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Polarization Spectroscopy and Collisions in NaK

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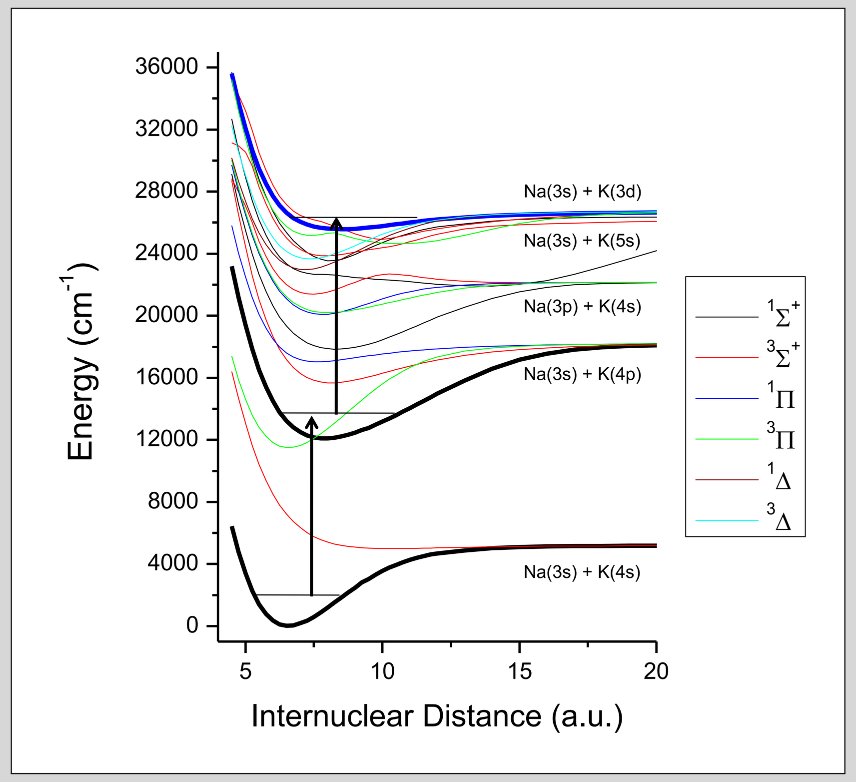
Abstract

We report current work to study transfer of population and orientation in collisions of NaK molecules with argon and potassium atoms using polarization labeling (PL) and laser-induced fluorescence (LIF) spectroscopy. In the PL experiment, a circularly polarized pump laser excites a specific NaK $A^1\Sigma^+(v=16, J) \leftarrow X^1\Sigma^+(v=0, J\pm1)$ transition, creating an orientation (non-uniform M_J level distribution) in both levels. The linearly polarized probe laser is scanned over various $3^1\Pi(v=8, J'\pm1)\ A^1\Sigma^+(v=16, J')$ transitions. The probe laser passes through a crossed linear polarizer before detection, and signal is recorded if the probe laser polarization has been modified by the vapor (which occurs when it comes into resonance with an oriented level). In addition to strong direct transitions ($J' = J$), we also observe weak collisional satellite lines ($J' = J \pm n$ with n = 1, 2, 3, ...) indicating that orientation is transferred to adjacent rotational levels during a collision. An LIF experiment (with linear polarized pump and probe beams) gives information on the collisional transfer of population. From these data, cross sections for both processes can be determined. We experimentally distinguish collisions of NaK with argon atoms from collisions with alkali atoms.

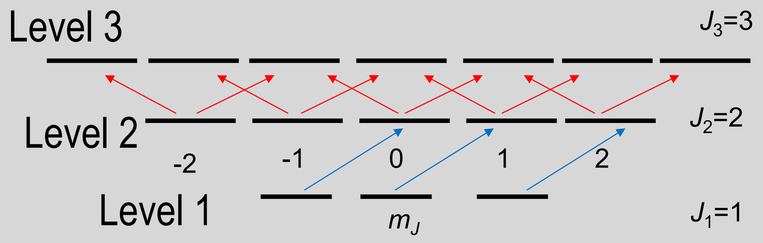
NaK



Theoretical potential energy curves for all $^1\Sigma^+$, $^3\Sigma^+$, $^1\Pi$, $^3\Pi$, $^1\Delta$, and $^3\Delta$ electronic states up to the Na(3s) + K(3d) asymptote

