Edge-Limited Valley-Preserved Transport in Quasi-1D Constriction of Bilayer Graphene

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ABSTRACT: We investigated the quantization of the conductance of quasi-one-dimensional (quasi-1D) constrictions in high-mobility bilayer graphene (BLG) with different geometrical aspect ratios. Ultrashort (a few tens of nanometers long) constrictions were fabricated by applying an undercut etching technique. Conductance was quantized in steps of \(\sim 4e^2/h\) (\(\sim 2e^2/h\)) in devices with aspect ratios smaller (larger) than 1. We argue that scattering at the edges of a quasi-1D BLG constriction limits the intervalley scattering length, which causes valley-preserved (valley-broken) quantum transport in devices with aspect ratios smaller (larger) than 1. The subband energy levels, analyzed in terms of the bias-voltage and temperature dependences of the quantized conductance, indicated that they corresponded well to the effective channel width of a physically defined conducting channel with a hard-wall confining potential. Our study in ultrashort high-mobility BLG nano constrictions with physically tailored edges clearly confirms that physical edges are the major source of intervalley scattering in graphene in the ballistic limit.

KEYWORDS: Bilayer graphene, conductance quantization, intervalley scattering, undercut etching, valleytronics

Bilayer graphene (BLG) has been proposed as a promising platform for realizing valleytronics because it can exhibit in situ tunable valley polarization by opening a bandgap under applied vertical electric fields. It is essential to effectively suppress intervalley scattering in valleytronic applications. One suggested major source of intervalley scattering is short-ranged sharp defects, which enable a large momentum transfer that is required for mixing to occur between valleys K and K'. Thus, for ballistic graphene, the intervalley scattering at atomically disordered physical edges is believed to dominate over bulk scattering. In recent studies, it has been suggested that the intervalley scattering length \(l_v\) in graphene is comparable to the size of micrometer-scale systems. Meanwhile, electrostatic gate-confined carrier guiding without physical edges has enabled valley-preserved transport in monolayer graphene, and BLG with \(l_v\) that is not limited by electrostatically defined confining-potential boundaries. Despite the predicted crucial role of the physical edges for intervalley scattering in graphene, systematic investigation of \(l_v\) governed by physical edges is lacking, as are quantitative estimations of \(l_v\) especially in the case of high-mobility graphene systems.

In this study, we investigated the conductance of physically tailored quasi-one-dimensional (quasi-1D) channels in ballistic BLG to gain insight into the intervalley scattering arising from disordered edges. A conducting channel, with a width in the range of the Fermi wavelength of carriers, exhibited quantized conductance in steps of \(\sim 4e^2/h\) or \(\sim 2e^2/h\) with spin-degeneracy, depending on the conservation of the valley symmetry. Here, \(e\) is the electron charge and \(h\) is Planck’s constant. Therefore, the value of the quantized conductance steps can serve as an indicator of the effect of edge scattering on preservation of the valley symmetry. Taking account of \(\lambda\), according to the system size, we prepared five nanoscale BLG constrictions with different geometrical aspect ratios, which correspond to the length \((L)\) of the constriction to its effective channel width \((W^*\)). We fabricated short nano constrictions beyond the resolution limit of electron-beam lithography using plasma reactive ion etching with an undercut electron-beam resist. The quantized conductance steps of the nano constrictions exhibited valley-preserved and valley-broken behavior when their aspect ratios \((L/W)\) were smaller and larger than 1, respectively. Devices with small aspect ratios \((L/W^* < 1)\) exhibited valley-preserved conductance quantization in steps of \(\sim 4e^2/h\), while the steps were smaller than \(2e^2/h\) in devices with large aspect ratios \((L/W^* > 1)\). This indicates that \(\lambda\) was limited by the channel width due to short-ranged sharp scatterers located dominantly at the disordered edges, which caused large-momentum-transfer scattering. The results of the bias-voltage spectroscopy measurements of the quasi-1D...
energy subbands formed in the BLG constrictions matched well with the hard-wall confinement model for the physical edges. This study gives systematic confirmation that the scattering at the physical edges of a BLG sheet in the ballistic limit is the major source of the intervalley scattering, with its scattering length limited by the spacing between the neighboring physical edges. Better understanding on valley-related transport in bilayer graphene can promote its valleytronic applications and can help resolve the controversies in existing observations.\textsuperscript{21,22} This result can easily be extended to monolayer graphene.

