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High Resolution Nonlinear Spectroscopy of 2D Excitons in Monolayer $MoSe_2$ JOHN SCHAIBLEY, TODD KARIN, University of Washington, HONGYI YU, University of Hong Kong, JASON ROSS, PASQUAL RIVERA, AARON JONES, MARIE SCOTT, University of Washington, JIAQIANG YAN, Oak Ridge National Laboratory, DAVID MANDRUS, University of Tennessee, WANG YAO, University of Hong Kong, KAI-MEI FU, XIAODONG XU, University of Washington — Monolayer transition metal dichalcogenides (mTMDs), such as $MoSe_2$, have emerged as the first truly 2D semiconductors, exhibiting a wide range of novel electro-optical phenomena which arise from the material's graphene-like honeycomb lattice. The optical response of mTMDs is dominated by strongly bound excitons which are localized in momentum space to two sets of inequivalent valleys (+K, -K) at the edge of the Brillouin zone. We report the first high resolution nonlinear spectroscopy measurements on monolayer $MoSe_2$. Differential reflection measurements reveal that the degenerate nonlinear optical response agrees with the previously reported photoluminescence and reflectivity measurements, showing a broadened linewidth on order of 5 meV. Non-degenerate spectral holeburning measurements reveal narrow optical resonances approximately 1000 times narrower (order of 2 micro eV), indicating that excitons in mTMDs are dominantly inhomogenously broadened and exhibit an intrinsic lifetime on the order of 1 ns, over an order of magnitude longer than all previously reported lifetimes obtained through time domain techniques. Polarization dependent spectral holeburning measurements probe valley dependent processes such as the intervalley relaxation rates.

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