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Experimental Studies of NaCs

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Abstract

We present experimental studies of excited electronic states of the NaCs molecule that are currently underway in our laboratory. The optical-optical double resonance method is used to obtain Doppler-free excitation spectra for several excited states. These data are being used to obtain Rydberg-Klein-Rees (RKR) or Inverse Perturbation Approach (IPA) potential curves for these states. Bound-free spectra from single ro-vibrational levels of electronically excited states to the repulsive wall of the $1(a)^3\Sigma^+$ state are also being recorded. Using the previously determined excited state potentials, we can fit the repulsive wall of the $1(a)^3\Sigma^+$ state to reproduce the experimental spectra using LeRoy's BCONT program. A slightly modified version of BCONT will also be used to fit the relative transition dipole moments, $\mu_e(R)$, as a function of internuclear separation, R, for the various bound-free electronic transitions.

Why Study NaCs?

Large permanent dipole moment

Permanent Dipole Moments of Heteronuclear Alkali Molecules in $X^1\Sigma^+$ State

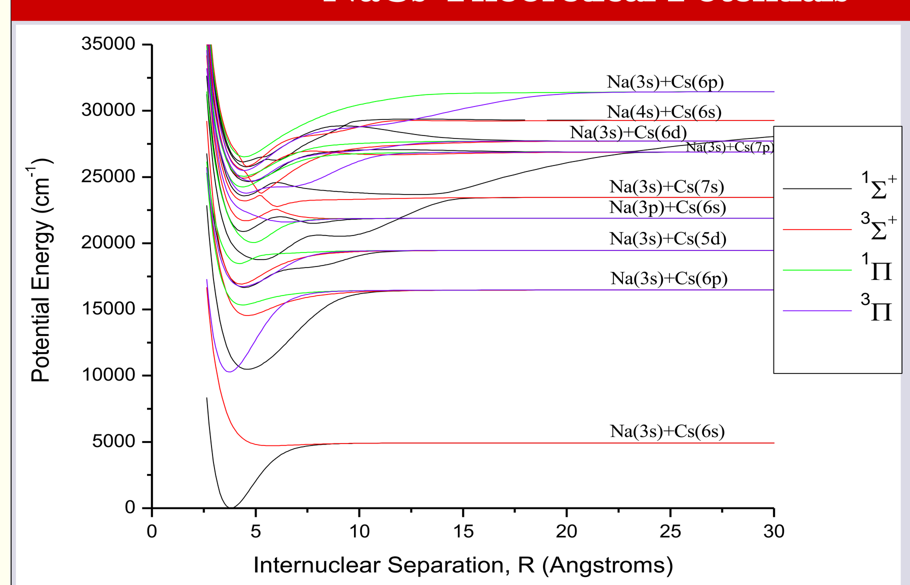
Molecule	Expt. Dipole(Debye)
LiNa	0.47
LiK	3.45
LiRb	4.05
LiCs	6.3
NaK	2.73
NaRb	3.1
NaCs	4.75
KRb	0.2
KCs	2.58
RbCs	2.39

Large spin-orbit interactions:

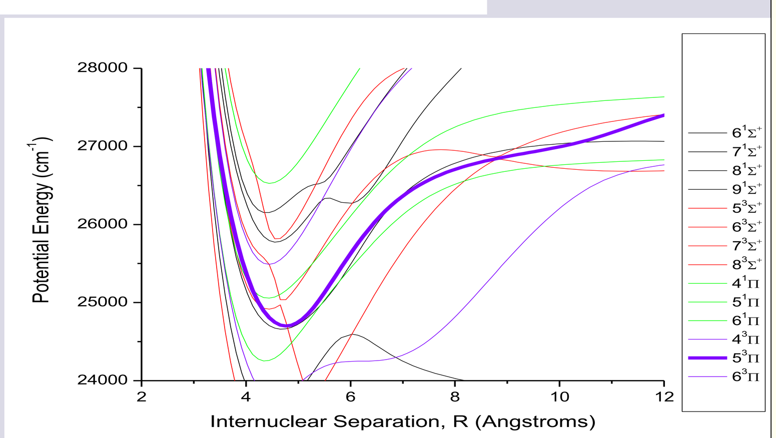
Spin-Orbit Splitting of Alkali Atoms (E[P(J=3/2)] - E[P(J=1/2)])

Atomic symbol	Splitting (cm^-1)
Li	0.335
Na	17.196
K	57.71
Rb	273.595
Cs	554.0393
Fr	1686.589

