Angular Momentum Orientation in Molecules using the AT-effect

J. T. Stahovich[1], B. A. Rowe[1], J. P. Huennekens[2], A. M. Lyyra[1], and E. H. Ahmed[1]

[1]Physics Department, Temple University, Philadelphia, PA 19122, USA

[2]Physics Department, Lehigh University, Bethlehem, Pennsylvania 18015, USA

TEMPLE UNIVERSITY

Abstract

We report an experimental demonstration of *state selective angular momentum orientation of nonpolar molecules* using dressed states created by a strong *cw* control laser. Our results show that the *M*-dependent Rabi frequency of the Autler-Townes effect for circular polarization allows for *M*-state selective molecular angular momentum orientation, where *M* is the projected angular momentum onto a lab fixed axis. Our results also show the square-root relationship between the splitting of adjacent *M*-levels and the power of the control laser, and thus the requirement for a strong control field to achieve *M*-state selectivity. The effect was observed using Li_2 molecules and a combination of left- and right-handed circularly polarized lasers.

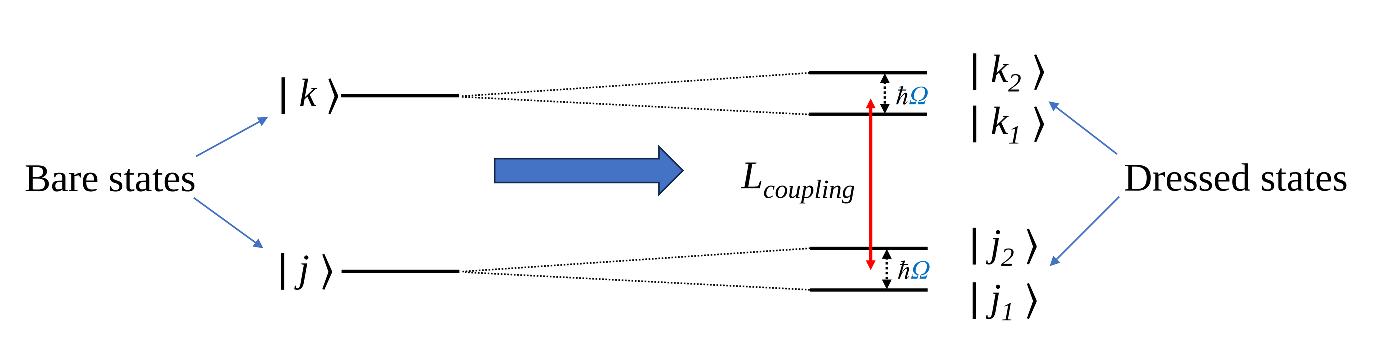
Autler-Townes (ac Stark) effect[1]

A dynamic Stark effect - corresponding to the case when a *strong* oscillating electric field (e.g., that of a laser) is tuned in resonance (or close) to the transition frequency of a given spectral line, resulting in a *change of the shape* of the absorption/emission spectra of that spectral line.

In this case, the alternating field has the effect of splitting the two bare transition states into doublets, or "dressed states", that are separated by the Rabi frequency,

$$\Omega = \frac{\mu E}{\hbar};$$

The bare molecular states are no longer eigenstates of the *molecule–field* Hamiltonian



Dressed state formalism[2]

Hamiltonian: $\mathcal{H} = H_{molecule} + H_{field} + H_{interaction}$,

- $H_{mol} = \sum_k \varepsilon_k |k\rangle\langle k|$,
  - here $H_{mol}|k\rangle = \varepsilon_k|k\rangle$
- $H_f = \hbar\omega_c(a^\dagger a + \frac{1}{2})$,
  - $a^\dagger$ and a are the photon creation and annihilation operators
  - ω_c is the coupling laser (L_c) frequency
- $H_{int} = \frac{1}{2}\sum_k \hbar\Omega_k (|l_k\rangle\langle m_k| + |m_k\rangle\langle l_k|)(e^{i\omega_k t} + e^{-i\omega_k t})$,
  - summation is over all external laser fields L_k
    - w/ frequency ω_k, e-field amplitude E_k and Rabi frequency $\Omega_k \equiv \frac{\mu E_k}{\hbar}$
  - $|l_k\rangle$ and $|m_k\rangle$ represent the l^{th} and m^{th} states coupled by laser L_k

