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**Breaking the O–H bond**

IR pulse parameters:  $E_{0,\text{IR}} = 3.2$  GV/m,  $\omega_{\text{IR}} = 1565$   $\text{cm}^{-1}$ ,  $\varphi_{\text{IR}} = 0$ ,  $t_{0,\text{IR}} = 0$  fs, and  $t_{p,\text{IR}} = 50$  fs.

UV pulse parameters:  $E_{0,\text{UV}} = 5.0$  GV/m,  $\omega_{\text{UV}} = 28\,228$   $\text{cm}^{-1}$ ,  $\varphi_{\text{UV}} = 0$ ,  $t_{0,\text{UV}} = 19$  fs, and  $t_{p,\text{UV}} = 5$  fs.

**Breaking the H–F bond**

IR pulse parameters:  $E_{0,\text{IR}} = 5.0$  GV/m,  $\omega_{\text{IR}} = 1565$   $\text{cm}^{-1}$ ,  $\varphi_{\text{IR}} = 0$ ,  $t_{0,\text{IR}} = 0$  fs,  $t_{p,\text{IR}} = 50$  fs.

UV pulse parameters:  $E_{0,\text{UV}} = 8.0$  GV/m,  $\omega_{\text{UV}} = 52\,423$   $\text{cm}^{-1}$ ,  $\varphi_{\text{UV}} = 0$ ,  $t_{0,\text{UV}} = 29$  fs and  $t_{p,\text{UV}} = 5$  fs. . . . . 138

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**Breaking the O–H bond:**

(a) Few-cycle IR + UV laser pulses achieve maximum O + HF fragmentation.

IR pulse parameters:  $E_{0,\text{IR}} = 3.2$  GV/m,  $\omega_{\text{IR}} = 1565$   $\text{cm}^{-1}$ ,  $\varphi_{\text{IR}} = 0$ ,  $t_{0,\text{IR}} = 0$  fs, and  $t_{p,\text{IR}} = 50$  fs.

UV pulse parameters:  $E_{0,\text{UV}} = 5.0$  GV/m,  $\omega_{\text{UV}} = 28\,228$   $\text{cm}^{-1}$ ,  $\varphi_{\text{UV}} = 0$ ,  $t_{0,\text{UV}} = 19$  fs, and  $t_{p,\text{UV}} = 5$  fs. (b) Time evolution of the branching ratio of the O + HF products (solid) versus OH + F (dotted).

**Breaking the H–F bond:**

(c) Few-cycle IR + UV laser pulses achieve maximum OH + F fragmentation.

IR pulse parameters:  $E_{0,\text{IR}} = 5.0$  GV/m,  $\omega_{\text{IR}} = 1565$   $\text{cm}^{-1}$ ,  $\varphi_{\text{IR}} = 0$ ,  $t_{0,\text{IR}} = 0$  fs,  $t_{p,\text{IR}} = 50$  fs.

UV pulse parameters:  $E_{0,\text{UV}} = 8.0$  GV/m,  $\omega_{\text{UV}} = 52\,423$   $\text{cm}^{-1}$ ,  $\varphi_{\text{UV}} = 0$ ,  $t_{0,\text{UV}} = 29$  fs, and  $t_{p,\text{UV}} = 5$  fs. (d) Time evolution of the branching ratio of the O + HF products (solid) versus OH + F (dotted). . . . . 139



- 4.25 Generation of a vibrational wave packet from the few-cycle IR pulse shown in Figure 4.24(a). IR pulse parameters are:  $E_{0,\text{IR}} = 3.2$  GV/m,  $\omega_{\text{IR}} = 1565$   $\text{cm}^{-1}$ ,  $\varphi_{\text{IR}} = 0$ ,  $t_{0,\text{IR}} = 0$  fs, and  $t_{p,\text{IR}} = 50$  fs. Dominant contributions are from vibrational eigenstates  $v_{00}$ ,  $v_{01}$  (asymmetric stretch),  $v_{02}$  (first excited asymmetric stretch), and  $v_{11}$  (symmetric + asymmetric). Depletion of population in the anion ( $\text{OHF}^-$ ) ground state begins shortly after  $t = 19$  fs, when the UV pulse is fired. The UV pulse parameters are  $E_{0,\text{UV}} = 5.0$  GV/m,  $\omega_{\text{UV}} = 28\,228$   $\text{cm}^{-1}$ ,  $\varphi_{\text{UV}} = 0$ ,  $t_{0,\text{UV}} = 19$  fs, and  $t_{p,\text{UV}} = 5$  fs. After vertical excitation of population to  $V_n$ , anion population (norm) drops to  $\sim 0.98$ . . . . 140
- 4.26 Schematic representation of the bending vibration of a triatomic molecule. . . . . 141
- 4.27 The electric field profile of the laser pulses consisting of very few optical cycles depends on the phase  $\varphi$  of the carrier frequency with respect to the pulse's envelope; the maximum of the electric field points in opposite directions at  $t = 10$  fs for  $\varphi = 0$  and  $\varphi = \pi$ . . . . . 144
- A.1 A linear triatomic molecule ABC with bond lengths  $R_{\text{AB}}$  and  $R_{\text{BC}}$  and masses  $m_{\text{A}}$ ,  $m_{\text{B}}$ , and  $m_{\text{C}}$ , rotates around an axis  $Z_{c.o.m}$  which passes through the center of mass of the molecule. . . . . 156

