Contribution of resonance excitation on ionization of OCS molecules in strong laser field

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Synopsis The resonant enhanced multiphoton ionization (REMPI) of OCS molecules has been studied by tracing the electron energy shift with the laser intensity dependent photoelectron energy spectra, we find two excited states ($^1\Delta$ and $^1\Pi$) have been involved in the ionization of OCS at 800 nm and the REMPI through a certain excited state can be selectively controlled by varying the laser intensity. Furthermore, the REMPI of OCS through 4sσ Rydberg states at 400 nm laser fields has also been identified by angular distribution of electron.

Strong field laser interaction with molecules has attracted extensive attention in recent years. Abundant information on electronic and nuclear dynamics of molecules is encoded in the photoelectron moment distribution, including the contribution of excited states, multi-orbital effect and electron rescattering in photoelectron holograph. Several experimental observations on ionization of OCS molecules show obvious different angular distribution from theoretical calculations. The observed maximum ionization occurs at 90 degrees (i.e., the polarization of ionization laser perpendicular to the molecular axis) [1] that cannot be reproduced by the calculations such as MO-ADK, adiabatic SFA, TD-DFT, and WAFT. In general the influence of excited states in the process has been proposed for interpreting this discrepancy.

We experimentally investigate ionization of OCS molecules in strong laser fields using velocity-map imaging. A 50 *fs*, 800 nm linearly polarized laser is used to ionize OCS molecules from a pulsed beam. The laser pulses centered at 400 nm are generated by passing an 800 nm linear polarized pulse through a BBO crystal. The obtained electron energy spectra under different laser intensities (10~100 TW/cm^2) are shown in Figure 1. Several peaks generated from above-threshold ionization (ATI), is signed by n=9-13, which energy decreasing as the laser intensity increasing. Three REMPI peaks with the energies of 0.7, 1.2 and 2.2 eV are observed, which energy is not change with the laser intensity. The electrons from Peak1 and Peak2 appear at different laser intensity are generated from resonant ionization from different electronic excited states (Freeman resonance [2]). The Peak1 appeared at laser intensity between 55 and 82 TW/cm^2 is generated from ground state $X^1\Sigma^+$ absorbing 6 photons to excited state $^1\Delta$, and then ionized by absorbing another 4 photons. While the peak 2, appeared above 80 TW/cm^2, can be assigned to transit from the ground state $X^1\Sigma^+$ absorbing 8 photons to excited state $^1\Pi$, and then ionized by absorbing another 3 photons. Peak 3 is the five-photon ionization through the $^1\Delta$ state. Furthermore, we also find Rydberg state 4sσ is involved in REMPI of OCS molecules at 400 nm at laser intensity between 3.5 and 34 TW/cm^2. Therefore, the present results confirm the resonance excitation contribution on strong-field ionization of OCS molecules.

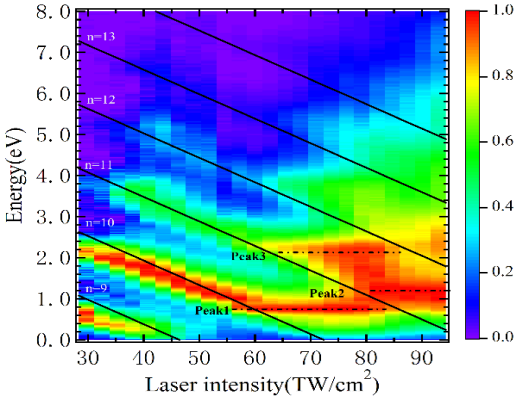

Figure 1. Laser intensity dependent photoelectron energy spectra of OCS molecules obtained from 800 nm.

References

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