

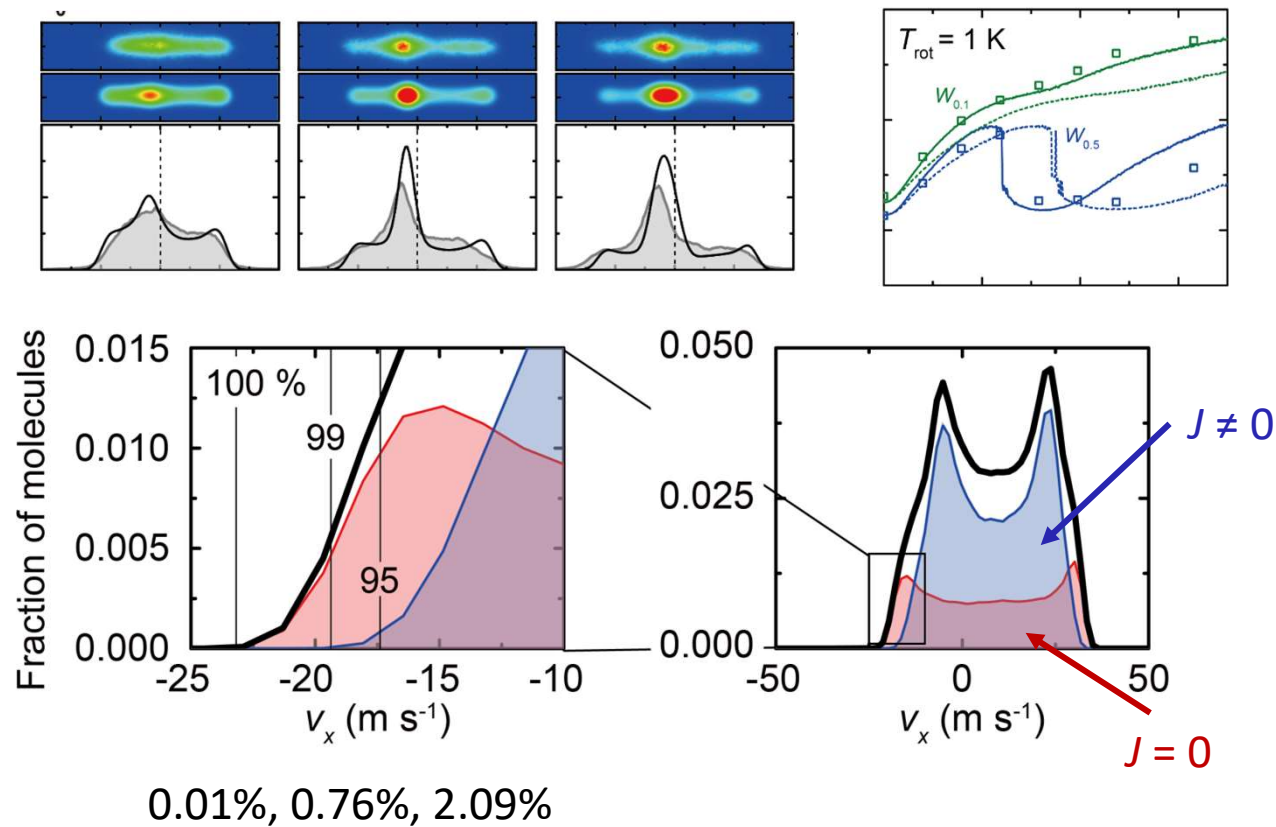
Optical dipole force on aligned molecules

Bum Suk Zhao

Ulsan National Institute of Science and Technology, Ulsan, Korea

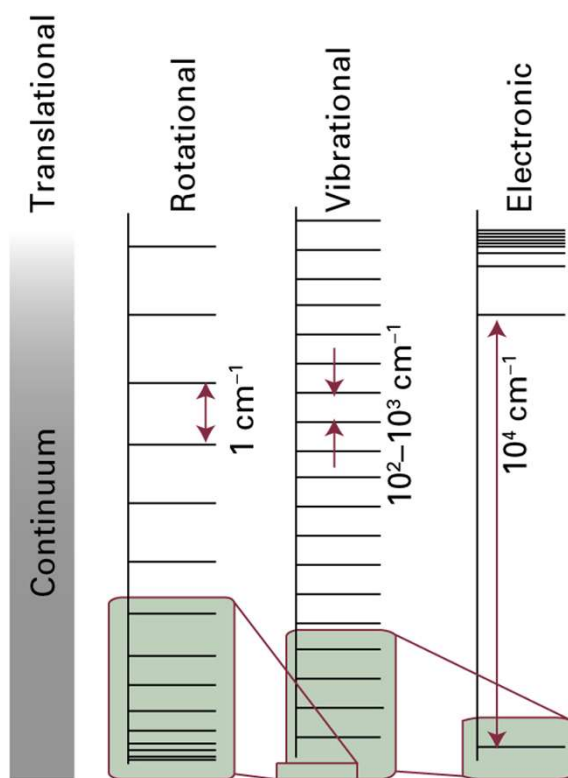
Take home messages

State-dependent molecular alignment is crucial to interpretate optical dipole force

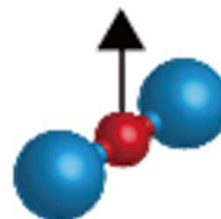


+ alpha

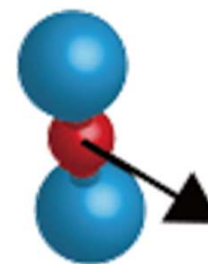
When we go from atoms to molecules



$|j, j\rangle$

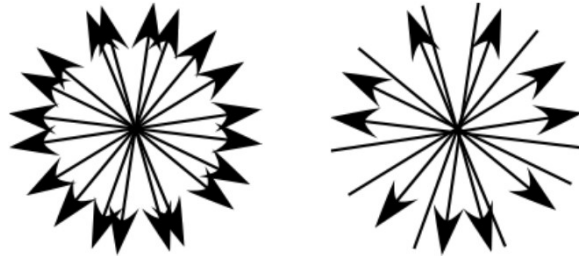


$|j, 0\rangle$



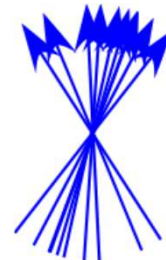
When we go from atoms to molecules

무작위
배향



입체 특이성 약화

정렬 혹은 배향
(alignment or
orientation)



입체 특이성 약화
감소

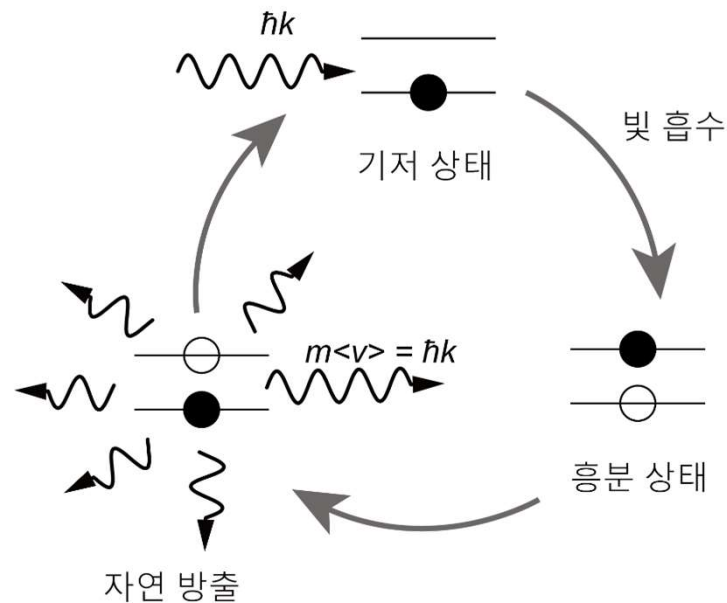
단일 양자
상태



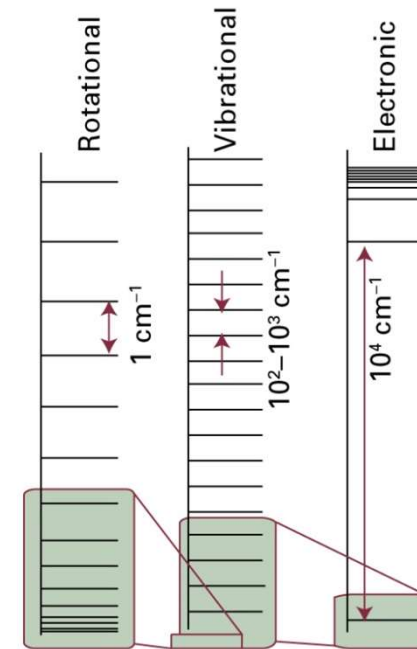
입체 특이성 보존

When we go from atoms to molecules

분자에 적용 불가능한 원자 제어 방법



- 빛의 흡수와 방출 반복
- 원자에 전달되는 **빛의 운동량으로 제어**

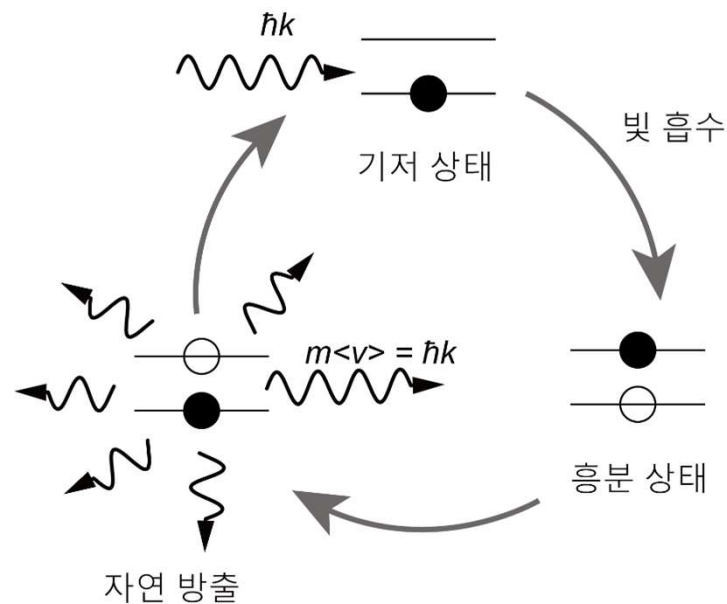


- 원자에 없는 양자 상태들이 존재
(진동과 회전 양자 상태)

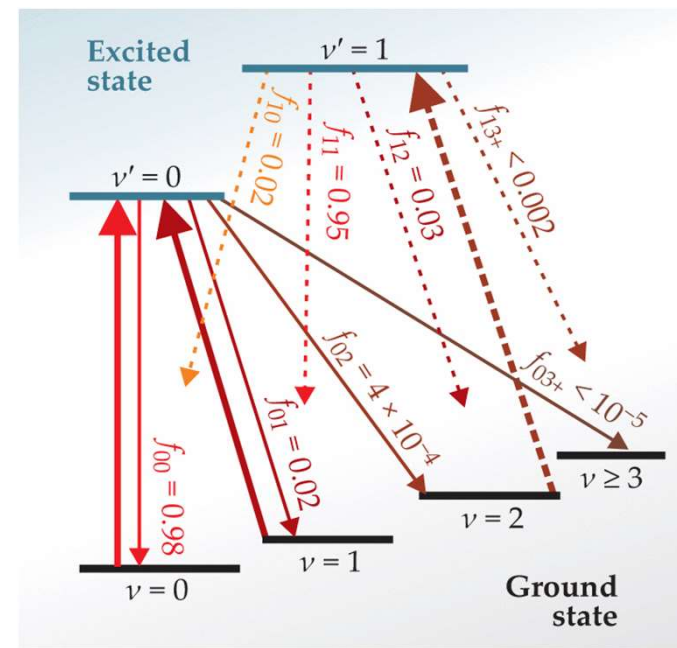
∴ 반복적인 빛의 흡수와 방출이 어려움

When we go from atoms to molecules

분자에 적용 가능한 방법들



- 빛의 흡수와 방출 반복
- 원자에 전달되는 **빛의 운동량으로 제어**

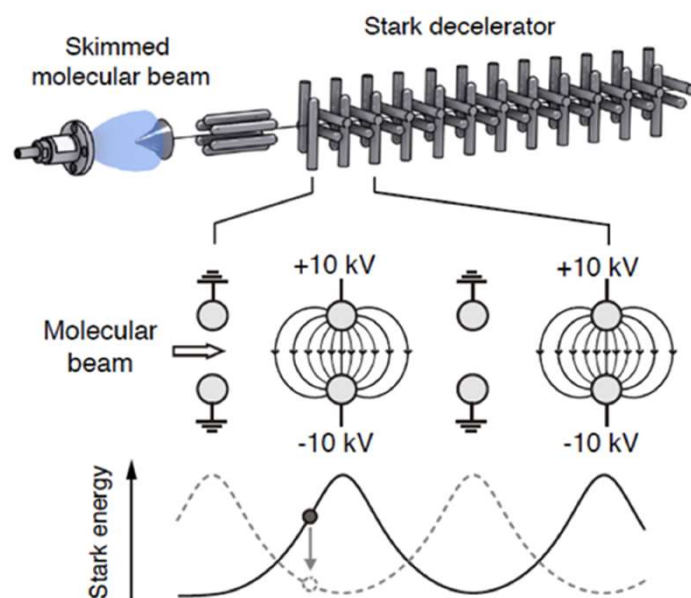


- repumping

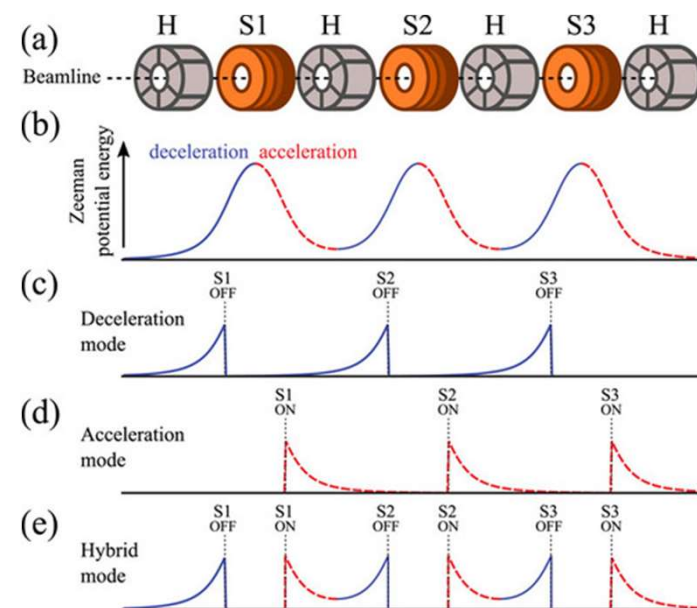
∴ 반복적인 빛의 흡수와 방출 가능

When we go from atoms to molecules

분자에 적용 가능한 방법들



Stark decelerator



Zeeman decelerator

When we go from atoms to molecules

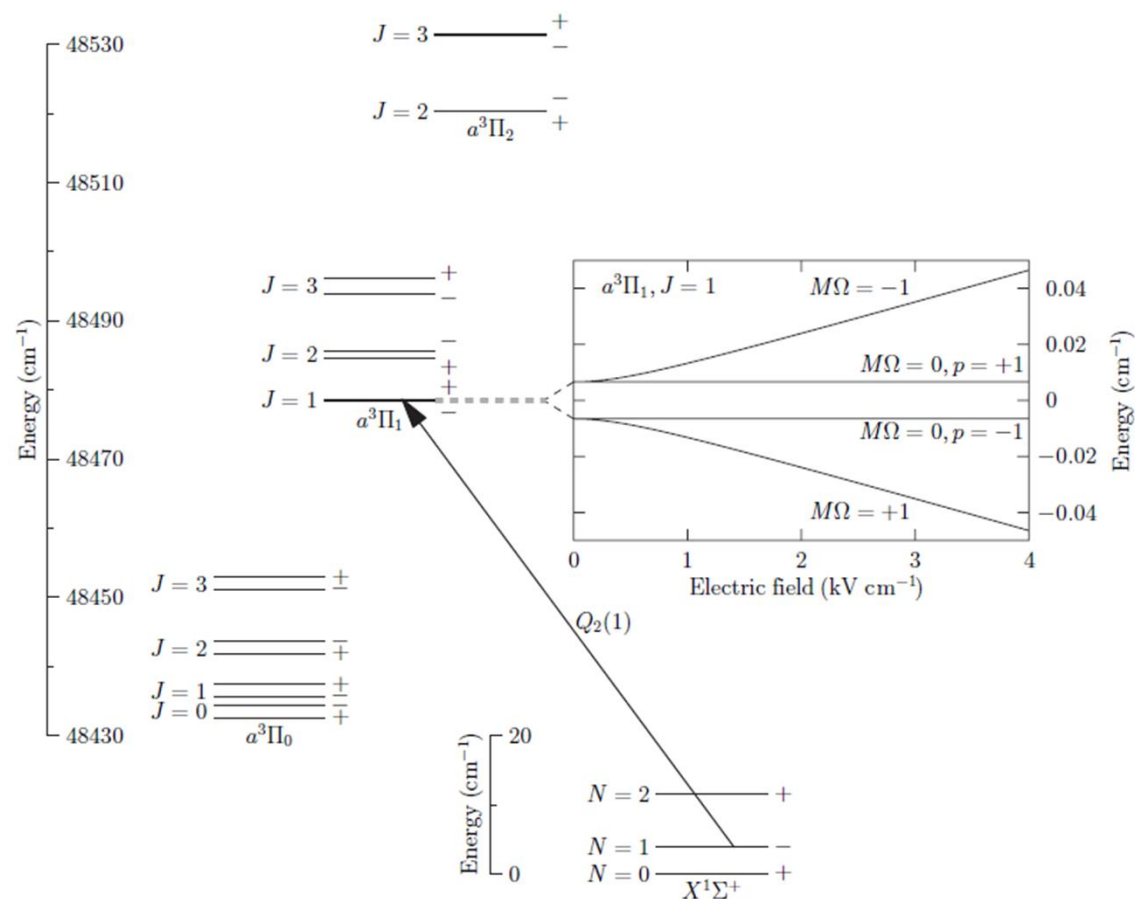
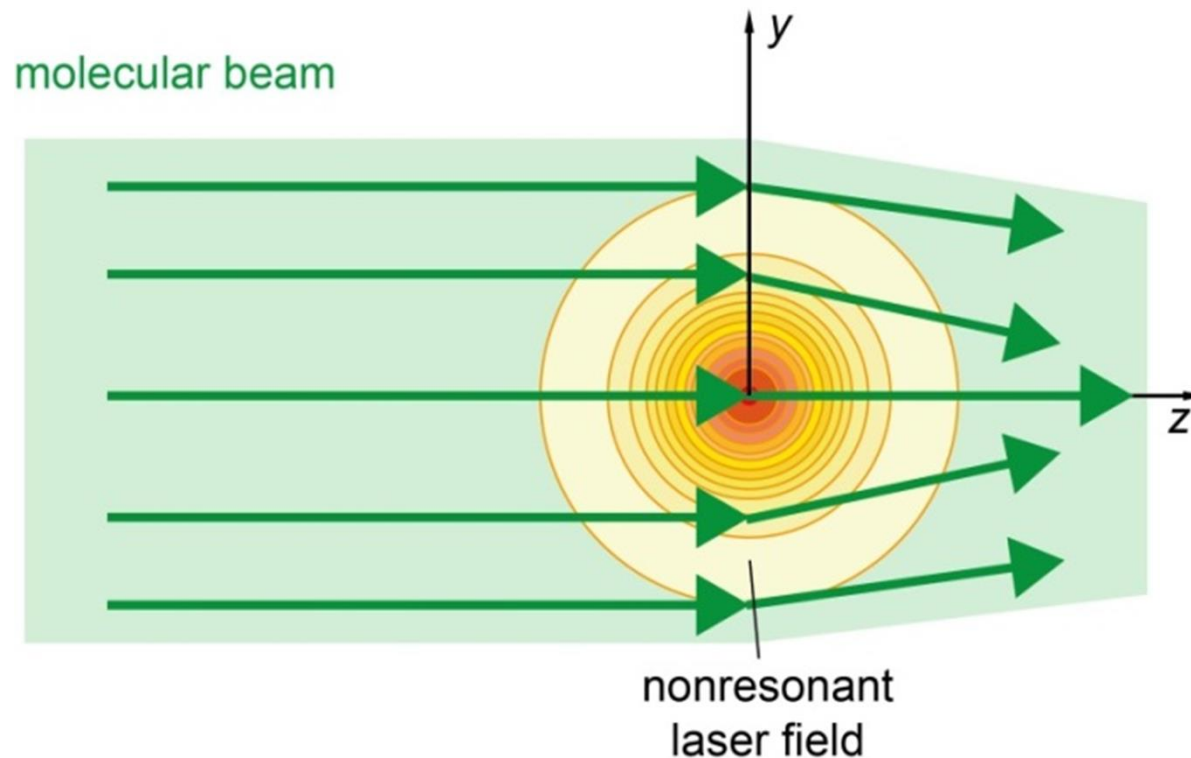


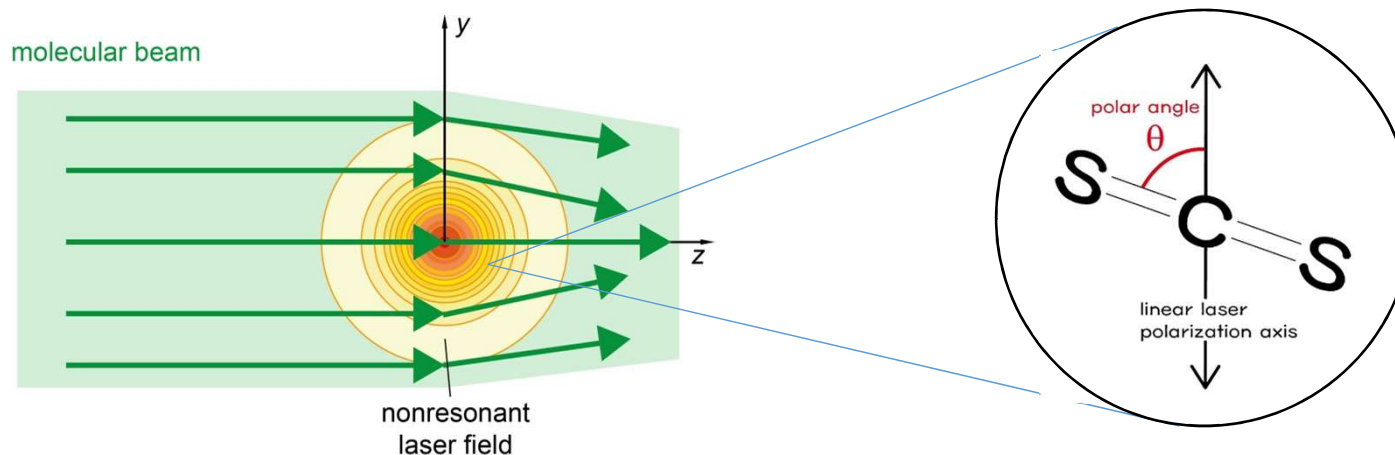
Figure 2. Rotational energy level structure of the $X^1\Sigma^+$, $v'' = 0$ ground state and the $a^3\Pi$, $v' = 0$ metastable state of CO. Molecules are prepared in the upper Λ -doublet component of the $a^3\Pi_1$, $v' = 0$, $J' = 1$ level by direct laser excitation on the $Q_2(1)$ transition around 206 nm. This spin-forbidden transition becomes weakly allowed via spin-orbit coupling of the $a^3\Pi_1$ states with higher-lying $A^1\Pi$ states. In the inset, the energy of the $a^3\Pi_1$, $v' = 0$, $J' = 1$ level is shown in electric fields up to 4 kV cm^{-1} .

