

STATE-DISTRIBUTION CONTROL OF LARGE AMPLITUDE VIBRATION IN SUBSTITUTED BIPHENYLS WITH INTENSE LASER PULSES

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With recent advances in ultrashort laser technology, many studies using intense nonresonant laser fields have been conducted to control vibrational or rotational wave packets. In particular, control of large-amplitude low-frequency vibration is important because such vibration may cause a substantial change in molecular structure. For instance, torsional motion of biphenyl has deserved much attention since chirality and physical properties of the molecule depend on its torsional angle. In this study, we coherently excite torsional vibration of substituted biphenyl derivatives by the interaction with ultrashort laser fields and the resultant vibrational excitation is monitored by recording resonant two-photon ionization (R2PI) spectrum. We further adopt double-pulse excitation to control vibrational state distribution via wave-packet interference.

Adiabatically cooled molecular sample of 2-fluorobiphenyl is irradiated by the fundamental output from a fs Ti:Sapphire laser. This pump pulse induces vibrational excitation through impulsive Raman process. With an appropriate delay after the pump-pulse irradiation, the S_1-S_0 excitation spectrum of the molecules is recorded via R2PI with the doubled output of a nanosecond dye laser (~ 280 nm). A progression with almost constant spacings appears in the R2PI spectrum without the pump pulse. It has been assigned to that of the torsional mode from vibrational ground state, *i.e.*, $v = 0$ (v being the quantum number of the torsional mode in the electronic ground state).^a When the pump pulse is introduced, the intensity of each band is reduced and new bands appear. These bands are assigned to the progression from $v = 1$. These observations indicate that impulsive Raman excitation of torsional vibration is realized. We also conduct a double-pump pulse experiment, where a pair of pulses are implemented for excitation. In this experiment, we succeeded in controlling the state distribution of torsional vibration by adjusting the time delay between the two pump pulses.

^aH. S. Im and E. R. Bernstein, J. Chem. Phys. **88**, 7337 (1988).