Full electric-field tuning of the nonreciprocal transport effect in massive chiral fermions with trigonal warping

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In a noncentrosymmetric system, an intrinsic electric polarization is allowed and may lead to unusual nonreciprocal charge transport phenomena. As a result, a current-dependent resistance, arising from the magnetoelectric anisotropy term of $k \cdot \vec{E} \times \vec{B}$, appears and acts as a current rectifier with the amount of rectification being linearly proportional to the magnitude of both current and applied magnetic field. In this work, a different type of nonreciprocal transport effect was demonstrated in a graphene-based device, which requires no external magnetic field. Due to the unique pseudospin (valley) degree of freedom in chiral fermions with trigonal warping, a large nonreciprocal transport effect was uncovered in a gapped bilayer graphene, where electric-field tunabilities of the band gap and valley polarization play an important role. The exact cancellation of nonreciprocal effect between two different valleys is effectively removed by breaking the inversion symmetry via electric gateings. The magnitude of the current rectification appears to be at a maximum when the Fermi surface undergoes a Lifshitz transition near the band edges, which is proportional to the current and the displacement field strength. The full electric-field tuning of the nonreciprocal transport effect without a magnetic field opens up a new direction for valleytronics in two-dimensional-based devices.

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I. INTRODUCTION

From a symmetry argument, a magnetoelectric anisotropy term in the form of $k \cdot \vec{E} \times \vec{B}$ can generally exist in all systems [1], which gives rise to additional contributions to various magnetoelectric properties in matter [2–5]. Among them, an intriguing subject that has been studied recently is the nonlinear and nonreciprocal transport effect (NRTE). An unusual current-direction dependence on the charge transport is a direct consequence of the NRTE. Phenomenologically, it can be described by a current-dependent resistance of $R = R_0[1 + \gamma \vec{I} \cdot (\vec{P} \times \vec{B})]$, where γ is the NRTE coefficient and $\vec{P}$ is the charge polarization of the system. As is evident from the equation, the NRTE appears only when the current ($\vec{I}$), the magnetic field ($\vec{B}$), and $\vec{P}$ are all nonzero and also perpendicular to each other, setting stringent criteria for the experimental realizations. So far, a number of discoveries for NRTE have been made in various systems, such as bulk-Rashba polar semiconductors [6], chiral magnets [7], noncentrosymmetric superconductors [8–12], and magnetic bilayer heterostructures [13–15]. In theories, the microscopic mechanism of NRTE is typically associated with the velocity asymmetry and the scattering lifetime asymmetry in the Boltzmann transport equation, giving rise to a nonzero transport coefficient in the E^2 term. Therefore, such a NRTE can be readily probed by

measuring the second-harmonic responses to an alternating driving current with a frequency of ω . In general, the nonlinear (E^2 term) resistance ($R_{2\omega}$) is negligibly small as compared to E -linear resistance (R_ω), but the ratio of $R_{2\omega}/R_\omega$ can be as large as 0.01 or higher in systems with NRTE.

As it turns out, the Dirac-like systems with chiral fermions are good platforms for the study of NRTE. The trigonal warping effect [16,17] and the chiral property with (pseudo)spin-momentum locking give rise to the asymmetries of energy dispersion and velocity on the Fermi surface. A large current-dependent resistance was reported in a magnetic topological insulator (TI)/TI heterostructure, which was attributed to the Dirac surface states and also magnon-driven asymmetric scattering [14]. On the other hand, in a thin film of nonmagnetic Bi_2Se_3 , a sizable $R_{2\omega}$ also appears when the charge transport is dominated by the Dirac surface states [18]. The Dirac Fermi surface with trigonal warping is further distorted by an applied magnetic field parallel to the film surface, giving rise to the observed $R_{2\omega}$ signals. In other words, the applied magnetic field acts effectively as a spin-to-charge converter via the nonlinear resistance channel due to the spin-momentum locking. The aforementioned systems all require an external magnetic field for the appearance of NRTE, which put a severe limitation for the practical device applications. Here, we report a type of NRTE that is rooted in the valley-momentum locking effect of massive and trigonal-warped chiral fermions with Berry phase 2π in a dual-gated bilayer graphene (BLG) device. The valley degree of freedom in a BLG is a counterpart of the spin degree of freedom, but the manipulation of the valley polarization, in principle, can be made possible by electric gating only. Large $R_{2\omega}$ signals were revealed in a dual gate BLG device whenever the Fermi energy (ϵ_F) was tuned to be near the band edges.

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FIG. 1. The setup of a dual-gated BLG device and the measured R_ω and $R_{2\omega}$ signals. (a) An optical image of a dual-gated BLG device with a schematic for a measurement circuit. (b) An illustration of the current-dependent resistance in a gapped BLG. The inset cartoon shows the AB-stacked BLG, where A1 and B2 are the two sublattices on different layers in a reduced two-component pseudospin representation. Parts (c) and (d) are the R_ω and $R_{2\omega}$ signals, respectively, as a function of V_{bg} and V_{tg} at $T = 5$ K. The corresponding R_ω - V_{bg} and $R_{2\omega}$ - V_{bg} curves at different V_{tg} values are shown in (e) and (f), respectively. The curves at different V_{tg} values are shifted for clarity.

Its magnitude grows larger with increasing displacement field strength $|D|$ and also the bias current. We note that the $|D|$ not only opens up a band gap in a BLG but also effectively polarizes the valley degree of freedom, and thus the exact cancellation of the NRTE from two different valleys of $K+$ and $K-$ is removed to result in a large $R_{2\omega}$ response. This effect is fully adjustable by electric gating alone and requires no external magnetic field. Those findings can be understood qualitatively from the velocity asymmetry in the Boltzmann transport equation for massive chiral fermions with trigonal warping.

II. EXPERIMENTAL RESULTS

Figure 1(a) shows an optical image of a dual-gated BLG device we fabricated, where the measurement circuits were also illustrated (see Appendix A for more information). A BLG was sandwiched between two hexagonal boron nitride (hBN) microcrystals with thicknesses of about a few tens of nanometers, as illustrated in Fig. 1(b). The device geometry was designed to give more than 80% of the BLG area between the voltage leads to be top-gate effective. Therefore, the measured voltage signals are mostly dominated by the BLG region that can be simultaneously tuned by the top-gate voltage (V_{tg}) and bottom-gate voltage (V_{bg}), enabling the introduction of a large displacement field D on a BLG. By neglecting the screening effect, the strength of the displacement field can be calculated via $D = [\epsilon_b(V_{bg} - V_{bg0})/d_b - \epsilon_t(V_{tg} - V_{tg0})/d_t]/2$,

