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Probing the limitations of using adiabatic passage with short XUV pulses in a Raman process[1] XUAN LI, Lawrence Berkeley National Laboratory, C. WILLIAM MCCURDY, Department of Chemistry, University of California, Davis, DANIEL J. HAXTON, Lawrence Berkeley National Laboratory — With the aid of recently developed *ab initio* implementation of the multiconfiguration time-dependent Hartree Fock method, we investigate quantum control of population transfer with a Raman process between two bound electronic states via intermediate states that are doubly excited core hole states well above the ionization threshold. We treat the example of the Li atom with intermediate states of the configuration 1s2s2p. A $\pi + \pi$ pulse approach is shown to yield a population transfer efficiency of 53% at relatively low laser intensities, while, surprisingly, the Stimulated Raman Adiabatic Passage (STIRAP) approach leads to much lower transfer efficiency ($< 5\%$), and its characteristic robustness at high laser intensities disappear. Further analysis of competing photo-ionization processes reveals the mechanism behind the failure of the STIRAP approach in this case.

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