Molecular tunnelling ionization of allyl halides induced by phase-controlled two-colour laser fields

Hideki Ohmura and Naoaki Saito

National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, Japan

E-mail: hideki-ohmura@aist.go.jp

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Abstract

We have investigated the molecular tunnelling ionization (TI) of four allyl halide molecules (C₃H₅X; X = F, Cl, Br, I) induced by phase-controlled two-colour laser fields consisting of a fundamental light and a second-harmonic light with a pulse duration of 130 fs and an intensity of 10¹²–10¹³ W cm⁻². The geometric structure of the highest occupied molecular orbital (HOMO) of the four allyl halides can be systematically changed by replacing the halogen atom. We observed phase-dependent orientation-selective molecular ionization in the four methyl halide molecules. The mechanism of molecular TI is discussed in connection with the geometric structure of the HOMO.

Keywords: tunnelling ionization, intense laser fields, phase-controlled laser fields

1. Introduction

Tunnelling ionization (TI) of atoms and molecules induced by intense laser fields occurs when the binding potential of an electron is transformed and suppressed by the electric field of the laser so strongly that the wavefunction of the outermost electron penetrates the potential barrier and the electron is removed from the atom or molecule through the tunnelling process [1–7]. Recent studies have revealed that TI induced by laser fields with an intensity of more than around 10¹³ W cm⁻² occurs mainly in the attosecond (1 as = 10⁻¹⁸ s) time region, when the electric field of the laser reaches its maximum value because of a highly nonlinear optical response [8, 9]. TI is one of the most important and fundamental phenomena for observing and controlling physics in the attosecond time regime [10–12].

The investigation of molecular TI has made rapid progress both theoretically and experimentally since the introduction of molecular manipulation by intense laser fields. In molecular systems, the concept of the molecular orbital is a basic idea in quantum chemistry [13]. In the molecular orbital picture, the outermost orbital, called the highest occupied molecular orbital (HOMO), plays an important role in understanding not only the diversity of chemical bonds and resultant molecular structure but also the properties of chemical reactions [13]. Recent studies have found that the HOMO is also a major contributor to molecular TI. Ammosov–Delone–Krainov (ADK) theory [3], which is often applied to atoms to compare the absolute values and electric field dependence of ionization rates between theory and experiment, has been extended to treat molecular systems (the molecular ADK model) [14, 15]. A qualitative consequence of the molecular ADK model is that the TI rate reflects the geometric structure of the HOMO [16–20]. We can make the physical interpretation that photoelectrons are much more likely to be extracted via tunnelling from the large-amplitude lobe of the HOMO in the opposite direction of the electric field vector. As a consequence of the angular dependence of the TI rate between the electric field vector and the molecular axis, molecules aligned in a certain direction are selectively ionized in a randomly oriented gas-phase molecular ensemble, and the photofragment-emission pattern induced by molecular TI reflects the asymptotic structure of the HOMO [16, 17]. As examples of homonuclear diatomic molecules, O₂ and N₂ have,
respectively, a butterfly-shaped pattern reflecting the structure of the \( \pi \) orbital and a dumbbell-shaped pattern reflecting the \( \sigma \) orbital structure, as indicated by two-dimensional photofragment-emission pattern imaging [16].

Recently it has been pointed out that another contribution plays an important role in molecular TI: the Stark shift of the ionization potential. The Stark shift reduces or raises the ionization potential when the laser field vector is parallel or antiparallel, respectively, to the dipole of the HOMO, which leads to a maximum ionization rate for the antiparallel configuration where more electrons are ionized from the smaller-amplitude side of the HOMO [21–23]. Thus, the effect of the Stark shift trends opposite to that of the geometric structure of the HOMO [21–23] (an induced dipole synchronized with oscillating laser fields are always parallel with electric field vector so that the induced dipole has no influence for the orientation-dependent ionization). A theory for molecular TI that considers the Stark effect, called the weak-field asymptotic theory (WFAT) based on the Siegert states in a static electric field, has been developed within the single-active-electron approximation [24, 25]. It has been shown that the dominance of either the Stark shift or the shape of the HOMO depends on the molecule and cannot be determined by just looking at the shape of the HOMOs [24, 25].

