A simulation and experimental study of dual-wavelength dye laser amplifiers

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Abstract. AVLIS requires a high average power laser beam of multiple wavelengths. There are two general ways to achieve such a beam. One is generating different wavelength laser beams by individual dye laser MOPA chains, then spatially combining them by a beam combiner. The other is combining different wavelength laser beams into one beam first, then co-amplifying it by a common MOPA chain. The latter one is more efficient. To further develop the method, this paper demonstrates a simulation and experimental study of dual-wavelength dye laser amplifiers. The one of the most critical parameters of a dual-wavelength laser beam is the power ratio of the two constituent beams, which can vary during the co-amplification. The simulation results show that for two laser beams with small wavelength difference, the spatial combination mismatches of two beams are the more dominant factor than the spectral differences in affecting the power ratio during the co-amplification. The result of the experiment is in good agreement with the simulation.

1. Introduction

Atomic vapor laser isotope separation (AVLIS) is a promising technology for the production of Uranium-235 and many other types of isotopes. [1-5] Basically, AVLIS utilizes narrow bandwidth, high average power, tunable lasers to selectively excite and eventually ionize the target isotope atoms by two or three step photoionization. Therefore, a combined laser beam of multiwavelength is required for AVLIS.

High average power, tunable dye laser systems have been developed for decades and they are still the most suitable laser systems for AVLIS up to now. Conventionally, high power lasers of different wavelengths are generated by individual Master Oscillator Power Amplifier (MOPA) chains and then combined by dichroic mirror or other methods. An alternative way is that the laser can be combined first before they are amplified by a common power amplifier chain. The co-amplification scheme has many advantages over the conventional method, such as lower combination energy losses[6]. However, the output power ratio between the different wavelength laser beams is hard to control because of gain competition. In multi-wavelength amplifiers, combined laser beams with separated wavelengths share the same gain medium, so their gains are not independent of each other. A direct result from the gain competition is that the power ratio between the laser beams, which is a key parameter for subsequent photoionization, could change during the amplification.
Multi-wavelength lasers have been studied a lot for other applications, such as element detections [7] and coherent control of molecules, [8,9] in which gain competition effects have been discussed. However, these results cannot be directly applied to multiwavelength laser systems of AVLIS, since laser systems of AVLIS have some unique properties. First, for AVLIS it is very common to combine lasers with less than 5 nm [4] or even 1 nm of wavelength shift, while other applications do not require such small differences. Second, the lasers of AVLIS would be amplified by several amplifiers to meet the power requirements in comparison to other applications where just one amplifier or no amplifier is used. More cascade amplifiers in AVLIS might lead to a cumulative effect of gain competition. There are a few works about multiwavelength lasers relevant to the AVLIS application. Agarwalla and his colleagues [10] presented a theoretical study of multiwavelength amplification process in liquid dye gain medium by rate equations. Their results showed that gain competition was affected by both variation of wavelength and input signal power. Schütz et al. [7] and Rana et al. [6] developed a dual-wavelength dye laser system composed of one amplifier. The wavelength difference was around 6 nm in both works but their results were different. In the work of Schütz, the energy converted from pump laser to dye laser was equally divided between the two-wavelength components when the input power ratio equaled 1. In contrast, in Rana’s research, the output power ratio was of 0.3 in the case of input power ratio of 1. We consider this contradiction between the two results is due to the fact that gain competition could be affected by more than one factor. The wavelengths were carefully tuned and controlled by Schütz and Rana but another important factor, spatial effects, is not mentioned in their works. It is obvious that gain competition could be affected by the difference in position and beam size in gain mediums between the two component laser beams. We will discuss this in detail in this work. Therefore, in this paper, we present a study of dual-wavelength dye laser system composed of 5 amplifiers by simulations and experiments. We mainly focus on the variation of output power ratio during amplification process affected by wavelength effects as well as spatial effects. The theoretical model and simulation results are first introduced. Then the experimental setups and their results are presented.

2. Theoretical model
As shown in figure 1, the complete model consists of a 5-stage amplifier chain with a dual wavelength signal input. Each stage is double side longitudinally pumped by the second harmonic of a Nd:YAG laser (532 nm) and the pump powers are also shown in figure 1.

![Figure 1. Scheme of dye laser amplifier chain. Amp: amplifier.](image)

The gain medium used in the simulations and experiments is Rhodamine 6G in ethanol. Other major parameters of the simulated amplifiers are listed in table 1.

