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Ion momentum spectroscopy of N$_2$ and O$_2$ molecules irradiated by EUV free-electron laser pulses

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Abstract
We have investigated dissociative ionization of N$_2$ and O$_2$ molecules by 52 nm extreme-ultraviolet light pulses at the free-electron laser facility in Japan. Distributions of kinetic energy release were measured at two different laser power densities below $3 \times 10^{13}$ W cm$^{-2}$ and intermediate and final states of the sequential two photon transitions were identified.

A free-electron laser (FEL) based on self-amplified spontaneous emission (SASE) in the extreme ultraviolet (EUV) region below $\lambda = 100$ nm was first developed in Germany and has proven to be a very powerful tool to explore the interaction of strong EUV laser pulses with atoms [1–4], molecules [5–7] and clusters [8–11].

In May 2008, a new facility, the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan started operation for users [12]. It provides linearly polarized EUV-FEL pulses ($\sim$30 $\mu$J per pulse, $\sim$100 fs pulse width, 10–20 Hz repetition rate) in the wavelength region 51–61 nm. In this energy regime, all atoms and molecules can be ionized by just a single photon with huge photoionization cross sections. We have demonstrated that this source is indeed ideal to investigate multiphoton multiple ionization of clusters [13, 14]. In the present work, we have investigated dissociative ionization of N$_2$ and O$_2$ molecules caused by two-photon absorption of the EUV-FEL pulses at 52 nm (photon energy of 24 eV) from this light source, using a dead-time-free multi-particle detection system [15]. The photon energy employed in the present experiment was set to be between single ionization thresholds and double ionization thresholds of N$_2$ and O$_2$, and twice the photon energy is above double ionization thresholds. Thus, N$_2$ and O$_2$ are ionized by single-photon absorption, and ionized N$_2$ and O$_2$ can be ionized or excited by absorption of another photon. Using tightly focused intense FEL pulses, first and second photoabsorption processes can occur with a single FEL pulse duration. Jiang et al also investigated sequential ionization of N$_2$ by 44 eV FEL pulses at FLASH [7]. Because of difference in the photon energy, we found different intermediate and final states and decay pathways. The first evidence of multiphoton multiple ionization of N$_2$ by the EUV-FEL pulses from the SCSS test accelerator was reported by Sato et al [16].

A new experimental setup employed here is similar to the previous one described and employed in [13, 14, 17]. We briefly describe the difference from the previous setup. To reduce background signals from residual gases, the base
pressure at ion momentum spectrometer chamber has been improved by one order and kept below \(10^{-9}\) Pa. The time-of-flight axis of the spectrometer is vertical and thus perpendicular to the horizontal axes of the FEL beam and of its polarization. The supersonic molecular beam is horizontal and crosses the FEL beam at 45°.

The 52 nm FEL beam was steered by two upstream Au plane mirrors, skimmed by a 5 mm hole 1.2 m upstream the entrance of the experimental chamber, and then introduced to the chamber placed \(~26\) m downstream from the radiation source point. To focus the FEL beam onto the molecular beam of \(~1\) mm in diameter, we used a homemade concave mirror at normal incidence [13]. The FEL beam was partially blocked by a 1.5 mm wide horizontal beam stopper so that the non-focused beam did not irradiate directly the molecular beam. The intensity of the FEL beam was adjusted using a gas attenuator, i.e., an absorption cell filled with the Ar gas.

Using electrostatic fields, ions are projected onto a 120 mm diameter microchannel plate (MCP) in front of a delay-line anode. The determination of the ion momentum is based on the measurements of the time-of-flight (TOF) and the detector hit position for each ion (see, e.g. [18]). Here, a three-layer type delay-line anode (Roentdek HEX120) is used to minimize the dead time. The design of the ion spectrometer with a two-stage acceleration section is similar to the one described in [19]. The lengths and fields are chosen to be 40 mm, 128 V mm\(^{-1}\) and 52 mm, 226 V mm\(^{-1}\), respectively. The acceleration is followed by a 308 mm long field-free drift tube with the ion detector mounted at its exit. All signals are fed to an eight-channel digitizer (Acqiris DC282 \times 2) and software CFD is employed for extracting the timing signals [15].

