Time-resolved photoelectron angular distributions from nonadiabatically aligned CO₂ molecules with SX-FEL at SACLA

Shinichirou Minemoto, Hiroyuki Shimada, Kazma Komatsu, Wataru Komatsu, Takuya Majima, Soichiro Miyake, Tomoya Mizuno, Shigeki Owada, Hirofumi Sakai, Tadashi Togashi, Makina Tabashi, Piero Deleva, Mauro Stener, Shota Tsuru, and Akira Yagishita

1. Introduction

Recent developments of x-ray free electron lasers (XFELs) [1–4] and table-top laser sources [5] are bringing off ultrafast imaging of molecules with femtosecond temporal and sub-Ångstrom spatial resolutions [6–9]. The first imaging of atomic motions during photochemical processes has been achieved by the ultrafast electron diffraction (UED) [10–12] and the ultrafast x-ray diffraction (UXD) [13–16]. Other imaging techniques such as the Coulomb explosion imaging (CEI) [17–20], the laser-induced electron diffraction (LIED) [21–23], and the ultrafast x-ray photoelectron diffraction (UXPD) [24–36] are also in progress. Among them, characteristic features of the XPD are element-specific and chemical-state-specific. More importantly, the x-ray photoionization cross sections are of four to six orders of magnitude larger than those for x-ray scattering [37]. From these reasons, the XPD is widely used for short-range surface structure analysis [38–40].

However, the UXPD for gas-phase molecules is only possible by fixing the molecules in space [41]. For instance, a spatiotemporal control of the axis distribution (alignment) for the molecular ensemble in the laboratory frame (LF) can be obtainable due to a rotational wave packet created with femtosecond IR laser pulses. Hence, at any given delay time between the IR pump and ionizing probe laser pulses, a set of different molecular alignment and polarization geometry is probed. That is, by preparing an aligned sample of molecules, the axis distributions and photoelectron angular distributions (PADs) of the CO₂ molecules aligned in the laboratory frame can be obtainable due to a rotational wave packet created with femtosecond IR laser pulses (LF). The axis distributions and PADs from the relevant molecular ensemble can be observed in the LF [42–45]. Up to now, such ultrafast time-resolved LFPADs have been intensively studied to gain insight into intramolecular dynamics [46–52]. In contrast to this, ultrafast molecular frame PADs (MFPADs) are now in progress, so that the present status of the UXPD is limited [29–34]. Namely, the ultrafast
MFPADs are necessarily demanding to determine ultrafast atomic motions during photochemical processes. In other words, the ultrafast processes such as dissociation, isomerization and intersystem crossing are best understood by MFPADs.

In the above mentioned context, prior to the final goal of the UXPD, here we present the time-resolved PADs from an aligned CO\textsubscript{2} molecular ensemble with the femtosecond soft x-ray (SX) pulses from SACLA SX-FEL [53]. The aligned molecular ensemble was prepared using the SACLA in-house Ti:sapphire NIR laser [54], and then evolves under field-free conditions. On one hand, the degree of alignment for the CO\textsubscript{2} molecular ensemble was determined by one of the facing velocity map imaging spectrometers (VMIs) [32, 34, 55], which recorded the momentum distribution of CO\textsuperscript{+} fragment ions resulting from Coulomb explosion of the doubly-charged CO\textsuperscript{2+} ions produced through double ionization by SX-FEL. On the other hand, using the other VMI, the time-resolved valence orbital PADs from the aligned molecular ensemble by SX-FEL was observed in the LF. Because the evolution of the prepared molecular ensemble is independent of ionization by the probe femtosecond SX pulses, one can consider such time-resolved PADs to be those from the partially aligned molecular ensemble, the degree of which was simultaneously determined by our facing VMIs. In the end, we will make the differences between the LFPADs for partially aligned molecules and the randomly aligned MFPAD clear, based on numerical simulations, since the interpretations of such PADs are often confused in the literatures [31, 49, 51].

