XPAL theory and predictions

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ABSTRACT

The exciplex pumped alkali laser (XPAL) system has been demonstrated in mixtures of Cs vapor, Ar, with and without ethane, by pumping Cs-Ar atomic collision pairs and subsequent dissociation of diatomic, electronically-excited CsAr molecules (exciplexes or excimers). The blue satellites of the alkali D₂ lines provide an advantageous pathway for optically pumping atomic alkali lasers on the principal series (resonance) transitions with broad linewidth (>2 nm) semiconductor diode lasers. The development of a simple theoretical analysis of continuous-wave XPAL systems is presented along with predictions as a function of temperature and pump intensity. The model predicts that an optical-to-optical efficiency in the range of 40-50% can be achieved for XPAL.

Keywords: exciplex pumped alkali laser, XPAL, diode pumped alkali laser, DPAL, excimer

1. INTRODUCTION

Approximately nine years ago, Krupke et al. [Krupke, 2003] demonstrated an optically-pumped atomic Rb laser operating on the resonance line at 795 nm. This scheme is commonly referred to as a diode pumped alkali laser (DPAL), and an equivalent pictorial representation for the Cs version of DPAL is illustrated in Fig. 1. Several other important studies followed the initial DPAL demonstration including theoretical modeling [Beach, 2004; Hager, 2010; Komashko, 2010], demonstration of lasing with other alkalis [Beach, 2004; Zhdanov, 2007a], lasing with helium as the 2P₃/2 → 2P₁/2 relaxer gas rather than ethane [Zhdanov, 2007b; Zweiback, 2009], a multi-diode pump scheme has produced 48 W of output power at 894 nm [Zhdanov, 2008], and a high power diode stack has produced an average output power of 145 W at 794 nm [Zweiback, 2010]. The level of interest in the laser community has risen because it appears that this laser may offer a route to extremely high power levels. The primary reason for the interest is that it allows one to use high power semiconductor laser diodes as the pump source to drive a gas laser. The disadvantage of the basic DPAL pumping scheme is that it must be pumped in a spectrally very narrow (≈ 10 GHz, or equivalently ≈ 0.02 nm) range such that only a limited portion of the semiconductor laser power will be absorbed by the alkali vapor because common semiconductor lasers, which typically emit with spectral widths of > 1000 GHz (roughly 2 nm). The ways to address this issue are to add high pressures (> 10 atm) of He gas to broaden the linewidth of the transition [Krupke, 2004], to narrow the linewidth of the pump laser, or a combination of pressure broadening and narrow linewidth diodes. While significant advances have been made in the area of narrow linewidth diode stacks [Podvyaznny, 2010], this approach dramatically increases the cost, and scaling of such narrow linewidth diode stacks to very high power is still uncertain.

The exciplex pumped alkali laser (XPAL) system approach takes a different course to this problem by invoking a molecular interaction to allow one to pump away from the atomic resonance in a broadband absorption blue satellite created by naturally occurring collision pairs such as Cs-Ar. XPAL was 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 four- and five-level variations of the pulsed XPAL system have also been modeled by Palla et al. [Palla, 2010; Palla, 2011]. A more detailed discussion of XPAL, as well as differences and similarities to DPAL systems may be found in [Readle 2008, 2009a-b, 2010a-b; Heaven, 2010; Galbally-Kinney, 2010].
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 Fig. 2. The four- and five-level laser operation mechanisms are further illustrated in Fig. 3. In this paper we present a simple theoretical model to make preliminary predictions of continuous-wave (cw) laser performance and to investigate the expected conditions to reach optical transparency in different gas mixtures and temperatures.

