Narrow-Band Pulsed Dye Laser System for Precision Nonlinear Spectroscopy

G. L. EESLEY, M. D. LEVENSON, D. E. NITZ, AND A. V. SMITH

***Abstract*—We report a CW oscillator/pulsed amplifier dye laser system that produces 20 kW of diffraction limited radiation with a linewidth of 17 ± 4 MHz. The key component is a unique long pulse Nd:YAG pump laser.**

PRECISION experiments in nonlinear spectroscopy often require power levels greater than can be produced by CW lasers and linewidths less than those of typical pulsed dye laser systems [1]-[4]. Systems incorporating a single-mode CW dye oscillator and pulsed amplifiers have been shown to overcome many of the deficiencies of earlier systems. The most widely used design of this type employs nitrogen laser pumped dye amplifiers [5], [6]. Such a system has two drawbacks: the resolution is limited to the transform limit of the 10 ns long pumping pulse—roughly 100 MHz—and the output beam is not diffraction limited.

We have developed a dye amplifier system pumped by the second harmonic of a unique Nd:YAG laser, which amplifies the output of a Coherent Inc. Model 599-21 CW dye laser to 20 kW while producing a diffraction limited beam and a resolution of 17 MHz. The key feature of this system is the long pulse Nd:YAG pump laser which produces peak powers of 3 MW in pulses 100 ns long. Up to 300 kW of second harmonic is available for pumping the dye amplifiers.

The laser system is diagrammed in Fig. 1. The Nd:YAG oscillator cavity is a negative branch unstable resonator with a magnification of 2.34 [7]. At one end of the cavity is a convex mirror of radius 2.09 m, while at the other end is a concave mirror of radius 15 m. The mirror separation is 3.7 m, and the thermal lensing action of the 7.6 cm long 0.6 percent Nd:YAG rod acts to superimpose the foci of the two mirrors. Sixty-five percent of the circulating power is coupled out by a flat 45° scraper mirror with a 6.4 mm diameter central hole. The Nd:YAG rod is also 6.4 mm diameter and is placed directly behind the scraper mirror. A glan-laser polarizer and an electrooptic Q-switch are placed between the rod and the convex mirror. To minimize losses, the scraper mirror is located as close as possible to the convex mirror. Quarter wave plates at either end of the oscillator rod (not shown) produce circularly polarized light in the rod to suppress spatial hole burning and increase output power by 15 percent.

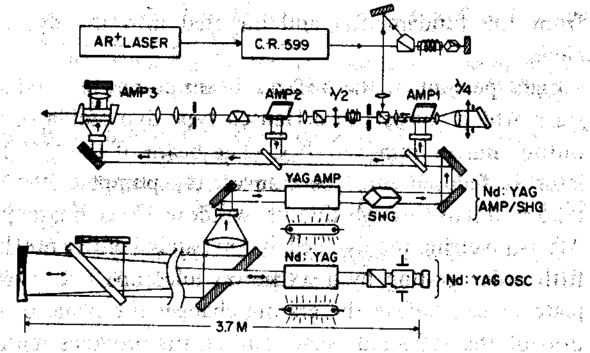

Fig. 1. The tunable laser system. The entire device is mounted on a 16 ft × 5 ft optical table.

In order to suppress self mode locking a 45° mirror coated for 50 percent reflectivity is placed 0.7 m from the 15 m radius concave mirror. The reflected beam is incident upon a nominal 13 m radius concave mirror the position of which is adjusted to produce phase fronts matching those reflected from the 15 m radius mirror. The combination of these three mirrors acts as a Michelson interferometer and reduces the amplitude modulation at frequencies below 250 MHz.

Even with the Michelson mode selector properly aligned considerable high-frequency amplitude modulation due to mode beating remains. Much of this modulation is suppressed when an optical element (not shown) having one flat uncoated surface and one low reflection coated surface is placed 2 cm from the convex mirror, between the convex mirror and Q-switch. When properly aligned, this element acts as a resonant reflector, suppressing amplitude modulation at frequencies below 1 GHz.

With 50 J input to the single flashlamp, this oscillator produces 80 mJ output in an annular beam 1.5 cm in diameter. At 10 pps the average power is 0.6 W. The pulse length is 110 ns at the half-power points. The oscillator beam is demagnified to a diameter of 6 mm by a Galilean telescope and amplified by a 3 in long 6.4 mm diameter 1.2 percent doped Nd:YAG rod. With 70 J loading of the amplifier flashlamp, the output at 1.06 μm is 3 MW peak, with a pulse length of 100 ns.

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The amplified beam is frequency doubled in a 4 cm long type II KD*P crystal. To improve the harmonic generation efficiency, the beam is first flattened to a ribbon 1 mm thick and 6 mm wide by a cylindrical telescope (not shown) [8]. The polarization of the 1.06 μm light is at 45° to the plane of the ribbon, and the SHG crystal is oriented so that the additional divergence introduced by the one dimensional compression does not affect the phase matching. The second harmonic is recollimated by a second cyclindrical telescope, separated from the fundamental and directed into the dye amplifier chain.

Eight percent of the 532 nm beam pumps the first amplifier stage which consists of a Hansch type dye oscillator with the output mirror removed [9]. The beam of the CW dye oscillator is focused into the transversely pumped dye cell by a 0.25 m fl lens through the side window of a glan-laser polarizer. After traversing the dye cell, it is expanded onto a 600 lines/mm fifth-order grating by a six power telescope. A quarter wave plate placed before the grating changes the plane of polarization of the reflected beam, and an iris provides spatial filtering. The telescope then refocuses the beam into the gain cell, and the horizontally polarized output of the first amplifier stage is transmitted through the glan polarizer.

