

Optical gain and loss studies in $\text{Ce}^{3+}:\text{YLiF}_4$

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Transient and stable color centers created by optical pumping at 308 nm have been studied in $\text{Ce}^{3+}:\text{YLiF}_4$. Since the color centers are absorptive at the cerium emission wavelengths, they degrade the $\text{Ce}^{3+}:\text{YLiF}_4$ laser performance. The color centers are produced following a multiphoton ionization of the Ce^{3+} ions. Rate equations that incorporate both creation and photobleaching processes are developed to model the observed kinetics and intensity dependence of the color-center density. Measurements of the single-pass gain at 325 nm as a function of the repetition rate of the pump laser are interpreted in terms of the relaxation time of the transient centers.

INTRODUCTION

The broad $5d \rightarrow 4f$ vibronic emission bands of trivalent cerium-doped crystals make these materials ideal candidates for tunable solid-state lasers. Such cerium lasers would usually operate in the blue and near UV, at much shorter wavelengths than typical $3d$ systems such as Cr^{3+} and Ti^{3+} . However, $\text{Ce}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$ shows a strong excited-state absorption¹⁻⁵ at the Ce^{3+} emission wavelengths that quenches the gain, and $\text{Ce}^{3+}:\text{CaF}_2$ is troubled by the formation of photochromic centers⁶ following an excited-state absorption at the UV pump wavelengths. Both $\text{Ce}^{3+}:\text{LiYF}_4$ (YLF) and $\text{Ce}^{3+}:\text{LaF}_3$ do show optical gain on the $5d \rightarrow 4f$ transition,^{7,8} but these two systems seem to have problems that severely limit their laser performance. In this paper we report on the origin of these performance-limiting characteristics in $\text{Ce}^{3+}:\text{YLF}$.

The absorption and emission spectra of $\text{Ce}^{3+}:\text{YLF}$ have been presented by Ehrlich *et al.*⁷ The broad absorption bands that peak at 195, 205, 240, and 290 nm result from transitions from the $4f$ ground state to the crystal-field split $5d$ levels of the Ce^{3+} ion. The emission bands peak at 310 and 325 nm and involve transitions from the lowest $5d$ level to the $4f(^2F_{5/2})$ and $4f(^2F_{7/2})$ Stark manifolds. The 40-nsec radiative lifetime of the lowest $5d$ level results from the electric-dipole-allowed character of the $5d \rightarrow 4f$ transition. Stimulated emission and laser action from 306 to 330 nm occur on the transition between the lowest $5d$ level and the $4f$ levels. However, the operation of the $\text{Ce}^{3+}:\text{YLF}$ laser is hampered by several poor performance characteristics.⁷ These include an early onset of saturation and rolloff in the above-threshold gain and power output as well as a drop in the output for pulse repetition rates above 0.5 Hz. Our effort here is to show that an excited-state absorption of the UV pump light is responsible for a photoionization of the Ce^{3+} ions, which in turn leads to the formation of transient and stable color centers. The color centers have a deleterious effect on the lasing characteristics of $\text{Ce}^{3+}:\text{YLF}$ since they absorb at the cerium emission wavelengths. The growth and relaxation of these centers influence the gain saturation and pump rate limitation of the $\text{Ce}^{3+}:\text{YLF}$ laser.

Following a brief description of the experimental methods, measurements of the characteristics of color centers

created by the 308-nm pumping of the cerium ions are presented. The creation of the centers is shown to arise from a nonlinear absorption of the UV pump light, mediated by a simultaneous photobleaching process. Rate equations are then developed to model the observed intensity dependence of the color-center absorption. The marked decrease in the single-pass gain at 325 nm with increasing pulse repetition rate indicates the significant absorptive loss due to the transient centers.

EXPERIMENTAL METHODS

The sample used in our measurements was an 8 mm \times 8 mm \times 13 mm block of LiYF_4 containing a nominal 1% concentration of substitutional Ce^{3+} ions and was oriented such that the c axis was normal to the 8 mm \times 8 mm face. An additional sample of undoped YLF was also studied in order to separate the effect of the cerium ions from any purely host lattice phenomena.

