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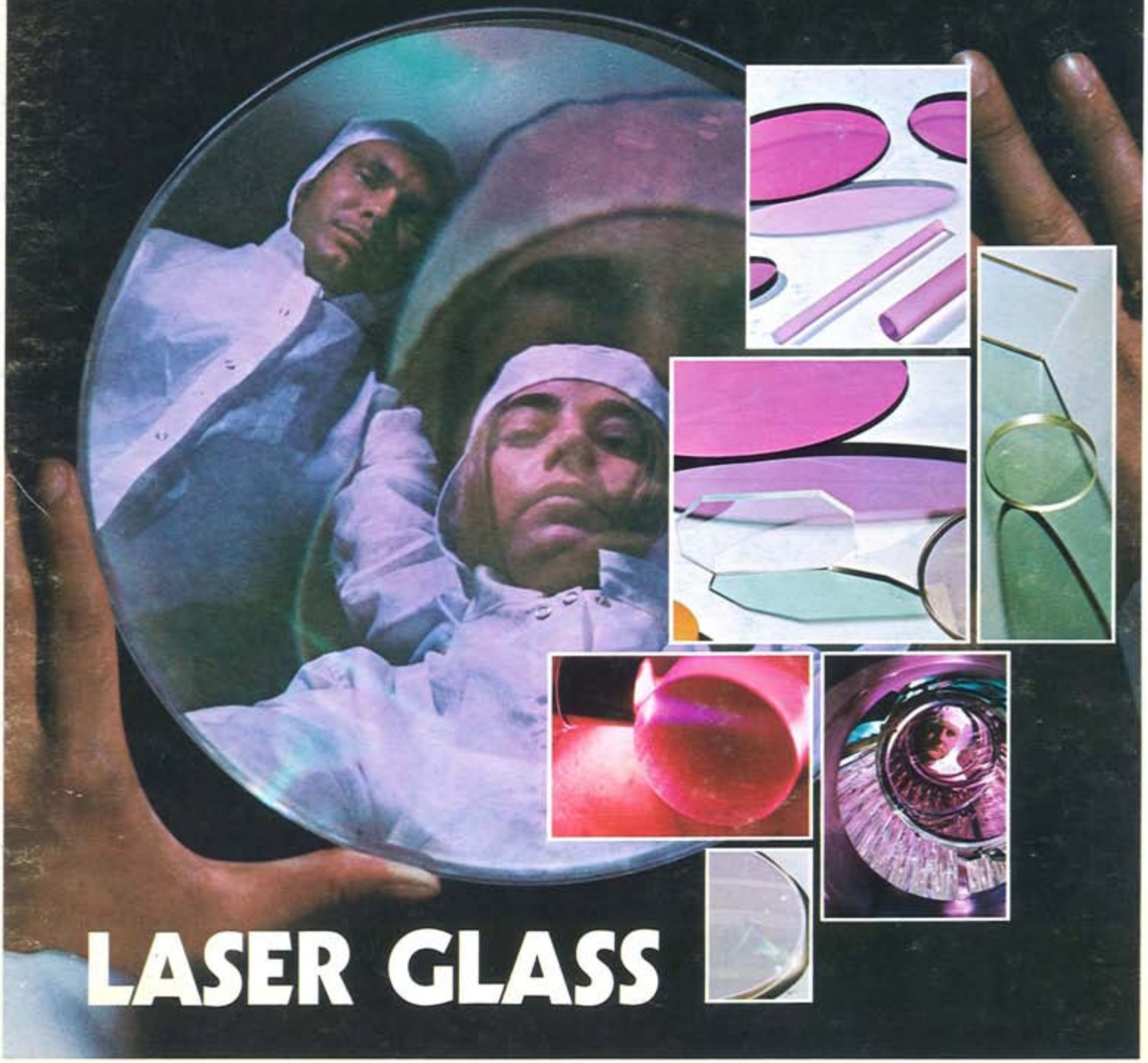

LASER GLASS

NATIONAL SECURITY

Glasses for High-Power Fusion Lasers

Neodymium-doped glass lasers, such as those in LLL's Shiva laser system, offer the best chance for early demonstration of the scientific feasibility of inertial confinement fusion. The output of these high-power lasers is determined by the media used for their amplifying and transmitting components. Our research has shown that significant increases in performance are possible with new fluoride-base glasses that have low refractive indices. These new glasses form the basis for more powerful experimental solid-state lasers and are potentially useful for advanced fusion laser systems.

