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RADIATION EFFECTS IN GLASS LASERS

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Experiments are in progress to study the effects of gamma ray irradiation on the stimulated emission properties of laser glasses.

The effects of high energy radiation upon glass are complex. The two principal processes are ionization (leading to color or defect center formation) and displacement of atoms from their position in the lattice. Of particular interest are induced optical absorption effects in commercially available laser glasses of different composition and dopant ions, starting with neodymium doped silicate glass (Eastman Kodak ND 11). Such changes as light absorption in glass produced by ionizing radiation may improve the optical efficiency for a laser by increased effective coupling between an optical pump source and the active medium. Improved laser efficiency was reported for irradiated crystal systems. However, a degradation of laser performance may also occur depending on the glass network and modifier composition, especially when metallic ion impurities (lead, iron, zinc) are present.

The effect of dose rate, total dose, selective bleaching and temperature will be related to optical absorption and emission measurements, laser tests, optical quality and magnetic resonance studies. Comparison against both a non-radiated and an undoped laser glass sample will be made.

INTRODUCTION

One of the important objectives in laser research is to imthis paper is directed towards increasing the optical pumping efficiency of neodymium doped laser glasses, since these are some of the most useful and practical laser media as yet realised.

The efficiency, i.e. laser radiation out-to-electrical energy supplied, of glass lasers has so far not exceeded a few percent. One of the reasons for this low efficiency has been the poor spectral match between the broad band laser pump lamps and the relatively few absorption bands found in glasses doped with trivalent rare earth ions. One way of increasing efficiency is to add another ionic species to the crystal which has broad absorption bands and which will transfer the energy they absorb to the rare earth ions (i.e. a sensitization or activation process). For example, the transfer of excitation radiation from Cr^{3+} to Nd^{3+} ions in various host media (including glass) has been observed (1). However, the inclusion of additional dopants is not always successful as the material quality and therefore, the optical homogeneity may deteriorate. Our approach for increasing the optical pump efficiency of Nd doped glass is to modify the intrinsic absorption characteristics and induce new absorption bands in the laser material by the formation of color centers under the influence of gamma rays and X-rays (2,3). Therefore, the objective of the work performed has been to examine whether or not this method is a possible mechanism for increasing the overall glass laser efficiency by transfering directly or indirectly some of the energy absorbed by optical excitation of the color centers to the Nd^{3+} system. In this manner a kind of percharged laser system is obtained which can be utilized upon optical pumping. This technique then represent a marriage of two established technologies: lasers, on the one hand, and photon irradiation of glasses, on the other. The usefulness of this approach has been explored to some extent with ruby (4,5,6). So far no definite conclusions have been obtained because the mechanisms for gamma ray excitation of fluorescence in solids is not well understood. In addition to improved optical pumpability, irradiation of a solid state laser system may have certain other practical merits in terms of:

1) direct pumping of a laser material by X-ray or gamma ray excitation

2) generation of giant laser pulses by saturable absorption of induced short lived color centers (7).

EXPERIMENTAL APPROACH

Our experimental program is directed towards studying to what extent the stimulated emission and spectroscopic characteristics of commercially available laser glasses are affected by energetic photon irradiation. As this program is not completed, only preliminary results are reported here so far.

The most convenient experimental approach for irradiating different configurations (i.e. rods and slabs) of laser glass has been to use a 4 kilocurie Co^{60} radioactive isotope source. This source emits gamma rays at a rate of 3.9×10^5 rads per hour with a photon energy of 1.1 - 1.3 MeV. Two glass types have been available to us so far:

1) Eastman Kodak ND 11 laser glass in slab form -- (a sodium silicate glass containing lanthanum)

2) Schott LG 55 silicate glass in rod form.

The former was exposed to different amounts of radiation varying from 10^5 up to 10^8 rads at room temperature before strong coloration effects appeared. The Schott glass required only 5×10^5 rads to produce considerable coloration. The irradiated glass was then examined under various experimental conditions.

Laser Tests

The most direct test to study the interaction of gamma rays and laser glasses is to compare the threshold energy of laser oscillations, near field and far field patterns, spiking behavior, energy and power output of an irradiated with a normal laser glass rod.