2. Experiment

The experiments were performed at the SX beamline BL1 at SACLA [53]. The SX-FEL was operated at the wavelength of 22.4 nm (55.4 eV) and the bandwidth of \(~\sim\)2% (\(~\sim\)1 eV). The duration of the SX-FEL pulses, \(\tau_\text{SX}\), was estimated as \(~\sim\)100 fs (FWHM) and the pulse energy was \(~\sim\)50 \(\mu\)J with a diameter of \(~\sim\)10 \(\mu\)m. For the pump laser pulses, we used the optical laser system synchronized to SX-FEL at SACLA [54]. The Ti:sapphire NIR laser was operated at the central wavelength of 793 nm (1.56 eV) and the bandwidth of \(~\sim\)30 nm. The NIR laser pulse duration, \(\tau_\text{L}\), measured by a single-shot autocorrelator was 300 fs (FWHM), which was slightly stretched from the Fourier-limited pulse to reduce the effects of timing jitter between the NIR and SX-FEL pulses. The NIR pulse energy was \(~\sim\)5 mJ; the peak intensities on the order of \(10^{14}\) W cm\(^{-2}\) could be reached at the focus of \(~\sim\)100 \(\mu\)m. The NIR laser beam was combined with the SX-FEL beam by a holey mirror in a vacuum chamber almost collinearly, and the spatial overlap between the two beams was monitored by a Ce:YAG screen inserted in the center of the facing VMI spectrometers, see [32, 34] for details. The delay time \(t\) between the NIR and SX-FEL pulses was controlled by a translational stage inserted in the NIR beam path. Firstly, the delay time \(t\) was roughly scanned with an interval of 0.67 ps between \(t = -2\) ps and 47 ps. From this, overall behavior of the degree of alignment, \(\langle \cos^2 \theta \rangle\), during the first rotational period, \(T_{\text{rot}} = 42.7\) ps, was examined, including the timing of the rapid modulations around \(t = 0\) and \(T_{\text{rot}}\) and the degrees of so-called permanent alignment (the DC level due to higher J states [36]). Next, the delay time \(t\) was finely scanned with an interval of 0.2 ps at around delay times when \(\langle \cos^2 \theta \rangle\) exhibits rapid modulations; near the pump pulse irradiation (from \(t = -1.6\) to 2.0 ps), around the half revival (from \(t = 18.7\) to 24.6 ps), and around the full revival (from \(t = 40.8\) to 46.8 ps). The pulsed molecular beam was crossed by the collinear laser beams. Then, the electrons produced in the interaction region were drawn into the lower VMI, while the ions were drawn into the upper VMI: the former recorded two-dimensional (2D) photoelectron momentum images, and the latter recorded 2D ion momentum images. Here, for the ion detection, we applied a pulsed high voltage of \(~\sim\)2 kV to the microchannel plate at the flight time corresponding to a specific mass-to-charge ratio. By this gating procedure, we measured only the desired CO\textsuperscript{+} fragment ions, which best represent the angular distributions of the molecular axis.

On one hand, both the SX-FEL and the synchronized NIR laser were operated at a repetition rate of 30 Hz. On the other hand, the pulsed valve for the molecular beam was operated at a 15 Hz repetition rate. Thus, the 2D images (signal + background) with and those (background) without the sample molecules were alternately measured, and then the 2D images without both the noise from the residual gas and from the scattered XUV were obtained by subtracting the background from the (signal + background) 2D images. The 2D data set consists of 2,500 images at each delay time.

