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High-order fractal quantum oscillations in graphene/BN superlattices in the extreme doping limit

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Abstract

Recent studies of van der Waals (vdW) heterostructures and superlattices have shown intriguing quantum phenomena such as correlated insulating states, unconventional superconductivity and quantum anomalous Hall effects. However, most of these phenomena have been explored only in the moderate carrier density regime using field effect gating which is restricted by dielectric breakdown. Here, we report the probe of high-temperature fractal Brown-Zak (BZ) quantum oscillations through magnetotransport in the extreme doping regimes by applying a newly developed electron beam doping technique. This technique gives access to both ultrahigh electron and hole densities beyond the dielectric breakdown limit in graphene/BN superlattices, enabling the observation of non-monotonic carrier-density dependence of fractal BZ states and up to fourth-order fractal BZ features despite strong electron-hole asymmetry. Theoretical tight-binding simulations qualitatively reproduce all observed fractal BZ features and attribute the non-monotonic dependence to the weakening of superlattice effects at high carrier densities. Our results thus demonstrate electron-beam doping is a powerful method for unveiling new physics in previously inaccessible high carrier density regimes in vdW heterostructures and superlattices.

Main Text

Van der Waals (vdW) heterostructures formed from disparate materials provide an exciting platform for unparalleled materials design and device innovation [1–3]. Recent advances in controlling the rotational orientation of vdW layers have led to moiré superlattices and associated interaction driven phenomenon such as Hofstadter’s butterfly, the fractional fractal quantum Hall effect, high-temperature quantum oscillations, correlated insulating states, magic-angle

superconductivity and quantum anomalous Hall effects [4–14]. A key control parameter is the carrier density, most commonly altered via field effect gating. However, conventional field effect gating (by applying a gate bias through the solid dielectric layer) has unavoidable limitations, such as those imposed by dielectric breakdown, which hinders further exploration of ultrahigh carrier density regimes. More effective doping techniques are called for in exploring new physics. For example, high-temperature quantum oscillations have been reported in graphene/h-BN superlattices [8,9]. These so-called Brown-Zak (BZ) oscillations originate from periodic emergence of delocalized Bloch states when the magnetic length is commensurable with the lattice periodicity, and can in principle occur for all rational fractions of magnetic flux $\phi = \phi_0 p/q$, where ϕ_0 is the flux quantum, and p and q are integer numbers [9,15,16]. In order to probe high-order fractal BZ states, high carrier densities and high magnetic fields are preferred. Kumar and collaborators achieved high electron densities by using a photo-doping technique and revealed fractal BZ states up to fourth-order with magnetic fields up to 30 T [9]. However, further observation of these high-order fractal states in the extreme doping limit has not been demonstrated due to the inefficiency of hole-doping for the photo-doping technique [17,18] and low visibility of BZ states originated from electron-hole asymmetry in graphene/h-BN superlattices [8].

Here we explore the extended carrier density parameter space in graphene/h-BN superlattices accessed by a recently developed electron-beam doping technique [19]. We uncover fractal BZ oscillations up to fourth order in the previously inaccessible highly electron and hole doped regimes with a moderate magnetic field of up to 9 T. A wide-range carrier density spectrum of the fractal BZ features is established, revealing unexpected non-monotonic carrier-density dependence and strong electron-hole asymmetry of fractal BZ states. Our tight-binding calculations qualitatively reproduce all observed fractal BZ features and attribute the non-

monotonic behavior to the weakening of superlattice effects at high carrier densities. Our results demonstrate the effectiveness of the electron-beam doping technique and provide new opportunities in exploring vdW heterostructures and superlattices in previously inaccessible high carrier density regimes.

The experimental scheme for electron-beam induced doping in the graphene/h-BN superlattice is shown in Fig. 1(a). We use standard dry transfer and e-beam lithography techniques to fabricate h-BN encapsulated graphene/h-BN heterostructures with edge contacts [20] and a standard Hall bar geometry. During the dry transfer process, we intentionally align the graphene's crystallographic axes with the bottom h-BN layer, but misalign it with the top h-BN layer. The graphene moiré superlattice is subsequently characterized by transport measurement. As illustrated in Fig. 1(a), charge doping is induced by electron beam irradiation in a standard scanning electron microscopy (SEM) for a few seconds while holding the back-gate $V_G = V_{\text{SET}} (\neq 0)$. By changing the sign and magnitude of V_{SET} during exposure, specific electron or hole doping can be controllably induced. The doped state persists even after the electron-beam and back-gate are removed. The process is reversible and repeatable so we can successively induce highly electron or hole doped states in a single device and systematically investigate the transport properties [19].

