Section 2
ADVANCED TECHNOLOGY DEVELOPMENTS

2.A Multiphoton Ionization Experiments

In recent years, a great deal of progress has been made in the study of atomic and molecular matter under the influence of high-intensity radiation. Multiphoton ionization (MPI) studies, in particular, have been greatly advanced in several respects. Multiple-electron ionization of different atomic species has been reported with laser intensities up to $I \approx 10^{17}$ W/cm^2 with pulses lengths from 22 fs to 10–100 ns with wavelengths from 193 nm to 10.6 μm.[1] Nine times ionization of the commonly used atomic target xenon has been reported with both Nd:YLF (1.053 μm) and excimer (248-nm) lasers.

At LLE, we are studying MPI in noble gases at the highest intensities available in the 1-μm-wavelength range. We are using the so-called T^3 (table-top terawatt) laser system,[2] which operates on the principle of chirped pulse amplification (CPA) and produces up to several hundred millijoules in 1-ps pulses at a wavelength of 1.053 μm. We have observed charge states in xenon up to and including Xe^{12+} with intensities of 5×10^{16} W/cm^2 using the T^3 fundamental frequency. Xe^{8+} has been observed at 3×10^{16} W/cm^2 using frequency-doubled, 0.527-μm laser light.

Multiphoton Ionization Experiments

An atom, ion, or molecule can be ionized by the absorption of N or more photons where $N\hbar\omega$ equals the ionization potential

$$N\hbar\omega + A^{+i} \rightarrow A^{+(i+1)} + e^- \qquad (1)$$

Many authors have observed highly charged ion states produced by high-intensity laser pulses. For example, L'Huillier *et al.*[3] observed Ar^{3+}, Kr^{4+}, and Xe^{+4} ions with 50-ps, 1.06-μm, 10^{14} W/cm^2 laser pulses and Perry *et al.*[4] observed Ar^{4+}, Kr^{5+}, and Xe^{+6} ions with 1- to 2-ps, 0.58-μm, 10^{15} W/cm^2 laser pulses. Luk *et al.*[5] observed Ne^{2+}, Ar^{6+}, Kr^{6+}, and Xe^{8+} with a 193-nm ArF laser with a pulse length of 5 ps and peak intensities of 10^{17} W/cm^2. Recently, Rhodes[6] reported He^{2+}, Ne^{4+}, Ar^{8+}, Kr^{8+}, and Xe^{9+}, using a KrF laser at 248 nm, with a pulse length of 0.5 ps and intensities of roughly 10^{16} W/cm^2.

We are carrying out multiphoton ionization experiments using the T^3 laser at 1.053-μm wavelength with an 1-ps pulse duration and intensities up to 5×10^{16} W/cm^2 using *f*/4 optics. Experiments were also performed up to 3×10^{16} W/cm^2 with the second harmonic of the laser wavelength at 0.527 μm.

The laser is focused into a vacuum tank, which has a base pressure of 1×10^{-8} T and is typically filled to 1×10^{-5} T of a noble gas. The ions are detected by applying a bias voltage across the focal region and using a time-of-flight technique to obtain energy resolution. The resolution of the time-of-flight spectrometer (TOF) is sufficient to identify Xe^{13+}. The ions are detected with a dual microchannel plate with a gain of 10^6, and the spectra were recorded using a digitizing oscilloscope. An example of the relative number of argon ions detected versus laser intensity is shown in Fig. 36.25. A complete spectrum, such as this, can be obtained in approximately 3 h of run time.

We have routinely detected Ne^{5+}, Ar^{8+}, Kr^{8+}, and Xe^{9+} ions at the highest intensities. The production of each of these ions requires a last-step ionization potential of 126 eV, 143 eV, 123 eV, and 202 eV, respectively. The production of the next highest charge states Ar^{9+}, Kr^{9+}, and Xe^{10+} require ionization potentials of 422, 231, and 233 eV, respectively, whereas the production of Ne^{6+} requires an ionization potential of 156 eV. The Ar, Kr, and Xe charge states achieved are consistent with the statement that the ionization state achieved depends primarily on the ionization potentials of the atoms. An intensity of 5×10^{16} W/cm^2 appears to be sufficient to ionize atoms or ions with ionization potentials of $\sim$200 eV. This is shown in Fig. 36.26 for argon and krypton, where the appearance intensity, corresponding to the production of $\sim$40 ions of a particular charge state, or 1/250 of the number of ions at saturation of the first state, is plotted versus the ionization potential of that charge state. This figure is similar to one shown by Perry *et al.*[4] and suggests that ionization is independent of the atomic structure. It should be noted that a smooth curve of appearance intensity can also be drawn versus the sum of the ionization potentials of that particular charge state suggesting that such a plot will not discriminate between sequential and direct ionization.

