INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **36** (2003) 1121–1127 PII: S0953-4075(03)53141-8

# Abnormal pulse duration dependence of the ionization probability of Na atoms in intense laser fields

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Received 29 August 2002, in final form 20 January 2003
Published 26 February 2003
Online at stacks.iop.org/JPhysB/36/1121

**Abstract**

We studied the ionization probability of Na atoms in an intense Ti–sapphire laser at 800 nm versus the laser pulse duration. The ionization probabilities for the Na(3s) and Na(3p) states were found to vary non-monotonically with the pulse duration at a constant peak laser intensity. The abnormal (non-monotonic) pulse duration dependence was traced to the competition between resonant and non-resonant multiphoton ionization processes, which occurs when the frequency broadening due to the finite pulse duration is compatible with the energy difference between the excitation energy and multiphoton energy.

## 1. Introduction

Ionization of rare gas atoms in an intense laser field has been studied extensively over the passing decades both experimentally and theoretically (Piraux and Rzazewski 2000). The measured ionization rates of rare gas atoms are well explained by the atomic tunnelling model (Ammosov *et al* 1986) and the ionization rate is not sensitive to the detailed atomic structure, the laser pulse duration and laser central frequency. In contrast to the rare gas atoms, the alkali atom has a small ionization potential and the ionization is dominated by the multiphoton process. Since the multiphoton process is sensitive to the atomic structure, especially when resonance occurs, one can expect ionization rates of alkali atoms to vary significantly, not only between neighbouring systems but also with laser parameters. For sodium atoms, single-photon ionization from the ground state has been studied both experimentally (Hudson 1964) and theoretically (Fink and Johnson 1986), and more recently, from the excited states as well (Petrov *et al* 2000). All of these investigations were carried out in the weak field regime where perturbation theory is applicable. Jones (1995) first measured the multiphoton ionization of Na in a two-pulse laser field and studied the delay-time dependence. Resonant multiphoton ionization of Na in an intense laser field has been studied theoretically by Haan and Geltman (1982). The non-resonant multiphoton ionization in a weak field has been studied experimentally by Leuchs and Smith (1982). With the advance of laser technology, a short-pulse femtosecond laser with high peak laser intensity is now available. It is desirable to investigate the

0953-4075/03/061121+07$30.00  

ionization of alkali atoms in a short-pulsed intense laser field, in particular, with respect to the pulse duration. For a short pulse, the laser cannot be treated as monochromatic. Rather one has to consider its spectral frequency distribution. The relative importance of resonant and nonresonant multiphoton ionization can be explored simply by varying the pulse duration.

Traditionally one uses the ionization rate to describe ionization in a long-pulse laser field. In such a case, the total ionization probability is proportional to the pulse duration. For a short-pulse laser, the ionization rate is no longer a constant and the ionization probability should be used to describe the ionization process. If we fix the peak laser intensity and change the pulse duration, generally speaking, a longer pulse contains more power and should result in a larger ionization probability. In the multiphoton ionization of atoms by a short laser pulse, however, this is not always the case, as illustrated by this work. We found that the ionization probability of Na(3s) *decreases* first, then increases as the pulse duration increases. For Na(3p), we found that the ionization probability increases rapidly to reach a peak first, then decreases to a minimum, followed by a monotonic increase with the pulse duration. We showed that this abnormal behaviour is due to the competition between non-resonant and resonant ionizations which depends on the pulse duration. Such an 'abnormal' behaviour is believed to be prevalent in the multiphoton ionization of atoms in a short-pulse laser field and the details depend on the atomic structure as well as on laser parameters, namely, the pulse duration and laser central frequency.

In the following the theoretical method and the numerical procedure are described briefly in section 2. The results and the interpretations are presented in section 3. A short summary is given in section 4.

