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New Class of Excimer-Pumped Atomic Lasers (XPALS)

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This program hns pursued the investigation or XPAL properties, and new pumping schemes, ns well ns modeling, nnd measuring critical photoioniz ntion nnd cxcitcd state-excited state reaction rates. We ore pleased to report that the main goal orthis program, the viability of nn atomic laser having a quantum efficiency greater thnn one, has been demonstrated. We believe this laser to represent a breakthrough in laser technology because the system appears to be scnlable in power but, more importBntly, the loser gain medium is cooled by the emission orevery laser photon. The smallest energy derect that can exist between the pumped and lasing states in a three-level loser system has also been experimentally measured. The photoionization processes in nlkali-noble gas mixtures hns been modeled and experimentally studied. Detailed modeling or XPAL nnd OPAL systems and plasma formation mechanisms during high power pumping has been performed.

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The focus of this 3 year AFOSR/HEL-JTO – sponsored research program was the investigation of XPAL properties, and new pumping schemes, as well as modeling, and measuring critical photoionization and excited state-excited state reaction rates in order to improve the performance of XPALs.

The first several experiments were conducted to measure the smallest energy defect that can exist between the pumped and lasing states in a three level laser system. This is a problem critical to the cooling of the laser medium by pumping it through the lower lying states. Results obtained during this program were described in detail in previous reports. In short, Rb-Xe gas mixtures were used to demonstrate the collapse of a three level laser when the thermal energy kT reaches approximately the energy defect between the laser and pumped levels.

Further experiments conducted at the University of Illinois have demonstrated the viability of an atomic laser having a quantum efficiency greater than one. We believe this laser to represent a breakthrough in laser technology because the system appears to be scalable in power but, more importantly, the laser gain medium is cooled by the emission of every laser photon.

Our experiments show that the suggested two color pumping scheme shown in fig. 1 allows the laser's efficiency to be increased by a factor of 1.7 (fig. 2). In addition, the

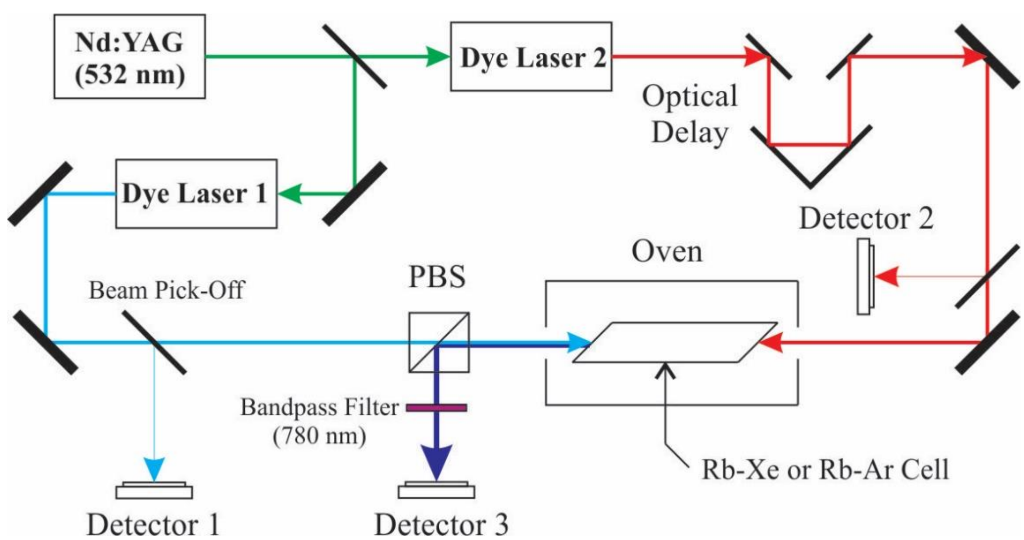

Figure 1. Schematic diagram of the laser system.

