Ultrafast coherent dynamics of a hybrid WS_2/plasmon structure probed by two-dimensional electronic spectroscopy

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Transition metal dichalcogenide (TMD) monolayers (1L) have been established as important building blocks for quantum materials. Hybridization between light and matter states, in particular in plasmonic nanostructures, offers great opportunities to tailor their optical and electronic properties on the nanoscale [1]. To probe and distinguish ubiquitous coherent and incoherent energy transfer processes, ultrafast two-dimensional electronic spectroscopy (2DES) has proven to be a valuable tool [1,2]. Here, we investigate a hybrid plasmonic nanostructure comprised of 1L-WS_2 [2] placed on a periodic silver nano-slit array using ultrafast 2DES with 10-fs time resolution (Fig. 1a).

When brought into contact, the A exciton of 1L-WS_2 (red line in Fig. 1b) hybridizes with the surface plasmon polariton (SPP) resonance (black line) of the nanostructure to form upper and lower polariton states (blue line). We observe a 20-fold increase of the optical nonlinearity compared to bare 1L-WS_2 and ultrafast plexciton dynamics during the dephasing time (~50 fs). 2DES maps in Fig. 1c-f show a rapid evolution from a sub-peak structure (diagonal and cross peaks, panels c,d) to more featureless vertical stripes (panels e,f) within the dephasing time.

We model our nonlinear spectra using a three-oscillator model [3] that accounts both for far field coupling of the SPP to momentum bright excitons and near field couplings to momentum dark excitons of the TMD and consider Pauli blocking and excitation-induced dephasing as the main origins for the optical nonlinearity. This allows us to explain our experimental data as a transfer from coherent polaritons at early times to incoherent polaritons and dark state populations. Our findings highlight the important role of many-body interactions and dark states in the collective response of TMD-based polariton systems. Using ultrafast 2DES, we obtain access to probe and disentangle the coherent and incoherent dynamics in TMD-based plasmonic systems.

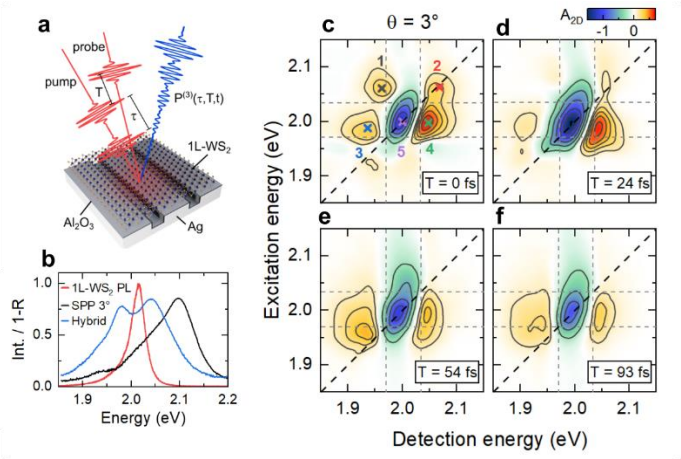

Figure 1: a) Sketch of the sample consisting of 1L-WS_2 on a Al_2O_3-coated silver nanoslit array and pulse ordering for 2DES. b) Linear spectrum of the SPP without 1L-WS_2 (black line) and photoluminescence spectrum of the A exciton of the bare TMD (red line) at ~2 eV. The SPP resonance shifts closer to 2 eV when changing the metal interface to TMD. The hybrid nanostructure shows upper and lower polariton resonances (blue line). c-f): Ultrafast 2DES of the hybrid structure. During the dephasing time, coherent polaritons are probed. At later delays, essentially featureless stripes along the excitation probe incoherent polaritons and dark states.

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References

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