The experimental arrangement used to investigate the characteristics of the UV-induced absorption is shown schematically in Fig. 1. The cerium ions were excited by the unpolarized 308-nm output of a XeCl excimer laser having an 8-nsec pulse width. The 308-nm wavelength is in the low-energy tail of the lowest $4f \rightarrow 5d$ absorption band of the Ce^{3+} ions. At this wavelength, the optical density of the $\text{Ce}^{3+}:\text{YLF}$ sample is 1.1 along the 13-mm path length. A 35-cm focal-length quartz lens was used to focus the 308-nm beam to a 2-mm-diameter spot. Three different low-power cw lasers, He-Cd at 325 and 442 nm and He-Ne at 633 nm, were used to generate the σ -polarized probe beams, which interacted with the pumped volume of the sample. A coaxial pump-probe geometry was used, with the probe beam being focused to a 0.3-mm-diameter spot that was concentric with the pumped volume. When the 325-nm probe laser was used, two quartz prisms and a 0.5-m spectrometer were required to separate the transmitted probe beam from the intense cerium fluorescence. For the transient absorption measurements, a LeCroy 3500 transient digitizer with a signal averager was used to record the transmitted probe beam intensity, which was detected with a photomultiplier tube. The UV-induced stable absorption spectra were measured with a Perkin-Elmer Lambda 4 spectrophotometer both be-

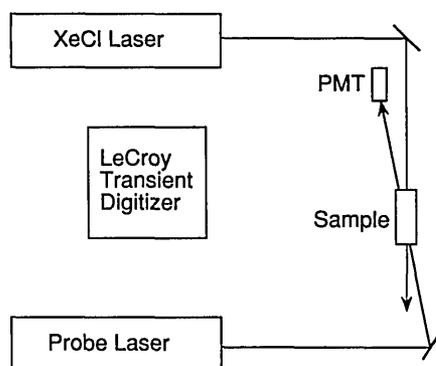


Fig. 1. Experimental arrangement used for the transient absorption measurements. The crossing angle between the pump and probe beams is approximately 4 deg. PMT, photomultiplier tube.

fore and after the exposure of the sample to the 308-nm laser.

EXPERIMENTAL RESULTS

Transient Absorption

A transient reduction in the transmitted intensity of a 633-nm probe beam following a 249-nm pump pulse in Ce^{3+} :YLF was reported by Ehrlich *et al.*⁷ This reduction was tentatively ascribed to thermal lensing due to the large quantum defect for the 249-nm excitation. By exciting the cerium ions at 308 nm, the thermal lensing effects should be significantly reduced owing to the smaller difference between the energies of absorbed and emitted photons and the near-unity quantum efficiency of fluorescence. However, our measurements indicate that even with this reduced thermal loading for a 308-nm pump pulse, a 10-mJ-per-pulse excitation resulted in a nearly 40% reduction in the transmitted intensity of a σ -polarized 633-nm probe beam. By using a set of different-diameter apertures at varying distances between the sample and the probe-beam detector, we have been able to rule out thermal lensing as the cause of the reduction in the detected probe-beam intensity. Since this loss was independent of the diameter and position of any aperture, there must be an absorptive process in the pumped volume of the sample.

This absorptive loss is directly related to the excitation of the cerium ions. Using an undoped YLF sample and similar 308-nm pulsed excitation, we observed no transient absorption following the pump pulse. Thus the transient centers responsible for the absorptive loss are not created in the absence of the cerium ions. Pumping the cerium-doped sample at 337 nm with a N_2 laser or at 355 nm with the third harmonic of a Nd:YAG laser, which are off resonance with the cerium $4f \rightarrow 5d$ transition, again does not lead to any observable transient absorption. From these two observations we conclude that the creation of the color centers responsible for this transient absorption is related to the optical excitation of the cerium ions and not to any direct UV-induced color-center formation in the YLF lattice.

