PHYSICAL REVIEW A VOLUME 59, NUMBER 5 MAY 1999

Production of high-charge states of Xe in a femtosecond laser pulse

A. Becker and F. H. M. Faisal
Fakultät für Physik, Universität Bielefeld, Postfach 100131, D-33501 Bielefeld, Germany
(Received 12 August 1998; revised manuscript received 3 December 1998)

In a recent experiment for ionization of the neutral Xe atom with a femtosecond Ti:sapphire laser pulse, large yields of high charge states up to sixfold ionizations have been observed. Here, we present a dynamical model of laser-induced nonsequential multiple ionization of a complex atom. The calculated results for the ion yields are found to agree remarkably well for the six charge states measured. Analyses show a significant presence of nonsequential (or direct) escapes of up to four electrons in the experimental data. For future experimental references, predictions for the sevenfold and eightfold ionizations are also made. [S1050-2947(99)50705-1]

PACS number(s): 32.80.Wr, 42.50.Hz, 34.50.Rk

About 15 years ago L'Hullier *et al.* [1] were the first to suggest the possibility of nonsequential (NS) or joint escape of two electrons for double ionization of Xe in intense picosecond laser fields. This was motivated by their observation of a kneelike structure in the intensity dependence of the ion yields. Subsequently such a structure in Xe, in the frequency domain between 527 and 532 nm, was confirmed [2] and interpreted to be due to an intermediate resonance effect [2,3]. Very recently, experiments with a femtosecond Ti:sapphire laser at a wavelength $\lambda = 800$ nm, which is away from the resonance, have been carried out and have shown copious productions of charge states up to sixfold ionization in the case of Xe [4,5]. This led the authors to surmise that the observed production of the high charge states might be due to high-order NS ionizations involving direct escape of several electrons.

In view of the complexity of the many-body problem, in general, and its highly nonperturbative nature, in particular, as yet very little theoretical progress has been made toward the quantitative analysis of particular cases. Some of the data in the multiple ionization of atoms in the CO_2 [6] and infrared [7] frequency regime could be understood in the past, using the so-called Ammosov-Delone-Krainov model [8,9] and the ''barrier-suppression'' model [6,9], as due to stepwise (or sequential) ionizations. However, these models do not so far agree with the multiple-ionization experiments in the field of femtosecond laser pulses of current interest (e.g., [4,5]).

Recently, new insights have been gained (e.g., [10] and the related references therein) into the NS double ionization in femtosecond laser pulses. Thus, using a dynamical model based on the *S*-matrix theory of NS two-electron escape, we have analyzed the double ionization of He [10–13] and reproduced the accurate double-ionization yields measured in the field of a femtosecond Ti:sapphire laser at $\lambda = 780$ nm [14,15] and other wavelengths. This result strongly suggests that dynamic electron correlation dominates the NS double-ionization process in He.

The purpose of this Rapid Communication is, first, to present an analogous dynamical model of the more challenging problem of nonresonant *n*-fold ionization of a complex atom; second, to apply it to the multiple ionization yields up to the sixth charged state of Xe that have been recently observed in the field of a femtosecond Ti:sapphire laser pulse at $\lambda = 800$ nm [4,5]; and, finally, to assess the order of the direct (or NS) escape process involved in the observed high charge states.

The essential ingredient for the present dynamical model of *n*-fold ionization of a many-electron atom is given by the Feynman diagram shown in Fig. 1(a). This diagram also provides immediately a physical picture of the NS process for the direct escape of *n* electrons in an intense laser field [16]. Thus, at first an ''active'' electron ''1'' interacts with the field, absorbs the field energy, and propagates in an intermediate field-modified state (the Volkov state, e.g., [17]), while the other electrons propagate in the virtual states of the ion. Then the active electron couples to the $n-1$ other electrons by the electron correlation and shares its energy with them, until they may escape together from the binding force of the residual ionic core. This diagram should be considered in conjunction with the usual intense field diagram, Fig. 1(b), for the ejection of a single electron from an *n*-electron configuration of the system (atom or ion).

