Computer Controlled Intracavity Frequency Doubling
of a CW Dye Ring Laser

by
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Sponsored and Monitored by:

Applied Photochemistry Division
Los Alamos Scientific Laboratory
Department of Energy
under grant No. E(29-3)3744

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1. ABSTRACT

The development of a computer controlled tunable ultraviolet spectrometer utilizing intracavity second harmonic generation in a single frequency CW dye ring laser is reported. With 5 W of argon ion laser pump power at 514.5 nm, the spectrometer can generate typical single mode and multimode UV output powers of 20 mW (linewidth \pm 40 MHz) and 40 mW (linewidth \sim 80 GHz) respectively at the peak of the Rh6G laser power spectrum. Wavelength tuning extends from 287 to 305 nm utilizing the organic dye Rh6G and employing temperature phase matching of the nonlinear optical crystal ADA. The performance characteristics of the laser spectrometer have been evaluated by studying the absorption and fluorescence spectra of \text{SO}_2 around 295 nm. Detailed Doppler limited continuous single frequency UV scans of up to 1600 GHz have been routinely achieved.
11. REPORT ON RESEARCH ACTIVITIES (June 15, 1978 - Sept. 30, 1979)

A. Introduction

We report here the results of the third stage of our research into the development of a computer controlled high resolution, high power cw laser spectrometer, tunable in the visible and ultraviolet. During the first stage of this program cw ultraviolet radiation, tunable from 211 to 410 nm, was generated by mixing the outputs of various dye lasers and high power ion lasers [1,2]. The second phase of this effort entailed the development of a computer controlled laser spectrometer consisting of a high power cw argon or krypton ion pump laser, a linear or ring cavity cw dye laser, an extracavity nonlinear optical crystal, diagnostic instrumentation to monitor wavelength and output powers and a DEC PDP-11 V03 minicomputer with a CAMAC interface system [3,4].

The objective of the third stage of this research was to increase by an order of magnitude the ultraviolet output power of the spectrometer through the substitution of intracavity second harmonic generation (ISHG) for the less efficient extracavity SHG used previously. During the past year we have (1) reviewed the techniques for ISHG in cw dye lasers developed by Gabel et.al. [5], Frolich et.al. [6] and Fergusson et.al. [7], and in cw Nd:YAG lasers by Hon [8]; (2) consulted with two commercial firms, Spectra Physics [9] and Coherent Radiation [10], which have performed research on ISHG with their cw ring dye lasers, and (3) developed a modified version of the Rice-LASL spectrometer suitable for ISHG which meets or exceeds our design goals.
B. Design of a CW Ring Dye Laser Suitable for Intracavity Second Harmonic Generation

The original LASL laser spectrometer [3,4] has been modified for intracavity frequency doubling by providing an additional beam waist for efficient SHG in a nonlinear optical crystal as shown in Fig. 1. The cavity has been designed to provide optimum focusing of the beam on the dye jet and inside the SHG crystal using readily available optics. It was necessary to position the crystal focus sufficiently far from mirrors M3 and M5 to provide space for insertion of an intracavity nonlinear optical crystal oven. A Fortran IV program was used to determine stable cavity dimensions and beam parameters for each possible configuration. Due to the strong interdependence between the dye and crystal focal radii it is difficult to achieve small beam waists at both positions. The dye jet focal radius has been slightly enlarged compared to the previous configuration [4] in order to achieve a crystal focal radius of the optimal value. The final configuration yields beam radii ($\frac{1}{\varepsilon}$) of 12 μm at the jet and 45 μm at the nonlinear crystal focus (Fig. 2). The slight enlargement of the jet radius (3 μm) over the previous configuration should have little, if any, effect on laser efficiency. The 45 μm crystal waist corresponds to an optimal crystal length of about 10 cm [11]. However, the crystal lengths we considered were in the 1.5 cm to 3 cm range which represented a practical compromise. This is not a serious design flaw since conversion efficiency is only a weak function of focus near the optimal value. Attempts to decrease the crystal beam waist would have increased the dye focal waist, placed the focus too close to mirror M2 or greatly increase cavity dimensions. It should also be noted that both crystal absorption and thermal distortions
increase at higher fundamental intensities.

The UV output reflector (M5) is a mirror highly reflecting in the visible (R = 99%) while transmitting 80% of the UV. This output coupling method has several advantages over other methods. It does not require additional cavity elements while still retaining the advantage of separating the UV from the fundamental output. In addition, the reflector allows the variation of the fundamental output coupler M4 to optimize both the UV and visible output powers. One minor problem with mirror M5 is the leakage of the fundamental (~ 20 mW) when using a low transmittance fundamental output coupler for M4. The UV output is also diverging and requires refocusing with a f = 30 cm lens.

The wavelength selective elements of the laser are a birefringent filter, a thin c-axis quartz etalon, an air-spaced etalon and for high resolution, a PZT translated cavity mirror as for the previously reported LASL laser [3,4]. As an alternative tuning method electrooptic tuning was evaluated as described in Appendix A. A Brewster cut Faraday glass rod (L = .9 cm, H0 = 3.5 k gauss) with the c-axis etalon ensures unidirectional travelling wave single frequency operation of the ring laser. Mirror feedback for unidirectional operation is used for multimode laser action since the Faraday isolator considerably reduces output power (~25%) and requires tuning of the c-axis etalon (FSR = 200 GHz, L = 0.5 mm). The Faraday isolator provides a single mode output suitable for coarse resolution scanning over large frequency ranges (up to 1600 GHz). However, the effective linewidth is of the order of 2 GHz. Insertion of the thick air-spaced etalon (FSR = 30 GHz, L = 5 mm) provides a stable tuning single mode output.
DYE JET BEAM WAIST
R = 12 μm

