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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

研究代表者

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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

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Schematic diagram of the experimental setup for molecular orientation. It shows a laser system with a pump laser (800 nm, 100 fs ~ 1 ps) and a probe laser (400 nm, 100 fs). The pump laser is used to create a directional pulse (方向制御パルス) and an imaging pulse (イメージングパルス). The probe laser is used to measure the molecular orientation. The setup includes a camera (カメラ) and a laser source (レーザー) that cannot be placed directly due to the ion electrode (イオン電極). The diagram also shows the molecular orientation (分子の瞬間的配向) and the resulting signal (分子の向きを決定できない).

Detailed description of the experimental setup and results. The diagram shows the laser system, the camera, and the ion electrode. The pump laser is used to create a directional pulse (方向制御パルス) and an imaging pulse (イメージングパルス). The probe laser is used to measure the molecular orientation. The setup includes a camera (カメラ) and a laser source (レーザー) that cannot be placed directly due to the ion electrode (イオン電極). The diagram also shows the molecular orientation (分子の瞬間的配向) and the resulting signal (分子の向きを決定できない).

既存のカメラアングル
 →分子の向きを決定できない

カメラ
 本研究のカメラアングル
 →分子の瞬間的配向を直接画像化

イオン電極
 レーザーが入射するため配置できない

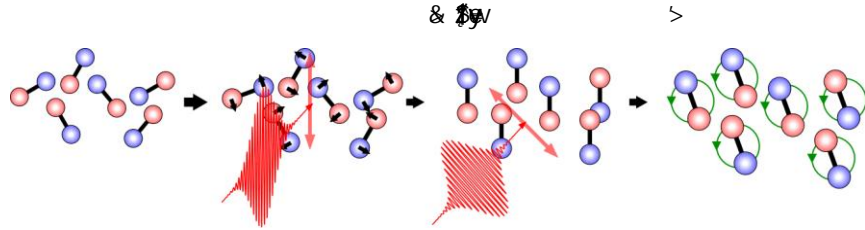
パルス電場
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pump
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 probe 400 nm, 100 fs

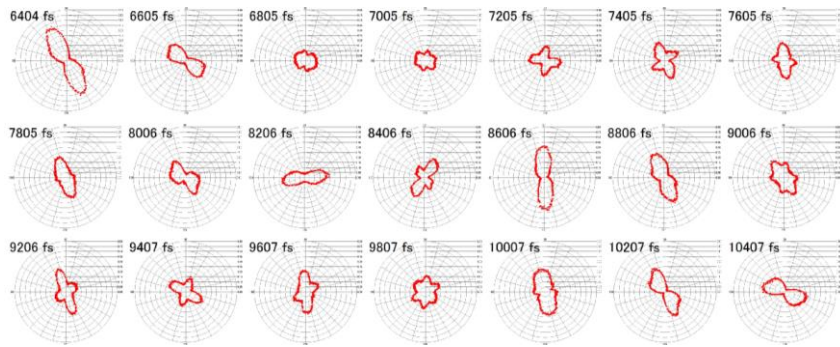
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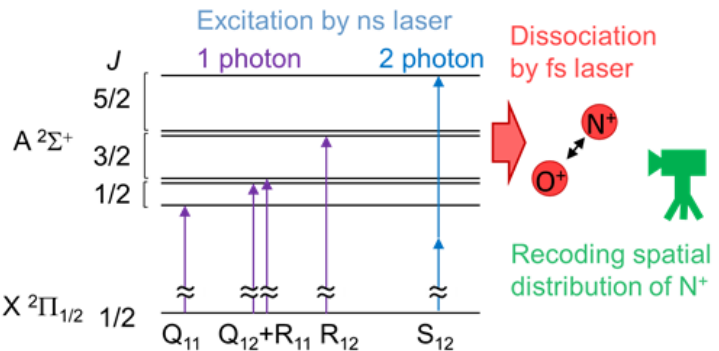
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 NO
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NO²⁺

S N⁺

□ □

□ □

M

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M

S □

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bs

ZcG3?

