

# 論文の内容の要旨

## 論文題目

Studies on ionization and coherent rotational motion of linear triatomic molecules induced by intense laser pulses

(高強度レーザー光により誘起される直線三原子分子のイオン化とコヒーレント回転運動の研究)

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Interaction between molecules and intense femtosecond laser pulses induces various phenomena, such as molecular alignment, high-order harmonic generation, ionization, Coulomb explosion, and structural deformation. These phenomena have attracted much attention because most of their characteristics cannot be understood within the perturbation theory. The ionization of molecules by intense femtosecond laser pulses, often referred to as strong-field ionization, is important among these phenomena because this process often triggers other phenomena. Revealing characteristics of the strong-field ionization is desired to understand the rich dynamics of molecules induced by the intense femtosecond laser pulses. In this thesis, three experiments are conducted to investigate the strong-field ionization of linear triatomic molecules.

In the first work of this thesis, double- and triple-ionization processes of axis-controlled OCS induced by the intense femtosecond laser pulses are investigated using a pump-probe method, in which the pump pulse creates a rotational wave packet and the probe pulse induces multiple ionization. The yields of  $\text{OCS}^{2+}$  and  $\text{OCS}^{3+}$  measured as a function of the delay between the pump and probe pulses,  $\tau$ , oscillate periodically as shown in Figs.1 (a) and (b), respectively. In Fig.1 (a), strong transient signals are clearly observed at  $\tau = 41.1$  ps and  $82.2$  ps. The observed strong

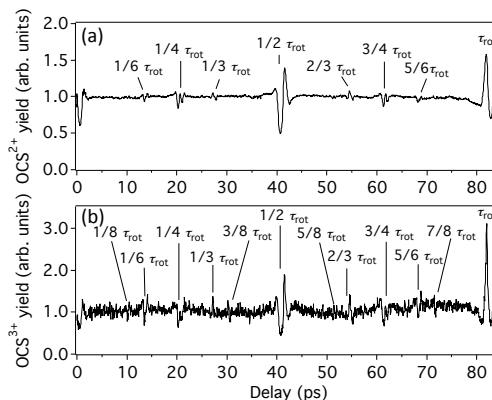


Figure 1: The dependence of the ion yields on the delay  $\tau$ . (a)  $\text{OCS}^{2+}$ . (b)  $\text{OCS}^{3+}$ .

signals are attributed to the time evolution of the created rotational wave packet because these signals appear at fractions of the rotational period of OCS ( $\tau_{\text{rot}} = 82.2$  ps). In addition to the strong transient signals, weak transient signals are also observed at  $\tau = 1/6 \tau_{\text{rot}}$ ,  $1/4 \tau_{\text{rot}}$ ,  $1/3 \tau_{\text{rot}}$ ,  $2/3 \tau_{\text{rot}}$ ,  $3/4 \tau_{\text{rot}}$ , and  $5/6 \tau_{\text{rot}}$ . These weak transient signals are assigned to high-order fractional revival structures. In addition to the revival structures observed in the measured signals of  $\text{OCS}^{2+}$ , the dependence of the  $\text{OCS}^{3+}$  yield on  $\tau$  shows higher-order fractional revival structures at  $\tau = 1/8 \tau_{\text{rot}}$ ,  $3/8 \tau_{\text{rot}}$ ,  $5/8 \tau_{\text{rot}}$ , and  $7/8 \tau_{\text{rot}}$ .

