

Optimal control on alkali systems

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Control of photo-induced molecular processes has attained considerable success in recent years. It became most exciting when self-learning feedback loop algorithms were employed where tailored laser pulses can be generated, which drive the induced processes at a maximum yield along desired paths. An important issue in this regard is the information coded in the optimized laser pulse shape which supplies insight about the underlying processes. Small alkali systems are suitable for this aim since they exhibit a number of bound states available for resonant transitions with weak fields which aids the theoretical description and hence the interpretation.

Closed loop optimizations on alkali dimers in a supersonic beam are first demonstrated for maximization of the ionization yield. Next, isotopomer selective optimizations are presented to examine the efficiency of the optimization procedure for the weak differences between the isotopic species. Surprisingly large enrichment factors are found and information about the dynamics on the involved vibrational states is extracted from the optimal pulse shapes, which provides a new spectroscopical approach of yielding distinct frequency pattern on fs-time scales. Both experiments are compared with optimal control calculations in order to decipher the underlying processes in detail.

A main aspect of this contribution is the development of novel optimal control methods to extract the most relevant information from the optimized laser field. One approach examines the implementation of genetic pressure within the algorithm for performing control pulse cleaning where extraneous pulse features were removed to expose the most important structures. This leads also to first investigations on multi-criteria optimizations. Another method addresses parametric optimization by introducing physically relevant pulse parameters employing a computer assisted sub pulse encoding in order to narrow the search space and to aid the interpretation. Moreover, novel pulse shaper schemes for combined phase, amplitude, and polarization pulse control were developed and applied on alkali dimers, even in a parametric encoding. The results demonstrate the perspectives of adding a new dimension by including also the polarization and hence all properties of the light field in the pulse modulation.

Currently, coherent control is applied to ultracold trapped ensembles motivated by the perspective to perform photoassociation and photostabilization of alkali systems. First results are received regarding optimized multi-photon excitation to molecular ions and pump-probe experiments exposing signal oscillations. They provide indications for photoassociation and open the perspective for transitions to lower vibrational levels in the electronic ground state, which would be a first step to an internally cold molecular Bose Einstein condensate.