An experimental, medium energy pulsed laser facility with an external laser cavity arrangement and linear pump lamp configuration is now under construction, and will allow us to observe in detail any changes in the stimulated emission properties of photon irradiated glass systems. In the meantime, an assembly with a helical flash lamp was used to examine a Schott LG 55 laser glass rod. The threshold for the rod was found to be 110 joules. Irradiation with a dose of 3.9×10^5 rads increased the threshold beyond the maximum rating of the laser device (i.e. 175 joules) but heating normalised the threshold again. For example, the rod lased with an input energy of 155 joules after heating it in ½ hour intervals up to 250°C and reached its initial values of 110 joules again around 400°C. This is depicted in Fig. 1. On the same figure the laser output energy is displayed for an input energy of 175 joules. As one might expect from the threshold data, the energy

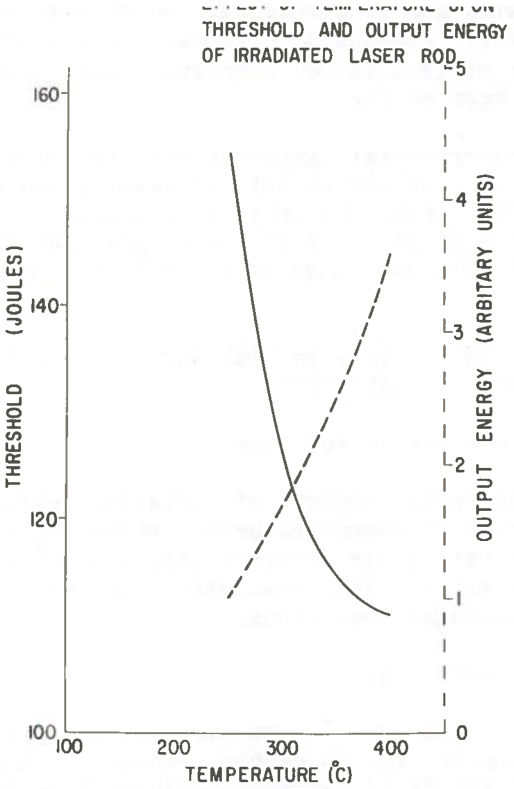


Fig. 1. Effect of temperature upon threshold and output energy of irradiated laser glass rod.

decreases with increasing exposure dose and increased again with bleaching temperature contrary to our desired objective of improved laser efficiency.

Another important design consideration will be the stability of the irradiated laser system, especially when exposed to high optical pumping powers which will tend to bleach induced color centers. However, for a single or even several laser pulses under favorable operating conditions (e.g. filtering) the intrinsic problem of color center fading may be minimised.

Optical Studies

Optical measurements are particularly useful in evaluating locally prepared laser glass samples in slab form, supplementing the previously described laser tests. Two effects have been studied:

Optical Absorption. A Beckman recording spectrophotometer was used to monitor transmittance and particularly spectral absorbance of the two available glass types (i.e. EK and Schott) in the wavelength range from 3500 and 9000 A^{o} where most of the intrinsic absorption occurs for rare earth doped glasses.

The measured absorption data can be related to laser performance criteria from which threshold energy and other data may be predicted on the basis of a simple theoretical model in the absence of saturation effects.

$$P_{output}\ (4\ level) = \Delta N\ \frac{h\gamma}{\tau_{spont}}$$

where the number of excited atoms generated by optical pumping is given by

$$\Delta N = \frac{kV\ \tau_{obs}}{\gamma_p}\ \ _{\Delta\lambda_p}\ \ P(\lambda,t)\ \alpha(\lambda)\ \eta(\lambda)\ \ \Delta\lambda_p\ \ e^{-\alpha(\lambda)z}$$

and where

V	is the active volume
$\eta(\lambda)$	the quantum efficiency for the pump light ($\sim$ 0.65)
k	is the coupling efficiency of the light source to the medium
γ and γ_p	the emitting laser and pump frequency respectively.
h	is Planck's constant
τ_{obs}	$= \tau_{spont}\ \phi$ = radiative lifetime of metastable atoms
ϕ	quantum efficiency
$\Delta\lambda_p$	absorption bandwidth
$P(\lambda,t)$	normally incident pump light available per band from high pump power Xenon pump source, which can be characterized in terms of a black body radiator at (say) 10,000^{o}K
$\alpha(\lambda)$	absorption coefficient, cm^{-1}
z	thickness of absorbing medium

