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Ultrafast F_2^- photodissociation by intense laser pulses: a time-resolved fragment imaging study

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Abstract: We present time-resolved coincidence imaging of F_2^- photodissociation by 400nm and intense 800nm ultrafast pulses. Coincidence fragment imaging reveals parallel and perpendicular single photon dissociation on $^2\Sigma_g^+$ and $^2\pi_g$ states, and additional intense-field dissociation features.

1 Introduction

Compression of light into femtosecond laser pulses towards time-resolved examination of ultrafast phenomena allows achieving high field intensities at the peak of the pulse. High peak intensities facilitate a broad range of non-linear phenomena like above threshold ionization, multiphoton ionisation, bond softening, Coulomb explosion etc.[1-4] Intense field interaction with neutral systems has been studied extensively, leading to important developments such as high order harmonic generation of attosecond pulses, along with the understanding of non-sequential electron dynamics leading to efficient double ionization.[3] However, despite the noteworthy implementation of femtosecond pulses in time-resolved experiments initiated from anionic precursors,[4,5] much less is known about the interaction of intense laser pulses with negatively charged systems. Negatively charged systems are intrinsically different from the neutral and cationic systems due to the absence of an attractive Coulomb potential in the rescattering process and expected to exhibit new mechanisms in intense field interactions.[6–8] In our laboratory, we explore intense laser pulse interactions with atomic, molecular and cluster anions. Our previous investigations provide evidence for an efficient non-sequential mechanism, which is not based on the rescattering dynamics that dominates intense field interactions with neutral systems.[9–13] In a recent study of the relatively simple F_2^- system, double photodetachment and Coulomb explosion were shown to successfully compete with photodissociation on low lying dissociative states.[12] In this paper, we focus on the linear versus the non-linear effects observed in the ultrafast photodissociation of F_2^-.

2 Experimental Method

The experimental setup has been recently described in detail.[13] A dedicated fast beam fragment imaging spectrometer allows simultaneous detection of all the possible anionic,

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cationic, as well as neutral fragments resulting from interaction of ultrafast intense laser pulses with mass selected atomic, molecular and cluster anion systems.[11,13] 3D coincidence imaging of all atomic and molecular fragments allows disentangling of the competing mechanism for each molecular dissociation event at a time. The F_2^- anions are prepared in an Even-Lavie cold ion source, accelerated and mass selected before reaching a dedicated photofragment spectrometer. In the photofragment spectrometer entrance side, the mass selected parent anions are further accelerated towards a field-free laser-anion interaction region. While parent anions and anionic fragments are decelerated as they exit the spectrometer, cationic products are accelerated, and neutral fragments maintain their velocity. Thus, the time of flight to a time and position sensitive detector clearly distinguishes product charge over mass ratios. Furthermore, fragment recoil in time as well as position allows 3D reconstruction of the dissociation velocities and removal of background processes by applying center of mass considerations.

