Energy Storage and Heating Measurements in Flashlamp-Pumped Cr:Nd:GSGG and Nd:YAG

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Abstract—We have experimentally measured the energy stored and the heat generated in flashlamp-pumped Cr:Nd:GSGG for three Cr3+ concentrations in the range of 1-2 × 1020 ions/cm3. We find that the energy storage efficiency in these samples is 1.7 times greater than that of our Nd: YAG sample, and the normalized heating parameter χ, defined as the heat deposited per unit of stored energy, is 2.5 under the specified pumping conditions, with no evident dependence on the Cr3+ concentration. We further find that the measured χ value for our sample of Nd: YAG is 2.9 for the same pumping conditions. These observed χ values exceed expected values by factors of 1.1 and 2 for the Cr: Nd: GSGG and Nd:YAG samples, respectively. We have also directly measured the thermal focal length in the two materials; the lensing is shorter in GSGG by a factor of 2.6 for the same available output power or a factor of 4.5 for the same input pump power. Finally, we have calculated the expected thermal lensing using our measured heat loads with no adjustable parameters, achieving satisfactory agreement with our measured lensing values.

I. INTRODUCTION

This paper describes our research into the properties of Cr: Nd: GSGG, a material which has recently become the subject of intense interest because it offers a significant efficiency increase relative to Nd: YAG in advanced laser designs. Since the first experimental verification [1], [2] of the potential benefits of this new material, many independent measurements [3]-[7] have been reported regarding the lasing performance of individual rods in various pump cavity configurations. However, in order for this or related materials to be fully understood and exploited in the design of high-average-power solid-state lasers, a more detailed, systematic study is required to establish the simultaneous heat loading and energy storage efficiency factors. In a series of experiments, we have measured both of these interrelated parameters in Cr: Nd: GSGG laser rods with chromium concentrations in the range of 1-2 × 1020 ions/cm3 and a fixed Nd3+ concentration of 2 × 1020 ions/cm3. The findings are of significance for several reasons. First, they provide accurate quantitative pumping efficiency data that are essential when specifying an optimum Cr3+ concentration to resolve the pumping efficiency-gain uniformity tradeoff [8] which arises in practical laser designs. Second, an accurate measure of the heat load is required to determine the appropriate cooling requirements for any specific average output power requirement. Previous reports [5], [9] on thermal lensing disagree near a factor of two. Third, any anomalous Cr3+ dependence in the pumping efficiency or heat load would indicate the presence of one or more mechanisms preventing complete energy transfer from the Cr3+ to the Nd3+

In this work, we have used small-signal gain measurements to determine the dependence of Cr: Nd: GSGG pumping efficiency on Cr3+ concentration, while an interferometric calorimeter was employed to determine the heat deposited. These two numbers yield the normalized heating parameter χ, which is defined as the heat generated per unit of stored energy at 1.061 μm. This parameter is relatively independent of the pumping efficiency of the laser, but increases linearly with flashlamp pulse length [10] when it is longer than the fluorescence lifetime of Nd3+. We find that χ in our GSGG samples is ≤2.5 for ~250 μs long flashlamp pulses and that no Cr3+ dependence is evident. Hence, we conclude no Cr3+ concentration-dependent energy sinks exist within the concentration range studied. Using our sample of Nd: YAG in the same experimental apparatus yields a larger χ of ~2.9. These experimental values for χ assume stimulated-emission cross sections [11] of 3.1 × 10−19 cm2 and 6.5 × 10−19 cm2 for GSGG and YAG, respectively. While the higher χ for YAG may seem surprising in view of the enhanced short-wavelength pumping of GSGG relative to YAG, it is important to realize that the observed value of χ for our Nd: YAG sample is considerably higher than the "intrinsic" value. By intrinsic value, we are referring to a calculated heat generation value based on the quantum defect or energy mismatch between absorbed pump photons and the emitted laser photon; this heating mechanism is inherent in all solid-state materials under flashlamp-pumped conditions. Apparently, additional mechanisms cause χ for YAG to be higher than its intrinsic value, whereas they are less effective in GSGG. As a consistency check of the heating load measurements, we have directly measured the thermal lensing of the laser rods and find satisfactory agreement with thermal lensing calculations in which there are no adjustable parameters. The measurements show the thermal lensing of the 0.43 cm diameter Cr: Nd : GSGG rods to be approximately 2.6 times worse than that of Nd : YAG for the same available output power or approximately 4.5 times worse for the same input pump power.
The detailed experimental results are presented below following a brief discussion of the nature of \( \chi \) and a calculation of the expected values for Cr: Nd: GSGG and Nd: YAG.

II. CALCULATIONS

The fundamental cause of heat generation in flashlamp-pumped solid-state lasers is the energy difference or quantum defect between the absorbed pump photon and the emitted laser photon. This intrinsic heat load can be calculated using the relevant spectroscopic parameters of the materials of interest. A variety of processes, such as impurity absorption or quenching, that can lead to additional heating have been studied [12], [13]. These heat sources are difficult to predict and are most easily quantified by experimental measurements. A previous study [10], which considered \( \chi \) for Nd: YAG and several Nd-doped phosphate glasses, found that the actual values were consistently larger than the intrinsic values and that there was considerable host dependence.

