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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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 producing tunable optical sources by means of non-linear optical, and acousto-optical techniques. During this period work was done in the following areas: electronic tuning and short pulsing of dye lasers, laser stabilization studies, and locking of optical parametric oscillators to atomic transitions. Progress reports on these topics are given in the following sections.

1. 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 the design considerations for our first experiment, including the properties of the pump laser, the dye laser, preparation of the filter crystal $CaMoO_4$, generation of the desired acoustic shear wave by longitudinal-to-shear mode conversion, and experimental configuration. During this report period the various components have been fabricated, assembly and alignment techniques have been developed, and partially successful experimentation has been performed.

In addition, a theoretical analysis of the dye laser and acoustic filter has indicated that the resulting output may be 100 times narrower than the bandwidth of the acoustic filter alone. This analysis is presented in Appendix A, and has been submitted for publication.

The $CaMoO_4$ filter crystal was cut and polished according to the design previously presented. Hardware constructed for this experiment included the filter-and-dye holder and a pair of adjustable mirror holders for the end mirrors of the dye laser cavity. The piezoelectric transducer material used for the generation of a longitudinal acoustic wave is a thin (~ 75 μm for fundamental resonance at the acoustic frequencies of interest) slice of $LiNbO_3$ oriented with its surface normal at 35.8° to the y-axis in the y-z plane. This crystal and orientation offer a large electromechanical coupling coefficient (0.485) to an almost pure longitudinal mode with no coupling to any shear modes. The transducer is bonded to the $CaMoO_4$ by a thin layer of phenyl benzoate, with typical one-way insertion loss of 8 dB from input electrical power to acoustic power in the $CaMoO_4$ crystal. The major problems with the acoustic transducer are the large insertion loss, the narrow frequency band for which efficient coupling can be achieved, and the mechanical fragility of the thin slice. To overcome the handling problems, some of the transducers we used were sliced for third harmonic resonance (~ 225 μm thick); however, these transducers have only one-third the bandwidth of a fundamentally resonant slice. The process of preparing transducers after they have been cut, including metallization of the top surface, ball-bonding a wire for top-surface contact, phenyl benzoate bonding, and coating the bond with Sylgard to protect the phenyl benzoate from the ethanol-dye solution, has required considerable development and practice.

Our first experiments were made with the dye Rhodamine 6G, 10^{-5} molar in ethanol, at a center wavelength of 5900 Å, rather than with sodium fluorescein at 5370 Å as first planned, because we already had appropriate dye laser mirrors at that frequency and the corresponding filter transducer is thicker. The first step was to set up a dye laser with the filter element outside and to check that the filter, with crossed polarizers on both sides, can tunably filter the dye laser light. However, it was not possible to measure polarization conversion efficiency as a function of acoustic drive level, since the incident light was broadband compared to the filtered band, resulting in negligible depletion at the incident polarization even if 100% conversion in the narrow filter band could be attained. This first test with the filter external to the dye laser also serves to align the filter for maximum overlap of the acoustic and optical beams. We then placed the filter element inside the dye laser (moving the second dye cavity mirror to account for the dogleg so introduced) and, with no acoustic power applied, got the dye laser running with no polarization change; the Brewster-plane faces of the filter dictate that the resulting polarizations at both ends of the filter crystal are extraordinary. The resultant dye laser output is still quite wide (~ 200 Å). With this configuration an interesting test of the filter can be made. As the acoustic drive is increased from zero at a frequency corresponding to a line within the dye laser spectrum, a point is reached at a relatively low polarization conversion efficiency (perhaps 5%, depending upon how far above threshold the dye laser is operating) where there is enough loss via polarization diffraction and subsequent refraction at the second crystal face into an angle outside the cavity's acceptance angle, that the frequency band within the filter bandwidth can no longer reach threshold. The dye laser spectrum consequently appears to have a line quenched, which is tunable by changing the acoustic filter frequency. The preceding steps were all successful. The final step toward obtaining an electronically tunable dye laser is to set the acoustic drive for 100% polarization conversion at a frequency within the dye laser spectrum, increase the pump laser power (to overcome the higher dye cavity loss when the polarization at the second $CaMoO_4$ face is horizontal and the second face is no longer a Brewster-plane face), and rotate the second dye cavity mirror to accept the horizontally-polarized beam. The resultant dye laser operation would be polarized (N_e, N_o) at the $CaMoO_4$ faces and would be narrow band (at least as narrow as the filter bandwidth) with its center frequency electronically tunable by changing the acoustic frequency. This last step was attempted many times under many different conditions of acoustic drive, pump laser power, etc., but tunable operation was not observed. With so many variables in that final step, we decided to modify the experimental configuration.

Our next experiments were at a center wavelength of 6328 Å, where a He-Ne laser could be used to predetermine the acoustic power level and optimum filter alignment for 100% polarization conversion and to aid in dye cavity alignment. For operation at 6328 Å, a new acoustic transducer, standard 6328 Å mirrors and the dye Rhodamine B(10^{-3} molar in ethanol) were used. The major problem with this configuration was the much lower gain of the dye than in the previous experiment. Furthermore, although the dye would lase in a band centered about 6328 Å with the filter absent from the cavity, the additional loss introduced by the filter tended to shift the dye spectrum up in frequency, usually so far that 6328 Å was not within the dye laser spectrum. It proved difficult to obtain an acoustic transducer and a dye laser spectrum that both centered at 6328 Å. The first step described above, except with the substitution of the He-Ne laser for the dye laser, could be performed if the transducer centered at, or at least reasonably overlapped, 6328 Å. (Reversion to fundamentally resonant transducers helped considerably here.) The second step was often difficult due to the low gain of the dye laser, and usually the frequency shift in the dye laser spectrum prevented observation of a quenched line at 6328 Å. Even when the first two steps were accomplished successfully, we could not perform the final step successfully, probably because we did not have enough pump power to reach threshold for the case of polarization (N_e, N_o) at the $CaMoO_4$ faces.

At present we are reviewing our experimental configuration and planning our next experiment. We are investigating methods of achieving higher dye gains, even if this requires changing dye and losing the convenience of the He-Ne alignment laser. To overcome the uncertainty involved with required higher pump power to reach threshold for tunable oscillation with (N_e, N_o) polarization after achieving (N_e, N_o) oscillation we plan to cut a new $CaMoO_4$ crystal with its two end faces cut in orthogonal planes so that with (N_e, N_o) polarization both faces are Brewster-plane faces. To aid in final alignment, it may be fruitful to employ a three-mirror, two-dye cell cavity. Filter alignment and acoustic level adjustment for 100% conversion, which requires narrow band incident light, may be obtained by several possible routes: using a spectrometer or another acousto-optic filter with incident white light, or by constructing another dye cavity with an internal diffraction grating.

2. Laser Stabilization

(S. C. Wang)

Work on this project during this period is described in the attached papers: Appendix B, which will be published in the September issue of the IEEE Journal of Quantum Electronics; and Appendix C, which has been presented at the 1970 Device Research Conference in Seattle, and will be published in the August 1 issue of Applied Physics Letters.

3. <u>Locking of Optical Parametric Oscillators to Atomic Transitions</u>
(R. B. Miles, J. F. Young and S. E. Harris)

The goal of this project is to achieve the power of efficient solid-state lasers within the bandwidth of any cold gas absorption line. The technique and theory have been described by S. E. Harris in Applied Physics Letters, 14, 335 (June 1969), and involve the use of a parametric oscillator with two reversed nonlinear crystals separated by a cell containing the gas. In this case the gain characteristic of the collection is essentially identical to the absorption linewidth of the low pressure gas. Thus the oscillator should produce high power coherent radiation locked to, and coincident in frequency with, the gas absorption. Such a source will have uses in studying gas absorption, saturation, and relaxation processes, as well as detection of low level atmospheric pollutants over long path lengths by resonant absorption, and tracking gas clouds or concentrations by resonant scattering.

During this period work has been directed at constructing a stable, reliable single crystal optical parametric oscillator having an output in the vicinity of 3.0 μ. Many gases of interest have vibration-rotation bands in this region. A reliable laser pump source using a Nd:YAG laser operating at 0.946 μ and internally doubled to 0.473 μ has been completed. The second harmonic pulse lengthening techniques developed in the last reporting period on the ruby laser have been used in this system to obtain 500 nsec, 3 kW pulses at 0.473 at rates of 5 pps.

