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Advanced Crystalline Laser Materials

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ADVANCED CRYSTALLINE LASER MATERIALS*

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Lawrence Livermore National Laboratory (LLNL) has recently undertaken an effort to develop new solid state lasers capable of operating at considerably higher performance levels (efficiency, average power, wavelength diversity, etc.) than attainable with current technologies. To guide this effort, we have performed analyses to establish the fundamental limits on the performance levels of solid state lasers in terms of known laser physics and design principles (gain, gain saturation, amplified spontaneous emission, parasitics, nonlinear self-focusing, etc.) and the physical properties of crystalline and amorphous materials.^{1,2} A technical summary of these results, and the salient conclusions drawn from them, was presented by John Emmett at this conference. To realize the substantially higher performance levels permitted within these fundamental limits, we conclude: 1) that thin-plate geometries (zig-zag³ or flow-cooled disk⁴) must be adopted and fully exploited through innovative engineering design solutions; 2) that efficiencies can be greatly improved by developing new types of efficient, high-irradiance pump sources and especially by instituting measures that rigorously maximize the utilization of the pump radiation generated by presently available efficient flashlamps; and 3) that new solid state laser gain materials must be developed which possess laser properties (stimulated emission cross-section, energy storage lifetime, saturation fluence, etc.) and bulk properties (mechanical, thermal, and optical) dictated by the applications of interest. To guide the laser materials research and development effort, we have analyzed the scaling laws for zig-zag and flow-cooled disk amplifiers, defined materials figures of merit, and

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developed laser performance applications maps.² Within this technical framework, we have embarked on an effort to synthesize, characterize, select, and develop new materials useful for a variety of robust laser applications.

To illustrate the leverage of identifying new laser gain materials responsive to operational requirements, consider the problem of developing an efficient, long-lived multijoule/pulse tunable blue laser. One generic approach is based on the use of a suitable Cr^{3+} or Ti^{3+} doped crystal in a flashlamp-pumped laser, efficiently generating multijoule pulses across the 800-1000 nm region, with a pulse duration sufficiently short to enable efficient harmonic generation in the blue. The technical properties of a gain medium capable of this service are highly coupled to one another. One logical string on technical imperatives is as follows: to be long lived, the pump flashlamp must be operated at a loading small compared to the explosion limit; to provide a significant amount of pump energy at low load, the flashlamp pulse duration cannot be very short; to be efficient, the gain medium needs to couple well to the pump spectrum and to store energy for a time long compared to the pump pulse duration; however, the energy storage time, τ_f is inversely related to the stimulated emission cross-section, σ_L , the fluorescence (tuning) bandwidth, $\Delta\nu$, and the square of the index of refraction, n , according to the Fuchtbear-Ladenburg expression

$$\tau_f \leq \lambda_L^2 / 8\pi c n^2 \Delta\nu \sigma_L \quad (1)$$

where λ_L is the mean wavelength of the fluorescence emission band, and we have assumed that the fluorescence lifetime is equal to the radiative lifetime. Thus, long storage time and broad tunability drive σ_L to lower values. However, one cannot permit σ_L to fall below that value which results in adequate small signal gain and efficient energy extraction. For the latter, the output fluence of the laser must be set at one or two times the saturation fluence of the medium, $r_{\text{sat}} = hc / \lambda_L \sigma_L$; and for long-lived operation, r_{sat} must be at least several times smaller than the single shot damage fluence, r_d .

How do present materials stack up against these requirements and what is the prospect for materials with significantly more favorable technical properties? Figure 1 shows graphs of Equation 1, assuming $\lambda_L = 750$ nm and $\Delta\nu = 1700$ cm^{-1} , typical of chromium doped crystals.⁵ Curves are drawn for $n = 1.44$ and $n = 1.85$, typical of fluoride and oxide crystals, respectively. Points for a number of chromium doped oxide^{5,6} and fluoride^{7,8,9} crystals are also shown in Figure 1 (it is assumed here, reasonably, that the radiative lifetime is equal to the fluorescence lifetime). Experience to date with flashlamps and optical damage suggests that τ_f values of at least tens of microseconds and r_{sat} values < 10 J/cm^2 are required. The index dependence of Equation 1 favors fluoride materials by roughly a factor of two in meeting these conflicting requirements (fluorides also appear to be more immune to deleterious excited state absorption processes than oxides¹⁰; at the same time, fluorides generally are less thermomechanically robust than oxides and can sustain less thermal loading¹). In earlier work,⁹ we examined the elpasolite (K_2LiGaF_6) and the fluoride garnet ($\text{Li}_3\text{Na}_3\text{Ga}_2\text{F}_{12}$) possessing highly symmetric O_h sites for the chromium substitution, which yield low σ_L transitions and relatively high τ_f values. More attractive properties are found for the perovskite^{7,9} KZnF_3 , presumably from a lowering of the site symmetry from O_h due to charge compensation. Although these fluoride materials may be quite useful for applications requiring laser pulses of long duration ($> \mu\text{sec}$), for the present interest one needs to use materials with yet more distorted sites for the chromium ion. The tetragonal crystal¹¹ SrGaF_5 (and the aluminum¹² analog, as well) offers this possibility, according to published crystallographic studies.¹³ Figure 2 shows the absorption and emission spectra of an SrGaF_5 crystal doped with nominally 2 percent chromium. The peak fluorescence wavelength lies at 890 nm and the half-width extends from 800 to 980 nm, providing an attractive tuning range. The fluorescence lifetime is somewhat nonexponential with a mean value of approximately 50 microseconds (the two types of gallium sites, with slightly differing amounts of distortion, may account for this behavior). If this lifetime indeed turns out to be the radiative lifetime, then the corresponding laser parameters are estimated to be $\sigma_L \sim 5 \times 10^{-20}$ cm^2 and $r_{\text{sat}} \sim 4$ J/cm^2 . These values appear to be quite promising for a material

to be used in the type of laser under discussion. Since SrGaF_5 melts congruently¹¹ at the modest temperature of 812°C , and since gallium and chromium have nearly the same ion size, the prospect for growth of large, high-quality doped crystals would appear promising. Efforts in this direction are proceeding; at the same time, the average power handling capacity of this crystal will likely prove to be less than desired for all applications, and we are continuing the search for additional fluoride and oxide crystals with more robust thermo-mechanical properties. Using the technical methodology illustrated by the example above, we are identifying and assessing novel solid state gain media for a variety of applications.

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Figure 1. Parametrics of Cr^{3+} laser gain media.

Figure 2. Absorption and emission spectra of Cr:SrGaF . (Crystal grown by H. Guggenheim, AT&T Bell Laboratories; spectral data by M. Shinn, Lawrence Livermore National Laboratory).