Exotic Ultrafast Band Renormalization Induced by Excitonic Correlations

Yi Lin[1], Yang-hao Chan[1], Woojoo Lee[2], Li-Syuan Lu[4,5], Zhenglu Li[1], Wen-Hao Chang[4,5], Chih-Kang Shih[2], Robert A. Kaindl[1], Steven G. Louie[1,3], Alessandra Lanzara[1,3]

[1] *Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

[2] *Department of Physics, The University of Texas at Austin, Austin, Texas 78712, USA*

[3] *Department of Physics, University of California, Berkeley, California 94720, USA*

[4] *Department of Electrophysics, National Yang Ming Chiao Tung University, Hsinchu 30010, Taiwan*

[5] *Research Center for Applied Sciences, Academia Sinica, Nankang, Taipei 11529, Taiwan*

Author Email: yilin@lbl.gov

Optical excitation has been demonstrated as a powerful approach for the ultrafast control of electronic structure in 2D materials by inducing many-body interactions (such as screening effects [1]) and driving coherence (such as optical Stark [2] and Bloch-Siegert [3] effects). Among these, photoexcited electrons and holes (e-h) that forming excitonic bonds have been theoretically predicted to drive an exotic type of quasi-particle band renormalization [4], featuring an increased bandgap and effective mass. However, observing such exciton-driven band renormalization has been experimentally elusive and challenging, largely due to the masking by the screening effects from free carries and the limitations of optical probes for accessing momentum-resolved single-particle bands.

In this work [5], we apply XUV-trARPES, with supreme data fidelity and low pump fluence, to study the ultrafast band renormalization in monolayer (ML) MoS_2 on a conducting polygraphite (HOPG) substrate. Following the photoexcitation, we observe a surprising ultrafast increase of the quasi-particle bandgap up to 40 meV and an ultrafast enhancement of band effective mass at the K valley of the ML MoS_2. Combined with many-body theoretical calculations, we reveal explicitly how the effects of excitonic e-h correlations and conducting substrates should be responsible for our observations. Our findings provide critical insights for understanding the emerging non-monotonic bandgap dynamics and transient binding energy of excitonic quasi-particles in 2D materials, leading to new pathways for engineering band structure via excitons.

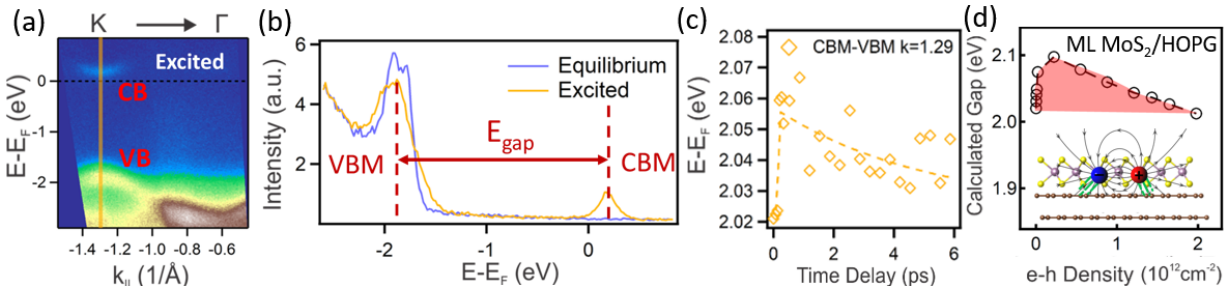

Figure 1. Band structure measured at K valley for photoexcited ML MoS_2/HOPG. Vertical lines in (a) denotes the cut at K point, where energy distribution curves (EDCs) are extracted and displayed in (b). The energy separation between the VBM and CBM is defined as the energy of the bandgap. (c) Measured bandgap size dynamics in time following the photoexcitation. (d) Calculated band gap for photoexcited ML MoS_2/HOPG in the presence of bound electron-hole gases with excitonic correlations.

References

[1] F. Liu, M. E. Ziffer, K. R. Hansen, J. Wang, and X. Zhu, Phys Rev Lett **122**, 246803 (2019).

[2] E. J. Sie, J. W. McIver, Y.-H. Lee, L. Fu, J. Kong, and N. Gedik, Nature Materials **14**, 290 (2015).

[3] E. J. Sie, C. H. Lui, Y.-H. Lee, L. Fu, J. Kong, and N. Gedik, Science **355**, 1066 (2017).

[4] H. Stolz and R. Zimmermann, physica status solidi (b) **94**, 135 (1979).

[5] Y. Lin et al., arXiv preprint arXiv:2205.05821 (2022).