where ϵ_b (ϵ_t) is the relative dielectric constant for the bottom (top) gate, and d_b (d_t) is the dielectric layer thickness of the bottom (top) gate. The operation of a dual-gated device not only capacitively varies the density of the system, but it also changes the D values that act on the BLG [19,20]. As demonstrated in Fig. 1(b), a band gap ϵ_g appears due to a finite potential difference (Δ) between the upper and lower layer of BLG due to D , giving rise to a rapid increase of the zero-density sheet resistance ($R_{\omega 0}$) with increasing D as shown in Figs. 1(c) and 1(e). The color codes in Fig. 1(c) represent the values of sheet resistance R_ω in logarithmic scale at $T = 5$ K, and the corresponding R_ω - V_{bg} curves at different V_{tg} values ranging from $+3$ to -3 V are shown in Fig. 1(e), where V_{bg0} and V_{tg0} were determined to be ≈ -1.86 and -0.15 V, respectively. The $R_{\omega 0}$ equals about 2.2 k Ω at zero V_{tg} , and it increases by more than two orders of magnitude of $R_\omega \approx 285.6$ k Ω at $V_{tg} = -3$ V, which corresponds to $D \approx +0.58$ V/nm. The large variation in R_ω indicates a high device quality, which is also supported by the high Hall mobility of $\approx 10\,000$ cm²/V s as determined from the Hall measurements (see Appendix B). Surprisingly, large second-harmonic signals $R_{2\omega}$ were uncovered and shown in Figs. 1(d) and 1(f). The color codes in Fig. 1(d) indicate the $R_{2\omega}$ values in linear scale, and the corresponding $R_{2\omega}$ - V_{bg} curves at different V_{tg} values are shown in Fig. 1(f). Pronounced $R_{2\omega}$ values appear near the zero-density regime with magnitudes rapidly growing for $|V_{tg}| \geq 2$ V. As V_{bg} sweeps from $+50$ V to -50 V, $R_{2\omega}$ first reaches a local maximum and then rapidly falls off to

FIG. 2. The current (I)-dependence and D -dependence of the NRTE. (a) The upper panel and lower panel show the R_{ω} - V_{bg} and $R_{2\omega}$ - V_{bg} curves, respectively, with different bias currents ranging from 20 to 200 nA at $T = 5$ K. The local extremal values of $R_{2\omega}$ ($R_{2\omega p}$ and $R_{2\omega v}$) are practically linear with I [lower panel of (b)], which is in big contrast to the nearly I -independent $R_{\omega 0}$ [upper panel of (b)]. The corresponding D -dependence of local extremal values of $R_{2\omega}$ and R_{ω} is plotted in the upper and lower panel, respectively, in (c). The inset cartoon of the lower panel illustrates the cross-sectional view of a dual-gated device with a given displacement field D .

a local minimum with an opposite sign. At $V_{tg} = -3$ V, the magnitude of $R_{2\omega}$ is as large as 5.0 k Ω . The bias current I equals 100 nA for all the data shown in Fig. 1, and the curves at different V_{tg} values in Figs. 1(e) and 1(f) are shifted for clarity.

The current dependence of R_{ω} and $R_{2\omega}$ with $V_{tg} = -2.5$ V and $T = 5$ K is shown in the upper panel and lower panel, respectively, of Fig. 2(a). By setting different I values ranging from 20 to 200 nA, the local extremal values in $R_{2\omega}$ - V_{bg} curves increase in magnitude with increasing I , where $R_{2\omega p}$ and $R_{2\omega v}$ are defined as the values at the peak and valley, respectively, of the $R_{2\omega}$ - V_{bg} curve as shown in the lower panel of Fig. 2(a). On the contrary, the R_{ω} data in the upper panel of Fig. 2(a) show negligible variations with I , where $R_{\omega 0}$ is defined as the peak value in an R_{ω} - V_{bg} curve. The nearly I -independent $R_{\omega 0}$ thus suggests an insignificant Joule heating effect during the measurements with a current up to 200 nA. The detailed I -dependence of $R_{\omega 0}$ and the magnitude of $R_{2\omega p,v}$ are plotted in the upper and lower panels of Fig. 2(b), respectively. For a 10-fold increase in I , the variation of $R_{\omega 0}$ at $V_{tg} = -2.5$ V [solid circles in the upper panel of Fig. 2(b)] is less than $\approx 3\%$, which is in large contrast with the practical I -linear dependence of $|R_{2\omega p(v)}|$ as shown in the lower panel of Fig. 2(b). The $R_{2\omega p}$ at $V_{tg} = -2.5$ V increases from a value of 1.08 k Ω for $I = 20$ nA up to 7.22 k Ω for $I = 200$ nA. $R_{2\omega v}$ [open circles in the lower panel of Fig. 2(b)], on the other hand, shows a consistent behavior with a slight difference in magnitude. Upon reversing the sign of V_{tg} , similar behaviors were observed for $V_{tg} = +2.5$ V. As revealed in Fig. 1(f), we remark that the same $R_{2\omega}$ - V_{bg} profiles as that in the lower panel of Fig. 2(a) are found, regardless of the sign of the V_{tg} and

thus D . By collecting all the data with different V_{tg} values, the D -dependence of $|R_{2\omega p(v)}|$ and $R_{\omega 0}$ in logarithmic scales is shown in the upper panel and lower panel, respectively, of Fig. 2(c). Both $|R_{2\omega p(v)}|$ and $R_{\omega 0}$ are exponentially growing with D in the low- D regime, and it slows down for $|D| \geq 0.4$ V/nm. By tuning D up to about a magnitude of ≈ 0.58 V/nm, we remark that the $|R_{2\omega p(v)}|$ spans more than four orders of magnitude, which is more than twofold larger than that for $R_{\omega 0}$. The inset cartoon in the lower panel of Fig. 2(c) illustrates the sign of D in relation to the device geometry.

In a dual-gated BLG device, the unscreened D values as a function of V_{bg} and V_{tg} can be calculated as shown in Fig. 3(a). The sweeping of gate voltages along a D -constant contour line ensures the shifting of only the ϵ_F in BLG while keeping the degree of inversion asymmetry unchanged. As an example, a sweeping along a black line, connecting point 1 and point 2 in Fig. 3(a) along a D -constant contour line, will result in a shifting of ϵ_F from the conduction band to the valence band as illustrated in the lower panel of Fig. 3(a). We carried out such sweepings along D -constant contour lines at 10 different D values ranging from $+0.5$ to -0.5 V/nm. The upper and lower panels of Fig. 3(b) show the measurement results for R_{ω} and $|R_{2\omega}|$, respectively, as a function of the corresponding sheet density n_{2D} . When $\epsilon_F = 0$ ($n_{2D} = 0$), R_{ω} attains a maximum value of $R_{\omega 0}$, but $R_{2\omega}$ is nearly zero. As ϵ_F moves toward the conduction (valence) band with $n_{2D} \leq 0$ ($n_{2D} \geq 0$), $|R_{2\omega}|$ first increases rapidly to a local maximum and then falls off to a vanishingly small value for $|n_{2D}| \geq 3 \times 10^{11}$ cm $^{-2}$. Δn_{2D} is the sheet density at which $|R_{2\omega}|$ reached a maximum, as illustrated in the lower panel of Fig. 3(b). An enlarged view of the $R_{2\omega}$ - n_{2D} data in a linear scale is shown in Fig. 3(c),

FIG. 3. The tuning of Fermi energy (ϵ_F) with a constant D . (a) The D values vs V_{bg} and V_{tg} . A D -constant sweep from point 1 to point 2 in (a) corresponds to a shift in ϵ_F from the conduction band to the valence band across a band gap. (b) The R_{ω} and the magnitude of $R_{2\omega}$ ($|R_{2\omega}|$) in logarithmic scales are plotted as a function of the sheet density n_{2D} with various D values ranging from +0.5 to -0.5 V/nm. The Δn_{2D} , as defined in the lower panel of (b), is the sheet density at which $R_{2\omega}$ attains a local extremal value. Part (c) shows an enlarged view of $R_{2\omega}$ - n_{2D} , which shares the same color codes and lines denoted in (b). The corresponding D -dependence of Δn_{2D} is shown in (d).

which shares the same color codes as in Fig. 3(b). As noted before, by reversing the sign of D [dashed lines in Figs. 3(b) and 3(c)], consistent behaviors of R_{ω} and $|R_{2\omega}|$ are observed with a minor difference in magnitude, but no sign changes in $R_{2\omega}$ were found due to the sign reversal of D . The extracted Δn_{2D} is plotted as a function of D , as shown in Fig. 3(d). For $|D| \leq 0.4$ V/nm, Δn_{2D} tends to drop slightly with increasing $|D|$ to a value of $\approx 3.7 \times 10^{10}$ cm⁻² at $D = 0.4$ V/nm.

