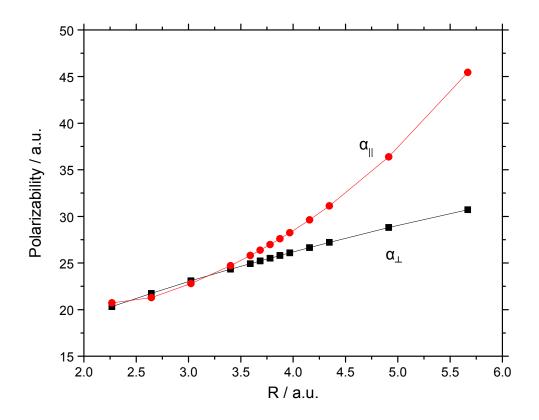
## Quantum Optimal Control of Rovibrational Excitations of a Diatomic Alkali Halide: One-Photon vs. Two-Photon Processes

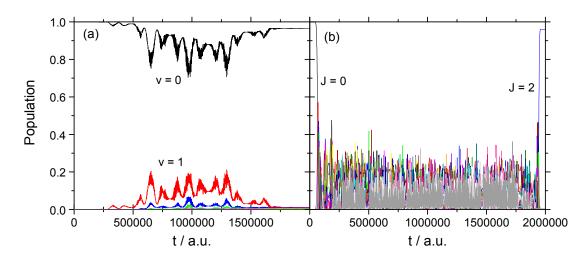
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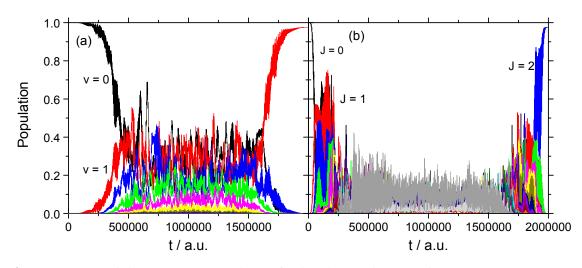
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**Figure 1.** Components of polarizability of LiCl parallel ( $\alpha_1$ ) and perpendicular ( $\alpha_{\perp}$ ) to the molecular axis, calculated at the RMP2/aug-cc-pVTZ level of theory.



**Figure 2.** Temporal changes in state populations for the rotational excitation,  $(v = 0, J = 0) \rightarrow (v = 0, J = 2)$ , caused by the optimal field shown in Figure 5, in the strong-field regime: (**a**) vibrational states; (**b**) rotational states.



**Figure S3**. Temporal changes in state populations for the vibrational-rotational excitation, (v = 0, J = 0)  $\rightarrow$  (v = 1, J = 2), caused by the optimal field shown in Figure 10, in the strong-field regime: (**a**) vibrational states; (**b**) rotational states.