2. THEORETICAL MODEL

2.1 Theoretical Model and Assumptions

To enable quick studies for cw performance, a preliminary analytic model was created for the four-level XPAL system. Assumptions in the theory include:

(i) that the equilibrium between the Cs(6^2S_{1/2}) and CsAr(X^2Σ^+_{1/2}) states is maintained in steady-state lasing,
(ii) that the equilibrium between the Cs(6^2P_{3/2}) and CsAr(B^2Σ^+_{1/2}) states is maintained in steady-state lasing,
(iii) the CsAr(A^2Π_{1/2}) and CsAr(A^2Π_{3/2}) states do not play a significant role,
(iv) the Cs(6^2P_{1/2}) state does not play a significant role in the four-level system,
(v) and spontaneous emission from the CsAr(B^2Σ^+_{1/2}) state is neglected.
The species in the model are denoted as: \( N_0 \) for Cs(6\(^2\)S\(_{1/2}\)), \( N_1 \) for CsAr(X\(^2\)\(\Sigma^+\)\(_{1/2}\)), \( N_2 \) for CsAr(B\(^2\)\(\Sigma^+\)\(_{1/2}\)), \( N_3 \) for Cs(6\(^2\)P\(_{3/2}\)), and \( M \) for Ar, Fig. 4. Note that the precise species of alkali and rare gas can be replaced by other alkalis or rare gases, e.g., one could use Rb in place of Cs, and Kr in place of Ar. First we define the equilibrium fractions \( f_{10} \) and \( f_{23} \), which provide relations between \( N_j \) and \( N_0, N_0 \) and \( N_1, N_2 \)

\[
f_{10} = \frac{N_1}{N_0} = \frac{[\text{CsAr}\left(X^2\Sigma^+_{1/2}\right)]}{[\text{Cs}\left(2S_{1/2}\right)]} = \frac{g_1}{g_0} \frac{4\pi R_0^2}{\Delta R} \exp\left(-\frac{\Delta E_{10}}{k_B T}\right) [M] \quad E.1
\]

\[
f_{23} = \frac{N_2}{N_3} = \frac{[\text{CsAr}\left(B^2\Sigma^+_{1/2}\right)]}{[\text{Cs}\left(2P_{3/2}\right)]} = \frac{g_2}{g_3} \frac{2\pi R_0^2}{\Delta R} \exp\left(-\frac{\Delta E_{23}}{k_B T}\right) [M] \quad E.2
\]

where \( k_B \) is the Boltzmann constant, \( R_0 \) is the optimal internuclear separation (4.5 Å for Cs-Ar) for the blue satellite, \( \Delta R \) is the range of distances over which the resonance absorption condition is maintained (1 Å), and \([M]\) is the rare gas concentration. Note that \( R_0 \) and \( \Delta R \) are implicitly dependent on the bandwidth of the excitation source. These expressions come from quasi-static approximations for the collision pairs as done by [Hedges, 1972] and [Eden, 1976]. The factor of 2 difference in \( E.2 \) arises because collisions between Cs(6\(^2\)P\(_{3/2}\)) and Ar sample both the B\(^2\)\(\Sigma^+\)\(_{1/2}\) and A\(^2\)\(\Pi\)\(_{3/2}\) potential energy curves [Palla, 2010]. The exponential terms in \( E.1 \) and \( E.2 \) involve \( \Delta E_{ij} \), which is the difference in the potential energy between the collision pair state at \( R_0 \) and the unbound state at \( R = \infty \), and are defined by

\[
\Delta E_{10} = U_1(R = R_0) - U_0(R = \infty) \quad E.3
\]

\[
\Delta E_{23} = U_2(R = R_0) - U_3(R = \infty) \quad E.4
\]

At the peak of the blue satellite (4.5 Å), the CsAr(X\(^2\)\(\Sigma^+\)\(_{1/2}\)) potential is very slightly repulsive, \( \Delta E_{10} = 10 \text{ cm}^{-1} \) [Merritt, 2009], and the CsAr(B\(^2\)\(\Sigma^+\)\(_{1/2}\)) potential is entirely repulsive, such that \( \Delta E_{23} = 249 \text{ cm}^{-1} \) at \( R_0 = 4.5 \text{ Å} \).