Using this pump we have built a singly resonant oscillator tunable from 2.45 μ to 3.2 μ in the infrared, with a corresponding range of 0.555 μ to 0.580 μ in the visible. It has been found that in oscillators of this type there is a significant advantage in resonating the longer wave output. This is described in the attached paper entitled "Pump Linewidth Requirement for Optical Parametric Oscillators," (Appendix D) which has been submitted to Applied Physics Letters. The linewidth of the resonant infrared idler is less than 1/4 wavenumber and theoretically could be made narrower.

We plan to simplify the experimental procedure by replacing the two inverted oscillator crystals by a double pass through a single crystal. with a relative π phase shift of pump, signal, and idler for the second pass. A double pass oscillator without a gas or phase shift has been successfully built, and works well. Introduction of the phase shift using the electro-optic effect of a small section of the $LiNbO_3$ oscillator crystal should extinguish the oscillation. Insertion of the gas will counteract this gain cancellation within the gas line only and locked oscillation should commence. Gases which may be used include methane, CO, HBr, and HCl. Temperature tuning of the oscillator crystal will cause the oscillator to sequentially lock to successive, adjacent rotation lines, spaced by several wavenumbers.

APPENDIX A

ANALYSIS OF DYE LASER TUNED BY ACOUSTO-OPTIC FILTER

by

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ABSTRACT

A model of a dye laser electronically tuned by an acousto-optic filter is analyzed. Chirping modes are superposed to yield a steady-state periodic solution which predicts a shift of the spectrum to the high end of the filter band, and appreciable spectral narrowing. For the example considered, the theory predicts a narrowing factor of 100 .

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I. INTRODUCTION

S. E. Harris has proposed that a dye laser be tuned by an electronically-tunable acousto-optic filter of the type proposed by Harris and Wallace[1] and demonstrated by Harris, Nieh, and Winslow.[2] The proposed arrangement, diagrammed in Fig. 1, shows a tunable filter of the transmission type placed within the laser cavity. Filters of this type recently studied by Harris and Nieh[3] are of $CaMoO_4$ and have optical bandwidths of a few angstroms with tuning over the visible range produced by varying acoustic frequency from 50 to 100 MHz, approximately. It will be shown that a simplified model of this system not only predicts electronic tuning over the broad dye linewidth, but also an appreciable line narrowing over that predicted simply by the filter bandwidth.

A characteristic of special interest for the transmission filters made to date is a shift in the optical frequency which is in the same direction for a wave propagating to the right and for one to the left.

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(Configurations can be devised to give other behavior, but the described property is of special interest to this paper). We take the shift to be an upshift, so that an optical wave of frequency f_0 is shifted to $f_0 + 2f_a$ after a round-trip through the filter, where f_a is the acoustic frequency. The change in frequency disturbs the feedback condition as normally conceived, so it is not immediately obvious that there will be any useful output from the laser.

The initial behavior of this simple model can be deduced by a physical reasoning. There are typically a few thousand longitudinal modes of the unperturbed laser within the few-angstrom band of the filter. Each is continuously swept upward in frequency after each pass through the filter, and increased in amplitude by the gain of the dye. Since unsaturated gain of the dye is appreciable (approximately 3) the modes will build up from noise to appreciable levels in the several thousand passes they undergo before being swept out of the high-frequency edge of the filter. There is thus a marked shift of the spectrum to the high-frequency end of the filter which produces a consequent line narrowing. Furthermore, the analysis given below shows that there is a steady-state periodic solution for the model.

Consider a cavity with plane parallel mirrors of separation L and a device which upshifts frequency by f_a each pass. Normal or self-consistent modes for such a cavity must chirp at the rate f_a per pass or $2\nu f_a$ per second. Such behavior is exhibited by functions in the form

$$\exp\left[j\theta_q(t)\right] = \exp\left\{2\pi j\left[(\nu q - f_a)\, t + \nu f_a t^2\right]\right\} \quad , \qquad (1)$$

where $\nu = c/2L$ is the longitudinal mode separation and q is an integer which denotes the longitudinal mode number. In a time interval $1/\nu = 2L/c$ equal to a round-trip transit the frequency

$$f_q(t) = \frac{1}{2\pi} \frac{d\theta_q}{dt} = \nu q - f_a + 2\nu f_a t \qquad (2)$$

has shifted by $2f_a$. Note too that in $T = 1/(2f_a)$ the frequency changes by ν so that the q-mode at $t + T$ has the same frequency as the $(q + 1)$ - mode at t. Because of the chirping modes the observed spectrum is expected to be smeared rather than discrete.

We expand the cavity fields in the chirping modes

$$\mathcal{E}(t) = \sum_q \mathcal{E}_q(t) \exp [j\theta_q(t)] \quad , \qquad (3)$$

and seek a steady-state periodic solution by requiring that $\mathcal{E}(t + T) = \mathcal{E}(t)$. Since during T each mode chirps in frequency by ν, we find

$$\mathcal{E}_q(t + T) = \mathcal{E}_{q+1}(t) \exp (j\gamma_q) \quad , \qquad (4)$$

where γ_q is a constant phase factor.

The electric field amplitudes at $t + T$ are also related to their values at t as follows. For amplitudes let

$\mathscr{G}$ = saturated dye gain/pass

$\mathscr{H}(f)$ = filter transmission/pass

$\mathscr{L}$ = resonator factor/pass

where $\mathscr{L}$ includes the effects of all other losses including output coupling. Since the time per pass is $1/(2\nu)$ the rate of change of mode amplitudes is

$$\frac{d\mathcal{E}_q}{dt} = \mathcal{E}_q \left[\mathscr{G}\mathscr{H}(f)\mathscr{L} - 1\right] 2\nu \quad . \tag{5}$$

The solution of (5) is

$$\mathcal{E}_q(t + T) = \mathcal{E}_q(t) \exp\left\{ \int_t^{t+T} \left[\mathscr{T}(f) - 1\right] 2\nu dt \right\} \quad , \tag{6}$$

where $\mathscr{T}(f) = \mathscr{G}\mathscr{H}(f)\mathscr{L}$. Since the time interval is short and $\mathscr{T} \approx 1$ we find

$$\mathcal{E}_q(t + T) \approx \mathcal{E}_q(t) \left\{ 1 + \left[\mathscr{T}(f) - 1\right] (\nu/f_a) \right\} \quad . \tag{7}$$

Then by combining (4) and (7), disregarding the phase and setting $t = 0$, we obtain

$$\mathcal{E}_{q+1} = \mathcal{E}_q + \mathcal{E}_q \left[\mathscr{T}(f) - 1\right] (\nu/f_a) \quad . \tag{8}$$

Considering q a continuous variable with $f = \nu q$, expanding in a Taylor series and substituting in (8) yields

$$\frac{d\mathcal{E}(f)}{df} = \frac{\mathcal{E}(f)}{f_a} \left[\mathscr{T}(f) - 1\right] \quad . \tag{9}$$

An alternate derivation of Eq. (9) follows by noting that the power spectral density, $P(f)$, is upshifted by f_a during one traverse of the cavity, while the power is modified by $\mathcal{T}^2(f)$, i.e.,

$$P(f + f_a) = \mathcal{T}^2(f)\, P(f) \quad ,$$

or

$$\mathcal{E}(f + f_a) = \mathcal{T}(f)\, \mathcal{E}(f) \quad .$$

Using the Taylor expansion as above yields Eq. (9).