The dominance of either the Stark shift or the geometric structure of the HOMO in molecular TI can decisively influence experimental molecules with asymmetric structure, where the ionization rate depends on the head–tail order of the molecule. Recent experiments with carbon monoxide (CO) on TI induced by intense linearly polarized (LP) two-colour phase-controlled laser fields consisting of a fundamental light and its second-harmonic light (‘phase-controlled’ means that the relative phase difference is stable and can be externally controlled but not stabilized actively such as phase-stabilized few-cycle pulses. hereafter, LP \( \omega + 2\omega \) laser fields) [26, 27] and cold target recoil ion momentum spectroscopy (COLTRIMS) [28] have demonstrated that more tunnelling ionization is observed from the C-side, which has the large-amplitude lobe of the HOMO, than the O-side and that the Stark effect plays a minor role.

On the other hand, the angular dependence of the TI rate for carbonyl sulfide (OCS) molecules exposed to single-frequency circularly polarized laser fields shows that the ionization rate is maximum when the laser field points from nuclei O to S, indicating that more TI occurs from the O-side, where the HOMO has a smaller amplitude, and that the Stark effect makes a major contribution [21]. Furthermore, it was found that molecular TI in OCS induced by LP \( \omega + 2\omega \) laser fields is dominated by the geometric structure of the HOMO, which is different from the results obtained with single-frequency circularly polarized laser fields [29]. Although molecular TI is an important and fundamental phenomenon in the field of molecular science using intense laser fields, molecular TI is still controversial in that the mechanisms extracted from some experiments have not yet converged. Considering this situation, it is important to explore various molecules and accumulate experimental results regarding molecular TI.

We have experimentally investigated orientation-selective molecular TI (OSM-TI) induced by LP \( \omega + 2\omega \) fields for various molecules [26, 29–33]. The method is capable of discriminating the head–tail order of the molecules, which is generally difficult to achieve by single-frequency laser fields. Furthermore, this method can be applied to a broad range of molecules, including the relatively large molecules that accompany heavy fragmentation [33].

In this study, we investigated the OSM-TI of allyl halides by using LP phase-controlled \( \omega + 2\omega \) laser fields to explore molecular TI for relatively large molecules. The geometric structure of the HOMO of four allyl halides (C₃H₅X; X = F, Cl, Br, I) can be systematically changed by replacing the halogen atom [31]. The mechanism of molecular TI will be discussed within the context of WFAT.

2. Experiment

Generally, a single-frequency LP laser field—with a waveform that is symmetric with respect to the negative and positive directions along the laser polarization—cannot discriminate the head–tail order of molecules because the ionization rate of molecules with negative orientation along the direction of polarization is equal to the rate of those with positive orientation. Meanwhile, the total electric field of an LP laser field consisting of two frequencies, the fundamental \( \omega \) and its second harmonic \( 2\omega \), is given by \( E(t) = E_1 \cos(\omega t) + E_2 \cos(2\omega t + \phi) \), where \( E_1 \) and \( E_2 \) are the amplitudes of the electric fields and \( \phi \) is the relative phase difference between the fundamental and its second harmonic. The amplitude of the electric field in the positive (negative) direction is twice that in the negative (positive) direction when \( \phi = 0 \) (\( \pi \)) and \( E_1 = 2E_2 \). When atoms and molecules are ionized by phase-controlled \( \omega + 2\omega \) laser fields, directionally asymmetric TI is expected to be induced. The TI rate of molecules with negative orientation is different from the rate of those with positive orientation if the relevant ionization is a nonlinear process such as TI. As a result, the asymmetric waveform of an LP \( \omega + 2\omega \) laser field can selectively ionize molecules while discriminating the head–tail order of molecules with asymmetric structure. We have experimentally demonstrated that OSM-TI is induced as a consequence of the directionally asymmetric TI of molecules with an asymmetric HOMO [26, 29–33].

