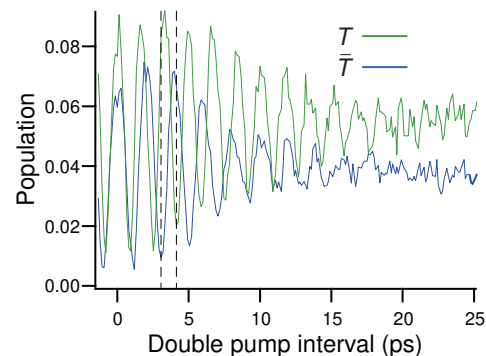


ULTRAFAST MODE-SELECTIVE POPULATION CONTROL OF LARGE AMPLITUDE VIBRATION IN DIPHENYLMETHANE

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Large-amplitude motion (LAM) may induce a substantial conformational change in molecules, which is deeply relevant to molecular functionality e.g., in biomolecules. However, the creation and manipulation of vibrational wave packets pertinent to LAM in isolated molecular systems have been rarely realized, and most studies on molecules in the electronic ground states deal with one-dimensional LAM^{ab}. Here we study diphenylmethane (DPM), which has two degrees of freedom for LAM, i.e., symmetric and anti-symmetric torsional vibration, T (20 cm^{-1}) and \bar{T} (16 cm^{-1}). Adiabatically cooled DPM was irradiated by a pair of femtosecond laser pulses (pumps) to excite modes T and \bar{T} through impulsive stimulated Raman scattering. Then, resonance two-photon ionization (R2PI) spectra were obtained by nanosecond UV pulses ($\sim 268\text{ nm}$, probe). Populations in $\nu = 1$ of both vibrational modes were evaluated from the integrated intensities of hot bands appearing in the R2PI spectra. The experimental results show that the populations in $\nu = 1$ of mode T and \bar{T} oscillate against the double pump interval as a result of the wave-packet interference (Figure). By utilizing the difference in these oscillation periods, mode selective excitation was realized (dashed lines). We also conducted semiclassical calculations, where molecular vibration is treated quantum mechanically while molecular rotation is simulated by classical trajectory calculation, to show a good match-up with the observed time-evolution of populations.



^aC. B. Madsen, et al., J. Chem. Phys. 130, 234310 (2009).

^bM. Nikaïdo, et al., 2021 International Symposium on Molecular Spectroscopy, RH06 (2021).