High carrier densities beyond $\pm 10^{13} \text{ cm}^{-2}$ can be reliably induced with this technique in graphene/h-BN superlattice devices. This exceeds the conventional electrostatic gating limit of $\sim \pm 5 \times 10^{12} \text{ cm}^{-2}$. Fig. 1(b) shows the e-beam doping effects of a graphene/h-BN superlattice (Device A) after 30 keV e-beam irradiation at $V_{\text{SET}} = 40 \text{ V}, 30 \text{ V}, -30\text{V}$ and -40V (from red to blue curves, skipping the black curve), respectively. The black curve is measured from the pristine device before the e-beam exposure. From the transfer curves, the primary charge neutrality point (CNP) positions (marked by the dashed lines) are shifted simultaneously with the secondary CNPs to

positive and negative sides after irradiation, indicating effective hole and electron doping, respectively. The large CNP shift ΔV_{CNP} over 160 V clearly demonstrates strong tunability of carrier density across a range more than 10^{13} cm^{-2} by the electron-beam doping technique, which typically cannot be achieved with a conventional SiO_2 back-gated device. Remarkably, the graphene device still maintains high mobility after doping even though the CNP peak slightly broadens as the doping increases [19]. The doped device is subsequently transferred into a cryostat for temperature and magnetic field dependent transport studies. Fig. 1(c) shows the temperature dependent transfer curves of another Gr/h-BN superlattice (Device B) after hole doping induced by 30 keV e-beam irradiation at $V_{\text{SET}} = -30 \text{ V}$. The primary CNP shifts to $\sim 60 \text{ V}$ and remains unchanged while the secondary CNP in the hole side becomes more pronounced as temperature decreases, in agreement with an earlier report [4]. It demonstrates that the 30 keV e-beam induced doping is robust against temperature variations and preserves the moiré superlattice. From the primary and secondary CNP positions, we derive the full-filling carrier density for the superlattice $n_0 = 2.3 \times 10^{12} \text{ cm}^{-2}$ and $3.8 \times 10^{12} \text{ cm}^{-2}$ for Devices A and B, respectively (Fig. S1). From the relation $n_0 = 4/S = 8/(\sqrt{3}a^2)$ [8,9], the superlattice a is estimated to be 14 nm (Device A) and 10.9 nm (Device B). Here, S is the area of the superlattice unit cell.

We present magneto-transport data for a highly electron-doped graphene/BN superlattice induced by the electron-beam doping technique. Fig. 2(a) shows the longitudinal resistivity R_{xx} for the graphene superlattice (Device B) at a high electron density of $n = 4.6n_0 = 1.7 \times 10^{13} \text{ cm}^{-2}$ as a function of magnetic field B at various temperatures. A strong periodic oscillation of the magnetoresistance is clearly seen even at 300 K with a magnetic field up to 9 T and it becomes further enhanced as temperature decreases. These high temperature magneto-oscillations are a manifestation of BZ oscillations, which have been described previously but characterized only in

a much lower carrier density regime [8,9]. At temperatures below 20 K, Shubnikov-de Haas (SdH) oscillations arise and overlay with the BZ oscillations evidenced by the appearance of additional oscillation peaks as shown in Fig. 2(a). One essential feature of BZ oscillations that differs from SdH oscillations is that the BZ oscillation periodicity is independent of the carrier density. This can be seen from Fig. 2(b), which shows R_{xx} as a function of B for the same graphene superlattice device at various electron densities from $n = 0.8 n_0$ to $4.6 n_0$. We see clearly that the oscillation periodicity remains unchanged across a wide carrier density range up to $1.7 \times 10^{13} \text{ cm}^{-2}$. The oscillation amplitude increases as n increases beyond the secondary CNP ($n = n_0$) [8,9] and becomes most pronounced near the third-generation CNP ($n = 2n_0$). Additionally, we observe second-order fractal oscillations as the electron density nears the third-generation CNP but the fractal periodicity becomes less visible at higher density at $n = 4.6n_0$. This is due to smearing between different minibands, which we discuss in more detail below.