The T -dependent measurements were carried out to extract the band gap ϵ_g using a thermal-activation model for the conductance. The normalized zero-density conductance of $R_{\omega 0}(D=0)/R_{\omega 0}$ versus $1/T$ curves for different D values up to ≈ 0.58 V/nm is plotted in Fig. 4(a), where the red lines are fitting curves using a formula of $R_{\omega 0}(D=0)/R_{\omega 0} = A \exp[-\epsilon_g/2k_B T] + C$. The extracted band gap ϵ_g as a function of the corresponding D is shown in Fig. 4(b), and ϵ_g is practically linear with D , giving $\epsilon_g \approx 60$ meV at $D = +0.58$ V/nm. The NRTE turns out to diminish faster with increasing temperature as compared to $R_{\omega 0}$. Figure 4(c) shows the T -dependence of the ratio for local extremal $R_{2\omega}$ to the corresponding R_{ω} , where it decays from a value of about 2.8% at $T = 5$ K to 0.28% at $T = 80$ K. The ΔV_{bg} , defined as the V_{bg} difference between the local extremes in $R_{2\omega}$ - V_{bg} curves, is shown in Fig. 4(d), which shows similar behavior to Δn_{2D} in Fig. 3(d). At $T = 5$ K, ΔV_{bg} first decreases with increasing $|D|$ up to a critical value of $D_c \approx 0.4$ V/nm. For $D \geq D_c$, ΔV_{bg} shows a small increase with $|D|$. We note that D_c turns out to

move to a higher value as T increases. For all the T -dependent data, the $R_{2\omega p(v)}$ versus the corresponding R_{ω} collapsed on a single curve, as demonstrated in Fig. 4(e), giving a relation of $R_{2\omega p(v)} \propto R_{\omega}^{2.02}$.

III. NUMERICAL CALCULATIONS OF NRTE

As illustrated in the lower panel of Fig. 5(a), the sublattices A1 and B2 on different layers form an effective honeycomb lattice in the reduced two-component pseudospin representation with two different valleys of $K+$ and $K-$. The reduced 2×2 Hamiltonian of a BLG [21] can be described by $\mathcal{H}_{BLG} = -\frac{\hbar^2}{2m^*} \sigma \cdot \vec{k}^2$, where $\vec{k}^2 \equiv [(k_x^2 - k_y^2)\hat{x} + 2k_x k_y \hat{y}]$, m^* is the effective mass, σ is the Pauli spin matrices, and $\hat{x}$ ($\hat{y}$) is a unit vector along the x (y) axis. The resulting two-component pseudospin states, referring to the electron probabilities on sublattices A1 and B2, are thus chiral fermions with the valley-momentum locking effect. By further considering trigonal warping and an asymmetry parameter Δ [22], the energy dispersion can be calculated and expressed as

$$\begin{aligned} \varepsilon_{\alpha, \xi}^2(p, \phi) &= \frac{\gamma_1^2}{2} + \frac{\Delta^2}{4} + \left(v^2 + \frac{v_3^2}{2}\right)p^2 + (-1)^\alpha \sqrt{\Gamma}, \quad \text{and} \\ \Gamma &= \frac{1}{4}(\gamma_1^2 - v_3^2 p^2)^2 + v^2 p^2 [\gamma_1^2 + \Delta^2 + v_3^2 p^2] \\ &\quad + 2\xi \gamma_1 v_3 v^2 p^3 \cos(3\phi), \end{aligned} \quad (1)$$

FIG. 4. The temperature dependence of R_{ω} and $R_{2\omega}$. (a) Normalized sheet conductance $[R_{\omega 0}(D=0)/R_{\omega 0}]$ as a function of $1/T$ at different D values. The red lines are the fitting curves using the thermal-activated conductivity formula. The extracted band gap ϵ_g is nearly linear with D as shown in (b), giving a band gap of $\epsilon_g \approx 67$ meV at $D = 0.58$ V/nm. (c) The ratio of local extremal $R_{2\omega}$ to the corresponding R_{ω} as a function of T for different D values. The ΔV_{bg} values are obtained from the difference of the V_{bg} values at which the $R_{2\omega}$ attains local extremal values in each $R_{2\omega}$ - V_{bg} curve. At $T = 5$ K, ΔV_{bg} first drops with increasing D up to a turning point of $D_c \approx 0.4$ V/nm, and then it starts to increase in the high- D regime. D_c turns out to shift to a bigger value with increasing T as shown in (d). (e) The local extremal $R_{2\omega}$ values as a function of the corresponding R_{ω} in logarithmic scales, giving a relation of $R_{2\omega} \propto R_{\omega}^{2.02}$. The data points share the same symbol and color code as that in (d).

where $(p = \hbar k, \phi)$ is the momentum in polar coordinate, γ_1 is the interlayer nearest-neighbor hopping amplitude, $\xi = +1$ (-1) refers to the $K+$ ($K-$) valley, and v is the Fermi velocity. The trigonal warping derives from the additional velocity term of $v_3 \equiv \sqrt{3}a\gamma_3/(2\hbar)$, where a is the lattice parameter for the sublattice and γ_3 is the next-nearest interlayer hopping amplitude as illustrated in the inset cartoon of Fig. 1(b). As revealed from Eq. (1), there are four bands as shown in the upper panel of Fig. 5(a) for $\Delta = 100$ meV, where the color codes represent the energy. The black and red contours for the lower energy band ($\alpha = 1$) are at $\epsilon = +300$ and $+100$ meV, respectively, where the corresponding constant energy contours as a function of k_x and k_y are shown in Fig. 5(b). Apparent distortions of the Fermi surface from symmetric circles (dashed lines) are observed, and we note that the distorted Fermi surfaces resemble rounded triangles pointing to the opposite direction for $K+$ and $K-$. Similar behaviors are found for $\epsilon = -300$ and -100 meV as shown as green and blue contours in Fig. 5(c). The calculated velocity $(\partial\epsilon/\partial k_x)$ and $\partial^2\epsilon/\partial k_x^2$ as a function of k_x for $\Delta = 100$ meV are shown in the upper and lower panels of Fig. 5(d), respectively, for lower energy bands of $K+$ and $K-$ valleys. For each valley, there are asymmetries in both $\partial\epsilon/\partial k_x$ and $\partial^2\epsilon/\partial k_x^2$ with respect to k_x , but it cancels exactly while summing up contributions from both valleys of $K+$ and $K-$. Therefore, an additional breaking of the valley symmetry (i.e., valley

polarization) is needed in order to have a nonzero contribution due to the asymmetry terms of $\partial\epsilon/\partial k_x$ and $\partial^2\epsilon/\partial k_x^2$, which is required for the appearance of NRTE to be shown as follows.

