Multi-Dimensional Modeling of the XPAL System

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The exciplex pumped alkali laser (XPAL) system has been demonstrated in mixtures of Cs vapor, Ar, and ethane, by pumping Cs-Ar atomic collision pairs and subsequent dissociation of diatomic, electronically-excited CsAr molecules (exciplexes or excimers). Because of the addition of atomic collision pairs and exciplex states, modeling of the XPAL system is far more complicated than classic diode pumped alkali laser (DPAL) modeling. In this paper we discuss BLAZE-V time-dependent multi-dimensional modeling of this new laser system including radiative transport and parasitic loss effects and compare with pulsed experiments. Predictions of continuous wave (cw) system performance are presented.

1. Introduction

The exciplex pumped alkali laser (XPAL) system has been demonstrated by Readle et. al. [Readle 2008, 2009a-b, 2010a-b] in mixtures of Cs vapor, Ar, and ethane, by pumping Cs-Ar atomic collision pairs and subsequent dissociation of diatomic, electronically-excited CsAr molecules (exciplexes or excimers). The 5-level variation of the pulsed XPAL system has also been modeled by Palla et. al. [Palla, 2010]. Because of the addition of atomic collision pairs and exciplex states, modeling of the XPAL system is far more complicated than classic diode pumped alkali laser (DPAL) modeling. The development of a time-dependent finite-volume model including transport, thermal, radiative, and kinetic effects appropriate for the simulation of a cylindrical closed cell XPAL system is presented. The model equations and solution scheme are described in detail and a structured tessellation of the relevant physical domain is demonstrated as an appropriate grid for the required simulations. An improved kinetic set appropriate for modeling XPAL systems is presented. A two-dimensional, time-dependent baseline simulation of a pulsed XPAL is presented and compared to data. Good agreement is achieved on the time gap between pump and laser pulses, laser pulse full width at half maximum, and laser pulse rise time. Parametric simulations of pulsed XPAL system configurations similar to that of the baseline case, given both four and five-level laser operation, are presented in which good agreement is obtained with outcoupled laser energy as a function of absorbed pump energy data. Various system configurations based on four or five-level laser operation and two possible pumping schemes are examined in detail. The potential impact of parasitic losses on certain system configurations is modeled and discussed. Performance potential of a continuous wave four-level operation system is predicted.

2. Model Description

BLAZE-V [Palla, 2010], the multi-physics model described and employed in the current study, is a multi-dimensional, transient, finite-volume, plasma-kinetic, fluid-dynamic model and was adapted and extended to model time-dependent specie molecular continuity, including kinetics and diffusion, energy conservation, and radiation transport including interaction with the gain media and boundaries over a two or three-dimensional computational domain appropriate to a non-flowing XPAL configuration. The model is capable of generating and solving over appropriate Cartesian (structured) and unstructured grids. The transient finite volume model functions in part by

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iteratively solving the molecular specie diffusion–reaction continuity equation (advective molecular transport is assumed insignificant in this case) for all finite control volumes at each successive iteration of each time step until a convergence criterion has been satisfied,

\[
\frac{\partial N_i}{\partial t} = \nabla \cdot \mathbf{D}_i \nabla N_i + \sum_r \alpha_i \beta_j \sum_j N_j \beta_{j,r} \quad E.1
\]

simplified as,

\[
\frac{\partial N_i}{\partial t} = \mathbf{D}_i \nabla N_i + \sum_r \alpha_i \beta_j \sum_j N_j \beta_{j,r} \quad E.2
\]

where \(N_i\) is the concentration of the \(i\)th specie, \(\phi\) is a closed control volume, \(\mathbf{D}_i\) is the diffusion coefficient of the \(i\)th specie on the closed boundary of the control volume, \(A\) is the closed boundary of the closed control volume, \(f_i\) is a source term coupling the \(i\)th molecular continuity equation to the radiation fields, the final summation term represents a summation over all \(r\) reactions, \(\alpha_i\) is the stoichiometric coefficient of the \(i\)th specie in the \(r\)th reaction, \(\beta_{j,r}\) is the exponent of the \(j\)th specie on the left hand side of the \(r\)th reaction. Note that, in general, Equations 1 and 2 are non–linear as an exponent of the \(i\)th specie in any of the \(r\) reactions, \(\beta_{j,r}\), in a transport equation for the \(i\)th specie, may exceed unity. The equations are linearized via,

\[
\forall \beta_{j,i,r} > 1, \quad N_j^{\beta_{j,i,r} - 1}
\]

where the product of more than one specie concentration at the next time step in an equation for that specie is approximated by the product of the specie concentration at the next time step and the specie concentration at the current time step (a fixed constant), \(N_j^*\), to the power \(\beta_{j,i,r} - 1\). The molecular/radiation interaction term, \(f_i\), is defined as a sum over all modeled photon types via,

\[
\Psi_i \equiv \sum_k \chi_{k,i} A_k N_k^u p_k \phi_k \sigma_k \left( N_k^u - \frac{G_{u,k}}{G_{l,k}} \prod_j N_j^l \right)
\]