3. Results and discussions

3.1. Alignment of CO\textsubscript{2} induced by NIR laser

In order to obtain the alignment states of the molecular axis, we analyzed the concentric parts of the CO\textsuperscript{+} fragment ion images, which correspond to the Coulomb explosion channel of CO\textsuperscript{+} + O\textsuperscript{−} from the doubly ionized molecular ions CO\textsuperscript{2+}. Then, we introduced the orientation distribution function \(P(\theta, t)\) [57], which is defined by
$F(\theta, t) = \frac{I(\theta, t)}{I_{\text{ran}}(\theta)}$  \hspace{1cm} (1)

where $I(\theta, t)$ is the CO$^+$ ion signal intensities at the delay time $t$, and $I_{\text{ran}}$ the intensities at $t = -0.4$ ps, which corresponds to the random orientation. The angle $\theta$ is the ejection angle of the CO$^+$ fragment ion relative to the polarization vector of the NIR laser pulse. The function $F(\theta, t)$ is normalized to $\int_{-\pi}^{\pi} F(\theta, t) \sin \theta d\theta = 1$, so that the time-dependent degree of alignment $\langle \cos^2 \theta \rangle(t)$ can be described by

$\langle \cos^2 \theta \rangle(t) = \int_{-\pi}^{\pi} F(\theta, t) \cos^2 \theta \sin \theta d\theta$. \hspace{1cm} (2)

Since the FEL pulses are generated by the self-amplified spontaneous emission (SASE) process [58], their intensities and temporal and spectral shapes vary significantly from shot to shot, and moreover the FEL pulses have a finite timing jitter on the order of $\sim 1$ ps with the synchronized optical pulses [55]. Thus, in our experiment, the degrees of alignment were much decreased due to the inevitable timing jitter between the SX-FEL and NIR pulses, i.e., the fluctuation of $t$ [55]. To remove the degradation of the alignment due to the timing jitter, we tried to rearrange the single-shot ion images by referring to the theoretical values for the degree of alignment: Firstly, the degrees of alignment were calculated by solving the time-dependent Schrödinger equation according to [59]. The calculated results under the rotational temperature of 6 K and the peak intensity of the NIR laser of $5 \times 10^{13}$ W cm$^{-2}$ are shown in figure 1. Among the parameters investigated, the results with these values best reproduce the maximum $\langle \cos^2 \theta \rangle(t)$ of 0.540 at around $T_{\text{rot}}$ and the permanent alignment with $\langle \cos^2 \theta \rangle(t) = 0.377$ simultaneously, both of which are experimental observations. Secondly, at $\sim T_{\text{rot}}/2$ and $\sim T_{\text{rot}}$, the delay time $t$ for each ion image was ordered within 0.7 ps by comparing the degrees of alignment determined from the image with the calculated ones. Thus, the 2,500 images for a single nominal delay time were distributed to seven different delay times. Finally, we obtained the reasonably ordered results of the time evolution of $\langle \cos^2 \theta \rangle(t)$, which is depicted in figure 1. By this cumbersome procedure, we got rid of the timing jitter effects on the time evolution of the alignment degrees partially.

Just after the irradiation of the NIR pump pulse, the rotational wave packet is created, showing maximum $\langle \cos^2 \theta \rangle$ of 0.540 at $t = 0.4$ ps. Following the dephasing, the rotational wave packet shows a permanent alignment with $\langle \cos^2 \theta \rangle = 0.377$. The degree of alignment $\langle \cos^2 \theta \rangle$ modulates with an interval of 10.6 ps, which corresponds to $T_{\text{rot}}/4$, and the rotational wave packet repases at 22.0 ps ($\sim T_{\text{rot}}/2$) and 43.6 ps ($\sim T_{\text{rot}}$) showing the minimum $\langle \cos^2 \theta \rangle = 0.255$ and the maximum $\langle \cos^2 \theta \rangle = 0.540$, respectively. A slight mismatch between the experimental and theoretical results may be due to the incomplete arrangement of the delay time and to the uneven detection efficiencies when the desired ions were gated. Since the duration of the gating pulse was as short as the width of the flight time of the fragment ions, the detection efficiencies might depend on the azimuth angles of ion ejection, by which $\langle \cos^2 \theta \rangle$ might be slightly underestimated.