## 2. Theoretical method

We calculate the ionization probability of alkali atoms in a pulsed intense laser field by solving the time-dependent Schrödinger equation (atomic units $m = \hbar = e = 1$ are used throughout the paper unless otherwise indicated)

$$\mathrm{i}\frac{\partial}{\partial t}\psi(\boldsymbol{r},t) = H(t)\psi(\boldsymbol{r},t) \tag{1}$$

directly, where

$$H(t) = -\frac{\nabla^2}{2} + V(\boldsymbol{r}) + V^{ext}(\boldsymbol{r},t). \tag{2}$$

Here $V^{ext}(\boldsymbol{r},t)$ is the interaction of the electron with the external laser field, taken to have the following form:

$$V^{ext}(\boldsymbol{r},t) = \boldsymbol{r}\boldsymbol{E}_0 \mathrm{e}^{-(2\ln 2)t^2/\tau^2}\cos(\omega_0 t + \delta), \tag{3}$$

with $\delta$ being the absolute phase. In this work we choose $\delta = 0$ since our test calculations showed that the qualitative behaviours presented in what follows do not depend sensitively on the value of $\delta$. $V(\boldsymbol{r})$ is a model potential used to describe the interaction of the outermost electron with the ion core. Here we choose

$$V(\boldsymbol{r}) = -\frac{X}{r} - \frac{Z-X}{r}\mathrm{e}^{-a_1 r} - a_2\mathrm{e}^{-a_3 r}. \tag{4}$$

Here $X$ is the charge state, $Z$ the atomic number and $a_1$, $a_2$ and $a_3$ are parameters that have been tabulated by Schweizer *et al* (1999). The time-dependent Schrödinger equation is solved with a generalized pseudospectral method in the energy representation (Tong and Chu 1997, 2000). In the present method, the time-dependent wavefunction is expressed as

$$\psi(\boldsymbol{r},t) = \sum_{l=0}^{l_{max}} g_l(r,t)P_l(\cos\theta). \tag{5}$$

The radial part is discretized in the pseudospectral method. After diagonalizing the radial field-free Hamiltonian, we obtained the eigen-energies and eigen-functions of the bound as well as continuum pseudostates (Tong and Chu 1997). We propagated the radial wavefunction $g_l(r, t)$ on this 'complete' basis set. After propagating each time step, we remove the outer part of the wavefunction by a filter function

$$f(r) = \begin{cases} 1 & r < r_c; \\ 1 - \left(\dfrac{r - r_c}{r_{max} - r_c}\right)^4 & r_c \leqslant r < r_{max}; \\ 0 & r \geqslant r_{max}, \end{cases} \tag{6}$$

to avoid reflection from the boundaries. In the present calculation, we choose the number of radial grid point $N = 300$, $l_{max} = 10$, $r_{max} = 150$ au and $r_c = 100$ au. The convergence of the calculation is checked by using larger $N$ and $l_{max}$, by varying $r_c$ and $r_{max}$. The detailed numerical procedure can be found elsewhere (Tong and Chu 1997, 2000).

When the laser pulse is over at $t \to \infty$, the time-dependent wavefunction $\psi(\boldsymbol{r}, \infty)$ contains all the physical information regarding the system, namely the ionization probability

$$P_i = 1 - \int |\psi(\boldsymbol{r}, \infty)|^2 \, \mathrm{d}^3\boldsymbol{r} \tag{7}$$

or the excitation probability to a bound state

$$P_b = |\langle \psi(\boldsymbol{r}, \infty) | \varphi_b(\boldsymbol{r}) \rangle|^2, \tag{8}$$

where $\varphi_b(\boldsymbol{r})$ is the bound state wavefunction. Alternatively one can extract the ionization probability from

$$P_i = 1 - \sum_b P_b. \tag{9}$$

The two methods (equations (7) and (9)) give essentially identical results. In the following discussion, we use the ionization probability obtained from equation (7). For later discussion, we also define an effective ionization rate as

$$\text{Rate} = -\frac{\ln(1 - P_i)}{\tau}, \tag{10}$$

which should depend weakly on the pulse duration $\tau$ for a long pulse.

In this study, we focused on the ionization of the valence electron of Na atoms below the saturation intensity ($10^{12}$ W cm$^{-2}$). In this laser intensity regime, inner-shell ionization can be totally neglected. Thus a single-active-electron model (L'Huillier *et al* 1991, Krause *et al* 1992) should be adequate without the need to resort to the more complicated time-dependent density functional approach (Tong and Chu 1998).