THIRD BEAM WAIST
R = 380 μm

M4
R = ∞

M2
R = 10 cm

M3
R = 5 cm

M5
R = 30 cm

CRYSTAL BEAM WAIST
R = 45 μm

80 cm

AXIAL MODE SPACING, $\frac{c}{L} \approx 167$ MHz
Typical operating characteristics for the laser are shown in Table 1. A 6% visible output coupler (H4) is used to obtain the highest fundamental powers with the greatest single mode stability. A 2% output coupler considerably reduces the fundamental power (up to 40%), but sufficiently enhances intracavity power to provide greater UV powers. However, using a maximum reflector for mirror H4 did not produce significantly more UV. Figure 3 shows the experimental dependence of the UV power on the fundamental output power of the dye laser. The curve is nearly parabolic at low intracavity powers, but shows a saturation effect as the intracavity power increases. This drop in efficiency is primarily caused by thermal effects in the ADA crystal. High fundamental and UV powers create radial (and to a lesser extent longitudinal) temperature gradients which eventually saturate the nonlinear optical conversion process [13].

Maximum multimode UV powers are obtained by using the birefringent wavelength tuning filter and mirror feedback. A record 63 mW was obtained with 6 W of argon laser pump power at 514.5 nm, and 57 mW output for 5 W of pump power. However, these powers were unstable for reasons previously stated, and rapidly decayed by about 30%. These peak powers could only be reobtained by either momentarily tuning the laser away from resonance, blocking the pump beam, or by spatial translation of the crystal. It should be noted that there are considerable UV losses in the laser cavity due to reflection at the Brewster surface of the ADA crystal (25%) and the dichroic mirror H5 (20%).

The most important consideration after optimization of the fundamental dye laser power is the positioning of the crystal at the beam waist. Once the
<table>
<thead>
<tr>
<th>Specification</th>
<th>Visible:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength Range (20% points)</td>
<td>570 nm to 615 nm</td>
</tr>
<tr>
<td>Multimode Output Power</td>
<td>1.1 W at 5 W pump</td>
</tr>
<tr>
<td>Linewidth 40 GHz</td>
<td></td>
</tr>
<tr>
<td>Single Mode Output Power</td>
<td>800 mW at 5 W pump</td>
</tr>
<tr>
<td>Linewidth 40 MHz</td>
<td></td>
</tr>
<tr>
<td>Ultraviolet:</td>
<td></td>
</tr>
<tr>
<td>Brewster Surface Crystal:</td>
<td></td>
</tr>
<tr>
<td>Multimode Output Power</td>
<td>40 mW 5 W pump</td>
</tr>
<tr>
<td>Linewidth 40 GHz (visible)</td>
<td></td>
</tr>
<tr>
<td>Single Mode Output Power</td>
<td>18 mW 5 W pump</td>
</tr>
<tr>
<td>Linewidth 40 MHz (visible)</td>
<td></td>
</tr>
<tr>
<td>Normal Incidence Crystal:</td>
<td></td>
</tr>
<tr>
<td>Single Mode Output Power</td>
<td>25 mW 5 W pump</td>
</tr>
<tr>
<td>Linewidth 40 MHz</td>
<td></td>
</tr>
<tr>
<td>Angle-Matched Normal Incidence ADP Crystal:</td>
<td></td>
</tr>
<tr>
<td>Single Mode Output Power</td>
<td>10 mW 5 W pump</td>
</tr>
<tr>
<td>Linewidth 40 MHz (visible)</td>
<td></td>
</tr>
</tbody>
</table>

1 utilizing a $2 \times 10^{-3}$ molar solution of R6G in Ethelene Glycol
2 for the 514.5 nm line of an argon ion laser
3 ADA crystal purchased from INRAD, L = 15 mm
4 ADA crystal purchased from INRAD, L = 30 mm
5 ADP crystal, 65° cut, from Quantum Technology, L = 25 mm
6 available only temporarily due to UV decomposition of index matching material
FIG. 3

UV OUTPUT POWER, mW

FUNDAMENTAL OUTPUT POWER, mW
crystal is nearly centered at the focus, translations of $\pm 20\%$ of crystal length will have a negligible effect. It was found that if mirror feedback is being used, the retroreflected beam must be aligned for maximum UV power rather than maximum fundamental power.
C. ISHG Crystal Considerations

The nonlinear optical crystal ammonium dihydrogen arsenate (ADA) has been used for most of our investigations. The principal reason for this choice of crystal is that ADA is the only KDP isomorph that can be noncritically (90°) temperature phase matched over nearly the entire range of Rh6G from 565 to 610 nm (see Fig. 4) and 90° phasematching is more efficient than angle tuning. However, for some applications, such as generating wavelengths beyond the 610 nm high temperature limit (100°C) of ADA, RDP or angle tuned crystals may be necessary. We have thus briefly investigated angle-matched ISHG in a 65° cut normal incidence ADP crystal and found the technique to be feasible.

The SHG crystal can be obtained with either a Brewster cut or flat end surface. Normal incidence crystals require antireflection coatings which must be transmitting to both the fundamental and the second harmonic. The AR coatings should not be applied directly to the crystal surface since this has been shown to lower the crystal damage threshold by an order of magnitude. For an AR coated window to be effective it must be attached to the crystal surface with index matching fluid. However, index matching fluids tend to be lossy both to the fundamental and the UV, and introduce thermal lensing effects at high power densities. These problems can be avoided by using Brewster cut crystals. The only significant problem with a Brewster cut rod is the uncompensated UV reflectance at the crystal surface.