An exact evaluation of the NS diagram, including all orders of correlation, is practically an impossible task. However, as in the case of the double ionization, and following the same approximation procedure [10,12,13], a model rate formula can be constructed [18]. Thus, for the direct emission of *n* electrons from a given initial configuration *i* (of an

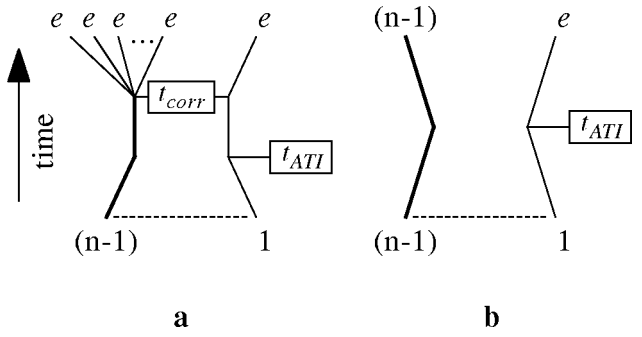

FIG. 1. Leading Feynman diagram for laser-induced nonsequential *n*-fold ionization (panel a) and single ionization (panel b). The lines stand for the states of the atomic electrons. t_{ATI} is the *T* matrix for a virtual ATI-like transition and t_{corr} is the *T* matrix for the e-ne transition.

atom or ion), into a final ionic configuration f, we get the NS rate of n-fold ionization in the form [atomic units are used below, $e=m=\hbar=1$; t (a.u.) $=2.42\times10^{-17}$ s]

$$\Gamma_{fi}^{(ne)}=\sum_{N=N_0}^{\infty}\sum_{j}\Gamma_{f,j}^{(e\text{-}ne)}(\mathcal{E}_N)\frac{\pi k_N}{2}\Gamma_{j,i}^{(ATI)}(\mathbf{k}_N\|\boldsymbol{\epsilon}), \quad (1)$$

with

$$\mathcal{E}_N=\frac{k_N^2}{2}+4k_N\sqrt{U_p}+8U_p \quad (2)$$

and

$$\frac{k_N^2}{2}=N\omega-U_p-E_B. \quad (3)$$

$\Gamma_{j,i}^{(ATI)}(\mathbf{k}_N\|\boldsymbol{\epsilon})$ is the differential above threshold ionization (ATI) rate *per* electron (atomic or ionic) along the polarization axis $\boldsymbol{\epsilon}$ for absorption of N photons, $\mathcal{E}_N$ is the dominant intermediate electron energy after a single backscattering [19], and $\Gamma_{f,j}^{(e\text{-}ne)}(\mathcal{E}_N)$ is the rate of the so-called e-ne process, i.e., n-fold ionization by electron collision (e.g., [20]) at the ''incident'' electron energy $\mathcal{E}_N$. $U_p=I/4\omega^2$ is the mean energy of oscillation of a free electron in a laser field, and $E_B=k_B^2/2$ is the binding energy of the ''target'' atom or ion. The physics of the model formula in Eq. (1) includes the rather analogous concepts of the so-called ''antenna'' picture [21] and the ''classical rescattering'' picture [22], the similarity and difference of which with the present picture have been discussed recently in the context of nonsequential double ionization [13].

For many-electron systems, the total n-fold ionization rate *per* species depends significantly on the ''statistical factor,'' namely, the number of ways the n electrons can be ejected from the initial occupied configuration i, and also on the contributions of all possible final ionic configurations f that can arise from the ejection of the n electrons. Thus we finally get the nonsequential n-fold ionization rate per species:

$$\Gamma_i^{(n+)}=\sum_f P_{fi}\Gamma_{fi}^{(ne)}, \quad (4)$$

where the number of ways to connect the configurations i and f differing by n electrons is

$$P_{fi}=\prod_s\binom{n_i^s}{n_f^s}; \quad (5)$$

n_i^s or n_f^s is the number of electrons in the sth subshell of the initial or the final configuration, respectively.

