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Local control of nonadiabatic photodissociation dynamics using Møller operators

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Synopsis We implement a local control strategy based on the use of Møller operators and use it to control the photodissociation of diatomic molecules in the presence of nonadiabatic interactions.

Various schemes have been proposed to design laser fields for the control of chemical reactions. In local control theory, the field is tailored so as to ensure that the expectation value of an observable increases or decreases, based on the instantaneous dynamics of the system. This observable must commute with the Hamiltonian of the isolated system [1]. In the case of a photodissociation reaction in presence of nonadiabatic couplings, it thus cannot be chosen as the population of a diabatic or adiabatic electronic state corresponding to a specific kind of fragment.

We overcome this difficulty by defining our observable as a superposition of diffusion eigenstates correlated to the fragments we want to produce. We implement a local control strategy based on the Møller operators, in which the control field is obtained through the Fourier Transform of the wavepackets propagated from the interaction zone to the asymptotic (dissociation) region for each time step of the control.

This method is applied to the control of the photodissociation of the HeH^+ molecular cation to favour its dissociation into a specific couple of fragments : either He(1s, nl) + H^+ or He^+(1s) + H(nl). Control fields are obtained for the photodissociation of HeH^+ in its fundamental $^1\Sigma^+$ state into its dissociation channels of the same symmetry, using a single parallel control field, and its dissociation in the Π channels by using a perpendicular and a parallel control field successively [2]. The Fourier transform of the laser fields is relatively simple to interpret in terms of the photodissociation cross sections.

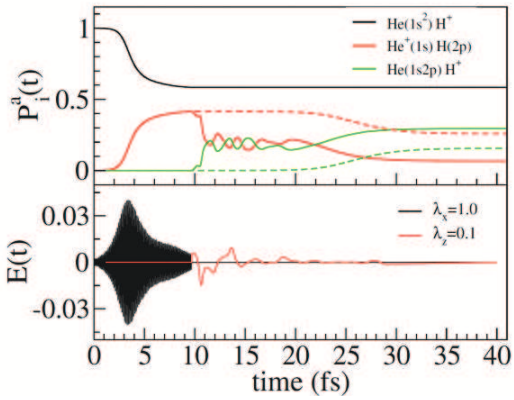

Figure 1. Upper part : Adiabatic electronic state populations with (full lines) and without laser pulse (dashed lines) for the optimization of H(2p)
Lower part : Parallel and perpendicular laser pulses for this case (in 1 GV/m)

We also discuss the application of this method to the CO^{2+} dication, a species of particular interest experimentally as it can be produced in the lowest vibrational level of its fundamental state almost exclusively.

References

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