where \(\chi_{k,i}\) is the stoichiometric coefficient of the \(i\)th specie with respect to emission of the \(k\)th photon type by the upper state \(N_k^u\), \(A_k\) is the spontaneous emission coefficient of the upper state \(N_k^u\), \(p_k\) is the number of possible modes into which \(N_k^u\) may spontaneously emit a photon, \(\phi_k\) is the flux per area of the \(k\)th photon type, \(\sigma_k\) is the collisionally broadened stimulated emission cross-section for the upper state \(N_k^u\) with respect to the \(k\)th photon type, \(G_u\) and \(G_l\) are the upper and lower states’ statistical weights respectively, and the final finite product term is present to account for the inclusion of the photo-association pump mechanism, i.e. \(\text{Ar} + \text{Cs} + \hbar\nu\) requires the product of Cs and Ar concentrations (this term simplifies to the concentration of the single lower state for all other cases).

The time–dependent radiation transport equation is applied to each control volume and solved iteratively in succession with the specie molecular continuity equation,

\[
\frac{\partial \phi_k}{\partial t} = \nabla \cdot \left( \mathbf{D}_k \nabla \phi_k \right) \quad E.5
\]

where \(\zeta\) is a constant vector indicating the direction of propagation for the \(k\)th photon type and \(\Omega_k\) is a source term coupling the radiation fields to the kinetics (physics similar to \(f_i\) defined in Equation 4).

The time–dependent energy transport equation is also applied to each control volume and solved iteratively in succession with the other conservation equations (again advective transport is assumed insignificant),

\[
\frac{\partial (\rho e)}{\partial t} = \nabla \cdot \left( \mathbf{D}_e \nabla e \right) - \nabla \cdot \left( \kappa \nabla T \right) - \nabla \cdot S_{\text{re}} \quad E.6
\]

where \(\rho\) is the gas density, \(e\) is the gas specific energy density (energy per volume per mass unit), \(\phi\) is the control volume, \(\kappa\) is the gas thermal conductivity, \(T\) is the gas temperature, and \(S_{\text{re}}\) is a constant volumetric reaction energy source term. Given the null velocity field, the divergence of the gas kinetic energy is uniformly zero over all space
and time and kinetic energy may be excluded from the gas specific energy density variable, thereby making it solely a function of thermal energy via

\[ e \equiv C_p T \]  \hspace{1cm} E.7

where \( C_p \) is the gas specific heat for constant pressure and \( T \) is the gas temperature. Equation 6 may be simplified to

\[ \int \int \int \int \int \int \frac{\partial}{\partial t} \rho (\rho e) \, d\varphi = \int \int \int \int \int \left( \kappa \nabla T \right) \cdot \mathbf{n} \, dA - \int \int \int \mathbf{S}_{i} d\varphi \]  \hspace{1cm} E.8

The sparse linear systems constructed as described above for each variable at each iteration of each time step are solved using the SuperLU library sparse linear system sequential solver [Demmel, 1999] and either linear or inertial under-relaxation is typically applied to each variable to ease convergence.

XPAL simulations described in the present work are based on the experimental configuration used by Readle et al. [Readle 2008, 2009a-b, 2010a-b] which is illustrated in Figure 1. There are several elements of this configuration critical to the modeling work presented here.

1. The system is pumped by a several mJ tunable pulsed laser with short (several ns) pulse widths.
2. The gain cell is held at a fixed temperature of interest and is immediately adjacent to the total reflector.
3. The output coupler is on the order of a meter distant from the gain cell, thereby limiting its interaction with the gain media over the very short durations of many of the experiments and simulations.

![Figure 1 Experimental pulsed XPAL configuration [Readle, 2009a] modeled in the research presented herein.](image)

For conceptual clarity, Cs-Ar interaction potentials including illustrations of the two Cs-Ar XPAL system pumping pathways and the four- and five-level laser operation mechanisms are shown in Figure 2. The four- and five-level laser operation mechanisms are further illustrated in Figure 3. A more substantial discussion of these mechanisms and their development and similarities to DPAL systems may be found in [Readle 2008, 2009a-b, 2010a-b].
Figure 2 Interaction potentials of Cs-Ar. The arrows indicate the various pumping pathways, with two variations of the XPAL scheme shown. Laser action at 852.1 nm and 894.3 nm correspond to four- and five-level operation respectively.

Figure 3 Pictorial representations of four- and five-level XPAL operation respectively using either pumping pathway.

