Vol 10 No 5, May 2001
1009-1963/2001/10(05)/0403-04
Chinese Physics
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# PHOTOEXCITATION OF ATOM WITH ULTRASHORT LASER PULSES*

DAI CHANG-JIAN(戴长建)[a] and CHEN ZI-DONG(陈子栋)[a,b]
[a] *Department of Physics, Zhejiang University, Hangzhou* 310027, *China*
[b] *Department of Physics, Shaoxing College of Arts and Sciences, Shaoxing* 312000, *China*

(Received 4 September 2000; revised manuscript received 15 November 2000)

The uncoupled basis set is used to describe the interaction between the ultrashort pulses and the Na atom. Several polarization configurations of laser pulses have been investigated, which produce different effects on the two-step photoexcitation process. The study shows that the final states of the transitions driven by ultrashort laser pulses have a strong dependence on the laser polarization configuration. A comparison of the two different basis sets shows that the uncoupled basis set may provide more information than the coupled basis set, such as angular distribution of the atom in the final states of the transitions.

**Keywords:** ultrashort laser pulse, polarization configuration, wave packet evolution
**PACC:** 3270, 3280, 3130J

## I. INTRODUCTION

The development of laser pulses with a duration as brief as 1 fs has led to a revolution in experimental atomic physics. Previously, the vast majority of experimental studies involved the stationary states of the quantum system of interest in the frequency domain by considering the amplitudes in various stationary states.[1−3] However, most physical phenomena (e.g. collisions between electrons within multi-electron atoms) are inherently time dependent. Therefore, it is often more straightforward to study these processes in the time domain by directly monitoring the evolution of the wavefunction in analogy with classical dynamics. For the last few years, there have been a great number of studies devoted to the dynamics of transitions driven by ultrashort laser pulses.[4−6] Coupling strengths between the atomic ground states and various excited states are different so that photoexcitation and photoionization can be significantly affected by the way of laser excitation and by the polarization configuration of laser fields. In this paper, we report on the investigation of the interaction between the ultrashort laser pulses and the Na atom, making use of the full information of the interaction between the nanosecond laser pulses and the atom, obtained in our previous studies.[7,8]

## II. GENERAL METHOD

When an atom in its ground state interacts with two successive ultrashort laser pulses with a time delay $\tau$, an electronic wave packet $|\varphi_i(r,t)\rangle$ can be created by each laser pulse, where $i$=1,2 corresponds to the first and second laser pulses. The excited wavefunction can be written as the sum of two wave packets,

$$|\varphi(r,t)\rangle = \sum_{i=1}^{2} |\varphi_i(r,t)\rangle.$$

Each wave packet $|\varphi_i(r,t)\rangle$ is a superposition of several stationary states. During the delay time between the two pulses, the wave packet $|\varphi_1(r,t)\rangle$ created by the first pulse evolves freely and then interferes with the second wave packet.

For short enough laser pulses, one can separate the act of excitation from the time evolution of the wave packet $|\varphi_1(r,t)\rangle$. Consider the first-step excitation of the Na atom from its ground state 3s to the two 3p fine-structure states. There are three states involved in this process: the ground state $|g\rangle$ and the two excited states $|e_k\rangle$ with $k$=1,2. Since the two $|e_k\rangle$ are excited simultaneously by the first laser pulse, the final state of the transition must be a superposition of both $|e_k\rangle$. Therefore, there are two kinds of linear combinations of excited states, which are orthogonal to each other. One corresponds to the linear combination of excited states having the highest coupling with the $|g\rangle$ via an electric dipole transition, called the bright state $|B\rangle$; the other involves all linear com-

*Project supported by the National Natural Science Foundation of China (Grant No. 19774048), the Inertial Confinement Fusion Foundation of the China National High Technology Development Program, and the Natural Science Foundation of Zhejiang Province, China (Grant No. 197024).

binations of excited states not coupled to the ground state, called the dark states $|D\rangle$. Specifically, for a two-level system, the bright and dark states can be expressed as

$$\begin{bmatrix} |B\rangle \\ |D\rangle \end{bmatrix} = \begin{bmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{bmatrix} \begin{bmatrix} |e_1\rangle \\ |e_2\rangle \end{bmatrix}. \quad (1)$$