It is also possible to rule out excited-state absorption originating on the lowest $5d$ state as the mechanism responsible for the transient losses. Such a process would be characterized by the 40-nsec radiative lifetime of the lowest $5d$ state of

the cerium ions. Instead we have observed much longer recovery times for the transmitted 633-nm probe beam.^{9,10} Although the relaxation is nonexponential, it is characterized by three distinguishable time constants of 50 nsec, 200 μ sec, and 50 msec at room temperature. In addition, the value of each time constant is strongly temperature dependent, whereas the 40-nsec cerium lifetime is observed to be approximately temperature independent between 77 and 400 K. Thus the transient absorption is thought to originate from color centers whose decay is thermally activated and not from the bound excited states of the isolated trivalent cerium ion.

These transient color centers absorb not only at 633 nm but at shorter wavelengths as well. Most color centers are strongly coupled to the lattice and show broad absorption bands, and such seems to be the case here as well. We have completed detailed measurements of the creation and relaxation characteristics of these absorption bands, using laser probe wavelengths at 325 nm and 442 nm as well as at 633 nm.^{9,10} The absorption band that gives rise to the 50-nsec decay component at 633 nm becomes much weaker in strength at the shorter probe wavelengths. At the He-Cd laser wavelength of 325 nm, this 50-nsec absorption component is no longer observable. On the other hand, the absorption bands with the longer lifetimes grow in strength as the probe wavelength moves to the blue. At 325 nm, a new absorption band is observed whose decay time at room temperature is approximately 30 sec.

In order to determine how these transient centers are formed, we have measured the strength of the transient absorption of a σ -polarized probe beam at 633 nm as a function of the 308-nm laser intensity. As illustrated in Fig. 2, two markedly different intensity relationships are observed. The strength of the 50-nsec component of the transient absorption increases quadratically with laser fluence. In contrast, the longer time components of the transient absorption at 633 nm show a near-linear behavior. The quadratic dependence of the 50-nsec component is characteristic of a two-photon process. As mentioned above, the transient center responsible for the 50-nsec component is

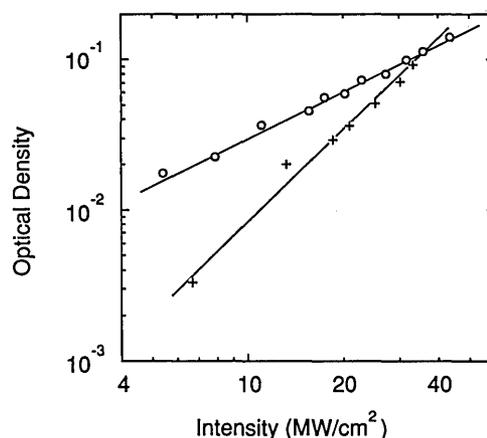


Fig. 2. Transient absorption at 633 nm as a function of the XeCl laser intensity. The circles represent the absorption at 50 μ sec after the laser pulse and are shown along with a straight line whose slope is 1. The absorption due to just the shortest-lifetime (less than 50-nsec) decay component is indicated with the crosses along with a straight line of slope 2.

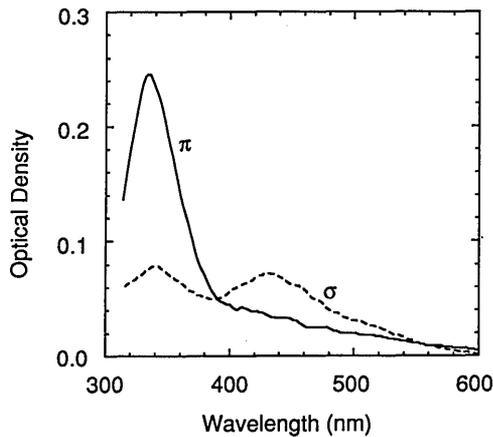


Fig. 3. Polarized absorption spectra of the Ce:YLF sample following irradiation at 27 MW/cm² for approximately 2000 laser pulses at room temperature.