Based on the Boltzmann transport theorem [23], the nonequilibrium distribution function (g_k) up to the second order of driving electric-field (E^2) can be described by $g_k = \frac{e\tau E_x}{\hbar} \frac{\partial f_0}{\partial k_x} + (\frac{e\tau E_x}{\hbar})^2 \frac{\partial^2 f_0}{\partial k_x^2}$, where we used $\vec{E} = E_x \hat{i}$. The resulting second-harmonic conductance $\sigma_{2\omega}$ due to NRTE at $T = 0$ K for the low-energy bands ($\alpha = 1$) can be expressed as (see Appendix C)

$$\sigma_{2\omega, \xi}(\epsilon) = \frac{q^3}{2\pi\hbar^2} \int d\phi \tau^2 p \cos^3 \times \phi \left[3 \frac{|v_{k, \xi}|}{v_{k, \xi}} \frac{\partial v_{k, \xi}}{\partial p} - \frac{\partial |v_{k, \xi}|}{\partial p} \right]_{\epsilon}, \quad (2)$$

where q is the charge of carriers, τ is the transport lifetime, $p = \hbar k$, and $v_{k, \xi} = \partial\epsilon_{1, \xi}/\partial p$. For simplicity, we neglect the complex asymmetric scattering effects for chiral fermions in a BLG, and Eq. (2) reduces to a simple form of $\sigma_{2\omega, \xi} = \frac{q^3 \tau^2}{2\pi\hbar^2} I_{2\omega, \xi}$, where the ϵ_{ξ} -dependent $I_{2\omega, \xi}$ term with a unit of velocity ($s^{-1} m$) can be numerically calculated from Eq. (1) and shown in Fig. 6(a) for $\Delta = 50$ meV (see Appendix C). For $\xi = +1$ ($K+$ valley), as ϵ decreases from 55 meV, the magnitude of $I_{2\omega, +1}$ increases, reaching a maximum value near the

FIG. 5. Calculated band structures and velocity asymmetry of a gapped BLG with trigonal warping. (a) The band structure of a BLG with asymmetric parameter $\Delta = 100$ meV. The lower panel of (a) shows the A1 and B2 sublattices in a reduced honeycomb lattice depicted by black dashed lines, comprising the two different valleys of $K+$ and $K-$. The corresponding constant energy contour plots of the low-energy band for $K+$ and $K-$ are shown as solid lines in the right panel and left panel of (b), respectively. Part (c) shows the results for the negative low-energy band. The color codes for the solid lines in (b) and (c) are the same as that in (a), indicating Fermi surfaces at different energies. Those Fermi surfaces show apparent deviations from a perfect circle [dashed lines in (b) and (c)] due to the trigonal warping effect, which thus gives rise to the asymmetries in velocity ($\partial\epsilon/\partial k_x$) and $\partial^2\epsilon/\partial k_x^2$ at Fermi surfaces as demonstrated in the upper panel and lower panel of (d), respectively.

band edge at $\epsilon \approx 25$ meV [dashed line in Fig. 6(a)]. For $\epsilon \leq 25$ meV, a Lifshitz transition occurs [21,24,25], and the single rounded-triangle Fermi surface transformed into multiple separated ellipses as illustrated in the right panel of Fig. 6(a) (see Appendix C). The magnitude of $I_{2\omega,+1}$ drops rapidly to zero for $\epsilon \leq 24.3$ meV, where the total area of ellipses and thus the density of states are negligibly small. As expected, the $I_{2\omega,+1}$ turns out to be canceled exactly by $I_{2\omega,-1}$ for $K-$ at the same ϵ [red line in Fig. 6(a)]. We remark that such a Lifshitz transition is a direct consequence of the trigonal warping effect that becomes significant at lower energy, and it happens for all Δ values near the band edges [25]. The corresponding energy ϵ_L for the occurrence of the Lifshitz transition equals ≈ 1 meV for $D = 0$, and $\epsilon_L \approx \Delta/2$ for a finite D . For more rigorous calculations, the asymmetric scatterings in τ and the correlation effect should be considered [14,26,27], which is beyond the scope of the current work. However, we argue that the inclusion of the asymmetric scatterings in τ should further amplify the asymmetry terms in Eq. (2) and thus may not affect the qualitative scenario that we described here.

IV. DISCUSSIONS

The breaking of the inversion symmetry by D not only results in a finite ϵ_g but also gives rise to a valley polarization in a gapped BLG [28,29]. A large nonlocal resistance has been reported in a gapped BLG and is attributed to the inverse valley Hall effect with D -enhanced valley polarization [30–32]. As illustrated in Fig. 6(b), the D -induced

potential difference between the upper and lower layer of a BLG causes a small energy difference for the electrons in different sublattices (valleys). The energy difference can be described by $\Delta\epsilon \approx qDd\delta n/n_c$, where d is the distance between layers, $n_c \equiv \frac{\gamma_1^2}{\pi\hbar^2 v^2}$, and δn is the D -induced density difference between layers. The resulting Fermi energies in $K+$ (black solid line) and $K-$ (red solid line) are then shifted in the opposite direction, as illustrated in the right panel of Fig. 6(b). The total second-harmonic conductance is then described by

$$\begin{aligned} \sigma_{2\omega} &= \sigma_{2\omega,+1}(\epsilon_F + \Delta\epsilon) + \sigma_{2\omega,-1}(\epsilon_F - \Delta\epsilon) \\ &= \frac{q^3 \tau^2}{\pi \hbar^2} \left[\frac{\partial I_{2\omega,+1}}{\partial \epsilon} \right] \Delta\epsilon. \end{aligned} \quad (3)$$

As a result, a nonzero $\sigma_{2\omega}$ appears when $\Delta\epsilon$ and thus D is finite. The measured second-harmonic resistance can be linked to the calculated $\sigma_{2\omega}$ via $R_{2\omega} = -\frac{I_0}{W} \frac{\sigma_{2\omega}}{\sigma_{\omega}^3}$, where W is the width of the device, I_0 is the magnitude of the bias current, and $\sigma_{\omega} = 1/R_{\omega}$ (see Appendix C). It thus justified the I -linear dependence shown in the lower panel of Fig. 2(b). The calculated $\sigma_{2\omega}$ then reflects the behavior of $R_{2\omega}$. The numerical calculations of the $\partial I_{2\omega,+1}/\partial \epsilon$ term for the $K+$ valley are plotted as a function of shifted energy ($\epsilon - \Delta/2$) shown in Fig. 6(c) for three different Δ values of 10, 30, and 50 meV. For the $K-$ valley, it gives the same values with the opposite sign. The shaded area corresponds to a small energy window (≈ 0.2 meV) in which the Lifshitz transition occurs, where large and discontinuous values of $\partial I_{2\omega,+1}/\partial \epsilon$ are found due to the abrupt changes in the density of states

