Gate-tunable strong-weak localization transition in few-layer black phosphorus

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Abstract
Atomically-thin black phosphorus (BP) field-effect transistors show strong-weak localization transition, which is tunable through gate voltages. Hopping transports through charge impurity-induced localized states are observed at low carrier density regime. Variable-range hopping model is applied to simulate scattering behaviors of charge carriers. In the high carrier concentration regime, a negative magnetoresistance indicates weak localization effects. The extracted phase coherence length is power-law temperature-dependent \( T^{-0.48\pm0.03} \) and demonstrates inelastic electron–electron interactions and the 2D transport features in few-layer BP field-effect devices. The competition between localization and phase coherence lengths is investigated and analyzed based on observed gate-tunable strong-weak localization transition in few-layer BP.

Supplementary material for this article is available online

Keywords: black phosphorus, strong localization, weak localization, variable-range hopping, nearest-neighbor hopping

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

Introduction
Atomically-thin 2D materials, such as graphene and transition metal dichalcogenides, have provided new approaches for investigating physical property anomalies and potential applications in nanoelectronics and optoelectronics [1–6]. Single- or few-layer black phosphorus (BP) is another promising 2D material, which is characterized by high mobility at room temperature, a tunable direct band gap, and high in-plane anisotropy properties for fundamental studies of thermal, optical, and optoelectronic properties and technological applications [7–17]. Recently, Shubnikov–de Haas oscillations and quantum Hall effects have demonstrated interesting results in high-mobility field-effect transistors (FETs) based on boron nitride (BN)-encapsulated few-layer BP at cryogenic temperatures [18–23]. However, achieving BP FETs with ultra-high mobility at low carrier density remains a challenge in further research of physical properties, such as many-body phenomena and/or fractional quantum Hall effects.

In this study, we report experimental results for scattering behavior of charge carriers in few-layer BP at various charge carrier densities, temperatures, and magnetic fields. We discover that strong localization results in low carrier mobility of several \( \text{cm}^2 \text{V}^{-1} \text{s}^{-1} \) at low carrier concentration regime \( \sim 10^{11} \text{ cm}^{-2} \), and the observed localization effects can be fitted by the variable-range hopping (VRH) model in low-temperature regions [24]. At high carrier concentrations \( \sim 10^{12} \text{ cm}^{-2} \), weak localization occurs, and a high carrier mobility of up to \( 10^3 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1} \) is achieved. We discuss the
mechanism for gate-tunable strong-weak localization transition by extracting phase coherence lengths and inelastic scattering time in high carrier concentrations [25]. The power-law dependence of phase coherence length on temperature demonstrates a 2D transport feature in BP system.

**Experimental**

Few-layer BP and h-BN thin films are exfoliated from bulk crystals on heavily doped silicon substrates covered with 300 nm thick SiO2. Another h-BN flake is simultaneously prepared on poly (methyl methacrylate) (PMMA) thin film. The h-BN flake on PMMA film is used to collect BP flakes, and the formed h-BN/BP structure is then placed on h-BN flakes on a silicon substrate. Then, h-BN/BP/h-BN heterostructure is constructed. All these processes are performed in an inert gas environment to minimize degradation of BP quality. This method prevents direct contact between PMMA film and BP flakes. Annealing at 300 °C in Ar flow for 10 h is applied to stabilize heterostructure.

Reaction-ion etching (RIE) (recite: 4 sccm O2 + 40 sccm CHF3; RF power: 200 W) after electron-beam lithography (EBL) is used to define Hall structures. Then, a selective RIE etching is applied to etch the top h-BN at the contact areas, which are defined by the second EBL. The third EBL is then used to define contact metal patterns followed by electron-beam evaporation to deposit contact metals (Cr/Au = 5/60 nm).

