Direct Imaging of Exciton Coupling and Valley Polarization Dynamics in Monolayer WS2

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Abstract

Time- and angle-resolved photoemission (tr-ARPES) has emerged as a powerful tool for imaging momentum-resolved dynamics in small, monolayer transition metal dichalcogenide samples. In this talk, I will present our recent work applying tr-ARPES to study the dynamics of exciton coupling and ultrafast valley depolarization in monolayer WS2. After excitation of the nominal B exciton resonance, we observe strong intravalley coupling between the B1s exciton and An>1 exciton states in the initial photoexcited spectrum. Our measurements indicate that the dominant valley depolarization mechanism conserves the exciton binding energy and momentum. While this conservation is consistent with Coulomb exchange-driven valley depolarization, we do not observe a momentum or energy dependence to the depolarization rate as would be expected for the exchange-based mechanism. On longer timescales, we also observe that exciton relaxation is accompanied by contraction of the momentum space distribution.