The model uses a kinetic set including nine species: Ar, Cs(6^2S_1/2), Cs(6^2P_1/2), Cs(6^2P_3/2), CsAr(X^2Σ^+_1/2), CsAr(A^2Π^+_1/2), CsAr(A^2Π^+_3/2), CsAr(B^2Σ^+_1/2), and C_2H_6. All relevant thermodynamic data for the included species is contained in the model. The model further contains 12 reactions.

\[
\begin{align*}
\text{Cs}(6^2S_{1/2}) + \text{Ar} & \leftrightarrow \text{CsAr}(X^2Σ^+_1/2) & k_i = 1.67 \times 10^{-11} \ T^{1/2} e^{-14.39/T} & \text{Keq} = k_i/k_b = 2.47 \times 10^{-22} e^{-14.39/T} \ (R.1-2) \\
\text{Cs}(6^2P_{1/2}) + 2 \text{Ar} & \leftrightarrow \text{CsAr}(A^2Π^+_1/2) + \text{Ar} & k_i = 2.00 \times 10^{-32} & \text{Keq} = k_i/k_b = 1.84 \times 10^{-23} e^{681.31/T} \ (R.3-4) \\
\text{Cs}(6^2P_{3/2}) + 2 \text{Ar} & \leftrightarrow \text{CsAr}(A^2Π^+_3/2) + \text{Ar} & k_i = 1.00 \times 10^{-32} & \text{Keq} = k_i/k_b = 4.71 \times 10^{-23} e^{734.65/T} \ (R.5-6) \\
\text{Cs}(6^2P_{3/2}) + \text{CsAr} & \leftrightarrow \text{Cs}(6^2P_{1/2}) + \text{CsAr} & k_i = 8.20 \times 10^{-11} \ T^{1/2} e^{-358.13/T} & \text{Keq} = k_i/k_b = 1.27 \times 10^{-22} e^{358.13/T} \ (R.7-8) \\
\text{Cs}(6^2P_{1/2}) & \rightarrow \text{Cs}(6^2S_{1/2}) & k_i = 3.27 \times 10^7 & \ (R.9) \\
\text{Cs}(6^2P_{3/2}) & \rightarrow \text{Cs}(6^2S_{1/2}) & k_i = 2.87 \times 10^7 & \ (R.10) \\
\text{Cs}(6^2P_{3/2}) + \text{C}_2\text{H}_6 & \leftrightarrow \text{Cs}(6^2P_{1/2}) + \text{C}_2\text{H}_6 & k_i = 4.76 \times 10^{-11} \ T^{1/2} & \text{Keq} = k_i/k_b = 0.5 \ e^{797.351/T} \ (R.11-12)
\end{align*}
\]

(Units: cm^3 s^{-1} for R.1, R.7 and R.11. cm^6 s^{-1} for R.3 and R.5. s^{-1} for R.9 and R.10. cm^3 for R.2, R.4, R.6 and R.8)

The Cs + Ar \leftrightarrow CsAr association and dissociation rate constants were estimated using the quasi-static approximation and a simple collision frequency model. Consider the case where CsAr is excited by light at a wavelength near the maximum of the blue wing absorption feature (11960 cm⁻¹). The Mulliken difference potential [Merritt, 2009] for the B^2Σ^+_1/2 \rightarrow X^2Σ^+_1/2 transition, Figure 4, indicates that Cs–Ar collision pairs that are separated by \( R_0 = 4.5 \pm 0.5 \) Å will be excited. The quasi-static approximation gives the number density for the collision pairs that satisfy this condition [designated CsAr(X)] by the expression (as done by [Hedges, 1972] and [Eden, 1976]).

\[
\text{[CsAr}(X)\text{]} = 4\pi R_0^2 \Delta R [\text{Cs}(2S)] [\text{Ar}] \exp \left( \frac{\Delta E_X}{k_b T} \right) \tag{E.9}
\]

where \( k_b \) is the Boltzmann constant, \( R_0 \) is the optimal internuclear separation (4.5 Å for this example) and \( \Delta R \) is the range of distances over which the resonance absorption condition is maintained (1 Å). Note that \( \Delta R \) is implicitly dependent on the bandwidth of the excitation source. The exponential term in Equation 9 involves \( \Delta E_X \), which is the difference in the ground state potential energy defined by

\[
\Delta E_X = U_X(R = \infty) - U_X(R = R_0) \tag{E.10}
\]

Equation 9 can also be used to define an effective equilibrium constant for the absorbing dimer (\( K_{eq}^X \)) such that
The rate constants for association ($k_f^X$) and dissociation ($k_b^X$) are then related to the equilibrium constant by detailed balance, such that $K_{eq}^X = \frac{k_f^X}{k_b^X}$.

Using the parameters given above and a value of $\Delta E_X = -10 \text{ cm}^{-1}$ from the ground state potential energy curve of [Merritt, 2009], Equation 11 predicts an equilibrium constant of $K_{eq}^X = 2.5 \times 10^{-22} \text{ cm}^3$ at a temperature of 470 K (a typical temperature for the laser demonstration experiments). It is of interest to note that the constant from Eq. 11 is more than an order of magnitude greater than the equilibrium constant for formation of the bound Cs–Ar complex calculated using the methods of statistical mechanics [Merritt, 2009]. This difference occurs because the effective equilibrium constant of Eq. 11 counts both bound and unbound collision pairs.