We further know that the sum of the Cs containing species must obey conservation, therefore

\[
N_T = N_0 + N_1 + N_2 + N_3 \quad E.5
\]

Under lasing conditions the threshold gain \( g_{th} \) is defined by the gain-equals-loss relation

\[
g_{th} = -\frac{1}{2L_g} \ln\left(\frac{r_1 r_2}{\sigma_{30}}\right) = \sigma_{30} \left[N_3 - 2N_0\right] \quad E.6
\]

where \( L_g \) is the gain length, \( r_i \) are the mirror reflectivities, and the factor of 2 multiplier to \( N_0 \) is a result of the degeneracy ratio \( g_{th}/g_0 \) between the upper and lower states of the four-level system. The system of equations \( E.1 - E.6 \) can be solved to find the number densities of the species in the cw threshold lasing case to be:

\[
N_0 = \frac{N_T - (1 + f_{23}) g_{th} / \sigma_{30}}{3 + f_{10} + 2 f_{23}} \quad E.7
\]

\[
N_3 = \frac{g_{th}}{\sigma_{30}} + 2N_0 \quad E.8
\]

\[
N_1 = f_{10} N_0 \quad E.9
\]

\[
N_2 = f_{23} N_3 \quad E.10
\]

At conditions for optical transparency (OT), the gain is zero and therefore Eqs. \( E.7 \) and \( E.8 \) reduce to,

\[
N_{0,OT} = \frac{N_T}{3 + f_{10} + 2 f_{23}} \quad E.11
\]

\[
N_{3,OT} = 2N_{0,OT} \quad E.12
\]

An analytical rate equation approach can be used to better understand the expected laser output as a function of pump input. If we account for spontaneous radiative emission from the upper state \( N_3 \) having lifetime \( \tau_{30} \), but assume it is negligible for the intermediate exciplex state \( N_2 \), for steady state cw lasing the rate equations can be expressed as:
\[
\frac{dN_0}{dt} = 0 = \frac{\sigma_{IL}}{\nu_L} [N_3 - 2N_0] + \frac{N_3}{\tau_{30}} - k_{01}N_0M + k_{10}N_1 \quad E.12
\]

\[
\frac{dN_1}{dt} = 0 = k_{01}N_0M - k_{10}N_1 - \frac{\sigma_{IP}}{\nu_P} [N_1 - N_2] \quad E.13
\]

\[
\frac{dN_2}{dt} = 0 = \frac{\sigma_{IP}}{\nu_P} [N_1 - N_2] - k_{23}N_2 + k_{32}N_3M \quad E.14
\]

\[
\frac{dN_3}{dt} = 0 = k_{23}N_2 - k_{32}N_3M - \frac{N_3}{\tau_{30}} - \frac{\sigma_{IL}}{\nu_L} [N_3 - 2N_0] \quad E.15
\]

where \(\sigma_p\) is the cross-section for the pump beam, \(I_p\) is the intensity of the pump beam in the cell volume, \(\nu_p\) is the frequency of the pump beam, \(\sigma_L\) is the cross-section for the lasing beam, \(I_L\) is the intensity of the lasing beam in the cell volume, \(\nu_L\) is the frequency of the lasing beam, and \(\tau_{30}\) is the spontaneous emission lifetime of the upper state \(N_3\).

The cross-section for the pump beam \(\sigma_p\) is determined via knowledge of the absorption \(\alpha\) and the reduced absorption coefficient \(k\):

\[
\alpha = k[N_0][M] = \sigma_p[N_1] \quad \Rightarrow \quad \sigma_p = k \frac{[N_0][M]}{[N_1]} f_{10} \quad E.16
\]

It is interesting to note that \(\sigma_p\) becomes a simple function of the rare gas concentration \(M\), the reduced absorption coefficient \(k\), and the ratio of \(N_1\) to \(N_0\). Chen and Phelps [Chen, 1973] established a solid value of the reduced absorption coefficient of \(1.3 \times 10^{-36}\) cm\(^2\) at \(837\) nm for CsAr and this was confirmed by measurements using modern instrumentation by Readle [Readle, 2010c]. For a Cs-Ar gas mixture with \(500\) Torr of Ar at \(200^\circ\)C, \(\sigma_p = 6.5 \times 10^{-15}\) cm\(^2\).

