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N_2O ionization and dissociation dynamics in intense few cycle femtosecond laser radiation.

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Synopsis Using few cycle pulses, uniform-electric–field-ion imaging spectrometer and Dalitz plotting, we can identify two new stepwise processes, in the dissociation of the 3+ and 4+ states of N_2O in intense few cycle pulses at 4 10^{15} Wcm^{-2}. We observe suppression of selective channels at longer pulse length, which we attribute to molecular dynamics on the 2+ state and the shape of the 3+ potential, which influences propagation of the dissociating wave packet.

Ultrafast molecular imaging using femtosecond laser pulses of varying duration, from few cycle, to sub picoseconds is a promising method for revealing dissociation dynamics and ionization processes [1]. In the current study we focus on the linear triatomic molecule N_2O which is a challenge due to the fact that it possesses two identical nitrogen atoms [2].

The experiment was carried out at the Advanced Laser Light Source (ALLS) in Quebec. Few cycle laser pulses were generated using spectral broadening in a hollow core fiber filled with Argon at a pressure of 1.1 Atm. In the target chamber the laser pulses are focused by a parabolic mirror (f=10cm) on a well collimated supersonic jet of molecules inside a uniform-electric–field-ion imaging spectrometer. By employing a time and position sensitive delay-line anode detector at the end of the spectrometer, the resulting fragments are detected and their momentum determined.

To examine our results, we use the Dalitz plotting method, which is sensitive to the point of origin of each ion in the molecule. This has allowed us to distinguish between two channels (2,1,1) and (1,2,1) which contain the same ion species but give rise to different energy release and undergo different modes of break up. Figure 1 shows one sector of the Dalitz plot for the ions N^+, N^{2+} and O^+. The plot shows only coincidences which result from the (2,1,1) channel ie the terminal ions are N_t^{2+} and O^+ and the middle ion is N_m^+. The x axis represents the difference in energy of the two terminal ions, and the y axis represents the energy of the middle ion. The plot is capable of distinguishing two processes, the concerted process which involves stretching of both bonds equally (the intense region close to x=0.1) and the stepwise process which involves an N^{2+}+NO^{2+} intermediary process. This second process is represented by the diagonal region which can be modeled by the rotation of the NO^{2+}as it separates from the N^{2+} ion before undergoing Coulomb explosion. During this process the N_m^+ and the O^+ ions switch positions leading to the signature swapping of kinetic energy visible in the plot. Similar methods have been employed for the 3+ ion and for pulse lengths up to 500fs

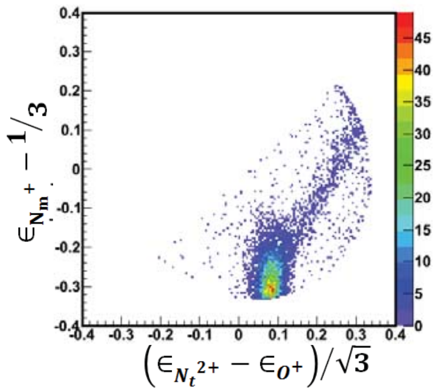

Figure 1. Dalitz plot showing break up of N_2O^{4+} into the (2,1,1) channel, with the intense region showing concerted break up and the diagonal region showing the stepwise channel N_2O^{4+}-NO^{2+}+N^{2+}-N^{2+}+N^++O^+

References

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