Rabi frequency[3]

$$\Omega_k \equiv \frac{\mu E_k}{\hbar} = \frac{\boldsymbol{\mu}_{ij}(\boldsymbol{r}) \cdot \boldsymbol{E}_k(\boldsymbol{r},t)}{\hbar} = \frac{\langle \hat{\varepsilon}_I \cdot \vec{\mu}\rangle * E_k}{\hbar}$$

where $\langle \hat{\varepsilon}_I \cdot \vec{\mu}\rangle =$

$$\begin{cases} \langle n_f\Lambda_i|\mu_z|n_i\Lambda_i\rangle\langle\Omega_i J_f M_f|\alpha_I^z|\Omega_i J_i M_i\rangle; & \Delta\Lambda = \Delta\Omega = 0, \\ \frac{1}{2}\langle n_f(\Lambda_i \pm 1)|\mu^\pm|n_i\Lambda_i\rangle\langle(\Omega_i\pm1)J_f M_f|\alpha_I^\mp|\Omega_i J_i M_i\rangle; & \Delta\Lambda = \Delta\Omega = \pm1 \text{ (with } \Lambda_i, \Lambda_f > 0), \\ \frac{1}{\sqrt{2}}\langle n_f(\Lambda_i \pm 1)|\mu^\pm|n_i\Lambda_i\rangle\langle(\Omega_i\pm1)J_f M_f|\alpha_I^\mp|\Omega_i J_i M_i\rangle; & \Delta\Lambda = \Delta\Omega = \pm1 \text{ (with either } \Lambda_i = 0 \text{ or } \Lambda_f = 0), \end{cases}$$

$\alpha_I^j \equiv \hat{I}\cdot\hat{j}$ are *direction cosine operator components* linking unit vectors in the space fixed (*XYZ*) and body fixed (*xyz*) coordinate systems; $\alpha_I^\pm = \alpha_I^x \pm \alpha_I^y$ and $\mu^\pm = \mp\frac{1}{\sqrt{2}}(\mu_x \pm i\mu_y)$.

$$\langle\Omega_i J_f M_f|\alpha_I^z|\Omega_i J_i M_i\rangle, \langle(\Omega_i\pm1)J_f M_f|\alpha_I^\mp|\Omega_i J_i M_i\rangle \propto \begin{cases} M^2, & \Delta M = 0 \quad (linear\ polarization) \\ M, & \Delta M = \pm1\ (circular\ polarization) \end{cases}$$

and $E_k = \sqrt{\frac{2}{c\varepsilon_0}}\sqrt{\frac{8P_k}{\pi w_k^2}}$ is the electric field amplitude.

here P_k is the power, and w_k is the beam waist spot size of the k^{th} laser

$$\therefore \Omega_k \propto \sqrt{\frac{P_k}{w_k^2}} * \begin{cases} M^2, & \Delta M = 0 \quad (linear\ polarization) \\ M, & \Delta M = \pm1\ (circular\ polarization) \end{cases}$$

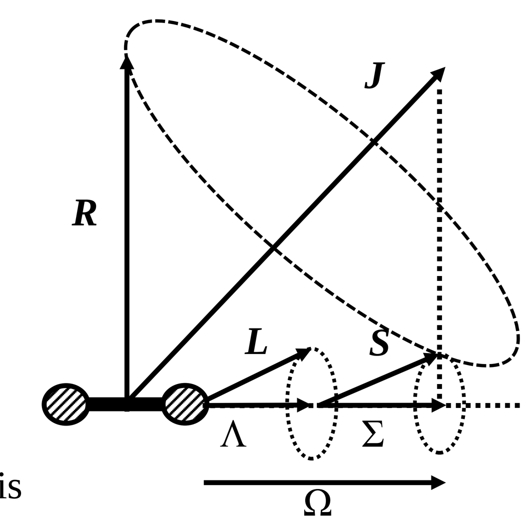
Orientation Factors[4] for ΔΩ = 0, ΔM = 0, ±1 (e.g. $^1\Sigma \leftarrow {}^1\Sigma$) transitions

