# Wavelength dependence of double ionization of xenon in a strong laser field

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(Received 1 July 2005; published 31 January 2006)

The wavelength dependence of double ionization of xenon in a 100-fs laser pulse ($2–4 \times 10^{13}$ W/cm$^2$) has been studied using photoelectron imaging and ion time-of-flight spectrometry. In the wavelength ranges between 1150 and 1560 nm and 792 and 803 nm a pronounced variation of the ratio of ion yields $Xe^{2+}/Xe^{+}$ is observed. We attribute this variation to the strong influence of a $5s^2 5p^5 \rightarrow 5s5p^6$ transition on the dynamics of double ionization.

DOI: 10.1103/PhysRevA.73.013413 PACS number(s): 32.80.Rm, 32.80.Fb, 32.90.+a

The characterization of double ionization of atoms in an ultrashort intense laser pulse is still a challenging task in strong field research today. Since the discovery of the so-called *knee structure* in the yield of doubly charged xenon ions produced in the focus of a 532-nm, 50-psec laser pulse [1] a number of theories and experiments attempted to describe the dynamics of the double ionization process. A stepwise (sequential) ionization process based on the Ammosov-Delone-Krainov (ADK) tunneling theory [2] failed to explain the enhanced double ionization at intensities at or below the knee [3–5]. Experiments confirmed this discrepancy for most rare gases and also for higher charge states [6,7].

In order to explain the observed data nonsequential (NS) models were introduced. These account for the influence of the first freed electron on the probability of emission of a second electron [8–10]. The role played by the electronic structure of the atom or ion core in resonant excitation was also considered as a source of enhancement of the double ionization rate [11–13].

The so-called rescattering model [14,15], took into account the influence of the electron generated in the first ionization step. This electron may be accelerated and driven back by the electric field of the laser pulse to its parent ion where it can free a second electron by inelastic scattering. The maximum kinetic energy an electron can classically gain in the external field is $3.17 \times U_P$ [16]. $U_P$ is the so-called quiver energy of a free electron. In the low intensity part of the knee-structure observed for $Xe^{2+}$, this energy is far too small to ionize $Xe^{+}$ or even prepare an excited state of the ion. Still, ionization or excitation might be due to multiple rescattering of the electron as it was proposed by Wiehle *et al.* [17] and recently reinvestigated classically in the calculations performed by Ho *et al.* [18].

Quantum-mechanical models based on many-body *S*-matrix theory were able to reproduce the enhanced ionization rate of doubly charged ions at selected wavelengths (780 and 800 nm) and to predict the energy distributions of the electrons by including electron-electron interaction [9,10].

Chen *et al.* [19] studied the wavelength dependence of double ionization of neon using a semiclassical rescattering model in the high intensity region of the knee structure. They found, that the ratio $Ne^{2+}/Ne^{+}$ increases as the wavelength decreases from 1100 nm reaching a maximum at about 250 nm. The ratio drops sharply, when the wavelength is decreased further.

To date no experimental data exist on the wavelength dependence of the yield of doubly and singly charged atomic ions apart from the recent measurement of the yield of $Xe^{2+}$ and $Xe^{+}$ at 770 and 790 nm by Rudati *et al.* [13].

The influence of the electronic structure and of electron-electron correlation should be most sensitively probed by studying the wavelength dependence of double ionization.

In this paper we present an unexpected wavelength dependence of xenon double ionization which we observe in the knee region at moderate intensities ($2–4 \times 10^{13}$ W/cm$^2$). In one experimental series the wavelength of the laser is varied between 1155 and 1565 nm at intervals of 10 nm. Laser radiation in this range was generated using a traveling-wave optical parametric amplifier of superfluorescense (TOPAS) pumped by an 800-nm, 100-fs Ti:sapphire laser. In a second series we recorded data at 792, 797, and 803 nm. The linearly polarized beam is focused into the vacuum chamber with a parabolic mirror and the momentum distribution of the electrons is recorded with a photoelectron imaging spectrometer [20]. A separate multichannel plate detects the positive ions and a multistop multichannel timer (resolution 0.5 ns) records their time of flight. A variable leak is used to regulate the xenon gas pressure in order to ensure a linear response of the digital detection system for both the singly and doubly charged xenon ions.

