Time-resolved ARPES of excitons in a 2D semiconductor

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The unique optical properties of two-dimensional semiconductors, such as monolayer transition metal dichalcogenides, have provided deep insights into fundamental materials research and emerging nanotechnologies [1,2]. The strong Coulomb interactions and distinct structural symmetries in these materials give rise to a rich variety of photoexcited states, including bright and dark excitonic complexes that are tightly bound and valley-spin polarized [3]. However, directly accessing both bright and momentum-forbidden dark excitons and fully capturing their relaxation processes, occurring rapidly along both the energy and momentum axes, has not been possible with conventional experimental probes. In particular, a direct determination of the quasi-equilibrium exciton distribution over the entire Brillouin zone after the initial cooling is essential for future research and optoelectronic applications of two-dimensional materials. Here, by performing time- and momentum-resolved photoemission spectroscopy based on table-top MHz XUV source (21.7eV) on a micron-scale monolayer flake of WSe_2, we directly observe the exciton distribution at the K- and Σ-valleys and measure their dynamics and relaxation pathways under different excitation conditions across the Brillouin zone [4].

In contrast to the typical behavior in bulk semiconductors, we find that bound excitons form within a few hundred femtoseconds. These excitons are characterized by multiple near-degenerate species involving holes in the K-valley and electrons in both the K- and Σ-valleys of the Brillouin zone, with relaxation pathways dictated by the initial excitation conditions. For resonant excitation, we see the rapid formation of K-valley excitons, followed by scattering of the exciton population to the Σ-valley in 300fs. In contrast, for above-gap excitation at 2.5eV, we observe excitons forming within 500 fs, with electrons in the K- and Σ-valley, but with a larger initial density at Σ, and net scattering from the Σ- to K-valley. Our measurements provide the first global view of the ultrafast optical response of 2D semiconductors and demonstrate the impact of studying excited-state dynamics over the full Brillouin zone in condensed matter systems.

References

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