Let us first examine the case where we just achieve optical transparency and there is no lasing, i.e., \(I_L = 0\), and the number densities can be determined from \(E.9 - E.12\). Adding \(E.12\) to \(E.13\) and eliminating the lasing term yields

\[
0 = \frac{\sigma_{IP}}{\nu_P} (N_1 - N_2) - \frac{N_3}{\tau_{30}} \quad E.17
\]

or

\[
\frac{\sigma_{IP, OT}}{\nu_P} (f_{10}N_{0, OT} - 2f_{23}N_{0, OT}) = \frac{2N_{0, OT}}{\tau_{30}} \quad E.18
\]

Rewriting \(E.18\) for \(I_{P, OT}\) provides the following for the pump intensity for optical transparency (OT, gain=0) inside the gain volume:

\[
I_{P, OT} = \frac{2\nu_P}{\sigma_p\tau_{30} (f_{10} - 2f_{23})} \quad E.19
\]

Interestingly, the number densities have cancelled out of \(E.19\) due to the ability to define what they must be in steady state for cw OT conditions \((E.9 - E.12)\). The difference \(N_1 - N_2\) is now represented by the term \(f_{10} - 2f_{23}\). Note that \(E.19\) actually represents the “intracavity” or “gain volume” pump intensity that builds up in a multi-pass cell (whereas it may be possible to actually pump or “feed” the cell with considerably lower intensity). Alternately \(E.19\) can be expressed in terms of the rare gas density by utilizing \(E.16\).

\[
I_{P, OT} = \frac{2\nu_P}{kM\tau_{30} (f_{10} - 2f_{23})} \quad E.20
\]

\(E.19\) and \(E.20\) are examined in more detail in Sect. 3.2.

Now we will examine the lasing case. Adding \(E.12\) to \(E.13\) gives

\[
0 = \frac{\sigma_{IP}}{\nu_P} (N_1 - N_2) - \frac{N_3}{\tau_{30}} - \frac{\sigma_{IL}}{\nu_L} (N_3 - 2N_0) \quad E.21
\]
For simplification, let

\[ P = \frac{\sigma_P I_P}{h \nu_P} \tau_{30} \quad \text{and} \quad L = \frac{\sigma_L I_L}{h \nu_L} \tau_{30} \quad \text{E.22} \]

Substituting E.22 into E.21, utilizing E.9 and E.10, and collecting terms for \( N_0 \) and \( N_i \) gives

\[ (Pf_{23} + L + 1)N_3 = (Pf_{10} + 2L)N_0 \quad \text{E.23} \]

or

\[ N_3 = \frac{(Pf_{10} + 2L)}{(Pf_{23} + L + 1)}N_0 \quad \text{E.24} \]

Computing the gain gives

\[ g = \sigma_L (N_3 - 2N_0) = \sigma_L N_0 \left[ \frac{P(f_{10} - 2f_{23}) - 2}{Pf_{23} + L + 1} \right] \quad \text{E.25} \]

Note that setting \( L \) and \( g \) equal to zero for optical transparency results in the recovery of E.19 above for \( I_{P,OP} \). As the lasing term \( L \rightarrow 0 \), we have the small signal gain regime and \( g \rightarrow g_0 \), thus we can now define the small signal gain coefficient for a saturation law to be

\[ g_0 = \sigma_L N_0 \left[ \frac{P(f_{10} - 2f_{23}) - 2}{Pf_{23} + 1} \right] \quad \text{E.26} \]

The corresponding gain saturation law is then

\[ g = g_0 \left[ 1 + \frac{L}{Pf_{23} + 1} \right] \quad \text{E.27} \]

When \( g \rightarrow g_0 / 2 \) we define the saturation parameter \( L_{sat} \) and the saturation intensity \( I_{L,sat} \) to be

\[ L_{sat} = Pf_{23} + 1 \quad \text{E.28} \]

\[ I_{L,sat} = \frac{h \nu_L}{\sigma_L \tau_{30}} \left( 1 + \frac{\sigma_P \tau_{30}}{h \nu_P} f_{23} I_P \right) \quad \text{E.29} \]

