EFFECTS OF GAMMA IRRADIATION OF RUBY LASER CRYSTALS

by

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ABSTRACT

PHILBRICK, CHARLES RUSSELL. Effects of Gamma Irradiation of Ruby Laser Crystals. (Under the direction of WILLIAM ROBERT DAVIS and MARVIN KENT MOSS).

The energy output of ruby laser crystals (Al_2O_3 doped with Cr^{+++}) has been significantly increased after Co^{60} γ-irradiation. Increases in output energy greater than a factor of five have been obtained with the experimental arrangements employed.

The initial importance of radiation produced center formations and their possible usefulness as energy storage centers is discussed. The general ideas of color center formation are presented with special emphasis on center formation in Al_2O_3. Also, spectral effects of center formation in Al_2O_3 are studied.

The experimental results have supported the hypothesis that energy stored in the radiation created center formations could be advantageously employed in increasing the laser output energy.

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INTRODUCTION

The center formations created by ionizing radiation have been extensively studied for the simplier crystalline materials for a number of years. Although much work has gone into the study of characteristics of these centers, many properties remain to be investigated.

The idea of using radiation formed centers to store energy in certain crystals has been investigated. Fluorescence and phosphorescence studies have shown that in some crystals, transfer mechanisms exist that allow part of the stored energy to be released through luminescent transitions. The possibility of using the energy stored in gamma radiation produced center formations to populate the fluorescent level of Cr^{+++} in Al_2O_3 (ruby) was investigated. This investigation prompted further study of the possibility of using this stored energy to enhance the laser output energy as well as provide an example of a possible nuclear-coupled laser system.

A spectral investigation of γ-irradiation effects on ruby laser crystals gave interesting results regarding energy storage effects in ruby crystals. The experimental results which are here reported, showed that the ruby laser output could be significantly increased by the effective use of γ-radiation (For a discussion of various aspects of laser theory see Appendices A, B, and C).

EFFECTS OF IRRADIATION OF SINGLE CRYSTALS

The fluorescent and phosphorescent effects obtained in studies of materials under visible light, x-rays, and γ-radiation, are of particular interest in considering the possibility of producing laser action by employing high energy radiation. Radiation produced center formations have been studied for many years, however, they are still not well understood. This section will present a discussion of the general ideas of color centers in crystals, their formation by x- and γ-irradiation, and the spectral changes of Al_2O_3 due to this resulting center formation.

Color Center Formations

Of particular interest is the early work of Kallman and Furst (1951a, 1951b) studying fluorescence and phosphorescence of certain liquids and crystals after γ-irradiation. These studies showed that fluorescence and phosphorescence may be obtained from certain crystalline materials under various wavelength radiations.

Fluorescence due to high energy irradiation of various crystals has also been extensively studied by Przibram (1956) and his discussion of the presence of F-center[1] and V-center[2] formations are of particular interest. It is known that in crystals there is a possibility of two types of coloring; one is that of normal colloidal coloring, and the other is due to the F-center type of defects normally

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[1]This notation is adapted from the German word "Farbe". The F-centers are associated with the electron traps in the defect structure of various crystals. The presence of these centers produces absorption bands in the visible portion of the spectra of a crystal.

[2]The V-center bands are several in number and are generally associated with absorption bands produced in the ultraviolet region of the spectra by trapped positive holes.

formed in growing the crystal[3]. High energy radiation is one of the more important mechanisms for producing these color centers in crystals which are already formed. In general, longer wave length radiation has a bleaching effect on color centers that have been previously formed by more energetic radiation. In some cases, the bleaching and coloring regions of the electromagnetic spectrum overlap and both processess proceed simultaneously. After long exposure at wave lengths which produce coloration, a saturated condition is generally reached. In many cases, longer irradiations cause the number of color centers to decrease. Except for specially sensitive and sensitized materials, visible light only produces bleaching whereas ultraviolet or shorter wave length radiations tend to produce coloring. If in a radiation-colored material the absorption of the F-band is lowered on exposure to light, it is a matter of experimentation whether there is bleaching or whether new bands are formed. According to experimental evidence (Urbach, 1926), excitation of the F-band causes the formation of a new absorption maximum at a longer wave length. At lower temperatures, the two bands are separated while at higher temperatures the band of the excited states, the F'-band, is not easily distinguished spectrocopically from the F-band. The return to the normal state is accelerated at higher temperatures. In the formation of the F'-centers from the F-centers by irradiation of the F-band, it was observed by Przibram (1956), for example, that in KCl two F-centers disappear with each quantum absorbed. This indicates that an F'-center

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[3]For a discussion of the effects of Cr^{+4} in Al_2O_3 see Hoskins and Soffer (1964).

is formed by an electron, which is released from one F-center, and is again captured by another F-center. According to Seitz (1946), an F'-center is an anion vacancy with two electrons (also see Markham, 1952).

Figure 1 presents a pictorial description of the F- and V-centers as characterized by Schulman and Compton (1962). The color centers can be characterized as electron traps of varying depths. This can be seen by starting with a low temperature colored crystal such as a fluorite, gradually letting the temperature increase, and observing that the brightness of the fluorescence increases and decreases as the temperature rises (Randall and Wilkins, 1945). This suggests the idea of traps at various depths but clustered in a certain characteristic set of bands (related to the various color centers _i.e._ the F, V, M, N, and R-bands).

After radium (gamma)irradiation of CaF_2, Przibram (1956) observed a brilliant thermo-luminescence spectrum corresponding to the lines of the doping ions of Sm, Eu, Dy, Tb, and Er. In samples in which this phenomenon was particularly strong, the known emission lines of the trivalent rare earth ions could be experimentally established. These materials and others which exhibit fluorescence and phosphorescence after γ- and x-ray irradiation may be likely candidates for either energy storage or for direct nuclear pumping of lasing levels.

The work of Kallman and Furst (1951) on γ-irradiated NaCl crystals with an active ion of silver gave very interesting results. Their experiments indicated that the trap depth is dependent upon irradiation time, with the longer irradiation period yielding color center traps of the greater depth. Also, at a given temperature the lifetime of the deeper color centers is longer than that for the more shallow traps.

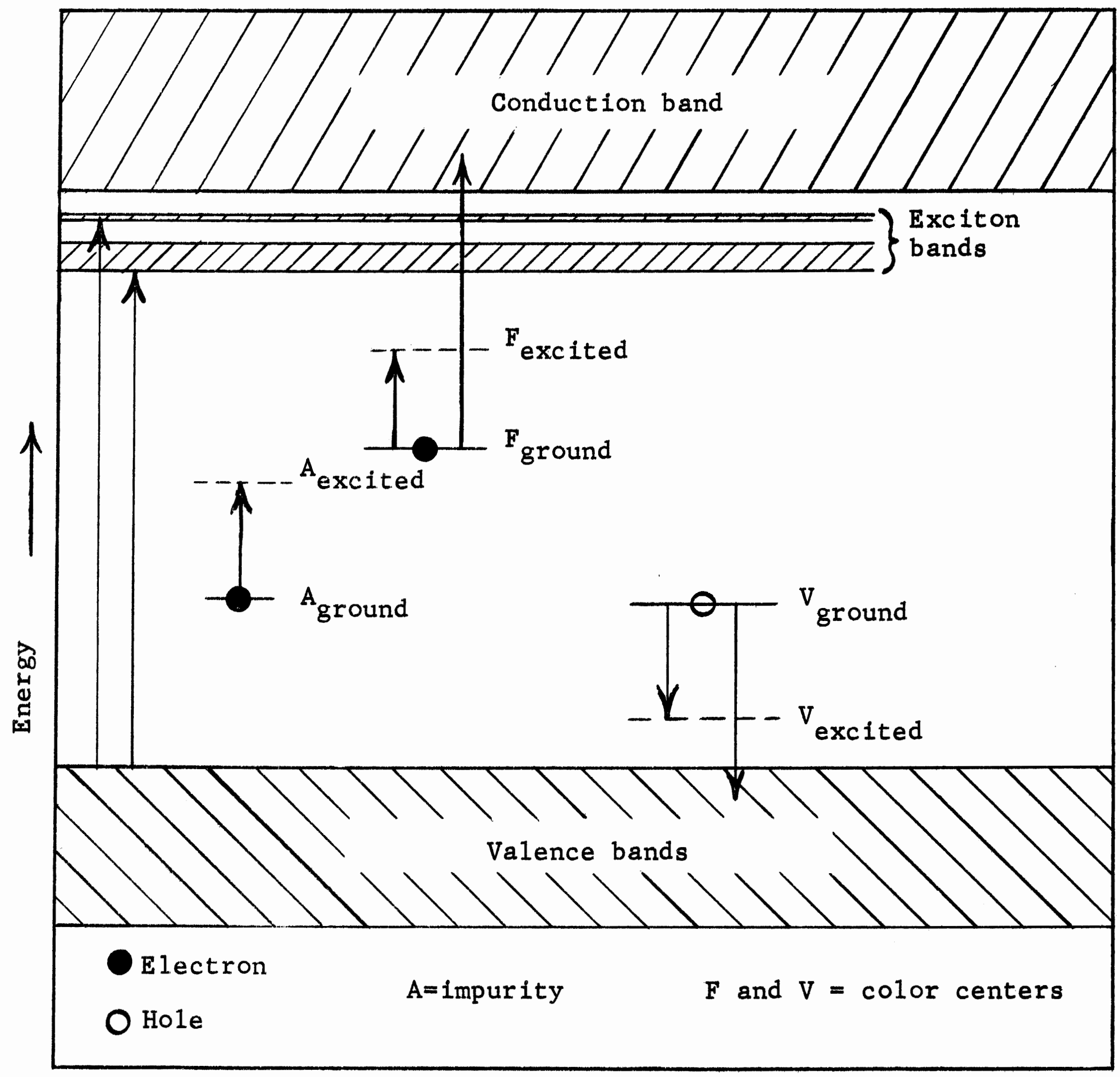

Figure 1. Band picture showing optical absorption due to excitons, impurities, and color centers (after Schulman and Compton (1962) p. 6.)