The present study considers Cr: Nd: GSGG in comparison to Nd: YAG. In addition, we wish to establish not only the magnitude of \( \chi \) for Cr: Nd: GSGG, but also to determine whether it depends on the Cr\(^{3+}\) concentration. In a pedagogical discussion of this potential concentration dependence, it is useful to establish an energy conservation equation for the laser medium. We begin by defining \( E_{\text{abs}} \) as the total energy absorbed by the laser crystal. Of the total energy absorbed by the two Cr\(^{3+}\) pump bands, a portion \( H_1 \) is converted to heat. The major portion of \( H_1 \) is generated as the Cr\(^{3+}\) relaxes to the \( \text{^{2}E} \) level, while the remaining contribution arises from the relaxation to the ground state following fluorescence. The \( \text{^{4}T}_2 \) level is thermally populated from the \( \text{^{2}E} \) and because of a favorable spectral match with the Nd\(^{3+}\) pump bands, as well as the fast transfer time compared to the Cr\(^{3+}\) fluorescence lifetime, energy is efficiently coupled from the \( \text{^{4}T}_2 \) level of Cr\(^{3+}\) to the Nd\(^{3+}\). Additional heat \( H_2 \) generated directly by the Nd\(^{3+}\) ions arises, first, as the ions relax to the \( \text{^{4}F}_{3/2} \) upper laser level and, second, as the ions relax to the ground state following either radiative emission at 1.061 \( \mu \)m or quenching. The total heat \( H \) due to the optical pumping of Cr\(^{3+}\) and Nd\(^{3+}\) is defined as the sum of \( H_1 \) and \( H_2 \). The primary objective of the optical pumping, of course, is to store energy in the \( \text{^{4}F}_{3/2} \) level of the Nd\(^{3+}\). Of the total stored energy generated during the entire pump pulse, an amount \( E_{\text{st}} \) remains at the measurement time, and an amount \( E_{\text{fl}} \) is lost to Nd\(^{3+}\) fluorescence. Using these definitions, we can write a schematic energy balance equation:

\[
E_{\text{abs}} = H + E_{\text{st}} + E_{\text{fl}} + E'
\]

where all the parameters are implicitly time dependent and \( E' \) represents an unspecified energy sink (or heat source) that may or may not be Cr\(^{3+}\) dependent.

For the laser rods used in this study, nearly all the available pump light is absorbed, even with the lowest Cr\(^{3+}\) concentration. Hence, changes in \( E_{\text{abs}}, H, \) and \( E_{\text{fl}} \) with Cr\(^{3+}\) concentration will be quite small. However, by measuring both \( H \) and \( E_{\text{fl}} \) to determine the normalized heating parameter \( \chi = H/E_{\text{fl}} \), we achieve a high sensitivity to \( E' \) since if \( E' \) becomes appreciable as the concentration is increased, \( H \) and/or \( E_{\text{fl}} \) must also change in order that (1) be satisfied. In other words, we are looking for changes in the way the total absorbed energy is distributed, rather than changes in its absolute magnitude.

This approach of searching for Cr\(^{3+}\) concentration-dependent energy sinks by measuring \( \chi \) only works, of course, if the fluorescence term \( E_{\text{fl}} \) is independent of Cr\(^{3+}\) concentration. We have verified that this condition is met by independently observing a constant Nd\(^{3+}\) fluorescence lifetime in spectroscopic samples (thickness \( \sim 1 \) mm) having different Cr\(^{3+}\) concentrations. This observation is consistent with the results reported in [3].

We have calculated the intrinsic value of \( \chi \) for Cr: Nd: GSGG following the method outlined in [10]. This calculation involves two steps. First, the instantaneous stored energy is related to the time-resolved excited state densities of the Nd\(^{3+}\) and Cr\(^{3+}\) ions as a result of flashlamp pumping, with proper account being taken of the effects of the energy transfer from the Cr\(^{3+}\) ions to the Nd\(^{3+}\) ions. Second, we find the heat generated as a result of the relaxation of the excited states.

We begin by expressing the excitation rate of the ions in terms of the emission spectrum of the flashlamp, the absorption spectra of the ions, and the energy transfer kinetics of the Cr\(^{3+}\). The emission spectrum of xenon flashlamps, \( I(\lambda, J(t)) \), as a function of time \( t \) can be quite accurately modeled [14], [15] given the lamp current density \( J(t) \) and the bore diameter. The absorption spectra of the Cr\(^{3+}\) and Nd\(^{3+}\) ions in GSGG, \( \beta_{\lambda}(\lambda) \) and \( \beta_{\lambda}(\lambda) \), respectively, have been measured [11]. The energy transfer kinetics from Cr\(^{3+}\) to Nd\(^{3+}\) ions were extensively analyzed by Ostroumov et al. [16] using short-pulse excitation of Cr\(^{3+}\) ions in garnets. They have shown that, at a Nd\(^{3+}\) concentration of \( 2 \times 10^{20} \) cm\(^{-3} \), a fraction \( a = 0.15 \) of the excited Cr\(^{3+}\) ions transfers its energy into the nearest and next nearest neighbor Nd\(^{3+}\) ions within a short time \(< 0.2 \) \( \mu \)s. The remaining fraction \((1-a)\) of the excited Cr\(^{3+}\) ions is deexcited either by direct Cr\(^{3+}\) fluorescence (\( \sim 110 \) \( \mu \)s) or by transferring its energy slowly (\( \sim 10 \) \( \mu \)s) to the Nd\(^{3+}\) ions via either i) long-range dipole–dipole interaction or ii) migrationally limited relaxation. Using this information, and assuming a uniformly illuminated Cr: Nd: GSGG slab extending from \( y = 0 \) to \( \infty \), we can calculate the excited Nd\(^{3+}\) ion density in the \( \text{^{4}F}_{3/2} \) state \( N(y, t) \) and the excited Cr\(^{3+}\) ion density \( C(y, t) \):

\[
C(y, t) = \int_0^t dt' \left(1-a\right)P_s(y, t') \exp \left\{ -[t - t']/\tau_l - \gamma \sqrt{t - t'} - \bar{w}(t - t') \right\}
\]
\[
\frac{d}{dt} N(y, t) = -\frac{1}{\tau} N(y, t) - \left[ \frac{d}{dt} - \frac{1}{\tau_f} \right] C(y, t) - P_e(y, t) + P_N(y, t)
\]  

where \( P_e(y, t) \) and \( P_N(y, t) \) express direct excitation of \( \text{Cr}^{3+} \) and \( \text{Nd}^{3+} \) ions, respectively, due to absorption of flashlamp photons and are given by

\[
P_e(y, t) = \int d\lambda \eta_e(\lambda) \beta_e(\lambda) I(\lambda, J(t)) 
\cdot K(\lambda, t) \frac{\lambda}{hc} \exp \left\{ -y[\beta_e(\lambda) + \beta_e(\lambda)] \right\}
\]