The time-dependent Schrödinger equation (TDSE) is numerically solved to reproduce the observed dependence of the  $\text{OCS}^{2+}$  and  $\text{OCS}^{3+}$  yields on  $\tau$ . The dynamics of the created rotational wave packet and the dependences of the double- and triple-ionization probabilities on the angle between the polarization direction of the probe pulse and the molecular axis of OCS,  $\theta$ , are taken into account in the calculations. The angular dependences of the double- and triple-ionization probabilities, which are denoted by  $W_2(\theta)$  and  $W_3(\theta)$ , respectively, are expanded by the Wigner  $D$  matrix elements with unknown expansion coefficients. These expansion coefficients are determined by comparing the observed revival structures with results of the calculations. The determined  $W_2(\theta)$  and  $W_3(\theta)$  show that  $\text{OCS}^{2+}$  and  $\text{OCS}^{3+}$  are preferably produced when the polarization direction of the probe pulse and molecular axis of OCS are set to be mutually perpendicular. The similar tendency has been already found in the single-ionization process of OCS [1]. The angular dependence of the single-ionization probability,  $W_1(\theta)$ , that has a large maximum at  $\theta = 90^\circ$ , was well explained using the tunneling ionization theory including the spatial information of the HOMO and effects of Stark shift and barrier-suppression ionization [2]. The large maxima of  $W_1(\theta)$ ,  $W_2(\theta)$ , and  $W_3(\theta)$  at  $\theta = 90^\circ$  are attributed to the fact that  $\text{OCS}^+$ ,  $\text{OCS}^{2+}$ , and  $\text{OCS}^{3+}$  are produced by removing electrons from the HOMO of OCS ( $3\pi$  orbital), which is occupied by four electrons. The ejection of electrons from the HOMO in the double- and triple-ionization processes is consistent with that the determined  $W_3(\theta)$  is sharper and more complex than  $W_2(\theta)$ .

The determined  $W_2(\theta)$  and  $W_3(\theta)$  include higher-order Wigner  $D$  matrix elements. It is found that the high-order fractional revival structures observed in the signals of  $\text{OCS}^{2+}$  and  $\text{OCS}^{3+}$  yields are originated from these high-order  $D$  matrix elements. The contribution of the high-order  $D$  matrix elements is an important factor of a sensitive method for probing the molecular axis distribution.

The second work of this thesis is about the enhancement and control of molecular orientation of OCS [3], which is a desired technique for investigating ionization of polar molecules. First, a well-known method of orienting molecules, in which polar molecules are irradiated with phase-locked two-color intense laser pulses consisting of the fundamental wave with a central frequency of  $\omega$  and its second harmonic, is adopted. However,

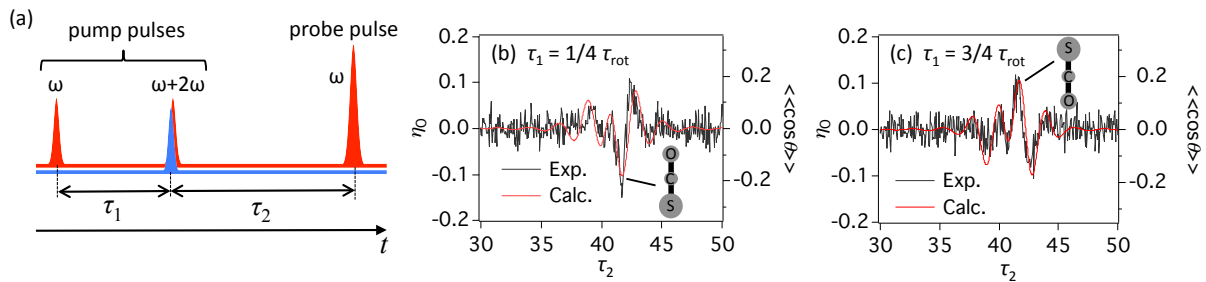


Figure 2: Molecular orientation of OCS. (a) The experimental scheme. (b) and (c): The experimental and calculated results ( $\tau_1 = 1/4 \tau_{\text{rot}}$  and  $3/4 \tau_{\text{rot}}$ ). The experimentally-determined and calculated orientation parameters are denoted by  $\eta_0$  and  $\langle\langle \cos \theta \rangle\rangle(t)$ , respectively.