**Results and discussion**

Our few-layer BP FETs are fabricated using exfoliated BP flakes. Primarily caused by oxidation, quality of BP flakes degrades in atmospheric conditions. To avoid the quality degradation, few-layer BP samples are encapsulated between two hexagonal BN (h-BN) sheets to form h-BN/BP/h-BN encapsulation structures [18, 19, 22, 23]. Exfoliation and encapsulation processes are performed in a glove box filled with nitrogen to protect the exfoliated BP flakes (substantial details regarding device fabrication are available in the experimental section). The thickness of encapsulated BP flakes is obtained (~8.2 nm) by subtracting thicknesses of top and bottom BN flakes from the total thickness of the h-BN/BP/h-BN structure (figure S3 is available online at stacks.iop.org/NANO/29/035204/mmedia). It was known that for BP flakes thicker than five layers (~2.7 nm), the band structure and effective mass of BP are weakly dependent on their thicknesses [9]. Therefore, any transport features detected in a thick BP device is largely contributed by its intrinsic properties, rather than its thickness. The electronic transport features have been reproducibly demonstrated in several devices with thicknesses ranging from 5 to 10 nm. In a recent report, 3D weak localization effect was detected in a n-type 20 nm thick BP flake [26]. This effect should be ascribed to the ultra-high carrier concentrations induced by ionic liquid gating instead of layer number in BP flakes. To design the conducting channel directions in our devices, crystal orientations of BP flakes are determined through angle-resolved Raman spectroscopy (figure S4). Unless otherwise specified, all our conducting channels are designed along the X direction (figure S5).

Figure 1(a) reveals variation in BP channel conductance for different gate voltages at 1.8 K. The observed p-type unipolar transport feature is due to the work function alignment between the contact metal Cr and BP crystals [19, 27]. The work function of Cr is well-aligned with the valence band, resulting in a good contact to BP holes. The green arrow indicates charge neutrality point obtained from linear fitting of charge carrier versus gate voltage (figure 1(c)). Charge carrier concentrations shown in figure 1(c) are determined by Hall effect measurement (figure S2). The equivalent capacitance obtained from the linear fitting result is $1.17 \times 10^{-5} \text{F m}^{-2}$ and agrees well with the geometric capacitance. The light blue color bar in figure 1(a) distinguishes the low-mobility region (Region I, $\mu \sim$ a few cm$^2$ V$^{-1}$ s$^{-1}$) from the high-mobility region (Region II, $\mu \sim$ 2000 cm$^2$ V$^{-1}$ s$^{-1}$) and indicates a soft boundary of transition between the two regimes. The different mobility demonstrate distinguishable charge carrier scattering mechanisms in the two regimes. Temperature dependences of mobility $\mu$ share the same characteristic with channel conductance $G$ which is quite intelligible since $G = W/L \cdot \mu n e$, where $W, L$ refer to the width and length of the conducting channels, respectively; $e$ is the elemental charge; $n$ is the carrier density and hardly exhibits any dependence on temperature (supplementary materials). Carrier mobility reaches $1300 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ and $900 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ for Region II and I, respectively.

To investigate scattering behaviors of BP devices, temperature dependencies of channel conductance $G$ with different gate voltages are measured (figure 1(b)). For low carrier concentrations ($V_g > -5 \text{V}$), channel conductance is strongly influenced by temperature variation, especially when temperature surpasses 100 K. In contrast, little impact is observed when temperature is lower than 40 K. This observation indicates a strong localization effect on charge carriers induced by charge impurities at low carrier concentrations. For high carrier concentrations ($V_g < -40 \text{V}$), temperature has little effect on the channel conductance. The measured negative magnetic resistance implies a weak localization effect in our devices at high carrier concentrations. Noticeably, strong-to-weak localization transition occurs when carrier concentration increases in our BP devices.

**Strong localization at low carrier concentrations**

Figure 2(a) shows channel resistance in log scale and plotted as a function of $T^{-1/3}$ at low carrier concentrations for temperatures ranging from 1.8 to 40 K. Linear dependence of log ($R$) on $T^{-1/3}$ agrees with the 2D VRH model: $R \propto \exp (\frac{E_0}{T})^{-\mu}$, where $T_0$ is the characteristic temperature [28]. Nonlinear dependence of log ($R$) on $T^{1/3}$ for $V_g = -10 \text{V}$ (green circle on figure 2(a)) demonstrates failure of 2D VRH model to describe scattering behavior of carrier at high carrier concentrations. Gate voltage dependence of $T_0$ (figure 2(b)) agrees with the VRH model in different systems.
and indicates hopping transport via localized states [5, 29]. Localization length \( \xi_{\text{VRH}} \) can be extracted from \( \xi_{\text{VRH}} = \sqrt{13.8/k_B\rho(E)T_0} \), where \( T_0 \) corresponds to the characteristic temperature obtained from the VRH model, and \( \rho(E) \) refers to density of state (DOS) at the Fermi level [30]. DOS of the 2D hole gas can be expressed as \( \rho(E) = m^*/\pi\hbar^2 \), where \( m^* = 0.26m_0 \) stands for effective mass of holes. \( m_0 \) is the electron rest mass. Figure 2(b) shows calculated gate voltage dependence of \( \xi_{\text{VRH}} \); \( \xi_{\text{VRH}} \) increases as gate voltage decreases (carrier concentration increases), showing a pattern that is widely observed in 2D electron systems [29, 31]. The increasing trend of \( \xi_{\text{VRH}} \) can be explained by the 2D hydrogen atom model. High energy states are occupied when density of localized charge carriers increases, forming 2D hydrogen atoms with a large radius (localization length). From another perspective, increasing localization length may also be explained by screening effect. Localized charge carriers screen the electronic field of localization center and contribute to weak localization strength, that is, large localization length [5, 32].