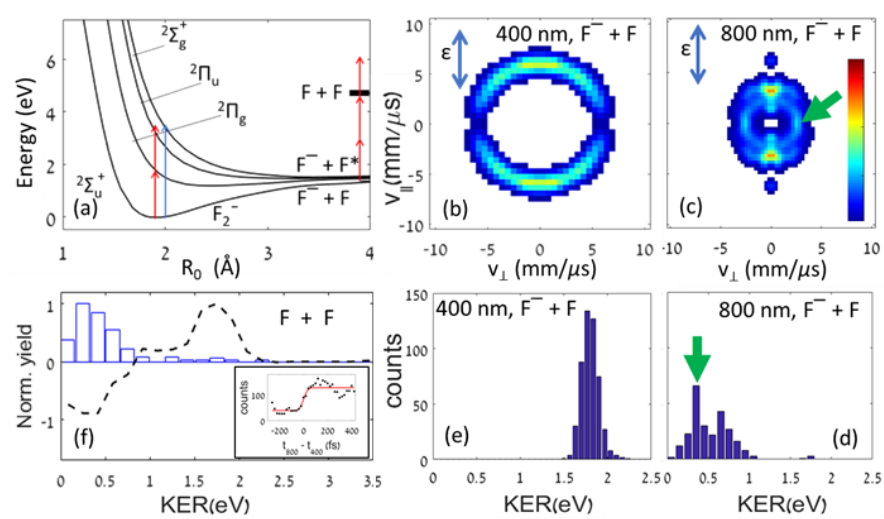

Fig. 1. a) Potential curves of the F_2^- anion, 800nm and 400nm photon energies are indicated by red and blue arrows. b, c) show angularly resolved dissociation velocity distribution between the F^- and F fragments for 400nm and intense (10^{14}-10^{15} W/cm^2) 800nm pulses, respectively. d), e) show the corresponding KER distributions. f) shows the measured KER between two neutral F atoms, bar chart represents KER due to intense 800nm alone, while the dashed line shows the difference KER spectrum due to a 400nm pump followed by 800nm probe pulse. The ultrafast time (in fs) onset of the 1.8eV KER peak is shown in the figure inset.

3 Result and Discussion

As can be seen in fig. 1a, photodissociation from the F_2^- ground state with 400nm photons is expected proceed on the $^2\Sigma_g^+$ state, while photodissociation with 800nm is most likely on the $^2\pi_g$ state. Dissociation on the $^2\pi_u$ state is forbidden for single photon transitions from the $^2\Sigma_u^+$ ground state. Fig. 1b shows the velocity distribution released between the F^- and F fragments, following photodissociation with 400nm. As can expected, the $^2\Sigma_u^+$- $^2\Sigma_g^+$ transition occurs when the molecular axis is parallel with respect to the laser polarisation. Fig. 1e shows the corresponding kinetic energy release (KER), centred at 1.794(5) eV. Considering energy conservation, as well as the 50meV spin-orbit splitting in the neutral atom, a F_2^- the measured KER indicates a dissociation energy of 1.256(5) eV, in agreement with previous theoretical and indirect experimental estimates.[14] Fig. 1c shows the relative velocity distribution released between the F^- and F products with intense 800nm pulses.

The green arrow indicates the expected perpendicular contribution from the $^2\Sigma_u^+$- $^2\Pi_g$ transition with the expected ~0.32 eV KER, as seen also in the KER distribution in fig. 1d. Also observed are additional components, which are strongly peaked at the parallel orientation with respect to the laser polarization. The ~1.8 eV KER events can be readily accounted for by considering two photons absorption onto the forbidden $^2\pi_u$ state. However, the broad feature at ~0.7eV KER, which is characterized by a structured angular dependence, parallel to laser polarization, must arise from a different non-linear mechanism that allows the dissociating molecule to gain additional energy from the intense laser field. Further theoretical work is underway to consider possible mechanisms such as dynamical stark shift of the potential curves, resonant Raman excitation or light-induced canonical intersections (LICIs)[15] that may account for the observed velocity distribution.

With the advent of neutral fragment imaging capability, it is also possible to characterize the KER of two correlated neutral fragments. In fig. 1f, the bar chart describes the KER obtained with 800nm pulse alone, which was assigned to dissociation followed by detachment of the atomic F^- anion.[12] The dashed line represents the difference KER spectrum, when a 400nm pulse precedes the 800nm pulse, showing on one hand depletion of the low KER peak due to 800nm alone and an onset of a high KER peak, corresponding to dissociation by 400nm followed by F^- detachment with the intense 800nm pulse. This clear spectral distinction allows following the dissociation on the $^2\Sigma_g^+$ state in a two-color pump probe fragment imaging experiment. Inset of fig. 1f shows the onset of the high KER peak, indicating ultrafast photodissociation within the 35fs FWHM of our laser pulses.

To conclude, time resolved fragment imaging provide insight into dynamical mechanisms. Furthermore, ultrafast intense field dissociation of the simple F_2^- anion exhibits surprising non-linear photodissociation velocity patterns.

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