Observation of ultrafast proton interactions in water using the Gemini Laser Facility

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Abstract

The solvated electron has been extensively studied in many areas of radiation chemistry. A large focus of this research has been in radiation biology as they are a product of ionising radiation reacting with water molecules that can cause damage to DNA. Many of these studies employ chemical scavenging techniques, where a chemical is added to the water which reacts in some measurable way to the ionising radiation. Using an optical streaking technique with a high degree of synchronicity and measuring changes in transmission of the pristine water sample, we use laser-driven protons to generate solvated electrons in water, whilst simultaneously probing their evolution on a sub-nanosecond (10^{-9} s) timescale.

1 Introduction

Free electrons present in aqueous solutions, also called solvated electrons, have been extensively studied since being first discovered in 1962 by Hart and Boag [1]. The formation of the solvated electron is a prototypical process for radiation chemistry in water and occurs when H₂O molecules orientate around free electrons Study of their formation is important to understand the effects of ionising radiation in the treatment of cancer, as the majority of energy deposited in a cell by radiation is absorbed by water as a typical cell consists of up to 80%water molecules [2].

Many of these studies employ a technique known as pulsed ion radiolysis. This is typically performed using pulses of ions from radio-frequency accelerators in combination with chemical scavenging techniques to determine radiolytic yields of reaction products in the water after exposure to the ionising radiation [3, 4, 5]. Although effective, there are some drawbacks to chemical scavenging. To obtain high temporal resolution in such experiments, a high concentration of scavenging agent is required. Under these conditions it becomes possible that the scavenger will begin to take part in the radiolysis processes, significantly increasing the uncertainty of the measurements [3].

One way to address this issue is by using ultrafast laser plasma accelerators. Laser-driven proton acceleration permits short, few-ps pulses of ions which are not Lehrstuhl für Medizinphysik, Fakultät für Physik, München, 85748, Garching b. München, Germany

routinely available through conventional radiofrequency accelerators [6]. Combining this with a probe originating from the same laser seed pulse, enables a high degree of synchronicity for pump-probe experiments. In comparison to electron radiolysis, protons have a much higher Linear Energy Transfer (LET) determined by the stopping power of the material. Traversal of the proton through the water creates tracks of very high ionisation density, ionising water molecules as they deposit their energy. This process generates secondary ionising species, such as electrons which are initially thermalised and then trapped by surrounding water molecules, becoming solvated.

Observation of the solvated electron dynamics is enabled due to its wide absorption band, the high absorptivity of which makes it easy to detect even at low concentrations [7, 8].

2 **Experimental Setup**

The experiment was performed in the Gemini TA3 chamber as shown in figure 1. The south beam (15 J, 35 fs)was focused using an f-2 off-axis parobolic mirror onto 4 µm thick aluminium targets to generate protons via the Target Normal Sheathe Acceleration (TNSA) mechanism [9, 10]. Protons were accelerated up to energies of $\approx 15 \,\mathrm{MeV}$ (verified with stacks of radiochromic film) and propagated 12 mm in vacuum before passing through the 200 µm wall of the polyether ether ketone (PEEK) cell and interacting with the water. The interaction region was probed with a chirped 1 ns pulse extracted after the final amplification stage of the north beamline and imaged onto the entrance of a Andor Shamrock 750i spectrograph. A motorised delay stage was used to control the timing between the main and probe laser pulses with a range of ± 3 ns. An optical streaking technique was then used to obtain time-resolved images of the evolution of the proton-water interaction. As the frequencies in the chirped pulse will arrive at different times, each frequency will encounter subsequent stages of the interaction, encoding the evolution of the sample in the frequency spectrum of the pulse, the spectrometer is used to spatially separate the frequencies, producing an optically streaked image showing both the spatial and temporal evolution of opacity in a small region of the sample [11].



Figure 1: Sketch of the experimental setup. The main beam (15 J, 40 fs) is focused by an off-axis parabolic mirror onto a 4 µm thick aluminium foil at 45° incidence. Protons are generated via the Target Normal Sheathe Acceleration mechanism and propagate through 15 mm of vacuum before passing through a 200 µm thick polyether ether ketone (PEEK) window on the water cell and stopping in the water contained inside. The interaction region is probed with a chirped laser pulse, extracted before the final compressor stage of the main pulse. Due to the chirp, different frequencies will reach the interaction region at different times, encoding the evolution of the interaction in the frequency spectrum of the pulse. This information is then extracted by imaging the region at 6X magnification onto the entrance of an Andor 750i imaging spectrometer, permitting a simultaneous spatial and temporal examination over the duration of the 1 ns probing window.

3 Results



Figure 2: Left: Experimentally obtained optical streak showing the response of H_2O to TNSA protons. The x-axis represents the temporal evolution of the opacity from time of the laser-foil interaction. The y-axis shows the spatial evolution along the central axis of the proton burst. Right: Differentiated image showing the rate of change of transmission with respect to time, highlighting the region of the streak where the ion pulse is depositing energy and the subsequent onset of the electron solvation process.

Figure 2 shows an experimentally obtained optical streak of opacity generated in water. The initial onset of opacity at 45 ps corresponds to the arrival of prompt electrons and X-rays generated by the laser-target interaction. This is followed by the arrival of the much slower moving proton burst. As the lifetime of generated solvated electrons in water is many tens of nanoseconds [12], the recovery of the opacity is not observed within the probing window.

4 Conclusion

Presented in this report is an introductory overview of the observation of solvated electrons generated by the interaction of TSNA proton pulses with pristine water. We have demonstrated how this technique not only provides an insight into the response of water to bunches of high energy protons through the absorption mechanisms of solvated electrons, but also in their formation during the proton interactions themselves.

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