Polarization Spectroscopy

Circularly polarized pump beam induces an uneven m_J population in both pump transition energy levels, making the gas birefringent for a linearly polarized probe beam sharing one or both of the pump transition levels



Equation for polarization spectroscopy lineshape:

Background Terms

$$I = I_0 e^{-\alpha L - \alpha_w}\left[\xi + (\theta')^2 + \left(\frac{1}{4}\Delta\alpha_w\right)^2 +\right.$$

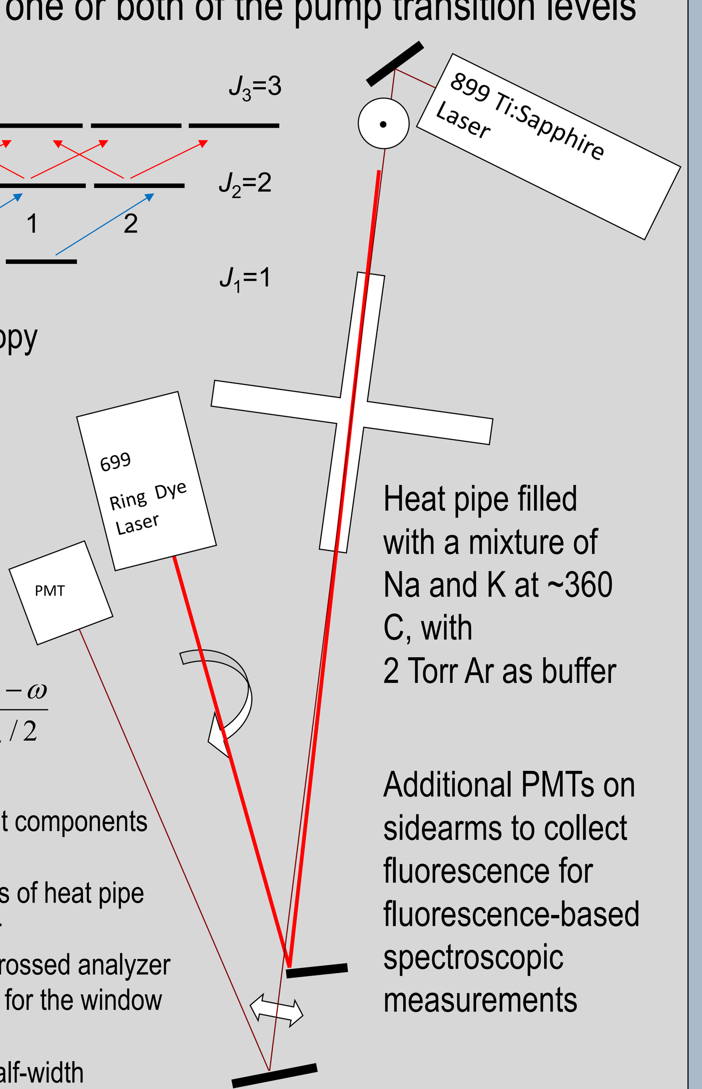
Lorentzian terms

$$\frac{1}{8}\frac{\Delta\alpha_0 \Delta\alpha_w L}{1+x^2} + \left(\frac{1}{4}\frac{\Delta\alpha_0 L}{1+x^2}\right)^2 +$$

Dispersion terms

$$\left.\frac{\theta'\Delta\alpha_0 L x}{1+x^2} + \left(\frac{1}{2}\frac{\Delta\alpha_0 L x}{1+x^2}\right)^2\right] \qquad x = \frac{\omega_0 - \omega}{\gamma_s/2}$$