FIG. 6. Numerical calculations for the nonreciprocal term of $\sigma_{2\omega} = \frac{q^3 \tau^2}{2\pi \hbar^2} I_{2\omega}$ in the Boltzmann transport equation. Part (a) plots the energy dependence of the calculated $I_{2\omega}$ with $\Delta = 50$ meV. The black and red denote $K+$ and $K-$, respectively. The six insets on the right (left) of (a) correspond to the Fermi surfaces at six different energies for $K+$ ($K-$). A transformation into three separated ellipses was found when ϵ_F is close to the band edge. Upon introducing D on a BLG, a small energy difference $\Delta\epsilon \approx qD\delta n/n_c$ appears as illustrated in (b). It thus breaks the exact cancellation of $I_{2\omega}$ terms for $K+$ and $K-$, giving rise to a nonzero $\sigma_{2\omega}$. The corresponding term of $\partial I_{2\omega} / \partial \epsilon$ as a function of shifted energy ($\epsilon - \Delta/2$) is shown in (c) for three different Δ values of 50, 30, and 10 meV. In general, the magnitude of $\sigma_{2\omega}$ is at maximum when ϵ_F is near the band edge, and it falls off rapidly as ϵ_F moves away from the Lifshitz transition energy [shaded region in (c)]. (d) The illustrations of the pseudospin states for electron-type carriers and the corresponding valley polarization in a BLG with $D \neq 0$.

at the Lifshitz transition (see Appendix C). This is an outcome from the band calculations at zero temperature, and, in principle, it can be smoothed out when taking into account a finite-temperature effect. In general, $\partial I_{2\omega, +1} / \partial \epsilon$ appears to be the largest when ϵ_F is close to the band edge with multiple separated Fermi surfaces. It then rapidly falls off as $\epsilon_F \geq \epsilon_L$. The decrease of $\partial I_{2\omega, +1} / \partial \epsilon$ with increasing ϵ becomes faster as D increases, which agrees qualitatively with the data shown in the lower panel of Fig. 3(b). We also note that the density for the maximum $R_{2\omega}$ shown in Fig. 3(d) gives $\Delta n_{2D} \approx (3-8) \times 10^{10} \text{ cm}^{-2}$, which is within the same order of magnitude for the density at $E_L \approx \Delta/2$ (see Appendix C). On the other hand, by observing Eq. (3), the sign of total $\sigma_{2\omega}$ is dictated by $q\Delta\epsilon \approx D\delta n$. When ϵ_F moves from the conduction band to the valence band with a constant D , δn changes sign, and thus $R_{2\omega}$ undergoes a sign reversal. On the contrary, reversing the sign for D also causes a sign reversal in δn , and it thus gives no sign changes for the resulting $R_{2\omega}$. This is also expected from the system symmetry point of view, where the longitudinal resistances of R_ω and $R_{2\omega}$ are symmetric with respect to D as shown in Fig. 2(c). Those behaviors are well consistent with our observations shown in Figs. 1(f) and 3(c). For an order-of-magnitude estimation of $R_{2\omega}$, we consider the case with $D = 0.6 \text{ V/nm}$ and a total density of $n_{2D} \approx 6 \times 10^{10} \text{ cm}^{-2}$ based on our experimental data, and it gives $\Delta\epsilon \approx 0.8 \text{ meV}$, which takes into account the self-consistent screening effect [21]. A rough estimation of the transport lifetime, using an effective mass of $0.024m_e$ [36] and a sheet resistance of $200 \text{ k}\Omega$, gives $\tau \approx 7 \text{ fs}$. From Eq. (3) and $R_{2\omega} = -\frac{I_0}{W} \frac{\sigma_{2\omega}}{\sigma_\omega}$, the calculated $R_{2\omega}$

for a bias current of 100 nA equals $\approx 2 \text{ k}\Omega$ with a ratio of $R_{2\omega}/R_\omega \approx 1\%$, which is in the same order of magnitude as the measured $R_{2\omega}$.

From $R_{2\omega} = -\frac{I_0}{W} \frac{\sigma_{2\omega}}{\sigma_\omega}$, the measured $R_{2\omega}$ is expected to scale with R_ω with exponent larger than 3. This is not the case, however, as shown in Fig. 4(e), giving $R_{2\omega} \propto R_\omega^\lambda$ with $\lambda \approx 2.02$. As it turns out, the exponent λ is device-dependent (see Appendix D). The charge-puddle effect is known to give rise to potential fluctuations and a finite minimum conductance at the charge-neutral point in graphene-based systems [33–35]. It may enhance the conductance near the band edges, resulting in a reduced R_ω and spoiling the scaling of $R_{2\omega} = R_\omega^\lambda$, particularly in the low- D regime. A rough estimate from the V_{bg0} value in our device gives an impurity-charge density $n_{\text{imp}} \approx 1.3 \times 10^{11} \text{ cm}^{-2}$, which is larger than the Δn_{2D} shown in Fig. 3(d). This indicates the importance of the charge-puddle effects in our device, and more rigorous theoretical descriptions that include charge-puddle effects are needed. On the technical side, we also noticed a ω -dependence on the measured $R_{2\omega}$ (see Appendix E). This is attributed to the current shunting effect due to the capacitance mainly from the macroscopic gold electrodes on $\text{SiO}_2(300 \text{ nm})/\text{Si}$, which can become larger as ϵ_g increases in the high- D regime. Such a finite capacitance in the measurement circuits can shunt the higher-frequency signals and thus cause a much reduced amplitude of $R_{2\omega}$ as compared to R_ω . On the other hand, we also note that the coexistence of top-gate effective and ineffective BLG regions as shown in Fig. 1(a) may lead to PN-junction-like rectification effects. In a control device with

FIG. 7. A demonstration of the current rectification in a dual-gated BLG device. Part (a) illustrates the device setup and measurement procedure. A dc current (I_{dc}) drawn by a biased voltage (V_{dc}) was measured with alternating V_{bg} values at which the $R_{2\omega}$ attains local extremal values. The upper panel and lower panel of (b) show the resulting time dependence of V_{bg} and I_{dc} signals, respectively, for different V_{dc} values.

a top-gate electrode fully covering the BLG (see Appendix D), consistent NRTE results were observed, thus excluding the rectifying contribution due to the trivial PN junction effect. Even though the issue of the sample-dependent exponent λ requires further investigations, the observed large $R_{2\omega}/R_{\omega}$ is well reproducible in all dual-gated BLG devices we fabricated, and, in principle, a larger $R_{2\omega}/R_{\omega}$ can be achieved with a further increase in D .

