# Ionization of Aligned $O_2$ by Intense Laser Pulse

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**Abstract:** Intense field ionization of aligned $O_2$ is investigated by a pump-probe method. The different behavior of $O_2^+$ and $O_2^{2+}$ yields against a pump-probe delay is ascribed to the angular dependence of the ionization probability.

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## 1. Introduction

Ionization of molecules by an intense laser pulse is one of the central topics in molecular science and has been intensively investigated in the recent decade. However, it has been a difficult task to investigate the dependence of an ionization probability of molecules on the angle between the molecular axis and the laser polarization direction because molecules rotate freely in space in the gas phase. One of the possible approaches to overcome this difficulty is to utilize a nonadiabatic molecular alignment technique [1].

An intense laser pulse induces an electronic dipole moment in a molecule. An interaction between the induced dipole moment and the laser pulse itself gives a molecule a torque to align it toward the laser polarization direction. When the pulse duration is shorter than a molecular rotational period, a coherent superposition of rotational eigenstates are prepared owing to the impulsive torque. Then, the distribution of molecular axes evolves in the time domain, and alignment as well as anti-alignment of the molecular axis distribution along the laser polarization is achieved periodically with the interval of the rotational period of molecules. This phenomenon has been known as nonadiabatic molecular alignment or field-free molecular alignment [1].

The intense field ionization of nonadiabatically aligned diatomic molecules has been investigated by the aid of high harmonic generation (HHG) [2,3]. However, the studies on the ionization process using the HHG have mainly been restricted to a single ionization process because the harmonics are generated by the recombination of the tunnel-ionized electron. On the other hand, the observation of parent and fragment ions generated by an intense laser pulse allows us to study a variety of ionization processes such as single ionization, multiple ionization, and dissociative ionization. In the present study, we investigate the intense field ionization of the $O_2$ molecule aligned in the field-free space using the nonadiabatic alignment technique by measuring $O_2^+$ and $O_2^{2+}$ generated by linearly polarized ultrashort intense laser pulses.

## 2. Experiment

A linearly polarized ultrashort laser pulse (110 fs, 800 nm, 2.0 mJ/pulse, 10 Hz) generated from a Ti:sapphire laser amplifier system was split into a pair of pump and probe pulses by passing through a Michelson-type interferometer. A rotational wave packet was created by the pump pulse. After the temporal delay, $O_2$ molecules were ionized by the probe pulse. The delay between the pump and the probe pulses was controlled by moving the position of a delay stage, which was placed in the optical path of the pump pulse in the interferometer. The pump laser power was reduced by changing the aperture size of an iris inserted just before the delay stage in order to avoid ionization. The probe power was controlled by a pair of a half-wave plate and a polarizer inserted in the probe arm of the interferometer. The pump and probe pulses were combined collinearly by using a half mirror, and were focused into a Wiley-McLaren type time-of-flight mass spectrometer (TOF-MS) by using a lens with a focal length of 200 mm. The intensities of the pump and probe pulses are estimated to be $0.3\times10^{14}$ and $1.0\times10^{14}$ W/cm$^2$, respectively. The polarization of the pump and probe pulses were set to be parallel to the TOF axis. A pulsed supersonic molecular beam of a pure $O_2$ gas with the stagnation pressure of 3.2 atm was introduced into the TOF-MS. The rotational temperature of $O_2$ was estimated to be 35 K by comparing the pump-probe delay dependence of the observed ion yield with those calculated as a temporal variation of the expectation value of the ionization yield described below. The generated ions at the laser focal spot were detected by a microchannel plate detector. The signals introduced into a digital oscilloscope were averaged over 1500 laser shots.

## 3. Results and Discussion

The ion species of $O_2^+$, $O_2^{2+}$, and $O^+$ were observed in the TOF spectrum. A pair of peaks with a small splitting is identified for $O^+$ thanks to the high mass resolution ($m/\Delta m$ = 650) of the TOF-MS. The peaks are securely assigned to the fragmentation pathways of $O_2^+ \rightarrow O^+ + O$ as described in the previous work [4].

