Subpicosecond radiolysis by means of terawatt laser wakefield accelerator

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ABSTRACT

A novel subpicosecond pulse radiolysis experimental system has been developed in Terawatt Ultrafast High Field Facility (TUHFF) at Argonne National Laboratory. TUHFF houses a 20 TW Ti:sapphire laser system that generates 2.5 nC sub-picosecond pulses of 4-25 MeV electrons at 10 Hz using laser wakefield acceleration. The system has been optimized for chemical studies. The subpicosecond electron pulses were used to generate hydrated electrons in pulse radiolysis of liquid water. Preliminary transient absorption spectroscopy data with picosecond resolution is presented.

Keywords: Laser wakefield electron acceleration, ultrafast pulse radiolysis, terawatt lasers.

1. INTRODUCTION

Currently, there is a strong interest among radiation chemists to develop subpicosecond and femtosecond pulse radiolysis techniques to study ultrafast chemical processes initiated by ionizing radiation. Proper understanding of these processes is important to a wide variety of basic and applied sciences[1]. When ionizing radiation (high energy ions or electrons) passes through condensed matter, it produces small volumes ("spurs") of highly concentrated excited and ionized molecules. For 5 MeV electrons spurs are a few to tens of nm in diameter and are separated by 100-200 nm along the radiolytic track[2]. The fundamental processes and rapid chemical reactions occurring in spurs on sub- and picosecond time scales (e.g., thermalization, solvation, and reactions of short-lived, energetic species (excited states and ions)) are very important for understanding the mechanism of radiation damage. These intraspur reactions involve exotic excited states and cross-reactions between ionized/excited molecules that far from equilibrium. Generation of such otherwise hard-to-access states, the unusual reaction regimes, and the inherently nonuniform deposition of the primary ionization/excitation events make it difficult if not impossible to study rapid radiolytic processes using all-optical techniques based on ultrafast laser sources.[3-5] Hence there is a great need for ultrafast high-energy electron pump sources.

The most common of radiation sources used in ultrafast pulse radiolysis experiments are radio frequency (RF) linear electron accelerators (linacs).[6] The time resolution of pulse radiolysis experiments employing these sources is typically limited to 20-30 ps at 10-100 Hz. Time resolution of ca. 5 ps was achieved at Argonne in 1986 in emission experiments only.[7-8] During the past five ye ars, new types of electron accelerators based on laser driven photocathode linacs have been developed. The best known of these are the facilities at Brookhaven National Laboratory (BNL),[9] Osaca University,[10] and the University of Southern Paris.[11] The low energy accelerator facility (LEAF) at BNL is now capable of generating 7 ps electron pulses at 10 Hz. This facility has been used to study pulse radiolysis of viscous (e.g., ionic) liquids[12-13] and supercritical Xe;[14] the short-lived species were detected using transient absorption (TA) spectroscopy (at delay times > 10-20 ps). A similar setup at the University of Southern Paris produces 4-6 ps electron pulses.[11] At Osaka University, Tagawa's group has succeeded in generating single electron pulses as short as 200 fs; but the time resolution is limited by the time jitter in the arrival of these short pulses, which is difficult to reduce below 10-15 ps. Improved time resolution is possible using streak cameras to clock the arrival times at the sample, but this method results in a time resolution reduced to a few picoseconds.

All three of these linear accelerators are photocathode based. The time resolution therefore depends upon the synchronization between the phase of the RF field and the laser pulse that generates the photoelectrons at the photocathode as well as the synchronization between the resulting electron pulse and the probe laser pulse. E.g., to generate an 800 fs electron pulse, the synchronization between the RF and the laser must be reduced to better than 500

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fs. This level of stability is difficult to achieve in practice and it is one of the major factors limiting the usefulness of this approach for ultrafast pulse radiolysis studies.

