**A HIGH POWER PULSED DYE LASER
WITH FOURIER LIMITED BANDWIDTH**

G. Bollen, H.-J. Kluge* and K. Wallmeroth

Institut für Physik, Universität Mainz,
Federal Republic of Germany

H.W. Schaaf

Institut für Physik, Universität Mainz and Coopervision,
Laser Division, Santa Clara, California, USA

R.B. Moore

McGill University, Montreal, Canada .

ABSTRACT

A high peak power, narrow linewidth light source with a homogeneous beam profile has been constructed by modifications to a commercially available pulsed dye laser system. Output pulses of up to 10 mJ were generated with linewidths of about 50 MHz for 12 ns pulses. The pulse to pulse frequency stability was better than the linewidth and the center frequency could be scanned over frequency range of 142.5 GHz at a wavelength of 600 nm. The performance of the system was demonstrated by observing the $6s^2\ ^1S_0 - 6s7s\ ^1S_0$ transition in atomic mercury at $2\lambda = 312.8$ nm and the $6s\ ^2S_{1/2} - 8s\ ^2S_{1/2}$ transition in atomic gold at $2\lambda = 308.9$ nm using up to 1 mJ of frequency doubled output for two photon nonlinear absorption spectroscopy. The lifetimes of both excited states, the magnetic hyperfine splitting of the $8s\ ^2S_{1/2}$ state in Au and the isotope shift of the $6s7s\ ^1S_0$ state of Hg were determined.

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1. INTRODUCTION

The laser system described in this paper was developed to extend our measurements of atomic isotope shifts in long isotope chains. As an example, it was first used to extend the measurements of isotope shift in the stable isotope ^{197}Au and the long-lived radioisotope ^{195}Au ($t_{1/2} = 183$ d) to the short-lived radioisotopes ^{193}Au through ^{190}Au , as has been reported elsewhere¹. For such measurements, high sensitivity and resolution are necessary, which can be accomplished with non-linear two photon absorption spectroscopy, a technique requiring a powerful, tunable narrow band light source. To measure a wide range of elements, the tuning range of this source should cover the entire spectral range from ultraviolet to near infrared. Therefore our primary objective in this work was to narrow the bandwidth of a commercial system to its Fourier limit while generating a power greater than 1 MW.

The goal of narrow bandwidth was realized by inserting a high finesse Fabry-Perot etalon in the laser oscillator cavity and adding external spectral filtering³. Amplification of the narrow band radiation with a four stage optical travelling wave amplifier resulted in the desired output power. Frequency calibration was provided by an optical spectrum analyzer, a tellurium and an iodine absorption cell.

Frequency scanning was carried out by varying the pressure in the laser oscillator cavity and the container of the external etalon. Fig. 1 shows the complete set-up consisting of a pump laser, a Molelectron Nd-YAG laser (MY32), a modified Molelectron dye laser (DL16P), our three added amplifier stages and the frequency calibration unit. In this paper we describe this system and report on its performance. Also included are some results from two photon spectroscopy on mercury and gold atoms and some subsequent improvements to the system that have been carried out at Coopervision Inc.

2. THE DYE LASER SYSTEM

The Molelectron MY-32 Nd-YAG pump laser with an unstable polarization coupled oscillator delivers about 220 mJ at $\lambda = 1064$ nm, 60 mJ at $\lambda = 532$ nm, and 30 mJ at $\lambda = 355$ nm in 12 ns pulses. The harmonics are separated by dichroic beam splitters. When using the green output light, 11% is sent into the dye laser oscillator and the first dye laser amplifier (the amplifier of the Molelectron DL16P dye laser system). When using the ultraviolet, 20% is sent to these two stages. The power distribution for the different amplifier stages is tabulated in Table 1. The pump power distribution into the different amplifier and oscillator dye cells was empirically optimized for the best performance, i.e. minimum broadband background.