The ratio of yields of doubly to singly charged xenon ions observed as a function of the wavelength is shown in Fig. 1. A pronounced and asymmetric minimum appears near 1275 nm and maxima near 1185 and 1475 nm. For wavelengths near 800 nm we find that the ratio decreases rapidly as the wavelength increases. The absolute yields of singly and doubly charged ions decrease with increasing wavelength. At 40 TW/cm$^2$, between 1160 and 1525 nm the $Xe^{+}$ yield decreases by a factor of 16.7 and appears only weakly

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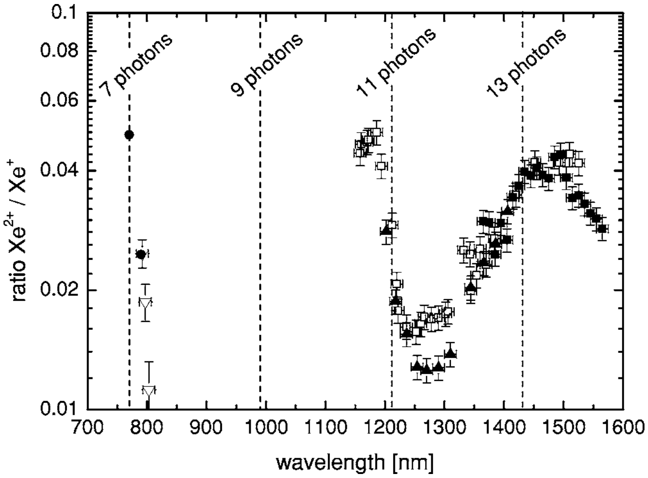


FIG. 1. The ratio of doubly to singly charged xenon ions is shown as a function of wavelength for various peak intensities. The peak intensities are 40.0±1.9 (open triangles), 38.9±1.9 (open squares), 25.2±1.2 (solid triangles), and 21.8±1.1 (solid squares) $TW/cm^2$. The ratio for 790 and 770 nm measured by Rudati *et al.* [13] at an intensity of 40 $TW/cm^2$ (multiplied by a factor of 4.1) is shown by the solid circles.

structured. Over the same range the $Xe^{2+}$ yield decreases by a factor of 18.6 with a pronounced minimum near 1275 nm. The overall decrease in ion yield with increasing wavelength at constant peak intensity is not unexpected as the number of photons required increases from 14 at 1155 nm to 22 at 1565 nm for production of $Xe^+$ and from 23 at 1155 nm to 32 at 1565 nm for the production of $Xe^{2+}$ at an intensity of 22 $TW/cm^2$.

The surprisingly strong variation of the ion yield ratio shown in Fig. 1 is not immediately obvious. This strong variation of the ion yield ratio cannot be explained by a single rescattering of the first electron, since the classical rescattering energy is too small for direct ionization of $Xe^+$ at our intensities and because the rescattering energy monotonously increases with increasing wavelength. A clue to interpret the observed variation with wavelength may be found in the work of Charalambidis *et al.* [12] who solved rate equations for the double ionization yield of xenon at 531 nm and pulse lengths of 30 ps. These authors included stepwise ionization processes as well as the direct production of doubly charged ions with 15 photons. When including excitation of the $5s5p^6\ ^2S_{1/2}$ ion state as a resonant intermediate setup, their rate model qualitatively reproduced the knee structure in the ion yield as a function of laser intensity. Also Walker *et al.* proposed a scenario with an excited state [11], but they excluded direct double ionization of the xenon atom.

The wavelength dependence in Fig. 1 lends strong support to the importance of the transition $5s^25p^5 \rightarrow 5s5p^6$ of the $Xe^+$ ion for the enhancement of ionization in the knee region. Figure 2 illustrates the likely scenario how this transition can enhance double ionization. First an electron is freed by an $n$-photon MPI process followed by the $5s^25p^5 \rightarrow 5s5p^6$ transition through absorption of $m$ photons by a $5s$ electron. From the ionic state $5s5p^6$ a two-electron process is required to reach the double ionization continuum $5s^25p^4+e$. This

