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RESEARCH ON PROPERTIES OF ASER DEVICES (U)

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RESEARCH ON PROPERTIES OF EASER DEVICES (U)

TRG, Incorporated Syosset, New York

June 10, 1961

Prepared for OFFICE OF SCIENTIFIC RESEARCH AIR RESEARCH AND DEVELOPMENT COMMAND UNITED STATES AIR FORCE Washington, D. C.

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ABSTRACT

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Experiments were performed to detect LASER oscillations at 3u and at 7μ from <u>cesium vapor optically pumped</u> with light from helium discharge lamps. No oscillations were observed. Fluorescence measurements have led to the construction of an improved system. Work was carried out on several possible LASER media utilizing <u>collisions of the</u> <u>second kind</u> in a gas discharge. Studies of the spacial and temporal behavior of the <u>ruby (R line) LASER</u> was investigated and crystal boules of calcium tungstate containing various concentrations of <u>rare-earth</u> ions were grown. Cscillation was attempted, unsuccessfully in each of these schemes. Studies of crystal perfection indicate that the internal quality of these crystals is not yet adequate. Two <u>new forms of the</u> <u>Fabry-Perot interferometer</u> have been constructed, which are superior in adjustment tolerances. Several aspects of the <u>optical behavior of</u> <u>Fabry-Perot interferometers</u> which are of importance for the understanding of F.P. LASERs have been studied theoretically.

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P. Rabinowitz and R. Targ.

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I. Introduction and Summary of Results

The immediate objective of this research is to produce working models of LASER devices, together with the experimental and theoretical information necessary to describe and predict their performance.

In this section we summarize the results that have been obtained to date.

1.0 Optical Pumping of Cesium Vapor by Helium Lamp

In the last report we described the apparatus constructed to look for 3μ and 7μ oscillations from cesium vapor in a Fabry-Perot cavity. Optical pumping was tried with modulated r-f helium discharge lamps and also with the much brighter helium flashlamps. No oscillations were observed.

We concluded that the losses must be higher than anticipated due to turbulence in the air path caused by thermal gradients and inhomogeneities in the sapphire and silicon windows.

In the present period we repeated the earlier Cs fluorescence measurements, extending them to include the 3.2μ (proposed IASER) line as well as other infrared lines, and re-calculated the gain expected from the above results. Reassured by these measurements and calculations we began construction of a modified system with the entire cavity in vacuum (eliminating air path) and a 10-fold increase in IASER length (increasing gain). The increased length also facilitates tuning of the cavity.

2.0 Direct Discharge Excitation of LASER Media

Work was carried out on several possible LASER media utilizing collisions of the second kind in a gas discharge.

Six different gas discharge processes were analyzed theoretically (an asterisk indicates an excited atom):

a) Kr^{*} (meta) + Hg \Rightarrow Kr + Hg b) Kr^{*} (meta) + Xe \Rightarrow Kr + Xe c) Hg^{*} + Zn \Rightarrow Hg + Zn d) He^{4*} + $He^{3} \Rightarrow$ He^{4} + He^{3*} e) Rb^{*} + $Rb^{*} \Rightarrow$ Rb + Rb^{**} f) Zn^{*} (meta) + Zn^{*} (meta) \Rightarrow Zn + Zn^{**}

For process (a), a gain coefficient, $\alpha_{G^{2}} 1\%$ per meter at $\lambda = 1.8\mu$ was computed from measured fluorescent intensities.

Variations in populations of interesting states were measured as a function of power input and pressures of the following gases:

- a) Krypton-mercury mixtures
- b) Krypton-xenon mixtures
- c) Pure krypton

The population changes were observed in spectral absorption and emission.

3.0 Optical Pumping of Ruby (R line)

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Studies of the spacial and temporal behavior of the mode characteristics of the ruby (R line) LASER have been carried out. Work has been initiated toward obtaining a relative figure of merit of the optical quality of ruby crystals.

4.0 Optical Pumping of Ruby (Neighbor Lines)

The "neighbor line" spectrum of dark ruby war investigated to determine the feasibility of using exchange displaced levels in a low power pulsed or C.W. LASER. The energy level diagram was found to be exceedingly complex but nonetheless appeared to fulfill the requirements for excitation of oscillation. An experiment was conducted in which the dark ruby sample, cooled to 2°K and pulse-excited, was observed to emit fluorescent radiation of sufficient spectral intensity to cause oscillation. Oscillation was not produced, however, because of the poor crystal quality of the samples and the resultant high scattering losses which it causes.

5.0 Optical Pumping of Rare Eciths

Single crystal boules of calcium tungstate containing various concentrations of trivalent europium, terbium, dysprosium, and praseodymium were grown. The crystals containing Pr have not been evaluated. Difficulties were encountered in obtaining boules containing Tu and Tb with the high emission intensity characteristic of flux grown crystallites of the same material. Crystals containing Dy however grow well and fluoresce strongly.

Some of the latter crystals were ground and polished to LASER tolerances and oscillation attempted on a pulsed basis. While the brightness of the output was more than adequate for LASER oscillation, no oscillation was observed. Studies of crystal perfection indicate that the internal quality of these crystals is not yet adequate.

A solution containing Europium (Eu:TTA) was evaluated as the active media of a LASER oscillator. The engineering difficulties with this system were found to be very great. Work on this solution, though still promising, was dropped in view of the encouraging results with the tungstate crystals.

Some effort on other host media than the tungstates was continued and has, as yet, not given positive results.

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6.9 Optical Measurements and Devices

Two new forms of the Fabry-Perot interferometer have been constructed which are far superior to the F.P. in adjustment tolerances. Haidinger fringes have been observed in both the retroreflective cornerreflector-vs-flat and crossed-roof interferometers.

A relatively simple optical system has been devised to indicate the optical quality of crystals to be used for LASER operation.

A method has been devised to measure the orientation of the c axis of uniaxial crystal boules with irregular sides.

7.0 Theory of Resonator Modes

Several aspects of the optical behavior of Fabry-Perot interferometers of importance for the understanding of F.P. LASERs have been studied. Firstly we have investigated the nature of the modes of a Fabry-Perot cavity in terms of the general theory of leaky wave or radioactive state resonances of open electromagnetic structures. Secondly we have derived an exact theory of the F.P. cavity and shown how this reduces in an appropriate approximation to the previously reported theory. Thirdly, we have studied the optical mode structure of a F.P. interferometer via the variational principle described in earlier reports. This has led to analytic expressions for the quantities of practical interest, e.g. the mode shpaes, resonant frequencies, and diffraction losses.

II. Optical Pumping of Cesium by Helium Lamp

1.0 Summary of Work Performed

In the last report we described the apparatus constructed to look for 3μ and 7μ oscillations from cesium vapor in a Fabry-Perot cavity. Optical pumping was tried with modulated r-f helium discharge lamps and also with the much brighter helium flashlamps. No oscillations were observed.

We concluded that the losses must be higher than anticipated due to turbulence in the air path caused by thermal gradients and inhomogeneities in the sapphire and silicon windows.

In the present period we repeated the earlier Cs fluorescence measurements, extending them to include the 3.2μ (proposed LASER) line as well as other infrared lines, and re-calculated the gain expected from the above results. Reassured by these measurements and calculations we began construction of a modified system with the entire cavity in vacuum (eliminating air path) and a 10-fold increase in LASER length (increasing gain). The increased length also facilitates tuning of the cavity. 2.0 Analysis of Work Performed

2.1 Cs/He Fluorescence

Figure 2.1 shows the experimental arrangement used for the fluorescence measurement. The Cs cell had sapphire windows. The radiation

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7. EXPERIMENTAL ARRANGEMENT FOR FLUDRESCENCE MEASUREMENT He LAMP FOCUSING CAUTY CS CELL Stoblaaten pi 1/2-CS TEMP Figure 2.1 Humming Annumule. -SPECIPOMETER (munut)

detectors were a 6911 phototube and PbS. The spectrometer was a Perkin-Elmer Model 98 grating instrument. Figure 2.2 shows the Cs cell inside a transparent heating tube, located at one focus of glass cylindrical cavity. A water-cooled helium lamp is at the other focus.

Figures 2.3 and 2.4 show the measured fluorescence intensities of 26 lines, and their variation with Cs temperature. Analysis of these measurements shows that (see Figure 2.5)

1. The $8P_{1/2}$ population is greatest at 175°C for 4 mm diameter tube (2.7 x 10^8 atoms/cm³).

2. The populations of the Cs 4F, 5D and 7D states, increase with temperature far beyond amount predicted for radiative processes. Thermally induced collisions must be responsible *.

3. The gain coefficient, given by the expression

$$K_{0} = \frac{ln2}{16\pi^{3}c^{2}} = A_{21} \lambda^{3} \frac{\lambda}{\Delta\lambda} = (N_{2} - \frac{g_{2}}{g_{1}} - N_{1}),$$

is equal to 5.8 x 10^{-3} cm⁻¹ for the $\lambda = 3.2\mu$ transition and is equal to 3.8 x 10^{-2} cm⁻¹ for the $\lambda = 7.18\mu$ transition. For L = 9 cm; $\lambda = 7.18\mu$, as in the previous LASER experiments,

$$\frac{I}{I_{o}} = e^{k_{o}L} = e^{.34} = 1.40, \text{ or } 40\% \text{ gain.}$$

* A detailed discussion of these results was presented at the Spring - Meeting of the Optical Society.



