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COMPUTER CONTROLLED
INTRACAVITY SECOND HARMONIC GENERATION
IN A CW DYE RING LASER

by

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ABSTRACT

The purpose of this research has been to develop a source of high power narrowline ultraviolet radiation suitable for spectroscopic applications. This was accomplished by using intracavity second harmonic generation in a computer controlled continuous wave dye ring laser. A DEC PDP 11/03 minicomputer was used to control laser tuning elements and nonlinear crystal temperature to provide constant UV output powers of over 10 mW with a short term linewidth of ± 50 MHz at the peak of the Rh6G gain curve. The computer system with a CAMAC interface provided control of laser calibration and scanning as well as data acquisition and storage over the wavelength range from 285 to 305 nm. The spectrometer development included the design of an astigmatically compensated ring laser to provide optimal conversion of intracavity fundamental power to second harmonic UV output. Both Brewster cut and normal incidence crystals were analyzed to determine the most efficient insertion technique. The Brewster cut crystal was found to provide the highest output powers with the fewest complications. The system development also included techniques for long range temperature tuning of the nonlinear crystal to provide constant UV
output over continuous frequency intervals of 3200 GHz. The performance of the spectrometer was evaluated through studying the absorption spectra of SO\textsubscript{2} around 300 nm. A simultaneous visible output at 600 nm was used to provide a reference spectra of the well documented fluorescence of I\textsubscript{2}.
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CHAPTER I

INTRODUCTION

The purpose of this research has been to develop a computer controlled high power ultraviolet laser spectrometer suitable for spectroscopic applications. High power cw output in the UV has been obtained through the use of second harmonic generation (SHG) in a dye ring laser cavity. A calibrated and tunable UV source has been produced by interfacing the laser tuning elements and the crystal second harmonic resonance temperature controller to a DEC PDP 11/03 mini-computer. Extensive software programs enable the calibration and tuning of the laser as well as the acquisition, storage and display of spectroscopic data. The system reliability has been proven through high resolution spectroscopic studies of the absorption spectra of \( \text{SO}_2 \) in the wavelength region near 300 nm.

The need for a source of high power cw UV radiation has been well established. A tunable UV source has numerous applications in spectroscopy such as in investigations of Rydberg states, and saturation spectroscopy. Other potential uses include studies of photo-induced processes, optical recording with UV photosensitive materials and isotope separation. However, tunable cw radiation substantially below 400 nm has not been achieved directly and must await the discovery of a suitable lasing medium. For this reason interest has centered on optical mixing where a nonlinear media is combined with the tunability.
of high power dye lasers to produce cw radiation in the range from 220 to 400 nm. A significant problem with this technique has been the low output powers resulting from the small conversion efficiencies of the nonlinear crystals. Since second harmonic power is at worst linear with fundamental power, the most efficient method of enhancing output has been to utilize the high fundamental powers available inside the dye laser cavity.

There has been considerable activity in dye laser intracavity second harmonic generation over the past decade. In 1972 Gabel and Hercher [1] succeeded in obtaining 1.1 mW of intracavity second harmonic power in a linear Rh6G dye laser. Several groups [2-6] over the next few years refined intracavity techniques by examining the limits on second harmonic power and optimizing laser design to obtain the maximum possible powers. However, single mode output powers did not exceed 5 mW until the dye ring laser came into intracavity use in 1976 [7-8]. This development increased by an order of magnitude the maximum single mode powers due to the high unidirectional fundamental power obtainable with the ring configuration. None of this previous research had been directed at developing a system capable of generating both a stable and widely tunable single mode UV output at these high powers.

The underlying aim of this project has been to demonstrate the feasibility of combining the high power capability of intracavity
SHG with the technological achievement of the computer controlled laser to produce a UV source capable of wide range single mode scanning. It had not been proven that it was possible to use intracavity SHG over substantial wavelength intervals with continuous output without frequency instability resulting from the dispersive loss and birefringence of the nonlinear crystal. There was also the technological question concerning the feasibility of utilizing temperature tuning to phase match the fundamental over substantial wavelength intervals. A computer controlled system is uniquely suited to solving these questions because of its capability for precise control over the many parameters involved.

This project has been a logical continuation of the development of the laser spectrometer developed by Pollock et.al. [9]. This spectrometer utilized extracavity SHG and angle tuned phase matching of the fundamental to obtain a scannable UV output for spectroscopic purposes. The system included multimode and single mode scanning with laser tuning, calibration and data acquisition under computer control. However, single mode scans never exceeded 1 mW in output power and this power was strongly dependent on the tuning angle. Through use of a suitable intracavity nonlinear crystal this output could be increased by one to two orders of magnitude over the same wavelength region. By incorporating temperature tuning into the system the maximum possible efficiency is maintained independent of the laser wavelength.

This dissertation has been divided into three sections. Chapter
II deals with the theory of intracavity second harmonic generation. This includes a discussion of SHG by Gaussian beams as well as an analysis of the complications introduced by intracavity doubling. It also focuses on determining the conditions for maximizing second harmonic power and possible reasons why theoretical limits on second harmonic power are not achieved experimentally. In Chapter III the design of the spectrometer will be discussed. This will include the modification of the ring laser, nonlinear crystal performance and ring laser performance. The final chapter (IV) reviews the software aspect of the system design including the scanning techniques as well as a discussion of the problems associated with computerized intracavity SHG. Chapter IV concludes with a presentation of the system capabilities through UV absorption spectra of varying resolution obtained with the spectrometer.

In summary, the spectrometer developed at Rice over the past few years has been modified to increase by more than an order of magnitude the UV output powers. This has included the redesign of the ring laser for optimal intracavity SHG and the development of the hardware and software necessary to temperature phase match the second harmonic and fundamental over the wavelength range of the dye. The spectrometer has been used to obtain simultaneous spectra of the fluorescence of I$_2$ in the visible and the absorption of SO$_2$ in the ultraviolet. This simultaneous UV and visible output has proven to be a useful addition to the spectrometer
system, which may have further applications in spectroscopy. The spectrometer has been utilized to obtain continuous UV scans of up to 1600 GHz at 300 nm with a short term linewidth of ± 50 MHz and a constant output power of 10 mW.
CHAPTER II
THEORY OF INTRACAVITY SECOND HARMONIC GENERATION

In this chapter the theory of intracavity second harmonic generation will be reviewed. The first part of the chapter will deal with the work of Boyd and Kleinman [10] with special attention given to non-critical (90°) phase matching. This review will be aimed at providing actual computations of the theoretical limits on second harmonic power as well as the consequences of deviations from optimal conditions. The second section of the chapter will examine the intracavity aspect of SHG. The theoretical maximum second harmonic power will be presented as well as the determination of optimal second harmonic output coupling. The analysis will include the effects of crystal losses and thermal absorption on second harmonic conversion efficiency and laser output. The results analyzed in this chapter will then be used in Chapter III to optimize the design of the spectrometer and also to analyze its final performance.

11.1 Second Harmonic Generation by Plane Waves

The theory of nonlinear optical mixing is well developed and excellent discussions are available in a number of sources [11-13]. This section will present the results which are pertinent to SHG in KDP isomorphs such as ammonium dihydrogen arsenate (ADA). Sum and difference frequency mixing will not be explicit-
ly discussed although the former is frequently present in actual SHG experiments due to multimode laser operation.

A nonlinear medium is characterized by a polarization:

\[ P_i = \varepsilon \chi_{ij} E_j + d_{ijk} E_j E_k + \ldots \]

where \( \chi_{ij} \) is the usual susceptibility tensor, \( \varepsilon_0 \) the vacuum permittivity and \( d_{ijk} \) the first nonlinear susceptibility constant (which is also frequently defined by \( \varepsilon_0 d_{ijk} \)). The indices \( ijk \) refer to an arbitrary set of coordinate axes in the crystal, and, for simplicity, will be assumed to be along the crystallographic axes. For KDP isomorphs, which are negative uniaxial crystals \( n_e < n_o \) with \( \bar{4}2m \) symmetry, the nonlinear coefficient can be expressed as

\[
\begin{bmatrix}
P_x \\
P_y \\
P_z
\end{bmatrix} =
\begin{bmatrix}
0 & 0 & 0 & d_{14} & 0 & 0 \\
0 & 0 & 0 & 0 & d_{14} & 0 \\
0 & 0 & 0 & 0 & 0 & d_{36}
\end{bmatrix}
\begin{bmatrix}
E_x^2 \\
E_y^2 \\
E_z^2 \\
2E_x E_y \\
2E_x E_z \\
2E_y E_z
\end{bmatrix}
\]

In the visible region of the spectrum KDP isomorphs are essentially lossless so that the Kleinman conjecture [14] is applicable and \( d_{36} = d_{14} \).

We will consider the interaction of a focused Gaussian beam in a nonlinear crystal as shown in Figure 2.1. The angle \( \rho \) in this figure is the usual double refraction angle for the second harmonic.
Relationship of focused beam to the nonlinear crystal
given by
\[ \tan p = 2(n_0^1)^2 \left( \frac{1}{(n_e^2)^2} - \frac{1}{(n_o^2)^2} \right) \sin 2\theta \]  \hspace{1cm} 2.2

where \( n_0 \) is the ordinary index of refraction at the fundamental wavelength, \( n_0^2 \) and \( n_e^2 \) the ordinary and extraordinary indices of refraction at the second harmonic wavelength and \( \theta \) the angle between the optic axis and the fundamental direction of propagation. The TEM\(_{00}\) Gaussian mode is characterized by an electric field distribution
\[ E(x,y,z) = \frac{E_o}{1 + i\tau} \exp[ i k z - \frac{x^2 + y^2}{\omega_o^2 (1 + i\tau)} ] \]  \hspace{1cm} 2.3

where \( \tau = (z - f)/b \); normalized \( z \) coordinate
\( f \) = focal position
\( b = k \omega_o^2 n \); confocal distance between the points where
the beam radius \((1/e)\) increases by \( \sqrt{2} \) and
\( k \) is the propagation constant
\( \omega_o \) = beam radius at the focus \( f \)
\( E_o \) = field amplitude constant

The far field half angle of divergence is defined by
\[ \delta_o = \lambda/\pi \omega_o n \]  \hspace{1cm} 2.4