A set of the absorption curves of ND 11 glass with the main absorption peak centered at 5800^{o}A is shown in Fig. 2. The effect

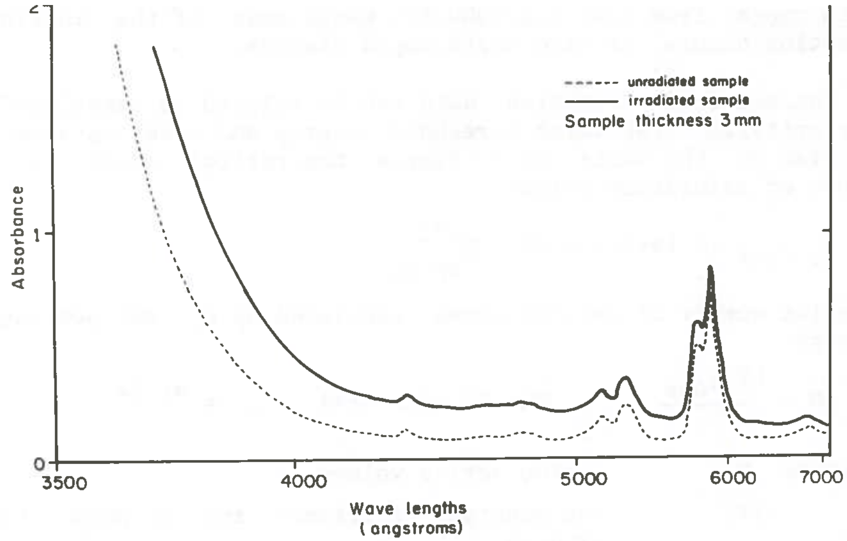


Fig. 2. Visible absorption spectra of neodymium laser glass (EK - ND 11)

of irradiation upon the absorption characteristics is apparent as Figure 3 shows the absorbance for both the irradiated and unirradiated samples. (Undoped ND 11 base glass irradiated under the same conditions will be investigated later). As one may anticipate the effective absorption crossection is increased. The net change in optical density may be obtained from these curves and plotted as function of photon energy or wavelength for low and high levels of irradiation.

Not only the laser threshold values can be predicted from the absorption data but also the number of color centers giving rise to induced absorption can be obtained using a well developed procedure due to Smakula (8).

If the centers which give rise to the induced absorption are sufficiently dispersed that they do not interact with one another, the area under the absorption curve is directly proportional to the concentration of the absorbing centers. Applying classical dispersion theory, one can write

$$Nf = \frac{9\ m\ c}{2\ e^2} \cdot \frac{n}{(n^2+2)^2} \quad \alpha_{max}\ \Delta\gamma$$

$$= 0.87 \times 10^{17} \cdot \frac{n}{(n^2+2)^2} \alpha_{max} \Delta\gamma \text{ (Gaussian)}$$

where N is the number of color centers per cm^3.

n the refractive index of glass at the wavelength of the absorption band m,e,c, have the usual meanings, the mass and charge of electron, and velocity of light respectively.

f is the oscillator strength of the optical transition, producing absorption ($\Sigma f = 1$)

α_{max} the absorption coefficient in cm^{-1} at the peak of the absorption and $\Delta\gamma$ the full width of the absorption band in eV measured at the half maximum points.

The validity of this approach could be checked using electron paramagnetic resonance techniques (9).