The association rate constant can be estimated using $R_0$ to define the collision radius. With this approximation the rate constant is given by

$$k_f^X = \sigma \langle v \rangle \exp \left( \frac{\Delta E_X}{k_b T} \right)$$

where $\sigma$ is the collision cross section ($\pi R_0^2$) and $\langle v \rangle$ is the average collision velocity. The latter is given by

$$\langle v \rangle = \frac{8 k_b T}{\pi \mu}^{1/2}$$

where $\mu$ is the reduced mass of the collision pair. For $T=470 \text{ K}$, Eq. 12 predicts an association rate constant of $k_f^{X} = 3.5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. Combined with the effective equilibrium constant, this yields a rate for dissociation of $k_b^{X} = 1.4 \times 10^{12} \text{ s}^{-1}$.

The same method can be applied to the formation and dissociation of the excited CsAr($B^2\Sigma^+_{1/2}$) collision complex. The differences between the rate constants for CsAr($X^2\Sigma^+_{1/2}$) and CsAr($B^2\Sigma^+_{1/2}$) derive from the differences in the interaction potential energy curves and the atomic state degeneracies. The effective equilibrium constant for CsAr($B^2\Sigma^+_{1/2}$) can be approximated by the equation

$$K_{eq}^B = \frac{[\text{CsAr}(B)]}{[\text{Cs}(6^2P_{3/2})][\text{Ar}]} = 2\pi R_0^2 \Delta R \exp \left( \frac{\Delta E_B}{k_b T} \right)$$

The factor of two difference between this expression and Equation 11 arises because the collisions between Cs($6^2P_{3/2}$) and Ar sample both the $B^2\Sigma^+_{1/2}$ and $A^2\Pi_{3/2}$ potential energy curves. The CsAr($B^2\Sigma^+_{1/2}$) potential is entirely repulsive, such that $\Delta E_X = -249 \text{ cm}^{-1}$ at $R_{eq}=4.5 \text{ Å}$. Application of Equation 14 then yields $K_{eq}^B = 5.9 \times 10^{-21} \text{ cm}^3$. As the electronic transition does not significantly perturb the radial momenta of the nuclei (Franck–Condon principle), the dissociation rate for CsAr($B^2\Sigma^+_{1/2}$) will be closely similar to that for CsAr($X^2\Sigma^+_{1/2}$). Combining $k_b^{B} = 1.4 \times 10^{12} \text{ s}^{-1}$ with the effective equilibrium constant provides an estimate for the association rate constant of $k_f^{B} = 8.5 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$. The CsAr($A^2\Pi_{3/2}$) and CsAr($A^2\Pi_{1/2}$) rates as a function of temperature are shown in Figure 5 and are computed from a statistical mechanics approach [Merritt, 2009] since these states are not directly involved in the photoexcitation process when the system is pumped at 837 nm. The formation of the CsAr($A^2\Pi_{1/2}$) and CsAr($A^2\Pi_{3/2}$) states is via three-body collisions and a gas kinetic rate of $2 \times 10^{-32}$ is assumed for the CsAr($A^2\Pi_{1/2}$) state while a rate of $1 \times 10^{-32}$ is assumed for the CsAr($A^2\Pi_{3/2}$) state as CsAr($A^2\Pi_{3/2}$) and CsAr($B^2\Sigma^+_{1/2}$) both correlate to the Cs($6^2P_{3/2}$) state.
3. Grid Study

The cell geometry of interest in the current study is cylindrical with the pump and laser beam optical axes perpendicular to the circular cross-section. The diameter of the circular face is 2.5 cm for all cases in the present study. Cell lengths of 6 and 10 cm are examined in the present study. Given that the pump beam is radially non-uniform and that the model considers radiative transport in the direction of the optical axis, a computational domain radially symmetric about the optical axis was chosen. A coarse example grid is shown in context with key model features highlighted, Fig. 6.

A previous grid study for the system configuration presently under consideration [Palla, 2010] indicated that 75 cells in the radial dimension were sufficient to limit the error in calculated system performance metrics due to radial discretation to less than 0.1%. The previous study also indicated that grid density requirements were most severe for the lowest efficiency cases. In order to determine axial grid density requirements, calculations using a 10 cm long gain cell initially at 435 K were performed in which five-level XPAL lasing was modeled with a 836.7 nm pump beam for various axial grid densities. The pump beam intensity was specified in order to achieve a low time integrated efficiency of ~0.1%. The relative variation in absorbed pump energy, outcoupled laser energy, and efficiency were calculated as the axial cell count was increased in increments of 10. Calculations indicated that the variation in these performance metrics with varying axial grid density was limited to less than 0.1% for an axial cell count of 150, Fig. 7. The resulting r×z, 75×150 cell grid is comprised of 11250 rectangular cells.
4.1. Pulsed XPAL Baseline Calculation

