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for Inertial Fusion: Present and Future

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**Insulator Materials in High Power Lasers for
Inertial Fusion: Present and Future***

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ABSTRACT

Dielectric insulator materials have played a critical role in the development of high peak power solid state lasers for use in inertial confinement fusion research: as laser gain media; as transparent substrates for lenses, mirrors, and polarizers; and as active optical materials in nonlinear harmonic generators and electro-optical devices. Materials have been developed which have exceptionally high resistance to damage in the presence of intense optical beams ($> \text{GW}/\text{cm}^2$) and which possess other properties which optimize their functions in the laser systems. Fusion lasers built to date have been designed for "single shot" operation, and the dielectric insulator materials developed for use in them have had to function under only extremely low average power loading. As we look to the future, fusion laser systems will be required to operate repetitively (few Hz) and deliver high average power output

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(> 10 Mw) at an efficiency greater than 10 percent. Insulator materials for use in these systems must be selected and developed on the basis of their combined mechanical, thermal, and optical properties. In this presentation, I will summarize the important characteristics of currently used insulator materials, identify figures of merit for materials needed in future systems, and outline a methodology for identifying and evaluating new materials meeting the stringent performance requirements of future fusion laser systems.

INTRODUCTION

The principal goals of the U. S. Inertial Confinement Fusion (ICF) Program are to produce in the laboratory energy-efficient, high-gain ($> 100X$) implosions of fusion pellets; to apply ICF techniques and facilities to nuclear weapons physics problems; and ultimately, to produce cost-effective, central-station electric power. The quintessential issue being addressed in the Program is this: can fusion pellets be designed which will produce useful thermonuclear yield (e.g. $> 100X$) while sufficiently minimizing the laser driver requirements (energy, power, wavelength, efficiency, cost, etc.) that practical applications can be realized. This question is being addressed with the aid of complex computer simulations of implosion processes and data obtained from laboratory implosion experiments. Using the data available presently, we project nominal performance parameters of a laser system suitable for central-station electric power production shown in Table I.

TABLE I
Nominal performance parameters of a laser system suitable for central-station electric power production

The short wavelength ($\lambda < 1000$ nm) implosion experiments conducted to date have been enabled as the result of the development of flexible, "single-shot," high-peak-power neodymium glass (Nd:glass) laser systems during the past decade. Figure 1 summarizes the performance (energy, peak-power, and pulsewidth) capabilities of a sequence of Nd:glass laser systems designed, constructed, and operated at the Lawrence Livermore National Laboratory (LLNL). Dielectric insulator materials are utilized for several critical functions in these laser systems [1]: as active laser gain media; as transparent substrates for lenses, mirrors, and polarizers; and as active optical materials in nonlinear harmonic generators and electro-optical devices. Materials have been developed which have exceptionally high resistance to damage due to the presence of intense optical beams ($> \text{GW}/\text{cm}^2$), while having properties which optimize their laser system functions. Note that the lasers built to date (Figure 1) have been designed for "single shot" operation (e.g. 1/hr) and the materials developed for use in them have had to function under only extremely low average power loading. From Table I, we see that future laser systems will be required to operate repetitively (at a few Hz rate) and to deliver high average power output ($> \text{MW}$) at an efficiency greater than 10 percent and at a relatively short wavelength (< 500 nm). Thus optical dielectric materials for use in these systems must be selected and developed on the basis of their combined mechanical, thermal, and optical properties. The types of materials presently being utilized and the performance levels achieved are first summarized below. Functional figures of merit for future materials are then described, particularly for solid state laser gain media which, perforce, must sustain a substantial thermal loading.

OPTICAL SUBSTRATES (MIRRORS, POLARIZERS, LENSES)

To implement the large lasers described above, transparent optical elements with aperture diameters in excess of one meter have been developed [1] using materials such as BK-7 glass, characterized by a high bulk damage fluence. These materials are used as substrates for high reflectivity mirrors and polarizers and as blanks for spatial filter and focusing lenses. The mirror and polarizer functions are achieved by depositing multiple thin film layers of dielectric materials on polished substrate surfaces; to greatly reduce the loss of beam energy due to Fresnel reflection from the surfaces of the spatial filter lenses, anti-reflection (AR) thin film coatings have been applied to these surfaces as well. Laser design analyses show that in the most cost-effective (J/\$) Nd:glass laser chain, the surfaces of the input lenses of the spatial filters are subjected to the greatest optical fluences and that these optical components generally limit the performance and cost effectiveness of modern Nd:glass lasers. In some of our experimental damage research studies [2], it was noted that the damage thresholds of AR surfaces of glasses prepared by gradient index techniques [3] were much higher than those of thin film AR surfaces. During the past few years, we have worked together with Corning, Hoya Corporation, Schott Optical Company, Owens-Illinois, and Westinghouse to evaluate and develop large-aperture AR surface optical elements with high damage thresholds. Various techniques for producing gradient index surfaces have been investigated, including chemical leaching [4], neutral solution [5], and sol-gel [6] processes. Figure 2 shows in perspective the roughly five-fold improvement in the damage threshold of critical AR surfaces

achieved with gradient index optical structures. The challenge in the future will be to extend these results to shorter wavelengths and to develop components that will have an extremely long shot life. It is believed that surface chemistry techniques have barely been tapped so far and that they will play an even larger role in the future development of such optical materials.