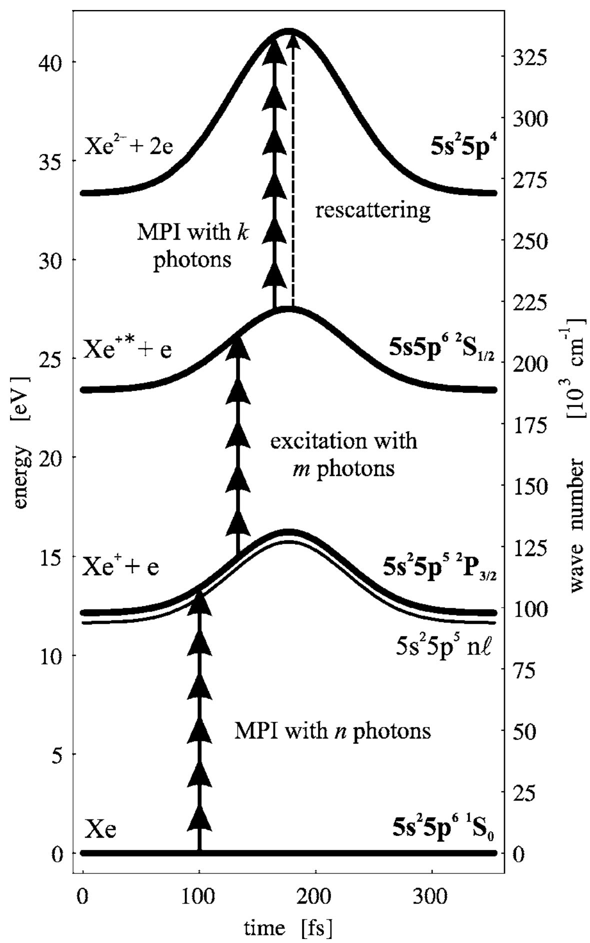


FIG. 2. Schematics of the proposed double ionization mechanism of xenon atoms based on resonance enhancement by the ionic transition and a two electron transition involving the rescattered electron. The graph is drawn for a laser pulse with a peak intensity of $3\times10^{13}$ $W/cm^2$ and a wavelength of 1200 nm. The excited states and continua follow the temporal development of the laser intensity.

final step is likely to occur in a combination of absorption of $k$ additional photons and a rescattering process of the electron from the first ionization step.

A crucial test of this model shown in Fig. 2 is the parity requirement of the ionic transition. Since an electron is transferred from an $s$ to a $p$ state, this transition is only allowed for an odd number of photons. The corresponding wavelengths of the $n$-photon transition in a free Xenon ion (11.27 eV, 90 873.83 $cm^{-1}$) are given by the dashed lines in Fig. 1 and they are listed in Table I. Evidently, the two maxima of the curve fall into the vicinity of the 11- and

TABLE I. Wavelengths for which the energy of the transition $Xe^+5s^25p^5\ ^2P_{3/2} \rightarrow Xe^+5s5p^6\ ^2S_{1/2}$ corresponds to the energy of an odd number $m$ of photons of the wavelength given. The wave number of this transition is 90 873.83 $cm^{-1}$ [21].

| $m$ | 3 | 5 | 7 | 9 | 11 | 13 | 15 |
|---|---|---|---|---|---|---|---|
| λ (nm) | 330 | 550 | 770 | 990 | 1211 | 1431 | 1651 |

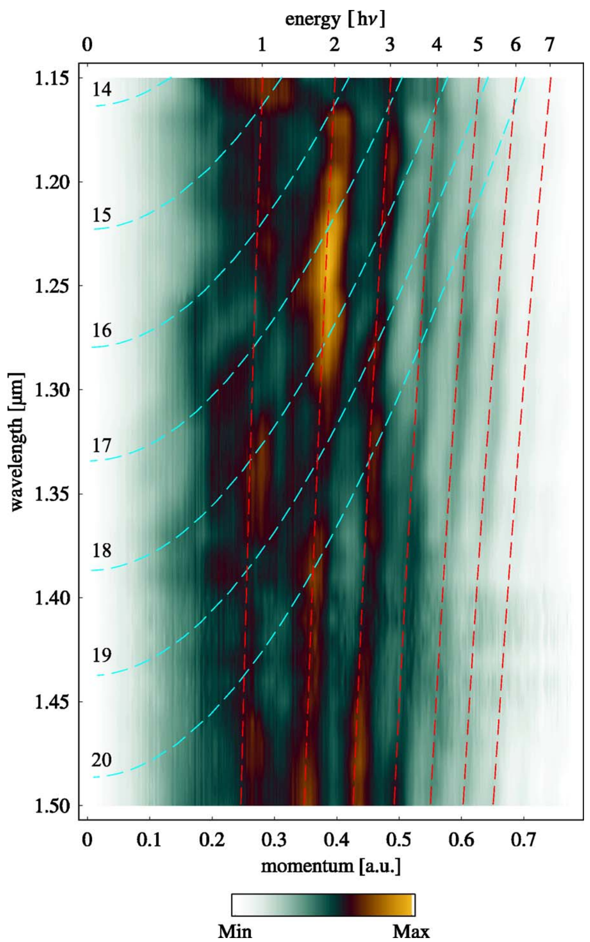


FIG. 3. (Color online) Contour plot of the photoelectron momentum distribution as a function of wavelength at an intensity of 22 TW/cm$^2$. Momentum spectra were recorded at discrete wavelengths spaced by 10 nm. At each wavelength the total electron yield was normalized to 1. The spectra were then splined to show a continuous graph with wavelength. The red dashed lines indicate the corresponding energy of one up to seven photons as a function of the wavelength. The light blue dashed lines show the corresponding energy of 14 (top line) up to 20 (bottom line) photons minus the ionization potential including the ac Stark shift at 22 TW/cm$^2$.