As temperature increases, resistance deviates from the 2D VRH model and fits the Arrhenius behavior \( R \propto \exp(T_f/T) \), as shown in figure S1(a), where \( T_f \) represents the characteristic temperature reflecting excitation energy. Normally, linear dependence of \( \log(R) \) on \( T^{-1} \) indicates a crossover from VRH to nearest-neighbor hopping model. However, the excitation energy extracted from fitting results is lower than the physical temperature in our device (figure S1(b)). Scattering process is dominated by high-temperature incoherent diffusive transport than nearest-neighbor hopping [33].

**Weak localization at high carrier concentrations**

As carrier concentration increases, dependence of channel resistance on temperature becomes increasingly weak (figure 1(b)). The VRH model fails to describe variation in channel resistance after entering region II (figure 1(a)). To investigate transport behavior of charge carriers at high carrier concentration regime \( \sim 10^{12} \text{ cm}^{-2} \), we measure magnetoconductance of our devices under different gate voltages and temperatures (figures 3(a) and (b), respectively). The applied perpendicular magnetic field results in a positive magnetoconductance (negative magnetoresistance), which is consistent with the features of weak localization [18, 34, 35]. Weak localization is a quantum correction to conductance of diffusive systems originating from phase interference of
charge carrier wave functions. For a 2D electron system with zero Berry phase $\phi$, constructive interference effects of wave functions by backscattered charge carriers will increase the probability of backscattering events. Thus, weak localization effect generally induces negative quantum corrections to channel conductance. An applied perpendicular magnetic field induces additional changes to $\phi$, which breaks constructive interference of backscattering events and consequently leads to positive magneto-conductance. Weak localization strongly depends on concentration and temperature when $T > 2$ K. Magneto-conductance will decrease with increase in temperature when $T > 2$ K. To investigate carrier concentration and temperature dependencies of weak localization, we simulate the measured magneto-conductance with the Hikami–Larkin–Nagaoka (HLN) model:

$$\Delta \sigma = \sigma(B) - \sigma(0) = \frac{e^2}{\pi h} \left[ \frac{1}{2} + \frac{B_0}{B} \right] - \ln \left( \frac{B_0}{B} \right),$$

where $\sigma$ represents channel conductivity; $h$ is Planck’s constant; and $e$ corresponds to elemental charge. $\Psi$ refers to digamma function. $B_0 = \hbar / 4eL_0^2$ calculates the phase coherence magnetic field, and $L_0$ is phase coherence length [36–39]. Green solid lines in figures 3(a) and 3(b) represent carrier concentration and temperature fitting results, respectively, of the HLN model. The HLN model reproduces experimental features of magneto-conductance between $-0.4$ and $+0.4$ T, especially the deep dips at zero magnetic field for gate voltage lower than $-30$ V. When gate voltage is higher than $-30$ V, gradual disappearance of deep dips at zero magnetic field demonstrates that the HLN model fails to describe the measured magneto-conductance, signaling the unsuitability of weak localization model at low carrier concentrations where strong localization occurs.

