

論文の内容の要旨

論文題目 Field-effect transistors and photodetectors based on bilayer graphene in all-two-dimensional layered heterostructures

(完全2次元層状ヘテロ構造における2層グラフェン電界効果型トランジスタ及び光検出器に関する研究)

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1. Introduction

Due to their stability at an atomic layer limit which can suppress short-channel effect, two-dimensional layered (2D) materials are considered to be successor to Si field-effect transistors (FETs). Graphene has the highest carrier mobility in 2D materials, because its conduction band is from p_z orbital, unlike transition metal dichalcogenides whose conduction band are from d orbital, making graphene suitable for high-performance FETs. Although monolayer graphene does not have band gap, in bilayer graphene (BLG) band gap can be opened and tuned by applying electric field perpendicular to the graphene layers (Displacement field, \bar{D}). This can suppress off-current (I_{off}) and increase on/off current ratio ($I_{\text{on}}/I_{\text{off}}$) in FET operation. Conventionally, high- k gate stacks have been utilized to provide electric field required to open maximum band gap of ~ 0.3 eV. However, due to potential fluctuations from charged impurities in oxide insulators, large amount of interface trap density of $10^{13} \text{ cm}^{-2}\text{eV}^{-1}$ was detected even in high quality $\text{Y}_2\text{O}_3/\text{BLG}$ gate stack^[1] and I_{off} remained large even at maximum band gap (Fig. 1(a)). In conventional semiconductor system, intrinsic band gap of channel materials is sufficiently large compared to potential fluctuations from fixed charges in oxide insulators. Therefore, performance has been improved by reducing interface trap density at semiconductor/gate insulator interface. On the other hand, band gap in BLG is comparable to potential fluctuations of 20-30 meV from oxide insulators. Moreover, band gap in BLG is formed by inducing different carrier density in two graphene layers. As a result, potential fluctuations can cause spatial variation of band gap in BLG channel, leading to high I_{off} (Fig. 1(a)). Therefore, in order to suppress I_{off} and increase $I_{\text{on}}/I_{\text{off}}$ for BLG, potential fluctuations must be reduced. It is known that using h -BN as gate insulator for graphene FETs can increase carrier mobility because h -BN has atomically flat surface and has small charged impurities. Although h -BN itself has small charged impurities, using $\text{SiO}_2/n^+\text{-Si}$ as back gate can still propagate charge inhomogeneity in SiO_2 through h -BN onto the channel. Therefore, graphite gate electrodes have been used in gate stack, creating all-2D layered heterostructures (Fig. 1(a)). Although this kind of structure has been mostly used to study quantum transport phenomena in graphene, no investigation on electrical response of h -BN/BLG heterointerface has been conducted from the perspective of device application yet.

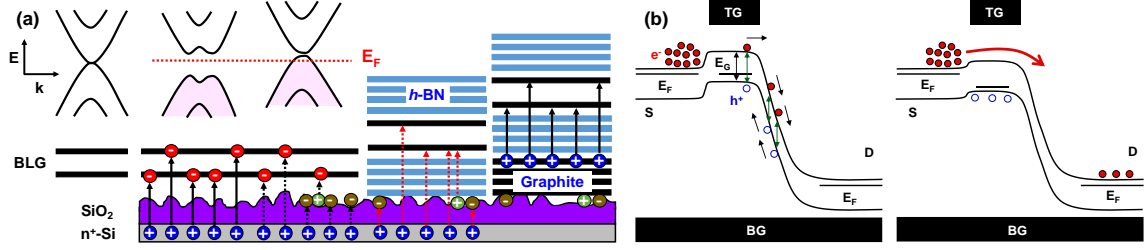


Figure 1. (a) Band gap opening in BLG, band gap variation by charged impurities and reduction of potential fluctuations by all-2D heterostructure. (b) Phototransistor action in dual gate BLG-FET.

Moreover, electrically tunable band gap of BLG can also be used for infrared detection. Recent development in self-driving technology further requires infrared detectors with faster response and room temperature operation. Although fast response can be achieved in quantum type HgCdTe and quantum-dot infrared photodetector, they require cooling as thermal noise greatly reduces detectivity in narrow gap semiconductors at room temperature. Although optical absorption is only 2.3% in each graphene layer, BLG phototransistor with electrically tunable band gap is theoretically suggested to show infrared detectivity even at room temperature because the phototransistor action due to photoexcited carriers can amplify the photocurrent and this amplification can overcome the thermal noise.^[2] In dual gate BLG-FETs with top gate positioned within back gate region shown in Fig. 1(b), gate action which imitates phototransistor action is expected when the position of Fermi energy in dual gate region is set within band gap and the region modulated only by back gate is doped. This relation of band gap and Fermi energy can create potential barrier for electrons and holes for either of the carriers to be swept out of dual gate region and the other to accumulate in dual gate region. Carrier accumulation in dual gate region will lower potential barrier for opposite carrier from source, amplifying the current. This concept has not been experimentally realized.