A. The Dye Laser Oscillator

Originally, the Molelectron dye laser consisted of a Hänsch type oscillator³, with cavity length reduced by the installation of a Molelectron four prism beam expander instead of the conventional telescope. The overall cavity length of the oscillator is about 25 cm. The linewidth of the system, with a low finesse etalon, is approximately 1 GHz. Usually three cavity modes contribute significantly to the output, these cavity modes being separated by the free spectral range (FSR) of $c/2L$ (L = cavity length) or 600 MHz. Due to the low Q value of the cavity, the modes are not clearly resolved and therefore with time averaging the etalon bandpass envelope is filled with a smooth distribution of intensities.

To reduce the linewidth of the emitted radiation, the low finesse (F) etalon (FSR = 30 GHz, F = 15 to 20) was replaced by a high finesse etalon (FSR = 30 GHz, F = 100) so that the single pass linewidth of the oscillator becomes⁴

$$\Delta\nu = \frac{\text{FSR}}{1.2\sqrt{2F}} = 177 \text{ MHz} \quad (1)$$

However, there are two other important consequences of such a simple change. First, the free spectral range of the cavity is significantly altered due to the extra effective optical length from the increased number of round trips of the light in the etalon. Secondly, the extra losses caused by the narrower bandwidth of the intra cavity etalon, which acts only as a passive filter, require the reflectivity of the output coupler to be much higher than the usual 4% in order to sustain laser action. This increases the lifetime of the stored light in the cavity or, in other words, raises the Q of the cavity. The higher Q of the cavity allows the individual axial modes of the cavity to clearly develop and be resolved within the free spectral range of the cavity.

In our set-up the high finesse etalon decreased the free spectral range of the cavity from 600 MHz to about 250 MHz, corresponding to an increase in the effective optical length of the cavity from 25 cm to about 60 cm. Good laser action was achieved in the resulting cavity with an output coupler of 65% reflectivity. From this one calculates axial mode resonances of FWHM about 50 MHz, corresponding to mode life-times in the cavity of about 3 to 4 ns, sufficiently shorter than the pump pulse duration of 12 ns for the dye laser pulse to fully develop.

The clear resolution of the axial modes of the cavity means that there can be severe problems with maintaining stability in the output power of the dye laser pulse if the frequency of the axial mode of the cavity is not synchronized with the center frequency of the internal etalon filter. The situation in our case is summarized in Fig. 2 which shows the relative widths of the etalon filter response, the axial modes and the intermode frequency spacing. If a particular mode moves off frequency from the etalon response center, the overall gain of that mode will diminish while the gain for another adjacent mode will grow to eventually take its place.

This situation can arise when the frequency of the laser output pulse is scanned by pressure variation of the index of refraction of the gas forming the medium of the etalon. The center frequency of the etalon filter decreases with the increasing index of refraction of the gas in the etalon but the frequency of the axial modes decreases more slowly because of the portion of the optical path in the cavity which is not in the gas itself (glass elements and air outside the pressure controlled cavity). The axial mode frequencies therefore move upward relative to the envelope of the etalon filter. The effect of this movement is to cause the sort of oscillation in dye laser output power which is clearly seen in Fig. 3.

The frequency interval of 1.0 ± 0.1 GHz can easily be explained. The change in the center frequency of the etalon response is related to the change in index of refraction of the gas by

$$\Delta f_{\text{etalon}} = f_0 \Delta n \quad (2)$$

where f_0 is the center frequency of the etalon and n is the index of refraction of the gas. The change in frequency of the axial modes near the center frequency is reduced by the fraction of the optical path length for which the index of refraction is changing, or

$$\Delta f_{\text{axial}} = f_0 \frac{L_{\text{var}}}{L_{\text{var}} + L_{\text{fix}}} \Delta n \quad (3)$$

where L_{var} is the optical path length which is changing and L_{fix} is the optical path length which stays fixed.