The pump-probe delay dependence of the $O_2^+$ ion yield is shown in Fig. 1(a), where the horizontal axis is the temporal delay $\tau_{delay}$ in units of the rotational period $\tau_{rot}$ of $O_2$, which is derived by $\tau_{rot} = 1 / (2B_0c) = 11.6$ ps, where $B_0$ is the rotational constant of $O_2$ in the vibrational ground state of the electronic ground state and $c$ is a velocity of light in vacuum. It is found that the spiky transient peaks appear at $\tau_{delay} = j\tau_{rot}/4$ ($j$ = 1, 2, 3, 4), indicating that the ionization is sensitive to the molecular axis distribution. The weak spiky peaks are also recognized at $\tau_{delay} = k\tau_{rot}/8$ ($k$ = 1, 3, 5, 7).

In order to investigate the influence of the molecular alignment on the observed ion yield, the expectation value of $\cos^2\theta$, $\langle\cos^2\theta\rangle = \langle\Psi(t)|\cos^2\theta|\Psi(t)\rangle$, was calculated by solving numerically the time-dependent Schrödinger equation, where $\theta$ is the angle between the molecular axis and the polarization direction, and $|\Psi(t)\rangle$ is a rotational wave packet. The calculated $\langle\cos^2\theta\rangle$ is shown in Fig. 1(c). It seems that the calculation cannot reproduce the weak peaks at $\tau_{delay} = k\tau_{rot}/8$. Since the observed ion yield depends on the ionizatoin probability as well as on the molecular axis distribution, this disagreement is ascribed to the dependence of the ionization probability on $\theta$, $W(\theta)$.

The observed ion yield is proportional to the product of the molecular axis distribution and the ionization probability integrated over $\theta$ = 0 - π, which is equal to the following expectation value of of $W(\theta)$,

$$\langle W(\theta)\rangle = \int_0^{\pi} W(\theta)|\Psi(\theta,t)|^2 \sin\theta \, d\theta. \quad (1)$$

We adopted $W(\theta)$ [5] calculated by the molecular orbital Ammosov-Delone-Krainov (MO-ADK) theory. The expectation value of eq. (1) obtained by using the rotational wave packet is shown by the dotted curve in Fig. 1(a). The overall feature of the calculated curve agrees with the observed $O_2^+$ signal except for the slight differences at $\tau_{delay} = j\tau_{rot}/4$. This result indicates that the ionization from the HOMO plays an important role in $O_2 \rightarrow O_2^+ + e^-$ as has been reported before [6].

The pump-probe delay dependence of the $O_2^{2+}$ yield is shown in Fig. 1 (b). It is found that the transient peaks appear only at $\tau_{delay} = j\tau_{rot}/4$. In addition, the phases of the respective peaks for $\langle\cos^2\theta\rangle$ and the observed $O_2^{2+}$ signal are out of phase by π. The observed $O_2^{2+}$ signal seems to behave like $-\langle\cos^2\theta\rangle$, i.e. $\langle\sin^2\theta\rangle$, showing that $O_2^{2+}$ is produced preferably when the laser pulse polarization is perpendicular to the molecular axis. This behavior cannot be reproduced by the MO-ADK theory based on the HOMO, suggesting that other effects such as the ionization from inner molecular orbitals (HOMO-1, HOMO-2, …) need to be considered.

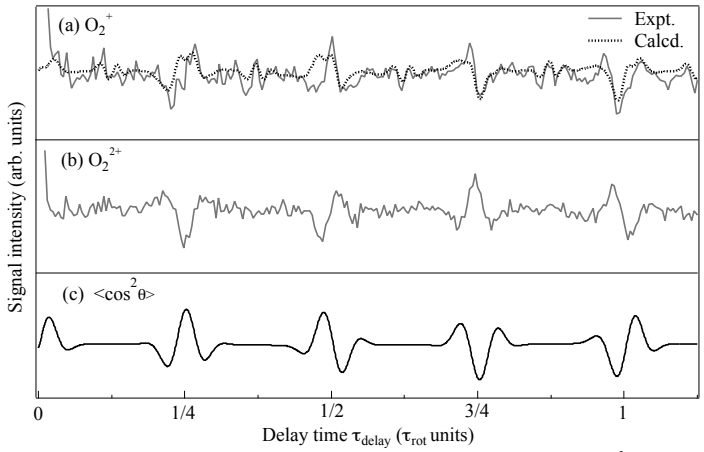


Fig. 1. The pump-probe delay dependences of (a) $O_2^+$ yield, (b) $O_2^{2+}$ yield, and (c) calculated $\langle\cos^2\theta\rangle$. In panel (a), the solid gray and dotted black curves represent the experimental and calculated signals, respectively.

## 4. References

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