Spin-Valley Relaxation of Rydberg Excitons in 2D Semiconductors

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Optical properties of atomically thin transition metal dichalcogenides (TMDs) are controlled by robust excitons characterized by a very large oscillator strength [1]. The use of fully encapsulated TMD monolayers (ML) in hexagonal boron nitride (hBN) yields narrow optical transitions approaching the homogeneous exciton linewidth [2]. In this work we have measured the optical and spin/valley properties of excitons in a very high-quality WSe_2 ML allowing the observation of excited excitonic states up to n=5 by photoluminescence or reflectivity spectroscopy. This offers a unique possibility to investigate the spin relaxation of excited excitonic states. Rydberg excitons, characterized by large spatial extension and reduced electron-hole overlap, must have spin-valley dynamics different from that of ground state excitons [3,4]. Here we report a direct measurement of spin relaxation of Rydberg excitons in high-quality monolayer WSe_2 using continuous-wave and time-resolved optical orientation experiments. Remarkably, we measure exceptionally large photoluminescence circular polarization, approaching 90% for the 3s state. Time-resolved measurements reveal a strong increase of the spin relaxation time with the principal quantum number, from ~2 ps for the 1s exciton to ~75 ps for the 3s exciton. To our knowledge, this is the first time that the Rydberg exciton spin relaxation time has been measured directly, and the results will be discussed in the framework of the exciton spin relaxation mechanism driven by the long-range exchange interaction between the electron and the hole.

References

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Figures

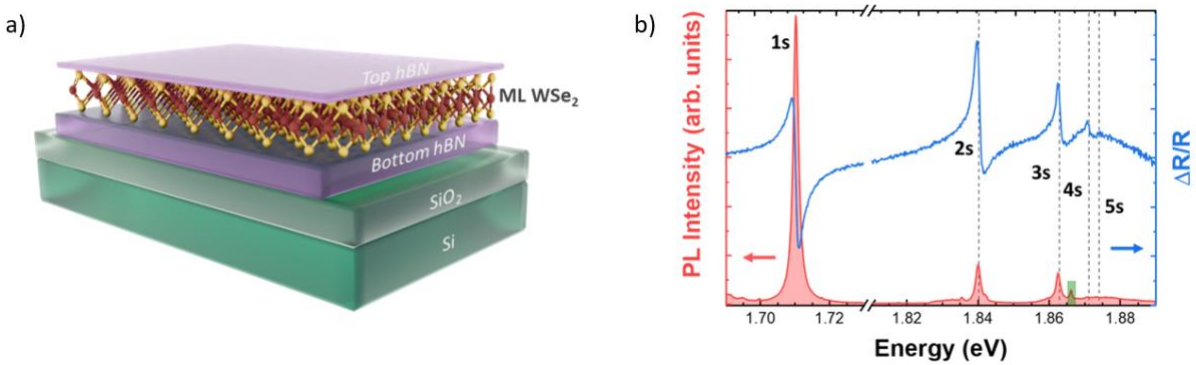

Figure 1: a) Schematics of the investigated samples, b) Photoluminescence (red) and reflectivity contrast (blue) measurements on monolayer WSe_2 at 4 K, with signature of excited exciton states up to n=5.