$f(J'J)g_z h_I$	$\hat{\varepsilon}_I = \hat{Z}$, $M' = M$	$\hat{\varepsilon}_I = \frac{1}{\sqrt{2}}(\hat{X}+i\hat{Y})$, $M' = M+1$	$\hat{\varepsilon}_I = \frac{1}{\sqrt{2}}(\hat{X}-i\hat{Y})$, $M' = M-1$
$J' = J+1$	$\frac{\sqrt{(J+1)^2-M^2}}{\sqrt{(2J+1)(2J+3)}}$	$-\frac{\sqrt{(J+M+1)(J+M+2)}}{\sqrt{(2J+1)(2J+3)}}$	$\frac{\sqrt{(J-M+1)(J-M+2)}}{\sqrt{(2J+1)(2J+3)}}$
$J' = J$	0	0	0
$J' = J-1$	$\frac{\sqrt{J^2-M^2}}{\sqrt{(2J+1)(2J-1)}}$	$\frac{\sqrt{(J-M)(J-M-1)}}{\sqrt{(2J+1)(2J-1)}}$	$-\frac{\sqrt{(J+M)(J+M-1)}}{\sqrt{(2J+1)(2J-1)}}$

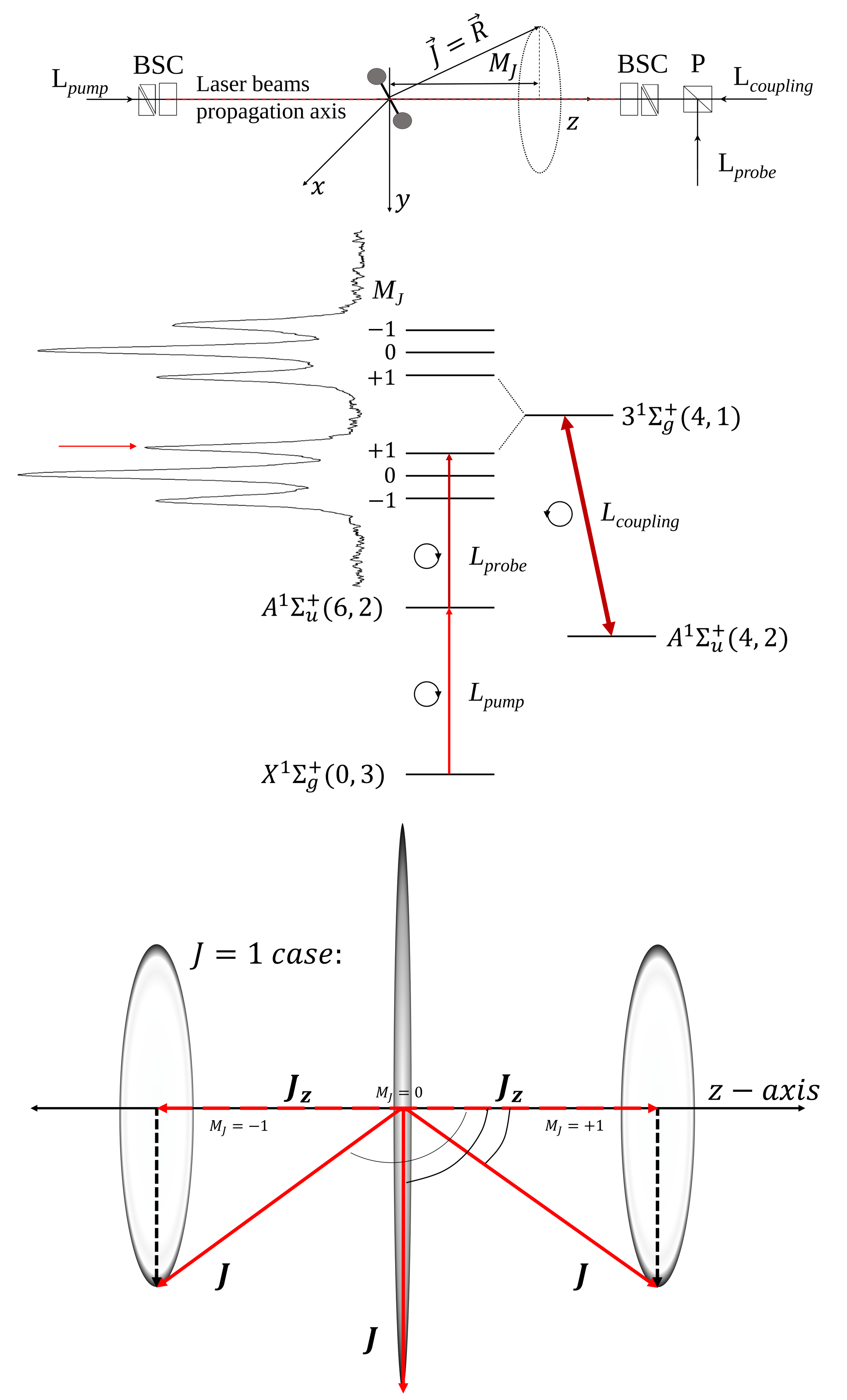
- **These are independent of *v* and proportional to the overall splitting.**