A novel fabrication method was used to prepare ultrashort constrictions, as illustrated in Figure 1. We started with a BLG sheet encapsulated between a pair of hexagonal boron nitride (BN) flakes using a multistage dry-transfer technique.\textsuperscript{23} In this heterostructure, BLG was protected from charged impurities introduced during device fabrication. Graphene-encapsulated by BN is known to have an enhanced mean free path, reaching tens of micrometers.\textsuperscript{23} Moreover, we used a graphite bottom...
gate to avoid charged impurities at the interface between the bottom BN and the bottom gate. This reduced the hysteresis with respect to the electrical bottom gating. Thus, the heterostructures in this study consisted of a stack of top-BN/BLG/bottom-BN/graphite from top to bottom.

To obtain an ultrashort constriction, we used conventional electron-beam lithography and reactive ion etching. Our fabrication method was unique because it used the undercut of an electron-beam resist (poly methyl methacrylate, PMMA) layer and the metallic top-gate layer as the etching stencil. First, we deposited a metal mask of Cr/Au (3 nm/12 nm) layers by electron-beam evaporation (Figure 1a) followed by spin-coating of PMMA. After electron-beam patterning and developing, an undercut was formed along the PMMA boundary due to backscattered electrons from the substrate (Figure 1b).

The range of the undercut could be controlled depending on the type of PMMA, energy of the electron beam, and resist-development time. After depositing a 30 nm-thick Al₂O₃ mask layer, the unmasked part of the heterostructure around the PMMA undercut was etched using O₂/CF₄ plasma. During the plasma etching, the heterostructure was protected by Al₂O₃ and Cr/Au mask layers (Figure 1c). As a result, the width of the metal mask and the range of the undercut determined the width and length of the constriction, respectively. Figure 1e shows a scanning electron microscopy (SEM) image of a device. A cross-sectional transmission electron microscopy (TEM) image along the white dotted line in Figure 1e shows that the constriction was about 50 nm long (Figure 1f). The constriction length ranged from 20 to 110 nm for different devices. This novel undercut-etching technique enabled us to create an ultrashort constriction, which was shorter than or comparable to the electron-beam lithography limit.

Figure 2a shows schematics of the measurement configuration of the BLG constriction, including the top and bottom gates. Electrical contacts were made of Cr/Au (10 nm/90 nm) layers. A metallic layer was used as an etching mask and top gate electrode, which allowed dual-gate operation in combination with the graphite bottom gate. Transport measurements were performed using low-frequency lock-in techniques with a root-mean-square bias current of 100 nA at a frequency of 17.7 Hz. A four-probe configuration was used to exclude the contact resistance.

Five devices were examined in this study. The discussion is focused primarily on two devices of different widths (170 and 110 nm for devices A and B, respectively) and lengths (40 and 110 nm for devices A and B, respectively). The width (W) and length (L) of the constrictions were determined using SEM images. Each device consisted of two BLG reservoirs and a constriction. Wider BLG reservoirs made a negligible contribution to the series resistances; thus, the measured conductance solely represents the value of a constriction. Figure 2b shows the conductance of device A with varying top (\(V_{TG} \)) and bottom (\(V_{BG} \)) gate voltages. Using a parallel plate capacitor model, the total carrier density \(n_{tot} \) of the constriction can be expressed as 

\[
\begin{align*}
 n_{tot} &= \frac{\varepsilon_{BN} \varepsilon_0 (V_{BG} - V_{bot}^0)}{W L},
\end{align*}
\]

Figure 3. Quantized conductance steps. (a) Conductance \(G \) of devices A and B with different widths (\(W \)) and lengths (\(L \)), determined by SEM images, as a function of the Fermi wavenumber \(k_F \). \(G \) was measured at 0.16 K for device A and at 4 K for device B. The gray lines indicate a linear relationship between \(G \) and \(k_F \). The magenta lines are to guide the eye. (b) Conductance \(G \) of devices A and B as a function of the total carrier density induced by gate voltages. The magenta curves represent the modified relationship between \(G \) and \(n_{tot} \), taking into account the trap states. (c) Quantized conductance steps of device A are denoted as a cyan trace in part b. The minima of transconductance \(dG/dn_{tot} \) versus \(G \) correspond to the conductance plateaus, with a modulation period of \(\sim 4e^2/h \). (d) Quantized conductance steps of device B are denoted as a blue trace in part b. The minima of transconductance \(dG/dn_{tot} \) versus \(G \) correspond to the conductance plateaus, with a modulation period of \( \sim 1.7e^2/h \). (e) Quantized conductance steps versus aspect ratios of the constrictions for the five devices used in this study.
The temperature dependence and bias voltage dependence of the conductance quantization steps to estimate the size of the energy spacing between subbands. Figure 4a shows transconductance $dG/dn_{tot}$ for device A at temperatures ranging from 0.16 to 15 K. Increasing temperature diminishes

$ed_{bot} + \varepsilon_{BN} n_{tot}(V_{TG} - V_{top}^{0})/d_{top}$ where $\varepsilon_{BN}$ is the relative dielectric constant of BN, $d_{bot}$ ($d_{top}$) is the thickness of the bottom (top) BN sheet, and $V_{top}^{0}$ ($V_{bot}^{0}$) is the bottom (top) gate voltage for the CNP. In addition, the displacement field, $D = \varepsilon_{BN}(V_{TG} - V_{top}^{0})/2d_{top} - \varepsilon_{BN}(V_{TG} - V_{bot}^{0})/2d_{bot}$ opens the energy gap ($E_{gap}$) by breaking the inversion symmetry of BLG.