VISIBLE UV WAVELENGTH OPERATION

In inertial-confinement fusion research, the trend in recent years has been to shorter and shorter wavelengths. This shift is driven by the combination of improved absorption of the light in the plasma, a substantial reduction in the production of hot and superhot electrons, and an increase in the production efficiency of thermal x rays exhibited for irradiation wavelengths shorter than ~ 500 nm. Harmonic conversion of existing Nd:glass laser systems operating at 1060 nm has proven to be a low-cost, efficient way of providing the shorter-wavelength radiation. Extremely high harmonic-conversion efficiencies (50 to 90%) have been demonstrated for conversion of intense beams ($I \geq 1 \text{ GW/cm}^2$) to the second, third, and fourth harmonics [7-9] using KH_2PO_4 (KDP) and its crystal isomorphs. The Nova-system beam diameter is 74 cm and therefore required 74-cm-diameter harmonic-conversion crystal assemblies. Fortunately, KDP is relatively easily grown from water solutions, and the scale-up of crystal size by about an order of magnitude has proceeded well, as shown in Figure 3. From the larger of these crystals, we are able to fabricate 27-cm-square plates, shown in Figure 3. Since the growth of crystals large enough for 74-cm-diameter plates would require a

prohibitively long time, the decision was made to develop crystal-array technology. [10] The array concept was tested with nine 5-cm-square crystals, shown in Figure 4. The output 2ω beam is also shown in this figure, indicating uniform harmonic-conversion efficiency and little distortion of the input beam. The array technology is made possible by extremely precise diamond-point machining and precise measurement of the KDP crystals [11], so that the phase-matching direction and the surface normal are coincident to an accuracy of ± 30 μ rad. The diamond-point machining produces such a finely finished surface that no subsequent grinding or polishing is required. The crystal-array technology is now being implemented on Nova at the 74-cm diameter using the 27-cm-square crystal plates.

For the development of future harmonic-generation capabilities at very large apertures, there appear to be several interesting directions to pursue. First, if materials that exhibit 90° phase matching at a convenient temperature (such as deuterated cesium dihydrogen arsenate, CD*A) could be grown in large sizes, the greatly reduced angular sensitivity would result in greatly reduced fabrication and alignment-system costs. Further in the future, the extremely high nonlinear susceptibilities of many organic materials [12] encourage us to consider the growth of large oriented films on transparent substrates. Such oriented-growth techniques might also be applied profitably to more conventional materials. Yet another way to achieve nonlinear properties over large areas is the use of liquid crystals, which, with material development, may ultimately prove useful without conventional crystal growth.

HIGH AVERAGE POWER GAIN MEDIA

Solid state laser gain media, and in particular Nd:glass, are not usually thought of as media suitable for use in high-average-power laser systems. Indeed, in the form most usually considered--that of large-diameter rods--solid state media are certifiably not useful for high average power; the combination of low thermal conductivity with temperature gradients normal to the propagation of an optical beam render high average power unattainable. However, Figure 5 shows an amplifier geometry that is simultaneously capable of high average power per unit beam area, high peak power per unit beam area, and large beam diameters. In this case, a single disk of a conventional disk amplifier is divided into two or more parallel plates. A gas coolant, typically helium or nitrogen, is flowed through the channels made by the parallel plates so that the heat in the plates is removed by conduction through the large flat surfaces of the plates, producing a thermal gradient normal to the surface of the plate. If the plates are infinite in lateral extent, the thermal gradient produces no optical distortion, since all rays of the beam will sample equally the stress field of the plate. In practice, finite plates will show little distortion except within one or two plate-thicknesses of an edge.

To get a feel for the power handling capacity of solid state media configured in this manner, consider the classic case of Nd:glass pumped with a broadband xenon flashlamp. Roughly speaking, about half of the energy absorbed in a unit volume of glass is converted to heat and about half is retained in the upper laser level of the neodymium ion and is potentially available as 1060 nm laser light. To achieve a useful small

signal gain coefficient (e.g. a few percent per centimeter) roughly a J/cc of thermal energy is deposited in the glass. Since a solid possesses a rather high product of density and specific heat, deposition of a joule of thermal energy per cubic centimeter will raise the temperature by only a fraction of a degree. Thus, repetitively pulsed operation can be considered as a steady state condition once the operating temperature is reached.