To clearly see the fractal oscillations, we carefully tune the carrier density of the graphene/h-BN superlattice across a broad carrier density range, from high electron-doping to high hole-doping using the electron-beam doping technique. We measure the magneto-transport of Device A at various carrier densities between $3.7n_0$ to $-4.0n_0$ (negative for hole-doping), and plot a map of longitudinal conductivity $\Delta\sigma_{xx}$ as a function of n and $1/B$ (in units of ϕ_0/ϕ) with a maximum B field up to 9 T as shown in Fig. 2(c). Here $\Delta\sigma_{xx}$ is obtained by subtracting a smooth background from the longitudinal conductivity σ_{xx} . The magnetic flux through the superlattice unit cell is $\phi = BS$. The characteristic carrier density for the superlattice is $n_0 = 2.3 \times 10^{12} \text{ cm}^{-2}$ as mentioned earlier. The data are taken from three sets of experiments after electron-beam doping on the same device, which results in varied backgrounds but the oscillation features match up with each other. We observe a set of dark vertical streaks (marked by the red arrows) indicating local

maxima of conductivity located at $\phi/\phi_0 = 1/q$ (q is an integer) in both highly electron-doped and hole-doped regimes, which corresponds to a magnetic flux quantum ϕ_0 per q superlattice unit cells. The oscillations are periodic in $1/\phi$ (i.e. $1/B$) with an oscillation frequency $B_0 = \phi_0/S = n_0\phi_0/4 \sim 24$ T, which is consistent with the value calculated from the Fourier transform of the magnetoconductivity data. Additional local maxima at $\phi/\phi_0 = p/q$, where p magnetic flux quantum ϕ_0 pierces through q superlattice unit cells mimicking zero effective magnetic field for the transport [8], are also observed as thinner vertical streaks (marked by the blue, green and pink arrows) between the thick streaks at higher magnetic field regime, independent of the carrier density. These extra fractal features located at $\phi/\phi_0 = p/q$ are caused by high-order magnetic Bloch states with $p = 2, 3$ and 4 .

To better resolve these high-order fractal oscillations, we plot the original longitudinal magnetoconductivity σ_{xx} (without background subtraction) as a function of ϕ_0/ϕ for Device A at various electron and hole densities in Fig. 3(a) and (b). The data are taken at 100 K and a magnetic field up to 9 T. The electron density, n , increases from $1.6 n_0$ to $2.6 n_0$ with a step of $\sim 0.14 n_0$ (from the bottom to top in Fig. 3(a)). We see that extra conductivity maxima gradually emerge as the electron density increases and become prominent near the third-generation CNP with $n \sim -2n_0$. We have identified these states (marked by vertical dash lines and arrows) with exact p and q values up to the fourth order ($p = 4$). At 100K, as the electron density further increases up to $3.7 n_0$, these higher-order fractions are less visible or are mixed with additional peaks and thus difficult to resolve (see Supplementary Fig.S2). This is consistent with previous observation in Device B (Fig. 2(b)). Note that higher-order magnetic Bloch states with $p > 2$ are not seen in Device B possibly because its moiré lattice is short (corresponding to larger B_0) and the magnetic field is not

sufficiently high in our measurements (maximum field up to 9T). Kumar's report that revealed the third- and fourth-order magnetic Bloch states used a high magnetic field up to 30 T [9].

Next, we focus on the high-order fractal BZ states in the highly hole-doped regime. At low hole density below the third-generation CNP with $n \sim -2n_0$, we observe only the primary maxima located with $p = 1$ (see Supplementary Fig. S2). As the hole density further increases, extra local maximas with $p = 2, 3$ and 4 gradually appear, corresponding to second, third and fourth-order fractal BZ oscillations. Here, the third and fourth-order BZ oscillations with $p = 3$ and 4 are seen for the first time (yellow and red curves), which is remarkable considering the much lower visibility of BZ oscillations for hole doping due to electron-hole asymmetry of electron-phonon scattering in graphene/BN superlattices [8]. Notably, these high-order fractal features are only visible at sufficiently high hole densities around -10^{13} cm^{-2} induced by the e-beam doping technique, which is not accessible by conventional back gate tuning nor the photo-doping technique.

In addition, these high-order fractal states for electrons and holes become smeared as the temperature increases and are more pronounced when temperature decreases, as shown in Fig. 3(b) and (d). At $T = 200 \text{ K}$, only the $p = 1$ oscillations are visible while the higher order states with $p = 2, 3$ and 4 are more prominent at $T = 50 \text{ K}$. These high-order fractal BZ features also show non-monotonic carrier density dependence. In Fig. 3(e), we plot the conductivity peak amplitude $\Delta\sigma_{xx}$ versus carrier density for certain fractal BZ states with $p = 2$ and 3 by subtracting a smooth background from σ_{xx} in Fig. 3(a) and (c). These peak amplitudes firstly increase as the carrier density increases and then gradually decreases or saturates above certain densities (at around $-3n_0$ for holes and around $2n_0$ for electrons). The peaks corresponding to fractal BZ features for holes

appear at higher densities and have less maximum amplitude, also indicating the lower visibility of BZ states for hole-doping.