## 3. Results and discussion

### *3.1. Na(3s) ionization*

Figure 1 shows the ionization probabilities $P_i$ of Na(3s) as a function of laser pulse duration at two peak intensities ($10^{11}$ and $5 \times 10^{11}$ W cm$^{-2}$). If the ionization rate is a constant, the ionization probability should vary linearly with the pulse duration. From figure 1, this appears to be the case for long pulses, but not for short pulses. In fact, the ionization probability shows a minimum at $\tau = 15$, and 10 fs, respectively, at the two peak intensities as shown in figure 1. The rise of ionization probability at short pulse durations is the focus of the following discussion.

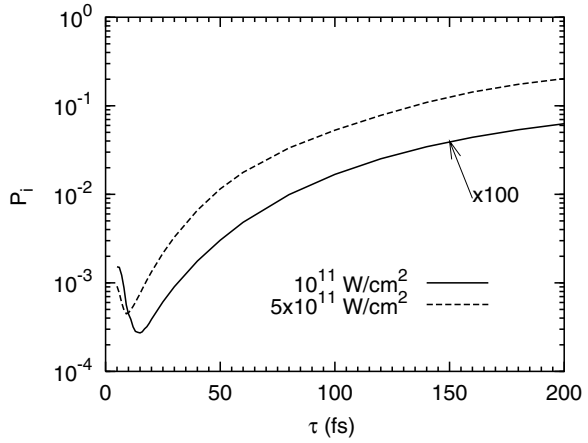


**Figure 1.** Ionization probabilities of Na(3s) in a pulsed laser field as a function of pulse duration for peak laser intensities of $10^{11}$ W cm$^{-2}$ (solid curve) and $5 \times 10^{11}$ W cm$^{-2}$ (dashed curve), respectively.

We have traced the rise of ionization probability for the shorter pulse duration to the contribution of two-photon resonant excitation to the 4s state. The energy separation between the 3s and 4s states of Na is 3.191 eV, which is close to twice the mean photon energy 3.10 eV of an 800 nm Ti:sapphire laser. For a short pulse, the frequency spread would make two-photon resonant transition to 4s very probable. Figure 2 shows the populations of the 4s state as a function of laser pulse duration for the two peak intensities. The maximum 4s population occurs at the pulse duration where the ionization probability is at the minimum. We analyse the resonant two-photon excitation to 4s and its contribution to the ionization of Na(3s). For the pulse given by equation (3), the power spectrum can be expressed as

$$P(\omega) \propto \tau \mathrm{e}^{-\tau^2(\omega-\omega_0)^2/(8\ln 2)}. \tag{11}$$

In the perturbation limit, the 4s population via resonant two-photon excitation can be estimated as

$$P_{4s} \propto \tau \int P(\omega_1) P(\omega_2) \delta(\omega_{4s} - \omega_1 - \omega_2)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2 \propto \tau^2 \mathrm{e}^{-\tau^2(\omega_{4s}-2\omega_0)^2/(16\ln 2)}. \tag{12}$$

Here $\omega_{4s}$ is the 4s excitation energy and $\omega_{4s} - 2\omega_0$ is the amount of two-photon off-resonance energy. Equation (12) shows that the 4s population first increases and reaches a maximum, and then follows an exponential decrease as the pulse duration increases, which is in qualitative agreement with the simulated results as shown in figure 2. Once the 4s electron is populated it can be ionized by one or two photons. Within this model, the total ionization probability can be expressed as

$$P_i \propto P_{4s}(P_1 + P_2) + P_n. \tag{13}$$

The first part is the contribution through 4s resonant excitation, followed by one-photon ($P_1$) and two-photon ionization ($P_2$). The single ionization energy of Na(4s) is 1.947 eV which is about 0.3 eV off resonance from the mean laser energy if the Stark shift of the level position is not included. Since only the tail of the power spectrum in equation (11) has enough energy to ionize 4s, $P_1$ will increase exponentially as the pulse duration is decreased. This would explain the ionization probability increase shown in figure 1 as the pulse duration is decreased, assuming that multiphoton contribution $P_n$ decreases smoothly with the shorter pulse duration.

