

Annual Research Highlights

(1) “Long-lived neutral H₂ in hydrogen migration within methanol dication”

The ejection of triatomic hydrogen molecular ions HD₂⁺ and D₃⁺ from partially deuterated methanol cation, CD₃OH²⁺, was investigated by first-principle molecular dynamics simulation. It was found from the numerical simulations that two C-D chemical bonds in the methyl group are broken first to form a neutral D₂ moiety within CD₃OH²⁺, and then, the neutral D₂ moiety vibrates and rotates for a relatively long period of time (20~330 fs) while moving towards a transition state leading to the ejection of HD₂⁺ or D₃⁺. The formation of such a long-lived neutral D₂ moiety within a hydrocarbon dication was commonly identified in all the trajectories yielding the formation of HD₂⁺ and D₃⁺. The formation of such a long-lived neutral D₂ moiety within a hydrocarbon dication explains well the recent experimental findings of the long lifetime of doubly charged energized hydrocarbon molecules prior to the ejection of H₃⁺.

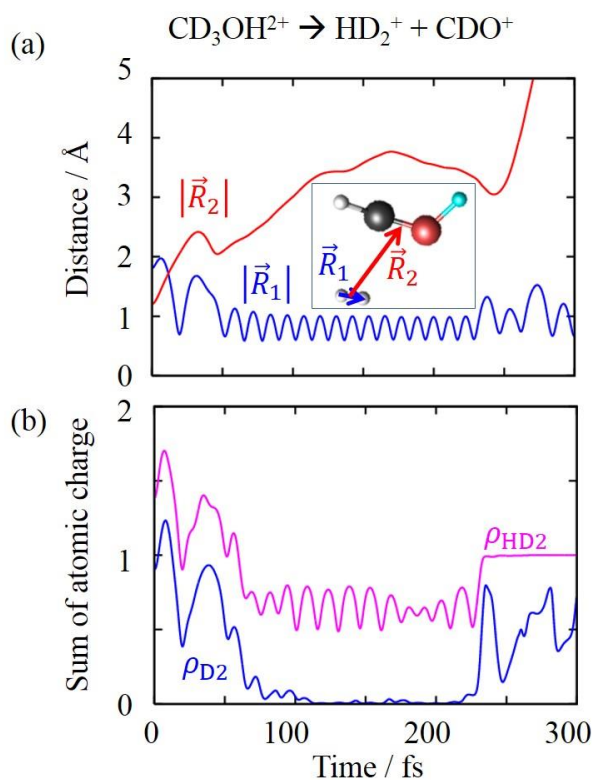


Fig. 1. (a) Time evolution of $|\vec{R}_1|$ and $|\vec{R}_2|$ for $CD_3OH^{2+} \rightarrow HD_2^+ + CDO^+$. (b) Time evolution of (i) the sum of the total charge ρ_{D_2} of two D atoms forming a neutral D₂ moiety and (ii) the total charge ρ_{HD_2} of two D atoms and one H atom forming HD₂⁺ as a final product.

1.(1)-1) *J. Chem. Phys.*, **139**, 181103 (2013)

(2) “Full-coherent free electron laser seeded by 13th and 15th-order harmonics of near-infrared femtosecond laser pulses”

In order to produce full-coherent and intense ultrashort light pulses in the extreme ultraviolet (EUV) wavelength region, the 13th-order harmonic and the 15th-order harmonic of Ti:Sapphire laser pulses at 800 nm were introduced into an undulator of the SCSS (Spring-8 Compact Self-Amplified Spontaneous Emission (SASE) Source) test accelerator at RIKEN with the accelerated electron bunch. The generated seeded SASE-free electron laser (FEL) light pulses were monitored with a single shot EUV spectrometer.

From numerical simulations of the amplification processes of high-order harmonic pulses in the undulator, optimum conditions of the electron bunch duration interacting with the high-order harmonic pulses were investigated for generating full-coherent and intense pulses in the EUV wavelength region.

The obtained spectral profiles of the FEL seeded with the 13th and 15th of harmonics are shown in Figs. 2(a) and 2(b), respectively. In contrast to the spike like spectrum of SASE-FEL (Fig. 2(a) middle), the single peak spectrum with the 30-fold amplification was recorded by the seeding with the 13th harmonic (Fig. 2(a) upper). Similarly, in contrast to the spike like spectrum of SASE-FEL (Fig. 2(b) middle), the single peak spectrum with the 80-fold amplification was recorded by the seeding with the 15th harmonic (Fig. 2(b) upper).