For the actual computations it is found to be most convenient to make use of the so-called Keldysh-Faisal-Reiss formula [23] for the ATI rates, corrected approximately for the Coulomb effect by a WKB estimate (cf. [9]) in the simple form,

$$\Gamma_{j,i}^{(ATI)}(\mathbf{k}_N)=C(Z,E_0,E_B)\Gamma_{j,i}^{(KFR)}(\mathbf{k}_N), \quad (6)$$

where

$$\Gamma_{j,i}^{(KFR)}(\mathbf{k}_N)=\frac{k_N}{(2\pi)^2}(U_p-N\omega)^2J_N^2\left(\boldsymbol{\alpha}_0\cdot\mathbf{k}_N;\frac{U_p}{4\omega^2}\right)\times|\langle\Phi_j(\mathbf{k}_N,2,\ldots,m)|\Phi_i(1,2,\ldots,m)\rangle|^2, \quad (7)$$

$J_N(a,b)$ is the generalized Bessel function of two arguments (e.g., [17]), and $\alpha_0=\sqrt{I}/\omega^2$ is the quiver radius. Φ_i is the wave function of the initial species and Φ_j is that of the intermediate Volkov ionic state arising from the ejection of one electron from the initial species. The Coulomb correction factor is given by

$$C(Z,E_0,E_B)=\left(\frac{4E_B}{E_0r_c}\right)^{2Z/\sqrt{2E_B}}, \quad (8)$$

where $r_c\approx2/k_B$ is a measure of the atomic size in the initial state; Z and E_0 are the residual charge and the peak field strength, respectively. Since generally the ATI rates decrease greatly with the excitation of the ionic states j compared to the ground state $j=0$, we have found it sufficient to retain in Eq. (1) the contribution of the intermediate ground state only.

Note that the rates $\Gamma_{f,j}^{(e\text{-}ne)}$ are related to the e-ne cross sections $\sigma_{f,j}^{(e\text{-}ne)}$ by the electron flux in the intermediate states,

$$\Gamma_{f,j}^{(e\text{-}ne)}(\mathcal{E}_N)=\frac{\sqrt{2\mathcal{E}_N}}{(2\pi)^3}\sigma_{f,j}^{(e\text{-}ne)}(\mathcal{E}_N). \quad (9)$$

In the actual computations for the latter cross sections for $n=2$, we have adopted the formula due to Lotz [24]:

$$\sigma_{f,0}^{e\text{-}2e}(\mathcal{E}_N)=a_{f,0}\frac{\ln(\mathcal{E}_N/E_{f,0})}{\mathcal{E}_NE_{f,0}}\{1-b_{f,0}\times\exp[-c_{f,0}(\mathcal{E}_N/E_{f,0}-1)]\}, \quad \mathcal{E}_N\geqslant E_{f,0}. \quad (10)$$

The parameters $a_{f,0}$, $b_{f,0}$, and $c_{f,0}$ are tabulated in [24] for various atoms and ions; $E_{f,0}$ is the threshold energy for the $(e,2e)$ transition from the intermediate ionic ground state into the final ionic configuration f. For $n\geqslant3$, we have adopted the prescriptions given by Fisher *et al.* [25],

$$\sigma_{f,0}^{(e\text{-}ne)}(\mathcal{E}_N)=\sigma_0^{(e\text{-}ne)}(\mathcal{E}_N)=\frac{a\pi}{17^{n-2}(2E_{min})^2}\times\frac{\ln x}{x^b}(1-2\exp^{-0.7x}), \quad \mathcal{E}_N\geqslant E_{min}, \quad (11)$$

where the parameters a and b are given in [25] for different many-electron systems and orders of ionization, $x=\mathcal{E}_N/E_{min}$ and E_{min} is the lowest ionization energy for the (e,ne) transition from the intermediate ionic ground state, $j=0$. To be consistent with their prescription for computing the total rate per species it is also necessary to replace the factor $P_{f,i}$ in Eq. (4) by the reduced factor

