Multiphoton Ionization of Xe and Kr with Intense 0.62-μm Femtosecond Pulses

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Multiphoton ionization of Xe and Kr is investigated with intense 0.62-μm pulses of 900, 90, and 22 fsec duration. Deviation from the scaling of lowest-order perturbation theory is observed. For the shortest pulses a modified Keldysh theory closely fits the experimental data for xenon. The importance of dynamic resonances and the shifting ionization potential is discussed.

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Multiphoton multiple ionization of atoms has been extensively studied over a wide range of frequencies and pulse durations.[1] With the recent advance in femtosecond technology these investigations can be extended to much shorter pulses. Ultrashort pulse measurements are of particular current interest for at least three reasons. First, nonlinear optics[2,3] and electron spectroscopy[4,5] show that ionization of xenon and krypton with ultrashort pulses is difficult relative to extrapolations of longer pulse data.[6,7] Second, recent experiments[4] have demonstrated the importance of transient resonances caused by the relatively large ac Stark shifts (for high-lying states the ac Stark shift is equivalent to the ponderomotive potential)[4,8] characteristic of the interaction of 0.62-μm pulses in the intensity range $I > 10^{13}$ W/cm^2 (energy shifts $\simeq$0.35 eV). Qualitatively, one might expect transient resonances to enhance the ionization cross section of otherwise nonresonant multiphoton ionization. However, multiphoton ionization of xenon has been investigated with 2-psec pulses at approximately the same wavelength and the results correspond to a theory that ignores the internal atomic structure.[9] Third, there have been suggestions that nonsequential multiphoton ionization, if significant, will only be observable with pulses shorter than a few optical cycles.[10]

This paper reports the first systematic investigation of the pulse-duration dependence of multiphoton ionization in the femtosecond domain. Well-characterized 900-, 90-, and 22-fsec (full width at half maximum) 0.62-μm pulses are used. These pulses range from the duration at which transient resonances have been reported[4] (900 fsec) to the limit of current high-power ultrashort-pulse laser technology[11] (20 fsec). Ionic species as high as Xe^{5+} are observed and the ion curves are compared with results of a modified Keldysh theory.[12]

Laser pulses were produced by amplification of the output of either a spectrally filtered[3] synchronously pumped dye laser (900 fsec) or a colliding-pulse mode-locked dye laser (90 fsec). The temporal, spatial, and spectral characteristics of the pulses have been fully described previously.[3,13] The wavelength of the 900-fsec pulse was centered at 616 nm and its bandwidth was slightly greater than the transform limit ($\Delta t \Delta \nu = 0.52$). The 90-fsec pulse was centered at 625 nm and had $\Delta t \Delta \nu = 0.5$. The pulse durations were measured by autocorrelation and fitted by a sech^2 (90 and 22 fsec) or by a Gaussian (900 fsec) pulse shape. After amplification, the 90- and 900-fsec pulses were spatially filtered[13] to ensure diffraction-limited beam profiles.

The 22-fsec pulses were created from the 90-fsec pulses with large-aperture pulse-compression techniques.[11] The resulting 100-μJ pulses were diffraction limited with a signal-to-background power contrast ratio of approximately 30:1 (5:1 in energy). Compensation for the dispersion in all optical elements (lenses, windows, beamsplitters, etc.) was accomplished by our predispersing the pulse. Thus, the pulse measured 22 fsec only in the target chamber and at the autocorrelation crystal. Since the 350-Å bandwidth[13] of the 22-fsec pulse gives rise to serious chromatic aberration in a single-element lens, an achromatic lens ($f = 14.3$ cm) was used to focus the pulses into the vacuum chamber (and onto the autocorrelation crystal).

All focal-spot measurements were made by either scanning a 5-μm pinhole (900- and 90-fsec pulses) through the focus or by observation of the portion of the energy transmitted through a pinhole (22-fsec pulse) of known diameter. Within the accuracy of the scans, the beam profiles were Gaussian. The optical system was unchanged between the Xe and Kr measurements, ensuring that the relative intensities are accurately known for each pulse duration.

The background pressure of the vacuum cell was $\simeq 5\times10^{-9}$ Torr. The operating pressure during the experiment was $\leq 4\times10^{-6}$ Torr. Ions were extracted with an $\simeq$80-V/cm static field into a time-of-flight mass spectrometer.

A microcomputer, coupled to a boxcar integrator, was programmed to accept only laser pulses within a narrow energy range ($\pm$2.5%). The computer recorded and averaged the associated ion signals. The intensity in the vacuum chamber was varied by rotation of a $\lambda/2$ plate placed in front of a polarizer (reflection from a germanium plate was used as a dispersion-free polarization selec-

tor for the 22-fsec pulse).

Figure 1 is a graph of the number of ions as a function of the peak laser intensity for both rare gases and all three laser-pulse durations. (Some ionization states were observed over too small an intensity range to plot. They are indicated on the graph by arrows showing their appearance intensities.) The solid curves were obtained from a modified Keldysh theory.[9,12] Although we did not measure the absolute number of ions, we estimate the threshold sensitivity of our ion collector to be less than 10 ions. In Fig. 1, the relative scaling between experimental (right scale) and calculated (left scale) ion signals is consistent with this estimate.

