Strong-field ionization and dissociation of linear triatomic molecules irradiated by 800-nm and 400nm femtosecond laser fields

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Synopsis: Ionization of molecules in strong femtosecond laser fields is a key process for understanding a varietyof strong-field molecular physical phenomena. In this study, we performed an experimental study on strong-field ionization and dissociation of three linear triatomic molecules, CO_2, CS_2, and OCS, using time-of-flight mass spectrometer. We measured the yield of different parent ions and various fragmentions as a function of laser intensity in the range of 2.0×10^{13}W/cm^2 to 8.0×10^{14} W/cm^2, in both 800-nm and 400-nm strong laser fields. The mechanism of strong-field single and double ionization, as well as fragmentation, was discussed based on the experimental results. In addition, the angular distributions of fragment ions were obtained. The results show the anisotropic distributions in the formation of highly charged atomic fragment ions, which may be attributed to the alignment of the molecules in strong laser fields before dissociation.

Strong-field ionization of molecules is believed to be the fundamental process leading to a variety of strong-field physical phenomena, such as non-sequential double ionization (NSDI), high-order above threshold ionization, high-order harmonic generation, and laser-induced electron diffraction. Understanding the strong-field molecular ionization process is also of vital importance in application of tomographic imaging of molecular orbitals, molecular coherent control, and attosecond pulse generation. Comparing to atoms, strong-field ionization of molecules exhibit novel phenomena due to their diverse structure and additional nuclear degree-of-freedom. The mechanism of molecular dissociative ionization in strong laser fields as well as the competition between different channels is still a challenging task, especially for polyatomic molecules.

We have measured and analyzed the TOF mass spectra of three triatomic molecules, CO_2, CS_2, and OCS, after strong-field ionization by IR and UV femtosecond laser pulses that were linearly polarized along the TOF axis. For each of the molecules, various kinds of ions, including single and double charged parent ions, along with various fragment ions from dissociative ionization and Coulomb explosion, have been observed in both 800nm and 400nm laser fields. However, the relative intensities are quite different in 800-nm laser fields comparing to those in 400-nm laser fields, indicating that the channels involved in tunneling ionization and multiphoton ionization could be different [1].

We further investigated the dependence of the ion yields on laser intensity and polarization. In linearly polarized 800-nm laser fields, clear “knee” structure, which is the signature of NSDI in atomic strong-field physics, has been observed for CO_2 and CS_2 molecules. In addition, the ion yields at the laser intensity at “knee” show strongly dependence on the laser ellipticity [2][3]. These observations demonstrate that the NSDI processes in strong-field ionization of triatomic molecules. With a joined study using classical ensemble calculations, we indicated that, similar to that of atoms, NSDI of molecules is produced via laser-driven electron recollision with the ion core and presents electron-electron correlations in the process. Analysis indicates that both mechanisms in atomic strong-field NSDI, i.e., recollision impact ionization and recollision excitation with subsequent ionization, may also be contributed to NSDI of molecules [3]. Our study should shed some light on the mechanisms of strong-field ionization and dissociation of molecules.

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References

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