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Losses due to silicon windows, tuning diagonal and reflectors amounted to less than 20% per pass. Apparently, immerfect optical flatness, thermal turbulence and other inhomogeneities amounted to more than 20%.

2.2 Long Tube System

a) Lamp

A 90 cm long r-f helium discharge tube has been constructed and operated at saturation intensity. Two transmitters supply a total of 800 watts at 30 mgc. The lamp is operated in a water jacket.

b) Pumping Cavity

In order to avoid the difficulty of making a meter-long cylindrical ellipse we constructed and evaluated a diffuse cavity to contain both lamp and LASER. The diffuse reflecting walls consist of MgO powder behind transparent vycor. Figure 2.4 shows construction, which includes heater coils to prevent Cs condensation when LASER cell is present.

Performance of the diffuse cavit" was evaluated by spacially scanning the interior thru a small hole. A Hilger f/10 monochromator was set at 3888A+ and three scans were made (see Figure 2.7). The measured cavity intensity was given by the relation

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 $J_{cavity} = 36\% J_{lamp}$



Figure 2.6





CS LASER CELL-DYNAMIC PUMPING SYSTEM



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From this result we calculate that the effective reflectivity of this cavity is about 88%. When the IASER cell is introduced into the cavity, J_c will drop to the value given by the expression

J_{cavity} = 31% J_{lamp}

provided the nichrome windings are concealed.

c) LASER Cell

Due to the large size of the Cs cell (1 cm I.D. and 90 cm long) absorption of cesium by walls becomes important along with outgassing. Therefore a dynamic system is being built. Cesium vapor migrates from a hot pot through an oven to a cold trap. Provision is made to reverse the flow when the reservoir is empty. See Figure 2.8

d) Interferometer

Tuning of the 1 meter interferometer will be nearly automatic in the 3.2μ case, where cavity modes lie so close together that they always overlap Cs Doppler width (Figure 2.9).

A new type of interferometer with much improved mechanical stability has been developed at TRG for use with LASERs. This is a crossed roof prism interferometer and is discussed in Section VII -Optical Mernurements and Devices. At the time of this writing, we have not yet found a suitable prism material to adapt the crossed roof

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int ferometer to use with the 7μ LASER. NaCl and BaF₂ both fail the heat and cesium test. We are considering MgO for 3.2μ LASER prisms. Because of this material problem we may have to use one of the unattacked metallic reflectors, such as chromium or iron. These reflect fairly well at 7μ , but their visible reflectivity is so low as to make impossible al ament of a roof prism interferometer, (with 8 reflections between successive emergent beam). The plane Fabry-Perot is however, still suitable.

3.0 Work Planned for Next Period

We are nearing completion of 1 cm diameter Cs/He LASER tube. We expect a gain coefficient $K_0 \approx .02$ cm⁻¹ for $\lambda = 7\mu$. This corresponds to a gain of 600% in one pass of 90 cm length.

During the next period we expect to measure the gain at 3.2μ directly and, if the result is positive, begin construction of a meter-long 7μ LASER.

Note that K₀ is smaller than for the 4 mm tube mentioned earlier.
This is because in a 1 cm diameter tube the pumping light absorption coefficient must be reduced to insure uniform pumping. This means LASER will be operated at a lower temperature and 8P_{1/2} population will be reduced.

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III. Direct Discharge Excitation of LASER Media

1.0 Summary of Work Performed

Work was carried out on several possible LASER media utilizing collisions of the second kind in a gas discharge.

1.1 Theoretical

Six different gas discharge processes were analyzed theoretically (an asterisk indicates an excited atom):

a) $Kr^{*}(meta) + Hg \rightarrow Kr + Hg^{*}$ b) $Kr^{*}(meta) + Xe \rightarrow Kr + Xe^{*}$ c) $Hg^{*} + Zn \rightarrow Hg + Zn^{*}$ d) $He^{4*} + He^{3} \rightarrow He^{4} + He^{3*}$ e) $Rb^{*} + Rb^{*} \rightarrow Rb + Rb^{**}$ f) $Zn^{*}(meta) + Zn^{*}(meta) \rightarrow Zn + Zn^{**}$

For process (a), a gain coefficient, $\alpha = 1.8 \mu$ Was computed from measured fluorescent intensities.

1.2 Experimental

Variations in populations of interesting states were measured as a function of power input and pressures of the following gases:

- a) Krypton-mercury mixtures
- b) Krypton-xenon mixtures
- c) Pure krypton

The population changes were observed in spectral absorption and emission.

2.0 Analysis of Work Performed

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a) Study of the krypton-mercury system was confined to gain computations from the latest measured fluorescent intensities. As indicated in Figure 31 selective excitation of Hg (9¹P₁) results from collisions of Hg (6¹S₀) with Kr (1s₅ - metastable) in a discharge. The relative intensities of the lines, $\lambda = 6234$ and $\lambda = 4916$, together with the calculated emission rates, enables one to compute the substate ppulation density ratio $\frac{n}{g}$ (9¹P₁) $/\frac{n}{g}$ (8¹S₀) within 10%. From measurements made in January, 1961, $\frac{n}{g}$ (9¹P₁) $/\frac{n}{g}$ (8¹S₀) 1.5. The absolute value of the excess population is $\frac{n}{g}$ (9¹P₁) $-\frac{n}{g}$ (8¹S₀) 3.3 x 10⁶ cm⁻³. The computed gain for a light wave inducing transitions from Hg (9¹P₁) to Hg (8¹S₀) is $\alpha_{G} = (1.0 \pm 0.5)\%$ per meter. This is marginally sufficient to generate oscillations in a 1 meter IASER at $\lambda = 1.8\mu$.

Figure 32 shows the population ratio per quantum state of the Hg (9^1P_1) and Hg (8^1S_0) levels as a function of Kr pressure. These population ratios were computed from fluorescent intensities measured through an f/10 Ebert grating monochromator employing a lead sulphide detector calibrated by a National Bureau of Standards tungsten lamp. During these runs, the mercury pressure was not well known. The pressure could not be less than the vapor pressure corresponding to the well temperature. However, mercury is driven into the walls by





(1) the discharge . Then the heating of the tube causes mercury outgassing and a non-equilibrium excess pressure. Thus we may be sure that the measured enhancement is not optimum.

POPULATION RATIO

I.D

O P

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KRYPTON

PRESSURE

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DISCHARGE

TUBE

In order to maintain a known mercury pressure, an allquartz, oven-heated and temperature-controlled system would be required. Before building this new apparatus, it was decided to examine a krypton-xenon gas mixture, which presumably would not present the same uncertainty in pressures.

b) The important krypton and xenon energy levels are shown in Figure 3.3. Adequately close coincidences occur between both the upper (1s₃) and lower (1s₅) krypton metastable levels, and several appropriate xenon levels.

Fluorescent intensities have been measured for the lines $\lambda = 8952A$, 9162A, 9923A, which originate on Xe $2p_6$, $2p_7$ and $2p_9$ levels as well as for the "control" lines $\lambda = 8576A$ and 7967A, which originate on the xenon $3p_5$ and $3p_7$ levels. Enhancements in intensity as large as (4.5%) were observed for certain transitions in Kr-Xe as compared with a pure menon discharge at a nominal power input density of 0.3 watt/cm³ (see Table 3.1).

A comparison was made of the rentive intensities of the five xenon lines in a discharge containing pure xenon at 30± to the [1] This refractory phenomenon has been investigated by U. F. Meggers and F. O. westfall in "Lamps and Wavelengths of Mercury 193" Journal of Research of The National Bureau of Standards 44, 447 (1950) TECHNICAL RESEARCH GROUP



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TABLE	3.1
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	Enhancement in of the Xenon 2 Collisions of Atoms Excited	n a Xenon-Krypton 2p - 1s Transitio the Second Kind to the Metastabl	Discharge ons by with Krypton e ls ₅ State
Enhanced Xe Transition	Wavelength A	ΔΕ	$\frac{I}{Xe + Kr}$
$^{2p}_{6} - 1s_{4}$	8952	-3.1 kT	75%
$2p_7 - 1s_4$	9162	-4.2 kT	61%
2p ₉ - 1s ₄	9923	-7.6 kľ	45%
Control Xe Transitions			
$^{3p}5 - 1s_2$	8576	+40.0 kT	167.
$3p_7 - 1s_2$	7967	+35.9 kT	187.

where ΔE is the resonance energy defect of the xenon levels from the krypton level

$$\Delta E = [E_{Xe 2p} - E_{Kr 1s_5}]$$

where I Xe+Kr

I_{Xe}

is the ratio of the relative intensities of emission of the five xenon lines in a discharge containing 10μ of Xe in 100μ of Kr to a discharge containing 100μ of Xe.
ABSORPTION OF SPECTRAL LINES BY EXCITATION FROM VARIOUS KRYPTON LEVELS (PROP. TO LEVEL POPULATION)



discharge containing pure xenon at 100μ . The intensities of the three "enhanced" xenon lines were reduced by an average of 75% of the original intensity, those of the two control xenon lines by an average of 75%.

Preliminary investigations indicate enhancement of the xenon $2s_5 \cdot 2p$ infrared transitions, resulting presumably from the excellent energy coincidence for collisions of the second kind between the metastable krypton is, and the zenon $2s_5$ levels.

