Rydberg Excitation in Xenon by the Rising Edge of a Femtosecond Laser Pulse

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Photoelectron spectra in xenon are obtained for (6+1)-photon ionization with intense laser pulses ($\lambda = 594$ nm, $I = 5\times10^{13}$ W/cm^2). By comparing spectra obtained with 300-fs and 600-fs pulses it is shown that the ground state is depleted by nondiabatic passage through high lying Rydberg states (nf) during the rise of the pulse. Spectra are well reproduced by a simple theory including both six-photon excitation to the Rydberg states and one-photon ionization out of these states.

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Since the first observation of resonances in the photoelectron spectrum from multiphoton ionization of xenon [1] in 1987, a large number of experiments with atoms and intense femtosecond laser pulses have been carried out [2]. In the seven-photon ionization process of xenon with femtosecond pulses ($\lambda \sim 600$ nm, linear polarization) Rydberg levels at the six-photon level play an important role. The ac Stark shift of the Rydberg levels in the intense laser field is nearly equal to the shift of the ionization threshold, while the ground-state shift is negligibly small (see Fig. 1). During the pulse the intensity changes and thereby the shift of the Rydberg states. When the intensity is such that the energy difference between the ground state and a shifted Rydberg state equals the energy of six photons (I_{res}), population can be transferred resonantly. Freeman *et al.* have shown [1] that such resonances enhance the photoionization signal: There are peaks in the photoelectron spectrum corresponding to one-photon ionization out of the Rydberg states.

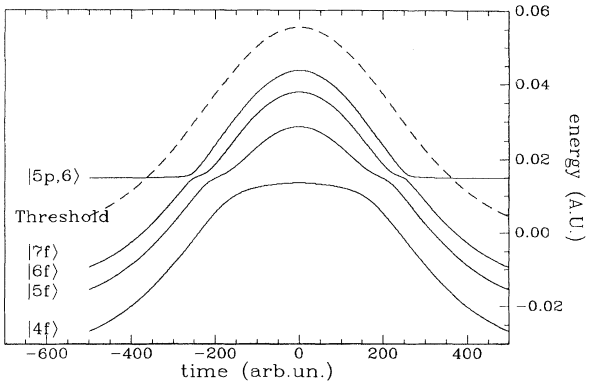

FIG. 1. Energy level diagram as a function of time, with a Gaussian laser pulse that peaks at $t=0$. The drawn levels are the eigenenergies at constant intensity. The ground state dressed with six photons lies above the ionization threshold at zero intensity. At nonzero intensity the ionization threshold shifts up in energy due to the ac Stark shift. The high lying Rydberg states gain nearly the same energy as the ionization threshold. The shift of the ground state is negligibly small. At higher intensities successive Rydberg states undergo avoided crossings with the dressed ground state. Since the laser pulse has a finite rise time these crossings are not passed totally diabatically and population can be transferred into the excited state. The population that is transferred into the Rydberg state is subsequently ionized by one-photon ionization during the rest of the pulse.

A still open question is *when* the excitation of the Rydberg states actually takes place. Intuitively it is expected that most of the population transfer occurs at the peak of the pulse, because the resonance time is the longest there [3]. As a consequence it was argued that the photoelectron spectrum from multiphoton ionization is dominated by those regions in space where the resonant intensity (I_{res}) of one of the Rydberg states corresponds to the local peak intensity.

In this paper we present experimental evidence that this scenario does not always apply. We present data in which the photoelectron spectrum from (6+1)-photon ionization of xenon is strongly influenced by resonant population transfer during the rising edge of the pulse, instead of at the peak of the pulse.

We tuned our femtosecond laser system [4] to a central wavelength of 594 nm, and obtained femtosecond photoelectron spectra of xenon with a magnetic bottle spectrometer (for experimental details see, e.g., Ref. [4]). By changing the bandwidth of the pulses we varied the duration of the pulses ($t_p \sim 1/\Delta\omega$). Great care was taken to minimize the Fourier product $\Delta\omega t_p$ in order to avoid effects induced by frequency chirp of the pulses [5]. We present photoelectron spectra obtained with 300-fs and 600-fs pulses without chirp. We will show that the dramatic differences can only be explained by assuming that a substantial amount of population is transferred to the Rydberg states during the rise of the pulse.