The nonlinear crystal quality is the most important factor in high power intracavity SHG. Crystals of poor quality deplete the intracavity power through absorption and scattering. Crystal absorption creates thermal effects
which in turn destroy the phase matching condition and defocus the fundamental beam. There is a strong relationship between crystal length, crystal absorption, the linear loss of the laser and optimal SH output coupling [7]. In general, the lower the absorption, the longer the crystal that can be used for optimal SH output coupling. A longer crystal has a higher conversion efficiency and a less severe fundamental focusing requirement. With no method of prejudging crystal quality we opted to use shorter crystals with small cross-sections. The small cross-section facilitates uniform heating and requires a smaller oven.

ADA crystals from INRAD (5 x 5 x 15 mm, 5 x 5 x 30 mm), Quantum Technology (5 x 5 x 5 x 15), and Cleveland Crystals (5 x 5 x 20 mm) were purchased. The 30 mm long crystal was the only normal incidence crystal obtained. The 15 mm INRAD Brewster cut crystal provided the largest UV powers primarily because it had the lowest insertion loss. The Quantum Technology crystal had a slightly larger insertion loss and the same conversion efficiency. The Cleveland crystal had twice the insertion loss of the INRAD crystal, but also a larger conversion efficiency. These qualities were apparently related to the longer crystal length. However, the conversion efficiency was less than what might be expected by direct comparison. As a result the Cleveland crystal produced the lowest UV powers. Also the Cleveland crystal had the poorest optical quality. Fundamental power varied by a factor of two with very small lateral translations of the beam over the crystal surface. This made optimization of the laser alignment extremely difficult. The lower crystal quality also considerably reduced the tuning range of the laser. With the Cleveland crystal scan-
ning above 590 nm was difficult due to occasional frequency hops of around 15
nm. This appeared to result from competition between the nonlinear crystal
and the birefringent filter in wavelength selection. In general, we found
INRAD ADA crystals to be the most suitable.

As mentioned previously, the use of an index matching fluid between the
crystal and the AR coated windows is desirable with normal incidence crystals.
This fluid must be compatible with the water soluble crystal and must transmit
without damage intense visible and ultraviolet radiation. Four materials
(fluorocarbon FC-104, silicon oil, carbon tetrachloride and vacuum leak seal-
ing resin) were tested, but none withstood the UV output for more than a few
minutes without substantial decrease in UV power. However, the SHG efficiency
for the normal incidence ADA crystal was significantly higher for brief peri-
ods than that observed with the Brewster cut crystal. The lower efficiency
of the Brewster cut crystal is presumably due to its shorter crystal length,
UV reflection at the crystal surface and uncompensated astigmatism. Thus the
normal incidence configuration would be superior if a suitable index matching
fluid was available.
D. Temperature Phase-Matching of ISHG Crystal

Reliable and convenient temperature control of the nonlinear optical crystal is essential for a tunable spectrometer employing ISHG. Figure 5 depicts UV power output at constant ADA crystal temperature (55°C) for a scan of 1500 GHz in the visible near 295 nm. The FWHM of 80 GHz for ADA places an upper limit on the useful range of a continuous single mode UV scan at any one temperature. The computer controlled LASL-Rice laser system has the unique advantage of precise frequency control through the computer interfacing of the laser elements. Hence, tunable UV output is only dependent on providing an equally precise method of controlling crystal temperature.

The temperature bandwidth of ADA is shown in Figure 6. The FWHM of .35°C requires a temperature stabilization of at least ± .02°C for useful and stable UV output. We initially investigated two commercial crystal ovens (from Quantum Technology and INRAD controlled by a YSI Model #72 Temperature Controller). Both ovens had large thermal time constants and lacked temperature reproducibility. The temperature stability of each of these ovens was also an order of magnitude lower than required for our purposes. In addition, neither of the ovens was easily adaptable to a Brewster cut crystal. Hence we designed our own compact oven for placement in the tight configuration of the ring laser cavity (see Fig. 7). A long term stability of ± .01°C from -25°C to 100°C has been achieved for this oven when used with an Artronix Model #5301 Temperature Controller. Temperatures below +10°C are not recommended due to condensation and frost on the oven windows. At the higher temperatures windows are not necessary which yields greater UV powers. An advantage of cooling is increased
UV POWER NEAR 295.0 NM
oven response below 40°C where convection cooling is very inefficient.

The Artronix controller has been interfaced to the computer through a CAMAC controlled stepping motor. The stepping motor has higher resolution and lower noise than can be conveniently obtained by direct electrical interfacing. The present resolution varies from 0.02°C (25°C) to 0.06°C (100°C) but can easily be increased by suitable gearing of the stepping motor (See Appendix B). The maximum slew rate (3.5°C/min.) has an overshoot and settling time of around .3°C and 1 minute, respectively. No active computer monitoring of oven temperature has been found to be necessary.

The amplitude stability of the UV output is primarily limited by the noise of the fundamental. However, at high output powers the amplitude fluctuations of the UV can be significantly less than the fundamental noise due to saturation effects. There are also several systematic sources of UV noise that result from the computerized frequency scanning. The UV output depends critically on the synchronization of frequency and temperature during scans. Frequency instability and occasional mode hops can have a significant effect on the UV output. Noise due to temperature fluctuations and the incremental steps of the stepping motor is virtually eliminated by the large thermal mass and continuous temperature tracking of the oven.

A significant problem in producing uniform UV powers has been the initial lag in the oven response to a computer imposed temperature change. This lag time is typically about 15 sec but obviously depends on the rate at which the temperature is being changed. This lag time cannot be recovered and primarily determines the degree to which temperature and frequency are mismatched. The oven response time can only be eliminated through the use of a smaller oven
which would tend to increase thermal gradients across the crystal. We have made allowances for this time lag by referencing the temperature scan to a dummy scan which leads the actual laser frequency by 15 seconds. Useful UV output powers of 10 mw with a maximum deviation of only 2% can be generated over frequency ranges of 1600 GHz with an effective linewidth of about 2 GHz.