At Argonne, a different approach of ultrashort electron pulse generation employing a laser wakefield acceleration technique has been pursued. [15-19] This technique is based on the use of table-top terawatt (T3) ultrafast solid state lasers (for reviews see Ref. 20) for electron acceleration. By focussing terawatt (10^{12} W) laser pulses to peak intensities exceeding 10^{18} W/cm^2 at supersonic helium gas jets, it is possible to generate subpicosecond electron pulses with a charge of a few nanoCoulomb (10^{-9} C) and to accelerate these electrons to energies in the MeV range.[15-19] There are several groups in the world developing laser wakefield accelerators. The main interest of these groups is the mechanism of laser-plasma interaction, optimization of the electron beam for high energy physics applications, and development of next generation X-ray light source[21-24]. In this paper we show that these accelerators are also suitable for subpicosecond pulse radiolysis experiments. These "chemical applications" present additional challenges to the laser wakefield acceleration method. It has to provide continuous operation at a reasonable high repetition rate (conducive for pump-probe kinetic measurements, e.g. 1-10 Hz) and sufficiently large electron charge (> 1 nC per pulse). In most of the physics experiments these objectives were not the goals (the focus being, for example, monochromaticity of the electron energy, more efficient electron and proton acceleration, etc.) and most of the experiments were carried out in what is, essentially, single pulse mode with poor reproducibility of the electron pulse characteristics. By contrast, for radiation chemistry applications, it is important to have a source with reproducible parameters operating at a rate that is comparable to that shown by competing methods (to at least 10 Hz). Otherwise, neither the pump-probe method nor even data averaging are possible.

Before the construction Argonne's ultrafast radiolysis system, in collaboration with the University of Michigan (Center for Ultrafast Optical Studies) we have demonstrated that T3 accelerators, operated in the "single pulse" mode regime (at 0.5 mHz) can produce sufficient charge per pulse for detection of radiolysis products using TA laser spectroscopy.[25] In these experiments, subpicosecond electron pulses were generated by focusing terawatt laser pulses into a He jet and subsequently used to ionize liquid water. The ionization of water results in the generation of metastable "hydrated electrons" in which the s-function of the excess electron fills a void between 6-10 water molecules that "hydrate" the cavity electron by their hydroxyl groups.[26] This species plays the pivotal role in water radiolysis and it strongly absorbs in the visible and near infrared, exhibiting the extinction coefficient of 2×10^4 M^{-1}cm^{-1} (1 M = mol dm^{-3}) at the maximum at ca. 720 nm. Hydrated electron concentrations as high as 22 μM were generated, which is sufficient for reliable detection of the decay kinetics using TA spectroscopy. More recently, the technique has been used on the picosecond timescale.[27] Below we describe the next generation setup that is capable of producing 10 Hz train of 2.5 nC subpicosecond pulses for hours at a time, which enabled us to perform the first pulse-probe measurements of chemical kinetics on a picosecond time scale using a laser wakefield accelerator. In addition to the production of femtosecond electron pulses, this setup can be used for the generation of narrow and broadband femtosecond X-ray pulses (up to 20 keV).[24]

Another important advantage of laser wakefield accelerators is the inherent synchronization of electron and laser pulses, so the time jitter typical of linac based accelerators is not an issue, and the ultimate time resolution depends only upon the cross correlation between the laser and electron pulses and the physics of the electron beam interaction with the sample. The limiting factors of the time resolution under these conditions are 1) the fact that the electrons move through the sample at a velocity close to the speed of light, c, whereas the probe photons travel at a substantially lower velocity, c/n (where n is the refractive index of the medium); 2) unlike the linacs, the present laser wakefield accelerators do not produce monoenergetic beams[15-16] (although that may change in the near feature).[17-19] The typical electron spectrum is Maxwellian, with the median energy corresponding to that of the electron plasma in the jet. This dispersion in energy results in the dispersion of arrival times at the sample as the electron pulse travels through space; furthermore, since the electrons of different energy are stopped differently by the sample, via scattering and loss of energy to the medium, there is additional temporal spread of the pulse in the sample. Thus, the time resolution is limited by the nature and the physical dimensions of the sample, to a much larger degree than in laser experiments: superior time resolution requires thin samples. On the other hand, since the linear energy transfer (LET) for a typical sample is 2-5 MeV/cm (for 1-100 MeV electrons) and the typical radiation yield of the species of interest is < 5 per 100 eV of absorbed energy, to have a detectable TA signal from the radiation-induced species one has to have a sufficiently thick sample to stop the electrons. This is not the case for low energy photons and electrons where very thin samples can absorb most of the energy. Thus, ultrafast pulse radiolysis using fast electrons (of the range that is important for applications) is always a compromise

between time resolution and detectability. The more sensitive is the detection, the better time resolution is possible. In our experiments, the time resolution is limited not by the characteristics of the source or the synchronization issues but by this fundamental constraint that is inherent to the measurement and the physics of electron interaction. For other high energy electron sources, that is not the case. Below, we describe the laser wakefield accelerator that provides subpicosecond electron pulses for chemical physics research and demonstrate its use for ultrafast studies of aqueous radiolysis.