(4)

\[
P_N(y, t) = \int d\lambda \eta_N(\lambda) \beta_N(\lambda) I(\lambda, J(t)) 
\cdot K(\lambda, t) \frac{\lambda}{hc} \exp \left\{ -y[\beta_N(\lambda) + \beta_e(\lambda)] \right\}
\]

(5)

Equation (2) is a straightforward generalization of eq. (1), given in [16], to include long-pulse pumping of the \( \text{Cr}^{3+} \) ions by the flashlamp. The terms in the exponential describe, respectively, the deexcitation of the \( \text{Cr}^{3+} \) ions by fluorescence with a lifetime \( \tau_f \), dipole-dipole energy transfer processes characterized by the parameter \( \gamma \), and the migrationally limited energy relaxation rate \( \tilde{\gamma} \). In this case, we have assumed the fast energy transfer times (\( \sim 0.2 \mu s \)) to be instantaneous compared to the dipole-dipole and migrationally limited relaxation energy transfer times (\( \sim 10 \mu s \)) and fluorescence decay (\( \sim 110 \mu s \)), so \( C(y, t) \) describes only the \( \text{Cr}^{3+} \) ions which lose energy slowly.

To obtain (3), we modified eq. (6) of [16] by adding the terms related to \( P_e(y, t) \) and \( P_N(y, t) \). The first term on the right-hand side of (3) represents the exponential decay of the \( \text{F}_{3/2} \) state with a lifetime \( \tau \). In GSGG, a nonexponential decay of the \( \text{F}_{3/2} \) state is not observed [11] for \( \text{Nd}^{3+} \) concentrations of less than \( 7 \times 10^{-5} \) ions/cm\(^3\). The following term, in square brackets, accounts for both the fast and slow mechanisms of energy transfer from \( \text{Cr}^{3+} \) to \( \text{Nd}^{3+} \) ions. This term simply states that the rate at which \( \text{Nd}^{3+} \) ions are pumped by energy transfer from the \( \text{Cr}^{3+} \) ions is equal to the negative of the time derivative of the excited \( \text{Cr}^{3+} \) ions after the fluorescence loss and direct pumping factors are removed. In other words, each time the \( \text{Cr}^{3+} \) population changes by a rate \( \epsilon \), a corresponding change in the \( \text{Nd}^{3+} \) population must occur.

In (4) and (5), the integrals over wavelength \( \lambda \) extend throughout the 340-900 nm absorption bands of \( \text{Cr}^{3+} \) and \( \text{Nd}^{3+} \); \( I(\lambda, J(t)) \) represents the light intensity emitted by the flashlamp into the wavelength interval between \( \lambda \) and \( \lambda + d\lambda \) at time \( t \) with current density \( J(t) \), and \( K(\lambda, t) \) represents the way that the reflectors in the laser head modify the flashlamp spectrum [14]. We also accounted for the UV cutoff (340-400 nm) by the cerium-doped quartz flashlamp envelope, although no attempt was made to include the small cerium fluorescence in the visible. \( \eta_e(\lambda) \) and \( \eta_N(\lambda) \) are the quantum yields of \( \text{Nd}^{3+} \) and \( \text{Cr}^{3+} \), respectively. \( \eta_N(\lambda) \) is the probability that the absorption of a pump photon produces a \( \text{Nd}^{3+} \) ion in the \( \text{F}_{3/2} \) state. Similarly, \( \eta_e(\lambda) \) is the probability that the absorption of a pump photon produces a \( \text{Cr}^{3+} \) ion in the \( \text{F}_{3/2} \) state. For generality, we have written the quantum yields as functions of wavelength. Although there is no reported direct measure for GSGG, it is generally agreed that \( \eta_e(\lambda) \) is independent of wavelength and is equal to unity (the value used in our analysis). In an earlier paper [10], we incorrectly assumed \( \eta_N(\lambda) \) for \( \text{Nd}^{3+} \) ions to be the same as the quantum efficiency \( \eta_0 \), which is the ratio of the decay rate constant \( \tau^{-1} \) attributable solely to fluorescence to the total decay rate constant \( \tau^{-1} \) of the \( \text{F}_{3/2} \) state. In our \( \text{Cr}: \text{Nd}: \text{GSGG} \) samples, \( \tau = 230 \mu s \), and from [11], \( \tau_R = 280 \mu s \) at room temperature. We have also assumed \( \eta_e(\lambda) \) to be unity and independent of wavelength.

The energy storage density \( \epsilon(y, t) \) can be determined from small-signal measurements; it represents the stored energy per unit volume which is available as photons at the output wavelength and is related to \( N(y, t) \) by

\[
\epsilon(y, t) = \frac{hc}{\lambda_0} N(y, t)
\]

where \( h \) is Planck's constant, \( c \) is the speed of light, and \( \lambda_0 = 1.061 \mu m \) is the laser wavelength.

The second step in the calculation involves the heat generated per unit volume, \( H(y, t) \):

\[
H(y, t) = H_f(y, t) + \int_0^t dt' \left[ q_N(y, t') + q_e(y, t') \right]
\]

(7)

In this equation, the function \( q_N(y, t) \) represents the rate of heat generation during direct flashlamp pumping of the \( \text{Nd}^{3+} \) ions due to the nonzero quantum defect between the absorbed photon energy and the energy \( hc/\lambda_1 \) of the \( \text{F}_{3/2} \) state; \( \lambda_1 = 880 \) nm for GSGG. Similarly, the function \( q_e(y, t) \) accounts for the quantum defect in \( \text{Cr}^{3+} \). The function \( H_f(y, t) \) is the heat generated due to fluorescence or quenching of \( \text{Nd}^{3+} \) ions in the \( \text{F}_{3/2} \) state. These three functions will now be defined in terms of other parameters.

\[
H_f(y, t) = \frac{1}{\tau_R} \left[ r + \frac{\lambda_0}{\lambda_1} \left( \frac{\tau_R}{\tau} - 1 \right) \right] \int_0^t dt' \epsilon(y, t')
\]

(8)