The paper of Boyd and Kleinman analyzed here \[10\] is primarily concerned with the interaction of focused Gaussian beams in the nonlinear medium. However, considerable insight into SHG can be obtained from a plane wave approximation for which \( \tau \) goes to zero over the region of interest. The resulting equation for \( E(x,y,z) \)
is of plane wave nature (infinite radius of curvature) with a Gaussian radial field distribution. For this simplified configuration it is straightforward to show from Maxwell's equations (assuming no crystal absorption or depletion of the fundamental by the second harmonic) that the second harmonic power $P_2$ as a function of the fundamental power $P_1$ is given by [15]

$$P_2 = \frac{2}{\pi} \frac{\mu_0^2}{\varepsilon_0^2} \frac{\mu_0^2}{\varepsilon_1^2} \frac{2}{\omega_1^2} \frac{1}{d_{36}^2} \frac{1}{\mu_0^2} \frac{L^2}{\pi} \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2}$$

where $\Delta k$ is the phase mismatch parameter defined by

$$\Delta k = 2k_1 - k_2$$

and $L$ is the crystal length, $\omega_1$ the fundamental frequency and $k_2$ the second harmonic propagation constant. The coefficient $d_{36}^1$ is related to the nonlinear coefficient by

$$d_{36}^1 = d_{36} \sin \theta \sin 2\varphi$$

where $\varphi$ is the angle between the projection of the fundamental electric field in the $ij$ plane and the $i$ axis. Hence, the coefficient is maximized by $\varphi = 45^\circ$ and $\theta = 90^\circ$ so that propagation is in the $[110]$ direction. It is also clear that the second harmonic is maximized by $\Delta k = 0$, or $2k_1 = k_2$. Since $\omega_2 = 2\omega_1$ and $k = \omega n/c$ it follows that

$$\Delta k = \frac{2\omega_1}{c} (n_1 - n_2)$$

where $n_1$ and $n_2$ are the effective indices of refraction at the two wavelengths. The crystal length which maximizes equation 2.5 is referred to as the coherence length.
\[ L_c = \pi/\Delta k \]  \hspace{1cm} 2.9

or equivalently

\[ L_c = \pi c/\omega_2 (n_1 - n_2) \]  \hspace{1cm} 2.10

In physical terms, the coherence length corresponds to the maximum crystal length which is useful in producing second harmonic power. If the length is exceeded the second harmonic produced at \( L < L_c \) will be reconverted to the fundamental at \( L > L_c \) and overall second harmonic output will be reduced.

From geometrical considerations it is straightforward to show that the index of refraction for the second harmonic is given by

\[ \frac{1}{n_2^2} = \frac{\cos^2 \theta}{n_0^2} + \frac{\sin^2 \theta}{n_e^2} \]  \hspace{1cm} 2.11

For the phase matching case \( n_2 = n_1 = n_0 \) and it follows that

\[ \sin^2 \theta = \frac{(n_e^2/n_0^2)^2 [n_0^2/2 - n_1^2]}{(n_0^2)^2 [n_0^2/n_1^2 - n_2^2]} \]  \hspace{1cm} 2.12

This relationship clearly demonstrates the effect of angle tuning of the crystal on the second harmonic power. If this equation is satisfied, the coherence length is infinite and the second harmonic intensity increases as the square of both the fundamental intensity \( \sim P_1^2/\omega_0^4 \) and the crystal length. This equation also justifies the polarization of the fundamental in the \( ij \) (or \( xy \)) plane since in this orientation the second harmonic is produced along the optic axis and the phase matching condition can be achieved for SHG.
The phase matching condition can also be satisfied by temperature tuning the crystal if
\[ \frac{d}{dT}(n_1 - n_2) \neq 0 \]  
This method will be of particular importance for intracavity SHG where angle tuning would be prohibitively complex. Finally, we note that a simple variation of wavelength will also change \( n_1 - n_2 \). Hence, there are three possible methods of disrupting the optimal phase matching condition. This plane wave approximation is useful for a general understanding of SHG but is of little use for the interaction of focused Gaussian beams in the nonlinear medium. Hence, we must turn to the more complete analysis of Boyd and Kleinman.

11.2 Second Harmonic Generation by Focused Gaussian Beams

The second harmonic field produced by the unapproximated distribution of equation 2.3 at each point in the crystal can be determined by utilizing equation 2.1. By summing the contribution to the second harmonic field outside of the crystal from each point in the crystal and taking into account double refraction the second harmonic power can be shown to be (in mks units)
\[ P_2 = KL P_1^2 e^{-\alpha L} h(\sigma, \beta, \xi, \mu) \]  
where
\[ K = \frac{k_1}{2\pi} \frac{1}{2} \frac{1}{n_1 n_2} \frac{1}{(\varepsilon_0)^{3/2}} \frac{3/2}{d^{12}} \frac{1/2}{\omega} \approx \frac{16\pi^2 c^3}{\mu_0 d^{36}} \]
And the following set of definitions have been made

\[ \alpha = \alpha_1 + \frac{3}{2} \alpha_2, \quad \alpha' = \alpha_1 - \frac{3}{2} \alpha_2; \quad \text{absorption parameters} \]

\( \alpha_1, \alpha_2 \quad \text{fundamental and second harmonic crystal absorption} \)

\( S_x = \frac{x - p(L - f)}{w_o} \quad \text{scaled "walkoff" x coordinate} \)

\( \sigma = \frac{3}{2} \Delta \kappa \quad \text{phase mismatch} \)

\( \sigma' = \sigma + 4 \beta S_x \quad \text{phase mismatch and walkoff} \)

\( \xi = L/b \quad \text{focusing parameter} \)

\( \mu = \frac{(L - 2f)}{L} \quad \text{normalized focal position} \)

\( \kappa = \frac{3}{2} \Delta \) \quad \text{scaled absorption parameter} \)

The factor \( h \) is a numerical integral which contains the optimizable parameters of the Gaussian interaction. A numerical evaluation of the integral has been performed and graphically shown by Boyd and Kleinman for varying values of the parameters above. The graphs can be utilized in performing numerical evaluations on the relative effects of the variation of these optimizable parameters.

Evaluation of the \( h \) integral gives a maximum value of \( h = 1.028 \) for \( \mu = \alpha_1 = \alpha_2 = p = 0, \xi = 2.84 \) and \( \sigma = .46 \). Since \( \xi = L/b \) optimal conversion efficiency is achieved for intermediate focusing in the crystal. For weak focusing the crystal length is much shorter than the confocal distance (\( \xi \ll 1 \)) and
the previously derived plane wave result can be obtained from $h$. The case of strong focusing is discussed below. The maximum value for $h$ stated above is obtained by assuming no crystal absorption. If losses are included, $h$ is obviously decreased and the focus is shifted away from the crystal center (i.e., $u = 0$) in a direction depending on whether fundamental or second harmonic absorption dominates.

The integral value of $h$ depends strongly on the walkoff angle $\theta$ and hence $\theta$. As $\theta$ is moved away from $90^\circ$ the conversion efficiency decreases not only due to the decrease in $d_{36}'$, but also because of walkoff of the second harmonic with the resulting phase incoherence. This walkoff is referred to as the aperature effect and is characterized by a length

$$L_a = \sqrt{\pi} \frac{w_0}{\rho}$$

When the crystal length exceeds $L_a$ the second harmonic is displaced by an amount exceeding the fundamental beam radius and considerable spatial spreading of the second harmonic output occurs.

The integral $h$ can also be evaluated at different values of the phase mismatch parameter $\sigma$. For weak focusing the sine function of the plane wave interaction results as $\sigma$ is varied. As the focusing parameter $\xi$ is increased the sine function becomes asymmetrical and optimal phase matching occurs for $\Delta k > 0$. This is a direct result of the divergence of the fundamental beam which makes it impossible to satisfy the phase matching condition over the entire beam. The beam can be considered as a diverging set of rays for which only a
small subset at the center can satisfy the relation $2k_1 = k_2$. The relative importance of these optimizable parameters can best be examined by a straightforward numerical analysis. For example consider the variation of $\mu, \xi, \text{ and } \theta$ which permits $h$ to remain within 80\% of its maximum value. For a laser with a fundamental output at 600 nm, $\alpha = 0$ and a crystal length of 1.5 cm, this requires that $\rho < 2.5$ mrad. For ADA with $n_1^o = 1.576$, $n_2^o = 1.634$ and $n_2^e = 1.573$ and using equation 2.2 the restriction on $\theta$ is $88.1^\circ < \theta < 90.0^\circ$. For optimum focusing the variation on $\xi$ for a beam waist of 45 m is $1 < \xi < 10$ or $3.2 < L < 32$ cm. Finally, for positioning of the crystal in the beam $|u| < .6$ or $f \approx \pm .45$ cm from crystal center for a 1.5 cm crystal length. Hence, under optimal focusing conditions second harmonic power is only weakly dependent on focal positioning and beam focusing. However, the power is a strong function of the angle $\theta$ (even when ignoring the dependence in $d_{36}^1$) which is a well known problem of angle tuning near $90^\circ$ phase matching.

For the optimal doubling case we can calculate the second harmonic conversion coefficient defined by $c = \frac{P_2}{P_1^2}$

For ADA with $d_{36} = 4.4 \times 10^{-24}$ mks the coefficient $K = 2.86 \times 10^{-2}$ W$^{-1}$ m$^{-1}$ and assuming a crystal length of 1.5 cm the coefficient is
\[ c = 4.3 \times 10^{-4} \text{ W}^{-1} \]

For a reasonable intracavity power of 20 W as much as 170 mW of intracavity second harmonic output could be produced at 300 nm.

In summary, there are three interrelated phenomena which are primarily responsible for reducing second harmonic conversion efficiency. The first of these is the focusing of the fundamental and it is optimized through careful design of the doubling system. The second effect is the aperture effect and it is corrected through noncritical (\( \theta = 90^\circ \)) temperature tuning of a properly chosen nonlinear crystal. The final effect is the phase matching itself and its optimization will be discussed in detail in later chapters.

11.3 Intracavity Aspects of Second Harmonic Generation

The previous analysis is valid for intracavity SHG since this primarily entails only higher fundamental powers. It is precisely this power enhancement which makes intracavity doubling so attractive as a method of providing cw laser radiation below 400 nm. However, the maximum obtainable second harmonic power is limited by the laser design and ultimately by the pumping power. This section will examine intracavity SHG to determine both the maximum second harmonic power which can be achieved and the reasons why, in practice, this theoretical limit will not generally be obtained.