Figure 4(a) displays temperature dependence of $L_0$ for various gate voltages. A maximum $L_0$ of 202 nm is obtained at $T = 1.4$ K and $V_g = -60$ V. For temperatures lower than 2 K, $L_0$ exhibits weak dependence on temperature. We fit $L_0$ with a power-law formula $L_0 \propto T^{-\beta}$ from 2.3 to 40 K. A universal $\beta = 0.48 \pm 0.03$ for different gate voltages is obtained from fitting results. Specifically, temperature dependence of inelastic scattering time $\tau = L_0^2 / D \propto T^{-\alpha}$ distinguishes different scattering mechanisms, where $\alpha = 2/3$, and $D = \sigma \hbar^2 / m^* e^2$ calculates the diffusion constant. $\tau$ is determined as 5.3 ps at $T = 1.4$ K and $V_g = -60$ V. Linear dependence of $\tau$ on $T$ (figure 4(b)) results in $\alpha = 1$, and this result agrees fairly with the obtained $\beta$ value demonstrating a 2D weak localization model in few-layer BP system [39]. The power-law dependence of phase coherence length on temperature observed by Hensworth et al may arise from the combination effect of strong and weak localization effects considering the low mobility of their devices [39]. Inelastic electronic interactions with small momentum transfer can be described by the Altshuler–Aronov–Khmelnitsky (AAK) theory $\tau_0 = \hbar / L_0^2 \ln(\hbar / e^2)$, and linear dependence of $\tau_0^{-1}$ on $T$ is expected from the theory by considering that channel conductance is independent of temperature. The observed deviation of $\beta$ from 0.5 can be explained by temperature dependence of $\sigma$. Figure 4(c) depicts gate voltage (or carrier concentration) dependence of $L_0$. $L_0$ decreases with decreasing carrier concentration, and this condition can be ascribed to carrier concentration dependence of $\sigma$. The gate-tunable coherence length in bilayer graphene is determined to be an important parameter [40]. Both bilayer graphene and BP exhibit weak spin–orbit coupling (SOC), implying that the gate-tunable coherence length represents a common phenomenon in the 2D systems with weak SOC. The effects of SOC on charge carrier scattering have been discussed theoretically [38]. A strong SOC generally causes an additional scattering, which may dominate over the weak localization effect and thus result in a negative magneto-conductance [38, 40].

In order to determine the anisotropic field-effect mobility $\mu_{\text{FET}}$, we have fabricated a special BP device with eight electrodes in different orientations (figure S5). Consistent with previous experimental results and theoretical predictions, the maximum value of $\mu_{\text{FET}}$ is obtained in the $X$ direction and the value is doubled that in $Y$ direction. The minimum value is achieved at the $Y$ direction as large effective mass is present along the $Y$ direction.
At low carrier concentrations, $f$ increases and reaches $f_{\text{VRH}}$ at high carrier concentrations. In this case, electrons > $f_{\text{VRH}}$ concentrations increase, and the Hall mobility of this device reaches approximately $900 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. A phase coherence length of 109 nm is obtained from the HLN model at $T = 2 \text{ K}$, as shown in figure 5. The short coherence length in the devices along the Y direction is resulted from the combination of low mobility and large effective mass. The mobility effect has been explained by the AAK model. In the strong localization regime (low carrier concentration), a large effective mass generally leads to a small localization length and thus strong localization strength. Strong localization may survive even in a high carrier density range. Overall, a large effective mass will shift the transition point (higher carrier concentration) between strong and weak localizations.

The observed gate-tunable strong-weak localization transition with transition range from $V_g = -10 \text{ V}$ to $V_g = -30 \text{ V}$ (corresponding carrier density $9.8 \times 10^{11} - 2.4 \times 10^{12} \text{ cm}^{-2}$) can be ascribed to the competition between $L_0$ and $\xi_{\text{VRH}}$. At low carrier concentrations, $\xi_{\text{VRH}} < L_0$ leads to a suppression of electronic wave function interference, whereas strong localization dominates weak localizations caused by inelastic electron–electron interactions [33, 41, 42]. As carrier concentrations increase, $\xi_{\text{VRH}}$ increases and reaches $\xi_{\text{VRH}} > L_0$ at high carrier concentrations. In this case, electronic wave function interference dominates scattering of charge carrier and suppression of strong localization [33, 41, 42]. In transition regime, strong and weak localizations influence transport properties of BP samples.

Conclusions

This study examines the distinguishable transport behaviors of charge carriers under varying carrier concentrations in few-layer BP. Strong and weak localization models are proposed to simulate the scattering behavior of charge carriers at low and high carrier concentrations, respectively. Strong localization is confirmed by the observation of VRH at low temperatures. A weak localization model is confirmed by magneto-transport features. Competition behavior between strong localization length and phase coherence length is concluded to occur due to gate-tunable strong-weak localization transition.

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