Angular Momenta:

L: electron orbital angular momentum
Λ: magnitude of projection of ***L*** onto internuclear axis
S: electron spin
Σ: magnitude of projection of ***S*** onto internuclear axis
R: nuclear rotational angular momentum
J = L + S + R **:** total angular momentum
Ω = | Λ + Σ |: magnitude of total angular momentum projection onto internuclear axis excluding nuclear spin



Excitation Scheme

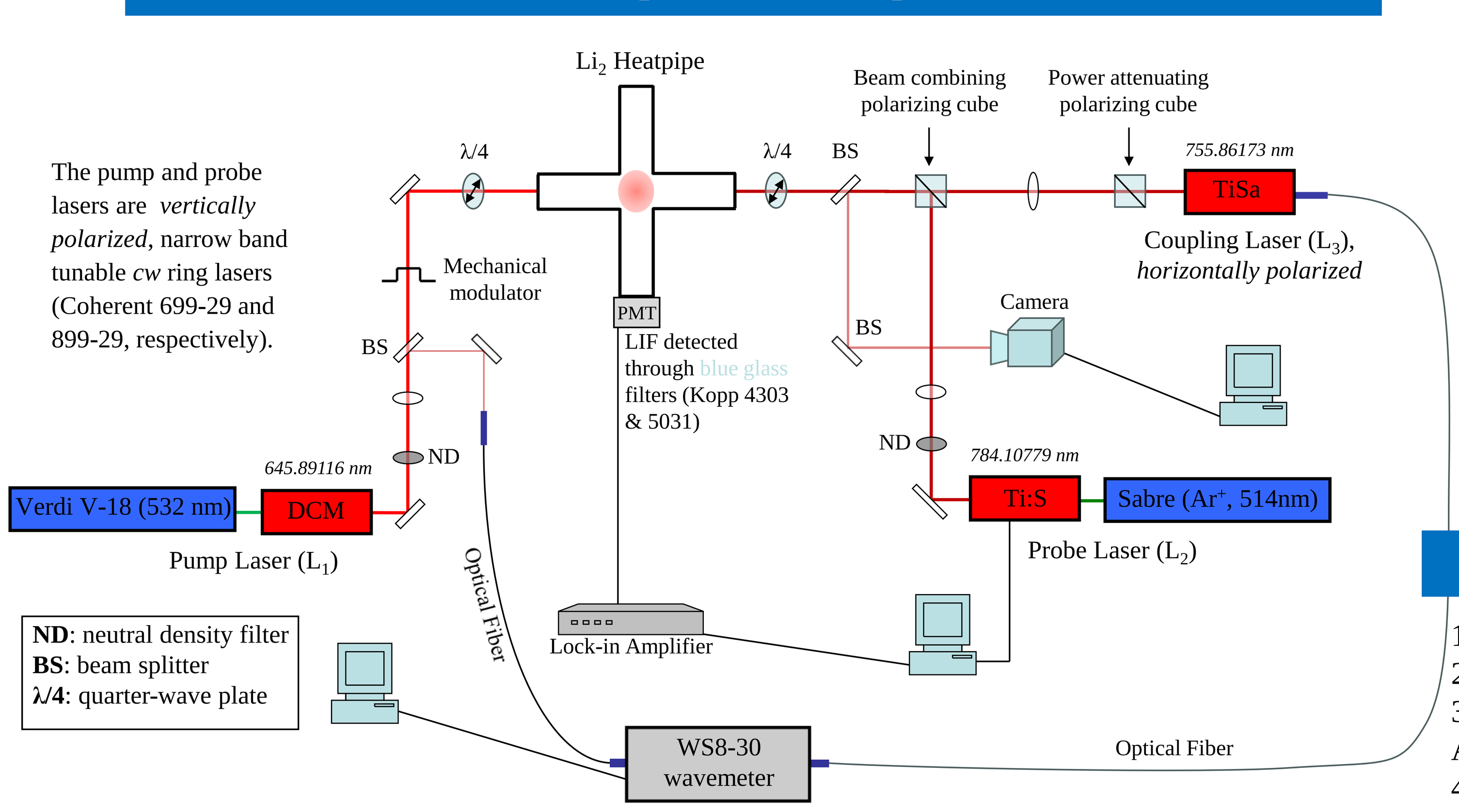


A strong (RHCP) coupling laser tuned to the $3^1\Sigma_g^+(4,1)$ - $A^1\Sigma_u^+(4,2)$ transition *splits both levels* into individual M_J components.

A weak (LHCP) pump laser tuned to $A^1\Sigma_u^+(6,2)$ - $X^1\Sigma_g^+(0,3)$ selectively *excites a narrow velocity group* of molecules from the ground state to in the intermediate state.

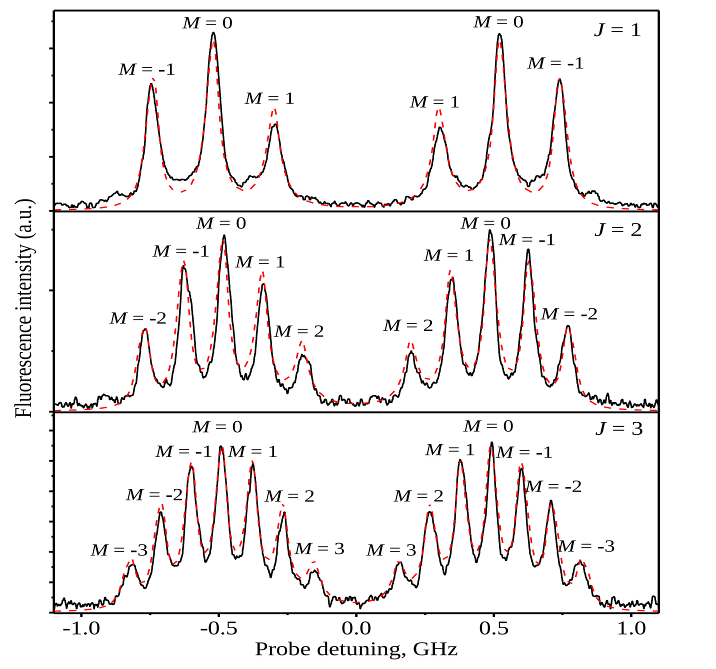
Tuning the weak (LHCP) probe laser to individual M_J components of the upper AT-split $3^1\Sigma_g^+(4,1)$ state **produces magnetic sublevel selective molecular orientation.**