We also have had limited success with a new technique for temperature control which would partially eliminate the need for computerized tracking of the oven temperature. An electrooptic modulation of the crystal indices of refraction [8] is used to determine the phase and magnitude of the crystal temperature deviation from the optimal phase matching condition. A small modulation voltage (~ 100 v) is applied along the optic axis of the ADA crystal and the resulting modulation in the amplitude of the UV output is monitored by a lock-in amplifier. This modulation is not of sufficient amplitude (< .1%) to interfere with spectroscopic measurements. The output of the amplifier is used to control crystal temperature via the Arttronix temperature controller. This method of temperature control has proved to be less effective than active computer control due to the thermal time constant of the oven and to a lesser extent laser noise. These problems could be overcome by an improved feedback control loop. However, this method can only be used to supplement computer control since the technique is only applicable for short frequency scans at slow scanning speeds.
E. Computer Operation of the CW Visible-UV Laser Spectrometer

The LASL-Rice laser spectrometer is capable of continuous frequency or wavelength scanning over the entire tuning range of the ring dye laser [3,4]. Each laser tuning element has been interfaced to the DEC PDP 11 V03 minicomputer through a CAMAC controller to provide simultaneous tuning of each element (Figures 8 and 9). An extensive software package has been developed to provide complete operator control over the wavelength scanning and calibration of the laser spectrometer as well as the acquisition, storage, and display of data. The software package consists of thirty commands which control all of the operations of the spectrometer.

The laser tuning elements are controlled by six motors: three stepping motors for adjustment of the birefringent filter, the monochromator and the crystal temperature, and three voltage controlled motors for adjustment of the two intracavity etalons and the cavity length. Calibration or cross referencing of the spectrometer consists of determining wavelength or frequency as a function of motor position for each tuning element. The software has been designed to perform this cross-referencing of each element with a minimum of effort on the part of the operator. Once the spectrometer has been calibrated computer controlled scanning is performed over the calibrated region by simultaneous tracking of each relevant motor.

The spectrometer is capable of scanning in a wavelength or frequency mode of operation depending on the number of intracavity elements. Coarse wavelength scanning is performed with only the birefringent filter. The resolution is then limited by the linewidth of the laser to about .5 Å (40 GHz).
When the Faraday isolator is inserted into the cavity the thin etalon must be frequency calibrated along with the birefringent filter to perform continuous scans of up to 1600 GHz with a resolution of about 2 GHz. The air-spaced etalon can be used externally for calibration of the thin etalon and as a marker cavity. When the air-spaced etalon is inserted into the cavity, frequency scans of 50 to 1600 GHz can be performed with a resolution determined by the ring cavity mode spacing of 170 MHz. Finally, a PZT translator can be used to alter cavity length in order to perform scans with a resolution limited by the linewidth of the laser. Up to 1000 data points can be taken from each of four ADC channels during scans. Software adjustment of the scanning speed and a time constant for digital filtering optimizes scanning resolution over any frequency or wavelength interval. Data can be displayed on either a video terminal or an X-Y recorder after acquisition and storage.

The software developed previously for extracavity angle tuned SHG has been updated to permit intracavity temperature tuning of the nonlinear crystal. As described in Section II C, the crystal temperature is controlled directly by a stepping motor interface with no active monitoring of the crystal temperature by the computer. Calibration of the crystal temperature and phase matching wavelength to motor position is performed by the software from the known characteristics of the temperature controller and the ADA crystal. The software has been designed for continuous UV frequency scans of up to 3200 GHz.

Figure 5 demonstrates the resolution obtainable when scanning the laser spectrometer without the intracavity air-spaced etalon. In this mode of operation resolution is limited to 2 GHz due to axial mode hops. The fluorescence
spectrum of I₂, in the visible and the absorption spectrum of SO₂, in the UV, have been recorded with the laser spectrometer. Figure 10 is a typical long range scan (over 800 GHz) obtained with the intracavity air-spaced etalon. Figure 11 is a higher resolution absorption spectrum taken over 320 GHz of SO₂ (5 torr, path length 25 cm, laser probe power 5 mw, linewidth ± 20 MHz).
300.04 to 300.14 nm
UV Power
Marker Cavity
8 GHz Fundamental
I2 Fluorescence
Spectrum
SO2 Absorption
Spectrum

Intensity [Arbitrary Units]

32  96  160  224  UV Freq [GHz]
F. Summary

A list of our accomplishments includes the following:

(1) Design of a ring laser configuration for single frequency, high power CW dye laser operations.

(2) Generation of over 50 mw of UV power by means of intracavity second harmonic generation in a CW dye ring laser spectrometer. Typical useful tunable single frequency UV powers of 15 mw with a narrow linewidth (± 20 MHz) in the tuning range 287 nm to 305 nm.

(3) Evaluation of optimum nonlinear optical crystal and phase matching for the dye ring laser.

(4) Development of a computer controlled CW dye laser spectrometer, which features automatic calibration, low and high resolution wavelength scans, data acquisition and display. Typical single frequency scans of 100 GHz to 1600 GHz have been routinely achieved.

(5) The spectrometer has been used to study SO₂ absorption around 295 nm.