To provide rationale to the experimental observations of non-monotonic carrier density dependence, i.e. the relatively stronger fractal BZ oscillations for intermediate charge carrier densities and weakened features for high densities, we simulate a similarly-aligned system using a tight-binding (TB) implementation of the Kubo formula for the longitudinal conductivity through wave-packet evolution [21–23]. This method allows us to straightforwardly probe qualitatively different transport behavior in the ballistic regime by following the mean quadratic displacement of the wavepackets [24]. Note that the ballistic regime is often mostly avoided because the extracted conductivity will depend on the specific timestep of the evolution method making it difficult to make quantitative predictions. However, in this work we focus on this unusual regime allowing us to avoid the effects of disorder yet leveraging the fact that even in the ballistic regime, the conductivity captures the BZ oscillations qualitatively. We are not aware of other studies extracting the BZ oscillations in this way.

Our simulation reproduces nearly all primary and fractal BZ features that occur in experiment. Fig. 4(a) and Fig. 4(b) show the calculated conductivity as a function of ϕ_0/ϕ and n/n_0 for hole-doped and electron-doped graphene/h-BN superlattice with a lattice periodicity of 13.5 nm. As indicated by the vertical lines, high-order fractal states with $p = 2, 3$ and 4 are clearly resolved for high hole and electron doping. In agreement with our experimental observations, the fractal BZ oscillations are less likely to be resolved at lowest charge carrier densities, due to the weaker density of states and reduced group velocity. This observation is slightly obfuscated by the robust Landau levels in our zero-temperature simulations. As the carrier density increases, we further observe an increase in the strength of the fractal BZ features, whereas it gradually becomes

smearred at the highest charge carrier densities (also supported by Supplementary Fig. S3), in consistent with our experiment observation. This is because the larger the density of states (DOS), the fewer the superlattice minibands carrying the BZ oscillations with specific values of p and q are separated in energy. The BZ oscillations occur at frequencies where the cyclotron orbit length scale becomes commensurable with the periodicity of the system. The existence of the former length scale gradually disappears at larger charge carrier density (cfr. the gradually weaker Landau level quantization signatures), thus requiring larger magnetic fields to resolve them.

To verify this, we calculate the DOS with respect to the energy E (in proportional to \sqrt{n} , n is the carrier density) and the magnetic field B for pristine graphene as well as the graphene/h-BN superlattice system. We subtract the former from the latter to remove the Landau level features coming from the primary Dirac cone in order to keep only the superlattice features as shown in Fig. 4(c). This subtraction is not perfect due to the superlattice induced broadening of the Landau levels, but it is sufficient to capture how the superlattice effects evolve with the energies (or carrier density) and magnetic fields. Fig.4(d) shows a series of line cuts at 0 T, 5 T, and 10 T from Fig. 4(c). As most of visible fractal BZ states appear in the magnetic field range of 4 - 8 T (Fig. 3), the line cut at 5 T represents the energy (or carrier density) dependence of superlattice effects in our experiments. It illustrates that the superlattice effects are weak close to the primary Dirac point energy (± 0.16 eV), become maximal around the secondary Dirac point energy and weaken again at higher energy (or carrier density) due to the increasing number of states with energy. This provide rationale for the weakened fractal BZ oscillations at high energies as the presence of fractal BZ is sensitive to the superlattice effects. The simulations also suggest that for higher magnetic field range, this weakening might only happen at even higher carrier density regime, which might be the reason it did not probed by previous experiments [9]. In this study with a moderate magnetic

field and high enough carrier density induced by e-beam doping technique, we demonstrate that such a non-monotonic behavior can be observed.

In summary, we have achieved high carrier densities in graphene/h-BN moiré superlattices by an electron-beam doping technique and have thereby revealed high temperature fractal BZ oscillations up to fourth-order both on electron and hole sides. We show that BZ oscillations exist in high electron density up to $n/n_0 = 4.6$ through magneto-transport while the fractal BZ oscillations are most visible near the third-generation CNP ($n \sim 2n_0$) and gradually become smeared as electron densities further increases. High-order fractal signatures with $p > 2$ are also clearly observed for the first time in highly hole-doped regime enabled by the electron-beam doping technique despite the masking effect of electron-hole asymmetry. Our qualitative magnetoconductance simulations using a tight-binding model shows good agreements with our experimental observations on the non-monotonic carrier density dependence of the fractal BZ oscillations, which is related to the weakening of superlattice features at high carrier densities. Our work thus expands the observation and understanding of fractal BZ oscillations in graphene/h-BN superlattices, and also proves the electron-beam doping technique is a reliable way to extend the study of quantum behavior to a broader carrier density regime in vdW heterostructures without extra fabrication processes.