Smith [16] has examined intracavity SHG with respect to a linear
Nd:YAG laser to show that the theoretical second harmonic output power is equal to the power available at the fundamental for optimal output coupling. The analysis for a homogeneously broadened dye ring laser is similar to Smith's analysis and will be presented here. The insertion of the nonlinear crystal into the cavity can be accounted for by introducing phenomenologically a power reflection coefficient defined by

\[ R' = R + cS \]

where \( c \) is a second harmonic conversion coefficient (in units of area/power), \( R \) the round trip reflection coefficient (which is lost to the ring laser) and \( S \) the intracavity power density in the laser. The round trip gain coefficient is given by

\[ g = g_0 / (1 + \frac{S}{S_0}) \]

where \( g_0 \) is the small signal unsaturated gain and \( S_0 \) the saturation parameter. For simplicity no fundamental output coupling will be assumed (which can simply be added to the linear loss under the assumption below). For the small signal gain typical of a dye laser the gain can be equated to the cavity loss to find the optimal second harmonic power density given by

\[ S_2 = cS^2 = g_0 S_0 (1 - \sqrt{\frac{L}{g_0}})^2 \]

where \( L \) is the total round trip linear loss. This equation for the ring laser is identical to the well known equation for the optimal fundamental output power of the linear laser. In both cases the
cavity losses must be minimized if high second harmonic powers are to be achieved.

The optimal value of the conversion coefficient is given by
\[ c = \frac{\ell}{S_0} \]
which is surprisingly independent of both the laser gain and the fundamental power. This optimal coupling coefficient is equal to half the optimal coefficient of the linear laser. Equation 2.18 demonstrates that optimal coupling can be achieved at all power levels for constant linear loss. However, in practice, it is usually not possible to increase \( c \) or decrease \( \ell \) to provide optimal coupling. If \( c \) is less than the optimal value second harmonic power will be less than the maximum possible power. It is interesting to note that the opposite case of overcoupling is of little practical importance in actual SHG. If \( c \) is too large it is easily reduced by adjustment of the phase mismatch and second harmonic power will be optimized at two values of the phase matching parameter \( \sigma \).

This simple but illustrative model fails to account for many of the complications of intracavity SHG. First, the model does not explicitly take into account the cross sectional variation of the fundamental and second harmonic. Variations of fundamental intensity can only be accounted for through an adjustment of the constants. Second, and more important, the model does not include the effects of crystal absorption. Since in this case both \( c \) and \( \ell \) are dependent on the crystal length, the optimal coupling condition above is no longer
valid. Crystal absorption also leads to thermal effects resulting from a radial temperature gradient in the crystal. This gradient disrupts the phase matching condition and thermally defocuses the fundamental because of the temperature dependence of the crystal indices of refraction.

The most complete analysis of intracavity SHG has been presented by Ferguson and Dunn [5]. They have postulated that the second harmonic power is given by

\[ P_2 = K h h L P_1^2 \]  

where \( K, h \) and \( L \) have been already defined and \( h_T \) determines the decrease in \( P_2 \) due to thermal effects. This equation does not take into account lensing effects in the crystal resulting from the radial temperature gradient. However, since \( \frac{dn}{dT} \) is less than zero in KDP isomorphs, the refractive index increases away from the beam axis and a slight diverging effect would occur. The magnitude of the change in second harmonic power resulting from this effect would be difficult to determine in an actual laser system. However, at high powers, it certainly contributes to the overall decrease in the laser efficiency.

The lowering of the conversion efficiency by \( h_T \) is of primary importance at high fundamental intensities. Okada and Ieiri [17] have approximated the temperature profile of a weakly focused Gaussian beam as

\[ T(r) = P_1 \frac{5}{2\pi} \exp\left(-r^2/\omega_0^2\right) \]
where $\kappa$ is the crystal thermal conductivity and $\delta$ is the absorption coefficient. As a first approximation we can Taylor expand $\Delta k$ and assume a Gaussian plane wave for the fundamental to calculate the expression for $h_T$ as

$$h_T = \frac{P_2}{P_{\text{opt}}} = \frac{8}{\theta} \left[ \mathcal{B} S_1(\theta) - (1 - \cos \theta) - 3(\gamma - C_1(\theta) + \ln(\theta)) + 3(\theta - \sin \theta)\theta \right]^{2.21} - \left( \frac{\theta}{\beta} - \sin \beta \right) / \beta - (1 - \cos \beta) / \beta^2$$

where

$$\theta = \frac{2P_1}{P_T}$$

$$S_1(\theta) = \int_0^\theta \left( \frac{\sin x}{x} \right) dx$$

$$C_1(\theta) = \int_0^{\infty} \left( \frac{\cos x}{x} \right) dx$$

$$\gamma = \text{Euler's constant} = .577$$

$$P_T = \frac{\lambda K}{5L} \left[ \left| \frac{dn_2}{dT} - \frac{dn_1}{dT} \right| \right]^{-1}$$

The parameter $P_T$ determines the strength of $h_T$ in reducing the second harmonic power. For $P_1 \ll P_T$, $h_T \rightarrow 1$ and the second harmonic is dependent on the square of the fundamental power. However, if $P_1 \gg P_T$, then $h_T \rightarrow \pi/\theta$ and $P_2$ becomes linear with intracavity power. For ADA with $\left| \frac{dn_2}{dT} - \frac{dn_1}{dT} \right| = 4 \times 10^{-5} \deg^{-1}$, $\kappa = .02 \text{W/cm}$, $\lambda = 600 \text{ nm}$, $\delta = .005 \text{ cm}^{-1}$ and $L = 1.5 \text{ cm}$

$$P_T = 4 \text{ W}$$

This analysis ignores the absorption of the second harmonic with its contribution to the temperature gradient. In general, the second
harmonic is one to two orders of magnitude greater than the fundamental. However, since the second harmonic power is typically two to three orders of magnitude less than the fundamental power, second harmonic absorption will have a smaller effect.

The most complete analysis of a high power cw dye laser has been performed by Teschke, Diennes and Whinnery [18]. Their theory has included triplet effects as well as excited state absorption of the pump and laser radiation by the dye. The intracavity power can be shown to be approximately equal to

\[ P_1 = \eta (P - P_{th}) \]

where \( \eta \) is the slope efficiency, \( P \) is the pump power and \( P_{th} \) is the threshold pump power. Both \( \eta \) and \( P_{th} \) have been determined in terms of the dye and laser parameters. This expression permits the determination of second harmonic conversion efficiencies and optimal crystal lengths as a function of pump power. The conversion efficiency \( E \) and nonlinear coupling \( T_2 \) are defined by

\[ E = \frac{P_2}{P} \quad T_2 = \frac{P_2}{P_1} = K_{hh}LP_1 \]

Hence, \( T_2 \) is equivalent to the term \( cS \) discussed previously. For long crystals or high pump powers we find that

\[ T_2 = \frac{\eta}{2} K_{hh} L P_T \]

and \( T_2 \) can be expected to be functionally equivalent to a fundamental output mirror with the same constant transmission.
This set of equations has been evaluated as a function of pump power, cavity loss and crystal absorption to determine the optimal crystal length and conversion efficiency. However, both \( \eta \) and \( P_{\text{th}} \) are dependent on \( T_2 \) and an iterative numerical analysis must be performed. The basic results (depicted graphically by Ferguson and Dunn) will be highlighted in the following paragraphs.

As is obvious from a purely qualitative argument, there will always exist an optimal crystal length which will give the maximum second harmonic power. When excited state absorption by the dye and thermal effects are neglected, this length is usually dominated by the crystal absorption (\( L \sim L_0/\delta \), where \( L_0 \) is the laser linear loss excluding absorption). When both of these effects are included, the optimal length and conversion efficiency decrease considerably with the thermal effect dominating the dependence. The optimal crystal length is increased at higher values of the laser linear loss, and decreased by higher crystal absorption. The first situation results in lower intracavity powers such that thermal effects become important at higher pump powers. However, higher crystal absorption predominately results in a stronger thermal effect and optimal crystal length must then decrease. In either case, once thermal effects become important, the optimal length decreases as the pump power increases, reflecting the shorter coherence length created through increasing power absorption.
The conversion efficiency (as well as the optimal crystal length) increases strongly with the pump power until thermal effects become important and \( T_2 \) becomes independent of fundamental power. At this point, \( E \) becomes essentially constant with pump power, but decreases with increasing crystal length because of the increase in \( h^T \). As is expected, the efficiency decreases with increasing crystal absorption and linear loss.

As an illustration, consider a typical Rh6G dye laser with an intracavity ADA crystal. For a linear loss of 5\% , crystal absorption of \( 0.005 \text{ cm}^{-1} \) and pump power of 5 W, the optimal crystal length is 1.5 cm with a corresponding efficiency of 2\%. If crystal absorption is increased to \( 0.01 \text{ cm}^{-1} \), the optimal length and conversion efficiency decrease to 1 cm and 1\%, respectively. However, if thermal effects are neglected, \( L \) and \( E \) increase to about 7.5 cm and 18\%, respectively, for \( \delta = 0.005 \). This represents a 9 fold increase in second harmonic power over the previous case. At 10 W of pump power and using the first case above, the optimal length and \( E \) are essentially unchanged from the 5 W case, which gives a second harmonic power of 200 mW. If \( L \) is now increased to 7 cm, the second harmonic power drops to 160 mW (or a 20\% decrease). On the other hand, if \( L \) is decreased to 0.5 cm, a similar 20\% drop in second harmonic power occurs. Thus, at this level of crystal absorption, the penalty is not severe for vastly exceeding the optimal length. However, as absorption is increased, \( h^T \) becomes increasingly impor-
tant, the total linear loss of the laser is significantly increased, and
the penalty will become increasingly severe.