Figure 1(a) shows kinetic energy release distributions (KERDs) from O\(_2\) (i.e. twice the kinetic energy of O\(^+\)) irradiated by the FEL of 3 and 1 \(10^{13}\) W cm\(^{-2}\). The ratio of the FEL power was determined by the ion yields due to residual gases which are proportional to the FEL power. The O\(^+\) ions were also produced from the residual H\(_2\)O. Thus, we recorded the signals with and without the molecular beam of O\(_2\). The spectra in figure 1 are the results after subtraction of the background contributions. For the spectrometer setting described above, we could collect all O\(^+\) ions emitted into 4 sr with kinetic energies up to \(~20\) eV. The O\(_2^+\) in the B\(^2\)Σ\(_g^+\) state is known to emit O\(^+\) of 0.8 eV kinetic energy and ionization threshold of this state is 20.3 eV [20]. Thus, the sharp peak at 1.6 eV in figure 1(a) is assigned to single photoionization to the B\(^2\)Σ\(_g^+\) dissociative state. The KER in the 0–3 eV range does not exhibit significant FEL power dependence, indicating that one-photon processes are saturated in the present experimental conditions.

The ion yields with KER above 3 eV decreases almost linearly with decreasing the FEL power. This power dependence indicates that the ion yields with KER above 3 eV is mostly due to two-photon processes. Close inspections reveal that the ion yields with KER above 13 eV decrease more rapidly with decreasing the FEL power, indicating contributions from the processes that involve more than two photons. The KERD depicted in figure 1(b) is extracted from the O\(^+\)–O\(^+\) coincidences. It has a shape similar to the one in figure 1(a) above 3 eV. This indicates that the ion yields with KER above 3 eV are mostly attributed to O\(^+\)–O\(^+\) pair formation from the dicaticonic states O\(_2^{2+}\) produced via sequential photo-doubleionization by sequential two-photon absorption.

Hsieh et al measured KERD for O\(^+\)–O\(^+\) pair using He II light [21]. The KERD for O\(^+\)–O\(^+\) in figure 1(b) is in good agreement in shape with the KERD measured at a photon energy of 48.4 eV. Thus O\(^+\)–O\(^+\) pairs in figure 1(b) are also released via the same dicaticonic states as in [21]. Dicaticonic states contributing to the KERD measured in [21] lie between 42 and 48 eV within the Franck–Condon region. These states cannot be reached by sequential two-photon absorption via X\(^2\)Π\(_g\) and a\(^4\)Π\(_g\) states of O\(_2^+\), because vertical ionization potential to these states are 12.307 and 16.703 eV, respectively [22]. The first step of the sequential two-photon absorption we observed are thus the excited states of O\(_2^+\) such as b\(^4\)Σ\(_g^+\) and B\(^2\)Σ\(_g^+\). Partial photoionization cross sections of O\(_2\) to b\(^4\)Σ\(_g^+\) and B\(^2\)Σ\(_g^+\) at 24 eV are indeed in the same order of that of X\(^2\)Π\(_g\) [23].

KERD for O\(_2^{2+}\) is also shown in figure 1(b). It required three or more photons because the lowest dissociation limit for O\(_2^{2+}\) + O is 53.85 eV [26, 27]. Saito and Suzuki measured kinetic energy of O\(_2^{2+}\) [24] and for O\(_2^{2+}\)–O\(^+\) coincidence [25] caused by the O K-shell excitation/ionization and following Auger decay. They described that KERD for O\(_2^{2+}\)–O\(^+\) coincidence have a broad peak at about 20 eV and a full width at the half maximum of about 12 eV [25]. In contrast, non-coincident KERD for O\(_2^{2+}\) has another peak at about 12 eV. Thus O\(_2^{2+}\) detected in this work is likely due to dissociation from O\(_2^{2+}\) to O\(_2^{2+}\) + O.
photon processes in N2 are not completely saturated because processes in which three or more photons are involved: one-due to two-photon processes and those above 12 eV are due to this indicates that the ion yields with KER up to 12 eV are mainly linear but weaker than quadratic. The ion yields with KER the same in the KER range up to 12 eV: it is stronger than this. Note that the KERD for 0.4 × 10^{13} W cm^{-2} is multiplied by 5. (b) The KERDs for N^+–N+ coincidences and N2+ at 2 × 10^{13} W cm^{-2}.

