# Controlling the Vibrational and Dissociation Dynamics of the Hydrogen Molecular Ion with Intense Infrared Laser Pulses

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**Synopsis** Using intense infrared laser pulses, we provide theoretical results for controlling the bound vibrational dynamics, the dissociation process and ionization of the hydrogen molecular ions $H_2^+$ and $D_2^+$.

The time–resolved imaging of the vibrational dynamics of the hydrogenic molecular ions $H_2^+$ and $D_2^+$ has seen great interest over the last decade, and became directly observable in the time domain using ultra–short laser pulses [1]. In particular, the time–dependent kinetic energy release of time–series pump–probe studies has been utilized to reveal intrinsic characteristics of the vibrating wave packet, such as laser induced potential curves, and the vibrational state distribution using internuclear–distant dependent Fourier analysis [2]. Moreover, the addition of one or more precisely timed ultra–short control laser pulses has been shown to be a suitable way to selectively manipulate the vibrational state distribution of the molecular ion and to direct the dissociation pathway [3].

In addition to our previous studies [2, 3] which were limited to the motion of the nuclear wave packet on the coupled $1s\sigma_g^+$ and $1s\sigma_u^+$ adiabatic potential curves of the molecular ion, we now explicitly include all electronic degrees of freedom to account for non Born–Oppenheimer (BO) effects and the electronic interaction with the intense laser field. We restrict the nuclear motion to the internuclear separation and assume alignment of the molecule along the electric field vector of the laser pulse. Taking advantage of the cylindrical symmetry for the electronic motion, we solve the resulting three–dimensional Schrödinger equation on a numerical lattice using the split–operator Crank – Nicolson scheme for the time propagation.

By projecting the propagated wave function $\psi(R, \vec{r}, t)$ onto the lowest few eigenvectors $\psi_\nu(R, \vec{r})$ of the full Hamiltonian after the external interaction has stopped $t > t_0$, we obtain the probabilities

$$P_\nu(t) = \left| \langle \psi_\nu(R, \vec{r}) | \psi(R, \vec{r}, t) \rangle \right|^2,$$

of finding the system in the $\nu$-th bound molecular state (i.e. a non-BO state, described by the combined nuclear and electronic motion). The Fourier transform of the autocorrelation function

$$a(t) = \left| \langle \psi(R, \vec{r}, t_0) | \psi(R, \vec{r}, t) \rangle \right|^2,$$

for $t > t_0$ provides the kinetic energy spectrum of the dissociating nuclei and the ejected photo-electrons [4].

We show results for a series of infrared (800 nm), intense ($\sim 10^{14}$ W/cm$^2$), femtosecond laser pulses. The first pulse launches a vibrational wave packet from the neutral molecule, and the precise timing of the subsequent control laser pulses allows for the directed manipulation of the bound vibrational state composition and the dissociation dynamics of the ion. We will address the influence of the laser parameters (wavelength, intensity, pulse length) on the extent of controlling the vibrational dynamics of the ion, and compare with previous model calculations [3].

## References

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