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

under the direction of

S. E. Harris

Quarterly Status Report

(Report No. 10)

for

NASA Grant NGL-05-020-103

National Aeronautics and Space Administration

Washington, D. C.

for the period

1 February 1970 – 30 April 1970

M. L. Report No. 1849

April 1970

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NASA Grant NGL-05-020-103

for the period

1 February 1970 – 30 April 1970

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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 non-linear optical techniques. During this period, work was active in the following areas. These were: first, pulse lengthening via overcoupled internal second harmonic generation; second, electronic tuning and short pulsing of dye lasers; third, laser stabilization studies; and fourth, backward wave oscillation. Progress reports on these topics are given in the following sections.

During this period the following publications have appeared in the literature:

S. E. Harris, "Tunable Optical Parametric Oscillators," Proc. IEEE 57, 2096 (December 1969).

S. E. Harris, S.T.K. Nieh, and D. K. Winslow, "Electronically Tunable Acousto-Optic Filter," Appl. Phys. Letters 15, 325 (November 1969).

J. E. Murray and S. E. Harris, "Pulse Lengthening Via Overcoupled Internal Second Harmonic Generation," J. Appl. Phys. 41, 609 (February 1970).

1. Pulse Lengthening Via Overcoupled Internal Second Harmonic Generation

(J. E. Murray and S. E. Harris)

The experimental effort on the KDP-ruby laser system has been completed. The fundamental pulse lengthening experiment was successful and indicated substantial agreement with the previously presented theory. The results of the second harmonic experiment, however, were disappointing due to the low damage threshold of the laser mirrors to the second harmonic radiation.

The experiment at the fundamental laser frequency was repeated with considerably improved results. The cavity geometry used was essentially the same as that presented in the last report; the mirror spacing was near to hemispherical to give a large mode volume at the ruby crystal and a small mode at the second harmonic generator.

The data for 24 consecutive firings of the laser at constant pumping power are presented in Fig. 1. The values of β less than its maximum at $\Delta k = 0$ were obtained by tuning with angle. Since the cavity mode apparently changed as the second harmonic crystal was tuned away from $\Delta k = 0$, it was necessary to experimentally obtain this tuning curve of β vs angle. For this, the geometry of the pulse lengthening experiment was used with the laser operated in the normal mode rather than in the Q-switched mode.

The theoretical curves on the figure are based on a 25^{th} laser shot with the second harmonic crystal detuned to give a β of essentially zero. Its pulse length of 35 nsec and the best estimate of the cavity lifetime, $\tau_c = 3.45$ nsec, gave a value for the initial inversion of $n_0 = 1.5$; this was sufficient to establish the theoretical curves.

