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Specific Heat Loading in Nd:Glass Lasers[†]

by

William F. Krupke

Summary

The specific thermal load parameter, χ , for xenon flashlamp-pumped Nd:glass gain media is written as a function of neodymium concentration, pump pulse duration, and energy extraction efficiency. The currently available data on radiative and nonradiative decay probabilities of several commercial Nd:glassses are used to calculate and graph specific thermal load parameter values. By factoring these results into performance scaling relationships for zig-zag and disk lasers, specific Nd:glassses can be selected for optimized laser performance.

Introduction

The average power capability of repetitively pulsed Nd:glass lasers is directly related to the specific thermal load parameter, χ . This parameter is defined as the heat energy density released in the medium during pumping and after extraction, divided by the stored energy density at the time of energy extraction. The magnitude of χ depends on the radiative and nonradiative transition probabilities for the relaxation of pump-excited electrons in the glass, on the duration of pump pulse relative to the energy storage lifetime of the medium, and on the degree of gain saturation (or extraction efficiency). Now, in order to limit the temperature rise in the working medium of a high average power solid state laser, relatively thin slabs of glass must be used.¹ This, in

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[†] Author's Note: This UCID is derived from a Lawrence Livermore National Laboratory Internal Memorandum dated March 30, 1983, WFK 83-129.

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turn, dictates the use of high neodymium concentrations in order to achieve a high absorbed fraction of the pump radiation from xenon flashlamps. However, since the nonradiative (heating) rate increases with neodymium ion concentration, one is constrained in neodymium concentration. To optimize laser performance in the face of this constraint, one needs to know the value of the specific thermal load parameter as a function of neodymium ion concentration, normalized pump pulse duration, and extraction efficiency.

Heating Mechanisms

Heating of Nd:glass takes place when an excited neodymium ion relaxes from one J-level to the next lower lying J-level, with the emission of one or more phonons. (It is properly assumed here that pump radiation is filtered in such a manner that direct heating via UV and IR transitions of the glass host is precluded.) The multiphonon heating pathways for Nd:glass are shown schematically in Figures 1 and 2. Note that the amount of heating associated with each decay channel is expressed here as a fraction of the energy of a 1060 nm photon (9400 cm^{-1}). Figure 1 shows the multiphonon heating channels operative in the limit of zero neodymium concentration. A xenon flashlamp pump photon, with an average wavelength of 650 nm ($16,000 \text{ cm}^{-1}$), excites a neodymium ion with this average energy. The ion relaxes in about 10 ns to the upper laser J-level ($^4F_{3/2}$) with the release of 0.426 units of heat. Based on a limited amount of experimental data,² it is assumed here that the quantum yield of producing an $^4F_{3/2}$ excited state is unity for excitation in any of the higher lying Nd^{3+} ion bands. A number of decay pathways for the $^4F_{3/2}$ ion are operative, each with a different amount of heating associated with it. The ion may decay radiatively with probability A_{4j} to one of the ^4I J-levels by emission of a photon at 880, 1060, or 1350 nm. These decay processes are immediately followed by nonradiative relaxation to the $^4I_{9/2}$ J-level, producing 0.00, 0.22, 0.44 units of heat, respectively. Alternatively, the $^4F_{3/2}$ ion may relax nonradiatively to the $^4I_{15/2}$ J-level by emission of phonons,

followed by successive phonon relaxation through each of the lower ^4I J-levels to the ground J-level. This totally nonradiative process produces 1.22 units of heat. The average amount of heat produced per ion decaying from the $^4F_{3/2}$ J-level is the sum of the heat energies per channel, each weighted by the probability of decay via that channel. The branching ratios for radiative decay are virtually independent of glass composition and are 0.40, 0.50, and 0.10 for the 880, 1060, and 1350 nm transitions, respectively. (The branching ratio to the $^4I_{15/2}$ J-level is less than 0.005 and can be neglected.) The radiative lifetime of the $^4F_{3/2}$ J-level, of course, depends on the glass composition. The transition probability for multiphonon relaxation of the $^4F_{3/2}$ J-level depends on glass composition and is typically about 200 per second for silicate glasses³ and about 500 per second for phosphate glasses.^{4,5} In the presence of intense radiation at 1060 nm, the probability that the excited $^4F_{3/2}$ ion decays by emission of a 1060 nm photon becomes very large compared to all other decay probabilities, and the ensuing heat fraction is 0.22 units.

When the concentration of neodymium is increased above some relatively small value (typically 0.5 wt-%), an additional heating decay channel becomes increasingly important. Figure 2 shows this heating channel schematically. A neodymium ion in the metastable $^4F_{3/2}$ J-level interacts with a ground state neodymium ion, producing two neodymium ions in excited $^4I_{15/2}$ J-levels. The latter ions relax nonradiatively to their $^4I_{9/2}$ ground levels by phonon emission. Relaxation of a $^4F_{3/2}$ ion by this process results in the production of 1.22 units of heat. The rate of heating via this quenching mechanism, $W_q(\rho)$, depends on the concentration of neodymium ions as well as on the composition of the glass.