When we go from atoms to molecules

분자에 적용 가능한 방법들



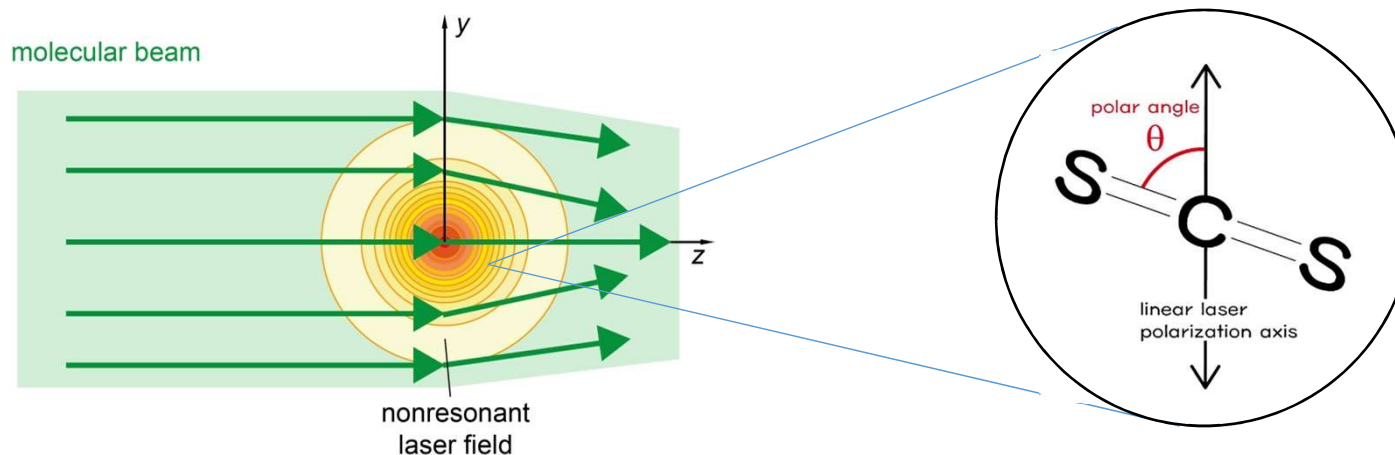
Molecules in the laser field



- The interaction potential between ground state CS₂ molecules and a laser field of intensity I

$$U = -\frac{1}{2} Z_0 I [(\alpha_{\parallel} - \alpha_{\perp}) \cos^2 \theta + \alpha_{\perp}] = -\frac{1}{2} Z_0 I \alpha_{\text{eff}}$$

Molecules in the laser field



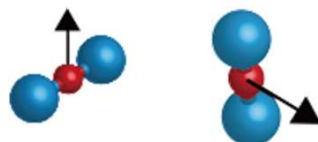
- The interaction potential between ground state CS_2 molecules and a laser field of intensity I

$$U = -\frac{1}{2} Z_0 I [(\alpha_{\parallel} - \alpha_{\perp}) \cos^2 \theta + \alpha_{\perp}] = -\frac{1}{2} Z_0 I \alpha_{\text{eff}}$$

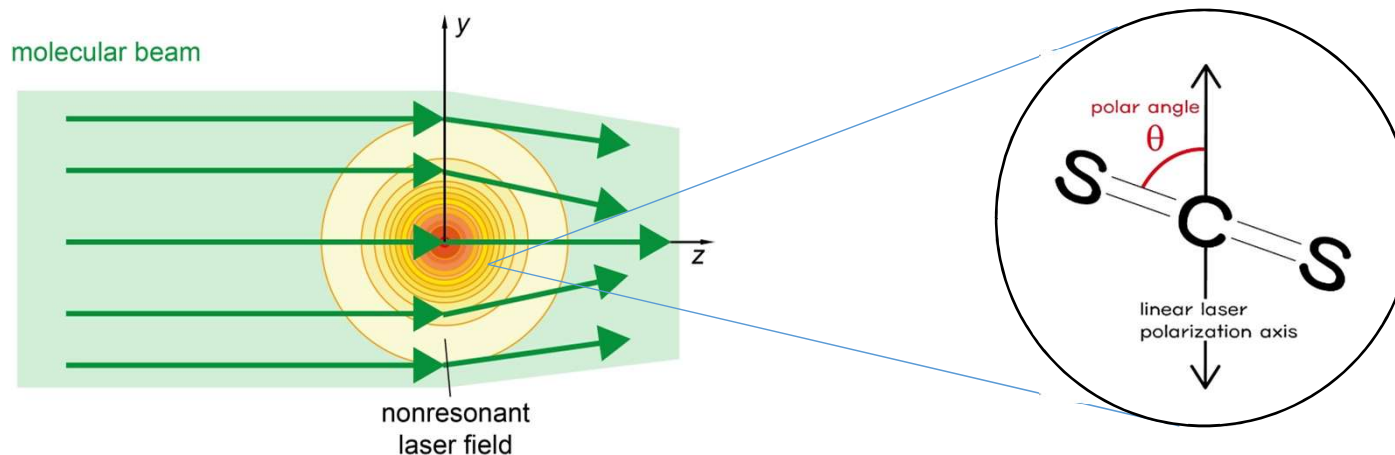
- $l = 0 \rightarrow$ spherical harmonics

$$\mathbf{H}_0 = B\mathbf{J}^2$$

$$Y_{j,M} = |j, M\rangle$$



Molecules in the laser field



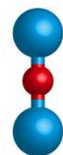
- The interaction potential between ground state CS_2 molecules and a laser field of intensity I

$$U = -\frac{1}{2} Z_0 I [(\alpha_{\parallel} - \alpha_{\perp}) \cos^2 \theta + \alpha_{\perp}] = -\frac{1}{2} Z_0 I \alpha_{\text{eff}}$$

- $l = 0 \rightarrow$ spherical harmonics
- $l > 0 \rightarrow$ pendular states

$$\mathbf{H}_0 = B\mathbf{J}^2$$

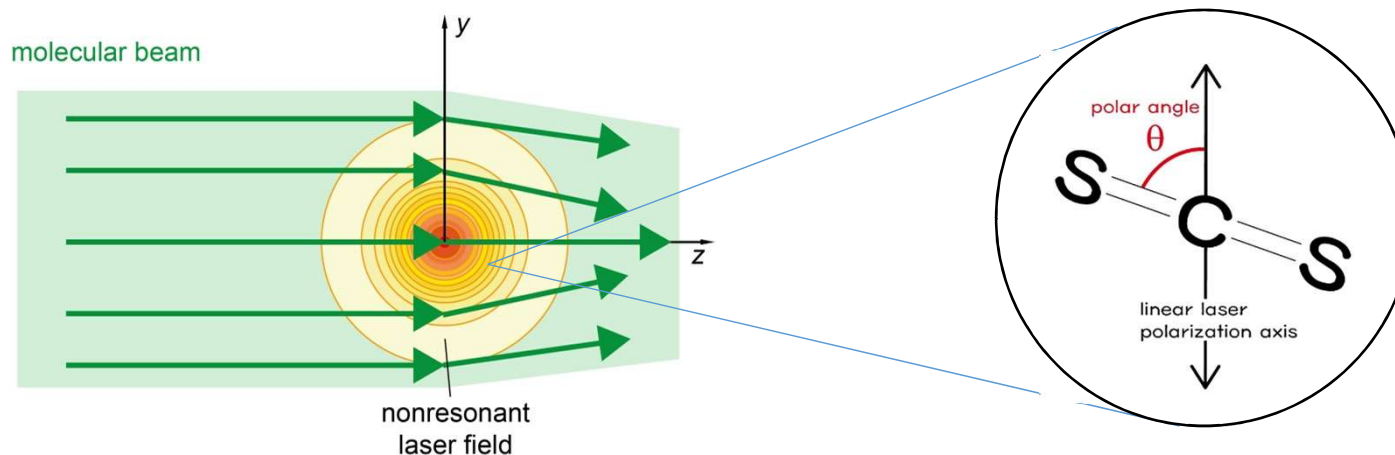
$$Y_{j,M} = |j, M\rangle$$



$$\mathbf{H} = B\mathbf{J}^2 + U$$

$$\Psi_{J,M}(I) = \sum_j C_j^{J,M}(I) |jM\rangle$$

Molecules in the laser field



- The interaction potential between ground state CS_2 molecules and a laser field of intensity I

$$U = -\frac{1}{2} Z_0 I [(\alpha_{\parallel} - \alpha_{\perp}) \cos^2 \theta + \alpha_{\perp}] = -\frac{1}{2} Z_0 I \alpha_{\text{eff}}$$

- $l = 0 \rightarrow$ spherical harmonics
- $l > 0 \rightarrow$ pendular states

$$\mathbf{H}_0 = B\mathbf{J}^2$$

$$\mathbf{H} = B\mathbf{J}^2 + U$$

$$Y_{j,M} = |j, M\rangle$$

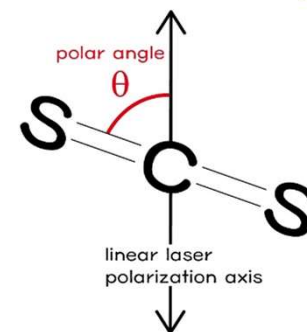
$$\Psi_{J,M}(0) = |jM\rangle$$

$$\Psi_{J,M}(I) = \sum_j C_j^{J,M}(I) |jM\rangle$$

Molecular alignment

- Expectation value of $\cos^2 \theta \rightarrow$ degree of alignment

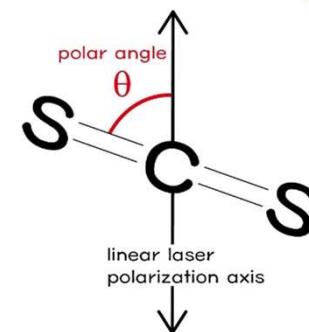
$$\langle \cos^2 \theta \rangle_{J,M} = \langle \cos^2 \theta \rangle_{J,M} (I) = \langle \Psi_{J,M}(I) | \cos^2 \theta | \Psi_{J,M}(I) \rangle$$



Molecular alignment

- Expectation value of $\cos^2 \theta \rightarrow$ degree of alignment

$$\langle \cos^2 \theta \rangle_{J,M} = \langle \cos^2 \theta \rangle_{J,M}(I) = \langle \Psi_{J,M}(I) | \cos^2 \theta | \Psi_{J,M}(I) \rangle$$



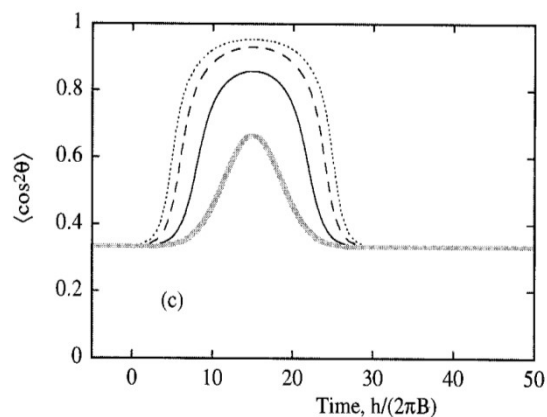
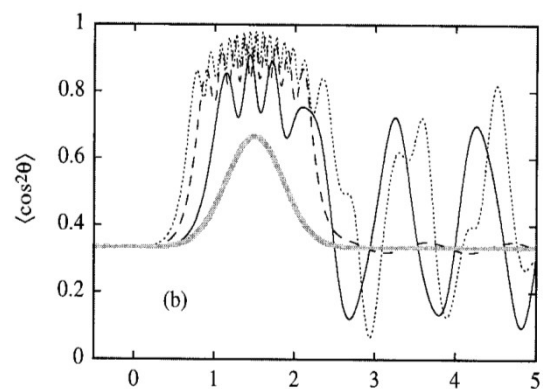
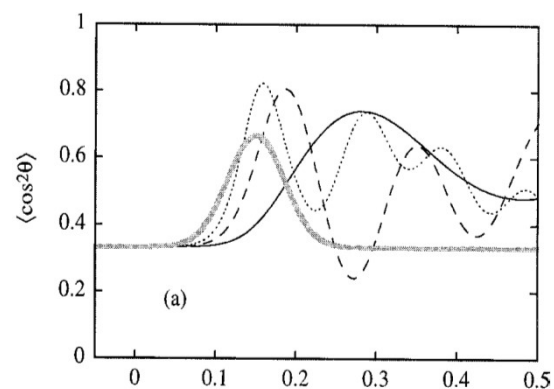
- Approximated interaction potential at the adiabatic regime

$$U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M} [I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0 \quad \alpha_{J,M}(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M} + \alpha_{\perp}$$

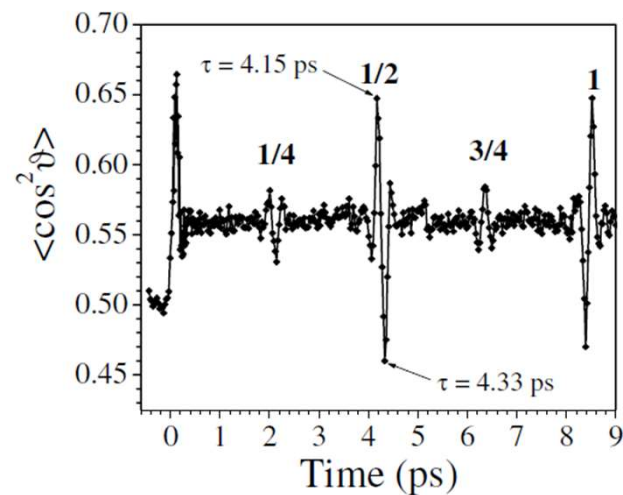


$$U = -\frac{1}{2} Z_0 I [(\alpha_{\parallel} - \alpha_{\perp}) \cos^2 \theta + \alpha_{\perp}] = -\frac{1}{2} Z_0 I \alpha_{\text{eff}}$$

Adiabatic and nonadiabatic alignment



TDSE



TISE

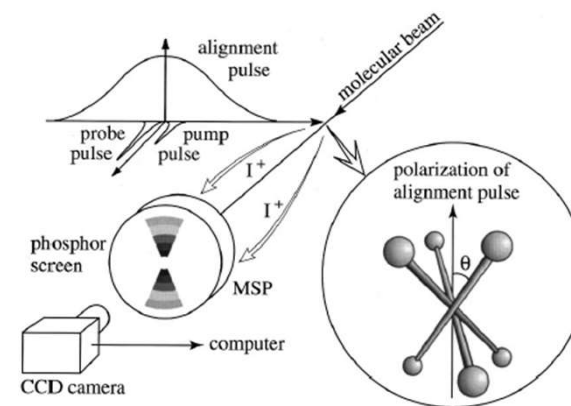


FIG. 1. A schematic of the experimental setup. The polarization of the lasers are indicated by the arrows (upper left-hand side). The inset shows the aligned molecules making an angle θ with the polarization axis of the alignment laser.

Molecular alignment and optical dipole force

- Expectation value of $\cos^2 \theta \rightarrow$ degree of alignment

$$\langle \cos^2 \theta \rangle_{J,M} = \langle \cos^2 \theta \rangle_{J,M}(I) = \langle \Psi_{J,M}(I) | \cos^2 \theta | \Psi_{J,M}(I) \rangle$$

- Approximated interaction potential at the adiabatic regime

$$U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M} [I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0 \quad \alpha_{J,M}(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M} + \alpha_{\perp}$$

- Optical dipole force

$$\mathbf{F}_{J,M}(\mathbf{r}, t) = \frac{1}{2} Z_0 \nabla \{ \alpha_{J,M}(I) [I(\mathbf{r}, t)] I(\mathbf{r}, t) \}$$

$$\Delta v_x = \int \frac{1}{m} F_{J,M}^x(x, y, z, t) dt$$

Molecular alignment and optical dipole force

- Expectation value of $\cos^2 \theta \rightarrow$ degree of alignment

$$\langle \cos^2 \theta \rangle_{J,M} = \langle \cos^2 \theta \rangle_{J,M}(I) = \langle \Psi_{J,M}(I) | \cos^2 \theta | \Psi_{J,M}(I) \rangle$$

- Approximated interaction potential at the adiabatic regime

$$U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M} [I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0 \quad \alpha_{J,M}(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M} + \alpha_{\perp}$$

- Optical dipole force

$$\mathbf{F}_{J,M}(\mathbf{r}, t) = \frac{1}{2} Z_0 \nabla \{ \alpha_{J,M}(I) [I(\mathbf{r}, t)] I(\mathbf{r}, t) \}$$

$$\Delta v_x = \int \frac{1}{m} F_{J,M}^x(x, y, z, t) dt$$

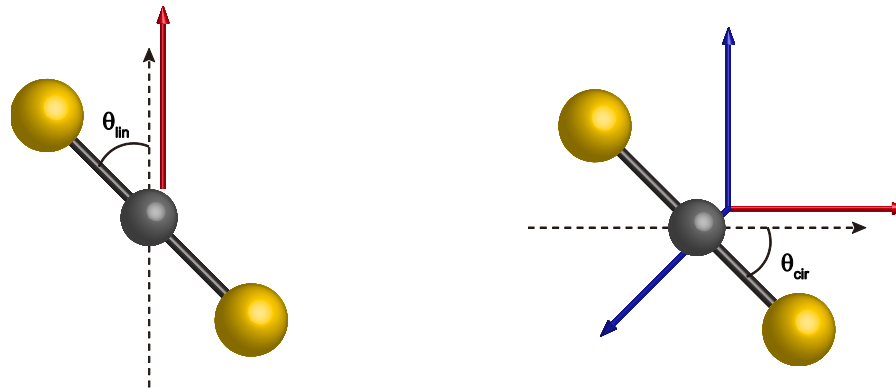
Molecular alignment and optical dipole force

$$\alpha_{J,M}^U(I) = \Delta\alpha \langle \cos^2 \theta_{\text{lin}} \rangle_{J,M} + \alpha_{\perp} \quad (4a)$$

and [Eq. (4b)]:

$$\alpha_{J,M}^U(I) = \frac{1}{2} (\alpha_{\parallel} + \alpha_{\perp} - \Delta\alpha \langle \cos^2 \theta_{\text{cir}} \rangle_{J,M}) \quad (4b)$$

for the linear and circular polarizations, which are the polarizability components along the y axis and in the y-z plane, respectively.^[23] Then, the resulting dipole force ($-\nabla U_{J,M}(y,z,t)$) is [Eq. (5)]:



First experimental study

Deflection of Neutral Molecules using the Nonresonant Dipole Force

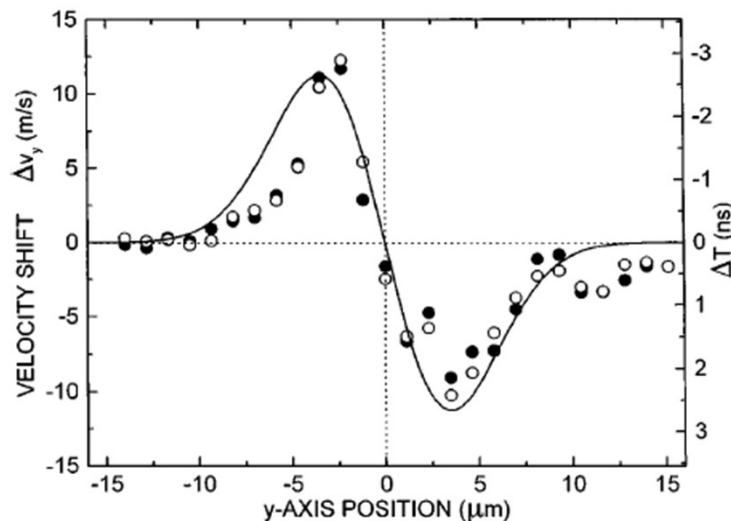
H. Stapelfeldt,^{1,*} Hirofumi Sakai,^{1,2} E. Constant,^{1,3} and P. B. Corkum¹

