Nd:YLF Laser System for the Study of Termolecular Reactions and Metastable Isomers of Polyatomic Molecules

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The DURIP grant F49620-97-1-0255 has supported acquisition of a state-of-the-art regeneratively amplified Ti:Sapphire laser system. This laser system is the primary laser for our new multi-particle photoelectron-photofragment coincidence spectrometer, and is currently being used to carry out experiments on three-body dissociation dynamics in clusters of oxygen and ozone. These experiments make use of a new particle detector built in our laboratory capable of recording the time- and position-of-arrival of up to eight photofragments in a period of 20-500 ns following a laser pulse. The initial DURIP proposal discussed the purchase of a Nd:YLF psec regenerative amplifier, however, the Ti:Sapphire laser purchased is a much more flexible and powerful laser, producing both psec and fsec pulses with a higher peak power. The initial experiments with this laser will use the fundamental and the 2nd, 3rd and 4th harmonics. Future applications will include the generation of tunable radiation with an optical parametric amplifier and ultrafast experiments using 100 fs pulses. We took delivery of the laser December 1, 1997, with initial installation December 12, 1997.
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**1. Equipment Acquired under DURIP Grant**

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**Acquired Equipment:** CPA-2000 kHz Ti:Sapphire regenerative amplifier, producing either 0.4 W, 1.4 ps pulses at 1 kHz or 0.8 W, 120 fs pulses at 775 nm

**Manufacturer:** Clark-MXR, Inc.

**Cost (Including CA State Tax and Shipping):**

- **DURIP Funds** $122,268
- **UCSD Matching Funds and other non-Federal Funds** $ 60,222
- **Total** $185,653

**2. Research Projects**

The new photoelectron-multiple-photofragment coincidence spectrometer in our laboratory allows a new generation of experiments on three-body dissociation dynamics to be carried out. The CPA-2000, with its picosecond and femtosecond pulse capabilities, plays an integral role in these new experiments. We plan on applying this apparatus to a study of some of the three-body association reactions important in atmospheric and combustion phenomena, such as the recombination reaction that produces ozone: \( \text{O} + \text{O}_2 + \text{M} \rightarrow \text{O}_3 + \text{M} \). In the initial experiments using the new laser, the dissociative photodetachment and photodissociation of \( \text{O}_4^- \) and \( \text{O}_6^- \) have been studied at 775 nm and 388 nm. The results at 775 nm show that while \( \text{O}_4^- \) only undergoes dissociative photodetachment at that wavelength, \( \text{O}_6^- \) only exhibits photodissociation processes yielding \( \text{O}_2^- + \text{O}_2 + \text{O}_2 \). This suggests that either a breaking of the high symmetry of the \( \text{O}_4^- \) core or the presence of a new charge-transfer-to-solvent band in \( \text{O}_6^- \) has a profound impact on the photochemistry at this long wavelength. At 388 nm, the results confirm our previous observations at 532 and 266 nm that while the addition of \( \text{O}_2 \) to \( \text{O}_4^- \) to form \( \text{O}_6^- \) only slightly perturbs the dynamics of dissociative photodetachment, significant changes in
the photodissociation dynamics occur, yielding highly vibrational excited \( \text{O}_2^\cdot \) products. We are now beginning our studies of the \( \text{O}_3^\cdot \cdot \text{H}_2\text{O} \) system, which will provide a model for the ozone recombination reaction.