If the two large, flat surfaces of the plate are held at constant temperature by the coolant flow, and if the energy-deposition profile through the thickness of the plate is uniform, then the temperature profile in the slab is just

$$T = T_0(1 - 4z^2/t^2), \quad (1)$$

where t is the thickness of the plate and z is distance measured normal to the plate with $z = 0$ at the mid-plane. T_0 is given by

$$T_0 = \frac{P_v t^2}{8 \kappa}, \quad (2)$$

where P_v is the thermal power deposited per unit volume and κ is the thermal conductivity. The temperature rise T_0 affects both the bulk of the glass and the Nd ion. The plate is compressively loaded at the center, while the surface of the plate is in tensile stress. Since glass is strong in compression but comparatively weak in tension, it is the tensile stress at the surface that concerns us. It is easy to show that the peak tensile stress occurs at the surface and is

$$\sigma_t = \frac{2\alpha T_0 E}{3(1 - \nu)}, \quad (3)$$

where α is the coefficient of thermal expansion, E is the modulus of elasticity, and ν is Poisson's ratio. Table II contains values for the relevant thermal and mechanical parameters needed to calculate average power capacity, for some typical amorphous and crystalline materials. Figure 6 plots lines of constant surface tensile stress (and surface mid-plane temperature difference, T_0) for various plate thicknesses and thermal deposition loads, for a glass and for crystalline MgF_2 . For a T_0 of $90^\circ C$, the tensile stress in glass is 8 kpsi ($\sim 6 \text{ kg/mm}^2$), or approximately 50% of the yield stress of unstrengthened glass. Many glass compositions can have surface compressive layers applied. For example [13], an ion-exchange process applied to ED-2 laser glass produces a surface compression of approximately 40 kpsi (30 kg/mm^2) with no loss of optical quality of the finished plate. Thus, operation with glass for values of T_0 in the range of 60 to $180^\circ C$ is possible. These T_0 values will allow for thermal loading of roughly 5 watts/cc for 1 cm thick plates, an operating point of utility for fusion-like lasers.

The heating of Nd:glass induces the thermal population into the terminal state of the 1060 nm laser transition at 2000 cm^{-1} . This both reduces the gain coefficient of the material and reduces the availability of the energy stored in the upper laser level. Simple calculations show that temperatures above ambient by more than $100^\circ C$ would be very deleterious (however, other laser ions may be more forgiving in this respect).

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A useful figure of merit for the thermal loading capacity of a solid is the "thermal shock" parameter R_T , defined by the ratio of strength to stress.

$$\frac{S_T}{\sigma_T} = \left\{ \frac{S_T \kappa (1-\nu)}{\alpha E} \right\} \left(\frac{12}{P_v t^2} \right) = R_T \left(\frac{12}{P_v t^2} \right) \quad (4)$$

For a specific uniform thermal loading, P_v , of a slab of thickness, t , a large value of R_T will lead to a more favorable S_T/σ_T ratio. Since the ratio S_T/E tends not to vary widely, there are two main strategies for achieving a large R_T and thus a high-average-power handling capability. First, materials could be chosen which have coefficients of thermal expansion a factor of 10-100 below conventional laser glasses. Second, crystalline materials can be chosen which exhibit thermal conductivities 10 to 100 times larger than conventional laser glasses. Table II summarizes the properties of both types of materials.

It should be noted here, however, that these two strategies are not equivalent. The low coefficient of thermal expansion lowers the surface tensile stress of the slab without lowering the centerline temperature. Thus, increasing the average power per unit volume increases the centerline temperature of the slab. In contrast, a high thermal conductivity lowers the tensile stress by lowering the temperature gradient and thus the centerline temperature. This is illustrated in Figure 6 by the constant surface stress plotted for crystalline MgF_2 .

Little effort to date has been expended on minimizing the coefficient of thermal expansion of laser glasses, yet there are well known glass compositions with low coefficients. Table II shows some of the extremely low coefficients of thermal expansion materials such as fused silica and

glass ceramics of which Zerodur is an example. If glass ceramics [14,15] are to be used as laser materials, significant compositional development would be required to control light scattering and to optimize the laser properties of the active ion. However, it is probable that unless the active ion can be induced to deposit preferentially in the crystalline phase, the choice of laser active ions would be limited to those that perform well in glassy or poorly ordered media--nameiy, neodymium and ytterbium.