The difference of the two frequency changes is

$$\Delta f_{etalon} - \Delta f_{axia} = f_0 \frac{L_{fix}}{L_{var} + L_{fix}} \Delta n \quad (4)$$

When this equals the separation between the axial modes, the output intensity will have gone through one complete cycle:

$$\frac{c}{2(L_{var} + L_{fix})} = f_0 \frac{L_{fix}}{L_{var} + L_{fix}} \delta n \quad (5)$$

The frequency interval between intensity maxima is given by $f_0 \delta n$. This is seen to be given by the simple relation;

$$f_0 \delta n = \frac{c}{2L_{fix}} \quad (6)$$

Hence the frequency interval between intensity maxima can be thought of as the "free spectral range" of the fixed part of the optical cavity. For a frequency interval of 1 GHz, the optical length of the fixed part of the cavity becomes 15 cm, a reasonable value for our apparatus.

To correct for this effect, the optical path length of the axial modes must increase by the same fraction as the increase in the index of refraction of the cavity gas. This can be accomplished by inserting a thin quartz plate into the optical cavity and changing the angle of incidence of the laser beam as the gas pressure changes. This approach was chosen because an angle scanner was already available in our laboratory and the required modifications to the mechanical configuration of the commercial laser were only minor.

It can be shown that for an angle of incidence α on a plate of thickness t and of index of refraction n , the rate of change of the angle of incidence with pressure in the cavity that is required to keep the mode frequency in step with the etalon center frequency is;

$$\frac{d\alpha}{dp} = \frac{1}{(\tan\alpha - \tan\beta)\cos\alpha} \times \frac{L_{fix}}{t} \times \left(\frac{dn}{dp} \right)_{gas} \quad (7)$$

where β is the angle of refraction for angle of incidence α . By setting the angle of incidence to the Brewster angle for the plate, reflection losses can be kept to a minimum and Eq. (7) becomes

$$\frac{d\alpha}{dp} = \frac{(1+n^2)^{1/2}}{(n^2-1)} \times \frac{L_{fix}}{t} \times \left(\frac{dn}{dp} \right)_{gas} \quad (8)$$

In our apparatus, a quartz galvoplate of 2 mm thickness was inserted into the dye laser cavity with a tilt servomechanism of the appropriate gain. By fine tuning the gain the frequency interval between intensity maxima could be increased to well beyond the range of frequencies covered in a pressure scan of the dye laser frequency.

As a consequence of the above changes to the dye laser system, the linewidth of the cavity output was narrowed from about 1 GHz to typically 90 to 125 MHz. Installation of an external etalon reduced this value to approximately 50 MHz. The frequency reproducibility was also greatly improved since the laser oscillator can now be forced to start oscillating on a particular axial mode near the maximum of the etalon transmission curve. (Frequency stability was also improved by using Zerodur spacers with an expansion coefficient of less than 10^{-8} per $^{\circ}\text{C}$ for the etalon).

The remaining deficiencies of the system were its relatively low power output and large amplitude fluctuations (up to 25%) even at these modest power levels. These difficulties arise because the oscillator must be pumped at low power levels to avoid excessive background light due to spatial hole burning and superfluorescence. To achieve a stable and sufficiently powerful light source for spectroscopy, the narrow band radiation from the dye laser cavity was amplified by a four stage travelling wave optical amplifier with an external etalon placed after the first stage.

B. External Etalon

It was found that the linewidth of the laser light could be filtered down to about 50 MHz by an external Fabry-Perot etalon. The etalon used consisted of two flat plates separated by 25-mm Zerodur spacers, having a flatness better than $\lambda/200$ and a coating which gave a reflectivity of 98.5%. The measured finesse and bandpass were 100 and 60 MHz respectively. The external etalon is placed in a very rigid, kinematically stable mount in a large vacuum vessel which is connected to the same gas source as the laser oscillator cavity. In this way the center frequency of this external etalon was kept in step with the laser cavity etalon.

This filter was placed in the system after the first stage of amplification (the amplifier stage built into the Molelectron DL16P dye laser). By carrying out the filtering at this stage, there was enough intensity of filtered light to achieve a good signal to noise ratio in the output. According to Ganiel et al⁵, the subsequent three amplifier stages should not change the signal to noise ratio of the output beam when appropriately isolated from the superfluorescence of each stage.

