Appendix G

Derivation of the time-dependent dissociation probability

The dissociation probability, DP, is derived by employing the usual flux expressions as calculated at \( R = R_0 \) where \( R_0 \) is a point at the asymptotic region, in front of the region where the absorbing potential \( U(R) \neq 0 \) (namely, the interval \( R > R_0 \)). The calculation yields, for a given electron-photon state \( n \) and initial state \( \psi_i \), the following DP, \( P_n(t|\psi_i) \):

\[
P_n(R_0, t|\psi_i) = \frac{\hbar}{M} \text{Im} \int_0^\infty dt' \left\{ \psi_n^*(R, t'|\psi_i) \frac{\partial \psi_n(R, t'|\psi_i)}{\partial R} \right\}_{R=R_0}.
\]  

(G.1)

Next, in order to get the total DP, \( P_d(t|\psi_i) \), at time \( t \) we sum over all the \( J \) electron-photon states

\[
P_d(t|\psi_i) = \sum_{n=0}^{J-1} P_n(t|\psi_i). 
\]  

(G.2)
A similar expression holds for the final DP calculated at time $t = T$ where $T > \tau_p$ (it is recalled that $\tau_p$ is the duration time of the electric pulse).

Finally, to calculate the DP for a given population of vibrational states, e.g., a Franck-Condon (FC) population of the original ion, $H_2^+$, we perform the following summation:

$$\hat{P}_d(t) = \sum_{\nu_i} p_{\nu_i} P_d(t | \nu_i)$$

where $p_{\nu_i}$ is the (FC) population for the $\nu_i$ - state.