We have applied the same criteria to fit the Kr and the Xe data. For xenon the calculated number of ions as a function of the peak laser intensity fits the experimental data remarkably well. The theory does not agree as well with the experimental data for krypton. However, the fit

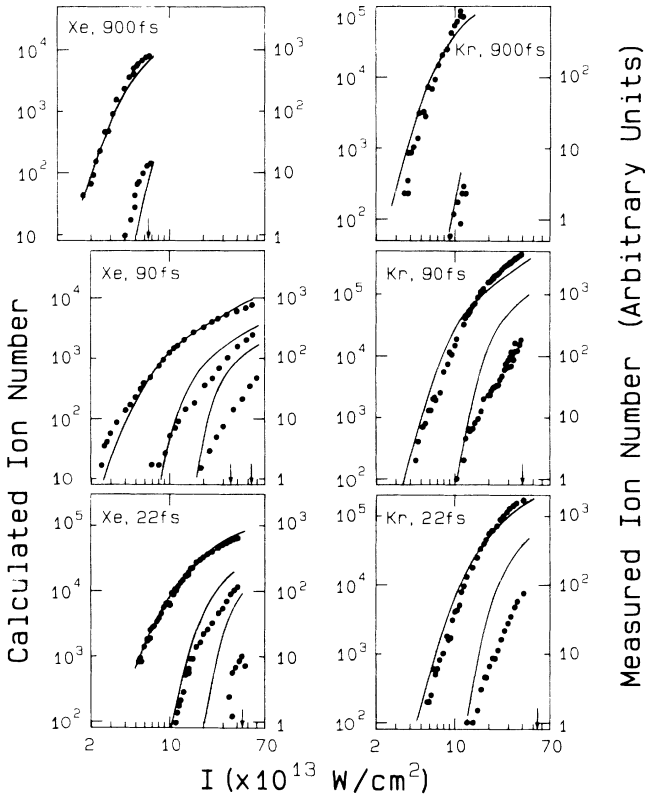

FIG. 1. Ion yield of Xe and Kr. The solid curves are calculated from a modified Keldysh theory (Ref. 12). The calculations give an absolute number of ions for the measured focal geometry and the neutral-gas density. The experimental number of ions is plotted in relative units. The standard error of the experimental data is too small to plot. The scatter of the data is an indication of the standard error. The left curve on each panel is the singly ionized species. The ion species increases by one for each adjacent curve. The arrows indicate the appearance intensity for ion species that were observed over too small an intensity range to plot.

(not shown) becomes comparable to that of xenon (and the relative scaling is more consistent with the sensitivity of the electron multiplier) if the energy scale of the theoretical or experimental data is changed by 25% (the same for all three pulse durations). There is no experimental justification for this procedure without an equivalent change for Xe. The intensities of krypton relative to xenon are accurately known ($< \pm 5\%$) since the pulse durations and focusing optics are the same.

We have also compared the data with the original Keldysh theory[14] (Fig. 2). Although many of the qualitative features observed experimentally for the lowest ionization state are predicted, and the ionization rates for Xe^+ production are close to the experimental values, the unmodified Keldysh theory does not fit the experimental data as well, especially for the highly ionized species.[9] Even for the modified theory the agreement in Fig. 1 between experiment and theory is not as good for the higher ionization states as it is for the singly ionized species. Not only do the intensities at our detection threshold differ but, more importantly, the slopes of the curves differ as well. It is known from tunneling theory that the ground-state wave function strongly affects the tunneling probability of highly ionized Xe.[15] The deviations between theory and experiment that we observe for high ionization states may reflect the use of inappropriate ground-state wave functions in the numerical calculations.

A 22-fsec pulse (only 11 optical cycles) with its associated high saturation intensity ($> 10^{14}$ W/cm^2) should produce nearly optimum conditions for nonsequential ionization. A necessary, but not sufficient, condition for nonsequential ionization is the observation at the same (below saturation) intensity for both the parent and daughter species. Experimentally we see, for example, the presence of both neutral and doubly ionized xenon at the same intensity for both 22- and 90-fsec pulses. However, the modified Keldysh theory (solid curves in Fig.

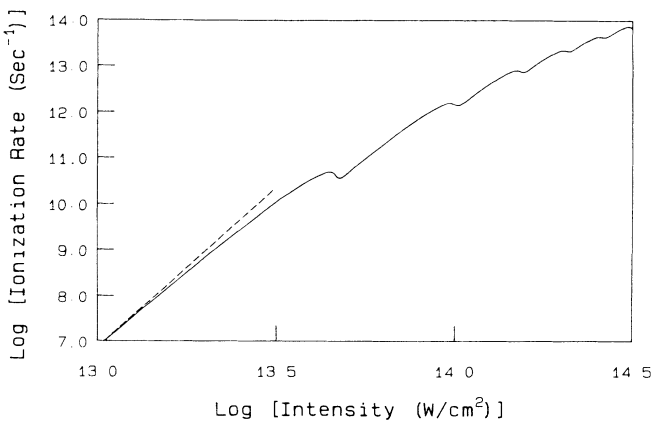

FIG. 2. Graph of the ionization rate of neutral xenon as a function of the intensity for 625-nm radiation as calculated from Eq. (1) of Ref. 14. The dashed line shows a slope of 7.

