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Neutral and Charged Inter-Valley Biexcitons in Monolayer Transition Metal Dichalcogenides KAI HAO, LIXIANG XU, University of Texas at Austin, JUDITH SPECHT, Institut fr Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universitt Berlin, PHILIPP NAGLER, University of Regensburg, KHA TRAN, AKSHAY SINGH, CHANDRIKER DASS, University of Texas at Austin, CHRISTIAN SCHLLER, TOBIAS KORN, University of Regensburg, MARTEN RICHTER, ANDREAS KNORR, Institut fr Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universitt Berlin, XIAOQIN LI, University of Texas at Austin, GALAN MOODY, National Institute of Standards Technology — In monolayer transition metal dichalcogenides, tightly bounded exciton and trion dominate the optical properties. Higher-order correlated states, such as biexciton, are possible but are difficult to unambiguously identified with linear optical spectroscopy alone. With polarization resolved two dimensional coherent spectroscopy, unambiguously signatures of neutral and charged inter-valley biexcitons are observed in monolayer MoSe2 with ˜20 meV and ˜3meV binding energies, which are consistent with variational and Monte Carlo calculations. These higher–order correlated states consist of quasiparticles formed at opposite valleys with large crystal momentum difference, making them a unique type of bound states with no direct analog in conventional semiconductors. Our findings offer new opportunities for developing ultrathin biexciton lasers andpolarization-entangled photon sources and for creating exotic exciton-polariton condensates.

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