

# Mid-infrared photoresponse due to cyclotron resonance absorption in graphene/h-BN van der Waals heterostructures

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## Abstract

Charge carriers in graphene, massless Dirac fermions, form a unique sequence of the Landau levels in high magnetic fields. Thus, the cyclotron resonance (CR) in graphene is distinctly different from that in conventional two-dimensional electron systems based on semiconductors. In this work, we study mid-infrared/THz photoresponse due to CR in graphene/h-BN van der Waals heterostructures. In particular, we focus on CR in a dual-gated trilayer graphene/h-BN [1] and a monolayer graphene under double-moiré potentials [2].

## 1. CR in dual-gated trilayer graphene

ABA-stacked trilayer graphene (TLG) possesses both monolayer graphene (MLG)-like and bilayer graphene (BLG)-like bands. In a high perpendicular magnetic field  $B$ , the energy interval of Landau levels (LLs) in TLG is in a wavelength range from mid-infrared to near-infrared, which is technologically important for optoelectronics application. By applying a perpendicular electric displacement field  $D$ , a band gap is induced, and  $D$  hybridizes MLG-like and BLG-like bands in TLG, which appears as anti-crossings of LLs. Thus, ABA-stacked TLG has an electrical tunability of LLs in a large extent, which enables electrical tuning of cyclotron resonance (CR) magnetic field in TLG.

To demonstrate the electrical tunability of CR signals in TLG, we fabricate a dual-gated TLG device in which we can control  $D$  by applying bias voltage to the top and bottom gate electrodes. Figs. 1a-c show the schematic illustrations and optical image of TLG device in the present work. After exfoliation of h-BN flakes onto a 290-nm-thick SiO<sub>2</sub>/p-doped Si substrate, TLG was transferred onto h-BN using a method based on polypropylene carbonate (PPC) [3,4]. Before capping it with another h-BN flake, we put electrodes on TLG by metal deposition of Pd 15 nm. Then the top h-BN layer was transferred onto the stack and a graphite (6~8 layers) was put on the top of it as a transparent top gate. Figs. 1d-g show color maps of the photo-induced voltage  $V_{\text{photo}}$  at the charge neutrality point  $\nu = 0$  irradiated by laser light of different wavelengths of  $\lambda = 9.250, 9.552, 10.247$ , and  $10.611 \mu\text{m}$ , respectively, as a function of  $D$  and

$B$ . We set the Fermi energy at the charge neutrality point by sweeping  $V_{\text{BG}}$  and  $V_{\text{SD}}$  along the line of  $R_{xx}$  peak in  $V_{\text{SD}}-V_{\text{BG}}$  plot. In these maps, the peak position of  $V_{\text{photo}}$  depends on  $D$ , drawing a curve which is symmetric to  $D = 0$ . These curves of  $V_{\text{photo}}$  evidently shows that CR signals in TLG is distinctly modified by  $D$ .

To give an account for  $D$  dependence of CR signals, we calculated LL diagram by tight-binding approximation with different  $\Delta_1$  ranging from 0 to 40 meV, where  $\Delta_1$  is proportional to  $D$ . Using the calculated LLs, we obtained a position of  $B$  for each  $\Delta_1$  where the LL interval is equal to the energy of irradiated laser, where CR transition between LLs is allowed. The numerical plot reproduces the experimental results quite well. These agreement of calculation and experiment demonstrate that LLs in TLG is successfully tuned in our dual-gated TLG device. At the same time, CR signals directly reflects the energy interval of LLs, thus it can be utilized as a powerful probe to study LLs in TLG quantum Hall systems.

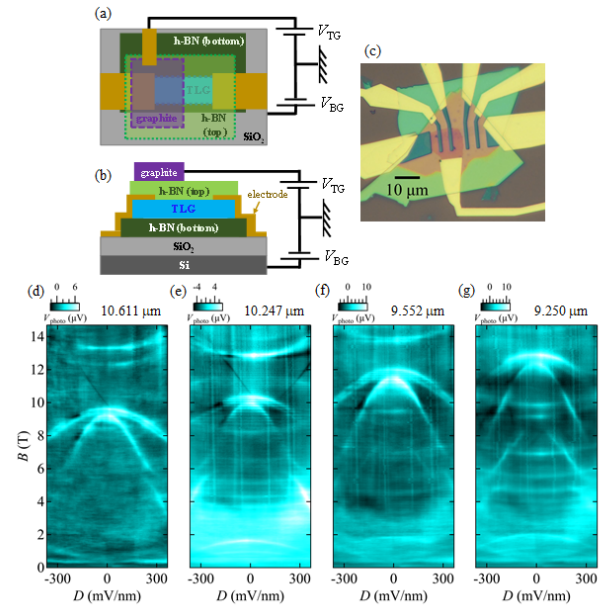


Fig. 1 (a) Top- and (b) side-view schematics of the device structure. (c) Optical image of the device. (d-g) Color map of  $V_{\text{photo}}$  as a function of  $D$  and  $B$  at  $\nu = 0$  (experiment).

