Pulsed amplification of cw dye laser with undetectable amplified spontaneous emission

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We have constructed a cw dye master oscillator/dye pulse-amplifier system that generates 4 ns, 100 mJ pulses with a repetition rate of 30 Hz. The output pulse has a bandwidth of 275 MHz. Backward-stimulated Brillouin scattering is used to control the growth of amplified spontaneous emission (ASE). The content of ASE in the final output is under our detection limit ($<10^{-4}$) for the entire tuning range. © 2000 American Institute of Physics.

I. INTRODUCTION

A spectroscopic light source of narrow bandwidth, high pulse energy, diffraction-limited beam quality, and wide tunability has been proven to be extremely powerful for use in the investigation of atomic, molecular, and optical processes that require a very high degree of sensitivity and selectivity. For many years, high brightness laser systems based on tunable dye lasers have satisfied these needs.1–5 These lasers are still in heavy use in frontier spectroscopic research. Standard approaches to the generation of pulsed transform-limited radiation have used the pulsed amplification of a cw single-frequency argon-ion-pumped ring dye laser,5 external filtering of a multimode laser oscillator,6,7 or development of a cavity oscillator incorporating a highly frequency-selective element.8

Pulsed amplification of a well-controlled cw laser remains one of the best means to obtain high power, narrow bandwidth with broad tunability that extends to the IR and UV regions, and controlled scanability. The method is simple and gives good frequency stability when scanning. Typical cw single-mode lasers operate at powers of 0.1–1.0 W. Since it is desirable to have high peak power, a minimum net or total gain of $10^8$ is required from the amplifier system.9 Then the small-signal unsaturated single-pass gain easily exceeds $10^{10}$. Any small amount of feedback, either intentionally when multipassing a single amplifier or unintentionally through scattering or incidental backreflection, will cause amplification of collinear spontaneous emission to reach superradiant intensity. Hence amplified spontaneous emission (ASE) is a primary limiting factor in efficient high-power laser pulse amplification by a high gain medium and in maintaining high spectral purity and broad tuning in pulse-amplified cw lasers.5,10,11 Elaborate dispersion and (or) spatial filtering schemes are being used to suppress the significant ASE content in these amplifiers.

We have demonstrated12,13 that this ASE can be efficiently suppressed by backward-stimulated Brillouin scattering (SBS) phase conjugation in a low gain Ti:sapphire amplifier laser system. The content of ASE increases rapidly when the amplifier is changed from low gain medium of Ti:sapphire to high gain medium of dye solution. In this article, we report the use of backward-stimulated Brillouin scattering to control the growth of amplified spontaneous emission in a pulse-amplified cw dye laser system. In this system, well controlled, single-mode scanning is provided by a cw ring dye oscillator. The power amplification is provided by three dye amplifiers that boost the overall energy gain in excess of $10^8$ to an output power of over 20 MW in the primary tuning range. The unwanted background radiation is suppressed to below the detection limit of our photodiode detectors ($<10^{-4}$) for the entire tuning range.

The next section gives a detailed description of the laser system, followed by a section that discusses the spectral, temporal, and energy characteristics of the laser output which show that this system should be useful in many applications.

II. SYSTEM DESCRIPTION

The basic arrangement of the dye amplifier system is similar to that described earlier.5 That system was a three-stage four pass amplifier where the intermediate dye amplifier was double passed to produce sufficient pulse energy to saturate the final amplifier. The ASE content was typically 15% or higher when the laser was tuned away from the wavelength where the dye gain was the highest. The present system differs from the previous system in a significant way. It is recognized that in order to reach well above the threshold for SBS phase conjugation, three passes through the dye amplifiers are required prior to sending the beam into the phase-conjugate cell or mirror. Then, in order to compensate for the reflection loss at the mirror, one more amplification stage is necessary to saturate the final amplifier. This is accomplished by double passing both the first dye amplifier and the second amplifier, with the high-reflection mirror in the previous system replaced by a SBS phase-conjugate mirror. The pump source for these amplifiers is the second harmonic of an injection-seeded Q-switched Nd:YAG laser with output energy of 600 mJ/pulse at 532 nm and pulse duration of 5 ns. A schematic of the system is shown in Fig. 1. The following is a detailed description of the entire system.
The starting point of the system is a Coherent 699-29 Autoscan ring dye laser pumped by an argon ion laser. This laser gives a well-characterized light beam of <10 MHz bandwidth, ~350 mW beam power and continuous tunability from 540 to 800 nm. However, the laser output beam moves periodically during each 10 GHz frequency scan, caused by the sweeping of an intracavity etalon in the cw dye laser. This movement is reduced to <5% of the beam diameter by sending the laser beam through a 100 μm diam pinhole filter. This filter also optically isolates the cw laser from the amplifier chain and prevents mode hopping induced by the feedback of ASE from the amplifiers.