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Figures

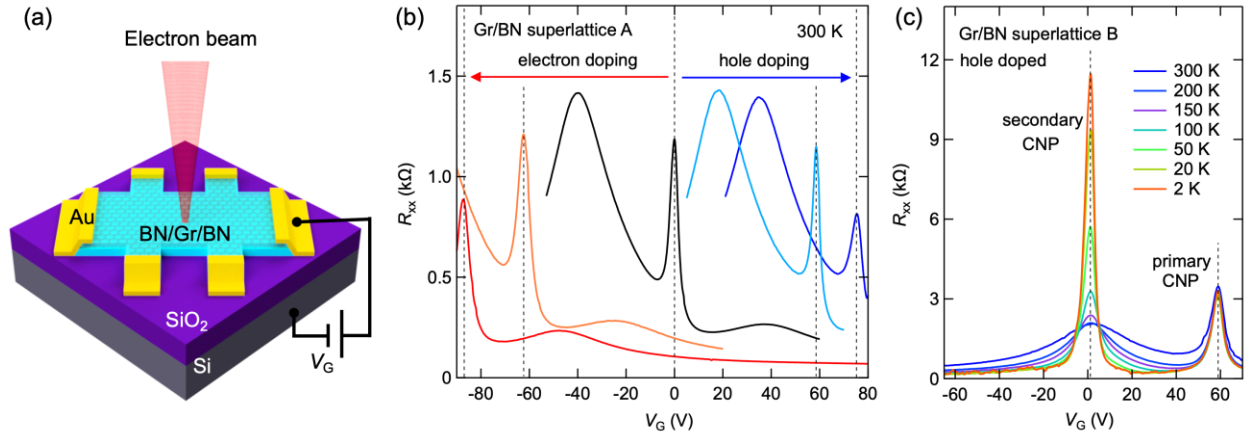


FIG. 1. (a) Experimental scheme for the electron-beam induced doping in graphene/h-BN superlattice device. Charge doping is induced in graphene by electron beam irradiation in a standard SEM for a few seconds while holding the back-gate $V_G = V_{SET} (\neq 0)$. (b) Transfer curves of a graphene/h-BN superlattice (Device A) after e-beam doping by controlling the e-beam energy and the V_{SET} value. For the data indicated, e-beam energy is 30 keV and $V_{SET} = 40$ V, 30 V, -30V and -40V (from red to blue curves, skipping the black curve), respectively. The black curve is measured for the pristine device before any e-beam exposure. Highly electron or hole doped states can be obtained as seen from the large shift of the charge neutrality point (CNP, identified with dashed vertical lines) from zero. (c) Transfer curves of another graphene/h-BN superlattice (Device B) after highly hole-doped measured from 300 K to 2 K. The primary CNP peak remains at the same position while the secondary CNP peak becomes more pronounced at low temperatures.

