Rydberg series of excitons in WSe_2 multilayers

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Semiconducting transition metal dichalcogenides of group VI exhibit strong excitonic effects and a transition from an indirect to a direct band gap when reduced to monolayer thickness. While extensive studies have characterized the Rydberg series of excitons in monolayers, a comprehensive understanding of their behavior in multilayers remains incomplete. In multilayer structures, despite the presence of an indirect band gap, momentum-direct excitons continue to play a dominant role in shaping the optical response. Understanding the interplay between interlayer coupling, hybridization effects, and excitonic properties in these systems is crucial for advancing their potential applications in optoelectronics and valleytronic devices.

In this work, we employ magneto-reflectance spectroscopy in conjunction with theoretical modeling based on the $\mathbf{k}\cdot\mathbf{p}$ approach to explore the excitonic landscape of WSe_2 bi-, tri-, and quadlayers. Our reflectance measurements reveal a series of excitonic resonances across all investigated thicknesses, beginning with a ground-state transition whose intensity is comparable to that of the monolayer $1s$ exciton. Higher energy excitonic features exhibit a reduction in intensity, consistent with the expected behavior of the Rydberg series. However, in tri- and quadlayers, the absence of a significant increase in diamagnetic shift across these states suggests the coexistence of multiple distinct excitonic series rather than a single one. Additionally, the observed variations in oscillator strengths indicate strong layer-dependent modifications to the electronic wavefunctions, further emphasizing the role of interlayer coupling.

Theoretical analysis of the $\mathbf{k}\cdot\mathbf{p}$ model for N-layer ($N = 2, 3, 4$) WSe_2 reveals a fine structure in the electronic bands at the $K^{\pm}$ valleys. The conduction band states become N-fold degenerate, while hybridization in the valence band leads to the emergence of distinct optical transitions. Consequently, N types of excitonic states are expected at the $K^{\pm}$ points, with some exhibiting degeneracy. Our experimental findings confirm these predictions, revealing a single Rydberg series in bilayers, two in trilayers, and four in quadlayers. The theoretical model further allows us to estimate the binding energies and spatial distributions of the excitonic states, providing deeper insight into their quantum nature. This detailed comparison between experiment and theory provides new insights into the nature of excitonic states in multilayer WSe_2, highlighting the complex interplay between interlayer hybridization and excitonic structure, and offering potential avenues for tunable optical properties in layered materials.

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