To test the performance of the new model, the baseline case simulation used in [Palla, 2010] was reproduced. The 5-level XPAL baseline simulation was performed for a 2.5 cm diameter, 6 cm long cell heated to 423 K. A 836.7 nm pump beam with a Gaussian temporal profile with a 4.3 ns FWHM and an elliptical spatial profile with a FWHM of 3.5 mm in the X direction and 2.5 mm in the Y direction was assumed (X and Y are mutually orthogonal and orthogonal to the optical axis here). In order to facilitate radially symmetric modeling, the elliptical spatial profile was translated into a radially symmetric profile by using a radial FWHM corresponding to a circle with the same area as the ellipse defined by the reported orthogonal FWHM values. The pump pulse was specified as a function of time in the model such that the time at which the peak intensity occurs coincides with the time at which the peak pump intensity was measured in the corresponding experiment [Readle, 2009b]. The normalized predicted pump pulse and laser pulse intensities are compared to data in Fig. 8. Note that the modeled laser pulse results were time shifted to the right by an amount corresponding to an estimate of the time required for the travel time between the gain media and the detector. The model and data are in good agreement.

In particular the data indicates a pump peak to laser peak time of 7.06 ns while the model predicts 5.79 ns, the data indicates a laser pulse rise time of 1.3 ns while the model predicts 0.59 ns, and the data indicates a laser pulse FWHM of 1.8 ns while the model predicts 2.00 ns, Fig. 8. The inaccuracy in predicted pump peak to laser pulse peak time may be partially attributed to inaccuracy in the estimate of the gain media to detector travel time. Pump power into the cell, pump power reflected out of the cell, and outcoupled laser power recorded at the z=0 plane in addition to the volumetric power absorption rate are plotted as a function of time in Figure 9. The time shift between the plotted pump pulses corresponds to the round trip travel time through the cell, Fig. 9. Calculations indicate that the peak power absorption rate is approximately 2-3% of the peak pump pulse power flux, and that the peak laser power is approximately 10% of the peak power absorption rate, Fig. 9. In the illustrated calculation, the applied pump pulse energy is 1.33 mJ, the total absorbed pump energy is 38.96 μJ and the total extracted laser energy is 1.99 μJ. Laser efficiency, as characterized by extracted laser energy divided by absorbed pump power, was predicted to be 5.11%. It is important to note that the XPAL experiments modeled here are initial experiments with a low absorption cross-section gas mixture and only a double pass pump; we estimate that dramatic improvements in efficiency are achievable with multi-pass cell configurations that will be employed in future experiments.
A critical difference between the simulation illustrated above and the similar case in [Palla, 2010], is that while the previous model used the gain equals loss condition and an $r_1r_2 = 0.5$ value (the product of resonator mirror reflectivities) as a lasing condition, the present model correctly predicts system behavior assuming only a total reflector and no outcoupler mirror. As stated earlier, in the experimental configuration, the outcoupler mirror is situated sufficiently far from the gain media that it does not likely participate in the lasing process over the measured time scale. For significant coupling of the outcoupler mirror and the laser radiation field to occur, initial laser radiation would need to travel from the gain cell to the detector, back to the gain cell, and again to the detector over the very short time scale between the measured pump pulse peak and laser pulse peak. Therefore, data, and now the present model imply that feedback from the single total reflector is sufficient to saturate the gain media and give rise to laser action. Furthermore, if the single total reflector is disabled in the model, significant (i.e. a value that would be measurable experimentally) laser action is not predicted to occur. However, it should be noted that while the outcoupler mirror does not appear to couple to the induced laser radiation field experimentally, recorded laser power and energy values are still transmitted through the outcoupler mirror. Given that all pulsed experiments modeled in this paper used a 50% outcoupler, when subsequent calculated pulsed system laser powers and energies are compared to data, they are reduced by 50% in order to simulate transmission through the outcoupler mirror. Surface plots of gain at various time steps of interest illustrate the effect described above, Fig. 10. While lasing in the core of the cell, the gain is predicted to saturate to $\sim 114 \text{ cm}^{-1}$, while the previous model [Palla, 2010], using the gain equals loss condition, predicted a saturated gain value in the cell core of $\sim 6 \text{ cm}^{-1}$. Note that if lasing is disabled in the model, peak small signal gain would rise to several thousand cm$^{-1}$ for the present case.
Figure 10  BLAZE-V calculated gain as a function of radial and axial location for various relevant time steps for the 423K, 6 cm cell, five-level, 836.7 nm pump, ~40 µJ absorbed pump energy, and 0.2958 cm radial FWHM pump baseline case. (Note that axial location is increasing left-to-right and radial location is increasing bottom-to-top)

Surface plots have also been used to illustrate the pump pulse traversing the cell, Fig. 11.