Experimental setup

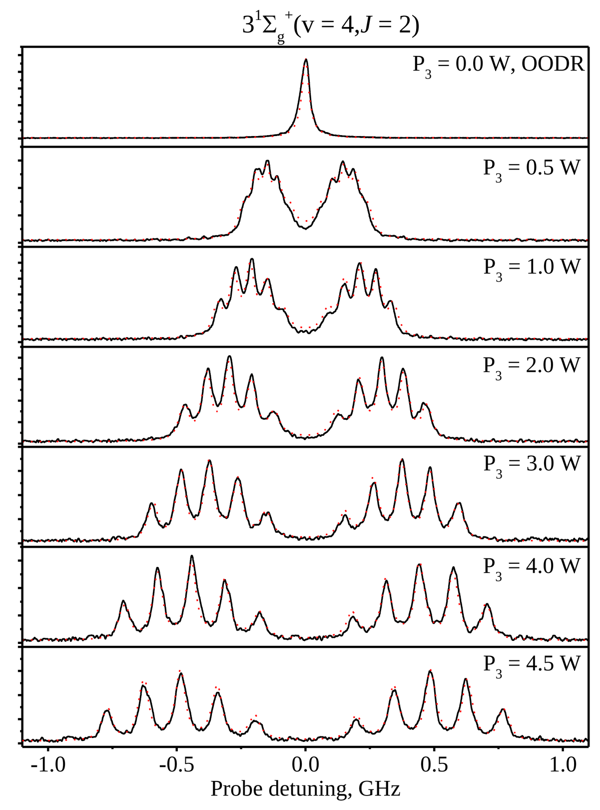


Probing higher *J* in $3^1\Sigma_g(v = 4)$

Experimentally fully resolved individual M_J levels for the lowest three rotational levels with non-zero angular momentum (J = 1, J = 2, and J = 3) of the Li_2 $3^1\Sigma_g^+(v = 4)$ vibrational level (**black**) compared with theoretical predictions (**red**).



