Development of a New Ti:Sapphire Laser System for Femtosecond Laser Ionization at kHz Repetition Rates

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Abstract. We have developed a new laser system which operates at a repetition rate of one kHz and generates 3.5 mJ/pulse with 85 fs pulse widths at 800 nm. It is composed of a self-mode locked oscillator followed by a pulse stretcher, a regenerative amplifier and post-amplifier which are pumped by frequency doubled Nd:YAG lasers, and a compressor. The new post-amplification stage has a four-pass, nearly collinear geometry. This allows for improved power and shorter pulses while still providing high repetition rates. Fast repetition rates are essential for imaging experiments using a time of flight (TOF) mass spectrometer since a fast rate decreases the probability of sample drift during image acquisition and decreases the amount of time needed per image.

LASER SYSTEM

The lasing process is initiated by a cw Argon Ion laser pumping a Ti:Sapphire crystal at 3 W at all lines. The oscillator self-mode locks due to Kerr lens modelocking (1) and produces an output of 3 nJ with a 60 fs pulse width. The pulse width is then stretched to 300 ps by multiple passes on a 1400 lines per millimeter grating.(figure 1) The stretched pulse is amplified to approximately 1.5 mJ/pulse in a Ti:Sapphire crystal which is pumped with a frequency doubled,

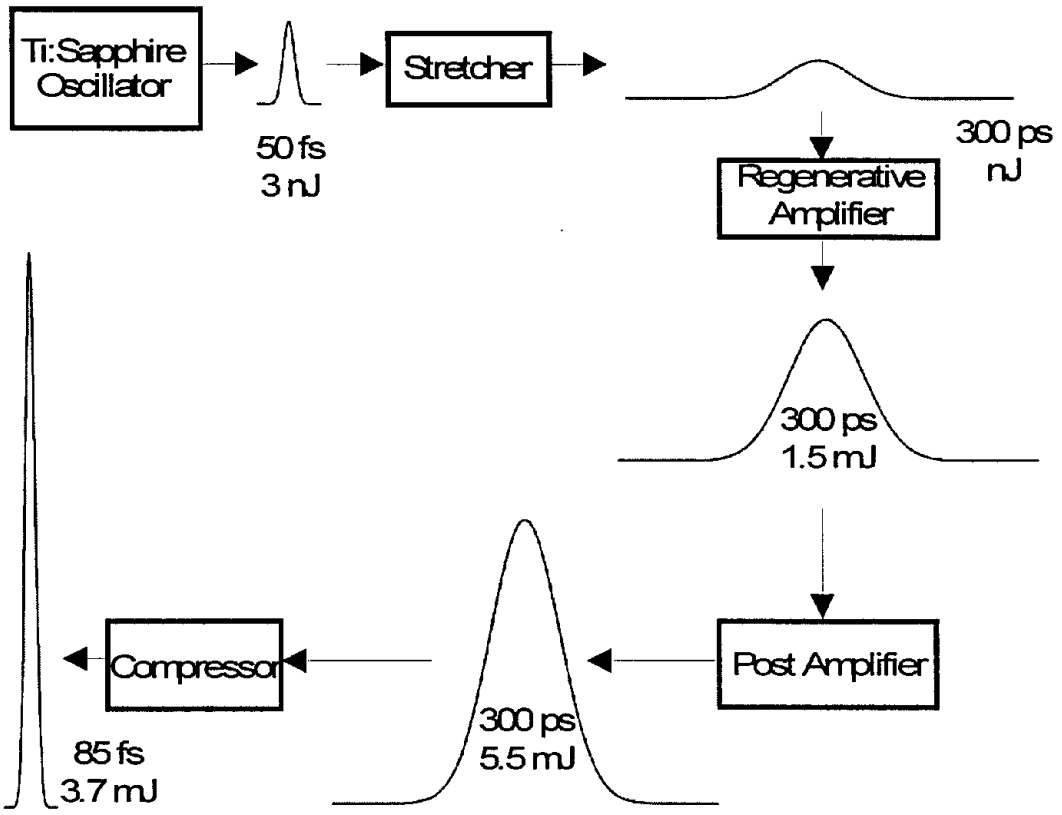

FIGURE 1. Overview of laser system pulses

Q-switched Nd:YAG laser running at an average power of 8.5 W. A Pockels cell injects pulses into the regenerative amplifier cavity and dumps the selected amplified pulse. This output is then directed into the post-amplifier by the isolation stage of the stretcher. The light is amplified to 5.5 mJ/pulse by passing four times, nearly collinearly through a third Ti:Sapphire crystal which is pumped with 14 W average power from a doubled Nd:YAG. The beam from the post-amplifier is expanded with a telescope and directed into the compressor. After compression the fundamental can be used directly or can be frequency doubled or tripled.