Phosphorescence, which decays slowly as long as the temperature remains unchanged, was observed after γ-irradiation. When activated with visible light after irradiation with gamma rays, the NaCl crystals yielded an increased phosphorescence which could be attributed to the raising of electrons from the F-center traps to the conductivity band, then the energy proceeds through nonradiative transport processes to the activator ions. Kallman and Furst estimated that in NaCl about 20% of the stored energy which could be emitted as light can be released by light stimulation.

Effects of γ-Irradiation on Al_2O_3

Attention will now be limited to the reported effects of γ-irradiation on the ruby lattice. The studies of Hunt and Schuler (1953) points out several interesting results. They found that x- and γ-ray induced coloration in the Al_2O_3 lattice produced broad absorption bands at approximately 230 and 400 mμ. Two corundum (α-Al_2O_3) crystals, the second of which had higher transmission in the region around 230 mμ, were irradiated. After irradiation, the absorption band at 230 mμ was much stronger in the second crystal. Thermoluminescence of low intensity and pinkish in color was observed when the crystals were heated. Stronger luminescence was observed originating from the second crystal.

The most complete work on color center formation in Al_2O_3 is that by Levy (1961) in which he reports on gamma and reactor induced centers. The bands reported by Hunt and Schuler were found by Levy to be centered at 227 and 405 mμ (full widths at half maximum of approximately 50 and 210 mμ respectively) with an indication of an additional band at 290 mμ (Figure 2).

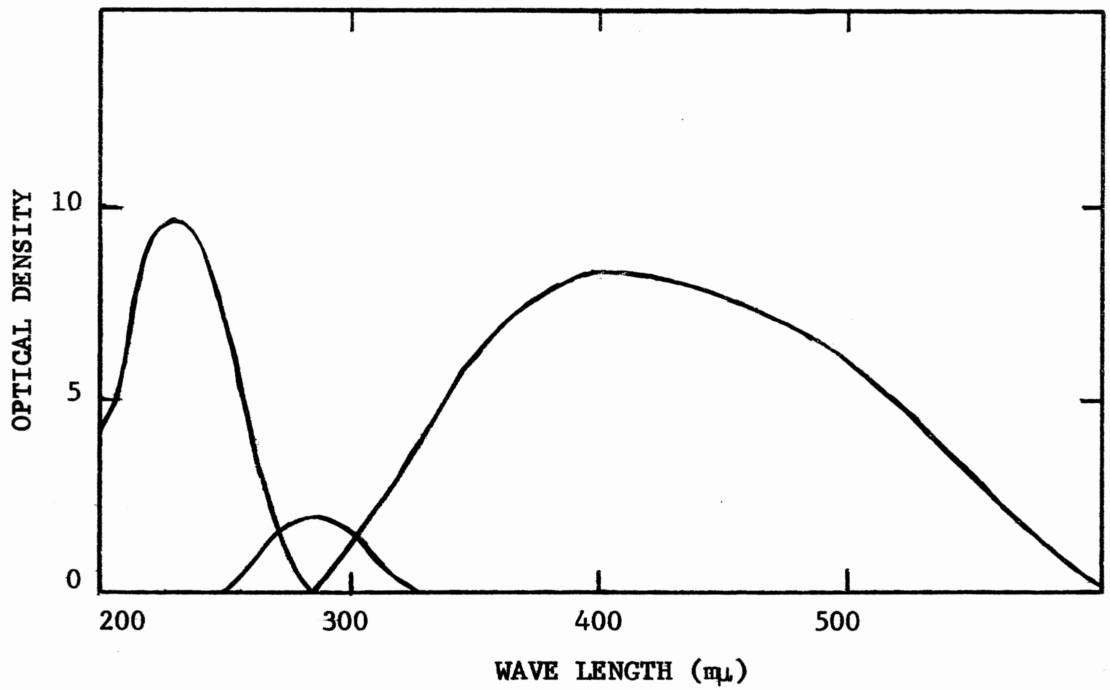

Figure 2. Gamma induced absorption in Al_2O_3 (dosage of 10^9 R-saturation occurs at approximately $3x10^4$ R, after data given by Levy, 1961)

Gabrysh et al. (1962) have studied thermoluminescence in α-Al_2O_3 and in ruby crystals after γ-irradiation. Their studies were primarily concerned with light induced luminescence build up and decay rates after γ-irradiation. The fact that irradiation at liquid nitrogen temperatures produced (Gabrysh et al., 1962, p. 3391) "phosphorescent after glow, long after the crystal has reached room temperature, indicates an energy-storing property in γ-ray damaged ruby, which might be of use to favorably affect 'memory' capabilities and optical laser properties" (also see Davis, et al., 1962, for an earlier study).

No specific models for the gamma induced absorption bands have been given which agree completely with experimental results (see Schulman and Compton, 1962). However, the recent experiments and discussions of Gamble et al. (1964) provide considerable insight into the problem.

THE POSSIBILITY OF NUCLEAR PUMPING OF LASER SYSTEMS

The effects of radiation induced defect structures[4] or various types of center formation in laser crystals providing the possibility of energy storage for laser output enhancement prompted the investigations to be discussed. As has been noted, the center formations appear to be a very likely mechanism by which energy storage can be obtained. However, the problem of utilizing the stored energy for either totally or partially driving a laser system has not been previously reported.

The problem basically involves studies of radiation produced defect structures of crystals in order to select those which (a) possess characteristics that may lead to useful energy storage in the various centers formed and, (b) also possess the combination of nonradiative transitions necessary for placing a large number of the activator ions in a metastable state such that the lasing process could occur. As a first criterion, the crystals chosen for study should be known to fluoresce and phosphoresce upon irradiation with x-rays, gamma-rays, or other high energy radiation. In order to be useful in increasing laser output, it is necessary that the newly-formed conduction band electrons and excitons contribute energy to the pumping absorption bands of the material to be lased. The energy stored in the centers would in turn be transferred by nonradiative transitions to the fluorescent levels. It is then necessary that these processes contribute to a population inversion of the lasing fluorescent level by this or some other means.

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[4] Irradiation of single crystals produces or ionizes lattice defects. These ionized defects are often referred to as various types of centers (Gamble, et al., 1964).

Also, it may be remarked that irradiation by neutrons could possibly be valuable if it is found desirable to create a larger number of vacant lattice sites which could give rise to additional centers.

An idealized energy diagram for coupling gamma irradiation with laser systems is given in Figure 3. The probably transitions are indicated with the color center bands shown as a single band. The color center band could also be considered as a set of bands, slightly below the energy of the conduction band, which depends on the color center trap depths.

In Figure 3, the W's represent transition probabilities per unit time for energy transport to the indicated level; the Q's represent quenching transition probabilities per unit time; the T's represent nonradiative energy transport probabilities; the A's represent the transition probability per unit time for spontaneous emission; and the S represents the transition probability per unit time for the stimulated or induced emission. In order to make quantitive use of this scheme, one would need to determine the various transition probabilities involved and then formulate a set of rate equations similar to the familiar steady state rate equations (Maiman, 1961). However, in general, the steady state conditions do not exist. This approach could give an approximation to the minimum condition for population inversion for a nuclear energy coupled laser system.