¹*Steacie Institute for Molecular Sciences, National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6*

²*Electrotechnical Laboratory, 1-1-4, Umezono, Tsukuba, Ibaraki 305, Japan*

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(Received 3 April 1997)



always larger than the averaged α . Figure 3 compares the results obtained with linearly (solid circles) and circularly (open circles) polarized light. The lack of alignment may be due to our multilongitudinal mode YAG laser. Mode beating can produce transient spikes as short as 30 ps which is comparable to or even shorter than the rotational period τ_{rot} of the CS_2 molecules ($\tau_{\text{rot}} \sim 34$ ps for a rotational quantum number $J = 4$). Under these

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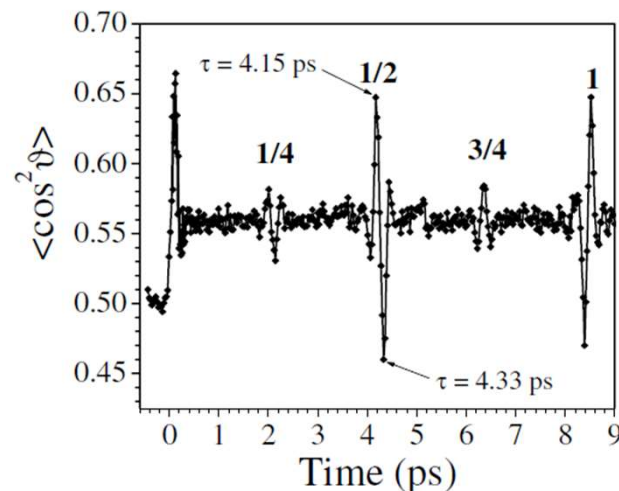
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Single longitudinal mode

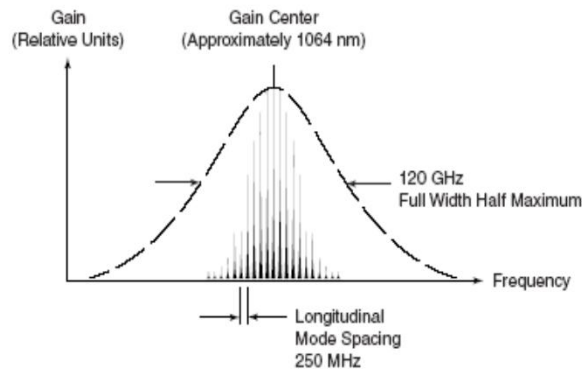


Figure 3-2: Longitudinal Modes of the Host Laser

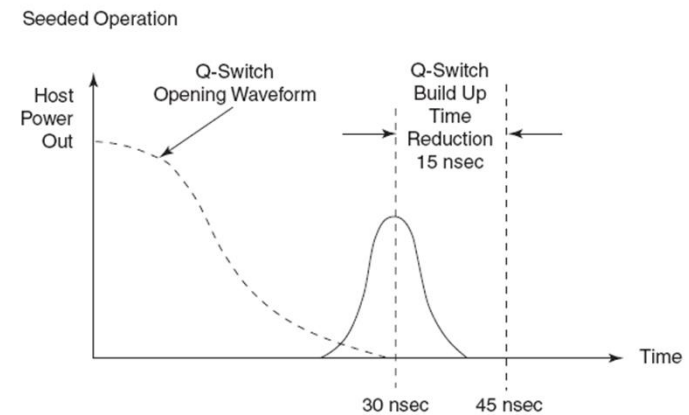
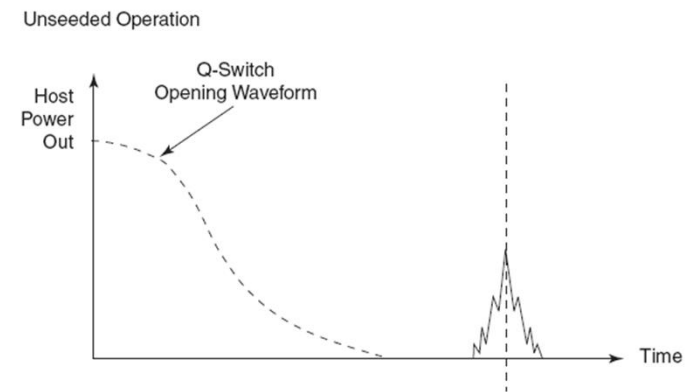
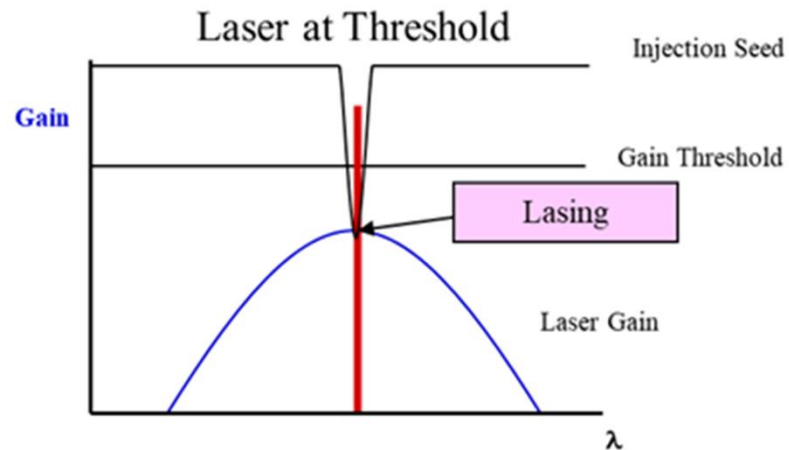
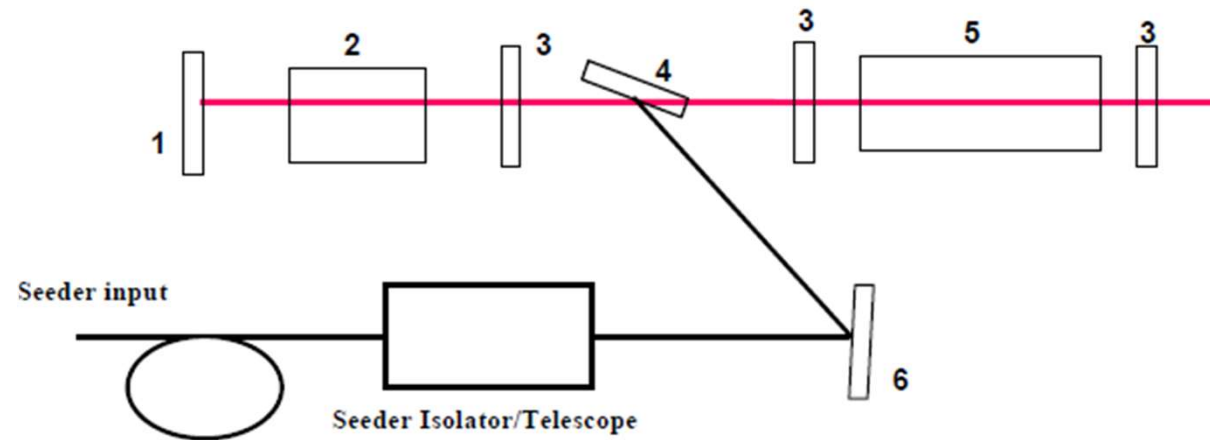


Figure 3-3: Q-Switch Buildup Time Reduction

Single longitudinal mode

- When the seed laser emission frequency is within the bandwidth of a pulsed laser cavity mode, a Q-switched pulse will develop more rapidly at this frequency than at a frequency created from background spontaneous noise emission.
- Consequently, the pulse developing from the seed signal will saturate the gain medium and extract the energy at the seed laser frequency before any pulse can develop from noise emission.
- This essentially inhibits the amplification and growth of any pulse that might develop from noise emission.

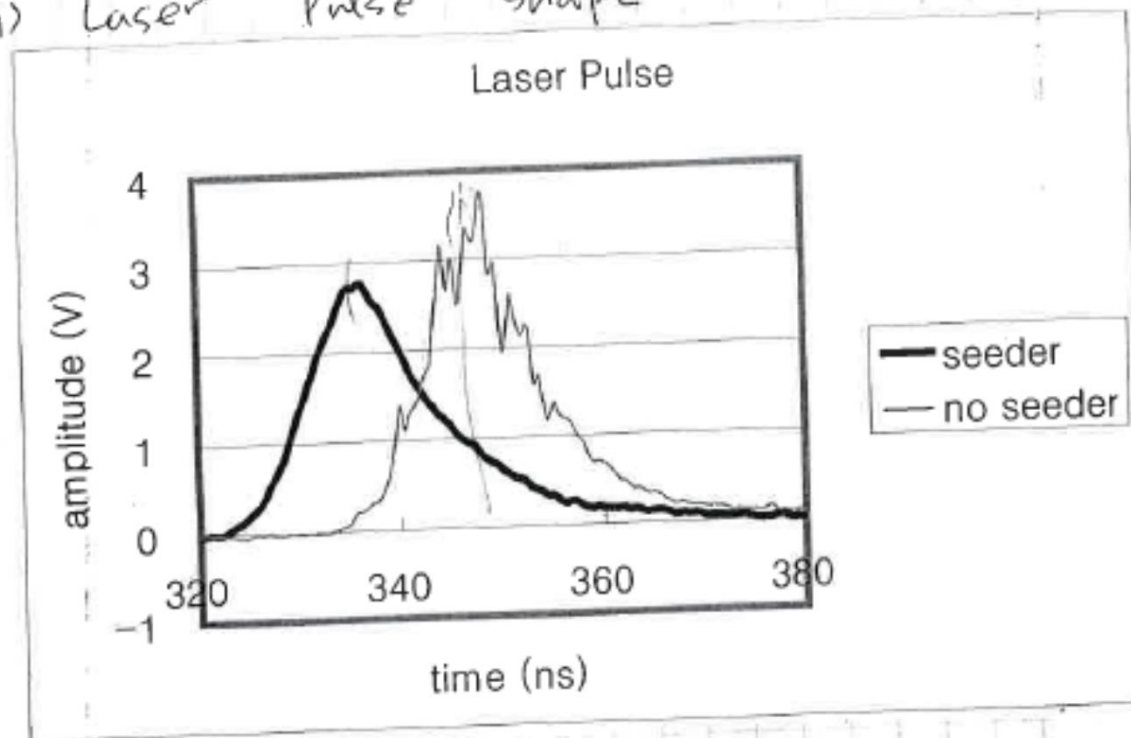
Our efforts (2003. 5. 16)



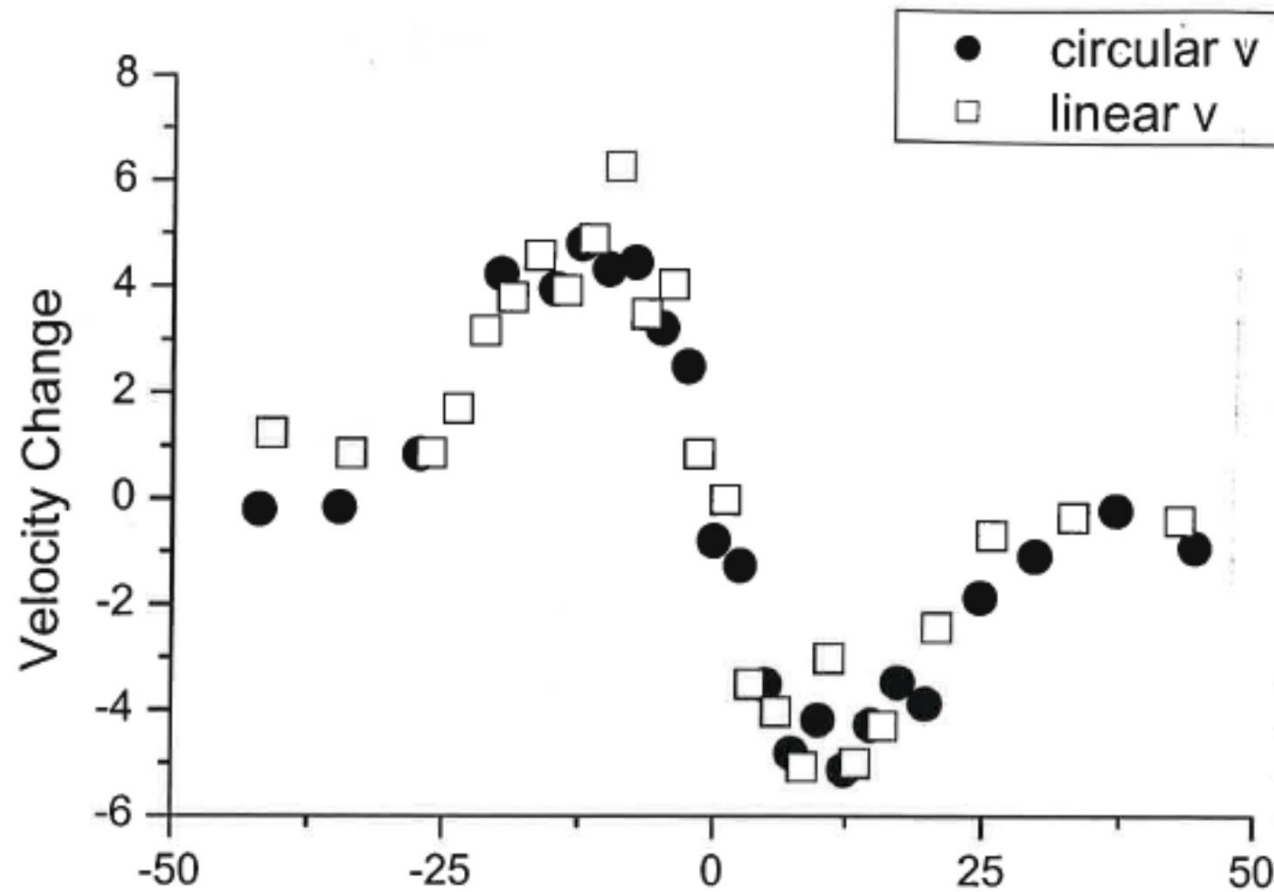
5/16 Korea-Laser.
"Seeder 전략!"

Our efforts (2003. 10. 28)

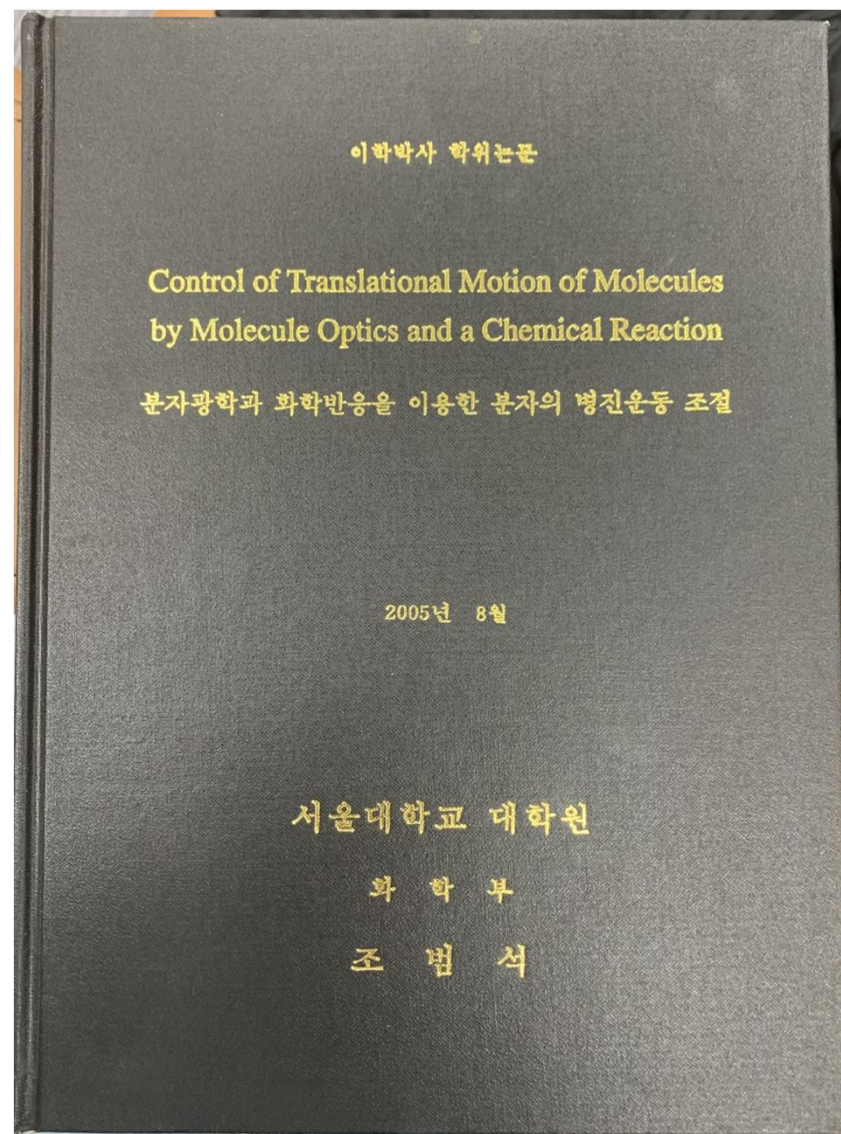
i) Laser Pulse shape



Our efforts (2003. 11. 6)



Our efforts



Rotational temperature matters

Role of rotational temperature in adiabatic molecular alignment

Vinod Kumarappan

Department of Chemistry, University of Aarhus, DK 8000 Aarhus C., Denmark

Christer Z. Bisgaard^{a)} and Simon S. Viftrup

Department of Physics and Astronomy, University of Aarhus, DK 8000 Aarhus C., Denmark

Lotte Holmegaard and Henrik Stapelfeldt^{b)}

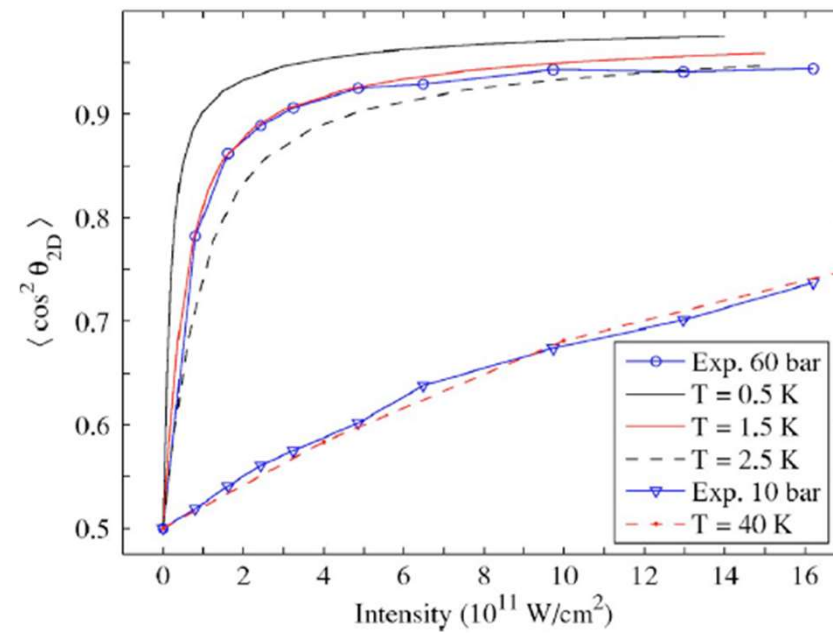
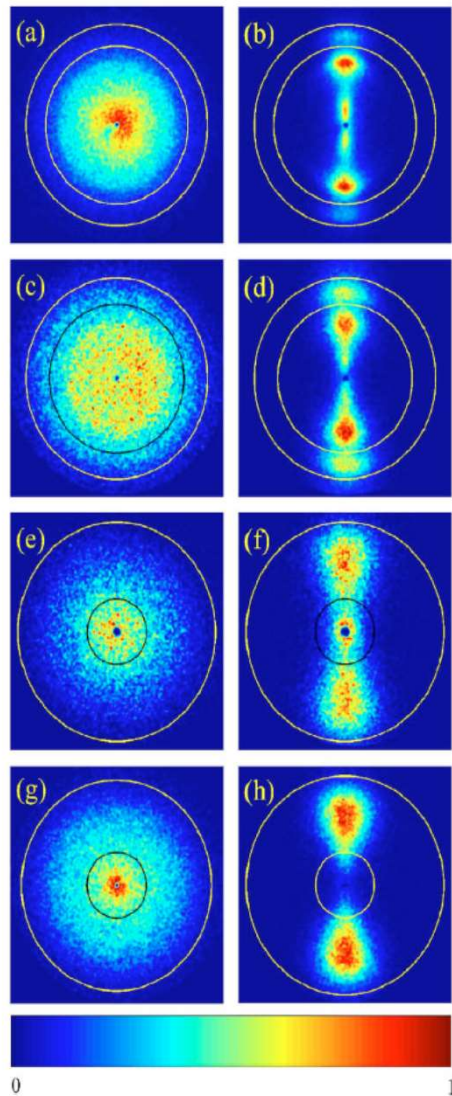
Department of Chemistry, University of Aarhus, DK 8000 Aarhus C., Denmark

(Received 17 August 2006; accepted 13 October 2006; published online 16 November 2006)

One-dimensional alignment of molecules in the adiabatic limit, where the pulse duration greatly exceeds the molecular rotational periods, is studied experimentally. Four different asymmetric top molecules (iodobenzene, *p*-diiodobenzene, 3,4-dibromothiophene, and 4,4'-dibromobiphenyl), rotationally cooled through a high pressure supersonic pulsed valve, are aligned by a 9-ns-long pulse. Their orientations are measured through Coulomb explosion, induced by a 130-fs-long pulse, and by recording the direction of the recoiling ions. The paper focuses on the crucial role of the initial rotational temperature for the degree of alignment. In particular, we show that at molecular temperatures in the 1 K range very strong alignment is obtained already at intensities of a few times 10^{11} W/cm² for all four molecules. At the highest intensities ($\sim 10^{12}$ W/cm²) the molecules can tolerate without ionizing $\langle \cos^2 \theta \rangle \geq 0.92$ in the case of iodobenzene. This is the strongest degree of alignment ever reported for any molecule. © 2006 American Institute of Physics.

