Wavelength and Intensity Dependence of Short Pulse Laser Xenon Double Ionization between 500 and 2300 nm

G. Gingras, A. Tripathi, and B. Witzel*

Centre d'Optique, Photonique et Laser, Université Laval, Pavillon d'optique-photonique Québec (Québec), Canada, G1V 0A6

(Received 28 January 2009; published 22 October 2009)

The wavelength and intensity dependence of xenon ionization with 50 fs laser pulses has been studied using time-of-flight mass spectrometry. We compare the ion yield distribution of singly and doubly charged xenon with the Perelomov-Popov-Terent'ev (PPT) theory, Perelomov, Popov, and Terent'ev, Zh. Eksp. Teor. Fiz. **50**, 1393 (1966) [Sov. Phys. JETP **23**, 924 (1966)], in the regime between 500 and 2300 nm. The intensity dependence for each wavelength is measured in a range between 1×10^{13} and 1×10^{15} W/cm^2. The Xe^+-ion signal is in good agreement with the PPT theory at all used wavelengths. In addition we demonstrate that ionic $5s5p^6\ ^2S$ state is excited by an electron impact excitation process and contributes to the nonsequential double ionization process.

DOI: 10.1103/PhysRevLett.103.173001 PACS numbers: 32.80.Rm, 32.80.Fb, 32.90.+a

The characterization of double ionization of atoms in ultrashort intense laser fields is still discussed controversially. 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 ps laser pulse [1] a number of theories [2–8] have been developed and experiments [9,10] have been carried out, to understand the dynamics of this double ionization process. Two different types of processes are known in strong field laser ionization. One process is a sequential ionization, where in a first step the neutral atom is ionized and a single charged ion is created. After the removal of the first electron the remaining ion is further ionized by a second step. The ion yield can be calculated with models taking exclusively this sequential process into account, such as the PPT theory [11]. A second type of ionization process can lead to a dramatic increase of the doubly charged xenon ions yield at relative low laser intensities. The so-called nonsequential ionization process consists of a simultaneous emission of two electrons. As a result of these two different ionization channels L'Huillier *et al.* [1] have observed a knee structure in the Xe^{2+} ion yield curve for 532 nm and 1.06 μm at 50 ps pulse duration. In this particular case, ionization dynamics could be explained by direct process from the xenon ground state by taking ionic excited states into account. Contrary to experiments carried out at pulse lengths of 50 ps, nonsequential double ionization [9,10] in the infrared regime at shorter pulse duration (e.g., 800 nm, 100 fs) is widely interpreted by the rescattering model [2]. In a first step an electron is set free by an ionization process with linearly polarized light. This electron is accelerated and driven back by the laser field to its parent ion and induces an impact ionization or an excitation. The maximal electron impact energy can be achieved when the electron is set free in the first step with a minimal kinetic energy either from a tunnel ionization process or from a nonresonant ionization process [12]. Quantum-mechanical calculation [7] and experimental data from Bhardwaj [13] show experimentally in neon at 800 nm that the cross section of an impact ionization or impact excitation can be enhanced by the Coulomb field of the remaining ion. In addition to impact ionization, measurements from Rudati *et al.* [14] demonstrated that the knee-structure changes for 0.77 and 0.78 μm. The authors interpret their results with the field independent resonant ionization (FIRE—model) via the $5s5p^6\ ^2S$ channel. Kaminiski *et al.* [15] confirmed this measurement, by measuring the ratio of doubly charged xenon to singly charged xenon in the wavelength regime between 1150 and 1560 nm at four different laser peak intensities between 2 and 3×10^{14} W/cm^2. We show for the first time that this excited state can also be reached by electron impact excitation with an intensity-dependent ion yield measurement of the doubly charged ion between 0.5 and 2.3 μm. Our measurements are in good agreement with ion momentum spectra measurements for He, Ne, and Ar from de Jesus *et al.* [16]. The cross section of a possible impact excitation as a result of a laser driven rescattering depends strongly on the electron impact energy.

$$E_{\max}[\mathrm{eV}] = 3.17 \times 9.33 \times 10^{-14} I[\mathrm{W/cm^2}]\lambda^2[\mu\mathrm{m}]$$

I is the maximal laser intensity at focus in units of [W/cm^2] and λ is the laser wavelength. The energy diagram in Fig. 1 shows the two possible scenarios. One possibility is the multiphoton driven [14] excitation (A) and impact excitation (B) via the ionic $5s5p^6\ ^2S$ channel. We confirm our experimental results with calculated impact excitation cross section, performed with the Lotz formula [17].