The linewidth of the light filtered by an etalon depends critically on the degree of collimation of the incident light. Therefore the light beam emerging from the first amplifier stage was expanded and collimated to a divergence of better than 0.03 mrad before passing it through the etalon.

C. Four Stage Travelling Wave Optical Amplifier

As mentioned above, the first stage of the four stage travelling wave optical amplifier is the stage which is built into the Molelectron DL16P dye laser system. After the etalon filter, the light was focused on the second amplifier dye cell by a telescope (two lenses of focal length 80 mm and -30 mm separated by 54 mm). The pump light for this stage was focused on the dye cell by a cylindrical lens of focal length 100 mm, giving an active diameter of 0.2 to 0.3 mm depending on dye concentration and lens position.

A lens of focal length 80 mm focused the amplified light from the second stage on a pinhole of 100 microns diameter acting as a spatial filter. The light filtered by this pinhole was focused on the third stage dye cell by a lens of focal length 80 mm. Again, the pump light for this stage was focused on the cell as in the previous stage.

The output of the third amplifier stage was expanded in diameter by a two lens

system (focal lengths 40 and 150 mm) and collimated by a diaphragm before entering the dye cell of the fourth and final stage of amplification. The pump light for this stage was focused on the dye cell by a combination of a negative and a positive cylindrical lens giving an illumination width of from 1 to 2 mm.

The output beam of the final stage was 1 to 2 times the diffraction limit in divergence, with a nearly Gaussian beam profile.

The dye concentrations in the different amplifier stages were adjusted to give an optimum contrast ratio of laser intensity to superfluorescence. Therefore the dye concentrations in the last three amplifier stages had to be reduced to accommodate the higher pump beam intensities without increasing the broadband background. In order to improve the contrast ratio, several other measures were taken. These include delaying the pump light relative to the dye laser pulse by 1 to 2 ns, installing iris diaphragms before and after each amplifier stage, and inserting prism polarizers into the beam before each amplifier stage to suppress the superfluorescent background. Typical contrast ratios of laser light to superfluorescence are better than 20:1.

The performances obtained with this system are listed in Table 1.

3. FREQUENCY CALIBRATION SYSTEM

To have a complete dye laser spectrometer, two optical branches were incorporated to determine the spectral purity and the wavelength of the radiation. The spectral purity was determined by a confocal Fabry-Perot spectrum analyzer with a photodiode and a diode array (Reticon model no 1024G) as separate detectors coupled to the analysed beam by a beam splitter. In this way the frequency components of a pulse could be observed as well as the shot to shot frequency jitter.

For accurate wavelength determination, an iodine (or tellurium) cell was used. The observed absorption lines were compared with the known tabulated iodine (tellurium) absorption lines⁶ using a minicomputer (DEC LSI-11).

4. FREQUENCY SCANNING SYSTEM

As already mentioned, the tuning of the laser frequency was carried out by varying the refractive index of the medium of the laser cavity and the etalon filters. This was done by introducing nitrogen gas from a 10 Atm supply into the two chambers through 4 glass capillaries of different diameters. To obtain different scanning rates, solenoid operated valves were placed in series with each of the capillaries. These valves could be operated manually or by the minicomputer which controlled and monitored the data taking phase of the experiment (the same minicomputer which carried out the frequency calibration). The pressure in the vessels during a frequency scan was also monitored by the computer through a capacitance manometer (Datametrix model 1402). To provide an accurate monitor of the frequency changes of the laser light during a pressure scan, the beam from a stabilized HeNe laser was observed through a confocal Fabry-Perot etalon mounted in the larger vacuum chamber connected to the laser pressure chamber. The exact knowledge of the wave numbers of the tellurium (iodine) absorption lines and the hfs splitting in the ground - and excited state of stable ¹⁹⁷Au allowed us to determine the laser wavelength with an accuracy of better than ± 0.01 Å. The linearity of the pressure scan was estimated by comparing the measured frequency differences between the tellurium absorption lines with the published values and by comparing the frequency spacing of consecutive spectrum analyzer transmission maxima. The non linearity over a frequency scan of typically 70 GHz was less than 50 MHz.