NaCs Theoretical Potentials*



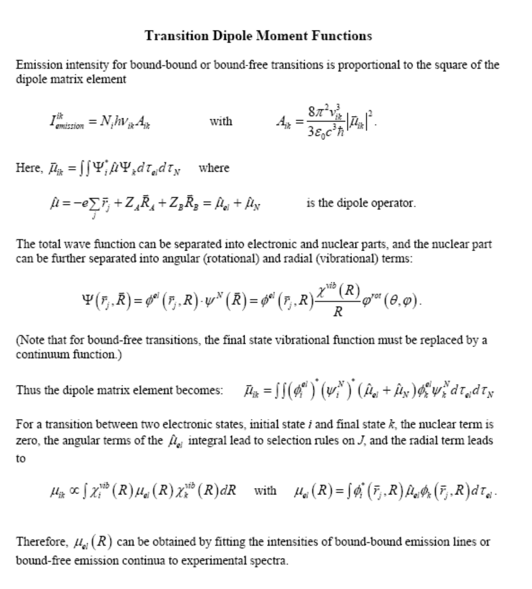
*M. Korek et al., JCP **126**, 124313 (2007)



Goals

- Map excited state potentials
- Map repulsive wall of the $a^3\Sigma^+$ state
- Determine transition dipole moment functions, $\mu_e(R)$
- Study collisional energy transfer
- Measure spin-orbit coupling interaction parameters as functions of R
- Study hyperfine structure

Transition Dipole Moment and Selection Rules



Selection Rules for Electronic Transitions

Δv = anything

$\Delta J = 0, \pm 1$ with restriction that $\Delta J = 0$ forbidden for $\Sigma \leftrightarrow \Sigma$ transitions

Hund's case a)

