Ultrafast mid-infrared 1*s* intraexcitonic spectroscopy in monolayer MoS_2

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Abstract

Ultrafast optical Photogenerated electron-hole pairs in solids create bound states whose elementary quasiparticle state is called 1*s* exciton in a Wannier-Mott exciton model. Above the fundamental 1*s* exciton, recent visible and near-infrared investigations revealed the excited excitons are much richer, exhibiting a series of Rydberg-like states [1,2]. Probing internal transition between these non-hydrogenic series, however, demand a fundamentally different experimental tool, capable of probing optical transitions from 1*s* "bright" to n*p* "dark" states [3,4]. Here, we employed ultrafast mid-infrared spectroscopy to explore the 1*s*-intraexcitonic transitions in monolayer MoS_2 [5]. As shown in Fig. 1, we observed two-folded 1*s*→3*p* intraexcitonic transitions within A and B exciton and the 1*s*→2*p* transition between A and B exciton. Our time-resolved analysis revealed that it takes about 0.7 ps for the 1*s* A exciton before reaching quasi-equilibrium whose characteristic time is associated with a rapid population transfer from the 1s B exciton. Our experiment, otherwise hidden in linear or nonlinear spectroscopy, may provide a second look for understanding the many-body exciton dynamics in two-dimensional materials.

References

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Figures

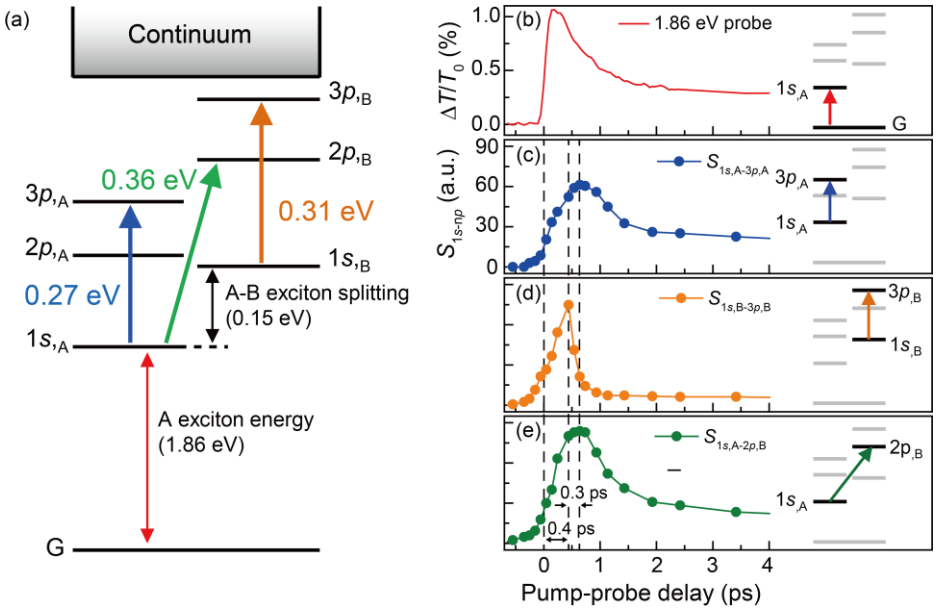

Figure 1. (a) Energy diagram of the excitonic Rydberg series in MoS_2. **(b)** transient band-to-band dynamics probed y 1.86 photon energy. **(c-e)** Transient dynamics of the intraexcitonic transition for each three oscillator are shown at each row: **(c)** $1s_{,A}$→$3p_{,A}$, **(d)** $1s_{,B}$→$3p_{,B}$, and **(e)** $1s_{,A}$→$2p_{,B}$, respectively. Dashed lines show the maximum peak for each intraexcitonic transition.