[DOI: [10.1063/1.2388273](https://doi.org/10.1063/1.2388273)]

Rotational temperature matters



The first potential project as a PI

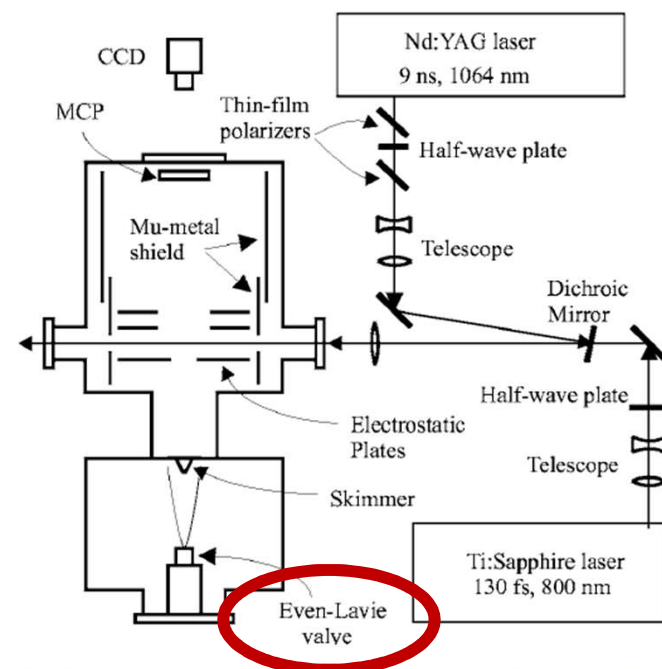
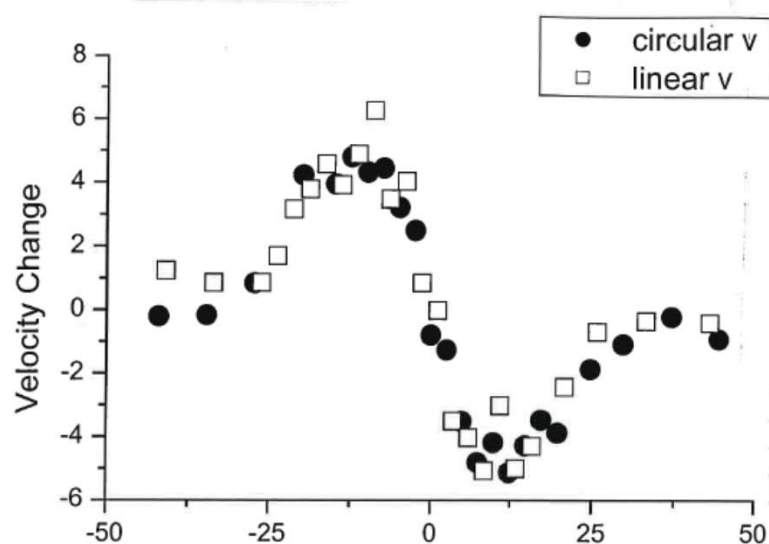


FIG. 1. Schematic of the experimental setup showing the molecular beam apparatus and the laser beam lines.

Other can do what you plan to do

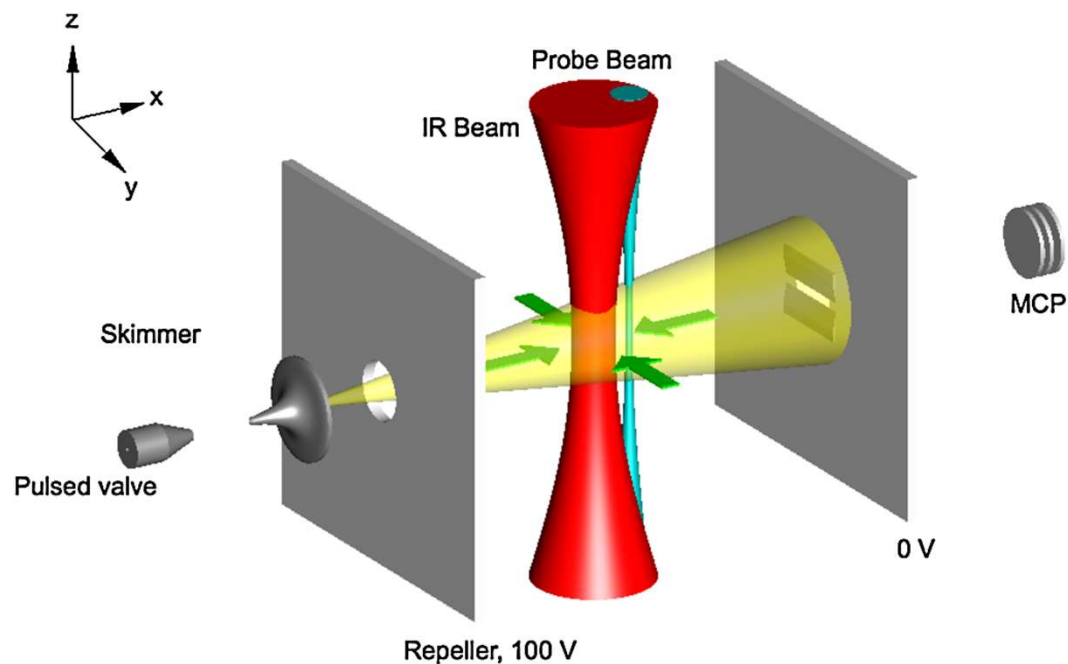


Tailoring the Optical Dipole Force for Molecules by Field-Induced Alignment

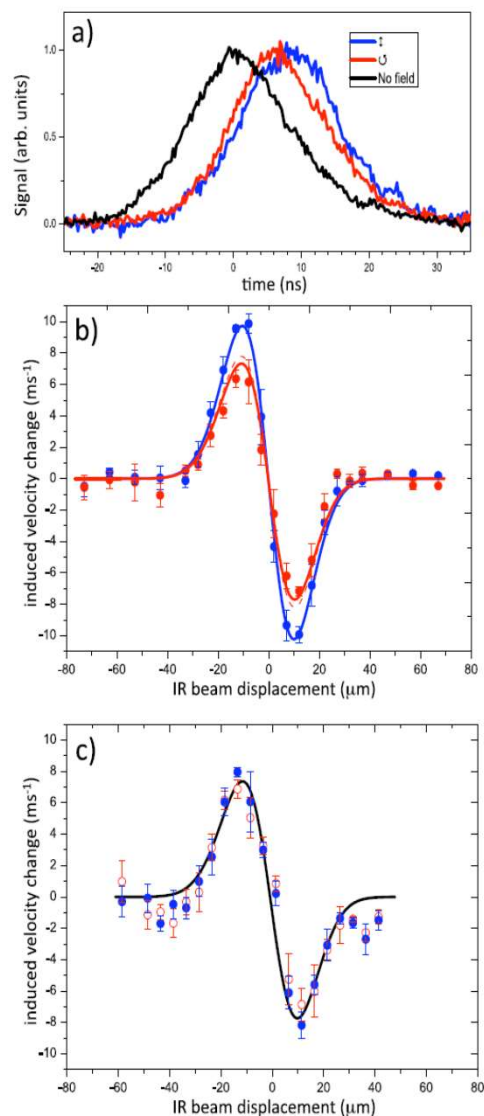
S. M. Purcell and P. F. Barker

Department of Physics and Astronomy, University College London, London, WC1E 6BT, United Kingdom

(Received 17 March 2009; revised manuscript received 7 June 2009; published 5 October 2009)



Other can do what you plan to do



We have measured the velocity imparted to CS_2 molecules for two different field polarizations. A cold beam of CS_2 molecules is produced by expanding 7 mbar of CS_2 in 1800 mbar of argon into a vacuum chamber. This beam intersects with a pulsed optical field of intensity within a time-of-flight (TOF) mass spectrometer as shown in Fig. 2.

ment. We also measured the velocity shifts for each polarization using a molecular beam seeded with 450 mbar of CS_2 where rotational cooling is not effective. The rota-

Which you could have done

Aligning molecules with intense nonresonant laser fields

Jakob Juul Larsen

Institute of Physics and Astronomy, University of Århus, DK-8000 Århus C, Denmark

Hirofumi Sakai

*Department of Chemistry, University of Århus, DK-8000 Århus C, Denmark
and Electrotechnical Laboratory, 1-1-4, Umezono, Tsukuba, Ibaraki 305-8568, Japan*

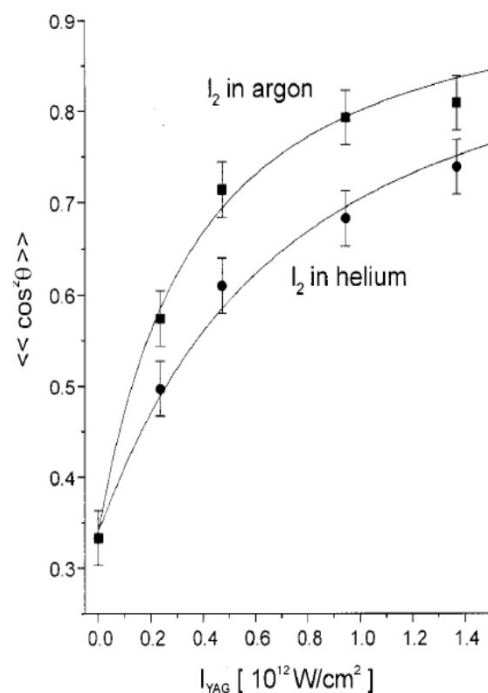
C. P. Safvan

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(Received 29 June 1999; accepted 10 August 1999)



$$\langle \langle \cos^2 \theta \rangle \rangle = \sum_{\tilde{J}} w_{\tilde{J}} \sum_{M=-\tilde{J}}^{M=\tilde{J}} \langle \cos^2 \theta \rangle_{\tilde{J},M}, \quad (3)$$

with the weighting factors given by

$$w_{\tilde{J}} = \frac{\exp[-B\tilde{J}(\tilde{J}+1)/kT]}{\sum_{\tilde{J}} (2\tilde{J}+1) \exp[-B\tilde{J}(\tilde{J}+1)/kT]}. \quad (4)$$

Because of the competition between the initial rotational energy $B\tilde{J}(\tilde{J}+1)$ and the laser induced potential the best aligned states are the ones with the lowest \tilde{J} values and it is thus crucial for the achievement of alignment that the rotational temperature is as low as possible. This property will be used to identify alignment later in the paper.

Rotational temperature matters

Alignment and Trapping of Molecules in Intense Laser Fields

Bretislav Friedrich and Dudley Herschbach

Department of Chemistry, Harvard University, 12 Oxford Street, Cambridge, Massachusetts 02138

(Received 12 December 1994)

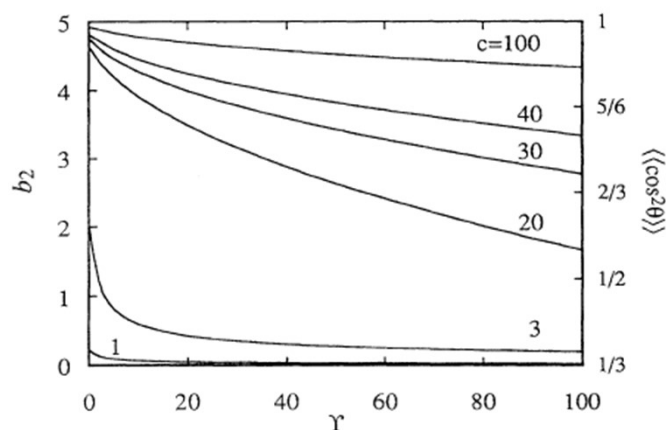


FIG. 2. Dependence of ensemble-averaged alignment parameters b_2 and $\langle \cos^2 \theta \rangle$ of Eq. (8) on anisotropy parameter $c = (\Delta\omega)^{1/2}$ of Eq. (5) and reduced rotational temperature $\gamma \equiv kT/B$.

$$\langle \cos^2 \theta \rangle = \sum_{\tilde{J}} w_{\tilde{J}} \sum_{M=-\tilde{J}}^{M=+\tilde{J}} \langle \cos^2 \theta \rangle_{\tilde{J},M}, \quad (7)$$

where $w_{\tilde{J}} = \exp[-\tilde{J}(\tilde{J} + 1)/Y]/Q_r$, with Q_r the rotational partition function and $Y \equiv kT/B$ the reduced rotational temperature. This determines the second Legendre moment

Which you could have done

Manipulating external degrees of freedom with intense light: Laser focusing and trapping of molecules

Tamar Seideman

Steacie Institute, National Research Council, Ottawa, Ontario K1A 0R6, Canada

(Received 11 July 1996; accepted 15 November 1996)

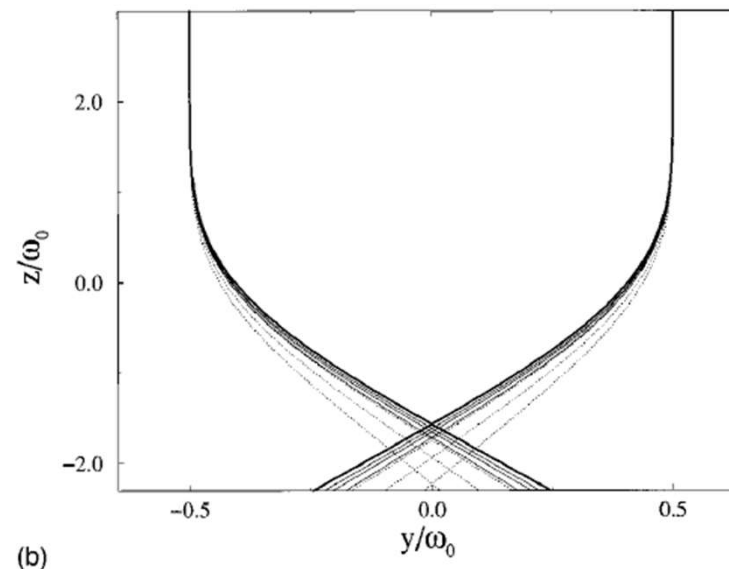
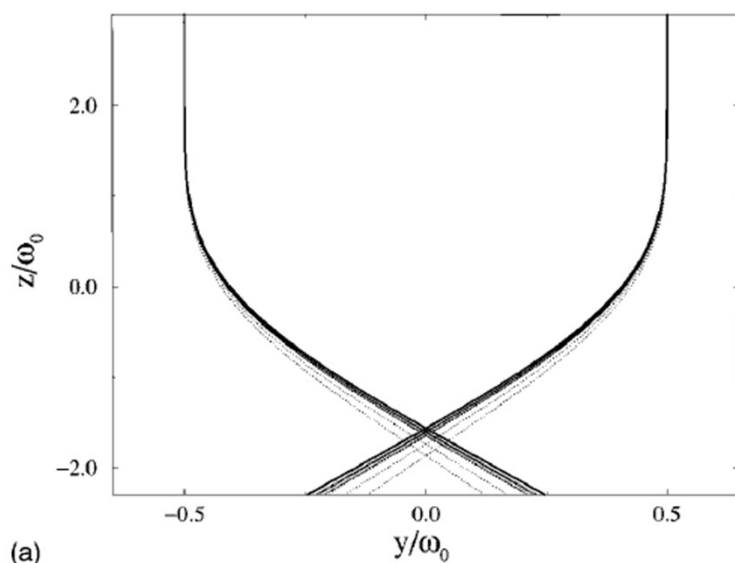


FIG. 7. Classical trajectories at a rotational temperature of (a) 5 K and (b) 10 K. The line type gives a rough measure of the Boltzmann population density. $J_i = 0$ (bold curves) $\{J_i = J_i^{\max}/2, |M| = 0, J_i/2, J_i\}$ (solid curves) $\{J_i = J_i^{\max}, |M| = 0, J_i/2\}$ (dotted curves). J_i^{\max} is the highest significantly populated rotational state at the beam rotational temperature, $J_i^{\max} = 14$ at 5 K and 20 at 10 K.

State-dependent molecular alignment

- Expectation value of $\cos^2 \theta \rightarrow$ degree of alignment

$$\langle \cos^2 \theta \rangle_{J,M} = \langle \cos^2 \theta \rangle_{J,M}(I) = \langle \Psi_{J,M}(I) | \cos^2 \theta | \Psi_{J,M}(I) \rangle$$

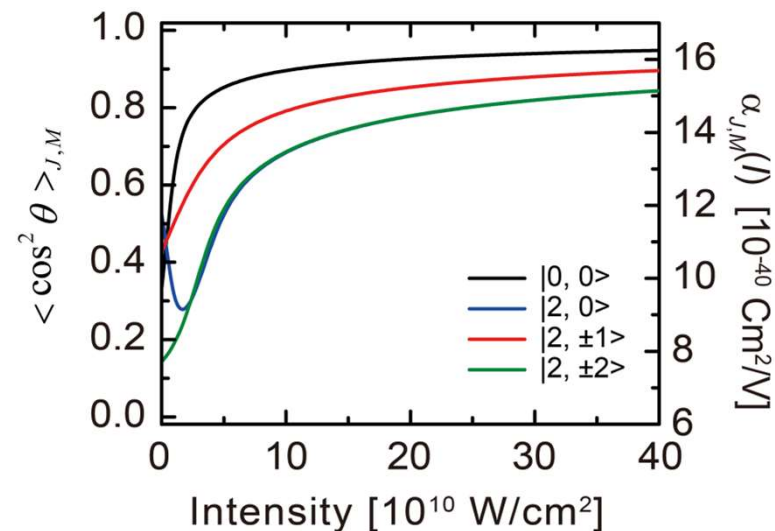
- Approximated interaction potential at the adiabatic regime

$$U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M} [I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0 \quad \alpha_{J,M}(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M} + \alpha_{\perp}$$

- Optical dipole force

$$\mathbf{F}_{J,M}(\mathbf{r}, t) = \frac{1}{2} Z_0 \nabla \{ \alpha_{J,M}(I) [I(\mathbf{r}, t)] I(\mathbf{r}, t) \}$$

$$\Delta v_x = \int \frac{1}{m} F_{J,M}^x(x, y, z, t) dt$$



State-dependent molecular alignment

- Expectation value of $\cos^2 \theta \rightarrow$ degree of alignment

$$\langle \cos^2 \theta \rangle_{J,M} = \langle \cos^2 \theta \rangle_{J,M}(I) = \langle \Psi_{J,M}(I) | \cos^2 \theta | \Psi_{J,M}(I) \rangle$$

- Approximated interaction potential at the adiabatic regime

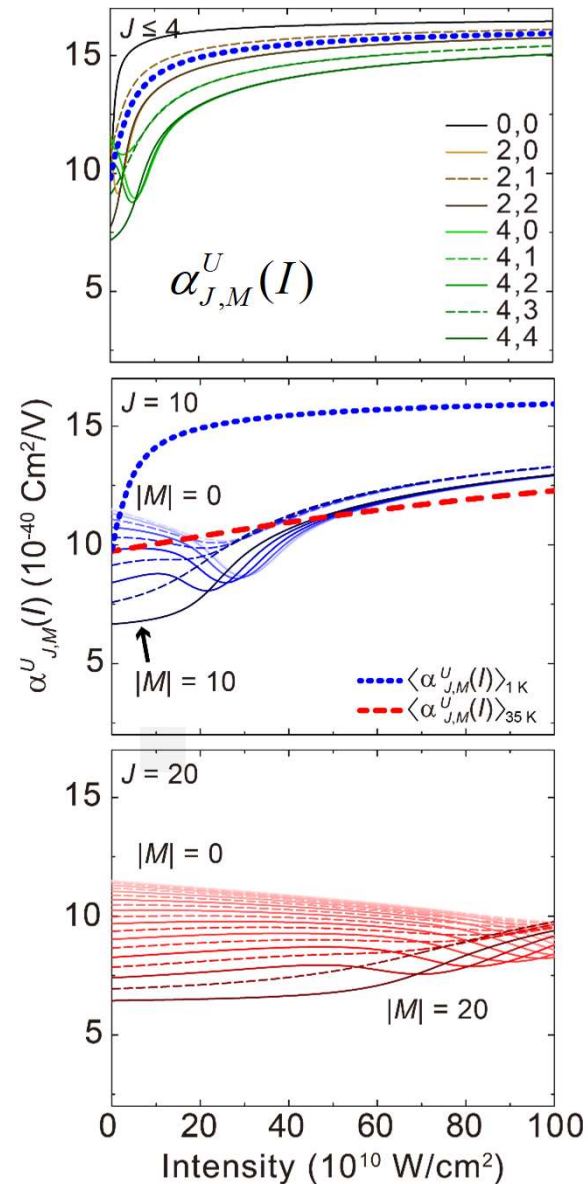
$$U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M}[I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0 \quad \alpha_{J,M}(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M} + \alpha_{\perp}$$

$$i\hbar \frac{\partial}{\partial t} |\Psi_{J,M}\rangle(t) = H(t) |\Psi_{J,M}\rangle(t), \quad H(t) = H_0 + V_{L,C}(t).$$

$$\Psi_{J,M}(I) = \sum_j C_j^{J,M}(I) |jM\rangle$$

$$i\hbar \frac{d}{dt} C_{J,M}(t) = \sum_{J',M'} C_{J',M'}(t) \langle J, M | H(t) | J', M' \rangle,$$