To evaluate the degree of alignment $\langle \cos^2 \theta \rangle$ by equation (2) in the above mentioned procedure, we derive the time-dependent orientation distribution function $F(\theta, t)$ from the CO$^+$ fragment ion images. Such functions are shown in figure 2. Taking the molecular symmetry of the CO$_2$ into account, this orientation
distribution function $F(\theta, t)$ can be expanded by the Legendre polynomials $P_K(\cos \theta)$ of even integers of $K$;

$$F(\theta, t) = \sum_k a_k(t) P_k(\cos \theta).$$  \hspace{1cm} (3)

Thus, to derive the coefficients $a_k(t)$, equation (3) was fitted to the experimental results at $t = -0.4$, 22.0 (about $T_\text{rot}/2$), and 43.6 ps ($T_\text{rot}$). The fitted curves are shown in figure 2 together with the experimental data. The obtained values of $a_k(t)$ are summarized in table 1 with the corresponding values of the degrees of alignment.

### 3.2. Photoelectron angular distributions from aligned CO$_2$

The 2D photoelectron images by 55.4-eV photons of the SX-FEL, which are the counterpart of the properly ordered ion images in the previous section, are Abel inverted by the pBASEX procedure [60] to obtain the 2D cut of the 3D photoelectron momentum distribution. Figure 3 displays the inverted Abel image of photoelectrons from the CO$_2$ molecules aligned along the polarization vector of the NIR laser pulse. In our experimental condition, the polarization vectors of both the NIR pump and the SX-FEL probe pulses are parallel to each other.

The lower panel shows the corresponding photoelectron spectrum, which was obtained by the integration over all the emission angles of the photoelectrons. The peak labeled by X at the kinetic energy of 42 eV corresponds to the ionization of the $1\pi_g$ level of the CO$_2^+$ ions [61]. The peak labeled by A, B, and C at 38 eV is the ionization of the $1\pi_u$, $3\sigma_u$, and $4\sigma_g$ levels, resulting in the lower excited states, A, B, and C of the CO$_2^+$. The energy resolution of our VMI, $\Delta E/E$ of about 5%, can distinguish the X state from the other excited states, while these excited states are merged to a single peak. The inner valence photoelectrons are observed at 18, 25, and 30 eV, which are obscured by the tail of back scattering electrons from the microchannel plate for the ion detection [91].

### Table 1. The coefficients $a_k(t)$ of the Legendre polynomials $P_K(\cos \theta)$ in equation (3).

| $t$   | $\langle \cos^2 \theta \rangle$ | $a_0$ | $a_2$ | $a_4$ | $a_6$ | $a_8$ | $a_{10}$ | $a_{12}$ | $a_{14}$ |
|-------|---------------------------------|-------|-------|-------|-------|-------|---------|---------|---------|
| -0.4 ps | 0.335                           | 1.000 | 0.001 |       |       |       |         |         |         |
| 22.0 ps  | 0.255                           | 0.986 | -0.583 | 0.159 | -0.002 | -0.008 |         |         |         |
| 45.6 ps  | 0.540                           | 0.999 | 1.549 | 0.674 | 0.181 | 0.074 |         |         |         |
| 11.2 ps $^a$ | 0.839                           | 0.997 | 3.372 | 5.816 | 5.555 | 7.810 | 7.605   | 9.901   | 9.940   |

$^a$ The delay time at which the theoretical calculation gives the highest value of $\langle \cos^2 \theta \rangle$ under the condition of the rotational temperature of 6 K and the peak intensity of $2 \times 10^{14} \text{ W cm}^{-2}$.

### Figure 2. The orientation distribution functions $F(\theta, t)$ of equation (1).

(a) random, (b) anti-aligned, and (c) aligned. Dots; experimental results, and solid lines; fitted results of equation (3). The statistical errors of the experimental results are ±0.5%.

Figure 2. The orientation distribution functions $F(\theta, t)$ of equation (1). (a) $t = -0.4$ ps, $\langle \cos^2 \theta \rangle = 0.333$, (b) $t = 22.0$ ps, $\langle \cos^2 \theta \rangle = 0.255$, and (c) $t = 43.6$ ps, $\langle \cos^2 \theta \rangle = 0.540$. Dots; experimental results, and solid lines; fitted results of equation (3). The polarization vector of the NIR laser is shown by the double-headed arrows. The distribution functions are used for the theoretical simulations of equation (4). The statistical errors of the experimental results are ±0.5%.