KS

8 GG[c

(

(J, |M|) = (1/2, 1/2)

~226 nm

• A-X 7

Q₁₁(1/2)

R₁₂(1/2)

G[NO b A

(1/2, 1/2) \ (3/2, 1/2)

biSXbG3? [wg

W

V

NO

3 > 0

J

V

(J, |M|) = (1/2,

6

IK

1/2), W

(3/2, 1/2)

W

IK

W

6

0

& 1 > 2x depolarization

(BMN)

x364(04.5b

G3? [wg

M]:

2x

depolarization

AS

2x

18

3

NO (G3? [wg

2x

depolarization PM

50

(564)

depolarization b

WZ0

& 2 > 3

z

>

G3? 5

N

¹⁴N₂

¹⁵N₂

8

7

KZ

0b

0

0Xb) [cGb

G[ASrSG3? b

W

& 3 > 4

θ

He p_s E

N₂⁺

(05/

c

9MG

' >

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(cos² θ)

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pump \ probe

4

G

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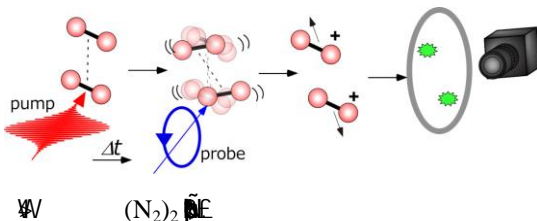
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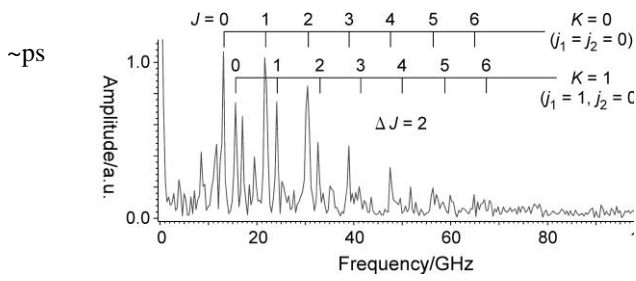
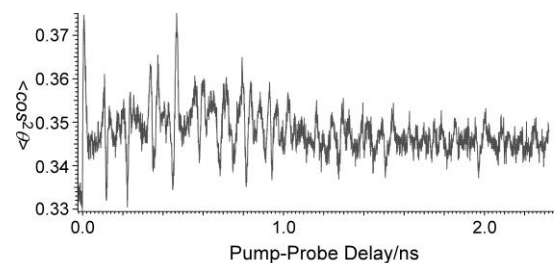


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$^{14}\text{N}_2\text{-PMO}\{Z\}'$
 $\text{S0}\{v/8453?g\}$
 $\text{AS}\{G3?X?\}$
 $\text{E}\{E(66270b)\}$
 $\text{vG}\{ASGb\}'$
 $5/_6M64_6M6uZb9_$
 $[6\sim G\{X\}\{E\}$
 $\text{EX8ZQb}\{M\}'$
 $[A\{X\}\{E\}$
 $\text{P}\{O\}\{Z\}$
 $\text{X}\{9\}\{B\}\{Q\}$
 $\text{G}\{b\}\{c\}\{E\}$
 $(6\{E\}\{3?\}\{P\}\{M\}\{v\}\{b\})'$
 $\text{S}\{P\}\{M\}\{k\}\{S\}$
 $\text{E}\{S\}\{S\}$
 $\text{b}\{E\}\{O\}\{I\}\{4\}\{E\}$
 $\text{X}\{E\}\{S\}\{S\}\{S\}\{9?\}$
 $0\{X\}\{b\}\{b\}\{N\}\{G\}\{E\}$
 $\text{E}\{M\}\{c\}\{E\}\{S\}$
 $\text{E}\{g\}\{K\}\{M\}\{v\}\{b\}\{6\sim 5\}\{E\}\{P\}\{S\}\{E\}$

$^{15}\text{N}_2$
 $E(\theta)$



$(\text{N}_2)_2$ b6EAO
 $\langle \cos^2 \theta \rangle (V) \setminus$
 $QbA (W)$

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