A means for the practical conversion of nuclear power to electrical power may be by inertial confinement fusion. We anticipate that the earliest demonstration of the scientific feasibility of this concept will be by means of laser-induced implosion of D–T pellets. In the past few years, considerable progress has been made in the development of powerful laser systems capable of igniting ever-increasing quantities of inertially confined nuclear fuel. [1] The most advanced lasers for this purpose use

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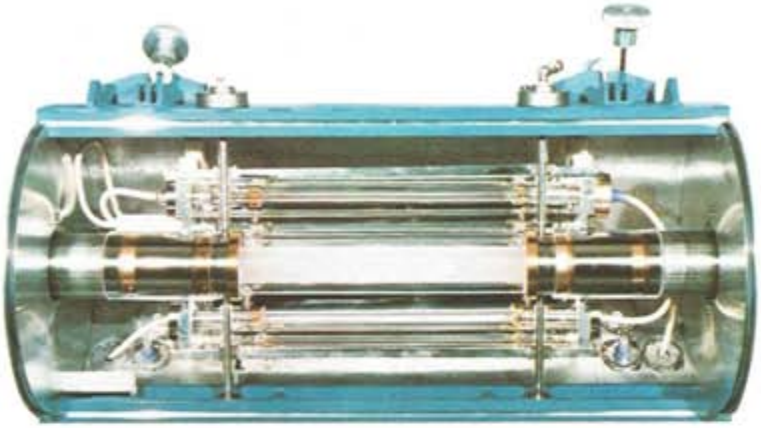
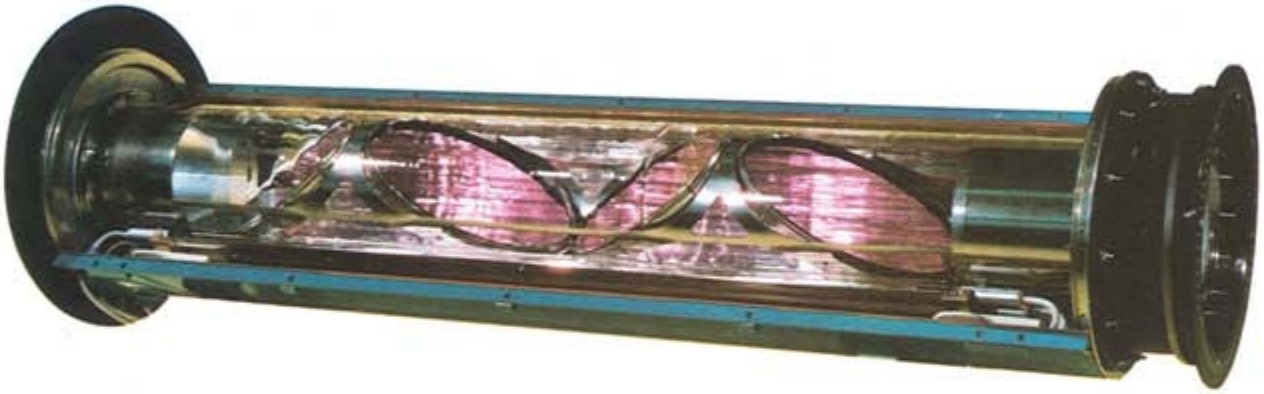


Fig. 1. **Fusion laser amplifiers. The amplifying medium is neodymium-doped silicate glass. (*Top*) Rod amplifier. The laser rod is 5 cm in diameter and is surrounded by an array of xenon flashlamps that serve as the optical pump. (*Bottom*) Disk amplifier. The elliptical laser disks are 15 × 30 × 3 cm and are oriented at Brewster's angle to minimize reflections.**



neodymium–doped glass as the amplifying medium. Typical rod and disk amplifiers are shown in Fig. 1. These components are arranged in chains of increasing apertures as shown in Fig. 2. The resulting systems are able to concentrate many terawatts of optical power onto targets of less than a millimetre in times of less than a nanosecond.

It became apparent at an early stage in the glass laser development program at LLL that future fusion laser development would be limited by the properties of the laser optics. Lasers based on both Nd–glass amplifier media, as well as gas amplifier media, use glass and crystalline optical materials extensively. The nonlinear propagation properties, the damage levels, and the ease of manufacture of large-aperture optics define the performance of all of these systems. In the case of Nd–glass laser systems, additional attention must be paid to the lasing properties of the glass amplifier media. When we began to study the effect that glass lasing properties and nonlinear propagation properties had on system performance, we found that remarkable increases in laser system performance were available if materials with high gain and low nonlinear refraction index could be found. These properties have now been realized with the fabrication of large pieces of fluoride-base laser glass. These materials have permitted us to extend our solid-state laser designs from the 30-TW Shiva laser to the 200- to 300-TW Nova laser and to conceive of a 1000-TW solid-state reactor-test laser.

Glass is used in several laser components.[2] The primary use is for the amplifying components—presently silicate glass with neodymium dopant. Glass is also used for light-transmitting components (such as lenses, windows, and substrates for thin-film polarizers and beam splitters) and for Faraday rotation in optical isolators. For laser beam diameters up to 50 cm, glass is an attractive medium since it can be made in large sizes with the requisite high optical quality; in fact, a limiting size for glass components has not been reached.