The experimental setup consists of a combination of an electron imaging spectrometer with a time-of-flight spectrometer for the ions [18]. The laser beam was focused with a spherical mirror into the spectrometer chamber. In order to cover different intensity regimes we have used mirrors

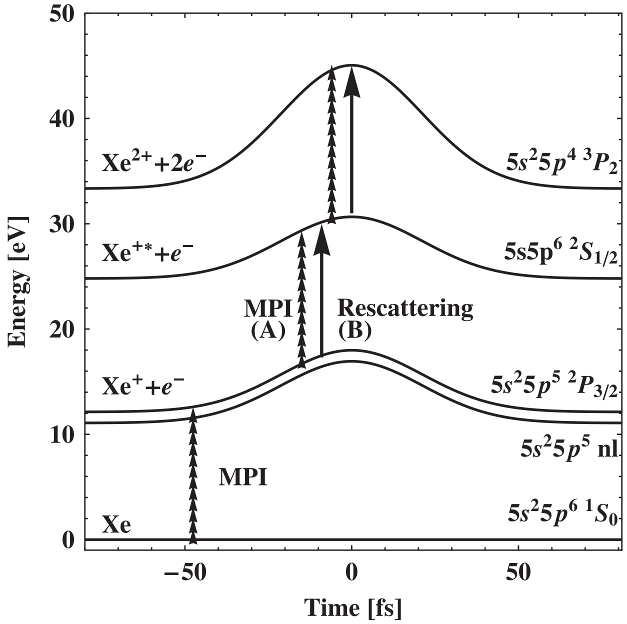

FIG. 1. Energy level diagram depending on the pulse duration. The ionic $5s5p^6\ ^2S$ state can be excited by the multiphoton transition [FIRE model (A)] or by an impact excitation (B).

with two different focal lengths (100 mm, 250 mm) resulting in a $1/e^2$ radius in the focus between 30 and 100 μm. Electrons as well as single and multiple charged ions are produced due to the high laser intensity at the focus regime and are separated in a homogeneous electric dc field. After passing an acceleration zone the ions go through a drift regime and their time of flight is detected with a 40 mm diameter chevron-type multichannel plate. We have recorded the ion yield with a digital oscilloscope. Depending on the laser intensity, the ion yield of every measurement is averaged over 2000 to 2500 single laser shots. Electrons are expanded from the interaction region (80 V/cm) onto a 40 mm diameter chevron-type set of multichannel plates with a phosphor screen attached. A CCD camera records the impact position of the electrons. A computer is used to accumulate the signal of 10^5 to 10^7 electrons to an image. An Abelian transformation is used [18] to extract the angle-resolved velocity distributions of the electrons. The background pressure was in the regime of 10^{-9} mbar. A variable leak valve allows us to apply a partial Xe pressure between 5×10^{-8} to 10^{-5} mbar. The experiment was carried out with the tunable HE-TOPAS system of the ALLS (Advanced Laser Light source Varennes, Québec, Canada) facility working with a repetition rate of 100 Hz. The optical spectrum was controlled with different computer controlled fiber based spectrometers (NIR256 and USB2000 from Ocean Optics, Inc.). The pulse length of the laser pulse (50–80 fs) is given by the specification of the laser system. Nevertheless, at certain wavelengths the pulse length has been experimentally confirmed with a FROG setup [19]. With the electron imaging spectroscopy [18] technique we have obtained the maximal peak intensity at a given laser pulse energy. We use the appearance of the opening and closing of ionization channels via the $P_{1/2}$ and $P_{3/2}$ continuum in the electron spectra as a function of the mean laser power and its wavelength [20,21]. We have compared our results with ion yield measurements performed during earlier experiments [12] and with those of other groups [22] and found good agreement between the intensity dependency at several isolated wavelengths.

We use PPT theory to calculate the ion yield of the xenon double and single ionization process [11]. The multiphoton ionization dynamic is described with the ionization probability of the bound state dependent on the frequency ω and the amplitude F of the incident radiation. The model includes the long range Coulomb correction term taking the interaction between electron and residual ion into account as well as the normalization constant for complex atoms [23]. All calculations have been performed for linear polarized laser light used in our experiment. For frequencies ω in the visible regime, the Keldysh parameter γ is much greater than unity and the model reflects well the multiphoton character of the ionization process. For ionization processes in the infrared the ionization probability $W(F, \omega)$ is reduced to the Ammosov-Delone-Krainov formula for the adiabatic approximation [23]. The rate equation of the xenon multiphoton ionization process is solved numerically in time steps of approximately 20 as for all wavelengths and intensities up to 10 times the saturation of the single and double ionization process. The laser intensity distribution at the focus of the spherical mirror is considered to be Gaussian. We obtained the total ion yield by integrating over isointensity shells up to 0.001 times the maximal peak intensity. For the double ionization process we use PPT theory to calculate the sequential ionization process. The difference between the experimental and the calculated result (Figs. 2 and 3) can be understood as a nonsequential contribution originating from the rescattering process. The Lotz formula [17] has been used to calculate the cross section of a possible impact excitation and ionization as a function of the wavelength and the laser intensity (Fig. 3).