not absorptive in the near UV and thus cannot be photo-bleached by the pump laser. On the other hand, the longer-lived transient centers, which show the linear intensity dependence, absorb strongly at the 308-nm laser wavelength and thus are susceptible to photobleaching by the 308-nm laser pulse. Significant photobleaching of these centers can compete with the creation mechanism and mask the expected quadratic intensity relationship.⁶

We have been unable to detect any afterglow phosphorescence that might accompany the decay of the centers that are responsible for the transient absorption. The subsequent charge-trapping process that returns the electron to the cerium ion does not apparently leave the Ce³⁺ ion with an excited 5*d* electron.

Stable Absorption Bands

Along with the transient absorptions, we have also observed temporally stable absorption bands created by the 308-nm excitation of the cerium ions. The lifetime of the stable centers is greater than a few days at room temperature. The absorption spectra due to these stable centers in Ce³⁺:YLF following the XeCl laser irradiation is displayed in Fig. 3. The π -polarized absorption band peaks near 340 nm and has an appreciable optical density at the 325-nm peak of the cerium emission band. The σ -polarized absorption spectrum shows a peak at 430 nm as well as at 340 nm, and this band also has some overlap with the cerium emission band. The polarized absorption bands of these UV-created color centers have also been seen in YLF following electron¹¹ or gamma-ray¹² irradiation and have been identified as F centers. Based on the similarity of the absorption spectra, we have concluded that the stable centers in Ce³⁺:YLF created by the near-UV pumping of the Ce³⁺ ions are also due to F centers.

Although these centers seem to be thermally stable at room temperature, it is possible to bleach them optically by exposing the crystal to the 266-nm light from a Nd:YAG laser. The photons at this wavelength are not absorbed by the cerium ions, but the color centers absorb and presumably photoionize them. However, exposure to the 355-nm light generated by the third harmonic of a Nd:YAG laser does not bleach out these centers, although they also absorb

at this wavelength. This observation would suggest that there is a photoionization threshold for the bleaching of these centers and that the color centers have bound excited states. It is also possible to bleach these color centers by illuminating the color centers at 308 nm, which is the same wavelength responsible for their creation. Following an exposure to a high-intensity 308-nm beam, the number of centers can then be reduced by continued exposure to a lower-intensity 308-nm beam. This photobleaching is more efficient at 77 K, where the Ce³⁺ absorption at 308 nm is significantly less than at room temperature because of the narrowing of the 4*f* → 5*d* absorption band.

The optical density at 325 nm due to these stable centers is plotted as a function of the accumulated number of 308-nm laser pulses for varying laser intensities in Fig. 4. The absorption is seen to grow with increasing exposure to the excimer laser and then to saturate at a steady-state value.

The rate of growth of the optical density in Fig. 4 is also intensity dependent. The initial growth rate as a function of laser intensity is displayed in Fig. 5. The data show a quadratic relationship, indicating that a two-photon process is responsible for the initial formation of these centers. The

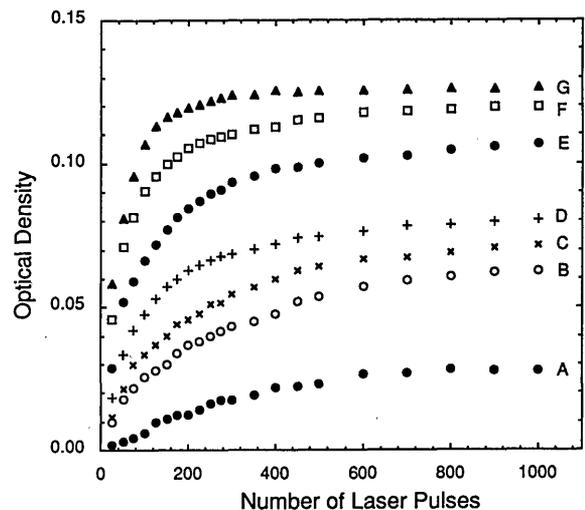


Fig. 4. Growth of the stable absorption at 325 nm. The XeCl laser intensity is A, 4.3 MW/cm²; B, 9.9 MW/cm²; C, 13 MW/cm²; D, 18 MW/cm²; E, 25 MW/cm²; F, 33 MW/cm²; and G, 44 MW/cm².