The term involving \( r/\tau_R \) accounts for the fraction of the energy stored in the \( \text{F}_{3/2} \) state that is converted to heat as a result of radiative decay to the \( \text{F}_{11/2}, \text{F}_{13/2}, \text{and F}_{15/2} \) states of the \( \text{Nd}^{3+} \) in GSGG. Here \( r \) can be expressed as
With $E_i$ and $B_i$ being the energy and fluorescence branching ratios, respectively, of these states. The second term in the brackets represents the heat generation as a result of nonradiative quenching of the $^4F_{3/2}$ state. Finally,

$$q_N(y, t) = \int d\lambda I(\lambda, J(t)) K(\lambda, t) \beta_n(\lambda)(f_1 + f_2)$$

\[ \times \exp \left[ -y(\beta_n(\lambda) + \beta_c(\lambda)) \right] \] (10)

with

$$f_1 = 1 - \eta_n(\lambda),$$

(11)

$$f_2 = \eta_n(\lambda)(1 - \lambda/\lambda_n),$$

(12)

and

$$q_c(y, t) = \int d\lambda I(\lambda, J(t)) K(\lambda, t) \beta_c(\lambda)$$

\[ \times (f_3 + f_4 + f_5) \exp \left[ -y(\beta_n(\lambda) + \beta_c(\lambda)) \right] \] (13)

with

$$f_3 = 1 - \eta_n(\lambda),$$

(14)

$$f_4 = \eta_n(\lambda)\left[ 1 - (\lambda/\lambda_n) \right] \eta_c,$$

(15)

$$f_5 = \eta_n(\lambda)\left[ 1 - (\lambda/\lambda_n) \right] (1 - \eta_c).$$

(16)

Since we have assumed the quantum yields of Nd$^{3+}$ and Cr$^{3+}$ to be unity, we have $f_1 = f_2 = 0$. The parameters $f_3$ and $f_5$ represent the two implicitly time-dependent pathways to consider when determining the energy defect due to the pumping of Cr$^{3+}$: 1) relaxation to the $^2E$ followed by energy transfer, with a probability $\eta_n$, to the Nd$^{3+}$, which subsequently ends up at the $^4F_{3/2}$ state, and 2) relaxation to the $^2E$ followed by Cr$^{3+}$ fluorescence at a wavelength $\lambda_n$ with a probability given by $(1 - \eta_c)$. Here, $\eta_c$ is the quantum transfer efficiency of Cr$^{3+}$ in GSGG, which is defined as

$$\eta_c = 1 - \frac{1}{\tau_c} \left[ \int_0^\infty C(y, t) \, dt \right] \left[ \int_0^\infty P_c(y, t) \, dt \right].$$

(17)

Equation (13) neglects the fact that the energy transfer and fluorescence do not occur simultaneously with the absorption of pump photons by Cr$^{3+}$ ions. However, this effect is not important since only the time-integrated heat rate in (7) is of practical interest. According to [16], for GSGG doped with $2 \times 10^{20}$ ions/cm$^3$ of Nd$^{3+}$, $\alpha = 0.15$, $\gamma = 220$ s$^{-1/2}$, and $\bar{w} = [\text{Cr}^{3+}] 4.4 \times 10^3$ s$^{-1}$; where the chromium concentration [Cr$^{3+}$] is in units of $1 \times 10^{20}$ ions/cm$^3$; hence, $\eta_c = 0.85$. We have assumed $\tau_c = 112$ $\mu$s, the same as in Cr$^{3+}$/GSGG [11].

In this model, we have neglected the reabsorption of the fluorescence light emitted by Cr$^{3+}$ at wavelengths that may overlap with any Nd$^{3+}$ absorption bands. Since about 15 percent of the excited Cr$^{3+}$ ions fluoresce and only a fraction of this emission band overlaps with the Nd$^{3+}$ absorption bands, the correction to $H$ and $E_n$ due to this process would be $\leq 5$ percent, and even less for $\chi$. Here we would like to point out that other simplifying approximations [10], [14]--[15] involving the model for the lamp emission spectrum and the head factor $K(\lambda, t)$ which we have assumed to be unity introduce a 5--10 percent uncertainty in the results. In our calculations, we have used a lamp current pulse shape approximated by a half sine-wave function of 325 $\mu$s full width at the base line, and a peak lamp current density of 3.9 kA/cm$^2$. These values closely approximate our typical experimental conditions. The Nd$^{3+}$ absorption cross-section data, energy levels, and branching ratios for GSGG were garnered from [11], as were the Cr$^{3+}$ absorption spectra.

In calculating $\chi$ for Nd$^{3+}$/YAG, (3) and (8) are replaced by (18) and (19), respectively, to account for concentration quenching of the $^2F_{3/2}$ state in Nd$^{3+}$/YAG, which manifests itself as a nonexponential fluorescence decay [12], [16]--[18] for concentrations above 0.4 percent [19]:

$$N(y, t) = \int_0^t dt' P_N(y, t')$$

\[ \times \exp \left[ -(t - t')/\tau_R - F(t - t') \right] \] (18a)

$$F(t) = \left\{ \begin{array}{ll} w_n & \text{for } t \leq t_1 \\ \eta_n \sqrt{t} & \text{for } t > t_1 \end{array} \right. \] (18b)

$$H_f(y, t) = \left[ \frac{\eta_c}{\tau_R} \frac{\hbar}{\lambda_1} \left( 1 - \eta_q \right) \frac{\hbar}{\lambda_1} \right]$$

\[ \times \left[ \int_0^t dt' P_N(y, t') \right] \] (19)

where the quantum efficiency is defined by

$$\eta_q = \frac{1}{\tau_R} \left[ \int_0^\infty N(y, t) \, dt \right] \int_0^\infty P_N(y, t) \, dt.$$

(20)

The terms in the exponential of (18a) describe, respectively, the deexcitation of the Nd$^{3+}$ ions due to fluorescence with radiative lifetime $\tau_R$ and the nonradiative quenching whose functional form [19] is given in (18b). The first term in square brackets in (19) accounts for heat due to energy relaxation following fluorescence, and the second term represents the heat generation as a result of nonradiative quenching of the $^2F_{3/2}$ state.