signals of molecular orientation are not observed within experimental uncertainty. The TDSE calculation shows that the degree of the molecular orientation is only  $|\langle\langle\cos\theta\rangle\rangle(t)| = 0.015$ , where the double brackets denote the thermally-averaged expectation value and  $\theta$  is the angle between the polarization direction of the two-color laser pulses and the molecular axis. In order to achieve the high degree of the molecular orientation, an intense femtosecond laser pulse with a central frequency of  $\omega$  and the two-color intense laser pulses are used as pump pulses (Fig.2 (a)). The TDSE calculation is systematically carried out at various sets of the relative phase of the two-color laser pulses,  $\phi_{\text{rel}}$ , and the delay between the two pump pulses,  $\tau_1$ . It is theoretically found that strongly-oriented molecules can be prepared by adjusting  $\phi_{\text{rel}}$  and  $\tau_1$  appropriately. On the basis of the theoretical predictions, signals of the molecular orientation are experimentally observed under the conditions of  $(\phi_{\text{rel}}, \tau_1) = (0, 1/4\tau_{\text{rot}})$ ,  $(0, 3/4\tau_{\text{rot}})$ , and  $(\pi, 3/4\tau_{\text{rot}})$ . It is possible to determine the experimentally-achieved value of the highest  $|\langle\langle\cos\theta\rangle\rangle(t)|$  because the observed signals agree well with the calculated  $\langle\langle\cos\theta\rangle\rangle(t)$ . The absolute value of  $\langle\langle\cos\theta\rangle\rangle(t)$  is about 12 times larger than that of  $\langle\langle\cos\theta\rangle\rangle$  achieved only by the two-color intense laser pulses. The molecular orientation of OCS is inverted by changing  $\phi_{\text{rel}}$  from 0 to  $\pi$ . In addition, it is experimentally shown for the first time that the orientation of molecules is inverted by changing  $\tau_1$  from  $3/4\tau_{\text{rot}}$  to  $1/4\tau_{\text{rot}}$  as shown in Figs. 2 (b) and (c).

In the last part of this thesis, ionization of  $\text{N}_2\text{O}$  induced by intense femtosecond laser pulses is investigated using a spectroscopic technique. In order to reveal the vibrational- and rotational-state distributions of  $\text{N}_2\text{O}^+$  in the electronic ground state produced by the intense femtosecond laser pulses, the  $\tilde{\text{A}}\ ^2\Sigma^+ - \tilde{\text{X}}\ ^2\Pi$  excitation spectra are recorded by measuring the yield of  $\text{NO}^+$  generated via the predissociation process in the  $\tilde{\text{A}}\ ^2\Sigma^+$  state as a function of excitation wavenumber of nanosecond laser pulses. It is concluded that the vibrational ground state of  $\text{N}_2\text{O}^+$  ( $\tilde{\text{X}}\ ^2\Pi$ ) is predominantly populated by the ionization of  $\text{N}_2\text{O}$  because only transitions from the vibrational ground state of this molecular ions are observed. The determined vibrational-state distribution of  $\text{N}_2\text{O}^+$  ( $\tilde{\text{X}}\ ^2\Pi$ ) is almost the same as the distribution predicted from Franck-Condon factors (FCFs) for the transitions between the vibrational ground state of  $\text{N}_2\text{O}$  ( $\tilde{\text{X}}\ ^1\Sigma^+$ ) and various vibrational states of this molecular ion. The similarity between the observed vibrational-state distribution and the FCFs indicates that structural deformation is not induced by the intense femtosecond laser pulses and that the dependence of the tunneling ionization rate on the structures of  $\text{N}_2\text{O}$  and  $\text{N}_2\text{O}^+$  is negligible.

In order to investigate the rotational-state distribution of  $\text{N}_2\text{O}^+$  in the  $\tilde{\text{X}}\ ^2\Pi_{3/2}$  (000) state, rotationally-resolved excitation spectra of the  $\tilde{\text{A}}\ ^2\Sigma^+$  (200) -  $\tilde{\text{X}}\ ^2\Pi_{3/2}$  (000) band are measured and analyzed. The observed spectral patterns show clear dependence on the laser intensity as shown in Fig.3. In fact, transitions from higher rotational states over  $J = 60.5$  of  $\text{N}_2\text{O}^+$  in the  $\tilde{\text{X}}\ ^2\Pi_{3/2}$  (000) state are observed by increasing the intensity of the intense femtosecond laser pulses from  $9.0 \times 10^{13} \text{ W/cm}^2$  to  $1.1 \times 10^{14} \text{ W/cm}^2$ . A simplified model based on the polarizability

