

LASER-MILLIMETER-WAVE TWO-PHOTON RABI OSCILLATIONS EN ROUTE TO COHERENT POPULATION TRANSFER

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Core-nonpenetrating Rydberg states of molecules are a relatively untapped resource in molecular physics. Due to the $\ell(\ell+1)/r^2$ centrifugal barrier, the Rydberg electron in high- ℓ states is essentially decoupled from the ion-core. This decoupling leads to the system becoming atom-like, with long lifetimes, an “almost good” ℓ quantum number, and “pure-electronic” transitions that follow $\Delta J^+ = 0$ and $\Delta v^+ = 0$ propensity rules. Access to these nonpenetrating states is generally blocked by the necessity that the multistep excitation scheme traverses a “zone of death” in which nonradiative decay mechanisms are prohibitively fast.

Coherent population transfer methods, such as STImulated Raman Adiabatic Passage (STIRAP), allow population of core-nonpenetrating states without even transiently populating states in the “zone of death.” We demonstrate coherent two-photon population transfer to Rydberg states of barium atoms using a pulsed dye laser and a chirped-pulse millimeter-wave spectrometer. Numerical calculations, using a density matrix formalism, reproduce our experimental results and provide insights into the fractional population transferred, optimal experimental conditions, and possibilities for future improvements, in particular extension to full STIRAP.