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High Gain Amplification of Femtosecond Pulses with Low Amplified Spontaneous Emission in a Multipass Dye Cell

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Abstract. We present a multipass femtosecond amplifier pumped by a Nd-YAG laser where the gain medium is given by a variable length dye cell. This system allows amplification factors up to $5 \cdot 10^6$ and output pulse energies of 150 μJ, while restricting the contribution of amplified spontaneous emission to 0.5% of the total output energy, without using any saturable absorber stage. A detailed study of the output pulsewidth as a function of the duration of the input chirped pulse is also presented.

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Amplification of femtosecond laser pulses is important whenever high intensity light pulses are required. This is the case, for example, in nonlinear frequency generation in dispersive media [1] or continuum generation [2]. Moreover, ultrashort amplified pulses are necessary in pump-probe experiments when one needs to increase the absorption coefficient with increasing peak pump intensity, while keeping low the average pump power. Femtosecond amplifiers have various configurations, which have been devised to suit the performance of the oscillators (synchronous pumped or CPM dye laser) and of the coherent source acting as a "pump". As far as the amplification of CPM dye laser pulses is concerned, two different configurations are frequently encountered: the multi-stage amplifier (pumped by a Q-switched Nd-YAG laser or by an excimer laser) and the multipass amplifier (pumped by a copper vapor laser, CVL). These two systems differ mainly in the peak intensity and the average power of the amplified pulse. It has been stressed [3] that no laser source can simultaneously provide large peak amplification and high average power so that a trade-off in repetition rate and pulse amplification is necessary when choosing the most convenient amplifier configuration. The main disadvantages of the multi-stage amplifier are the complexity of alignment, the relatively low quality of the amplified beam due to transverse pumping and the difficulty of eliminating nonlinear distortions of the beam. On the other hand, the performance of the multipass amplifier, in spite of its simple structure, can be limited by the somewhat unreliable performance of the CVL laser.

In this paper we describe a Nd-YAG amplifier with a multipass dye-cell as active medium. In spite of its low repetition rate (20 Hz) this system is a good alternative to the conventional multi-stage Nd-YAG amplifier and can be used in many applications where high energy pulses (greater than 100 μJ) are required. The main feature of our system consists in the use of a dye cell instead of a jet stream [4]. In this case it is possible to obtain an amplification factor larger than the conventional multipass jet amplifier. In particular, by taking advantage of the relatively high pulse energy of the pump laser, it is possible to properly adjust the cell length and the diameter of the active medium, obtaining in this way, without using any saturable absorber stage, a ratio of ∼200 between pulse and amplified-spontaneous-emission (ASE), larger than in other similar femtosecond amplifiers.

1. Experimental Apparatus

The femtosecond pulses to be amplified are generated by a CPM dye laser [5] pumped by a 5 W all-line argon laser, operating with Rh 590 as active medium and DODCI as saturable absorber. In this laser compensation of group velocity dispersion is provided by a four prism sequence [6]. The laser developed in our laboratory generates pulses close to 620 nm at a average power of 12 mW, tunable from 50 fs to 500 fs by increasing the negative dispersion with the four prism sequence [8].

As shown in Fig. 1, the multipass amplifier design is similar to that described in [8]. The multipass active