$\Delta S = 0$

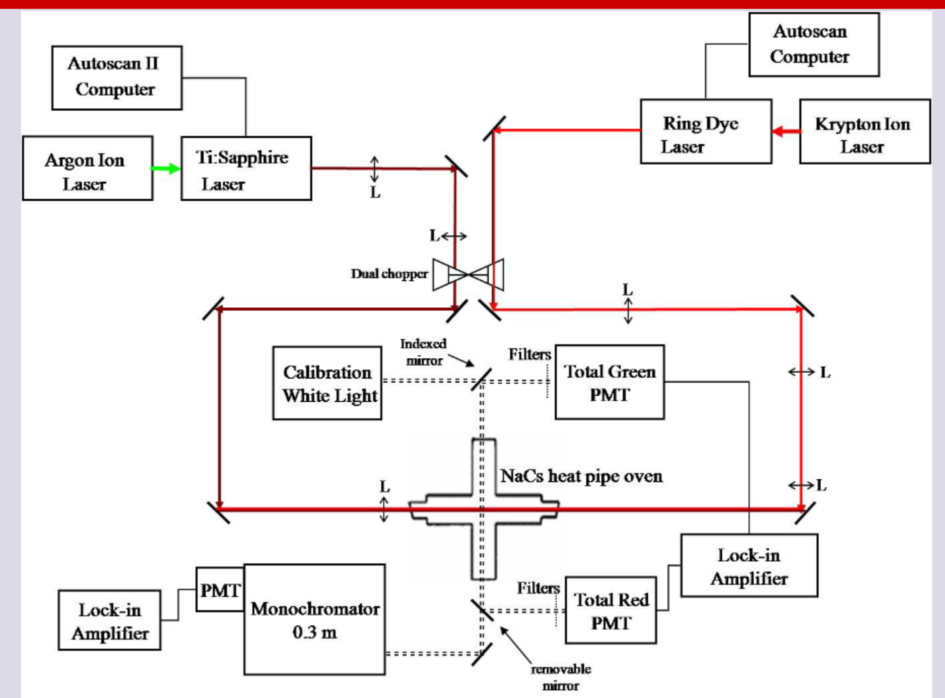
$\Delta\Lambda = 0, \pm 1$

$\Delta\Omega = 0, \pm 1$

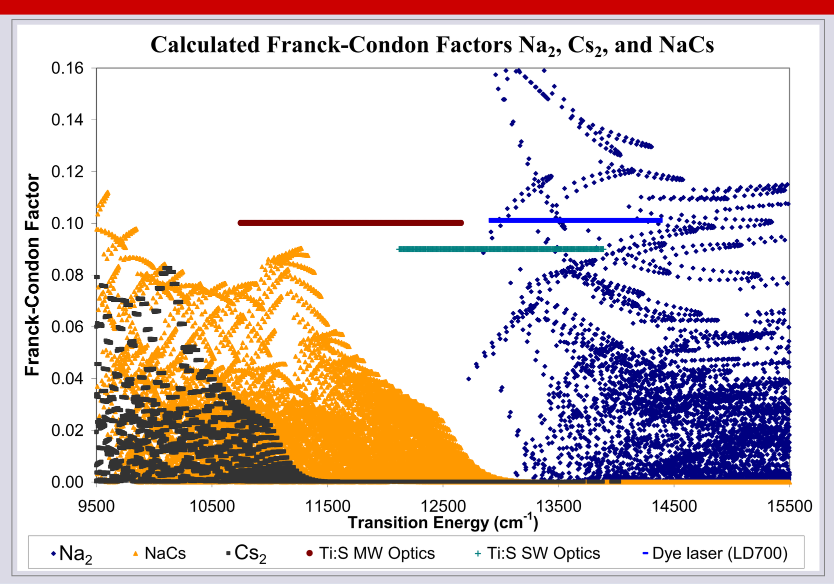
Hund's case c)

