Excitonic effects in optical-field-driven quasi 2D materials from first-principle time-dependent GW approach

Yang-hao Chan[1,2*]

[1]*Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei, Taiwan*

[2]*National Center of Theoretical Sciences, Physics Division, Taipei, Taiwan*

*yanghao@gate.sinica.edu.tw

Atomically thin quasi two-dimensional (2D) insulating materials exhibit novel exciton physics due to ineffective screening, quantum confinement, and topological effects. Such exciton physics has recently been studied in details experimentally and theoretically. Going beyond near-equilibrium set-up, one expects that excitonic effects also dominate the responses of out-of-equilibrium systems and can lead to interesting phenomena in optically-driven 2D materials. Using a newly developed real-time, non-equilibrium Green function method within the adiabatic GW approximation [1,2], we show that how excitonic effects give rise to strong nonlinear optical response such as shift current, and second harmonic generation [2-4]. Furthermore, we show that, in optical-field-driven angle-resolved photoemission spectroscopy (ARPES) experiments, the energy and wavefunction of excitons may be measured directly under achievable laboratory conditions. With optical pump frequencies close to the resonance frequency for exciton excitations, distinct excitonic features manifest themselves dramatically as modulated replicas of the involved valence band states. We also find that, at higher pump intensity, the quasiparticle band energies are strongly renormalized due to the driving optical fields. [5,6]

**We acknowledges financial support from the Institute of Atomic and Molecular Sciences at Academia Sinica, the National Science and Technology Council (NSTC), and the National Center for Theoretical Sciences (NCTS).

References:

[1] C. Attaccalite, M. Gruning, and A. Marini, Phys. Rev. B 84, 245110 (2011).

[2] Y.H. Chan, Diana Y. Qiu, Felipe H. da Jornada, and Steven G. Louie, PNAS 118 (25) e1906938118 (2021)

[3] Yi-Shiuan Huang, Y.H. Chan, and Guang-Yu Guo, Phys. Rev. B 108, 075413 (2023)

[4] Jiawei Ruan, Y.H. Chan, and Steven G. Louie, arXiv:2310.09674 (2023)

[5] Y.H. Chan, Diana Y. Qiu, Felipe H. da Jornada, and Steven G. Louie, PNAS 120 (32) e2301957120 (2023)

[6] V. Pareek, et al., arXiv:2403.08725