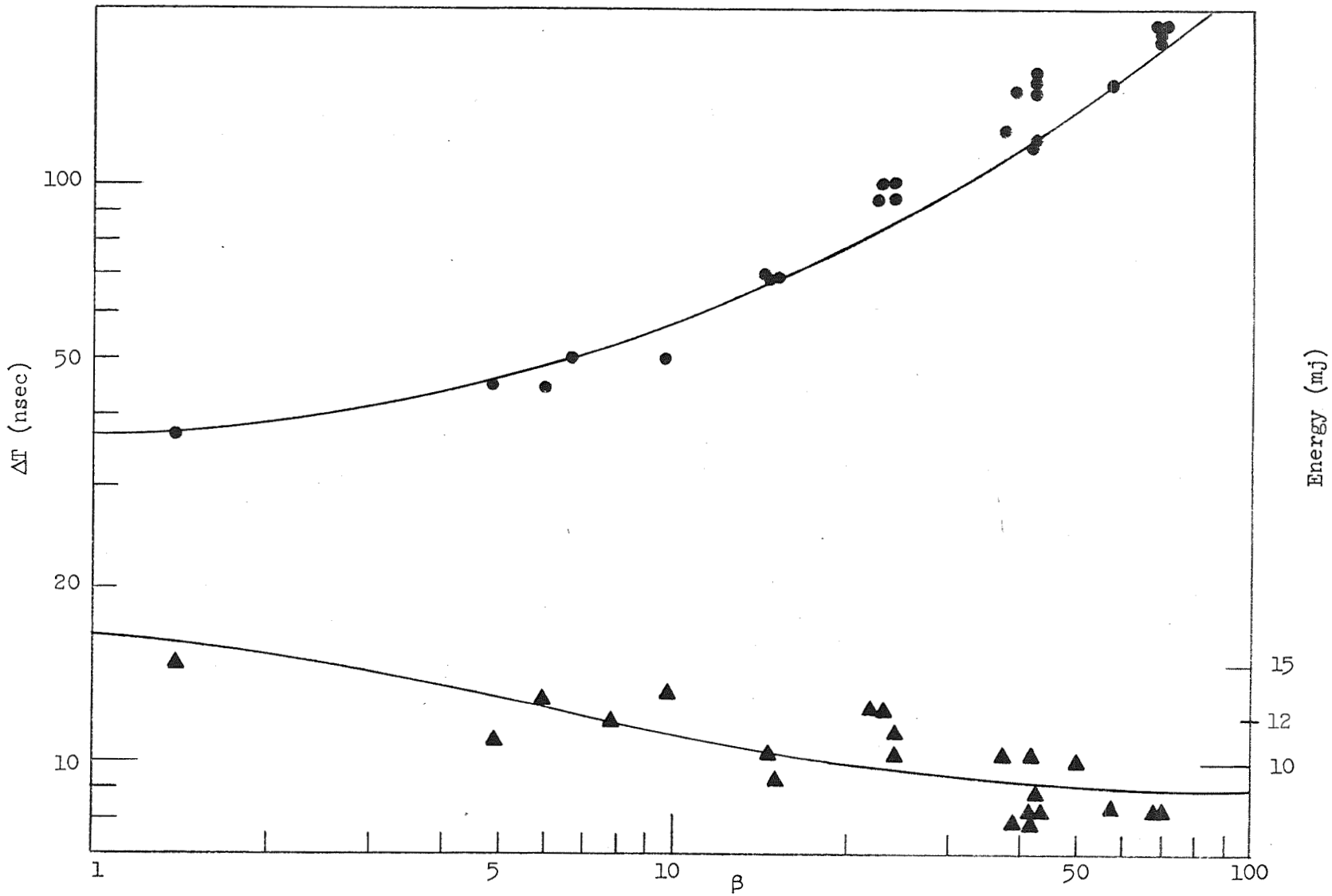

FIG. 1--Pulse width and energy vs β from the fundamental pulse lengthening experiment. The circles and triangles represent experimentally obtained pulse widths and energies, respectively, for 24 consecutive pulses at constant pumping power. The solid curves are theoretical fits based on a 25^{th} pulse with $\beta = 0$.

The decrease in pulse energy with β evident in the data was due to the change in the final value of the population inversion, n_f, with β. For $\beta = 0$, n_f is between zero and one, and approaches zero as the initial population inversion, n_0, is increased;[1] for this data with $n_0 = 1.4$ and $\beta = 0$, $n_f = 0.7$. For β much larger than one, $n_f = 1 - 1/\beta$. The total energy available for laser action is given by

$$E_T = \frac{h\nu}{2} N_{th}(n_0 - n_f)$$

and since this data was taken with $n_0 = 1.4$, the change in n_f from 0.7 to 0.98 gave rise to a substantial change in pulse energy. For larger values of n_0, the percentage change would be correspondingly less. However, for any value of n_0, the pulse energy becomes essentially independent of β for large values of β.

The calculated values of E_T and β based on the cavity geometry used were about three times larger than those observed experimentally; the fact that both values were too large by the same amount suggests that the error is in a factor common to both terms, the most likely candidate being the mode area in the laser crystal. Twyman-Green interferograms of the ruby rod indicate that there was considerable distortion in the vicinity of the beam path through the rod. This, coupled with the effects of thermal focusing in the rod, could possibly account for the above discrepancy by decreasing the effective cavity length and therefore reducing this mode size.

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[1] W. G. Wagner and B. A. Lengyel, J. Appl. Phys. 34, 2040 (1963).

These experimental results indicate that Q-switched ruby pulses with half-widths of 150 - 180 nsec and pulse energies of 10 millijoules or more can be obtained easily and repeatably. This is a pulse length of 6 - 10 times longer than is normally obtained in the lowest order transverse mode from ruby lasers with this pulse energy. Considerably longer pulses (e.g., 740 nsec at 16.5 millijoules) were obtained with tighter focusing geometries and higher pumping energies, but these results were not repeatable.