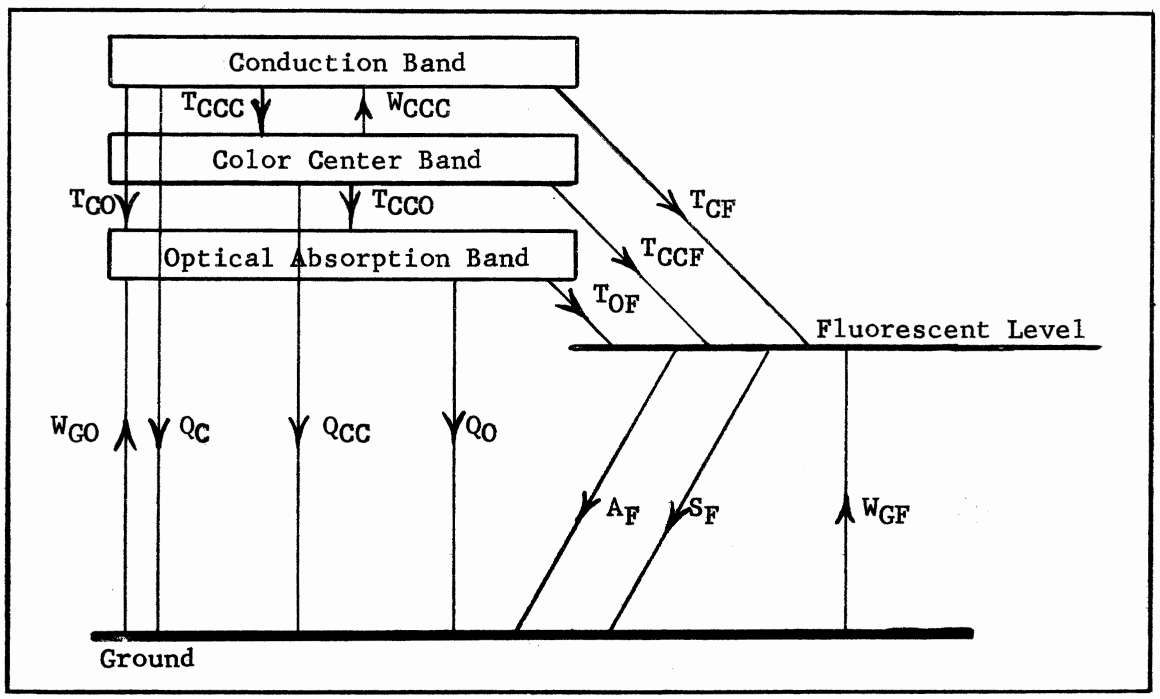

Figure 3. Idealized energy level diagram for partially nuclear driven laser

THE EXPERIMENTAL INVESTIGATION

The results of the experimental program which are reported in the remaining portion of this work utilized ruby laser crystals. Other laser materials were not available.

Early Experiments

Laser quality ruby crystals were irradiated with Co^{60} γ-rays. After a dosage of approximately 100,000 R, the transmission spectrum of the ruby laser crystal was considerably changed in the region below 550 mμ. However, the primary laser pumping band around 550 mμ as well as the band encompassing the ruby R_1 and R_2 lines was not significantly altered (see Figure 4).

Irradiation of the ruby laser crystal with Co^{60} γ-rays also resulted in a brilliant phosphorescence which decayed over a period of several hours after removal from the irradiation facility. The phosphorescence was of sufficient intensity for the ruby crystal to be used as the light source in a spectrophotometer to obtain an emission spectrum (Figure 5). The emission spectrum shows a relatively strong peak centered around the natural ruby lasing lines. Thus, energy was stored in the ruby laser crystal and, through what appear to be nonradiative transport processes, a part of it was transferred to the lasing levels producing the observed phosphorescent decay.

After the phosphorescence due to the γ-irradiation had decayed, it was found that excitation with an intense xenon light source caused the phosphorescence to again increase to near the original level (just after removal from the gamma source). However, the time decay on this phosphorescence was considerably more rapid. The phosphorescence could be

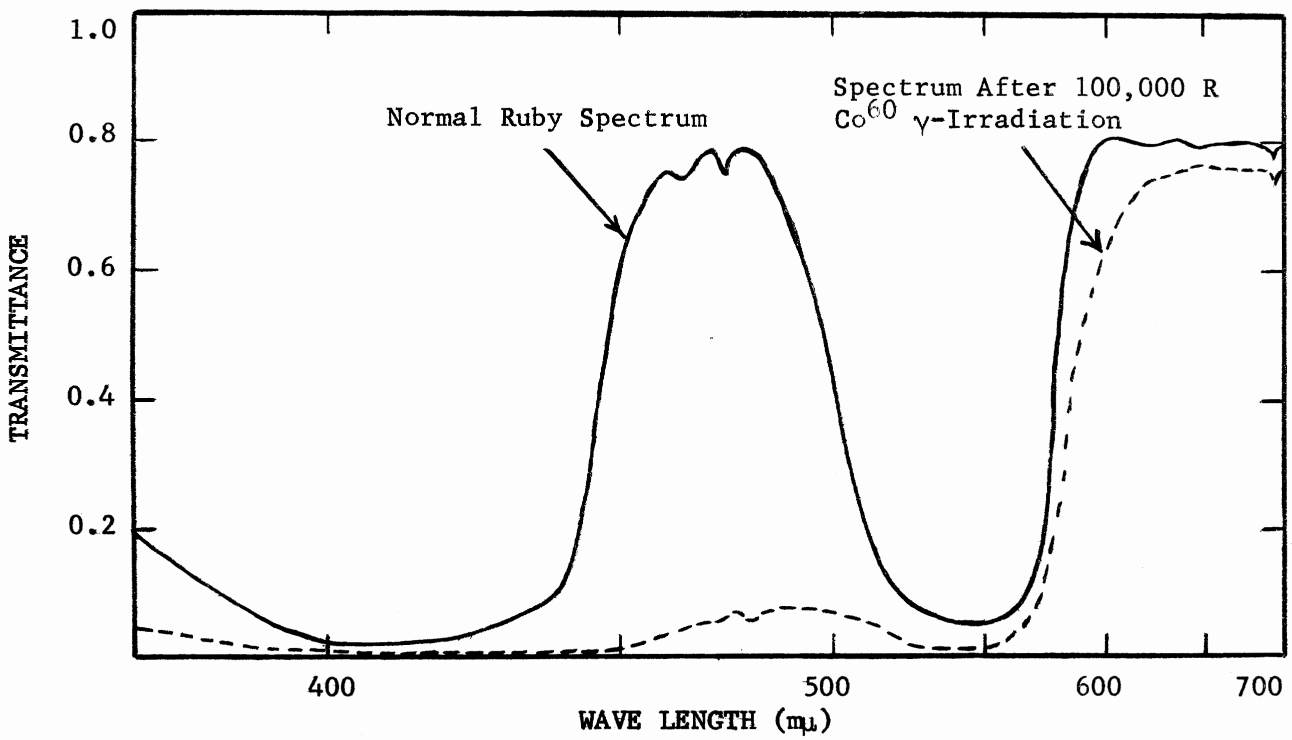

Figure 4. Typical spectra of ruby showing the effect of gamma-ray irradiation

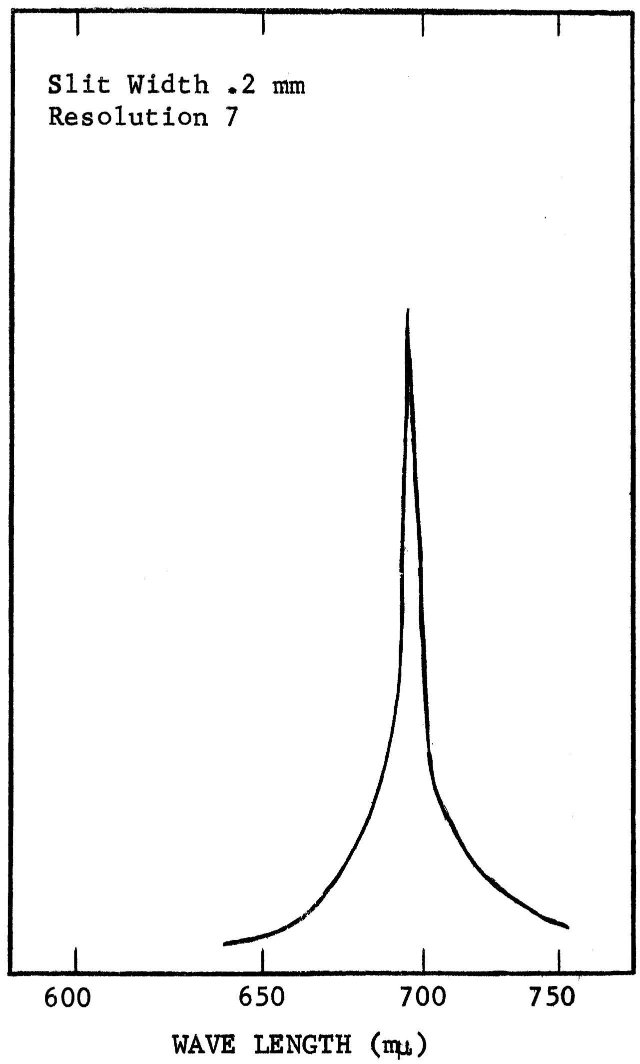

Figure 5. Phosphorescent emission spectrum of ruby after 100,000 R Co^{60} γ-dose

increased again upon each of several intense light flashes but to a lesser degree in each succeeding attempt. These early experiments with ruby indicated the need for a systematic investigation of radiation effects on ruby laser crystals and their subsequent laser action.