The solution of Eq. (9) is

$$\mathcal{E}(f) = \mathcal{E}(f_0) \exp\left\{\frac{1}{f_a} \int_{f_0}^{f} [\mathcal{T}(f') - 1]\, df'\right\} \quad , \tag{10}$$

where f_0 is the lowest oscillating frequency defined by

$$\mathcal{T}(f_0) = 1 \quad . \tag{11}$$

The pumping level and saturation parameter determine $\mathcal{E}(f_0)$ and $\mathcal{T}(f)$. We take $\mathcal{T}(f) = \mathcal{T}_0 \operatorname{sinc}[\alpha(f - f_c)]$, as in Refs. 1 and 2, where f_c is the filter center, α determines the filter bandwidth and $\mathcal{T}_0$ is the net gain per pass at f_c including losses and saturation. Equation (10) is then

$$\mathcal{E}(f) = \mathcal{E}(f_0) \exp\left\{\frac{\mathcal{T}_0}{\alpha f_a} \Big[\mathrm{Si}[\alpha(f - f_c)] - \mathrm{Si}[\alpha(f_0 - f_c)]\Big] - \frac{(f - f_0)}{f_a}\right\} \quad , \tag{12}$$

where

$$Si(y) = \int_0^y \mathrm{sinc}(y)\, dy \quad .$$

The maximum of the amplitude spectrum given by Eq. (12) occurs at $f = f_m = f_c + (f_c - f_0)$ (see Fig. 2). By employing a Taylor expansion about f_m that equation becomes

$$\mathcal{E}(f) \approx \mathcal{E}(f_m) \exp\left\{\frac{\alpha(f - f_m)^2}{2f_a} \quad \mathcal{T}_0 F\right\}. \quad , \tag{13}$$

where

$$F = \frac{\cos[\alpha(f_c - f_0)]}{\alpha(f_c - f_0)} - \frac{\sin[\alpha(f_c - f_0)]}{[\alpha(f_c - f_0)]^2} \quad . \tag{14}$$

Thus the amplitude has decreased by $\exp(-1)$ at

$$\Delta f = \left(\frac{-2f_a}{\alpha \mathcal{T}_0 F}\right)^{1/2} \quad . \tag{15}$$

The dependence on f_a and the filter bandwidth (inversely proportional to α) are clear.

The experiment presently in progress uses a 2 cm $CaMoO_4$ crystal with $f_a = 60$ MHz and $\nu \approx 1000$ MHz . The filter bandwidth is approximately 10 Å and $\alpha \approx 3 \times 10^{-12}$. Then if $\mathcal{T}_0 = 1.18$, $F = .3$, $f_m - f_c$ corresponds to 3.3 Å and $\Delta f = 10^{10}$ Hz or $\Delta\lambda = .1$ Å ; if $\mathcal{T}_0 = 1.015$, $F = .1$, $f_m - f_c$ corresponds to 1 Å and $\Delta\lambda = .17$ Å . The use of a 2 cm $LiNbO_3$

crystal with f_a and α increased by approximately 10 does not affect Δf, but in either case Δf is inversely proportional to $L^{1/2}$, where L is crystal length.

The above analysis considers only the amplitudes and not the phases. Clearly, if the modes are properly phased either because of dye nonlinearity or a relationship between ν and f_a pulse behavior may occur. The Gaussian form of (13) would predict Gaussian pulses.

Some limitations on this model are clear. Saturation has been considered as it affects gain, but not in its nonlinear interactions. Such nonlinear effects will produce a parametric gain contribution to the modes, and a coupling between modes that might favor a mode-locked solution over the steady-state solution considered here. In this context the interesting experiment of Smith[4] should be noted, in which he produced mode locking of a gas laser by continuous variation of the mirror spacing. His experiment produced a continuous shift of intermode spacing as well as of absolute mode frequencies, and is concerned with inhomogeneously broadened systems. His figures show a smeared spectrum as predicted for the chirped modes.[5] Time constants of the dye must also be considered more carefully in determining whether or not the steady-state solution can be realized.

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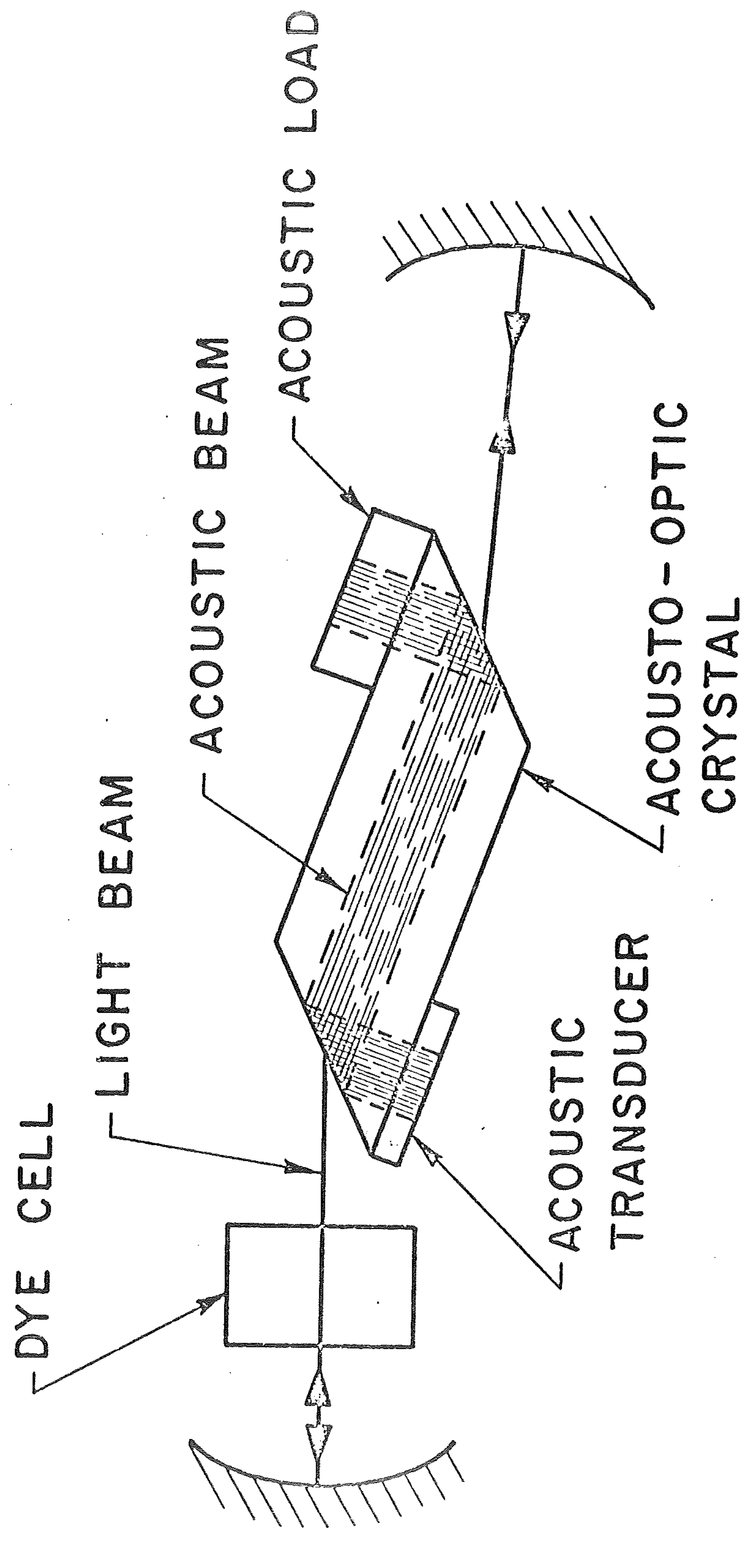
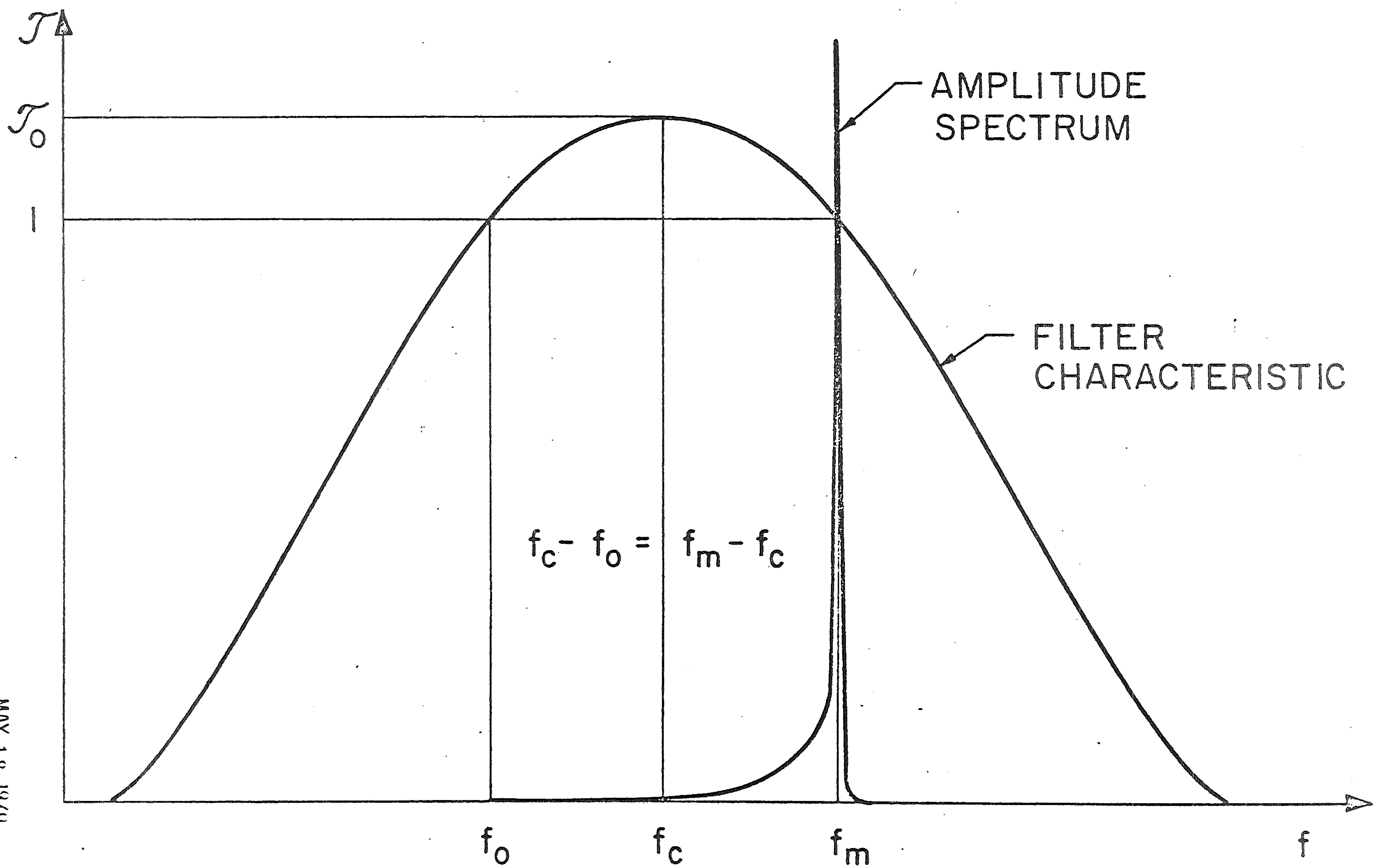
REFERENCES