The preceding discussion is valid for a linear Rh6G dye laser. For
the case of the ring laser some modifications must be performed. The
thermal effects for the linear and ring laser are the same for a given
pump power since intracavity power is identical in the two systems.
However, the ring laser operates at twice the intracavity one way power
of the linear laser. This does not vary the maximum obtainable second
harmonic power since the power available at the fundamental is the same
for both types of laser. However, it does increase by a factor of two
the second harmonic power produced in a cavity round trip. This then
requires a decrease by a factor of two in the optimal coupling coeffi‐
cient $T_2$ if output is to remain unchanged. It also increases laser
efficiency by a factor of two since Ferguson and Dunn assume the second
harmonic produced in one direction of the laser is lost. As a first
approximation these changes can be incorporated into the model of Fer‐
guson and Dunn by simply doubling the slope efficiency of equation 2.22
and recalculating utilizing $\eta$ and $P_{th}$ of Pike [19]. The net result
would be to decrease the optimal crystal length by more than a factor
of two and an increase in the laser efficiency $E$ by a similar amount.
The factors would result from both a reduced thermal effect and a more
efficient output coupling scheme at the optimal output power. These
modifications will have to be considered in the design and performance
of the doubling system in the next chapter.
CHAPTER III

SYSTEM DESIGN

In this chapter the design of the intracavity ultraviolet spectrometer will be reviewed. As previously stated, this spectrometer is a modified version of the spectrometer developed by Pollock [20] which included: 1) a high power Ar\(^+\) pump laser, 2) a cw dye ring laser, 3) an extracavity frequency doubling system, 4) diagnostic equipment for determining laser output power and frequency, 5) a PDP 11/03 minicomputer with a CAMAC interface, and 6) an extensive software package for frequency calibration of the spectrometer, and acquisition, storage and display of data. The modifications which have been made to the spectrometer include redesign of the ring laser, replacement of extracavity angle tuning with intracavity temperature tuning, and the development of the necessary hardware for computerized scanning of the second harmonic radiation. The development of the computer software will be discussed in the final chapter.

III.1 Ring Laser Design

The first step in the development of the spectrometer was the redesign of the ring laser described in reference [20]. The ring configuration was preferred for intracavity doubling because of its numerous advantages over the linear laser. First, the ring laser is capable of higher fundamental single mode output powers.
which leads to higher second harmonic powers. This higher power results from the travelling wave capability of the ring laser which requires fewer tuning elements to obtain a stable single mode output. This simplifies laser tuning as well as lowers the laser linear loss. Second, a unidirectional travelling wave laser produces the second harmonic in one direction only. This simplifies output coupling of the second harmonic and increases overall laser efficiency since no output coupling scheme is ideal. Furthermore, a lowered efficiency leads to an earlier onset of thermal effects for the same second harmonic laser output. Third, and most important, the one way power of the travelling wave ring laser is increased by a factor of two over the linear laser, which increases the second harmonic power by up to a factor of four. The doubling of the one way power also means that optimal coupling is achieved at half the conversion coefficient of the linear laser which is important for the reasons previously discussed.

The LASL-Rice ring laser needed to be modified to provide an optimally focussed beam for intracavity doubling and output coupling of the second harmonic. The most effective method for accomplishing this task is to replace one of the full reflectors (M4 or M5 in the first version) with a dichroic mirror (100% reflective to the fundamental, 100% transmittive to the second harmonic) of the properly chosen radius of curvature as shown in Figure 3.1. This technique has several advantages over other more complex output coupling schemes. First, and most important, for an ideally coated mirror this method adds no additional
losses to the laser cavity. Second, the mirror separates the second harmonic from the fundamental output which considerably simplifies experimentation and diagnostics. Finally, the mirror permits the variation of the fundamental output coupling to provide maximum second harmonic and fundamental output powers. The only minor problems with the technique turned out to be imperfect coatings (T = 0.14% at 600 nm, and T = 83% at 300 nm) and the divergence of the second harmonic output which requires recollimating with a f = 15 cm lens.

The radius of curvature of the dichroic mirror was chosen through a consideration of commercially available mirrors and optimal beam focusing in the dye jet and nonlinear crystal. The cavity also needed to be sufficiently large to provide space for the tuning elements in the long laser arm, and the crystal oven in the short arm preceding the dichroic mirror. However, an overly large cavity would make single mode operation more difficult to achieve due to the small cavity mode spacings (c/P, P = laser light path length). The beam radius in the tuning arm of the cavity was not explicitly used as a specification in the laser design. However, this beam should be as large as possible since thermal distortions and beam walkoff in the tuning elements increase with decreasing beam radius [21].
The analytic method for the determination of stable cavity dimensions has been developed by Kogelnik and Li [22]. The stability condition evolves from the propagation of Gaussian beams and the requirement that a beam reproduce itself after one cavity round trip. The complex beam parameter for the Gaussian TEM$_{00}$ mode is

$$1/q(z) = 1/R(z) - i\lambda/m\omega(z)$$ \hspace{1cm} (3.1)

where $R(z)$ and $\omega(z)$ are the radius of curvature and beam diameter (1/e) defined by

$$R(z) = z(1 + z^2/\omega_0^2)$$ \hspace{1cm} (3.2)

$$\omega(z)^2 = \omega_0^2(1 + z^2/\omega_0^2)$$

where $\omega_0 = m\omega_0 n/\lambda$ and $\omega_0$ is a constant.

Utilizing the ABCD law, (where $A,B,C,D$ are elements of the matrix equal to the product of all of the element ray matrices in the laser) the stability condition for a cavity round trip is

$$1/q_2 = 1/q_1 = \frac{Aq_1 + B}{Cq_1 + D}$$ \hspace{1cm} (3.3)

to give as the condition for a confined beam

$$|D + A| \leq 2$$ \hspace{1cm} (3.4)

The radius of curvature and spot size at the arbitrary position $q_1$ are given by

$$R = \frac{2B}{D - A}$$ \hspace{1cm} (3.5)

$$\omega = (\lambda/mn)^{1/2} |B|^{1/2}/[1 - ((D + A)^2/4)]^{1/4}$$

The ABCD law can then be used to find $R$ and $\omega$ at any other position in the cavity.
The modified ring laser has a minimum of six elements (three curved mirrors and three spaces) so that the ABCD matrix involves the product of six matrices. Because of this large number of elements and the number of mirror combinations which needed to be considered, a Fortran IV program was developed to determine stable cavity dimensions and the necessary beam parameters. We considered all possible combinations of 2.5 cm, 5 cm, and 15 cm focal lengths for M2, M3, and M5 as shown in Figure 3.2. The first criteria in choosing the optimal set of mirrors and distances was that the stability condition of equation 3.4 be satisfied. However, the stability range should be as large as possible if the design is to be usable, and fundamental and second harmonic laser efficiency is to be easily optimizable. Specifically, variations in mirror separation should have as small an effect as possible on laser stability, focal positioning and beam radius in the tightly focused arms of the cavity. Only the combination shown in the figure met this design criteria as well as those to be discussed in detail in the following paragraphs.

Figure 3.2 shows the cavity beam parameters and dimensions which are used in the spectrometer. Using the optimal beam focusing condition derived by Boyd and Kleinman [10] the 45 um crystal beam radius corresponds to an optimal crystal length of

\[ L = 2.8nk_0w^2 = 9.5 \text{ cm} \]

at 600 nm. The crystal lengths under consideration were in the 1.5 cm to 3 cm range. For a 1.5 cm crystal, the optimal value of the h
DYE JET BEAM WAIST
R = 12 μm

THIRD BEAM WAIST
R = 380 μm

CRYSTAL BEAM WAIST
R = 45 μm

M2
R = 10 cm

M3
R = 5 cm

M4
R = ∞

M5
R = 30 cm

26 cm

41 cm

16.5 cm

48 cm

θ = 9°

80 cm

AXIAL MODE SPACING, \( \frac{c}{L} \approx 167 \text{ MHz} \)

Ring Laser Dimensions and Beam Parameters

FIG. 3.2
integral is reduced by about 50% for this waist size. However, the optimal value of $w_o = 18 \mu m$ for a 1.5 cm crystal was unobtainable without increasing the dye jet beam radius or leaving insufficient room for a crystal oven. An increase in the dye jet beam radius would tend to lower the overall laser efficiency.

The present beam radius at the jet is about 12 \mu m, which is greater than the optimal value. The pump beam is focused by the $f = 3.75$ cm mirror to a beam radius of about 6 \mu m at the dye jet. Pike [19] has developed expressions for the change in slope efficiency and laser threshold as a function of

$$u = \frac{w_L^2}{w_p^2}$$

where $w_L$ and $w_p$ are the beam radii of the laser and pump, respectively, at the jet. Pike states that the threshold power is proportional to $(1 + u)/2$ while the slope efficiency is proportional to $u/(1 + u^2)^{2/3}$. For the modified spectrometer, $u$ is equal to approximately four, which results in a high laser threshold but also a high slope efficiency. The optimal value of $u$ can be calculated for a given pump power and threshold by maximizing the equation

$$P_1 = \frac{u^n}{(1 + u^2)^{2/3}} \left( \frac{p}{p_t} - \frac{1 + u}{2} \right)$$

with respect to $u$ to get

$$P_1 = \frac{u^n}{(1 + u^2)^{2/3}} \left( \frac{p}{p_t} - \frac{1 + u}{2} \right)$$
\[ u^3 + 2u + \left(1 + \frac{P}{P_t} \right) = 0 \]

If we assume that \( P = 0.5 \) W, and \( P_p = 5 \) W, then \( u = 2.4 \) and the reduction in power for \( u = 4 \) is about 5% independent of the slope efficiency. For the laser designed in reference [20], \( u = 1.8 \) and the laser output was about 98% of the maximum value under the same conditions. Hence, this enlargement of the laser beam size in the jet is not critical under the experimental high power conditions.

Of considerable importance in the design of the ring was the need to provide sufficient space for the crystal oven. A Brewster cut crystal must be tilted at a 25° angle with respect to the incident beam if the beam is to propagate down the crystal axis. This requires that the oven also be tilted and sufficient separation of the beams in the figure eight cavity is necessary. The separation of the beams at the focus is controlled by the beam angle of incidence on the mirror and the position of the crystal waist. The beam waist to mirror separation is the primary reason for the use of the \( f = 15 \) cm mirror. The angle of incidence on the mirror is determined by the need to compensate cavity astigmatism and optimize laser stability.

It is well known that mirrors used off axis as well as Brewster angled plates produce astigmatic distortions in Gaussian beams. This astigmatism can be compensated according to a method similar to that developed by Kogelnik [23]. A mirror used off axis (at an angle \( \theta \) with respect to the incident beam) focuses the sagittal and
tangential wave bundles with different effective focal lengths which are given by

\[
\begin{align*}
    f_s &= f / \cos \theta \\
    f_t &= f \cos \theta
\end{align*}
\]

for the sagittal and tangential planes respectively. Similarly, the effective path length in a Brewster plate of refraction index \( n \) is different for the two planes and is given by

\[
\begin{align*}
    d_s &= t (n^2 + 1)^{\frac{1}{2}} / n^2 \\
    d_t &= d / n^2
\end{align*}
\]

where \( t \) is the plate thickness. To compensate the cavity the astigmatism due to the off axis mirrors and Brewster plates must be equated to maximize laser stability. For the previous Rice design (or the linear laser of Kogelnik), the off axis mirrors focus the beam from a virtual infinity down to the mirror's focal length. For this case the sagittal and tangential mirror separation \( z \) which provides a stable cavity is given as

\[
\begin{align*}
    z_s &= \sum f_s i_s + \delta_s \\
    z_t &= \sum f_t i_t + \delta_t
\end{align*}
\]

where the summation is taken over the off axis cavity mirrors of the laser arm containing the Brewster plate. The additive constant \( \delta \) represents the variation in mirror separation over which the cavity remains stable. For the tight focusing case, \( \delta \) is essentially constant and equal for the two planes of incidence, and the cavity can be compensated.
by determining the angle $\theta$ such that

$$z_s - z_t = d_s - d_t$$

When this equation is satisfied both planes of incidence will remain stable over exactly the same range of mirror separation (excluding the differential length in the sagittal plane due to the Brewster plate), and laser stability will be optimized.