The main uncertainty in the calculated value of $\chi$ for YAG arises from the uncertainty in the value of $\tau_R$. Krupke [20] has used the Judd--Ofelt theory to predict $\tau_R = 259 \pm 25$ $\mu$s. Experimental fluorescence measurements as long as 280 $\mu$s have been reported [17], while [16] and [19] state $\tau_R = 250$ and 260 $\mu$s, respectively. We find that $\chi$ decreases by $\sim 15$ percent as $\tau_R$ is increased from 250 to 280 $\mu$s. We have used $\tau_R$ from [19] and the accompanying values of $\tau_N = 10^{-1/2}$, $w_n = 1 \times 10^3$ s$^{-1}$, and $t_1 = 90$ $\mu$s, for a Nd$^{3+}$ concentration of $1.5 \times 10^{20}$ ions/cm$^3$. Note that this parameter set predicts $\eta_q = 0.91$, which is inconsistent with generally quoted values [17] of
mine a value of $\chi$ averaged over the cross section of the 4.3 mm diameter rod. However, as is shown in Fig. 1, the theoretical depth dependence is comparable to our experimental and computational uncertainties and is only of minor practical consequence. Hence, we will use the $\chi$ value at a 1 mm depth (roughly half the radius of the uniformly pumped rod) for comparison to the experimental values; the appropriate values are 2.2 for Cr: Nd: GSGG and 1.5 for Nd: YAG. We find that calculated values of $\chi$ are insensitive to the uncertainties in many of the input parameters of the model: $\chi$ varies less than 5 percent as we separately vary the Cr$^{3+}$ concentration (1–2 $\times$ 10$^{20}$ cm$^{-3}$), pulse length (295–325 $\mu$s at the base), peak flashlamp current (3–3.9 kA/cm$^2$), and the Cr$^{3+}$–to-Nd$^{3+}$ energy transfer parameters $a$ (0.1–0.2), $\gamma$ (200–260 s$^{-1/2}$), and $w$ (0.4–4 $\times$ 10$^{-4}$ s$^{-1}$). Moreover, as a check on the sensitivity of the calculations to the Nd$^{3+}$ pump band linewidths, we artificially broadened the absorption spectra so that it resembled those of Nd$^{3+}$ doped in phosphate glass and found the calculated $\chi$ to vary by $\leq$ 5 percent.

### III. Experimental Results

The determination of $\chi$ requires the measurement of two parameters: the maximum upper-laser-level stored energy, and the heat deposited in the laser medium as a consequence of the optical pumping process. In the present study, the stored energy was calculated from small-signal gain measurements, while an interferometric calorimeter was used to determine the heat deposition by measuring the thermally induced optical path length changes of the laser rod under test. We studied three different co-doped GSGG laser rods having Cr$^{3+}$ concentrations of 1.0, 1.5, and 2.0 $\times$ 10$^{20}$ ions/cm$^3$ and a Nd$^{3+}$ concentration of 2 $\times$ 10$^{20}$ ions/cm$^3$. The Cr$^{3+}$ concentrations fall into the range that might be appropriate for typical laser hardware with active media having transverse dimensions of $\sim$ 1/2 to 1 cm. The Nd$^{3+}$ concentration was selected to be near the onset of concentration quenching [11]. The GSGG crystals were grown by the Airtron Division of Litton Industries, and all concentrations were verified by spectroscopic analysis at Lawrence Livermore National Laboratory. The Nd: YAG rod, also employed in an earlier study [10], was grown by Airtron in 1983. This rod was extracted from the top of the boule, and the nominal Nd$^{3+}$ concentration was 1.1 atomic percent (1.5 $\times$ 10$^{20}$ ions/cm$^3$), which is standard in the industry. Because concentration self-quenching in Nd: YAG would increase $\chi$, we concluded that at least an approximate verification of the concentration was appropriate. To this end, we measured the 1.06 $\mu$m fluorescence decay at one end of the rod, following excitation with a Q-switched 532 nm pulse. Pumping occurred a few millimeters from the rod face, perpendicular to the rod axis, and we monitored the fluorescence emitted through the face. This measurement technique avoids spurious decay rate results, which could arise from effects such as radiation trapping. For our Nd: YAG rod, the fluorescence decays with an initial time constant of 210 $\mu$s, while a longer time constant of 240
\( \mu s \) is observed after a \(-400 \mu s \) delay. Our results are fully consistent with other fluorescence decay measurements [17] for an identically sized Airtron Nd:YAG rod with the same nominal concentration. This consistency suggests that our YAG sample is typical. Reference [17] also presents the measured variation of the fluorescence decay rate along the length of the rod. Using their data and the published [18], [19] results for the variation of fluorescence lifetime with concentration, we estimate a concentration variation of \( \pm 10 \) percent throughout the length of their rod. We assume the same upper limit to the concentration variation would also apply to our rod sample. Our GSGG samples were nominally 4.6 cm long, and the YAG sample was 4.4 cm long; all the rods were 0.43 cm in diameter with fine-ground barrels. The GSGG rod faces were cut and polished at \( 2^\circ \) off normal to the rod axis, while the YAG rod faces were normal to the axis.

In order that meaningful comparisons could be made, we used the same flashlamp and pump cavity for all samples. The rod was pumped by a 0.3 cm bore, 3 cm arc length, and 450 torr xenon flashlamp. The cavity walls were coated with electrodeposited gold, and parts of the surface were covered with packed barium sulfate powder. The cerium-doped quartz flashlamp envelope (1 mm wall) minimized the UV emission below 400 nm. A 0.7 mm thick samarium-doped glass tube around the rod further filtered out the UV. Water could be passed between the rod and glass tube to cool the rod; the water also filtered out IR wavelengths longer than 1.4 \( \mu m \). A single-mesh pulse-forming network (PFN) provided \( \sim 250 \mu s \) long flashlamp current pulses (measured at the 20 percent points), which are comparable to the \( \sim 230 \mu s \) Nd\(^{3+} \) fluorescence lifetimes in the Cr: Nd: GSGG and Nd: YAG samples used. The PFN component values were \( C = 72.5 \mu F \text{ and } L = 98.1 \mu H \).