In this study, the possibility for FET application for BLG was investigated in all-2D layered heterostructure BLG-FETs based on transport characteristics and mobile carrier response at *h*-BN/BLG heterointerface, and phototransistor action in BLG-FETs was identified through observation of photoresponse in all-2D layered heterostructure BLG-FETs.

2. Transport characteristics

All-2D layered heterostructure BLG-FETs were fabricated. It was found that surface contact to BLG channel can be formed by selectively etching top *h*-BN layer by CF₄ plasma, achieving reliable ohmic contact with over 80 % yield. Device properties in all-2D layered heterostructure BLG-FETs was demonstrated to be significantly improved compared to *h*-BN-encapsulated BLG on SiO₂/Si as well as high-*k* Y₂O₃/BLG/SiO₂. For the transfer characteristics at 20 K, *I*_{off} in all-2D layered heterostructure BLG was reduced compared to *h*-BN-encapsulated BLG on SiO₂/Si as well as high-*k* Y₂O₃/BLG on SiO₂/Si at the same $|\bar{D}|$, as shown in Fig. 2(b), indicating drastic reduction of the charged impurities and hence the spatial uniformity of the band gap of BLG. As a result, *I*_{on}/*I*_{off} was greatly increased compared to other devices at the same $|\bar{D}|$, and the maximum *I*_{on}/*I*_{off} at 20 K was

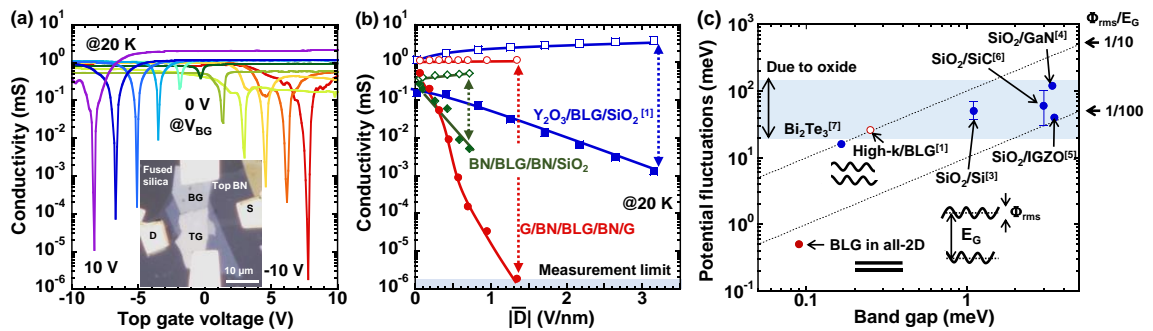


Figure 2. (a) Conductivity as a function of top gate voltage for each back gate voltage measured at 20 K. Inset is optical image of all-2D layered heterostructure BLG-FET. (b) Conductivity for *I*_{on} and *I*_{off} as functions of displacement field. (c) Comparison of potential fluctuations in various insulator/semiconductor gate stack structures.

4.6×10^5 , with the off-state resistivity of $\sim 5 \text{ G}\Omega$ even at $\bar{D} = -1.48 \text{ V/nm}$ where band gap was estimated to be 90 meV. Potential fluctuations were estimated from remnant band gap at $\bar{D} = 0 \text{ V/nm}$ to be 0.5 meV in all-2D layered heterostructure BLG. Potential fluctuations and band gap for various insulator/semiconductor systems are shown in Fig. 2(c). It can be seen that the effect of potential fluctuations due to amorphous oxide insulators is more significant when band gap is small.

