Spectroscopic Studies of Alkali Atom-Rare Gas Systems
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ABSTRACT
We describe a series of measurements of absorption and laser induced fluorescence on cells that contained cesium and rubidium and a rare gas: He, Ar, Kr, or Xe. These studies showed strong blue wing absorption to the short wavelength side of the alkali atom D_2 lines due to collisionally formed Cs- or Rb-rare gas excimers. We also have observed an efficient two photon excitation of higher lying states in Cs and Rb that produces both intense blue emission and IR atomic emission in the 1.3 to 3.8 μm spectral region.

Keywords: optically pumped alkali lasers, excimer absorption, photodissociation

1. INTRODUCTION
There has been a long standing interest in developing alkali atom lasers. For example, Krupke and coworkers demonstrated that excitation of the slightly higher energy ^3P_{3/2} level followed by rapid collisional energy transfer to the ^3P_{1/2} 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 ^3P_{3/2} state in Rb. Ethane in the Cs cell provided efficient energy transfer to the ^3P_{1/2} level and laser oscillation was observed on the Rb D_1 line at 794 nm. Diode laser pumped alkali lasers (DPAL) holds considerable promise for efficient, scalable lasers in the 760 to 894 nm range depending on the alkali atom.

High power diode lasers and laser arrays typically are spectrally much wider than the D_1 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 atoms absorption lines. Very recently Readle et al. described a novel approach that eliminates this concern by exciting the D lines of alkali atoms via broad band absorption on rare gas-alkali atom molecular complexes. They used the blue-wing satellite of the Cs D_2 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 ^3P_{3/2} atoms that were collisionally relaxed to the ^3P_{1/2} level and laser oscillation on the D_1 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. They have also recently demonstrated a Cs atom laser based upon Cs-Kr molecular absorption.

In this paper we describe spectroscopic studies of Rb- and Cs- absorption using both white light and tunable Ti:S laser radiation in order to further assess these systems as possible efficient gas phase optically pumped laser systems.

2. DESCRIPTION OF EXPERIMENTAL SETUP
Our experimental test bed is shown in Figure 1. The oven 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.

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For these experiments we used a Schwartz Electro Optic “Titan” Ti:S laser for our excitation source. This device that can be operated in both a stable cavity and ring configuration 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 have used 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 cooled, Si-CCD array that provides low dark count detection ideal for CW measurements. We used several 5 cm long, 2.5 cm diameter cells each containing Cs or Rb and buffered with 500 Torr of a rare gas species (Kr, Xe, Ar). Some cells also contained 75 Torr of ethane.

3. RESULTS AND DISCUSSION

One of our Cs cells contained enough Rb that we could observe both species simultaneously with the OMA equipped with a low dispersion grating (300 grooves/mm). Figure 2 shows absorption from the Rb-Kr excimer and from both D lines (780 and 794 nm) and part of the CsKr exciplex. Note that for the Cs-Kr case the D1 line at ~ 895 nm cannot be displayed simultaneously. These data clearly show the excimer formation as the alkali vapor pressure is increased. We also completed a short study using a Rb-He-ethane cell and saw much weaker blue wing absorption that implies a smaller excimer concentration as shown in Figure 3. 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.

Figures 4 through 7 show the absorption spectra as a function of Cs number density for the Cs/Kr/ethane; Cs/Ar/ethane, Cs/Ar; and Cs/Xe cells. Note that the Cs number densities were calculated using vapor pressure curves assuming that the Cs vapor is in equilibrium with the solid.
Figure 2. White light absorption of the Rb-Kr and Cs-Kr exciplex systems as a function of cell temperature

Figure 3. Absorption spectrum of Rb cell containing 500 Torr of He and 75 Torr of ethane
Figure 4. Absorption spectrum of Cs cell containing 500 Torr of Kr and 75 Torr of ethane

Figure 5. Absorption spectrum of Cs cell containing 500 Torr of Ar and 75 Torr of ethane
We have normalized all of the spectra to zero intensity where the D1 and D2 lines appear to be opaque. The data clearly show the formation of blue satellite wings at 837 nm for the CsAr excimer, and 841 nm for CsKr and CsXe. We also observed that ethane has some effect on the exciplex formation. Figure 8 shows a comparison of the CsAr excimer formation with (black trace) and without (dashed trace) ethane buffer gas in the cell. Both spectra were recorded at a cell temperature of 448 K ([Cs] = 6.75 x 10^{14} cm^{-3}). The blue wing peak and origin appear in the same location in both spectra, however the exciplex formation is greatly enhanced in the Cs/Ar/ethane cell as compared to the Cs/Ar cell. We observed a similar enhancement in the Cs/Kr/ethane system.
As we reported previously, we also observed an intense blue beam in the Cs-Kr cell and similarly in the Rb-Kr cell when exciting with the Ti:S laser near the D₂ absorption line. We identified the blue emission was due to transitions from the higher states of the alkali atoms: 7²P₃/₂,1/₂ for Cs and 6²P₃/₂,1/₂ for Rb to the respective ground states. We have also observed transitions in the 1.3 to 3.8 µm region as shown in Figures 9 and 10. This is similar to Sharma et al.⁸ who 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 CW 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 Figure 11. The emission lines that we observed must be formed by a two photon absorption, and we believe it is a very efficient, sequential pumping through real, excited states of the alkali-Kr complex.
4. SUMMARY

These studies further illustrate the spectroscopy of alkali-rare gas molecules and show how these species can be used to optically excite both the resonant $^2P_{3/2}$ and $^2P_{1/2}$ levels and higher lying levels in the alkali energy level manifold. These exciplex species provide broad band absorption features for high power diode lasers. In addition, efficient two photon absorption may produce a host of excited states that may serve as the upper levels of a host of laser lines in the near to mid-IR spectral region. We plan to investigate these phenomena in future studies.
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REFERENCES