The cavity geometry for the second harmonic experiment was similar to that for the fundamental except that the high transmission sapphire etalon was replaced with a second 60 cm dielectric coated mirror. The mirror coatings were highly reflecting at the fundamental frequency and about 85% transmitting at the second harmonic. Unfortunately, these mirrors damaged at second harmonic pulse energies of more than 2 to 3 millijoules. Although pulse lengthening was observed at these second harmonic pulse energies, this maximum set by the damage threshold of the mirrors was too low to be of practical interest to us. Also it prevented the acquisition of data such as that of Fig. 1 because it was too near the detectable minimum of the TRG thermopile used to monitor pulse energies.

2. Electronic Tuning and Short Pulsing of Dye Lasers

(D. J. Taylor and S. E. Harris)

The goal of this project is to obtain an electronically tunable coherent light source by inserting an acousto-optic filter element into the cavity of a dye laser. In our last report we discussed several aspects of the theory of this device. Since then considerable progress has been made in the design and construction of the experimental apparatus.

The configuration shown in Fig. 2 illustrates the components needed to obtain an acoustically tunable dye laser. The dye cell, which may be excited by flashlamps or by another laser of higher frequency, provides a large gain for a broad band of optical frequencies. A narrow frequency band of the y-polarized light incident on the acousto-optic element is diffracted into the orthogonal polarization by the collinear acousto-optic interaction and is passed by the z-polarizer, while for frequencies outside this band no diffraction occurs and the y-polarized light is blocked by the z-polarizer. After reflection from the mirror the narrow band is again diffracted, from z-polarized to y-polarized, and passes through the y-polarizer to experience gain in the dye cell. Thus only for this narrow band of frequencies is the loss outside the dye low enough to allow oscillations to build up.

There are several factors that render the simplified configuration considered above unacceptable. Most critical is the requirement that the acoustic wave travel collinearly with the optical wave, which must be achieved without placing the acoustic transducer, with its metal electrodes, in the optical path. This can be accomplished by reflecting the acoustic beam off an inclined free end surface through which the light also passes;

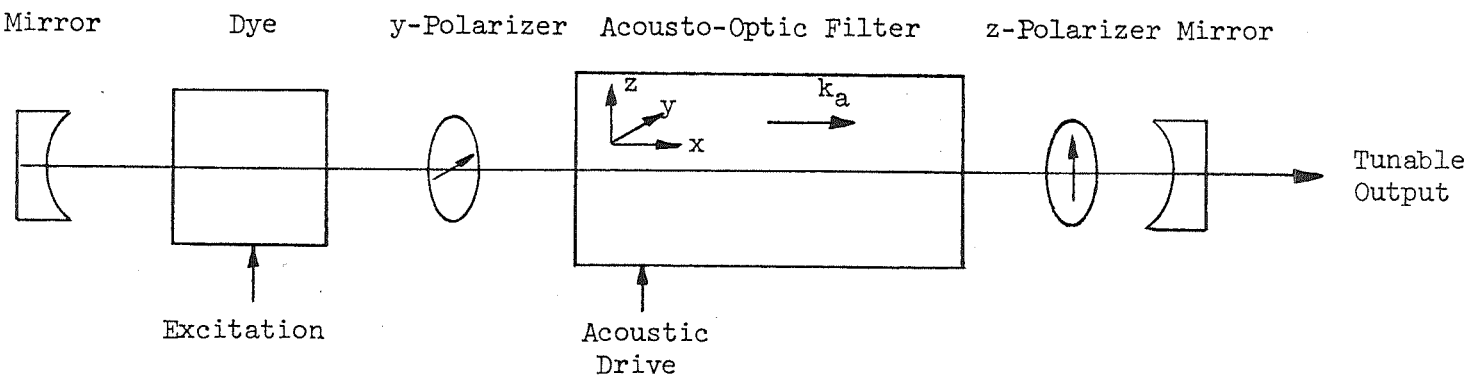

FIG. 2--Simplified acoustically tunable dye laser.

the reflection process will be discussed below. Other criticisms of the above configuration include the large number of surfaces, each with certain reflection losses which may represent cavity loss for the desired narrow frequency band, and the possibility of the normal surfaces of the y-polarizer reflecting enough light back into the dye, with its extremely large gain, to permit oscillation at undesired frequencies. The actual experimental configuration must avoid these problems.

