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CW all-Optical Quadruple Resonance Spetroscopy of Sodium Dimer[1] ERGIN AHMED, PENG QI, MARJATTA LYYRA, Temple University — We report the first cw all-optical quadruple resonance excitation experiment with all excitations steps being coherently driven by a combination of four tunable lasers. We have constructed a theoretical model to simulate the experimentally observed signal based on density matrix formalism, which can be used also to identify optimal laser wavelengths as well as laser propagation geometry for observation of coherence effects such as the Autler-Townes splitting. This excitation technique is very general and can be used to probe transitions to highly excited electronic states and their transition dipole moments. Of special interest are excited states with ion pair character. Transitions to such electronic states have unusual electronic transition dipole moment functions. The Autler-Townes effect associated with this technique can be used as a probe of transition dipole moment functions to facilitate a study of charge separation as a function of internuclear distance with well-defined initial conditions. This technique also allows for general access to highly excited electronic states at large internuclear distance with high resolution and thus can be used to probe Rydberg states in high vibrational levels, which are difficult to reach otherwise starting from the thermal population in the ground state. In ultracold samples this technique can be used in reverse order from intermediate to small internuclear distance to form cold ground state molecules through photoassociation.

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