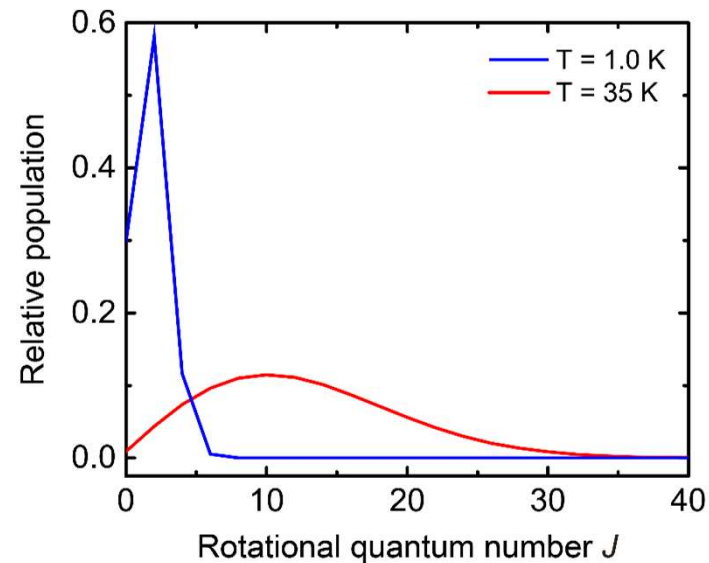
Effective polarizability



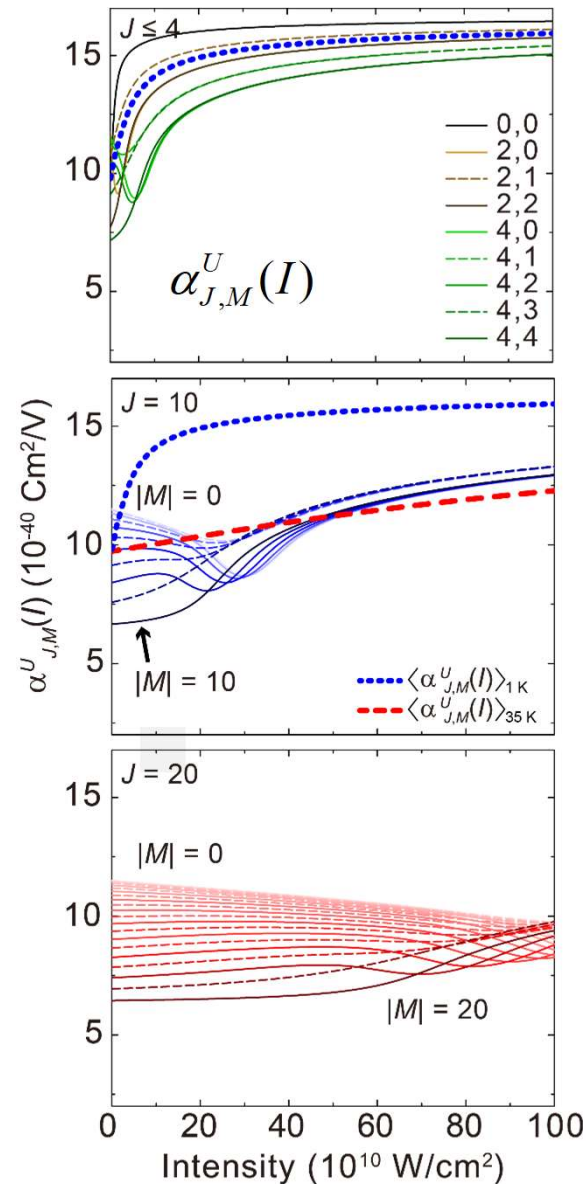
- alignment-including state-dependent polarizability

- $\alpha_{J,M}^U(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$

- $U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M}^U[I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0$



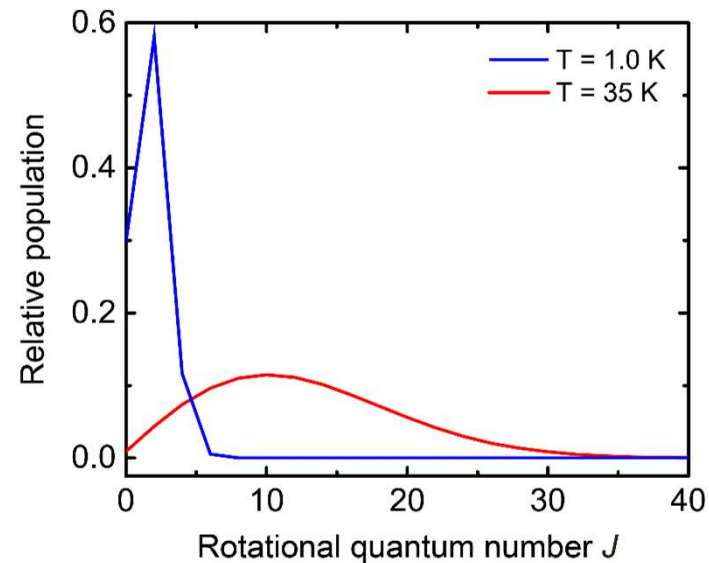
Effective polarizability



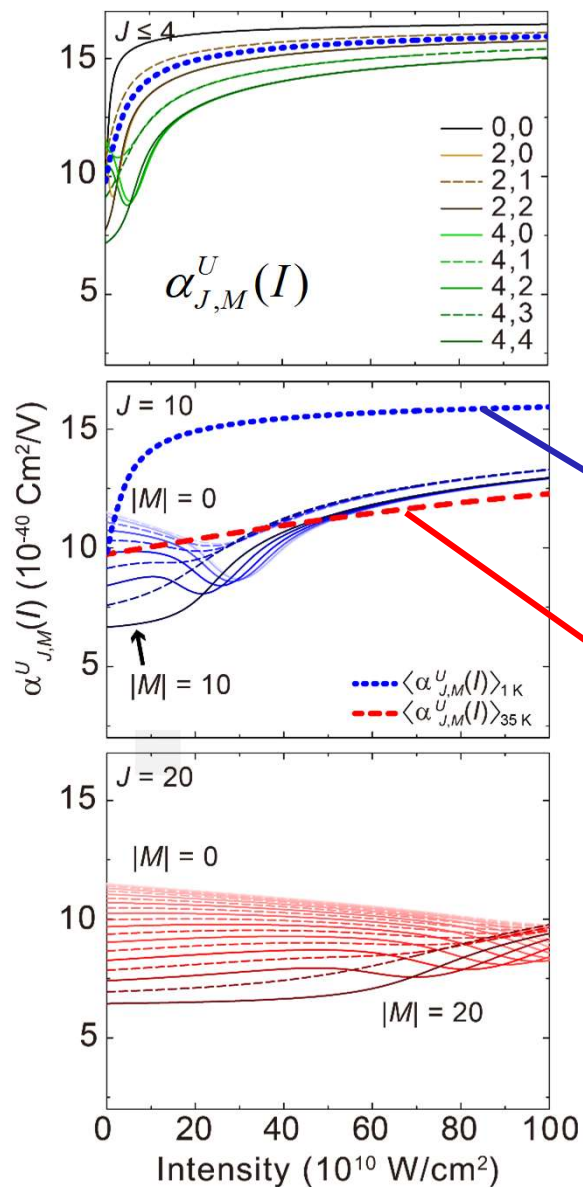
- alignment-including state-dependent polarizability

- $\alpha_{J,M}^U(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$

- $U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M}^U[I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0$



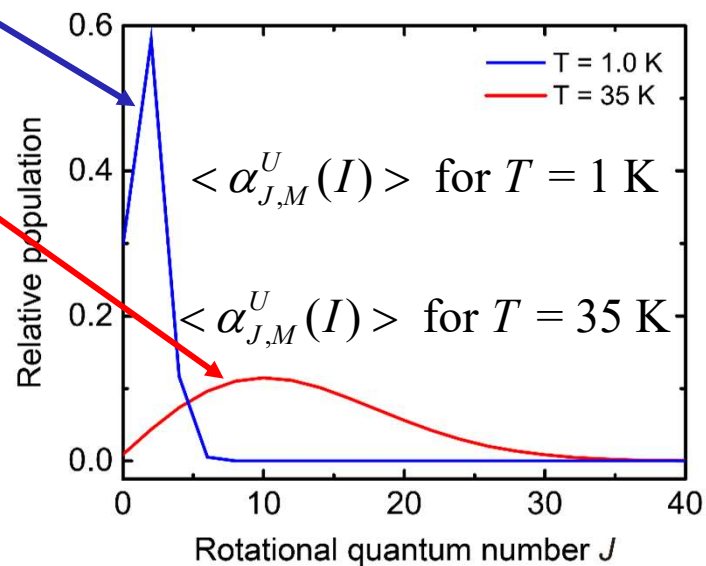
Effective polarizability



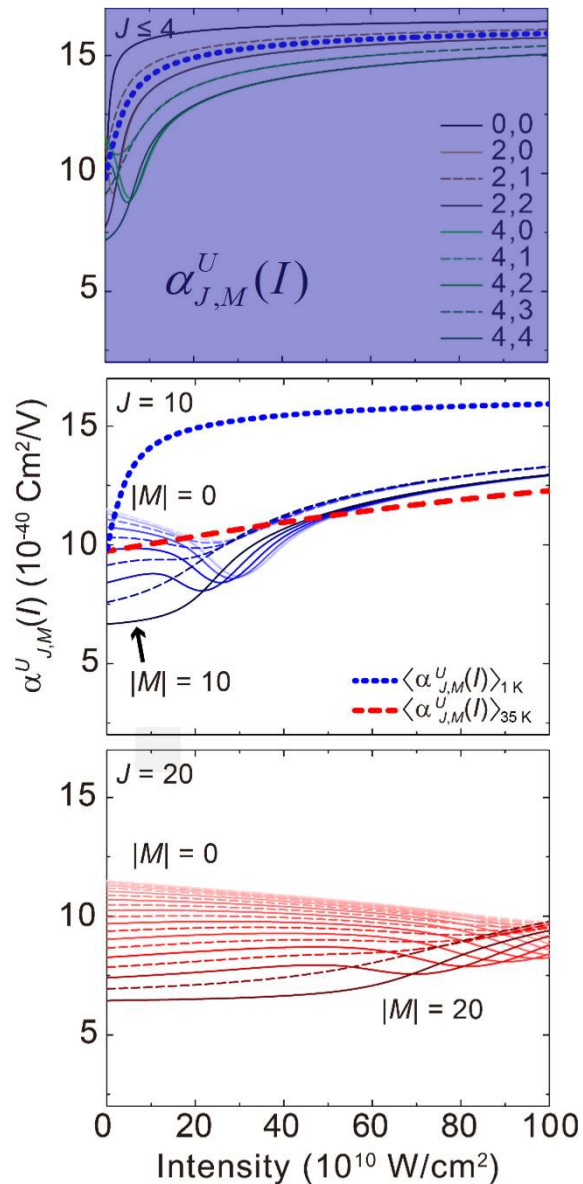
- alignment-including state-dependent polarizability

- $\alpha_{J,M}^U(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$

- $U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M}^U[I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0$



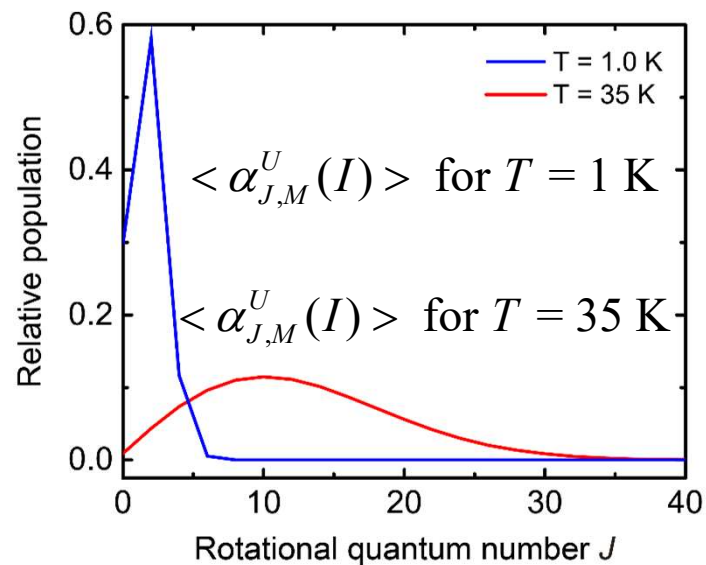
Effective polarizability



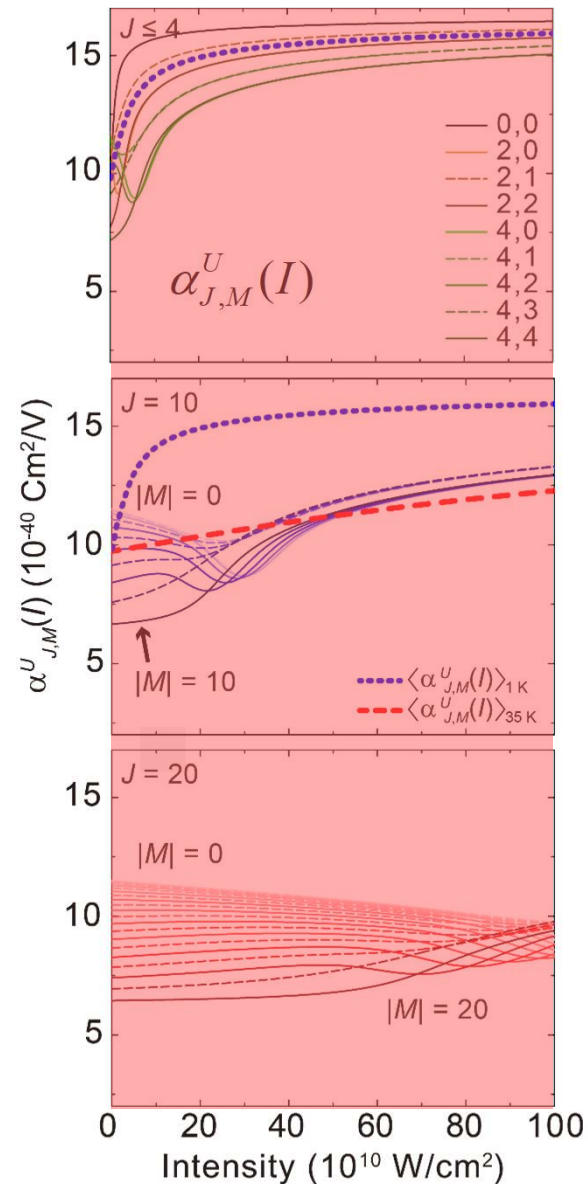
- alignment-including state-dependent polarizability

- $\alpha_{J,M}^U(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$

- $U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M}^U[I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0$



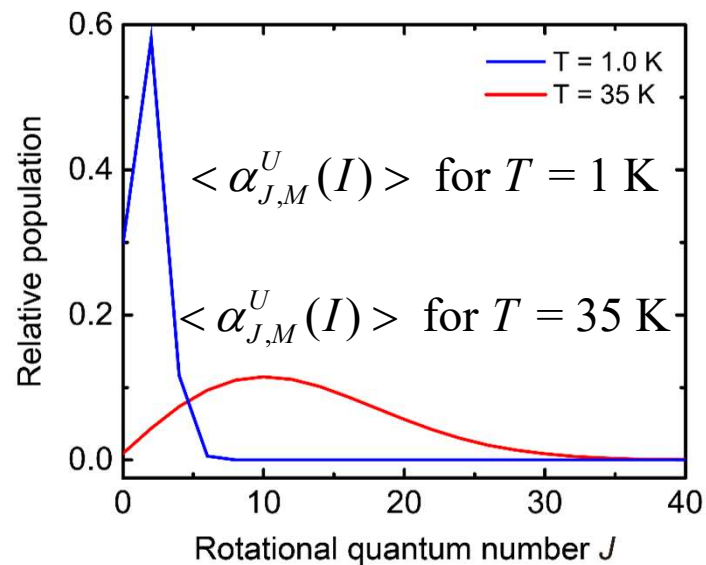
Effective polarizability



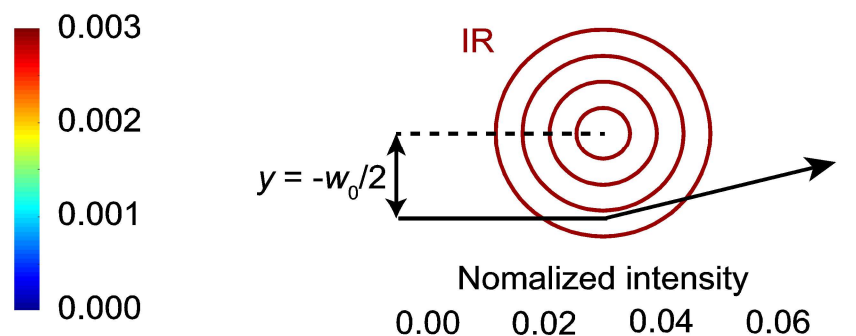
- alignment-including state-dependent polarizability

- $\alpha_{J,M}^U(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$

- $U_{J,M}(\mathbf{r}, t) = -\frac{1}{2} \alpha_{J,M}^U[I(\mathbf{r}, t)] I(\mathbf{r}, t) Z_0$