The acousto-optic filter will be a calcium molybdate ($CaMoO_4$) crystal, chosen because of its high elasto-optic coefficients and because its low birefringence permits tuning with relatively low acoustic frequencies, about 60 MHz. The development of the growth technique for high optical quality $CaMoO_4$ crystals is a continuing project at Stanford's Center for Materials Research, and boules up to 10 cm in length have been grown. One recurring problem in every a-axis crystal grown to date has been the existence of a filament of optical inhomogeneity in the cross-sectional center, extending over the length of the boule. During this last quarter we grew a boule of $CaMoO_4$ of sufficiently large transverse dimensions (2.1 cm $\times$ 1.6 cm) that it can be cut lengthwise into two usable crystals, each 3.4 cm long, with the filament of inhomogeneity essentially cut out. This crystal was evaluated optically by standard tests developed at this lab[2] for transmission (less than 2% variation over cross-section) and birefringence (uniform over the cross-section except at the center core, and uniform along the crystal length). This crystal, after being sliced lengthwise, will be used for the acousto-optic filter element.

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[2]R. L. Byer and J. F. Young, "Quality Testing of $LiNbO_3$ Crystals," Stanford University Microwave Laboratory Report No. 1749 (April 1969), to be published in Journal of Applied Physics.

During this last quarter we have constructed our own dye laser. Since we already had a high-power blue laser available to excite the dye, this actually proved to be relatively simple; because of the high gain of the dye cavity, mirror alignment is not too critical to achieve lasing. The excitation laser was developed here[3] and is a Nd^{3+}:YAG laser operating at the 0.946 μ line, which is internally doubled to 0.473 μ by a $LiIO_3$ crystal and Q-switched by an acoustic Q-switch to achieve high peak output power (5 kW max) in the blue, repetitively pulsed at 10 pps. We plan to use the dye sodium fluorescein (also called uranine), which can be made to lase in ethanol (easiest), methanol, or water (hardest), and has a broad gain band of 300-400 Å in the yellow-green. Experimentally, the most significant aspect of the dye is its very large gain, which we were able to measure by measuring its threshold energy both with and without a 3 dB neutral density filter placed inside the cavity. For 10^{-4} molar concentration in ethanol, the highest gain solution, we measured $\alpha = 1.07/\text{kW-cm}$; for this dye solution in a .9 cm cell with a 5 kW pump we should be able to obtain threshold even for single-pass cavity losses as high as 82% . In our experiment we will use a 10^{-4} molar concentration of sodium fluorescein in ethanol, with a center wavelength of 5370 Å.

For collinear interaction of the optical and acoustic waves along the x-axis in $CaMoO_4$ the necessary acoustic wave is an S_{13} shear wave, which as mentioned above, can be obtained by reflection from an inclined surface, with the incident wave traveling along the z-axis being either a shear wave or a longitudinal wave. If a shear wave is incident, all the acoustic

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[3] R. W. Wallace and S. E. Harris, "Oscillation and Doubling of the 0.946 μ Line in Nd^{3+}:YAG," Appl. Phys. Letters 15, 111 (August 1969).

energy is transferred to the reflected shear wave and the surface must be at 45° since the velocities of the incident and reflected shear waves are equal. Because of the high index of refraction of $CaMoO_4$, there is no incident angle for which light refracted at this 45° surface can propagate along the x-axis unless the crystal is surrounded by a liquid (which still allows the surface to be considered acoustically free) with an index greater than 1.422. For an incident longitudinal wave the process is called mode conversion; when a longitudinal wave is obliquely incident upon a free surface, there are in general 2 reflected waves, one longitudinal and one shear. Their directions and amplitudes depend upon the elastic properties (velocities of propagation) of the material and the angle of incidence, and can be determined by requiring that the normal component of the stress tensor $(\vec{n} \cdot \vec{T})$ vanish at a free surface. The mode conversion process in $CaMoO_4$ was analyzed using the isotropic approximation. Imposing the condition that the reflected shear wave propagate along the x-axis specifies the angle of incidence to be $61^{\circ}39'$, and the resulting 96.2% conversion of energy from the incident longitudinal wave into the reflected shear wave is very close to the maximum conversion that could be obtained in $CaMoO_4$ at the optimum angle of incidence. A calculation to estimate the maximum amount of light that could be deflected by the reflected longitudinal wave via other elasto-optic tensor elements indicated that even for the greatly exaggerated figure of 100 watts of incident longitudinal power, the deflected optical intensity is less than 10^{-8} of the incident light, and is therefore a negligible optical loss. Because the normal to the inclined surface is closer to the x-axis for the longitudinal-to-shear mode conversion case than for the shear-to-shear

case, it is easier to introduce the optical beam into the $CaMoO_4$ to propagate along the x-axis, and can even be done from air.

