X-ray Photoionization in the Presence of Strong Optical Fields

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Introduction

A new class of linac-based x-ray light sources is on the horizon in the United States with the commissioning of the Short Pulse Photon Source (SPPS) in May 2003 and the design and engineering of the Linac Coherent Light Source (LCLS), due to produce its first light in 2008. These light sources promise, respectively, 10^8 x-rays in a 80-fs pulse and 10^{12} x-rays in a 230-fs pulse at 8-keV photon energy. Exciting possibilities await because the pulse length is decreased 1000-fold and the intensity is increased by many orders of magnitude relative to the $\approx 10^7$ x-rays in ≈90-ps pulse available at the Advanced Photon Source (APS), a state-of-the-art, third-generation synchrotron optimized for hard x-ray production. For atomic physics, these light sources enable the study of high-field and nonlinear effects at short wavelengths. For chemistry, materials science, and biology, the ultrafast x-ray pulse will probe, on a subpicosecond timescale, atomic-scale motion initiated by a femtosecond laser [1].

Many proposed experiments for the next generation x-ray sources involve laser/x-ray pump-probe techniques on the ≈100-fs timescale. At the compressed timescale, the laser intensity used to initiate a dynamical process can easily approach 10^{14} W/cm^2 (i.e., a field strength of ≈3 volts per angstrom, which is comparable to the field that binds an electron to the nucleus. Therefore, it is important to understand how the x-ray physics of an atom is perturbed by the presence of a high-power laser. We plan to investigate how the field from an ultrafast laser modifies the x-ray photoionization and vacancy decay of an isolated atom. Theoretically, the problem simplifies to the behavior of an atom under the influence of a strong, low-frequency dressing beam (laser) as probed by a weak, high-frequency beam (x-ray) as discussed in the literature for the hydrogen atom [2]. Two major effects have been theoretically predicted and observed for valence electrons: (1) the ponderomotive shift of the ionization threshold [3] and (2) the appearance of sidebands in photoelectron [4] and Auger spectra [5].

Despite the “long” pulse length of 87 ps at the APS, we can already address these issues at intensities relevant to the next-generation sources. A combination of microfocusing of the x-ray beam to $\approx 1 \times 1$ μm^2 and tight focusing of the dressing laser to 10×10μm^2 is required. Clearly, shortening of the x-ray pulse length and tightening of the x-ray focus will extend the parameter space that we are able to access. Our initial goal is to search for the ponderomotive shift of the ionization threshold, which is 6 eV for a 800-nm laser dressing intensity of 10^{14} W/cm^2. This project can only be performed at beamline 7-ID at the APS, where a fully configurable Ti:sapphire-based ultrafast laser system is synchronized to an undulator beamline. The following is a progress report based upon two 5-day singlet mode beam times during 2003 on this technically challenging project.

Materials and Methods

Since we wish to probe the atoms in a well-defined electromagnetic field, the ideal situation is for the laser focus and pulse length to be larger than those of the x-rays. The achievable spot diameter for the x-ray focus using Kirkpatrick-Baez (K-B) mirrors is ≈1 μm [6]. For the laser, the diffraction-limited focused spot diameter, $2w_0 = 2(M^2\lambda f/\pi d)$, is ≈2 μm if a perfect Gaussian beam $M^2 = 1$ and f/4 optics are assumed, but we operate at larger spot sizes to ensure more uniform overlap. The standard operating mode for the laser system (Ti:sapphire oscillator, stretcher, multipass amplifier, compressor) produces ≈0.7 mJ in a 50-fs pulse [7]. However, this does not provide sufficient overlap with the x-ray pulse of 87 ps full width at half-maximum (FWHM). Bypassing the compressor stage yields a chirped pulse of 25 ps FWHM, still insufficient for complete overlap. Therefore, we incorporated a home-built stretcher using a 1200 L/mm grating to seed the amplifier with a pulse having a 30-nm bandwidth. The amplified pulse duration was measured to be ≈120 ps FWHM by using second harmonic autocorrelation. With x-ray and laser pulse lengths of 87 and 120 ps, respectively, a jitter of 20 ps between the two pulses degrades the laser intensity by <7% from the optimally overlapped case. The laser intensity is a critical parameter that we will calibrate with measurements of ion yields in rare gases.