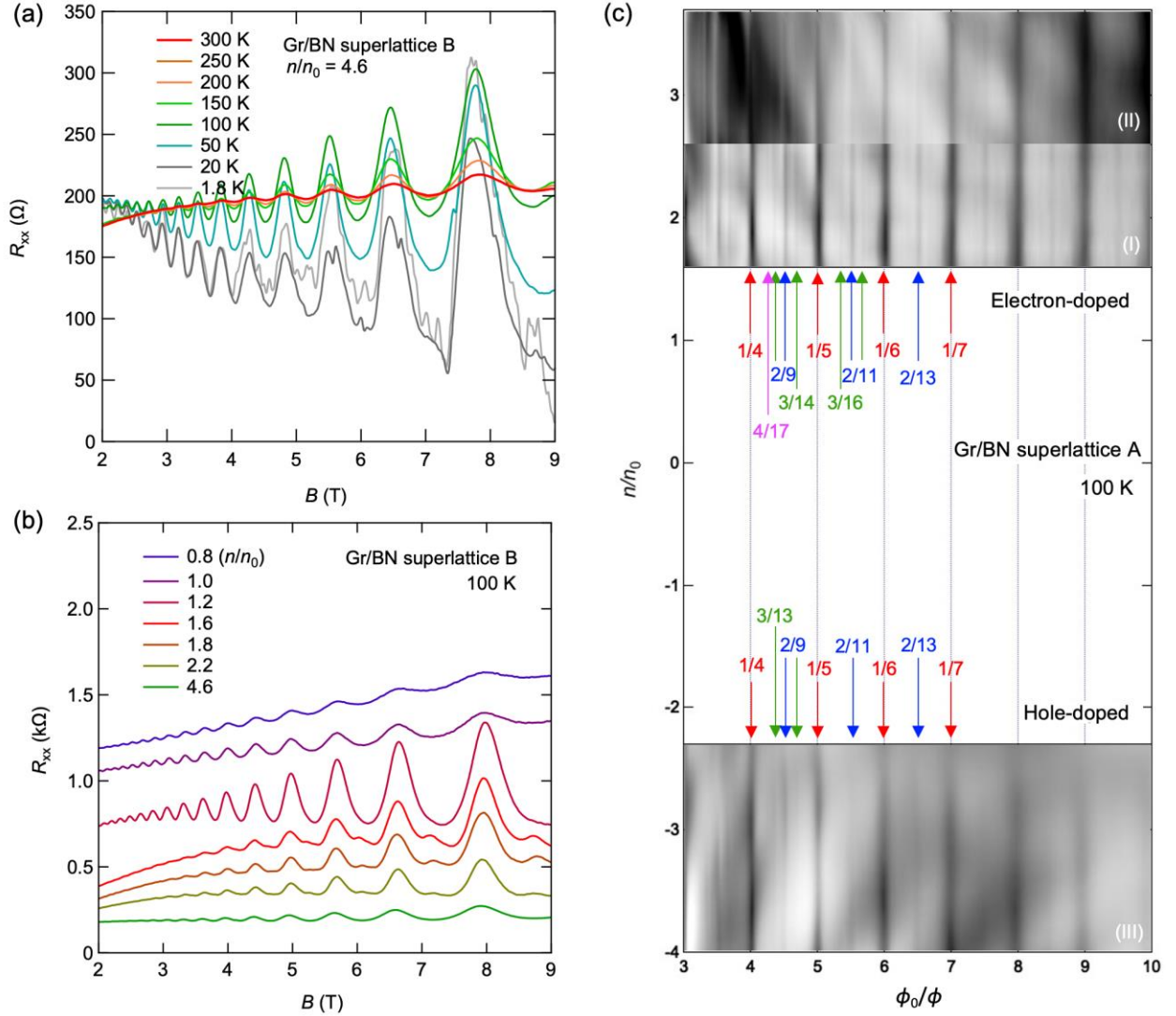


FIG. 2. Brown-Zak (BZ) oscillations in electron-doped and hole-doped graphene/h-BN superlattice device. (a) Longitudinal resistivity R_{xx} for the graphene superlattice (Device B) as a function of magnetic field B at various temperatures from 300 K to 1.8 K. The device is highly electron-doped by e-beam doping technique with a carrier density at $n = 4.6n_0$ ($\sim 1.7 \times 10^{13} \text{ cm}^{-2}$). (b) R_{xx} as a function of B for the same device at various electron density measured at 100 K. (c) Longitudinal conductivity $\Delta\sigma_{xx}$ for Device A as a function of ϕ_0/ϕ and n/n_0 for high electron and hole doping in B up to 9 T measured at 100 K. Here, the characteristic carrier density for the

superlattice $n_0 = 2.3 \times 10^{12} \text{ cm}^{-2}$. $\Delta\sigma_{xx}$ is obtained by subtracting a smooth background from σ_{xx} .

Logarithmic gray scale: white: -1.0 mS; black: 1.5 mS. The red, blue, green and pink arrows indicate the primary and fractal states with $p = 1, 2, 3$ and 4 , respectively.