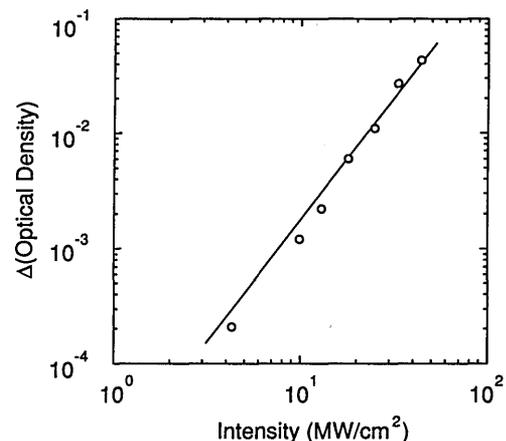


Fig. 5. Initial growth of the stable color-center formation.

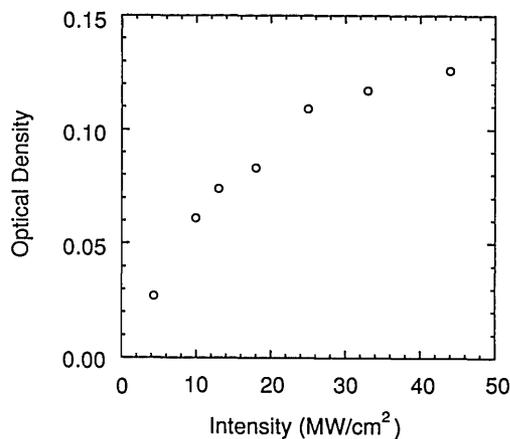


Fig. 6. Steady-state value of the absorption at 325 nm as a function of the XeCl laser intensity.

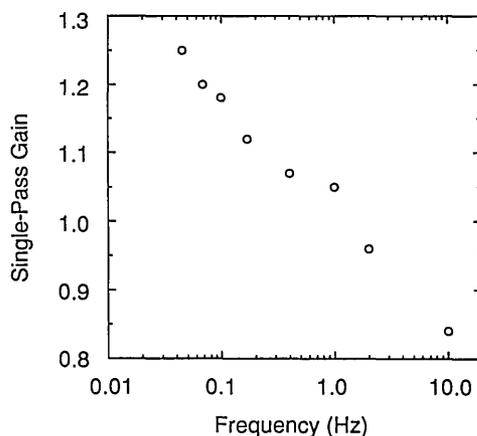


Fig. 7. Single-pass gain at 325 nm as a function of the repetition rate of the 308-nm pump beam.

steady-state values of the optical density of these stable centers saturate with increasing laser intensity, as illustrated in Fig. 6, indicating an upper bound for the number of deep trapping sites.

Single-Pass Gain Measurements

The transient and stable color centers play an important role in determining the Ce³⁺:YLF laser performance characteristics since both types of center have absorption bands that overlap the cerium emission spectrum. The influence of the longer-lived transient centers can be seen from our measurement of the single-pass gain as a function of the 308-nm pump repetition rate, which is displayed in Fig. 7. The baseline value of 1.0 is the transmitted 325-nm σ -polarized probe-beam intensity without 308-nm pumping. If the pulse repetition rate of the excimer laser is greater than approximately 0.5 Hz, the quasi-steady-state density of transient color centers is large enough to result in a net loss of the transmitted 325-nm probe beam. As the repetition rate is lowered, there is more and more time for the transient color centers to relax before the arrival of the next pump pulse, and thus the gain from the excited cerium ions becomes larger than the losses from the color centers. Thus the gain measured after any particular pump pulse is re-

duced by the loss due to steady-state density of the more stable centers, the loss due to the density of transient centers created by that pump pulse, and the density of transient centers that remain from all the previous pump pulses. Only the last mechanism is dependent on the pulse repetition rate. We estimate from the stimulated-emission cross section⁷ at 325 nm that the single-pass gain in the absence of these color-center losses would be approximately 1.7 for the 5 mJ/7 mm² excitation pulse.