As is obvious from the dimensions of Figure 3.2, the three mirror cavity does not focus the beam to the focal length of the mirrors. As a result the stable mirror separation and compensation angle are a more complicated function of focal length than given in the previous equations. As a first approximation, the analysis first given in [23] and described above can be extended to the three mirror cavity. This would involve a determination of the mirror separation in one cavity arm as based on the position of the focal plane in the remaining arms of the cavity. This type of analysis ignores the strong coupling between all three mirrors in the cavity and, for proper compensation, an exact solution should be utilized.

As previously detailed, a fully compensated cavity is one in which the cavity is equally stable in the sagittal and tangential planes for a given mirror separation. Using equation 3.4, the center of the laser stability range can be determined from the cavity ABCD matrix as the mirror separation where

$$A + D = 0$$

A complete analysis of all the laser beam parameters throughout
the stability region shows that at the center of the stability range, beam waists at the foci in the dye jet and nonlinear crystal have a maximum value and hence minimum slope. Equation 3.12 eliminates the need to directly consider changes in the stability limits (e.g. $\delta$), which result from astigmatic variations in focal length and mirror separation.

To determine the mirror separation in any one arm of the ring cavity which satisfies equation 3.12, consider a three mirror cavity with focal lengths $f_1$, $f_2$, and $f_3$, and mirror separations $x, y$, and $z$ where $x$ is the separation between mirrors $f_1$ and $f_2$, $y$ the separation between $f_2$ and $f_3$, and $z$ the separation between $f_3$ and $f_1$. If the ABCD matrix of the cavity is determined analytically using these parameters, and equation 3.12 is utilized to solve for $z$ in the center of the stability region, the solution is

$$z = \frac{2 + (x + y)(A + B + C) + xyB(A + C)}{(A + B + C) + xA(B + C) + yC(A + B) + xyABC} \tag{3.13}$$

where $A = -1/f_1$, $B = -1/f_2$, and $C = -1/f_3$ (not to be confused with the ABCD parameters of previous equations). This equation is completely general for any laser arm of a three mirror ring cavity provided proper sequencing of the cavity parameters as defined above is maintained (i.e., $f_1 - x - f_2 - y - f_3 - z$). Equation 3.13 can be used to determine the difference in mirror separation at the center of the stability range for the sagittal and tangential planes. If $\gamma = \cos \theta$, then for the sagittal plane the mirror
separation \( z(\gamma) \) is

\[
z(\gamma) = \frac{M + Ny + O\gamma^2}{Py + Q\gamma^2 + R\gamma^3}
\]

where \( M, N, O, P, Q \) and \( R \) are defined by comparison to equation 3.13, and the angle of incidence on each mirror is assumed equal. As in equation 3.11, astigmatic compensation is achieved by equating the difference in mirror separation in the two planes of incidence to the differential path length due to a Brewster plate in the same cavity arm. This requires that

\[
z(\gamma) - z(\theta) = (d_s - d_t)z
\]

in any one laser arm.

Figure 3.3 shows a plot of plate thickness \( t \) as a function of the mirror angle \( \theta \) for each arm of the laser cavity. The figure demonstrates clearly that a given Brewster plate thickness \( t \) has the most significant effect on laser stability in the laser arm with the tightest focusing and smallest stability range. (The stability ranges of the dimensions of Figure 3.2 are approximately \( 3.81 \pm .17 \) cm, \( 42.0 \pm 2.7 \) cm, and \( 128. \pm 96 \) cm.) Equivalently, at any angle \( \theta \), a Brewster plate of thickness \( t \) is least effective in compensating astigmatism in the essentially unfocused laser arm. For the present laser system, with a dye jet thickness of .03 cm, an angle of incidence of 1.92° is required on each cavity mirror to compensate the cavity.

The laser cavity which contains more than one Brewster plate is of substantial interest for the intracavity doubled laser. If the
BREWSTER PLATE THICKNESS $t$, cm

COMPENSATION ANGLE $\theta$, deg

128 cm ARM

42 cm ARM

8.8 cm ARM

SINGLE PLATE COMPENSATION FOR RING CAVITIES

FIG. 3.3
plates are in the same cavity arm, the plate thickness can be summed provided the plates are oriented in the same plane [24]. If the plate is in another cavity arm, then the mirror separation in this arm must be corrected in the sagittal plane by an amount equal to the differential path length. With this added consideration, equation 3.15 can be expressed in its final form as

\[ z(Y, x + (d - d_s t_y), y + (d - d_s t_y)) - z(Y, x, y) = (d - d_s t_y) \] 3.15

As an example, consider a laser with a dye jet thickness of .03 cm and a nonlinear crystal with an optical path length of 1.5 cm configured as shown in Figures 3.1 and 3.2. A crystal with an optical path length \( \xi \) of 1.5 cm has an effective thickness perpendicular to the plane of incidence [23]

\[ t = \frac{n \xi}{\sqrt{n^2 + 1}} \] 3.17

or \( t = 1.27 \) cm for a refractive index \( n \) of 1.58. If \( z \) and \( x \) represent the stable mirror separations of the jet and crystal arm respectively, then equation 3.16 yields a compensation angle of about 3.68°. If the birefringent filter (\( t \approx .8 \) cm) and Faraday rotator (\( t \approx .7 \) cm) are included in this calculation the required angle increases by only a few hundredths of a degree since these elements are contained in the unfocused laser arm. This angle is considerably less than the required compensation angle obtained for very tight focusing as analyzed by Kogelnik and employed in four mirror ring lasers [8]. (The ring laser described in [8] is
actually overcompensated for astigmatism when the exact analysis described above is extended to the four mirror cavity. The overcompensation results from the degree of focusing and the interaction between all laser mirrors in the cavity as well as the changes in the stability regions. These are factors that are not included in the approximate analysis of Kogelnik).

The actual angle used in the laser system was 4.0° to 4.5° as dictated primarily by the space requirements of the crystal oven. A computer analysis of the beam parameters in the compensated and uncompensated laser cavity shows that astigmatism can be expected to decrease both laser and second harmonic efficiencies. This decrease results from the differences in focal positions and beam waists in the sagittal and tangential planes which occur even in a compensated cavity. For example, in the compensated cavity described above, it is impossible to focus both the sagittal and tangential planes at the center of the nonlinear crystal. For the overcompensated cavity, this type of distortion increases along with the decrease in laser stability which results at angles other than the compensation angle. However, both analytical analysis and comparison of the conversion efficiencies of normal incidence (for which the compensation angle should be 1.92°) and Brewster cut crystals indicates that the effect on second harmonic efficiency is minor in comparison with the other considerations discussed previously.
III.2 Laser Tuning

Tuning of the laser spectrometer, as well as single mode operation, was accomplished in precisely the same manner as reported in reference [20]. Insertion of a high quality temperature tuned crystal into the laser cavity was not found to interfere with laser operation. We will briefly discuss the tuning of the laser here as well as the operation of a newly acquired Faraday isolator. The operation of an electro-optic tuner (EOT) in the spectrometer will also be evaluated as a possible replacement for the birefringent filter.

Traveling wave operation of the ring laser was obtained through either mirror feedback or a Faraday isolator. In mirror feedback, one of the output beams is retroreflected back into the laser cavity, which sufficiently biases one direction of oscillation to produce quasi-unidirectional operation. However, mirror feedback tends to produce a less stable single mode (a true travelling wave is not produced) and also enhances laser noise. In multimode operation, mirror feedback is still preferred because of the high insertion loss of the isolator used in the spectrometer.

A Faraday isolator (or "optical diode") consists of a reciprocal and a non-reciprocal polarization rotator which discriminates between the two possible directions of laser oscillation. For only one direction of oscillation will the rotations add (for the other they cancel) and this direction will then suffer greater losses at the laser Brewster surfaces. Johnston and Proffitt [25] have determined
that a differential loss between directions of oscillation of only 0.4\% is necessary to obtain stable single mode travelling wave operation. By assuming that the component of the rotated field perpendicular to the laser polarization is lost, this requires a net rotation of only 3.6°.

A two element isolator is used in the spectrometer which consists of an optically active c-axis quartz plate and a Faraday effect rotator of Hoya FR-5 glass. The 0.5 mm thick quartz plate provides a rotation proportional to its thickness (20°/mm at 633 nm) and inversely proportional to the square of the wavelength. The rotation of the Faraday rod is given by

$$\theta = VHL$$

where $V$ is the Verdet constant (4.3°/kG cm), $H$ the magnetic field, and $L$ the crystal length. Two samarian-cobalt magnets are used to provide a field of about 3 kG in the crystal to produce a rotation of about 11° at 633 nm. Hoya FR-5 glass is widely used as a Faraday material because of its high Verdet constant, but other suitable materials are available (such as SF-2 glass [25]) which have higher ratios of rotation to absorption. The Hoya glass was Brewster cut to increase the losses of the rotated wave and to minimize etalon effects as well as AR coating losses which were noticeable in the previous version of the spectrometer.

The Faraday isolator discussed above gives a net rotation in excess of 20° which is considerably greater than actually re-
quired. This large a rotation is not desirable because of the high absorption (0.03 cm\(^{-1}\) for FR-5 glass) associated with materials with substantial Verdet constants. However, small rotations are difficult to achieve because of the difficulty in fabricating thin c-axis quartz planes. Jarret and Young [26] use a Faraday isolator similar to the one discussed above except the element lengths are decreased by a factor of four to give a net rotation of only 5° at 633 nm. Since rotation decreases as the inverse square of the wavelength this rotation is close to the minimum necessary to cover the range of visible dyes. The small rotation also minimizes any net rotation of the preferred direction of oscillation which would increase the insertion loss of the isolator. A 1.8° rotation corresponds to a loss of 0.1% as determined by the approximation of Johnston and Proffitt.

