様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　　元 年　　６ 月 １１ 日現在

機関番号：１２６０８
研究種目：基盤研究(B)（特設分野研究）
研究期間：2015～2018
課題番号：１５ＫＴ００６０

研究課題名（和文）単分子反応遷移状態の精密時空間イメージング

研究課題名（英文）Precise spatiotemporal imaging of transition states in unimolecular reactions

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交付決定額（研究期間全体）：（直接経費）　　13,900,000円

研究成果の概要（和文）：化学反応は、遷移状態における原子の配置や運動によって決定付けられるため、遷移状態近傍における波動関数の形状を直接観測する意義は大きい。本研究課題は、振動や回転に対する波動関数・状態確率分布を実空間で可視化することを目指し、独自のコンセプトに基づいた２次元イオン画像観測装置を製作した。時間分解クーロン爆発イメージング法と組み合わせることにより、高い時間・空間分解能と高検出効率を両立させた観測を実現し、１方向にそろって回転する分子集団が示す量子論的挙動を初めて明らかにした。さらに、電子励起分子に対する回転固有状態の観測や、分子運動の実時間観測を活用した分子クラスターの分光学的研究へと展開した。

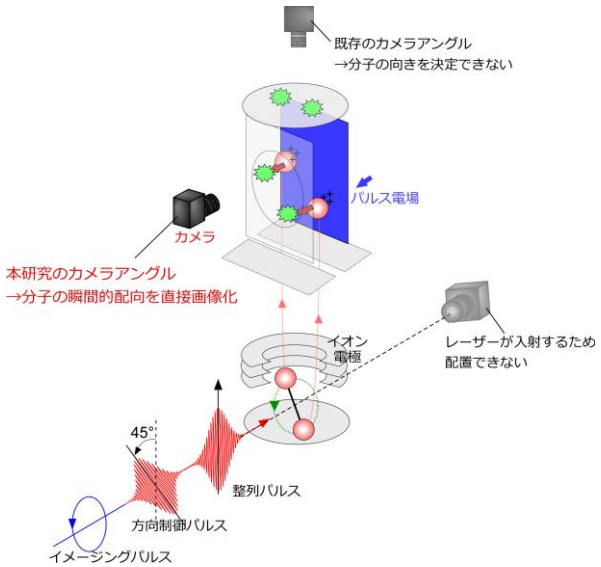
研究成果の学術的意義や社会的意義
本研究課題は、もっぱら理論計算の結果としてのみ表示されてきた波動関数や存在確率分布という存在を実際に測定された物理量として提示するものである。この結果は、科学に興味を持つ一般の方々に対してまでも躍動する分子の姿をvividに伝えるものであり、そのインパクトは物質科学全領域をカバーする。また、分子運動に関する波動関数の時空間発展を実験的に計測することにより、振動や回転のエネルギー準位を明らかにし、分子構造や相互作用ポテンシャルまでも特定する道を拓いたという点でも、大きな意義を有する。

研究成果の概要（英文）：The fates of chemical reactions are determined by arrangement and motion of atoms in the transition-state region, and experimental characterization of wave functions near the transition states is of significant importance. This research project has aimed to visualize wave functions or probability distributions pertinent to molecular vibration and rotation, and we have constructed a new two-dimensional ion imaging setup with novel concept for monitoring the arrangement of atoms in molecules. Its high spatial and temporal resolution as well as high data throughput has allowed us to unveil quantum-mechanical behavior of unidirectionally rotating molecular ensembles. Rotational eigenstates of electronically excited molecules have also been visualized and spectroscopic investigation on molecular clusters has been conducted via spatiotemporal tracking of rotational and intermolecular motion of the clusters.

