ANL/CHM/CP-97338

Invited Contribution - MO4025
Linac98 Conference
Fairmont Hotel
Chicago, IL
August 23-28, 1998

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Laser System for a Subpicosecond Electron Linac*

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Abstract

At the Argonne Chemistry Division efforts are underway to develop a sub-picosecond electron beam pulse radiolysis facility for chemical studies. The target output of the accelerator is to generate electron pulses that can be adjusted from 3nC in .6ps to 100nC in 45ps. In conjunction with development of the accelerator a state-of-the-art ultrafast laser system is under construction that will drive the linac's photocathode *and* provide probe pulses that are tunable from the UV to IR spectral regions.

1 Introduction

From the start time-resolved investigations of fast chemical processes have been part of radiation chemical studies. Recent advances in accelerator technology are making it possible to generate subpicosecond electron pulses. Additionally, solid state laser development is making it more routine to generate subpicosecond optical pulses. A merging of these two technologies will provide an invaluable tool for probing ultrafast radiation induced processes. In this contribution the development of a state-of-the-art laser system that will be capable of both driving the photocathode of a subpicosecond linac and simultaneously and provide a widely tunable (UV to mid-IR) stroboscopic detection capability will be described

2 FEMTOSECOND LASER SYSTEM

2.1 Femtosecond Laser System

In order to generate subpicosecond electron pulses a state-of-the-art laser systems must be developed to drive the photocathode. The accelerator under consideration at Argonne National Laboratory requires the laser operate within very strict specifications[1]. The following key problems must be addressed:

- Development of an intense (>3mJ) 5ps UV laser pulse that is needed to drive the photocathode.
- A .1ps probe pulse must be derived from the same laser and be independently tunable from the UV to the IR.
- The UV pump and the probe laser pulses must be synchronized to the RF of the accelerator to within .5ps.
- Development of novel, sensitive and specific detection techniques to make the fullest use of the potential information.

For the photocathode the laser must provide high energy UV (~260nm) pulses that can be varied in energy from .1mJ to 3mJ. The low UV pulse energy corresponds to operation of the accelerator in the low charge short pulse mode (i.e., 3nC, .6ps), while the high UV pulse energy corresponds to operation in the long pulse high charge mode (i.e., 45ps, 100nC) mode. In addition to variable UV pulse energy the UV pulsewidth must be made variable from 5ps to 60ps. Some of the laser pulsewidth and energy requirements are summarized in Table 1.

To make use of the full capabilities of the experimental system, the laser must also produce pulses with less than 0.1 ps duration with enough energy (~1mJ) to pump a series of widely tunable optical parametric amplifiers that will provide probing wavelengths from the UV to IR spectral regions. The laser is to operate from single shot to 60 Hz with a pulse-to-pulse jitter less than .5ps with respect to a 1.3 GHz master clock. *Minimization of the jitter is the is the most crucial and difficult aspect of the laser design.*

Table 1: Laser Energy and Pulsewidth Specifications

Linac Pulsewidth	Charge	Laser Pulsewidth	Laser Pulse Energy
.6ps	3nC	5ps	.1mJ
3.0ps	10nC	15ps	.3mJ
8.0ps	50nC	45ps	1.5mJ
45ps	100nC	60ps	3mJ

2.2 Generation of Intense UV Laser Pulses

A block diagram of the laser amplification scheme is shown on the following page. Briefly, a diode-pumped frequency-doubled Nd:YVO laser will be used to pump a femtosecond Ti:Sapphire (Ti:S) laser that will produce a

* Work performed under the auspices of the Office of Basic Energy Sciences, Division of Chemical Science, US-DOE under contract number W-31-109-ENG-38

81.25MHz pulsetrain of 3nJ 50fs pulses centered at 800nm. The 50fs pulses will be passed through a grating pulse stretcher which will increase the pulsewidth to 500ps. This will reduce the peak power of the laser pulses enough to prevent optical damage in the following two stages of amplification (chirped pulse amplification). After the stretcher a polarizer-Pockels cell-polarizer combination will be used to select pulses out of the 81.25MHz pulse train at 60Hz (linac repetition rate).

The first preamplifier stage (AMP1) is a multipass (8 pass) Ti:S amplifier that will produce ~5mJ pulses at 800nm[2]. AMP1 is pumped with 50mJ pulses from a high power Nd:YAG laser (HP Nd:YAG). The output of AMP1 will be used to seed a power amplifier (AMP2) that will boost the energy up to >100mJ. AMP2 is also a multipass amplifier (4 pass) and will be pumped by 450mJ from a HP Nd:YAG laser.