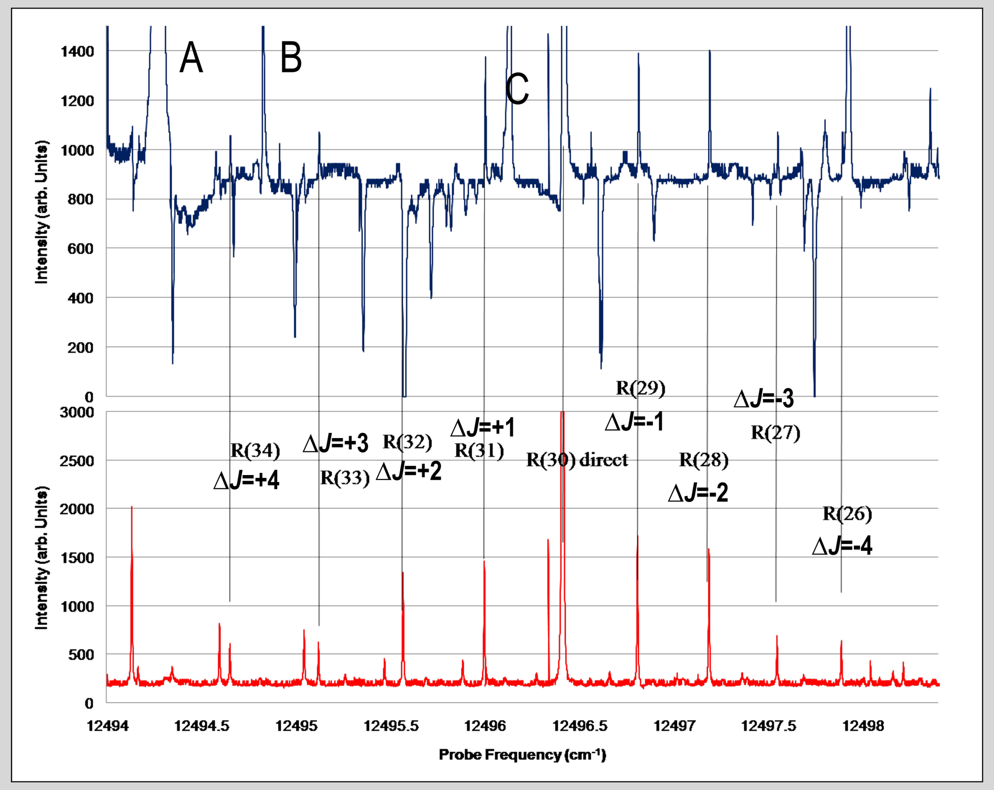
$\Delta\alpha_0$ = Difference of absorption for left/right components of probe beam in vapor
$\Delta\alpha_w$ = Difference of absorption in windows of heat pipe
L = Interaction length of beams with vapor
ζ = Residual transmission of completely crossed analyzer
ϑ' = Crossing angle of analyzer corrected for the window birefringence
x = Laser detuning divided by transition half-width



Heat pipe filled with a mixture of Na and K at ~360 C, with 2 Torr Ar as buffer

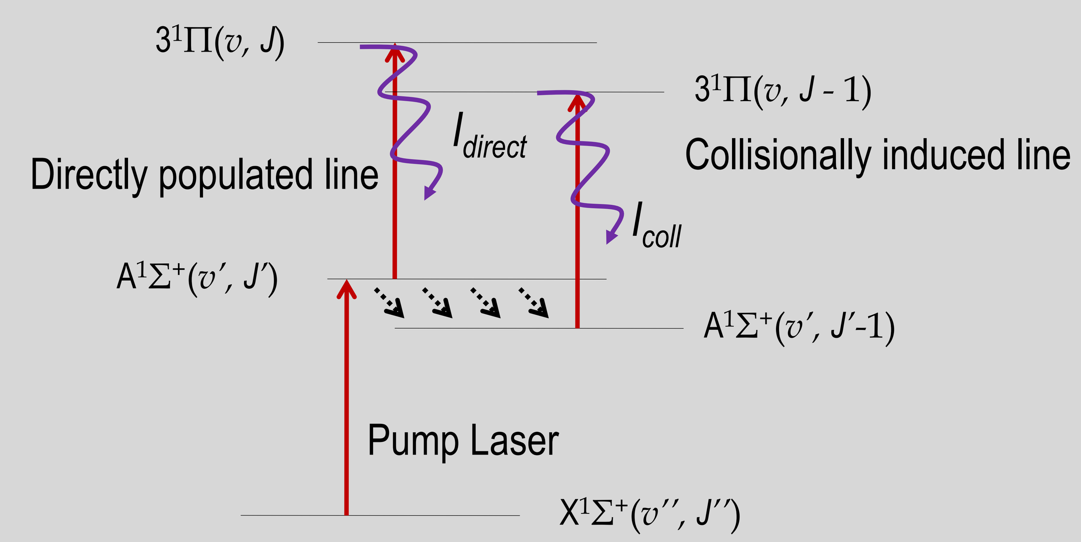
Additional PMTs on sidearms to collect fluorescence for fluorescence-based spectroscopic measurements