研究分野：物理化学

キーワード：コヒーレント制御　遷移状態　量子波束　固有状態　イオンイメージング　分子クラスター

[1] H. Eyring, J. Chem. Phys. **3**, 107 (1935). [2] A. H. Zewail, J. Phys. Chem. A **104**, 5660 (2000). [3] D. M. Neumark, Acc. Chem. Res. **28**, 119 (1993). [4] M Stumpf, *et al.*, Faraday Discuss. **102**, 193 (1995). [5] A. Bach, *et al.*, J. Chem. Phys. **116**, 9739 (2002).



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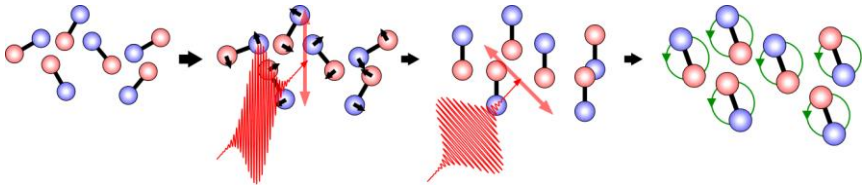
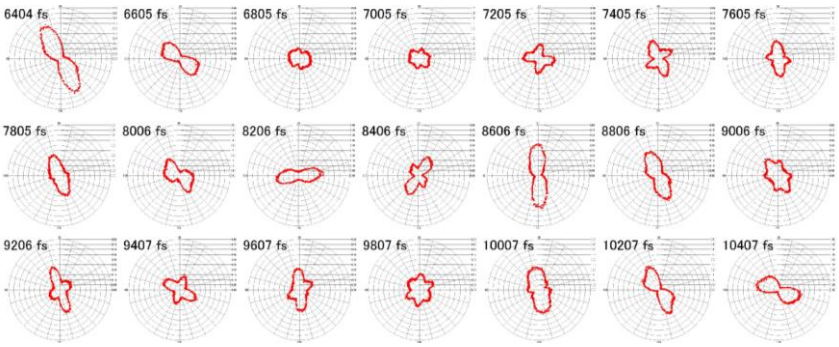
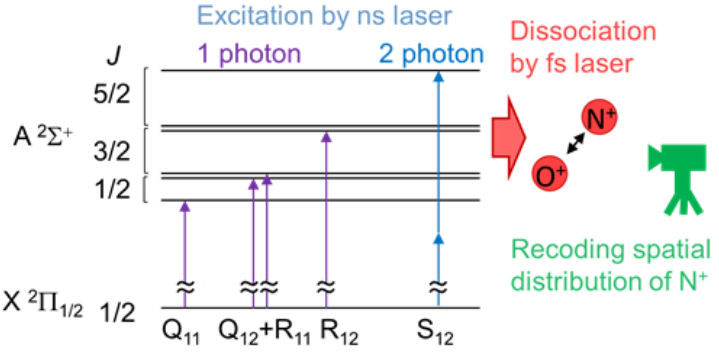




[6] K. Kitano, H. Hasegawa, and Y. Ohshima, Phys. Rev. Lett. **103**, 223003 (2009).



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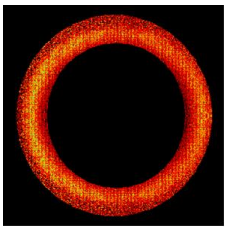