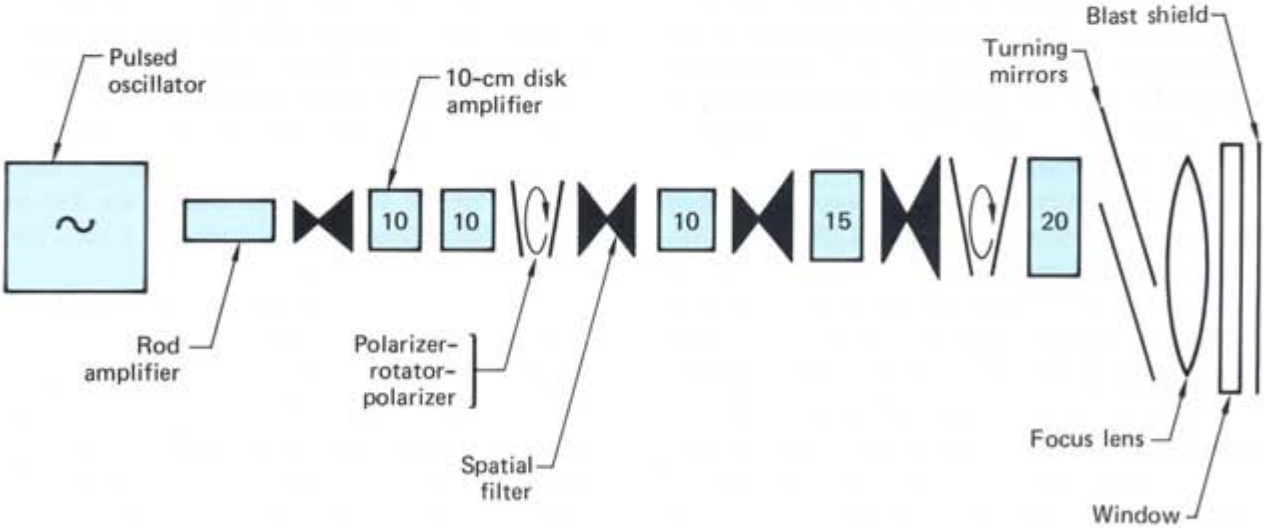


Fig. 2. Typical staging sequence of fusion laser components. The actual installation of components in the Shiva laser system is shown in the photograph above. A schematic representation of a single chain is shown in the line drawing below. Each component in the chain must be optimized for the increasing beam-power levels (left to right). Numbers represent the minor axis diameter of the elliptical glass disks in centimetres. The total length of the chain is about 50 m.

The ability of the lasing ions to absorb, store, and release energy depends on the properties of the glass in which they are suspended. A useful laser glass must have all the properties of high-quality optical glass: high transmission and index homogeneity, low thermally induced distortions (such as birefringence), high mechanical strength, and easy workability to stringent standards of surface flatness. It must also be easy to coat with an index-matched, absorbing, parasitic-suppressing edge coating. The capacity of a glass to sustain very high power beams is dependent on its nonlinear optical response, or nonlinear index of refraction. Thus, glasses that have low nonlinear response can sustain very high power densities throughout. Finally, the glass must withstand the intense ultraviolet flux from the flashlamp without darkening or incurring physical damage.

Because the physical properties of glass are affected in varying degrees by composition changes, it is possible to achieve wide variations in the characteristics of glass. The choice of characteristics, in turn, makes it possible to tailor glass performance to a specific component function and, thereby, to optimize laser design.

LASER-GLASS RESEARCH PROGRAM

Glass is required for several components in a laser amplifier chain, such as that shown in Fig. 2. Current glass laser technology uses silicate glasses. For each different component function and for increasing power levels, the required glass properties change. These variable properties as well as cost factors demand that we do optimization studies for both the glass elements and the component staging.

In the solid-state physics effort at LLL, we are methodically exploring an immense parameter field to achieve optimum lasing and optical performance from the glass. Commercial glass manufacturers are supplying glass samples formulated, melted, and configured to our specifications. We measure the physical and optical properties of these samples and then use this empirical information in a set of computer codes to model the glass performance under laser operating conditions.

The resulting laser-glass research program has shown that, within limits, we can tailor and optimize the properties of glass for fusion lasers. By changing the glass network-former (silicate or borate) and network-modifier (alkali or alkaline-earth oxide) ions, we can achieve large variations in optical properties (such as the linear and nonlinear refractive indices) and in lasing properties (such as energy levels, transition probabilities, and radiative and nonradiative processes).