Energy levels of Rhodamine 6G are shown in figure 2. Here, S₀, S₁, and S₂ are the singlet states of the dye molecule, whereas T₁ and T₂ are triplet states, and kₘₙ represents their intersystem crossing rate. σₐ, σₑ, and σₘₙ are absorption cross section between S₀ and S₁, stimulated emission cross section from S₁ to S₀ and absorption cross section from S₁ to upper energy levels, respectively. τ₁ is the lifetime of state S₁.
### Table 1. Basic parameters of dye cell and light

| Parameters                                      | Value       |
|------------------------------------------------|-------------|
| Active Length                                  | 3 mm        |
| Pulse repetition Rate                          | 10 kHz      |
| Pumping light wavelength                       | 532 nm      |
| Input light wavelength of \( \lambda_1 \)     | 580 nm      |
| Input light wavelength of \( \lambda_2 \)     | 581 nm (in the experiment) |
| Pulse duration                                 | 30 ns       |
| Laser beam spot size in the amplifier 1 - 5   | 0.6mm, 1mm, 1.2mm, 1.5mm, 2mm |
| Dye concentration                              | 100 mg/L    |

### Figure 2. Energy levels of simulated dye molecules

The rate equations used to describe the process above are as follows:

\[
\frac{dN_i(t, x)}{dt} = \left[ \sigma_a(\lambda_p)N_0(t, x) - \sigma_e(\lambda_p)N_i(t, x) \right] \frac{I_{\text{pump}}(t, x)}{h\nu_{\lambda_p}} - \left[ \sigma_e(\lambda_{L1})N_1(t, x) - \sigma_a(\lambda_{L1})N_0(t, x) \right] \frac{I_{L1}(t, x)}{h\nu_{L1}} - \left[ \sigma_e(\lambda_{L2})N_1(t, x) - \sigma_a(\lambda_{L2})N_0(t, x) \right] \frac{I_{L2}(t, x)}{h\nu_{L2}} - \frac{N_i(t, x)}{\tau_1} - k_{ST}N_1(t, x) \tag{1}
\]

\[
\frac{dN_T(t, x)}{dt} = k_{ST}N_1(t, x) - \frac{N_T(t, x)}{\tau_T} \tag{2}
\]

\[
\frac{1}{c} \frac{\partial I_{\text{pump1}}(t, x)}{\partial t} + \frac{\partial I_{\text{pump1}}(t, x)}{\partial x} = \left[ \sigma_e(\lambda_p)N_1(t, x) - \sigma_a(\lambda_p)N_0(t, x) \right] - \sigma_a(\lambda_p)N_1(t, x) - \sigma_a(\lambda_p)N_0(t, x) \tag{3}
\]

\[
\frac{1}{c} \frac{\partial I_{\text{pump2}}(t, x)}{\partial t} - \frac{\partial I_{\text{pump2}}(t, x)}{\partial x} = \left[ \sigma_e(\lambda_{L1})N_1(t, x) - \sigma_a(\lambda_{L1})N_0(t, x) \right] - \sigma_a(\lambda_{L1})N_1(t, x) - \sigma_a(\lambda_{L1})N_0(t, x) \tag{4}
\]

\[
\frac{1}{c} \frac{\partial I_{L1}(t, x)}{\partial t} + \frac{\partial I_{L1}(t, x)}{\partial x} = \left[ \sigma_e(\lambda_{L1})N_1(t, x) - \sigma_a(\lambda_{L1})N_0(t, x) \right] - \sigma_a(\lambda_{L1})N_1(t, x) - \sigma_a(\lambda_{L1})N_0(t, x) \tag{5}
\]
\[
\frac{1}{c} \frac{\partial I_{L2}(t, x)}{\partial t} + \frac{\partial I_{L2}(t, x)}{\partial x} = [\sigma_e(\lambda_{L2})N_1(t, x) - \sigma_a(\lambda_{L2})N_0(t, x) - \sigma_s(\lambda_{L2})N_1(t, x) - \sigma_T(\lambda_{L2})N_T(t, x)]I_{L2}(t, x) \quad (6)
\]

\[
N_1(t, x) + N_0(t, x) + N_T(t, x) = N \quad (7)
\]

Here, \(N_0, N_1, \) and \(N_T\) are the population densities of state \(S_0, S_1,\) and \(T_1\) respectively, and their unit is \(\text{m}^{-3}\). \(N\) is local dye molecule number density. \(\tau_1\) and \(\tau_T\) are the lifetimes of state \(S_1\) and \(T_1\), respectively. \(I_{L1}\) and \(I_{L2}\) are the intensities of the pump beam along the \(+X\) axis. \(I_{Pump1}\) and \(I_{Pump2}\) are the intensities of the pump beams propagating in the direction of the \(+X\) axis. The unit of light intensity is \(\text{W/m}^2\). The values of the parameters in in Equations (1) – (7) are listed in Table 2.