\( Q \)-switched Cr: Nd: GSGG and Nd: YAG lasers were employed as probes to measure the small-signal gain of the Cr: Nd: GSGG and Nd: YAG samples, respectively. Probe pulses were adjusted for temporal overlap with the occurrence of maximum gain in the test rod. The probe fluence, even following amplification, was always less than 1 percent of the saturation fluence, thereby guaranteeing operation in the small-signal regime. By simultaneously measuring the transmitted probe pulse through the rod along with a 4 percent sample of the incident pulse, the shot-to-shot energy fluctuations were normalized out. Any passive insertion losses were also taken into account in these measurements by calculating the ratio of the normalized amplifier-active and amplifier-passive pulse energies. Thus, we accurately obtained the small-signal gain \( G \). The energy stored in the rod \( E_{st} \) is obtained from \( G \) using the relation

\[
E_{st} = \frac{h \nu}{\sigma k} A \ln G = \frac{h \nu}{k} A \int_0^l N(z) dz \tag{21}
\]

where \( h \nu \) is the photon energy, \( \sigma \) is the stimulated-emission cross section of the particular sample, \( A \) is the rod cross-sectional area, \( l \) is the length of the rod, and \( N(z) \) is the inversion density at the position \( z \) along the rod axis. In calculating \( E_{st} \), we used stimulated-emission cross-section values [11] of \( 3.1 \times 10^{-19} \) and \( 6.5 \times 10^{-19} \) cm\(^2\) for GSGG and YAG, respectively. While the precise values of these cross sections are difficult to specify, due to such a wide range of values being quoted in the literature, the values chosen for use in this analysis have been shown to be consistent (to within \( \pm 15 \) percent) with amplifier energy extraction measurements performed in our laboratory. The factor \( k \) in (21) accounts for the fact that only a fraction of the total population inversion is in the upper \( 2F_{5/2} \) subgroup which participates in the stimulated emission. The level splittings are 60 and 86 cm\(^{-1}\) for Cr: Nd: GSGG [11] and Nd: YAG [21], respectively, leading to room-temperature values for \( k \) of 0.43 and 0.40, respectively. Using a video system [22] to display the intensity distribution of the amplified probe beam, we estimate that the transverse gain distribution was uniform to within \( \pm 15 \) percent.

Table I presents the measured values of the energy storage efficiency \( \eta_{st} \), which we define as the ratio of laser photon energy stored in the rod \( E_{st} \) to the electrical energy stored in the PFN. For each rod sample, \( \eta_{st} \) was calculated from a graph of \( \ln G \) versus pump energy in the linear region (\( G \leq 4 \)) well below the ASE-limited gain saturation regime (\( G_{max} \leq 20 \)). We observed that the Cr: Nd: GSGG is approximately 1.7 times more efficient than Nd: YAG. Notice that, within the experimental error, the measurements indicate virtually no dependence on the Cr\(^{3+} \) concentration. This behavior is due to "pump light saturation"; even for the lowest Cr\(^{3+} \) concentration studied, the pump light absorption coefficient is high enough that essentially all of the available light is absorbed, so doubling the Cr\(^{3+} \) concentration cannot increase the pumping efficiency.

The heat deposited in the test rod during flashlamp pumping was deduced from the observed temperature rise of the uncooled rod following the flashlamp pulse. The rod temperature change was obtained by measuring the shift of the interference fringes of a Michelson interferometer with the test rod in one of the two legs. One advantage of this method is that unavoidable pumping nonuniformities that arise along the length of the rod due to the finite lengths of the flashlamp and rod are averaged out in the same way as the small-signal gain measurement averages out longitudinal stored energy nonuniformities. Thus, the experimental value of \( \chi \) is not affected by pumping nonuniformities along the length of the rod. During all these flashlamp-induced temperature changes, the test rod barrel was in contact with a bath of static water.

Fig. 2 shows a schematic of the experimental apparatus used to quantify the fringe shifts. A HeNe probe laser is used to illuminate the Michelson interferometer, which contains the test rod in one of the legs. HeNe wavelengths of 543 and 633 nm were used for Cr: Nd: GSGG and Nd: YAG, respectively. The fringe pattern is directed into a TV camera and a video recording system to allow fringe shifts to be stored for later quantification. The use of the video system provides a temporal resolution of 1/30 s. Because the rod is not actively cooled during the heat den-
TABLE I

MEASUREMENTS OF THE NORMALIZED HEATING PARAMETER $\chi$ AND THE ENERGY STORAGE EFFICIENCY $\eta_s$ FOR Cr: Nd: GSGG AND Nd: YAG ($\sigma = 3.1 \times 10^{-19}$ cm$^2$ for GSGG, $\sigma = 6.5 \times 10^{-19}$ cm$^2$ for YAG), PUMP PULSE LENGTH = 250 $\mu$s (20 PERCENT POINTS), 450 TORR XENON LAMP, CERIUM-DOPED ENVELOPE, AND WATER-COOLED LASER ROD

<table>
<thead>
<tr>
<th>MATERIAL</th>
<th>CONCENTRATION ($10^{20}$/cm$^3$)</th>
<th>$\eta_s$ (PERCENT)</th>
<th>$\chi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr: Nd: GSGG</td>
<td>1</td>
<td>3.5$\pm$0.05</td>
<td>2.5$\pm$0.2</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
<td>3.7$\pm$0.11</td>
<td>2.4$\pm$0.2</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>3.6$\pm$0.14</td>
<td>2.4$\pm$0.2</td>
</tr>
<tr>
<td>Nd: YAG</td>
<td>----</td>
<td>2.1$\pm$0.2</td>
<td>2.0$\pm$0.3</td>
</tr>
</tbody>
</table>

*Nominal boule concentration.
$^a$In [10], we previously used $\sigma = 7.6 \times 10^{-19}$ cm$^2$ and pulselength = 150 $\mu$s (20 percent points).

Fig. 2. Schematic of Michelson interferometer for heat deposition measurements in flashlamp-pumped laser rods.