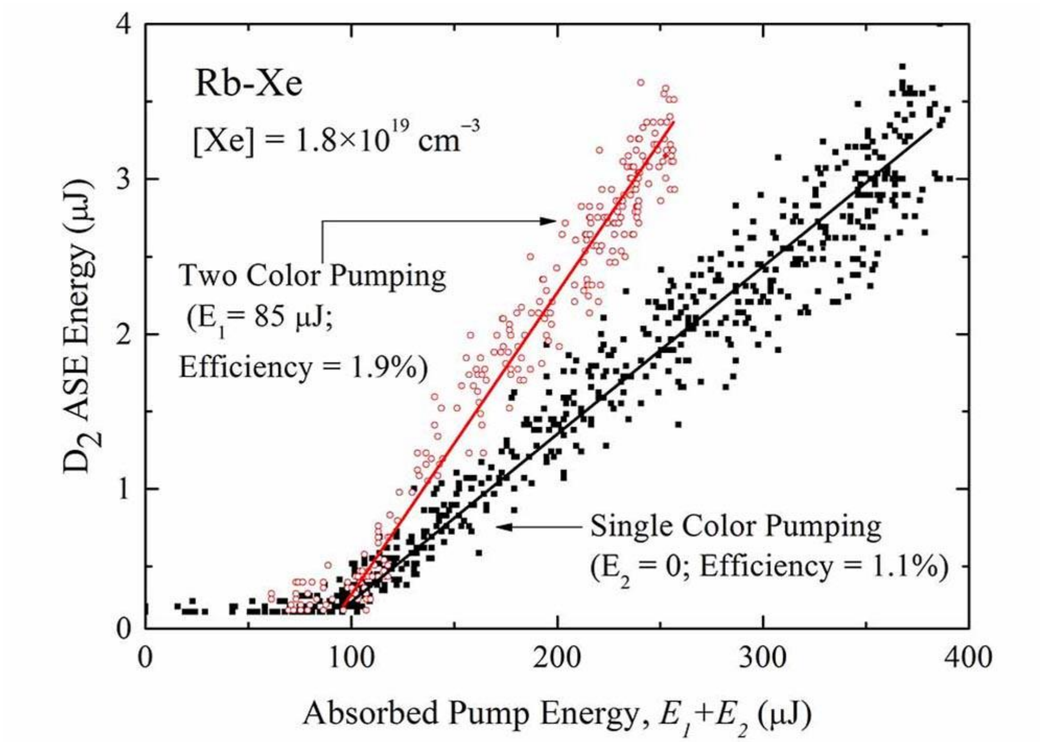

Figure 2. (Red circles) Dependence of D_2 line (780 nm) ASE on $E_1 + E_2$, the sum of the energies absorbed by the first and second pump pulses. For all measurements, [Rb]=$9.2*10^{14}$cm^{-3}, [Xe]=$1.8*10^{19}$cm^{-3}, λ_1=759.95 nm, and λ_2=794.76 nm. E_1 was fixed at 85 µJ and the time delay Δt was maintained at 8 ns; (Black Squares): Similar data were recorded when E_2=0 and the laser is pumped only at 760 nm (Rb-Xe blue satellite).

pump absorption coefficient was increased by more than an order of magnitude, as compared to the classical XPAL pumping scheme. These results were published in Applied Physics Letters [1].

The experimental setup (Fig. 1) was described in detail in a previous report to the HEL-JTO. Briefly, a quartz cell filled with an alkali-noble gas mixture was pumped longitudinally with two dye lasers. The first pump pulse was used to prepare the system and populated the upper laser level to a point slightly below the lasing threshold. The second (main) pump pulse created the population inversion and triggered lasing on the Rb D_2 line transition at 780 nm.

The most important feature of this pumping scheme is that the wavelength of the main pump was longer than the wavelength of the Rb D_2 line laser. In other words, the energy of the emitted photons was higher than the energy of the pump photons. This fact implies the extraction of thermal energy from the gain medium to compensate for the energy difference between the ASE and excitation photons.

It has been demonstrated (Fig. 2) that, in addition to the quantum efficiency being above one, the two-color pumped system exhibits higher slope efficiency as compared to the single color pumped case. It is important to note that the absolute efficiencies shown in Fig. 2 describe the ASE signals due to the specific experimental arrangement and, therefore, the actual D_2 line *laser* efficiency is significantly higher.

During the same reporting period, the kinetics of the higher energy excited states of Rb and Cs were studied in laser excitation experiments. In previous work, we have seen that the higher excited states are significantly populated by energy pooling processes when the D_1 or D_2 lines are strongly excited (e.g., optical pumping conditions). For the XPAL system, these higher excited states may be photoionized, resulting in loss of both input energy and metal atom number density. In a conventional DPAL system the higher excited states may react with hydrocarbons, producing metal hydride particles and carbon deposits.