Analogous to the classic Rigrod derivation [Rigrod, 1965], \( I/I_{L,sat} = L/L_{sat} = (L^+ + L^-)/L_{sat} = \beta^+ + \beta^- \), and one can utilize Rigrod’s formulation to obtain a first order model of laser output power. Because of the presence of intracavity optical elements in the XPAL systems to date (windows on the alkali cell and a polarizing beam splitter for the pump beam), we will implement a modified version of Rigrod’s formulation that includes intracavity optical losses (which can be significant) [Carroll and Verdenyen, 2009]. (Note that Carroll and Verdenyen originally considered small intracavity diffraction losses, but the formulation is appropriate for any discrete intracavity optical elements that create losses inside the resonator.)

\[ \frac{I_{out}}{I_{L,sat}} = \left( \frac{1 - \delta_t}{\delta_t} \right) \left( \frac{1 - \delta_t}{\delta_t} \right) \left( \frac{1 - \delta_t}{\delta_t} \right) \left( \frac{1 - \delta_t}{\delta_t} \right) \left[ g_0 L_{eq} + \ln \left( \frac{1 - \delta_t}{1 - \delta_t} \right) \right] \quad \text{E.30} \]

where \( L_{eq} \) is the gain length, \( r_t \) are the mirror reflectivities, \( t_i \) are the mirror transmissivities, \( \delta_t \) are the loss terms from intracavity elements on either side of the gain medium, and \( I_{out} \) is the total outcoupled intensity. Note that Rigrod’s original formulation only included mirror absorption losses, but not losses from intracavity optical elements. Also observe that a simple substitution of \( R_t = (1 - \delta_t) t_i \) results in the recovery of Rigrod’s expressions.

A critical realization that must be included into the model is to recognize that the pump beam will be absorbed as it traverses through the gain medium and hence reduce the pump intensity. The reduction in pump intensity becomes progressively more pronounced with higher temperature and ultimately leads to an optically thick gain medium that cannot be lased for reasonable power inputs. To treat this aspect of XPAL, let us consider a two-pass pump beam through the gain medium, and make the simplifying assumption that the integrated average pump intensity through the
gain medium will be representative of the true intracavity intensity for the purpose of computing the laser performance. Therefore, \( E.22 \) becomes

\[
\bar{P} = \frac{\sigma_P T_P}{h \nu_P} \tau_{30} \tag{E.31}
\]

and we must use these integrated numbers in equations \( E.23 \) – \( E.30 \). Figure 5 illustrates the geometry for computing the average pump intensity. Expressions \( E.32 \) – \( E.34 \) provide information relating the intensities of the forward and backward running waves to the actual input pump beam:

\[
I_{P1}^+ = \eta_{del} I_{w1} I_{P,inp} \tag{E.32}
\]

\[
I_{P2}^+ = I_{P1}^+ e^{-\sigma_P (N_1-N_2) L_g} \tag{E.33}
\]

\[
I_{P2}^- = \frac{t_w^2 r^2 L_P}{I_{P2}^+} \tag{E.34}
\]

where \( \eta_{del} \) is the fraction of the input beam that is delivered to the front face of the alkali cell via the beam train of optics, the \( t_w \) are the alkali cell window transmissivities, and \( I_{P,inp} \) is the pump intensity input into the optical system.

Making use of \( E.32 \) – \( E.34 \), the integrated average intracavity pump intensity can then be computed as

\[
\bar{I}_P = \frac{1}{L_g} \int_0^{L_g} \left[ I_{P1}^+(z) + I_{P2}^-(z) \right] dz
\]

\[
= \frac{1}{L_g} \int_0^{L_g} I_{P1}^+ e^{-\sigma_P (N_1-N_2) z} + I_{P2}^-(z) e^{-\sigma_P (N_1-N_2) (L_g-z)} dz
\]

\[
= \frac{I_{P1}^+}{L_g} \int_0^{L_g} e^{-\sigma_P (N_1-N_2) z} + \frac{t_w^2 r^2}{L_g} e^{-\sigma_P (N_1-N_2) L_g - \sigma_P (N_1-N_2) (L_g-z)} dz
\]

\[
= I_{P,inp} \eta_{del} t_{w1} \left[ \frac{1}{\sigma_P (N_1-N_2) L_g} \right] \left[ 1 + \frac{t_w^2 r^2}{L_g} \right] \tag{E.35}
\]