2. TERAWATT LASER SYSTEM

Our 20 TW laser system is described only briefly here (see Ref. 28 for more detail). The laser system consists of a mode-locked Ti:Sapphire oscillator (KM Labs Basic), a stretcher, three multi-pass Ti:Sapphire amplifiers and a compressor. The oscillator is pumped with the 4.75W output from a diode pumped frequency doubled Nd:YVO_4 laser (Spectra Physics Millennia V). The output of the oscillator exhibits a broad spectrum (FWHM ~50nm) that is centered at 775nm. The oscillator produces 5nJ pulses at a repetition rate of 100MHz. The pulses are then passed through a 1:1 imaging single grating stretcher to obtain a pulsewidth of ~300ps. The pulse train is then passed through a 10Hz pulse picker. The latter consists of a Pockel cell polarizer (Medox). The first amplifier has a multi-pass ring design. It houses a 1cm thick Brewster angle amplifier rod (Crystal Systems) that is pumped using 25mJ (532nm) light that is split off of the output of a frequency-doubled Nd:YAG (Spectra Physics PRO-270). After six passes through the amplifier the IR pulse is passed through a second pulse picker that eliminates any amplified spontaneous emission background. The pulse energy at this point is 2.5mJ; the beam diameter is 0.8 cm ($1/e^2$). The second amplifier rod (Crystal Systems) is an anti-reflection coated 2cm cylinder. Two frequency-doubled beams from Nd:YAG lasers (Spectra Physics LAB-190 and Spectra Physics PRO-270) are relay imaged onto each face of the amplifier rod. The total pump energy is 850mJ. This amplifier has a four pass bow tie configuration. A -1.5m lens is placed after the first pass to compensate for the thermal lensing that occurs in the amplifier rod. The output of the second amplifier is passed through a spatial filter that expands the beam diameter to 1.2cm ($1/e^2$). At this point, the pulse energy is 330mJ. A beamsplitter is used to divert ~60mJ of this beam which is passed through a second compressor and used as a probe. The third amplifier rod (Crystal Systems) is an anti-reflection coated 3cm cylinder. The third amplifier is pumped on each face by a relay imaged frequency-doubled Nd:YAG laser (Spectra Physics PRO-350). The total pump energy is 2.4J. To eliminate thermal lensing in the third amplifier the rod is mounted in a liquid nitrogen cryostat (Janis Cyrogenics). Two passes through the amplifier boost the pulse energy to 1.1J. After the first pass slight aberrations in the beam profile are cancelled out by rotation of the beam profile by 90° before the second pass. After the third amplifier, the beam is expanded to 5cm and directed into a vacuum chamber that houses a two-grating pulse compressor. The output pulse has a spectrum that is centered at 800 nm and has a width of 34 nm FWHM. The compressed pulse is 600 mJ, 35fs FWHM. The repetition rate of the setup is 1-10 Hz. Ultrashort electron pulses are generated by focusing the resulting laser beam onto the front surface of a 1.2 mm supersonic He jet using a 50 cm focal length off-axis gold-plated parabolic mirror. The jet characteristics are crucial for obtaining the high electron output. We have tested different types of nozzle designs and selected the one which produces the highest electron charge. This nozzle has a similar design to the one used by the group of Prof. Umstadter at the University of Michigan.[29] For electron charge measurements a home-built 4.5 cm diameter Faraday cup was used; this cup was placed ca. 2 cm after the jet. In order to block the laser light and low energy electrons the Faraday cup is screened using a 80 μm thick Al window. The total electron charge is 2-3 nC per pulse with a pulse-to-pulse variation of 15-30%. This charge is distributed over a wide cone of almost 20° (Fig. 1). Within this cone there is a narrower 6° cone of electrons that have somewhat higher energies (see below). The electrons generated in the plasma undergo multiple elastic scattering events as they emerge from the jet. This scattering is stronger for low energy electrons and it results in a steep angular distribution of electron energies.

