INVESTIGATION OF LASER DYNAMICS, MODULATION AND CONTROL BY MEANS OF INTRA-CAVITY TIME VARYING PERTURBATION

under the direction of

S. E. Harris

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# STAFF

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# PRINCIPAL INVESTIGATOR

S. E. Harris

PROFESSORS

A. E. Siegman

R. L. Byer

RESEARCH ASSISTANTS

A. Kovrigin

J. E. Murray

J. F. Young

#### INTRODUCTION

The work under this Grant is generally concerned with the generation, control, and stabilization of optical frequency radiation. In particular, we are concerned with obtaining tunable optical sources by means of nonlinear optical techniques. During this period work was active in three particular areas. These were: first, on a novel technique to produce long, high-power optical doubled pulses from Q-switched lasers; second, on a ring-type optical parametric oscillator which has allowed very substantial depletion of the driving argon laser, and third, on a proposed new method which should allow the output frequency of an optical parametric oscillator to be locked to the absorption line of an atomic transition. The effective linewidth of the oscillator should become approximately that of the absorbing transition. Work on the tunable acoustooptic filter has also continued, but the bulk of this work has now been transferred to Navy Research Grant NO0014(67-A-0112)0036.

During this period the following publications have been submitted for publication:

Joel Falk and James E. Murray, "Single Cavity Noncollinear Parametric Oscillation," Appl. Phys. Letters 14, 245-247, April 1969.

A. I. Kovrigin and R. L. Byer, "Stability Factor for Optical Parametric Oscillators," to be published in J. Quant. Electr.

S. E. Harris, "Method to Lock an Optical Parametric Oscillator to an Atomic Transition," submitted to Appl. Phys. Letters.

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R. L. Byer, A. I. Kovrigin, and J. F. Young, "A Ring Cavity CW Parametric Oscillator," submitted to Appl. Phys. Letters.

S. E. Harris, "Nonlinear Optical Materials," a talk delivered before the 1969 Conference on Laser Engineering and Applications, Washington, D. C., May 26-28, 1969.

# Pulse Lengthening Via Overcoupled Internal Second Harmonic Generation (S. E. Harris and J. E. Murray)

A definite need has arisen for an effective means of extending the length of the Q-switched laser pulses which are used to drive optical nonlinear devices. In particular, longer pumping pulses are required to allow these interactions to reach true steady state operating conditions and to reduce the danger of burn damage to optical components. Several authors<sup>1-8</sup> have proposed schemes for pulse lengthening where the fundamental laser frequency is the useful output. This scheme offers controlled Q-switched pulse lengths of the second harmonic of the laser frequency. It also has the advantage that the pulse lengthening is accomplished in a conservative manner.

The method proposed here utilizes a second harmonic generator internal to the laser cavity. This acts as a power dependent loss to the fundamental laser frequency, and if the coupling to the second harmonic is much stronger than that required to maximize the second harmonic power, it limits the peak amplitude of the Q-switched pulse and extends its length in time. This scheme is conservative in that the second harmonic pulse length can be varied while keeping its energy constant. For other pulse lengthening schemes, such as those which utilize Raman, Brillouin, or Rayleigh scatters<sup>2,3</sup> as the power dependent losses, or those which

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use two photon absorption,<sup>4-7</sup> the energy lost to the scattering mechanism is not useable. For the proposed scheme, second harmonic generation acts as both the power dependent loss and the means of output coupling for the laser; thus the energy lost to second harmonic generation is the useful output.

The theoretical analysis of the problem<sup>9-11</sup> begins with the usual rate equations for a laser system with an additional term in the photon density equation representing the loss to the second harmonic generator:

$$dN/dt = W - (\sigma c/LA)uN - N/\tau_{tot}$$
$$du/dt = -u/\tau_c + (\sigma c/LA)N(u + 1) - Ku^2$$

where

N	=	$N_2 - N_1 = Total population inversion$
W	-	Rate of population inversion due to pumping
u	=	Total number of photons per pertinent cavity mode
τc	==	Photon lifetime in optical resonator
$^{\tau}$ tot	<u>,</u> ==	Total lifetime in upper state
K	Ħ	Coupling coefficient between fundamental and second harmonic
L	=	Optical length of resonator
A	=	Beam area in laser material
σ	nia	Transition cross section for the laser line of interest
	-	$\lambda^2/4\pi^2 \epsilon_{\tau} \rho \Delta \nu$
		$\epsilon$ = Relative dielectric constant of laser material
		$\tau_{\ell}$ = Radiative spontaneous lifetime in upper state
		$\Delta v$ = Full linewidth of transition at half power points