DISCUSSION

The quadratic dependence of the initial growth of the stable centers and the strength of the shortest-time-component transient absorption on the laser intensity indicate that two 308-nm photons are required to create both types of color center. With a band gap in YLF corresponding to approximately 120 nm, the fact that two photons at 308 nm can photoionize the Ce³⁺ ion suggests that there is a significant energy gap between the top of the valence band and the 4*f* level of the Ce³⁺ ion. Since the centers are formed only at wavelengths resonant with the 4*f* → 5*d* absorption of the Ce³⁺ ions, this two-photon process involves an excited-state absorption on the Ce³⁺ ions. For an excited-state absorption that reaches the conduction band states of the YLF lattice, the electron will have sufficient mobility to find and become trapped at alternate lattice sites. Different charge-trapping sites will result in different binding energies and energy-level structures for the color center. In addition to creating the color centers, the 308-nm laser pulse can also annihilate them by a photobleaching process. Thus the description for the growth, intensity dependence, and saturation characteristics of the color-center population must incorporate both a quadratic creation term and a simultaneous photobleaching term.

The kinetics of the number of transient color centers n of the i th type of electron trapping site can be described by a rate equation of the form⁶

$$\frac{dn}{dt} = Nn_a A I^2 - nBI, \quad (1)$$

where N is the density of Ce³⁺ ions, n_a is the density of available charge-trapping sites, and I is the laser intensity. The value of the creation coefficient A will depend on the product of the absorption coefficients from the 4*f* state and from the 5*d* state and the electron capture probability at the electron acceptor site. The linear photobleaching coefficient B involves the absorption and photoionization of the color centers at 308 nm. Since the density of transient centers does not seem to saturate with increasing laser intensity, we have assumed that $n \ll n_a$ and thus have treated n_a as a constant. Equation (1) can be integrated over the j th square laser pulse¹³ of duration Δt to give

$$n(j^+) = n(j^-) \exp(-BI\Delta t) + \frac{Nn_a A I}{B} [1 - \exp(-BI\Delta t)], \quad (2)$$

where $n(j^+)$ is the density of transient centers after the j th laser pulse and $n(j^-)$ is the density immediately before. If the lifetime of the particular color center is τ and the time interval between laser pulses is T , then

$$n(j^-) = n(j^+ - 1)\exp(-T/\tau). \quad (3)$$

The expression for $n(j^+)$ can be written in terms of $n(1^-)$, the number of color centers before the first pulse, which is assumed to be zero. The resulting recursion relation is a geometric series that can be easily summed to give

$$n(j^+) = \frac{Nn_a AI[1 - \exp(-BI\Delta t)][1 - \exp(-BIj\Delta t - jT/\tau)]}{B[1 - \exp(-BI\Delta t - T/\tau)]}. \quad (4)$$

For $T \gg \tau$, Eq. (4) reduces to the expression in Eq. (2) with $n(j^-) = 0$ and is thus independent of the number of accumulated laser pulses j .

The steady-state value of the number of transient centers can be obtained by taking the limit of Eq. (4) as $j \rightarrow \infty$,

$$n(\infty) = \frac{Nn_a AI[1 - \exp(-BI\Delta t)]}{B[1 - \exp(-BI\Delta t - T/\tau)]}. \quad (5)$$