The general ion yield dependency on the wavelength and the intensity is shown as a two-dimensional plot (Fig. 2) for single and double charged xenon. More than 2000 ion measurements have been included in these images. The image on the bottom side shows the results obtained from the PPT theory calculated for pulse duration of 50 fs. The black solid curves on top of the ion yield distribution exemplify the theoretical saturation intensity. At lower wavelength the saturation appears as a steplike function resulting from a continually increasing number of photons needed for the ionization. This feature vanishes at longer wavelengths because of the large number of photons needed for the ionization process where the Keldysh parameter is significantly smaller than 1. The experimental result for the single ionization process [Fig. 2(a)] is in good

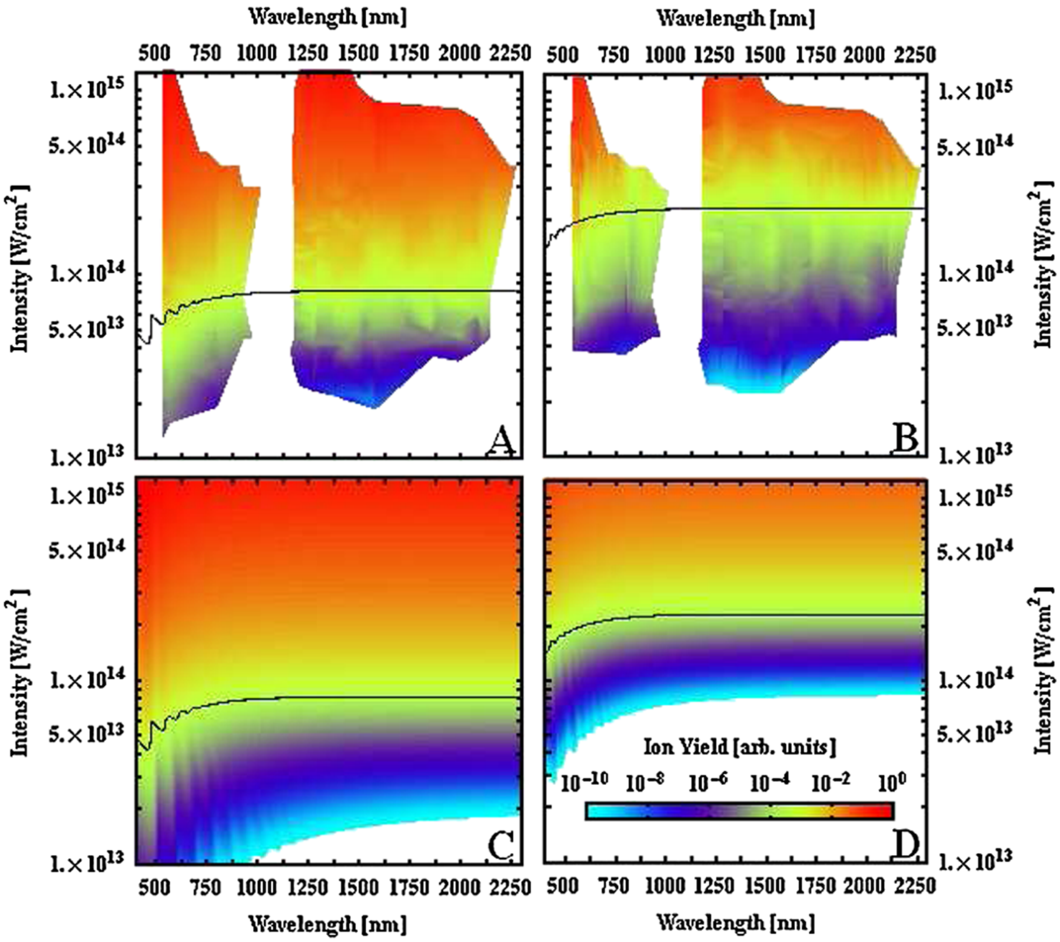

FIG. 2 (color online). Experimental ion yield (in false color representation) of Xe^+ (A) and Xe^{2+} (B) as a function of the intensity (vertical axis) and the wavelength (horizontal axis). The solid line represents the saturation threshold (calculated with the PPT theory). The lower figures show the calculated ion yield for Xe^+ (C) and Xe^{2+} (D). (We use for experiment as well as for theory the same color scale.)