$\Delta\Omega = 0, \pm 1$

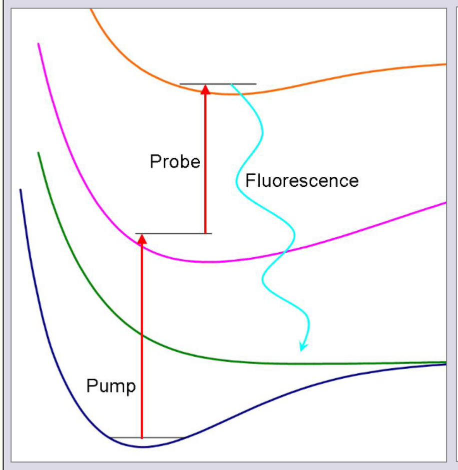
Experimental Setup

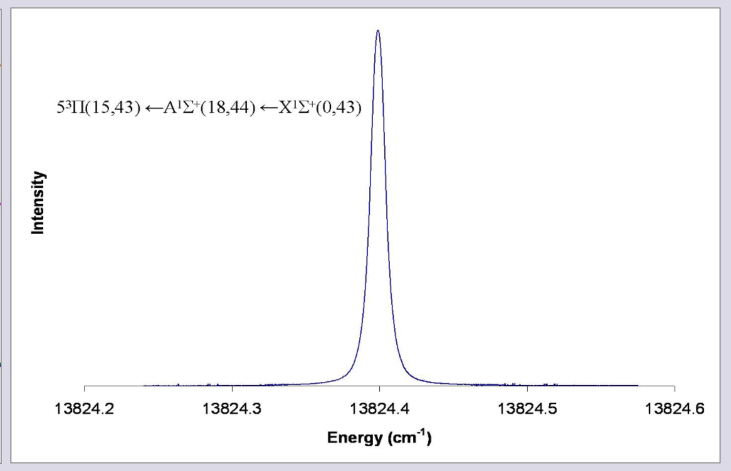


Franck-Condon Factors $A^1\Sigma^+ \leftarrow X^1\Sigma^+$

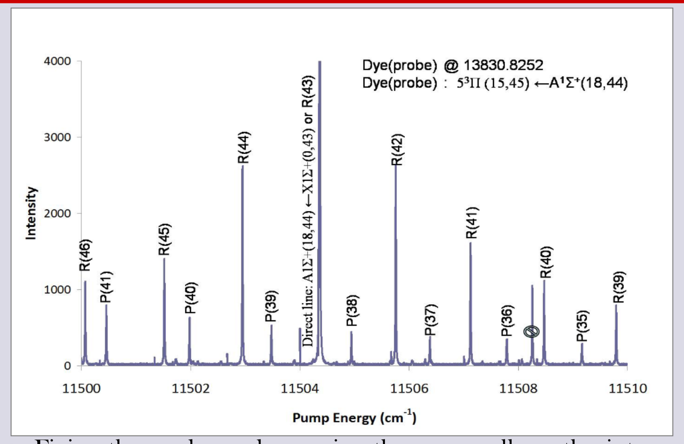


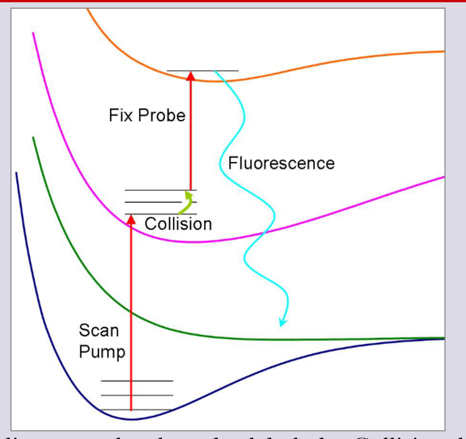
Double Resonance Excitation



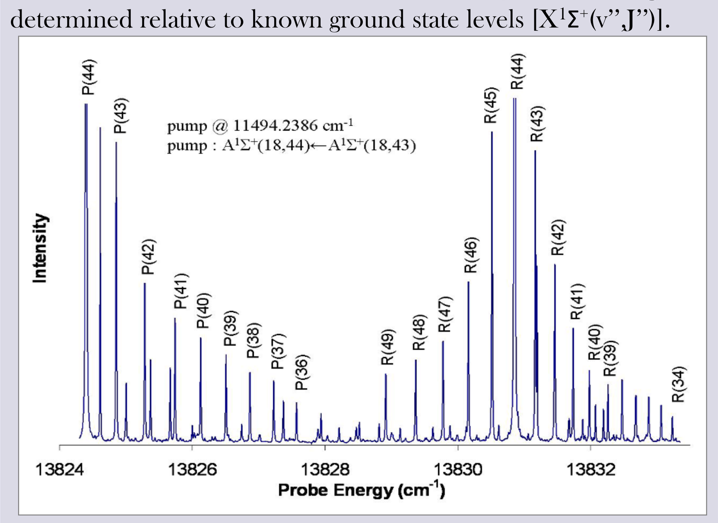


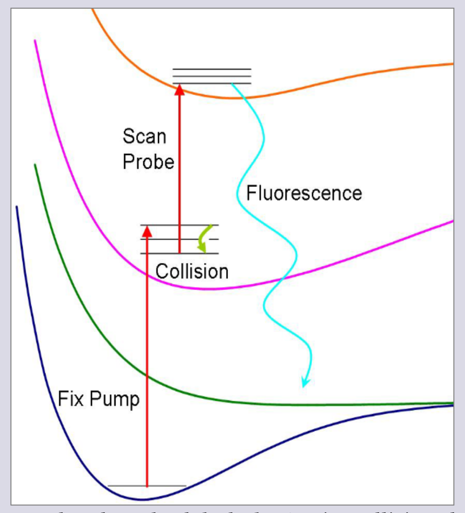
Collisional Population Transfer





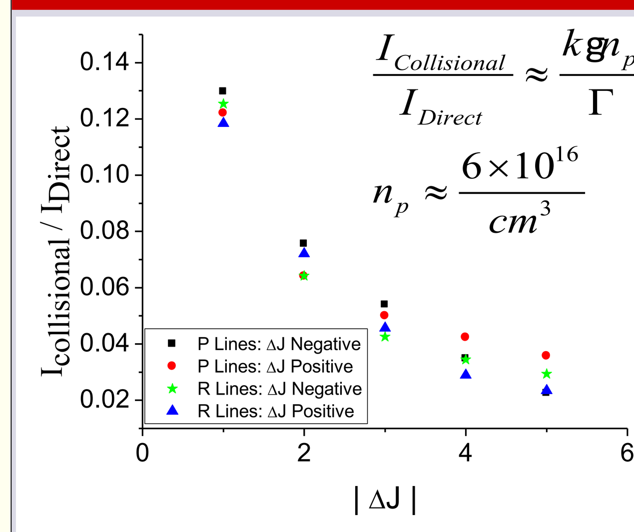
• Fixing the probe and scanning the pump allows the intermediate state levels to be labeled. Collisional satellite lines allow intermediate state $[A^1\Sigma^+(v',J')]$ level energies for a large number of rotational levels to be determined relative to known ground state levels $[X^1\Sigma^+(v'',J'')]$.





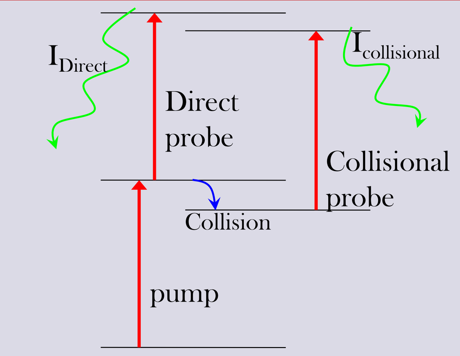
•Then fixing the pump and scanning the probe allows the excited state levels to be labeled. Again collisional satellite lines allow the upper state level energies for a large number of rotational levels to be accurately determined relative to $A^1\Sigma^+(v',J')$ levels.

