Multiphoton Ionization of Xenon with 100-fs Laser Pulses

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Seven-photon ionization of xenon is investigated for the first time with 100-fs, very intense (1.7×10^{14} W cm^{-2}) pulses. The electron spectra show a large number of peaks due to above-threshold ionization. Because of the short pulse duration, large threshold shifts (up to 5.6 eV) are directly observable in the electron spectra. The magnitude of the ionization signal reveals that the ionization probability is much lower than expected at such a high intensity. By a linking of the observed shift to the laser intensity, the latter can be obtained with high accuracy.

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Multiphoton ionization (MPI) has a very low probability and is observable only if the atom is submitted to an intense electromagnetic field. As a consequence, MPI usually takes place in a strongly distorted atom. If the atom can be ionized by absorption of at least N photons, this N-photon transition usually goes along with a number of "higher-order" effects. Above-threshold ionization (ATI), the process in which a number of photons are absorbed in excess of the minimum required by the ionization potential, is one of these effects. The resulting electron-energy spectra consists of a series of peaks evenly spaced by the photon energy. Another effect of the intense light on the energy levels of the atom is the so-called light shift or ac Stark shift, which distorts the energy spectrum[1] of the atom. This distortion is particularly important for the continuum states, which are upshifted by the so-called ponderomotive potential. This "potential" is, in fact, the average kinetic energy associated with the forced oscillatory motion of the free electron inside the electromagnetic field $E_q=(eE)^2/4m\omega^2$. Apart from the energy in the drift motion of the electrons, the ionizing field has to supply this "quiver energy" as well, leading to smaller than expected drift energies.

Although the ponderomotive potential can be very large (3.4 eV at 10^{14} W cm^{-2} and 620 nm), its effects on the electron-energy spectra were not directly observed until recently. As it was pointed out by Muller and coworkers[2] the reason for this is that the energy in the oscillatory motion of the photoelectrons tends to be transferred to the drift motion when the electron travels through the region of strong intensity gradients at the perimeter of the laser focus.

It was already anticipated by Kibble[3] that the situation is drastically different if the pulse duration is so short that the light pulse is switched off before the photoelectron can move appreciably. More precisely, as shown in experiments by Agostini *et al.*[4] and Freeman *et al.*,[5] the electron in such a situation only partly recovers the quiver energy. For extremely short pulses this recovered part approaches zero and we measure the drift energy as it really was inside the light beam.

Energy shifts have also been reported by Luk *et al.*[6] for the MPI of xenon at 248 nm. Other measurements of electron spectra generated with subpicosecond pulses have been reported by Freeman *et al.*[5] Energy shifts are clearly observed in this experiment.

The current paper presents observations of ATI spectra obtained with femtosecond pulses over a significant intensity range and displaying the whole above-threshold ionization phenomenology: a large number of excess photon peaks, peak suppression, and large shifts. The electron measurements are complemented by measurements of the total ionization rate, performed by our monitoring the Xe^+ and Xe^{2+} ion signals as a function of the laser intensity.

The experiment uses the following setup: A pulsed laser beam is focused into a cell with static xenon gas of adjustable pressure. The laser consists of a colliding pulse mode-locked oscillator with intracavity group-velocity compensation[7] that can generate pulses as short as 40 fs, and four Nd-doped yttrium aluminum garnet (Nd:YAIG) pumped dye amplifiers, separated by saturable absorber jets to suppress fluorescence.[8] The central wavelength of the light is 620 nm, corresponding to a photon energy of 2.00 eV. Temporal broadening due to group-velocity dispersion in the amplifiers is compensated for by means of a four-prism arrangement. This sys-

tem produces bandwidth-limited pulses with a duration of typically 100 fs and an energy up to 1 mJ. The beam was focused into the vacuum chamber by a 15-cm focal-length spherical lens. A combination of a half-wave plate and a Glan polarizer was used to define the polarization and to vary the power of the laser.

The background pressure inside the vacuum chamber was less than 2×10^{-8} Torr. The pressure of the xenon gas was varied in such a way that not more than around ten ionization events per laser shot were detected.

The electron spectra were recorded with a magnetic bottle spectrometer, with acceptance angle 2π sr. Therefore the measurements are free of systematic errors due to effects of the laser light on the angular distribution of the photoelectrons[9] and of space charge. The energy scale of the spectrometer was calibrated by means of the well-known ATI spectrum of xenon at 532 nm, using the nanosecond pulses from the Nd:YAlG pump laser. Ions charge spectra were obtained through a time-of-flight spectrometer.

