

Visualizing exciton coupling dynamics in transition metal dichalcogenide heterostructures in space and time

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Abstract: We demonstrate coherent coupling between excitons in a MoSe₂/WSe₂ heterostructure, as well as rapid interlayer electron and hole transfer. We visualize the spatial homogeneity of the coupling in the presence of significant sample inhomogeneities. © 2022 The Author(s)

Transition metal dichalcogenides (TMDs) have received considerable attention over the past decade for their efficient light-matter coupling, which has rendered them a prime materials platform for applications from solar-energy and lasers to quantum light-emitting diodes. Recently researchers in the TMD community have paid considerable attention to their rapid charge transfer [1], coherent coupling [2], and high tunability for Moiré excitons [3]. However, the intricate dynamics and interactions between excitons in heterostructures of these materials are elusive due to limitations in the employed experimental techniques. Here, we spatially map coherent coupling between excitons and rapid electron and hole transfer in an MoSe₂/WSe₂ heterostructure. We visualize the robustness of coherent coupling and charge transfer across the heterostructure, even in the presence of significant strain, using multi-dimensional coherent imaging spectroscopy (MDCIS). We further show that a common technique for characterization of these samples, photoluminescence (PL), suggests a very heterogeneous sample and fails to capture the good overall quality and robustness of key sample properties.

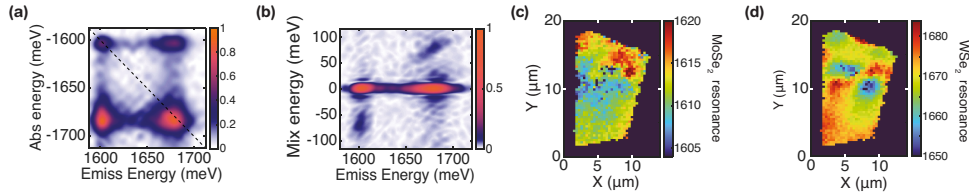


Fig. 1: (a) Low-power absorptive MDCS spectrum of a MoSe₂/WSe₂ heterostructure. (b) Zero-quantum spectrum showing signatures of coherent coupling between MoSe₂ and WSe₂. (c) Energy map of MoSe₂ resonance (d) Energy map of WSe₂ resonance

The sample studied in this work is a MoSe₂/WSe₂ monolayer, encapsulated in hexagonal Boron Nitride. Using multi-dimensional coherent spectroscopy (MDCS), we correlate absorption and emission energies, with details about the methods found elsewhere [2, 4]. A characteristic low-power absorptive MDCS spectrum for the sample is shown in Fig. 1(a). Here, the two on-diagonal peaks (dashed line) are due to the MoSe₂ and WSe₂ intralayer excitons, while the presence of the two off-diagonal peaks suggest coupling - incoherent or coherent - between excitons in the MoSe₂ and WSe₂ monolayers. Incoherent coupling is expected to occur due to rapid charge transfer in these samples. Coherent coupling, or coherent interactions, between the excitons, can be caused by static dipole-dipole, exchange interactions, or transition dipole (Förster) coupling [5], among other things. We distinguish between coherent and incoherent coupling by performing zero-quantum spectroscopy [2], where instead of the absorption energy, the mixing energy between the two resonances is resolved by observing the signal along the pump-probe delay T . The zero-quantum spectrum shown in Fig. 1(b) clearly shows two peaks at mixing energies around +71 meV and -74 meV, the energy difference between MoSe₂ and WSe₂ resonances in this sample, and is thus a clear indication of coherent coupling. We have previously resolved the incoherent electron and hole transfer dynamics in this sample using MDCS [2].

The resonance energies are plotted in Fig. 1(c) for the MoSe₂ exciton and Fig. 1(d) for the WSe₂ exciton. Res-

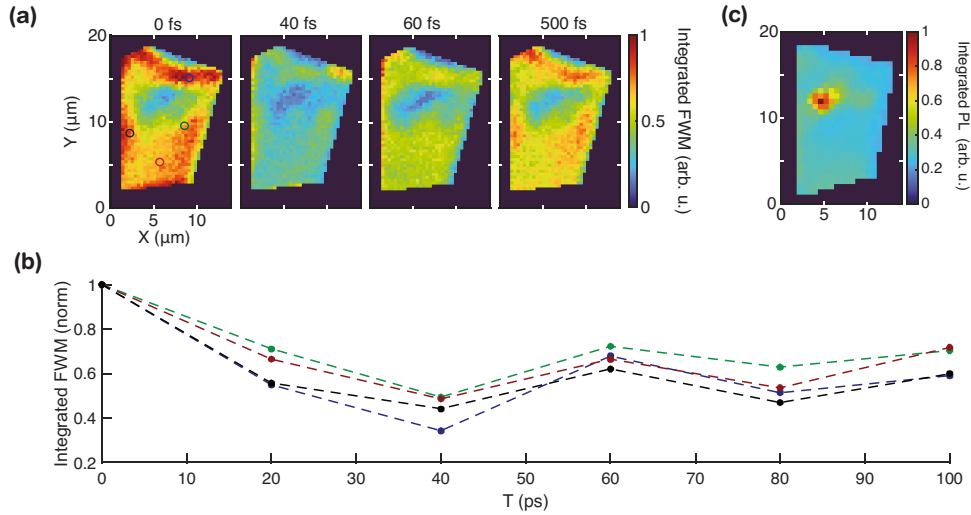


Fig. 2: **(a)** Integrated FWM signal for the MoSe₂/WSe₂ coupling peak for varying T delay across the sample. **(b)** T -dependent integrated FWM signal for the MoSe₂/WSe₂ coupling peak for four sample points marked in (a). **(c)** Integrated PL of the interlayer exciton across the sample.

onance shifts of up to 15 meV can be observed across the sample. Most importantly, there are areas of correlation and anti-correlation between the resonance shifts, suggesting complex local strain dynamics across the sample that are expected to change the coupling dynamics across the sample.

However, despite these notable inhomogeneities, MDCIS experiments show both coherent coupling and charge transfer are mostly unaffected across the sample. In Fig. 2(a) the integrated four-wave mixing amplitude of the MoSe₂/WSe₂ coupling peak (upper right in Fig. 1(a)) as a function of pump-probe delay T is shown. Both the initial decay and recovery of the signal due to the coherent coupling oscillation along T , as well as an increase in signal for longer T delays due to charge transfer are apparent. Most strikingly, both coherent coupling and charge transfer are mostly homogeneous across the sample, with the temporal dynamics for early T delays at four select sample points being shown in Fig. 2(b). The comparison between the four sample points, marked in Fig. 2(a), and the respective resonance shifts shown in Fig. 1(c,d) show that despite notable resonance shifts, the coherent oscillation is strongly visible in all regions of the sample, with similar amplitude, albeit coherent coupling at the very top part is somewhat weaker. Most notably, this is not what would inherently be expected by looking at the integrated interlayer exciton PL, which strongly varies across the sample and is maximized at a strained area of the sample, showing suppressed FWM intensity. Indeed, this is common in TMD monolayers, where FWM is suppressed due to a reduced dipole moment [6] in strained areas, but these areas show increased PL.

The future of TMDs in device applications is inherently coupled to the scalability and quality of fabricated devices. This work shows a reproducibility of crucial physical properties across the sample, laying the groundwork and strengthening the case for TMDs as a next generation materials platform.

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