In particular, if the gamma irradiated ruby transmission spectra changed considerably in the region around the R_1 and R_2 lines, or even around the pumping region of 550 mμ, then further experiments with ruby would not appear interesting.

Initial Experimental Investigation of the Effects of γ-Irradiation on Laser Action

The question of using the energy stored in the ruby crystal by γ-irradiation to enhance the laser energy output was the object of a series of experiments. The results of these experiments are discussed in the following section.

Experimental Arrangement. For the first series of experiments, a Maser Optics Laser System (Model 600) with a modified head assembly was employed with a dielectric coated 1/4 x 2 in. ruby rod (0.05% Cr^{+++}) with a 90° orientation to the crystal axis. The threshold for the ruby crystal was approximately 230 joules. All of the experimental data was taken at an energy input level of 270 joules. A laser detector system employing an RCA 925 phototube was used. The output of the ruby laser spikes detected by the phototube was also electronically integrated (Figure 6) to give an oscilloscope trace which is proportional at any instant to the total energy output. The output of the phototube was displayed on a Tektronix 555 dual beam oscilloscope with one channel displaying the pulses of the laser output and the other channel showing an integrated output of the ruby laser spiking.

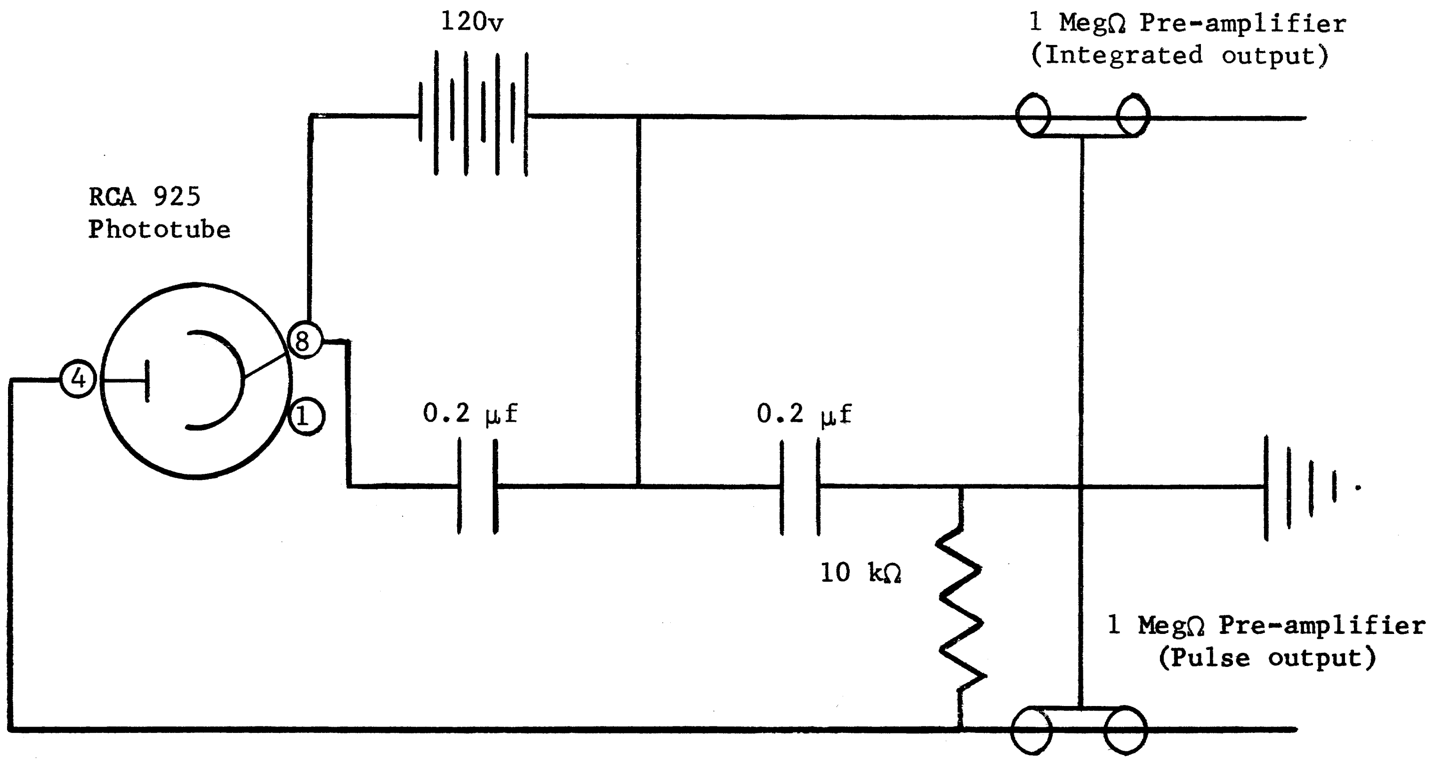

Figure 6. Schematic diagram of the phototube detector

The laser head employed with the Maser Optics System used two linear FX-38 xenon flash tubes. The flash tubes were positioned adjacent to the ruby laser crystal and 180° apart. They were closely coupled with the ruby laser to form the optical pumping cavity. A high voltage pulse was used to discharge the flash tubes which were connected to a 50 μf capacitor bank charged to a potential of 3.28 kilovolts. Therefore, the energy input to the flash tubes was 270 joules for these experiments. The head was cooled by an air blower and five minutes was allowed for cooling after each firing.

Figure 7 presents a diagram of the optical system. Two Fresnel plates were used to intercept the laser beam and direct a fraction through a 10 mμ wide filter centered about 694 mμ. This portion of the beam then fell on the phototube to yield the pulsed and integrated output signals.

Spectrographic data were taken on a Perkin-Elmer Model 350 recording spectrometer. Visible and ultraviolet transmission spectra of the ruby crystal were taken before and after γ-irradiation in an effort to determine the effect of γ-irradiation in forming centers. Spectra were also taken after laser firings in an effort to determine the effectiveness of the pumping light of a normal laser system in releasing the energy stored in the radiation produced centers. However, the spectral data obtained has not given sufficient information to allow interpretation of these problems.

Increase in Laser Output after γ-Irradiation. Intensive studies of the ruby laser crystal were made before γ-irradiation. Spectroscopic analysis of the ruby laser crystal was performed and calibration firings of the laser were made to investigate reproducibility of the measurements.