Achieving high average power by virtue of a high thermal conductivity requires the choice of ordered or crystalline media. As shown in Table II, representative oxide and fluoride crystals exhibit thermal conductivities which are typically 10 to 100 times that found in typical laser glasses and thus have a potential for operating at a correspondingly higher average power. Also attractive is the fact that, in general, the thermal conductivity of crystalline materials increases as the operating temperature of the materials is lowered, which further enhances the average power handling capability. To construct highly energetic lasers using crystals, it will of course be necessary that large, doped, high-optical-quality crystals are growable with high yield. It seems to us that the prospect for successful crystal growth scaling would be greatest for crystals that melt congruently; and those with relatively low melting temperatures (e.g. $T_m \leq 1500^\circ\text{C}$). Such temperatures permit the use of a wide variety of crucible materials and growth methods. We are greatly encouraged by the progress made in recent years in the growth of high-quality, large diameter (> 10 cm) LiYF_4 crystals doped with trivalent rare earth ion activators. Effort is now also being directed toward the

growth of large aperture MgF_2 crystals doped with Co^{2+} ions. Growth of large-aperture oxide crystals has also been achieved in recent years (Nd:GGG up to 15 cm diameter and undoped Al_2O_3 up to 30 cm diameter).

While the prospect for large scale growth is positive, development of large-aperture doped crystals will require the investment of considerable resources and will take several years to accomplish. It is therefore extremely important that candidate systems be thoroughly evaluated on the basis of small-scale spectroscopic and laser experiments coupled with extensive modeling, before a specific crystal system is selected for scaled development. Recently, an assessment [16,17] has been made of optical pump sources, laser ions, and solid state host materials which, when combined, may form a fusion laser driver responsive to the performance requirements given in Table I. Identified pump source types with usefully high efficiency and spectral brightness include conventional xenon flashlamps, surface discharges, semiconductor diode arrays, and e-beam pumped semiconductor lasers and epitaxial phosphor layers. The electronic level structures and anticipated dynamical properties of rare earth and transition metal ions were reviewed with respect to their use as possible laser and/or sensitizer ions, and desirable microscopic properties of the host lattice were indicated. Consideration of laser amplifier geometries, laser kinetics and energy extraction physics, and laser beam quality issues led to the conclusion that cubic or uniaxial crystals with high-symmetry sites for the laser ions should have particularly favorable laser properties. Furthermore, these hosts must also exhibit a ratio of nonlinear to linear refractive indices (n_2/n) of $\approx 10^{-13}$ esu and have damage threshold fluences in excess of 50 J/cm^2 for pulses 10 nsec long. As a class, fluorine-based crystals

have been found to have attractively low nonlinear refractive indices [18] and high damage fluences [19]. While less robust thermally and mechanically than the refractory oxides (see Table II), selected fluoride crystals appear to have adequate thermal shock parameters (i.e., a few hundred W/m). Accordingly, a number of cubic and uniaxial fluoride crystal types, doped with various rare earth and transition metal ions, are now being examined for their laser properties. Specific crystal types include elpasolites, perovskites, and garnets. It is expected that this search and evaluation effort will be expanded to include additional fluorides as well as selected oxide crystals.

Table I
Fusion Laser Driver Requirements (1 GWe)

Laser Energy, E_L	1-3 MJ
Pulse Width*, t_p	~ 5 nsec
Wavelength, λ	≤ 500 nm
Efficiency,** η	$\geq 10\%$
Pulse Repetition Rate, R_p	~ 10 Hz
Intensity of Target, I_t	~ 400 TW/cm ²

* Highly shaped laser pulse is required for high yield; duration quoted is for delivery of the bulk of the energy.

** A theoretically perfect (Fermi-degenerate) pellet implosion could allow a driver efficiency as low as 1%; the 10% value given accounts for pellet performance degradation from the ideal.

FIG. 1. Energy, peak power, and pulse width capabilities of Nd:glass laser systems at LLNL. The Nova system is capable of operating at 1050, 525, and 350 nm; the others operate at wavelengths of 1050 nm only.

A = $\text{TiO}_2/\text{SiO}_2$ thin film coating — conventionally polished substrate

B = $\text{TiO}_2/\text{SiO}_2$ thin film coating — super polished substrate

C = Neutral solution gradient index surface, as prepared

D = Neutral solution gradient index surface, laser cleaned

FIG. 2. Improvements in damage threshold fluence of anti-reflection (AR) surfaces achieved during the past half-decade.

Solution Grown Boule

Finished Plate TYPE-II

FIG. 3. Large crystal of KDP grown by Cleveland Crystals, Inc., Cleveland, Ohio. Crystal measures 30 x 32 cm in cross section at the base and is 64 cm long. The 27 cm crystal plate on the right was fabricated using diamond point machining techniques.

FIG. 4. Experimental nine-crystal array of 5-cm-square KDP crystals. Representative second-harmonic output beam.

FIG. 5. Brewster-angle disk amplifier geometry suitable for attaining high-peak-power pulses and high-average-power output, simultaneously.

$$P_v = \frac{8\kappa T_o}{t^2}$$

$$\sigma = \frac{2\alpha T_o E}{3(1-\nu)}$$

FIG. 6. Thermal loading capacity of solids.

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