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In terms of the second harmonic crystal parameters, K can be defined as follows:

$$K = \frac{8\pi\hbar}{w^2} (\eta \nu)^3 (d\ell)^2 \left(\frac{c}{L}\right)^2$$
$$= \xi h \nu \left(\frac{c}{L}\right)^2$$

where  $\xi$  characterizes the second harmonic crystal and is defined according the following equation:

$$P_{SH} = \xi P_{Fund}^2$$

and where

w = Beam radius in second harmonic crystal

 $\eta$  = Impedance of second harmonic crystal to the fundamental wave-length

v = Fundamental frequency of laser

- $\ell$  = Effective interaction length in second harmonic generation.
- d = Effective electro-optic constant for second harmonic generation.

For Q-switched operation, the pumping term and the spontaneous emission terms can be neglected. With these simplifications, the equations can be normalized as follows with only one parameter,  $\beta$ , representing the coupling to the second harmonic frequency:

$$dn/dT = - \phi n$$
  
$$d\phi/dT = \phi(n-1) - \beta \phi^2 ,$$

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where

$$n = N/N_{th} = (\sigma c/LA)\tau_c N$$
  

$$\phi = (\sigma c/LA)\tau_c u$$
  

$$T = t/\tau_c$$
  

$$\beta = (LA/\sigma c)K$$

The second harmonic coupling parameter,  $\beta$ , can also be defined in the following way in terms of parameters which are more characteristic of laser systems and second harmonic crystals:

$$\beta = \frac{(\xi c/L)}{G/E}$$

where

 $\xi = P_{SH}^2/P_{Fund}^2$ .

G = Gain per pass through laser medium

E = Energy stored in inversion to give this value of G .

To obtain the time evolution of the Q-switched pulses we programmed these equations on a digital computer. Figure 1 shows the computer generated second harmonic pulse shapes for  $n_0 = 2.0$  and for five different values of the coupling parameter  $\beta$ . The normalized amplitudes of the pulses can be related to second harmonic power by the following relation:

$$P_{SH} = \left(\frac{hvLA}{\sigma c\tau_c^2}\right)\beta \phi^2$$

The initial value for the normalized photon density,  $\phi_0$ , used in each case was approximately 1/100 of its corresponding peak value rather than

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FIG. 1--Computer solutions for second harmonic pulse shapes for  $n_0 = 2.0$ , and five different values of the coupling parameter,  $\beta$ . The upper figure shows the  $\beta = 10^2$  and  $10^3$  pulses with an expanded time scale.

the appropriate spontaneous noise power. This was done to reduce computing time.

In order to check the computer solutions, an attempt was made to solve for some characteristic of the pulses analytically. By dividing the equation for  $\phi$  by the equation for n , the variable T can be eliminated and the resulting differential equation can be solved in closed form. Its solution is:

$$\phi(n) = \left(-\frac{1}{\beta} + \frac{n}{\beta-1}\right) + \left(-\phi_0 + \frac{1}{\beta} - \frac{n_0}{\beta-1}\right)\left(\frac{n}{n_0}\right)^{\beta}$$

where  $\phi_0$  and  $n_0$  are the initial values of the variables. From this, an expression can be obtained for the peak amplitude of the fundamental pulse:

$$\phi_{\max} = -\frac{1}{\beta} + \frac{n_0}{\beta} \left[ \frac{n_0}{\beta n_0 - (\beta \phi_0 + 1)(\beta - 1)} \right]^{\frac{1}{\beta - 1}}$$

This solution was checked against the computer solutions and indicated that the program was working correctly.

We intend to compute solutions to these normalized equations for as many cases of interest as possible. The results will be presented in terms of graphs of pulse length vs initial inversion, peak power vs initial inversion, etc., with different values of the coupling parameter  $\beta$  giving families of curves on each plot. We will also attempt to demonstrate the effect experimentally.