The primary criterion for the experimental configuration must be the minimization of single-pass optical loss outside the dye, with secondary criteria of simplicity of construction and ease of alignment. The configuration that best satisfied these criteria is shown in Fig. 3; its overall single-pass transmission, assuming 100% optical diffraction efficiency in the $CaMoO_4$ and neglecting mirror losses, is 83%. The $CaMoO_4$ crystal will be immersed in the dye solution to reduce the number of surfaces in the cavity and to provide better matching of indices of refraction (at 5370 Å ethanol n = 1.3639). Longitudinal-to-shear mode conversion will be accomplished at the surface cut at angle α_1 . The other surface is cut at angle $\alpha_2 \neq \alpha_1$ to permit the z-polarized beam leaving the α_2 surface to be parallel to the y-polarized beam leaving the α_1 surface; having $\alpha_2 \neq \alpha_1$ also ensures against acoustic resonances, which can limit the optical diffraction efficiency. With this configuration there are no normal surfaces off which reflected light could allow the dye to lase at undesired frequencies. Finally, there is the possibility of obtaining the output coupling by allowing somewhat less than 100% optical diffraction from z- to y-polarized light and taking advantage of the double refraction phenomenon to couple the remaining z-polarized light out of the cavity; in this way output coupling could be varied electronically within the filter element by changing the acoustic drive amplitude.

At this time the $CaMoO_4$ crystal is being cut and polished, and the other components are being assembled, and during the next quarter the electronically tunable dye laser will be tested.

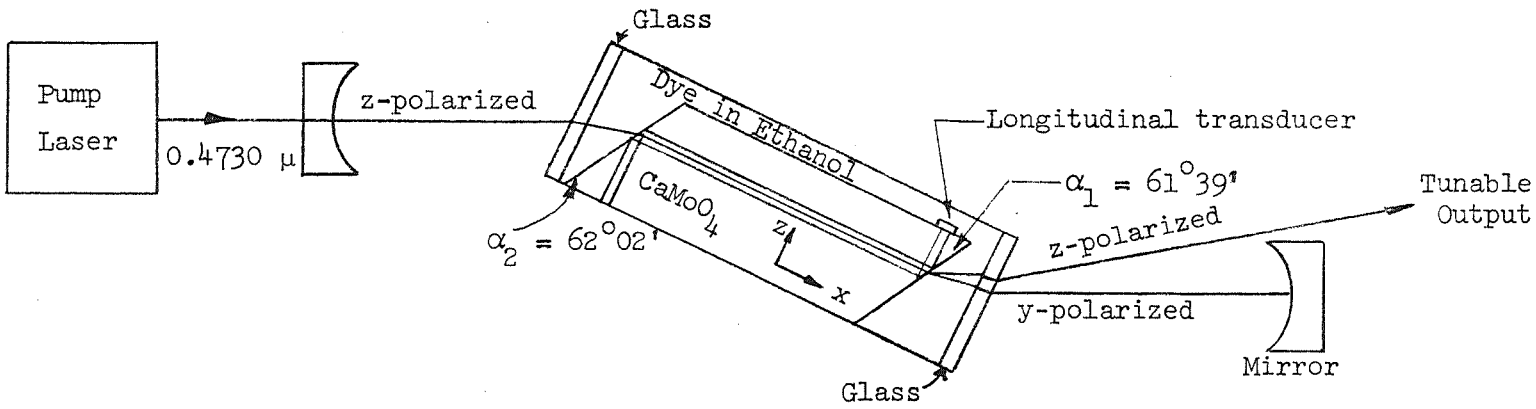

FIG. 3--Experimental configuration for acoustically tunable dye lasers.

During this last quarter we began evaluation of a new nonlinear material, $Gd_2(MoO_4)_3$, which may prove to be useful for parametric oscillators in the infrared. It is a ferroelectric crystal, with a Curie temperature of 160°C, and at room temperature it is slightly orthorhombic, but almost uniaxial. Because of its symmetry properties (mm 2 point group at room temperature) and the fact that $n_e > n_o$ for this material, only Type II phasematching, in which the signal and idler are orthogonally polarized, is possible, and a higher birefringence is needed than for Type I phasematching (signal and idler identically polarized). We measured the indices of refraction of a sample of $Gd_2(Mo_4)_3$ at several wavelengths, using an apparatus similar to that described by W. L. Bond,[4] and found that phasematching does not appear to be possible for the degenerate parametric process $1.06\mu \rightarrow 2.12\mu$ or higher frequency degenerate processes. Investigation into the properties of this material is continuing.