Collision Rates



$$\frac{I_{Collisional}}{I_{Direct}} \approx \frac{k n_p}{\Gamma}$$

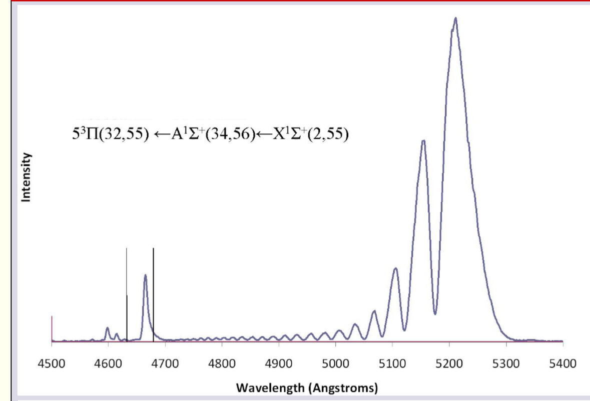
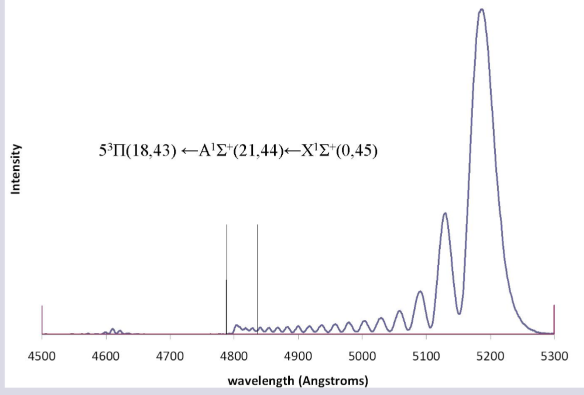
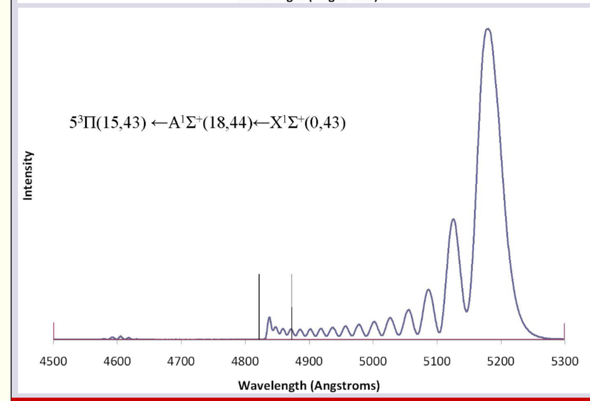
$$n_p \approx \frac{6\times10^{16}}{cm^3}$$



Using $\Gamma \approx 3\times10^7$ Hz and $I_{Collisional}/I_{Direct} = 0.1$, the collisional rate coefficient k can be calculated:

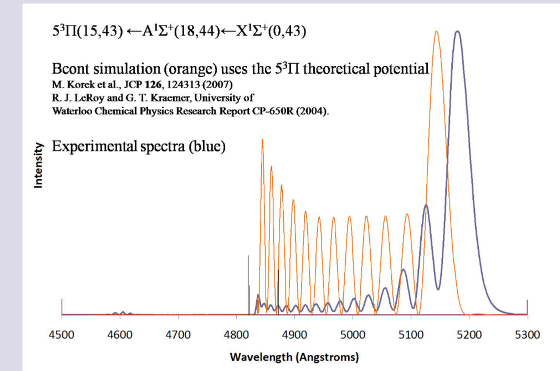
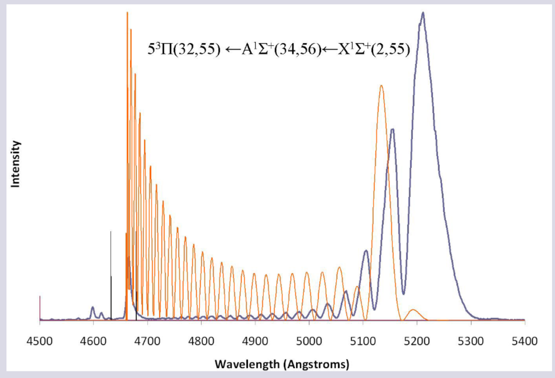
$$k = \frac{\left(\frac{I_{Collisional}}{I_{Direct}}\right)^{|\Delta J|=1}_{Average} \times \Gamma}{n_p} = 5\times10^{-11}\,\frac{cm^3}{s}$$