3. Mobile carrier response at *h*-BN/BLG interface

Capacitance measurement and conductance method were employed to investigate mobile carrier response in electrical response at *h*-BN/BLG heterointerface. Total capacitance (C_{Total}) between top gate and source was measured in a device fabricated on fused silica substrate where parasitics are greatly reduced. C_{Total} is composed of serial capacitance of geometric capacitance of top gate and quantum capacitance of BLG. Density of state can be extracted from quantum capacitance. The obtained quantum capacitance of gapped BLG shows two sharp van Hove singularity at gap edges, suggesting low disorder (Fig. 3(a)). In conductance method, capture and emission process of mobile carrier trap levels in the band gap can be detected as a deviation from the ideal carrier response seen in frequency dependence of C_{Total} . No frequency dependence of C_{Total} was observed in all-2D layered heterostructure BLG-FETs suggesting negligible interface trap density in the present frequency range (Fig. 3(b, c)). These results suggest that *h*-BN/BLG heterointerface is electrically inert, which is mainly attributed to the drastic reduction of the potential fluctuation to $\sim 0.5 \text{ meV}$ and hence the spatial uniformity of band gap of BLG. This quite low potential fluctuations can be achieved only in all-2D layered heterostructure BLG-FETs, not in conventional semiconductor systems with high-*k* gate stacks. Therefore, all-2D layered heterostructure is suitable for FET application in BLG.

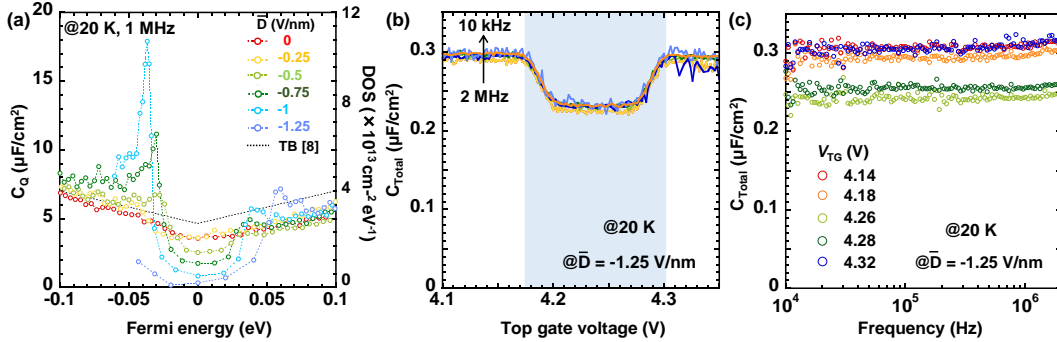


Figure 3. (a) Quantum capacitance for each \bar{D} measured at 20 K. (b) Frequency dependence of C_{Total} as a function of top gate voltage measured while keeping \bar{D} constant. (c) C_{Total} as a function of frequency measured at each top gate voltage in (b).

4. Photodetection

Next, photoresponse in all-2D layered heterostructure BLG-FETs was investigated. Gate length was considered as an important parameter in realizing phototransistor action because phototransistor action requires carrier diffusion through top gate region (Fig. 1(b)). If top gate length is much longer than carrier diffusion length which is calculated to be $\sim 1.3 \mu\text{m}$ in BLG, the phototransistor action will be lost. First, basic photoresponse mechanism without phototransistor action was investigated in a device with $5 \mu\text{m}$ long top gate. Distinct photoresponse was observed at Dirac point (DP) (Fig. 4(a)). There was no consistent photocurrent sign reversal at DP (Fig. 4(b)). Therefore, photovoltaic effect and photothermoelectric effect are considered negligible^[9, 10] and basic photoresponse mechanism in BLG-FETs without phototransistor action was associated with the remaining possible mechanism: bolometric effect. Bolometric response in all-2D layered heterostructure BLG-FET taken as resistivity change at Dirac point (DP) was observed up to 80 K (Fig. 4(c)) which is quite higher than in previous study^[11] (15 K), implying improvement in photoresponse by reducing potential fluctuations in BLG band gap.

Photoresponse to visible light in device with $0.25 \mu\text{m}$ long top gate was investigated. Significantly increased photocurrent compared to bolometric response was observed on the right side of band gap region in boundary of npn and nnn marked by red dotted circle in Fig. 5 (a). There was