Figure 6 illustrates the absorption in a two-pass Cs-Ar system as a function of temperature, and Fig. 7 shows the ratio of the integrated average two-pass intensity versus the input pump intensity (for good quality optics) as a function of temperature. An interesting feature to observe is that the integrated double-pass intensity can actually be higher than the input pump for temperatures below approximately 260°C (533 K). With creative pump and cell design it may be possible to arrange for multi-pass configurations (> 2 passes) that further enhance the integrated intracavity pump intensity. It is important to note that \( E.35 \) must be used in \( E.23 \) – \( E.31 \) for reasonable results, especially at elevated temperatures above 200°C.
2.2 Predictions from CW Theoretical Model for CW Performance

We will now examine the theoretical model in terms of comparing to available data. For a Cs-Ar laser with $r_1=0.50$ and $r_2=1.0$, the number densities of the different species $N_0 - N_3$ are shown in Fig. 8. Note that this simple analytical approach predicts number densities at 464 K of $N_0=4.24\times10^{14}$, $N_1=2.36\times10^{12}$, $N_2=1.59\times10^{12}$, and $N_3=8.48\times10^{14}$, which are very similar to the values predicted by the detailed BLAZE-V model [Palla, 2010].

One of the challenges for XPAL will be to properly absorb the broadband pump radiation on the $N_2\leftarrow N_1$ ($\Lambda\leftarrow X$) transition and a multi-pass pump configuration may be required. Using Beer’s Law, one can make an estimate of how...
many passes would be required to absorb 90% of the pump radiation. If we make the assumption that we would like 1-10 passes in a 10 cm cell, Fig. 9 shows that our operating range should likely be at a temperature of 450-550 K.

One of the critical questions for XPAL is whether or not it can be scaled to have good optical-to-optical efficiency. If we consider a double-pass system with excellent optical elements having low losses (for example, a high quality polarizing beam splitter and Brewster angle cell windows that are anti-reflection coated), then the values for the $\delta_i$ loss terms are small ($\delta_i$ of 0.059 and 0.02 were used in these calculations). Figures 10 and 11 show predicted cw performance of a 4-level XPAL system as a function of temperature and pump intensity with a 10 cm Cs-Ar cell having 1 atm of Ar. The model predicts that an optical-to-optical efficiency in the range of 40-50% can be achieved for XPAL. Figure 11 shows that there is an optimum temperature for the available pump intensity, and that in general higher pump intensity should lead to higher XPAL performance.

![Fig. 10. Predicted 4-level XPAL optical-to-optical efficiency versus input pump intensity as a function of alkali cell temperature using a 25% outcoupler.](image1)

![Fig. 11. Predicted 4-level XPAL optical-to-optical efficiency versus alkali cell temperature as a function of input pump intensity using a 25% outcoupler.](image2)

It is clear that such high pump intensities are not desired, and very hard (if not impossible) to achieve in cw systems. However, there are some options to help alleviate the high pump intensity:

1. Some alternate alkali and rare gas mixtures have better blue wing absorption characteristics than Cs-Ar (discussed further below).
2. The intensity required to reach optical transparency is inversely proportional to rare gas pressure, $E.20$, therefore higher rare gas pressure could be utilized to increase absorption and decrease intensity.
3. Multi-pass optical arrangements can be used to increase the integrated average intracavity intensity that drives the gain medium.
4. A shorter gain length cell may enable high temperature cells to be used more efficiently in multi-pass pump configurations.

Through a combination of the above options, one could likely push the pump intensities significantly lower.