As a result, lines 1 and 2 in Figure 2b correspond to the variation in $n_{tot}$ and $E_{gap}$ respectively. $E_{gap}$ and $n_{tot}$ are fixed in the respective cases. BLG reservoirs were only affected by the bottom gate.

Therefore, npp or nnp junctions formed between the reservoir and the constriction in the gating regions $\alpha$ of the conductance map in Figure 2b, where the transmission probability was reduced by the presence of the pn potential barriers. We measured quantized conductance with the same corresponding transconductance $dG/dn_{tot}$ measured at 4 K for device B. Each curve was taken at a fixed gate voltage, corresponding to the curve in Figure 3d. Emergence of $G_{diff}$ plateaus at a higher bias of $V_{dc} = 7$ mV leads to a subband energy spacing of 14 meV (i.e., the spacing between the dotted vertical lines).

Figure 4. Energy dependence of the quantized conductance steps. (a) Variation of the transconductance $dG/dn_{tot}$ for device A with increasing temperature for $T = 0.16, 2, 6, 10,$ and $15$ K from left to right. Quantized conductance steps survive even at $T = 15$ K. (b) Voltage-bias dependence of the differential conductance ($G_{diff}$) measured at 4 K for device B. Each curve was taken at a fixed gate voltage, corresponding to the curve in Figure 3d.

$G = 4e^{2}/h \cdot W^{*} \cdot k_{F}' \cdot \sqrt{n_{tot} - n_{F}(E)}$ (magenta curves in Figure 3b). The modified G value agrees well with the measured data, except near the CNP. Parts c and d of Figure 3 show zoomed-in views of G in Figure 3b as a function of $n_{tot}$ with the corresponding transconductance $dG/dn_{tot}$. The conductance steps are emphasized by the vanishing $dG/dn_{tot}$. For device A, we observed distinct conductance quantization steps, $\Delta G$, close to $4e^{2}/h$ (Figure 3c). With preservation of both the spin and valley symmetry, $\Delta G$ should be $(4e^{2}/h)\pi$, but it becomes $(2e^{2}/h)\pi$ for spin symmetry only. As $t \leq 1$, a $\Delta G$ value near $4e^{2}/h$ indicates that both the spin and valley symmetries are preserved with $t \sim 1$. This suggests that the channel length of device A ($\sim 40$ nm) was shorter than $\lambda_{v}$. We also observed valley-symmetric conductance steps in two other devices with ultrashort lengths ($L < 60$ nm) and similar widths ($170-190$ nm). However, device B was longer ($L \sim 110$ nm) and showed conductance steps of $\Delta G \sim 1.7e^{2}/h$. To fit the conductance using the modified relationship that accounts for trap states near the edge disorders (magenta curves in Figure 3(b)) to extract an effective channel width of $W^{*} \sim 130$ nm for device A and $W^{*} \sim 14.5$ nm for device B. We attribute the discrepancy between $W^{*}$ and $W$ to overetching of the graphene edges by reactive ion etching and/or disorder-induced localization of carriers at the edges.

When $\Delta G$ is less than $2e^{2}/h$, it is not clear whether valley symmetry is preserved or broken, where $\Delta G < 2e^{2}/h$ can be either due to small $t < 0.5$ with preserved valley symmetry, or to broken valley symmetry. However, previous studies have suggested that $\lambda_{v}$ is comparable to the width of graphene, because intervalley scattering occurs predominantly at the edges of graphene rather than the interior. $L > W^{*}$ at $V_{dc} = 20$ mV for device B. We used the measured $\Delta G$ value near $4e^{2}/h$ to obtain the value of $t \sim 0.85$ for the measured $\Delta G = 1.7e^{2}/h$. As shown in Figure 3e, devices with smaller aspect ratios have $\Delta G$ values close to $4e^{2}/h$ for preserved valley symmetry, which confirms that $\lambda_{v}$ is comparable to $W^{*}$.
the conductance plateaus due to thermal broadening of $k_B T$, where $k_B$ is the Boltzmann constant. The conductance plateaus survived at 15 K, which was the highest temperature reached in our measurements; thus, the energy spacing was larger than $\sim 1.3$ meV. Voltage bias spectroscopy was carried out to extract the energy spacing of the subbands for device B at $T \simeq 4$ K (Figure 4b). Each trace shows differential conductance with respect to the voltage bias at a fixed gate voltage. Zero-bias differential conductance exhibits quantized conductance steps corresponding to the trace in Figure 3d. At a finite bias of $V_{dc} \sim \pm 7$ meV, additional plateaus appear between quantized conductance steps at zero-bias, $V_{dc} \sim 0$.