A number of electron-energy spectra are obtained at pulse energies varying from 11–35 μJ and are displayed in Fig. 1. Note that the structure due to ATI is clearly observable in all spectra exactly as with longer pulses: discrete peaks spaced by 2 eV. In this energy range, the number of excess photons increases from one to about ten. The peaks in Fig. 1 are clearly shifted in energy, but to determine by how much they have to be assigned first.

Firstly, the Xe ions can be left either in the $P_{3/2}$ or in the $P_{1/2}$ state, which are separated by 1.3 eV as a result of spin-orbit interaction. The doublet structure with the splitting of 1.3 eV is clearly visible in spectrum (a) and (b) in Fig. 1. Thus the high-energy (large) peak of the doublet corresponds to the low-energy Xe^+ state, i.e., $P_{3/2}$. This fits with the observation that nanosecond pulses (too long to give shifts) unambiguously produce ions almost exclusively in the $P_{3/2}$ state, probably because of resonance enhancement by a Rydberg state with a $P_{3/2}$ core after absorption of six photons.

After identification of the main peak recorded at the lowest intensity as a $P_{3/2}$, we need to determine the corresponding excess photon order S to complete the assignment. If we assume $S=0$, then it is shifted by 0.3 eV towards the high energies. There is no identified mechanism to account for a shift in this direction, and we con-

