

Kilohertz dye laser system for high resolution laser spectroscopy

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We have electro-optically sliced the output light of a commercial Coherent Evolution Nd:YLF laser to pump a kilohertz repetition rate nanosecond dye laser system. Simple and highly adjustable, this laser system can easily be used for initial state preparation for ultrafast systems as well as high resolution spectroscopy. © 2010 American Institute of Physics. [doi:10.1063/1.3462978]

I. INTRODUCTION

Over the past 40 years, narrow-linewidth pulsed dye lasers have been used in investigations of many atomic and molecular systems.¹ The broad tunability and versatility of dye lasers allow for a wide range of applications, including high resolution spectroscopy, laser isotope separation, and initial state preparation for experiments with ultrafast laser systems. Although many types of pulsed dye lasers exist, the most widely used laboratory dye lasers are optically pumped, usually by *Q*-switched, frequency doubled or tripled Nd:yttrium aluminum garnet (YAG) lasers or excimer lasers.²⁻⁴ These pump lasers have repetition rates of 10–100 Hz and pulsewidths of approximately 10 ns and produce dye laser pulses of similar widths. An attractive feature of such short laser pulses is that it is possible to excite an atom or molecule through comparably short lived states with synchronized pulses from several dye lasers. In fact, as many as six sequential dye laser pulses have been used to excite atoms to high lying states.⁵ A more recent use of pulsed dye lasers is to prepare atomic and molecular states for subsequent exposure to intense picosecond or femtosecond pulses.^{6,7} In these experiments the intense short pulses were generated by regenerative Nd:YAG or Ti:sapphire amplifiers which operated at 10–20 Hz repetition rates.

The advent of commercially available kilohertz repetition rate pump lasers, typically a frequency doubled, *Q*-switched Nd:yttrium lithium fluoride (YLF) laser, has led to the development of kilohertz repetition rate Ti:sapphire regenerative amplifiers for mode locked Ti:sapphire oscillators, bringing a new level of sophistication to strong field laser experiments.⁸ The existence of these kilohertz ultrafast Ti:sapphire lasers has fueled interest in the development of kilohertz repetition rate dye lasers for preparation of excited atomic and molecular states for subsequent exposure to the femtosecond laser pulses. Unfortunately, the kilohertz doubled Nd:YLF lasers used to pump the regenerative amplifiers produce 527 nm pulses at least 100 ns long, far too long to pump a conventional nanosecond dye laser.^{2-4,9} One approach to using such a long pump laser is to use a dye

laser cavity more like that of a continuous wave laser. The most important difference from a nanosecond dye laser is that the laser is longitudinally pumped and does not have the high gain associated with pulsed dye lasers. To accommodate the lower gain the dispersive element is a set of prisms instead of a diffraction grating,^{10,11} with the result that linewidths in excess of 30 GHz are typical unless an etalon is inserted into the cavity.

Here we report a different approach, using a high gain transversely pumped dye laser. Our kilohertz repetition rate pump laser is a Coherent Evolution 30, which has a pulse width of roughly 100 ns. If the entire 100 ns pulse is used to pump the dye laser we observe relaxation oscillations with a period of 40 ns in the dye laser output and dye lifetimes of several hours, even with rhodamine dyes. These observations suggest that we slice the pump pulse into several shorter pulses of usable length. This approach works quite well, enabling us to pump three dye lasers of conventional design.²⁻⁴ In fact, the lasers are the same lasers we use with 20 Hz Nd:YAG pump lasers having 8 ns long pulses. Since these dye lasers use gratings as the dispersive elements, they have excellent linewidths, in some cases better than 3 GHz. The fact that this approach is simple and offers excellent dye laser linewidths suggest that it may be more generally useful than simply preparing targets for further exposure to intense Ti:sapphire pulses.

II. DESIGN

The pump laser is a Coherent Evolution-30 diode pumped solid state, frequency doubled, *Q*-switched Nd:YLF laser. The Evolution-30 laser produces horizontally polarized pulses of 527 nm light less than 250 ns long at a 1 kHz repetition rate and can deliver up to 20 W of average power. The initial 20 mJ pulse, shown in Fig. 2, is 100 ns long at full width at half maximum, with a problematic long tail. This long tail, decaying from 90% to 10% in 130 ns, is not particularly useful for pumping the dye, but it does heat the dye, which is probably a factor in the short dye lifetimes with long pump pulses.