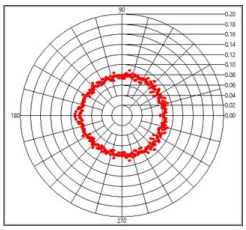

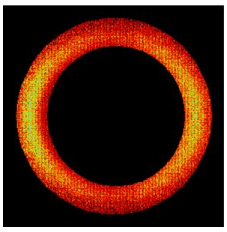

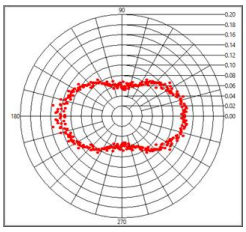

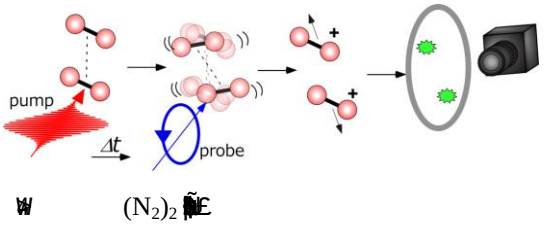
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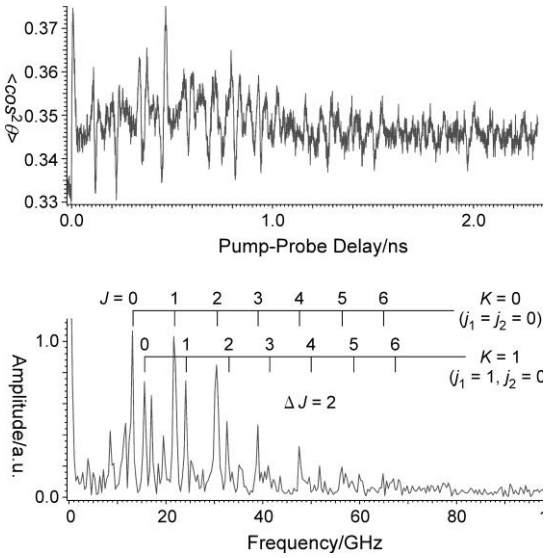

M. Hayashi and Y. Ohshima, "Sub-Doppler electronic spectrum of the benzene–D_2 complex," J. Chem. Phys. **150**, 014301-1–10 (2019). DOI: 10.1063/1.5077028

H. Akagi, T. Kumada, T. Otobe, R. Itakura, H. Hasegawa, and Y. Ohshima, Isotope-selective ionization utilizing field-free alignment of isotopologues with rapidly-turned-off laser field," Appl. Phys. B **24**, 14-1–8 (2018). DOI: 10.1007/s00340-017-6884-7

K. Mizuse, N. Chizuwa, D. Ikeda, T. Imajo, and Y. Ohshima, "Visualizing rotational wave functions of electronically excited nitric oxide molecules by using ion imaging technique," Phys. Chem. Chem. Phys. **20**, 3303–9 (2018). DOI: 10.1039/c7cp06347d

66, 54–57 (2017). https://www.bunkou.or.jp/prints/prints_1.html

66, 214–232 (2017). https://www.bunkou.or.jp/prints/prints_1.html

C. Harthcock, J. Zhang, W. Kong, M. Mitsui, and Y. Ohshima, "Electronic spectra and excited-state dynamics of acridine and its hydrated clusters," J. Chem. Phys. **146**, 134311-1–8 (2017). DOI: 10.1063/1.4979631

K. Mizuse, R. Fujimoto, N. Mizutani, and Y. Ohshima, "Direct imaging of laser-driven ultrafast molecular rotation," J. Vis. Exp. **120**, e54917 (2017). DOI: 10.3791/54917

S. Kunishige, T. Katori, M. Baba, M. Hayashi, H. Hasegawa, and Y. Ohshima, "Spectroscopic study on deuterated benzenes. II. High-resolution laser spectroscopy and rotational structure in the S_1 state," J. Chem. Phys. **143**, 244303-1–8 (2015). DOI: 10.1063/1.4937950

K. Mizuse, K. Kitano, H. Hasegawa, and Y. Ohshima, "Quantum unidirectional rotation directly imaged with molecules," Sci. Adv. **1**, e1400185-1–8 (2015). DOI:10.1126/sciadv.1400185

H. Akagi, T. Kasajima, T. Kumada, R. Itakura, A. Yokoyama, H. Hasegawa, and Y. Ohshima, "Isotope-selective ionization utilizing field-free alignment of isotopologues with a train of femtosecond laser pulse," Phys. Rev. A **91**, 063416-1–7 (2015). DOI:10.1103/PhysRevA.91.063416

