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Optical spectroscopy and imaging of the higher energy excitons and bandgap of monolayer MoS_2 NICHOLAS BORYS, WEI BAO, EDWARD BARNARD, Molecular Foundry, Lawrence Berkeley National Lab, CHANGHYUN KO, University of California Berkeley, SEFAATIN TONGAY, Arizona State University, JUNQIAO WU, University of California Berkeley, LI YANG, Washington University in St. Louis, P. JAMES SCHUCK, Molecular Foundry, Lawrence Berkeley National Lab — Monolayer MoS_2 (ML-MoS_2) exhibits a rich manifold of excitons that dictate optoelectronic performance and functionality. Disentangling these states, which include the quasi-particle bandgap, is critical for developing 2D optoelectronic devices that operate beyond the optical bandgap. Whereas photoluminescence (PL) spectroscopy only probes the lowest-energy radiative state and absorption spectroscopy fails to discriminate energetically degenerate states, photoluminescence excitation (PLE) spectroscopy selectively probes only the excited states that thermalize to the emissive ground state exciton. Using PLE spectroscopy of ML-MoS_2, we identify the Rydberg series of the exciton A and exciton B states as well as signatures of the quasi-particle bandgap and coupling between the indirect C exciton and the lowest-energy A exciton, which have eluded previous PLE studies. The assignment of these states is confirmed with density functional theory. Mapping the PLE spectrum reveals spatial variations of the higher-energy exciton manifold and quasi-particle bandgap which mirror the heterogeneity in the PL but also indicate variations in local exciton thermalization processes and chemical potentials.

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