1. S. E. Harris and R. W. Wallace, J. Opt. Soc. Amer. 59, 744 (1969).
2. S. E. Harris, S. T. K. Nieh, and D. K. Winslow, Appl. Phys. Letters 15, 325 (1969).
3. S. E. Harris and S. T. K. Nieh, to be published.
4. P. W. Smith, Appl. Phys. Letters 10, 51 (1967).
5. B. Alpiner and A. Korpel have considered placing a Bragg scatterer inside a laser cavity to sweep the laser frequency and thus utilize the inversion more completely.

FIGURE CAPTIONS

1. Schematic of the tunable laser. One mirror is aligned to the ordinary wave, the other to the extraordinary.
2. Schematic of the laser spectrum.


DYE CELL
LIGHT BEAM
ACOUSTIC BEAM
ACOUSTIC LOAD
ACOUSTO-OPTIC CRYSTAL
ACOUSTIC TRANSDUCER


$\mathcal{T}$
$\mathcal{T}_0$
1
AMPLITUDE SPECTRUM
FILTER CHARACTERISTIC
$f_c - f_o = f_m - f_c$
f_o
f_c
f_m
f

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APPENDIX B

ABSORPTION COEFFICIENT, TRANSITION PROBABILITY, AND COLLISION BROADENING FREQUENCY OF DIMETHYLETHER AT He-Xe LASER 3.51μ WAVELENGTH

by

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ABSTRACT

The pressure dependence of the absorption coefficient of dimethylether (DME) gas at He-Xe laser 3.508μ wavelength is measured. The experimental results are in fairly good agreement with theory. A collision broadening frequency of $1.6 \pm .1 \times 10^8$ sec^{-1} at 15 Torr, 293^oK and a transition lifetime of $2.2 \pm .2$ sec are obtained. A saturation intensity of about 2 mW/cm^2 for 1 Torr of DME is also estimated.

ABSORPTION COEFFICIENT, TRANSITION PROBABILITY, AND COLLISION BROADENING FREQUENCY OF DIMETHYLETHER AT He-Xe LASER 3.51μ WAVELENGTH

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Experiments on the resonance absorption in certain gases of particular laser oscillation lines have been reported recently.[1-4] By using such an absorption gas inside the laser cavity as a saturable absorber, the laser can be highly stabilized at the atomic line center of the absorption gas. Also, the saturable absorption can provide unusually high resolution spectra for spectroscopy studies. We report here some experimental results on the measurement of absorption coefficients, transition probabilities, and collision broadening frequency of dimethylether (DME), (CH_3-O-CH_3) at the He-Xe laser oscillation wavelength of 3.5080μ . This is part of our attempt to investigate DME as one possible candidate for frequency stabilization purposes.

The He-Xe laser used in this experiment consists of a 20 cm long, 4mm diameter discharge tube filled with 2 Torr of He and ~20 m Torr of single isotope Xe^{136} . The output mirror of the 30 cm long laser cavity is flat with about 50% transmission, and the other mirror is 97.5% reflection. The Doppler width of the laser transition is about 100 MHz and we believe the cavity should provide a single longitudinal oscillation mode. Two irises were placed inside each end of the cavity for partial transverse mode control, but whether the output was limited strictly to the lowest-order transverse mode was not carefully checked. During the absorption measurement the laser output power was kept constant at ~ 720 μwatts and the pressure of the DME was changed from 0.25 Torr to about 70 Torr .

The absorption tube consists of a 35 mm long, 6 mm diameter pyrex tube with quartz windows at Brewster angle placed outside the cavity and left at room temperature during the measurement period. The DME gas has a purity of 99% and was obtained from Matheson Company. The power of the laser with and without absorption by DME gas was measured by an InSb cooled detector calibrated against an Eppley thermopile power meter. The change in intensity of the laser beam due to absorption by the DME varied from a few percent to nearly 90% .

The absorption coefficient at line center for the low-pressure case, where the Doppler broadening of the line is dominant, is given by the expression[5,6]

$$\alpha_0 = \frac{1}{8\pi} \frac{\lambda^3}{\tau} \left(\frac{M}{2\pi kT} \right)^{1/2} \left(\frac{g_2}{g_1} N_1 - N_2 \right) ,$$

where M is the mass of the molecule; λ is the transition wavelength; T is the gas temperature; k is the Boltzmann constant; τ is the radiative lifetime of the transition; and N_1 and N_2 are the populations in the lower and upper levels of the transition, respectively; while g_1 and g_2 are the degeneracy factors of the two levels. At constant temperature and equilibrium condition the absorption coefficient depends linearly on the population difference between the two levels, or in other words, depends linearly on the pressure of the gas. At high pressure, where the collision broadening becomes dominant, the absorption coefficient at line center becomes[5,6]

$$\alpha_0 = \frac{1}{8\pi} \frac{\lambda^2}{\tau \Delta\nu_c} \left(\frac{g_2}{g_1} N_1 - N_2 \right) \quad ,$$

where $\Delta\nu_c$ is the collision broadening frequency. Since this frequency is proportional to the density of molecules, and thus to the pressure of the gas, the absorption coefficient in this region is independent of pressure or density of gas. The measured results of absorption coefficients for DME as shown in Fig. 1, are in fairly good agreement with the theoretical argument of pressure dependence.