As illustrated in Fig. 6(d), the directions of the eigen pseudospin states in the reduced 2×2 Hamiltonian for a BLG are determined when a bias current and thus a propagation wave vector k are given, which make ϕ and $\pi - \phi$ angles to the current direction due to its unique chiral property. For electron-type carriers ($\varepsilon_F > 0$), the two eigenstates of pseudospin-up (S_e^+) and pseudospin-down (S_e^-) correspond to electrons fully on A1 sites (upper-layer) and B2 sites (lower-layer), respectively [29]. Therefore, the valley polarization S can be defined as the charge-density difference between the two layers of BLG with a direction along the eigen pseudospin states, i.e., $S = \delta n / 2 (S_e^+ - S_e^-)$. Nevertheless, a nonzero D is required to break the inversion symmetry and thus give rise to a finite value for $S \times I$. Phenomenologically, the observed NRTE in a gapped BLG can then be described by a current-dependent resistance in the form of $R = R_0 [1 + (-1)^\beta \gamma' I \cdot (D \times S)]$, where γ' and S are the NRTE coefficient and valley polarization, respectively. β equals 0 and 1 for electron-type and hole-type carriers, respectively, to account for the opposite signs of the $q\Delta\varepsilon$ term for the conduction band and the valence band. Upon reversing D , the term of $S \times I$ changes sign as required by the system symmetry, where the roles of A1 and B2 sites are exchanged. Therefore, there is no sign

change for $R_{2\omega}$. On the other hand, when ε_F moves to the valence band (hole-type carriers) at a constant D , S attends the same sign, and the prefactor of $(-1)^\beta$ gives rise to the sign reversal of $R_{2\omega}$. Finally, a scheme for a practical electric-field tuning of the current rectification effect is demonstrated in Fig. 7(a). By sourcing a dc voltage (V_{dc}), the measured dc current (I_{dc}) is found to give a high-current and low-current state, corresponding to the setting values of V_{bg} for extremal $R_{2\omega p}$ and $R_{2\omega v}$, respectively. As shown in Fig. 7(b), the differences in the high-current and low-current states scale linearly with the magnitude of V_{dc} and thus I_{dc} . Upon reversing the sign of V_{dc} , the high-current and low-current states are also reversed, as expected.

V. CONCLUSION

In conclusion, we uncovered an unusually large $R_{2\omega}$ signal in dual-gated BLG devices, which turns out to grow with increasing bias current and also displacement field D . Such a NRTE in a zero magnetic field sets a clear distinction from other NRTE systems that require the breaking of the time-reversal symmetry by magnetic field. The largest ratio of $R_{2\omega}/R_{\omega}$ for $I = 100$ nA was found to be about 2.8% at $T = 5$ K and $D = +0.58$ V/nm. The extremal $R_{2\omega}$ occurs whenever the Fermi energy is shifted to the band edges, and its magnitude quickly declines when Fermi energy moves away from the Lifshitz transition energy. Those behaviors agree qualitatively well with the calculated velocity asymmetry contribution in the Boltzmann transport equation, giving a total second-harmonic conductance $\sigma_{2\omega} = \frac{q^3 \tau^2}{\pi \hbar^2} \left[\frac{\partial I_{2\omega, +1}}{\partial \varepsilon} \right] \Delta \varepsilon$ with $\Delta \varepsilon \approx q D d \delta n / n_c$. The breaking of the inversion symmetry by

D not only removes the exact cancellation between $I_{2\omega,+1}$ and $I_{2\omega,-1}$ but also results in valley polarization, which is crucial for the appearance of NRTE in a BLG. The scaling relation of $R_{2\omega} \propto R_{2\omega}^\lambda$ gives device-dependent values for the exponent λ ranging from 1.3 to 4.17, which infers a somewhat reduced increase of $R_{2\omega}$ with D in our dual-gated BLG devices. Nevertheless, the revelation of a large $R_{2\omega}$ for the massive chiral fermions with trigonal warping enables a full electric-field tuning of the NRTE and opens up a new possibility for the valleytronics in graphene-based 2D devices.

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APPENDIX A: METHODS

High-quality natural graphites and hexagonal boron nitrides were mechanically exfoliated by tapes and then transferred onto 300 nm SiO_2/Si wafers. From optical contrast analyses and atomic force microscope scanings, high-uniformity flakes with proper thicknesses were selected and sequentially stacked using the dry transfer technique with a polycarbonate/PDMS stamp. For the device shown in Fig. 1(a), the thicknesses for the top hBN-layer and bottom hBN-layer are about 22 and 6 nm, respectively. Contact electrodes of Ti(5 nm)/Au(30 nm) were then fabricated by an electron beam lithography facility. The device measurements were carried out in a closed-cycle cryostat that provides stable sample temperature above $T = 5$ K. A lock-in amplifier was used for the measurements of R_ω and $R_{2\omega}$ simultaneously with a bias current at a low frequency of 18.111 Hz.

APPENDIX B: THE SHEET DENSITY AND HALL MOBILITY

To characterize the properties of the bilayer graphene sample, we have performed Hall measurements at $T = 5$ K with $V_{\text{tg}} = 0$. The Hall resistance was found to be field-linear in weak fields up to 0.5 T, and the corresponding sheet density n_{2D} was obtained from the Hall coefficient $R_H = \frac{1}{n_{2D}e}$, where e is the elementary charge. We measured the Hall resistance at 0.5 and -0.5 T and estimated the carrier density with respect to V_{bg} , as shown in Fig. 8(a). We have also tried to evaluate the capacitance of the bottom-gate in our dual-gated device. The capacitance $C = \frac{\Delta n_{2D}e}{\Delta V_{\text{bg}}}$ can be determined from the slope for the curve of carrier density as a function of voltage of the bottom-gate. The capacitance values in the electron-doping and hole-doping regions are around 91.34 and 109.72 aF/ μm^2 , respectively, which are well consistent with the nominal value of around 110 aF/ μm^2 estimated from a simple parallel plate capacitor model. The corresponding mobility μ as a function of the bottom-gate is shown in Fig. 8(b), using the relation of conductivity $\sigma = n_{2D}e\mu$. The mobility is more than 10 000 $\text{cm}^2/\text{V s}$ in the electron-doping region, and it is slightly higher for the hole-doping region. We do note some anomaly in the hole-doping region near $n_{2D} = (0.5-1.0) \times 10^{12} \text{ cm}^{-2}$, which is likely due to the contribution from top-gate ineffective regions between the voltage leads.

APPENDIX C: CALCULATIONS OF $\sigma_{2\omega}$ BASED ON THE BOLTZMANN TRANSPORT EQUATION

The equilibrium Fermi-Dirac distribution function is given by $f_0 = \frac{1}{e^{(\epsilon-\mu)/k_B T} + 1}$. The total electron distribution is $f_{\vec{k}} = f_0 + g_{\vec{k}}$, where $g_{\vec{k}}$ refers to the nonequilibrium distribution function. Using a relaxation approximation with $(\frac{\partial f_{\vec{k}}}{\partial t})_{\text{collision}} = -\frac{g_{\vec{k}}}{\tau}$, we set $\vec{E} = E_x \hat{i}$ and keep terms up to E^2

FIG. 8. Hall effect measurements. (a) The sheet density n_{2D} as a function of V_{bg} . The red dashed line is a curve with a capacitance of ≈ 110 aF μm^{-2} via the bottom gate. (b) The extracted mobility μ as a function of V_{bg} .