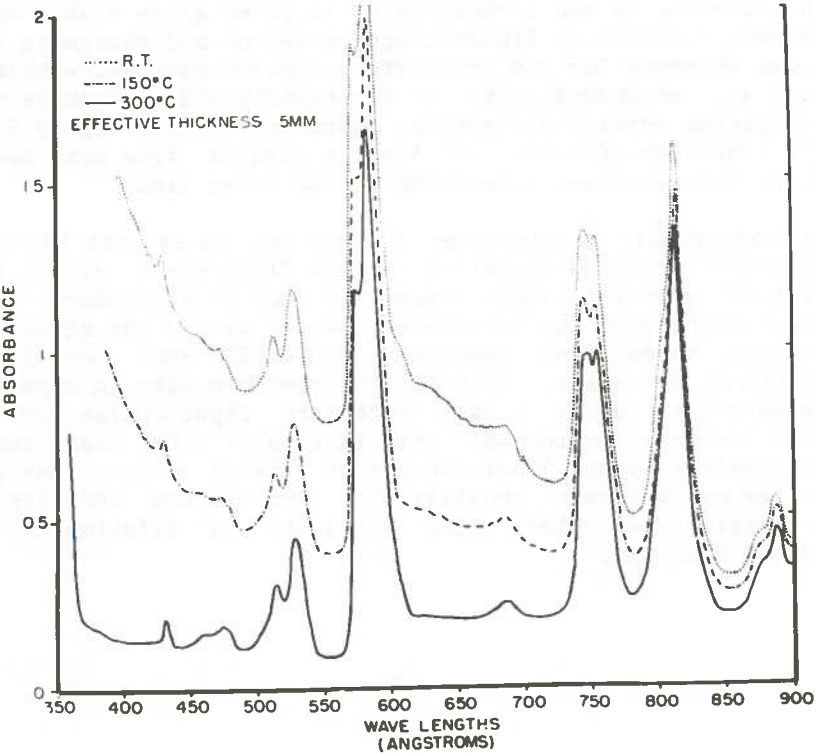


Fig. 3. Effect of bleaching temperature on spectral absorbance of irradiated laser glass rod.

Irradiated glass can be bleached and returned to its normal state either by heating or by light excitation. Fig. 3 shows the thermal bleaching effect upon the absorption characteristics of ND 11 laser glass.

Fluorescence Measurements. Two alternative methods for measuring relative fluorescence intensity levels and lifetime are used. One method is a spectroscopic method analysing the spontaneous emission of an optically excited sample. Figure 4 shows the fluorescent spectrum obtained with an optimized Leitz prism monochromator system employing a PbS detector, lock-in amplifier and pen recorder display arrangement. The glass sample is optically excited with a 650 watt quartz tungsten iodine lamp. For laser action the main fluorescent line located at 1.06 microns is utilized. This line is traced out in detail as shown in Fig. 5, both for an unirradiated and irradiated ND 11 laser glass slab. So far no apparent increase in fluorescence emission and change in linewidth was observed for the irradiated specimen examined within the accuracy and reproducibility of the experimental parameters despite improved initial absorption characteristics. Figure 5 also shows a linewidth of about 350 A which results from both homogeneous and inhomogeneous broadening of the laser line.

A considerable disadvantage of this method is that the excitation level required to obtain a good fluorescent signal (good in terms of signal-to-noise ratio) is such as to produce considerable bleaching of the irradiated sample within the actual time of sweeping through the spectrum, especially when scanning the wavelength at low speed. This will be overcome with an experimental arrangement using a high intensity light pulse of short duration from say a strobotac unit instead of a CW light source. The fluorescent decay signal can now be picked up by a fast photo multiplier or phototube sensitive at 1.06 microns and displayed on an oscilloscope. Fluorescent intensity and lifetime may then be deduced directly.

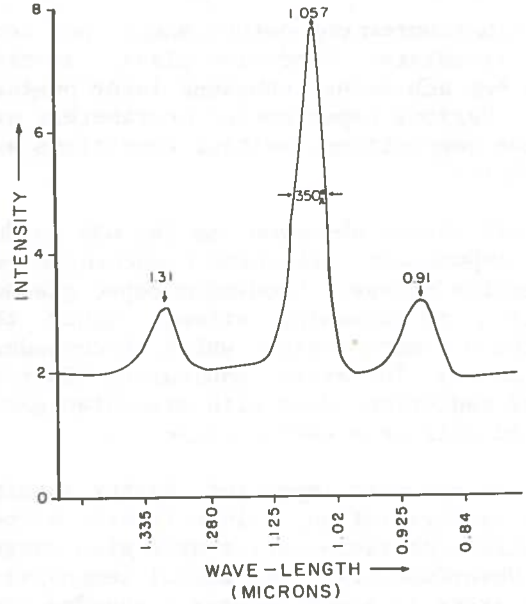
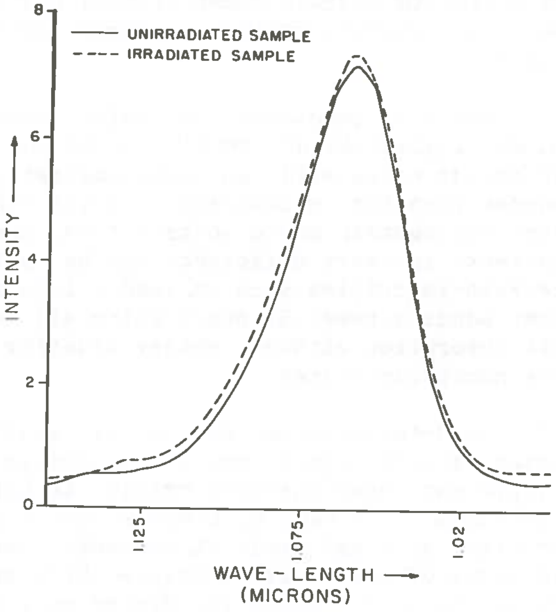


