Coherence in semiconductor nanostructures inferred with nonlinear spectroscopy

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Optical spectra of semiconductors display narrow resonances below the absorption edge, which are attributed to the **excitons**, i.e. Coulomb correlated excitations of electrons and holes. When shining laser light onto a semiconductor, the induced excitonic polarization collectively oscillates in phase - this phenomenon is known as the exciton **coherence**. The coherence decays in time owing to various processes occurring in a solid: radiative and nonradiative decays, scattering with carriers and phonons.

Over the last 15 years, by employing tools of nonlinear spectroscopy, I have been exploring coherent dynamics and control of excitons in various semiconductor nanostructures. In this talk, I will highlight the results obtained on $MoSe_2$, which is an iconic layered semiconductor from the family of transition metal dichalcogenides. In a monolayer form, which can be obtained either via mechanical exfoliation or the epitaxial growth, $MoSe_2$ hosts robust excitons of enhanced binding energy and high oscillator strength. Owing to the strong Coulomb interactions, they can generate giant optical nonlinearities, like harmonic generation series and wave-mixing responses [1, 2].

As an example, I will explain how nonlinear spectroscopy, which retrieves such responses, permits us to accurately assess homogeneous and inhomogeneous broadenings from the spectral line width in a presence of disorder. I will then argue that, owing to the exciton localization via disorder, these quantities are anti-correlated [3]. I will demonstrate that with nonlinear spectroscopy one can monitor the crossover from the toward 'clean' samples [4], by inspecting coherent ultrafast transients.

The last part of this presentation will tackle a more involved concept of coherent coupling between individual quantum systems. I will show that by performing two-dimensional coherent spectroscopy, we achieved controlled coupling in advanced quantum dot molecule devices.

The talk will be concluded by sketching emerging research opportunities and collaborations. In particular, in the context of the J-Fast International Research Laboratory, recently launched between Tsukuba and Grenoble.

[1] T. Jakubczyk *et al.*, *Radiatively limited dephasing and exciton dynamics in $MoSe_2$ monolayers revealed with four-wave mixing microscopy. Nano Lett.* **16**, 5333 (2016).

[2] T. Hahn *et al.*, *Destructive photon echo formation in six-wave mixing signals of a $MoSe_2$ monolayer. Adv. Sci.* **9**, 2103813 (2022).

[3] T. Jakubczyk *et al.*, *Coherence and density dynamics of excitons in a single-layer MoS_2 reaching the homogeneous limit. ACS Nano* **13**, 3500 (2019).

[4] C. Boule *et al.*, *Coherent dynamics and mapping of excitons in single-layer $MoSe_2$ and WSe_2 at the homogeneous limit. Phys. Rev. Materials* **4**, 034001 (2020).