As mentioned earlier, the essential idea is to split each pulse into shorter pulses. Accordingly, each pulse from the Evolution-30 pump laser is spatially split into three temporally distinct pulses using a system of two deuterated potassium dihydrogen phosphate (DKDP) Pockels cells, schemati-

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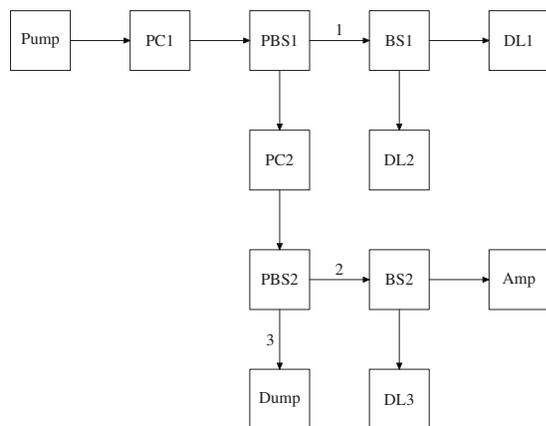


FIG. 1. Laser system schematic. Shown are the pump laser, Pockels cells (PC), polarizing beam splitting cubes (PBS), beam splitting cubes (BS), dye lasers (DL1–DL3), single-pass dye amplifier (A1), and beam dump.

cally shown in Fig. 1. The Pockels cells are switched from zero retardation to $\lambda/2$ retardation in 2 ns, a time that is short compared to the laser pulse length. The half-wave voltage $V_{\lambda/2}=3.9$ kV is generated by a Marx-bank circuit using Motorola 2N5551 transistors.^{12,13} The two Pockels cells are switched at 48 and 81 ns after the beginning of the laser pulse at $t=0$. A SRS DG535 digital delay/pulse generator, triggered by the pump laser Q -switch output, is used to trigger the high-voltage switches. Light exiting the first Pockels cell (PC1) is sent to a polarizing beam splitting cube (PBS1). Horizontally polarized light passes straight through PBS1 becoming what we term the first pulse, whereas the rotated, vertically polarized light is sent through the second Pockels cell (PC2). Light exiting the second Pockels cell is sent to the second polarizing beam splitting cube (PBS2). Vertically polarized light is reflected to produce the second pulse and horizontally polarized light retarded by PC2, the third pulse, passes straight through to a beam dump. Figure 2 shows how the pump pulse is split into three pieces, and the figure inset shows the original pump pulse. The first, second, and third pulses have widths of 16, 35, and 45 ns, respectively. Note, however, that the Pockels cell timing may be tailored to fit the needs of the experiment. The pulse energies of the three pulses are 4.25, 3.75, and 3.5 mJ, respectively.

The first pulse, further split by a 50-50 beam splitter (BS1), pumps two single grating Littman-type dye lasers,³ DL1 and DL2 of Fig. 1. Both dye lasers were constructed using 1200 lines/mm diffraction gratings at grazing incidence with the output taken from the zeroth order reflection. DL1 uses DCM in dimethyl sulfoxide (DMSO) at a molar concentration of 1×10^{-3} . DL2 uses LDS-821 dissolved in methanol at a molar concentration of 1.5×10^{-4} . The second pulse, also split by a 50-50 beam splitter (BS2), pumps a double grating Littman-type dye laser,¹⁴ DL3 of Fig. 1, and a single-pass amplifier (A1). DL3 was constructed using two 1800 lines/mm diffraction gratings. Rhodamine-640 in molar concentrations 5×10^{-4} (oscillator) and 1×10^{-4} (amplifier) was used to characterize the dye laser. The third pulse, with an average power of 3.5 W, is sent to a beam dump but could be used to pump additional dye lasers or amplifiers.

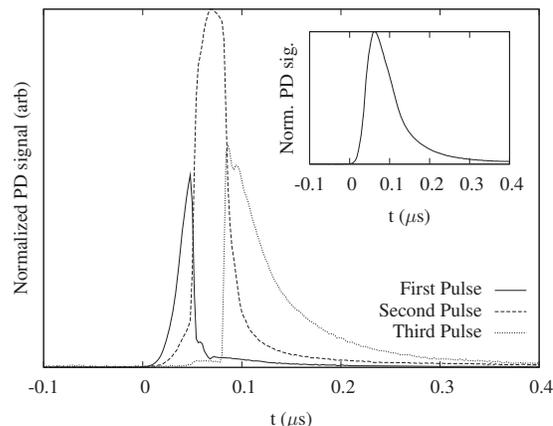


FIG. 2. Coherent Evolution-30 Nd:YLF laser pulse shape before (inset) and after Pockels cell system.

III. PERFORMANCE

When pumped with 16 ns 2.2 mJ pump pulses, DL1 produces 12 ns laser pulses over a tuning range from 625 to 695 nm, centered around 660 nm. The measured temporal pulse shape and the measured tuning curve are shown in Fig. 3. Typically, this laser produces 21.4 μJ per pulse at 671 nm with a linewidth less than 10 GHz. Transmission through a 1 cm (10 GHz free spectral range) solid etalon produces a visible fringe pattern. When tuned to 813 nm, DL2 produces 14.4 μJ per pulse delivered in 12 ns. The pulse shape is shown in Fig. 3. The tuning range of this laser is 800–840 nm with a linewidth less than 10 GHz, as measured using a 1 cm solid etalon. The third dye laser oscillator produces 3.72 μJ per pulse at 615 nm before the amplifier stage when pumped with 35 ns 2.1 mJ pulses, and 36 μJ per pulse after the amplifier stage. The postamplifier pulse shape is shown in Fig. 3. Although we did not conduct systematic tests versus pump pulse length, as mentioned above, we encountered relaxation oscillations with a 40 ns period with a long pump