13-photon transition of the free Xe ion. Likewise the appearance of the pronounced minimum near 1275 nm may be attributed to enhancement of the ionic transition at odd-number resonances.

The ac Stark shifts of the continua $5s^2 5p^5 (^2P_{3/2}) + e$ and $5s5p^6 (^2S_{1/2}) + e$ should be nearly identical. As a consequence the energy difference between these states will not change during the temporal evolution of the intensity of the laser pulse and at a given wavelength the detuning character from $m$-photon resonance will not change during the laser pulse or when the intensity is changed. This supports our observation that the ratio of yields does not significantly depend on intensity.

Additional support to our model comes from the variation of the ion yield ratio near 800 nm. The observed decrease in yield near 800 nm is consistent with the presence of the seven-photon resonance (expected near 770 nm) and suppression of this process at the eight-photon level (expected at 880 nm). The experimental data reported by Rudati *et al.* [13] are fully consistent with our observations. These authors studied the intensity dependence at 770 and at 790 nm and found that the ratio of doubly to singly charged xenon ions is higher at 770 nm compared to 790 nm for intensities between 2 and $5 \times 10^{13}$ W/cm$^2$. Their results at $4 \times 10^{13}$ W/cm$^2$ are included in Fig. 1 (the two points are shown as solid circles). We have normalized their data to our values at the common wavelength 790 nm by multiplying them with a factor of 4. The origin for the difference in the ion yield ratios of the two experiments is likely due to different detection sensitivities of the ion counting detectors used in the two experiments. We also measured the ratio at 792, 797, and 803 nm for various intensities between 20 and 200 TW/cm$^2$ and found an analogous behavior of the knee structure as that observed by Rudati *et al.* [13].

The model which we outline in Fig. 2 appears at first similar to the field-independent resonant excitation model of Rudati and co-workers [13], however, these authors rule out rescattering in the low intensity part of the knee structure in the ratio, an essential step to drive the two-electron transition from the excited ion into the $Xe^{2+}$ continuum with absorption of $k$ photons.

In addition to recording the ion yield we studied the wavelength dependence of the momentum distribution electrons formed. Note that the imaging spectrometer, as operated in this experiment, sums over all electrons. That is between 2% and 10% of all electrons recorded stem from double ionization. Figure 3 gives a contour plot of the photoelectron momentum distributions recorded at an intensity of 22 TW/cm$^2$ at discrete wavelength steps of 10 nm. The red dashed lines correspond to the energy of 1,2,3, … photons as a function of the wavelength with reference to zero momentum. The light blue dashed lines correspond to the energy of 14 (top line) up to 20 (bottom line) photons minus the ionization potential including the ac Stark shift. A clear above-threshold ionization (ATI) structure can be seen at each wavelength. The most surprising feature in this image is the bright ATI peak between 1200 and 1300 nm in the momentum interval [0.35, 0.4 a.u.]. This electron group arises from the absorption of 18 photons. It is in this wavelength region that the minimum of the ratio of doubly to singly charged ions occurs. The exact meaning of a concentration of electron yield near an energy of 2 eV, at wavelengths for which double ionization is suppressed, is unclear at this point. We assume that this electron signature will certainly serve as an important measure in judging the validity of empirical models and of theory.

In conclusion, our measurements of the double ionization of xenon in strong laser fields between 1150 and 1560 nm suggest that the $Xe^+ 5s^2 5p^5\ ^2P_{3/2} \rightarrow Xe^+ 5s5p^6\ ^2S_{1/2}$ transition is responsible for the strong wavelength dependence of the ratio of doubly to singly charged ions. The importance of this specific transition may be appreciated by its strong transition matrix element. This transition is the equivalent of the Rb $D$ line; it accounts for nearly all of the oscillation strength of the ion.

This work was supported by Grant No. SFB C14/UP III of the Deutsche Forschungsgemeinschaft.

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