In order to optimize the krypton pressure to first order, the populations of the krypton levels were observed in absorption. The populations of Kr ls_2 , ls_3 (meta), ls_4 and ls_5 (meta) levels were found to be maximum at $P_{Kr} = 0.1$ mm Hg (see Figure 3.4).

In the Hg - Kr experiment, one may recall that the Kr states that pump the Hg 9^{1} P and 10^{3} P states were observed to be depopulated by the addition of Hg to the pure Kr discharge. Similarly in the Xe - Kr experiment the addition of Xe to the Kr discharge is seen to reduce the population of those Kr metastables that pump the "enhanced" Xe states.

The absorption measurements, by which the krypton metastable populations tore monitored, were made on the double beam apparatus shown in Figure 3.5.



c) A mercury-zinc gas mixture was reported to exhibit a large gain (a 2 cm⁻¹) in a Soviet publication.^[2] A difference in energy of only 133 cm⁻¹ $\frac{1}{2}$ kT exists between the Hg (7³S₁) and Zn (4¹D₂) levels. Thus the Zn (4¹D₂) \Rightarrow Zn (4¹P₁) transition at $\lambda = 6362$ A was presumably enhanced by collisions of the second kind.

However, Hg $(7^{3}S_{1})$ is neither trapped nor metastable. Kenty has shown that the population of this state is very low in a discharge^[3]. Furthermore, the rate of radiative decay of the lower LASER state -Zn $(4^{1}P_{1})$ - will be reduced, by trapping of radiation for zinc densities greater than $n = 3 \times 10^{11}$ atoms/cm³. This density is only 10 times larger than that supposedly observed in the excited Zn $(4^{1}D_{2})$ state. Thus calculations and previous work cast grave doubts on the report.

A consultant to TRG, Dr. Patrick Thaddeus, visited the Lebedev Institute just outside of Moscow in February 1961. He reports that the experiment was crudely carried out and that zinc fluorescence was probably confused with amplification. G. Gould, of TRG, discussed the experiment briefly at the March, 1961 Quantum Electronics Conference in Berkeley with N. G. Basov, Assistant Director of the Lebedev Institute. Dr. Basov stated categorically that gain due to coherent induced emission was not cbserved.

- [2] V. K. Ablekov, M. S. Pesin, and I. Fabelinski, Soviet Physics, JETP, (USSR) <u>39</u>, 892 (1960)
- [3] C. Kenty J. Appl. Phys. <u>21</u>, 1309 (1950)

d) A pure helium discharge as a possible LASER medium was investigated at TRG both theoretically and experimentally between July and December 1959. The spontaneous emission rate, $A(2^{1}P_{1} \rightarrow 1^{1}S_{0})=2.3 \times 10^{9}$ sec⁻¹ is so much larger than the rate $A(3^{1}S_{0} \rightarrow 2^{1}P_{1}) = 5.4 \times 10^{7}$, that an inversion in the populations of $(3^{1}S_{0}$ and $2^{1}P_{1})$ might occur under certain discharge conditions (see Figure 3.6).

Calculations on the trapping of the resonance radiation were made using the theory of Holstein^[4]. These indicated that the pressure of helium could not be greater than $P_{He} = 0.1\mu$ at which pressure the populations would be too small to give appreciable gain.

Experiments showed no inversions in several pairs of levels at somewhat higher pressures. This result was confirmed by the work of Sanders at Bell Laboratories.^[5]

In February a possible improvement over pure helium gas was conceived and analyzed at some length. The isotope shift of the resonance line, He $(2^{1}P_{1}) \Rightarrow$ He $(1^{1}S_{0})$ is large enough to allow He³ radiation to escape without reabsorption by He⁴. Thus if He³ were at P_{He}³ 0.1µ, an inversion might be possible by collisions of the second kind between the two isotopes.

[4] T. Holstein, Phys. Rev. <u>83</u>, 1159 (1951)
[5] J. H. Sanders, Nature <u>183</u>, 312 (1959)

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HELIUM SINGLET ENERGY LEYELS



Figure 3.6

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Further calculations revealed this difficulty: the wing of the He⁴ $(3^{1}S_{0}) \rightarrow He^{4} (2^{1}P_{1})$ transition slightly overlaps the corresponding (IASER) transition in He³. Since no inversion would exist in the denser He⁴, the LASER light would be absorbed (a 1 cm⁻¹) too strengly to permit a net gain. For this reason, no additional experimental work was done with helium.

e) Zn atoms may be selectively excited by the following collision process in a discharge:

$$Zn (4^{3}P_{2}-meta) + Zn (4^{3}P_{2} - meta) + Zn (8^{3}S_{1}) + Zn (4^{1}S_{0}-ground).$$

The energy defect is less than $\frac{1}{2}$ kT. This mechanism has been observed in mercury vapor, but a multiplicity of upper states reduces the efficiency. Assuming a reasonable cross-section for the process (σ 30A²), the upper state population should be greater than n = 10⁸ cm⁻³. Several LASER transitions are possible with a gain α 0.50 meter⁻¹

A similar process should occur in rubidium. The first excited state (5^2P) is severely trapped, giving rise to lifetimes greater than 10^{-4} second (observed in the sister element, sodium).

A single component system would relieve some of the experimental difficulties encountered in maintaining adequate partial pressure control in two-component gas discharges.

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3.0 Work Planned for Next Period

I).

a) The presence or absence of a population inversion in a Kr-Xe mixture will be established.

b) With the recent improvement in lead sulphide infrared detectors, a gain measurement has become possible at $\lambda = 1.8\mu$ in the Kr-Hg system. This may be attempted in an improved, temperature controlled cell.

c) A possible enhancement in a pure zine discharge will be examined.

d) A He- Ne LASER will be fabricated, using the self-adjusting prism cavity recently reduced to practice at TRG. (see Section VII).

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IV. Optical Pumping of Ruby (R line)

1.0 Summary of Work Performed

Studies of the spacial and temporal behavior of the mode characteristics of the ruby (R line) LASER have been carried out. Work has been initiated toward obtaining a relative figure of merit of the optical quality of ruby crystals.

2.0 Analysis of Work Performed

The ruby LASER was made to oscillate approximately one year ago. Some of the characteristics were given in the report of January 9, 1961. At that time there was some indication of a Fabry-Perot mode pattern in the output beam and a suspicion that the beam divergence of 15 to 45 minutes of arc was determined by the mode spectrum rather than by diffraction.

A study of the mode characteristics of the LASER has been carried out and shows that the off-axis Fabry-Perot modes play an important part in determining the beam divergence.

A study of the variation of the mode characteristics during the pulse was carried out using a 16 mm high speed streak camera. Pictures obtained showed that:

- a) The area of the filamentary region increased during the pulse.
- b) Two separate oscillating regions appeared to undergo relaxa tion oscillations together.

c) When Fabry-Perot type ring patterns were observed, the entire pattern appeared on every spike of the relaxation oscillations. These results indicate a coupling between modes within the crystal, and also between separate oscillating regions within the crystal. This coupling is very likely due to scattering from defects, strains, or inhomogeneity.

The results of these two investigations were presented at the Optical Society Meeting in Pittsburgh, March 2nd through the 4th. Papers are being prepared for publication and preprints will be submitted as soon as they are completed.

Work has been initiated toward obtaining a relative figure of merit of the optical quality of ruby crystals. The method, which involves photographing a resolution pattern in polarized light through the crystal, will be discussed in Section VII. Figure 41 shows some of the patterns photographed. The comparison between the resolution obtained and the performance of the crystal to obtain some standard criteria is a continuing project. The results to date are not conclusive.

Efforts to obtain higher power and improved performance are continuing. Figure 4.2 shows three ruby boules. The short boule with the rough finish is a standard ruby boule. The boule with the polished surface is 1" diameter by 2-1/4" long. It was grown (by the Linde Co) by a horizontal rotating and reciprocating method. The physical

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a) 190 nm x 9 mm



b) 25 mm x 5 mm





c) 25 mm x 5 mm

d) 10 mm x 10 mm

Resolution Charts for Ruby Figure 4.1



appearance of this crystal is excellent. However close examination indicates growth rings of varying index around the 1/4" clear sapphire center making this crystal not useful for LASER application. The lone (7") boule, grown by another method (also by the Linde Co), appears to be of good optical quality. This crystal and similar ruby rods obtained from the Adolph Meller Co (grown in Switzerland) will be useful for ruby amplifier applications and for ruby oscillators of increased output power.

3.0 Work Planned for Next Period

- Construction of ruby amplifier and investigation of its properties. High output power from long ruby rods is expected and pulse sharpening due to saturation will be looked for.
- 2. Continuation of the study of the quality of ruby crystals by comparison of their resolution with polarized light to their LASER performance.

Optical Pumping of Ruby (Neighbor Lines) V.

1.0 Summary of Work Performed

The "neighbor line" spectrum of dark ruby was investigated to determine the feasibility of using exchange displaced levels in a low power pulsed or C.W. LASER. The energy level diagram was found to be exceedingly complex but nonetheless appeared to fulfill the requirements for excitation of oscillation. An experiment was conducted in which the dark ruby sample, cooled to 2°K and pulse-excited, was observed to emit fluorescent radiation of sufficient spectral intensity to cause oscillation. Oscillation was not produced, however, because of the poor crystal quality of the samples and the resultant high scattering losses which it cause ...