The evolution of $|\varphi_1(r,t)\rangle$ before the arrival of $|\varphi_2(r,t)\rangle$ corresponds to an oscillation between $|B\rangle$ and $|D\rangle$. The period of oscillation depends on the energy difference $\hbar\omega_{21}$ between the two $|e_k\rangle$, namely, $T = 2\pi/\omega_{21}$. For the Na atom, $|g\rangle = |3s_{1/2}\rangle, |e_1\rangle = |3p_{1/2}\rangle, |e_2\rangle = |3p_{3/2}\rangle$ and $\omega_{21}$=17.2cm$^{-1}$, which is much less than the typical bandwidth (135cm$^{-1}$) of an ultrashort pulsed laser, e.g. a passively mode-locked titanium–sapphire oscillator. Thus a photon from an ultrashort laser cannot resolve the two wavelengths $\lambda_1$=589.6nm and $\lambda_2$=589.0nm, corresponding to the two transitions from the ground state to the two $|e_k\rangle$. Then an ultrashort laser pulse excites a collection of final levels constituting a coherent superposition or the wave packet, which is non-stationary as time evolves. For reference, the time scales of evolution for a Rydberg wave packet can be in the ps regime.[9] In contrast, the time scales of evolution for the ground and low-lying excited states are of the order of 1 fs.[5,6] The interference between the two wave packets $|\varphi_1(r,t)\rangle$ and $|\varphi_2(r,t)\rangle$ depends on their overlap at time $t = \tau$. Specifically, in the case $|\varphi_1(r,t)\rangle = |D\rangle$ when $t = \tau$, there is no interference between $|\varphi_1(r,t)\rangle$ and $|\varphi_2(r,t)\rangle$; while in the case of $|\varphi_1(r,t)\rangle = |B\rangle$ at $t = \tau$, the interference between $|\varphi_1(r,t)\rangle$ and $|\varphi_2(r,t)\rangle$ will reach its maximum, giving rise to highest contrast of the interference pattern. The variations of the contrast will reflect the properties of oscillation of the wave packets of excited states.

In order to examine the effects of polarization configuration clearly, it is appropriate to use the uncoupled $|l, m_l, s, m_s\rangle$ basis set in which the $m_l$ and $m_s$ are shown explicitly. Furthermore, in an electric dipole transition the spin of the electron is unaffected by the laser excitation, thus it is more convenient to use $|l, m_l, s, m_s\rangle$ than a coupled $|l, s, j, m\rangle$ basis set. On the other hand, the results are independent of the initial orientation of the electronic spin. For example, we may assume that the ground state is an eigenvector of $S_Z$, namely, $|0, 0, 1/2, 1/2\rangle$. In the coupled $|l, s, j, m\rangle$ basis set, however, the same state is described as $|0, 1/2, 1/2, 1/2\rangle$.

If one defines the dipole moments of transitions from the state $|g\rangle$ to $|e_1\rangle$ and $|e_2\rangle$ as $\mu_{1g} = \langle e_1|\mu|g\rangle$ and $\mu_{2g} = \langle e_2|\mu|g\rangle$, respectively, the definition of a dark state gives

$$\langle g|\mu|D\rangle = \mu_{1g}\left(\frac{\mu_{2g}}{\mu_{1g}}\cos\theta - \sin\theta\right) = 0$$

with

$$\mathrm{tg}\,\theta = \mu_{2g}/\mu_{1g}. \quad (2)$$

On the other hand, one has

$$\frac{\mu_{2g}}{\mu_{1g}} = \sqrt{\frac{f_{2g}}{f_{1g}}}\frac{(3j)_{2g}}{(2j)_{1g}}, \quad (3)$$

where $(3j) = \begin{pmatrix} j & k & j' \\ -m & q & m' \end{pmatrix}$ is the $3j$-symbol of the transition, and $f$ is the corresponding oscillator strength. Considering the fact that the ratio of two oscillator strengths is equal to the ratio of the quantum weights of two final states of the transition, we have

$$\frac{f_{2g}}{f_{1g}} = \frac{2j_2+1}{2j_1+1} = 2 \quad \text{with} \begin{cases} j_1 = 1/2 & \text{for} \quad 3p_{1/2} \\ j_2 = 3/2 & \text{for} \quad 3p_{3/2} \end{cases}.$$

Thus we obtain

$$\mu_{2g}/\mu_{1g} = \sqrt{2}\frac{(3j)_{2g}}{(3j)_{1g}}. \quad (4)$$

## III. RESULTS AND DISCUSSION

Evaluation of all the $3j$-symbols for the linear and circular polarization schemes ($q = 0, \pm 1$ and $\Delta m = 0, \pm 1$, respectively) will determine the value of tg$\theta$ in Eq.2. In Table 1, we tabulate all the expressions corresponding to different laser polarization schemes for $|B\rangle$ and $|D\rangle$ states in terms of $|e_1\rangle$ and $|e_2\rangle$ states.

