Controlled Coherent Coupling and Dynamics of Excitons in a Monolayer Semiconductor

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We perform coherent nonlinear spectroscopy of an electrostatically-gated monolayer of $MoSe_2$. This iconic material from the family of semiconducting transition metal dichalcogenides hosts correlated electron-hole pairs, called excitons (X), of enhanced binding energy well beyond 100 meV and high oscillator strength yielding sub-picosecond radiative lifetimes. In particular, Coulomb interactions between Xs can generate giant optical nonlinearities, like harmonic generation series and wave-mixing responses. In addition, the surface protection thanks to the encapsulation by thin flakes of hBN eradicates the structural disorder, allowing to approach the homogeneous linewidth (γ) of the X transitions. This enabled to redefine the state-of-the-art in the field of correlated systems in condensed matter, with such landmark achievements like observation of the Wigner crystal state and the optical sensing of quantum Hall effect in graphene.

In that context, it is of crucial importance to ascertain in these devices the influence of the free carrier density on γ. To accomplish that, we perform four-wave-mixing micro-spectroscopy at 8 K, directly quantifying not only the X dephasing, but also an amount of residual disorder through the photon-echo formation, and the X population dynamics with 100 fs resolution.

We find that with increasing the gate-voltage governing the electron density, due to X interaction with the Fermi sea, γ increases from 1 meV to 6 meV. This goes in tandem with the shortening of the X population lifetime from 0.9 ps to 0.2 ps. For the charged exciton (CX) the measured dephasing is faster than for X, and also an increase of γ from 2 meV to 4 meV is observed. Conversely, the CX lifetime, which equals 6 ps, is independent of the gate-voltage.

Finally, by performing two-dimensional four-wave-mixing (2D FWM) we show that X and CX are coherently coupled, and with the coupling strength which is directly controlled with the gate voltage. This observation brings new fundamental insights into the optoelectronics of $MoSe_2$.

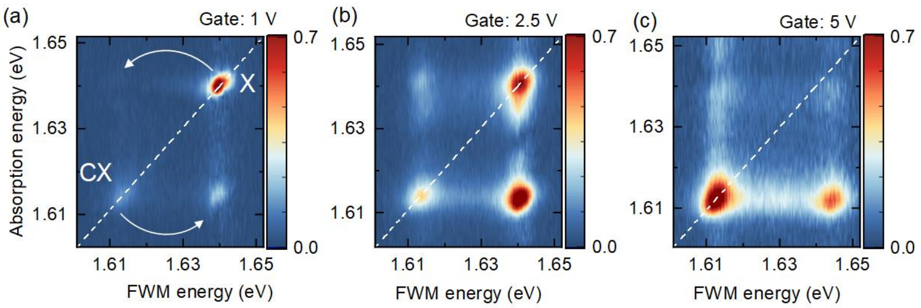

Fig.1 *(a-c) 2D FWM spectra for different gate voltages. Diagonal peaks correspond to X, CX signal. Off-diagonal peaks indicate coherence transfer that strongly depends on carrier density. (arrows mark transfer direction)*