The electron beam spectrum was determined using a spectrometer in which two permanent magnets were used to bend the electron beam. The electrons were imaged on a luminescent screen and the emitted light was observed using a camera. Copper apertures were used to dissect the electron beam. The magnetic field of the spectrometer was mapped and relativistic equations solved numerically to reconstruct the energy spectrum. Figs. 2a and 2b show the spectra obtained for electrons scattered into 2° and 14° cones, respectively. In the latter figure, the most probable electron energy is ca. 4 MeV. There is also a sloping "tail" extending to at least 25 MeV. In the 2° cone, the most probable energy is increased to 7-8 MeV and the fraction of low-energy electrons is greatly reduced. Very similar energy distributions can be obtained theoretically assuming Maxwell distribution for the electron energies and small angle multiple elastic

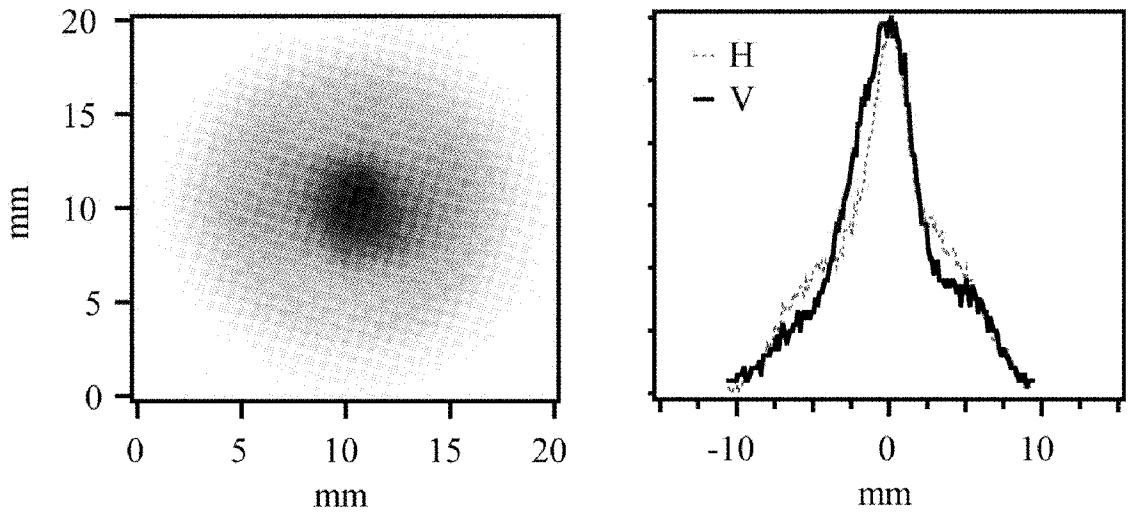

Figure 1: Transverse profile of the electron beam measured at 27 mm after the jet. The beam profile is almost identical in both the vertical (V) and horizontal (H) planes.

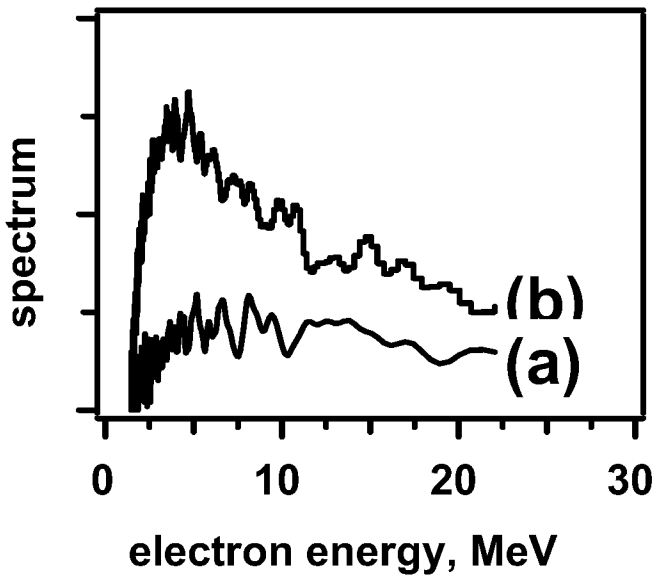

Figure 2: The energy spectrum of the electron beam measured for (a) 2^{o} and (b) 14^{o} cones.