[4] W. L. Bond, "Measurement of the Refractive Indices of Several Crystals," J. Appl. Phys. 36, 1674 (May 1965).

3. Laser Stabilization (S. C. Wang)

The goal of this project is to stabilize a laser using internal saturable absorption. In particular, we are continuing work with the He-Xe laser (3.51 μ) using DME (di-methyl-ether) and low pressure xenon as saturable absorbers.

The experimental work on DME has now been completed. From the measurement of the absorption coefficient vs pressure of the DME gas at 3.51 μ, we have obtained the following significant information. The low pressure (< 5 Torr) absorption coefficient is approximately 4×10^{-3} cm^{-1} $Torr^{-1}$, and the transition lifetime is approximately 2 seconds; the collision broadening frequency at 15 Torr and 293°K is 1.6×10^{8} sec^{-1}, and a saturation intensity of about 2 mW/cm^2 for DME at 1 Torr has also been obtained. The detailed results have been submitted for publication.[5]

An attempt has also been made to obtain a qualitative interpretation of the inverted Lamb dip spectrum observed with a DME absorption cell inside the laser oscillator.[6] A number of closely spaced absorption lines have been observed, and the origin of these may be one or more of the following:

(1) Splitting of the top doublet due to the slight asymmetry of the DME molecule. This splitting will be a few tens of MHz depending on the J values.

(2) Overlap of absorption band due to high order effects such as the overtone of the ν_4 oscillation, and coriolis coupling.

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[5] S. C. Wang and A. E. Siegman, "Absorption Coefficient, Transition Lifetime and Collision Broadening Frequency of DME at 3.51 μ He-Xe Lasers," submitted to IEEE J. Quant. Electr.

[6] R. I. Myers, private communication.

(3) Internal rotation of the molecule.

Due to the asymmetric and complicated structure of the DME molecule, the assignment of J numbers, and quantitative analysis is very difficult.

In this part of the work on DME, we have demonstrated the existence of the inverted Lamb dip, which is essential for frequency stabilization, and have obtained all the necessary information to use DME as a saturable absorber. However, due to its asymmetric and complicated molecular structure, the usefulness as a frequency stabilizing absorption gas is limited.

Preliminary results on a second method of frequency stabilization, using a low pressure pure isotope Xe gain cell, have been very promising. In this system, the low pressure Xe cell provides an enhanced Lamb dip. The Xe cell is cooled to liquid nitrogen temperatures and a pressure of approximately 5 μ is maintained. The enhanced Lamb dip observed was approximately 5 MHz wide. The dip was strong and easy to observe. This experiment will be continued during the next quarter.

4. Backward Wave Oscillation (J. Falk and S. E. Harris)

The backward wave oscillator project is an attempt to produce coherent far infrared radiation by means of a temporarily unstable, three photon parametric interaction.

Attempts at backward wave oscillation this quarter used 10° and 15° cut lithium niobate and a TEM_{oo} mode ruby laser. These crystal cuts were designed to provide phase matching well below the lowest vibrational mode of $LiNbO_3$ at 109 μ. The 10° cut crystal was expected to phase match at 2 mm where the calculated absorption loss of $LiNbO_3$, based on measured vibrational mode strengths, is 0.20 cm^{-1}. The 15° cut crystal was expected to phase match at 1 mm where the expected E field losses were believed to be 0.76 cm^{-1}.

The ruby laser, which has a nominal 2 mW output of 20 nsec duration, was focused with 45 cm and 19 cm lenses to provide a beam waist at the center of the $LiNbO_3$ crystal. In spite of the fact that pump densities were well above the 160 mW/cm^2 (100 mW/cm^2) calculated threshold for the 10° (15°) cut crystal oscillation was not observed. Detection equipment consisted of a one-meter Spex spectrometer (resolution $\leq$ 0.25 Å) set to photograph the idler output which was expected to be several angstroms from the ruby pump.

We believe that our failure to observe backward wave oscillation in $LiNbO_3$ was due to the strain induced birefringence wander commonly observed in all but a and b cut niobate crystals.

Attempts at backward wave oscillation are now being suspended pending the improvement of the quality of off-angle lithium niobate crystals or the development of other materials suitable for backward wave oscillation.