Y. Ohshima, "Taking a Movie of Rotating Molecules," *Bimala Churn Law Memorial Lecture*, Indian Association for the Cultivation of Science, Kolkata, India, 2018.2.19. (Invited)

Y. Ohshima, "Spatiotemporal Imaging of Rotational/Vibrational Motion of Molecules and Clusters," *SDMC2018*, Sinclairs Retreat Dooars, Jalpaiguri, India, 2018.2.2.15-18. (Invited)

Y. Ohshima, “Creation and observation of coherently rotating/vibrating molecular ensembles,” *WRHI International Workshop on Advanced Laser Spectroscopy for Soft Molecular Systems*, Tokyo Institute of Technology, Yokohama, Japan, 2017.10.6. (Invited)

t Y. Ohshima, “Spatiotemporal imaging of rotational/vibrational motion of molecules and clusters,” *International Symposium on "Diversity of Chemical Reaction Dynamics"*, Jibasan Building, Himeji, Japan, 2017.7.14-15. (Invited)

u [illegible] 2017.3.27-28.

v [illegible] 2016.11.18-20.

w Y. Ohshima, “Creating and observing coherently rotating/vibrating molecular ensembles,” *Indo-Japan Discussion Meeting on ‘Frontiers in Molecular Spectroscopy: From Fundamentals to Applications on Material Science and Biology’*, Indian Institute of Technology, Kanpur, India, 2016.11.13-17. (Invited)

x Y. Ohshima, “High-resolution UV laser spectroscopic study of vdW clusters containing benzene,” *WRHI International Workshop on Advanced Laser Spectroscopy for Soft Molecular systems*, Tokyo Institute of Technology, Yokohama, Japan, 2016.9.26. (Invited)

y Y. Ohshima, “Creating and observing coherently rotating/vibrating molecular ensembles,” *24th International Conference on High Resolution Molecular Spectroscopy*, University of Chemistry and Technology, Prague, Czech, 2016.8.30-9.3. (Plenary)

z K. Mizuse and Y. Ohshima, “High-resolution spatiotemporal imaging of molecular rotational wave packets,” *Computational Chemistry (CC) Symposium-The main symposium of ICCMSE 2016-*, Metropolitan Hotel, Athens, Greece, 2016.3.17-20. (Invited)

{ Y. Ohshima, “Frequency- and time-domain investigation on intermolecular vibrations of molecular clusters,” *PACIFICHEM 2015*, Honolulu, Hawaii, USA, 2015.12.15–20. (Invited)

| [illegible] 2015.11.20.

} Y. Ohshima, “Coherent nonlinear optical manipulation of molecular vibration and rotation,” *5th Asian Spectroscopy Conference*, University of Sydney, Sydney, Australia, 2015.9.29-10.2. (Invited)

[illegible]

http://www.chemistry.titech.ac.jp/~ohshima/index.html
Ohshima Group, Tokyo Institute of Technology,
http://www.chemistry.titech.ac.jp/~ohshima/index_e.html

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	2015 º 7 v 17
The Japan Times	2015 º 7 v 18
	https://www.japantimes.co.jp/news/2015/07/18/national/science-health/surfing-waves-two-worlds/#.XP4Dp-S7qUk
EurekAlert!	http://www.eurekalert.org/pub_releases/2015-07/nion-ao070215.php
	http://www.eurekalert.org/pub_releases_ml/2015-07/aaft-_1070215.php
Phys.Org	http://phys.org/news/2015-07-movie-ultrafast-rotating-molecules-billion.html
ScienceDaily	http://www.sciencedaily.com/releases/2015/07/150703150209.htm
Nanotechnology Now	http://www.nanotech-now.com/news.cgi?story_id=51826
Media INAF	http://www.media.inaf.it/2015/07/03/quantum-movie/
YouTube	https://www.youtube.com/watch?v=NcUucMFUedE

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