Table 1. The coefficients $a_k(t)$ of the Legendre polynomials $P_K(\cos \theta)$ in equation (3).

| $t$   | $\langle \cos^2 \theta \rangle$ | $a_0$ | $a_2$ | $a_4$ | $a_6$ | $a_8$ | $a_{10}$ | $a_{12}$ | $a_{14}$ |
|-------|---------------------------------|-------|-------|-------|-------|-------|---------|---------|---------|
| -0.4 ps | 0.335                           | 1.000 | 0.001 |       |       |       |         |         |         |
| 22.0 ps  | 0.255                           | 0.986 | -0.583 | 0.159 | -0.002 | -0.008 |         |         |         |
| 45.6 ps  | 0.540                           | 0.999 | 1.549 | 0.674 | 0.181 | 0.074 |         |         |         |
| 11.2 ps $^a$ | 0.839                           | 0.997 | 3.372 | 5.816 | 5.555 | 7.810 | 7.605   | 9.901   | 9.940   |

$^a$ The delay time at which the theoretical calculation gives the highest value of $\langle \cos^2 \theta \rangle$ under the condition of the rotational temperature of 6 K and the peak intensity of $2 \times 10^{14} \text{ W cm}^{-2}$.

Due to the voltage settings of the VMIs, some of electrons produced at the surface of the microchannel plate for the ion detection are accelerated toward that for photoelectron detection. They are detected as signals at later flight times and with broad and smaller kinetic energies than proper photoelectron signals.
spectra integrated over all the angles, i.e., the photoionization cross sections for each level, were independent of the delay time \( t \), in other words, the degree of alignment, although they are not shown here, as can be expected.

In what follows we consider the photoelectron angular distributions (PADs) from the valence levels of the aligned CO\(_2\) molecules. The time-dependent PADs from the 1\( \pi \)g level of the CO\(_2\) are demonstrated as the polar plots in figure 4; (a) \( t = -0.4 \) ps (randomly aligned), (b) \( t = 22.0 \) ps (aligned perpendicular to the polarization vector), and (c) \( t = 43.6 \) ps (aligned along the polarization vector). Such laboratory frame PADs (LFPADs) from the aligned molecular ensemble are described by the sum of the molecular frame PADs (MFPADs) [62] weighted by the orientation distribution function of equation (3) [63]. Taking both the polarization vector of the SX-FEL along the LF \( z \)-axis and the symmetry of the CO\(_2\) molecules into account, the time-dependent LFPAD can be expressed by [63]

\[
I (\theta_e, t) \propto \sum_{L, L', K, M} \left( 10, 10|L, 0 \right) A_{LM}^{L'} (-1)^{M}(I - M, L, M|K0)(L0, L, 0|K0) \times \frac{\sqrt{2L + 1}}{(2K + 1)} a_k(t) P_L (\cos \theta_e)
\]

Figure 3. Abel inverted image of photoelectrons from CO\(_2\) molecules at \( t = 43.6 \) ps (upper) and the corresponding angle-integrated photoelectron energy spectrum (lower). Polarization vectors of the NIR and SX-FEL are parallel to each other as shown by the double-headed arrows. The peak labeled A at 42 eV are attributed to the ionization of 1\( \pi \)g level and those labeled C, B, and C at 38 eV to the 1\( \pi \)u, 3\( \sigma \)u, and 4\( \sigma \)g levels. The statistical errors are less than \( \pm 1\% \).