In the analysis of the data shown in Fig. 1, we have assumed the resonance absorption of DME at 3.508μ to be from the ground state with a rotational quantum number of about 18, corresponding to maximum intensity. This was obtained from the known rotation constant of DME.[7] The absorption is considered to be within the IR absorption band of CH_3 symmetric stretching.[8,9] The exact transition level involved is difficult to obtain due to the rather complicated spectrum of DME. It is possible that an error could arise in interpretation of the data based on this assumption. From the experimental data the collision broadening frequency was obtained from the point at transition region where the two extrapolated lines intercepted. This yields a collision frequency of $1.6 \pm .1 \times 10^8$ sec^{-1} at 15 Torr , $293^\circ K$. The transition lifetime which best fits the data is $2.2 \pm .2$ sec .

In the low-pressure region, which is of most interest from the frequency stabilization point of view, the absorption coefficient is

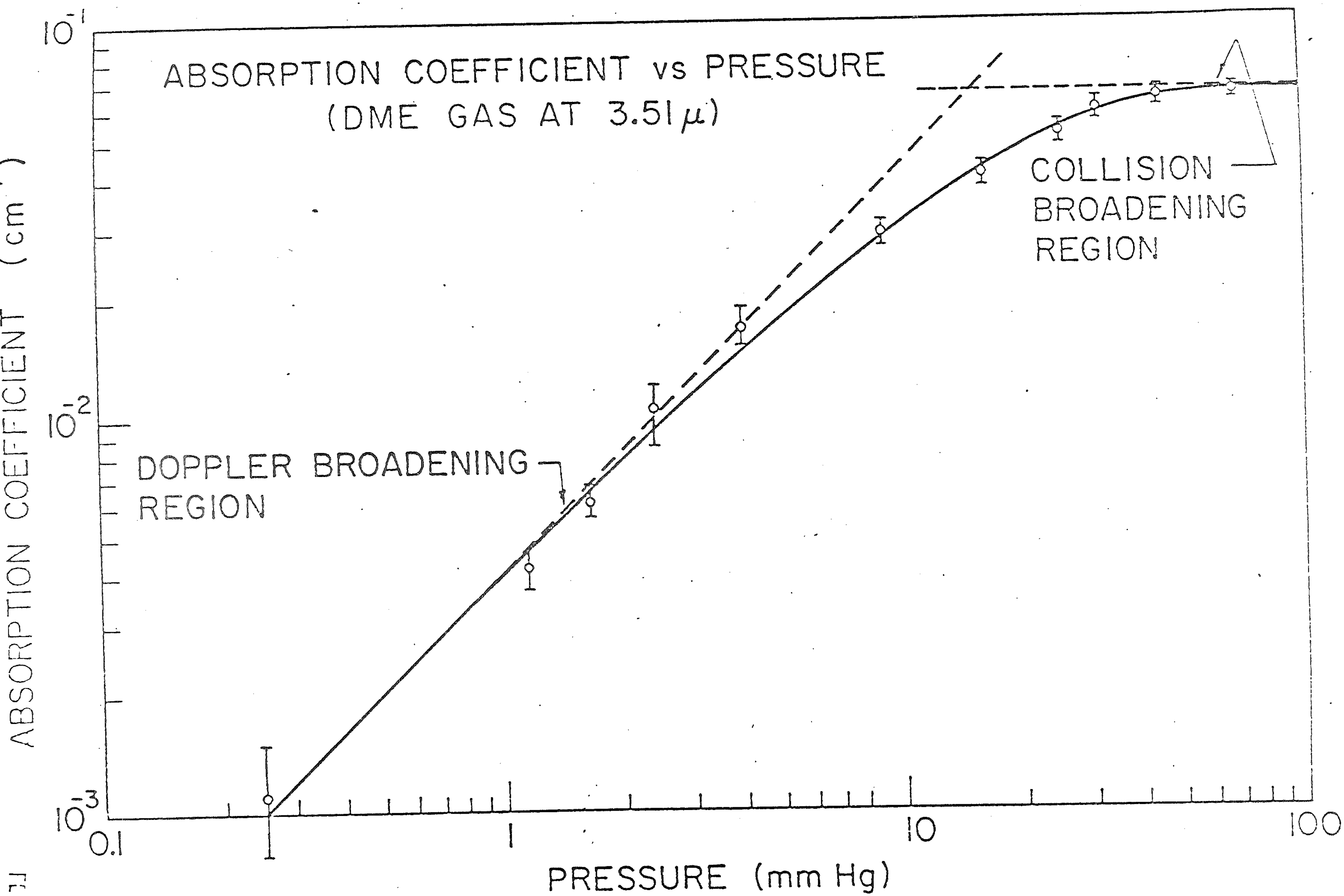

FIG. 1--Absorption coefficient of DME gas for 3.51μ He-Xe laser as a function of DME gas pressure at 293°K .

$4.0 \pm 0.2 \times 10^{-3}\ \mathrm{cm}^{-1}\ \mathrm{Torr}^{-1}$. The saturation intensity of the gas can be calculated using the definition,[10] $I_s = \epsilon_0 c \gamma_a \gamma_b \hbar^2/p^2$, where γ_a and γ_b are the relaxation rates of the upper and lower levels of the transition; c is the velocity of light in the vacuum; ϵ_0 is the permittivity of free space; and p is the electric dipole moment of the transition for DME, which is about 1.3 debye.[7] At a pressure of 1 Torr the fastest relaxation rate would be the collision rate, which is available from our data. If we set γ_a and γ_b equal to $3 \times 10^6\ \mathrm{sec}^{-1}$, the saturation intensity of DME is about 2 $\mathrm{mW/cm}^2$. This is a value obtainable from the laser we used. The results of saturable absorption of DME when placed inside the laser cavity will be reported in a separate paper.

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REFERENCES

1. P. H. Lee and M. L. Skolnich, Appl. Phys. Letters 10, 303 (1967).
2. R. L. Barger and J. L. Hall, Phys. Rev. Letters 22, 4 (1969).
3. G. R. Hanes and C. E. Dahlstrom, Appl. Phys. Letters 11, 362 (1969).
4. R. G. Brewer, M. J. Kelly, and A. Javan, Phys. Rev. Letters 23, 559 (1969).
5. S. S. Penner, Quantitative Molecular Spectroscopy and Gas Emission, (Addison-Wesley, 1959), Chapters 2 and 3.
6. E. T. Gerry and D. H. Leonnard, Appl. Phys. Letters 9, 227 (1966).
7. V. Blukis, P. H. Kasai, and R. J. Myers, J. Chem. Phys. 38, 2753 (1963).
8. H. A. Szymanski, Interpreted Infrared Spectra (Plenum Press, 1967), Vol. 3, p. 88.
9. H. A. Szymanski, Theory and Practice of Infrared Spectroscopy (Plenum Press, 1964), Chapter 5.
10. W. E. Lamb, Phys. Rev. 134 A1429 (1964).

APPENDIX C

OBSERVATION OF AN ENHANCED LAMB DIP WITH A PURE Xe GAIN CELL INSIDE A 3.51μ He-Xe LASER*

by

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ABSTRACT

The observation of an enhanced Lamb dip is reported for a pure Xe^{136} gain tube in a $He\text{-}Xe^{136}$ laser cavity. The pure Xe gain cell is operated at a pressure of 5 mTorr so that pressure broadening effects are negligible. The observed enhanced Lamb dip has a 5 MHz width and a depth which is 10% of the peak power. It is estimated that frequency stabilization at 3.51μ to one part in 10^{11} is possible using the enhanced Lamb dip as a reference.

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Observation of an inverted Lamb dip due to an absorbing cell inside the laser cavity has been reported previously,[1-3] and it has been suggested that the inverted dip may be used for frequency stabilization purposes. In previous experiments, the absorption gas was either an organic compound whose absorption peaks were near the laser frequency or a cell using the same atom as the laser, such as Ne in a He-Ne laser.

We report the first observation of an enhanced Lamb dip in a laser oscillator using an internal gain cell. In our case, the pure Xe tube serves to provide additional gain, instead of loss at the He-Xe 3.51μ transition. With this arrangement, the usual Lamb dip of the He-Xe laser, which operates at a 2 Torr pressure, is enhanced by the narrow Lamb dip of the pure Xe cell which is operated at a pressure of a few millitorr. Thus the Xe gain cell provides a narrow Lamb dip which can be used for a frequency marker while at the same time using the higher output power and gain of the He-Xe laser tube.

