Spectroscopic investigations of Rb- and Cs- rare gas systems

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ABSTRACT

We describe a series of measurements of absorption and laser induced fluorescence using cells that contained cesium and rubidium and krypton as a bath gas. These studies showed strong blue wing absorption to the short wavelength side of the alkali atom D₂ lines due to collisionally formed Cs-Kr or Rb-Kr excimers. These studies indicate that these species may be appropriate candidates for optically excited Rb and Cs atomic lasers.

Keywords: optically pumped alkali lasers, excimer absorption, photodisociation

1. INTRODUCTION

For more than 30 years there has been interest in developing gas phase lasers based on the strong D-line radiation in alkali atoms. For example, White1 observed amplified spontaneous emission at 589.6 nm from the Na D₁-line (3p²P₁/₂ → 3s²S₁/₂) subsequent to photodissociation of NaI by the fifth harmonic of a Q-switched Nd:YAG laser (212.8 nm). The excitation wavelength connected the ground state of the NaI molecule to an excited state that had the excited Na 3p²P₁/₂ and the ground state I atom as the separated atom limit. Thus, the photodissociation products provided the selective excitation of the Na atom. While this result provided a model for producing lasers from other alkali species, the quantum efficiency was poor and the method required ultra-violet excitation sources. Direct optical excitation of alkali atoms via the ²P₁/₂ ← ²S₁/₂ transition would remove the quantum efficiency issue, but consideration of pumping a two level system shows that one would only be able to obtain equal populations of the ²P₁/₂ and ²S₁/₂ levels. Recently, Krupke and coworkers2,3 demonstrated that excitation of the slightly higher energy ²P₃/₂ level followed by rapid collisional energy transfer to the ²P₁/₂ level could provide the basis of an efficient three level laser. This initial work used a CW Ti:S laser at 760 nm to excite the ²P₃/₂ state in Rb. Ethane in the cell provided efficient energy transfer to the ²P₁/₂ level and laser oscillation was observed on the Rb D₁ line at 794 nm. Diode laser pumped alkali lasers (DPAL) hold considerable promise for efficient, scalable lasers in the 760 to 894 nm range with the wavelength depending on the choice of alkali atom.2-5

High power diode lasers and laser arrays typically are spectrally much wider than the D₁ absorption lines in alkali atoms, and this can significantly reduce the efficiency of the optical excitation process. Collisional broadening of the alkali atom absorption lines and spectral narrowing of the diode laser output using Bragg gratings are two approaches being pursued to improve the spectral overlap of the pump laser and the alkali atom absorption lines cause inefficient optical excitation. Very recently Readle et al.6 described a novel approach for exciting atomic alkali lasers using rare gas-alkali atom complexes originally observed by Chen and Phelps7. They used the blue-wing satellite of the Cs D₂ line to absorb pulsed dye laser pulses covering the spectral region of ~ 840 to 852 nm. Rapid dissociation of the Cs-Ar excimer produced Cs ²P₃/₂ atoms that were collisionally relaxed to the ²P₁/₂ level and laser oscillation on the D₁ line was observed. The broad absorption wing of the Cs-Ar excimer may provide a path for using spectrally broad high power diode lasers to produce alkali atom D-line lasers. In this paper we describe spectroscopic studies of RbKr and CsKr absorption using both white light and tunable Ti:S laser radiation in order to assess possible laser candidates analogous to the recent results on CsAr.

2. DESCRIPTION OF EXPERIMENTAL SETUP

We designed and built an oven that allows us to insert sealed Pyrex cells that contain alkali species buffered with a rare gas. The cell chamber includes four cartridge heaters that have the capability for operating the cells from room temperature to over 250°C. The oven chamber also contains ports that allow both optical absorption and fluorescence measurements to be completed as shown in Figs. 1 and 2.

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For these experiments we used a Schwartz Electro Optic “Titan” Ti:S laser for our excitation source. This device can be operated in both a stable cavity and ring configuration, and provides tunable, CW output covering the range from 750 to nearly 900 nm. The Ti:S laser is pumped by a Coherent Verdi diode laser-pumped, frequency-doubled Nd:YAG laser that produces 5W of 532nm output.

We also developed a single-mode optical fiber-coupled white light excitation source for absorption studies. This device provides a collimated beam convenient for absorption studies using the heated alkali atom cells. We used an optical multi-channel analyzer (OMA) to detect the white light absorption and laser induced fluorescence (LIF) spectra from the alkali atom cells. The heart of this detection system is a Princeton Instruments, liquid nitrogen, Si-CCD array that provides low dark count detection ideal for CW measurements.