The output from the pinhole filter is an Airy pattern with a central lobe and a set of diffraction rings. The lens after the pinhole filter is chosen such that the size of the central lobe matches with the bore size of the first dye cell. Only the central lobe is sent to the second stage of amplification. The beam is expanded by a cylindrical lens onto the dye cell. Use of relay lenses are used to form a relay image telescope to produce a homogeneous pump beam profile near the dye cell. 30% of the pump beam energy is extracted by a beam splitter, and is efficiently suppressed by SBS phase conjugation in a Ti: Al2O3 amplifier chain. The output after the single-pass amplification in the second stage dye amplifier is then sent to a phase-conjugate mirror, the key component of this new system. The phase-conjugate mirror operates by SBS. By focusing the laser beam using a lens with a 15 cm focal length into a cell containing the SBS medium FC104, a fluorocarbon liquid (PCR Inc.), as much as 45% of the incident narrow bandwidth beam is reflected. The energy dependence of the reflectivity, as demonstrated in Fig. 2, shows that the threshold of the reflectivity is 0.25 mJ at 570 nm and it reaches a maximum reflectivity of 45% at 1.5 mJ. For comparison, in the Ti:sapphire system, the SBS threshold was 1.6 mJ and the highest reflectivity achieved by the mirror was 75% for FC104 at 810 nm. We have not tried other SBS medium since FC104 is stable and possesses a sufficiently high reflectivity for the present purpose.

The reflected beam is the phase conjugate of the original beam. Insertion of a SBS phase-conjugate mirror in the amplifier chain therefore provides effective discrimination between the signal pulse and the ASE background in the high-gain dye-amplifier system. It retraces the beam path through the second amplifier, easily permitting additional amplification and efficient extraction of the energy stored in the amplifier.

Upon extraction by a polarizer, this beam is then sent to the third stage dye amplifier. The third amplifier is a single-pass amplifier. The dye cell in the third stage has a bore diameter of 6 mm and is 50 mm long, and it is pumped by
the remainder of the second harmonic of an injection seeded Nd:YAG laser. An optical delay of the pump beam is set after the beam splitter to ensure the signal beam and the pump beam arrive at the dye cell at the same time. After completion of the final amplification the output is analyzed.

III. PERFORMANCE

A. Energy

A gain of approximately \(2 \times 10^4\) is obtained from the first dye amplifier with a Nd:YAG pump laser energy of 24 mJ per pulse at 532 nm. The output energy, measured after the second pinhole filter, is about 120 \(\mu\)J per pulse at 570 nm at the center of the dye gain curve (R6G). The content of ASE is about 7%. About 300 mW beam energy from the ring laser is enough to saturate the gain of the first dye amplifier. Since less than 60% of the cw beam passes through the first spatial filter and only central lobe enter the dye cell, the actual power of the cw beam needed to saturate the amplifier is estimated to be less than 140 mW.

After the amplification of the first pass in the second dye amplifier, we obtain about 10 mJ per pulse at 570 nm for a pump laser energy of 180 mJ per pulse at 532 nm. At the end of two passes in the second dye amplifier, about 12 mJ measured after the extraction by the polarizer is obtained at 570 nm. After the completion of third stage amplifier, as much as 100 mJ is obtained at the center of the dye gain curve and about 40 mJ at the edge of the gain curve with a final pump beam energy of 360 mJ. A total power gain of \(10^8\) is obtained from the amplifier chain. Figure 3 shows the final output energy of this system at various wavelengths when R6G dye is used.

