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Ionization of neutral D_2 molecules by a short and intense pump laser pulse may create a vibrational wave packet on the lowest $(1s\sigma_a^+)$ adiabatic potential curve of the D_2^+ molecular ion. We investigate the possibility of manipulating the bound motion, dissociation, and vibrationalstate composition of such nuclear wave packets with ultra-short (6 fs) intense $(1 \times 10^{14} \text{ W/cm}^2)$ near infrared (800 nm) control laser pulses [1]. The influence of such control pulses has been shown to lead to vibrational cooling and to modify dissociation yields [2]. We show numerically that a single control pulse with an appropriate time delay can quench the vibrational state distribution of the nuclear wave packet by increasing the contribution of a selected stationary vibrational state of D_2^+ to more than 50%. We also demonstrate that a second control pulse with a carefully adjusted delay can further squeeze the vibrational state distribution, thereby suggesting a multi-pulse control protocol for generating almost stationary excited nuclear wave functions. The quality of this Raman–control mechanism can be tested experimentally by fragmenting the molecular ion with an intense probe pulse and by identifying the nodal structure of the surviving vibrational state in the kinetic energy release spectrum of the molecular fragments [3, 4].

The probability $P_{\nu}(t)$ for finding the system at time t > 0 in the ν -th vibrational state is the quantum mechanical overlap of the calculated bound wave function $\chi_g(t)$ with the known eigenfunctions χ_{ν} of the $1s\sigma_g^+$ curve,

$$P_{\nu}(t) = \frac{|\langle \chi_{\nu} | \chi_g(t) \rangle|^2}{\langle \chi_g(t) | \chi_g(t) \rangle}.$$

For our specific set of laser parameters (see Figure), we find that a second control pulse vibrationally cools the wave packet due to Raman transitions, mainly from the $\nu = 3$ to the $\nu = 2$ state, with over 97% of the molecular ions remaining bound and 78% in the $\nu = 2$ vibrational state. In addition we show that the small discrepancy between the measured [3] and calculated [4] nuclear revival times is due to vibrational Raman-transitions, in the tail of the pump pulse.



Fig. 1. Time evolution of the nuclear wave function probability density (top) and the few lowest vibrational states (bottom, logarithmic scale) for two 6 fs, 1×10^{14} W/cm² control pulses with delay times of $\tau_1 = 70.7$ fs and $\tau_2 = 136.8$ fs relative to the creation of the wave packet at $\tau = 0$. The superimposed curve in the top graph shows the center motion $\langle R \rangle$ of the bound wave packet.

References

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