

Summary of AMO/Chemical Physics Work Group

The work group on atomic, molecular, optical, and chemical physics consists of Mark Knickelbein (Argonne National Lab.), Steve Pratt (Argonne National Lab.), Laurie Butler (University of Chicago), and Cheuk Ng (Univ. of California at Davis). Each member of the work group has proposed experiments, which require specific characteristics of the VUV ALFF as summarized in the table below.

PI/ Affiliation	Experiment	λ -range (nm)	Pulse energy (μ J)	Pulse length (fs)	Spot size (μ m)	Energy FWHM (%)	2 nd laser
Mark Knickelbein/ANL	Photo- absorption of clusters	55-200	100	500	5000	2.5	ArF
Laurie Butler/U. Chicago	Photo- dissociation of radicals	65-180	200	500 or longer	<500	1.0	ArF
Steve Pratt/ ANL	Double photo- ionization	10-100	200	<300	<500	0.3-1.0	
Cheuk Ng/ UCDavis	Spectroscopy of radicals and ions	55-200	200	200-500	2000	0.3-2.5	IROPO

In order to carry out these experiments, the work group also proposes to construct a portable, multi-purpose experimental apparatus as shown in Fig. 1. The apparatus consists of a pulsed molecular beam production system, a time-of-flight (TOF) mass spectrometer, and a TOF electron detector. The molecular beam production system can be used to prepare metal clusters by laser ablation and radicals by laser photodissociation method.

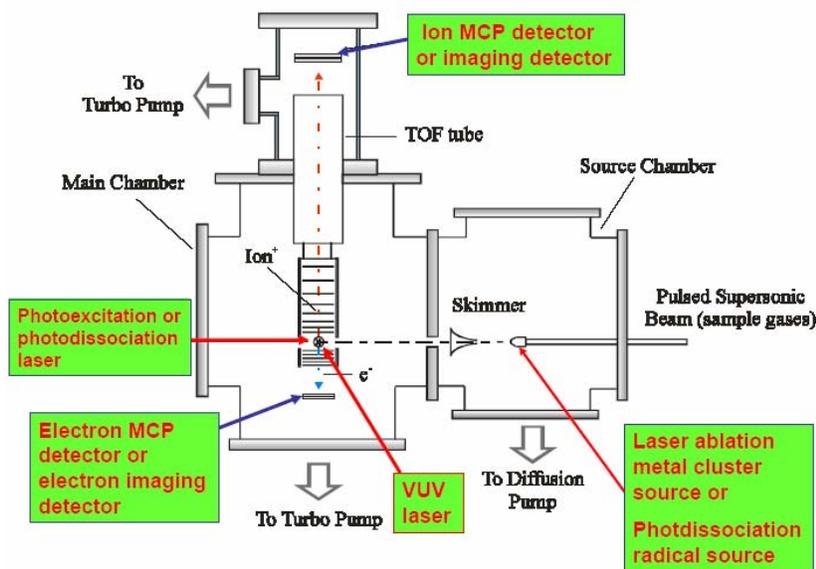


Figure 1. Molecular beam photoion-photoelectron apparatus

Furthermore, the ion and electron multichannel plate (MCP) detectors can be replaced by ion and electron imaging detectors for velocity imaging experiments described below.

The types of experiments described below can be performed using the molecular beam photoion-photoelectron apparatus of Fig. 1.

1. Single-photon VUV photoionization efficiency (PIE) measurements

In this experiment, the PIE of a specific ion is measured as a function of VUV photoionization energies. Mark Knickelbein is interested to determine the IEs of metal clusters and transient complexes produced in the metal cluster ablation and radical sources can be measured. Photoionization cross sections and IEs for radicals produced by the

2. Photodissociation dynamics studies

In this experiment, a molecular sample in the form of a supersonic beam is intersected by a dissociation laser. The photofragments thus formed are sampled by VUV photoionization and detected by velocity ion imaging scheme. By selecting the VUV photoionization energy, structures of photoproducts, including isomeric radicals, can be determined. The velocity imaging technique allows the determination of product angular distributions and translational energy distributions, from bond dissociation energies and potential barriers for dissociation can be deduced. By momentum matching, correlated product pairs can be unambiguously identified.

3. Ultrafast Pump-probe studies

The VUV ALFF source is unique for ultrafast pump-probe studies due to the tenability of the ALFF source. The detailed photodissociation dynamics or energy transfer processes induced by photoexcitation using a femtosecond laser can be followed by probed by delayed VUV photoionization using the ALFF source. In this experiment, the resulting photoions or photoelectrons are detected. The photoelectron distribution provides information about the state of the excited molecular complex along the dissociation coordinate and the wave-packet dynamics for the fragments.

4. Double photoionization via three VUV photons excitation

By selecting the proper pulse energy, pulse width, and spot size of the VUV photon, it is possible to examine double ionization via three-VUV-photon excitation. The competition and mechanisms of single and double photoionization can be examined by velocity imaging of photoelectrons.

5. Two-color IR-VUV photoionization

By scanning the IR laser frequency with the VUV photoionization fixed below the ionization threshold of a molecular species, the IR spectrum of the neutral molecules can be measured with high sensitivity by detecting the photoions formed due to IR laser excitation. To obtain the absorption spectrum, the photoelectron signal at the ionization threshold can be monitored as the excitation laser frequency is scanned. Since the VUV ALFF is used as the probing photoionization light source, high optical resolution is not needed.

6. Two-color VUV-IR photoionization

Infrared spectroscopy of ions can be studied by using the VUV-IR photoionization scheme. In this experiment, the low resolution VUV ALFF source is used to excite the molecular to high-n Rydberg states prior to IR excitation of the ion core to discrete rovibrational states. The detection of the electron or ion resulting from the IR excitation provides spectroscopic information on the corresponding ion. It has been demonstrated that the IR spectrum is independent over a wide range of n-values. Thus, a high-resolution VUV source is not needed. However, the high VUV intensity offered by the VUV ALFF source is expected to make the IR-VUV scheme highly sensitive.