Resolved $5^3\Pi_0$ Fluorescence Spectra

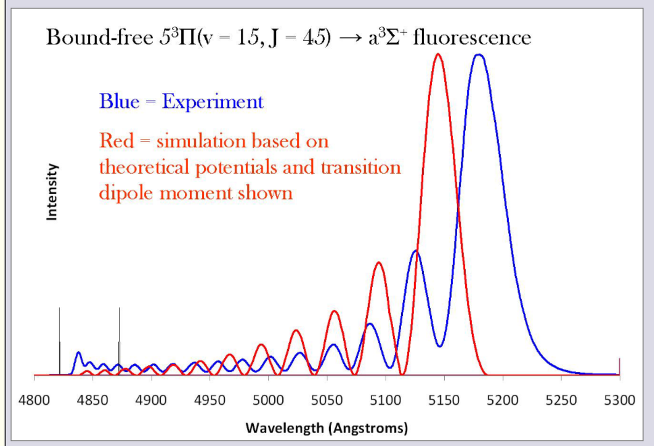




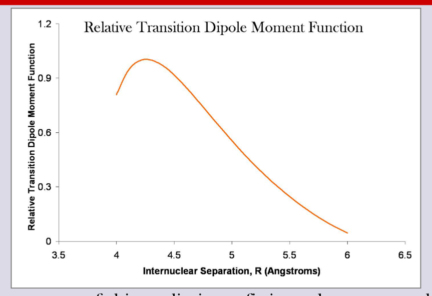
The black vertical lines in each scan denote the difference in energy between the selected $5^3\Pi$ level to the $a^3\Sigma^+$ asymptotic limit or to the bottom of its shallow well. Thus the part of the spectrum between the vertical lines represents unresolved bound-bound transitions. A monotonic difference potential produces simple reflection spectra, which provide an easy method to determine the upper state vibrational quantum number.

Spectra Simulation using BCont: Constant Transition Dipole Moment

Spectra Simulation using BCont: Fit of Relative Transition Dipole Moment Function





The purpose of this preliminary fit is to demonstrate that the increasing amplitude of the experimental bound-free intensity with increasing wavelength can be explained by a transition dipole moment function that generally decreases with increasing R. A final fit of the transition dipole moment will be carried out once accurate experimental potentials for the $5^3\Pi$ and $a^3\Sigma^+$ potentials have been obtained. (The latter require more data than we have acquired so far.) Positions of the peaks and nodes of the oscillating spectra depend on the upper and lower state potential energy curves while peak amplitudes depend on the transition dipole moment function.

Preliminary Dunham Coefficients for $5^3\Pi_0$ State

Parameter	Energy* ± Error** (cm^{-1})	Theoretical values*** (cm^{-1})
Te	24525.5 ± 6.2	24880
Y(1,0)	61.18 ± 0.55	58.6
Y(2,0)	-0.270 ± 0.013	-
Y(0,1)	0.0398 ± 0.0020	0.0394
Y(1,1)	$-(2.4 \pm 0.95) \times 10^{-4}$	-
Y(0,2)	-2.0279×10^{-8} (fixed in fit)	-