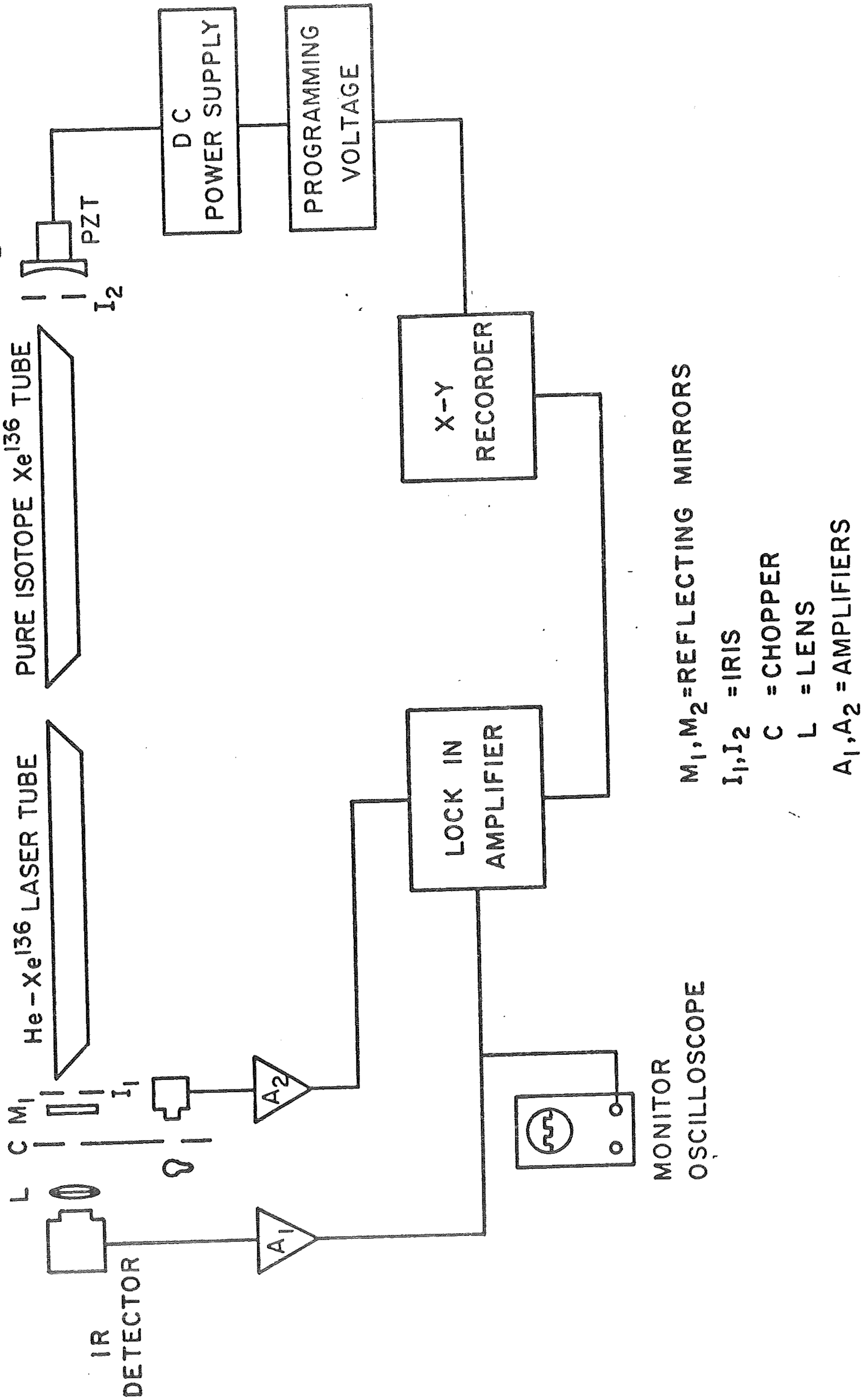
The He-Xe laser used in this experiment is a 20 cm long, 4 mm diameter discharge tube filled with 2 Torr of He and approximately

20 mTorr of single isotope Xe^{136}. The Xe gain cell is a 30 cm long, 6 mm diameter discharge tube filled with isotopic Xe^{136} to a pressure of about 5 mTorr. The Xe pressure in both tubes is controlled by a liquid nitrogen cooled pressure adjustment system. This allows the tubes to be operated over long lifetimes by providing a large reservoir of Xe to counteract the gas clean up problem.

Figure 1 is a block diagram of the experimental set-up. The cavity is 83 cm long with invar bar spacers. The air path of the laser beam inside the cavity is enclosed to avoid any fluctuations due to air motion. The output mirror is a flat with about 50% transmittance and the other mirror is a 400 cm radius of curvature high reflector. Both the He-Xe and the pure Xe tubes are designed for dc and rf excitation. In the present experiment the He-Xe tube was rf excited, while the pure Xe gain cell was both rf and dc excited to maintain a uniform discharge.

The doppler width of the He-Xe laser at 3.51μ is about 110 MHz. Thus the present cavity configuration provides operation on only a single longitudinal mode. Single transverse mode operation was obtained by the use of two apertures inside the laser cavity. A single isotope of Xe was used to eliminate the possibility of an isotope effect on the output power vs. frequency tuning of the laser.

The output power as a function of laser frequency was recorded in two steps. First the curve was taken with the Xe gain cell tube turned off to show the pressure broadened slightly asymmetrical Lamb dip. The curve was then taken with the gain cell discharge on but with the laser output power adjusted to the previous level to avoid power broadening effects. The results are shown in Fig. 2. The dotted portion of the



IR DETECTOR
L
C
M_1
I_1
A_1
A_2
$He-Xe^{136}$ LASER TUBE
PURE ISOTOPE Xe^{136} TUBE
I_2
M_2
PZT
D C POWER SUPPLY
PROGRAMMING VOLTAGE
X-Y RECORDER
LOCK IN AMPLIFIER
MONITOR OSCILLOSCOPE
M_1, M_2 = REFLECTING MIRRORS
I_1, I_2 = IRIS
C = CHOPPER
L = LENS
A_1, A_2 = AMPLIFIERS

