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Ultrafast dynamics of methyl iodide with XUV Free Electron Laser

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The development of the free electron laser (FEL) gives us a new way to investigate the interplay between electrons and nuclear motions with intense short wavelength pulses [1]. The pulse duration of current FELs varies from a few femtosecond of hard X-rays to around 100 femtoseconds of extreme ultraviolet (XUV) radiation. Recent works showed that the multiphoton ionization process allows to investigate nuclear dynamics with hard X-rays, by creating highly multiply charged states [e.g 2]. We extend here this approach by using multiphoton ionization at a longer wavelength FEL for studying methyl iodide fragmentation.

The experiment was carried out by using a moderate intensity of XUV FEL source at 90 eV in FLASH BL1 with a reaction microscope (REMI), providing a complete kinematical information for different charged carbon-iodine pairs up to C³⁺ and I⁷⁺. To understand our observation, we develop a classical wave-packet dynamics model based on Coulomb explosion, including sequential photoionization, Auger decay and charge transfer. The charge transfer mechanism is described with the classical over-barrier (COB) model [3,4] and is prohibited when the carbon-iodine bond becomes over-elongated. From this model, we can derive measured physical quantities from the experiment.

The agreement between simulation and experiments is in general very good despite the complex ultrafast dynamics. Two important factors to control the dissociation dynamics are found. First, the charge delocalization in the methyl group on hydrogen atoms influences the Coulomb repulsion force between carbon, C^{p+}, and iodine, I^{q+}, and thereby the momentum correlation between measured ions. Second, the Auger decay time of ionic iodine strongly affects the branching ratio of higher carbon charge states. Since both factors play a role in the earlier stage of the dissociation process, < 10 fs, it suggests that the initial dissociation dynamics of the methyl iodide is driven to a great extent by the electronic dynamics.

References

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