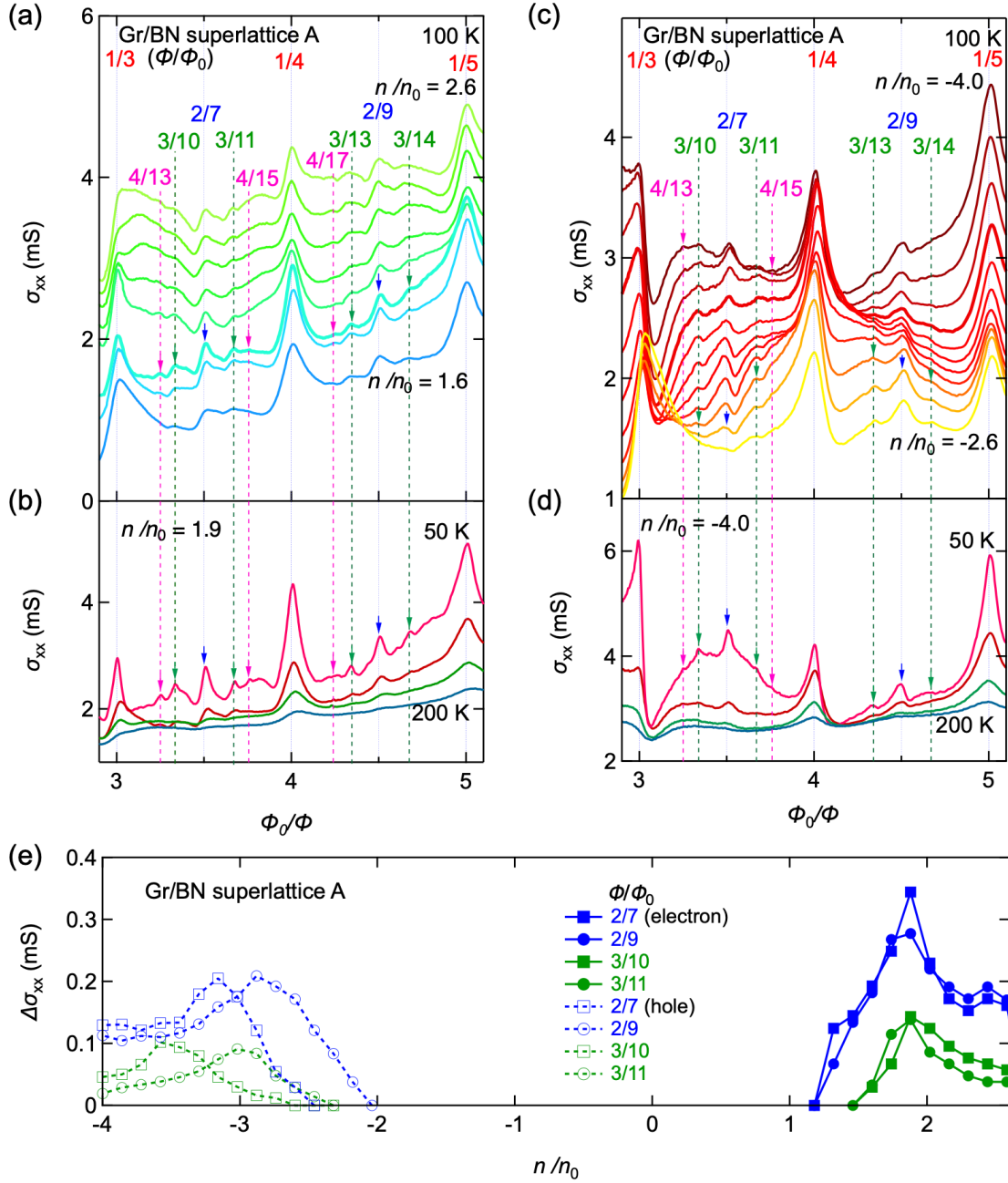


FIG. 3. High-order fractal BZ oscillations in highly electron-doped and hole-doped graphene/h-BN superlattice (Device A). (a) Longitudinal conductivity σ_{xx} as a function of ϕ_0/ϕ measured at various electron density n from $1.6n_0$ to $2.6n_0$ with a step of $\sim 0.14n_0$. Here, the characteristic carrier density for the superlattice $n_0 = 2.3 \times 10^{12} \text{ cm}^{-2}$. (b) σ_{xx} as a function of ϕ_0/ϕ for electron density $n = 1.9n_0$ at various temperatures from 200 K to 50 K. (c) σ_{xx} as a function of ϕ_0/ϕ measured at various hole density n from $-2.6n_0$ to $-4.0n_0$ with a step of $\sim 0.14n_0$. (d) σ_{xx} as a function of ϕ_0/ϕ for hole density $n = -4.0 n_0$ at various temperatures from 200 K to 50 K. The vertical dashed lines and arrows indicate the magnetic field positions where the fractal BZ states with $p = 2, 3$ and 4 . (e) Conductivity peak amplitude $\Delta\sigma_{xx}$ versus carrier density for fractal BZ states with $p = 2$ and 3 . $\Delta\sigma_{xx}$ is obtained by subtracting a smooth background from σ_{xx} in (a) and (c).