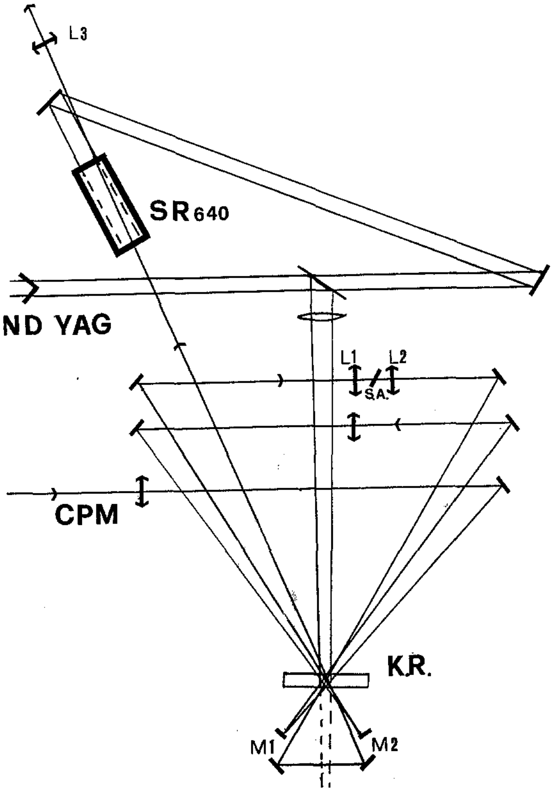

Fig. 1. Schematic of the femtosecond dye amplifier

medium is given by a dye cell with very thin (~100 μm) low-dispersion windows where a solution of $5 \cdot 10^{-4}$ M of Kiton Red in ethylene glycol is kept flowing. This dye was chosen for its strong absorption at 532 nm. In order to prevent self-lasing effects from surface reflections, the optical windows are antireflection coated. The thickness of the dye cell can be adjusted continuously between 0.5 and 2.5 mm in order to get the optimum gain length. Due to the low repetition rate, no problem arises from the flow speed of the solution in the cell.

The gain medium is excited by the doubled output (532 nm) of a 20 Hz repetition rate, self-filtering unstable resonator (SFUR) *Q*-switched Nd-YAG laser (Quanta System model S.Y.L. 202). The pumping energy is ~ 30 mJ for a pulse duration and shape that can be adjusted from 3 ns to 8 ns by properly adjusting the discharge time of the HV avalanche circuit driving the Pockels Cell. A synchronization circuit operating on the *Q*-switch unit provides the temporal coincidence between the femtosecond pulses and the pump pulse. The jitter between YAG and CPM pulses, measured with a fast photodiode and a sampling oscilloscope, is 100 ps. Thanks to the good flatness ($\lambda/4$) of the windows of the active cell of the amplifier, it is possible to pump the active medium over a large region. This is an advantage over the commonly used jet-stream amplifier where the surface inhomogeneities restrict the useful active area. The advantages of the dye cell vs the jet are in fact evident: first, it is possible to align the femtosecond beam in different portions of the pumped area during successive passes; secondly, and very important, this configuration is found to minimize the ASE contribution. It has in fact been demonstrated that, for a cylindrical amplifier, the fraction per solid angle of ASE is lowered upon increasing the pumped area [9]. In our case the pump beam is focused on a region of $7 \cdot 10^{-2}\,\mathrm{cm}^2$ with an overall intensity of $\sim 80\,\mathrm{MW/cm}^2$. This intensity, easily obtainable with a *Q*-switched Nd-YAG laser, is chosen to obtain a suitably high value of the amplification, and is ~3 times larger than the typical value obtained in a similar amplifier pumped by a focused CVL laser. We took particular care in the choice of the lenses used to focus the red beam in order to reduce gain saturation and self-phase-modulation (SPM) in the final passes. The diameter of the spot on the three pairs of passes is 400 μm, 600 μm, and 1 mm respectively. On the other hand, for the first four passes (Fig. 1), the beam is focused on the mirrors M_1 and M_2 which are close to the cell in order to keep the same beam diameter in the first and second pair of passes. Following the fourth stage a Malachite green jet can be used to suppress ASE. However with our system, very high ratios $I_{\mathrm{peak}}/\mathrm{ASE}$ are usually obtained without a saturable absorber. The transverse beam dimension in the last two passes can be varied by carefully adjusting the spacing between lenses L_1 and L_2.

After the multipass amplifier the beam can eventually be injected through a 10 cm cell pumped by 70 mJ of the Nd-YAG laser beam in a counterpropagating longitudinal configuration. The two beams travel in slightly different directions in this cell in order to take advantage of spatial discrimination between amplified pulse and ASE. Sulforhodamine 640 ($1.5 \cdot 10^{-5}\,M + 1.5\%$ Ammonix LO H_2O) was chosen as a gain medium because it has a fluorescence peak at 615 nm, close to the wavelength of the CPM laser. The output of the amplifier is collimated by the lens L_3. The total length of the multipass amplifier is 219 cm (measured from the first to the last pass), and the portion of the pump pulse passing through the longitudinal cell is properly delayed in order to maximize the pulse amplification. The polarization of the pump electric field is taken parallel to the amplified electric field in order to increase the amplification efficiency in the multipass cell [10].

