

Gate tunable spin-singlet/triplet charged interlayer excitons in atomically thin heterostructures

Joe, Andrew¹, Jauregui, Luis A.¹, Pistunova, Kateryna¹, Lu, Zhengguang^{2,3}, Wild, Dominik S.¹, Scuri, Giovanni¹, De Greve, Kristiaan^{1,4}, Gelly, Ryan J.¹, Sung, Jiho^{1,4}, Mier Valdivia, Andrés⁵, Sushko, Andrey¹, Taniguchi, Takashi⁶, Watanabe, Kenji⁶, Smirnov, Dmitry^{2,3}, Lukin, Mikhail D.¹, Park, Hongkun^{1,4}, Kim, Philip^{1,5*}

¹ Department of Physics, Harvard University, Cambridge, Massachusetts, USA

² National High Magnetic Field Laboratory, Tallahassee, Florida, USA

³ Department of Physics, Florida State University, Tallahassee, Florida, USA

⁴ Department of Chemistry and Chemical Biology, Harvard University, Cambridge, Massachusetts, USA

⁵ John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts, USA

⁶ National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

*e-mail: philipkim@g.harvard.edu

Abstract: Strong Coulomb interactions across the atomically separated electrons and holes can create neutral and charged excitons with long lifetimes in van der Waals (vdW) heterostructures [1]. Large spin-orbit coupling in transition metal dichalcogenides (TMD) provide helical coupling of photons and excitons with different spin states. Here, we fabricate a dual-gated MoSe₂/WSe₂ vdW heterostructure optoelectronic device and demonstrate electrically tunable optical helicity using spin-singlet and spin-triplet charged interlayer excitons. We use electrostatic gate doping to reach the spin-split conduction band, accessing the spin-triplet exciton state in TMDs (Fig. 1a) and show they still have lifetimes significantly longer than intralayer excitons (Fig. 1b). By applying a magnetic field, we measure the g-factors for the two states and show that they have opposite signs (Fig. 1c). Due to a non-trivial phase introduced in rotation by the in-plane displacement of the layers, the singlet and triplet interlayer excitons have opposite valley characteristics when emitting the same circular polarization. Electrically controlled and optically addressed singlet/triplet charged excitons provide a route for optoelectronic valleytronic devices that can dynamically switch between the two valley states without changing the helicity of incoming light.

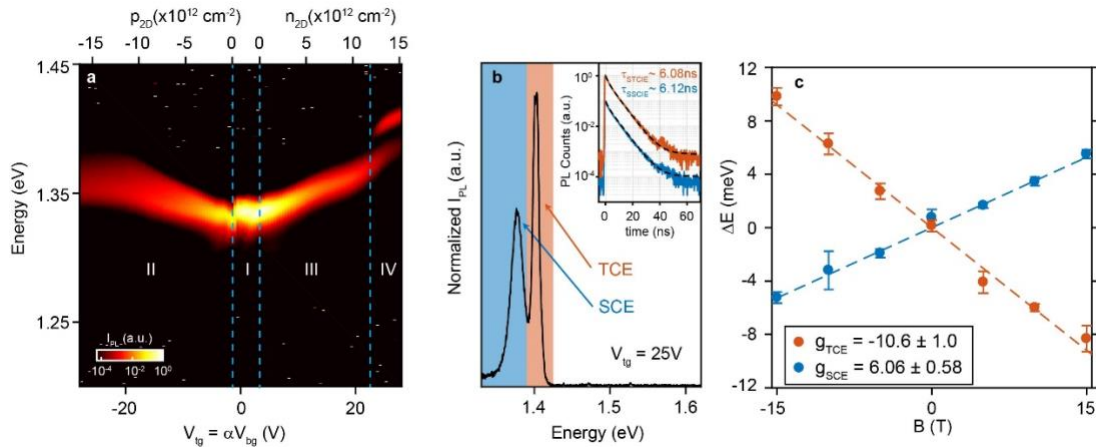


Fig. 1. (a) Electrostatic doping dependence of the photoluminescence spectra that shows spin-singlet and spin-triplet charged interlayer excitons at high n -doping. (b) Single spectra at $V_{tg} = 25$ V and lifetimes of the two peaks. (c) Energy splitting ($\Delta E = E_{\sigma^+} - E_{\sigma^-}$) of the photoluminescent energy for the triplet and singlet with calculated g-factors.

References

- [1] L. A. Jauregui *et al.*, “Electrical control of interlayer exciton dynamics in atomically thin heterostructures,” *arXiv*, p. 1812.08691, 2018.

