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Collisional Transfer of Population and Orientation in NaK

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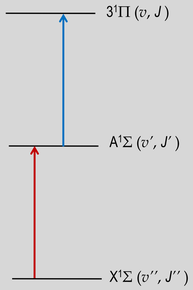
Abstract

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 $A^1\Sigma^+(v'=16, J') \leftarrow X^1\Sigma^+(v''=0, J' \pm 1)$ transition, creating an orientation (non-uniform m_J level distribution) in both levels. The linearly polarized probe laser is scanned over various $3^1\Pi(v, J'\pm1) \leftarrow A^1\Sigma^+(v''=16, J')$ 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). Using both spectroscopic methods, analysis of weak collisional satellite lines adjacent to these directly populated lines, as a function of argon buffer gas pressure and cell temperature, allows us to discern separately the effects collisions with argon atoms and potassium atoms have on the population and orientation of the molecule. In addition, code has been written which provides a theoretical analysis of the process, through a solution of the density matrix equations of motion for the system.

Background

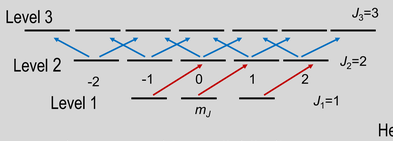


- We use Optical-Optical Double Resonance (OODR) to study the NaK molecule in both Polarization Labeling and Laser-Induced Fluorescence spectroscopy
- Quantitative study of collisional population and orientation transfer
- Separately study the effects of collisions with Argon vs. Potassium

Polarization Spectroscopy

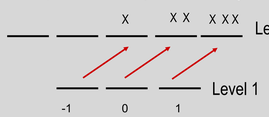
Due to asymmetric m_J distribution created by circular polarized pump beam (Δm = +1 for right circular polarization), vapor is birefringent for any probe that shares upper, lower, or both levels of pump transition

Note: linearly polarized probe can be thought of as equal components of left and right circular polarization. (Δm = -1 and +1)



Red: Pump
Blue: Probe

Pump creates asymmetric m_J distribution



Left circular polarization, Δm_J = -1
Right circular polarization, Δm_J = +1

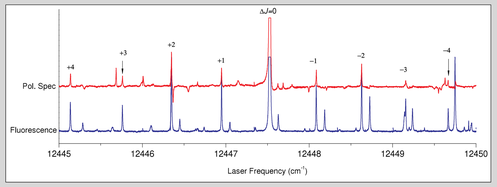
Xs show relative population in m_J sublevel

Left and right circular components of probe are unequally absorbed

Heat pipe filled with a mixture of Na and K at 280-380 °C, with 0.5-8 torr Ar as buffer

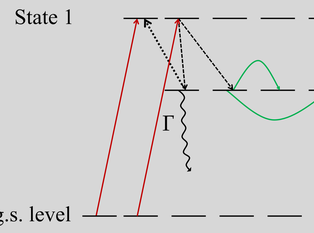
Additional PMTs on sidearms to collect fluorescence for fluorescence-based spectroscopic measurements

Comparison Spectra



Comparison excitation scan showing both polarization spectroscopy (top) and laser-induced fluorescence spectroscopy (bottom)
Direct pump-probe transition is
$3^1\Pi$ (7, 29) ← $A^1\Sigma$ (16, 30) ← $X^1\Sigma$ (0, 29)
Collisionally populated and oriented levels $A^1\Sigma$ (16, 30 + ΔJ) are labeled by ΔJ

Collisional Transfer

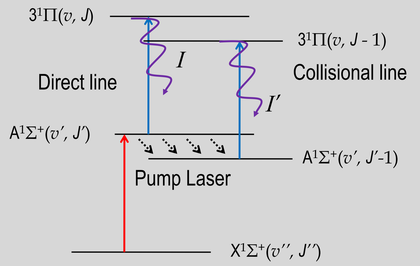


Collisional effects on population and orientation can be divided into two types:

1. Population and orientation are transferred from state 1 to state 2
2. Population and orientation are lost from state 2 due to subsequent collisions and radiative decay

Empirical models have been developed to study population and orientation transfer, separating the effects of argon and potassium

Transfer of Population (Fluorescence Spectroscopy)