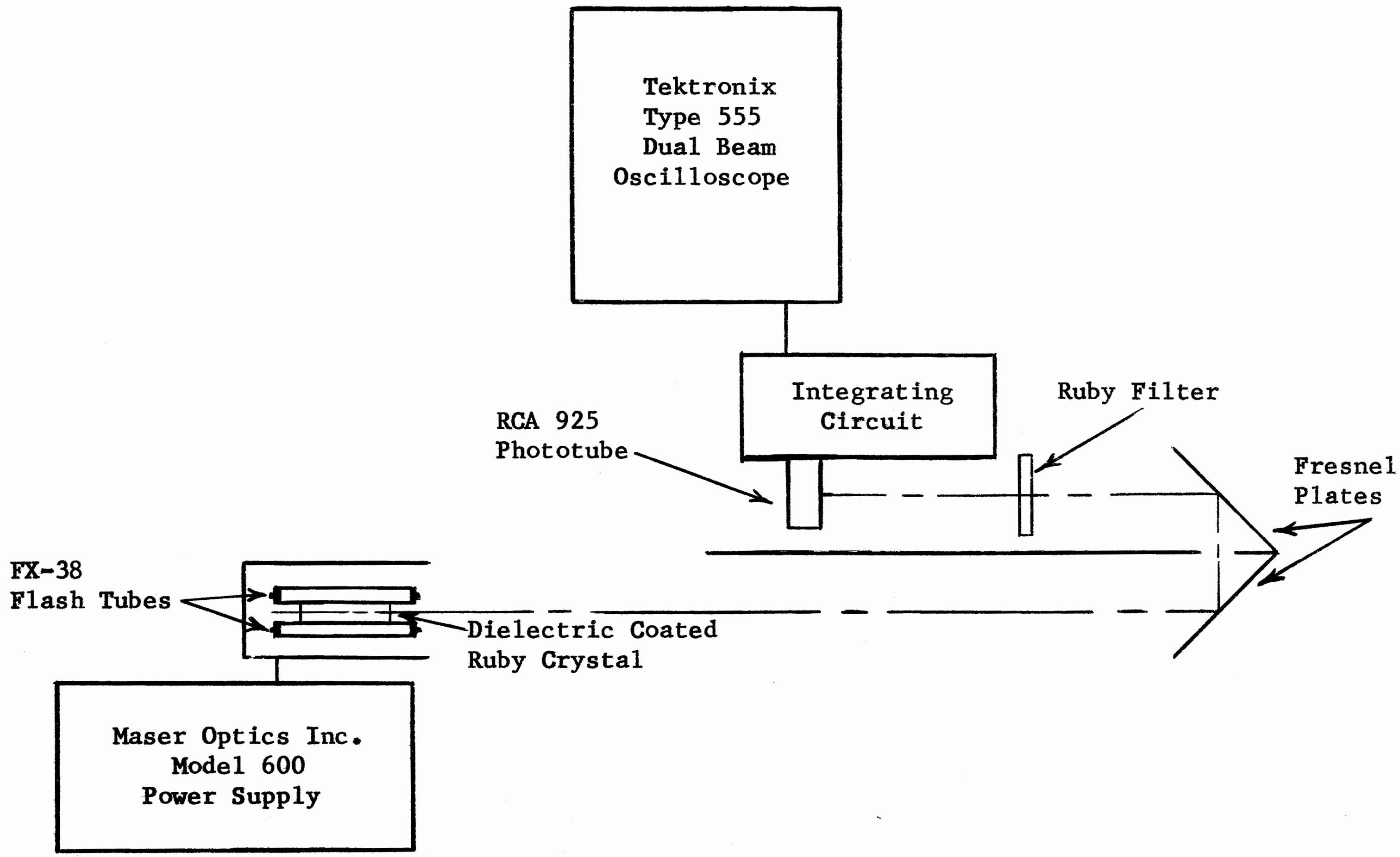

Figure 7. Diagram of experimental system

The calibration firings of the laser prior to γ-irradiation were quite reproducible with no detectable change in the integrated output of the detector. After a consistent set of calibration data was obtained (see Figure 8(a) for a typical calibration firing), a five second irradiation in the Co^{60} gamma facility was made. The total dose from this irradiation was approximately 450 roentgens. The first lase after irradiation revealed that a large increase in the laser output had been obtained (see Figure 8(b)). The integrated output and the individual ruby laser spikes are increased to the point that they are off scale. Figure 8(c) shows the laser output after several more firings with the gain of the vertical amplifiers reduced by a factor of five in order to reduce the signal sufficiently for displaying the trace on scale. Several additional laser firings were made with a slight decrease in energy output apparent in each lase. A single firing of the ruby laser crystal in a larger head at 5,000 joules input significantly reduced the subsequent energy output when the crystal was again fired in the smaller head assembly[5]. A number of firings of the laser crystal were made and the laser output was reduced to about one fourth of the magnitude of the output after the first irradiation. By significantly increasing the γ-dosage, the laser energy output was eventually reduced below its pre-irradiation level, indicating the existence of competing detrimental effects at higher dosage levels. Although the desired effect had definitely been obtained, the need for additional and more carefully controlled experiments was evident.

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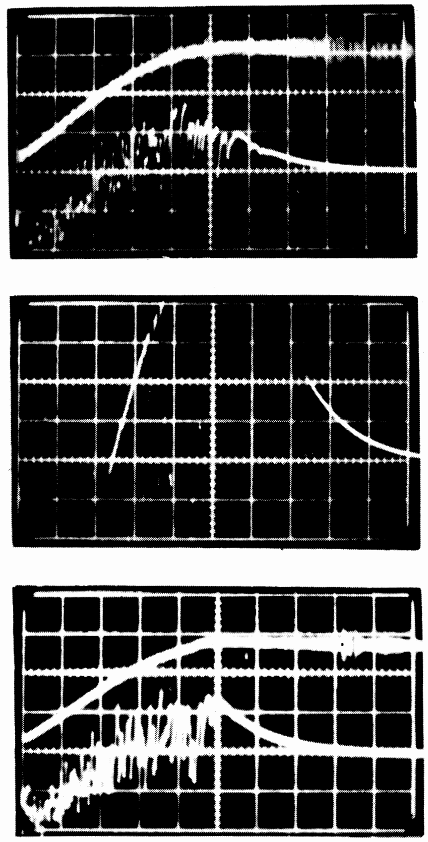
[5]When the crystal was fired in the larger head, the laser output was considerably larger than would be normally expected. The fact that the laser output was subsequently reduced would be an expected effect of an energy storage mechanism.



(a)

(b)

(c)

Figure 8. Ruby laser output showing the increase in energy output due to γ-irradiation: (a) calibration (b) after 450 R γ-irradiation (c) same as (b) but vertical scale reduced by a factor of five

Other Experimental Investigations of γ-Irradiation Effects on Laser Action

Some of the results of further experiments are presented in this section along with additional data giving more detail than the previously described measurements. Also, these results were obtained with a different experimental arrangement which minimized possible undesirable effects due to system misalignment.

Experimental Arrangement. In order to eliminate any effect which radiation might produce on ruby dielectrics, external dielectric reflectors of 99% and 75% reflectivity were employed. A single FX-100 flash tube was used with power provided by a Laser Systems Center LS-4 system employing a capacitor bank of 585 μf charged to 1,000 volts (total energy of 293 joules input). The flash tube and crystal were tightly coupled and the flash tube was triggered by the ordinary capacitive coupling method. The ruby crystal was of a 0° orientation cut to the crystal axis, 1/4 x 3 in., containing 0.04-0.05% Cr^{+++}. The energy threshold for lasing action was approximately 140 joules.

The measurements made during this set of experiments employed the same pulse and integration circuit used in the earlier experiments. In addition, a separate detector in the form of a TRG Thermopile Model 101 was used. The TRG Thermopile makes calorimetric measurements of the energy of the laser beam by means of absorption of the beam energy by a Mendenhall wedge. The instrument was earlier calibrated against a National Bureau of Standards source. The dielectrics and the laser crystal were aligned with a Davidson Optronics Autocollimator. Figure 9 gives a diagram of the optical set up.

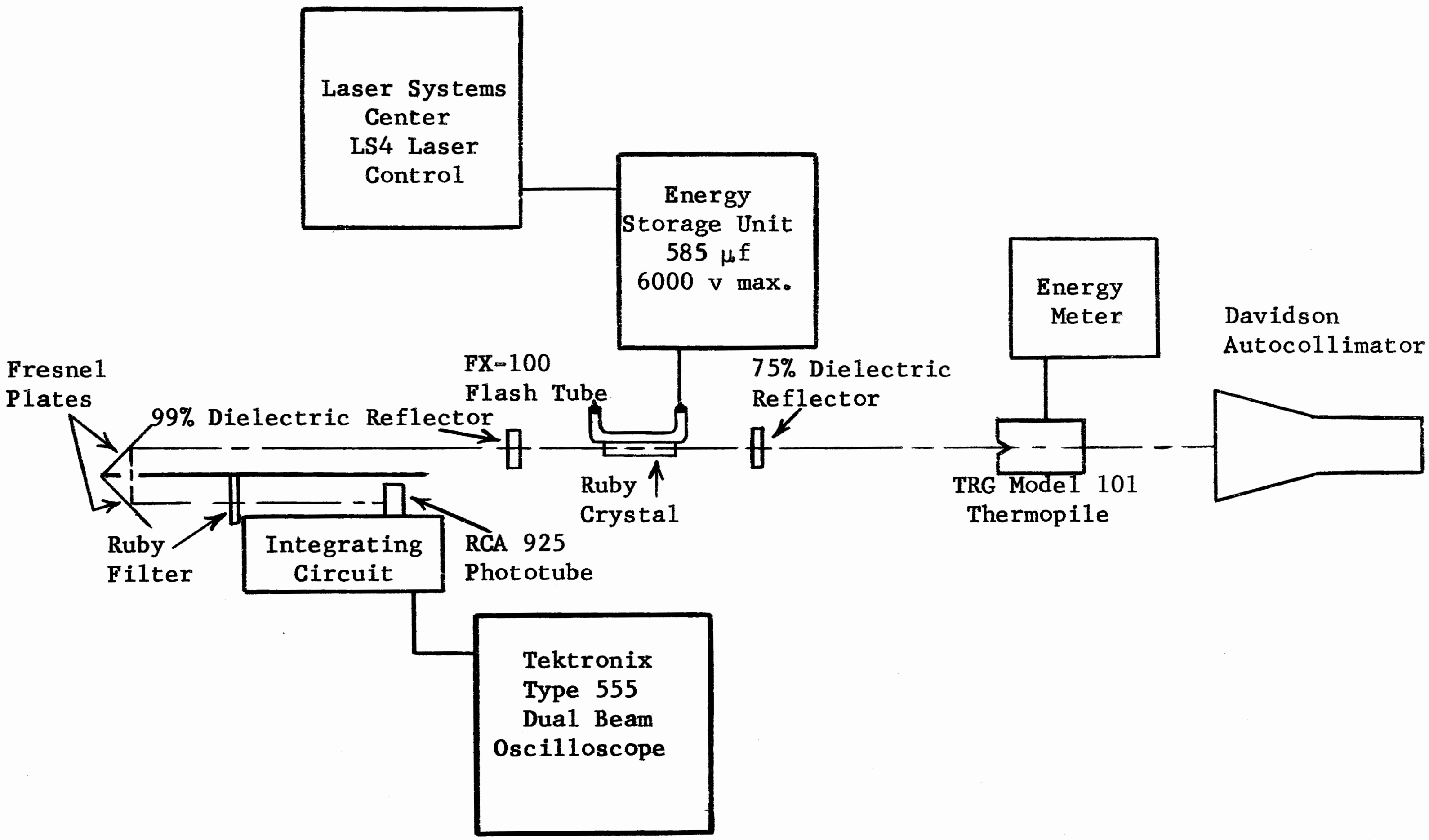
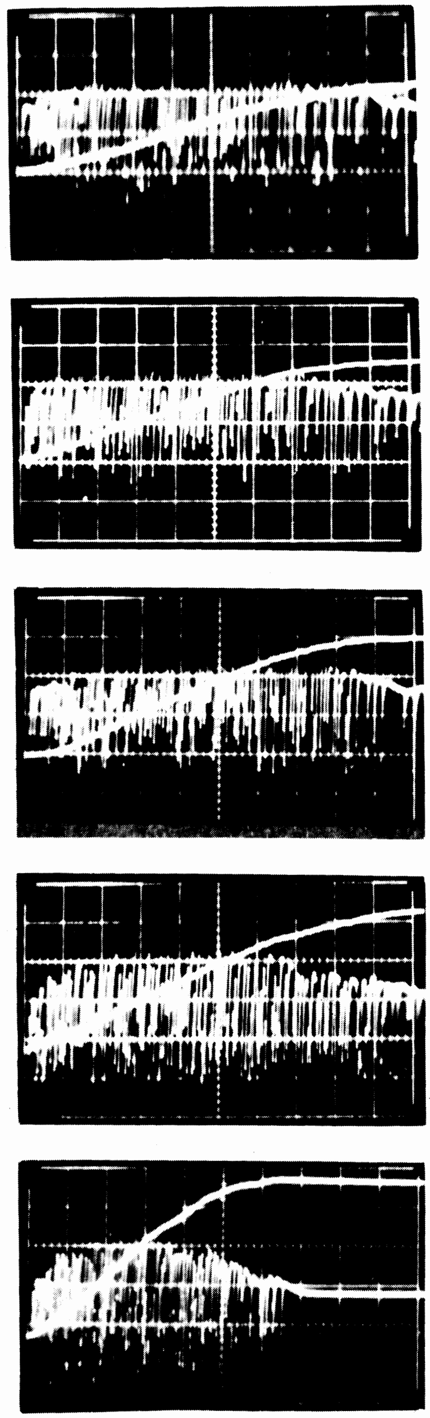