Comparing our results with those of L'Huillier *et al.*[3] at 1.064 μm, we find that our appearance intensities are almost an order of magnitude larger for Ar, Kr, and Xe, whereas they are a factor of 2 larger for He. Chin *et al.*[7] showed that there can be changes in the

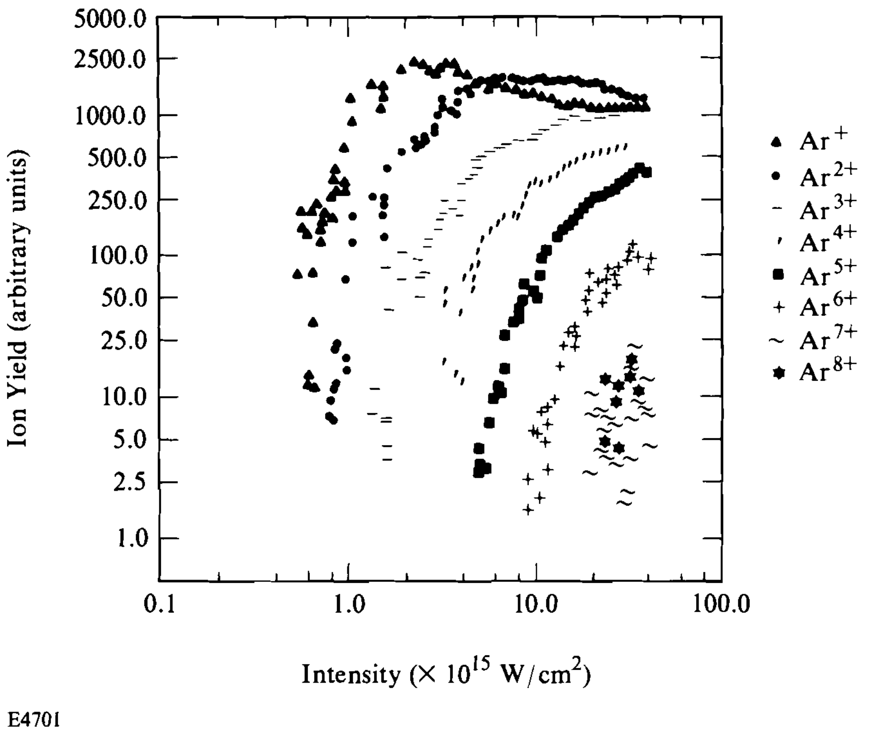


Fig. 36.25
Relative argon ion yield versus 1-μm laser intensity. The pulse width is 1 ps.

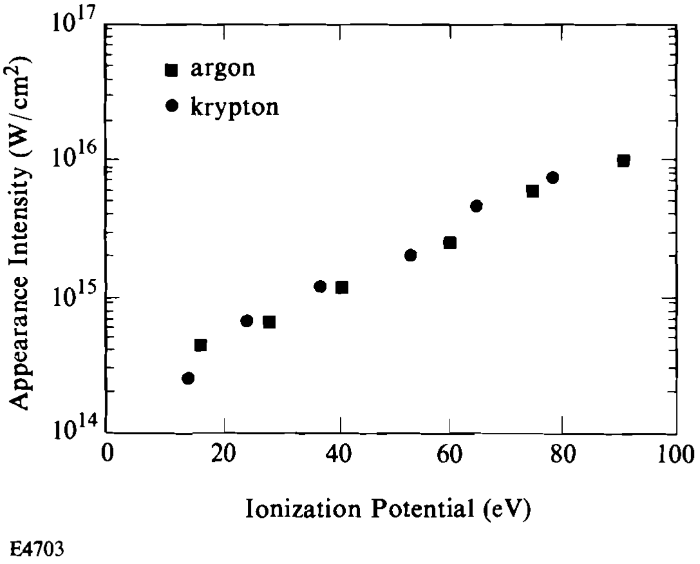


Fig. 36.26
Appearance intensity of various argon and krypton charge states versus the ionization potential of the state.