2.0 Analysis of Work Performed

2.1 Introduction

The two prominent red R line's at $\lambda 6920A$ and $\lambda 6934A^{\star}$ in the fluorescent spectrum of pink ruby arise from a level of the free Cr ion which is shifted and split by the internal electric field of the corumdum lattice . The chromium ion which has replaced an aluminum ion in the crystal lattice finds itself in an electric field of predominately octehedral symmetry with an additional trigonal component. * Wavelength and wavenumber are vacuum designations for $T < 77^{\circ}K$. pink" ruby signifies a chromium oxide content of up to 0.1 Wt%. [6] "standard"ruby from 0.1 to 0.6Wt% and "dark" ruby indicates higher concentiations.

[7] Y. Tanabe and S. Sugano, J. Phys. Soc. (Japan) 9, 753, 766 (1954)

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Crystal field theory shows that the 9 fold orbitally degenerate ${}^{2}G$ level of the free ion at 3,000 cm⁻¹ will be split by the octehedral component of the crystal field into four levels: an orbital doublet, two orbital triplets and an orbital singlet. The lowest of these is the doublet which is further split by 29 cm⁻¹ into two components by the trigonal components of the crystal electric field at the cation site. It is the transition from these two doublet components to the ground state that produces the strong red fluorescence at 14,418 cm⁻¹ and 14,447 cm⁻¹.

It is observed^[7] that as the concentration of chromium is increased new fluorescent lines appear slightly to the red of the "R" lines. These new lines are not accounted for in the theory of Sugano and Tanabe. It is also observed that in absorption some of these lines vanish at low temperatures (<77*K) indicating that the transition terminates on a level lying above the ground state by 10 to 100 cm⁻¹ [8].

Because of the concentration dependence ^[9] of the intensity
of the neighbor lines, their explanation must necessarily involve some
motual interaction between near neighbors. One explanation which has been
advanted ^[9] ^[10] supposes an anti-ferro magnetic exchange interaction between neighboring pairs through the interposed oxygen ions. It is likely,
[*] Under good conditions each line is seen to be further split into doublets with spacing approximately 0.3 cm⁻¹. This is due to residual spinorbit coupling which splits the spin-doublet ground state.
[7] O. Deutschbein, Annal der Physik
[8] S. F. Jacobs, unpublished Thesis, Johns Hopkins U. 1954
[9] A. L. Schawlow, D. L. Wood, A. M. Clagston, Phys. Rev. Letters <u>3</u> 271, (1959)

[10] L. Remai, et al, Phys. Rev. Letters 4, 125 (1960)

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that the largest term in the ground state exchange interaction Hamiltonian has the form $J(S_1, S_2)$ where S_1 is the vector spin of the ith member of the pair. This term is diagonalized by eigen functions of total spin $S = S_1 + S_2 = 3,2,1,0$. The eigenstates have energies J, 3J, 6J with respect to the ground level, S = 0. They obey the Lande interval rule. Because in the ground state the exchange interation is expected to be larger than the residual spin-orbit coupling that is, greater than 1 cm⁻¹ we are led to expect that the lower satellite levels will tend to follow the interval rule. In the excited levels however, the larger trigonal field splitting will cause devictions. Support for the exchange hypothesis comes from the anti-ferromagnetic property of pure chromic oxide . Paramagnetic resonance data^[10] taken at room temperatures and lower also seems to support this hypothesis and indeed gives the interaction energy for nearest neighbors, third nearest neighbors, fourth and so on.^[*]

2.2 Determination of the Energy Level Diagram for Ruby Neighbor Lines

It is observed that of the more than 30 neighbor lines to be seen at temperatures higher than 77°K, two are especially bright and in fact can be seen weakly even in pale ruby. They are found at λ 7010.3A and λ 7042.5A. At the temperature of pumped liquid He (T<2°K) all but [*] The experiments performed failed to reveal the resonance due to 2nd nearest neighbors. Reasons for this failure are not known.

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Il lines vanish while the two mentioned increase in intensity. It was initially supposed by us that the λ 7010A and λ 7042A fluorescence represented transitions out of the lowest metastable state of non-equivalent pairs, and that higher levels of each pair were coupled strongly via lattice interactions. The remaining lines at 2°K were assumed to be transitions out of these metastable levels to other members of the ground complex.

To verify these assumptions the experiment shown in Figure 5.1 was arranged. With this equipment, the monitor spectrometer was tuned to one of the strong satellite lines emitted by the ruby sample inside the dewar vessel. The sample was excited by light from the monochromator whose wavelength drive was programmed to scan slowly over the region about 14,000 cm⁻¹.

With the sample at a low temperature, such that the population of the lowest excited member of the ground complex is not populated significantly, fluorescence was detected by the spectrometer each time the monochromator passed light corresponding to a transition from ground to a member of the excited complex above the emitting metastable level.

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TABLE 5,1

Details of Equipment of Figure

Spectrometer: JACO, Model 82000 0.5 m Ebert; 1800 *l*/mm Grating, 1st order linear dispersion 9A/mm; slit height 3 mm.

Monochromator: Ferkin-Elmer Model 9803 automatic scanning; 1200 2/mm; 52 x 52 mm Bausch and Lomb grating, 1st order linear dispersion 33A/mm; slit height 3 mm.

Arc Source: Bausch and Lomb automatic feed 5 Amp A. C. Cored Carbon Arc or Hanovia No. 538-C high pressure xenon arc.

Detector: EMI 9558-B photomultiplier.

Recording System:

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Monochromator output light mechanically chopped at 35 cps; chopper reference and signal demodulated in Sanborn Servo Monitor preamplifier driving a Bristol Recorder.

<u>Sample:</u> 400 mesh chips of approximately 0.6% Cr₂0₃ ruby packed into 1 mm I. D. 5 mm length capillary immersed in the cryogenic fluid.

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The procedure described was repeated for five of the neighbor lines which appear in emission at 2°K. Others emitted too weakly to permit a reliable determination of levels. At 2°K it was certain that all fluorescence exciting transitions were from the ground state or from states lying within 10 cm⁻¹ of ground.

From the experimental data we may determine the position of the metastable level from which the neighbor line under investigation originates. Thus we find that three metastable levels are involved suggesting the occurence of three non-equivalent pairs. Although there is yet some uncertainty, we show in Figure 5.2, a set of three metastable levels and ground complexes which fit the data. Figure 5.2 does not show all the levels of each system, but it should be noted that in the case of (a) and (c), a Lande interval spacing is suggested, the former being inverted with respect to the latter. Levels of system (b) higher than 50 cm⁻¹ above ground would not appear in the experimental data because 77° K was the highest temperature of the sample.

2.3 Stimulated Emission of the Ruby Neighbor Lines

The energy level diagrams of Figure 22 shows that the strong neighbor lines λ 7010 and λ 7042 terminate on levels which are not thermally excited at liquid helium temperatures (T < 4°K). Also, these lines are strongly excited by absorption of light in the well known "green" band

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of ruby. Thus it should be possible to excite stimulated emission and LASER oscillation by the absorption of relatively small amounts of pump power.

An experiment to measure the peak spectral intensity of the neighbor lines under flash excitation was conducted. The equipment used was similar to that shown in Figure 5.1 except that four 100 joule flash lamps were focussed through the dewar ports on the sample (a rod, 2 mm diameter x 25 mm length). It was not possible to completely evaluate the results because we do not yet know the quantum degeneracy of the metastable and terminal levels. On the basis that they are equal, sufficient intensity was observed to expect LASER oscillations. When the rod ends were polished flat and mirrored, no evidence of oscillation was seen. This result is attributed mainly to the large scattering loss which a light ray suffers in traversing the length of the rod, requiring a higher gain through the rod than would be expected.

3.0 Plans for Next Period

As ruby crystal quality improves, it becomes meaningful to repeat the neighbor line experiments. It is planned to obtain such improved samples, if possible and make a further effort toward producing oscillation with low power excitation.

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VI. Optical Pumping of Rare Earths

1.0 Summary of Work Performed

Single crystal boules of calcium tungstate containing various concentrations of trivalent europium, terbium, dysprosium, and praseodymium were grown. The crystals containing Pr have not been evaluated. Difficulties were encountered in obtaining boules containing Eu and Tb with the high emission intensity characteristic of flux grown crystallites of the same material. Crystals containing Dy however grow well and fluoresce strongly.

Some of the latter crystals were ground and polished to LASER tolerances and oscillation attempted on a pulsed basis. While the brightness of the output was more than adequate for LASER oscillation, no oscillation was observed. Studies of crystal perfection indicate that the internal quality of these crystals is not yet adequate.

A solution containing Europium (Eu:TTA) was evaluated as the active media of a LASER oscillator. The engineering difficulties with this system were found to be very great. Work on this solution, though still promising, was dropped in view of the encouraging results with the tungstate crystals.

Some effort on other host media than the tungstates was continued and has, as yet, not given positive results.