For the Kr rare gas studies we used three cells: one contained Rb and 500 Torr Kr, one contained Cs and 500 Torr Kr, both were 5 cm long, and a third that contained Rb and 500 Torr Kr that was 2.5 cm long. None of these cells contained ethane, thus we expect a severely reduced energy transfer from the $^3P_{3/2}$ to $^3P_{1/2}$ state of the alkali atom utilized in
traditional DPAL devices. We also used a 5 cm long cell that contained 500 Torr of He, and 75 Torr ethane to investigate Rb-He excimer formation and absorption.

3. RESULTS AND DISCUSSION

Our Cs cell contained enough Rb that we could observe both species simultaneously with the OMA equipped with a low dispersion grating (300 grooves/mm). Figure 3 shows absorption from the Rb-Kr excimer and from both D lines (780 and 794 nm) and part of the Cs-Kr excimer. These data clearly show that the excimer absorption increases as the alkali vapor pressure is increased. We also completed a brief study using a Rb-He cell and saw much weaker blue wing absorption that implies a smaller excimer concentration as shown in Fig. 4. While there is some evidence for blue wing satellite absorption, it is much weaker than for the Rb-Kr case as might be expected since He is much less polarizable than Kr.

![Fig. 3. White light absorption of the Rb-Kr and Cs-Kr exciplex systems as a function of cell temperature.](image)

For the data shown above in Figs. 3 and 4, we used a 300 groove/mm grating in the OMA spectrometer. The relatively low resolution allowed us to record both the Rb and Cs spectral features simultaneously. In order to more accurately characterize the excimer absorption wings we used a 1200 groove/mm grating. Figure 5 shows the Cs D2 line and the
Cs-Kr blue wing at higher resolution. We have normalized this spectrum to zero intensity where the D₁ line appears to be "black". Comparison of this spectrum to that for the Cs-Ar system reported earlier by Readle et al. shows similarities in spectral shape. The blue wing origin and peak for Cs-Ar appear to be slightly blue shifted compared to the Cs-Kr spectrum.

To further explore the Cs-Kr system we performed some spectrally resolved laser induced fluorescence experiments using the Ti:S laser as the excitation source. The top half of Fig. 6 illustrates spectrally resolved LIF spectra for the Cs-Kr complex when excited near the Cs D₂ line. These data were obtained using the OMA described above. The data on the bottom of this figure illustrate the absorption spectrum obtained with the white light source. The major features are very similar in both. We plan to examine these excitation spectra under higher spectral resolution and for other bath gas partners in the near future.

The feature at 795 nm is the Rb D₁ line also excited when pumping the Cs-Kr system presumably by energy transfer.
When the region of the excimer absorption wings was probed at several discrete wavelengths by the tunable Ti:S laser, both D1 and D2 emission were observed for the respective Cs and Rb sample cells. Figure 7 clearly shows that as the laser is tuned through the blue wing of the Cs-Kr excimer and through the region of between the Cs D lines, emission from the two D lines persist, consistent with Readle et al.’s model for the excimer absorption followed by rapid dissociation into excited alkali atoms. Note that as the Ti:S laser was tuned to the short wavelength side of the Cs-Kr band origin, the excitation of the D lines vanished.

![Fig. 7. Spectrally resolved LIF spectrum of Cs-K cell for four excitation wavelengths.](image)

We also observed an interesting spectral feature while recording the data shown in Fig. 7. We saw visually an intense blue beam in the Cs-Kr cell and similarly in the Rb-Kr cell. Spectral analysis (see Fig. 8) demonstrated that these features were from higher states of the alkali atoms: 7^2P_{3/2,1/2} for Cs and 6^2P_{3/2,1/2} for Rb. This implies at least a two step or two photon process where the excited excimer may be an intermediate state. Admittedly, we were operating at fairly high Cs and Rb number densities (~ 0.5 to 1x10^{15} cm^{-3}), but this process appears to warrant further investigation. The important point is that it may allow excitation of alternative states of alkali atoms with broad band sources, and these may lead to other laser transitions. For example, Sharma et al. observed mirrorless, c-w lasing from many transitions in both Cs and Rb when the same manifolds that we observed were excited directly with a c-w dye laser. These laser transitions varied in wavelength from 1.32 to 3.09 µm. The energy levels and laser transitions observed by Sharma and co-workers are shown in Fig. 9.
4. SUMMARY

Using white light absorption and spectrally resolved laser induced fluorescence we have observed excimer formation and absorption in the Rb-Kr and Cs-Kr systems. We observed D$_1$ and D$_2$ emission from both Rb and Cs when exciting the within the excimer wings. These results show considerable promise for development of broadband pumped Cs and Rb lasers using the Kr induced absorptions. We also observed intense blue emission from higher lying levels in the Cs and Rb energy level manifolds.

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