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Unraveling the Valley Depolarization dynamics in WS_2 with TR-ARPES

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In two-dimensional (2D) transition metal dichalcogenides (TMDCs), strong Coulomb interactions and their lack of inversion symmetry making them has led to tightly bound excitons which are ideal for valleytronics applications. However, the valley information encoded in bright excitons is rapidly lost due to the presence of many scattering channels such as intervalley exchange interactions as well as phonon-induced scattering. Additionally, the complexity of the exciton landscape, which hosts a variety of bright and dark excitons species, has further hindered any comprehensive understanding of valley depolarization dynamics.

Time- and angle-resolved photoemission spectroscopy (TR-ARPES) has emerged as a powerful tool for studying excitons in 2D semiconductors. By directly imaging excitons in energy and momentum space while simultaneously resolving their constituent electrons and holes, we could study different excitonic species within the material [1–4]. Here in this presentation, we will demonstrate how we can employ TR-ARPES to track the evolution of valley-polarized excitons in monolayer WS_2 across the entire Brillouin zone. By extracting exciton populations and key scattering timescales, we identify excitation conditions under which intervalley exchange scattering is suppressed. This suppression leads to the formation of a momentum-dark exciton that retains its valley selectivity for more than two orders of magnitude longer than its bright exciton counterpart. These findings offer new insights into controlling valley polarization, paving the way for future valleytronic applications.

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