5. EXPERIMENTAL DATA AND PERFORMANCE

The performance of the laser system was first checked by recording the transmission signal of a spectrum analyzer. The line width of the spectral profile of the laser was found to range between 50 MHz with external etalon to 100 MHz without external etalon.

In order to test the applicability of the laser system to the determination of the hyperfine structure (HFS) and isotope shift (IS) of unstable isotopes, measurements were performed on the $6s^2\ ^1S_0 - 6s7s\ ^1S_0$, $2\lambda = 312.8\text{ nm}$ transition of Hg and the $6s\ ^2S_{1/2} - 8s\ ^2S_{1/2}$, $2\lambda = 308.9\text{ nm}$ transition of Au. In both cases the technique of two-photon spectroscopy² with frequency doubled laser light was used, a technique which was found during the test experiments to be extremely sensitive to the spectral purity of the laser light.

Besides being Doppler free, two-photon spectroscopy offers some attractive advantages for the investigation of short-lived isotopes:

- (i) All atoms interact with the laser light, irrespective of their velocities.
- (ii) Wall scattered laser light can be blocked by filters since the wave length used for excitation is very different from that of the fluorescence.
- (iii) In $ns^2\ ^1S_0 - nsn's\ ^1S_0$ transitions such as those of Hg studied in this work, the optical transition strength is combined in one resonance line, allowing a direct measurement of the IS between different isotopes. The IS's of the neutron deficient isotopes of Hg and Au with mass number $A \leq 185$ are very interesting from the nuclear physics point of view because of a sudden nuclear shape change taking place in this mass region^{7,8}.

A. Measurement of the Lifetimes

In a first experiment, the lifetimes of the $6s7s\ ^1S_0$ state of Hg and of the $8s\ ^2S_{1/2}$ state of Au were determined. These data are needed to calculate the transition probabilities for the two-photon excitation. Fig. 4 shows the signal for the case of Au. An ultrafast multichannel analyzer⁹ with a dwell time of 2 ns was started by the laser pulse and the single photon events detected by a photomultiplier were stored in the channels of the multichannel analyzer according to their delay with respect to the laser pulse. Nine measurements each were carried out on Hg and Au. The best fit of exponential functions to the data resulted in the following values of the lifetimes of the states:

$$\tau(6s7s\ ^1S_0, \text{Hg}) = 31.0(2)\text{ ns} \quad (9)$$

$$\tau(8s\ ^2S_{1/2}, \text{Au}) = 36.9(4)\text{ ns} \quad (10)$$

The lifetime of Au in the 8s state was previously unknown. The result for Hg agrees very well with the previously published values of 30.3(8) ns (Ref. 10) and 31.0(5) ns (Ref. 11).

B. Hyperfine Structure and Isotope Shift

These measurements were performed on thermal atomic beams of Hg and Au. A typical scan is shown in Fig. 5. During the experiment it was found that better signal to noise and signal to background ratios were obtained by detecting the ions created by the absorption of a third photon rather than the resonance fluorescence after just the two photon transition. In order to obtain a well defined time overlap of the two laser beams needed for the two photon process, the frequency doubled laser beam was split into two beams of identical pulse energy but of opposite circular polarization. These two beams were sent collinearly in opposite directions through the atomic beam. This arrangement assures that due to the selection rules no Doppler broadened signal can be obtained since the excitation cannot occur for two photons coming from the same direction. Indeed the resonance signal disappeared completely when one of the circularly polarized laser beams was blocked.

Nevertheless, it is clear from Fig. 5 that while there is no Doppler broadened background, there is a broad background under the narrow two photon resonance. An explanation of this is given in section 5.C.