REFRACTIVE INDEX

The nonlinear refractive index is the most important of the many optical properties affecting the performance of short-pulse, high-power fusion lasers. The total refractive index is composed of linear (normal) and nonlinear components; the nonlinear refractive index is dependent on the material's response to intense light. Although the intensity-dependent change in the index is small (on the order of parts per million at intensities of 10 GW/cm^2), the optical path length through a series of amplifiers in a fusion laser may be long enough (about a metre) to cause phase shifts of several wavelengths in the optical wavefront. These phase shifts can destroy the uniformity of the beam intensity, increase the beam divergence, and even cause destructive self-focusing. The existence of large, positive nonlinear refractive indices is, therefore, a major problem to be solved for the improvement of laser glass.

Our early experiments revealed that material with a small linear refractive index, n, and a small dispersion (change in n with change in wavelength) also had a small nonlinear index coefficient, n_2. We have obtained further data on the correlation between linear and nonlinear refractive indices by using time-resolved interferometry on a wide variety of glasses.[3] With these empirical results, we can estimate the nonlinear index from measurements of the linear index and the dispersion.

Optical glasses are conveniently grouped using as coordinates the refractive index, n, and the Abbe number, ν, where ν is the reciprocal of dispersion. Regions of n_d and ν_d of known optical glasses are shown in Fig. 3. (The subscript d signifies that the properties were measured at the wavelength of the

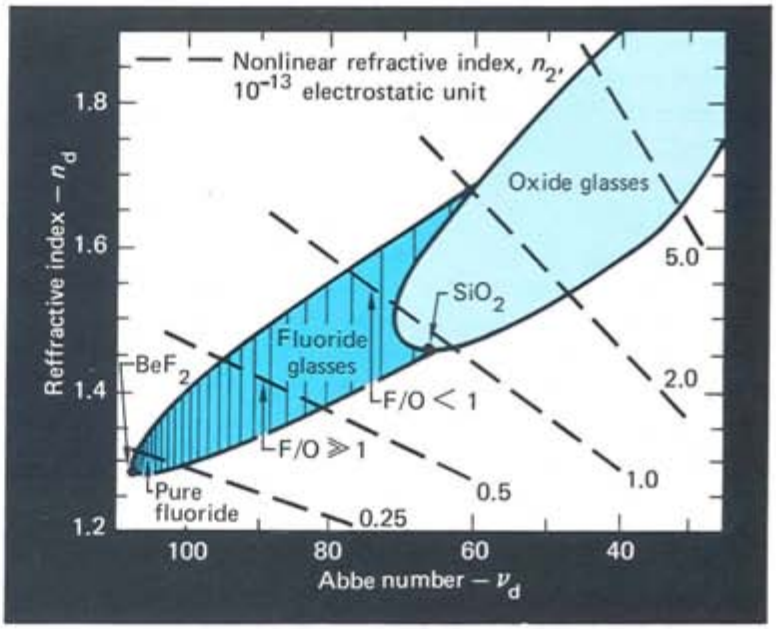


Fig. 3. Optical glasses of varying composition are mapped by their refractive index and Abbe number (reciprocal of dispersion). Superimposed are dashed lines of constant nonlinear index, n_2, based on our measurements. The nonlinear index, n_2, is reduced as the fluorine-to-oxygen ratio (F/O) is increased. The most promising glasses for fusion lasers are those toward the lower left-hand corner.

helium d line—587.6 nm.) Superimposed in Fig. 3 are dashed lines of constant n_2 predicted from the n_d and ν_d values. To propagate laser beams with small distortions, we need low-n_2 glasses; these are the fluoride-containing glasses in the lower left-hand corner of the figure. Various types of fluoride glasses are listed in Table 1.

Table 1. Various types of fluoride-containing glasses. We have investigated those marked with an asterisk for possible fusion laser applications.

Former	Modifier ions
Pure Fluoride Glasses*	
BeF_2	MF (M^+ = Li,Na,K,Rb,Cs)
AlF_3	MF_2 (M^{2+} = Mg,Ca,Sr,Ba,Zn,Cd,Pb)
ZrF_4	MF_3 (M^{3+} = Al, Y, La, Bi)
	MF_4 (M^{4+} = Zr, Th)
Fluoride Glasses Containing Oxygen ($F/O^a > 1$)*	
Fluoride glasses containing small mole % phosphate	
Oxide Glasses Containing Fluorine ($F/O \lesssim 1$)	
Fluoroborate	
Fluorosilicate*	
Fluorophosphate*	
Fluorogermanate	

[a]F/O represents the ratio of fluorine to oxygen ions.