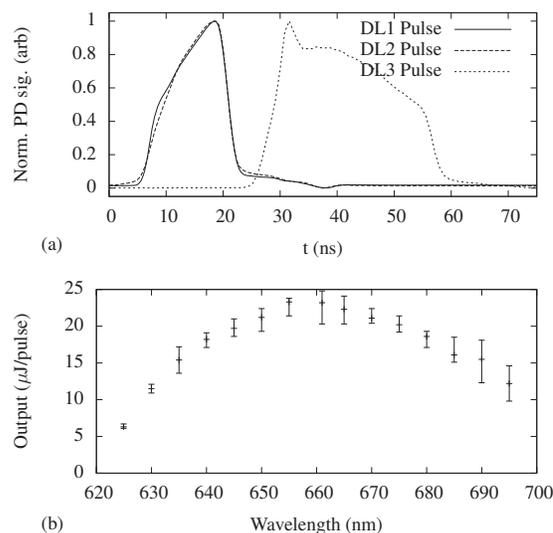


FIG. 3. Dye laser characteristics. (a) Dye laser pulse shapes for DL1–DL3. DL1 and DL2 were both pumped by a 16 ns 2.2 mJ pump pulse, DL3 was pumped by a 35 ns 2.1 mJ pump pulse. These pump pulses are shown in Fig. 2. (b) Tuning curve for DL1 characterized with DCM in DMSO at a molar concentration of 1×10^{-3} .

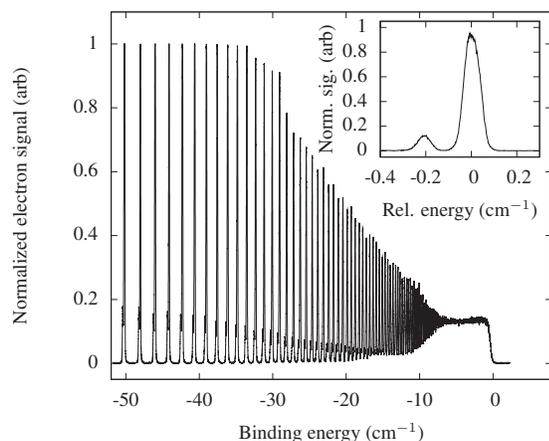


FIG. 4. Rydberg lithium signal scan as a function of laser photon energy. 100-point moving average, from $n=46$ to the ionization limit. The inset is the signal scan relative to the ${}^7\text{Li}$ $34p$ state.

pulse, but this laser shows that a pulse length up to 35 ns works quite well. The use of a second diffraction grating of high groove density, combined with the slightly longer pump pulse, provides a narrower laser linewidth of 3 GHz, determined from the observed width of an atomic line, as described below. The improved linewidth is due primarily to the use of the double grating laser design.

Ultimately, the test of such a laser system is its performance in an experiment. Accordingly, we have used the dye laser system shown in Fig. 1 to excite Li atoms to Rydberg states. Specifically, a thermal beam of ground state lithium atoms was excited to np Rydberg states by the scheme: $2s \rightarrow 2p \rightarrow 3s \rightarrow np$. The ground state atoms pass between two parallel field plates separated by 1.9 cm. The three dye laser pulses, corresponding to 670.776, 812.645, and ≈ 615 nm, propagate perpendicularly to the atom beam in the central region of the parallel plates. Subsequent to the laser pulses a voltage pulse is applied to the lower plate to field ionize the Rydberg atoms and drive the electrons from field ionization through a small hole in the upper field plate to a dual microchannel-plate detector. The signal from the detector is recorded with a gated integrator as the frequency of the third laser is scanned.

Scanning the frequency of the third laser produces a lithium Rydberg spectrum, as shown in Fig. 4. Expanding the

scale of a smaller energy region, such as the inset of Fig. 4 which shows the $34p$ state, reveals the 6.6 GHz isotope shift between ${}^7\text{Li}$ and ${}^6\text{Li}$,^{15,16} from which we determine the linewidth of DL3 to be approximately 3 GHz.

IV. CONCLUSION

We have constructed and used a kilohertz repetition rate dye laser system using a pump laser with ~ 100 ns long pump pulses. Such pump lasers are routinely used to pump regenerative Ti:sapphire amplifiers for ultrafast lasers. By slicing the long pump pulse into pulses from 12 to 35 ns long we are able to pump several dye lasers of conventional design. This approach should be useful for preparing excited states of atoms or molecules for exposure to femtosecond pulses or as a way of conducting experiments presently done at a 20 Hz repetition rate.

ACKNOWLEDGMENTS

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