In addition, the boundary and initial conditions are given by:
\[
I_{L1}(t, -L/2) = I_{in1}(t) \quad (8)
\]
\[
I_{L2}(t, -L/2) = I_{in2}(t) \quad (9)
\]
\[
I_{Pump1}(t, -L/2) = I_{p1}(t) \quad (10)
\]
\[
I_{Pump2}(t, L/2) = I_{p2}(t) \quad (11)
\]
\[
N_0(0, x) = N \quad (12)
\]

\(I_{in1}(t), I_{in2}(t), I_{p1}(t)\) and \(I_{p2}(t)\) are light intensities at the entrance of dye cell. Their temporal profiles are the same as our previous work [11]. \(N\) is the dye molecule number density. \(L\) is the active length.

| Quantities | Values | Unit |
|------------|--------|------|
| \(\sigma_a(\lambda_{L1})\) | \(3 \times 10^{-22}\) | \(\text{m}^2\) |
| \(\sigma_a(\lambda_{L2})\) | Depends on the wavelength | \(\text{m}^2\) |
| \(\sigma_a(\lambda_p)\) | \(4 \times 10^{20}\) | \(\text{m}^2\) |
| \(\sigma_a(\lambda_{L1})\) | \(1.6 \times 10^{20}\) | \(\text{m}^2\) |
| \(\sigma_a(\lambda_{L2})\) | Depends on the wavelength | \(\text{m}^2\) |
| \(\sigma_s(\lambda_p)\) | \(6.3 \times 10^{21}\) | \(\text{m}^2\) |
| \(\sigma_s(\lambda_{L1})\) | \(1 \times 10^{21}\) | \(\text{m}^2\) |
| \(\sigma_s(\lambda_{L2})\) | Depends on the wavelength | \(\text{m}^2\) |
| \(\sigma_T(\lambda_{L1})\) | \(4 \times 10^{21}\) | \(\text{m}^2\) |
| \(\sigma_T(\lambda_{L2})\) | \(1 \times 10^{21}\) | \(\text{m}^2\) |
| \(\sigma_T(\lambda_p)\) | Depends on the wavelength | \(\text{m}^2\) |
| \(\tau_1\) | \(3.5 \times 10^9\) | s |
| \(k_{ST}\) | \(2 \times 10^7\) | s\(^{-1}\) |
| \(\tau_T\) | \(0.5 \times 10^{-7}\) | s |
| \(h/\text{Planck constant}\) | \(6.62 \times 10^{-34}\) | \(\text{m}^2\cdot\text{kg} \cdot \text{s}^{-1}\) |
| \(c, \text{(light speed in dye solution)}\) | \(2.2 \times 10^8\) | \(\text{m/s}\) |

Equation (5) and (6), which are very similar, present the gain process of laser beams of \(\lambda_1\) and \(\lambda_2\). Actually, if the wavelength difference is very small, the cross sections of \(\lambda_1\) and \(\lambda_2\) will be approximately the same: \(\sigma_a(\lambda_{L1}) \approx \sigma_a(\lambda_{L2}), \quad \sigma_e(\lambda_{L1}) \approx \sigma_e(\lambda_{L2}), \quad \sigma_s(\lambda_{L1}) \approx \sigma_s(\lambda_{L2}), \quad \sigma_T(\lambda_{L1}) \approx \sigma_T(\lambda_{L2})\).
\[ \sigma_T(\lambda_{L,2}) \]. In addition, the left side of Equation (5) and (6) can be rewritten as the total derivative at time \( t \). Then the following relationship can be derived by dividing equation (5) by (6):

\[
\frac{dI_{L,1}(t,x(t))}{dI_{L,2}(t,x(t))} \approx \frac{l_{l,1}(t,x(t))}{l_{l,2}(t,x(t))}
\]  

Equations (13) suggest that the output power ratio should be the same as the initial input power ratio in the case of small wavelength difference.

With the increase of wavelength difference, the equation (13) will no longer be valid, and this is how wavelengths affect the gain competition. Next, we will quantitatively evaluate the influence of wavelengths with numerical simulations.