As previously stated, insertion of a nonlinear crystal into the cavity did not create frequency instability in single mode operation as the crystal was tuned through resonance. This considerably simplified development of the spectrometer since no additional cavity elements were necessary. The crystal at resonance is a highly wavelength selective loss and hence could be expected to produce instabilities such as those reported by Schroder et al. [7]. Nonlinear crystals are also birefringent and might be expected to have a small effect on laser tuning and traveling wave selection when the crystal is not precisely ori-
ented with respect to the incident beam. This interaction would be further complicated by the changes in the indices of refraction due to temperature tuning of the crystal. None of these complications have been found to occur for the high quality Brewster cut crystals. However, for the normal incidence crystal and the poor quality Brewster cut crystal (to be discussed) some frequency instability was evident when changing the crystal temperature by large amounts.

The wavelength selective elements of the laser are the birefringent filter, the Faraday isolator, an air-spaced etalon and, for very high resolution, a PZT translated mirror. As noted previously, the Faraday isolator was used primarily for single mode operation of the laser. In multimode operation the isolator was not convenient because the 200 GHz free spectral range (FSR) of the uncoated c-axis etalon limited the scanning resolution of the spectrometer. Mirror feedback and the three plate birefringent filter provided tunable output with a linewidth of 0.5 Å at 600 nm. Insertion of the Faraday isolator permitted higher resolution scans if the etalon was tilted to scan through its FSR. In this mode of operation the laser oscillated in an unstable single mode with frequency hops of 2-3 GHz during scanning. Tilting of the c-axis plate was not found to introduce any noticeable detrimental effects on the laser operation due to the plate birefringence. An angle of only 1.7° is sufficient to scan through several FSR's at 600 nm. Finally, addition of the air-spaced etalon (FSR = 30 GHz, R = 33%) to the cavity provided stable unidirectional single mode operation with a linewidth
of about 40 MHz.

The performance of two electro-optic tuners were also evaluated as possible substitutions for the birefringent filter in some applications. The EOT has the advantage of very rapid tuning without the mechanical complications of the birefringent filter. The tuners used in the spectrometer (Ithaca Research Corporation models LS-14K and LS-24) are similar in design to that described by Telle and Tang [27]. 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 (or optic) 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 then discriminate against all of the rotated elements. The pass wavelength of the tuner is varied electrically by application of a voltage across the crystal which changes the refractive indices through the linear electro-optic effect. The variation of the pass wavelength \( \lambda \) depends on the angle \( \theta \) and voltage \( V \) according to the relation

\[
\frac{d\lambda}{dV} = \frac{3}{L} \frac{Ln}{w} r_{41}
\]

where \( L \) and \( w \) are the crystal length and width, \( n \) the refractive index, \( r_{41} \) the electro-optic tensor and \( N \) the integer order of the pass wavelength.

According to the tuner specifications, the LS-14 tuner is designed
to yield a large tuning rate (~0.15 nm) when used as the only cavity tuning element. The LS-24 is supposed to be used in conjunction with the LS-14 to provide a narrow linewidth (~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. When using the LS-14 alone, a voltage of 0-4 kV is sufficient to tune through the wavelength range of Rh6G.

The experimental tuning curve of the LS-14 tuner obtained with the spectrometer is shown in Figure 3.4. The tuning rate of 14 nm/kV was found to be angle dependent and very linear in accordance with theory. However, the linewidth of 0.2 nm was greater than specification and the tuner reduced the fundamental output power to 85% of that obtainable with the birefringent filter. (The birefringent filter typically decreased output power by less than 10% at the peak wavelength.) Insertion of the LS-24 into the cavity with the LS-14 reduced the linewidth to 0.025 nm with 40% of the power available with the birefringent filter. However, removal of the LS-14 did not substantially increase the linewidth or increase the laser tuning range which was limited to 570-605 nm by the LS-24. This is in contrast with specifications which indicate that simultaneous operation of the tuners should provide the smallest linewidth. Hence, there is little to be gained by the complications involved in simultaneous scanning of both tuners.
Electro-Optic Tuning Curve

LS-14 TUNING CURVE
To summarize the results for the tuners, they are not a suitable substitute for the birefringent filter for application in the spectrometer. Their assets of rapid tuning and high linearity are more than offset by high insertion losses, large linewidths and increased laser noise. However, we realize that for specialized laser applications requiring rapid tuning and high reproducibility these drawbacks can be acceptable [28].

11.3 Nonlinear Crystals

The choice of nonlinear optical crystal is based entirely on its wavelength tuning properties. Ammonium dihydrogen arsenate (ADA) is the only KDP isomorph which can be noncritically phase matched over nearly the entire range of the organic dye Rh6G (565 to 610 nm). Figure 3.5 shows the phase matching temperature for the wavelength range of ADA as determined by Stickel [28]. Wavelengths beyond 610 nm can not be obtained with ADA because of crystal decomposition at high temperature. However, wavelengths in this range can be obtained through the use of a properly cut ADP crystal (θ = 65°), although the efficiency will be lowered. One problem with KDP isomorphs is that they are extremely fragile, hygroscopic and sensitive to thermal shock. This requires careful design of the crystal oven and a reliable method of temperature control.

The crystal can be obtained with either a Brewster cut or flat end surface for insertion into the laser cavity. Normal incidence
ADA Phase Matching Relationship
crystals require antireflection coatings which must be transmitting to both the fundamental and second harmonic, and can not be applied directly to the crystal surface. For an AR coated window to be effective it must be attached to the crystal face with index matching fluid. However, index matching fluids can be lossy both to the fundamental and second harmonic and, more importantly, can introduce thermal lensing effects at high intensities. For these reasons use of a conventional crystal tuning cell [30], typically utilized in angle tuning, was never considered. The only significant problem with the Brewster cut crystal is the uncompensated second harmonic reflection at the crystal surface of about 17%. However, even AR coatings tend to have a high second harmonic reflectance because of the difficulty in fabricating coatings transmittive to both wavelengths.

The nonlinear crystal quality is the most important factor in high power intracavity SHG. As detailed in Chapter II, thermal effects due to crystal absorption can considerably reduce second harmonic output power. In addition, crystals of poor optical quality can have higher reflectance and scattering losses which increase the laser linear loss. With no method of prejudging crystal quality, crystals of optimal length based on crystal absorption published in the literature (~ 0.005 cm⁻¹) were obtained. This absorption required a crystal length of about 1.5 cm according to the analysis of Ferguson and Dunn. Since a ring laser
would require shorter lengths, and the effects of overcoupling are not severe, crystals in the 1.5 to 3 cm length range were obtained. The crystals were also obtained with small cross sections to facilitate uniform heating and use of a smaller oven.

ADA crystals from INRAD (5 x 5 x 15 mm, 5 x 5 x 15 mm), Quantum Technology (5 x 5 x 15 mm), and Cleveland Crystals (5 x 5 x 20 mm) were purchased. The 30 mm crystal was the only normal incidence crystal obtained. The 15 mm INRAD Brewster cut crystal provided the highest UV powers primarily because it had the lowest insertion loss. The Quantum Technology crystal had a slightly higher insertion loss with the same conversion efficiency. The Cleveland crystal had a considerably higher insertion loss, but also a higher conversion efficiency. As a result, the Cleveland crystal produced the lowest second harmonic powers. It was difficult to determine to what extent this reflected the longer crystal length of the Cleveland crystal since determination of absorption and reflectance losses is difficult. However, because the crystal transmission was strongly dependent on the spatial position of the fundamental beam in the crystal, this crystal probably was of lower optical quality. This lower quality also reduced the tuning range of the laser and single mode operation above 595 nm became difficult due to frequency hops of about 15 nm. There was no reduction in the tuning range of the laser for the low loss INRAD crystal.

Only a limited investigation was performed into the use of AR
coated normal incidence crystals because of the advantages of the Brewster cut. The primary difficulty with the normal incidence crystal is in the availability of a suitable index matching material. Four materials were investigated (fluorocarbon FC-104, silicon oil, carbon tetrachloride and a vacuum leak sealing resin). However, none withstood high UV output powers for prolonged periods of time without substantial decreases in UV power or thermal defocusing. The 30 mm crystal used in the investigation yielded a higher peak UV laser efficiency than the Brewster cut crystals by an amount commensurate with its longer length and higher transmittance to the second harmonic (due to the lack of Brewster surface loss). Hence, this method would be superior if the index matching material difficulty could be overcome. The fact that the longer crystal did not produce significantly greater UV output powers even with its closer to optimum focusing, indicates that the crystal length of 1.5 cm is close to the optimum value. This is an experimental confirmation of the weak dependence of the laser efficiency on crystal length once the optimal length is exceeded. However, a disadvantage of exceeding the optimal length is the decrease in the fundamental laser efficiency resulting from the increased laser linear loss.

III.4 Temperature Phase Matching and Scanning

Reliable and convenient temperature control of the nonlinear crystal is essential for a tunable spectrometer employing noncritical
phase matching. Figure 3.6 depicts UV output power at constant ADA cry-
stellar temperature (35°C) for a scan of 1500 GHz near 295 nm. The FWHM
of 80 GHz for ADA places an upper limit on the useful range of a con-
tinuous single mode scan at only one temperature. Note that the figure
clearly demonstrates the asymmetrical sinc-like dependence that is
expected from the h integral (equation 2.14) as the phase match is
varied. (The side lobe results from nonuniformities in optical length
resulting from inconsistent crystal quality or longitudinal thermal
gradients.) A tunable UV output requires a very precise method of con-
trolling the phase match condition as the wavelength is being scanned.
In this respect the LASL-Rice spectrometer has the unique advantage of
precise frequency control over long ranges through the computer inter-
facing of laser elements.