Dependence on Normalized Pump Pulse Duration

For purposes of illustration, consider the case of the Nd:glass medium being pumped with a constant pump intensity, P , for a duration,

τ_p . Let the medium be characterized by a fluorescence lifetime, τ_f . The ${}^4F_{3/2}$ ion density at any time is given by

$$n_4(t) = P\tau_f[1 - e^{-t/\tau_f}]; \quad 0 \leq t \leq \tau_p \quad (1)$$

$$n_4(t) = P\tau_f[1 - e^{-\tau_p/\tau_f}]e^{\tau_p/\tau_f}e^{-t/\tau_f}; \quad \tau_p \leq t \quad (2)$$

Since the total number of ${}^4F_{3/2}$ ions produced during time, t , is simply Pt , the fraction of ${}^4F_{3/2}$ ions remaining at time, t , is

$$f_r = (\tau_f/t)[1 - e^{-t/\tau_f}]; \quad 0 \leq t \leq \tau_p \quad (3)$$

$$f_r = (\tau_f/t)[1 - e^{-\tau_p/\tau_f}]e^{\tau_p/\tau_f}e^{-t/\tau_f}; \quad \tau_p \leq t \quad (4)$$

In practice, energy is extracted at time $t = \tau_p$ before fluorescence decay decreases the achieved excited population. The fraction remaining at $t = \tau_p$ is

$$f_r(\tau_p) = (\tau_f/\tau_p)[1 - e^{-\tau_p/\tau_f}] \quad (5)$$

which is graphed as a function of the normalized pump pulse duration τ_p/τ_f in Figure 3. For comparison, the more realistic case of a half-period sine pump pulse, calculated by Trenholme,¹ is also shown. Over the range of practical interest, the half-period sine pulse shape predicts a fraction stored which is 0-20 percent larger than that predicted by the square pump pulse shape.

Specific Thermal Load Parameter

If we let η_{ext} equal the extraction efficiency (defined as the number of 1060 nm photons extracted per unit volume divided by the number of 1060 nm photon equivalence stored per unit volume), we can write the specific thermal load parameter as the sum of four terms:

$$x = \frac{1}{f_r} \left\{ 0.426 + H(\rho)[1 - f_r] + \eta_{\text{ext}} f_r (0.22) + H(\rho)(1 - \eta_{\text{ext}}) f_r \right\} \quad (6)$$

The first term gives the heat fraction generated in producing ${}^4\text{F}_{3/2}$ ions. The second term gives the incremental heating fraction which occurs due to spontaneous decay during pumping. The third term gives the heat fraction generated by the fraction of ions which undergo stimulated emission at the end of the pump pulse. The fourth term gives the fractional heat generated by the unextracted ions as they decay spontaneously back to equilibrium. In this expression, the quantity $H(\rho)$ is defined by

$$H(\rho) = \frac{\sum_{j=0}^2 A_{4j} \Delta E_{j0} + \Delta E_{40} [W_{43} + W_q(\rho)]}{\sum_{j=0}^2 A_{4j} + W_{43} + W_q(\rho)} \quad (7)$$

where ρ is the neodymium concentration and the other parameters are defined in Figures 1 and 2. This equation expresses the decay-probability-weighted sum of the heat fractions for the various decay pathways shown in Figures 1 and 2.

The concentration dependent decay probabilities and values for $H(\rho)$ are summarized in Tables 1-5 for two silicate (ED-2, LG-650) and three phosphate (LHG-5, LG-750, S-16) glasses. It should be noted that both radiative and nonradiative transition probabilities vary from site to

site in a given glass³ and that one must adopt some appropriate average values for the ensemble of excited ions. The silicate glasses tend to have a fairly wide variation of radiative and nonradiative rates and a fairly low $^4F_{3/2}$ multiphonon decay rate, W_{43} , of about 200 per second.^{3,5} The phosphates tend to have a narrower variation of transition probability values, but are reported to possess a fairly high $^4F_{3/2}$ multiphonon decay rate, W_{43} , of 500 per second.^{4,5} Many of the phosphate glasses studied at LLNL are reported⁶⁻⁸ to have measured $^4F_{3/2}$ fluorescence lifetimes significantly in excess of the calculated radiative lifetimes. This is not only internally inconsistent, but it is also at odds with the presence of a significant multiphonon decay probability. In Tables 3-5, I have adopted W_{43} multiphonon decay rates of 260 per second for LHG-5, 145 per second for LG-750, and zero per second for S-16. Radiative rates are based on the best, low-water-content data available.^{7,8} However, the specific thermal load parameters may have to be significantly revised if more accurate measurements show that $^4F_{3/2}$ multiphonon decay rates are substantially larger than those adopted here, as is suggested by the Soviet work.⁴ With the aforementioned caveats, the data from these tables are used to construct the specific thermal load parameter plots shown in Figures 4-7.