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# 2. <u>A cw Ring Cavity Parametric Oscillator</u>, (R. L. Byer, A. Kovrigin, J. F. Young)

The ultimate usefulness of tunable optical sources will depend considerably on their efficiency and stability. We have just constructed a new type of parametric oscillator using a three mirror ring cavity having greater stability and conversion than a conventional two mirror linear cavity oscillator. In the two mirror oscillator, the resonant signal and idler waves traveling through the nonlinear element in the backward direction (opposite the pump wave) are phased matched for the sum frequency generation of a backward wave at the pump frequency. This back generated pump wave interferes with the input pump beam and reduces the effective pump field. Recently Bjorkholm<sup>12</sup> pointed out that a parametric oscillator operating without a back generated pump wave has the potential of 100% conversion efficiency when operated 4 times above threshold, in contrast to a maximum of 50% in the presence of the backward pump wave.<sup>13</sup> The singly resonant oscillator eliminates the backward pump wave at the expense of a considerably increased threshold. Tn the ring cavity, the resonated signal and idler travel through the nonlinear crystal in the forward direction only, thus eliminating the backward generated pump wave. In addition, the off angle ring configuration provides very effective isolation between the oscillator and the pumping The elimination of the back generated pump wave and of direct laser. specular reflections from oscillator component surfaces should result in improved pump and oscillator stability.

Figure 2 shows the components of the ring cavity oscillator. The oscillator was pumped with a single frequency 5145 Å argon ion laser which

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M2: flat gold mirror

FIGURE 2

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was properly mode matched into the parametric cavity to achieve maximum parametric gain<sup>14,15</sup> and was continuously monitored with a confocal scanning Fabry-Perot interferometer to insure that it was single frequency. Operation was far from degenerate with a tuning range from 6600 Å to 7000 Å at the signal and a corresponding idler tuning range near 2.0  $\mu$ . A three to one ratio of signal to idler frequencies was chosen to simplify the fabrication of the dielectric mirrors and the LiNbO2 antireflection coatings, as described previously.<sup>16</sup> Both the signal and idler waves were resonated in the three mirror cavity consisting of two 5 cm radius dielectric morrors and a flat gold mirror. The gold coated flat was mounted just above the crystal inside the oven so that the dielectric mirrors were used at nearly normal incidence. Alignment was accomplished using the pump beam and the small reflectivity of the oscillator components at 5145 Å. We found that the difficulty of aligning the three mirror cavity was comparable to that of the two mirror cavity. The nonlinear element of the oscillator was a 34 mm crystal of LiNbO3 grown at the Center for Materials Research, Stanford University.

The single pass cavity loss at the signal and idler was approximately 2% which, with confocal mode radii of 40  $\mu$  and 70  $\mu$  at the signal and idler respectively, gives a theoretical threshold of 50 mW. We believe that the discrepancy between this and the observed value of 150 mW can be accounted for by incomplete overlap of the pump, signal, and idler waves in the crystal, and by errors in the cavity loss measurements. In particular, a linear cavity oscillator constructed with the same crystal and dielectric mirrors had a threshold of 60 mW, indicating that the gold flat at grazing incidence was more lossy than estimated.

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We observed up to 60% pump depletion indicating operation near two times above threshold. Operation further above threshold was not possible due to limited pump power. No precautions were taken to stabilize the oscillator or pump cavities, and the pulsing behavior typical of the phase and frequency sensitive, doubly resonant oscillator was observed. The ring cavity oscillator exhibited on-times as much as ten times longer than the linear cavity oscillator constructed using the same components. The Fabry-Perot interferometer clearly showed the improved frequency stability of the pump due to the effective isolation of the ring cavity configuration. In spite of the large pump depletion of the output power at the signal was relatively low due to the large ratio of internal losses to coupling losses, and peak output powers of only a few milliwatts were observed.

In conclusion, the operation of this parametric oscillator verified the increased pump depletion predicted for a parametric oscillator operating without the back generated pump wave, and also showed the improved stability which can result from the optical isolation afforded by the ring cavity configuration. These advantages were obtained with only a very moderate increase in construction and alignment difficulty.

# Method to Lock an Optical Parametric Oscillator to an Atomic <u>Transition</u>, (S. E. Harris)

Work on this project is described in the attached paper, Appendix A, which has been submitted for publication to Applied Physics Letters.

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 R. L. Byer, M. K. Oshman, J. F. Young, and S. E. Harris, Appl. Phys. Letters <u>13</u>, 109 (1968). APPENDIX A

# METHOD TO LOCK AN OPTICAL PARAMETRIC OSCILLATOR

TO AN ATOMIC TRANSITION

## METHOD TO LOCK AN OPTICAL PARAMETRIC OSCILLATOR

TO AN ATOMIC TRANSITION

by

S. E. Harris

In recent years optical parametric oscillators which are tunable over most of the visible and near infrared spectra have been constructed.<sup>1-6</sup> The linewidth of these oscillators is determined by the length and type of nonlinear crystal, and by how close to degeneracy they are operated. Observed linewidths have been as small as 3 cm<sup>-1</sup> and have more typically been between 30 and 100 cm<sup>-1</sup>.