agreement with the PPT—calculation and shows all major features [Fig. 2(c)]. The laser did not offer sufficient laser peak intensity in the focus for wavelengths between 950 and 1150 nm to drive a double ionization process. The intensity and wavelength dependency of Xe^{2+} [Fig. 2(b)] yield shows differences between theory and experiment. At low laser intensities 2×10^{13} W/cm^2 to 8×10^{13} W/cm^2 we observe for all wavelengths more ions than predicted by the PPT theory [Fig. 2(d)]. For 533, 571, 1181, and 1579 nm the yield of single and double charged xenon as a function of the intensity is shown in the lower part of (Fig. 3) in comparison with the PPT theory (solid curves). The experimental data set of the Xe^{2+} yield at 533 nm is always above the calculated curve. This effect is even more prominent at 571 nm and is in deviation to the PPT theory. As opposed to earlier approaches at this wavelength in the ps range [1], no knee structure at laser peak intensities down to 3×10^{13} W/cm^2 can be observed. This is in good agreement with the observation in the lower wavelength regime (248 nm, 500 fs) where single frequency measurements by Charalambidis *et al.* [24] do not show any knee structures as well. We found the first evidence of a knee structure at about 634 nm. At 1181 nm the knee structure becomes more prominent and the curve allows us to distinguish between the sequential (greater than 2×10^{14} W/cm^2) and the nonsequential regime for the doubly charged ion yield. At higher wavelengths the effect will be more and more prominent. At 1579 nm the Xe^{2+} yield at low laser intensities becomes several orders of magnitude higher than predicted by the PPT theory (e.g., at 4×10^{13} W/cm^2 the ratio between the observed Xe^{2+} yield and the theoretical prediction is 8 orders of magnitude). At 2.077 μm the knee becomes already visible at about 1×10^{14} W/cm^2. The behavior of the nonsequential part of the ionization curve has been compared with the cross section of the rescattering electron with the remaining ionic core. We made use of the Lotz formula [17] and calculated the cross section for electron impact excitation of the first ionic excited state $Xe^{+*}(5s5p^6S_{1/2})$. We assume that ions in this excited state will be further ionized to doubly charged xenon in the laser field either with a multiphoton—or with a second rescattering process [14]. The electron im-

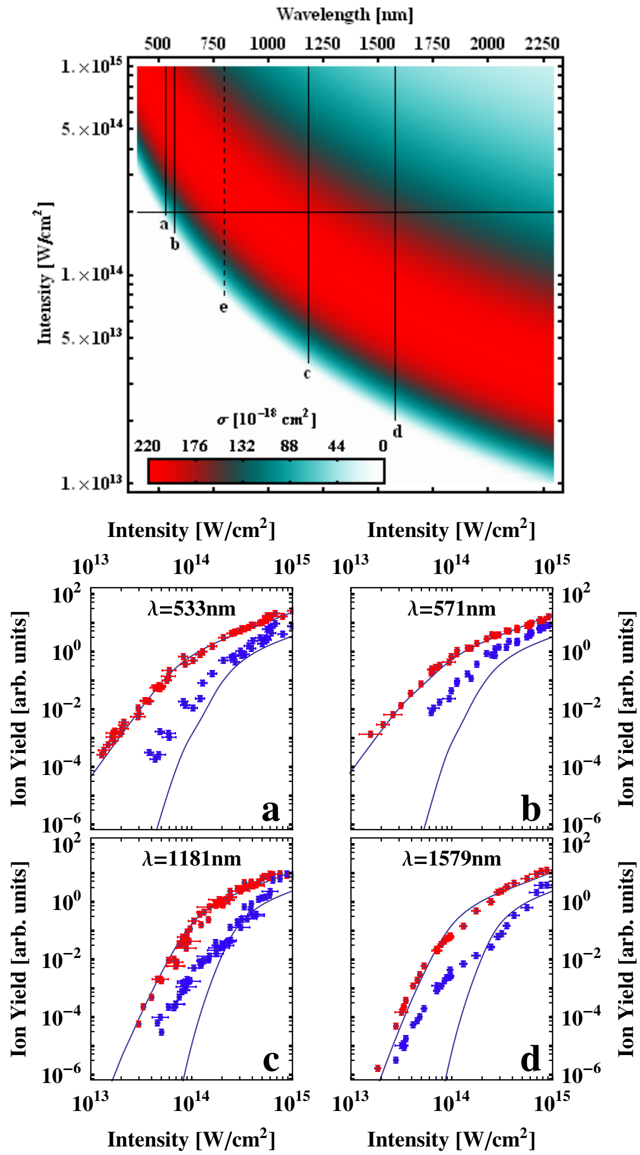

FIG. 3 (color online). Cross section of the electron impact excitation, calculated with the Lotz formula [17] for the maximal rescattering energy. The horizontal line is the intensity where approximately the sequential ionization processes start. The vertical lines correspond to ion yield measurements at selected wavelengths (a),(b),(c),(d) shown at the lower part of the graphic. [(e), upper graphic, refers to 800 nm.]