scattering in the jet. To characterize the average energy of the electrons across the whole beam the latter was passed through a 1 cm thick stack of aluminum plates and the distribution of the dose was determined using a radiochromic film inserted between the plates. The dose distribution was consistent with a Maxwell distribution of electron energies with average energy of 2.3±0.3 MeV.

The narrow cone contains the electrons that would be optimum for doing ultrafast experiments, but the fraction of these high-energy electrons in the beam is too low for 90^{o} crossed-beam spectroscopy described below. The scattering in the

jet can be reduced by optimizing the jet parameters (e.g., by reducing the gas density and increasing the gas velocity), which will be done in the future.

3. ULTRAFAST PULSE RADIOLYSIS

In the pulse radiolysis experiments (Fig. 3), a 80 μm thick Cu foil is placed at 1.7 cm after the jet to block the laser light and transmit MeV electrons. The transmitted electron beam is used to radiolize and pump the sample in a pump-probe experiment. It passes through a 1 cm path Suprasil cell filled with liquid water; the front window of this cell is 1 cm away from the Cu shield. The electron beam is crossed at 90° inside the cell with two collinear probe beams: 800 nm 35 fs beam and a 670 nm cw laser beam. The first probe beam is derived from the 2nd amplifier. After passing through a variable length compressor, and a motorized delay stage this probe beam is split into two beams by a 50% beam splitter. The transmitted beam is used as a reference, while the reflected beam is focused into a sample cell and used as a probe "signal" beam. The second probe beam, derived from a 670 nm cw diode laser, is used to compensate for pulse-to-pulse variations in the absorbed dose. It propagates along the same optical path as the 800 nm probe beam. Both of these probe beams are used to measure transient absorption of solvated electrons generated by the electron pulse. However, the 800 nm beam is used to obtain the transient absorption on the picosecond time scale in a stroboscopic fashion, whereas the 670 nm cw beam registers the same absorption on the nanosecond to microsecond time scales using a 30MHz photodiode and a fast transient digitizer. The solvated electron in water is long lived and both absorbance signals scale linearly with the dose. If both of the beams probe the same region of the sample, the two TA signals track each other. This proportionality can be used to compensate the variation in the dose deposition.