Figure 2(a) shows KERDs from N2, obtained from the N+ kinetic energy distributions, recorded at 2 and 0.4 × 10^{13} W cm^{-2}. The vertical ionization potential of the lowest dissociative ionic state is 24.29 eV [26, 27] and thus there is no dissociative ionic state reach by a one-photon absorption. The FEL power dependence of the ion yields is approximately the same in the KER range up to 12 eV: it is stronger than linear but weaker than quadratic. The ion yields with KER above 12 eV sharply drop by reducing the FEL power. This indicates that the ion yields with KER up to 12 eV are mainly due to two-photon processes and those above 12 eV are due to processes in which three or more photons are involved: one-photon processes in N2 are not completely saturated because FEL power is weaker than in the case of O2.

The KERD for N^+–N+ coincidences is also shown in figure 2(b). The comparison between coincidence and non-coincidence KERD indicates that the ion yields forming the two peaks at 4 eV and 10 eV are mainly due to dissociation from N_2^+ ionic states produced via sequential two-photon absorption. By the first one-photon absorption at 24 eV, three ionic states X^2\Sigma^+_g, A^2\Pi_u and B^2\Sigma^+_g are mainly populated at vertical ionization potentials of 15.580, 16.926 and 18.751 eV, respectively [28]. The branching ratios for the production of these states are about 37, 54 and 9%, respectively [29]. Via the second photon absorption by these ionic states, the highly excited ionic states at ionization potentials of 40, 41 and 43 eV are expected to be produced.

Gagnon et al measured photoelectron–photoion correlation by 43 eV light generated by high-harmonic upconversion of a femtosecond laser [30]. They found that there are states around 40 eV dissociating to N + N^+ and that the sum of KER and photoelectron kinetic energy KE correlated to these states split into two peaked distributions, 12–18 eV and 5–9 eV, reflecting two peaks in the KERD. The two different KERD peaks correspond to the two different dissociation limits N+(2s^22p^2) + N(2s^22p^3) and N^+(2s^22p^2) + N(2s^22p^33\ell). The reason why two dissociation channels appear is because the two different characters are embedded in the states at energies around 40 eV, i.e., inner-valence single-hole character responsible for dissociating to N+(2s^22p^2) + N(2s^22p^3) and two-hole one-electron character responsible for N+(2s^22p^2) + N(2s^22p^33\ell). The two peaks at 10 eV and 4 eV in figure 2 correspond to these two dissociation channels, respectively.

The KERD for N^+–N^+ coincidence is extended up to 17 eV. N^+–N^+ ion pair with KER above 10 eV can be released from the D^1\Sigma^+_g state at the vertical ionization potential of 50.826 eV [31]. This state may be produced via the processes in which three or more photons are absorbed. The lowest N_2^+ dissociating into N^+ + N^+ is the a^1\Pi_u state with the vertical ionization potential of 45.195 eV. In point of excitation energy, N^+–N^+ ion pair thus can be produced by two-photon absorption of the 24 eV photons, whereas it cannot be produced by sequential two-photon absorption via X^2\Sigma^+_g, A^2\Pi_u and B^2\Sigma^+_g states. Contributions from C^2\Sigma^+_u and 2^2\Pi_g states should be small at the photon energy of 24 eV because these vertical ionization potentials are 25.514 and 24.788 eV, respectively, although the adiabatic ionization potential of these states are 23.583 and 23.755 eV, respectively. Thus, in the present study, the N^+–N^+ ion pairs are mostly produced via the processes in which three or more photons are absorbed.

The KERDs for N2+ in figure 2(b) are mainly located below 10 eV with peak position of about 4 eV. These relatively low energy N2+ are released from N2^+ dissociating to N2^+ + N [7]. It required three or more photons because the lowest dissociation limit for N2^+ + N is 53.89 eV [26, 27].

In conclusion, we have investigated sequential two-photon dissociative ionization of N2 and O2 molecules irradiated by EUV-FEL by means of ion momentum spectroscopy. Intermediate and final states produced by the first and second photon absorption are partially identified. To discuss in further detail, electron spectroscopy and/or electron–ion coincidence spectroscopy are indispensable. Such experiments are under considerations.

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