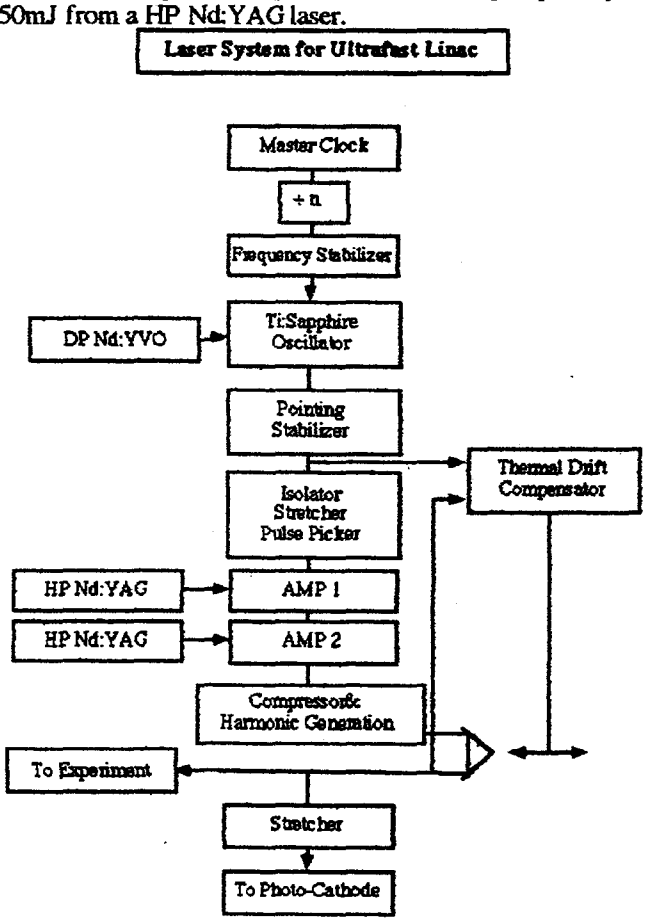

Fig. 1. Block diagram of the amplified femtosecond laser system. See text for details.

The amplifier output will be compressed in a standard grating pulse compressor to <.1ps and passed through a series of specially designed nonlinear crystals to generate the third harmonic at 260nm. The UV pulse energy is expected to exceed the required maximum of 3mJ. Dichroic mirrors will be used after the harmonic conversion step to divert the residual fundamental towards the optical parametric amplifiers while directing the third harmonic towards the linac cathode. Before illumination of the photocathode the UV will be temporally stretched by passing it through several centimeters of suprasil. The pathlength of the suprasil will be determined by the amount of UV pulse stretching that is required for the desired electron pulse charge and pulsewidth (see Table 1).

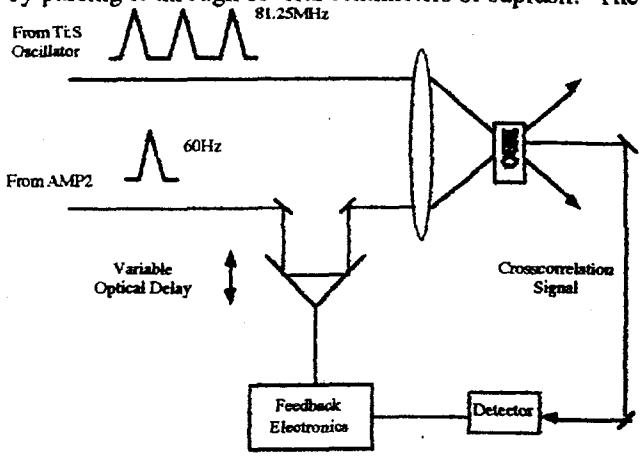

Fig. 2. Thermal drift compensation of the Ti:S amplifiers using the crosscorrelation technique. BBO (beta barium borate) is a nonlinear frequency mixing crystal. See text for details.

2.3 Minimization of Jitter

To generate .6ps electron pulses from a photocathode driven linac it is necessary to synchronize the phase of the RF that drives the photocathode and the UV laser pulses to within .5ps[1] of each other. The master clock for the RF will operate at a frequency of 1.3GHz. Expected sources for loss of synchronization will come from a variety of sources. Most importantly will be, drifting in the output frequency of the Ti:S oscillator, slow thermal drifting in the optical components, mechanical vibrations, and air currents.

