayer graphene (sample #1 and sample #2) under the conditions with and without the presence of an applied magnetic field. At lower temperature region (2–200 K) the measured resistance shows a gradual decrease (region A in Fig. 4(c)). However, as the temperature approaches 200 K the resistance of the graphene begins to rise (region B). This resistance climb continues as the temperature rises but after 280 K it starts to decrease gradually again (region C). The decreasing and increasing resistance behavior is a characteristic of semiconductor and metal property, respectively. Therefore, the resistance measurement of the monolayer graphene in the temperature region 2–340 K reveals a transition of semiconductor–metal–semiconductor. Such resistance transition behavior is attributed to the interplay between the Coulomb and short-range scatterings. The observation of the sudden rise of graphene resistance near at room temperature was also reported by Morozov et al. [36]. They attributed the resistance rise behavior to the intraripple flexural phonons scattering, where the ripples originate from the graphene corrugations following SiO₂ substrate roughness. However, when the temperature-dependent resistance measurement

is subject to an external perpendicular magnetic field $B = 12$ T, there is a striking change to the transition behavior, in addition to an increase of the overall resistance value. At lower temperature region 2–200 K, the measured resistance shows a gradual increase (region D in Fig. 4(d)). As the temperature approaches 200 K the resistance value increases rapidly and reaches its peak at 230 K. Then, the value drops rapidly from 230 to 270 K (region E) forming a resistance spike within a narrow temperature range (70 K), and shows a resistance change ratio of $\sim 70\%$. At higher temperature (>270 K) the resistance starts to increase gradually again (region F). Therefore, when under the influence of a perpendicular magnetic field the resistance measurement of the monolayer graphene indicates a metal–semiconductor–metal transition. The unusual transition behavior is attributed to the disorder of lattice defects as a short-range scattering in monolayer graphene and the Landau levels interaction [33,37–39]. The increase of the short-range scattering in monolayer graphene arises from the lattice defects being strongly influenced by the external magnetic field and it is dependent of temperature [33,40]. With the applied magnetic field, the formation of Landau levels increases the effect of interactions due to the quenching of the kinetic energy [41–44]. The energy splitting of the different broken symmetry phase is determined predominantly by lattice effects in the clean graphene [45,46]. This effect is more pronounced at low filling when the lowest levels are occupied. Interactions can result in a new phase when the effect overcomes that of disorder [47]. The increase of the resistance at all temperature range when an external field is applied indicates that an excitonic gap in the monolayer graphene can be tuned by a magnetic field. Inset shows the relative resistance change $\Delta R = R_{B=12T} - R_{B=0T}$ of the monolayer graphene as a function of temperature. The results clearly indicate that the gap is thermally activated due to the Coulomb interaction-driven electronic instabilities [15,28].

Fig. 5 shows the resistance measurements as a function of a lateral electric field in monolayer graphene under the conditions with and without the presence of an applied perpendicular magnetic field. The measurement show that, within the electric field $E < 0.01$ V/ μm , the resistance increases with the increasing applied electric field strength. One possible explanation for the resistance increase is that the application of electric field has caused heating effect that would enhance the SiO₂ substrate surface polar phonon scattering [35]. The interface polarization between graphene and SiO₂ substrate has induced an electric field, which is strongly dependent on temperature. Since the surface polar phonons scattering is proportional to the phonon population number [35,36], as the temperature rises, the resistance is expected to increase. However, when a magnetic field of 4 T is applied to the measurement, at lower applied electric field strength ($E < 0.02$ V/ μm), the resistance drop rapidly, particularly that with a higher measurement temperature. This is attributed to Coulomb scattering by impurities which is a strong function of temperature. On the other hand, when the electric field strength is further increased, the resistance gradually decreases or towards saturation. This behavior is due to the competition between the surface polar phonons and Coulomb scatterings. The increase in the overall measured resistance with the introduction of a magnetic field, irrespective of the

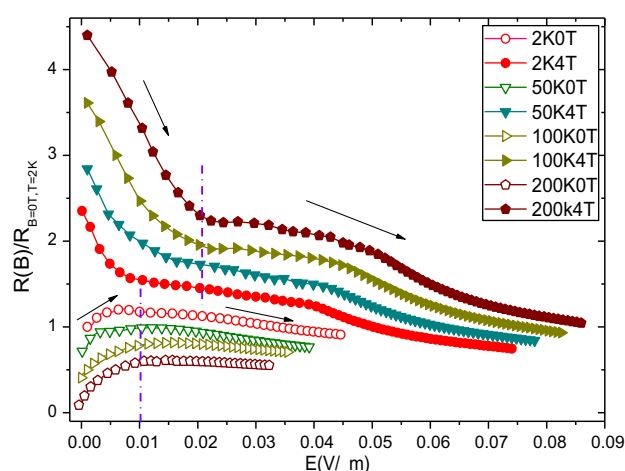


Fig. 5 – Resistance measurements as a function of lateral electric field of monolayer graphene. When a magnetic field is applied, the resistance of the monolayer graphene jumps to a higher value but decreases with increasing lateral electric field strength.

lateral electric field strength, also supports that magnetic field opens an excitonic gap in the monolayer graphene and the excitonic gap is dependent on temperature.

4. Conclusion

In conclusion, we have investigated temperature and magnetic field dependence of four-terminal resistance behavior of monolayer graphene. Our results reveal that the monolayer graphene exhibits a semiconductor–metal–semiconductor transition and reversal transition under the applied magnetic field condition. The novel transition property is attributed to the competition between the disorder of lattice defects as a short-range scattering in monolayer graphene and the Landau levels interaction. Magneto-transport measurement reveal that the excitonic gap induced by magnetic field in the monolayer graphene show an anomalous thermally activated property. The obtained results indicate that magnetic field could induce high resistance and is a qualitative proof for the opening of energy gap.

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