(6) Evaluation of an electrooptic birefringent wavelength tuning system for the dye laser spectrometer.
III. REFERENCES


APPENDIX A

Electro-Optic Tuner Performance

This appendix describes the performance of two electro-optic tuning devices (EOT's), in the modified cavity of the Rice-LASL ring laser. The tuners are similar in design to that described by Telle and Tang [12]. Briefly, the tuner consists of a uniaxial crystal (e.g. KD*P) which is placed in the laser cavity so that the beam propagates in the yz crystal plane at a small angle (~ 1°) to the z-axis. The beam is initially polarized along the (110) plane and due to the crystal birefringence only certain wavelengths emerge from the crystal in the initial polarization state. Polarization selective elements such as Brewster plates and the dye jet then discriminate against all other wavelengths. The pass wavelength of the tuner is varied electrically by application of a voltage to electrodes attached to the crystal which changes the refractive indices through the linear electro-optic effect.

Each tuner supplied is packaged in a cylindrical housing with a 2" concentric mounting ring slightly offset toward one end so that the cylinder is longer on one side of the ring than the other. A small aperture at each end of the housing permits propagation of the beam through the tuner only in or near the proper direction. The laser polarization direction should be along a line from the center of the cylinder to the high voltage pin at each end. The LS-14 tuner is designed to yield a large tuning rate (~ 15 nm/kV) and laser linewidth (~ 0.15 nm) when used as the only tuning
element in the cavity. The LS-24 tuner is to be used in conjunction with the LS-14 to provide a more narrow line (∼0.025 nm) with a reduced tuning rate (∼0.4 nm/kV). Since the maximum voltage which may be applied to each tuner is ±7.5 kV, electrical tuning is limited to ±3 nm about the zero voltage wavelength when both tuners are in use. The exact tuning rate is dependent on the orientation of the tuners with respect to the beam. However, when using the LS-14 alone, a monopolar voltage range of 0–4 kV is sufficient to tune through the useful wavelength range of Rh6G.

When evaluating the LS-14 alone, the tuner was placed in the collimated arm of the ring laser cavity with its long end toward the visible output coupler, M4, and as close to this mirror as possible. The housing was rotated so that the high voltage pins were vertically above the cylinder axis, and aligned so that the laser output wavelength with no voltage applied was ∼565 nm. The pin in the long end was grounded and a negative high voltage signal was applied to the short end connector. In this configuration the tuning rate was 13.9 nm/kV and the observed tuning curve is shown in Fig. 12. The output linewidth ∼120 GHz with the LS-14 as compared to ∼40 GHz with the standard birefringent filter. The maximum output power obtainable with the EOT was ∼85% of that available with the birefringent filter at the peak of the dye curve.

For the evaluation of both tuners together, the LS-14 was realigned to pass a wavelength near the center of the dye curve with no applied voltage. The LS-24 unit was then inserted in the collimated ring laser arm with its long end toward the LS-14 and its short end as close as possible to the UV
FIG. 12

LS-14 TUNING CURVE

POWER [MW]

LAMBDA [A]
out coupler, M5. The angular orientation of the LS-24 was adjusted to return the laser output to the wavelength passed by the LS-14 alone. Insertion of the LS-24 reduced the peak laser output to ~ 40% of the level obtained with the birefringent filter while narrowing the linewidth to ~ 20 GHz. Application of voltage to either the LS-24 or the LS-14 or to both tuners in synchronism resulted in a tuning rate of ~ 0.4 nm/kv.

In summary we find that the electro-optic tuner LS-14 is suitable for rapid long range scanning when only coarse resolution is required. However, this asset is offset by lower output powers as well as increased laser noise. The LS-24, or the LS-14, LS-24 combination is capable of higher resolution scanning but only over a limited range. In addition to the low tuning rate of the LS-24, the EOT's considerably reduce the wavelength range of the organic dye. However, for specialized laser applications requiring the rapid tuning capabilities of the EOT's, the reduced tuning and output power characteristics are acceptable [14].
APPENDIX B

Parts List of Components for ISHG Laser Spectrometer

1. Optical

1 M1:R = 7.5 cm, 1 cm dia. Total reflector for Argon Ion Laser
   Coherent Inc., 3210 Porter Drive, Palo Alto, Ca. 94304,
   (415) 493-2111

1 M2:R = 10 cm, 7.75 mm dia. Total reflector for Rh6G
   Spectra Physics, 1250 W. Middlefield Rd., Mountain View, Ca.,
   94042, (415) 961-2550 (coating: Perkin-Elmer, Electro-Optics
   Division, Norwalk, Conn. 08665, (203) 762-6304)

1 M3:R = 5 cm, 7.75 mm dia. Total reflector for Rh6G
   Spectra Physics

1 MR: Flat 1" dia. 2% output coupler
   CVI Laser Corporation, P.O. Box 11308, Albuquerque, NM 87112,
   (505) 296-9541

1 M5:R = 30 cm, 1" dia. Total reflector for Rh6G, maximum transmission
   285-310 nm
   coating: Perkin-Elmer

1 Birefringent filter, #598-02
   Coherent Inc.

1 .5 mm c-axis uncoated quartz etalon, 1" dia.
   CVI Laser Corporation

1 9 mm long Brewster cut Faraday rotator rod, 6.35 mm dia., FR-5 glass
   Hoya Corporation, 2200 Sand Hill Rd., Suite 200, Menlo Park,
   Ca. 94025, (415) 854-4680

1 Air-spaced etalon, TL-15, 33% broad band coatings CVI
   Burleigh Instruments, Burleigh Park, Fishers, NY 14453,
   (716) 924-9355
1 2-ring magnet stack, 3.5 kG
   Electron Energy Corporation, 329 Main St., Landersville, Pa.
   17538, (717) 898-2294

1 ADA crystal
   Inrad, 181 Legrand Ave., Northvale, NJ 07647, (201) 767-1910

Electro-Scan Tuners. LS-14K and LS-24
   Ithaca Research Corp., 225 Berkshire Rd., Ithaca, NY 14850,
   (607) 257-7074