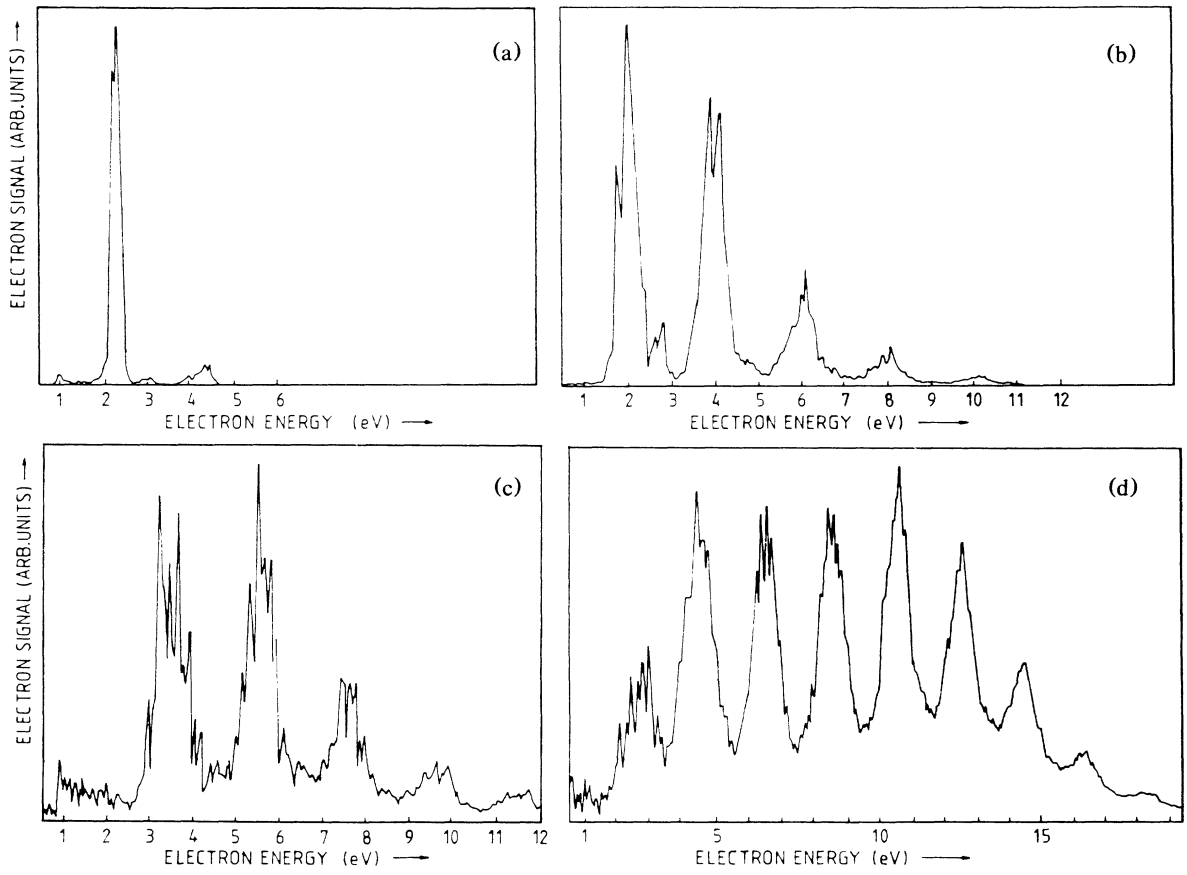

FIG. 1. Electron-energy spectrum measured after multiphoton ionization of xenon with 620-nm light pulses of 100 fs duration. The energy of the pulses is 11 μJ for spectrum (a), 24 μJ for spectrum (b), 29 μJ for spectrum (c), and 35 μJ for spectrum (d).

clude that $S \geq 1$. (The $S=0$ peak unfortunately falls below the low-energy transmission cutoff of our spectrometer.) The choice $S=1$ and the subsequent assignment for the other spectra is based on the following guide lines: the continuity and the relative-intensity dependence. The energy shifts which are reported in Fig. 2 are deduced from such assignments, the decisive justification of this choice being the linear intensity dependence. Any other assignment would produce a nonlinear dependence. The minimum shift observed is 1.6 eV, and at the maximum intensity, it is 5.6 eV so that two ATI peaks have been suppressed.

At the lowest intensity the peak width is about 0.25 eV. This broadening is very small compared to the observed shifts. This is, however, to be expected because of the large nonlinearity of the process: Only intensities close to the maximum intensity contribute to the ionization process. Therefore, not much broadening is expected from the spatial inhomogeneity of the intensity.

The linear dependence of the shift on intensity is expected from the ponderomotive mechanism of peak suppression, and the proportionality constant is known. It is the sum of the atom-independent quiver energy E_q and the small difference in ac Stark shift of the atomic ground state and the ionic state. The atomic Stark shift is 15.1%, [10] while the ionic Stark shift is estimated to be at least 11% of the quiver energy. So the shift of the observed ionization potential is at most 4% more than the quiver energy, a difference which we will neglect.

A direct estimate of the peak intensity can be obtained from the measured values of pulse energy and duration and an estimate of the focal size based on the characteristics of the spatial filter. It reasonably agrees with the one deduced from the shifts. This fact supports the peak assignment, which provides an absolute intensity scale on the right-hand axis in Fig. 2. Intensities deduced from the shift have an accuracy of a few percent. This could be easily reduced to 1% by our assuming that the relevant Stark shifts are known, a formidable improvement over the direct estimate.

The intensity corresponding to the maximum observed shift is 1.7×10^{14} W cm^{-2}. The large average number of excess photons absorbed also indicates an intensity of this order. When compared to a "typical" seven-photon-ionization saturation intensity (defined as the intensity that corresponds to an ionization probability of one for the considered pulse duration), 1.7×10^{14} W cm^{-2} is a large value. Nevertheless, the total electron count per laser shot compared to the total number of atoms in the interaction volume is small, indicating that we are far from saturation. The intensity dependence of the production of Xe^+ and Xe^{2+} is shown in Fig. 3. Xe^+ yield initially has the power-law dependence characteristic of unsaturated signals, with exponent 6.6 ± 0.5. The fraction of double ionization obtained at saturation is about 5%. Above saturation, the number of Xe^{2+} increases again, because of the sequential double-ionization process. At the highest intensity used to record the electron spectra, the fraction of Xe^{2+} never exceeded 5% of the Xe^+ ions, which supports the conclusion that at 1.7×10^{14} W cm^{-2} the ionization is not saturated. A number of qualitative arguments can be advanced to explain this: The use of shorter pulses compared to previous experiments [11] reduces the saturation intensity (which is proportional to the seventh root of the pulse duration), but only by a factor of 2. A more profound