The quality of the UV signal from the spectrometer is obviously
dependent on the temperature stabilization of the ADA crystal. The
required stabilization can be predicted from the theory of Boyd and
Kleinman. Assuming a crystal length of 1.5 cm, a beam radius of 45
μm and with \( d(n_1 - n_2)/dT = 4 \times 10^{-5} \text{ K}^{-1} \) a FWHM of 43°C (\( \Delta \sigma = 5.65 \))
for the output power as a function of temperature can be determined.
This value is in fair agreement with the experimental FWHM of .35°C
in Figure 3.7. (The difference between theoretical and experimental
widths is a measure of the effective interaction length of the cry-
stal.) ADA then requires a temperature stabilization of at least
± .02°C for a useful second harmonic output signal.
UV Power Dependence on Wavelength

FIG. 3.6
UV Power Dependence on Temperature

8.7 W FUNDAMENTAL POWER

UV POWER AT 293.9 NM

FIG. 3.7
We initially investigated two commercial ovens (Quantum Technology #T-926 and Lasermetrics #S-2CH controlled by a YSI #72 Temperature Controller). Both commercial ovens had large thermal time constants and lacked the necessary temperature reproducibility. The temperature stability of both ovens was also an order of magnitude lower than that required for ADA. In addition, neither oven design was easily adaptable to a Brewster cut crystal. Hence, a compact crystal oven was designed and built for placement in the tight configuration of the ring laser cavity (Figure 3.8). A long term stability of at least ±0.01°C from 20°C to 100°C has been achieved for this oven when used with the Artronix #5301 Temperature Controller. Temperatures below 10°C were not recommended with the design due to the possibility of condensation and frost on the oven windows. At higher temperatures (>20°C) windows were not necessary (reference [6] has reported rapid crystal degradation in air which we did not observe) which yielded higher powers because of the reduction in the cavity loss. The primary advantage of cooling was the increased oven response below 40°C where air convection cooling becomes inefficient.

The Artronix temperature 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 a direct electrical interface. The high reliability of the stepping motor has made active computer monitoring of the temperature unnecessary. The present resolution varies from
CRYSTAL OVEN

SIDE VIEW

THERMISTOR

BREWSTER CUT CRYSTAL

CARTRIDGE HEATER

TEFLON INSULATION

BREWSTER WINDOW

CERAMIC INSULATOR

ALUMINUM BLOCK

(2" x 1.25" x 1.25"

END VIEW

ALUMINUM BLOCK

CARTRIDGE HEATER

THERMISTOR

COPPER PLATE

FOR EO VOLTAGE

TEFLON INSULATION

WATER-COOLED

HEAT SINK

PELTIER HEAT PUMP

NONLINEAR CRYSTAL

CERAMIC INSULATOR

FIG. 3.8
.02°C (25°C) to .06°C (100°C) but can easily be increased by suitable gearing of the stepping motor. The maximum allowed slew rate of 3.5°C/min has an overshoot and settling time of about .3°C and one minute respectively (this software limit is set to be below the maximum slew rate allowed by the crystal of 5°C/min).

We have also had limited success with a new technique for temperature control which would partially eliminate the need for computerized tracking of the oven temperature. This method could considerably simplify scanning by conventional techniques since the temperature is automatically determined by laser frequency to optimize the second harmonic output power. An electro-optic modulation of the crystal refraction indices [31] is used to determine the phase and magnitude of the crystal temperature deviation from the optimal phase matching condition. A small ac voltage is applied along the optic axis of the ADA crystal (perpendicular to the direction of fundamental propagation). In 42m crystals this field rotates the two equal crystallographic axes by 45° and changes the indices to (for example, see reference [32])

\[
\begin{align*}
n^{0I} &= n^0 - \frac{n^0 r_{63}}{2} (V/w) \\
n^{eI} &= n^e
\end{align*}
\]

where V is the applied voltage, w is the crystal width and \( r_{63} \) the electro-optic tensor. Thus, the voltage modulates the second harmonic power by changing the phase matching parameter. The effectiveness of this voltage in modulating \( \Delta k \) can be determined by evaluating
\[ \frac{d(n^0 - n^e)}{dV} = n^0 r_{63}^3 / 2w \]

For ADA with \( w = 0.5 \) cm, \( r_{63} = 9.2 \times 10^{-10} \) cm/V and \( n^0 = 1.58 \) this becomes

\[ \frac{d(n^0 - n^e)}{dV} = 3.6 \times 10^{-9} \]

which can be compared to \( 4 \times 10^{-5} \) for a 1 K change in temperature. This small number makes electrical modulation of the refractive indices a relatively ineffective method of obtaining the phase matching condition. However, a modulating voltage of 100 V, while having a negligible effect on the output signal (it is comparable to a 0.01°C change in temperature), can easily be detected with a lock-in amplifier. The output signal of the phase sensitive amplifier can then be used to control the oven temperature through an electrical interconnection in the Artronix temperature controller. The feedback signal would then automatically insure that the oven temperature would always be adjusted to maximize the second harmonic signal during frequency scans of the fundamental.

In practice, this method of temperature control was less effective than the computerized technique. The problem resulted from the slow thermal response of the oven and the small FWHM of the crystal temperature dependence. These factors limited the scanning speed and resulted in an unacceptable oscillation of the oven temperature during scans. However, the technique is certainly feasible with a properly designed feedback system and/or use of a very low thermal mass oven (as, for example, designed by [33]). In any case, this method could only be used to supplement computer control in the LASL-Rice spectro-
meter since the technique requires very precise control of the laser frequency at all times. A second possible use for the electro-optic modulation would be as a "noise eater" for the second harmonic output. A feedback loop could be constructed such that modulation of the second harmonic by the electro-optic effect would minimize the amplitude fluctuations due to laser noise.

III.5 Laser Specifications

Typical operating characteristics for the laser are shown in Table 3.1. A 6% visible output coupler 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 power. However, use of a maximum reflector for M4 did not produce significantly greater UV power. This was due to the increased thermal losses as well as the high linear loss of the laser when all cavity elements are utilized. The cavity is also difficult to align with a full reflector since the wavelength dependent leakage from the dichroic mirror must be used to monitor fundamental power. Hence, the small advantage in output power when using a full reflector is not worth foregoing the usefulness of having a substantial fundamental output.

The second harmonic output power saturated at high fundamental powers as is evident in Figure 3.9. This experimental plot of
<table>
<thead>
<tr>
<th></th>
<th>Visible:</th>
<th>Ultraviolet:</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Wavelength Range (20% points)</strong></td>
<td>570 nm to 615 nm</td>
<td></td>
</tr>
<tr>
<td><strong>Multimode Output Power</strong></td>
<td>1.1 W @ 5 W pump&lt;sup&gt;2&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>Linewidth 40 GHz</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Single Mode Output Power</strong></td>
<td>800 mW @ 5 W pump</td>
<td></td>
</tr>
<tr>
<td>Linewidth 40 MHz</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Brewster Surface Crystal:</strong>&lt;sup&gt;3&lt;/sup&gt;</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Multimode Output Power</strong></td>
<td>25 mW @ 5 W pump</td>
<td></td>
</tr>
<tr>
<td>Linewidth 40 GHz (visible)</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Single Mode Output Power</strong></td>
<td>10 mW @ 5 W pump</td>
<td></td>
</tr>
<tr>
<td>Linewidth 40 MHz (visible)</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Normal Incidence Crystal:</strong>&lt;sup&gt;4&lt;/sup&gt;</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Single Mode Output Power</strong></td>
<td>12 mW @ 5 W pump</td>
<td></td>
</tr>
<tr>
<td>Linewidth 40 MHz</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<sup>1</sup> utilizing a 2 x 10<sup>-3</sup> molar solution of Rh6G in Ethelene Glycol

<sup>2</sup> for the 514.5 nm line of an argon ion laser

<sup>3</sup> ADA crystal purchased from INRAD, L = 15 mm

<sup>4</sup> ADA crystal purchased from INRAD, L = 30 mm
ULTRAVIOLET LASER OUTPUT POWER

FIG. 3.9
fundamental power (2\% coupler) versus the second harmonic power (as measured by United Detector Technology model #21A) clearly demonstrates the deviation from parabolic dependence at high powers. This saturation effect is due to the thermal effects discussed in the previous chapter, and, as is now evident, they considerably reduce second harmonic power. The flattening of the curve at the highest powers is primarily due to frequency instability in multimode operation created by the crystal dispersive loss. The cavity is also undercoupled with respect to the second harmonic since second harmonic output power had only a negligible effect on the fundamental power.

Maximum multimode powers were obtained by using the birefringent filter and mirror feedback. A record 32 mW was obtained with 6 W of argon laser pump power at 514.5 nm, and 29 mW for 5 W of pump power. However, these powers were unstable and rapidly decayed by over 30\% primarily due to frequency instability resulting from the high crystal resonance loss. The peak powers could only be reobtained by momentarily tuning the laser away from resonance, blocking the pump beam, or spatial translation of the crystal. This effect did not occur in single mode operation of the spectrometer because of the lower maximum power (12 mW) and higher frequency selectivity of the intracavity elements. Typical fundamental output powers with the INRAD Brewster cut crystal and 2\% coupler were 400 mW and 150 mW in multimode and single mode
operation respectively. However, with the normal incidence crystal typical single mode output was less than 100 mW at 5 W of pump power.

The linewidth of the fundamental did not experimentally have a significant effect on the second harmonic power as long as the phase matching condition was nearly satisfied over the entire bandwidth. However, it can be derived [34] that the second harmonic power should increase in multimode operation as \((2m-1)/m\) where \(m\) is the number of modes among which the fundamental power is equally divided. This factor results from a contribution to the second harmonic by sum frequency mixing, and the phase incoherence of the many oscillating modes. It is obvious that this can only be true if the effective linewidth is substantially less than the spectral acceptance of the crystal. Hence, with only the birefringent filter in the cavity, an increase in the second harmonic efficiency by nearly a factor of two as compared with single mode operation could have been observed. This increase was obscured by thermal effects and the crystal dispersive loss at resonance which prevented multimode laser operation at the phase matching wavelength.

The measured conversion efficiencies and the theory of Boyd and Kleinman can be used to calculate the nonlinear coefficient of ADA. When the output powers are corrected for the reflectance losses (17% in the Brewster crystal, 20% in the dichroic
mirror), the nonlinear coefficient for both the INRAD Brewster cut cry-
stals was found to be $d_{36} \approx 5 \times 10^{-24}$ mks. Of primary interest in this result is that the Brewster cut and normal incidence crystals are equally efficient and that astigmatic distortions of the beam did not have a major effect. A $65^\circ$ cut ADP was also used to determine the experimental magnitude of any distortions on conversion efficiency. Analysis of this crystal gave a nonlinear coefficient in excellent agreement with the published value. This indicates that residual astigmatism in the crystal arm has a negligible effect on conversion efficiency.
CHAPTER IV
SPECTROMETER COMPUTERIZATION AND PERFORMANCE

IV.1 System Overview

The LASL-Rice spectrometer is capable of continuous frequency or wavelength scanning over the entire tuning range of the dye ring laser. Each laser element has been interfaced to a DEC PDP 11/03 minicomputer through a CAMAC crate controller to provide simultaneous tuning of each cavity element. Figures 4.1 and 4.2 give a schematic overview of the spectrometer and CAMAC interface. An extensive software package has been developed to provide complete operator control over the wavelength scanning and calibration of the spectrometer, as well as the acquisition, storage, and display of data. The software program consists of thirty commands which control all of the operations of the spectrometer.