The experimental apparatus, which has been described previously [33], consisted of a Ti:sapphire laser system, a robust phase-controlled \( \omega + 2\omega \) laser field generator, and a time-of-flight mass spectrometer (TOF-MS) equipped with a supersonic molecular beam source. Briefly, the output beam of the Ti:sapphire laser system (800 nm wavelength, 130 fs duration, 1.0 mJ pulse energy, 20 Hz repetition rate) was delivered to the robust phase-controlled \( \omega + 2\omega \) laser field generator, which was designed so that both the \( \omega \) and \( 2\omega \) beams pass through almost identical paths [33]. This generator enabled us to provide stable phase-controlled \( \omega + 2\omega \) laser pulses and to control \( \phi \) between the \( \omega \) and \( 2\omega \) pulses by using a rotatable 1 mm-thick quartz plate with a resolution of about 30 as (0.05 \( \pi \)) after the second-harmonic generation (\( \beta\)-barium
borate, type I phase-matching, 1 mm thickness, conversion efficiency: 30%) [33]. The ratio \( I_2/I_1 \) was adjusted to around 0.25 (\( E_2/E_1 = 0.5 \)), where \( I_1 \) and \( I_2 \) are the intensities of the \( \omega \) and \( 2\omega \) pulses, respectively. The phase-controlled \( \omega + 2\omega \) laser beam was focused on a supersonic molecular beam of allyl halides in the TOF-MS by an aluminium concave mirror (200 mm focal length). The TOF-MS consisted of a Wiley–McLaren type two-stage accelerator, a field-free drift tube, and an ion detector. Ions generated by the \( \omega + 2\omega \) laser pulses were accelerated towards the ion detector by static electric fields. After passing through a drift tube, the photofragment ions were detected by the ion detector composed of a microchannel plate. The TOF spectra of parent ions and photofragment ions were recorded by a digital oscilloscope.

We used the definition of the experimental configuration based on the polarization direction, the detection axis, and the direction of the electric field maxima at relative phase difference \( \phi = 0 \) as reported previously [26]. The polarization direction of the \( \omega + 2\omega \) laser fields is set to be horizontal and parallel to the detection axis, and we define \( \phi = 0 \) to be the condition when the electric field maximum points towards the ion detector. In this configuration, we could simultaneously measure the phase dependence of all photofragment ions at identical \( \phi \) values and laser intensities. To calibrate \( \phi \), we performed simultaneous measurements using gas mixtures of target molecules and reference \( C_3H_5I \) molecules [34]. This method provides an accurate phase relationship between target molecules and reference molecules under identical experimental conditions, i.e., within the same experimental run. The duration of the laser pulses used in this study is 130 fs so that the ‘origin’ does not mean the absolute zero of \( \phi \) but, for convenience’s sake, within the temporal resolution of about 20 fs, and the influence of an unstable carrier–envelope phase can be neglected.

Gas samples of \( C_3H_5F \) were prepared by dilution (5%) with helium gas with a total pressure of 0.1 MPa. Other allyl halides were prepared by the evaporation of liquid allyl halides into helium gas with a pressure of 0.1 MPa.