Isolation between stages is essential in a high gain amplifier system of this sort. A terbium gallium garnet Faraday rotator isolator prevents the first amplifier stage from oscillating on the cavity formed by the grating and the output mirror of the CW oscillator and prevents spontaneous emission from the amplifier chain feeding back into the photodiodes of the oscillator stabilization servos [10]. A second isolator consisting of a Faraday rotator, glan polarizer, and a half-wave plate rotates the polarization of the light from the first amplifier stage to vertical and prevents light from the second and third stages feeding back into the first stage.

The output of the first stage is focused into a transversely pumped dye cuvette pumped by another 8 percent of the 532 nm beam. The output of the second stage is filtered spectrally and spatially by a direct vision prism and a lens and pinhole spatial filter. After the spatial filter, the beam is recollimated at a diameter of 3 mm to provide a location where a plane Fabry-Perot etalon can be inserted to provide additional spectral filtering. A 0.6 m fl lens then focuses the beam into the third dye amplifier.

The output amplifier is a transversely pumped 0.6 mm diameter precision bore capillary. The dye solution flows longitudinally through the 3.5 cm long cell, and a second dye channel is provided above the capillary through which bubbles can flow without blocking the beam. The dye concentration is adjusted so that roughly 10 percent of the pump light is transmitted through the dye stream. That light and the light missing the dye stream is refocused into the capillary by a lens and mirror on the far side.

The capillary dye cell produces a stable output only when a high viscosity solvent such as ethylene glycol is employed. With such a solvent, the flow through the capillary is slow (<1 cm/s) and laminar. Heat is transferred to the wall principally by conduction, resulting in a smooth variation in the index of refraction across the gain medium. Alignment of the cell and pumping optics is crucial as is the speed of dye flow. When all adjustments are correct, the cell acts as an amplifying spatial filter that produces an output beam with a far-field pattern similar to an Airy spot. Ninety percent of the output is in a bright round central spot surrounded by rings and fringes. The divergence of this central spot corresponds to that produced by a 0.6 mm aperture.

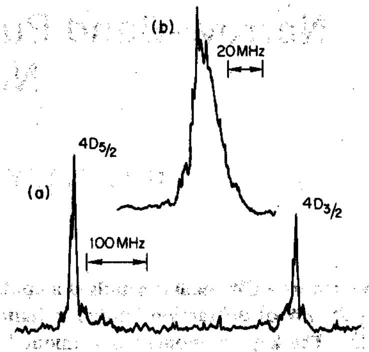

Fig. 2. Doppler-free two-photon absorption traces of the $3S$ $(F = 2) \rightarrow 4D_{3/2,5/2}$ transition in sodium.

Without injection from the CW oscillator, the output spectrum of this amplifier system has a width of 0.12 nm corresponding to the resolution of the grating in the first amplifier. When 40 mW of CW laser radiation is injected, the broadband background is reduced by a factor of 100 or more, and the narrow-band output power is 5-20 kW depending upon wavelength. The broad-band output is eliminated when a 3 cm^{-1} free spectral range Fabry-Perot is placed between the second and third amplifier. A plane Fabry-Perot interferometer with a 0.10 cm^{-1} free spectral range then detects only the narrow component.

A Doppler-free two-photon absorption experiment was performed in order to estimate the actual laser linewidth. The laser beam collimated at a diameter of 2 mm and attenuated to 2 kW was directed into a sodium vapor cell. The transmitted beam was reflected back into the cell at an angle of 4 mrad. The laser was tuned to the $3S$-$4D$ two-photon transition at 578.7 nm, and the fluorescence due to the $4P$-$3S$ decay channel was detected and plotted as a function of the frequency of the CW input.

Fig. 2 shows a typical result. The two peaks on trace *A* correspond to the $3S$ $(F = 2) \rightarrow 4D_{5/2}$, $4D_{3/2}$ two-photon transitions. The spacing of these lines is known to be 514 MHz. Trace *B* shows the $D_{5/2}$ peak at higher resolution, scanned at a rate of 0.5 MHz/s. The linewidth measured from such traces is 20 ± 4 MHz (FWHM). The main uncertainty results from residual thermal drifts in the oscillator. The linewidth of the transition was estimated as 7 MHz, 3 MHz due to the homogeneous linewidth and 4 MHz due to residual Doppler broadening. The actual laser linewidth can thus be estimated as 17 ± 4 MHz.

The transform limit for Gaussian dye laser pulses 60 ns long (FWHM) is approximately 7 MHz. Any additional linewidth of our system probably results from a chirp produced by the amplifier. In a preliminary experiment, in which the amplified output was compared with the CW input using an 8 GHz free spectral range scanning confocal interferometer, we found roughly a 15 MHz shift of the amplified output to higher frequencies. Larger shifts have been reported for nitrogen pumped amplifier systems [5].

When the light intensity was sufficient to power broaden the Doppler-free two-photon absorption lines, additional "ghost" lines appear at frequency shifts corresponding to the Fourier components of the amplitude modulation of the YAG pump. Mode beating in the pump laser modulates the gain of the dye amplifiers and creates sidebands on the output frequency. Scanning with an 8 GHz spectrum analyzer we find sidebands that contain less than 3 percent of the total laser power and occur at 1, 2, and 3 GHz. Higher frequency sidebands are suppressed by the finite lifetime of the upper dye laser level.

The laser system is remarkably reliable in spite of the apparent complexity. It has operated for over one year and requires cleaning and major realignment only once every several months.

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