Let us now explore the question: are there other alkali and rare gas mixtures that are more favorable in terms of the required pump intensity? To answer this we need to have measurements of the absorption of the collision pair, for which data is sparse. Chen and Phelps [Chen, 1973] established a solid value of the reduced absorption coefficient of $1.3 \times 10^{-36}$ cm$^5$ at 837 nm for CsAr and this was confirmed by measurements using modern instrumentation by Readle [Readle, 2010c]. Galbally-Kinney et al. [Galbally-Kinney, 2010] provide transmission data for a Rb-Kr gas mixture, and we extract a value of $1.66 \times 10^{-36}$ cm$^5$ from this data for the reduced absorption coefficient at 758 nm. However, we note that very little other transmission as a function of wavelength (blue satellite) data appears in the literature for alkali – rare gas mixtures. Blue satellite absorption data exist for Cs-Kr [Readle, 2010c], Cs-Xe [Eden, 1976; Readle, 2010c], Cs-Ar-Kr...
[Readle, 2010c], and mixtures that include an absorption enhancer such as ethane [Galbally-Kinney, 2010; Readle, 2010c]. While calculations can be made to provide estimates of these absorption coefficients [Baylis, 1969; Heaven, 2010; Readle, 2010c], the theoretical predictions appear to be good to only within a factor of 2 and therefore we recommend that more absorption measurements are needed for other alkali–rare gas mixtures to better understand which gas mixtures will be most suitable for scaling.

An interesting trade-off should be noted here: on first inspection of Figs. 6 and 9, it is easy to conclude that perhaps one would prefer a higher temperature to absorb all of the pump intensity in as few passes as possible, however, it may in fact be advantageous to work at lower temperatures where multiple passes through the gain volume would represent a higher integrated average pump intensity. There may be an optimal balance between temperature and integrated average pump intensity, which will in part be influenced by the number of passes. This possibility should be investigated in the future.

To get an idea of whether other gas mixtures can be useful, we compare the predicted pump intensity for optical transparency for Cs-Ar and Rb-Kr mixtures, Fig. 12. For these calculations the peak of the blue satellite for RbKr is at 4.4 Å [Heaven, 2010], the RbKr(X2Σ+1/2) potential is very slightly attractive, \( \Delta E_{10} = -25 \text{ cm}^{-1} \), and the RbKr(B2Σ+1/2) potential is repulsive with \( \Delta E_{23} = 444 \text{ cm}^{-1} \) at \( R_0=4.5 \text{ Å} \) [Heaven, 2010]. Clearly Rb-Kr appears to have a significantly lower required pump intensity than does Cs-Ar, thereby suggesting that this mixture and possibly others may be better candidates for a scaled cw XPAL system.

The results shown in Fig. 12 raise the question: what are the driving factors for the lower pump intensity required for Rb-Kr mixtures? The factors that influence \( I_P \) in E.19 are \( \tau_3 \), \( \nu_0 \), \( \sigma_{P}f_{10} \), and \( f_{23}. \) \( \tau_3 \) is 30.5 ns for Cs and 26.24 ns for Rb, which actually increases \( I_P \) for Rb by \( \approx 16\% \). \( \lambda_P = \frac{1}{v_P} \) is 837 nm for Cs-Ar and 758 nm for Rb-Kr, so this represents a 10% increase in \( I_P \) for Rb-Ar. At 200°C, \( \sigma_P \) is \( 6.5 \times 10^{-15} \text{ cm}^2 \) for Cs-Ar and \( 6.8 \times 10^{-15} \text{ cm}^2 \) for Rb-Kr, so this results in approximately a 5% drop in \( I_P. \) Essentially the net of these three factors is only a small change in \( I_P \), therefore the key factor(s) must be \( f_{10} \) and/or \( f_{23}. \) To examine these factors, they are plotted as a function of temperature in Fig. 13. Several things become clear:

1) \( f_{10} \) for Rb-Kr is larger than Cs-Ar indicating that there will be a greater concentration of the ground state Rb-Kr collision pair. This makes sense since the Rb-Kr pair is slightly attractive (\( \Delta E_{10} = -25 \text{ cm}^{-1} \)) in the blue satellite whereas the Cs-Ar pair is slightly repulsive (\( \Delta E_{10} = 10 \text{ cm}^{-1} \)). A larger concentration of the collision pair enables a larger absorption of the pump, i.e., faster pumping to the exciplex.