Figure 11  Sum of BLAZE-V calculated left and right running pump intensities as a function of radial and axial location for various relevant time steps for the 423K, 6 cm cell, five-level, 836.7 nm pump, ~40 µJ absorbed pump energy, and 0.2958 cm radial FWHM pump baseline case. (Note that axial location is increasing left-to-right and radial location is increasing bottom-to-top)

Plots of the sum of the left and right running laser intensity at relevant time steps illustrate laser action beginning at the core of the cell and then expanding slightly outward, Fig. 12. Also of interest is the manifestation of the single reflection laser mechanism where the laser intensity is much higher at the cell end opposite the total reflector, Fig. 12. This is the result of right running laser intensity amplification, reflection at the single mirror, and subsequent left running laser intensity amplification increasing right to left, Fig. 12.
Predicted time traces of the left and right running pump and laser intensities at the core of the cell are shown in Figure 13, corresponding to \( z=0 \), and in Figure 14, corresponding to \( z=6 \) and the surface of the total reflector. At \( z=0 \), Fig. 13, the right and left running pump intensities profiles are separated by the round trip transit time of the cell, while at \( z=6 \), Fig. 14, the right and left running pump intensities are equal as would be expected at the surface of a total reflector. At \( z=0 \) the right running laser intensity is negligible as there is nothing feeding it from the left, while the left running laser intensity is significant as it is fed by laser intensity amplified by a near full round trip through the cell, Fig. 13. At \( z=6 \), the right and left running laser intensities are nearly equal as would be expected, Fig. 14.

The time evolution of the various Cs states’ concentrations are nearly identical at \( z=0 \) and \( z=6 \), Figures 15 and 16 respectively, with the upper and lower laser levels’ concentrations reaching nearly identical values while lasing as would be predicted from their respective statistical weights. However, close examination will show while the
difference between the upper and lower laser levels' concentrations is negligible at \( z=0 \) in Figure 15, a small difference is visible at \( z=6 \) in Figure 16. This is the result of a less strongly saturated gain medium at the \( z=6 \) end of the cell as a result of the lower local laser intensity.

The time evolution of the various Cs-Ar states’ concentrations are also nearly identical at \( z=0 \) and \( z=6 \), Figures 17 and 18 respectively, although the concentrations of the CsAr(A\(^2\Pi_{1/2}\)) state are predicted to be higher than was predicted in [Palla, 2010]. This is likely the result of the switch to a three-body gas kinetic formation mechanism for CsAr(A\(^2\Pi_{1/2}\)) in the present work.
For further illustration of the varying degree of gain media saturation along the optical axis, the positive component of the temporal gain profiles at \( z=0 \) and \( z=6 \) are compared in Figure 19.

![Figure 19 BLAZE-V calculated gain as a function of time at \((z,r) = (0,0)\) and \((6,0)\) for the 423K, 6 cm cell, five-level, 836.7 nm pump, \(\sim\)40 \(\mu\)J absorbed pump energy, and 0.2958 cm radial FWHM pump baseline case. (Note that only the positive portions of the gain profiles are shown)](image)

Given the demonstrated capability of the model to accurately predict the behavior of the baseline case, the model was employed to study other cases of interest.

### 4.2. Parametric Study of Pulsed Five-Level XPAL Pumped at 836.7 nm

The model as described in the previous sections was used to perform a parametric study of five-level XPAL system performance as a function of absorbed pump energy for a 10 cm long cell at 435K case pumped at 836.7 nm. Although previous calculations assumed that the pump beam employed experimentally had a radial mean FWHM of 0.2958 cm, testing of the model with the system configuration described above indicated greater agreement with data using a radial FWHM value of 0.25 cm. Given the possibility of variations in experimental pump beam performance, this small relative change in the specification of pump beam characteristics was deemed reasonable. An initial parametric study was performed for the 435K, five-level, and 836.7 nm pump beam case in which pump beam intensity was varied over a range sufficient to produce a set of cases with absorbed pump energy values in a range similar to that of an experimental study in [Readle, 2009a]. A comparison of the calculated extracted laser energy values as a function of absorbed pump energy to data produced good agreement on threshold energy and slope efficiency, Fig. 20. An additional parametric study was performed for this configuration in which the pump beam was spatially uniform, Fig. 21. Comparing the two parametric studies, we find that the spatially uniform pump cases in which lasing occurs give rise to a greater slope efficiency as no pump energy is deposited in gain media regions not accessed by laser action. Further study indicated that as the pump intensities and the resulting absorbed pump energies are increased in the spatially non-uniform pumping case, the slope efficiency will asymptote to the slope efficiency of the uniform pump case as an increasing fraction of the cell and deposited pump energy is accessed by laser action.
The time evolution of both the Cs and Cs-Ar states’ concentrations at the cell core and $z=0$ for a 120 µJ absorbed pump energy example case, Figures 22 and 23 respectively, are similar to that found in the baseline case, Figures 15 and 17 respectively.

The time evolution of the right and left running pump and laser intensities at the cell core and $z=0$ for a 120 µJ absorbed pump energy example case, Figure 24, are similar to that found in the baseline case, Figure 13.