### 3. Results and discussion

Figure 1 shows the molecular structures and isocontours of the HOMOs of the four allyl halide molecules as determined by ab initio calculations using the Gaussian 03 W software package [28] (method: MP2; basis sets: 6–31++G(2df,p) for \( C_3H_5F \) and \( C_3H_5Cl \), and LanL2DZ augmented by polarization functions and diffuse functions for \( C_3H_5Br \) and \( C_3H_5I \)). The HOMOs of all the allyl halide molecules show a large amplitude around the \( C=\text{C} \) double bond. The amplitude of the wavefunction for the \( C=\text{C} \) double bond side becomes slightly smaller from \( C_3H_5F \) to \( C_3H_5I \). On the other hand, the amplitude of the wavefunction for the halogen-atom side becomes gradually larger from \( C_3H_5F \) to \( C_3H_5I \), reflecting the decrease of the ionization potential for the halogen atoms. The HOMOs of all the allyl halide molecules are symmetric with respect to the \( C=\text{C} \) plane (not shown). The amplitude of the wavefunction for the halogen-atom side is apparently larger than that of the \( C=\text{C} \) double bond in \( C_3H_5I \), leading to an asymmetric structure of its HOMO. The degree of asymmetry decreases gradually from \( C_3H_5I \) to \( C_3H_5Cl \), and then apparently reverses for \( C_3H_5F \). As shown in figure 1, the geometric structure of the HOMO of the four allyl halides can be systematically changed by replacing the halogen atom [31].

Figure 2 shows the TOF spectra of singly charged ions generated by the dissociative ionization of \( C_3H_5X \) (\( X = I, \text{Br}, \text{Cl}, \text{F} \)) with one-colour irradiation by an 800 nm laser pulse. We observed singly charged \( \text{CH}_3^+, \text{C}_2\text{H}_3^+, \text{C}_3\text{H}_3^+, \text{C}_3\text{H}_5^+, \text{X}^+ \), and parent \( C_3H_5X^+ \) ions in all the molecules. We adjusted the laser
intensity in the vicinity of the regime where doubly charged fragment ions due to Coulomb explosions were observed. The minimum intensity was $2.0 \times 10^{12}$ W cm$^{-2}$ for C$_3$H$_4$I, and the maximum intensity was $1.0 \times 10^{13}$ W cm$^{-2}$ for C$_3$H$_5$F, reflecting the difference in ionization potentials among the molecules. The spectra for C$_3$H$_5$Br and C$_3$H$_5$Cl are split due to the presence of isotopes for Br and Cl atoms (the abundance ratios are $^{79}$Br/$^{81}$Br = 50.7%/49.3% and $^{35}$Cl/$^{37}$Cl = 75.8%/24.2%). Each halogen photofragment exhibited a pair of peaks, the first peak resulting from ions emitted directly towards the detector and the second from those ions that were first emitted in the backward direction before being reversed by the extraction fields. The spacing of the forward and backward peaks reflects kinetic energy release. Each halogen ion showed symmetric forward–backward peaks under one-colour irradiation.

We believe that tunnelling ionization from HOMO is the dominant process. The orientation-dependent TI rate of CO molecules induced by single-colour CP laser pulses with an intensity of $4.0 \times 10^{14}$ W cm$^{-2}$ have been measured by COLTRIMS [28]. The contribution of HOMO-1 to the dissociative single ionization process in that experiment was $\sim$30% of the total signal in the experiment, with a laser intensity of about ten times higher than that used in our experiments. Therefore, we can safely say that the contribution of HOMO-1 seems to have been much smaller in the present study.

When the molecules were irradiated with the $\omega + 2\omega$ pulses, breaking of the forward–backward symmetry was clearly observed in the TOF spectra. Expanded views of typical TOF spectra of halogen ions and counterpart cations C$_3$H$_3^+$ and C$_3$H$_5^+$ with relative phase differences $\phi = 0$ and $\pi$ are shown in figure 3. In the case of C$_3$H$_5$F (figure 3(a)), the forward (backward) peak of the F$^+$ ions is predominant over the backward (forward) peak at $\phi = 0$ ($\pi$). This result shows that F$^+$ ions were emitted preferentially towards the forward (backward) direction when the optical electric field maximum pointed in the forward (backward) direction. In the cases of C$_3$H$_5$Br and C$_3$H$_5$I (figures 3(c) and (d)), forward–backward asymmetry was observed for Br$^+$ and I$^+$ but with a trend opposite to F$^+$ with respect to $\phi = 0$ and $\pi$. In the case of C$_3$H$_5$Cl (figure 3(b)), phase-dependent behaviour was not clearly observed in the TOF spectrum. However, the counterpart C$_3$H$_3^+$ and C$_3$H$_5^+$ cations were not split into forward–backward peaks in all the allyl halide molecules, and phase-dependent behaviour was not clearly discernible in the TOF spectra.