Polarization Spectra



Bottom (Red) – Laser-induced Fluorescence Spectroscopy
Top (Blue) – Polarization Spectroscopy
A : A(5,28) ⟵ X(0,29) B: A(16,28) ⟶ X(7,29) C: A(5,27) ⟵ X(0,28)

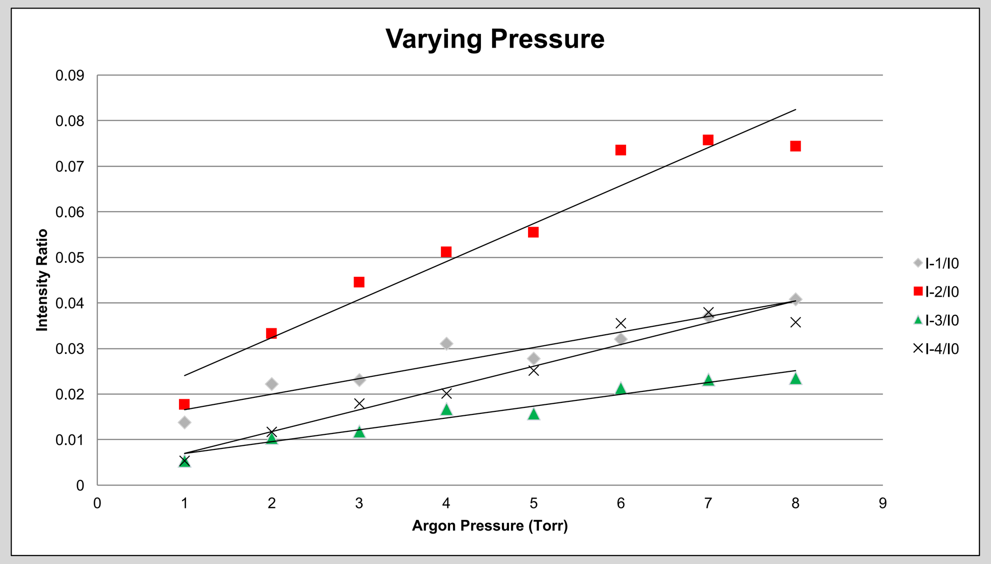
Transfer of Population (Fluorescence Spectroscopy)



Population transfer rates can be modeled using

$$\left(\frac{I_{coll}}{I_{direct}}\right)_{fluor} = \frac{n_{coll}}{n_{direct}} = \frac{k_{Ar}^{pop}[Ar] + k_K^{pop}[K] + other\ species}{\Gamma}$$

[Ar] – Argon density
[K] – Potassium density



Slope yields k_{Ar}, intercept yields k_K

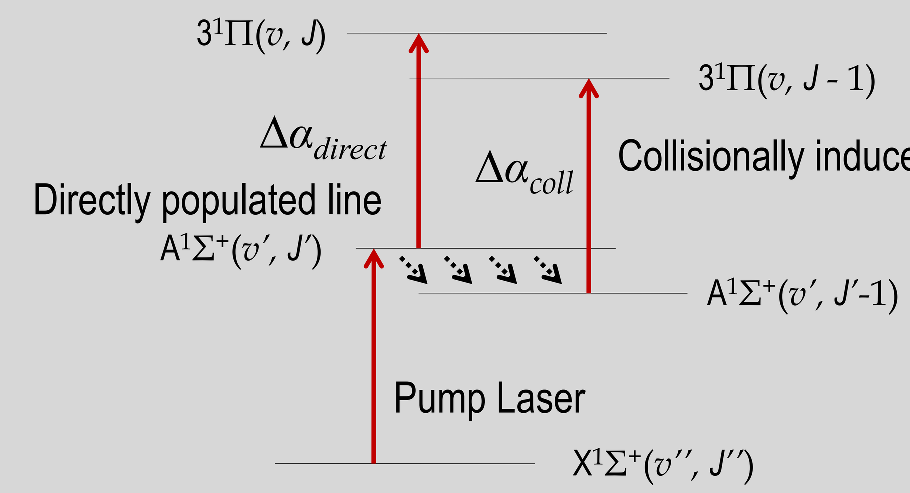
Preliminary values for population transfer rates using above data:

Collisions with Argon atoms seem to show a propensity toward even-numbered ΔJ transitions

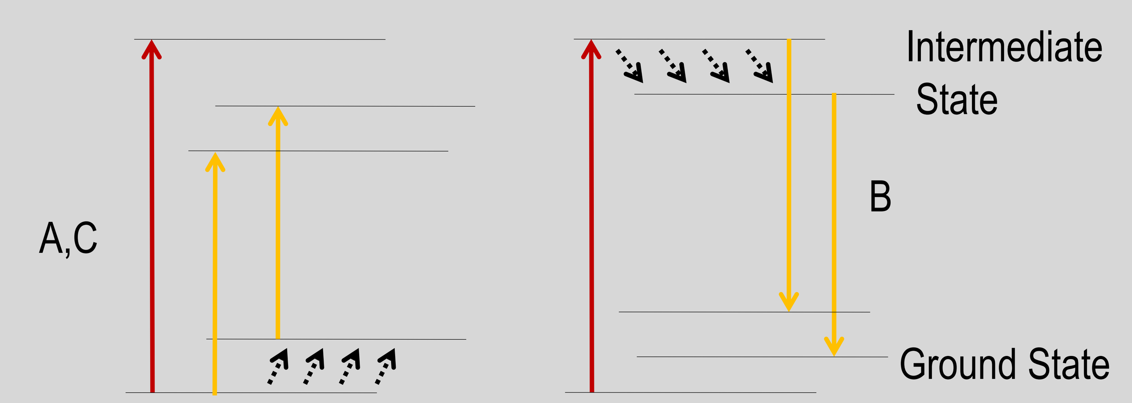
ΔJ (collision)	k_{Ar} (cm³ s⁻¹)	k_K (cm³ s⁻¹)
-1	9.8×10^{-12}	2.6×10^{-11}
-2	2.4×10^{-11}	3.2×10^{-11}
-3	7.5×10^{-12}	8.8×10^{-12}
-4	1.4×10^{-11}	4.4×10^{-12}

Transfer of Orientation (Polarization Spectroscopy)

In order to use polarization spectroscopy on collisionally populated level, collision must (at least partially) preserve m_J orientation



In addition to two-step excitation, can also get polarization signal due to ground-state and induced transitions



Red: Pump
Yellow: Probe

Polarization signal depends on the difference of absorption between left and right circular polarization components of linearly polarized probe beam

$$\Delta\alpha_0 = \alpha_+ - \alpha_-$$

Can model this using:

$$\Delta\alpha_0 = \sum_{m_i}\left(f_{J_f,J_i}^{m_i+1,m_i} - f_{J_f,J_i}^{m_i-1,m_i}\right)n_{m_i}$$

Where f is defined by:

$$\left|\left\langle \beta_f, J_f, m_f \left|\vec{r}\right| \beta_i, J_i, m_i \right\rangle\right|^2 = f_{J_f,J_i}^{m_f,m_i}\left|\left\langle \beta_f, J_f \left\|r\right\| \beta_i, J_i \right\rangle\right|^2$$

Difference in f factors is proportional to m

$$f_{J_f,J_i}^{m_i+1,m_i} - f_{J_f,J_i}^{m_i-1,m_i} \propto m_i$$

So the ratio between polarization signal for collisional line and main line gives

$$\left(\frac{I_{coll}}{I_{direct}}\right)_{pol} = \frac{\Delta\alpha_{coll}}{\Delta\alpha_{direct}} \approx \frac{\sum_{m_{coll}} m_{coll} n_{m_{coll}}}{\sum_{m_{direct}} m_{direct} n_{m_{direct}}}$$

Thus the ratio of ratios for polarization and fluorescence-based spectroscopy gives the ratio of orientations of collisional and direct levels

$$\frac{\left(\frac{I_{coll}}{I_{direct}}\right)_{pol}}{\left(\frac{I_{coll}}{I_{direct}}\right)_{fluor}} \approx \frac{\left(\frac{\sum_{m_{coll}} m_{coll} n_{m_{coll}}}{\sum_{m_{coll}} n_{m_{coll}}}\right)}{\left(\frac{\sum_{m_{direct}} m_{direct} n_{m_{direct}}}{\sum_{m_{direct}} n_{m_{direct}}}\right)} = \frac{\langle m_{coll}\rangle}{\langle m_{direct}\rangle}$$