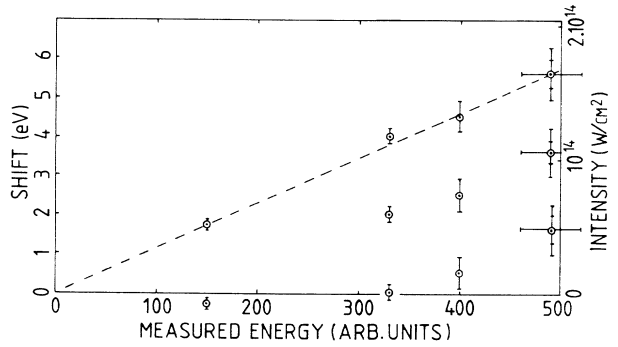

FIG. 2. Shift of the observed spectra (left-hand scale) as a function of measured light energy per pulse. At each intensity the shift corresponding to various peak assignments is indicated. There is a direct relation between the shift of the spectrum and the light intensity (right-hand scale), explained in the text. The vertical bars indicate the width of the peaks in the spectra. If the width is varying within one spectrum the largest and the smallest widths are indicated.

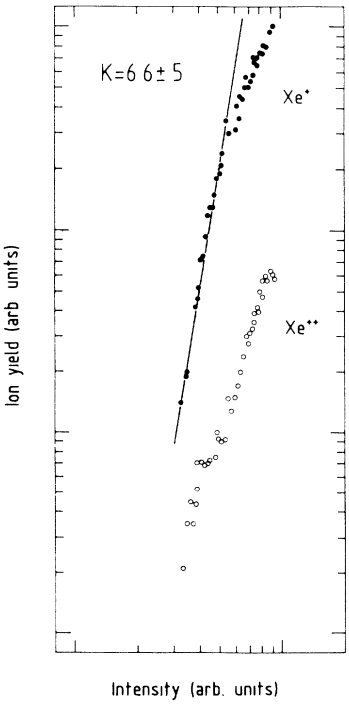

FIG. 3. Xenon-ion yields vs laser intensity.

argument is that, since at the lowest intensity used some peaks have already been suppressed, it is not the seven-photon saturation intensity that matters but the saturation intensity of the process producing the first observable peak. Most experiments as well as model calculations to date revealed that the order of nonlinearity stays at its lowest-order perturbative value even in the regime where higher-order processes dominate. Since this behavior is as yet not understood, there is, however, no guarantee that it will persist indefinitely. Estimates of saturation intensities based on power laws might therefore be misleading.

There is theoretical indication[12] that a typical cross section for ionization is 6 orders of magnitude lower than that of xenon at 532 nm, often used as a standard case. In Ref. 5, however, a saturation intensity of 3×10^{13} W/cm^2 was found at nearly the same wavelength (616 nm) but a pulse duration of 500 fs. Note, however, that in this case the clearly resonant nature of the ionization process greatly increases the cross section as compared to the nonresonant case presented here. It thus appears that it is not so much the wavelength that matters, but the fact that our short pulses quench all resonant behavior in this system. It is obvious that these explanations are rather tentative and the conclusion should be that this question will remain open.

Corkum, Rolland, and Srinivasan-Rao[13] deduce, albeit indirectly, from measurements at high pressure that 100-fs, 620-nm laser pulses are unexpectedly inefficient for the ionization of xenon atoms. It is reasonable to think that the same mechanism "inhibits" ionization in both experiments, namely a light-induced increase of the ionization energy and, consequently, of the number of photons necessary for the MPI process.

A very interesting feature has been reported by Freeman *et al.*[5] in the ATI spectra obtained with subpicosecond pulses: The spectra show a number of spikes which they connect to resonances with bound states upshifted by the ponderomotive potential. At an intensity close to theirs (as deduced from the shift of 1.6 eV common to both experiments), our low-energy peaks in Fig. 1 show a hint of structure, which sometimes appeared even more clearly than shown here. As our short pulse duration quenches resonances, the alleged structure was too weak to be identifiable as belonging to known states.

In conclusion, multiphoton ionization with ultrashort intense pulses (100 fs, 10^{14} W cm^{-2}) at 620 nm shows the following characteristics: (i) a large number of excess-photon peaks and peak shifts; (ii) a high saturation intensity together with a low double-ionization rate. The measurement of energy shifts of the electron spectrum gives direct evidence of the strong perturbation of the final state of the MPI process by the laser field; the use of ultrashort pulses also allows one to directly probe the electron drift energy after ionization. Finally, we have to leave open the question of the high saturation intensity measured in this experiment: Is it simply an effect of the atomic structure (at this laser wavelength) or a more profound effect of the pulse duration?

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