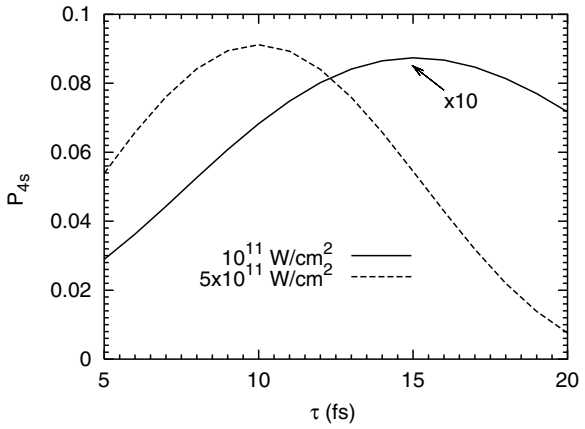


**Figure 2.** The Na(4s) excitation probability from Na(3s) in a pulsed laser field, as a function of pulse duration. The laser peak intensities are $10^{11}$ W cm$^{-2}$ (solid curve) and $5 \times 10^{11}$ W cm$^{-2}$ (dashed curve).

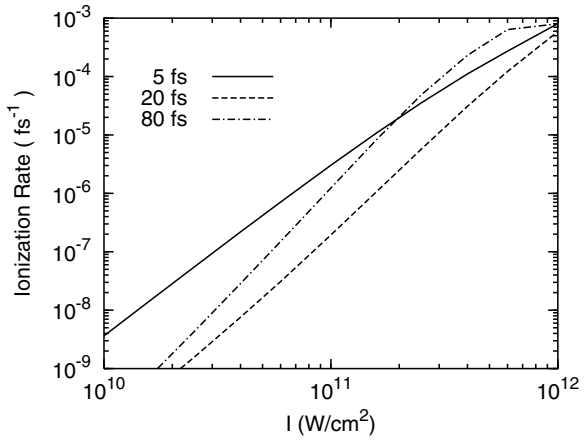


**Figure 3.** Effective ionization rates of Na(3s) atoms in an 800 nm pulsed laser field with different pulse durations.

The non-resonant part is more important at higher peak laser intensity. Thus the position of the minimum in ionization probability moves to a shorter pulse duration as the peak laser intensity increases, as shown in figure 1.

From the above discussion, we see that ionization of Na is dominated by three-photon absorption at small $\tau$ and by four-photon absorption for large $\tau$. This can be checked by studying the effective ionization rate defined in equation (10). Figure 3 shows the effective ionization rate of Na atoms in an 800 nm pulsed laser field as a function of laser peak intensity. Indeed, for a short pulse duration (5 fs), the ionization rate is proportional to $I^3$ and for a long pulse duration (80 fs), the ionization rate is proportional to $I^4$. For a medium pulse duration ($\tau = 20$ fs), the ionization rate shows the competition between the two processes.

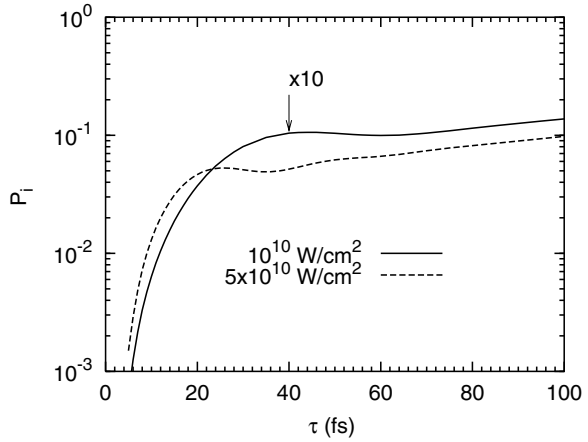


**Figure 4.** Ionization probabilities of Na(3p) in a pulsed laser field as a function of pulse duration. The peak laser intensities are $10^{10}$ W cm$^{-2}$ (solid curve) and $5 \times 10^{10}$ W cm$^{-2}$ (dashed curve), respectively.