pact excitation process depends on the rescattering energy and is a function of the wavelength as well as the intensity. Figure 3 shows the cross section as a function of both parameters. The horizontal line represents the intensity (2×10^{14} W/cm^2) for which we can observe the change from a nonsequential to a sequential ionization. The calculation shows that for 532 and 571 nm the cross section is not sufficient to trigger an excitation of the $Xe^{+*}(5s5p^6S_{1/2})$ state and higher laser intensities in the focus would be necessary to enable an excitation. At these intensities the sequential ionization process is already saturated and a knee structure cannot be seen at these wavelengths but it might lead to a small deviation of the Xe^{2+} calculation in respect to the ion yield measurement. At 800 nm many ion yield curves are reported in the literature and they do show a weak knee structure [12,22]. The knee structure could be seen more prominently at longer wavelengths. At this wavelength the energy of the rescattered electron becomes higher and as a result of this, the cross section of an impact excitation or ionization is sufficient enough to enable a nonsequential ionization process.

Single and double ionization processes have been studied experimentally as a function of the wavelength and the intensity. We use two-dimensional plots to compare our experimental result with PPT calculation. While the single ion yield follows the prediction of the PPT theory, double ionization has a wavelength dependent nonsequential contribution and cannot be understood with this theory. We used the Lotz formula to calculate the cross section of the electron impact excitation process and show its contribution to the nonsequential ionization. The huge numbers of experimentally obtained ionization yields allow now the comparison with other theories taking the nonsequential process into account.

The research was performed at the Advanced Laser Light Source facility of INRS—Énergie, Matériaux et Télécommunications, which is supported by the Canada Foundation for Innovation. The authors have been supported by NSERC individual grants (B. W.) as well as by an NSERC SRO grant (G. G., A. T.).

*BWitzel@copl.ulaval.ca

[1] A. L'Huillier *et al.*, Phys. Rev. A **27**, 2503 (1983).
[2] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[3] J. S. Parker *et al.*, J. Phys. B **34**, L69 (2001).
[4] K. T. Taylor *et al.*, J. Mod. Opt. **54**, 1959 (2007).
[5] A. Becker *et al.*, Phys. Rev. A **59**, R3182 (1999).
[6] A. Becker *et al.*, J. Phys. B **32**, L335 (1999).
[7] R. Kopold *et al.*, Phys. Rev. Lett. **85**, 3781 (2000).
[8] C. J. G. J. Uiterwaal *et al.*, Eur. J. Phys. D **30**, 379 (2004).
[9] D. N. Fiffinghoff *et al.*, Phys. Rev. Lett. **69**, 2642 (1992).
[10] B. Walker *et al.*, Phys. Rev. Lett. **73**, 1227 (1994).
[11] A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, Zh. Eksp. Teor. Fiz. **50**, 1393 (1966) [Sov. Phys. JETP **23**, 924 (1966)].
[12] R. Wiehle *et al.*, Phys. Rev. Lett. **89**, 223002 (2002).
[13] V. R. Bhardwaj *et al.*, Phys. Rev. Lett. **86**, 3522 (2001).
[14] J. Rudati *et al.*, Phys. Rev. Lett. **92**, 203001 (2004).
[15] P. Kaminski *et al.*, Phys. Rev. A **73**, 013413 (2006).
[16] V. L. B. de Jesus *et al.*, J. Phys. B **37**, L161 (2004).
[17] W. Lotz *et al.*, Z. Phys. **232**, 101 (1970).
[18] C. Bordas *et al.*, Rev. Sci. Instrum. **67**, 2257 (1996).
[19] F. Légaré (private comunication).
[20] P. Kaminski *et al.*, Phys. Rev. A **70**, 053413 (2004).
[21] T. Sudmeyer *et al.*, Nat. Photon. **2**, 599 (2008).
[22] A. Talebpour *et al.*, J. Phys. B **30**, 1721 (1997).
[23] M. V. Ammosov *et al.*, Sov. Phys. JETP **64**, 1191 (1986).
[24] D. Charalambidis *et al.*, Phys. Rev. A **50**, R2822 (1994).