Preliminary results

ΔJ (collision)	Ratio of ratios defined at left
+4	0.37
+3	0.36
+2	Insufficient data
+1	0.27
-1	0.25
-2	0.27
-3	0.31
-4	0.33

Thus ~1/3 of orientation is preserved

Density Matrix Model

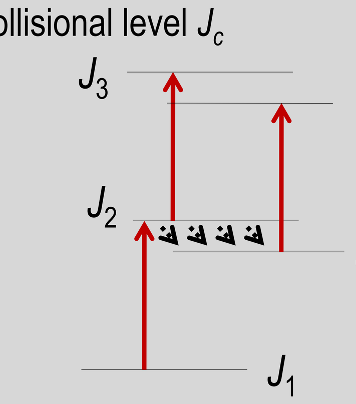
Necessary to develop a model for the transfer of orientation in a collision between the NaK molecule and an Argon or Potassium atom

We need to solve the density matrix equations of motion for the system of energy levels in question

$$\dot{\rho}_{nm} = -\frac{i}{\hbar}\sum_k\left(H_{nk}\rho_{km} - \rho_{nk}H_{km}\right) + relaxation\ terms$$

Diagonal elements represent population densities, off-diagonal element represent coherences between levels

A computer program has been developed which solves the density matrix for a general case of 4 energy levels J_1, J_2, and J_3 and a collisional level J_c



Computer model includes:

- Laser coherence terms
- Fluorescence into and out of various levels
- Transit relaxation
- Collisional excitation transfer
- Adjustable m_J dependent collisional rate
- Additional "dump" levels representing other ground and intermediate state levels

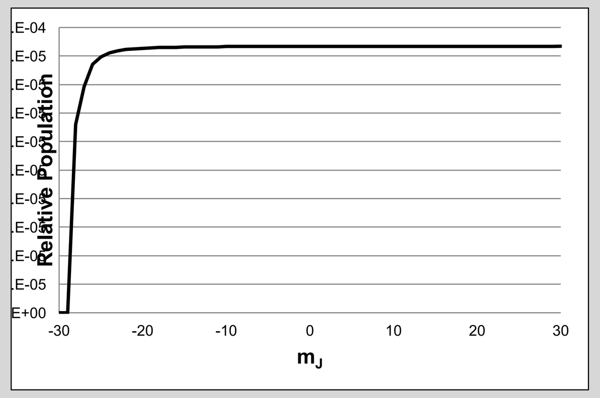
Model input includes collisional population transfer rates (k_{Ar} and k_K), along with various other critical experimental parameters (laser power, beam width, temperature, pressure, decay rates, etc.) and outputs m_J distribution of level 2 and collisional level. Input transfer rates are varied until agreement with experiment is reached

Current Computer Model Results

Calculation for m_J population distribution of directly populated level, given typical experimental parameters

Anisotropic distribution of magnetic sublevels is clearly shown

Probe beam is assumed to be weak compared to pump beam (consistent with polarization spectroscopy parameters)



Modeling of Collisional Orientation Transfer

It is necessary to develop an empirical model for the transfer of population between individual magnetic sublevels of directly populated level and collisionally populated level

- Adjustment of model must not affect total population transfer (would alter calculated fluorescence signal)
- Model must not create an additional alignment in the collisional level
- 2 models currently proposed

$$R(m_{direct}, m_{coll}) = \frac{e^{\frac{-|m_{direct}-m_{coll}|}{\Delta m_0}}}{\sum_{m_{coll}} e^{\frac{-|m_{direct}-m_{coll}|}{\Delta m_0}}}$$

$$R(m_{direct}, m_{coll}) = \begin{cases} f & \text{for } \Delta m = 0 \\ (1-f) & \text{for } \Delta m \neq 0 \end{cases}$$

Future Plans

Future plans for research include

- Find an appropriate model for the change in orientation during a collision
- Take precise data under more controlled conditions as a function of temperature and pressure in order to get more precise values for k_{Ar} and k_K
- Adjust collisional rate model in order to obtain observed experimental orientations of collisional levels
- Look for vibrational transitions in collisions
- Collaborating group at Temple U. has observed ground state collisional lines for large ΔJ in polarization spectroscopy on the Rb_2 molecule
    - Investigate these further