Multiconfiguration method applied to electron-nuclear dynamics of H_2

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The rapid technological progress in ultrashort intense light sources has triggered various research activities in the field of attosecond science, with the ultimate goal to directly measure and control electron and nuclear motion in atoms and molecules. From the theoretical side, ab initio simulations of the electronic and nuclear dynamics in atoms and molecules remain a challenge[1]. A fully general time-dependent multiconfiguration self-consistent-field (TD-MCSCF) method has been developed[2], which can be used for the investigation of a system comprising arbitrarily different kinds and numbers of particles, with all the constituent particles are treated on an equal footing.

In this contribution, we apply this method to the simulation of strong field-induced ionization and dissociation of H_2. In our approach, the total wave function is written as

$$\Psi(R, \boldsymbol{r}_1, \boldsymbol{r}_2, t) = \sum_I^M \sum_J^N C_{IJ}(t)\chi_I(R,t)\Phi_J(\boldsymbol{r},t), \quad (1)$$

where $C_{IJ}(t)$ is the configuration-interaction coefficients, $\chi_I(R,t)$ the protonic orbitals and $\Phi_J(\boldsymbol{r},t)$ the electronic Slater determinants with $\boldsymbol{r} = (\rho, z)$. In our calculation, both the molecular axis and the electric field vector of the laser pulse are assumed to be parallel to the z-axis. Figure 1 shows the integrated protonic density with different number of configurations. We can see that with only one configuration ($M = 1, N = 1$), only bound vibrational component of the protonic wave packet is reproduced. When the number of configuration is increased ($M = 4, N = 16$), a bifurcation of the vibrational wave packet into a dissociating component and a vibrational excitation component occurs, which implies the present method is well suited to describe not only the laser-induced ionization but also laser-induced dissociation and vibrational excitation.

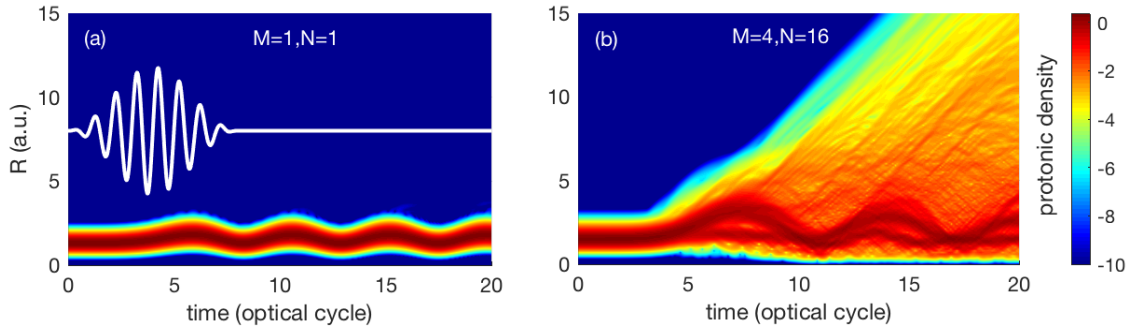

Figure 1: Protonic density for H_2 exposed to an intense laser pulse with (a) $M = 1, N = 1$ and (b) $M = 4, N = 16$. Electric field of the laser pulse is indicated by a white curve in (a).

References

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