B. Temporal profile

The temporal profiles of the output from various dye preamplifiers are shown in Fig. 4. The profiles significantly deviate from the near Gaussian profile of the pump beam. The deviation is due to the exponential gain of the amplifier, the excited state lifetime of the dye molecules, and the double passage in the dye cell. The most significant change to the pulse profile in this system comes from the SBS phase-conjugate mirror. One of the consequences of using a SBS mirror is that it compresses the pulse duration. The pulse duration of the reflected beam from the SBS mirror is a function of the input pulse duration, the SBS medium, and the focal length of the lens used to focus the beam into the SBS medium. For an input pulse duration of 5 ns and using FC104 as the SBS medium, a pulse duration of 2.7 ns is obtained when using a lens with a focal length of 15 cm. A lens with a shorter focal length produces a longer pulse duration. However, it tends to cause optical breakdown in the SBS medium. Subnanosecond pulse duration can be obtained by using a lens with a long focal length. For the purpose of producing the smallest bandwidth, a lens with a 15 cm focal length is used in this system. Second-pass amplification in the second dye cell and the final amplification of the third dye cell also caused changes in the temporal profile, and a final output pulse duration of 4.1 ns is obtained. All of these effects on the pulse duration contribute to the spectral broadening which is discussed below.

C. Amplified spontaneous emission

ASE in this system originates from spontaneous emission in the first dye amplifier. The emission is amplified first in the two-pass first amplifier and then by the subsequent amplifiers. Due to the double passes in the first dye cell, a significant amount of ASE is produced. However, after pass-
ing through the spatial filter, most of the ASE is blocked. Essentially, only the collinear ASE is left in the beam. The output of the first dye amplifier at the center of the dye gain curve, measured after the first spatial filter, has about 7% of its energy as ASE at 570 nm for dye R6G.

After the first pass in the second dye amplifier, the percentage of the ASE is increased significantly. This percentage grows higher as the wavelength is changed from the center of the dye gain curve to the edge of the gain curve. However, when focusing the laser beam by a lens with a 15 cm focal length into a cell containing FC104, the reflected SBS beam contains an amount of the ASE that is virtually nondetectable. To analyze the ASE content, a small portion of the output pulse from the system is dispersed with a 1200 grooves/mm grating and then sent to two calibrated photodiodes. One photodiode is located in the direction where the dispersed ASE beam would traverse while the other photodiode is located at a position that intercepts the dispersed injection laser beam. The measurement is performed for several injection wavelengths. It is found that the ASE energy in the beam is less than the noise limit of 1 μW of the calibrated photodiode, or the ASE in the reflected beam is under our detection limit (<10⁻⁵).

**D. Frequency shift and bandwidth**

A consequence of using a SBS mirror is that it imposes a frequency shift onto the incident beam, in addition to the compression of the pulse duration. Using a spectrum analyzer with a free spectral range of 7.5 GHz, the signal beam before entering the SBS cell and the beam reflected from the SBS cell are measured simultaneously (without the refractive index effects in the amplifiers). We determined this shift to be 2510 MHz±20 MHz for FC104 at 574 nm. Figure 5 shows an analyzer scan of the pulse laser beam before and after the reflection by the SBS mirror simultaneously. The bandwidth of the beam is broadened from 200 to 275 MHz. This increase is a combination of the effect of pulse compression and the bandwidth imposed by the SBS medium. The analyzer scan of the final output shows that the bandwidth is not changed by the final amplification.

**E. Tuning and frequency extension**

The tuning range of this laser system is limited by the tuning range of the cw ring laser and the gain curve of dye amplifiers. With the use of different dyes, the system is continuously tunable from 540 to 800 nm.

The good beam quality and short pulse duration provides high conversion efficiency to other frequencies by doubling, tripling, and mixing techniques. Second harmonic generation is done with a KDP crystal (10×10×25 mm³). The conversion efficiency measured (after reflection losses at optical surface are accounted for) is more than 40% at 570 nm for an input energy of 100 mJ and a beam diameter of ~6 mm.

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