The most common commercial optical glasses are oxide glasses, principally silicates and phosphates (i.e., SiO_2- and P_2O_5-base). Current optical glasses with the lowest n_2 values are fluorine-containing oxide glasses, namely, fluorosilicates and fluorophosphates. The glass with the lowest index reported is beryllium fluoride (BeF_2) without any modifiers. BeF_2 forms a glass analogous to SiO_2, but it is more ionic and the BeF_2 bonds are weaker (therefore, it has a lower melting point). Pure BeF_2 and SiO_2 glasses are plotted in Fig. 3 and represent the lowest n_2 values of the fluoride and silicate glass types, respectively. The addition of network modifiers, such as alkali, alkaline earth, aluminum, and other cations, changes the optical properties and yields the n_d–ν_d regions shown.

Because of their small predicted n_2 values, fluoride glasses are the most promising candidates for both passive components (such as lenses, windows, and substrates) and active components (such as the laser-amplifier and Faraday-rotation media). Optical figures of merit two to three times that of present oxide laser glasses may be realized (see

Table 2. Figures of merit for the nonlinear refractive index of optical materials.[a]

Component	Figure-of-merit formula	Silicate glass (ED-2, LSG-91h)[b]	Phosphate glass (EV-2, LHG-7, Q-88)[b]	Fluoro-phosphate glass (LG-810, E-181, LHG-10)[b]	Fluoro-beryllate glass (B-101)[b]
Laser disk (parasitic-limited; at Brewster's angle)	$\frac{n^2}{n_2}$	1.0	1.4	2.3	3.1
Window	$\frac{n}{n_2}$	1.0	1.4	2.3	3.6
Lens	$\frac{n(n-1)}{n_2}$	1.0	1.3	2.0	2.2

[a]A smaller n_2 implies a larger figure of merit and, hence, a better glass. Silicate glasses are currently used in glass lasers at LLL.
[b]Manufacturer's designation.

Table 2). Since fluoride glasses generally have large band gaps, they transmit light over a large spectral range. Hence they are potentially useful as transmitting optical components for high-power lasers operating at various wavelengths with lasing media other than neodymium.

LASING PROPERTIES

In the amplifying element, the important factor is the influence of the host glass on the ability of the lasing ion to absorb light from the optical pumping source, to store this energy, and to release it to amplify the laser beam. Energy storage by the lasing ion is governed by its absorption properties, excited-state lifetimes, and quantum efficiency. The position of absorption bands of rare-earth ions (such as neodymium) in solids shows little change with host, but the relative strength of the bands does change. Thus, the absorption coefficient of the lasing ions is host-dependent.

Lasing ions excited to high-energy electronic states relax nonradiatively (heating the host) to the upper lasing energy level. The lifetime of this metastable state determines how fast the level must be pumped to achieve the excited lasing ion population necessary for lasing.

Also, for each absorbed pumping photon, one lasing photon should come out. The quantum efficiency can be degraded in some hosts if the ion-host interaction causes nonradiative losses. In addition, interactions between lasing ions can also cause self-quenching. This effect limits the useful dopant concentration in the glass; that is, the lasing ion concentration cannot be arbitrarily increased without causing self-defeating losses. Thus, the important parameters of net absorption, rate of excitation, and efficiency are all dependent on the host glass.

The rate of energy yield from a laser amplifier is governed by a product of the optical intensity of the extracting beam and the stimulated emission cross section of the lasing ions. As discussed earlier, the beam intensity is limited by the nonlinear refractive index of the host glass. The stimulated emission cross section is also host-dependent.

The emission cross section of a lasing ion determines the yield of the stored energy and, thus, the amount of its amplification. The best cross section to use depends on the intended operation of the laser. Short-pulse, unsaturated lasers need high-cross-section materials in order to get large gain coefficients with a minimum thickness of glass.

However, if the cross section is too high, parasitic losses (in large amplifiers) and saturation will set in and limit performance. On the other hand, long-pulse, saturated lasers require low cross sections so that large amounts of energy can be stored and extracted without parasitic losses. The cross section must not be too low because excessive flux, capable of causing nonlinear optical distortion, would be required to extract the energy. For any fixed pulse width, there is an optimum cross section. If the cross section is too low, gain is low and too much material (glass) is required in the beam path. If the cross section is too high, saturation and parasitic losses reduce the gain. The optimum cross section generally becomes smaller as the pulse width is increased.

The ideal glass is one for which the emission cross section can be varied by a variation in glass composition without degradation of other desirable properties. However, some interaction is unavoidable; for example, raising the cross section usually increases the fluorescent decay rate. Early studies have shown that in silicate glasses, variations by a factor of three in the cross section were possible by changing the network-modifier ions. Variations in cross section by a factor of five have been observed by varying glass network formers.[4]

An additional consideration is the terminal laser-level decay time. To extract all of the stored energy, the terminal laser state must rapidly relax to the ground state. If the stimulated emission rate is too fast, bottlenecking occurs at the terminal energy level, which reduces the useful energy yield. The relaxation rate of the terminal level is also governed by ion–host interactions and can be varied by selecting different glass compositions.