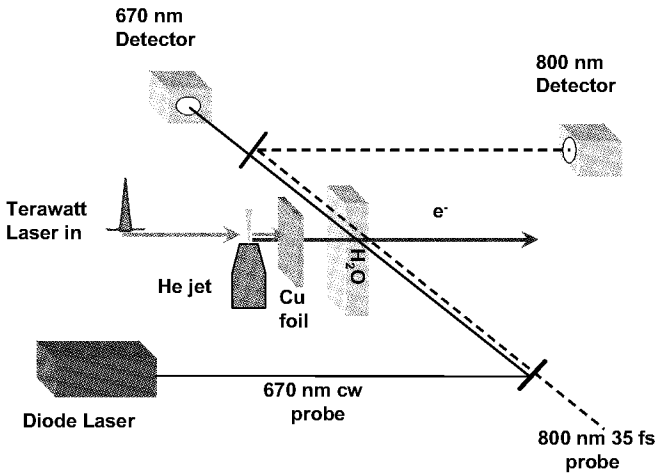

Figure 3: Ultrafast pulse radiolysis setup

In our first demonstration experiment, transient absorption of solvated electron in radiolized water was observed. The absorption spectrum and decay kinetics of these species are well known from a previous work.[26] Fig. 4a shows the decay kinetics of the solvated electron on the microsecond time scale following a 75pC electron pulse from TUHFF. This decay is due to the reaction of the solvated electron with electron-acceptor impurities in water, mainly traces of oxygen. Fig. 4b shows the early time kinetics of solvated electron in water measured by 800 nm 35fs probe. There are three factors determining the rise time of the TA signal in Fig. 4b: the formation time of the species and the time scale of their spectral evolution, the pulse duration (and dispersion) of the electron beam at different points of the sample, and the traveling time of the probe through the irradiated zone. On the time scale of Fig. 4b, the formation of e_{aq}^- can be considered as instantaneous. Assuming that the pulse duration for the electrons emerging from the jet is comparable to the duration of the TW laser pulse, and knowing the electron spectrum, the dispersion of the arrival times for the electrons at the sample was estimated as 3-4 ps. Given the large diameter of the electron beam at the sample (which is

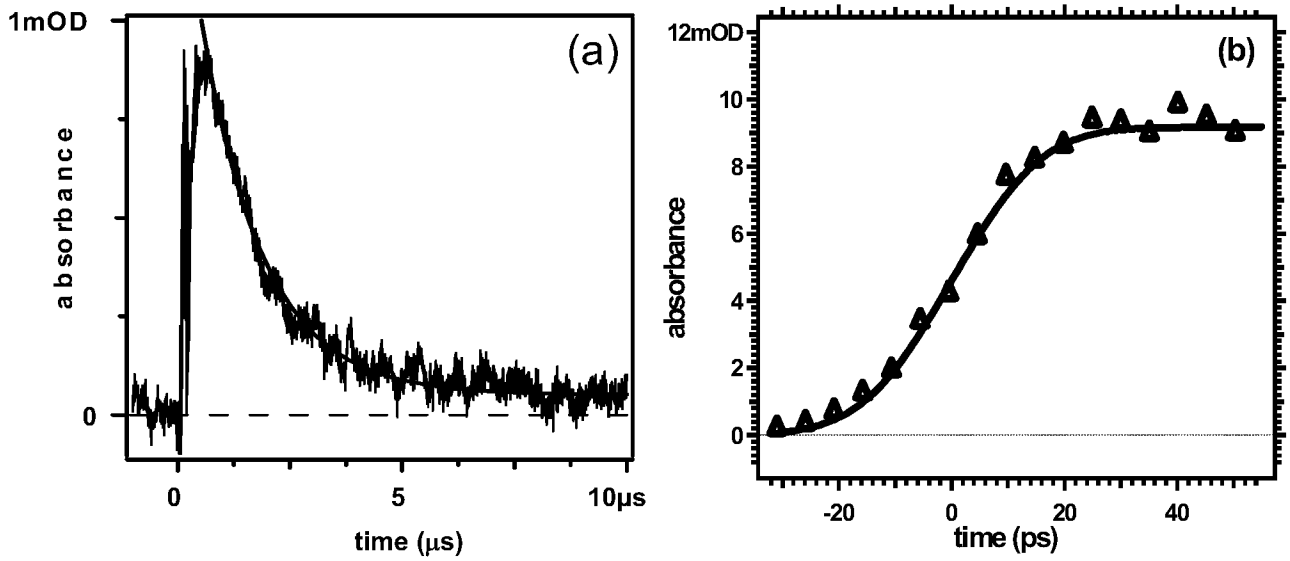

Figure 4: Kinetics of the solvated electron in water measured by (a) a 670 nm CW probe beam; (b) a 800 35 fs probe.

commensurate with the sample thickness) one would expect that the third factor determines the rise time of the TA signal observed. In principle, this rising part of the kinetics can be simulated from the calculated dose distribution (i.e., the distribution of e_{aq}^-) along the path of the probe beam. In reality, this distribution is not known exactly, and moreover it fluctuates from pulse to pulse. In order to make a comparison with other experimental systems the kinetics were fit by an error function which corresponds to a hypothetical experiment with a Gaussian pulse of electrons, infinitely thin sample and collinear beam geometry. The $1/e^2$ Gaussian time obtained from the data in this fashion is 17.9 ps for the 10 mm cell used. We have also performed measurements with thinner cells, which showed shorter rise times. The results of these measurements will be reported in Ref. 30. The best time resolution we have demonstrated so far[30] is on the order of a few ps. In principle, this time can be shortened further by the use of even thinner samples, better shaping of the electron spectrum, and collinear detection.

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