2. Experimental Results

The gain factor of the amplifier in the two configurations, with and without the longitudinal cell, was determined by means of a photodiode at its output and by measuring unamplified and amplified pulses using calibrated neutral density filters. The total gain factor of the 6-pass cell alone, adjusted to an optimum length of 2 mm, is $2.5 \cdot 10^5$. This corresponds to an average gain per pass of 8. We also found that some gain saturation occurs in the last two passes. Insertion of the longitudinal cell allows a further amplification by a factor ~20. The overall measured output energy is typically 150 μJ. This energy is largely sufficient to generate the "continuum" by focusing the amplified pulse in a water cell. The contribution of ASE can be made as low as 0.5% of the pulse energy by fine adjustment of alignment and by optimization of the spatial discrimination after the second cell. In particular, the ASE emitted by the multipass cell is strongly reduced by the longitudinal cell because of the absorption of Sulforhodamine 640 at the peak wavelength of ASE of Kiton Red. These results are obtained without using the saturable absorber jet (S.A.) shown in Fig. 1.

The stability of the amplified pulse is found to depend on the shape and duration of the YAG pulse. Varying the pump pulse duration from 3 to 8 ns, the amplified pulse stability changes from 20% to 12%.

The beam emerging from the multipass cell when expanded by a positive lens on a white screen, shows a good mode profile. This profile is slightly worse after the beam has propagated through the second (longitudinal pumped) cell, probably because of gain inhomogeneities along the cell length. We believe that the use of an optical waveguide [10] to get a uniform pump beam before entering the cell could be useful in this respect.

3. Pulsewidth Measurement

Two autocorrelation traces of the pulse were measured by background-free second harmonic generation at the output of the multipass amplifier and after further amplification on the longitudinal cell. They are shown in Fig. 2. The traces are the result of the average over two delay scans. The resulting durations are of 86 fs and 264 fs, respectively, assuming a sech^2 pulse shape. The two output pulses refer to the same input of 128 fs. We performed pulsewidth measurements of the amplified pulses as a function of the duration of the CPM pulses, varying the input pulsewidth from 50 to 500 fs. Figure 3 displays the output pulsewidth of the amplifier, with and without the longitudinal stage. It can be seen that a marked shortening of the pulse (up to a factor of 3) is possible for input pulses longer than 130 fs.

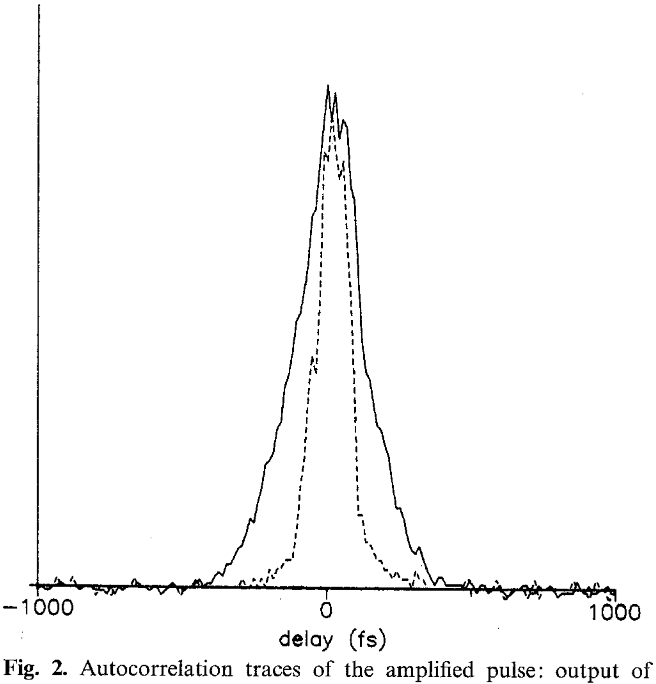

Fig. 2. Autocorrelation traces of the amplified pulse: output of the multipass cell (dashed line); output of the longitudinal cell (continuous line)