2.0 Analysis of Work Performed

2.1 Crystal Growth

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Measurements at low power on the fluorescent intensity of small, flux grown, single crystals of $CaWO_4$ containing Eu compared to the fluorescent intensity of ruby indicated that calcium tungstate crystals containing Eu⁺⁺⁺, Tb⁺⁺⁺, Dy⁺⁺⁺, and possibly others would oscillate at considerably lower power than ruby provided:

- a. Large single crystals had the same fluorescent efficiency as the small flux grown variety.
- b. Crystals obtained by pulling from the melt had adequate perfection and optical clarity.
- c. Unforseen difficulties did not arise such as pumping out of the excited state, etc.

The intensity measurements were made only on crystals containing Fu and the results extrapolated to Tb and Dy by using the data of Soden and Van Uitert [11].

Initially the trivalent ions of Eu, Tb, and Dy were considered most promising in that order. The level structure of Eu⁺⁺⁺ is most suitable because the emitting level (of the 6100A transition) is the singlet ${}^{5}D_{0}$ state and the lower level, ${}^{7}F_{2}$ at 1000 cm⁻¹ above ground, is neither excessively high nor too low to operate at room temperature. The emission intensity is high (in flux grown crystals) [11] L. G. Van Uitert and R. R. Soden, J. Chem. Phys. <u>32</u> 1161 (1960); J. Chem. Phys. <u>32</u>, 1687 (1960); also personal communication.

and there is no significant concentration quenching. Tb (at 5450A) also has a high emission intensity and no significant concentration quenching. However, the degeneracies involved in the transition, ${}^{5}D_{4}$ to ${}^{7}F_{5}$, are less favorable. The strong transition in Dy^{+++} (5750A, ${}^{6}F_{11/2}$ to ${}^{6}H_{13/2}$) is concentration quenched at about 1 mole per cent Dy^{+++} . This transition was observed to have a very narrow linewidth at low temperatures, however, and is considered very promising.

Crystals of CaWO₄ containing various concentrations of Eu, Tb, Dy, and Pr have been grown. Most of the problems of obtaining boules of adequate size have been overcome so that it is normal to obtain a reasonable boule on each attempt. A more substantial apparatus for pulling crystals has been constructed and put into use. The pulling apparatus is shown in Figure 6.1. The crystals grown have been encouraging but yet inadequate.

a. Europium

Boules were grown containing 5%, 2% and 0.16% Eu. These crystals were black to dark red and showed little fluorescence, in sharp contrast to the microcrystals of the same material grown from a flux. Difficulties were also encountered in cracking of the crystals upon cooling after the crystals were pulled. Very slow cooling alleviates this problem.



Figure 6.1 Crystal Pulling Apparatus TECHNICAL RESEARCH GROUP

Considerable changes in the color and the emission were observed when some of these boules were annealed. A second group of boules were grown with similar results except that these required considerably longer periods of annealing (5 days at 1450°C) before the changes in color and increased emission were observed. An X-ray analysis failed to show any differences between these two sets of boules.

The most likely explanation for these results on Eu in $CaWO_4$ is that the crystals grow with an oxygen deficiency. This would account for the coloration and the changes which occur after long periods of annealing. The ionic diffusion rate in calcium tungstate at elevated temperatures is well known. Oxygen could thus be absorbed during the annealing process. Future growth of Eu in CaWO₄ will be done in an oxygen atmosphere.

b. Terbium

One group of boules containing terbium in various concentrations was grown. These were optically clear but lacked the strong emission intensity characteristic of the flux grown crystals. Annealing these crystals failed to improve their emission intensity. The sintered powder of NaTb(WO_4)₂ used to dope the calcium tungstate powder for growing was not ' galy fluorescent and appeared to have a brownish tinge characteristic of four-valent terbium. This situation

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was corrected and doping material is now available which is white and highly fluorescent. Future growing and annealing of these crystals will be done in an argon atmosphere to prevent the absorption of oxygen. The terbium should thus be kept in the trivalent state.

c. Dysprosium

Many boules of calcium tungstate containing various concentrations of Dy⁺⁺⁺ have been grown. These crystals appear optically clear and their growth does not present the problems associated with the crystals containing Eu or Tb. They are highly fluorescent ($\lambda = 5740A$) and appear very promising as the active media for LASER oscillation. The evaluation of these crystals will be reported separately.

d. Praseodymium

One group of boules containing four different concentrations of Pr has been grown. The crystals were pulled in an argon atmosphere because of the well known tendency of Pr to be converted to the quadri valent state. The crystals were optically clear but have not yet been evaluated.

e. Growth of other crystals

Other host media for rare earth salts are being worked on with reduced priority as time permits. These include mainly anhydrous

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lanthanum chloride in single crystal form and a variety of sintered crystalline powders containing rare earth salts.

A relatively new approach has been adopted for the synthesis of lanthanum chloride crystals containing rare earth chlorides. Equipment has been set up for pulling a single crystal out of the melt in an atmosphere of hydrogen chloride. An initial run has been completed with partial success. If this method proves feasible the single crystals will be coated with a film to make them impervious to moisture and then evaluated for LASER possibilities.

Sintered powders of many mixed oxides, chlorides, and fluorides containing rare earths are being prepared. They have not yet been evaluated.

2.2 Evaluations of EusTTA

As reported earlier this solution has sufficient brightness of emission to give LASER oscillation at room temperature with pulsed excitation lower than required for ruby. The difficulties involved in the use of this solution were stated in previous reports on this program, namely

- a. Difficulty of containment of the liquid in the presence of the strong excitation flash, particularly in a tube with the required tolerances.
- b. Photosensitivity of the organic compound.

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It was felt that the photodissociation of the molecule could be overcome by circulation of fresh solution. The containment of the solution, however, has proved to require an effort beyond that which could reasonably be devoted to this problem, particularly in view of the encouraging results on the solid state systems.

The problem of containment is difficult because the solvent used (alcohol, ether, isopentane) has a boiling point of 29°C. Other solvents were tried but no other solution had as high an emission intensity and could not contain a sufficiently high concentration of TTA for LASER action. Freezing of the solution at liquid nicrogen temperatures failed since the resulting highly viscous glass-like liquid almost invariably cracked before it could be used. It then seemed that the shock wave generated by sudden local boiling must be contained. Capillary tubes were tried but none were obtained with the required tolerances. They presented one further problem--that of bubble formation in the capillary tube. After a flash of the excitation light it was difficult to clear the bubbles from the tube even with mighting with cooled liquid since new bubbles would invariably form.

The decrease in emission as a function of total energy of excitation was measured. The results of this measurement are shown in

Figure 6.1. Each point required care to remove the bubbles from the tube. The liquid, which was recirculated continuously, had a total volume of 220 cm³. The highest emission intensity observed with fresh solution indicated that this material would have given LASER oscillation at room temperature with a threshold energy to the lamp of 1/10 that required for ruby. These calculations are based on a capillary tube 1/8" inside diameter, 3" long, with 95% reflecting plates, assuming the solution of the engineering difficulties connected with its use.

Similar problems (in particular, photodis sociation) might be expected with other organic compounds in the presence of strong flashes of excitation light. The strong excitation is presently required because of non-radiative processes which reduce the quantum efficiency of most rare earth compounds. When these processes are understood and overcome the characteristic wide absorption bands of organic compounds should prove to be advantageous.

2.3 Evaluation of CaWO4: Dy for LASER Oscillation

Spectrographic analysis of the emission spectra of Dy⁺⁺⁺ in CaWO₄ indicated a very narrow linewidth at low temperatures. This fact along with the strong emission and the lack of some of the

56.



problems in growth associated with Eu and Tb in CaWO₄ made this crystal sufficiently promising to warrant further investigation.

a. Excitation and emission spectra

The emission spectra of the group of lines in the neighborhood of 5700A was taken at liquid nitrogen and liquid helium temperatures. These transitions are from the ${}^{6}F_{11/2}$ to the ${}^{6}H_{13/2}$ about 3500 cm⁻¹ above ground. (See level diagram of Figure 6.2.) The wavelengths of emission are tabulated in Table 6.1. The excitation wave numbers which are effective in exciting the ${}^{6}F_{11/2}$ level are tabulated in Table 6.2. These are so numerous as to preclude obtaining an accurate level designation for each transition without a great deal more work. The excitation bands are relatively narrow. The strongest, a group around 4500A, have individual widths of about 1.5 at helium temperatures. The narrow line absorption is a drawback as far as IASER oscillation is concerned. They are sufficiently numerous, however, to give an emission intensity of the 5740A line which appears adequate for LASER oscillations.

b. Linewidth and lifetime

13

The emission lines are not well resolved at room temperature. The width of the 5740A line is about 8-10A. At nitrogen temperature the lines are all resolved and the width decreases to 1A. The same

58.