FIGURE 1

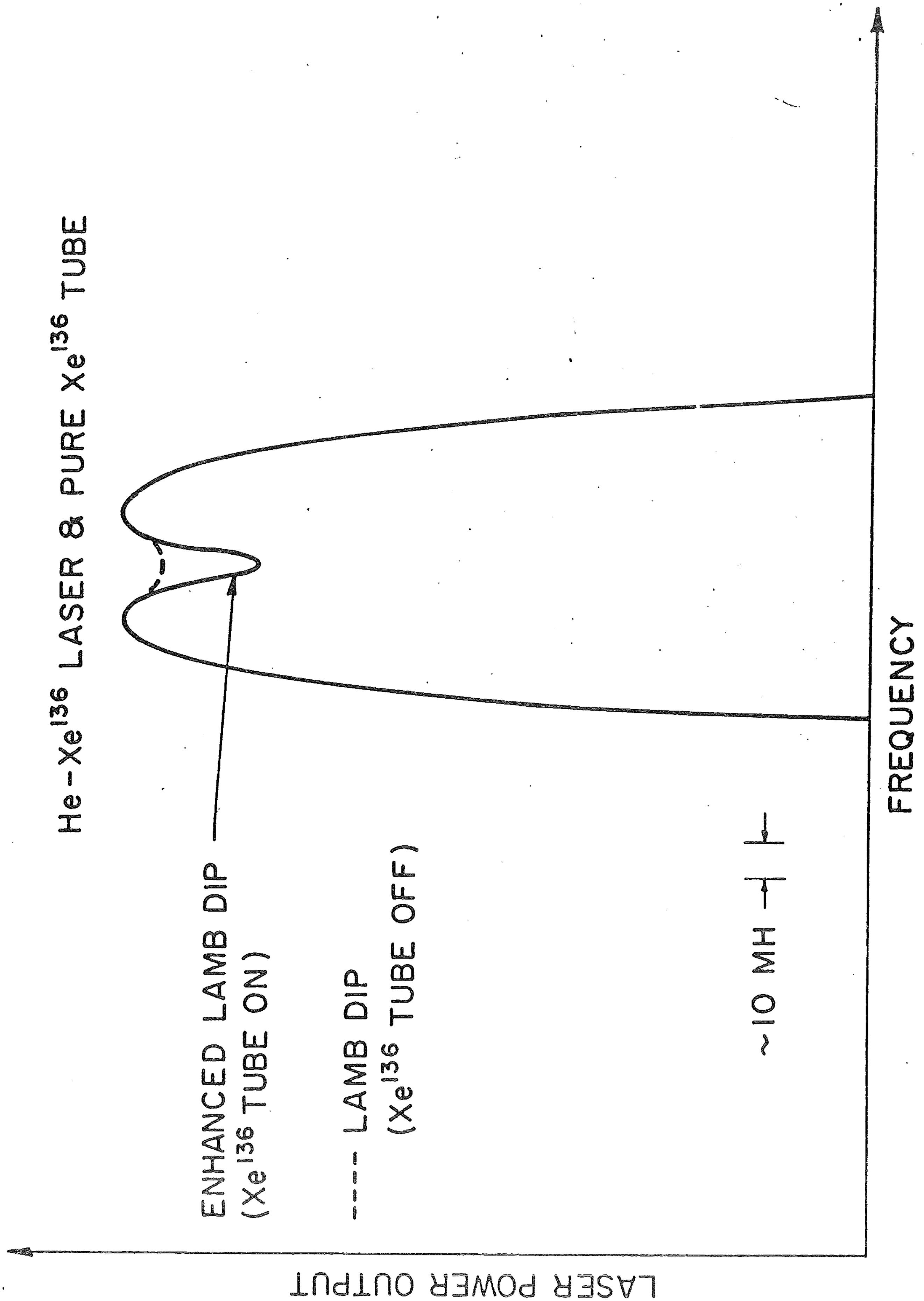

He-Xe^{136} LASER & PURE Xe^{136} TUBE
ENHANCED LAMB DIP
(Xe^{136} TUBE ON)
---- LAMB DIP
(Xe^{136} TUBE OFF)
~10 MH
FREQUENCY
LASER POWER OUTPUT

FIGURE 2

curve shows the He-Xe laser output only, and the solid curve shows the enhanced Lamb dip resulting from the Xe gain tube. The curves show that the enhanced dip is narrower than the laser Lamb dip due to the low pressure in the Xe gain cell, and that the enhanced dip is significantly deeper than the laser Lamb dip. For the present case, the width of the enhanced Lamb dip is 5 MHz and the total depth is about 10% of the peak power.

The intensity of the laser as a function of frequency with an additional gain cell or absorption cell inside its cavity can be derived from Lamb's theory[4] with slight modifications. If we neglect with asymmetry effects due to collisions[5] we find that

$$I(\Omega) = K_0 \frac{1 \pm K_1 - K_2 e^{(\Omega^2/\Delta\omega_D^2)}}{[1 + \gamma_1^2/(\gamma_1^2 + \Omega^2)] \pm K_3 [1 + \gamma_2^2/(\gamma_2^2 + \Omega^2)]} ,$$

where K_0 is a constant, and $\Omega = \omega - \omega_0$ is the detuning from line center. $K_1 = G_{20}/G_{10}$, $K_2 = R/G_{10}$, and $K_3 = (G_{20}/G_{10})(I_{s1}/I_{s2})$ are constants relating the unsaturated laser gain G_{10} , unsaturated gain or absorption of the gas cell G_{20} , the cavity loss R , the saturation intensities of the laser tube, and the gain or loss tube I_{s1} and I_{s2} , respectively. γ_1 and γ_2 are the linewidths of the laser and the gain cell and are related to the natural linewidth γ_n and to the collision linewidth γ_c of the transition by $\gamma = \gamma_n + \gamma_c$.

In this expression the numerator is the doppler broadened line shape function with a doppler linewidth $\Delta\omega_D$. The first term in the denominator is the usual laser Lamb dip while the second term is the enhanced Lamb dip or the inverted Lamb dip, depending on the sign. As can be seen from the expression, the depth of the enhanced Lamb dip depends upon both the relative gains of the two cells and upon their relative saturation parameters. In our present case the total He-Xe laser tube pressure was about 2 Torr, while the pure Xe^{136} tube pressure was 5 mTorr . For these pressures and the excitation conditions we expect that $\gamma_1 \gg \gamma_2$ and that $I_{s1} \gg I_{s2}$. With proper excitation conditions to adjust the relative gain values we, therefore, expect to observe a narrowed enhanced Lamb dip due to the pure Xe gain cell.

The nature linewidth of the Xe atom at 3.51μ is about 2 MHz which can be calculated from the known lifetimes of the upper and lower levels which are 1.36×10^{-6} sec and 4.5×10^{-8} sec , respectively.[6] The frequency broadening due to collisions is negligible compared to the radiative lifetime in the pure Xe tube. Thus, the enhanced Lamb dip should have a width of twice the natural linewidth.[7] With possible power broadening of the enhanced Lamb dip taken into consideration, the observed width of 5 MHz is in good agreement with the theoretical prediction.

By using the $He\text{-}Xe^{136}$ laser to provide gain and power, and the low pressure pure Xe^{136} tube to provide a narrow frequency discriminant, our present combination scheme should be very useful as a frequency standard in the near infrared. With appropriate feedback control techniques, the frequency of the laser can be stabilized to better than one part in a thousand of the enhanced Lamb dip of 5 MHz , and we thus estimate that for the 3.51μ Xenon laser frequency stabilization to one part in 10^{11} is possible. Furthermore, this technique could easily be extended to other lasers for which low pressure narrow linewidth gain tubes can be constructed.

We wish to thank K. R. Manes and D. J. Kuizenga for helpful suggestions during the course of this work.

REFERENCES

1. P. H. Lee and M. L. Skolinch, Appl. Phys. Letters 10, 303 (1967).
2. V. N. Lisitsyn and V. P. Chebotayev, Soviet Phys. JEPT 27, 227 (1968).
3. R. L. Barger and J. L. Hall, Phys. Rev. Letters 22, 4 (1969).
4. W. E. Lamb, Jr., Phys. Rev. 134, A1429 (1964).
5. A. Szoke and A. Javan, Phys. Rev. 145, 137 (1966).
6. L. Allen, D. G. C. Jones, and D. G. Schofield, J. Opt. Soc. Amer. 59, 842 (1969).
7. W. R. Bennett, Jr., Quantum Electronic Paris 1963, edited by P. Grivet and N. Bloembergen (Columbia University Press, 1964), p. 441.

FIGURE CAPTIONS

1. A schematic diagram of the enhanced Lamb dip He-Xe^{136} laser with the pure Xe^{136} gain cell. The fill pressure of the He-Xe^{136} laser tube is 2 Torr and the pressure of the Xe^{136} gain cell is about 5 mTorr.

2. The power output vs. frequency tuning curve (solid) show the enhanced Lamb dip observed in He-Xe^{136} laser oscillating at 3.51μ with an excited pure Xe^{136} gain cell inside the cavity. The dotted portion, recorded with Xe^{136} gain cell turned off, was superposed. Only the upper half of the whole power tuning curve is shown here.

APPENDIX D

PUMP LINEWIDTH REQUIREMENT FOR OPTICAL PARAMETRIC OSCILLATORS*

by

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ABSTRACT

A pumping laser at 0.473μ having a bandwidth of about 4 cm^{-1} has been used to construct a parametric oscillator having a bandwidth of about 1/4 cm^{-1} and tunable from about 2.45μ to 3.2μ . It is shown that for such an oscillator there is a significant advantage in resonating the I-R wave instead of the visible wave.

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It has been shown that singly resonant optical parametric oscillators have far greater spectral stability then the previously constructed doubly resonant oscillators.[1,2,3,4] The present Letter shows that, in addition, such singly resonant oscillators may be constructed using a relatively broad band multi-mode pumping laser, and still yield a narrow band spectral output. In particular a 4 cm^{-1} wide pump at 0.473μ has been used to construct a parametric oscillator having a 1/4 cm^{-1} wide spectral output tunable from 2.45μ to 3.2μ .