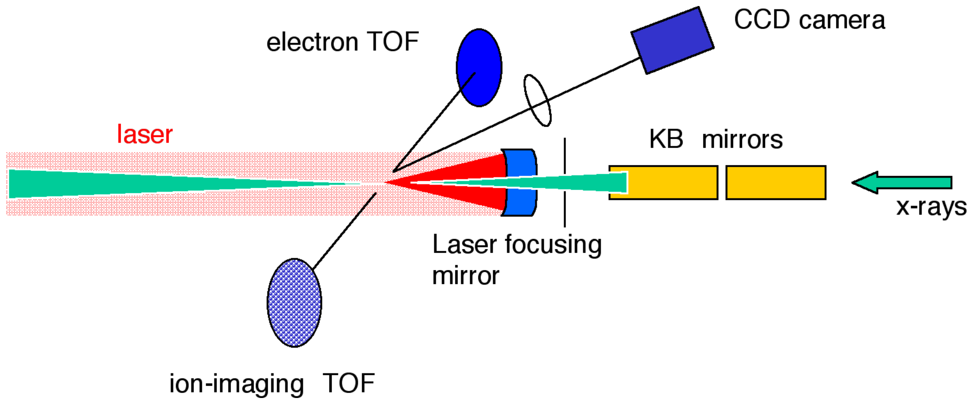

FIG 1. Schematic of the apparatus for overlapping the focused x-rays and laser in a gas jet (not shown). The interaction region is viewed by ion/electron spectrometers and a CCD camera.

We have constructed an atomic beam apparatus that permits the overlap of the laser and x-rays in time and space in an interaction region simultaneously viewed by ion-imaging and electron time-of-flight (TOF) spectrometers and a charge-coupled device (CCD) camera (Fig. 1). The ion-imaging TOF has 4π acceptance, and the electron TOF has 1/1800 of 4π. The x-rays are focused by a K-B mirror pair, and the laser is focused by using reflective optics. The two beams are overlapped in space and time with the assistance of 10-μm crosshairs, scintillating glass, CCD camera, avalanche photodiodes, and the photoelectron spectrometer. Either an in-beam x-ray chopper [8] or pulsed extraction is used to select ions coming from the chosen singlet bunches to reach the detectors. An event-mode data acquisition system is configured to simultaneously record laser-on/laser-off ion (electron) TOF spectra, ion position, the energy of each laser pulse, and the relative time delay between the laser and the x-rays. Recording these parameters allows postexperiment sorting of the data for laser intensity and temporal overlap.

We selected the krypton atom as our target (1) because we have studied it extensively in the weak-field regime by using x-rays from the APS [9, 10]; (2) because a chirped pulse amplified ultrafast Ti:sapphire-based laser system exists at the MHATT-CAT sector (7), which can, in principle, provide ≈1 mJ/100 ps at a repetition rate of 1 kHz; and (3) because the sector 7 undulator beamline is equipped with a Si(111) monochromator that provides excellent flux over an energy range of 8-25 keV (≈10^{13} x-rays/s/0.01% bandwidth at the Kr K edge, 14.326 keV, Kr 1s natural width = 2.75 eV). The focusable x-ray flux is paramount since only 1/5440 of the x-rays can be synchronized with the laser pulses. (In the special timing mode of the APS, we overlap a singlet x-ray bunch, isolated by ±1.6 μs, which contains 1/20 of the total flux and circulates at 272 kHz.)