Figure 9. Experimental arrangement for the experiments employing external dielectric reflectors

Data Presentation. A series of pre-irradiation calibration firings were made. No detectable variation in the laser output energy from the phototube detector could be noted and a maximum variation in the output energy as measured by the TRG calorimeter was 5% (see Figure 10(a)). The calorimeter yielded an average value of 0.076 joules for the calibration laser firings. After a five second γ-irradiation (450 R), the laser output increased slightly. The energy output was measured to be 0.083 joules (the maximum error for which the TRG calorimeter is rated is 5%). The output of the phototube detector appeared to have increased (see Figure 10(b)). After a waiting period of a few hours, the laser was again fired and the output had increased to 0.110 joules. The corresponding phototube detector output may be observed from Figure 10(c). After an additional 450 R of γ-irradiation, the laser output increased to 0.180 joules. The corresponding output from the phototube detector is shown in Figure 10(d). Thus, after a gamma dosage of 900 R, an increase in energy output of about 137% was observed. Another five second irradiation yielded an output of approximately 0.190 joules. Due to the dosage increments chosen in this experiment, it was thought that the optimum dosage (for maximum energy output) had been passed. The laser crystal was therefore bleached for five minutes under black light. The laser output on the next firing increased to 0.215 joules (19% over the previous firing and 183% above the pre-irradiation firing, see Figure 10(e)). Five minutes additional ultraviolet radiation under the black light caused a further increase of 8% to .233 joules (207% above the pre-irradiation firing).

Another series of experiments were performed using the same experiments just described but using a different ruby laser. The ruby used



(a)

(b)

(c)

(d)

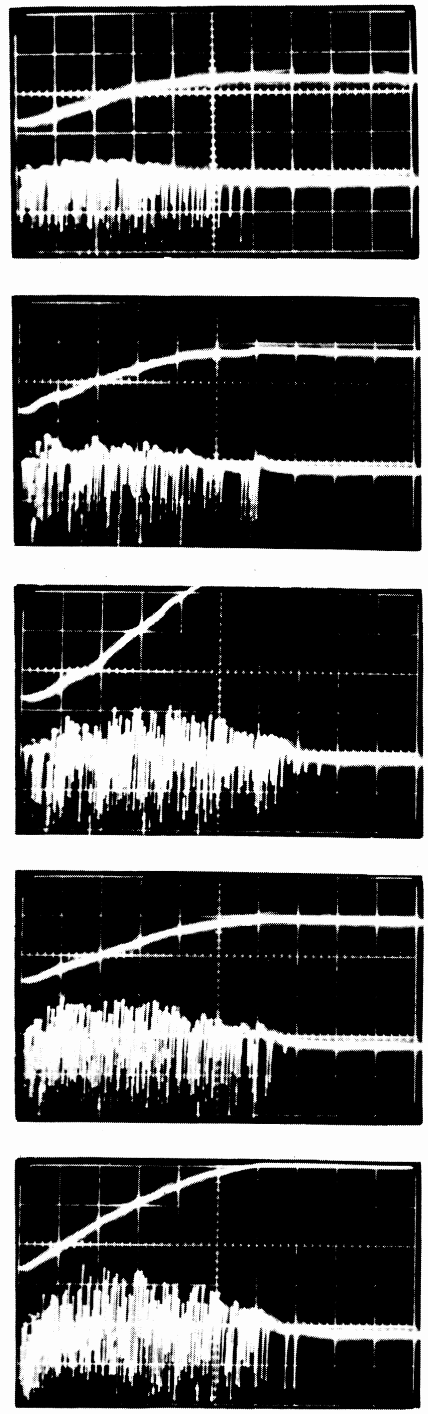
(e)

Figure 10. Phototube output showing the increase in ruby laser output with γ-irradiation--time scale reduced in (e) by factor of two

in this set of experiments was 3 in. long x 3/16 in. diameter with 0.04-0.05% Cr^{+++}. Figure 11(a) presents a typical pre-irradiation calibration firing of this ruby laser crystal. The energy output for this firing as measured by the TRG calorimeter was taken to be one unit. The same ruby crystal was then irradiated with Co^{60} γ-rays receiving a total dosage of approximately 800 R. Figure 11(b) presents a typical post-irradiation firing of the ruby crystal with the same input energy. The integrated energy output, shown in Figure 11(b), clearly indicates a definite increase in output energy. The relative energy output measured by the TRG calorimeter was for the firing shown in Figure 11(b), indicating an increase in laser output of approximately 50% after the first irradiation.

Figures 11(c), 11(d), and 11(e) contain the results of the laser output (as determined from the RCA 925 phototube) as a function of total accumulated Co^{60} gamma dosage — with constant energy input to the laser system. The magnitude of the individual ruby laser output spikes and their number per unit time are greatly increased after irradiation. Both of these effects cause the observed increase in the total integrated energy output as a function of the gamma dosage.

