# Coherent Coupling and Hybridization of Bright and Dark Excitons Enabled by Intense Magnetic Fields

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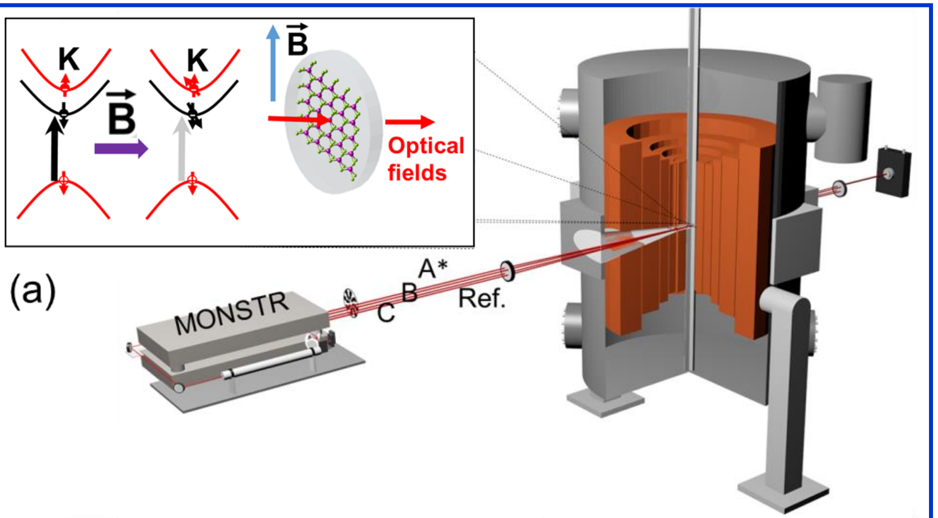
Transition metal dichalcogenide (TMD) monolayers exhibit unusual properties that arise directly from a band structure with spin-valley locking due to their broken inversion symmetry and strong spin−orbit coupling. Potential applications range from light emitting devices and displays, to electronic devices, to solar cells. The lack of inversion symmetry gives rise to spin-valley polarization, whereas *the strong spin−orbit interaction results in the splitting of the conduction and valence band extrema, forming spin-allowed "bright" excitons and spin-forbidden "dark" excitons*.

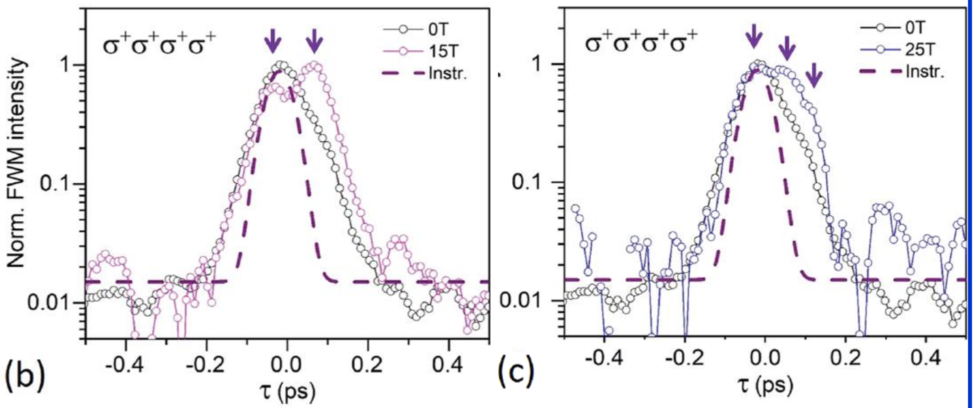
In monolayer $WSe_2$ - the two-dimensional (2D) TMD studied by this MagLab user collaboration - the dark excitons are optically inactive. However, they play a key role in the observed optical properties, as they provide alternative decay channels via many-body interactions that affect the lifetime and coherence of the bright excitons. As such, *probing the interaction of dark and bright excitons using coherent optical excitations is important for fundamental understanding and applications of 2D $WSe_2$*.

Magnetic fields up to 25T applied *parallel* to the $WSe_2$ plane lead to a partial brightening of the energetically lower lying exciton, leading to an increased coherence time. *Utilizing a broadband femtosecond pulse excitation, the bright and partially allowed excitonic state can be excited simultaneously, resulting in coherent quantum beating between these states.* Magnetic fields *perpendicular* to the $WSe_2$ plane energetically shift the bright and dark excitons relative to each other, resulting in the hybridization of the states at the K and K′ valleys. Each of these effects is accurately modeled by time-dependent density functional theory calculations. These experiments show that magnetic fields can be used to control the coherent dephasing and coupling of the optical excitations in atomically thin semiconductors.

**(a)** Experimental setup using the 25T Split-Helix magnet and the multidimensional optical nonlinear spectrometer (MONSTR). As shown in the inset, when applied in the 2D plane of the sample, a magnetic field tilts the spins. As a result, the spin forbidden transition (black arrow) becomes partially allowed or brightened (gray arrow).



The time-integrated Four Wave Mixing (FWM) intensity shows quantum beating at **(b)** 15T and **(c)** 25T, evidenced by the double and triple peaks arising (purple arrows) in the time evolution of the FWM signal. This is due to the coherent coupling of the bright and dark excitons enabled by the intense magnetic field.



**Facilities and instrumentation used:** 25T DC split helix optical magnet (Cell 5)

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