1. Isotope Shifts in the $6s^2\ ^1S_0 - 6s7s\ ^1S_0$ Transition of Hg

Eleven scans of the resonances in a mixture of natural Hg isotopes were performed. The frequency differences of the resonances relative to the resonance of ^{204}Hg were determined by a best fit of the data from each scan. The averaged values of the IS's obtained from these frequency differences are given in Table 2. Since the IS's of Hg have already been determined in the $6s^2 - 6s6p\ ^3P_1$ transition (Schweitzer¹²) and in the $6s6p\ ^3P_1 - 6s6p\ ^1S_0$ transition (Blaise¹³), the sum of these IS's can be compared with our measurements. However, Blaise gives values only relative to ^{200}Hg because the IS of ^{204}Hg could not be reliably determined. Hence in Table 2 the comparisons with the previous work is made through the differential IS's between adjacent isotopes.

The agreement is excellent except for the isotope pair 201,202 where the two measurements deviate by 1.5 standard deviations. The consistency of our data with the IS's measured very accurately in the intercombination line by Schweitzer¹²), can be verified by a King plot¹⁴. Fig. 6 shows the result of such a plot, indicating the validity of our results.

2. Hyperfine Structure of the $8s\ ^2S_{1/2}$ State of Au

Fourteen measurements were performed on the $6s - 8s$ transition in Au where the selection rules allow two resonances. The separation of these resonances;

$$\delta\nu = 2A(6s\ ^2S_{1/2}) - 2A(8s\ ^2S_{1/2}) = 5.96(10)\text{ GHz} \quad (11)$$

together with the well known $6s$ HFS¹⁵ allows the HFS of the $8s$ state to be evaluated. The result

$$A(8s\ ^2S_{1/2}, ^{197}\text{Au}) = 69(50)\text{ MHz} \quad (12)$$

can be compared to the value of $A = 148\text{ MHz}$ calculated from the Fermi-Goudsmit-Segre formula¹⁶. However, no definite conclusions can be drawn from the comparison because of the large experimental errors. These errors are due to the uncertainty of the frequency calibration caused by the availability of only two tabulated I_2 absorption lines⁶ in the neighbourhood of the two photon resonances.

C. The Broad Background Under the Two Photon Resonances

All the two photon resonances observed in this work consisted of a narrow resonance on top of a broad background. The ratio of heights of the narrow resonances to the broad background was found to depend critically on the alignment of the laser system components. As an example, the two photon signal of Hg is shown in Fig. 7 together with fitted Gaussian curves. The fitting parameters were 300 MHz FWHM for the narrow resonances, 11 GHz FWHM for the broad background and a ratio of 0.24 for the height of the broad background relative to the narrow resonance. As already mentioned, it was proved that the broad background must be true two photon resonances with each photon coming from the separate beams.

This unexpected effect is explained as follows. In the frequency doubling process, not only harmonic generation but also frequency mixing takes place. Assuming a spectrum of the laser intensity given by $I_L(\nu)$ at the fundamental frequency, the intensity profile after frequency doubling is approximated by

$$I(2\nu) = \int_{-\infty}^{+\infty} I_L(\nu-\Delta\nu)I_L(\nu+\Delta\nu)d\nu \quad (14)$$

Essentially the same mixing takes place again in the two photon excitation. As a consequence, the ratio of the narrow resonance to the broad background is drastically reduced from the ratio of narrow band light to broad background in the original laser light.

This phenomenon is illustrated in Fig. 8. Starting with a ratio of 20:1 in the primary light, the ratio decreases to 4:1 after frequency doubling and is further reduced to 1:2 in the two photon process. Hence, two photon spectroscopy with frequency doubled laser light is extremely sensitive to small admixtures of broad band laser light in the narrow band laser output. For example, the broad band background shown in Fig. 5 is explained by a ratio of 60:1 in the heights of the narrow band and broad band components of the laser light. A broad band background of this magnitude could be due to the Airy function of the etalon. Such small admixtures are difficult to observe with a spectrum analyzer and yet can severely affect the applicability of pulsed frequency doubled lasers to two photon spectroscopy.