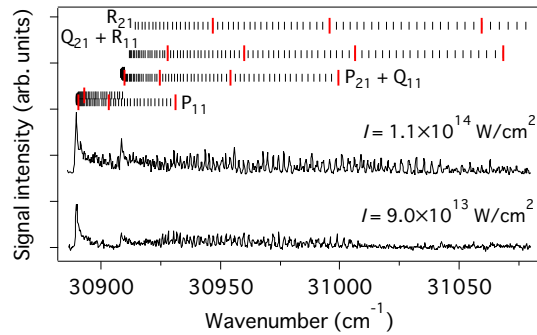


Figure 3: The  $\tilde{\text{A}}\ ^2\Sigma^+$  (200) -  $\tilde{\text{X}}\ ^2\Pi_{3/2}$  (000) excitation spectra of  $\text{N}_2\text{O}^+$  measured at different laser intensities. The transitions from  $J = 20.5, 40.5,$  and  $60.5$  are exaggerated by the thick red lines.

interaction is built to reproduce the measured excitation spectra and to extract the rotational-state distribution of  $\text{N}_2\text{O}^+$  in the  $\tilde{X}^2\Pi_{3/2}(000)$  state. In this model,  $\text{N}_2\text{O}$  molecules in the intense laser fields are rotationally excited by multiple Raman transitions induced by the polarizability interaction until the time  $t = t_{\text{switch}}$ , where  $t_{\text{switch}}$  is an adjustable parameter. After the ionization ( $t_{\text{switch}} < t$ ), the rotational excitation processes of  $\text{N}_2\text{O}^+$  in the  $\tilde{X}^2\Pi_{3/2}(000)$  state are induced in the intense laser fields. Excitation spectra calculated using this model agree well with the observed spectra. Therefore, it is concluded that the observed rotational excitation is attributed to the sequence of the rotational Raman excitation that occurs in the electronic and vibrational ground states of both  $\text{N}_2\text{O}$  molecules and  $\text{N}_2\text{O}^+$  ions. According to the determined rotational-state distribution, the maximum values of  $J$  are about 53.5 and 61.5 when using the intense femtosecond laser pulses with intensities of  $9.0 \times 10^{13} \text{ W/cm}^2$  and  $1.1 \times 10^{14} \text{ W/cm}^2$ , respectively. The higher rotational states are populated by stronger laser pulses because the polarizability interaction is enhanced as the laser intensity increases.

In addition to the rotational-state distribution, it is important to investigate whether the rotational states of  $\text{N}_2\text{O}^+$  in the  $\tilde{X}^2\Pi_{3/2}(000)$  state are coherently populated. It is known that the rotational coherence of neutral molecules can be investigated using two intense femtosecond laser pulses. The populations of rotational states change with the delay between the two laser pulses if the rotational states of the molecules are coherently populated by the first laser pulse [4]. In the present study, this experimental technique is applied to molecular ions produced by the intense femtosecond laser pulses. Two intense femtosecond laser pulses are used to investigate how a change of the delay between the two intense laser pulses,  $\tau_d$ , affects the rotational-state populations of the molecular ions generated by the first intense laser pulse. The nanosecond laser pulse mainly probes the population of  $J = 40.5$ . It is found that the population of this rotational state oscillates periodically by changing  $\tau_d$ . This oscillation of the rotational-state population is evidence that the several rotational states of  $\text{N}_2\text{O}^+$  in the  $\tilde{X}^2\Pi_{3/2}(000)$  state are coherently populated. The similar oscillation of the rotational-state population is observed even when the population of the neighboring rotational state,  $J = 39.5$ , is monitored by the nanosecond laser pulse. The observed oscillations of the populations of  $J = 39.5$  and  $40.5$  are in antiphase relation at delays of  $1/8 \tau_{\text{rev}}$  and  $3/8 \tau_{\text{rev}}$  even though both of the oscillatory signals are in phase each other at delays of  $1/4 \tau_{\text{rev}}$  and  $1/2 \tau_{\text{rev}}$ , where  $\tau_{\text{rev}}$  is the revival time of  $^2\Pi$  molecules (81.3 ps) and twice as large as the rotational period of  $^1\Sigma^+$  molecules. It is shown that the antiphase behavior is attributed to the different dependence of the alignment dynamics on  $J$ .

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