We consider the case where the signal frequency is fixed by the optical resonator and interacts with a broad band pump to generate a non-resonant broad band idler. The allowable pump bandwidth such that all modes of the pumping laser act in unison to produce gain is determined by the allowable momentum mismatch $\Delta kL \cong \pi$, where L is the length of the nonlinear crystal. For this case, the momentum mismatch Δk due to a pump bandwidth $\Delta\omega_p$ is given by

$$\Delta k = \left(\frac{\partial k_p}{\partial \omega_p} - \frac{\partial k_i}{\partial \omega_i} \right) \Delta\omega_p \tag{1}$$

and thus the allowable pump bandwidth is approximately

$$\Delta\omega_p = \frac{\pi}{L} \frac{1}{\frac{\partial k_p}{\partial \omega_p} - \frac{\partial k_i}{\partial \omega_i}} \tag{2}$$

As a result of normal dispersion $\Delta\omega_p$ is significantly larger if the free or non-resonated frequency is the frequency nearest to the pump. For the case of $LiNbO_3$ with a pump at 0.473μ , the signal and idler frequencies at 0.569μ and 2.79μ , respectively, evaluation of Eq. (2) using the Sellmeier equations for $LiNbO_3$,[5] indicates that the allowable pump bandwidth is about six times larger if the I-R frequency is resonated as it is if the visible frequency is resonated.

For our 3.2 cm $LiNbO_3$ crystal the allowable pump bandwidths are $\Delta\omega_p$(visible resonant) = 0.98 cm^{-1} , while $\Delta\omega_p$(I-R resonant) = 5.8 cm^{-1} . The spectral envelope of the doubled 0.946 line of our Nd:YAG pumping laser is about 4 cm^{-1} . Thus all the pump power should be effective in driving an oscillator which resonates the I-R wave, while only about one-fourth of the pump spectral power would be available to an oscillator resonating the visible wave.

A singly resonant parametric oscillator which resonated the I-R wave was successfully built. Figure 1 shows the spectral envelopes of the pump, non-resonant visible wave, and the resonant I-R wave as measured by a 1 meter

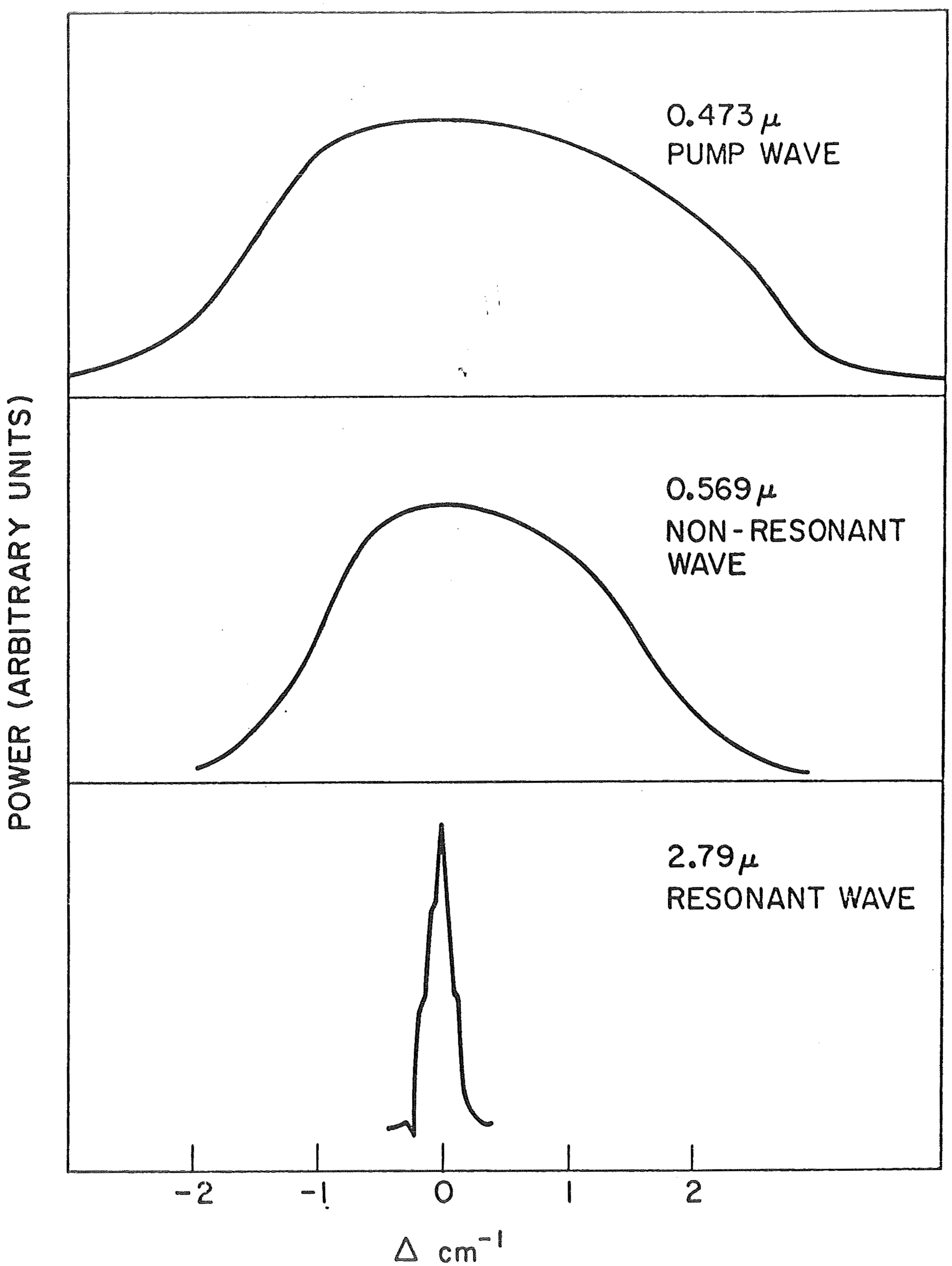

FIG. 1--Spectra of pump, non-resonant wave, and resonant wave.

Spex spectrometer having a resolution of about 0.4 cm^{-1} in the visible, and 0.1 cm^{-1} in the I-R . The spectrometer scan rate and the oscillator pulse rate were adjusted so that about 50 pulses were averaged within each resolvable frequency interval. As expected, the free idler has picked up the width of the pump while the resonated idler remains quite narrow. In general, the oscillator operated very stably with excellent pulse-to-pulse reproducibility, and a conversion efficiency of pump to tunable radiation of about 50% .[6]

An attempt to construct an oscillator which resonated the visible wave was unsuccessful until an internal etalon was used to narrow the laser spectrum. Even then the oscillator was only marginally above threshold and operated erratically at low power.

This work has thus shown that when constructing a singly resonant optical parametric oscillator with a relatively broad band pump, a significant advantage can be obtained by choosing the resonated wave to be that which is furthest in frequency from the pump. By using an etalon[2] inside the parametric oscillator cavity, still narrower I-R outputs will be obtainable; and will again utilize the full power of the relatively wide band pumping laser.

The authors thank R. L. Byer and R. C. Rempel for a number of helpful discussions.

REFERENCES

1. J. E. Bjorkholm, "Some Spectral Properties of Doubly and Singly Resonant Optical Parametric Oscillators," Appl. Phys. Letters 13, 399 (December 1968).
2. L. B. Kreuzer, "Single Mode Oscillation of a Pulsed Singly Resonant Optical Parametric Oscillator," Appl. Phys. Letters 15, 263 (October 1969).
3. Y. N. Belyaev, A. M. Kiselev, and J. R. Freidman, "Investigation of Parametric Generator with Feedback in Only One of the Waves," J. Expt. Theoret. Phys. Letters 9, 263 (April 1969).
4. S. E. Harris, "Tunable Optical Parametric Oscillators," Proc. IEEE 57, 2096 (December 1969).
5. M. V. Hobden and J. Warner, "The Temperature Dependence of the Refractive Indices of Pure Lithium Niobate," Phys. Letters 22, 243 (August 1966).
6. R. W. Wallace (to be published).