The energy output measured by the TRG calorimeter, which correspond to Figure 11(a)-(e), are given in Table 1. Figure 11(e) and Table 1 show that the ruby laser energy output increased by approximately a factor of three after a total Co^{60} gamma dosage of 1700 R. Other results using the same ruby are presented in graphical form in Figure 12. Indications are that a maximum laser output energy occurred, however, its exact value was not observed due to the dosage increments chosen. Measurements were also made at higher total radiation dosages. It was



(a)

(b)

(c)

(d)

(e)

Figure 11. Oscilloscope traces of the enhancement in ruby laser output with increasing Co^{60} γ-dosage. The upper traces in the sequences of figures compare the total or integrated energy of the ruby laser spikes. For (d) and (e) the integrated output is reduced by a factor of two from that of (a),(b) and (c). Horizontal scale 100 μ sec per major division

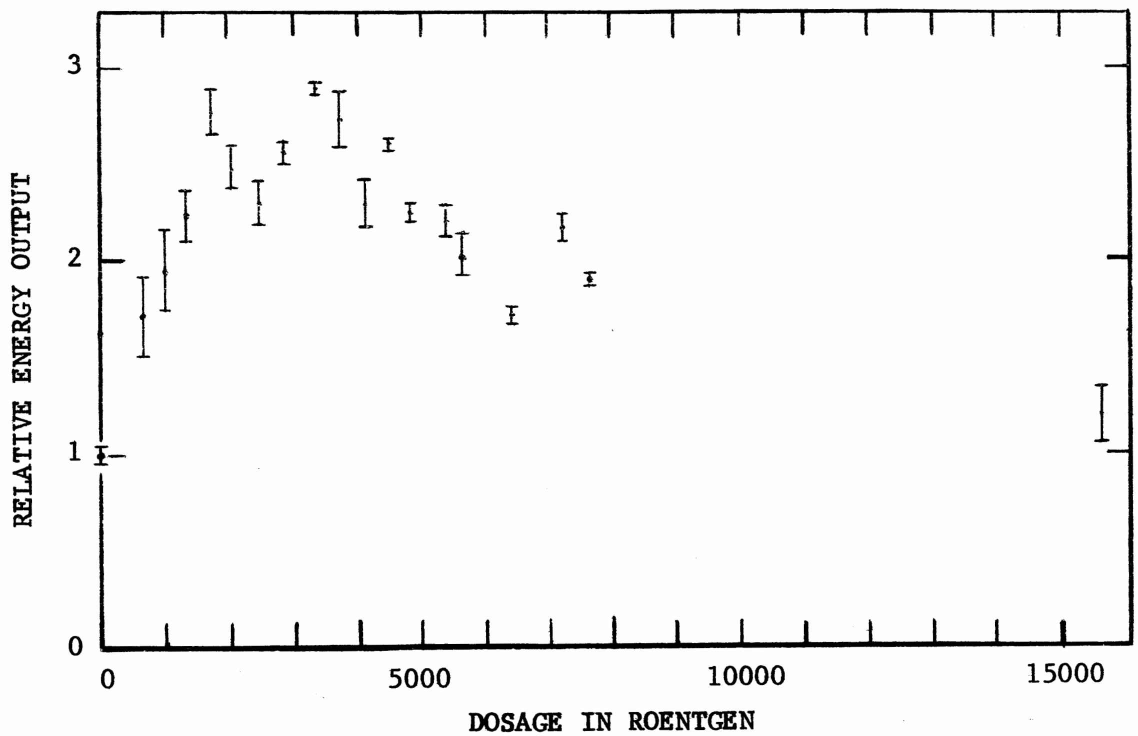


Figure 12. Ruby laser output as a function of Co^{60} gamma dosage

found that after a total accumulated dosage of approximately 15,000 R the ruby laser output was essentially reduced to its pre-irradiation output level. Qualitative experiments indicated that with large gamma dosages, laser action either ceases or the input energy threshold increases substantially.

Table 1. Energy output of ruby laser as a function of Co^{60} gamma dosage

Gamma Dose (Roentgen)	Calorimeter Energy Output (Relative)	Comparison Figure 12
0	1.0	a
700	1.7	b
1,000	2.0	c
1,400	2.2	d
1,700	2.8	e

It should be noted that the increased energy output which was observed in all of these experiments have been obtained by pumping only a part of the laser crystal. For example, in the last two experiments in which increased outputs of factors of about three were observed, the laser crystal was pumped with only one flash tube. Thus, with the close coupling between crystal and flash tube, only a portion of the laser was pumped. Note that in the first reported experiment, flash tubes 180° apart were used to pump the ruby — with the result that an increase in energy output by a factor of five was obtained. Thus, increases in energy output with heads offering improved pumping efficiencies should be substantially higher than has been obtained thus far. Further experiments to investigate this and other phenomena are now underway.

SUMMARY AND CONCLUSIONS

The results obtained show that the energy output of ruby laser crystals can be significantly increased through the effective use of γ-radiation. The laser output energy has been increased by approximately a factor of five — considering the phototube detector output as approximately linear with incident energy. In other series of experiments which were reported, the laser output was simultaneously monitored by two independent detector systems. In these experiments, the laser output energy was increased by approximately a factor of three. This is not, however, the maximum effect that can be realized. The maximum output was not observed due to the dosage increments chosen and also because of certain non-optimum experimental arrangements which were necessary at the time. Results have shown that at higher dosages, a competing effect occurs. At dosages on the order of 10^4R (corresponding approximately to the center saturation dosage) or greater, the competing effect is as strong as the useful effect of the stored energy.

The spectral results obtained have not been of sufficient quality to allow detailed study of the processes involved in the observed effects. However, the results obtained by Levy (1961) and shown in Figure 2 for the case of radiation induced color center saturation indicate possible bands which could contribute energy to the lasing process. The precise mechanism involved in the energy storage and transport process is not well understood. However, the mechanism may be tentatively explained by what has been called (Gabrysh et al., 1962, p. 3391) "light-induced thermoluminescence" or (Przibram, 1956, p. 118) "radio-photoluminescence."

RECOMMENDATIONS FOR FURTHER STUDY

The major problem remaining to be investigated is a precise explanation of the mechanism involved in the energy storage and transport mechanisms. Extensive defect structure studies of various crystalline materials will help in understanding the general character and effects of radiation induced centers. Also, the increase in energy output of ruby crystals has by no means been optimized. Other parameters which may possibly be key factors in optimizing the laser output are: the concentration of the Cr^{+++} doping ions, the operating temperature, the wavelength of the pumping energy, the laser head configuration, and the input energy level. The spectral changes due to center formation have not been adequately correlated to the optical bleaching caused by the laser firings. After an exhaustive study of the effects of ionizing radiation has been completed for ruby, further studies of other radiation induced effects in crystals (e.g. Nd doped glass and CaF_2 doped with various rare earth ions) would promise to be of interest.

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A P P E N D I C E S

APPENDIX A

BRIEF INTRODUCTION TO THE BASIC THEORY OF LASERS

Due to the very recent development of the laser and because of its importance to a wide variety of fields, an investigation of the general ideas involved will be given. A discussion of the possibilities of either partially or totally nuclear pumped laser systems can then be considered.

The basic ideas of the theory of spontaneous and stimulated emission of radiation are first presented. The approach taken here is essentially from thermodynamics, originally given by Einstein (1917) and interpreted by Troup (1963).

For a system of molecules in thermal equilibrium, the energy density of radiation per unit frequency range, u_ν, is related to the total energy density, u, by

$$\int_0^\infty u_\nu d\nu = u.$$

The Plank radiation formula relating the radiation frequency and energy density may be written as

$$u_\nu \, d\nu = \frac{8\pi\nu^2}{c^3} \left\{ \frac{h\nu}{\exp\,(h\nu/kT)-1} + \frac{h\nu}{2} \right\} d\nu \ ,$$

where $\frac{8\pi\nu^2}{c^3} d\nu$ is the number of modes of oscillation, the bracketed term is the average energy of the oscillation mode, and $\frac{h\nu}{2}$ is the zero-point energy. At optical frequencies the relation $h\nu \gg kT$ is valid for ordinary values of T. Therefore, the average energy per mode reduces to $\frac{h\nu}{2}$.

Einstein postulated that the Plank radiation law was obeyed by the energy state transitions for both emission and absorption. In making an energy transition between the states m and n, a quantum of radiation, whose energy is $h\nu_{mn} = E_m - E_n$, is either emitted or absorbed. In the equilibrium case $h\nu_{mn} = h\nu_{nm}$, and the probability of an atom being in the state m or in the state n is given by

$$P_m = a \exp(-E_m/kT),$$

$$P_n = b \exp(-E_n/kT).$$

The probability of a downward transition due to spontaneous decay from a metastable level may be written as

$$dP_{mn} = u_\nu B_{mn} dt,$$

and the probability for an upward transition is

$$dP_{nm} = u_\nu B_{nm} dt.$$

The thermal equilibrium condition along with the Plank radiation law then yields

$$B_{nm} = B_{mn}; \quad A_{mn} = \frac{8\pi h\nu^3}{c^3} B_{mn} ;$$

where A_{mn} and B_{mn} are the Einstein coefficients for spontaneous and induced emission respectively.