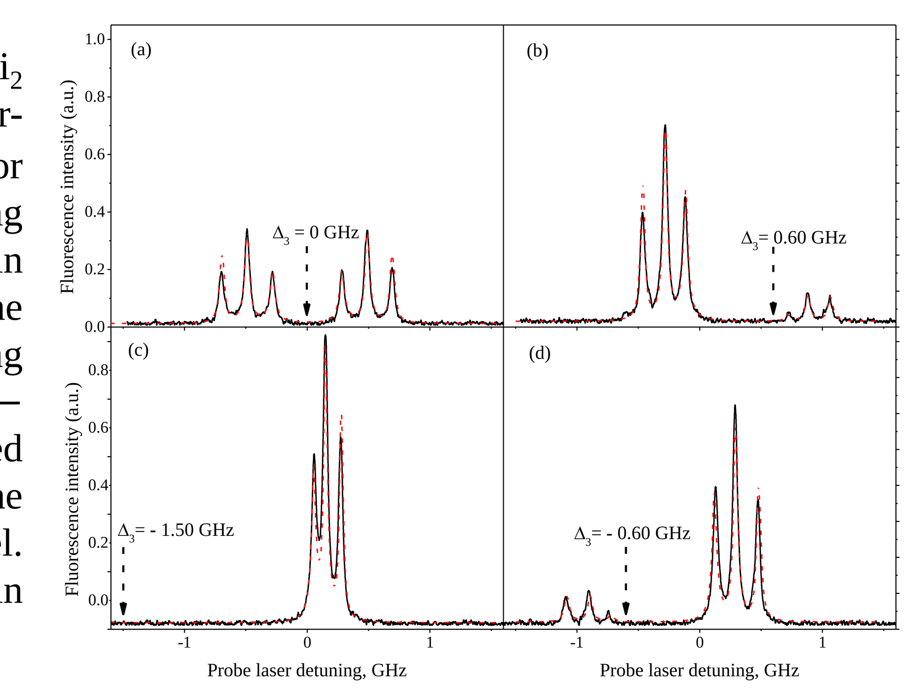
Dependence on coupling laser power



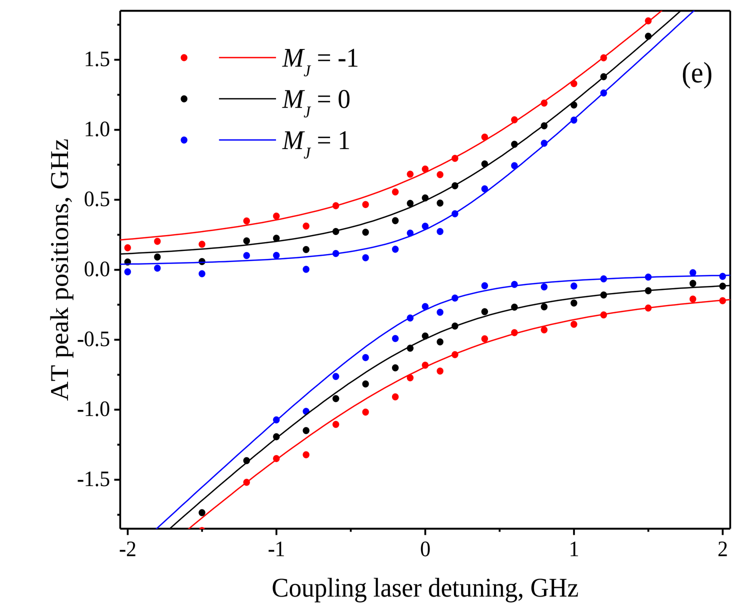
Dependence of the splitting of the M_J levels on the coupling laser power illustrated with spectra for the J = 2 rotational level of the Li_2 $3^1\Sigma_g^+(v = 4)$ state (**black**) compared with theoretical predictions (**red**). Similar behavior is observed also for the other rotational levels.

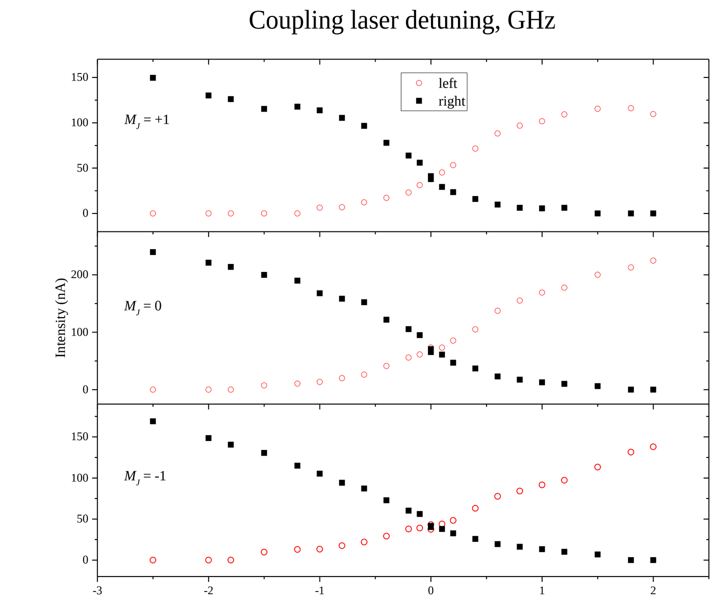
Coupling laser off-resonance

Comparison of the Li_2 $3^1\Sigma_g^+(v = 4, J = 1)$ Autler-Townes line shapes for selected values of coupling laser detuning is presented in panels (a) though (d). The positions of the coupling laser detuning from the |3⟩ − |4⟩ resonance are indicated with red arrows on the horizontal axis in each panel. Theoretical predictions are in red.



Coupling laser off-resonance





AT peak positions (relative to the OODR condition - *top*) and relative intensities (*bottom*) plotted as a function of the coupling laser detuning, Δ_3. The experimental measurements for the $M_J = -1, 0,$ and $+1$ levels are plotted with solid circles, while the solid lines represent the behavior predicted by $\Delta_{AT,M} = \frac{\Delta_3}{2} \pm \frac{1}{2}(\Delta_3^2 + \Omega_{3,M}^2)^{1/2}$ of a simple two-level atom model. The upper (lower) data sets in the top panel and black squares (red circles) on the bottom panel correspond to the AT-split peaks that are shifted higher (lower) in energy than the OODR position.

References and support

1. S.H. Autler and C.H. Townes, Phys. Rev. **100**, 703 (1955).
2. C. Cohen-Tannoudji et al., Wiley-VCH, Weinheim (2004).
3. H. Lefebvre-Brion and R.W. Field, Academic Press, Amsterdam (2004).
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