Ratio of fluorescence intensities is roughly equal to the ratio of population densities in the collisionally and directly populated level

$$R_f = \frac{I'}{I} = \frac{I_2^F}{I_1^F} = \frac{n_2}{n_1}$$

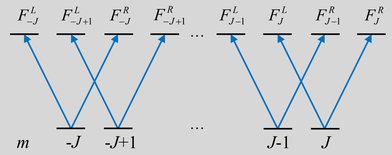
Using a rate equation approximation, we model this ratio as

$$\frac{n_2}{n_1} = \frac{k_{\mathrm{Ar}}^{\Delta J} n_{\mathrm{Ar}} + k_{\mathrm{K}}^{\Delta J} n_{\mathrm{K}}}{\Gamma + k_{\mathrm{Ar}}^{\mathrm{Q}} n_{\mathrm{Ar}} + k_{\mathrm{K}}^{\mathrm{Q}} n_{\mathrm{K}}}$$

Here $k^{\Delta J}$ is the rate of transfer of population from level 1 to the level characterized by ΔJ and k^Q is the total collisional quenching rate

Intensity ratios are fitted as a function of argon and potassium density in order to obtain population transfer rates

Transfer of Orientation (Polarization Spectroscopy)



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

Can model this using: $\mathrm{Signal} \propto \sum \mathrm{Right\ Absorptions} - \sum \mathrm{Left\ Absorptions}$

$$= \sum_{m=-J'}^{+J'} n_m F_m^R - \sum_{m=-J'}^{+J'} n_m F_m^L$$

$$= \sum_{m=-J'}^{+J'} n_m \left(F_m^R - F_m^L\right) = f(J, J') \sum_{m=-J'}^{+J'} m n_m = f(J, J')\langle m\rangle \sum_{m=-J'}^{+J'} n_m$$

In this expression, F factors are relative absorption coefficients:

$$\left|\langle \beta_1, J_1, m_1 | \vec{r} | \beta_2, J_2, m_2\rangle\right|^2 = F_{J_1,J_2}^{m_1,m_2} \left|\langle \beta_1 \| r \| \beta_2\rangle\right|^2$$

Evaluation of difference in F factors yields $F_m^R - F_m^L = f(J, J')m$

Thus the ratio of polarization spectroscopy signal strengths for the collisional and direct line are roughly equal to the ratio of orientation multiplied by the population of the levels. We can use a rate equation model to solve for the ratio of the intensities of the collisional and direct line:

$$R_p = \frac{\langle m_2\rangle n_2}{\langle m_1\rangle n_1} = \frac{k_{\mathrm{Ar}}^{\Delta J,\mathrm{O}}}{\Gamma} n_{\mathrm{Ar}} + \frac{k_{\mathrm{K}}^{\Delta J,\mathrm{O}}}{\Gamma} n_{\mathrm{Ar}}}{1 + \frac{g_{\mathrm{Ar}}}{\Gamma} n_{\mathrm{Ar}} + \frac{g_{\mathrm{K}}}{\Gamma} n_{\mathrm{K}}}$$

Here $k^{\Delta J,O}$ is the rate of transfer of orientation from level 1 to the level characterized by ΔJ, and g is the total collisional rate of destruction of orientation. Separating out the population and orientation dependence yields

$$k_{\mathrm{Ar}}^{\Delta J,\mathrm{O}} = k_{\mathrm{Ar}}^{\Delta J}(1 - f_{\mathrm{Ar}}^{\Delta J}) \qquad g_{\mathrm{Ar}} = k_{\mathrm{Ar}}^{\mathrm{Q}} + g'_{\mathrm{Ar}}$$

$$k_{\mathrm{K}}^{\Delta J,\mathrm{O}} = k_{\mathrm{K}}^{\Delta J}(1 - f_{\mathrm{K}}^{\Delta J}) \qquad g_{\mathrm{K}} = k_{\mathrm{K}}^{\mathrm{Q}} + g'_{\mathrm{K}}$$

$f_{Ar,K}$ represents the fraction of orientation destroyed in the initial collision with either Ar or K, and g' is the rate of destruction of orientation by subsequent collisions that do not change J. g' is small compared to k^Q, so we approximate

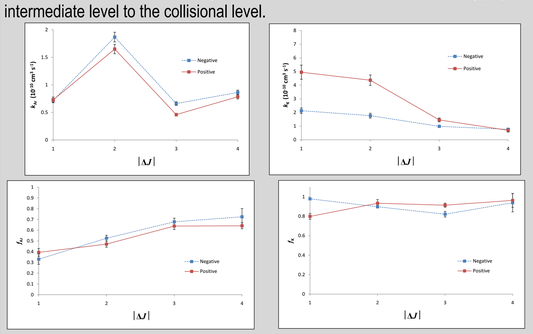
$$g'_{\mathrm{Ar}} = \frac{1}{2}\left(f_{\mathrm{Ar}}^{-1} k_{\mathrm{Ar}}^{-1} + f_{\mathrm{Ar}}^{+1} k_{\mathrm{Ar}}^{+1}\right)$$

$$g'_{\mathrm{K}} = \frac{1}{2}\left(f_{\mathrm{K}}^{-1} k_{\mathrm{K}}^{-1} + f_{\mathrm{K}}^{+1} k_{\mathrm{K}}^{+1}\right)$$

A fit of intensity ratios obtained from fluorescence and polarization spectroscopy, using our empirical formulas R_f and R_p, as functions of Ar or K number density has been carried out , for $\Delta J = \pm1, \pm2, \pm3, \pm4$

Fitting Results

The parameters k_{Ar} , k_K , f_{Ar} , and f_K give us information on the population transferred, as well as the orientation destroyed, in the initial collision from the directly populated intermediate level to the collisional level.