where the dynamical parameter \( A_{LM}^{L'} \) is described by the dipole matrix elements for photoionization [64], \( L \) the vector sum of the angular momenta of photoelectron partial waves in the MF, \( L_s (=0 \text{ and } 2) \) the vector sum of photon angular momentum, and \( \theta_e \) the angle between the photoelectron momentum and the LF \( z \)-axis. Since \( L_s \) and \( K \) are even integers, even integers of \( L \) are allowed by the non-zero conditions of the Clebsch–Gordan coefficients, \( (L0, L, 0|K0) \neq 0 \), in equation (4), and also the maximum integer of \( L \) is given by the condition.
Using the experimentally determined $a_k(t)$ values in equation (3) and the dipole matrix elements for photoionization of the $1\pi_g$ level calculated with the time-dependent density functional theory (TDDFT) [65], we have simulated the time-dependent LFPADs. The simulation results are shown in figure 4 to compare with the corresponding experimental data. In figure 4(a), the experimental LFPAD is perfectly reproduced by the simulation. It should be noted that the simulated LFPAD for the randomly aligned molecules in figure 4(a) is consistent with the LFPAD for the asymmetry parameter $\beta = 1.26$, which was calculated with the TDDFT. In figure 4(b), both the experimental and simulated LFPAD, for the molecules aligned perpendicular to the polarization vector, are slightly changed from that for the randomly aligned molecules in figure 4(a). In figure 4(c), one can see considerable change of the LFPAD, for the molecules aligned parallel to the polarization vector, from those in figures 4(a) and (b). This change is reasonably reproduced by the simulated LFPAD according to equation (4), which is the average of the MFPAD over the molecular orientation, as can be seen in the figure.

The LFPADs for the sum of the $1\pi_g, 3\sigma_u$, and $4\sigma_g$ levels didn’t exhibit the alignment-dependent nature, i.e., time-dependent nature, although they are not shown here.

**4. Summary and outlook**

Using quantum mechanical nature, i.e., revival of the rotational wave packets, which were induced by the femtosecond NIR laser, we have measured the time-dependent LFPADs from the partially aligned CO$_2$ molecules under the field-free condition. The time dependent LFPADs have been well reproduced by the TDDFT calculations considering the time-dependent degree of alignment, in other words, molecular axis distributions. In this context, higher degree of alignment is most desirable in developing the UXPD.

To get a feeling on the alignment effect on the LFPAD, we simulated the LFPADs for the $1\pi_g$ level of the CO$_2$ at the photon energy of 55.4 eV, under the condition of $\langle \cos^2 \theta \rangle = 0.84$ (see the last line in table 1). This degree of alignment was calculated under the extreme experimental condition of the rotational temperature of 6 K and the peak intensity of the NIR laser of $2 \times 10^{14}$ W cm$^{-2}$. The simulation results are shown in figure 5. As can be seen from the figure, under the present experimental condition of $\langle \cos^2 \theta \rangle = 0.54$, the characteristic feature of the MFPAD in figure 5(c) consisting of lobes and nodes are fully smeared out by the convolution of it with the molecular axis distributions. In contrast to this, under the promising condition of $\langle \cos^2 \theta \rangle = 0.84$, every lobe and node of the MFPAD is observed in the LFPAD. From this, we can say that when one can measure the LFPAD, for example, for the degree of alignment $\langle \cos^2 \theta \rangle > 0.8$, one can determine the matrix elements of the photoionization by the analyses of such experimental LFPAD, on the analogy of our previous works on the complete photoionization experiments, which derives the matrix elements from the MFPAD measured from the photoelectron–fragment–ion coincidence experiments [66]. Conversely, one can derive the unknown molecular geometry from the experimental MFPAD or LFPAD measured with the high degree of alignment, when the theoretical calculations give accurate matrix elements of the photoionization for a given molecular geometry. In this context, the geometrical change of laser-aligned molecules caused by another femtosecond optical laser can be probed by the SX-FEL. This is our scenario of the UXPD: we trigger the photochemical reaction by an optical
laser and determine ultrafast atomic motions by SX-FEL during dissociation, isomerization and intersystem crossing [25, 35, 36].