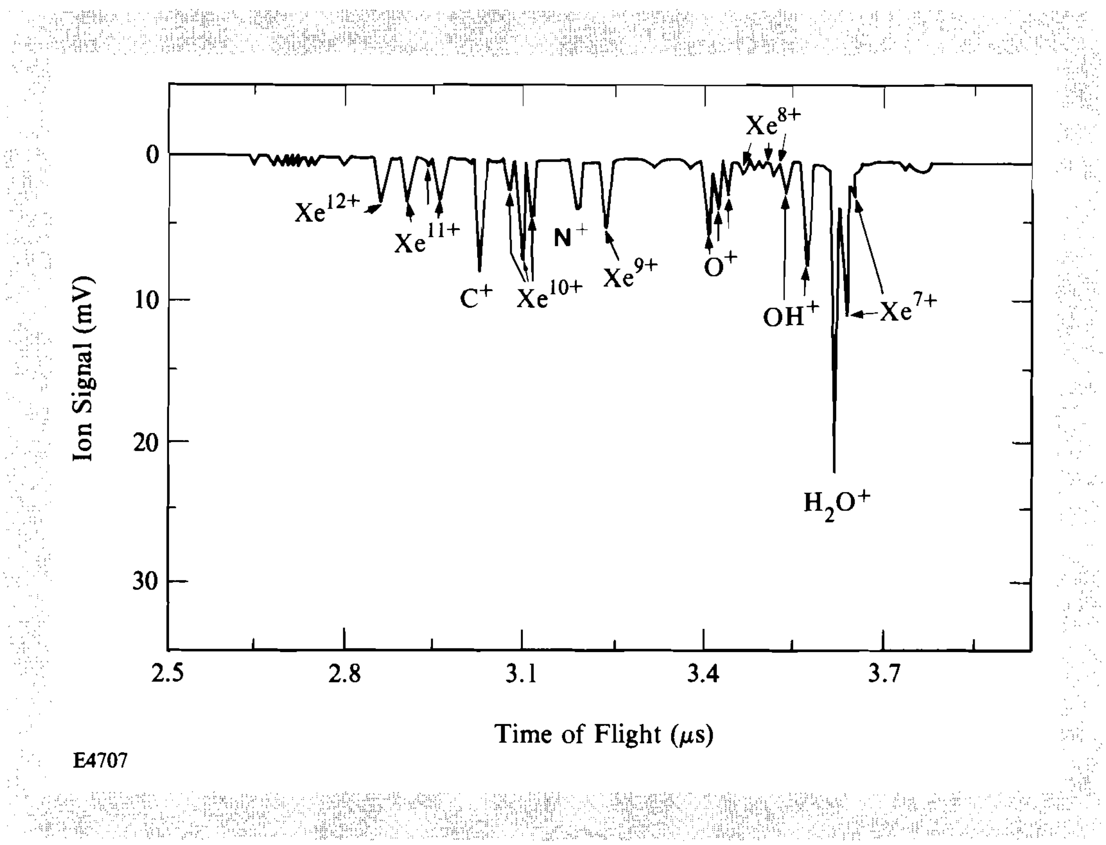
appearance intensity due to pulse-length changes; thus, we expect to find different results for L'Huillier *et al.*,[3] who worked with a 50-ps bandwidth-limited laser pulse.

While the ionization of Ar, Kr, and Xe appears to occur independently of the details of the atomic structure, Ne behaves differently. Based on the charge states achieved in the heavier elements, Ne^{6+} and perhaps Ne^{7+} (ionization potential = 207 eV) should have been observed routinely. Thus, there appears to be a significant difference between the multiphoton ionization in neon compared with the three heavier elements.

On occasional shots, Xe^{12+} was observed. The ionization potential from $Xe^{11+} \rightarrow Xe^{12+}$ is 294 eV and according to Fig. 36.26, this probably requires an intensity of 10^{17} W/cm^2, which may occur due to hot spots in the beam. The TOF spectrum showing Xe^{12+} is shown in Fig. 36.27. The observed Xe^{12+} spectra corresponds to ~ 10 ions. The identification is accurate to within about 3% due to space charge effects in the detected ions. We have also observed Ne^{6+} on occasional shots.

A preliminary experiment was carried out to test the effects of bandwidth versus pulse width. The xenon ion yields were compared for a 1-ps compressed pulse and a 200-ps uncompressed pulse. These two pulses have the same bandwidth and were found to give the same ion yield at the same intensity.

Fig. 36.27
Time-of-flight spectrum of multiphoton ionization of xenon showing Xe^{12+}.



Because of the relatively efficient frequency doubling of the laser light we were able to compare MPI at two different wavelengths. The qualitative results are that at low intensities it is easier to ionize the ions with the shorter-wavelength light, which is consistent with perturbation theory. At the highest intensities it appears that it is easier to ionize the ions with the longer-wavelength light. This is consistent with an increased importance of the pondermotive potential in the laser focus.

Summary

At LLE, we have begun multiphoton ionization experiments using the T^3 laser. This laser is currently the highest-intensity laser operating at 1-μm wavelength, with peak intensities observed up to 5×10^{16} W/cm^2. We have observed charge states up to 12 times ionized xenon with the fundamental frequency, and 8 times ionized xenon with frequency-doubled light at intensities of 3×10^{16} W/cm^2. Preliminary experiments suggest that at the highest intensities, it is easier to ionize the ions with the longer-wavelength light. We are continuing these experiments with emphasis on the effects of pulse width and bandwidth on the ionization process, and a continued comparison of the effects of different wavelengths on the process.

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