The tuning and calibration components of the spectrometer are controlled by six motors: three stepping motors for adjustment of the birefringent filter, the monochromator and the temperature controller; 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 the cross-referencing with a minimum contribution of effort and time.
CAMAC Crate Schematic
by the operator. Once the spectrometer has been calibrated, computer
controlled scanning is performed over the calibrated region by sim¬
taneous tracking of each relevant motor. For a complete discussion
of the calibration and scanning capabilities of the spectrometer the
reader is referred to reference [20].

The spectrometer is capable of scanning in a wavelength or fre¬
quency mode of operation depending on the number of intracavity ele¬
ments. Coarse multimode wavelength scanning is performed with only
the birefringent filter in the cavity. The resolution is then lim¬
it ed by the laser linewidth to .5 Å (or 40 GHz). When the Faraday
isolator is inserted into the cavity, the thin etalon must be fre¬
quency calibrated along with the birefringent filter to perform
continuous scans of up to 1600 GHz with a resolution limited to
2-3 GHz by frequency hopping. In this mode of operation, the air-
spaced etalon is used extracavity 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 the cavity length in
order to perform scans with a resolution limited by the laser line-
width. Up to 1000 data points can be taken from each of four
analog-to-digital (ADC) channels during scans. Software adjustment
of the scanning speed and a time constant for digital filtering
permits optimization of scanning resolution over any frequency or wavelength interval. Data can be displayed graphically on either a video terminal or an X-Y recorder after acquisition and storage.

IV.2 Software Development and Computerized UV Scanning

The system just described is the same as that described in reference [20] and is essentially unchanged by the presence of the nonlinear crystal in the cavity. To perform intracavity UV scanning a reliable method of calibrating crystal phase matching wavelength to laser frequency had to be developed. An initial goal was set of obtaining computerized single mode scanning over the range of fundamental frequency operation extending from 50 to 1600 GHz as described in the preceding paragraph. No attempt was made to calibrate the crystal and wavelength over the entire range of the dye laser (as was performed extracavity by Pollock) for reasons to be discussed shortly. At 600 nm, a 1600 GHz scan (3200 GHz at 300 nm) implied tracking of the crystal over 19 Å and 5.3°C. If tracking is to deviate less than 1/10 of the FWHM during the scan, this requires a total deviation of less than 0.6° over the maximum scan range.

The development of the software for intracavity tuning involved a two step process. First, a highly reliable method of changing and maintaining the crystal temperature had to be devised. This would then permit setting of the crystal temperature at the wavelength of interest so that scanning could be initiated. For the
second step, the laser frequency and phase matching wavelength had to be cross-referenced to permit simultaneous scanning. As described in Chapter III, the crystal temperature is controlled by a stepping motor interface with no active computer monitoring of the oven temperature. The high reliability and resettableness of the control system considerably simplified the software development since computer feedback is a relatively inefficient method of precise temperature control. However, active monitoring of crystal temperature should be utilized in a fully operational system if only to prevent crystal damage due to thermal shock.

The calibration of the temperature controller motor position to oven temperature was performed completely by the system software. Upon initialization of the system, a 100 entry table was generated from the known characteristics of the Artronix dial position (to which the motor was attached) and the oven thermistor. This table was then referenced to the current motor position supplied by the operator. The motor was not operated with the limit switch system devised by Pollock because of the need for constant crystal temperature. Once this table was generated, it was used by the software to change crystal temperature at any constant rate below the 5°C/min maximum required to prevent crystal damage.

The second phase of the software development involved the calibration of the crystal temperature and laser frequency. This task was considerably simplified by the almost linear dependence of the
phase matching wavelength on crystal temperature. As determined experimentally by Stickel [27] in a pulsed dye laser this dependence goes as

\[
\lambda = \lambda_o + AT + BT^2
\]

\[
\lambda_o = 577.71 \text{ nm}
\]

\[
A = 0.2309 \text{ nm/}^\circ\text{C}
\]

\[
B = 5.554 \times 10^{-4} \text{ nm/}^\circ\text{C}^2
\]

where the phase matching wavelength is accurate to \(\pm 2\) Å from \(-40^\circ\text{C}\) to \(100^\circ\text{C}\) at noncritical phase matching. This equation does not include the power dependence of the phase matching temperature (due to the low average power of the pulse system) which is essentially constant [6] and can be estimated from the temperature profile of equation 2.20.

Using equation 4.1 above and the temperature table, a second table of motor position versus frequency was generated. The table was then referenced to the actual laser frequency at a single point by maximizing the second harmonic output at constant temperature. The table was generally accurate over the limited frequency scan range without referencing to additional points. The software has been designed to permit scans to higher or lower wavelengths, or equivalently, to higher or lower temperatures. However, except at high temperatures, it was generally more effective to generate the table toward higher temperatures due to the uncertainties of relying on oven cooling (Peltier cooling was not generally employed).

As previously stated, the software has been designed for continuous single mode UV frequency scans of up to 3200 GHz. No attempt was made to cross-reference longer multimode scans as performed in
the earlier spectrometer version because of the difficulty in maintaining the phase matching condition. This difficulty arises from the complexities of intracavity temperature tuning as opposed to extracavity angle tuning as utilized in the previous system (which generated the cross-reference table of crystal angle motor position versus laser wavelength by maximizing the second harmonic output at intervals over the laser tuning range and then curve fitting the dependence). This is impractical for temperature tuning because heating and then cooling the crystal to set up the table would be prohibitively slow, as would be the scanning. (Generating the table and performing one scan would typically take 1½-2 hours.) However, if this type of long range scanning is to be performed, the only practical way to undertake the task is with computer control where the minimum operator intervention is required.

It is important to point out that temperature tuning over wide wavelength ranges is easier to perform than angle tuning if just the required precision is considered. For example, consider a direct comparison of ADP angle tuning versus ADA temperature tuning from 570 to 620 nm (assuming this wavelength (120°C) can be achieved with ADA). The respective FWHM are $\Delta \theta = 4$ mrad for ADP and $\Delta T = .35$°C for ADA (Blit [35] has shown that $\Delta \theta$ is essentially constant away from noncritical incidence.). To change the phase matching wavelength over this 50 nm interval requires a 10° change
in angle or a 150°C change in temperature. This dictates a precision (FWHM/total change) of .04% and .2% for angle and temperature tuning respectively to remain within the FWHM. Hence, temperature phase matching should be easier to achieve over the range of Rh6G. However, this analysis does not include the formidable problems of varying oven response and uncertainties in oven temperature which are non-existent problems in mechanical angle tuning.

A minor problem in developing the software for tracking of the crystal was the slow response of the oven to a computer imposed temperature change. The lag in the oven response was typically about 15 seconds, but obviously depended on the rate of temperature change as well as the oven temperature. The lag time was not recovered during a scan and primarily determined the degree of phase mismatch. The only way to reduce the response time would be to use a smaller oven which would tend to increase longitudinal temperature gradients in the crystal. A number of methods were considered to minimize the effects on output power of this slow oven response. The simplest solution was to reference the temperature-frequency cross-reference table to an equivalent dummy frequency which would lead the actual laser frequency during a scan by an amount equal to the lag. An experimental analytical equation for the delay as a function of temperature and scan rate was attempted but not implemented due to the substantial software modifications required. The lag time was usually determined by the
operator experimentally prior to scanning.

The synchronization of laser frequency and oven temperature during scans primarily determined the quality of the UV output signal. However, UV noise was also introduced by other sources such as fundamental frequency instability and noise. The second harmonic's critical dependence on fundamental wavelength made any frequency instability in single mode operation have a significant effect on second harmonic output power. The dependence on fundamental noise was of greater importance since frequency instability produces scans of limited usefulness under any circumstances. The dependence on single mode fundamental noise was not as substantial as expected, and at higher powers appeared to be no greater than the fundamental noise. This was attributed to the saturation effect discussed earlier. This result is in sharp contrast to the significant noise of the extracavity doubling system with its strong $P_2$ dependence. Additional noise from temperature fluctuations and the incremental steps of the temperature controller stepping motor was virtually eliminated by the large thermal mass of the oven and the continuous temperature tracking during scans.

IV.3 Ultraviolet Spectrometer Performance

Figures 3.5, 4.3 and 4.4 are typical examples of frequency scans which can be obtained with the spectrometer. Figure 3.5 has been discussed previously, and is mentioned here because it is a good example of the resolution obtainable by scanning the birefringent
800 GHz SO₂ Absorption Spectra

FIG. 4.3
400 GHz SO$_2$ Absorption Spectra

300.04 TO 300.14 NM

UV POWER

MARKER CAVITY
8 GHZ FUNDAMENTAL

I$_2$ FLUORESCENCE
SPECTRUM

SO$_2$ ABSORPTION
SPECTRUM

FIG. 4.4
filter and c-axis etalon without the intracavity air-spaced etalon. This technique has only limited application in spectroscopic measurements because of the previously mentioned 2-3 GHz frequency hops. Figure 4.3 and 4.4 demonstrate spectra which can be obtained with the air-spaced etalon in the cavity. (Both $\text{SO}_2$ spectra were obtained from a 25 cm path length cell at 5 torr.) The fundamental output is used during scans to provide both a marker signal and a simultaneous reference spectra of the well documented fluorescence of $\text{I}_2$. Figure 4.3 is a typical longer range scan (800 GHz) which demonstrates the stability of the UV output (note the reference signal). Figure 4.4 is a higher resolution scan taken near the same region of the spectra. The initial lag in the oven response is evident in the UV reference at the start of both scans. Spectra similar to these have been obtained with varying resolution and high reproducibility from 298 to 303 nm in $\text{SO}_2$. An attempt was also made to detect the fluorescence of $\text{SO}_2$ in the same region using the high power capability of the spectrometer. This attempt was unsuccessful because any possible fluorescence was dominated by UV scatter in the cell. However, the measurements did yield high quality absorption spectra from the strong dependence of the scatter on UV absorption at the pressures utilized (5 torr, 200 mtorr).
REFERENCES


29. R.E. Stickel, private communication.


32. A. Yariv, Quantum Electronics, pp. 327-343.