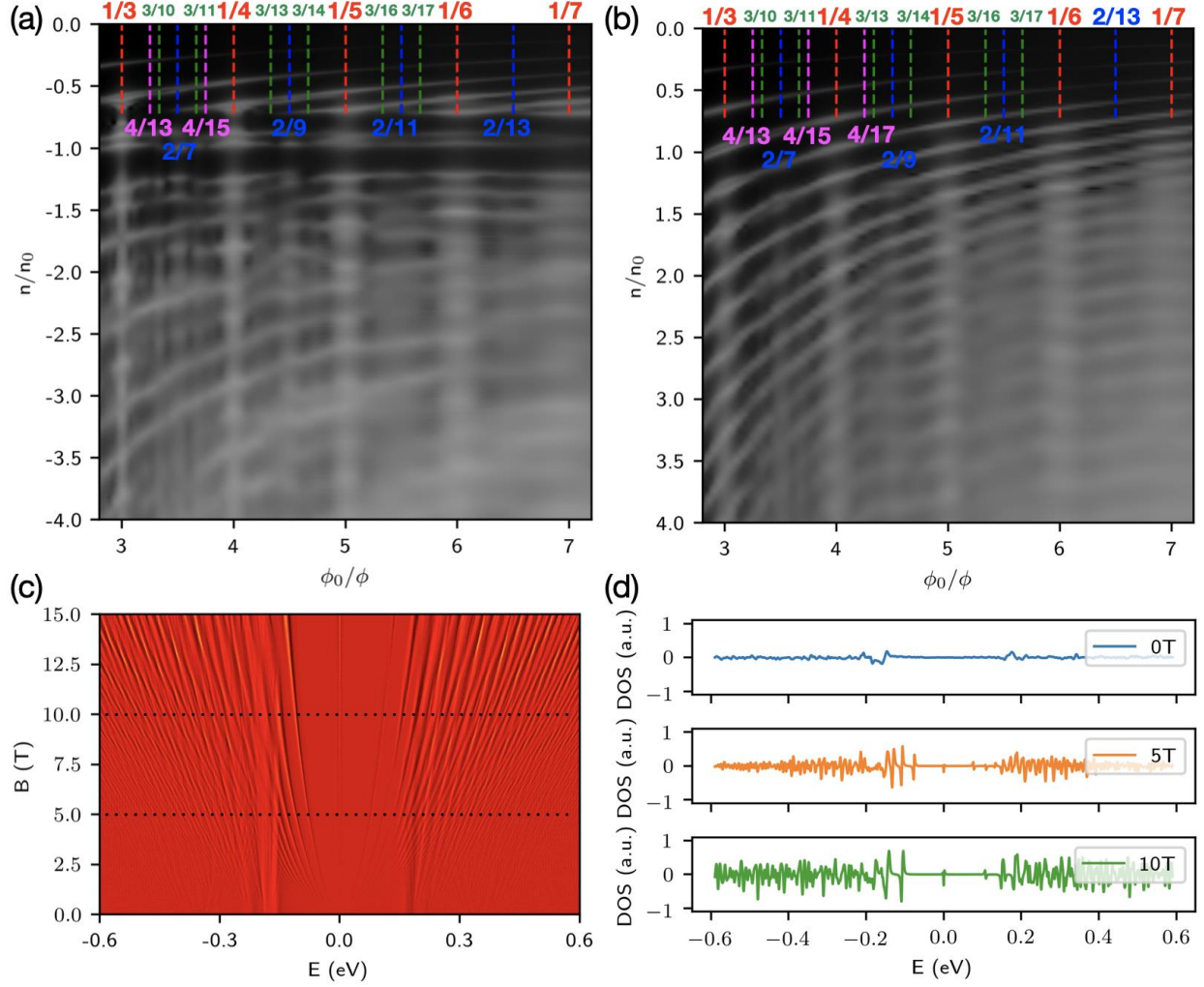


FIG. 4. Qualitative simulation of magnetoconductivity by a tight-binding method. The simulated conductivity of (a) hole-doped and (b) electron-doped graphene/h-BN superlattice plotted as a function of ϕ_0/ϕ and charge carrier density n/n_0 (negative for hole doping) clearly resolves the fractal BZ oscillations with $p = 2, 3$ and 4 , as indicated by the dashed vertical lines with the labels of corresponding q values. Landau level features are still seen in the background as the simulations occur at zero temperature. (c) The calculated density of states (DOS) with respect to the energy and the magnetic field for the graphene/h-BN superlattice after subtracting the DOS Landau level map of the pristine graphene system. (d) The DOS versus the energy at different magnetic fields,

obtained from the line cuts in (c). The secondary Dirac points with $n/n_0 = 1$ appear at the energy around ± 0.16 eV.