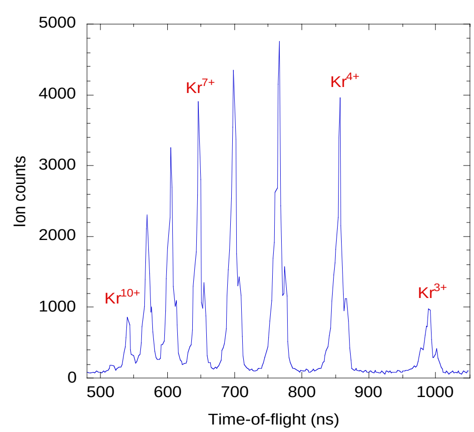

FIG. 2. Representative ion TOF spectrum of krypton induced by the 5-mA singlet bunch. The charge states are well resolved, as is the natural isotopic structure.

Results and Discussion

Preliminary tests conducted at the sector 7 beamline (Fig. 2) demonstrate feasibility. Monochromatized 14.453-keV x-rays were passed through four-jaw slits (320 × 320 μm) onto the K-B mirrors to yield 3 × 10^{11} x-rays per second in a spot size of ≈3 × 2 μm^2. With this flux, we obtained an ion count rate of 10 per second from the singlet bunch at a chamber pressure of 4×10^{-7} torr. A gain of 20× can be realized by increasing the gas density, and a further gain of 7× is expected from use of an improved acceptance K-B mirror pair. Thus, count rates of 1400 Hz for the singlet bunch and ≈5 Hz for laser-on bunches are expected. A laser-on spectrum with 10,000 events will therefore require about 30 minutes. For a complete mapping of the threshold shift, we need to scan the energy of the x-ray photon across the 1-second threshold (10 points) and vary the intensity of the laser beam (three intensities), after ensuring optimal overlap. With in-vacuum, scannable, electrically isolated crosshairs having a 10-μm diameter, we confirmed the spatial overlap of the beams (Fig. 3). X-rays and laser overlap were recorded simultaneously, by using the current to monitor the x-rays and by using the scatter into the CCD to monitor the laser. Temporal overlap was confirmed with avalanche photodiodes (APDs) to better than 100 ps, and it was simply a lack of beam time that prevented considerable improvement over this value.

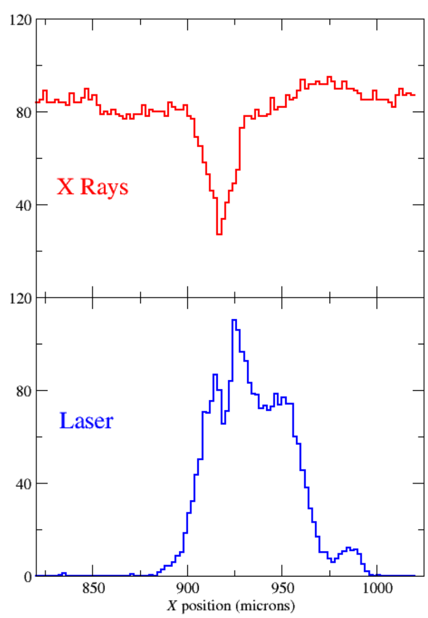

FIG 3. X-ray-induced current and laser-induced scatter as a function of crosshair position.

The initial two beam times have been very successful; we determined the feasibility of the experiment and developed methods to overlap the x-ray and laser pulses spatially and temporally. During the December 2003 singlet timing period of 11 days, we expect to be able to acquire meaningful data for a specific laser intensity. The largest uncertainty is the magnitude of the focused laser intensity. Using the home-built stretcher to seed the amplifier caused a degradation in pulse energy and mode, which may limit the achievable focusable intensity to $\leq 10^{13}$ W/cm^2.

Acknowledgments

We thank the MHATT-CAT staff, D. Arms and D. Walko, for assistance during the beam times. We also thank M. DeCamp, R. Crowell, and D. Gosztola for assistance with the ultrafast laser system. The Argonne and Michigan groups were supported by the Chemical Sciences, Geosciences, and Biosciences Division. Use of the APS was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. The Michigan group was also supported by the National Science Foundation (NSF) FOCUS physics frontier center.

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