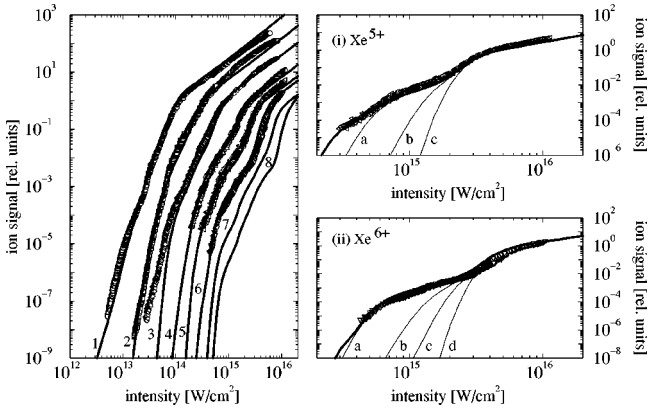

FIG. 2. Comparison of experiment and theory for n-fold ionizations ($n=1-8$) in Xe. Experiment: $\lambda=800$ nm, $t_{pulse}=200$ fs [5]. Theory: solid curves (full calculations). A one-point matching with respect to Xe^+ only fixed the relative scale. Panels (i) and (ii) show yields (thin curves) calculated by *neglecting* specific nonsequential channels (indicated parenthetically below): Xe^{5+}, a (0–5,1–5), b (0–5,1–5,2–5), c (0–5,1–5, . . . ,3–5); Xe^{6+}, a (0–6,1–6), b (0–6,1–6,2–6), c (0–6,1–6, . . . ,3–6), d (0–6,1–6, . . . ,4–6).

$$P'_{fi}=\binom{n_i^{s_1}}{n_f^{s_1}}\binom{\overline{n}+1-n_i^{s_1}}{n+n_f^{s_1}-n_i^{s_1}} \quad \text{for } n_f^{s_1}<n_i^{s_1} \tag{12}$$

and

$$P'_{fi}=0 \quad \text{for } n_f^{s_1}=n_i^{s_1}, \tag{13}$$

where s_1 stands for the outermost subshell and $\overline{n}$ is the number of electrons in the effective shells participating in the (e,ne) transition from the intermediate state according to the rules given in [25].

To determine the yields theoretically, we first calculated all the n-fold nonsequential rates, as well as the stepwise single-ionization rates that lead to a given charge state, as a function of the field intensity. They are then used in the rate equations that couple the various ion yields. To this end a laser beam with a Gaussian TEM_{00} mode and a Gaussian pulse profile (e.g., [26]) having the pulse width t_p of a given experiment are chosen. The final yields are obtained from the solutions of the rate equations at the end of the pulse and by integrating them along the longitudinal and transverse directions of the beam. The measured experimental yields are matched but at *one* point, namely, the single-ionization signal at the saturation intensity of the neutral atom. We may emphasize that this alone fixes the scales for the corresponding multiple-ionization yields.

In Fig. 2 we present the results of the calculation for the n-fold ionizations for $n=1-8$, and compare them with the experimental ion yields from Xe measured at $\lambda=800$ nm by Larochelle *et al.* [5]. The data can be seen to agree remarkably well with the full calculations (solid curves), virtually over the entire intensity range and for all the six charge states measured (except possibly for Xe^{3+} near the lowest intensities in the experiment [27]). Moreover, a weak structure in the case of Xe^{2+} and a number of relatively weak kneelike structures for the higher-charge states are also reproduced by the calculations [28].