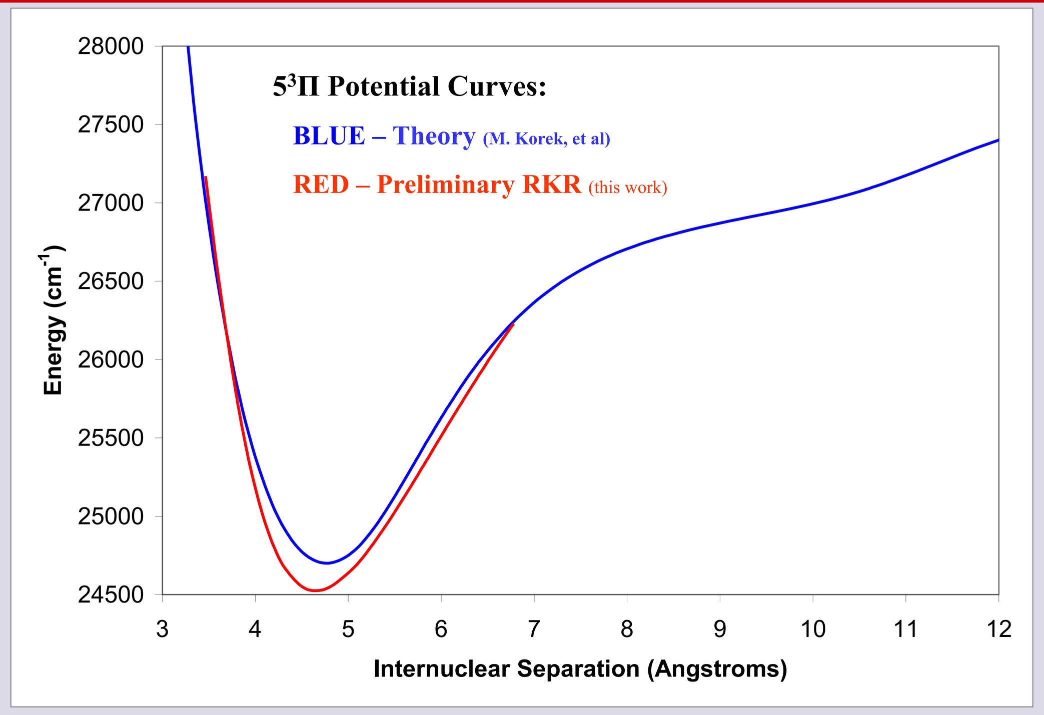
*Fit using DParfit program

R. J. LeRoy, DParFit 3.3: A Computer Program for Fitting Multi-Isotopologue Diatomic Molecule Spectra, University of Waterloo Chemical Physics Research Report CP-653 (2001)

**Statistical error only

*** M. Korek et al., JCP **126**, 124313 (2007)

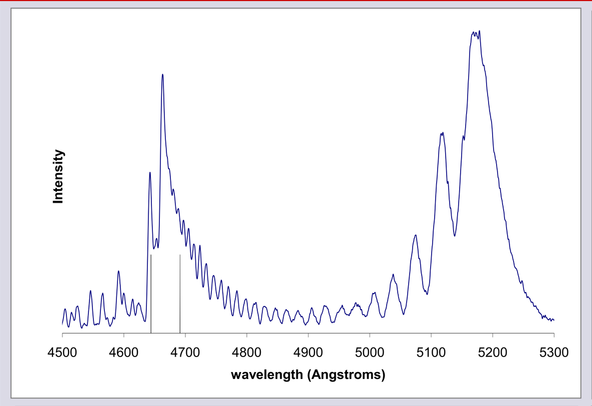
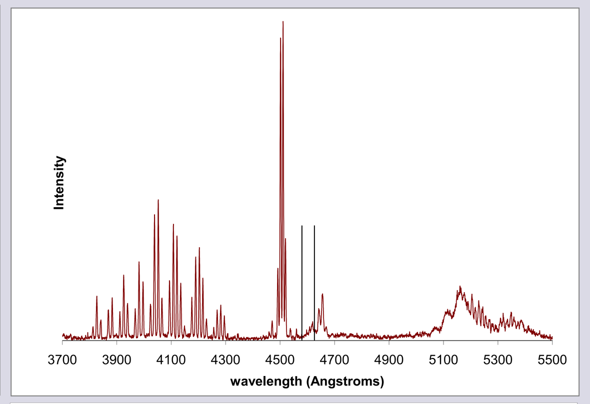
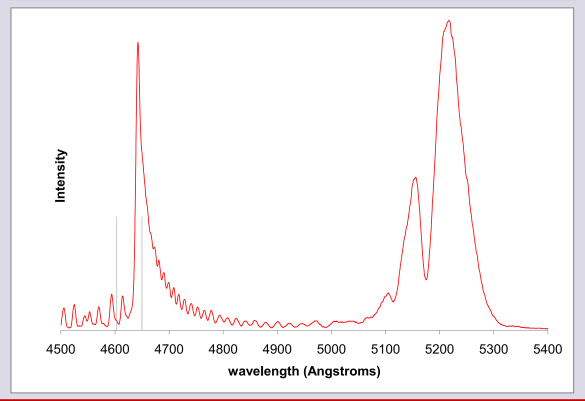
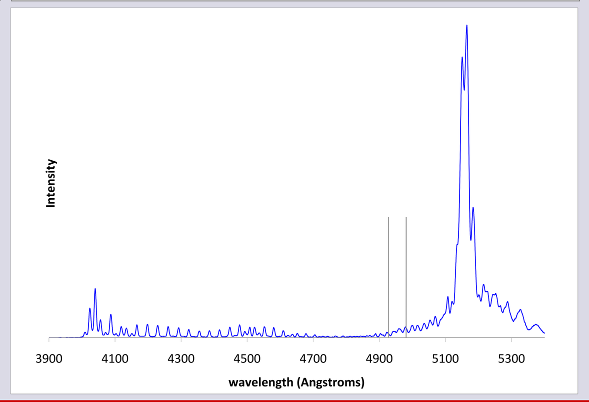
$5^3\Pi_0$ RKR and Theoretical Potentials