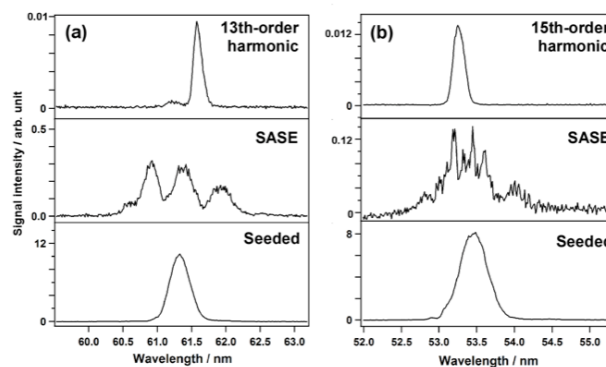


Fig. 2. (a) Typical examples of the spectral profiles of the 13th harmonic averaged over 30 shots (upper), the single-shot profile of SASE pulse at 61.7 nm (middle), and the single-shot profile of the FEL pulse seeded by the 13th-order harmonic (lower). (b) Typical examples of the spectral profiles of the 15th-order harmonic averaged over 30 shots (upper), the single-shot profile of SASE pulse at 53.5 nm (middle), and the single-shot profile of the FEL pulse seeded by the 15th-order harmonic (lower).

1.(1)-7) *J. Phys. B: At. Mol. Opt. Phys.*, **46**, 164006 (2013)

研究ハイライト

(1) 2価メタノールカチオン内長寿命中性水素分子

部分的に同位体置換されたメタノールの2価のカチオン CD_3OH^{2+} から3原子水素分子イオン HD_2^+ と D_3^+ が放出されるメカニズムを明らかにするために、第一原理分子動力学シミュレーションを用いて CD_3OH^{2+} の幾何学的構造と電荷分布の時間発展を計算した。メチル基の2つのC-D化学結合が解離した後、新たにD-D化学結合が生成し、中性の D_2 部分が生成した。 D_2 部分は、振動・回転をしながら HD_2^+ と D_3^+ が生成する遷移状態に向かって比較的長時間 (20-330 fs) をかけて移動することが示された。 D_2 部分の生成は、 HD_2^+ と D_3^+ が生成するトラジェクトリ全てにおいて共通に見られた。このような炭化水素分子の2価カチオン内での長寿命の中性 D_2 部分の生成は、 H_3^+ を生成する高い振動励起の炭化水素分子2価カチオンが長寿命であるという最近の実験結果を良く説明する。

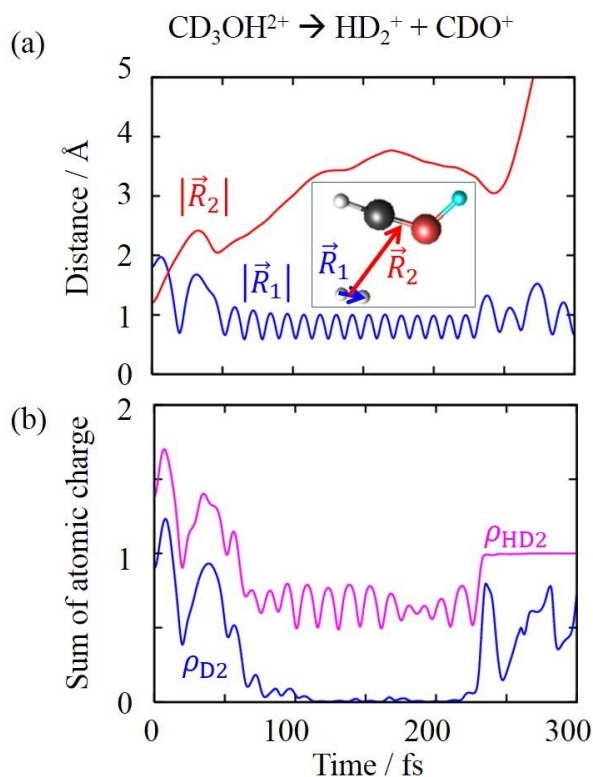


図1 (a) $|R_1|$ 、 $|R_2|$ で示す距離の時間変化, (b) 解離反応中に D_2 を構成する2つのD原子の電荷の総和 ρ_{D_2} と HD_2 になる3つの原子の電荷の総和 ρ_{HD_2} の時間変化

1.(1)-1) *J. Chem. Phys.*, **139**, 181103 (2013)

(2) レーザー高次高調波による極端紫外自由電子レーザーのシード化

フルコヒーレント高強度極端紫外光源の実現のため、レーザー高次高調波をシード光として SASE (Self Amplified Spontaneous Emission)-FEL (Free Electron Laser) である SCSS 試験加速器 (理化学研究所) のシード化実験を行った。Ti:Sapphire レーザーの第13次高調波および第15次高調波を SCSS 試験加速器の挿入光源に電子バンチと同時に入射し、発生した FEL 光を分光器によって観測した。

シード光と電子バンチの入射タイミングの揺らぎの影響と SASE 光成分の増幅を抑制するために、電子バンチ長を SASE 運転時の 300 フェムト秒から 600 フェムト秒に伸張した。

第13次高調波のシード光入射によって、スパイク状のスペクトル形状を持つ SASE 光に対して、シングルピークかつ約30倍の出力に増幅されたシード化 FEL 光が観測された(図2(a)上)。さらに第15次高調波を入射し、FEL の発振波長を 53.5 nm に調整したところ、同様にシングルピークを持ち、SASE-FEL 光に対して 80 倍に増幅されたシード化 FEL 光が観測された(図2(b)上)。この結果は、2010年に初めて第13次高調波によるシード化 FEL が達成された結果と比較して、シード化 FEL のシード化 FEL 光と SASE-FEL 光のコントラストが15倍向上し、さらなる短波長化が達成されたことを示している。