**Table 1.** The expressions of bright and dark states in terms of $|e_k\rangle$.

| | $\|B\rangle$ | $\|D\rangle$ |
|---|---|---|
| $\pi$ | $\frac{\sqrt{3}}{3}\|e_1\rangle - \frac{\sqrt{6}}{3}\|e_2\rangle$ | $\frac{\sqrt{6}}{3}\|e_1\rangle + \frac{\sqrt{3}}{3}\|e_2\rangle$ |
| $\sigma^+$ | $\|e_2\rangle$ | $\|0\rangle$ |
| $\sigma^-$ | $\frac{\sqrt{6}}{3}\|e_1\rangle - \frac{\sqrt{3}}{3}\|e_2\rangle$ | $\frac{\sqrt{3}}{3}\|e_1\rangle + \frac{\sqrt{6}}{3}\|e_2\rangle$ |

On the other hand, $|e_1\rangle$ and $|e_2\rangle$ can be expressed in the scheme $|l, m_l, s, m_s\rangle$, which are shown in Table 2.

**Table 2.** Expressions of $|e_k\rangle$ in the $|l, m_l, s, m_s\rangle$ basis set.

| | $\|e_1\rangle$ | $\|e_2\rangle$ |
|---|---|---|
| $\pi$ | $\frac{\sqrt{6}}{3}\|1,1,1/2,-1/2\rangle - \frac{\sqrt{3}}{3}\|1,0,1/2,1/2\rangle$ | $\frac{\sqrt{3}}{3}\|1,1,1/2,-1/2\rangle - \frac{\sqrt{6}}{3}\|1,0,1/2,1/2\rangle$ |
| $\sigma^+$ | $\|0\rangle$ | $\|1,1,1/2,1/2\rangle$ |
| $\sigma^-$ | $\frac{\sqrt{3}}{3}\|1,0,1/2,-1/2\rangle - \frac{\sqrt{6}}{3}\|1,-1,1/2,1/2\rangle$ | $\frac{\sqrt{6}}{3}\|1,0,1/2,-1/2\rangle + \frac{\sqrt{3}}{3}\|1,-1,1/2,1/2\rangle$ |

With all the expressions listed above, the bright and dark states can be described in the $|l, m_l, s, m_s\rangle$ basis set, which are shown in Table 3.

**Table 3.** The expressions of bright and dark states in $|l, m_l, s, m_s\rangle$ basis set.

| | $\|B\rangle$ | $\|D\rangle$ |
|---|---|---|
| $\pi$ | $-\|1,0,1/2,1/2\rangle$ | $\|1,1,1/2,-1/2\rangle$ |
| $\sigma^+$ | $\|1,1,1/2,1/2\rangle$ | $\|0\rangle$ |
| $\sigma^-$ | $-\|1,-1,1/2,1/2\rangle$ | $\|1,0,1/2,-1/2\rangle$ |

By observing the values of $m_s$ in Table 3, it is evident that the $|B\rangle$ state is strongly coupled with the $|g\rangle$ state, while the $|D\rangle$ state is not coupled with the $|g\rangle$ state. Namely, the $m_s$ values of the $|g\rangle$ and $|D\rangle$ states have opposite signs, making $\langle g|\mu|D\rangle = 0$. Furthermore, Table 3 shows that $|B\rangle$ and $|D\rangle$ are orthogonal to each other, i.e. $\langle B|B\rangle = \langle D|D\rangle = 1$, and $\langle B|D\rangle = \langle D|B\rangle = 0$.

Equipped with the above information, we study the dipole matrix elements of the various transitions in the $|l, m_l, s, m_s\rangle$ basis set. The dipole matrix element of the transition between $|a\rangle$ and $|b\rangle$ states is

$$D_{ab} = -Be\langle l|R|l'\rangle, \tag{5}$$

where $B=(-1)^{\frac{5}{2}l+\frac{1}{2}(l'+1)-m_l}\sqrt{l_{\max}}\begin{pmatrix} l & 1 & l' \\ -m_l & q & m_l \end{pmatrix}$, and $l_{\max}$ is the greater of $l$ and $l'$, $e$ is the electron charge, $q = 0, \pm 1$ depending on the polarization scheme, and the radial integral $\langle l|R|l'\rangle$ can be evaluated or can be found in the literature.[10,11]