Figure 4 shows χ as a function of neodymium ion concentration for a normalized pump pulse duration of 2 (upper cluster of curves) and of 0.7 (lower cluster of curves). Parameter values range from a low of about 0.8 to a high of about 2. For all the glasses save LG-650, χ varies almost linearly with neodymium concentration. LG-650 exhibits a relatively complex dependence of χ on neodymium concentration, dictated by its low radiative transition probabilities and its curious concentration quenching dependence (the latter may be an artifact of inaccurate fluorescence lifetime data).

Figure 5 shows χ as a function of normalized pump pulse duration for an assumed extraction efficiency of 0.5 and a fixed neodymium concentration of 6 wt-% in all glasses. The numbers shown along the curves give the pump pulse durations in microseconds. The panel at the top of Figure 5 reminds us of the fraction stored (for a half-period sine pulse shape) as a function of the normalized pump pulse duration. All of the glasses are well behaved, with a little "kink" occurring near unity pump pulse duration; no fundamental physical reason is readily obvious for this behavior, but the effect is of little consequence.

Figure 6 illustrates the dependence of χ on extraction efficiency, η_{ext} , assuming a normalized pump pulse duration of 0.7 (fraction stored of 0.78) and a neodymium concentration of 6 wt-% in all glasses. As noted previously, a high degree of gain saturation preferentially drives the excited electrons into the terminal $^4I_{11/2}$ J-level. Upon relaxation, this level produces relatively little heating (0.22 units/ion). High efficiency extraction thus "short circuits" the ion-ion quenching decay channel (with 1.22 units/ion), following energy extraction. Since ion-ion quenching and heating increases with neodymium concentration, we would expect high extraction efficiencies to be most effective in limiting the heating rate at high neodymium concentrations. This effect is shown in Figure 7, which plots the derivative of χ with respect to extraction efficiency, as a function of neodymium concentration. (A normalized pump pulse duration of 0.7, corresponding to a fraction stored of 0.78, is assumed.) Near a concentration of one wt-%, the χ parameters for phosphates exhibit virtually no dependence on extraction efficiency. However, at the higher concentrations, significant suppression of heating occurs at higher extraction efficiencies. The curves shown can be used to estimate the magnitude of a thermal gradient which would be imprinted in a glass slab or disk due to spatial variation of the intensity of an extraction beam.

Using Equations (6) and (7) and the data in Tables 1-5, other cross-cuts of the dependence of χ on neodymium concentration, normalized pump pulse duration, and extraction efficiency can be generated.

Comparison with Experiment

I know of only one (indirect) measurement of the specific thermal load parameter in a repetitively pulsed Nd:glass laser. J. Eggleston and B. Byer of Stanford have reported^{9,10} a specific thermal load parameter value of 1.9 using LHG-5 glass doped with 3.5 wt-% neodymium oxide and with a fluorescence lifetime of 240 microseconds. A pump pulse duration of 170 microseconds was used, which results in a fraction stored of 0.70 or 0.78, for square or half-period sine pulse shape approximations, respectively. From Figure 4, we predict a specific thermal load parameter value of 1.0; χ increases to 1.1 for $\eta_{\text{ext}} = 0$ appropriate to the measurement condition. Both of these values are about a factor of two lower than that reported. The value cited by Eggleston and Byer may be fairly inaccurate since it was deduced from a measurement of the thermally induced depolarization.¹⁰ At the same time, significant uncertainties remain as to the actual ${}^4F_{3/2}$ multiphonon decay rates in phosphate glasses. It is clear that multiphonon decay rates of candidate glasses should be accurately measured and that direct measurement of specific thermal load parameter values should be carried out on the same glasses. In the meantime, the graphical data presented here may be used to guide our design analyses of high performance, high-average-power Nd:glass lasers.