To gain a better insight into the above result, we have further calculated the hypothetical yields of the charge states by neglecting specific NS channels. We found for all cases that the neglect of one or more of the NS rates has a drastic effect on the magnitudes of the yields. In the right-hand panels, (i) and (ii), we show the results of the detailed calculations for the two highest charge states measured, i.e., Xe^{5+} [panel (i)], and Xe^{6+} [panel (ii)] and compare them with the experimental data. Note, for example, that for Xe^{6+} we show the results of the full calculation (bold solid curve) and four additional curves that are calculated by *neglecting* the following NS channels (thin curves): (a) 0–6,1–6; (b) 0–6, 1–6,2–6; (c) 0–6,1–6, . . . ,3–6; (d) 0–6,1–6, . . . ,4–6. (In the above notation the integers stand for the initial and final charge states involved; e.g., 1–6 stands for the channel in which an initial singly ionized atom becomes a final sixfold ionized atom.) And we have proceeded similarly for Xe^{5+} in panel (ii). The drastic reduction of yields for neglecting specific NS channels, mentioned above, is now apparent. In general, it is found that only specific NS channels contribute dominantly in different parts of a yield curve. We note from the figures that in general a higher-order NS channel becomes more important at the lower intensities than a lower-order channel. For example, we observe in panel (ii) that, in the intensity domain of the measurement, the rates for the NS transitions 0–6 and 1–6 (cf. solid curve and thin curve a) make no significant contribution but the channel 2–6 (cf. solid curve and thin curve *b*) becomes very significant at the lowest intensities. From this result we may also conclude that the contributions of up to the fourth-order NS channels, involving direct escape of up to four electrons, are significantly present in the measured yields of Xe^{6+}. Similar conclusions hold for Xe^{5+}, as can be seen from panel (i) (and correspondingly for the other charge states). One finds that the yield curve for a given charge state tends to change its slope in the intensity range between two neighboring dominant channels. This is due to the competition between the neighboring NS or sequential channels. Note, finally, that a significant kneelike structure, covering a broad range of intensity, arises only when one of the two neighboring channels contributes in the total yield more dominantly, over the intensity range, than the other. We may conclude by presenting the predictions of the yields for the sevenfold and eightfold ionizations of Xe (Fig. 2, curves 7 and 8) that are yet to be measured experimentally.

To summarize, we have given a dynamical model of laser-induced nonsequential *multiple* ionization of atoms, and applied it to analyze the production of up to the sixth charge state of Xe that has been observed recently with an intense femtosecond Ti:sapphire laser pulse ($\lambda=800$ nm). The calculated yields are found to reproduce the experimental results remarkably well. It is shown that the nonsequential processes up to the fourth order (direct escape of up to four electrons) contribute significantly to the formation of the high charge states observed in the experiment. Finally, predictions are made for the sevenfold and eightfold ionizations of Xe that are yet to be measured. Looking ahead, we may expect that an analogous nonsequential mechanism might provide a fruitful point of departure for a dynamical model of production of high charge states observed in more com-

plex systems such as large molecules [29] and/or clusters [30] in intense femtosecond laser pulses.

We are thankful to Dr. S. L. Chin and Dr. A. Talebpour for kindly communicating their data in numerical form and for stimulating discussions. We are grateful to Dr. D. Andrae for providing us with the Hartree-Fock wave functions for the noble gases and their ions. This work was partially supported by the DFG (Bonn) under SPP: ''Wechselwirkung intensiver Laserfelder mit Materie,'' FA 160/18-1.