Different glasses may be optimum for different amplifier stages for reasons other than optimum gain. But the wavelength of peak gain varies with the glass used. In the case of phosphate and fluorophosphate glasses, the wavelength differences of the two glasses are small, and their use can be mixed without a large sacrifice in gain performance. This statement is not true, however, for the combined use of, say, silicate and phosphate glasses or oxide and fluoride glasses. Nor does it apply to passive components.

Table 3 compares the range of spectroscopic properties observed in different glass types of interest. A wide variation in most parameters has been obtained and thus provides the laser designer with a range of material parameters to select in optimizing a laser design.

LASER-GLASS OPTIMIZATION STUDIES

Since the advent of lasers, thousands of glasses have been formulated to investigate the effects of changes in glass network and network-modifier ions on the spectroscopic and lasing parameters of neodymium. This work to a large degree was empirical and was hampered by two difficulties: first, the spectroscopic properties and interactions of rare-earth ions were not completely understood even in a crystal environment; second, the specific uses and applications of lasers were less well defined and, therefore it was not always clear which properties should be optimized. Today, both of these problem areas are better understood, and we have

Table 3. Measured range of variations of neodymium laser glass properties.

Glass type	Nonlinear index, 10^{-13} esu	Peak wavelength, nm	Cross section, 10^{-20} cm^2	Linewidth (FWHM), nm	Fluorescence lifetime, μs
Silicate	$\gtrsim$1.0	1057 to 1062	1.0 to 3.1	28 to 35	300 to 1000
Phosphate	$\gtrsim$0.9	1053 to 1056	2.0 to 4.5	19 to 28	300 to 500
Fluorophosphate	$\gtrsim$0.5	1050 to 1055	2.0 to 4.5	22 to 29	300 to 600
Fluoroberyllate	$\gtrsim$0.3	1046 to 1050	1.6 to 4.0	15 to 24	500 to 1000