The 435K, five-level, and 836.7 nm pump beam example case presented in Figures 22-24 with 120 µJ absorbed pump energy was used to demonstrate a new amplified spontaneous emission (ASE) model. Given that the region of the gain cell in which significant pump energy is deposited has a high aspect ratio geometry that is aligned along
the optical axis, any parasitic lasing mechanism acting on a substantially different axis would only be amplified over a comparatively short path length. In an attempt to model possible parasitic lasing mechanisms on an axis sufficiently close to the optical axis to access a large fraction of the high gain region but still not be measured as laser power, an additional pair of left and right running intensities was modeled at the same wavelength as the laser but specified to not reflect at the total reflector. The spontaneous emission coefficient for the added parasitic laser mechanism corresponds to the spontaneous emission coefficient for the standard lasing mechanism multiplied by $4\pi$ to simulate the hypothetical scenario of all solid angles of spontaneous emission divided by the number of possible modes feeding the parasitic mechanism. The resulting simulation did not predict sufficient intensity and energy in this additional mechanism to detract from the standard laser action, Fig. 25. More specifically, despite the increased spontaneous emission feeding this mechanism, the amplification associated with a single transit of the cell cannot compete with the amplification associated with the two-pass standard laser mechanism, Fig. 25. Further study using the new ASE model is suggested for envisioned high efficiency system configurations in which feedback of ASE intensities may be possible.

![Figure 24 BLAZE-V calculated right and left running (denoted + and – respectively) pump and laser intensities as a function of time at (z,r) = (0,0) for the 435K, 10 cm cell, five-level, 836.7 nm pump, 0.25 cm radial FWHM pump, and ~120 μJ absorbed pump energy case.](image)

![Figure 25 BLAZE-V calculated right and left running (denoted + and – respectively) pump, laser, and ASE intensities (with the ASE model enabled) as a function of time at (z,r) = (0,0) for the 435K, 10 cm cell, five-level, 836.7 nm pump, 0.25 cm radial FWHM pump, and ~120 μJ absorbed pump energy case.](image)

4.3. Parametric Study of Pulsed Five-Level XPAL Pumped at 853.5 nm

Initial data has been obtained for five-level XPAL laser operation using the 853.5 nm pump pathway [Readle, 2009a]. Although outcoupled laser energy as a function of absorbed pump energy data is not available, the parametric studies presented in the previous section were repeated assuming the 853.5 nm pump pathway. The model predicts a lower absorbed energy threshold of approximately 50 μJ for this case, Fig. 26, compared to approximately 70 mJ for the 836.7 nm pump case, Fig. 20. When the parametric study was repeated using a radially uniform pump profile, an absorbed energy threshold of approximately 3 mJ was predicted, Fig. 27, which is nearly identical to the absorbed energy threshold predicted for the corresponding 836.7 nm pump parametric study, Fig. 21. The predicted slope efficiency for the uniform 853.5 nm pump configuration, 83.5% from Figure 27, is greater than for the uniform 836.7 nm pump configuration, 82.3% from Figure 21. This is likely due to the slightly higher quantum efficiency of systems using the 853.5 nm pump.
Despite the pumping of a different state in the 853.5 nm, ~80 µJ absorbed pump energy example case, CsAr(A^3Π_{3/2}), as compared to the 836.7 nm, ~120 µJ absorbed pump energy example case, CsAr(B^3Σ^+_{1/2}), the time evolution of the Cs and Cs-Ar states are nearly identical, Figs. 28 and 29 compared to Figs. 22 and 23.

4.4. Parametric Study of Pulsed Four-Level XPAL Pumped at 836.7 nm

The parametric studies presented in the previous sections were repeated assuming four-level operation using the 836.7 nm pump pathway. This was achieved by removing the ethane from the modeled mixtures while holding the argon pressure constant. Calculations indicated greatest agreement with extracted laser energy as a function of absorbed pump energy using a radial FWHM of 0.19 cm. Calculated outcoupled laser energy as a function of
absorbed pump energy results are in good agreement with data from [Readle, 2010a], Fig. 30. Note that the model predicts a higher absorbed energy threshold of approximately 160 µJ for this case, Fig. 30, compared to approximately 70 mJ for the five-level, 836.7 nm pump case, Fig. 20. This is consistent with data [Readle, 2009a; 2010a], although further study is recommended. When the parametric study was repeated using a radially uniform pump profile, an absorbed energy threshold of approximately 12 mJ was predicted, Fig. 31, which is much greater than the absorbed energy threshold predicted for the corresponding five-level, 836.7 nm pump parametric study, Fig. 21. The predicted slope efficiency for the uniformly pumped four-level, 836.7 nm pump configuration, 82.9% from Figure 31, is slightly greater than for the uniformly pumped 836.7 nm pump configuration, 82.3% from Figure 21. A priori, a much larger increase in slope efficiency was predicted for the four-level system due to the increase in quantum efficiency. Therefore further study of the predicted small increase is recommended.

Figure 30 BLAZE-V parametric study of outcoupled laser energy as a function of absorbed pump energy compared to data for the 464K, 10 cm cell, four-level, 836.7 nm pump, and 0.19 cm radial FWHM pump case.