[1] A. L'Huillier *et al.*, Phys. Rev. Lett. **48**, 1814 (1982); J. Phys. B **16**, 1363 (1983); Phys. Rev. A **27**, 2503 (1983).
[2] B. Walker *et al.*, Phys. Rev. A **48**, R894 (1993).
[3] D. Charalambidis *et al.*, Phys. Rev. A **50**, R2822 (1994).
[4] A. Talebpour *et al.*, J. Phys. B **30**, 1721 (1997).
[5] S. Larochelle *et al.*, J. Phys. B **31**, 1215 (1998).
[6] S. Augst *et al.*, J. Opt. Soc. Am. B **8**, 858 (1991).
[7] T. Auguste *et al.*, J. Phys. B **25**, 4181 (1992).
[8] M. V. Ammosov *et al.*, Zh. Éksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].
[9] V. P. Krainov, J. Opt. Soc. Am. B **14**, 425 (1997).
[10] F. H. M. Faisal and A. Becker, Comments At. Mol. Phys. (to be published).
[11] A. Becker and F. H. M. Faisal, J. Phys. B **29**, L197 (1996); Laser Phys. **8**, 69 (1998).
[12] F. H. M. Faisal and A. Becker, in *Multiphoton Processes 1996*, edited by P. Lambropoulos and H. Walther, Int. Nat. Conf. Ser. No. 154 (IOP, Bristol, 1997), p. 118; Laser Phys. **7**, 684 (1997).
[13] A. Becker and F. H. M. Faisal, Phys. Rev. A **59**, R1742 (1999).
[14] B. Walker *et al.*, Phys. Rev. Lett. **73**, 1227 (1994).
[15] L. F. DiMauro and P. Agostini, Adv. At., Mol., Opt. Phys. **35**, 79 (1995).
[16] This is a direct generalization of the diagram for NS double ionization in intense fields analyzed by us recently [10–13].
[17] F. H. M. Faisal, *Theory of Multiphoton Processes* (Plenum Press, New York, 1987), p. 11.
[18] Thus the rate formula is obtained from the Feynman diagram, Fig. 1(a), by first expressing the latter analytically, using the Floquet representation of the Volkov Green's function in the intermediate states, evaluating the intermediate continuum integration by the pole approximation (an on-shell approximation), assuming the angular distribution to be dominant in the direction of the polarization, modulo squaring the amplitude, and neglecting the many rapidly oscillating off-diagonal terms (that tend to cancel out by interference) compared to the dominant sum of the diagonal terms.
[19] Quantum mechanically, the backscattering energy occurs significantly [12] up to the order of the associated Bessel functions comparable to their argument, since beyond that order their contributions fall off exponentially. Thus, for the absorption of N' photons by backscattering, the dominant order $N' = \alpha_0(k_N + k_{N+N'})$, where $k_{N+N'}$ is the backscattered momentum arising from the incident momentum k_N, and $\alpha_0 = \sqrt{I}/\omega^2$. Using this equation and the identity $k_{N+N'}^2/2 - k_N^2/2 = N'\omega$, one can easily find the energy after the backscattering, $k_{N+N'}^2/2 = k_N^2/2 + 4k_N\sqrt{U_p} + 8U_p \equiv \mathcal{E}_N$. We may note that $\mathcal{E}_N$ may also be considered as the maximum classical energy attainable in the field.
[20] J. H. McGuire, *Electron Correlation Dynamics in Atomic Collisions* (Cambridge University Press, Cambridge, England, 1997).
[21] M. Yu. Kuchiev, Pis'ma Zh. Éksp. Teor. Fiz. **45**, 319 (1987) [JETP Lett. **45**, 404 (1987)]; J. Phys. B **28**, 5093 (1995); Phys. Lett. A **212**, 77 (1995).
[22] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[23] L. V. Keldysh, Zh. Éksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)]; F. H. M. Faisal, J. Phys. B **6**, L89 (1973); H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).
[24] W. Lotz, Z. Phys. **216**, 241 (1968).
[25] V. Fisher *et al.*, J. Phys. B **28**, 3027 (1995).
[26] M. R. Cervenan and N. R. Isenor, Opt. Commun. **13**, 175 (1975); P. W. Milonni and J. H. Eberly, *Lasers* (Wiley & Sons, New York, 1988), p. 490.
[27] The deviation in the data of Xe^{3+} at the lower intensities from the trends appears at present to be rather puzzling. It may indicate the presence of other mechanisms (e.g., an induced resonance) that are not accounted for in the present model. This needs further investigation for its clarification.
[28] We have also obtained excellent agreement between the results of the present model and the experimental data for Ar at 800 nm, measured up to the triple ionization [5]. Due to the limitation of space, however, we restrict ourselves here to the analysis of the more demanding case of up to sixfold ionization of Xe only.
[29] A. Talebpour *et al.*, J. Phys. B **30**, L245 (1997); L. Bañares *et al.*, Chem. Phys. Lett. **267**, 1 (1997); C. Cornaggia and Ph. Hering, J. Phys. B **31**, L503 (1998).
[30] A. McPherson *et al.*, Nature (London) **370**, 631 (1994); E. Snyder *et al.*, Phys. Rev. Lett. **77**, 3347 (1996); T. Ditmire *et al.*, *ibid.* **78**, 2737 (1997); C. Rose-Petruck *et al.*, Phys. Rev. A **55**, 1182 (1997); B. Lang *et al.* Z. Phys. D **40**, 1 (1997); T. Ditmire, Phys. Rev. A **57**, R4094 (1998).