Figure 6.2

TABLE 6.1

Dy Emission Spectra in CaWO4

77°K	<u>4•K</u>
5763A	not measured
	5752
5750	5750
5737	5737
	5736
	5734
5732	
5730	5731
5727	5727

TABLE	6.2
	the second secon

Excitation Spectra for 5737 Line of Dy^{+++} in CaW0₄

Relative			Relative	Relative			Re
<u>77°K</u>	Intensity	<u>7 4°K</u>	Intensity	<u>77°K</u>	Intensity	<u>4°K</u>	In
-1	- 4	-			1	-1	
17461cm	24	1	-	24988cm	- 18	24988cm -	•
489	15	17492cm -	11	25031	15	25019	
20028	5	20028	5	082	18		
492	4	492	4	100	21	100	
820	3			170	28	170	
886	30	890	58	227	11	240	
907	5	907	6	426	30	432	
942	11	925	8	491	28	494	
		956	11	543	19	543	
978	6	982	7	628	25	628	
21000	3	21017	7	667	34	667	
039	28	044	> 90	694	42	694	
053	11			740	60	746	>
097	9	088	20	773	52	769	1
155	3	142	5	826	41	816	
240	4		•	860	20	847	
277	11	268	6	887	15	886	
211	• -	277	14		~ 3		
		313	4	26157	8	26144	
		368	2		Ŭ	254	
896	21	000	6	350	19	364	
954	32	952	2 90			413	
978	69	976	5 90	27137	9		
22012	32	22026	> 90	159	27		
065	15	075	14	211	74	214	
005	▲ 3 7	114	8		3 -1	228	
139	35	148	74	263	50	270	
188	47	193	> 90	300	18	270	
222	50	222	86	337	15	330	
		257	12	231	±./	345	
		282	7	793	5	809	
23226	6	~~~	•	28169	19		
269	18	272	49	20109	50		
	20	310	4	265	46	272	
337	10		~₹	297	62	297	
386	15	378	17	3/15	67	252	
411	20	416	46	747 701	67	202	*
457	5	460	8	433	26	/33	/
24802	14	802	37	455	17	433	
851	15	845	30	400	17	571	
863	10	875	10	5 2 1	0	5/T	
000	IV	2702	10 6	637	о с	629	
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61,
line at liquid helium temperatures has a linewidth of 0.15A in the more dilute samples and 0.23A in the 0.8% Dy sample. The peak emission elative ntensity intensity varies nearly inversely with linewidth. The measured lifetime of the $F_{11/2}$ level is 250 μsec independent of temperature.

Emission intensity с.

8

23

20

50

18

29 70

50 15

52 49

90 90

86 24

10

14 7

80 7

44 33

30

18 20

3

72

9

To check the absolute emission intensity a crystal was placed into the helical xenon flashlamp used to excite the ruby oscillator. The crystal was grown from a melt containing 1% molar concentration of Dy^{+++} , was cut 1" long, and had a diameter (somewhat uneven) of about 3/16". Both ends were polished for optical transmission, although not to the tolerances required for IASER oscillation. The crystal contained visible flaws which would make it inadequate for use as a IASER crystal. Scattering in the crystal however does not have much effect on the results of the present experiment since in a reasonably isotropic field as much light is scattered into the solid angle of observation as out of it. By the same token this experiment gives little indication of the optical perfection of the crystal.

The output was detected through a 0.5 M Jarrell-Ash monochromator by a photomultiplier. The response of the system was measured with a standard lamp calibrated by the National Bureau of Standards.

With an input of 500 joules into the flashlamp the measured brightness of the crystal was 0.26 watts/cm²/ster A at room temperature. The duration of the flash at this input energy was approximately 500 μ sec, or twice the lifetime of the upper level in Dy. Increase in energy of the flash by addition of capacitance mainly increased the duration of the flash rather than the peak power and thus should have only a small effect on the peak brightness of the Dy crystal. This analysis was verified experimentally. If, on the other hand, the flash duration were made shorter than the lifetime of the upper level of Dy the same energy input would give about a factor of two increase in brightness of the crystal.

The absolute brightness of a ruby crystal with unsilvered faces was checked in the same way with an input of 500 joules. Measured brightness was nearly one fourth that computed to be required for oscillation. Since the lifetime of the R_1 level in ruby is 4 msec an increase in lamp energy does increase the output brightness of the ruby crystal proportionately. The measured threshold of this particular crystal (now with silvered faces) was 2300 joules input to the lamp. The close agreement between the predicted and measured threshold of ruby, based on brightness measurements at lower power, give increased confidence on the validity of the brightness measurements.

The brightness required for LASER oscillation at 5740A, assuming 5% losses (in transmission, absorption, on reflection, scattering, etc.) is 0.94 watts/cm²/ster A. This also assumes an unpopulated lower level of the LASER transition (as is the case for Dy in pulsed operation). This value of required critical brightness must also be multiplied by the product of the degeneracies of upper and lower states which contribute to the measured brightness. In the case of Dy the narrowness and symmetrical shape of the 5740A line at low temperatures indicates that the transition may be between single nondegenerate states, or, at worst, between doubly degenerate upper and lower states. The required critical brightness for this transition in Dy, 0.94 watts/cm²/ster A, is thus uncertain by no more than a factor of 4.

or

r

From these measurements the expectations for these crystals, given as the ratio of input energy for threshold compared to that for ruby, is as follows:

<u> </u>	$\frac{(E_{Dy}/E_{R})}{R}$ crit
300*к	0.5 ~ 2
77 ° K.	0.05 - 0.2
4*K	0.012 - 0.05

These calculations are based on excitation by xenon flash-

lamps of 150 usec duration such as the straight lamps presently in use.

 * This brightness, as well as brightness measurements quoted, is that
just inside the output face of the crystal. The measured brightness in air will be reduced from this value by the refraction at the crystalair interface. TECHNICAL RESEARCH GROUP

d. Experiments with optically polished crystals

Two crystals of $CaWO_4$: Dy have been polished with ends flat to $\lambda/4$ and parallel to 15 seconds of arc. The finished crystals were 1/4" diameter and 7/8" long, and had a ground finish on the sides. One of these is shown in Figure 6.2. The concentrations were 0.033% and 0.8% Dy (in the melt), and neither was annealed. The crystals appeared optically clear in ordinary light. The resolution through the crystals with polarized light was reasonable for the 0.033% Dy crystal and poor for the 0.8% crystal. A great deal of scattering is thus indicated in the latter crystal. Nevertheless LASER oscillation was attempted in this crystal in the hope that filaments of good quality might exist which would not be observed by the resolution technique because of the aperture used. The intensity of emission of the crystal wich lower concentration was not adequace.

The first attempt was made with the 0.8% Dy crystal at nitrogen temperature in a small, specially designed glass dewar surrounded by 6 xenon flashlamps of the GE FT-91 type. The flash tubes were surrounded by a cylindrical reflector of Al foil outside a quartz tube. Precautions were taken to exclude moisture from condensing inside the enclosure. No bright yellow beam was observed when the lamps were flashed with an energy sufficient to cause oscillation

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Figure 6.2 Polished Crystal of CaWO₄:Dy

in ruby. The eye is more than 100 times as sensitive to light at 5740A as to the 6943A light from ruby so that this method would appear to be a sensitive test for detecting an oscillating beam. Subsequently the crystal cracked so that measurements could not be made to verify that the brightness was indeed 5 to 20 times the critical brightness.

The brightness of this crystal had been measured, however, in the helium dewar. With four FT91 lamps excited by a pulse energy of 180 joules/lamp the measured brightness was 0.67 watts/cm²/sterA at liquid helium temperatures. While this is not adequate it corresponds closely to expectations in this situation based upon the previous calculations. The excitation efficiency is low in this arrangement because of the necessity of focussing the lamp flashes into the dewar by means of lenses, with the resulting loss of solid angle of excitation.

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> The conclusions derived from these results are that the crystal did not oscillate because of the lack of optical perfection, that more perfect cyrstals will oscillate at nitrogen temperatures or somewhat above, and that oscillation at room temperature will be difficult. These conclusions will be checked on other crystals being grown or in the process of polishing. The scattering losses in the crystal were expected to be reduced by growing the crystals more slowly.

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e. Other experiments on Dy

No attempt has yet been made to orient the crystals grown. Orientations of some of the boules have been checked by a method similar [12] to that suggested by Strundberg. These boules have had the c axis oriented at various angles with respect to the boule axis. They were grown from seeds cut more or less randomly from other boules so that the direction of easiest growth is not well known. The two crystals which were polished had the c axis oriented at 32° and 38° from the rod axis.

As yet, the most desirable orientation is not known. Polarization of the emitted light has been observed and these effects are being studied. Absorption coefficients have not yet been measured. This also plays a part in determining the most desirable orientation of the crystal axis.

The effect of annealing on the properties of the crystal is also being studied. Preliminary studies indicate that annealing does not change the emission intensity at room temperature.

A wet chemical analysis is being conducted to determine the actual concentration of Dy in the crystal. In conjunction with this program the optimum concentration will be determined. It is possible [12] M.W.P. Strandberg and R. P. Mattuck, Rev. Sci.I. 30, 195 (1959)

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that a compromise may have to be made to achieve optical clarity since the latter often varies inversely as the impurity concentration.

3.0 Work Planned for Next Period

a) Continued growth and evaluation of $CaWO_4$ containing Dy. This program includes slower growth of oriented crystals and should lead to a pulsed LASER at 5740A.

b) Growth and annealing of CaWO₄ containing Eu and Tb in controlled atmospheres to maintain the rare earth ion in the trivalent state. Evaluation of crystals grown.

c) As time permits, the following programs will also be carried out:

- 1) Growth of anhydrous LaCl₃ by pulling from the melt. Doping with rare earths.
- 2) Growth of SrCl₂ by pulling from the melt and doping with rare earths, in particular with Sm^{++} .
- 3) Evaluation of sintered powders of mixed compounds containing rare earths.