Fig. 3. Time-lapse sequence of Michelson interferometer fringes for 8 J flashlamp-pumped Nd: YAG. The interferometer wavelength is 633 nm. (a) Immediately before the pump pulse ($\Delta t = 0$). (b) Two frames after the pump pulse ($\Delta t = 1/15$ s). (c) Eleven frames after the pump pulse ($\Delta t = 0.37$ s). The frame rate is 30 Hz.

\[
\Delta T_F = \frac{\lambda}{2I\frac{dn}{dT} + (n - 1)\alpha}
\]

where $\lambda$ is the appropriate HeNe wavelength, $I$ is the length of the laser rod, $n$ is the refractive index, $dn/dT$ is the change in $n$ with temperature, and $\alpha$ is the thermal expansion coefficient.

The quantity $[dn/dT + (n - 1)\alpha]$ in (23) is not available in the existing database on Cr: Nd: GSGG at 543 nm, although $dn/dT$ at 1.06 $\mu$m has recently been reported [23]. Thus, we performed the following experiment to measure $\Delta T_F$. With the flashlamp turned off, we slowly increased the temperature of the water flowing around the rod using a heater immersed in the water reservoir. A thermocouple measured the water temperature near the rod. The heating rate was kept small ($\sim 0.7$ K/min) in order that the rod temperature would always be uniform (the thermal time constant for this rod is $0.3$ s). Typically, we observed the temperature change corresponding to 30 fringe periods in the temperature region centered near 30°C. Then, the heater was turned off, the rod began to cool, and the fringes shifted the opposite way, corresponding to 30 fringe periods. The heating and cooling numbers agreed within a few percent. The average value was $(0.276 \pm 2$ percent) K/fringe for a probe wavelength of 543 nm. Based on these $\Delta T_F$ results, we have
calculated a $dn/dT$ of $(14 \pm 0.6) \times 10^{-6} \text{K}^{-1}$ at 543 nm using (23) with literature values of $n = 1.982$ and $\alpha = 7.5 \times 10^{-6} \text{K}^{-1}$ (see Table II). This is in reasonable qualitative agreement with [23], given the known dispersion for Cr:Nd:GSGG.

This immersion heater technique for measuring the fringe temperature coefficient was confirmed by using Nd:YAG with the same interferometers used: the Michelson and the Fizeau formed by the parallel end faces of the Nd:YAG rod itself (the latter approach was the one used in [10]). These complementary interferometric measurements allow the determination of not only $\Delta T_f$, but also the individual parameters $\alpha$ and $dn/dT$. The measured values of $\Delta T_f$ were $(0.385 \pm 2\text{ percent})$ and $(0.265 \pm 2\text{ percent})$ K/fringe for the Michelson and Fizeau interferometers, respectively. These values yield $\alpha = (6.95 \pm 0.6) \times 10^{-6} \text{K}^{-1}$ and $dn/dT = (9.49 \pm 0.8) \times 10^{-6} \text{K}^{-1}$ (the latter value assumes $n = 1.84$ at 543 nm as derived from published data [24] for undoped YAG). The measured value of $\alpha$ is well within 1 percent of the literature value [25], [26]. In addition, our value for $dn/dT$ agrees with the calculated value of $8.8 \times 10^{-6} \text{K}^{-1}$ derived from [27] within our experimental uncertainty.

The values of $\chi$ determined from our experiments are presented in Table I; the relevant material parameters used in the data reduction are summarized in Table II. First, we note that $\chi$ is independent of Cr$^{3+}$ concentration (within experimental uncertainties). We conclude, following the reasoning outlined in Section II, that there are no concentration-dependent energy sinks for Cr$^{3+}$ concentrations in the range studied. This conclusion is consistent with the suggestion in [3], which was based on Cr$^{3+}$ fluorescence lifetime measurements. The second important aspect of our results is that our samples of GSGG have a lower $\chi$ than our sample of YAG under similar pumping conditions.

The final experiments performed were aimed at actually measuring the thermal lensing of the GSGG and YAG rods. These measurements were motivated by two factors. First, they offer us the opportunity to check the overall consistency of our heating measurements by comparing measured and calculated values of the thermal lensing. Second, the lensing measurements will help eliminate uncertainties raised by the fact that previous estimates [5], [9] differ by nearly a factor of two. Our measurements were made by determining the effect of the thermal lens on a collimated HeNe beam using the experimental arrangement shown schematically in Fig. 4. The output of a 543 nm HeNe laser is directed through a pair of lenses having focal lengths of $f_1 = -40$ mm and $f_2 = +250$ mm, thereby expanding the beam size by a factor of $m = 6.25$. Upon passing through the test rod, which is mounted in a pump cavity, the collimated beam is focused by a lens of focal length $f_3 = 100$ cm onto a screen placed in the focal plane. With the test rod passive, the spacing between lenses $f_1$ and $f_2$ is adjusted to produce a minimum spot size on the screen, thereby ensuring a collimated beam. Any static spherical lensing in the test rod is compensated out in this manner. When the test rod is pumped (at 20 Hz) and water-cooled to produce a positive thermal lens, the size of the focused spot on the screen increases. The negative lens is then translated by a distance $\delta$ that collocates the beam, as determined by once again minimizing the spot size on the screen. Using this experimental approach with the indicated focal lengths, the displacement $\delta$ varies up to several centimeters, and it can be repeatedly determined to an accuracy of ~1 mm. The thermal focal length $f_T$ can be related to the parameter $\delta$ using simple ray tracing; the appropriate relationship is

$$\delta = -m^2 \frac{f_1^2}{f_T} \left[ \frac{1}{1 - m f_1 - \frac{L}{f_T}} \right]$$

where a positive value corresponds to a greater distance between lenses, $m$ is the magnification of the telescope, and $L = 9$ cm is the distance between the telescope output lens and the test rod. Referring to this relationship, we see that in addition to allowing long focal lengths to be conveniently measured on a standard laboratory bench, the accuracy is quite good, limited primarily by the <10 percent accuracy in determining $\delta$.