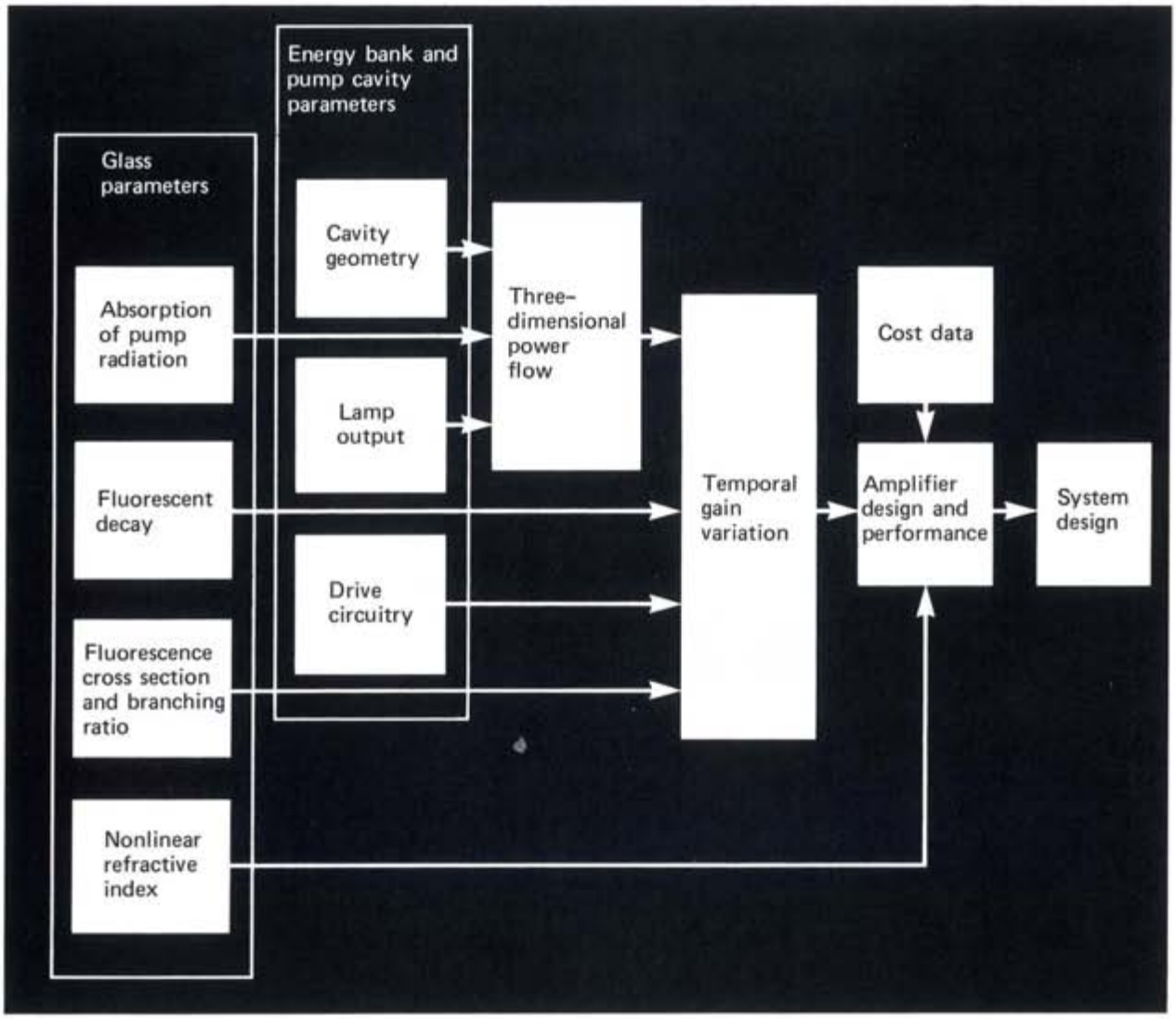


Fig. 4. Schematic flow chart showing laser-system modeling procedure. On the left, the parameters of a specific laser glass are entered into appropriate glass-modeling codes. The next block provides detailed information on the capacitor bank, laser heads, reflectors, flashlamps, and lamp circuitry, which input to the appropriate codes for modeling the power flow and gain variation. The final result of this modeling is the most cost-effective laser system design.

now evaluated over 200 glasses for fusion laser applications.

Predictions of the small-signal laser gain are based on measurements of absorption and fluorescence spectra and decay properties combined with calculations of the effects of flashlamp pumping. We have been successful in using data obtained from small glass melts to:

- Determine lasing parameters from spectroscopic properties.
- Estimate the nonlinear refractive index from linear refractive indices.
- Predict gain of amplifier stages and model the most cost-effective staging of an overall laser system.

From such work, the optimum glasses are selected for the design and staging of individual laser amplifiers in an overall fusion laser system. The lowest-cost system is then chosen for comparison or implementation.

A flow chart for modeling amplifier performance is shown in Fig. 4. The absorption spectrum of the candidate glass is recorded from the near-ultraviolet to the near-infrared. Using the measured line strengths, we derive a set of optical intensity parameters to calculate fluorescence line strengths and to determine the stimulated emission cross section. We also calculate the radiative lifetime and compare it with the observed fluorescence decay rate to determine the quantum efficiency. With data from the per-ion absorption spectrum, a computer program calculates the fractional absorption by the material of the xenon flashlamp radiation. The program contains a model of the flashlamp output spectrum that we modified by rerunning some of the light back through a layer of plasma to approximate the multiple passes that actually occur inside the laser pumping cavity. We then use the output curves of fractional absorption as a function of optical depth in conjunction with decay rate and cross section information to predict the temporal gain of different materials in disk laser amplifiers.[5]

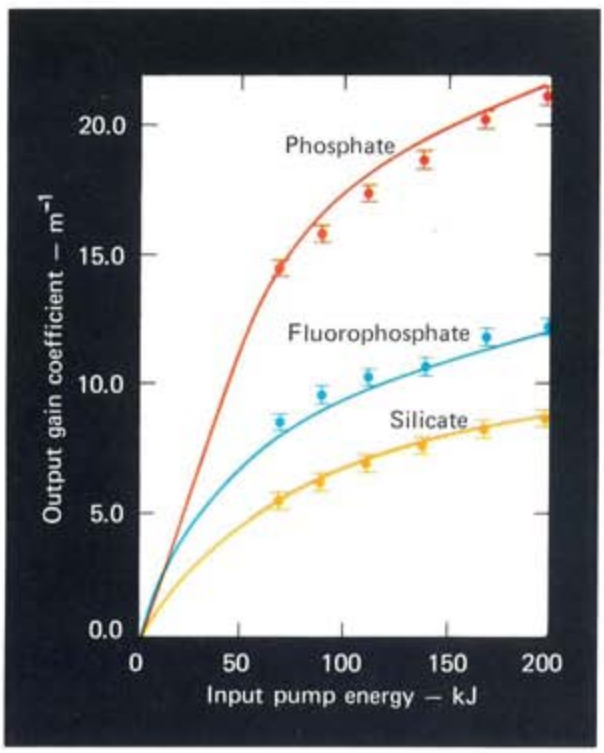


Fig. 5. Comparison of measured (points) and calculated (lines) gain coefficients for three neodymium glasses, using the laser-glass modeling procedure shown in Fig. 4. With all other conditions the same, the mere change to phosphate or fluorophosphate glasses will markedly increase the laser gain or provide the advantage of a much smaller nonlinear index of refraction.

How well does this work? Figure 5 shows the laser amplifier output versus pump energy input for three glasses: a high-gain phosphate, a fluorophosphate, and a low-gain silicate. Shown are a comparison of measured points and predicted gain curves. The overall agreement is within 10% and is typical of the results obtained in our study to date. Because of uncertainties in the accuracy of modeling the pump cavity configuration, data on a silicate glass are normalized to the predictions; however, once this normalization has taken place, there are no other adjustable parameters.