6. SUBSEQUENT IMPROVEMENTS TO THE SYSTEM

Subsequent to the work described above, at Coopervision Laser division Inc. an etalon with a higher surface finesse and slightly reduced reflectivity, resulting in the same total finesse of $F = 100$, was inserted in the dye laser oscillator system. The cavity losses were thereby reduced and narrower bandwidths of laser output were achieved. With an output coupler reflectivity of 40% the bandwidth was reduced to 120 MHz, with a reflectivity of 65% to 70 MHz and with a reflectivity of 85% to between 30 and 40 MHz.

To achieve these bandwidths, two major problems had to be addressed;

- i) excessive background and a tendency toward multimode operation due to the extra pump power required to overcome the extra filtering of the etalon
- ii) increased sensitivity to non-linearities in the tuning of the laser mode by the rotating galvoplate.

The problems due to higher pump power can be reduced by substituting a ring cavity for the linear cavity. Because of space limitations in the existing laser pressure chamber, a "pseudoring" cavity of the twisted mode type first proposed by Evtuhov and Siegman¹⁸ was used here. By placing the active medium between two crossed $\lambda/4$ plates, the linear polarization is split into two counterrotating polarizations thereby inhibiting the standing wave pattern in the active medium and leading to a more axially uniform energy density in the cavity. This therefore reduces the spatial hole burning which leads to the multimode operation.

A schematic of the pseudoring cavity is shown in Fig. 9. Using this cavity the output power of the laser oscillator was boosted to between 100 and 200 μJ . Comparing the transmitted intensity through a confocal spectrum analyzer in the transmission maximum to the background outside the peak gives a contrast ratio of about 1000:1 in narrow band light to broad background light.

The problems due to non-linearities in the mode tuning system were overcome by using an electronic compensator between the output of the pressure transducer and the servo-controlled galvoplate tuner (model G100PD of General Scanning Corp). With this modification, pressure scans over a range of 1000 mbar, i.e. a frequency range of 142.5 GHz at a wavelength of 600 nm, were achieved with neither mode hopping nor double mode operation. For long term stability however, the laser chassis and the dye have to be temperature controlled.

7. CONCLUSIONS

We have shown that a relatively broad band pulsed dye laser can be modified so that it delivers light of bandwidth as low as 30 MHz with contrast ratios of narrow band light to broad band background light as high as 1000:1 and with high power. The capabilities of this instrument make it a very versatile tool for nonlinear two photon absorption spectroscopy and two photon resonant, three photon ionization spectroscopy providing care is taken in suppressing the broad band background in the laser amplifier output.

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*) Present address: CERN, Geneva, Switzerland.

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Table 1:
Laser properties.

	Oscillator	1st Amp.	2nd Amp.	3rd Amp.	4th Amp.
Pump Energy:					
at $\lambda = 355$ nm					
(30 mJ pulse)	8%	12%	12%	20%	48%
at $\lambda = 532$ nm					
(60 mJ pulse)	3%	8%	8%	12-20%	62-70%
Dye conc.					
(molar):					
C540A	1.2×10^{-2}	5×10^{-3}	5×10^{-3}	1×10^{-3}	8×10^{-4}
R640	1×10^{-3}	5×10^{-4}	1×10^{-4}	5×10^{-5}	1×10^{-5}
Output:					
C540A	10 μ J	100 μ J	200 μ J	0.5-0.7 mJ	2-4 mJ
R640	1-10 μ J	100 μ J	500 μ J	2 mJ	10 mJ
Bandwidth:					
C540A with R of output coupler = 40% and no external etalon				-	180-220 MHz
R640 with R of output coupler = 65% and no external etalon				-	100 MHz
" with R of output coupler = 65% and external etalon				-	60 MHz

Table 2:

Isotope shifts in the $6s^2 \ ^1S_0 - 6s7s \ ^1S_0$ transition of Hg isotopes of mass number A relative to ^{204}Hg and relative to adjacent isotopes one mass unit greater. All values are given in MHz.