Our work on the optical force exerting on aligned molecules

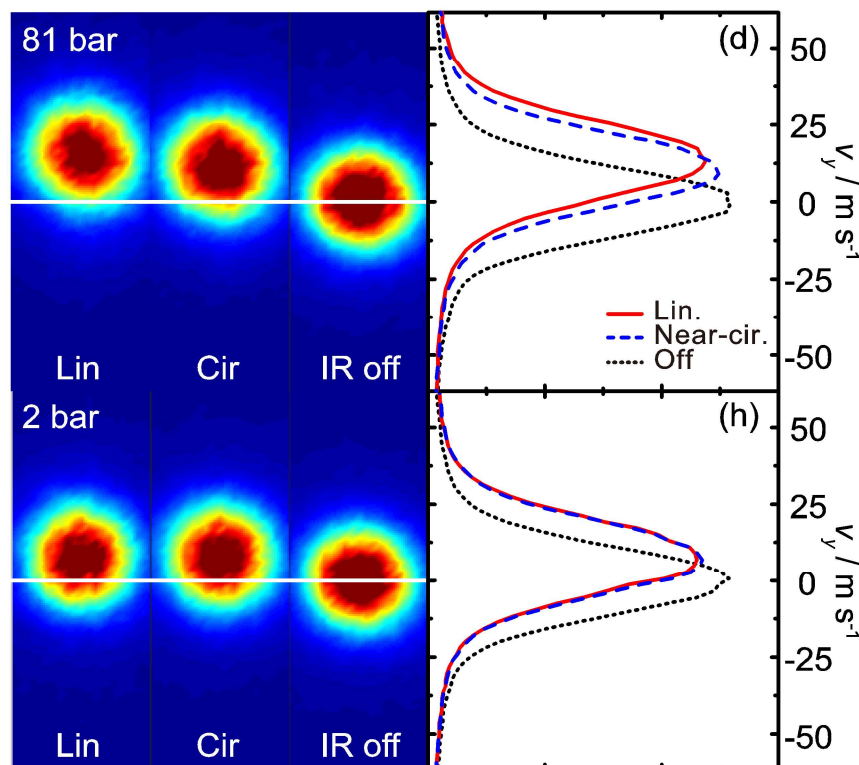


The peak intensity $I_0 = 140 \times 10^{10} \text{ W/cm}^2$

To measure the the maximum Δv_y ,

$$y \sim -w_0/2 = -9.8 \text{ } \mu\text{m}$$

Thus, $I = 0.61I_0 = 85 \times 10^{10} \text{ W/cm}^2$

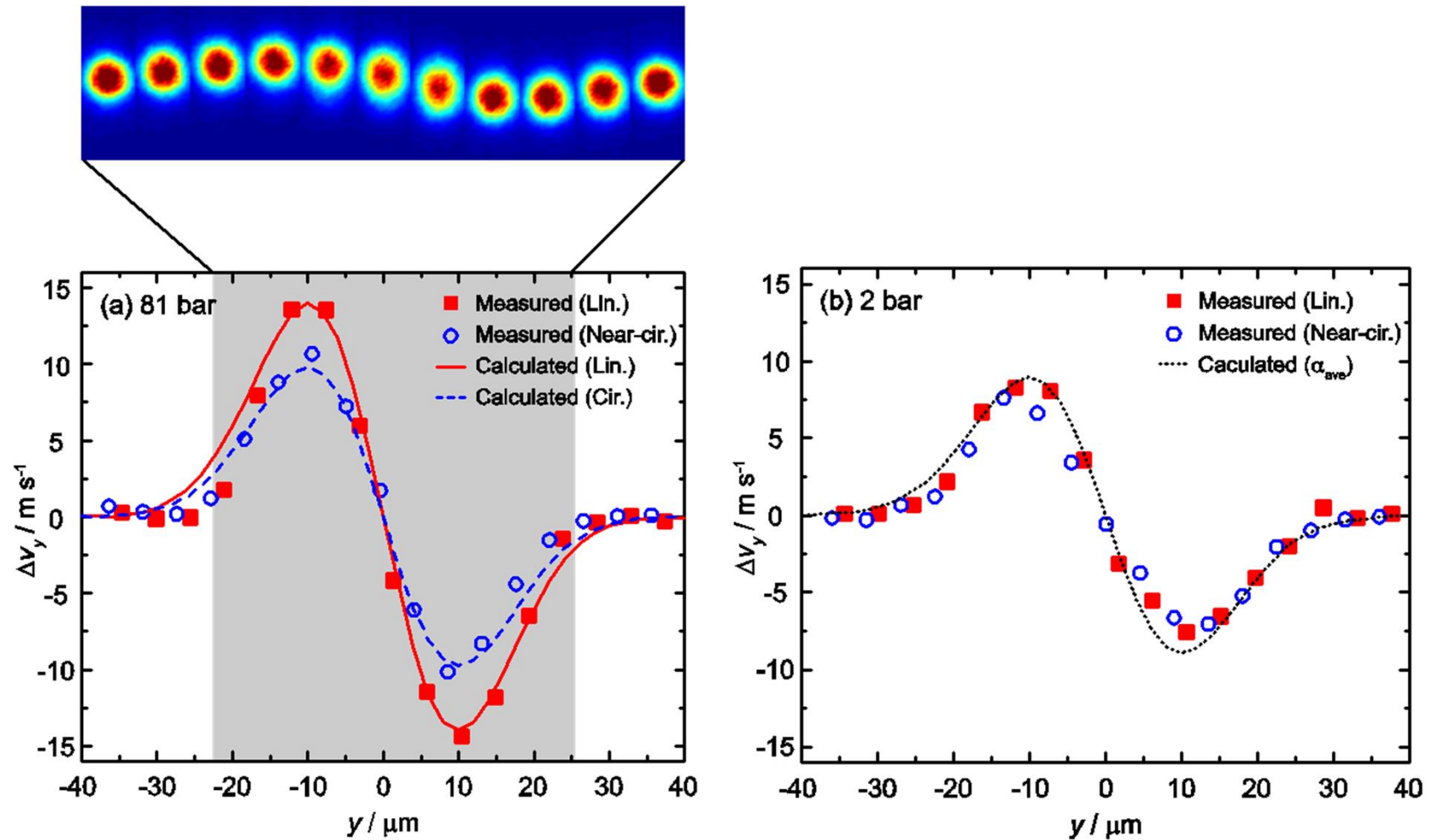


As a result, the estimated Δv_y

	Lin	Cir
81 bar	13.6	10.7
2 bar	8.3	7.6

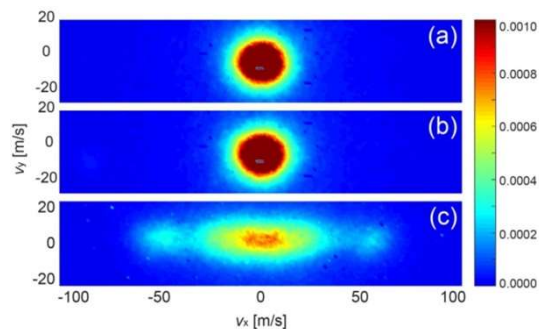
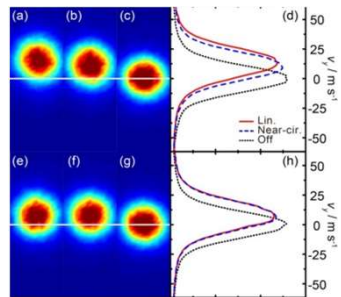
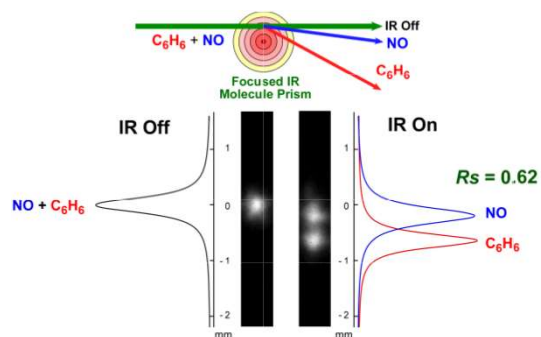
Unit: m/s

Our work on the optical force exerting on aligned molecules

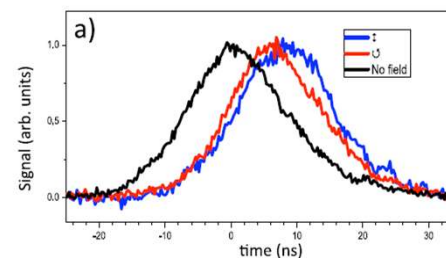
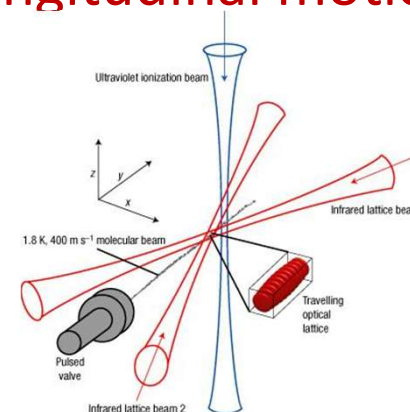


Manipulating translational motions of molecules (NR LF)

transverse motion



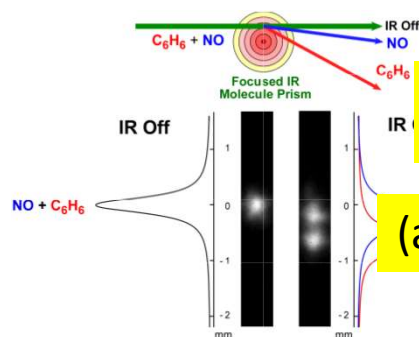
longitudinal motion



- H. Stapelfeldt, H. Sakai, E. Constant, and P. B. Corkum, *Phys. Rev. Lett.* **79**, 2787 (1997).
 B. S. Zhao et al., *Phys. Rev. Lett.* **85**, 2705 (2000).
 B.S. Zhao et al., *J. Chem. Phys.* **119**, 8905 (2003).
 X. N. Sun, B. G. Jin, L. Y. Kim, B. J. Kim, and B. S. Zhao, *Chem. Phys. Chem.* **17**, 3701 (2016).
 X. N. Sun, L. Y. Kim, B. S. Zhao, and D. S. Chung, *Phys. Rev. Lett.* **115**, 223001, (2015).
 R. Fulton, A. I. Bishop, and P. F. Barker, *Phys. Rev. Lett.* **93**, 243004 (2004).
 R. Fulton, A. I. Bishop, M. N. Shneider, and P. F. Barker, *Nat. Phys.* **2**, 465 (2006).
 S. M. Purcell and P. F. Barker, *Phys. Rev. Lett.* **103**, 153001 (2009)..

Manipulating translational motions of molecules

transverse motion

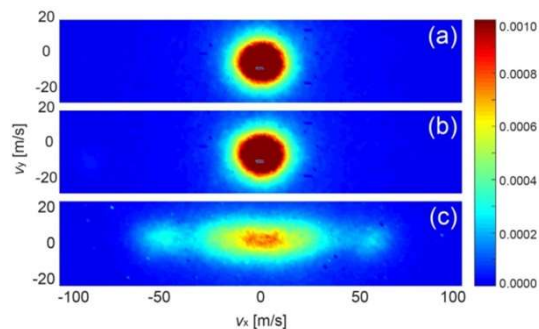
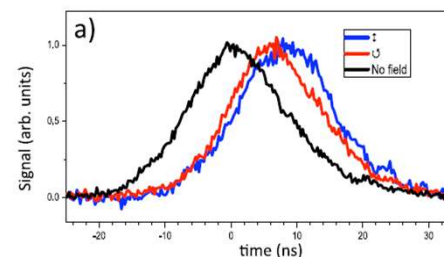
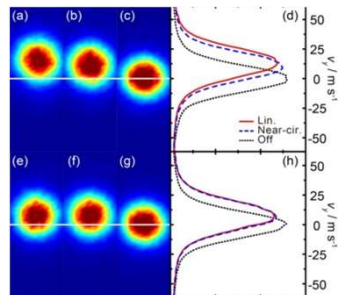
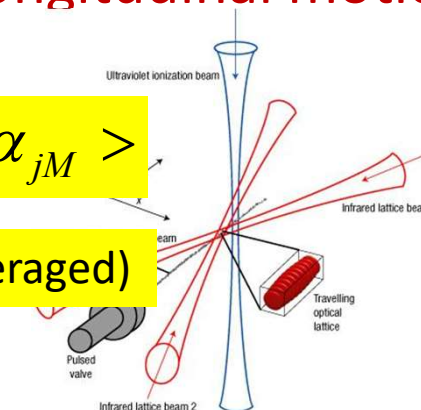


$$\alpha_{ave} = \langle \alpha_{JM} (I = 0) \rangle = \langle \alpha_{JM} \rangle$$

$$Rs = 0.62$$

(alignment-ignored and state-averaged)

longitudinal motion

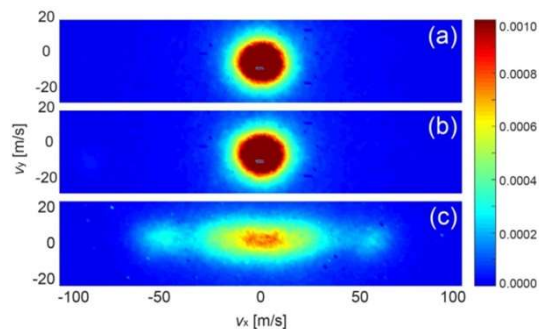
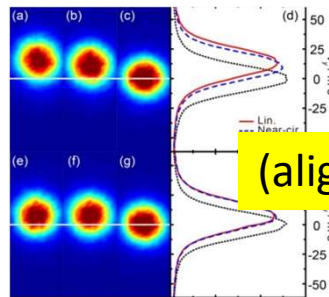
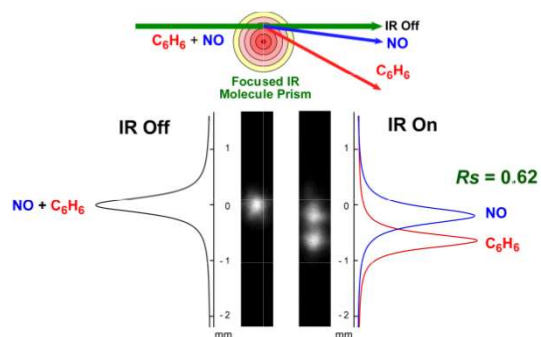


H. Stapelfeldt, H. Sakai, E. Constant, and P. B. Corkum, *Phys. Rev. Lett.* **79**, 2787 (1997).
 B. S. Zhao et al., *Phys. Rev. Lett.* **85**, 2705 (2000).
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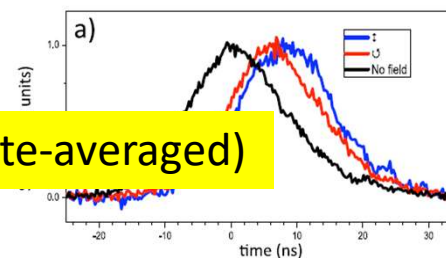
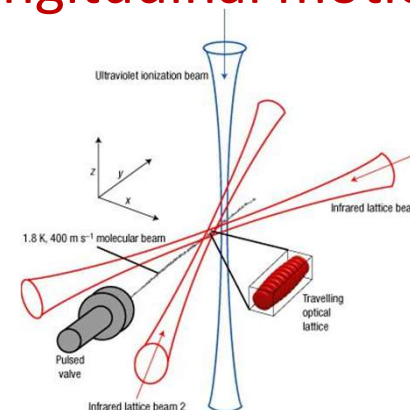
R. Fulton, A. I. Bishop, and P. F. Barker, *Phys. Rev. Lett.* **93**, 243004 (2004).
 R. Fulton, A. I. Bishop, M. N. Shneider, and P. F. Barker, *Nat. Phys.* **2**, 465 (2006).

Manipulating translational motions of molecules

transverse motion



longitudinal motion



$$\langle \alpha_{JM}(I) \rangle$$

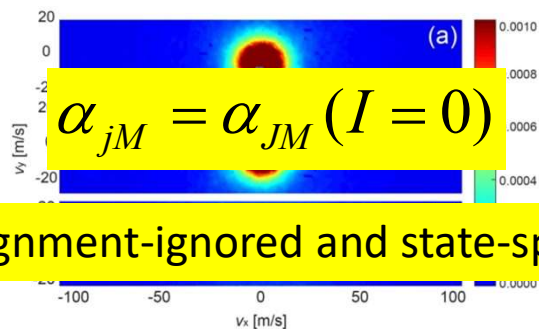
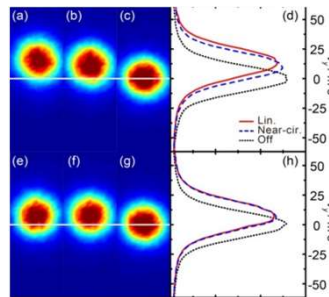
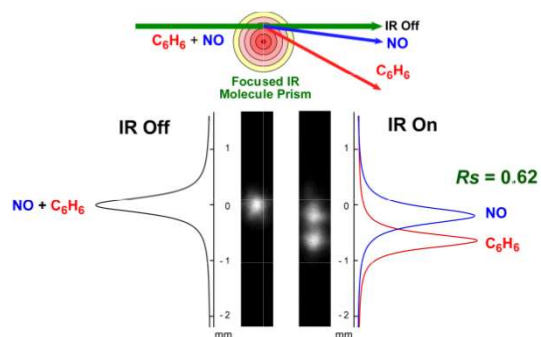
(alignment-considered and state-averaged)

X. N. Sun, B. G. Jin, L. Y. Kim, B. J. Kim, and B. S. Zhao, *Chem. Phys. Chem.* **17**, 3701 (2016).

S. M. Purcell and P. F. Barker, *Phys. Rev. Lett.* **103**, 153001 (2009)..

Manipulating translational motions of molecules

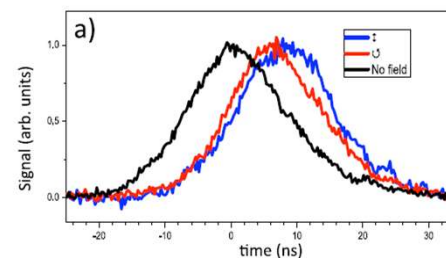
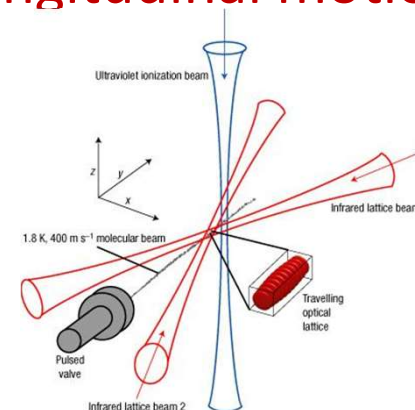
transverse motion



$$\alpha_{jM} = \alpha_{JM} (I = 0)$$

(alignment-ignored and state-specific)

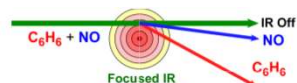
longitudinal motion



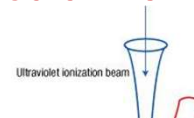
X. N. Sun, L. Y. Kim, B. S. Zhao, and D. S. Chung, *Phys. Rev. Lett.* **115**, 223001, (2015).

Manipulating translational motions of molecules

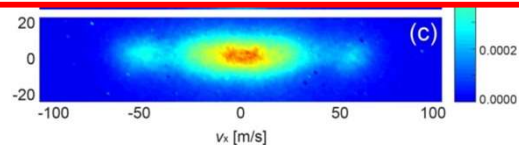
transverse motion



longitudinal motion

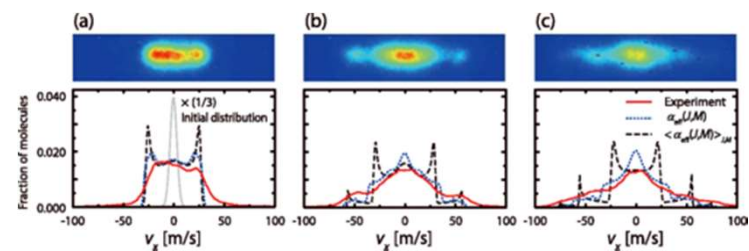
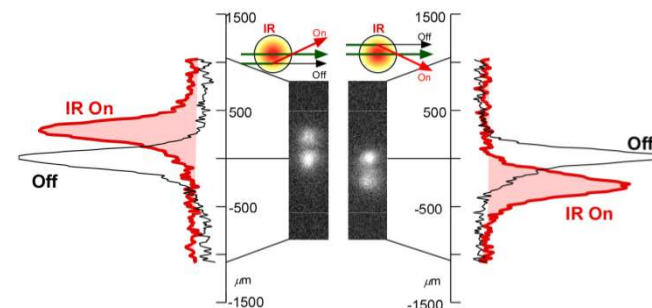
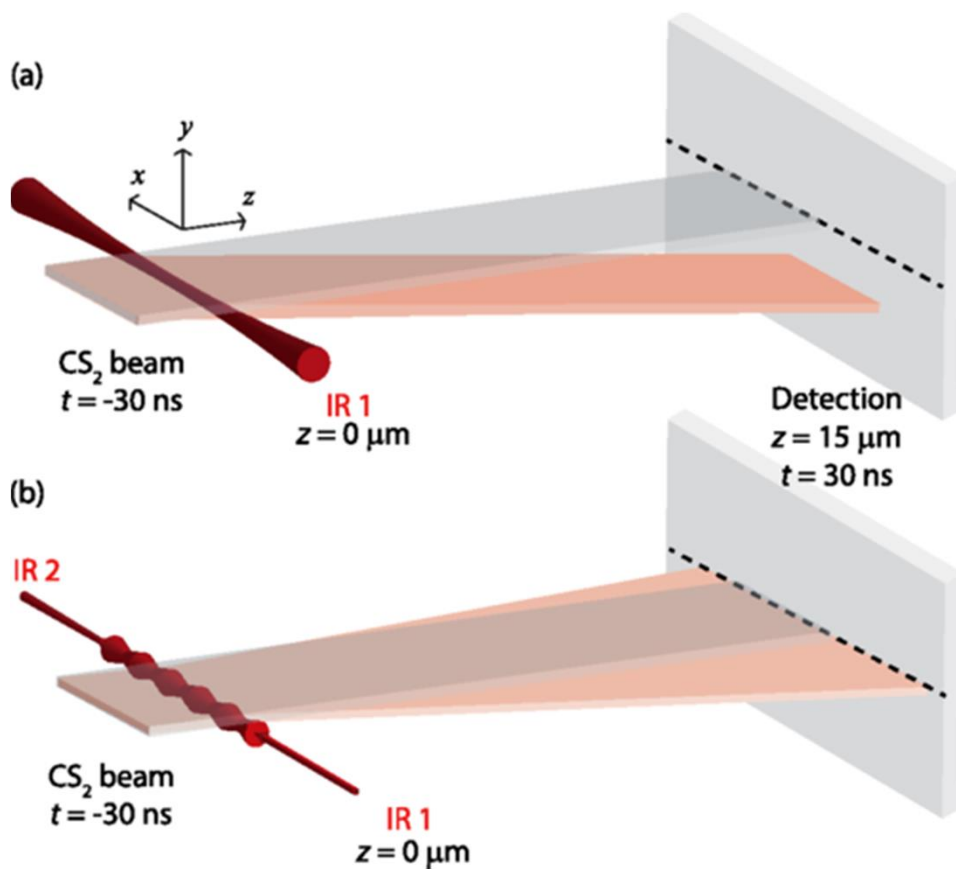


