Pulse width effect on the ionization and dissociation of Methyl Iodide in the intense femtosecond laser field

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The ionization rate of the diatomic molecule in an intense laser field has been observed to be enhanced for inter-nuclear separation greater than its equilibrium bond length ’RE’. This specific inter-nuclear separation is known as ’critical’ bond length ’R_c’, where the HOMO (Highest occupied molecular orbital) and LUMO (Lowest unoccupied molecular orbital) starts to overlap in the presence of the laser field [1]. Previous results on diatomic molecules showed that the typical ’R_c’ could be two to three times greater than the ’RE’. Several experiments have been performed on diatomic molecules to understand the ionization mechanism and the dissociation dynamics by changing various parameters such as intensity, pulse width of the laser field. It has been observed that enhanced ionization dominates for larger pulse duration (100fs) and suppressed for few-cycle pulses (~10fs), as the molecular ion does not have sufficient time to reach ’R_c’ within the duration of the pulse. To follow the appearance of the enhanced ionization and investigate its molecular origin, it would be better to vary the pulse duration from few-cycle to multi-cycle regime. Few experiments of this kind have been done for diatomic molecules and small polyatomic molecules having simple geometry, such as CO_2 [1], which has a linear molecular geometry. For polyatomic molecules with critical geometry the ionization mechanism and dissociation pathways remain matter of interest.

In the current work we have irradiated intense (~10^{12} W/cm2) laser pulse of 800nm on CH3I molecule, which has a tetrahedral geometry. We have varied the pulse width from 25fs to 1200fs to understand the ionization mechanism and the dissociation pathways in a systematic manner. Being a large molecule, the vibrational time period of the C-I bond is quite large (~100fs), so we can easily consider the C-I bond to be frozen for the pulse duration of 25fs. We have used velocity map imaging techniques [3] to collect all the fragmented ions. Here we have closely looked into the fragments originating from the ’Coulomb Explosion’ pathways, such as from $CH_3I^{++} \rightarrow CH_3^+ + I^+$ and $CH_3I^{+++} \rightarrow CH_3^+ + I^{++}$. Coulomb explosion in molecules is extensively studied to understand different dissociation pathways and to reconstruct the molecular structure at the moment of the explosion. The measured kinetic energy of the fragmented ions originating from Coulomb explosion processes has been observed to decrease with increasing pulse duration, which is a clear indication of increasing bond length. Molecules in laser field undergo rotation and align itself along the laser polarization axis, it is known as dynamical alignment. The observed angular distribution of the fragmented ions confirms the ’dynamical alignment’ and with increasing pulse width molecules become more aligned. Interestingly, we have also seen C^+, CH^+ and CH_2^+ fragments in coincidence with I^+ and I^{++}, and the yield of C^+, CH^+ and CH_2^+ fragments increase with increasing pulse duration. From the kinetic energy distribution of these fragments, we can confirm they are coming from further dissociation of CH^+.

References
[1] Bocharova. Irina, Karimi. Reza, Penka. Emmanuel F.,Brichta. Jean-Paul, Lassonde. Philippe, Fu. Xi- quan, Kieffer. Jean-Claude and Bandrauk, Andre´ D. and Litvinyuk. Igor, Sanderson, Joseph, Le´gare´, Franc¸ois, *Phys. Rev. Lett.* **107**,6, 063201 (2011).
[2] T Zuo, A D Bandrauk, *Phys. Rev. A* **52**, 4, R2511–R2514 (1995).

[3] R Gopal, A Sen and S R Sahu, A S Venkatachalam, M Anand, V Sharma, *Review of Scientific Instruments* **89**, 8, 086107 (2018).