In Fig. 2 the photoelectron spectra obtained with 300-fs pulses at various peak intensities are presented. At the highest intensities the well known signatures of the $4f$, $5f$, and $6f$ resonances are observed (see, e.g., [2]). The relative peak intensity was determined by measuring the energy of each shot (about 100 μJ/shot). In principle the absolute intensity can now be obtained by measuring the pulse duration (300 fs) and determining the focal spot

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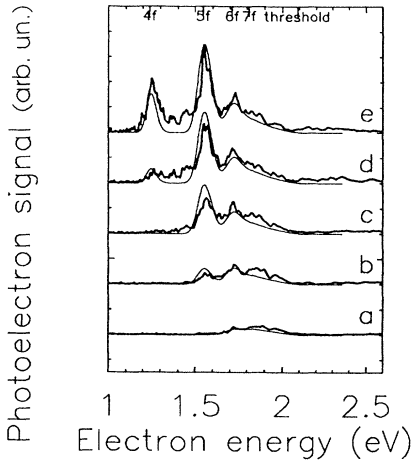

FIG. 2. Photoelectron spectra from (6+1)-photon ionization of xenon with 594 nm, linearly polarized light. The five bold solid traces show the experimental data with a 300-fs pulse at increasing peak intensities. The theoretical positions of the photoelectron peaks originating from the various Rydberg states that act as intermediate resonances are indicated on the horizontal axis. At higher peak intensities more deeply bound states shift into resonance with the six-photon-dressed ground state. The thin solid curve is the theoretical spectrum calculated with the model based on Landau-Zener crossings as described in the text. The labels a–e correspond to peak intensities of a, 2.9×10^{13}; b, 3.5×10^{13}; c, 4.0×10^{13}; d, 4.5×10^{13}; e, 5.1×10^{13} (W/cm^2).

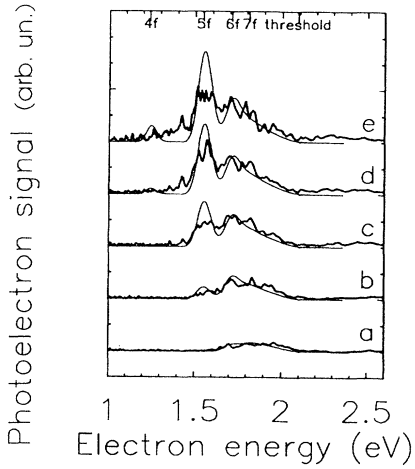

FIG. 3. Photoelectron spectra from (6+1)-photon ionization of xenon with 594 nm, linearly polarized light. The conditions are the same as described in Fig. 2, except for the pulse length which is now 600 fs.

size (radius $\approx 20\ \mu$m). From these numbers we estimate the intensity to be 8.3×10^{13} W/cm^2. However, the absolute intensity can be obtained much more accurately by inspection of the photoelectron spectra. The intensities at which the various peaks shift into resonance and thus become visible in the spectrum can be calculated. Using these intensities and identifying the peaks in the spectrum we were able to determine the actual peak intensity within 15%.

By reducing the spectral width of the femtosecond pulses, the pulse duration was doubled to 600 fs. In order to obtain the same peak intensities (within 15%) the energy of the pulses was also doubled. The photoelectron spectra obtained with the 600-fs pulses are presented in Fig. 3. The traces with the corresponding labels have approximately the same peak intensity. For the low traces (a,b) the electron spectra are similar to those obtained with the short pulses in Fig. 2: By increasing the peak intensity the signatures of lower Rydberg nf resonances appear as expected. Note that the $6f$ resonance appears in both trace $2a$ and $3a$ and the $5f$ resonance appears in both trace $2b$ and $3b$ indicating that the intensity is very similar indeed.

However, a remarkably different behavior is observed as we increase the intensity even further (traces c–e). With the longer pulse the low-lying $4f$ Rydberg state does not show up in the photoelectron spectrum and the $5f$ resonance remains somewhat less pronounced. From the short pulse spectrum we know that in certain regions of the focus the atoms are exposed to a sufficiently high intensity (3.8×10^{13} W/cm^2) to shift the $4f$ state into resonance. From its absence in the photoelectron spectrum we conclude that the ground state was depleted before this intensity was reached: In these regions the excitation of the Rydberg states must have occurred during the rise of the pulse.