II. Mechanical

3 Mirror Mounts, #S2P
   Custom Microwave, 7065 Oserbrook Dr., Longmont, Ca. 80501,
   (303) 652-2815

3 Translation Stages, #M50.16
   Klinger, 83-45 Parsons Blvd., Jamaica, NY 11432, (212) 657-0335

5 Flexible Shaft Couplers
   Servometer Corporation, #SC-9, 501 Little Falls Rd., Cedar
   Grove, NJ 07009, (201) 785-4630

1 Dye Jet
   Spectra Physics or Coherent

1 Dye Circulator, Model 376
   Spectra Physics

1 Dye Accumulator, Model 372A
   Spectra Physics

1 3:1 Precision Gear Box with anti-backlash, PIC #ES2
   PIC, P.O. Box 335, Benrus Center, Ridgefield, Conn. 06087,
   (203) 431-1500

1 8:1 Precision Gear Box with anti-backlash, PIC #ES7

1 15:1 Worm and Wheel Assembly, PIC #DQ1
III. Electrical

1 Scanner, #G208
   General Scanning Corporation, 150 Coolidge Ave., Watertown, Ma. 02172, (617) 924-1010

1 Amplifier, #AX200
   General Scanning Corporation

1 Ramp driver, RC42
   Burleigh Instruments

1 Temperature controller, #5301
   Artronix Instrumentation, 4390 Lindel Blvd., St. Louis, Mo. 63108, (314) 371-5314

3 Stepping motors, #M061-FC02 and driver cards
   Superior Electric

1 Operational amplifier - OPS 5000
   Kepco, 131-38 Sanford Ave., Flushing, NY 11352 (212) 461-7000
APPENDIX C

Publications - E(29-2)3744 "Development of a Tunable Ultraviolet CW Coherent Source of UV Light"


Papers Presented at Meetings


M. Sc. Thesis

1. "Generation of Broadly Tunable Coherent Radiation in the UV Using Nonlinear Optical Interactions." E. Weaver


Computer controlled cw laser spectrometer

C. R. Pollock, J. Kasper, G. K. Ernst, W. E. Ernst, S. Blit, and F. K. Tittel

A computer controlled cw UV-visible dye laser source for spectroscopic use has been developed. Computer control facilitates both continuous single-frequency scanning and data acquisition. With 4 W of Ar-ion laser pump power, such a spectrometer can generate in excess of 1 W of cw single-frequency power in the visible and up to 1 mW in the UV by using extracavity nonlinear optical mixing. The laser spectrometer has been tested by performing high-resolution measurements of the fluorescence spectrum of I₂ in the visible and of the absorption spectrum of SO₂ in the UV.

Introduction

The wavelength tunability and the narrow linewidth of the single-frequency output of cw dye lasers are useful in many spectroscopic applications. The spectral range of these lasers can be extended to both the UV and IR regions through use of nonlinear optical mixing techniques. In this paper the development of a versatile computer controlled cw visible-UV dye laser spectrometer will be described. Computer control facilitates a number of interrelated operations that are necessary for multimode and single-frequency scanning of the dye laser and synchronous tracking of the optimum orientation of the nonlinear optical crystal. In addition, the use of on-line computer control permits convenient wavelength and frequency calibration, monitoring and optimization of power, and spectral data acquisition and processing. In fact, several groups have applied computer control to tunable lasers in recent years. Examples include optical parametric oscillators, an electrooptically tuned cw dye laser, and pulsed dye lasers. This work describes a computerized cw laser spectrometer capable of long-range (>20 Å) single-frequency scans in the visible and UV. The scanning and resolution capabilities of the spectrometer have been tested on the I₂ spectrum in the visible region and on SO₂ in the UV region.

Experimental Details of Laser Spectrometer

The computer controlled laser spectrometer is shown schematically in Fig. 1. The spectrometer consists of several components: (a) a high power cw pump laser, either an Ar-ion or Kr-ion laser; (b) a cw dye laser in either a linear or ring cavity configuration; (c) a nonlinear optical crystal; (d) diagnostic instrumentation to monitor wavelength, frequency, and visible-UV power levels; and (e) a minicomputer system.

The central part of the spectrometer is the dye laser. A high-power single-frequency ring dye laser has been developed that is similar to the recently reported Spectra-Physics model 380A dye laser. This laser has a four-mirror figure-eight optical resonator geometry as shown in Fig. 2. One resonator arm has a tight focus and contains the dye jet stream, while the other arm is collimated for insertion of the various wavelength selection elements. Coarse tuning with a linewidth of 40 GHz is accomplished with a three-element quartz birefringent filter. High-power single-frequency scanning requires two additional intracavity optical elements. A coated airspaced etalon (free spectral range (FSR) = 30 GHz, 33% reflectivity) and an uncoated solid etalon (FSR = 170 GHz which limits lasing to a single transmission peak of the airspaced etalon) produced optimum output power and single-frequency scanning with a linewidth of ±20 MHz. The birefringent filter orientation is controlled by a stepping motor (Superior Electric type MO61-FC02) suitably geared to permit a frequency resolution of 6 GHz/step. The thick airspaced etalon is controlled by the voltage output from a digital-to-analog converter (DAC) amplified by a high-voltage ramp generator (Burleigh model RC-42). The thick solid etalon is angle tuned using a galvanometric scanner combination (General Scanning models 208-AX 200), which is controlled by a second DAC output. With such a birefringent filter-etalon combination, single-frequency scans of up to 20 Å with dye laser cavity mode hops at c/L = 200-MHz intervals can
be achieved. Continuous frequency scanning requires synchronous tracking of the effective cavity length (using either a PZT translator or a tilted scan plate responding to a third DAC output) with respect to the airspaced etalon spacing. In order to insure unidirectional laser output from such a traveling wave ring type laser either an external reflector or a nonreciprocal Faraday device with low cavity insertion loss is used. The compact optical isolator used in this ring dye laser consisted of an AR coated Faraday rotator glass rod, 1 cm in length, placed in a cylindrical stack of high field permanent magnets (2kOe) and a 0.5-mm thick c-axis quartz plate, which acted both as a polarization rotator and as an etalon.