Considering the fact that the transition matrix element from the $|g\rangle$ state to the $|B\rangle$ state depends on the laser polarization scheme, while the one from the $|D\rangle$ state to the $|g\rangle$ state is always zero, or $\langle g|R|D\rangle = 0$, we have
for the $\pi$ polarization,

$$\langle g|R|B\rangle = \frac{e}{\sqrt{3}}\langle 0|R|1\rangle = \frac{e}{\sqrt{3}}R_{01};$$

for the $\sigma^-$ polarization,

$$\langle g|R|B\rangle = \frac{e}{\sqrt{3}}\langle 0|R|1\rangle = \frac{e}{\sqrt{3}}R_{01};$$

and for the $\sigma^+$ polarization,

$$\langle g|R|B\rangle = -\frac{e}{\sqrt{3}}\langle 0|R|1\rangle = -\frac{e}{\sqrt{3}}R_{01}.$$

In the above three cases, the absolute values of the three transition matrix elements are the same, although the signs can be different for different polarization schemes. On the other hand, although the photon cannot excite the Na atom to the $|D\rangle$ state from $|g\rangle$ directly, the atom can oscillate between $|B\rangle$ and $|D\rangle$ states due to the coupling between them by the spin-orbit Hamiltonian, since the bright state is not an eigenstate of the free atom Hamiltonian. This coupling produces the oscillation between $|B\rangle$ and $|D\rangle$ during the free evolution of the system after the photoexcitation by the first laser pulse. Classically, this oscillation corresponds to the precession of the electronic spin $\boldsymbol{s}$ and of the orbital angular momentum $\boldsymbol{l}$ along the total angular momentum vector $\boldsymbol{j}$. Therefore, at the time $t = \tau$, when the second laser pulse excite the Na atom, the atom can be either in $|B\rangle$ or $|D\rangle$ state. Tables 4 and 5 list the results of dipole matrix elements from $|B\rangle$ and from $|D\rangle$ to the Rydberg states, from which the Na atom can be photoionized.

**Table 4.** The dipole matrix elements from the $|B\rangle$ state.

| | | |
|---|---|---|
| $\pi\,\pi$ | $\langle B\|R\|0,0,1/2,1/2\rangle = \frac{e}{\sqrt{3}}R_{01}$ | $\langle B\|R\|2,0,1/2,1/2\rangle = \frac{2}{\sqrt{15}}eR_{21}$ |
| $\sigma^-\sigma^-$ | $\langle B\|R\|2,2,1/2,1/2\rangle = \frac{10}{\sqrt{5}}eR_{21}$ | |
| $\sigma^+\sigma^+$ | $\langle B\|R\|2,-2,1/2,-1/2\rangle = -\frac{10}{\sqrt{5}}eR_{21}$ | |

**Table 5.** The dipole matrix elements from the $|D\rangle$ state.

| | |
|---|---|
| $\pi\,\pi$ | $\langle D\|R\|2,1,1/2,-1/2\rangle = -\frac{\sqrt{5}}{5}eR_{21}$ |
| $\sigma^-\sigma^-$ | $\langle D\|R\|2,2,1/2,1/2\rangle = 0$ |
| $\sigma^+\sigma^+$ | $\langle D\|R\|2,-1,1/2,-1/2\rangle = \frac{\sqrt{5}}{5}eR_{21}$ |

From the results shown in Tables 4 and 5, it is clearly seen that the final states of the photoexcitation are highly dependent on the polarization configurations of two laser pulses due to the selection rules of the dipole transitions. On the other hand, the number of the possible transitions from $|B\rangle$ is also different if one uses different polarization schemes. Even the polarization configuration is fixed, the number of possible transitions from $|B\rangle$ could be different from that of $|D\rangle$. Furthermore, the values of matrix elements are different from case to case, although they sometimes only have different signs. Especially, for the $\sigma^-\sigma^-$ polarization configuration, the transition from $|D\rangle$ is forbidden. With the help of all the information provided by the tables in the uncoupled $|l, m_l, s, m_s\rangle$ basis set, we cannot only obtain the orbital and spin angular momentum of the final states from the $l$ and $s$ quantum numbers, but also know the properties of the precession along the $oz$ axis. Therefore, the results provide the information of angular distribution of the Na atom in the final states of the transitions, which is one of the advantages compared with the coupled basis set where only $m$ is given.

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