$\alpha_{JM}(I)$



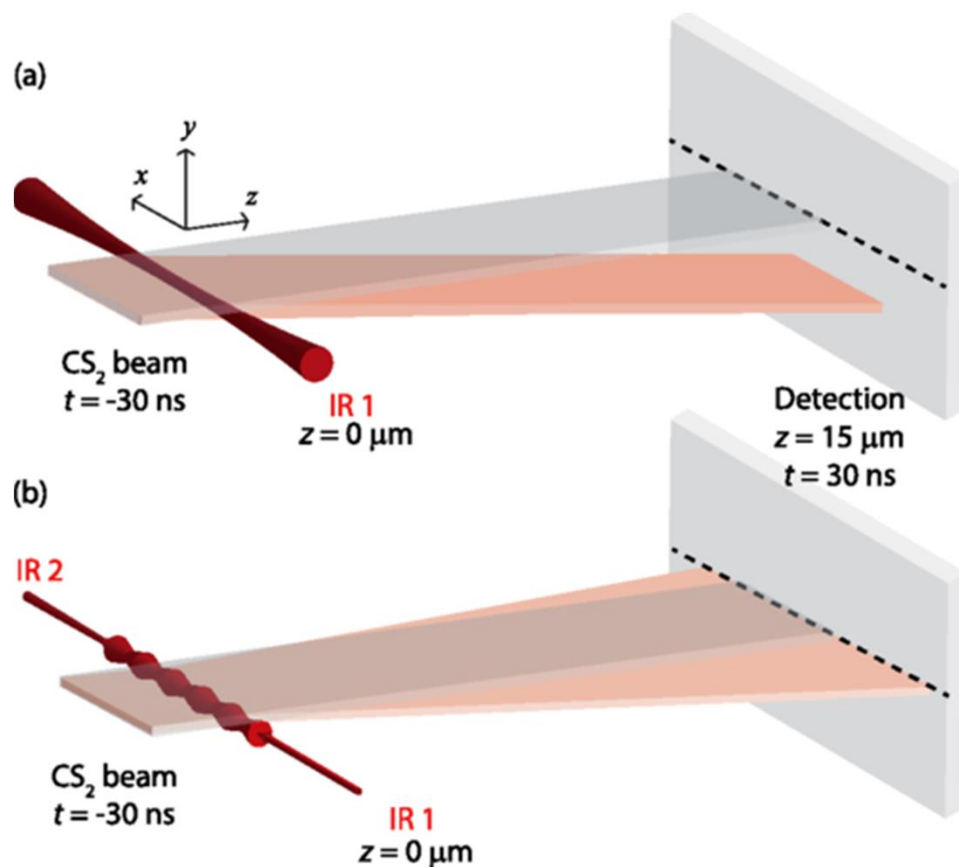
Trajectory simulation

Calculation schemes



Trajectory simulation

Calculation schemes

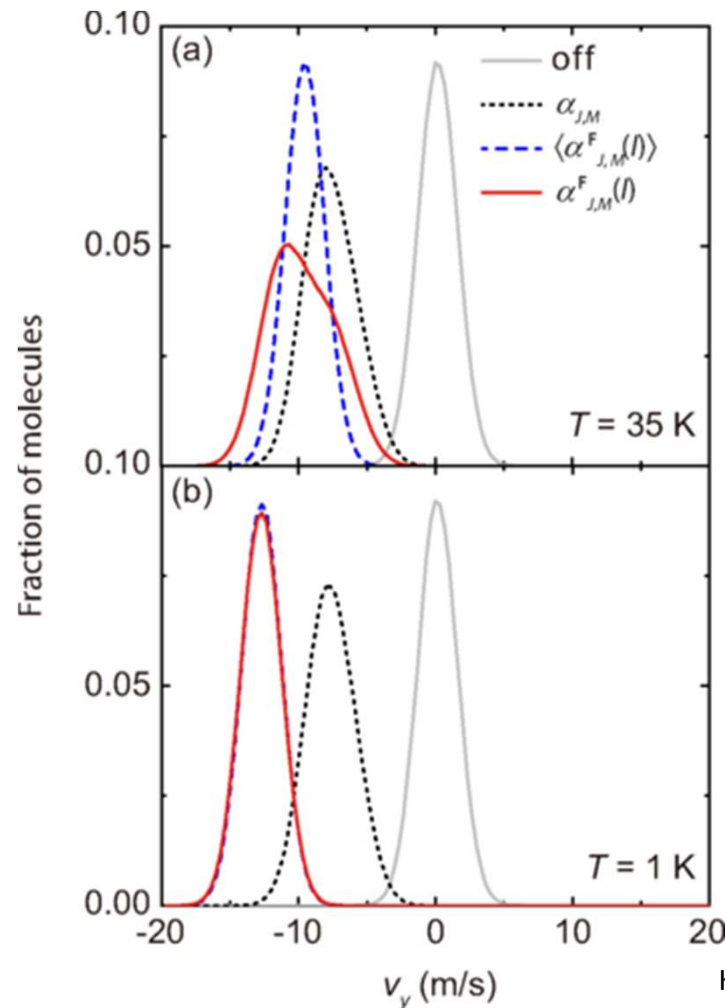


- IR beams
pulse width $\tau = 10$ ns
waist radius $\omega_0 = 23.5$ μ m
- molecular beams (¹²C³²S₂)
 $v_{z,mp} = 560$ m/s
 $\Delta v_z = 56$ m/s
 $T_{rot} = T = 1$ and 35 K

Calculated velocity change by trajectory simulation

Deflection

$$\Delta v_y = \int \frac{1}{m} F_{J,M}^y(x, y, z, t) dt \quad I_0 = 6.1 \times 10^{11} \text{ W/cm}^2$$



$$\alpha_{j,M} = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{j,M} + \alpha_{\perp}$$

$$\langle \alpha_{J,M}^U(I) \rangle = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$$

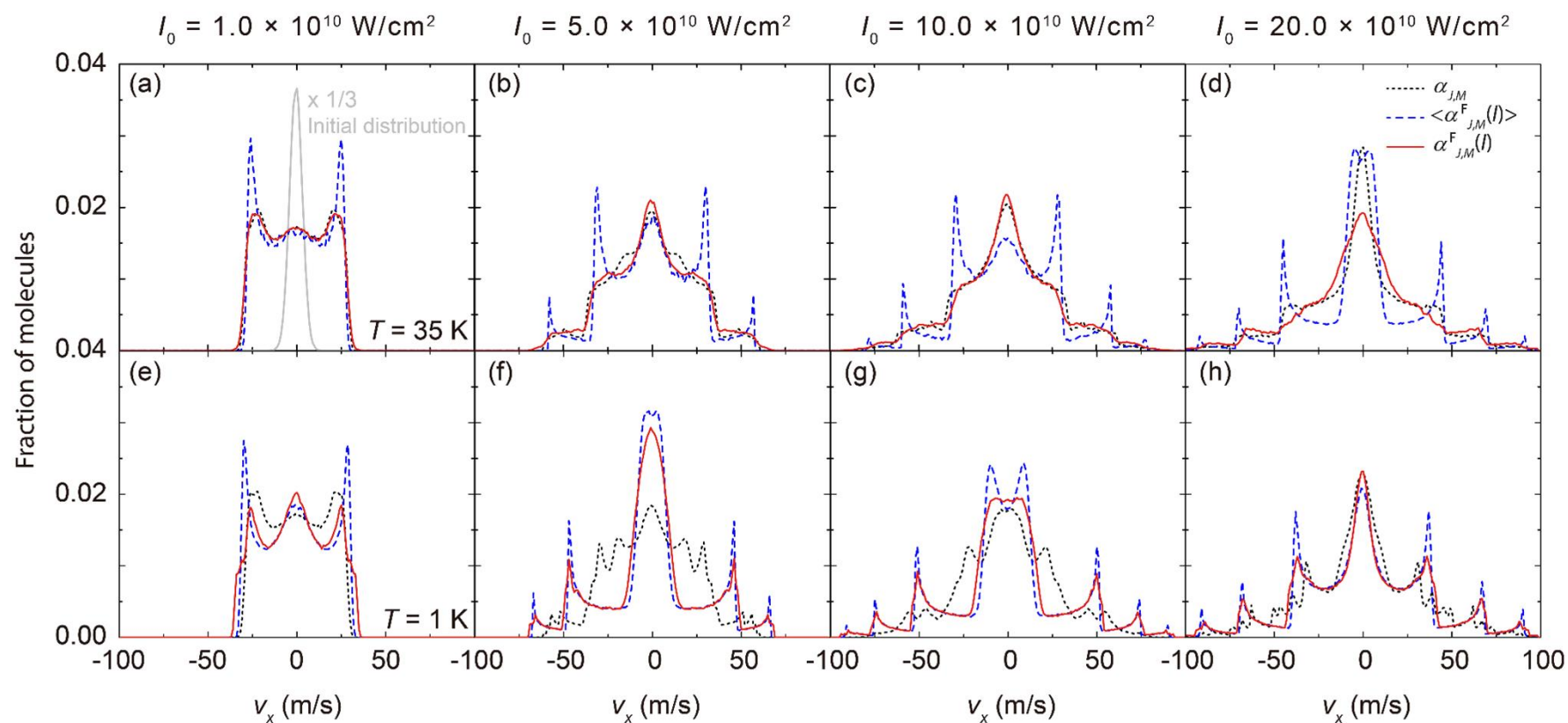
$$\alpha_{J,M}^U(I) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{J,M}(I) + \alpha_{\perp}$$

- $\langle \alpha_{J,M}^U(I) \rangle$ is good approximation
 - for $T = 1 \text{ K}$
 - for $T = 35 \text{ K}$ when only velocity changes are considered

Calculated velocity change by trajectory simulation

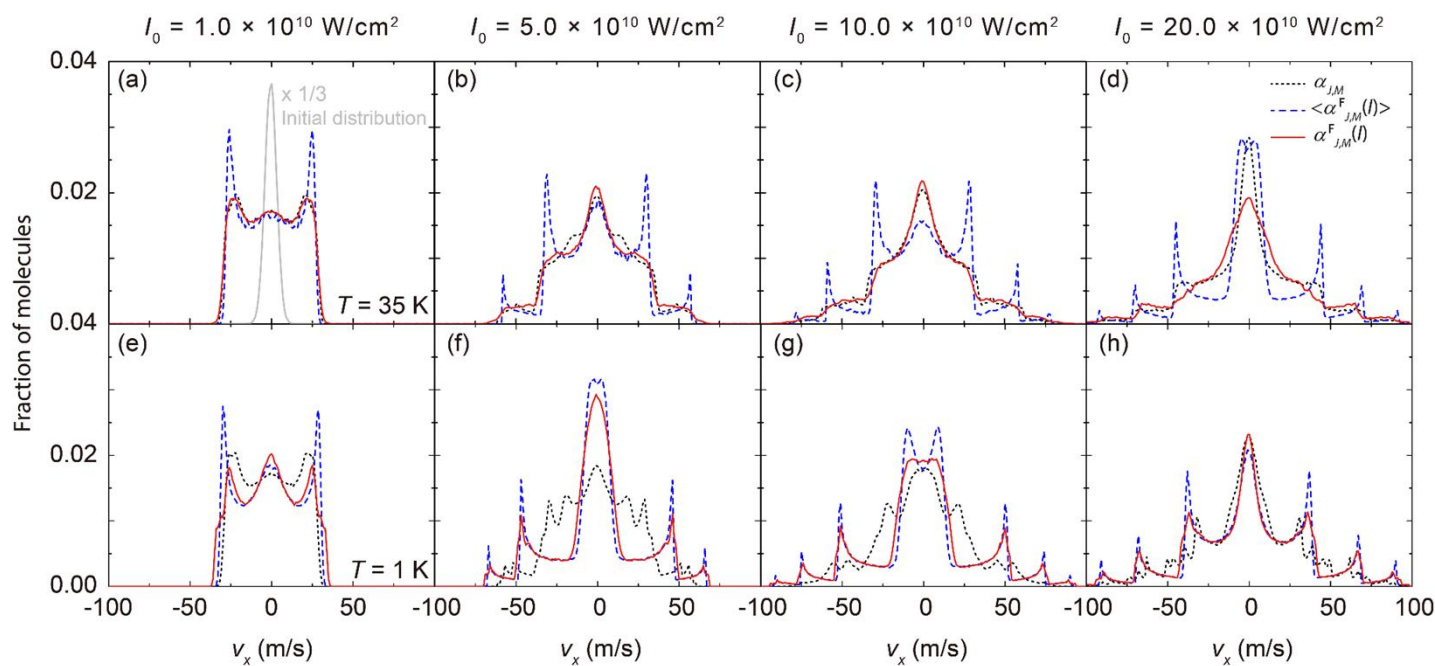
Dispersion

$$\Delta v_x = \int \frac{1}{m} F_{J,M}^x(x, y, z, t) dt$$



Calculated velocity change by trajectory simulation

transverse motion



$$\alpha_{jM}$$

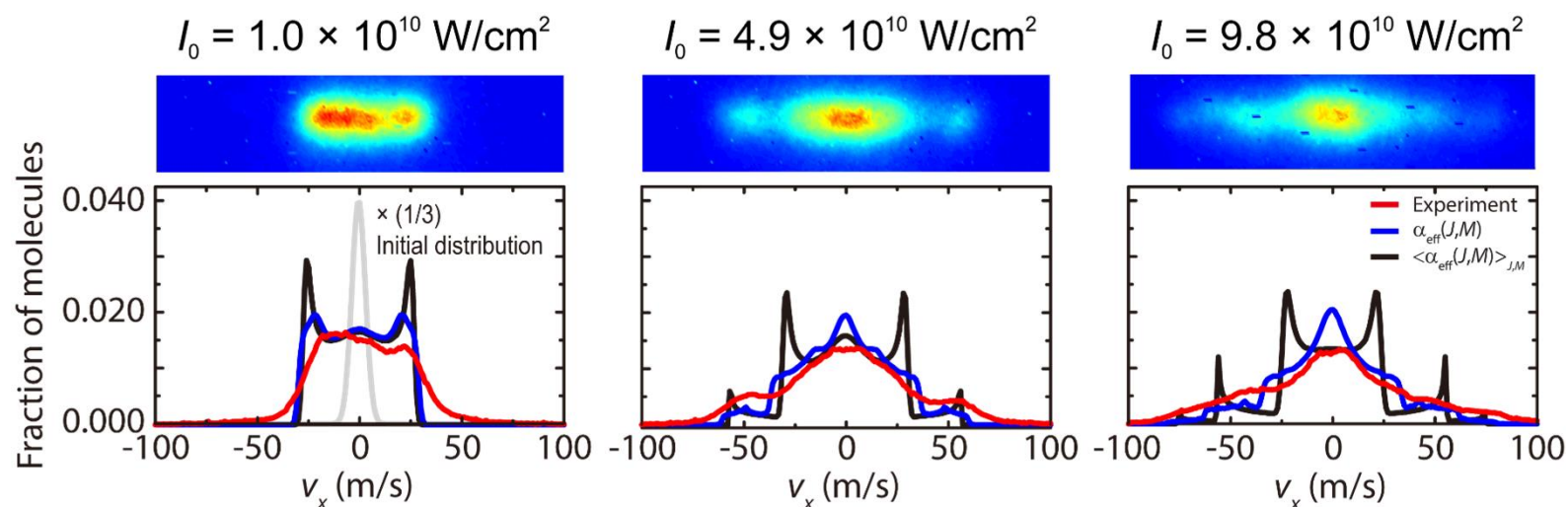
$$\langle \alpha_{JM}(I) \rangle$$

$$\alpha_{JM}(I)$$

Good approximations

Alignment-neglecting state-dependent polarizability

$$\alpha_{j,M}(0) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{j,M}(0) + \alpha_{\perp}$$



- good approximation for low rotational temperature ($T = 35 \text{ K}$) and low laser intensity ($I_0 < 10^{11} \text{ W/cm}^2$)

Good approximations

Alignment-neglecting state-dependent polarizability

$$\alpha_{j,M}(0) = (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle_{j,M}(0) + \alpha_{\perp}$$

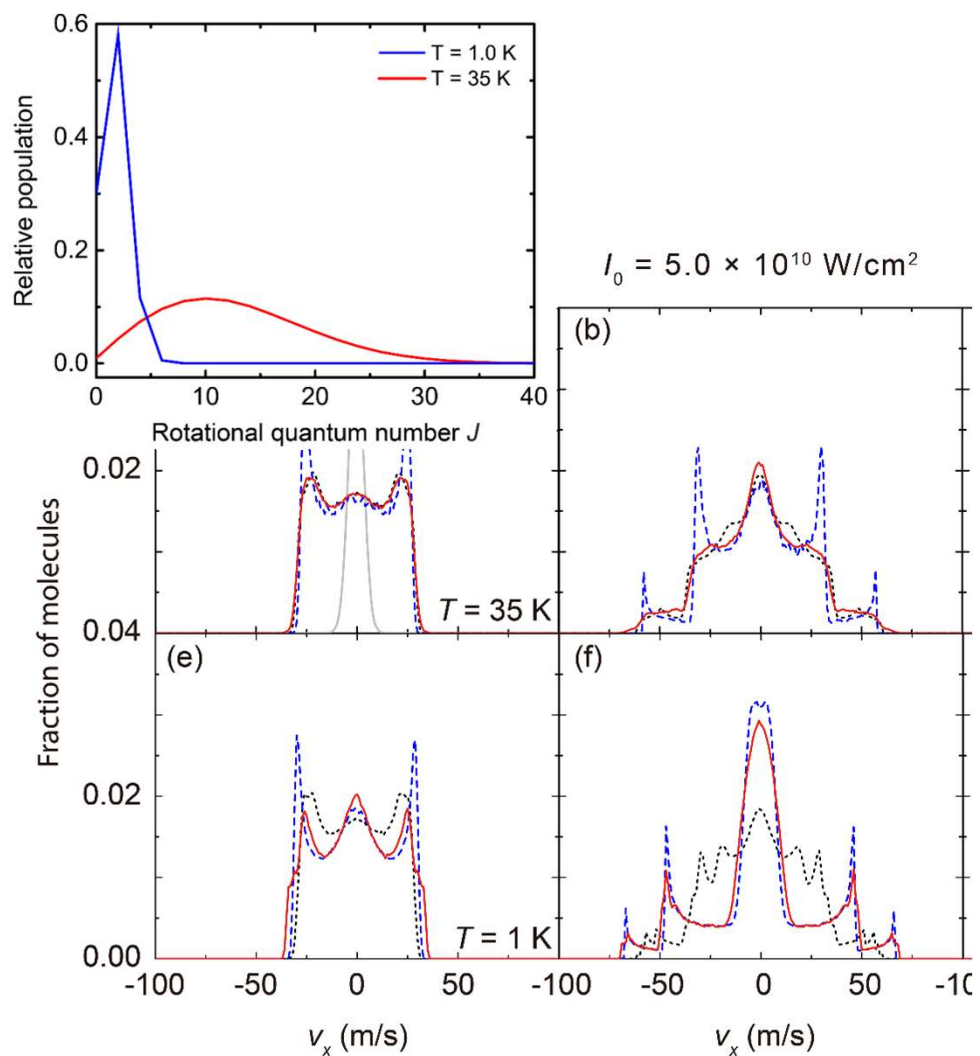
$$\overline{\cos^2 \theta} = \langle J, M | \cos^2 \theta | J, M \rangle = \frac{1}{3} + \frac{2 J(J+1) - 3M^2}{3(2J+3)(2J-1)}. \quad (2)$$

$$D_{M0}^J(\varphi, \theta, \xi) = \left(\frac{4\pi}{2J+1} \right)^{1/2} Y_{JM}^*(\theta, \varphi), \quad \cos^2 \theta = \frac{1}{3}(2P_2(\cos \theta) + 1) = \frac{1}{3}(2D_{00}^2(\varphi, \theta, \xi) + 1).$$

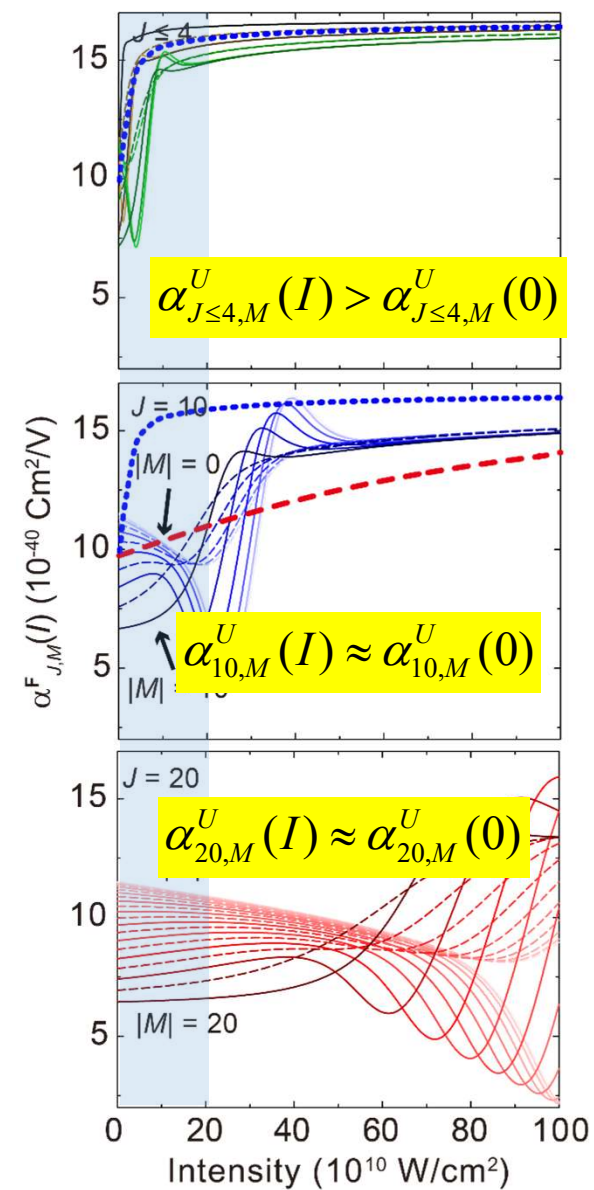
$$D_{00}^J(\varphi, \theta, \xi) = P_J(\cos \theta).$$

$$\begin{aligned} & \int Y_{l_1 m_1}(\theta, \varphi) Y_{l_2 m_2}(\theta, \varphi) Y_{l_3 m_3}(\theta, \varphi) \sin \theta \, d\theta \, d\varphi \\ &= \sqrt{\frac{(2l_1+1)(2l_2+1)(2l_3+1)}{4\pi}} \begin{pmatrix} l_1 & l_2 & l_3 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \end{aligned}$$

Good approximations



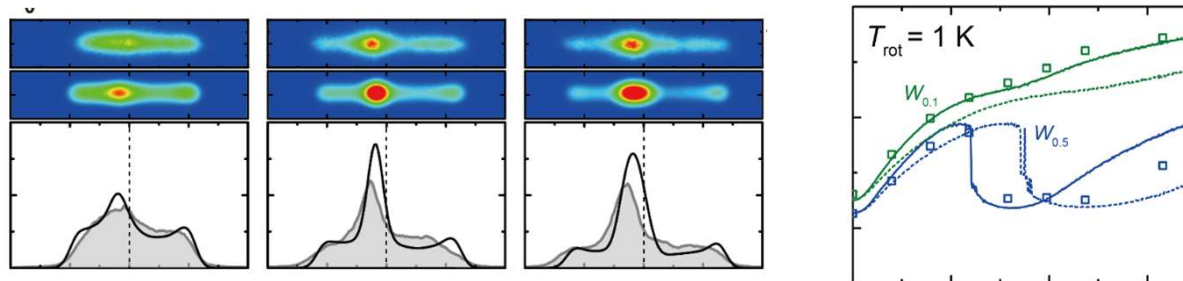
$$0 < I < 20 \times 10^{10} \text{ W/cm}^2$$



