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Optical Control of Population Flow between Molecular Electronic States of Different Spin Multiplicity ERGIN AHMED, XINHUA PAN, Temple University, JOHN HUENNEKENS, Lehigh University, MARJATTA LYYRA, Temple University — The adiabatic description of molecular electronic states in terms of potential energy surfaces, defined by the motion of the electrons, on which the slower nuclear motions (vibrations and rotations) occur, breaks down when relativistic effects such as the coupling between the electron spin and its orbital angular momenta (spin-orbit coupling) are taken into account. The result is that conical intersections (avoided crossings) develop between the adiabatic potential surfaces (curves) resulting in molecular states with mixed spin (multiplicity) character. Such intersections play a critical role in defining the pathways of nonadiabatic processes such as collisional quenching and intersystem crossings of excited states. In this work we demonstrate optical control of the singlet/triplet probability distribution in the outcome of a collisional process between Lithium dimers and Argon atoms. The control is achieved using the Autler-Townes effect to manipulate the spin character of a spin-orbit coupled pair of levels serving as a "gateway" between the singlet and triplet electronic state manifolds.

Ergin Ahmed
Temple University

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