We will first give a qualitative explanation of the observed absence of the $4f$ resonance. As can be seen in Fig. 1 the ground state dressed by six photons crosses the Rydberg series while the intensity of the pulse is increasing. As a result of the coupling of the ground state with the Rydberg series (approximately proportional to $n^{-3/2}$) none of the crossings is passed purely diabatically: some population is transferred to each Rydberg state. The mount of transferred population depends on the matrix element, the intensity, the rate at which the ground state is swept through the resonance, and the ground state population present as the resonance is reached. For the 300-fs pulse the rate of increase of light intensity was sufficiently fast that not all amplitude was transferred to the high lying Rydberg states, and some of the ground state population survived up to the peak of the pulse. Population of the $4f$ state was still possible near the peak of the pulse.

However, for the 600-fs pulses the high lying Rydberg states (in resonance well before the peak of the pulse) are crossed sufficiently slowly to become significantly populated. The successive population of these states is efficient enough to deplete the ground state by the time the $5f$ resonance is passed.

In order to confirm this qualitative explanation we have modeled the population of the Rydberg series by calculat-

ing the probability to populate the excited state for each crossing of the six-photon-dressed ground state with a Rydberg state. This probability is given by [6]

$$p_n = 1 - \exp\left(-\frac{2\pi|V_n|^2}{\partial\omega/\partial t}\right)$$

in which V_n is the six-photon coupling of the ground state to the excited state and ω is the energy difference between the six-photon-dressed ground state and the excited state. This energy difference is directly related to the intensity through the ac Stark shift. The singularity for the case where the peak intensity I_{peak} equals the resonance intensity I_{res} ($\partial\omega/\partial t|_{I_{\text{res}}}=0$) is avoided by including a term with the second derivative of ω with respect to time in the denominator (see Van Woerkom *et al.* [3]).

The various V_n approximately obey a scaling law: $V_n = \beta n^{-3/2}$ with β a constant. We varied β to get agreement with the experimental data. We used 200 f states from the $4f$ up to the $203f$ state and took into account both the rising and the falling edge of the pulse to reproduce the spectra. In principle such a combination of two processes would give rise to interference. We can neglect these interference effects between the population transfer at both crossings since they vanish in averaging over the focus.

The extension of the focus along the direction of propagation was cut off due to the geometry of our electron spectrometer. The situation is thus accurately described by averaging over a focal intensity distribution which is constant along the direction of propagation, Gaussian in the plane perpendicular to this axis, and Gaussian in time.

In calculating the photoelectron spectrum we do not assume immediate ionization of the transferred population, as Van Woerkom *et al.* do [3], since the signal from the high Rydberg states (which are hard to ionize) does not agree with such a model. We had to explicitly calculate the fluence that an excited state is exposed to after having been populated. This fluence determines which fraction of the transferred population is actually ionized. We used hydrogenic couplings from the Rydberg states to the continuum since f states of xenon have a negligible quantum defect. The results of the simulation are shown as thin solid curves in Figs. 2 and 3.

In summary, by comparing 300-fs and 600-fs photoelectron spectra of (6+1)-photon ionization of xenon, it can be concluded that at least in some cases a considerable amount of population is transferred to states that shift into resonance well before the peak intensity is reached. The actual amount of population transfer depends on the rate at which the resonances are crossed, the field strength at which the states shift into resonance (determined by the static detuning of the excited state with respect to the dressed ground state), and the coupling of the state with the ground state. Note that the photoionization does not occur instantaneously, and even a large fraction can survive the pulse [7]. The ground state is depleted, and a fraction of the population is trapped in Rydberg states rather than photoionized.

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Note added.— While preparing this manuscript we became aware of a similar experiment [8]. Story, Duncan, and Gallagher show that population is transferred during the rise of the pulse although their experimental circumstances were different. A (2+1)-photon ionization scheme in potassium was used. Since this is a lower order process, the involved intensities are also somewhat lower (8×10^{12} W/cm^2 vs 4×10^{13} W/cm^2 in Xe). Furthermore in potassium the first excited state is below the virtual one-photon level and the ground-state shift dominates over the Rydberg state shift. The net ac Stark shift of the Rydbergs in potassium is opposite to the shift in noble gases such as xenon. As a consequence at higher intensities in potassium the continuum shifts into resonance whereas in xenon at higher intensities more deeply bound states shift into resonance. It is remarkable that under such different experimental conditions still the same mechanism plays a role: So much population is transferred during the rise of the pulse that the ground state is depleted before the peak intensity is reached.

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