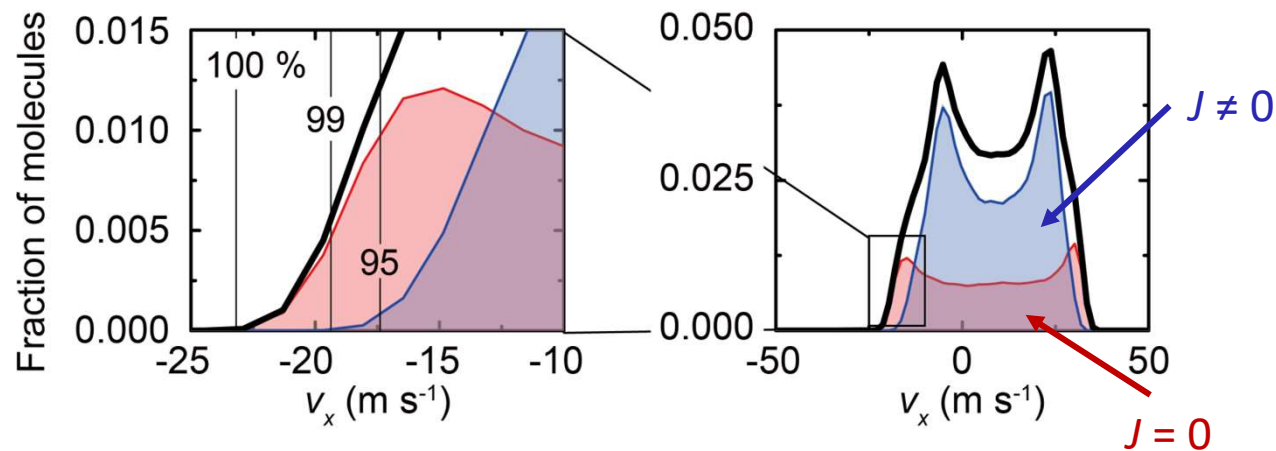
Limit of the approximation

Demonstration of the molecular alignment effect in their scattering
from an optical standing wave

$$\langle \alpha_{JM}(I) \rangle \quad \alpha_{JM}(I)$$

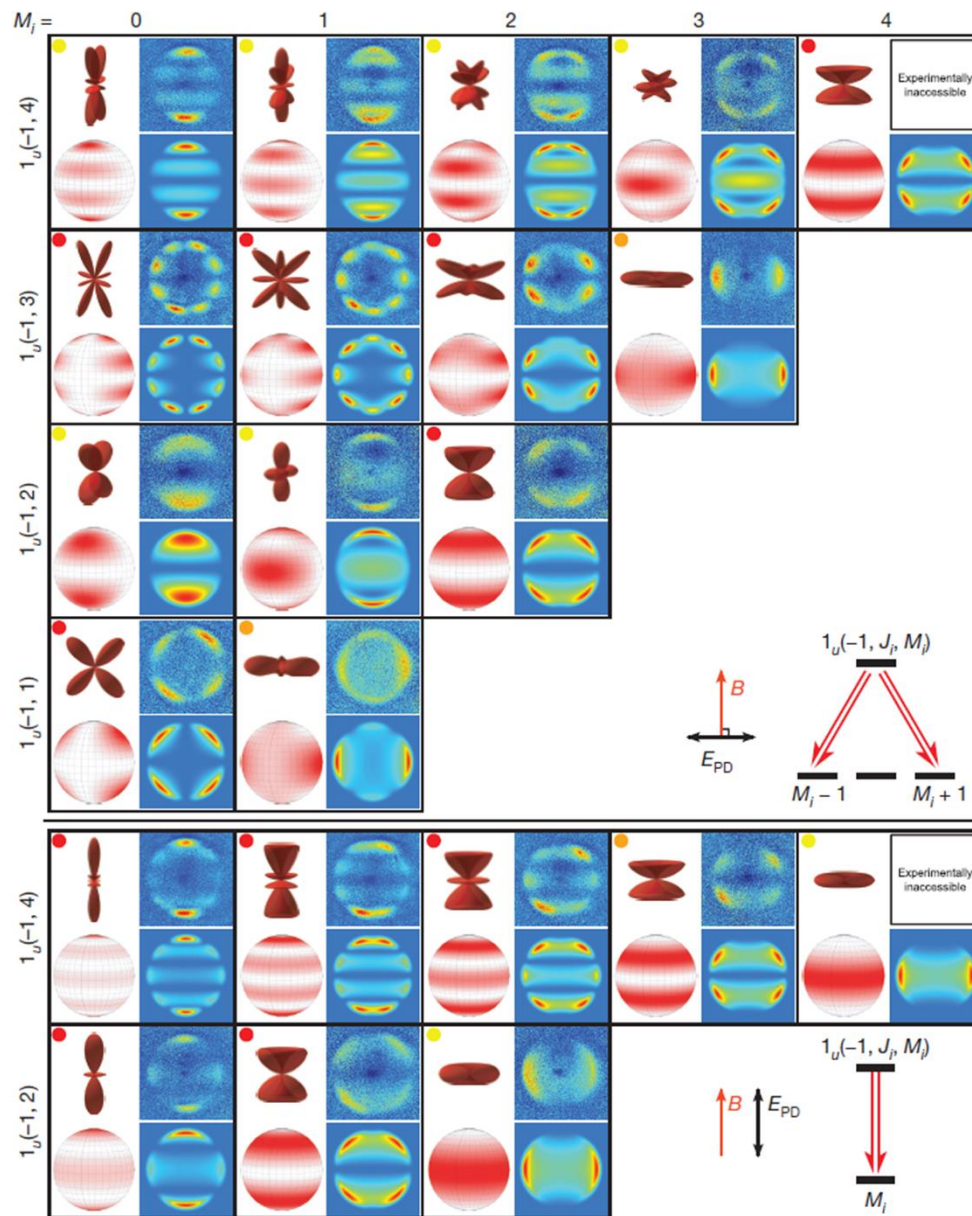


Rotational-state selection of nonpolar molecules



0.01%, 0.76%, 2.09%

Molecules in an optical trap and in a fast beam



ICAP 2016

Molecules in an optical trap and in a fast beam

Photodissociation of ultracold diatomic strontium molecules with quantum state control

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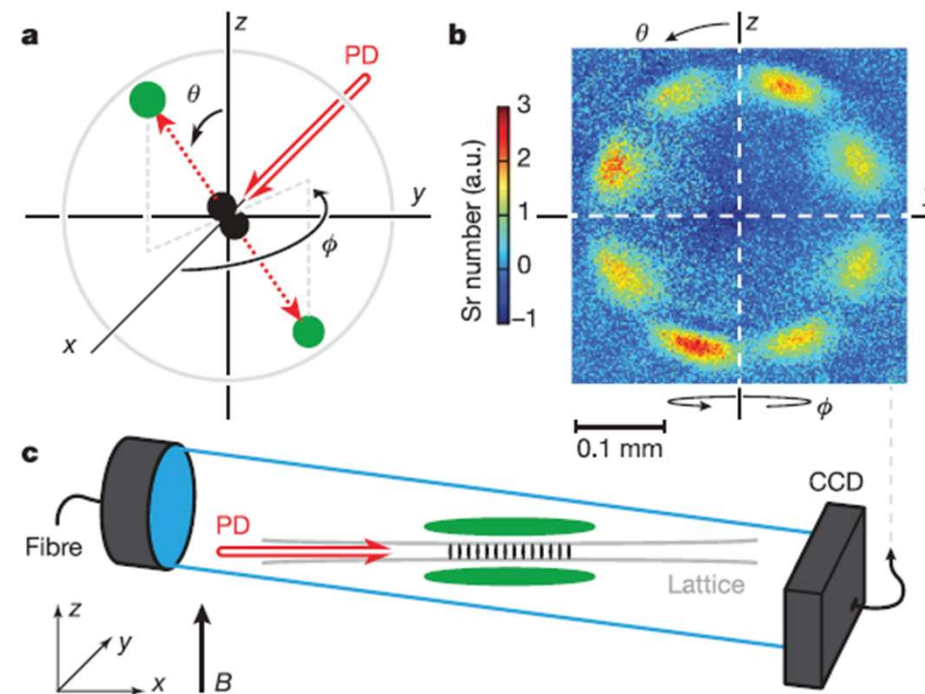
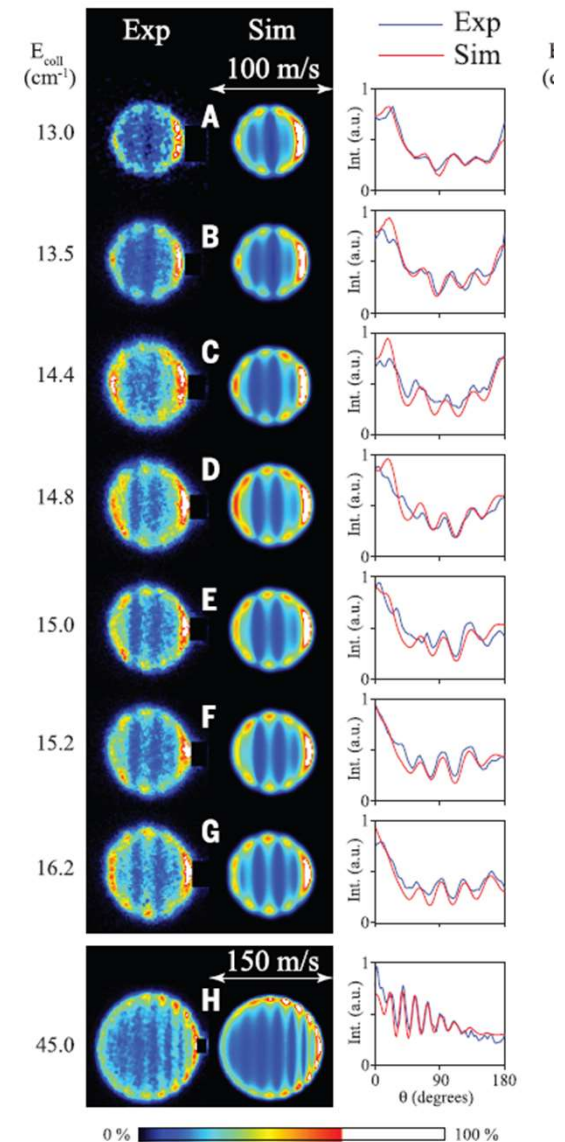
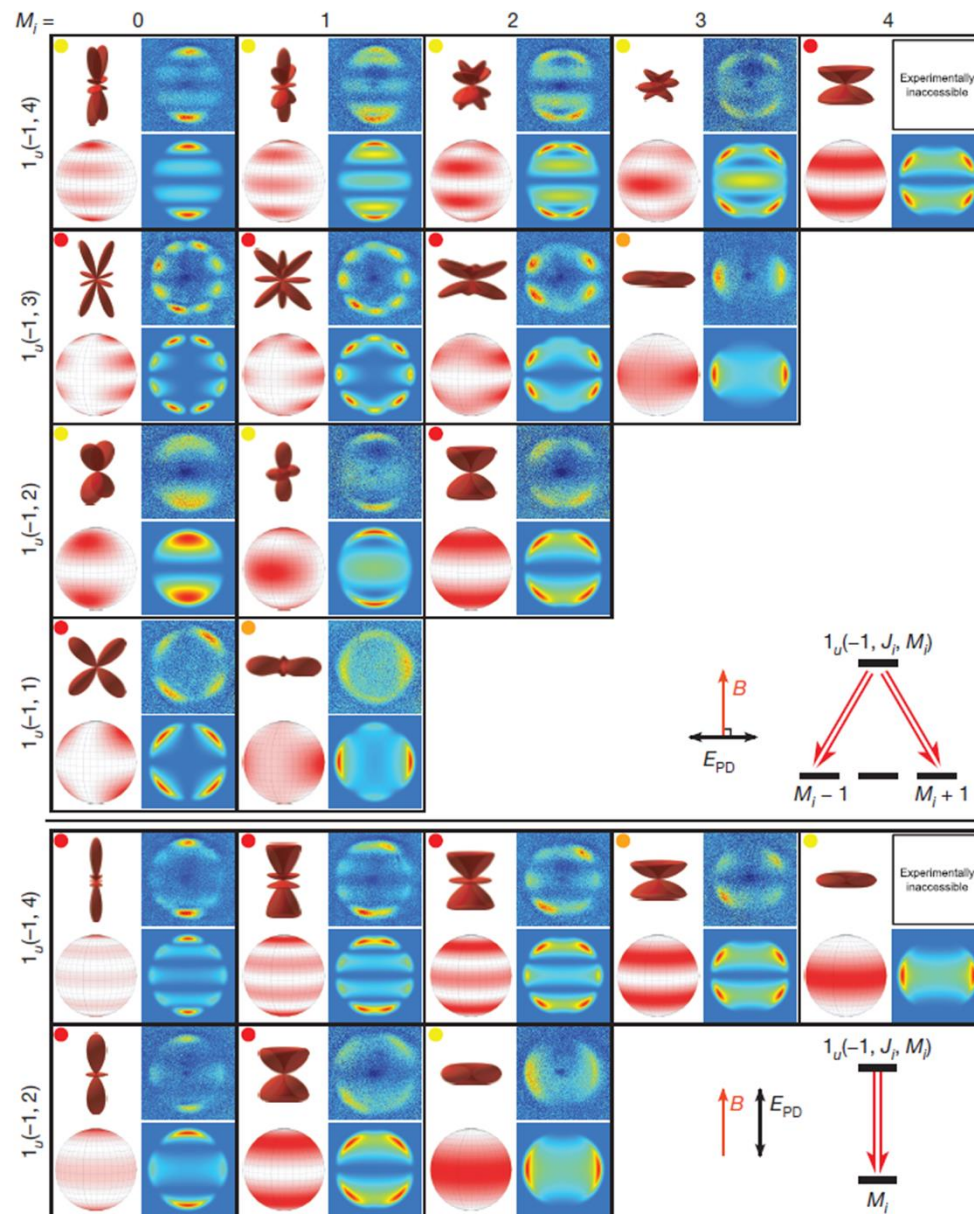


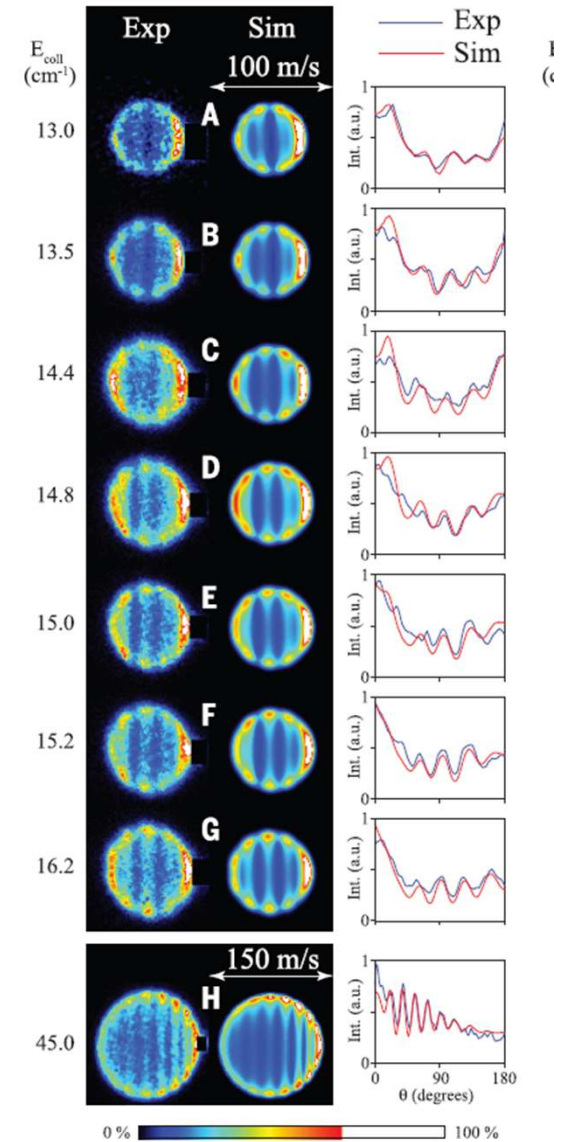
Figure 1 | Photodissociation of diatomic molecules in an optical lattice.

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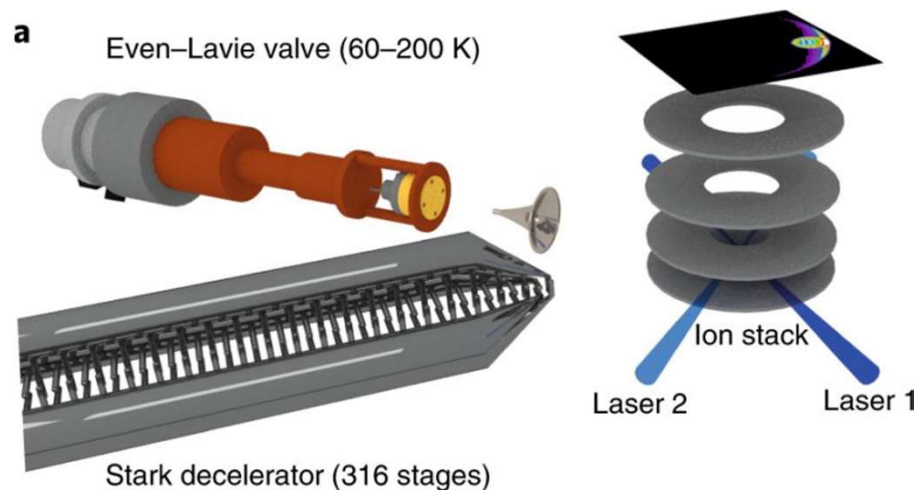


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Imaging resonances in low-energy NO-He inelastic collisions

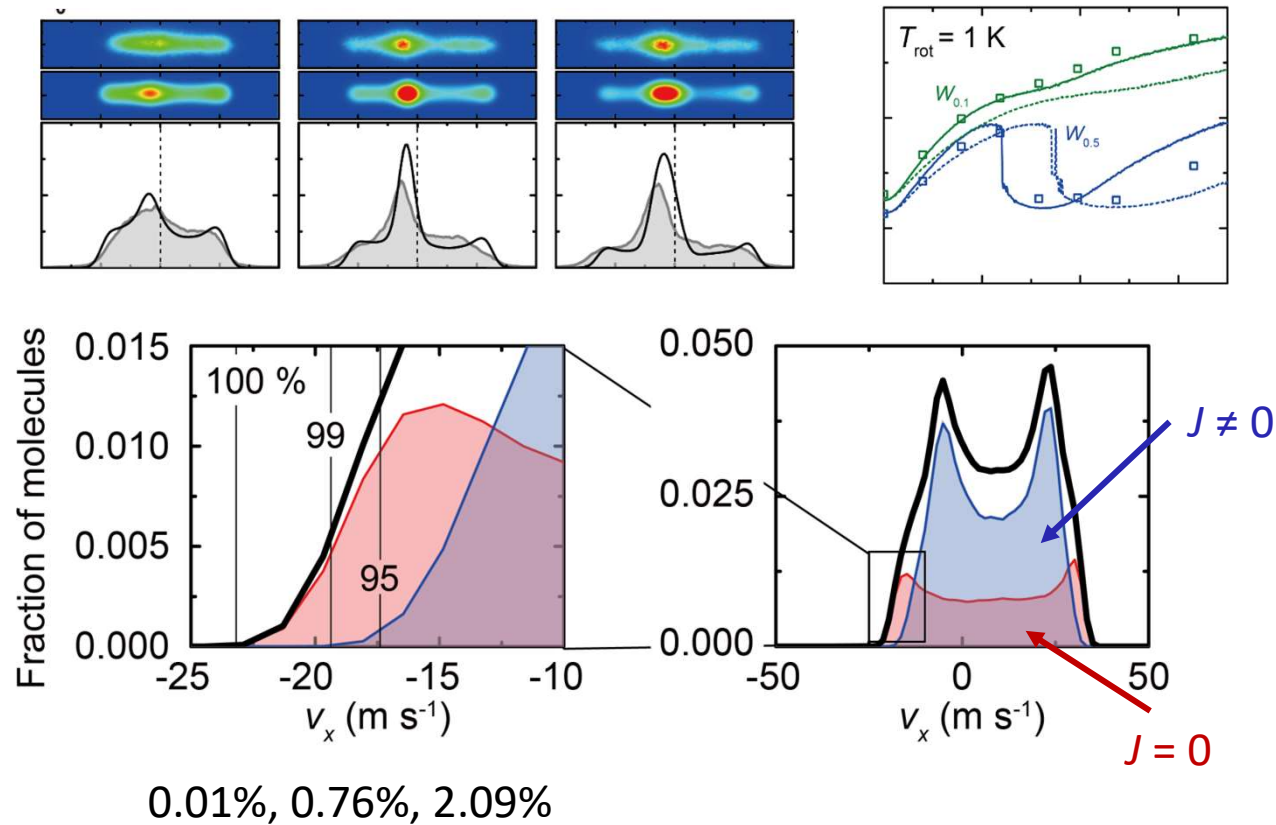
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Take home messages

State-dependent molecular alignment is crucial to interpretate optical dipole force



Literature survey is really important