PROGRAM STATUS AND PLANS

Significantly improved neodymium–glass laser performance is now possible with newly developed glasses. We have found that for short-pulse, high-power lasers, the output is nearly inversely proportional to the nonlinear refractive index of the glass, n_2. Glasses with n_2 from one-half to one-third lower than presently used glasses have been identified. The relative figure of merit of these glasses for several laser elements is given in Table 2. In addition, by using phosphate rather than silicate glasses, we have achieved amplifier gain increases greater than 50%.

Commercially available phosphate glasses can have very high gain, but their use in large disk amplifiers is limited because high-gain-induced parasitic losses become dominant. For these large amplifiers, fluorophosphate glasses with medium-high gain and low n_2 are currently the best choice and are the most likely medium for the next larger Nd–glass laser system at LLL (see Fig. 5). The predicted performance of this laser, which we call Nova, is based on the physical parameters of the glasses. Undoped fluorophosphate glasses have been commercially available for some time and are now being formulated and optimized for use in fusion lasers. Beryllium fluoride glass, the ultimate high-power laser medium, is being formulated and tested in small quantities; test results have been very

encouraging, but great uncertainties remain in the production of large, high-optical-quality pieces and in the yields and costs associated with the precautions of handling beryllium.

To ensure that improved glasses will be available in time for Nova, ERDA is supporting developmental research with glass manufacturers in close cooperation with LLL. New glasses are being evaluated in our computer-equipped spectroscopy laboratory[6] where more than a dozen spectroscopic, optical, and physical properties are measured on samples supplied by manufacturers. To hasten development, we provide this information to the manufacturers in the form of data books—to date, in six volumes describing about 200 laser glasses. Large melts of fluorophosphate laser glasses are now being cast into test disks for Nova prototype components.

New ideas in laser design indicate that 10-MJ, 1000-TW solid-state laser systems may be possible. Recent studies also point toward high-average-power systems with efficiencies greater than 1%. These concepts are optimistic, but they may be more reasonable extensions of established glass laser technology than we presently imagine. The accomplishment of laser fusion objectives with advanced lasers based on gaseous laser media will also require imaginative use of optical materials. These lasers are limited by nonlinear, damage, and manufacturing constraints just as solid-state lasers have been. Materials similar to those described in this report will be the basis for the design of future high power lasers.

Key Words: glass; glass lasers; neodymium-glass; lasers—materials.

NOTES AND REFERENCES

1. The laser fusion program at LLL has been reviewed in two full issues of the *Energy and Technology Review*: February 1976 (UCRL-52000-76-2) and August 1977 (UCRL-52000-77-9). More detailed information on the program is presented in the *Laser Program Annual Report* for 1974, 1975, and 1976, Lawrence Livermore Laboratory, Repts. UCRL-50021-74, UCRL-50021-75, and UCRL-50021-76.
2. M. J. Weber, "Optical Materials for Neodymium Fusion Lasers," in *Critical Materials Problems in Energy Production*, C. Stein, Ed. (Academic Press, New York, NY, 1976), pp. 261–279.
3. E. S. Bliss, D. R. Speck, and W. W. Simmons, "Direct Interferometric Measurements of the Nonlinear Refractive Index Coefficient N_2 in Laser Materials," *Appl. Phys. Lett.* **25**, 728 (1974); D. Milam and M. J. Weber, "Measurement of Nonlinear Refractive-Index Coefficients Using Time-Resolved Interferometry: Application to Optical Materials for High-Power Neodymium Lasers," *J. Appl. Phys.* **47**, 2497 (1976); and "Nonlinear Refractive Index Coefficient for Nd Phosphate Laser Glasses," *IEEE J. Quant. Electron.* **QE-12**, 512 (1976).
4. R. R. Jacobs and M. J. Weber, "Dependence of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ Induced-Emission Cross Section for Nd^{3+} on Glass Composition," *IEEE J. Quant. Electron.* **QE-12**, 102 (1976).
5. For a description of the computer-equipped spectroscopy laboratory, see R. A. Saroyan and M. J. Weber, *Unconventional Spectroscopy*, proceedings of a technical symposium held in San Diego, Calif., August 24–25, 1976 (SPIE, Palos Verdes Estates, Calif., 1976), vol. 82, p. 165.
6. G. J. Linford, R. A. Saroyan, J. B. Trenholme, and M. J. Weber, "Measurements and Modeling of Gain Coefficients for Neodymium Glass Lasers," *Digest of Technical Papers*, 1977 IEEE/OSA Conference on Laser Engineering and Applications, Washington, D.C., June 1–3 (to be published).