The 16th subharmonic (81.25MHz) of the master clock will be used as a reference frequency for the Ti:S oscillator. Using commercially available electronics (Spectra Physics, Mountain View, CA) it is possible to reduce the jitter/synchronization out of the Ti:S oscillator to better than .1ps rms.

During the amplification process the synchronization will degradedue to the slow thermal drifting of the optical components and vibrations that result from mechanical and audio interference. To reduce effect of the slow thermal expansion and contraction of the optical components the pathlength that the laser pulse travels through the amplifier must be minimized. This dictated the choice to use a multipass amplifier design rather that a more efficient regenerative amplifier for AMP1. A multipass amplifier should reduce the path length by ~20m relative to a regenerative amplifier.

Compensation for slow thermal drifts will be actively achieved by passing the pulse out of the amplifiers through a stepper motor/piezo driven optical delay that will compensate for thermal drifts on a shot-to-shot basis

will compensate for thermal drifts on a shot-to-shot basis (see Figure 2). The error signal for driving the optical delay will be derived from the crosscorrelation between the Ti:S oscillator and the output of AMP2 in a BBO crystal. This technique has been previously used to maintain subpicosecond synchronization between a Ti:S amplifier and an infrared optical parametric amplifier[3]. Specially designed optical tables and evacuated beam tubes will minimize the sensitivity of the system to mechanical vibrations.

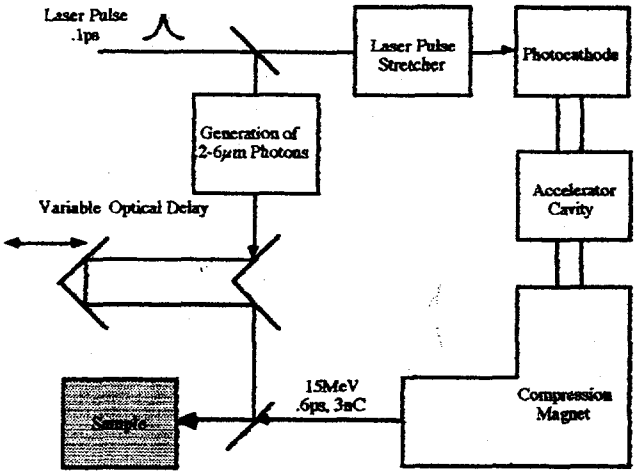

Fig. 3. Block diagram depicting a typical electron linac pump/laser probe experiment.

3 CONSEQUENCES OF LASER SYNCHRONIZATION

A significant advantage of this system is that the laser that "drives" the photocathode is inherently synchronized wish the electron pulse, with very little jitter in the synchronization. This fact translates into considerable improvements in sensitivity and capabilities for detection schemes. Figure 3 shows a the setup for a typical pump/probe experiment in which the subpicosecond electron linac acts as pump source to initiate a radiological event that is subsequently probed by a laser pulse.

Detection schemes can now be based on laser produced interrogating light pulses as opposed to linac produced light pulses (Cerenkov radiation) that are presently used. Thus, the intensity and wavelength range will be available to develop new detection schemes for examination of transient species that are impossible to study at this time.

A new type of detection that will become available is transient infrared spectroscopy, which can be used to determine the role of excess thermal energy and vibrational energy transfer in radiation chemistry. In addition to providing a window through which to view the role of vibrational dynamics, the structural selectivity of infrared spectroscopy can be used to follow the progression of and identification of transient species. To this extent we have recently developed a unique optical parametric amplifier based on potassium niobate that is tunable throughout the chemically important 2.5-5.5 micron spectral region.

Other new types of detection under development include time resolved coherent spectroscopy (e.g., CARS, transient grating spectroscopy, photon echo spectroscopy), and time-resolved resonance Raman spectroscopy. Such techniques will provide new information on energy levels, lifetimes of transient species, collisional processes, coupling strengths, and diffusion processes.

4 SUMMARY

A femtosecond laser system for driving a subpicosecond photocathode electron linac has been described. The main design criteria center around the necessity to achieve .5ps synchronization between the high energy UV pulses and the phase of the RF that drives the photocathode. As a consequence of the synchronization between the linac and the laser a wide variety of sensitive detection schemes now become available for probing radiation induced chemical events on fast timescales.

5 ACKNOWLEDGEMENTS

We gratefully acknowledge the input from Drs. Illya Shkrob, D. M. Bartels, and D. Werst in the preparation of this manuscript.

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