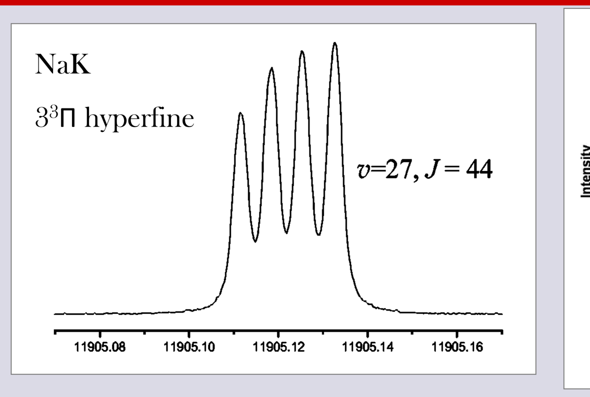
M. Korek et al., JCP **126**, 124313 (2007)

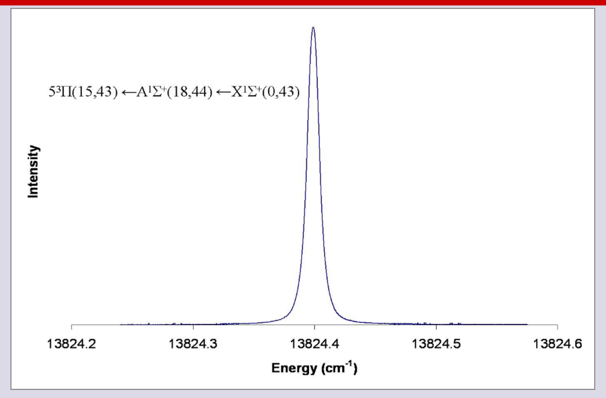
R. J. LeRoy, RKR1 2.0: *A Computer Program Implementing the First-Order RKR Method for Determining Diatomic MoleculePotential Energy Curves*, University of Waterloo, Chemical Physics Research Report CP657R (2004).

Spectra of Other NaCs Excited States Under Investigation

Hyperfine Structure





Previous work on NaK showed a rich variety of hyperfine structure. Ongoing studies of NaCs have yet to reveal clear hyperfine structure.

Hyperfine interaction in NaCs should be described by:

$$\mathbf{E_{hfs} = b_F\, I \cdot S}$$

$b_F^{Na} = 886$ MHz $\quad I_{Na} = 3/2$

$b_F^{Cs} = 2298$ MHz $\quad I_{Cs} = 7/2$

From this we expect a larger hyperfine interaction for NaCs. However, states of NaK can often be described by Hunds case $b_{\beta J}$ or $b_{\beta s}$, while NaCs has a much stronger spin-orbit interaction and is likely described by Hunds case c.

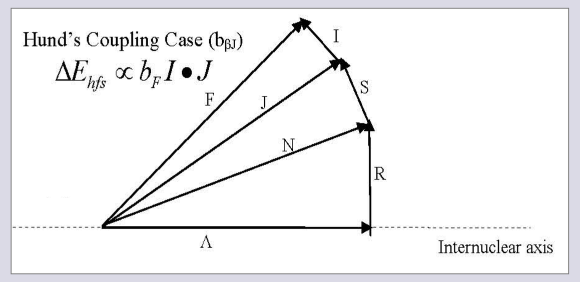
Hunds case $b_{\beta J}$

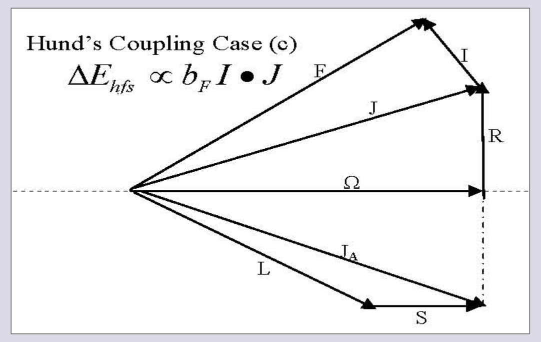
$$E_{hfs}^{b_{\beta J}} \approx \frac{b_F K}{4}\,\frac{J(J+1) - S(S+1) - N(N+1)}{J(J+1)}$$

$$K = [F(F+1) - J(J+1) - I(I+1)]$$

Hunds case c

$$E^c_{hfs} \approx \frac{b_F K}{2}\,\frac{\Sigma\Omega}{J(J+1)}$$





Therefore, the hyperfine structure might only be observable at lower J values than those we have studied.