VII. Optical Measurements and Devices

1.0 Summary of Work Performed

a) Two new forms of the Fabry-Perot interferometer have been constructed which are far superior to the F.P. in adjustment tolerances. Haidinger fringes have been observed in both the retroreflective cornerreflector-vs-flat and crossed-roof interferometers.

b) A relatively simple optical system has been devised to indicate the optical quality of crystals to be used for L\SER operation.

c) A method has been devised to measure the orientation of the c axis of uniaxial crystal boules with irregular sides.

2.0 Analysis of Work Performed

2.1 Crossed Roof Prism Interferometer

When a medium with a negative absorption coefficient is contained within a tuned optical cavity sustained oscillations are possible, provided the gain of the medium exceeds the losses in the cavity. The plane Fabry-Perot interferometer has been used in this way^[13], and is conceptually the simplest cavity. However, the stability of the F.P. leaves much to be desired.

[13] A. Javan, W. R. Bennett, Jr., D. R. Herriott, Phys. Rev. Letters <u>6</u>, 106, 1961.

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It would seem that corner reflectors could be substituted 14,15816 for the flats of the F.P. in much the same way that Peck and [17518] Murty have modifed the Michelson interferometer to relax the alignment tolerances. Peck has shown theoretically that, with corner reflectors used as a F.P. cavity, one obtains 6 eigen-polarizations, a difficulty with which one must contend when constructing a LASER cavity. We have succeeded in obtaining fringes with a corner vs flat reflector Figure 7.1, and have observed the polarizations predicted by Peck. (Figure 7.2) The corner reflector holds considerable promise as a stable F.P. cavity and further work is in progress to analyze its use as a cavity. The roof prism interferometer combines several features: a simpler pattern of eigen-polarizations than the corner cube, end plates easier to make than corner cubes and stability much improved over the plane F.P. interferometer.

As a first step toward attaining stability we set-up a folded Fabry-Perot interferometer (Figure 7.1) using 2 front surface mirrors, mounted at a right angle, and a multilayer coated flat. * Private communication ¹⁴ E. R. Peck, JOSA <u>38</u>, 1015, 1948 ¹⁵ E. R. Peck, JOSA <u>38</u>, 66, 1948 ¹⁶ E. R. Peck, JOSA <u>47</u>, 250, 1957 ¹⁷ M.V.R.K. Murty JOSA <u>50</u>, 7, 1960 ¹⁸ M.V.R.K. Murty JOSA <u>50</u>, 83, 1960

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- <u>FOLDED</u> FABRY-PEROT INTERFEROMETER

Figure 7.1





Polarized Fringes from One Sector of a Corner Cube Interferometer Figure 7.2 73.

Two sets of fringes were observed, one set polarized normal and one parallel to the plane of incidence. These were due to the different polarization phase changes upon reflection from the mirror pair. The multilayer used had 95% reflectivity at $\lambda = 5461$. We found that freshly evaporated silver coatings were required on the mirror surfaces. Aluminum had too low a reflectivity. (Fringes were also observed when we replaced the mirror pair with a roof prism, making use of total internal reflection). It was possible to rotate the mirror pair several degrees about the intersection line without losing the fringes.

We next replaced the flat with a second mirror pair, oriented at right angles to the first mirror pair, Figure 7.3 and observed a single set of fringes. In this case the two mirror pairs introduce compensating phase shifts, as the orientations of the two polarizations are interchanged with respect to the mirror pairs. The stability of this interferometer much surpasses that of the plane Fabry-Perot and will be discussed in detail later in the section.

Comparison of this interferometer with a plane F.P. shows that the free spectral range is equivalent to that of a F.P. with twice the vertex separation, d, of the mirror pairs. The resolving power differs from that of a plane F.P. since adjacent interfering beams suffer 8 reflections instead of 2. Each surface must therefore have an extremely

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high reflectivity to obtain sharp fringes. This apparent drawback is not so significant, however, when this interferometer is used as a resonator with an amplifying medium. In a resonator it is the reflection loss per pass that is important, and this interferometer has two reflections per pass, as opposed to one per pass for the plane F.P.

The inevitable losses of front surface reflectors can be avoided by employing total internal reflection. In order to obtain the advantage of total internal reflection without the disadvantage of Fresnel reflection losses in prisms we have constructed an interferometer with roof prisms whose entrance faces are so oriented that impinging rays strike them at or near Brewster's angle. Thus the cavity has a very low reflection loss for one polarization. The output beam is conveniently picked off by constructing one entrance face at an angle slightly different from Brewster's angle, providing 2% reflectivity. The fact that the cavity is lossy for the second polarization does not lessen its efficiency for LASER applications, but merely means that stimulated emission of radiation will build up only in one polarization.

a) Tolerance of Crossed Roof Prism Interferometer

Previously it was noted that the use of a Crossed Roof Prism interferometer led to greatly relaxed tolerances on the mechanical adjustments of the "LASER" resonator. To make a quantitative tolerance

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comparison between the plane Fabry-Perot and the Crossed Roof Prism interferometer, a rather simple criterion is available. i.e. comparison of the deviation angle between successive rays leaving the roof interferometer vs. the same in the case of the F.P.

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The deviation angle (in the plane F.P. is simply calculated to be -2ψ , where ψ is the tip angle between plates. In the case of the Crossed Roof Prism, the deviation is not simply derived. To facilitate the calculation, we consider two Cartesian coordinate systems each fixed to a roof prism, with one axis lying along the roof axis of the prism. After having lined the prisms up with their roof axes normal to one another, we now allow one prism to rotate through small angles about three mutually perpendicular axes. Since rotation about the roof axis produces no deviation, this property being basic to roof reflectors, we need only consider two rotations.

Before interfer the can occur between successive rays, the wave must travel 4 times through the cavity, compared to twice in the plane F.P. It is necessary to consider this complete trip to calculate the deviation. We now consider a unit vector n which represents the original ray in the fixed coordinates. Transforming this vector into the rotated coordinate system, we operate on it with a reflection

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operator and then transform it back into the original coordinate system where another reflection operation takes place. The procedure is then repeated since we must consider four trips. The result is a vector nⁱ which represents the deviated ray. The scalar product between n and nⁱ thus gives us the cosine of the deviation angle. The process can be represented formally as

$$n' = (RT^{-1} R'T)^2 n$$

$$\cos \delta = (RT^{-1} R'T)^2 n \cdot n$$

where R_s and R' are the reflection matrices in their respective coordinates systems, T and T⁻¹ being the transformation matrix and its inverse. The result of this calculation is that $\delta = 4 \ \Delta\theta \Delta \phi$, where $\Delta\theta$ and $\Delta \phi$ are small angles of rotation. (see Figure 73).

It is obvious that, for equal deviations in the Crossed Prism and plane F.P., $\psi = 2 \ \Delta \theta \Delta \Phi$. However, since the interference path and hence the gain of this device is twice that of a comparably spaced plane F.P., the tolerance δ , the angle of tip, should be compared with $\Delta \theta \Delta \Phi$ rather than twice that quantity.

To consider a specific example of relaxed tolerance, it has been shown that for the neon-helium LASER to operate the tip of plane F.P. plates cannot exceed 6 seconds of arc_{p} or 3 x 10⁻⁵ radians. For the prism instrument this would mean $\Delta \theta \Delta \phi \leq 3 \times 10^{-5}$. If we assume equal tolerance in $\Delta \theta$, and $\Delta \phi$ then $\Delta \theta = \Delta \phi \leq 5 \times 10^{-3}$ radians, 5×10^{-3} radians 16 minutes of arc. This means that the relaxation in tolerance would be greater by two orders of magnitude.

2.2 Optical Clarity of Crystals

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By use of a relatively simple optical system, a resolution chart and a camera, an indication of the optical quality of crystals to be used for LASER operation can be determined.

Collimated light from a white light source, passes through Nicol prisms between which the crystal to be investigated is placed as shown in Figure 7.4 .

Unfinished crystals are first prepared by immersion in a parallel sided container filled with a refractive liquid having an index equal to that of the crystal, to eliminate distortion due to the ends not being parallel. This also eliminates any lens effect the crystal might give. Exposures are then made viewing the resolution chart both with the crystal in and out of the system. It was found that refocussing after the crystal was inserted in the system did not improve the resolution, thereby indicating that the crystal was not acting as a lens. The smallest pattern that can be resolved gives an indication of the optical quality.

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The procedure described above gives only a relative indication of sality. These crystals are then compared with those in which oscillations have been observed in order that a standard may be determined.

2.3 Orientation of Uniaxial Crystal Boules

A simple method has been devised to measure the orientation of the c axis of uniaxial crystal boules with irregular sides. The method is essentially similar to me proposed by Mattuck and Strandberg^[12]. It is different in that one further measurement allows the orientation of the c axis to be read directly from a scale without the computation required in the method of Mattuck and Strandberg.

The boule is placed in a glass vessel with square sides. Liquid with the same index as the crystal is added to cover the boule. The boule is held by a device to allow it to be rotated about the boule axis with the vessel fixed. A scale is provided to read the amount of rotation.

The vessel and boule are then placed into a beam of parallel light between crossed Nicol prisms such that the light is incident normally to one surface of the vessel. No corrections for refraction are required because of this normal incidence.