1), which assumes sequential ionization, also predicts the simultaneous presence of all three species at these intensities. Although on purely experimental grounds we cannot eliminate the possibility of nonsequential ionization, we do not see a significant enhancement of highly ionized species compared with predictions of a sequential model.

In view of the transient resonance[4] induced in the medium by high-intensity light, there is a striking agreement between the experimental results for xenon and a theory that assumes a structureless atom. Small deviations appear only at relatively low intensities. Thus, it seems that transient resonances can play a significant role in the overall ionization rate over at most a limited intensity or time range.

To understand this observation, consider just how transient these resonances can be . Assuming that all high-lying states move with the ponderomotive potential,[4] we can write the maximum rate of change of the ponderomotive shift as

$$(dU/dt)_{\max}=2\sqrt{2}U_0[\ln(0.5)^{1/2}/T\exp(0.5)],$$

where U_0 is the maximum value of the ponderomotive shift during the pulse and T is the full-width at half-maximum duration of a Gaussian pulse. In the case of the 90-fsec pulse with a characteristic peak intensity of 10^{14} W/cm^2, $(dU/dt)_{\max}=0.1$ eV/optical cycle. In the even more extreme case of the 22-fsec pulse, the same peak intensity gives $(dU/dt)_{\max}=0.4$ eV/optical cycle.

The significance of such large ponderomotive shifts can be seen by consideration of a two-level system. For a two-level system, the pulse-duration and intensity dependence of the dephasing between the transition (ω_{ab}) and the nearly resonant harmonic of the laser frequency can be estimated. For a constantly shifting transition, $\omega_{ab}+(dU/dt)t/\hbar$, the dephasing time ($\tau$) is given by the condition that $\delta\phi\simeq 2\pi$. That is $\pi\simeq[2h/(dU/dt)]^{1/2}$, where dU/dt is assumed constant. If $dU/dt=(dU/dt)_{\max}$, then $\tau\simeq 13$ fsec for a 90-fsec pulse at 10^{14} W/cm^2 and 6 fsec for a 22-fsec pulse of the same intensity, i.e., dephasing occurs in only a few optical cycles. Resonances that last only a few cycles are hardly resonances at all and can be expected to have only minor effects on the overall ionization rate. Only for relatively small dU/dt can transient resonances play an important role. They may account for the small deviation of the experimental and calculated curves observed in the 900-fsec and the low-power 90-fsec xenon results.

The above discussion does not imply that transient resonances will not be observable in the electron spectrum, perhaps even for large dU/dt. The electron spectrum is more sensitive to relatively small changes in the ionization rate. Transient resonances should also have observable nonlinear optical signatures. In fact, nonlinear optics may provide a good method of observation of transient resonance through, for example, the bandwidth and time dependence of the harmonic radiation.

The slopes of the ion curves (Fig. 1) change as the pulse duration falls from 900 to 22 fsec. From the point of view of perturbation theory, the slopes in Fig. 1 imply the order of the process. For neutral xenon and krypton, the lowest possible order is 7 for 625-nm light. The 900-fsec data for both gases is adequately fitted by a slope of 7 while the characteristic slopes of the shorter-pulse curves is approximately 4 and 5 for xenon and krypton, respectively. The slower rise in the ion number with intensity is not a result of a four-photon process as can be seen from a plot of the Keldysh ionization rates (Fig. 2) for Xe^+. (Keldysh theory adequately describes neutral Xe ionization.) For low intensities, the ionization rate increases approximately as I^7 (dashed line). When the ponderomotive potential is sufficient to shift the minimum energy necessary for ionization to near the eight-photon limit, the ionization rate levels off and then falls as the seven-photon channel is blocked ($\simeq 6\times 10^{13}$ W/cm^2). At higher intensity, the ionization rate resumes its rapid rise but levels and falls again as the eight-photon channel closes. The slope of 4 observed for the 90- and 22-fsec pulses is the result of our averaging over the spatial and temporal profile of the beam. (Clearly the saturation intensity cannot be usefully defined as the intersection of the 7- and 1.5-power-law extrapolations of the experimental data when ultrashort pulses are used.)

Of course, Keldysh theory uses a $1S$ hydrogenic ground-state wave function. Qualitatively similar, but perhaps more complex, behavior will occur with a more appropriate wave function.

In summary, we have observed multiphoton ionization using ultrashort pulses containing as few as 11 optical cycles. The high "saturation intensities" characteristic of ultrashort pulse interaction with Xe and Kr are consistent with previous experiments.[2,3,5] Remarkable agreement is found between the experimental data for the production of singly ionized xenon and a calculation based on a modified Keldysh theory. Dynamic resonances have little effect on the overall ionization rate for ultrashort pulses. From the perspective of perturbation theory, the main feature that affects the intensity dependence of the ionization rate is the shift of the effective ionization potential with the ponderomotive potential of the light.

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