With a $2 \times 10^{-3}$ molar solution of the dye Rh6G in ethylene glycol, the dye laser spectrometer produces a $\text{TEM}_{00}$ multimode output power of 2 W with 7-W input pump power and a single-frequency power of 1.5 W with 6-W pump power at 514.5 nm ($\sim 25\%$ conversion efficiency). This represents a considerable improvement in dye laser efficiency as compared with the conventional linear type dye laser.

Generation of tunable cw UV radiation is accomplished conveniently external to the dye laser cavity as shown in Fig. 1 but less efficiently than for intracavity optical mixing. Details of various optimum second harmonic (SHG) and sum frequency (SFM) mixing schemes that generate radiation spanning the UV wavelength region from 410 nm to 211 nm are given in Refs. 9–11. For this system, UV output powers of 1.4 mW (multimode) and 1 mW (single frequency) have been obtained for a fundamental dye laser power of 1.1 W by SHG in a 25-mm long ammonium dihydrogen arsenate (ADA) crystal. The nonlinear optical crystal is placed in a special optical mount containing index matching fluid that minimizes beam displacement as the crystal is rotated to optimize the phase matching conditions. The nonlinear optical crystal is rotated by a stepping motor driven rotation stage with a resolution of 30 arc sec. A separate motor controlled prism serves as both a UV wavelength discriminator and a beam steering device to keep the output direction unchanged.

The dye laser wavelength is monitored by a monochromator. For convenience a 1/4-m Jobin Yvon H-20 monochromator with a spectral resolution of 1 nm is controlled by a geared down stepping motor with an accuracy of 0.3 A/step. A temperature stabilized optical spectrum analyzer with a FSR of 8 GHz served as a reference cavity for calibration of the airspaced etalon and to provide frequency markers during scans. Several visible and UV photodiode-operational amplifier detectors were used to monitor the power levels, as shown in Fig. 1. The signals from these detectors were input to a multiplexed analog-to-digital converter (ADC).
Minicomputer System

The various operations and elements of the laser spectrometer are controlled by a DEC PDP-11 VO3 minicomputer with a 28K word memory interfaced via a CAMAC system shown in Fig. 3. CAMAC interfacing provides a standardized and flexible method for transmitting digital control and data information between computer and various instrumentation modules. This approach allows the use of a wide variety of readily available modular interface hardware, which is compatible with most minicomputer systems.

A crate controller (Kinetic Systems model 3912) is used as the interface between the PDP-11 VO3 and the various CAMAC modules. Stepping motors are operated by a stepping motor controller (Bira Systems model 3101A) followed by stepping motor driver modules (Superior Electric STM 101). The photodiode detectors are connected to a sixteen-channel 12-bit differential input ADC (Bira Systems model 5301). The scanning and cavity length elements of the dye laser are controlled by the voltage outputs of an eight-channel 12-bit DAC (Kinetic Systems model 3112). A real-time clock (SEC model RTC-01) permits adjustment of the scan speed of the various stepping motors to eliminate undesirable resonances. A 24-bit input/output module (NEC model 9017) is used to read the position of all the limit switches associated with the various stepping motors. Several data display options are provided. An X-Y-Z graphics display driver (Bira Systems model 4301) allows convenient real-time monitoring of photodetector data on a Tektronix model 604 oscilloscope. The input/output module is also used to drive two 4-digit alphanumeric displays for readout of current data such as signal magnitude, motor position, wavelength, or power. Two channels of the DAC converter are used to drive an X-Y recorder (Houston Instruments model 2000) for generation of plots of data.

Software Considerations

The software consists of a main program and about forty subroutines, mainly written in Fortran IV, which operate under the DEC RT11 operating system. Some subroutines are written in assembler language to minimize computation time (Fortran versions thereof simply require too much time). Some of the important functions of the software are: (a) coarse wavelength tuning by setting the birefringent filter (BRF); (b) scan rate selection; (c) wavelength and frequency calibration; (d) optimization of UV power by appropriate orientation of the phase matching angle of the nonlinear optical crystal; (e) fine wavelength selection by adjustment of intracavity etalons; (f) tracking of the dye laser cavity length; (g) UV output direction control; (h) data acquisition and processing; and (i) data display, listing, and storage. The laser spectrometer is controlled with up to seven motors consisting of four stepping motors to adjust the BRF filter, the SHG crystal angle, the monochromator, and the UV separating prism and three voltage controlled scanners to adjust the two intracavity etalons and the cavity length. The software allows any motor (stepping or scanning) to be moved to a desired location within hardware or software limits. Data can be taken, filtered with a suitable time constant, stored, and displayed as a function of motor position from any of the various photodetectors as any motor is scanned.

The fundamental concept underlying the design of the software is that the positions of the various optical elements are stable and reproducible enough to permit open-loop operation. Tight closed-loop control of the elements is not necessary, and the software is thus significantly simpler. Initialization of the spectrometer requires the generation and storage of various cross-reference tables, which relate the positions of each motor to a corresponding set of wavelengths or frequencies. First, a table of the BRF position vs mo-
nochromator position is generated by setting the BRF to a number of positions (typically eleven) over the effective tuning range of a given organic dye and adjusting the monochromator for maximum output signal at the exit slit. The resulting data are fitted with a cubic polynomial. This fitted curve is used to create a cross-reference table between 100 BRF motor positions and 100 monochromator motor positions. Positions between these are readily determined by linear interpolation. Since the monochromator position is linear in wavelength with a known ratio, entry of the monochromator reading at only one point calibrates the BRF position in terms of wavelength throughout the dye laser tuning range. The laser spectrometer can now be operated in convenient wavelength units.