Fig. 4. Fluorescence spectrum of neodymium laser glass (fast scan).

Fig. 5. Main fluorescence line of neodymium laser glass (slow scan).



observed so far with gamma irradiated neodymium glass, several possibilities and approaches for achieving enhanced laser performance remain to be explored. Various experimental parameters are at our disposal: these include composition, melting conditions and controlled presence of impurities.

The decrease in laser efficiency observed so far may be due to unfavorable composition, impurities, lifetime quenching by in duced color centers and radiation damage. Neodymium doped glasses may be particularly sensitive to quenching effects since the spontaneous lifetime is in the 0.5 msec region which is considerably shorter than for ruby (3msec). Therefore, monitoring fluorescence lifetime as function of radiation dose with strobotac excitation as previously suggested will be a useful guide.

Favorable composition is the most important factor leading to enhanced laser operation by irradiation, since it will affect both fluorescence and absorption characteristics over wide ranges from one glass to another. Unfortunately, the actual composition of commercially available glasses is almost always a guarded company secret, so that a certain unknown factor will always exist. We plan to test numerous glass forming systems from which we hope to correlate certain compositional changes in glass and the effect of these changes on the spectral properties of neodymium laser glass.

The high threshold for color center formation observed for EK ND 11 glass is apparently due to the presence of a small amount of cerium which will suppress coloration and most likely inhibit energy transfer mechanisms. On the other hand the Schott glass does not contain any desolarization additives. In this case; the decrease in laser efficiency may be attributed to the presence of certain impurities such as lead, iron and zinc or ruptured covalent bonds between Si and O which all would tend to produce overall absorption without energy transfer from the color centers to the neodymium system.

No deterioration of optical quality of the two available laser glasses due to radiation damage has been observed using a simple gas laser interferometric technique (10). A visible He-Ne laser beam was used to compare quantitatively the optical homogeneity and local refractive index changes of irradiated glasses in terms of interference fringe shift and distortion. Furthermore, researchers (5) using irradiated ruby reported large increases in efficiency only when operating at very large optical pump energies, and this possibility will be checked for in our glass media as soon as the new laser facility now under consideration and capable of 2000 joules input is completed.

In brief, the possibility of improved laser performance induced by gamma irradiation may well be feasible despite our initial results for only two specific glass systems.

ACKNOWLEDGMENTS

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DISCUSSIONS

E. DEEG (American University in Cairo, U.A.R.): *What kind of induced color centers would you want to have in order to allow you to decrease the threshold of a* Nd^{3+} *doped laser glass?*

F. TITTEL: *Radiation color centers giving rise to increased and induced absorption within the visible wavelength range are desirable in order to achieve maximum coupling of available pump light to the laser glass.*

F. TITTEL: *The mechanism leading to energy transfer between induced color centers and the Nd^{3+} ions is probably quite complicated and so far not well understood. Some possible mechanisms are the following:*

a) Gamma rays create radiation centers and defects in the glass. Some of these centers emit light over a wide wavelength range. This emitted light may then be reabsorbed by the rare earth ions.

b) Photo-electrons, released by photon irradiation can excite all the electronic levels by inelastic scattering processes.

c) Electronic recombination processes of trapped electrons and positive holes in the neighborhood of the Nd ions may also assist population inversion of the Nd^{3+} laser system.

R. WOODCOCK (American Optical Co., Research Division, Southbridge Mass., U.S.A.): *Was any direct or extrapolated measurement of the transmission at 1.06μ made before or after irradiation?*

F. TITTEL: *So far no absorption measurements at 1.06 microns have been carried out. Certainly, this would be an additional method to determine whether photon irradiation will enhance or deteriorate the intrinsic gain coefficient of Nd laser glass.*