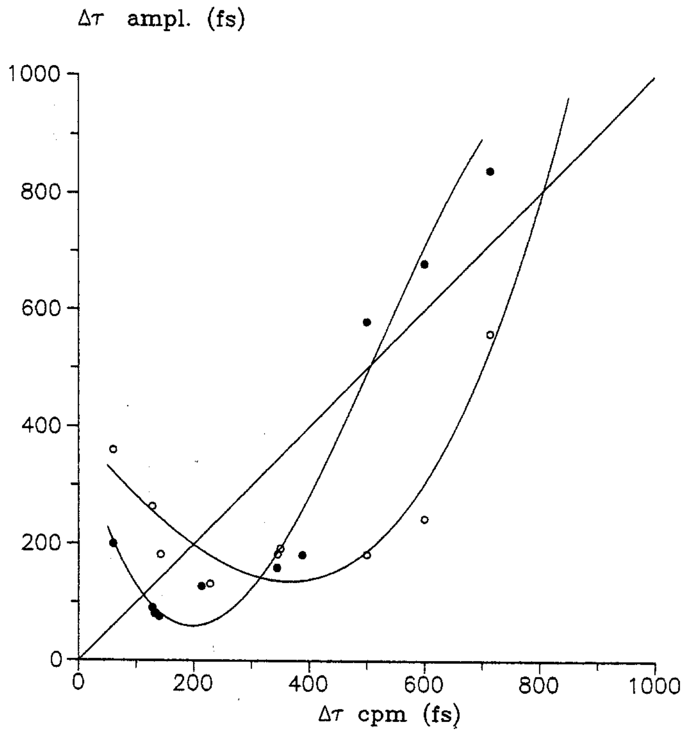

Fig. 3. Experimental values and best fit of amplified pulse duration versus CPM input pulse duration, with (white circles), and without (black circles) the longitudinal cell. On the region to the right of the 45° line $\Delta t\,(\mathrm{ampl}) < \Delta t\,(\mathrm{CPM})$

The values of $\Delta t \cdot \Delta \nu$ for the output pulse versus those of the input are plotted in Fig. 4.

The shortening effect is due to compensation of the negative chirp of the CPM pulse by the positive effects present in the amplifier. By assuming that these are mainly due to group velocity dispersion (GVD), we performed some calculations for the case of the two pulsewidths reported in Fig. 2. Knowing the bandwidth of the 128 fs CPM pulse ($\Delta\lambda = 5.5\,\mathrm{nm}$) it is possible to determine the size of its negative chirp. This, combined with the GVD present in the multipass amplifier (ethylene glycol and lenses), and in the longitudinal cell (10 cm of H_2O and windows), leads to the calculated pulse duration of 10 and 170 fs, respectively.

In order to give a more exact evaluation of the process, we determined the relative frequency chirp of the pulse by measuring its duration before and after passing through a water cell of known thickness. Knowing the dispersion of water ($d^2n/d\lambda^2 = 1.03 \cdot 10^{11}\,\mathrm{m}^{-2}$ at our wavelength) it is possible to evaluate the negative frequency chirp for each value of the pulse duration. For a pulse of 128 fs, generated by our CPM laser, this parameter is equal to $-1.1 \cdot 10^{24}\,\mathrm{s}^{-2}$ leading to the calculated output pulsewidth of 133 fs and 196 fs. Comparison of these results with the experimental ones of Fig. 2 shows that other effects must be considered; in particular anomalous dispersion of the amplifier gain saturation and self-phase-modulation (SPM) in the multipass cell.

An aproximate analysis of the spectra reported in Fig. 5 shows that the CPM pulse is at longer wavelength with respect the center of the Kiton Red flu-