In this Letter we propose a technique which allows the output frequency of an optical parametric oscillator to be locked to the frequency of an absorbing atomic transition. The effective linewidth of the oscillator becomes approximately that of the absorbing transition, which for a gas may be  $.03 \text{ cm}^{-1}$  or less.

The basic configuration for the proposed technique is shown in Fig. 1. The usual single nonlinear crystal is replaced by two nonlinear crystals which have the direction of their positive z axes reversed. For example, in LiNbO<sub>3</sub>, the positive end of the z axis is that end which becomes negative upon compression in the z direction. Between the reversed nonlinear crystals is placed a cell containing the gas to which it is desired to lock the output frequency of the oscillator. We term the frequency of the absorbing transition as the signal frequency, and assume the mirrors of the oscillator to have high reflectivity at

$$\begin{array}{c|c} P \\ i \\ i \\ -L_1 \\ -L_1 \\ -L_1 \\ -L_2 \\ -L$$

FIG. 1--Schematic of frequency locking technique. The letters s, i, and p denote the signal, idler, and pump respectively. The gas transition is assumed to absorb at only the signal frequency; and the mirrors are assumed to have high reflectivity at only the idler frequency. The vertical arrows in the LiNbO<sub>2</sub> crystals denote the direction of their positive z axes. only the idler frequency. Oscillators having only their signal or idler frequencies resonant have recently been successfully demonstrated.<sup>7-9</sup>

To understand the operation of the oscillator, first consider the case where the two nonlinear crystals are of equal length, and the absorbing gas is removed. At the center of the parametric linewidth where  $\Delta k = 0$ , the parametric gain of the first crystal is exactly cancelled by the second crystal. That is, as a result of the reversal of the +z axes of the two crystals, the relative phases of the signal, idler, and pump on entering the second crystal are such that instead of further growth, the signal and idler decay to the values which they had on entering the first crystal. Since relative phases are involved, the physical spacing between the two crystals is not of consequence. Suppose now that an absorbing gas is inserted into the gas cell and the pressure adjusted such that the gas is nearly opaque at the pertinent transition. The signal generated in the first crystal is now absorbed by the gas, and therefore cancellation of gain no longer occurs in the second crystal. The result is a sharply peaked gain function centered at the frequency of the atomic transition. It will be seen below that the peak height of this gain function is between fifty and one hundred percent (50 - 100%) of the gain which would exist if the +z axes of the two crystals were aligned instead of opposed. It will also be seen that it will be of advantage to make the second crystal a number of times longer than the first.

We will take the gas to have a loss  $\alpha(\omega)$  and phase shift  $\phi(\omega)$ such that the signal frequency  $\overline{E}$  field at its output is related to that at its input by exp -  $[\alpha(\omega) + j \phi(\omega)]$ ; and assume the gas to

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be completely transparent at the idler frequency. We take the first nonlinear crystal to have length  $L_1$  and the second to have length  $L_2$ . The magnitude of the parametric gain at the idler frequency for a single pass through the two reversed crystals separated by the gas cell is given by

$$\left| \frac{E_{12}}{E_{10}} \right|^{2} = \left| \left( \cosh sL_{1} + \frac{j \Delta k}{2s} \sinh sL_{1} \right) \left( \cosh sL_{2} + \frac{j \Delta k}{2s} \sinh sL_{2} \right) - \exp - \alpha(\omega) \exp j \phi(\omega) \frac{r^{2}}{s^{2}} \sinh sL_{1} \sinh sL_{2} \right|^{2}$$
(1)

where

$$s = \left(\Gamma^2 - \frac{\Delta k^2}{4}\right)^{1/2}$$

The wave vector mismatch  $\Delta k = k_p - k_s - k_i$  is zero at the center of the parametric gain curve and may be written  $\Delta k = b \Delta \omega_s$  where  $\Delta \omega_s$  is the excursion from line center of the signal frequency. For LiNbO<sub>3</sub> at 6328 Å, the constant  $b = 6.2 \times 10^{-10} \text{ sec/m} \cdot ^{10} \Gamma^2$  is the parametric gain constant and is dependent on the strength of the pump. To obtain Eq. (1), pump depletion was neglected and thus  $\Gamma$  was taken to be the same in both crystals. For 90° phase matching in LiNbO<sub>3</sub> with  $\lambda_s \cong \lambda_i \cong 1\mu$ ,  $\Gamma^2 \cong \cdot 1 P_p/A$  where A is the area of the pumping beam and  $P_p/A$  has units of MW/cm<sup>2</sup>.