FIG. 9. The numerically calculated $I_{2\omega}$ and n_{2D} for the $K+$ valley at different values of asymmetry parameter Δ . The calculated $I_{2\omega}$ and n_{2D} as a function of the Fermi energy ϵ_F are plotted in the upper panel and lower panel of (a) for $\Delta = 10$ meV, respectively. Part (c) shows the calculated $I_{2\omega}$ and n_{2D} for $\Delta = 30$ meV.

for $g_{\vec{k}}$, and then

$$g_{\vec{k}} = -\frac{q\vec{E}\tau}{\hbar} \frac{\partial(f_0 + g_{\vec{k}})}{\vec{k}}, \quad \approx -\frac{qE_x\tau}{\hbar} \frac{\partial f_0}{\partial k_x} + \left(\frac{qE_x\tau}{\hbar}\right)^2 \frac{\partial^2 f_0}{\partial k_x^2}. \quad (C1)$$

The current density for a two-dimensional system is then

$$\begin{aligned} j_x &= 2q \int v_{\vec{k}} g_{\vec{k}} \frac{d^2 \vec{k}}{(2\pi)^2} \\ &= \left[-\frac{q^2}{\hbar} \int \frac{d^2 \vec{k}}{2\pi^2} \tau v_{\vec{k}} \frac{\partial f_0}{\partial k_x} \right] E_x + \left[\frac{q^3}{\hbar^2} \int \frac{d^2 \vec{k}}{2\pi^2} \tau^2 v_{\vec{k}} \frac{\partial^2 f_0}{\partial k_x^2} \right] E_x^2 \\ &\equiv \sigma_{\omega} E_x + \sigma_{2\omega} E_x^2. \end{aligned} \quad (C2)$$

Therefore, the second-harmonic conductance can be expressed as [23]

$$\begin{aligned} \sigma_{2\omega} &= \frac{q^3}{\hbar^2} \int \frac{d^2 \vec{k}}{2\pi^2} \tau^2 v_{\vec{k}} \frac{\partial^2 f_0}{\partial k_x^2} \\ &= \frac{q^3 \tau^2}{2\pi \hbar} \oint dS_F \cos^3 \phi \left[3 \frac{|v_{\vec{k}}|}{v_{\vec{k}}} \frac{\partial v_{\vec{k}}}{\partial p} - \frac{\partial |v_{\vec{k}}|}{\partial p} \right]_{\epsilon_F} \\ &\equiv \frac{q^3 \tau^2}{2\pi \hbar^2} I_{2\omega}, \end{aligned} \quad (C3)$$

where $dS_F = p d\phi/\hbar$ at the Fermi surface, $v_{\vec{k}} = \partial \epsilon/\partial p$, and $I_{2\omega} \equiv \oint d\phi \frac{1}{p} \cos^3 \phi \left[3 \frac{|v_{\vec{k}}|}{v_{\vec{k}}} \frac{\partial v_{\vec{k}}}{\partial p} - \frac{\partial |v_{\vec{k}}|}{\partial p} \right]_{\epsilon_F}$ is the path integral along closed Fermi surfaces. Here, we assume that τ is independent of p and ϕ .

By numerically calculating $v_{\vec{k}}$ and $\partial v_{\vec{k}}/\partial p$ for a gapped BLG with trigonal warping, $I_{2\omega}$ and thus $\sigma_{2\omega}$ can be obtained. Using the energy dispersion for a gapped BLG with trigonal warping [Eq. (1)], the $I_{2\omega}$ term can be numerically calculated and shown in the upper panels of Figs. 9(a) and 9(b) for two

different asymmetry parameters of $\Delta = 10$ and 30 meV, respectively, where the corresponding calculated sheet densities n_{2D} are shown in the lower panel. The inset cartoons show the corresponding Fermi surfaces in different ϵ_F regimes. For $\Delta = 10$ meV [Fig. 9(a)], there are two Lifshitz transitions at energies of ϵ_{L1} and ϵ_{L2} . ϵ_{L1} equals ≈ 5.09 meV, corresponding to a transition from a single Fermi surface (black line) to four separated Fermi surfaces (blue line). As ϵ_F keeps decreasing, another transition into three separated Fermi surfaces (red line) occurs at $\epsilon_{L2} \approx 5.00$ meV. Figure 9(b) shows the results for $\Delta = 30$ meV, where only one Lifshitz transition from a single Fermi surface to three separated Fermi surfaces was observed at $\epsilon_L \approx 15$ meV. We note that both calculated $I_{2\omega}$ and n_{2D} values show abrupt changes at Lifshitz transition energies. The sheet density at ϵ_{L1} (ϵ_L) for $\Delta = 10$ meV (30 meV) equals $\approx 1.3(3.9) \times 10^{10} \text{ cm}^{-2}$, which turns out to increase gradually with Δ . Within our experimental D values, the sheet density at the Lifshitz transition is expected to be in the range of $\approx (1-10) \times 10^{10} \text{ cm}^{-2}$, which is in the same order of magnitude as the sheet density that we observed in experiment for the occurrence of a maximum $|R_{2\omega}|$ [Fig. 3(d)].

The measured quantity of $R_{2\omega}$ can be related to σ_{ω} and $\sigma_{2\omega}$ by inverting the current density equation shown in Eq. (C2) [14], giving

$$E_x \approx [1/\sigma_{\omega}] j_x - [\sigma_{2\omega}/\sigma_{\omega}^3] j_x^2. \quad (C4)$$

Therefore, the magnitude of the measured second-harmonic sheet resistance $R_{2\omega}$ can be shown to be

$$R_{2\omega} \equiv \frac{V_{2\omega} W}{I_0 L} = -\frac{I_0}{W} \frac{\sigma_{2\omega}}{\sigma_{\omega}^3}, \quad (C5)$$

where $V_{2\omega}$ is the measured second-harmonic voltage, I_0 is the magnitude of the driving current, and $W(L)$ is the device width (length between voltage electrodes).

FIG. 10. Semiconducting PN phase diagram of a dual-gated BLG device. (a) An illustration of a dual-gated BLG device with top-gate effective and ineffective regions. (b) Sheet resistance of BLG as a function of bottom gate and top gate at 5 K. The dashed black line is a visual guide showing the charge neutrality of the top-gate effective region. The dashed and solid black lines divide the phase diagram into four parts of different polarities. The observed sign of $R_{2\omega}$ is also marked in (b). The red dashed line indicates the D -constant line of $D = +0.2$ V/nm across three different PN phases.

APPENDIX D: DISCUSSIONS OF THE POSSIBLE PN JUNCTION EFFECT AND $R_{2\omega}$ IN OTHER DUAL-GATED BLG DEVICES

Electron/hole-doping concentration in BLG is manipulated by a top-gate and a bottom-gate. The silicon wafer is used as a bottom-gate, so the whole area of BLG is affected by the bottom-gate. However, the area of the top-gate is limited and only a part of BLG can be affected by the top-gate, as shown in Fig. 1(a). Thus, the BLG device is divided into three regions, as illustrated in Fig. 10(a), where only the middle region is top-gate effective and the other two regions are only bottom-gate effective (or top-gate ineffective). The sheet resistance (R_{ω}) as a function of V_{bg} and V_{tg} at 5 K is shown in Fig. 10(b). The dashed line shows the locations for R_{ω} peaks and corresponds to the charge neutrality for the top-gate effective region of a gapped BLG. The solid line represents $V_{bg} = V_{bg0} (\approx -1.86$ V), which gives a maximum R_{ω} at zero V_{tg} , which separates the P-type and N-type phases for the top-gate ineffective regions. These two lines divide the resistance map into four phases, representing four different combinations of NNN, NPN, PPP, and PNP based on the Fermi level location as illustrated in Fig. 10(a). It is known that a PN junction results in a current rectification effect. However, our dual-gated device comprises two PN junctions that work more like a transition with gate-tunable conductance across the device, and it is not likely to exhibit current-directional-dependent resistance for a single PN junction. Starting from NPN phase, for example, the conductance reaches a minimum near the dashed-line region when moving along the D -constant line to the NNN phase as expected for a transistor-like behavior. Supposedly, there is somehow a difference in the two top-gate ineffective regions [Fig. 10(a)], and it thus may cause certain current-directional-dependent effects. It still cannot explain the observed complex $R_{2\omega}$ behaviors as described in

the main text, including the sign changes and also the rapid decays of its magnitude away from the band edges. On the other hand, when moving along the $D = +0.2$ V/nm line [red dashed line in Fig. 10(b)], we shall observe another large $R_{2\omega}$ when moving from NPN to PPP phases, but this was not happening. As also shown in Fig. 11(a), a control device with the top-gate electrode fully covering the voltage leads was fabricated to ensure no PN junctions within the voltage leads, and a consistent NRTE effect was observed. We argue, therefore, that the observed large $R_{2\omega}$ is not likely due to the trivial PN semiconducting junction or diode-related effects.