The determination of the relation between the axis of rotation of the boule and the c axis θ_{p} and ψ consists of three operations:

a) A measurement to eliminate an ambiguity between θ_p and its compliment. This is made by setting the polarization of the E vector of the incident light at an angle of 45° to the axis of rotation (which is vertical). The analyser is rotated correspondingly to keep the polarizer and analyser always crossed. Now rotation of the boule about its axis determines whether θ_p is greater or less than 45°. If θ_p is greater than 45° there will be four positions at which a dark ileld is observed. These will occur in pairs symmetrically placed about the position at which the plane containing the axis of rotation and c axis is normal to the incident light. (The latter position could be noted and eliminate step b) below). If θ_p is less than 45° no dark field will be observed, but rather two positions of minimum intensity.

b) The polarizers are rotated so that the E vector of the incident light is now set either parallel or perpendicular to the axis of rotation. The crystal is rotated until a dark field is observed. The plane containing the axis of rotation and the c axis now contains the line of incidence. A further rotation by 90° now insures that this plane is perpendicular to the incident light.

c) Both Nicols are now rotated together until a dark field is observed. The amount of this rotation is either θ_p or its compliment (the ambiguity is removed by referring to step a) and can be read directly on a scale.

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The direction of the c axis is such that the plane of the c axis and the axis of rotation is normal to the incident light.

3.0 Work Planned for Next Period

We plan further work to investigate the eigen-polarization orientations within the interferometers and explore their usefulness as LASER resonators.

VIII. Theory of Resonator Modes

1.0 Summary of Work Performed

Several aspects of the optical behavior of Fabry-Perot interferometers of importance for the understanding of F.P. LASERs have been studied. Firstly we have investigated the nature of the modes of a Fabry-Perot cavity in terms of the general theory of leaky wave or radioactive state resonances of open electromagnetic structures. Secondly we have derived an exact theory of the F.P. cavity and shown how this reduces in an appropriate approximation to the previously reported theory of Kotik and Newstein. Thirdly, we have studied the optical mode structure of a F.P. interferometer via the variational principle of Kotik and Newstein. This has led to analytic expressions for the quantities of practical interest, e g. the mode shapes, resonant frequencies, and diffraction losses.

2.0 Analysis of Work Performed

The following preprint (submitted for publication as a Letter to the Editor of the Journal of Applied Physics) is a preliminary report on a theoretical study of the optical behavior of the F.P. interferometer. This study starts from an exact formulation of the F.P. cavity problem which is reduced in an appropriate approximation to the previously reported formulation of Kotik and Newstein.

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The variational principle of Kotik and Newstein is then employed to find analytic expressions for the mode shapes, resonant frequencies, and diffraction losses of a F.P. cavity. A much more comprehensive discussion of the resutls obtained is in the course of preparation.

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Submitted for publication as a Letter to the Editor of the Journal of Applied Physics

Resonances of the Fabry-Perot LASER

S. R. Barone Technical Research Group, Inc. Sycsset, New York

The Vabry-Perot (F.P.) interferometer has recently 1 become of interest as & resonator in LASER oscillators. For this purpose it is important to know the optical mode structure of such a cavity. From the mathematical point of view the problem of two (perfectly) conducting, finite, mirrors has only a continuous spectrum of proper i.e. bounded at infinity, modes; the associated frequency spectrum running continuously from zero to infinity. However physical intuition indicates that the modes of interest should have a discrete frequency spectrum determined approximately by the condition that the ratio of the separation of the F.P. mirrors to a half wavelength be integral.

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The mathematical realization of this physical idea lies in the occurrence of leaky wave or radioactive resonances² for the F.P. cavity. These resonant wave fields de not belong to the proper mode spectrum which is complete in itself. Also the associated frequency spectrum is complex which implies that the leaky wave fields grow exponentially at large distances from the F.P. Nevertheless for an open cavity in an infinite active madium these are the resonances which are analogous to the discrete resonances of a closed cavity.

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From this point of view we have studied the problem of a F.P. interferometer composed of two parallel infinite strip mirrors each with a reflection coefficient r, the angular dependence of which is neglected. Characterizing the mirrors by their scattering matrix in Kirchoff approximation, Kotik and Newstein have previously given an expression for r which is stationary with respect to variations of the angular spectrum of the mode field. We have assumed, for the dominant mode, a trial spectrum equal to the Fourier transform of:

$$P(x) = 1 - \rho |x|/d \quad x < d$$

= 0 $x > d$ (1)

where d is the half-width of the F.F. mirrors and ρ is a frue parameter to be determined by the variational principle. Physically P(x) represents the field traveling towards either mirror at the center plane of the F.F. In the limit of sufficiently small wavelengths (1) compared to both d and L, we find that to lowest non-vanishing order in the small parameter $\lambda L/d^2$

$$1 - \rho = 0.42 = \frac{\pi 1/4}{(\lambda 1/d^2)}$$
 (2)

where L is the separation of the V.P. mirrors. Thus we see that for optical frequencies the dominant mode field at the edge of the V.P. is a small fraction of its value at the center of a mirror and that the phase at the edge is v/4 relative to the phase at the center of a mirror.

In addition the variational principle yields a complex resonant frequency. Then since the time dependence of the field is given by

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where we and we are the real and imaginary parts of the angular frequency w, we can define a frequency shift (500) and lifetime (τ) for this mode by $\delta m = m_1 - m_0$, $\tau = -1/m_2$ where $m_0 L/\pi c$ is a large integer which determines the number of axial half wavelengths in the Y.F. In particular we find, again to lowest non-vanishing order in the small parameter $\lambda L/d^2$, that

$$\delta a^2 = 1.50 \ (c/m_o d^2)$$
 (3a)

$$\frac{1}{\tau} = \frac{c}{L} \left[\ln \frac{1}{\tau} + 0.12 \left(\frac{\lambda_0 L}{d^2} \right)^{3/2} \right]$$
(3b)

where for simplicity we have supposed that the phase of r is zero, and c denotes the speed of light. The resonator losses are equally well expressed in terms of $\alpha = 1/\tau c$, the amplitude gain coefficient which the medium filling the cavity must have in order to balance the losses and achieve a steady state resonance. Also the frequency shift is alternatively expressed in terms of bd = bnL/c the difference between the change in phase of the field in traveling from one mirror to the other and the geometric phase change for the frequency w_0 . These results are to be contrasted with the work of Kotik and Newstein wherein P(x) was taken to be a constant. The resulting overestimate of P(d) led to overestimates π' both the frequency shift and losses.

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The strip mirror F.P. interformeter has also been considered by Fox and Li⁴ who solved the appropriate integral equation by machine iteration under the restriction $(\lambda L/d^2)^{-L/2} \gg d/L$. Unfortunately this is a lower bound on the allowable wavelength which for optical frequencies restricts the F.P. to be unusually long and narrow. The results obtained under this restriction, however, should for decreasing wavelength asymptotically approach the present results which for fixed dimensions of the F.P. become increasingly accurate as the wavelength decreases. The comparison between these two complimentary results is shown in Figures 1 and 2 and Table 1. In connection with Table 1 it should be emphasized that P(x)refers to the center plane of the F.P. while the results of Fox and Li refer to a mirror plane. However the field at a mirror plane is not significantly different from P(x).

Further investigation of the variational principle has led to the conclusion that for a wide class of trial functions the resonator loss is proportional to the 3/2 power of the parameter ($\lambda_0 L/d^2$) while the phase shift is simply proportional to this parameter. Thus only the numerical coefficients in equation 3 depend on our special choice of trial

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function, and are consequently subject to some error due to the inadequacy of this crial function in representing the correct field distribution. Comparison with the materical results of Fox and Li indicates that thus error is not Large.

Also, we have derived an approximation to the variational principle of Kotik and Newstein which has the advantage that the corresponding Euler equations are capable of exact solution. In this approximation the above frequency shift for the dominant mode is reduced by approximately 18% to precisely the first order frequency shift for the closed cavity obtained by closing off the side walls of the F.P.. a result anticipated by _____ 7 4 Totmas. Also the losses are reduced by a factor of .79. These results are also shown in Figures 1 and 2 and Table 1 with the label "approximate variational principle". For the higher order modes, labeled by an integer n which equals unity for the dominant mode, wa find in this approximation that both the frequency shift and loss are increased by a factor of n^2 . This latter result is also in good agreement with the work of Pox and Li who have considered the first higher mode.

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We are indubted to M. Nowstein for suggesting that further work on this problem was necessary as well as for a number of helpful discussions. We also wish to thank J. Estik for a critical reading of the manuscript.

FOOTNOTES

*	Supported by the Advanced Research Projects Agency through the Air Force Office of Scientific Research
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4	A. G. Fox and Tingys Li, Bell System Teah. J. 40 453 (1961)

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LIST OF FIGUES

- Fig. 1 Comparison of the dominant mode phase shift per transit as given by the variational principle with the numerical results of Fox and Li.
- Fig. 2 Comparison of the dominant mode loss par transit due to diffraction as given by the variational principle with the numerical results of Fox and Li.

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Comparison of the dominant mode field as given by the variational principle with the pumperical results of Fox and Li.

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λ sufficiently Variational calculation							
small with one parameter trial							
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Approximite variational							
****	principle	0.55	45.				
a Estimated from Figure 7 of reference 4.							

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3.0 Work Planned for Next Period

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In the next period we plan to determine the effect of 7 non-uniform saturation and con-homogeneous broadening as the pumping power is increased beyond that required for oscillation in the mode of lowest loss.