Figure 31 BLAZE-V calculated extracted laser energy as a function of absorbed energy for the 464K, 10 cm cell, four-level, 836.7 nm pump, and ∞ radial FWHM pump case.

The time evolution of the upper and lower laser level Cs states’ concentrations for a ~170 µJ absorbed pump energy example case are consistent with theory as they reach values with a ratio near to the ratio of their respective statistical weights while lasing, Fig. 32. The time evolution of the various Cs-Ar states’ concentrations are predicted to be different for the four-level, 836.7 nm pump case, Fig. 33, compared to the five-level, 836.7 nm pump case, Fig. 23, with the CsAr(A^3Σ^1/2) concentration reaching greater values compared to the CsAr(B^3Σ^+ 1/2) concentration.
5. Parametric Study of cw Four-Level XPAL Pumped at 836.7 nm

Given the previously demonstrated ability of the BLAZE-V model to simulate critical XPAL physics with reasonable accuracy, the model was used to ascertain the continuous wave performance potential of the four-level, 836.7 nm pump system. To simulate cw, four-level operation, the configuration studied in the previous section was modified to include time invariant pump intensity and a radially uniform pump profile in addition to an 80% outcoupler mirror. Both mirrors were assumed to be 0.999 reflectors with respect to the pump beam, which was again applied along the optical axis. This was an attempt to model a higher number of pump passes, in a configuration similar to that used by [White, 1942]. Time integration was performed until steady-state was reached. This method is superior to the direct determination of a steady-state solution as it also provides information regarding the time-scales over which steady-state operation is achieved. A parametric study was performed in which the constant pump intensity was varied over a range sufficient to generate cases over a range from near threshold operation to near maximum achievable efficiency. Calculations indicate that system efficiencies near to and possibly in excess of 90% may be achievable, Fig. 34. In order to illustrate the time-scale over which steady-state operation is achieved, of interest in potential attempts to reach quasi-steady-state operation experimentally, three cases of interest with varying efficiency levels of interest were selected from the parametric study illustrated in Figure 34, and highlighted in Figure 35. The time-evolution of the laser efficiencies of cases that converge to efficiencies of 1, 10, and 50% respectively are presented in Figure 35. Calculations indicate that steady-state operation may be achieved for high efficiency cases on times scales on the order of 10’s of nanoseconds and may be achieved for lower efficiency cases on times scales on the order of 100’s of nanoseconds, Fig. 35.
Figure 34 BLAZE-V calculated steady state efficiency as a function of absorbed power for the 464K, 10 cm cell, four-level, 836.7 nm pump, and \( \infty \) radial FWHM pump case.

Figure 35 BLAZE-V calculated efficiency as a function of time and efficiency case of interest for the 464K, 10 cm cell, four-level, 836.7 nm pump, and \( \infty \) radial FWHM pump case.

For additional clarity the time evolution of the Cs and Cs-Ar states’ concentrations for the \(~10\%\) efficient steady-state case are presented in Figures 36 and 37. The results are consistent with earlier four-level pulsed results, Figs. 32 and 33.

Figure 36 BLAZE-V calculated Cs state concentrations as a function of time at \((z,r) = (0,0)\) for the 464K, 10 cm cell, four-level, 836.7 nm pump, \( \infty \) radial FWHM pump, and \(~10\%\) steady state efficiency case.

Figure 37 BLAZE-V calculated Cs-Ar state concentrations as a function of time at \((z,r) = (0,0)\) for the 464K, 10 cm cell, four-level, 836.7 nm pump, \( \infty \) radial FWHM pump, and \(~10\%\) steady state efficiency case.

6. Concluding Remarks

Diode pumped alkali lasers (DPALs) are receiving increased interest in the gas laser community as a way of taking electrically efficient diode lasers and using a gas vapor alkali cell as a brightness converter to achieve a scalable high energy laser (HEL) system. Recent experiments successfully demonstrated a variant of the DPAL system called XPAL in which mixtures of Cs vapor, Ar, and ethane create Cs-Ar atomic collision pairs that are
pumped in a blue satellite to create electronically-excited CsAr molecules (exciplexes or excimers) that subsequently dissociate to create the desired upper laser level of the alkali. Because of the addition of atomic collision pairs and exciplex states, modeling of the XPAL system is more complicated than classic DPAL modeling and in this paper we employ time-dependent, multi-dimensional modeling using the BLAZE-V code. BLAZE-V simulations of the XPAL system are in good agreement with recent pulsed experiments in which the XPAL concept was demonstrated. Further, modeling of pulsed and continuous wave cases with higher absorbed powers (simulating a multi-pass pump cell configuration) indicates that the XPAL system should be able to attain electrical-to-optical conversion efficiencies comparable to DPAL and likely even higher. Initial implementation of an ASE model has indicated that parasitic losses are likely not significant in low efficiency cases or when modeled as a single optical pass mechanism. Future modeling of these mechanisms should focus on likely high efficiency system configurations where multiple pass effects are possible. Overall, modeling results are highly encouraging for the potential of the XPAL system to scale to large alkali laser systems.

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