- Only $\Delta J = \pm 2, \pm 4, \ldots$ transitions observed in homonuclear molecules (like Rb_2)
- k_{Ar} values are much larger for even numbered collisional transitions than for odd numbered transitions
- k_K values do not seem to show such a propensity
- f values show that collisions with potassium atoms are more likely to destroy angular momentum orientation than collisions with argon atoms

$$k_{\mathrm{Ar}}^{\mathrm{Q}} = 1.16 \pm .07 \times 10^{-9}\ \mathrm{cm^3\ s^{-1}}$$

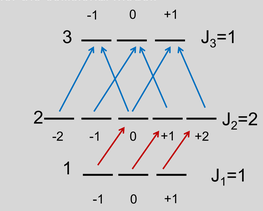
$$k_{\mathrm{K}}^{\mathrm{Q}} = 5.3 \pm .6 \times 10^{-9}\ \mathrm{cm^3\ s^{-1}}$$

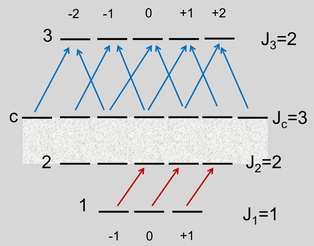
Density Matrix Model

To completely describe a system of quantum mechanical energy levels in a statistical manner, it is necessary to solve the density matrix equations of motion

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

A diagonal element ρ_{nn} represents the population of sublevel n, while an off-diagonal element ρ_{nm} represents the laser-induced coherence between sublevels n and m. A computer code has been written which solves the density matrix equations of motion for either three coupled angular momentum states for the direct probe model, or four states for the collisional model.





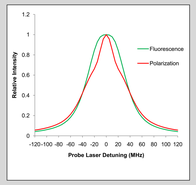
Model Includes

- Laser coherence terms
- Fluorescence into and out of various levels
- Transit relaxation
- Additional "dump" levels representing other ground/intermediate levels
- Velocity group and laser detuning loops

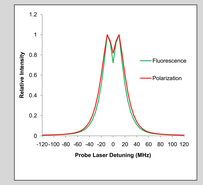
Collisional model is equipped to incorporate any theoretical model for collisional transfer of population and orientation linking states "2" and "c"

Sample Computer Model Results

Detuning Loop Calculations

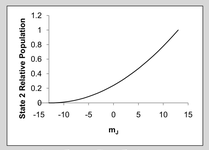


High Probe Power

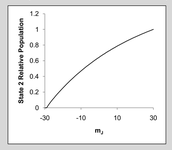


Low Probe Power

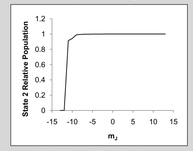
Intermediate state orientations predicted for various pump-probe power regimes



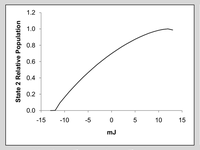
Low Pump, Zero Probe



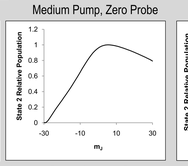
Medium Pump, Zero Probe



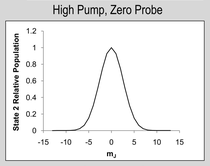
High Pump, Zero Probe



Medium Pump, Low Probe



Medium Pump, Medium Probe



Medium Pump, High Probe

Future Plans

- Find an appropriate theoretical model for the change in orientation during a collision to incorporate into density matrix
- Investigate ground state collisional lines
  - Thus far we have only investigated excited state collisional lines
  - v-changing collisions have been observed in ground state. We plan to investigate these in greater detail
- At Temple U., we have observed ground state collisional lines for very large ΔJ (up to ΔJ =58) in polarization spectroscopy of Rb_2
  - How can orientation be preserved in collisions where J changes by a very large amount?
- Effects on orientation/population by other collision partners (He, Xe, Kr)
- Propensity toward $\Delta J = \pm 2, \pm 4$ not observed in NaCs
  - Is this due to NaCs being "more heteronuclear" than NaK?