To avoid a possible PN-junction-like effect, we fabricated a dual-gated BLG device with the top-gate electrode fully covering the voltage leads as shown in Fig. 11(a). The Hall mobility is about 5000 $\text{cm}^2/\text{V s}$, which is slightly lower than the device shown in Fig. 1(a) due to the additional Hall-bar patterning process on BLG. The resulting $R_{2\omega}$ data are shown in Fig. 11(b), where $R_{2\omega}$ attends a peak near the conduction-band edge and then rapidly drops to a valley near the valence-band edge when moving along the D -constant contour line. The corresponding scaling plot of $R_{2\omega}$ versus R_{ω} in a logarithmic scale is shown in Fig. 11(c), giving a relation of $R_{2\omega} \propto R_{\omega}^{4.17}$ and a maximum ratio of $R_{2\omega}/R_{\omega} \approx 1.5$ %. Another dual-gated BLG device was fabricated with a much narrow top-gate electrode of only about 0.8 μm as shown in Fig. 11(d). The Hall mobility is about 10^5 $\text{cm}^2/\text{V s}$ at $n_{2D} \approx 1 \times 10^{11}$ cm^{-2} . The measured $R_{2\omega}$ are shown in Fig. 11(e) with an exponent $\lambda \approx 1.87$ [Fig. 11(f)]. The maximum ratio of $R_{2\omega}/R_{\omega}$ is about 7%. For more than five dual-gated BLG devices with Hall mobilities ranging from 3000 to 10^5 $\text{cm}^2/\text{V s}$, $R_{2\omega}$ signals appear in all the devices and also show consistent behavior and a similar order of magnitude for NRTE as described in the main text. However, the exponent λ is shown

FIG. 11. The measured $R_{2\omega}$ for other dual-gated BLG devices. (a) The optical microscope image of a dual-gated BLG with a top gate fully covering the voltage leads. The corresponding $R_{2\omega}$ as a function of V_{bg} and V_{tg} at $T = 5$ K is shown in (b), which shows a consistent behavior with that described in the main text. (c) The scaling plot of $R_{2\omega}$ vs R_{ω} gives an exponent $\lambda \approx 4.17$. On the other hand, the results for another dual-gated BLG device with a much narrower top-gate electrode is shown in (d), (e), and (f), giving an exponent $\lambda \approx 1.87$.

to be sample-dependent with values ranging from about 1.3 to 4.17.

APPENDIX E: THE FREQUENCY DEPENDENCE AND COMPARISON TO RESULTS FROM PULSED DC MEASUREMENTS

For ac measurements, a change in the driving frequency turns out to cause a variation in both measured R_{ω} and $R_{2\omega}$. The frequency dependences of R_{ω} and $R_{2\omega}$ are shown in Figs. 12(a) and 12(b), respectively. Both R_{ω} and $R_{2\omega}$ drop with increasing driving frequency, where the decay in $R_{2\omega}$ with increasing frequency appears to be more significant. This indicates the capacitance effect in our measurements circuits, which is mainly derived from the macroscopic gold electrodes. The whole dual-gated device comprises a template with macroscopic gold pads with size up to about 3 mm. An estimated capacitance of Au/SiO₂(300 nm)/Si can be as large as 150 pF, and it gives an impedance of about 1 MΩ at 1 kHz, which is comparable to the $R_{\omega 0}$ at low temperature. Therefore, the ac measurements for probing the NRTE in dual-gated devices should be carried out in the low-frequency regime to

avoid the shunting of the current via the capacitive coupling to electrodes. We note that this is particularly important for the measurements near the band edges in the high- D regime, where the sheet resistance of a gapped BLG rises up rapidly. For a low driving frequency of about 18 Hz, the imaginary component of $R_{2\omega}$ turns out to dominate when setting with the same phase value as that for the R_{ω} detection, which thus confirms a phase difference of about $\pi/2$ between the R_{ω} and $R_{2\omega}$ signals as expected for NRTE. We would also like to remark that Ti/Au are commonly used as contact electrodes for graphene-based devices for low contact resistance. In addition, the BLG regions that were in contact with Ti/Au are only coupled to the bottom gate (V_{bg}) and should be considered as heavily doped regions when the dual-gated device was set at a large D condition. Therefore, the Schottky barriers are not likely to form and give rise to trivial current rectification effects in our device.

We have also performed pulsed dc measurement to confirm the nonreciprocal transport effect. One of the concerns of dc pulse measurement is the onset of excitation. When the current excitation is suddenly lifted from zero to a set point, there is a finite rise time for the responding voltage.

FIG. 12. Frequency dependence of the measured signals. Parts (a) and (b) are the first-harmonic sheet resistance and the second-harmonic resistance, respectively, as a function of ac driving frequency in logarithmic scale.

To minimize the Joule heating effect, the proper width for the current pulse and also a suitable delay time are carefully chosen to ensure a reliable measurement. For consistency, at each V_{bg} set point, the resistance data are derived from statistical average data of 50 pulsed dc measurements, where each measurement comprises a current pulse of 10 ms width

and a voltage measurement with 8 ms source delay. The measured resistance as a function of the bottom-gate through dc pulse mode measurement using 20 and -20 nA under $V_{tg} = -2.5$ V is shown in Fig. 13(a), which demonstrates an observable difference in the resistance near the local maximum of R_{ω} . Thus, the current-direction-dependent resistance is again

FIG. 13. Pulsed dc measurements. (a) Sheet resistance as a function of bottom gate with the top gate fixed at -2.5 V using opposite current directions by a dc pulse measurement. Parts (b) and (c) show the difference of sheet resistances as a function of bottom-gate with top-gate values fixed at -2.5 and 2.5 V, respectively.

verified under pulsed dc measurement. The resistance change $\Delta R = R_{20\text{ nA}} - R_{-20\text{ nA}}$ as a function of the bottom-gate with $V_{\text{tg}} = -2.5$ and 2.5 V is plotted in Figs. 13(b) and 13(c), respectively. A noticeable ΔR happens near $V_{\text{bg}} = 35$ V with $V_{\text{tg}} = -2.5$ V and $V_{\text{bg}} = -39$ V with $V_{\text{tg}} = 2.5$ V. ΔR changes

sign when the type of carrier changes. The observed ΔR first attains a peak and then drops rapidly to reach a valley as gate voltage decreases. Those results are consistent with the behavior in ac measurements shown in the main text.

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