Ab initio Study of Charge Carrier Dynamics and Fragmentation of Gas-Phase Lanthanum Cyclopentadienyl Complexes

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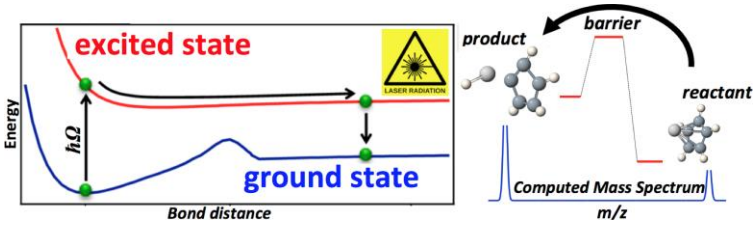
Lanthanide oxide thin films have attracted much attention due to their wide applications such as gate oxides, protective coatings, and phosphors. Lanthanide cyclopentadienyl complexes are potential precursors to fabricate lanthanide oxide thin films by laser-assisted chemical vapor deposition (LCVD). To better harness the LCVD technique, it is necessary to understand a range of photoinduced processes and dynamics of electrons and nuclei resulted in photolysis reaction of gas-phase lanthanide cyclopentadienyl complexes. In this study, photoinduced charge carrier dynamics are investigated by density matrix formalism with the combination of Redfield theory and on-the-fly coupling of electron-to-nuclear motion by the molecular dynamics trajectory.[1] It is found that electrons relaxation is much faster than holes relaxation. Time-dependent excited-state molecular dynamics (TDESMD) algorithm based on Rabi theory and trajectory surface hopping approximation is applied to the gas-phase metal-containing complexes to simulate the photolysis process.[2] The dynamical formation of multiple products such as LaC_3H_3 and LaC_5H_4 is observed in TDESMD trajectories. Simulated mass spectra are extracted from TDESMD simulations and compared to experimental photoionization time-of-flight (PI-TOF) mass spectra.[3] It is found that several features in the experimental mass spectra are reproduced by simulated one. In addition, the simulation results indicate the fragmentation patterns depend on the excitation energy and the oscillator strength of the transition.



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