The expected thermal focal lengths can be calculated using the formula

$$f = \frac{\kappa A}{P_n} \left[ \frac{1}{2} \frac{dn}{dT} + \alpha C_r \alpha n^3 + \frac{\alpha (n - 1)}{f} \right]^{-1}$$

Table II

<table>
<thead>
<tr>
<th>PARAMETER</th>
<th>Cr: Nd:GSGG</th>
<th>Nd:YAG</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho (g/cm^3)$</td>
<td>6.503$^a$</td>
<td>4.552$^a$</td>
</tr>
<tr>
<td>$C_r (J/Kg)$</td>
<td>0.246$^b$</td>
<td>0.060$^b$</td>
</tr>
<tr>
<td>$\Delta T_f (K^{-1}), \text{MICHELSON}$</td>
<td>0.276$^{+2}$</td>
<td>0.505$^{+2}\pm 0.356$</td>
</tr>
<tr>
<td>$\Delta T_f (K^{-1}), \text{FIZEAU}$</td>
<td>0.265$^{+1}$</td>
<td>0.986$^{+1}$</td>
</tr>
<tr>
<td>$dn/dT (10^{-6} K^{-1})$</td>
<td>1.982$^c$</td>
<td>1.823$^c$, 1.84$^c$</td>
</tr>
<tr>
<td>$C (J/0.426\alpha)$</td>
<td>7.5</td>
<td>6.90$^d* $</td>
</tr>
<tr>
<td>$\kappa (W/m)^{0.6}\alpha$</td>
<td>6.02, 7.0$^e$</td>
<td>9.76, 12.9$^d$</td>
</tr>
<tr>
<td>$C_r, \phi$</td>
<td>0.019, 0.001</td>
<td>0.019, 0.002</td>
</tr>
</tbody>
</table>

$^{a}$Averaged values from [11].
$^{b}$Calculated from Sellmeier formula in [28]. Sellmeier coefficients incorrectly listed in [11].
$^{c}$[25].
$^{d}$Estimated from [24] at $\lambda = 543$ nm.
$^{e}$[29].
TABLE III
THERMAL LENSGING COMPARISON OF Cr: Nd: GSGG AND Nd: YAG AT
A = 543 nm

<table>
<thead>
<tr>
<th>MATERIAL</th>
<th>EXPERIMENTAL MEASUREMENT</th>
<th>CALCULATION*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr: Nd: GSGG</td>
<td>7.2 ± 0.2</td>
<td>8.5, 7.3</td>
</tr>
<tr>
<td>Nd: YAG</td>
<td>1.6 ± 0.1</td>
<td>2.5, 1.9</td>
</tr>
</tbody>
</table>

*Based on Eq. (25), using C, value only.

where \( \chi \) is the thermal conductivity, \( A \) is the rod cross-sectional area, \( P_a \) is the total power absorbed as heat in the rod, \( n \) is the refractive index, \( \alpha \) is the thermal expansion coefficient, \( C_{\phi} \) are the photoelastic coefficients for radially and tangentially polarized light, respectively, \( r \) is the rod radius, and \( l \) is the rod length. The parameter values used in these calculations are listed in Table II. The thermooptic coefficients used in the calculation. Given the large uncertainties in the literature concerning the many relevant thermooptic coefficients, this agreement is quite satisfactory.

IV. CONCLUSION

Within the experimental error, our results indicate little or no change in the normalized heating parameter \( \chi \) for Cr: Nd: GSGG as a function of the \( Cr^{3+} \) concentration in the range of \( 1-2 \times 10^{20} \) ions/cm\(^3\). We conclude that concentration-dependent energy sinks and heat sources for \( Cr^{3+} \) have a minimal effect, if any, in these samples, and in fact, their existence appears unlikely for the concentration range studied.

The second aspect of our results is that \( \chi \) for our GSGG samples is less than for our YAG sample under similar operating conditions. This may be surprising in view of the fact that the enhanced short-wavelength pumping due to the \( Cr^{3+} \) pump bands increases the average quantum defect; this factor would tend to increase \( \chi \) in GSGG relative to YAG, as the calculated intrinsic values show. However, it is important to realize that the observed value of \( \chi \) for our YAG rod is twice the intrinsic value estimated in Section II. For GSGG, on the other hand, the observed \( \chi \) is very close to the intrinsic value. Therefore, it is appropriate to consider what factors might lead to a high measured value of \( \chi \) for YAG. One possibility is the quantum efficiency \( \eta Q \). Our fluorescence decay measurements place a lower limit on the decay time of 210 \( \mu \)s. Assuming a maximum value of \( \tau_g \) as long as 280 \( \mu \)s leads to a worst case \( \eta Q \) value of 0.75. Hence, we cannot justify an \( \eta Q \) less than the value 0.75 used in our calculations. Perhaps another possible explanation for our measured \( \chi \) value is transient color-center generation, which has been reported [31] in Nd: YAG during xenon flashlamp pumping (no UV blockage of lamp radiation). Those color-center absorption bands, which peak at 525 nm, would be expected to increase \( \chi \). In that study [31], however, the transient color-center formation was considerably reduced when the excitation radiation was filtered with a glass plate. Thus, we do not expect transient color-center absorption to be a contributing factor to our measured \( \chi \) value since our cerium-doped lamp envelope and Sm: glass flowtube substantially attenuate the UV portion of the flashlamp radiation. Hence, additional heating mechanisms must be active in our YAG sample. Examples may include effects such as dark \( Nd^{3+} \) ions [12] or impurities [13].

Considering the importance of Nd: YAG and the possible variations among Nd: YAG boules, it would be desirable to repeat these \( \chi \) measurements for a wide variety of samples in order to determine and, if it is hoped, to remove the additional heating source(s) and energy sink(s). Similar trends were reported previously for the measured and calculated values of \( \chi \) in various Nd: glass materials [10]. Finally, our results indicate that the thermal lensing in our GSGG samples is 2.6 times stronger than in our YAG sample for the same available output power, and our measured and calculated values agree within 15 percent.

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REFERENCES


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David A. Rockwell, for a photograph and biography, see this issue, p. 1140.

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