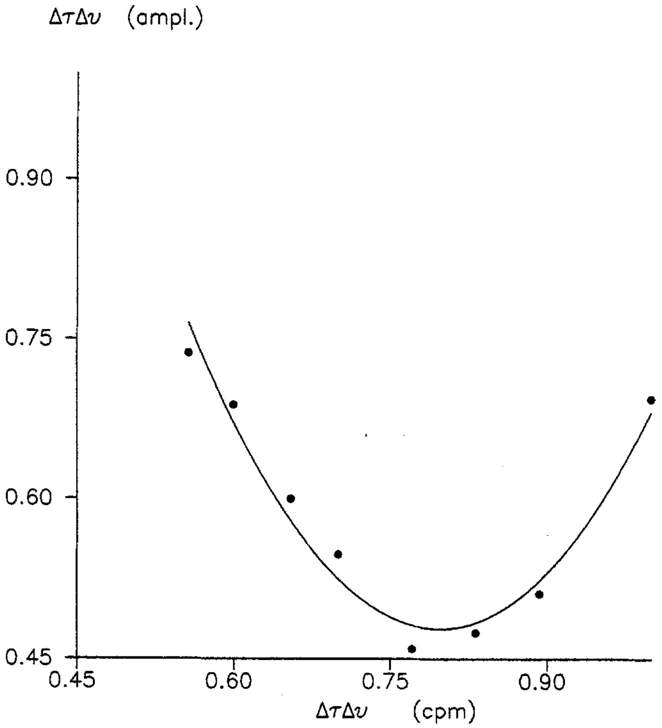

Fig. 4. Experimental values and best fit of the product $\Delta t \cdot \Delta \nu$ of the output pulse (amplifier with longitudinal cell) as a function of the same product for the input CPM pulse

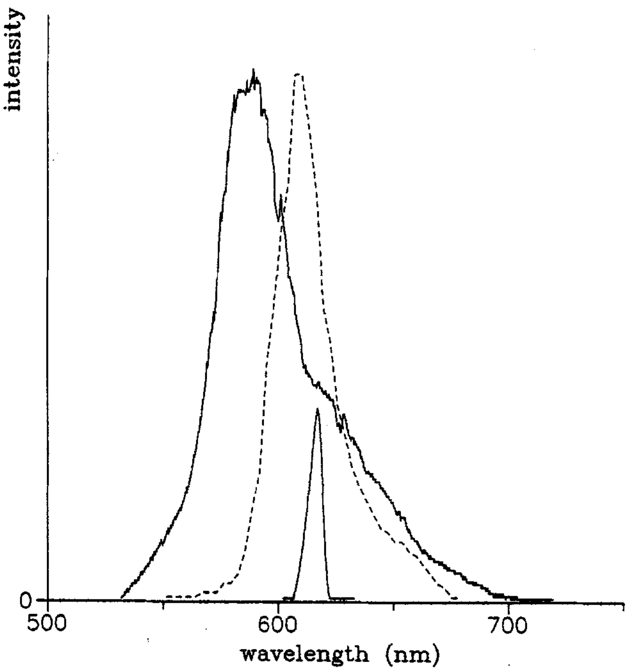

Fig. 5. Fluorescence spectra of Kiton Red in Ethylene glycol (continuous line) and Sulforhodamine 640 in H_2O (dashed line). As a reference the spectrum of the 50 fs CPM pulse is included

orescence band. In this way anomalous dispersion in the gain medium and gain saturation can give, together with GVD, a positive contribution to the frequency chirp which could be combined with the down chirp of the input pulse. The same effect is expected for SPM in the solvent. A similar mechanism has also been suggested in order to explain the results observed in another femtosecond amplifier [11]. This shortening effect can however be useful whenever a not already ultrashort, bandwidth-limited pulse is available from CPM.

These measurements are made without using any dispersive delay line at the output of the amplifier. The use of a four prism sequence to compensate for the linear pulse broadening in the amplifier should give a further reduction of the output pulsewidths. A detailed analysis of all contributions to the pulse shaping occurring in the amplifier for input pulses of different chirping will be presented in a forthcoming paper.

4. Conclusions

In conclusion, we have described a femtosecond amplifier consisting of a multipass dye cell and a longitudinal cell, pumped by the second harmonic of a 20 Hz Nd-YAG laser. In this system, the ASE contribution is restricted to 0.5% of the total output energy with no use of any saturable absorber stage. The amplification factor of the system described in this work is more than $5 \cdot 10^6$ with an overall output energy of 150 μJ at 20 Hz repetition rate. An experimental study of the output pulsewidth has shown that this can be made a factor three shorter than the pulsewidth of the input pulse. These characteristics and its simple configuration make this amplifier a good alternative to the traditional multi-stage Nd-YAG-pumped amplifier.

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