3. Simulation results

Equations (1) - (7) is calculated in the scheme of Fig. 1. The differences between the two wavelength lasers in \( \sigma_t \), \( \sigma_z \), \( \sigma_r \) are neglected because the values of these cross sections are at least one order of magnitude smaller than \( \sigma_e \). The results are summarized in Table 3. The wavelength of \( \lambda_1 \) is 580 nm. PA in the table represents power ratio (P1/P2).

### Table 3. Results of the simulation

| Wavelength of \( \lambda_2 \) | Percentage difference of \( \sigma_e \) | Input PA | PA after Amp 1 | PA after Amp 2 | PA after Amp 3 | PA after Amp 4 | PA after Amp 5 |
|-----------------------------|---------------------------------|----------|----------------|----------------|----------------|----------------|----------------|
| 580.2 nm                    | 0.3%                            | 1        | 1.01           | 1.01           | 1.02           | 1.02           | 1.02           |
| 580.5 nm                    | 1.0%                            | 1        | 1.02           | 1.05           | 1.06           | 1.07           | 1.07           |
| 581.0 nm                    | 2.5%                            | 1        | 1.06           | 1.13           | 1.16           | 1.19           | 1.2            |
| 582.5 nm                    | 5.0%                            | 1        | 1.12           | 1.25           | 1.32           | 1.40           | 1.44           |
| 585.0 nm                    | 10%                             | 1        | 1.25           | 1.54           | 1.73           | 1.92           | 2.03           |

The results in Table 3 show a cumulative effect of gain competition when dual wavelength lasers pass through a sequence of amplifiers. Although the variation of power ratio by a single amplifier is small, this change will increase stage by stage. A change in power ratio from 1 to 2 could happen after a 5-stage amplification in the case of wavelength difference of only 5 nm.

The results of \( \lambda_2 \) of 580.2 nm, 580.5 nm and 581 nm also suggest that the wavelength effects on power ratio is very limited when the two laser beams are separated by less than 1 nm.

The analysis above is based on the assumption that the two wavelength laser beams have perfect spatial overlap in the gain medium. In reality, this condition is difficult to satisfy when the two components of the incident dual-wavelength laser come from two individual oscillators. The deviation in divergence and pointing between the two initial beams could cause unexpected spatial combination mismatches in the amplifier, which would also affect the gain competition.

The simulation of spatial effects needs a 3-dimensional model. A rigorous 3D model must consider the 3D transport of the light, which would intensely complicate the calculation. To simplify the problem, all the laser beams are supposed to propagate along parallel axes. Thus, the 3D problem can be converted to a combination of an array of 1D problems, which have been described by equation (1) - (7). Take equation (5) as an example, in the 3D model, it should be rewritten as:

\[
\frac{1}{c} \frac{\partial I_{L,1}(t,x,y_j,z_k)}{\partial t} + \frac{\partial I_{L,1}(t,x,y_j,z_k)}{\partial x} = [\sigma_e(\lambda_{L,1})N_1(t,x,y_j,z_k) - \sigma_a(\lambda_{L,1})N_0(t,x,y_j,z_k) - \sigma_s(\lambda_{L,1})N_1(t,x,y_j,z_k)]I_{L,1}(t,x,y_j,z_k)
\]

At each point \((0,y_j,z_k)\), the boundary condition of \( \lambda_1 \) is given by:

\[
I_{L,1}(t,-L/2,y_j,z_k) = I_{L,1}^0(t)exp\left(-2\frac{(y_j-y_0)^2+(z_k-z_0)^2}{R_{L,1}^2}\right)
\]
It is assumed that the spatial intensity profiles of the laser beams form a Gaussian shape. \((y_0, z_0)\) is the center of the beam and \(R_{L1}\) is the beam spot size. \(I_{LL1}^0\) is the light intensity at point \((0, y_0, z_0)\).

The other equations are treated in a similar way. Then, the effects of two typical types of spatial combination mismatches are simulated. One observes the effect of the beam center deviation while the other one simulates differences in beam size, as demonstrated in figure 3.

**Figure 3.** A demonstration of the two typical spatial combination deviation.

The results of the simulation are summarized in table 4 and table 5. \(R_{L1}\) and \(R_{L2}\) are the beam spot size of \(\lambda_1\) and \(\lambda_2\), respectively.