Attempts were made to observe the ionization of Rb and Cs in the presence of 500 Torr of Ar. The D_2 lines were excited using pulsed laser intensities of approximately 0.5 MW/cm^2. A time-delayed, pulsed dye laser probe was used to detect metal vapor loss due to ionization. These measurements indicated that less than 1% of the neutral Rb or Cs was lost via ionization. Direct photoionization in the presence of 500 Torr of Ar, using 266 nm and 193 nm radiation ($P>1MW/cm^2$), was also found to be inefficient. Loss of ground state atoms was not detected, and there was negligible atomic line fluorescence.

Time-resolved fluorescence and laser pump-probe measurements were used to study the interactions of Rb(6p) with H_2, CH_4, and C_2H_6. At room temperature, the total removal, rate constants were found to be k_{H2}=(7.0±0.2)×10^{-10}, k_{CH4}=(6.2±0.2)×10^{-10}, and k_{C2H6}=(8.1±0.3)×10^{-10} cm^3 s^{-1}. These values are consistent with earlier determinations. Electronic structure calculations were used to investigate the deactivation mechanisms. Quenching of Rb(6p) by H_2 proceeds via a curve crossing with the potential energy curve of the Rb(6s)+H_2 collision pair, while CH_4 and C_2H_6 quench via electronic-to-vibrational energy transfer. Measurements of ground state population recovery were used to estimate the fraction of the quenching that could be attributed to chemical reactions. For H_2 the reaction channel accounted for 12% of the total removal rate constant. Production of RbH was confirmed by observing the laser induced fluorescence (LIF) spectrum of the $A^1\Sigma^+$-$X^1\Sigma^+$ transition. Reactive loss in the collisions with CH_4 and C_2H_6 accounted for 3% and 6% of the total removal rate constants. Surprisingly, searches for RbH produced by these reactions yielded negative results. Electronic structure calculations are currently being dedicated to examining the possible reactive channels for Rb(6p)+CH_4.

In summary, the main proposed milestones, and, in particular, the demonstration of the laser medium cooling with each laser photon produced, have all been achieved. The work conducted under this HEL-JTO program has demonstrated that the proposed two color pumping scheme of XPALs may be potentially used as the first high power laser system which is able to cool itself internally, thereby allowing its characteristics to be improved considerably as compared to other high power gas lasers.

We are grateful to the HEL-JTO for the support of this work.

List of publications:

1. A. E. Mironov, W. Goldshlag and J. G. Eden, “Two color pumping of the Rb D_2 line laser (780 nm) through the photoassociation of Rb-Ar or Rb-Xe thermal pairs: Realization of a quantum efficiency above one”, *Appl. Phys. Lett.*, 107, 041112 (2015).
2. J. D. Hewitt, T. J. Houlahan, Jr., J. E. Gallagher, D. L. Carroll, A. D. Palla, J. T. Verdeyen, G. P. Perram, and J. G. Eden, “Role of excited state photoionization in

the 852.1 nm Cs laser pumped by Cs-Ar photoassociation," Appl. Phys. Lett., vol. 102, 111104 (March 18, 2013).

3. B. D. Barmashenko, S. Rosenwaks, and M. C. Heaven, Opt. Comm. 292, 123-125 (2013).
4. A. E. Mironov, W. Goldshlag, and J. G. Eden. "Alkali D_2 line laser optically pumped by two color free-free absorption," Proc. SPIE (Submitted)

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Abstract

This program has pursued the investigation of XPAL properties, and new pumping schemes, as well as modeling, and measuring critical photoionization and excited state-excited state reaction rates. We are pleased to report that the main goal of this program, the viability of an atomic laser having a quantum efficiency greater than one, has been demonstrated. We believe this laser to represent a breakthrough in laser technology because the system appears to be scalable in power but, more importantly, the laser gain medium is cooled by the emission of every laser photon. The smallest energy defect that can exist between the pumped and lasing states in a three-level laser system has also been experimentally measured. The photoionization processes in alkali-noble gas mixtures has been modeled and experimentally studied. Detailed modeling of XPAL and DPAL systems and plasma formation mechanisms during high power pumping has been performed.

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1. A. E. Mironov, W. Goldshlag and J. G. Eden, "Two color pumping of the Rb D2 line laser (780 nm) through the photoassociation of Rb-Ar or Rb-Xe thermal pairs: Realization of a quantum efficiency above one", Appl. Phys. Lett., 107, 041112 (2015).
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