Figure 4 shows the ratio of the forward and backward yields ($I_f/I_b$) as a function of $\phi$. For the C$_3$H$_3^+$, C$_3$H$_5^+$ cations, which were not clearly split into forward–backward peaks, we obtained $I_f/I_b$ from the area of the forward and backward sides with respect to the peak centre. A clear periodicity of $2\pi$ was observed in the $I_f/I_b$ ratio for the halogen ions. On the other hand, we observed phase-dependent behaviour not by counterpart cations C$_3$H$_3^+$ but by C$_3$H$_5^+$ in all the allyl halide molecules. We examined why the C$_3$H$_5^+$ cations have no phase dependence in the molecules. One possible explanation is that it is due to a slow dissociation process, in which C$_3$H$_5^+$ is produced on a time scale longer than the rotational period, allowing orientation averaging. The experimental results show that prompt dissociation, which ensures the axial recoil and conserves information regarding the molecular orientation at a moment of dissociation, accompanies the dissociation of two

![Figure 3. The TOF spectra of singly charged ions produced by dissociative ionization of (a) C$_3$H$_5$F, (b) C$_3$H$_5$Cl, (c) C$_3$H$_5$Br, and (d) C$_3$H$_5$I irradiated with both 800 and 400 nm laser pulses at relative phase differences (1) $\phi = 0$ and (2) $\phi = \pi$. The solid lines indicate the pairs of forward and backward peaks. The dotted vertical lines indicate the centres of the peaks.](Image 320x194 to 536x448)

![Figure 4. Forward/backward yield ratios ($I_f/I_b$) as a function of the relative phase difference $\phi$: (open circles) halogen ions and (closed circles) C$_3$H$_5^+$ cations.](Image 320x531 to 536x771)
hydrogen atoms in addition to the halogen atom. The phase
dependences between the halogen ions and the counterpart
C3H5+ cations were completely out of phase with each other
for all the allyl halide molecules. This result shows that a
phase-controlled $\omega + 2\omega$ optical field discriminates the head–
tail order of oriented molecules.

As an additional experiment to calibrate $\phi$ and to
confirm the orientation direction of the ionized molecules,
we performed simultaneous measurements using gas mixtures of
C3H5F–C3H5I, C3H5Cl–C3H5I, and C3H5Br–C3H5I [29,31].
The forward/backward yield ratios $I_F/I_B$ of X+ generated from
C3H5X (X = F, Cl, Br) and I+ generated from C3H5I were
measured as a function of $\phi$. The F+ generated from C3H5F
and the I+ generated from C3H5I were completely out of phase
with each other. Furthermore, the Cl+ and Br+ generated from
C3H5Cl+ and C3H5Br+ and the I+ generated from C3H5I were
completely in phase with each other. (These results using
gas mixtures were used in the calibration of $\phi$ [34], and
are reflected in figure 4, in which the phase dependence of F+ is a cosine type whereas those of Cl+, Br+, I+ are ~cosine types). These results indicated that the direction of the oriented
molecules is the same in C3H5F, C3H5Br, and C3H5Cl and that
the C3H5F molecule is oriented in the opposite direction from
the other three allyl halides. We have experimentally succeeded in
observing the transition of the trend in the direction of
the ionized molecules by using a systematically changing
molecular system.