A	198	199	200	201	202
$\delta\nu^{A,204}$	12726(29)	12196(26)	8754(19)	7401(13)	4322(13)
$\delta\nu^{A,A+1} (\S)$	528(38)	3428(30)	1349(21)	3005(35)	4263(65)
$\delta\nu^{A,A+1} (\S\S)$	530(39)	3442(32)	1353(29)	3079(18)	4322(13)

$\S$ - Ref. 11 and 12

$\S\S$ - This work

FIGURE CAPTIONS

Figure 1:

Layout of the pulsed laser system. The overall path length from the first dye cell to final output beam splitter is about 4 m. The symbols on the diagram represent:

A	Amplifier for Galvoplate drive	GP	Galvoplate
BP	Brewster polarizer	L	Lens
BS	Beamsplitter	M	Mirror
C	Beam height compensator	OI	Optical isolator
CFP	Confocal Fabry-Perot etalon	P	90° Prism
CL	Cylindrical lens	PC	Pockels cell
CM	Capacitance manometer	PD	Photodiode
D	Diaphragm	PH	Pinhole
DA	Diode array	Po	Polarizer
DC	Dye cell	SHG	Second harmonic generator
E	Etalon	T	Telescope
G	Grating	THG	Third harmonic generator
		W	Window

Figure 2:

The etalon response curve compared to the axial modes of the cavity. The heavy arrows to the right refer to the relative motion of the axial mode frequencies under the etalon envelope as the index of refraction of the etalon medium is increased.

Figure 3:

The output intensity of a narrow-band filtered pulsed dye laser system as the cavity gas pressure is increased so as to scan the laser frequency downward (without Brewster galvoplate).

Figure 4:

Measurement of the life-time of the $8s^2S_{1/2}$ state of Au. The fluorescent light was time differentially recorded for 45000 laser pulses.

Figure 5:

Two-photon absorption spectrum of natural mercury, recorded with an external etalon and a laser cavity output coupler of 65% reflectivity. The frequency intervals shown are the isotope shifts for the mercury transitions. Since the spectrum is obtained with photons of double the laser output frequency, the laser frequencies will be 1/4 of these frequencies.

Figure 6:

Kingplot of isotope shift data on natural mercury for the $6s^2^1S_0 - 6s7s^1S_0$ transition (this work) and the $6s^2^1S_0 - 6s6p^3P_1$ transition¹¹. The scale of the insert diagrams is 40 times that of the full scale diagram.

Figure 7:

The signal for the $6s^2^1S_0 - 6s7s^1S_0$ transition in mercury with gaussians fitted to the narrow resonances for each isotope and to the broad background.

Figure 8:

The enhancement of the background in 2 photon spectroscopy caused by mixing of the background light and the narrow band light in the frequency doubling and the 2 photon processes.

(a) The spectrum of the original laser light with a relative height of narrow band peak to broad background of 20:1.

(b) The spectrum of the frequency doubled light. The relative heights of the narrow peak to the broad background is now 4:1.

(c) The 2 frequency-doubled photon signal. The relative heights of the narrow peak to the broad background is now 1:2.

Figure 9:

A schematic of the twisted mode "pseudoring" cavity. The grating, the etalon and the polarizing beam expander are contained within the pressure chamber. The symbols on the diagram represent:

G	Grating	E	Etalon
PBE	Polarizing Prism	DC	Dye Cell
	Beam Expander	GP	Galvoplate
BCP	Brewster Compensator	M	Mirror
	Plate		

Mailing Addresses:

H.-Jürgen Kluge
CERN
Geneva 23
Switzerland

Klaus Wallmeroth, Georg Bollen
Institut für Physik
Universität Mainz
D-6500 Mainz
Federal Republic of Germany

Horst W. Schaaf
Coopervision Inc., Laser Division
Santa Clara
California 95051
USA

Robert B. Moore
Foster Radiation Laboratory
McGill University
Montreal
Quebec, H3A2B2
Canada

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9