We first examine the shape of the parametric gain curve which is predicted by Eq. (1) in the absence of the absorbing gas, i.e., with

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 $\begin{aligned} \alpha(\omega) &= \phi(\omega) = 0 \quad \text{. Figure 2 shows the incremental power gain} \\ \left| \textbf{E}_{12} / \textbf{E}_{10} \right|^2 &= 1 \quad \text{plotted versus } \Delta \textbf{k} (\textbf{L}_1 + \textbf{L}_2) \quad \text{, for three cases where} \\ \textbf{FL}_1 &= .5 \quad \text{, } \textbf{FL}_2 = .5 \quad \text{; } \textbf{FL}_1 = .25 \quad \text{, } \textbf{FL}_2 = .75 \quad \text{; and } \textbf{FL}_1 = .2 \quad \text{, } \textbf{FL}_2 = .8 \quad \text{,} \\ \text{respectively. For the case of two crystals of equal length the gain is} \\ \text{zero at line center and rises off line center. Crystals of differing} \\ \text{length (note that the sum of the lengths is held constant) increase the} \\ \text{gain at line center and reduce the ripple.} \end{aligned}$ 

Next, consider the gain in the presence of the absorbing gas. We will assume that the linewidth of the absorbing transition is many times smaller than that of the parametric gain, and that the temperature of the nonlinear crystals have been adjusted such that their parametric linewidths center at approximately the atomic transition. For such cases  $\Delta k$  may be taken equal to zero in the vicinity of the atomic transition, and Eq. (1) gives

$$\left|\frac{E_{i2}}{E_{i0}}\right|^{2} = \left|\cosh \Gamma L_{1} \cosh \Gamma L_{2} - \exp - \alpha(\omega) \exp j \phi(\omega) \sinh \Gamma L_{1} \sinh \Gamma L_{2}\right|^{2}$$
(2)

For large  $\alpha(\omega)$ , i.e., for a nearly opaque gas,  $|E_{12}/E_{10}|^2 = \cosh^2 \Gamma L_1 \cosh^2 \Gamma L_2$ . This gain is shown plotted as a delta-function at  $\Delta k = 0$  for each of the three cases in Fig. 2. For the case of a four-to-one ratio of crystal lengths, i.e.,  $\Gamma L_1 = .2$ ,  $\Gamma L_2 = .8$ , the ratio of gain at the atomic transition to the maximum parametric gain which occurs elsewhere on the line is about 1.8. Choosing the second crystal longer than the first has the additional advantage that the signal power entering the gas from the first crystal varies as  $L_1^2$ , and thus the possibility of saturating the gas is reduced.

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The detailed shape of the parametric gain in the vicinity of the transition is determined by the relation of  $\alpha(\omega)$  to  $\phi(\omega)$ . For an ideal Lorentzian line  $\phi(\omega)/\alpha(\omega)$  is proportional to the detuning from the atomic line center. For small  $\alpha(\omega)$ , the peak parametric gain occurs at the center of the atomic line. However, for larger  $\alpha(\omega)$ , e.g., higher pressure or longer cell length, higher gain results toward the wings of the atomic line where the ratio of  $\phi(\omega)$  to  $\alpha(\omega)$  is larger. (Note that the most favorable case for Eq. (2) is  $\alpha(\omega) = 0$ ,  $\phi(\omega) = \pi$ .) In this case the gain becomes double peaked, dipping at the center frequency of the atomic transition. As a result of this phase contribution of the atomic line, the peak gain may exceed the height of the delta-functions shown in Fig. 2, and the width of the gain function may be a few times wider than that of the atomic transition.

In constructing an oscillator of the type described here, it may be desirable to make the c/2L frequency spacing of the idler modes less than the width of the atomic transition. Even though the pump frequency is randomly fluctuating, there would then always be at least one idler mode such that the difference between the pump frequency and its frequency falls within the width of the atomic transition.

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