Evidence of abnormal hot carrier thermalization at van Hove singularities of twisted bilayer graphene

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In the past decades, hot carriers in graphene have been exhaustively studied in many ways. From the broadband ultrafast photoluminescence (PL)[1, 2] , to the ultrafast and efficient hot carrier harvesting among graphene and various semiconducting materials. These intriguing carrier dynamic behaviours and the diverse ultrafast phenomena have addressed graphene an excellent platform for advanced optoelectronics and photonics.

As a unique engineering method for two-dimensional (2D) materials, interlayer twisting brings new opportunities in both fundamental physics and applications into graphene. The emergence of van Hove singularities (VHSs) endows graphene enhanced density of states, intensified light absorption and strongly bound excitons[3, 4]. Strong correlation of electrons in magic-angle TBG can further lead to more intriguing phenomena like correlated insulating states, superconductivity et al[5]. With all these fascinating optical and electronic properties, TBG-like 2D material systems have infused new vitality to the research frontiers of materials science and condensed matter physics. However, research until now on TBG is limited to the ground state, and the photoexcited state dynamical properties of TBG, representing one of the irreplaceable aspects of its physical picture and cornerstone for its application design, are still little studied.

Here, we propose the time-resolved PL autocorrelation method to unveil the ultrafast hot carrier dynamics in TBG. As ultrafast PL intensity is linearly correlated with the transient hot carrier density, time-resolved PL autocorrelation should be a faithful reflection to the carrier dynamics[6]. By extracting the peak components from PL spectra which represent the pure VHS contribution, we can recover the VHS carrier dynamics from the carrier mixture. Ultrafast hot carriers on the linear bands of TBG behave similarly to MLG, while the carriers captured by VHS possess a decelerated dynamic, which is 4-time longer than that of MLG. Further time-dependent density functional theory (TDDFT) suggests that the prolonged lifetime is attributed to the abnormal hot carrier thermalization brought by bottleneck effect at mini-gaps and interlayer charge redistribution process in real space.

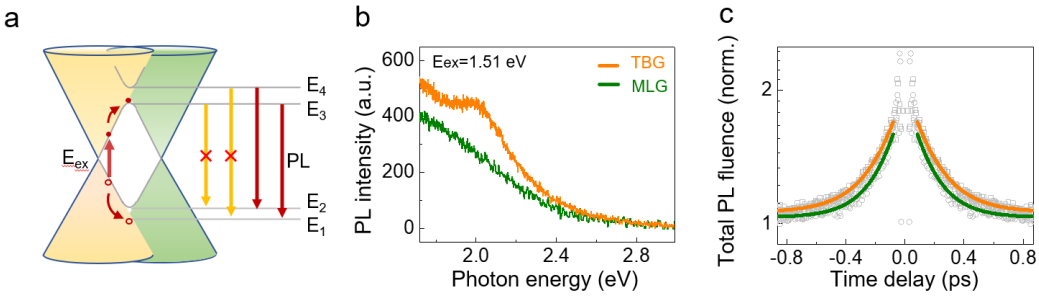

Figure 1 Ultrafast PL and PL autocorrelation of TBG. a. Bandgap opening at VHS of TBG and its optical transition selection rules. b. the ultrafast PL spectra of TBG and MLG. c. ultrafast PL autocorrelation of TBG and MLG, where TBG shows a longer lifetime.

Our exploration should have important implications for the design of TBG-based ultrafast photonics and optoelectronics devices, such as light-harvesting, light-sensing or light-emitting devices. Meanwhile, our technique should be a unique spectroscopy method to investigate the fundamental element interaction in TBG at small twist angles, when the system lies in a strong-correlation regime at a cryogenic environment. We expect that the abnormal dynamics at VHS will bring more intriguing physics to TBG and its family.

References

[1] C. H. Lui, K. F. Mak, J. Shan and T. F. Heinz *Phys. Rev. Lett.* **105**, 127404 (2010)
[2] W.-T. Liu, S. W. Wu, P. J. Schuck, M. Salmeron, Y. R. Shen and F. Wang *Phys. Rev. B* **82**, 081408 (2010)
[3] R. W. Havener, Y. Liang, L. Brown, L. Yang and J. Park *Nano Lett.* **14**, 3353 (2014)
[4] H. Patel, R. W. Havener, L. Brown, Y. Liang, L. Yang, J. Park and M. W. Graham *Nano Lett.* **15**, 5932 (2015)
[5] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras and P. Jarillo-Herrero *Nature* **556**, 43 (2018)
[6] T. C. O'Haver and J. D. Winefordner *Anal. Chem.* **38**, 1258 (1966)