This newly designed laser system has a pulse energy of 3.7 mJ with a pulse width of 85 fs at a 1 kHz repetition rate. Without a post-amplifier, the pulse energy is 1.5 mJ and the pulse width is 150 fs at the fundamental. The original plans for this laser system called for a bow-tie configuration (which is typically used in Clark-MXR's 10 Hz systems) in the post-amplification stage. However, at these high powers, the very first pass of the laser is close to the saturation intensity for the Ti:Sapphire crystal. These powers also generate thermal gradients causing thermal lensing within the crystal which can result in poor mode

quality and diffraction of the beam. The nearly collinear design helps minimize these problems, since each beam, passing through about the same point, is experiencing the same aberrations. In spite of this, there were still deleterious effects from thermal lensing in the crystal, so gain reshaping was employed by using a 532 nm pump beam which was smaller than the 800 nm beam. This amplified the most intense parts of the 800 nm beam and therefore eliminated many of the negative qualities of the beam which existed at the edges of the gaussian profile. At energy densities near the saturation point, the pump fluence dominates the amount of amplification that can occur. Ideally, the pump beam would be as small as possible, however, a compromise must be reached so the thermal gradients are not too large. The increased power of this system will lead to an increase in doubling and in tripling power.

POSTIONIZATION AND MASS SPECTROMETRY

The time of flight instrument used for our studies has been specially designed to incorporate laser ionization experiments. It is equipped with a leak valve to facilitate gas phase experiments. Moreover, we have incorporated a liquid nitrogen cooled sample stage which removes the background gases which are present and slows the sublimation rate of volatile components of the sample. This is critical to postionization studies.(2) The system is also equipped with a time to digital converter (TDC) and an analog detector which receive signal from a dual microchannel plate assembly. Previous experiments in our laboratory, on a similar machine, indicated that the detection system (TDC) was being saturated due to the large number of ions produced in postionization studies. The analog detector eliminates this problem and counts all of the ions hitting the detector giving a more quantitative representation of the mass spectrum.

The shorter pulses of this new laser increase the probability of outrunning neutral dissociation events and should aid in the detection of intact molecular ions.(3) Previous power dependence studies indicated that the ionization volume was not saturated. However, preliminary experiments with the new laser system indicate that the ionization volume is beginning to approach saturation.

A very fast repetition rate is essential to chemical imaging using mass spectrometry. It not only saves time by requiring less time per image, but also decreases the likelihood of sample drift during image acquisition which further improves the spatial resolution of imaging experiments. If we consider an area of 20 μm by 20 μm with 20000 pixels and an ion dose of 1% (static limit) with a Ga^+ beam at 60 pA, then a typical secondary ion mass spectrometry (SIMS) experiment with a primary ion pulse of 10 ns and a repetition rate of 5 kHz takes 33 minutes to reach the static limit. Our goal is to reach the static limit as quickly as possible so as to gain the most information in the shortest amount of time. In comparison, a laser running at 10 Hz with a primary ion pulse of 500 ns would

require 5.5 hours to reach the static limit. However, a femtosecond laser with a repetition rate of 1 kHz and a primary ion pulse of 500 ns only requires 3.2 minutes for the same area and conditions. A short primary ion beam pulse is necessary in SIMS because the pulse length is what determines the mass resolution. In laser postionization studies, the mass resolution is determined by the ability of the mass spectrometer to compensate for the energy dispersion introduced by the finite width of the laser beam so a much larger ion pulse can be used.

Images and spectra have already been obtained using this laser system and instrument. The image in figure 2 shows a map of pyrene butyric acid on silver beads. The image is 200 μm by 200 μm and was obtained by postionizing the neutral particles with 266 nm light that were produced as a result of Ga^+ ion bombardment. This map represents the m/z 215 ion (M-CH_2CH_2COOH) where the brightest areas are most intense. The postionized spectra show significant quantities of the molecular ion and fragments at m/z 215. In comparison, SIMS spectra show much fragmentation and very little intact molecular ion.

FIGURE 2. Map of fragment ion of pyrene butyric acid on ~70 μm silver beads.

CONCLUSION

The new laser system and time of flight mass spectrometer have resulted in improved chemical imaging capabilities. In addition, the increased peak power of the laser is leading to a further understanding of the mechanisms which govern the ionization process in the gas phase.

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