A similar procedure is used to construct the cross-reference tables of BRF position vs nonlinear optical crystal phase matching angle and vs prism angle by monitoring the UV power. The software then allows automatic tracking of both the crystal and the prism with the BR filter. For single-frequency operation of the spectrometer, two intracavity etalons are inserted in the dye laser. The proper control of these elements requires the generation of three additional cross-reference tables: the airspaced etalon position vs frequency, the solid etalon tilt position vs frequency, and the BRF position vs frequency. The airspaced etalon is cross referenced by monitoring the output of the 8-GHz marker cavity. Calibration of the position of the tilted etalon requires monitoring the modulation of the output power due to the intracavity 30-GHz airspaced etalon as the tilted etalon is scanned. Finally, a table relating the BRF position to a change in frequency is generated from the wavelength table for a 20-Å range.

Upon completion of these seven tables, which typically takes 30 min, the laser system is ready for high-resolution spectroscopic scanning. Single-mode scan speeds up to 0.2 Å/sec are possible, with adjustable time constants for each of the ADC inputs. Typical single-frequency scans of the order of 90 GHz are obtained by updating each motor position according to the cross-reference tables. The scans can be extended to about 1600 GHz by repetitive computer controlled resetting of the airspaced etalon and tilted etalon. Continuous high-resolution single-mode scans without cavity mode hops can be performed by tracking the dye laser cavity length simultaneously with both etalons. Scanning the dye laser cavity length with a mirror mounted on a PZT translator or a thin intracavity plate set at Brewster's angle allows generation of continuously tunable radiation over a 40-GHz frequency range. During each scan, data from up to four of the various signal sources can be acquired, digitally filtered, and stored in memory. At the end of the scan, these data may be displayed on the graphics terminal, plotted on the X–Y recorder, or stored on a diskette.

**Evaluation of Visible–UV Laser Spectrometer**

Typical multimode output of the laser spectrometer is shown as a function of wavelength in Fig. 4. The visible power for the R6G dye laser is between 0.6 W and 1.0 W for the region from 572 nm to 617.6 nm. Using a 2.5-cm long ammonium dihydrogen phosphate (ADP) crystal for doubling, useful UV output power (although lower than for ADA) is obtained from 286 nm to 308.8 nm. The narrow spike with a full width at half-maximum (FWHM) of 10 Å shows the UV output when the crystal and prism angles are held at fixed angles as the BRF is scanned. The broad UV curve in Fig. 4 shows the output when the crystal and prism angles are continuously tracked according to the cross-reference tables. This output is within a few percent of optimum for the entire wavelength scan. The long-term stability of the spectrometer is excellent, and these cross-reference tables are valid from day to day. In fact, these tables need to be regenerated only if an actual change is made to the spectrometer.

The fluorescence spectrum of I₂ in the visible and the absorption spectrum of SO₂ in the UV served as convenient spectroscopic calibration media for evaluating the laser spectrometer in terms of resolution capability, scan and tuning range, and amplitude and frequency stability. Figure 5 shows the fluorescence spectrum of room temperature I₂ obtained by computer controlled scanning of a multimode R6G dye laser. The experimental apparatus consisted of the laser spectrometer, the I₂ cell, and a photomultiplier. A Doppler linewidth limited high-resolution fluorescence spectrum of I₂ obtained from a single-frequency dye laser scan of 80 GHz (∼1 Å) is shown in Fig. 5. Rotational structure is clearly resolved for this mode of operation of the laser spectrometer.

![Diagram](image_url)
Fig. 5. Low-resolution I₂ fluorescence spectrum from 569 nm to 615 nm; I₂ vapor pressure 0.2 Torr; Rh6G dye laser linewidth = 40 GHz; peak power = 500 mW.

Fig. 7. Low-resolution SO₂ absorption spectrum from 294 nm to 309 nm. SO₂ pressure 2 Torr; cell length 25 cm, peak power 0.05 mW.

Fig. 6. High-resolution I₂ fluorescence spectrum near 600.2 nm; I₂ vapor pressure 0.2 Torr; Rh6G laser linewidth ±20 MHz; probe power 500 mW; scan interval 85 GHz.

Fig. 8. High-resolution SO₂ absorption spectrum near 305 nm, probe power 0.03 mW, UV scan interval 100 GHz.

The UV absorption spectrum of SO₂ at 2 Torr (25-cm path length) obtained by frequency doubling the multimode dye laser output using an ADP crystal is given in Fig. 7. For the absorption measurement, the above mentioned photomultiplier was replaced by a UV sensitive photodiode. A normalized SO₂ spectrum obtained by a single-frequency scanning over a 100-GHz (∼0.3 Å) range in the UV is reproduced in Fig. 8.

The authors thank Suhas Katkar for his helpful assistance and J. Aldridge, D. Milligan, and D. Taylor of the Los Alamos Scientific Laboratory for their stimulating discussions. This work was supported by the U.S. Department of Energy, the Robert A. Welch Foundation, and the National Science Foundation.

J. Kasper is on leave from the Department of Chemistry of the University of California at Los Ange-
les, and G. K. Ernst is on leave from the Institute of Plasma Physics of the Technical University of Hanover. W. E. Ernst is a German Research Foundation Fellow.

References

13. The software sources can be obtained by contacting J. Kasper at UCLA.