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Table 1
ED-2 Heating Function

ρ (wt-%)	τ_f (μsec)	A_f (sec^{-1})	$W_{43} + W_q(\rho)$	Heat Distribution			$H(\epsilon)$
				1060 nm	1350 nm	NR ₄₀	
0.03	360	2780	200	0.110	0.042	0.088	0.230
0.3	360	2780	200	0.110	0.042	0.088	0.230
1.0	330	3030	452	0.091	0.034	0.180	0.307
2.0	309	3236	658	0.085	0.032	0.248	0.365
3.0	240	4167	1590	0.066	0.025	0.465	0.556
4.3	199	5026	2447	0.055	0.021	0.594	0.670
6.0	127	7874	5296	0.035	0.013	0.821	0.869

$$A_{\text{rad}} = 2578 \text{ sec}^{-1}$$

$$\tau_{\text{rad}} = 388 \mu\text{sec}$$

$$W_{43} = 200 \text{ sec}^{-1}$$

Table 2
LG-650 Heating Function

ρ (wt-%)	τ_f (μsec) ^a	A_f (sec^{-1})	$W_{43} + W_q(\rho)$	Heat Distribution			$H(\rho)$
				1060 nm	1350 nm	NR ₄₀	
0.2	745	1342	262	0.080	0.028	0.238	0.346
1.0	678	1342	395	0.073	0.026	0.327	0.426
3.0	612	1634	533	0.066	0.023	0.398	0.487
5.0	608	1645	565	0.065	0.022	0.419	0.507
7.0	560	1786	704	0.060	0.021	0.481	0.562
8.0	435	2020	942	0.053	0.019	0.568	0.640
9.0	438	2283	1203	0.047	0.017	0.643	0.707
10.0	412	2427	1344	0.044	0.016	0.676	0.736

$$A_{\text{rad}} = 1080 \text{ sec}^{-1}$$

$$\tau_{\text{rad}} = 926 \mu\text{sec}$$

$$W_{43} = 262 \text{ sec}^{-1}$$

$$^a \tau_f = (\tau_1 + \tau_2)/2$$

Table 3
LHG-5 Heating Function

ρ (wt-%)	τ_f (μsec) ^a	A_f (sec^{-1})	$W_{43} + W_q(\rho)$	Heat Distribution			$H(\rho)$
				1060 nm	1350 nm	NR40	
0.3	319	3135	261	0.101	0.036	0.102	0.239
1.0	318	3134	271	0.101	0.036	0.105	0.242
3.0	275	3536	763	0.087	0.031	0.256	0.374
3.5	260	3846	972	0.082	0.030	0.302	0.415
6.0	199	5025	2152	0.063	0.023	0.522	0.608

$$A_{\text{rad}} = 2874 \text{ sec}^{-1} \quad \tau_{\text{rad}} = 348 \text{ } \mu\text{sec} \quad W_{43} = 261 \text{ sec}^{-1} \quad a_{\tau_f} = (\tau_1 + \tau_2)/2$$

Table 4
LG-750 Heating Function

ρ (wt-%)	τ_f (μsec)	A_f (sec^{-1})	$W_{43} + W_q(\rho)$	Heat Distribution			$H(\rho)$
				1060 nm	1350 nm	NR40	
0.5	356	2812	145	0.104	0.040	0.063	0.207
2.0	336	2972	305	0.099	0.037	0.125	0.261
3.0	315	3176	509	0.092	0.033	0.196	0.323
4.0	289	3461	795	0.085	0.032	0.280	0.392
6.0	234	4277	1610	0.069	0.026	0.459	0.554
8.0	185	5419	2752	0.054	0.021	0.620	0.695
10.0	145	6887	4220	0.043	0.016	0.748	0.807

$$A_{\text{rad}} = 2667 \text{ sec}^{-1} \quad \tau_{\text{rad}} = 356 \text{ } \mu\text{sec} \quad W_{43} = 145 \text{ sec}^{-1}$$

Table 5
S-16 Heating Function^a

ρ (wt-%)	τ_f (μsec)	A_f (sec^{-1})	$W_{43} + W_q(\rho)$	Heat Distribution			$H(\rho)$
				1060 nm	1350 nm	NR ₄₀	
0.0	(365)	2740	0	0.110	0.042	0.000	0.152
0.5	357	2801	61	0.108	0.041	0.027	0.178
5.0	272	3677	937	0.082	0.031	0.311	0.424
10.4	194	5155	2415	0.058	0.022	0.572	0.652
22.7	99	10,100	7361	0.030	0.011	0.889	0.930

$$A_{\text{rad}} = 2740 \text{ sec}^{-1}$$

$$\tau_{\text{rad}} = 365 \text{ } \mu\text{sec}$$

$$W_{43} \approx 0$$

^aReference 8

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 Figure 1: Multiphonon Decay Heating Channels in Nd:Glass
 (heat energy fractions in units of 9400 cm^{-1})

Figure 2: Ion-Ion Cross-Relaxation Heating Channel in Nd:Glass
(heat energy fraction ΔE_{40} in units of 9400 cm^{-1})

Figure 3

THERMAL LOADING PROPERTIES OF Nd:GLASS

Figure 4

Figure 5

SPECIFIC THERMAL LOAD χ vs η_{ext}

-17-

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Figure 6

$-dX/d\eta_{\text{ext}}$ vs ρ

Figure 7

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