We now discuss the mechanism of OSM ionization.
The directions of the ionized molecules obtained from phase
dependence of the forward–backward yield ratios ($I_F/I_B$) show
that (1) for C3H5F, electrons are much more likely to
be extracted from the allyl part and (2) for C3H5Cl, C3H5Br,
and C3H5I, in contrast, electrons are much more likely to
be extracted from the halogen atom by the phase-controlled
$\omega + 2\omega$ field at $\phi = 0$ (\pi). For C3H5F and C3H5I, we can
safely say that the directions of the detected molecules are consistent with those expected by the molecular ADK model,
where electrons are extracted from the large-amplitude parts
(allyl part for C3H5F and the halogen atom for C3H5I). For
C3H5Cl and C3H5Br, since the direction of the ionized
molecules cannot be anticipated by just looking at the shape of
the HOMOs due to the intermediate structure, a comparison
between experiment and calculation would be required.

Finally, we describe the most recent situation regarding the
development of a theory for molecular TI. A theory for
molecular TI that considers the Stark effect, WFAT has been
developed within the single-active-electron approximation
[24,25]. The dominance of either the Stark shift or the shape
of the HOMO has been shown to depend on the molecule
and cannot be determined by just looking at the shape of the
HOMOs [24,25]. For one of the simplest cases of the
heteronuclear diatomic molecule CO, however, the WFAT
calculates maximum ionization rates that imply that the
HOMO amplitude on the O-side is smaller, which is opposite to
the experimental results [35]. Since the WFAT contains all
the information on the TI rates from the HOMO, the main source of
the difference between theory and experiment is multi-electron
effects such as the dipole moment of the core cation of CO+.

A recent time-dependent Hartree–Fock calculation shows good agreement with experimental data for the circularly polarized laser field, and the importance of dynamic core
polarization effects is discussed [36]. We have reported that the mechanism of the OMS-TI induced by phase-controlled
$\omega + 2\omega$ laser fields can be explained by the geometric structure of the HOMO [26,29–33]; thus some kind of screening
due to multi-electron effects that cancel the Stark shift
could be induced. A comprehensive theory involving multi-
electron effects for any molecule is now in progress, and the experimental results of OSM-TI for C3H5Cl and C3H5Br with intermediate conditions will be a benchmark for the developed
theory of molecular TI.

4. Conclusions

We have investigated the orientation-sensitive molecular TI
induced by intense phase-controlled $\omega + 2\omega$ pulses for the case of systematically changing orbital asymmetry C3H5X (X = F, Cl, Br, I). The $\omega + 2\omega$ method is applicable to a broad range of molecules that show hard fragmentation processes. The orientation direction of the ionized molecules is the same for
C3H5F, C3H5Br, and C3H5Cl, and is in the opposite direction for
C3H5I. In the cases of HOMOs with large asymmetry, C3H5F
and C3H5I, the direction of the ionized molecules is consistent
with that expected by OSM-TI based on the molecular ADK
model, and the effect of the geometric structure of the HOMO
is stronger than of the Stark shift. In the cases of C3H5Cl and
C3H5Br, for which it is not so easy to anticipate the direction of
the ionized molecules just by looking at the shape of the
HOMOs with intermediate structure, the Stark effect might
make some contribution. Although the WFAT, which fully
contains the effects of the geometric structure and the Stark
shift of the HOMO, has been developed within the single-
active-electron approximation [35], it has been pointed out
that a comprehensive theory involving multi-electron effects
for molecules is required [36]. Since our previous experiments
have shown that the mechanism of the OMS-TI induced by
the a phase-controlled $\omega + 2\omega$ laser fields can be explained
by the geometric structure of the HOMO [26,29–33], some
kind of screening due to multi-electron effects that cancel the
Stark shift could be induced. Our experimental results of
OSM-TI for C3H5Cl and C3H5Br with intermediate conditions
will be a benchmark for the developed theory of molecular TI.
Moreover, as it has been reported experimentally that the
molecular TI process is dependent on laser polarization [37],
the accumulation of experimental results for various molecules
investigated by different methods such as cold target recoil
ion momentum spectroscopy [28] and the TI for oriented
molecules by static electric fields [21] is required for in-depth
understanding of molecular TI.

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