In the end, we can say that we stepped towards the final goal of the UXPD by the success of the time-resolved LFPAD measurement in the present work.

Acknowledgments

The authors appreciate fruitful discussions with J. Itatani (The University of Tokyo). They thank the operation and engineering staff members of SACLA for their support in performing the XFEL experiments, which were conducted at the BL1 of SACLA with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposal No. 2017B8011). They acknowledge the computer time in CINECA through the IseC_UTREP grant. This work is financially supported from JSPS KAKENHI (Nr. 16H02132, Nr. 16K05496, and Nr. 17H05214). K. K and W. K are financially supported by Advanced Leading Graduate Course for Photon Science (ALPS) of MEXT.

ORCID iDs

Shinichirou Minemoto @ https://orcid.org/0000-0001-5868-823X
Hiroyuki Shimada @ https://orcid.org/0000-0003-0207-2238
Takuya Majima @ https://orcid.org/0000-0003-2804-1915
Shota Tsuru @ https://orcid.org/0000-0003-0014-7873

References

[1] Emma P et al 2010 Nat. Photon. 4 641
[2] Ishikawa T et al 2012 Nat. Photon. 6 540
[3] Margaritondo G 2017 Riv. Nuovo Cimento 40 411–71
[4] Helml W et al 2017 Appl. Sci. 7 915
[5] Calegari F et al 2016 J. Phys. B: At., Mol. Opt. Phys. 49 062001
[6] Elsasser T E and Woerner M 2017 Appl. Sci. 7 915
[7] Xu J, Blaga C I, Agostini P and DiMauro L F 2016 J. Phys. B: At., Mol. Opt. Phys. 49 112001
[8] Miller R J D 2016 Faraday Discuss. 194 777
[9] Elsasser T 2017 Chem. Rev. 117 10621
[10] Yang J et al 2016 Phys. Rev. Lett. 117 153002
[11] Vecchioine T et al 2017 Rev. Sci. Instrum. 88 033702
[12] Zandi O, Wilkin K J, Xiong Y and Centurion M 2017 Struct. Dyn. 4 044022
[13] Głownia J M et al 2016 Phys. Rev. Lett. 117 153003
[14] Kirrander A and Weber P M 2017 Appl. Sci. 7 7534
[15] Kowalewski M, Bennett K and Mukamel S 2017 Struct. Dyn. 4 054101
[16] Chergui M and Collet E 2017 Chem. Rev. 117 11025
[17] Légaré F et al 2006 J. Phys. B: At., Mol. Opt. Phys. 39 S503
[18] Cornaggia C 2009 Laser Phys. 19 1660
[19] Karimi R, Liu W-K and Sanderson J 2016 Femtosecond laser-induced Coulomb explosion imaging (Advances in Multi-Photon Processes and Spectroscopy vol 23) ed S H Lin, A A Villays and Y Fujimura (Singapore: World Scientific)
[20] Burt M et al 2017 Phys. Rev. A 96 043415
[21] Ito Y et al 2016 Struct. Dyn. 3 034303
[22] Wolter B et al 2016 Science 354 308
[23] Morimoto Y, Kanya R and Yamanouchi K 2017 Laser-assisted electron scattering and diffraction for ultrafast imaging of atoms and molecules Progress in Photon Science ed K Yamanouchi (Berlin: Springer)
[24] Krasniqi F et al 2010 Phys. Rev. A 81 033411
[25] Kazama M et al 2013 Phys. Rev. A 87 063417
[26] Sun S X L, Kaduwela A P, Gray A X and Fadley C S 2014 Phys. Rev. A 89 053415
[27] Wang X et al 2016 Sci. Rep. 6 23635