In the degenerate case, Einstein's first relation takes the form

$$g_n B_{nm} = g_m B_{mn} \quad ,$$

where g_n and g_m are the multiplicities of the n and m levels respectively. To be more explicit, Lengyel (1962) includes the index of refraction, η, in Einstein's second relation,

$$A_{mn} = (8\pi h\nu^3 \eta^3/c^3)B_{mn} .$$

In thermal equilibrium a large number of atoms, N_o, obey the Boltzmann statistical distribution of atomic states;

$$N_m/g_m = N_n/g_n \exp\left(-\frac{E_m - E_n}{kT}\right),$$

where the g_m and g_n represent the multiplicities of the m and n states respectively. It is evident that N_m is less then N_n for m greater than n and that for this case the combined number of spontaneous and induced downward transitions, $(A_{mn} + u_\nu B_{mn})N_m$ quanta per second, exceeds the total number of induced upward transitions, $u_\nu B_{nm} N_n$ quanta per second. The net effect is the absorption of $(N_n - N_m)B_{mn} u_\nu$ quanta per second. However, if N_m is greater than N_n a population inversion exists and in the Boltzmann distribution above, there appears what is called a negative temperature or a negative temperature state (cf. Singer, 1960). In this case the above formulas give a negative absorption indicating that radiation may be amplified by stimulation when a population inversion exists. Therefore, by producing an inversion of the number of ions in a higher energy state over those in the ground state, radiation of the wavelength corresponding to the fluorescent line may be amplified for a time interval during the earlier part of the half-life of the fluorescent transition. The ions are normally placed in the higher energy states by a pumping light source of high intensity. The energy from the pumping light source is absorbed in a broad absorption band and non-radiative transitions to the narrow fluorescent line occur in a time interval which is neglible compared to the half-life of the fluorescent transition. The well-defined fluorescent levels correspond to a longer wavelength than the broad absorption band with the difference in energy being that of the radiationless transitions together with the differences in band widths.

The output obtained from a laser occurs as the laser light traverses the crystal, stimulating other active ions to emit their quanta of energy. Thus, the laser output is coherent since stimulated emission occurs in phase with the stimulating radiation. The small line widths associated with the fluorescence transitions of interest cause the laser output to be nearly monochromatic. According to Lengyel (1962), the line width of .01 mμ is attributed in part to a frequency shift due to warming during the laser firing. Also Zeeman broading of the levels due to the magnetic field generated from the current in the flash tube has been considered (see Heavens, 1964). Also, due to the configuration of the laser rod or radiation cavity, highly directional laser beams may be obtained. This allows the transport of high energy densities with little dispersion.

APPENDIX B

THE THREE STAGE RUBY LASER

There are two basic types of solid state lasers, the three and the four stage. The work which will be reported was performed with ruby, a three stage laser, whose active ions is Cr^{+++} in the Al_2O_3 lattice. In the energy level diagram for the three stage ruby laser (Figure 13), the broad $^4F_2$ and $^4F_1$ bands correspond to absorption maxima in the green and blue portion of the spectrum respectively.

The $^4F_2$ band centered in the green region of the spectrum around 550 mμ absorbs the pumping energy in the laser mechanism of ruby (Maiman et al., 1961). Immediately after the Cr^{+++} ions are pumped to the $^4F_2$ energy band, nonradiative processes allow transitions to the ^2E state. The ^2E state is composed of two close-lying states, the E state associated with the R_1 line of 694.3 mμ and the $2\bar{A}$ state associated with the R_2 line at 692.9 mμ. Normally the R_1 line, with a half-life of about 3 milliseconds, is more highly populated during the ruby laser process (cf. Lengyel, 1962). The metastable $\bar{E}$ state of the excited Cr^{+++} ions may be stimulated to emit its energy (at 694.3 mμ) during its short lifetime. The build up of this emitted radiation produces a laser beam.

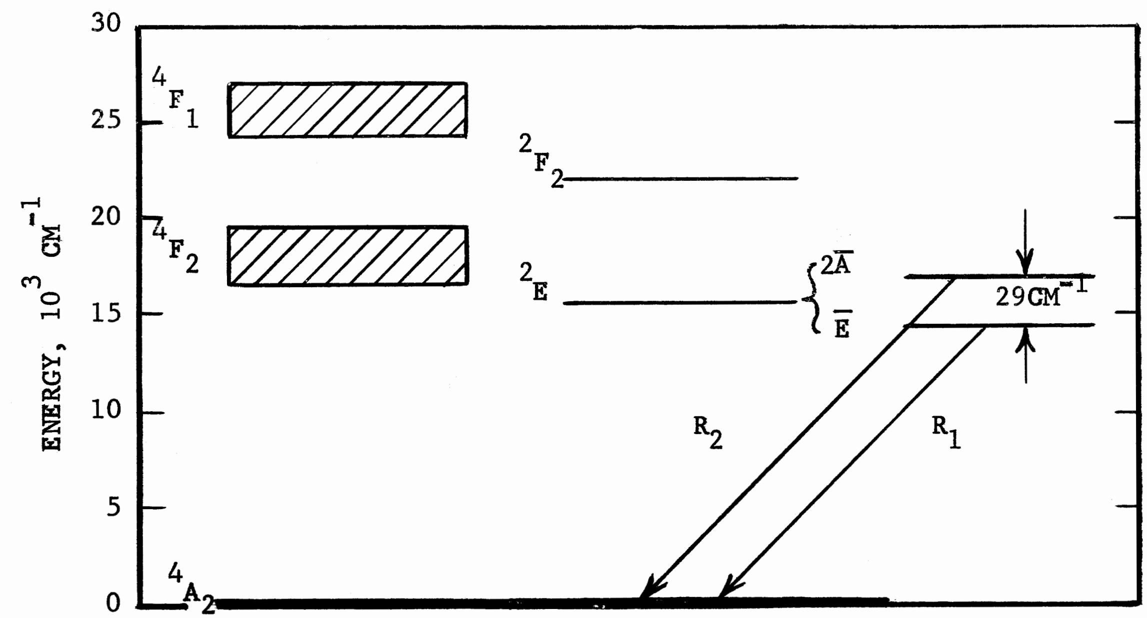

Figure 13. Energy level diagram for a ruby laser (after Maiman *et al.*, 1961)

APPENDIX C

USE OF STEADY STATE RATE EQUATIONS

It is interesting to look at the scheme of Maiman (1961) for writing steady state rate equations in order to obtain the minimum conditions for laser action in a three-level laser such as ruby. The steady state rate equations for a system (Figure 14) such as this are:

$$dN_3/dt = W_{13}N_1 - (W_{31} + A_{31} + S_{32})N_3 = 0,$$

$$dN_2/dt = W_{12}N_1 - (A_{21} + W_{21})N_2 + S_{32}N_3 = 0,$$

$$N_1 + N_2 + N_3 = N_0,$$

where S_{32} represents a probability per unit time of the non-radiative processes, the A's represent the Einstein coefficient for spontaneous emission, the W's represent the indicated transition probabilities per unit time and the N's represent the number of ions in each state at a particular instant of time. Solution of the above equations for the ratio of the population of the excited state to the population of the ground state yields:

$$\frac{N_2}{N_1} = W_{13} \quad \frac{S_{32}}{W_{31} + A_{31} + S_{32}} \quad + W_{12} \qquad (A_{21} + W_{21}).$$

In a material such as ruby, the fluorescent quantum efficiency is high, therefore, $A_{31} \ll S_{32}$ and $W_{31} \ll S_{32}$. Thus the expression above may be reduced to the form

$$\frac{N_2}{N_1} \simeq \frac{W_{13} + W_{12}}{A_{21} + W_{21}} \quad , \text{ or}$$

$$\frac{N_2 - N_1}{N_0} \simeq \frac{W_{13} - A_{21}}{W_{13} + A_{21} + 2W_{12}} \quad .$$

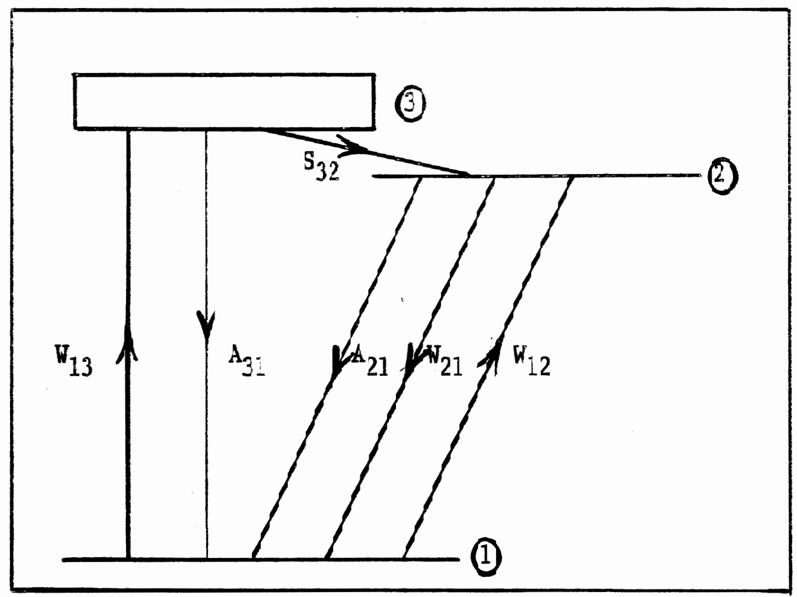

Figure 14. Typical transition diagram for energy levels of a three-stage laser.

and the minimum condition for a population inversion of N_2 over N_1 appears as $W_{13} > A_{21}$ (account for physical losses must also be made). Maiman et al. (1961) have determined the minimum condition for a typical ruby crystal to correspond to an energy of 555 watts/cm^2.