no photocurrent sign reversal at DP (Fig. 5 (b)), denying the presence of photothermoelectric effect and photovoltaic effect at BLG/metal contacts.^[9, 10] Therefore, the newly observed photocurrent was associated with phototransistor action. Although with phototransistor action, significantly increased photocurrent should be observed in all V_{TG} in gap region, the results show increased photocurrent only on the right side of band gap region in boundary of npn and nnn. Band structure and potential barrier in BLG in phototransistor regime were estimated at each V_{TG} and V_{BG} . It was found that potential barrier at V_{TG} that gives maximum photocurrent is lower than at DP. This contradicts with theoretical prediction where higher potential barrier is expected to give higher responsivity. The result suggests that when $V_{TG} = DP$, the potential barrier was too high for carrier accumulation under top gate to lower the barrier to amplify photocurrent. On the other hand, when potential barrier is initially set to lower value, carrier accumulation under top gate can lower the barrier and amplify photocurrent. Such behavior seems to be consistent with the mechanism of phototransistor action based on bipolar transistor. It should be noted that maximum photocurrent at similar position in $V_{BG} = 8$ V was smaller than in $V_{BG} = 6$ V band gap (Fig. 5 (c)). This similarly contradicts theoretical prediction where larger band gap which gives higher potential barrier should give higher responsivity. From these observations, it is suggested that effect of potential barrier lowering by carrier accumulation in gate region may be too small for conditions that gives highest potential barrier in present device. Despite higher dark current compared to bolometric response, significantly increased photocurrent compared to bolometric response was consistently observed up to 140 K (Fig. 5 (c)), implying a possibility of phototransistor action where current amplification can overcome thermal noise at higher temperature than bolometric effect. The observed phototransistor action suggests possibility for photodetection application for BLG-FETs. The observed non-ideal photocurrent suggests room for improvement such as using top gate with shorter length and higher optical transmission.

References [1] Sci. Rep. **5**, 15789 (2015). [2] PRB **79**, 245311 (2009). [3] JAP **45**, 2593 (1974). [4] IEEE T. Elec. Dev. **48**, 458 (2001). [5] APL. **65**, 2723 (1994). [6] APL **98**, 203508 (2011). [7] Nat. Phys. **7**, 939 (2011). [8] New J. Phys. **11**, 095010 (2009). [9] Nat. Nanotechnol. **3**, 487 (2008). [10] Science **334**, 648 (2011). [11] Nat. Nanotechnol. **7**, 472 (2012).

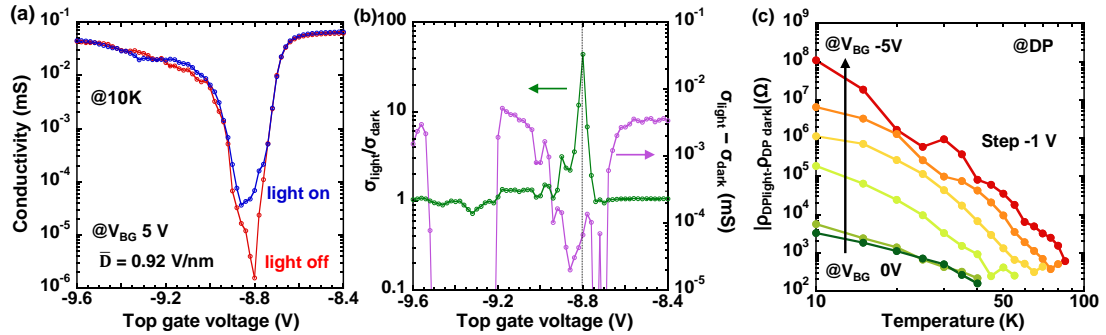


Figure 4. Photoresponse in device with 5 μm long top gate. (a) Conductivity as a function of V_{TG} measured at dark and light condition. (b) Conductivity ratio and difference taken from (a). Dotted line shows position of DP. (c) Temperature dependence of bolometric response.

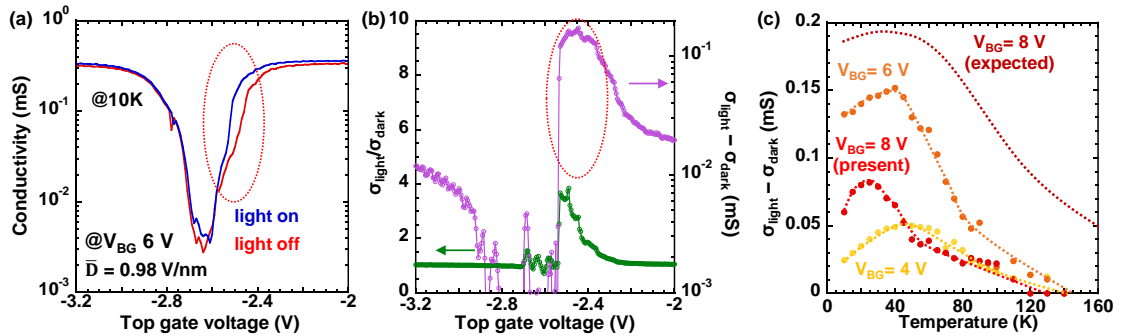


Figure 5. Photoresponse in device with 0.25 μm long top gate. (a) Conductivity as a function of V_{TG} measured at dark and light condition. (b) Conductivity ratio and difference taken from (a). (c) Temperature dependence of maximum photocurrent.