2) \( f_{23} \) for Rb-Kr is smaller than Cs-Ar indicating that there will be a smaller concentration of the exciplex state of Rb-Kr. This makes sense since the Rb-Kr exciplex is more repulsive (\( \Delta E_{23} = 444 \text{ cm}^{-1} \)) than the Cs-Ar exciplex (\( \Delta E_{23} = 249 \text{ cm}^{-1} \)). Therefore the Rb-Kr exciplex will break apart more rapidly. Faster dissociation of the exciplex provides rapid transfer of energy into the desired upper laser level and helps to prevent the pumping from overwhelming the dissociation rate.

3) The factor of \( (f_{10} - 2f_{23}) \) in the denominator of E.19 indicates that the effect of \( f_{23} \) is further amplified by a factor of \( 2 \), therefore the \( f_{23} \) factor becomes the more dominant term.

4) Figure 14 illustrates that the factor \( (f_{10} - 2f_{23}) \) for Rb-Kr is significantly larger than that for Cs-Ar and this quantity is the primary reason why the required pump intensity for Rb-Kr is lower, Fig. 12. Note that \( I_P \) is inversely proportional to \( (f_{10} - 2f_{23}) \), therefore as \( (f_{10} - 2f_{23}) \) decreases \( I_P \) will increase.

These results suggest that through proper choice of the alkali–rare gas pair, it may be possible to significantly reduce the required pump intensities for the XPAL system and thereby enable a more scalable device. It is noted that a mixture of multiple rare gases, as done by Readle et al. [Readle, 2010b; Readle, 2010c], may further enhance results by broadening the blue satellite.
2.3 Comparison of Theoretical Model with Absorbed Energy for Optical Transparency Data

A check on the accuracy of the theoretical model can be obtained by comparing to data, however we only have pulsed data available [Readle, 2010b] and our theory is designed for steady-state cw operation. Nevertheless, we can make some cursory checks. First, by recognizing that (i) the number density required for optical transparency is defined by $E.11$ and $E.12$, and (ii) because the energy absorbed by the exciplex is rapidly transferred to the $P_{3/2}$ state, the number density of the $P_{3/2}$ state to first-order defines the amount of energy that must be absorbed for a specified interacting gas volume (FWHM area of pump beam multiplied by length of gas cell), $V=AP_{cell}$. The energy absorbed by the gas medium can thus be estimated as

$$E_{abs,OT} = h\nu PN_{3,OT}AP_{cell}$$  \hspace{1cm} E.36

Using $E.11 – 12$ and $E.36$, we can now make a direct comparison between the theoretical estimation of the absorbed energy required for optical transparency and the experimental data of Readle et al. [Readle, 2010b], Fig. 15; the theory is in reasonable agreement with data as a function of gas cell temperature and provides some confidence that at least the non-lasing portion of the model has validity. Because of the non-equilibrium situation that exists in the short pulse situation, there is no easy way to make the simple CW theory developed herein model the existing pulsed data; therefore CW (or even quasi-CW) laser data is required to make a proper comparison against the simple theory. At this point, the best comparison we can make to the pulsed data is with the detailed model discussed by Palla et al. [Palla, 2011], where the detailed model shows good agreement with experiments. Therefore we believe that we have all of the critical physics included in this CW theory.
Fig. 15. Energy absorbed to reach optical transparency in pulsed operation; comparison between theory and data of Readle et al. [Readle, 2010b].

5. CONCLUDING REMARKS

A preliminary cw XPAL theory was developed and predicts that an optical-to-optical efficiency in the range of 40-50% can be achieved for XPAL, however high pump intensities appear to be required for high efficiency operation. The theory is also in reasonable agreement with experimental optical transparency data as a function of gas cell temperature. Rb-Kr appears to have significantly lower required pump intensity than does Cs-Ar, thereby suggesting that this mixture and possibly others may be better candidates for a scaled cw XPAL system. Thus, we recommend that more absorption measurements are needed for other alkali – rare gas mixtures to better understand which gas mixtures will be most suitable for scaling. The future simulation effort using the theoretical model will continue to refine and expand the capabilities of the theory, while at the same time expanding the predictions to help guide future experiments with long term scaling of the system in mind.

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