## 2. CR in graphene under double-moiré potentials

When the crystal axis of graphene is aligned to that of adjacent hexagonal boron nitride (h-BN), graphene's band structure is modified by the moiré potentials induced at the interface, exhibiting unique physical phenomena such as Hofstadter butterfly. Recently, it was found that two coexisting moiré potentials in a h-BN/graphene/h-BN heterostructure interfere with each other to generate second-order moiré potentials. Here, we study cyclotron resonance (CR) in graphene in double-moiré potentials, which provide us a direct insight on Landau levels (LLs) in graphene.

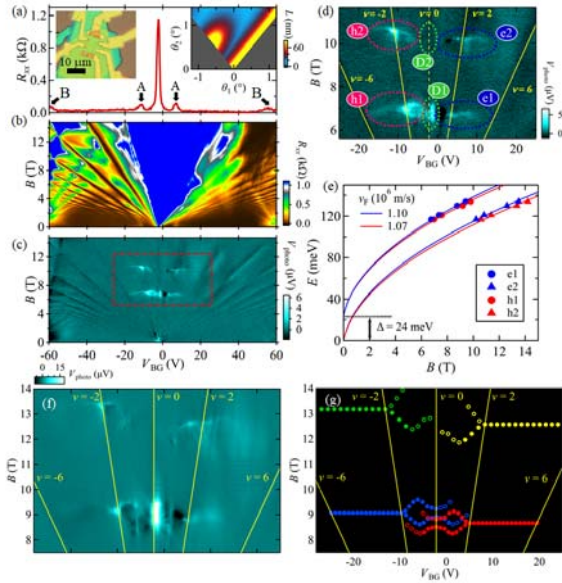


Fig. 2 (a)  $R_{xx}$  vs  $V_{BG}$  at  $T = 3$  K and  $B = 0$ . Left inset: photograph of the device. Right inset: second-order moiré wavelength as a function of  $\theta_1$  and  $\theta_2$ . (b)  $R_{xx}$  vs  $V_{BG}$  and  $B$  at  $T = 3$  K. (c)  $V_{photo}$  intensity vs  $V_{BG}$  and  $B$  with a light wavelength of  $\lambda = 10.611 \mu\text{m}$ . (d) Expanded view of the region in (c) indicated by the dashed-red outline. (e) Fitting to the experiment. (f) Experimental  $V_{photo}$  map as a function of  $V_{BG}$  and  $B$  with a light wavelength  $\lambda = 9.536 \mu\text{m}$ . Yellow lines indicate the locations of  $\nu = 0, \pm 2, \pm 6$ . (g) Calculated  $B_{CR}$  corresponding to the experimental CR map shown in (f).

We fabricated a h-BN/graphene/h-BN heterostructure by layer-by-layer assembly using a method based on polypropylene carbonate (PPC).  $R_{xx}$ - $V_{BG}$  plot and Landau fan at  $T = 1.6$  K show satellite peaks very close to the Dirac point, which is an indicative of a moiré potential with a large pe-

riod [Figs. 2a,b]. From the position of these  $R_{xx}$  peaks, we estimated the lateral orientation angle  $\theta$  between graphene and two h-BN sheets as  $\theta \sim 0.73^\circ$  and  $\sim 1.01^\circ$ . Next, we measured photo-induced voltage  $V_{photo}$  under the irradiation of mid-infrared light at  $T = 3$  K. By sweeping carrier density, we observed unique patterns of CR signals, which are distinct from those in conventional graphene without moiré potentials [Fig. 2c]. To account for the distinct signal patterns, we consider electron-hole asymmetry in the Fermi velocity  $v_F$  and a large bandgap  $\Delta$  induced at the Dirac point [Figs. 2d,e]. Based on this model, we fitted the CR magnetic field  $B_{CR}$  of these signals and derived  $v_F = 1.10 \times 10^6$  m/s for electrons,  $v_F = 1.07 \times 10^6$  m/s for holes, and  $\Delta = 24$  meV [Fig. 2f]. We could precisely determine  $\Delta$  and  $v_F$  because  $B_{CR}$  is highly sensitive to  $v_F$  and that we could observe inter-band and intra-band LL transitions by sweeping carrier density. Besides, CR signals have a unique ring-like shape, which we attributed to the enhancement of spin splitting owing to many-body interaction [Fig. 2g]. Our work will aid in understanding the band structure of double-moiré graphene and also demonstrate that CR is a powerful tool to reveal the LL structures.

## Acknowledgements

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