**Table 4.** Effects of the center deviation

| Deviation value | Input PA | PA after Amp 1 | PA after Amp 2 | PA after Amp 3 | PA after Amp 4 | PA after Amp 5 |
|-----------------|----------|----------------|----------------|----------------|----------------|----------------|
| 0.1\(\times R_{L1}\) | 1        | 1.02           | 1.02           | 1.03           | 1.04           | 1.05           |
| 0.2\(\times R_{L1}\) | 1        | 1.05           | 1.09           | 1.13           | 1.17           | 1.2            |
| 0.4\(\times R_{L1}\) | 1        | 1.19           | 1.4            | 1.59           | 1.79           | 1.92           |

**Table 5.** Effects of beam size difference

| \(R_{L2}/R_{L1}\) | Input PA | PA after Amp 1 | PA after Amp 2 | PA after Amp 3 | PA after Amp 4 | PA after Amp 5 |
|-------------------|----------|----------------|----------------|----------------|----------------|----------------|
| 1.1               | 1        | 1.02           | 1.02           | 1.03           | 1.04           | 1.05           |
| 1.2               | 1        | 1.05           | 1.09           | 1.13           | 1.17           | 1.2            |

The results above show that the center deviation of 0.2\(R_{L1}\) or the beam size ratio of 1.2 will lead to the similar effect as wavelength difference of 1 nm. Considering that these two types of spatial mismatches exist simultaneously in the amplifiers, and there are other factors leading to spatial mismatches, such as energy distribution, the spatial effects on gain competition is more important in the case of two component laser beams separated by less than 1 nm.

4. **Experiment results and discussion**

Our first experimental setup is similar to Rana’s work, except that we have more amplifiers as shown in figure 4. Briefly, a dye laser beam in the experiment operate in the high-pulse repetition frequency regime, with a repetition rate of 10kHz and a pulse duration of 32ns. It is pumped by a sequence of second harmonic Nd:YAG lasers.

**Figure 4.** Experimental setup for dual wavelength co-amplification. Amp: Amplifier
The dual wavelength laser beam is generated by the combination of lasers from two individual oscillators. Attenuator 1 is used to adjust the initial power ratio of the two laser beams. A sampling light which leaks through the main beam is diffracted into two beams by a grating. Then, the output power ratio can be measured and calculated by a power meter.

The wavelength difference in the experiment is 1 nm (\(\lambda_1\) is 580 nm and \(\lambda_2\) is 581 nm), and the results of the experiment is shown in table 6

| Input power of \(\lambda_1\) | Input power of \(\lambda_2\) | Input PA | Output PA (measured) |
|-----------------------------|-----------------------------|----------|----------------------|
| 180 mW                      | 740 mW                      | 0.24     | 1.0                  |
| 400 mW                      | 750 mW                      | 0.53     | 1.3                  |
| 400 mW                      | 500 mW                      | 0.80     | 1.4                  |
| 400 mW                      | 400 mW                      | 1.0      | 2.5                  |

According to the simulation results, wavelength difference of 1 nm can only contribute to about 1.2 times the variation of power ratio, which is not enough to explain the experiment results in table 6. So, the spatial effects must also take part in the process.

In order to confirm the dominant role of the spatial effect in this situation by the experiment, we replace the original two independent oscillators with a dual wavelength oscillator to produce a laser beam of dual wavelength. The layout of the dual wavelength oscillator is shown in figure 5

![Figure 5. Layout of the dual wavelength oscillator.](image)

This oscillator is able to offer a perfectly spatially overlapped dual wavelength laser beam, so the influence of spatial mismatch could be minimized. The results of this experiment are summarized in table 7.

| Input PA | Output PA (measured) |
|----------|----------------------|
| 0.27     | 0.33                 |
| 0.62     | 0.56                 |
| 1.00     | 1.08                 |
| 1.67     | 1.46                 |
| 3.42     | 3.04                 |

The results in table 6 and table 7 suggest that the wavelength effects on the variation of PA is limited when the wavelength difference is small, which are in good agreement with our prediction in
table 3. Besides, spatial mismatches are the dominant factors affecting the gain competition in this case.

These two experiment results are also a possible explanation of the different results of Schütz and Rana’s works.

5. Conclusion
This paper demonstrates a simulation and experiment study of dual-wavelength dye laser amplifiers. We mainly focus on the variation of power ratio due to gain competition during the co-amplification process. The wavelength effects and spatial effects are studied and discussed. It is found that spatial mismatches are important factors affecting the gain competition, especially when the wavelength difference is small. In this situation, the use of dual wavelength oscillators may provide a better control of the gain competition at the amplification stages.

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