This indicates that the energy spacing between subbands of device B was $\sim 14$ meV. In a single-particle configuration, each subband energy of BLG with a hard-wall potential is $E_N = \hbar^2 \pi^2 N^2 / 2m^* W^2$, where $m^* (=0.33 m_e)$ is the electronic effective mass of BLG and $N$ is a positive quantum number, yielding an increasing subband energy spacing with increasing quantum number $N$. However, voltage bias spectroscopy shows that the subband energy spacing was nearly equal for different $N$ (Figure 4b). We attribute this to the dispersion relationship in the band structure of BLG, which becomes almost linear for large wavenumbers. In the tight-binding model, the low energy band of BLG is approximated by a quadratic equation ($E \sim p^2 / 2m^*$) for small wavenumbers, but it becomes linear ($E \sim vp$) for large wavenumbers, with $v \sim 10^6$ m/s. Such a quadratic-to-linear crossover takes place for $k_F \sim \gamma_l / 2v \hbar = 260 \times 10^6$ m$^{-1}$, corresponding to $n_e \sim 3.5 \times 10^{12}$ cm$^{-2}$ for the device B, where $\gamma_l (=0.35$ eV) is the interlayer hopping parameter. Because voltage bias spectroscopy was performed above the crossover point, the dispersion relationship should be approximately linear, which results in equally spaced subband energies with $\Delta E = v \hbar / W^* = 42$ meV in a hard-wall potential. In the presence of valley mixing, however, the valley-degenerate subbands are split and valley is no longer a good quantum number, giving rise to the quantized conductance steps of $2e^2 / h$. This indicates that a modified subband energy spacing $\Delta E_{f}^*$ is needed when considering the valley mixing. Because the measured subband energies are equally spaced for different $N$ as shown in Figure 4b, the subband splitting energy is estimated as $\Delta E_{ff} = 0.5 \Delta E$. Thus, the modified subband energy spacing is predicted to be $\Delta E_{f} = 0.5 \Delta E = 21$ meV, which corresponds well to the observed value of $\sim 14$ meV.

In summary, to understand the role of physically etched constrictions, we studied quasi-1D transport of ultrashort constrictions in BLG fabricated via PMMA undercut etching. For devices with small aspect ratios ($<1$), we observed quantized conductance steps with 4-fold degeneracy close to $4e^2 / h$ in zero magnetic field, suggesting carrier transport with preserving valley symmetry through nano constrictions. On the other hand, for devices with large aspect ratios ($>1$), quantized conductance steps smaller than $2e^2 / h$ were observed with broken valley-symmetry. This result indicates that $L < W^*$ is essential for valley-symmetry-preserved transport in physically etched BLG because $\lambda_e$ is limited by $W^*$. This study gives solid confirmation of the general belief that the scattering at the physical edges of a graphene layer in the ballistic limit is the major source of the intervalley scattering. In the presence of valley mixing, the voltage bias dependence of quasi-1D transport shows almost equally spaced confinement subbands of $\sim 14$ meV. This agrees with the calculation for a hard-wall potential of width $W^*$, assuming a linear dispersion of the band structure. The latter is explained by the tight-binding model for large wavenumbers. In contrast to the gate-confined carrier guiding, a hard-wall confining potential of physically etched BLG is robust against the gate tuning, which provides the merits of generating valley polarized current near the CNP.\textsuperscript{1,5} Combining observed valley-preserved transport and gate-independent confining potential, dual-gated BLG with physically etched edges would create new opportunities for valleytronic devices with gate-tunable BLG.

- **ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.8b02750.

Ballistic transport of encapsulated bilayer graphene constrictions, quantized conductance steps for different aspect ratios of the BLG constrictions, and quantized conductance steps for different displacement fields (PDF)

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**Author Contributions**

H.L. and H.-J.L. conceived the idea and designed the experiments. H.L. prepared the devices and performed the measurements. All authors analyzed the data. H.L., G.-H.L., and H.-J.L. wrote the manuscript. H.-J.L. supervised the study. All authors contributed to the discussion and approved the final version of the manuscript. K.W. and T.T. provided high-quality hexagonal boron nitride substrates.

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**Notes**

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- **REFERENCES**


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