[28] Nguyen N T, Lucchese R R, Lin C D and Le A T 2016 Phys. Rev. A 93 063419
[29] Boll R et al 2013 Phys. Rev. A 88 061402(R)
[30] Rolles D et al 2014 J. Phys. B: At., Mol. Opt. Phys. 47 124035
[31] Boll R et al 2014 Faraday Discuss. 171 57
[32] Nakajima K et al 2015 Sci. Rep. 5 14065
[33] Yagishita A 2015 J. Electron Spectrosc. Relat. Phenom. 200 247
[34] Minemoto S et al 2016 Sci. Rep. 6 38654
[35] Tsutsumi S, T, Fujikawa T and Yagishita A 2017 Phys. Rev. A 95 043404
[36] Tsutsumi S et al 2018 J. Chem. Phys. 148 124101
[37] Stout G H and Jensen L H 1989 X-Ray Structure Determination: A Practical Guide 2nd edn (New York: Wiley)
[38] Berman H M et al 2002 Acta Crystallogr., Sect. D: Biol. Crystallogr. 58 899
[39] Rhodes G 2006 Crystallography Made Crystal Clear: A Guide for Users of Macromolecular Models 3rd edn (San Diego, CA: Academic)
[40] Fadley C S 2010 J. Electron Spectrosc. Relat. Phenom. 178–179 2
[41] Filsinger F et al 2012 Phys. Chem. Chem. Phys. 14 13570–7
[42] Johnson P et al 2009 J. Phys. B: At. Mol. Opt. Phys. 42 134017
[43] Kellensberg F et al 2011 Phys. Rev. A 84 051404(R)
[44] Marceau C et al 2017 Phys. Rev. Lett. 119 083401
[45] Reid K L et al 1999 J. Chem. Phys. 111 1438
[46] Tsutsumi S et al 2001 Phys. Rev. Lett. 86 4500
[47] Suzuki T 2006 Annu. Rev. Phys. Chem. 57 555
[48] Kumarappan V et al 2008 Phys. Rev. Lett. 100 093006
[49] Bisgaard C Z et al 2009 Science 323 1464
[50] Holmegaard L et al 2010 Nat. Phys. 6 428
[51] Maurer J et al 2012 Phys. Rev. Lett. 109 123001
[52] Rouzée A et al 2012 J. Phys. B: At. Mol. Opt. Phys. 45 074016
[53] Owada S et al 2018 J. Synchrotron Rad. 25 282
[54] Owada S et al 2018 J. Synchrotron Rad. 25 68
[55] Minemoto S et al 2018 J. Phys. B: At. Mol. Opt. Phys. 51 075601
[56] Sonderegger A A, Shepperson B and Stapelfeldt H 2017 J. Chem. Phys. 147 013905
[57] Minemoto S and Sakai H 2011 J. Chem. Phys. 134 214305
[58] Saldin E L, Schneidmiller E A and Yurkov M V 2000 The Physics of Free Electron Lasers (Berlin: Springer)
[59] Ortigoso I, Rodriguez M, Gupta M and Friedrich B 1999 J. Chem. Phys. 110 3870
[60] Garcia G A, Nahon L and Powis I 2004 Rev. Sci. Instrum. 75 4989
[61] Katsumata S, Achiba Y and Kimura K 1979 J. Electron Spectrosc. Relat. Phenom. 17 229
[62] Dill D et al 1978 Phys. Rev. Lett. 41 1230
[63] See for example Underwood J G and Reid K L 2000 J. Chem. Phys. 113 1067
[64] Motoki S et al 2000 J. Phys. B: At. Mol. Opt. Phys. 33 4193
[65] Yagishita A et al 2005 J. Electron Spectrosc. Relat. Phenom. 142 295
[66] Adachi J et al 2007 J. Phys. B: At. Mol. Opt. Phys. 40 29
[67] Stener M 2005 J. Chem. Phys. 122 234501
[68] Motoki S et al 2002 Phys. Rev. Lett. 88 063003
[69] Motoki S et al 2002 J. Phys. B: At. Mol. Opt. Phys. 35 3801
[70] Teramoto T et al 2007 J. Phys. B: At. Mol. Opt. Phys. 40 F241
[71] Teramoto T et al 2007 J. Phys. B: At. Mol. Opt. Phys. 40 4033