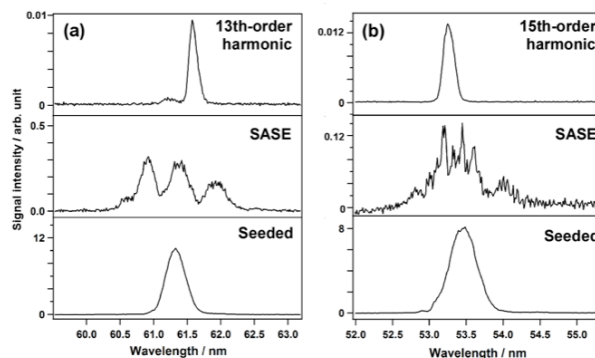


図2 (a)第13次高調波をシードした場合の典型的なスペクトル。レーザー高次高調波(上段)、同波長帯域の SASE-FEL(中段)、シード化 FEL(下段)。 (b)第15次高調波をシードした場合の典型的なスペクトル。レーザー高次高調波(上段)、同波長帯域の SASE-FEL(中段)、シード化 FEL(下段)

1.(1)-7) *J. Phys. B: At. Mol. Opt. Phys.*, **46**, 164006 (2013)

1. 原著論文

(1) Refereed Journals

- 1) K. Nakai, T. Kato, H. Kono, and K. Yamanouchi, "Communication: Long-lived neutral H₂ in hydrogen migration within methanol dication," *J. Chem. Phys.*, **139**, 181103 (2013).
- 2) E. Lötstedt, T. Kato, and K. Yamanouchi, "A classical model of H₃⁺ in an intense laser field," *J. Phys. B: At. Mol. Opt. Phys.*, **46**, 235601 (2013).
- 3) H. Zhang, C. Jing, J. Yao, G. Li, B. Zeng, W. Chu, J. Ni, H. Xie, H. Xu, S. L. Chin, K. Yamanouchi, Y. Cheng, and Z. Xu, "Rotational coherence encoded in an "air-laser" spectrum of nitrogen molecular ions in an intense laser field," *Phys. Rev.*, **X3**, 041009 (2013).
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- 5) S. Owada, A. Azarm, S. Hosseini, A. Iwasaki, S. L. Chin, and K. Yamanouchi, "Amplified spontaneous C³Π_u - B³Π_g emission and rotational and vibrational state distributions in C³Π_u state of N₂ in femtosecond laser induced filament in air," *Chem. Phys. Lett.*, **581**, 21-25 (2013).
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- 7) T. Sato, A. Iwasaki, S. Owada, K. Yamanouchi, E. J. Takahashi, K. Midorikawa, M. Aoyama, K. Yamakawa, T. Togashi, K. Fukami, T. Hatsui, T. Hara, T. Kameshima, H. Kitamura, N. Kumagai, S. Matsubara, M. Nagasono, H. Ohashi, T. Ohshima, Y. Otake, T. Shintake, K. Tamasaku, H. Tanaka, T. Tanaka, K. Togawa, H. Tomizawa, T. Watanabe, M. Yabashi, and T. Ishikawa, "Full-coherent free electron laser seeded by 13th and 15th-order harmonics of near-infrared femtosecond laser pulses," *J. Phys. B: At. Mol. Opt. Phys.*, **46**, 164006 (2013).
- 8) E. Lötstedt, T. Kato, and K. Yamanouchi, "Intramolecular electron dynamics in the ionization of acetylene by an intense laser pulse," *J. Chem. Phys.*, **138**, 104304 (2013).
- 9) R. Kanya, A. Iwasaki, T. Teramoto, and K. Yamanouchi, "Numerical simulations of time-domain interferometric soft X-ray microscope with broadband high-order harmonic light sources," *Appl. Phys. Lett.*, **102**, 071102 (2013).
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(2) その他

該当なし

2. 総説・解説

- 1) R. Kanya, Y. Morimoto, and K. Yamanouchi, "Laser-assisted electron scattering and diffraction in ultrashort intense laser fields," *Progress in Ultrafast Intense Laser Science X*, 1-16, Springer-Verlag (2014).
- 2) H. Xu, J. Yao, W. Chu, B. Zeng, J. Ni, G. Li, Y. Cheng, Z. Xu, D. Kartashov, S. Alisauskas, A. Pugzlys, A. Baltuska, K. Yamanouchi, and S. L. Chin, "Strong and coherent forward emissions from molecules driven by femtosecond infrared laser pulses," *Progress in Ultrafast Intense Laser Science X*, 95-109, Springer-Verlag (2014).
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3. 著書

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- 2) M. Chergui, A. Taylor, S. Cundiff, R. de Vivie-Riedle, and K. Yamanouchi, editors, “Proceedings of the XVIIIth International Conference on Ultrafast Phenomena,” The European Physical Journal, EPJ Web of Conferences 41, EDP Sciences, 2013.
- 3) K. Yamanouchi and K. Midorikawa, editors, “Progress in Ultrafast Intense Laser Science IX,” Springer-Verlag (Germany), 2013.

4. その他

該当なし