### *3.2. Na(3p) ionization*

We next examine whether there is also an abnormal pulse duration dependence for the ionization of the Na(3p) state by the 800 nm pulse laser field. The 3p $\rightarrow$ 3d energy separation in Na is 1.51 eV which is nearly in resonance with the mean photon energy of 1.55 eV. Figure 4 shows Na(3p) ionization probabilities as a function of pulse duration at two peak laser intensities, $10^{10}$ and $5 \times 10^{10}$ W cm$^{-2}$. For the longer pulse, say for $\tau$ greater than 40 fs, the ionization probability increases almost linearly with $\tau$. At small $\tau$, the ionization probability drops rapidly as $\tau$ is decreased. In other words, the ionization probability versus pulse duration also shows abnormal behaviour, but the dependence is different to the ionization of Na(3s). The abnormal behaviour can be attributed to the resonant transition between 3p and 3d, but the effect is different from the 3s $\rightarrow$ 4s two-photon resonant transition for the ionization of Na(3s).

Since the 3p $\rightarrow$ 3d transition energy is nearly equal to the mean single-photon energy, the 3d state is easily populated. The mean population of the 3d state during the time when the laser pulse is on is nearly independent of the pulse duration and laser intensity, i.e. there is a steady-state population of Na(3d). Once the electron is in Na(3d), which has a binding energy of 1.523 eV, it can easily be ionized by a single photon and thus the ionization *rate* should depend linearly on the intensity. Figure 5 shows the ionization rate versus laser intensity at different pulse durations. For the long pulses, namely, $\tau = 40$ and 80 fs, it appears that the ionization rate depends linearly on the intensity. In the same graph, we also note that the ionization rate depends almost quadratically on the laser intensity for the short pulses of 5 and 10 fs. From figure 4, we also note that for such short pulses the ionization probability drops quickly with the decreasing pulse duration. This sharp drop is related to the 3p $\rightarrow$ 3d resonant transition. For the short pulse, the energy spread of the pulse is large such that the spectral density at the 3p $\rightarrow$ 3d resonance energy decreases exponentially with the pulse length. Thus single-photon excitation to 3d would depend exponentially with the pulse length and linearly with the laser intensity. For the ionization from Na(3d), the yield would depend linearly with the intensity and the pulse duration. Thus we expect that ionization probability at short pulse duration to be decreasing exponentially with decreasing $\tau$, as shown from the calculated results presented in figure 4. Meanwhile, the ionization rate depends almost quadratically on the laser intensity, as shown in figure 5 for the 5 and 10 fs pulses.

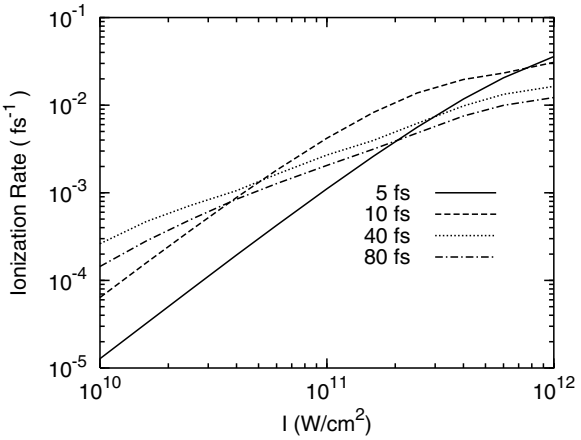


**Figure 5.** Effective ionization rates of Na(3p) in an 800 nm pulsed laser field with different pulse durations.

## 4. Summary

We have studied the ionization probability of Na atoms in a short pulsed laser field. An abnormal pulse duration dependence of the ionization probability has been found. The ionization probability does not *always* increase as the pulse duration increases. This abnormal behaviour exists in the ionization of the sodium 3s ground state as well as in the first excited state 3p. The abnormal pulse duration dependence was attributed to the competition of resonant and non-resonant ionizations. The 'abnormal' behaviour is expected to occur for other alkali atoms in an intense pulsed laser field, but the detailed behaviour of the ionization probability will depend on the atomic structure as well as on the laser parameters.

## Acknowledgment

This work was supported in part by Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, US Department of Energy.

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