In the absence of any significant photobleaching by the 308-nm laser pulse, $BI\Delta t \ll 1$ and Eq. (5) becomes

$$n(\infty) \approx \frac{Nn_a AI^2 \Delta t}{1 - \exp(-T/\tau)}, \quad (6)$$

which is quadratic in the laser intensity. If there is significant photobleaching of the transient color centers such that $BI\Delta t \gg 1$, then Eq. (5) reduces to a result that is linear in the laser intensity,

$$n(\infty) \approx \frac{Nn_a AI}{B}. \quad (7)$$

Although the creation rate depends on the square of the laser intensity, the competition between the quadratic creation and linear annihilation terms can result in a linear relationship of the transient center density to the laser intensity. The data presented in Fig. 2 for the transient absorption show both the linear and the quadratic dependence. Measurement of the rapidly decaying 50-nsec absorption is quadratic in the laser intensity, as predicted by approximation (6). This is expected since the color center responsible for this decay component is not absorptive in the near UV and hence is not photobleached by the 308-nm light. For the longer-lived transient centers that are absorptive at 308 nm, approximation (7) predicts the observed linear dependence of the color-center absorption on the laser intensity.

Equation (1) can also be used to describe the kinetics of the more stable color-center population. For these centers, the steady-state absorption saturates with increasing laser intensity, as displayed in Fig. 6. This saturation is due to the finite number of available charge-trapping sites responsible for the formation of these centers, which we have assumed to be fluorine vacancies. Accordingly, the expression for the density of available charge-trapping sites is

$$n_a = V_f - n_s, \quad (8)$$

where V_f is the initial density of fluorine vacancies and n_s is the density of the stable color centers. The rate equation for these stable centers is then

$$\frac{dn_s}{dt} = N(V_f - n_s)AI^2 - n_s BI. \quad (9)$$

This rate equation can be solved in a manner similar to that presented for the transient centers. Since $\tau \gg T$ for these stable centers, we have assumed that $n_s(j^-) = n_s(j^+ - 1)$. The solution to Eq. (9) is then

$$n_s(j^+) = \frac{NV_f AI}{NAI + B} \{1 - \exp[-(NAI^2 + BI)j\Delta t]\}. \quad (10)$$

At the initial stage of color-center formation, for which the argument of the exponential function in Eq. (10) is sufficiently small, the density of centers can be approximated by

$$n_s(j^+) \approx NV_f AI^2 j\Delta t, \quad (11)$$

which is quadratic in the laser intensity. This quadratic relationship is in agreement with the intensity dependence of the initial growth-rate measurements of the stable absorption presented in Fig. 5.

The steady-state solution to Eq. (9) is

$$n_s(\infty) \approx V_f(1 + B/NAI)^{-1}, \quad (12)$$

which is linear in the laser intensity I for small values of I and then tends to V_f with increasing laser intensity. The measured optical density of the stable color-center absorption as a function of laser intensity in Fig. 5 shows that this saturation behavior is due to the finite number of the deep charge-trapping sites. The number of these fluorine vacancies is estimated¹⁰ to be of the order of 10^{15} cm^{-3} .

The characteristics of the color-center formation described here will have an important effect on the the lasing properties of flash-lamp-pumped $\text{Ce}^{3+}:\text{YLF}$ and implications for other materials as well. If the spectral content of the flash lamp is broader than or is not matched to the Ce^{3+} absorption bands, then the photons outside the Ce^{3+} bands will not contribute to the excitation of the cerium ions but can promote excited-state absorption processes that may photoionize the cerium ions and lead to color-center formation. These spectrally nonmatched photons thus do not just lower the wall-plug efficiency of the laser but serve to increase threshold and saturation problems as well. Such considerations may be responsible for the inability to achieve a net stimulated gain in flash-lamp-pumped $\text{Ce}^{3+}:\text{YLF}$ crystals.¹⁴

CONCLUSION

We have demonstrated that the optical pumping of $\text{Ce}^{3+}:\text{YLF}$ in the near UV leads to the formation of both transient and stable color centers. These centers clearly play an important role in determining the device characteristics of a $\text{Ce}^{3+}:\text{YLF}$ laser. The relaxation of the transient centers governs the repetition rate limitation. Both stable and transient centers mediate the gain saturation and slope efficiency as well as the lasing threshold. The photoionization of the Ce^{3+} ion requires two photons at 308 nm to reach the conduction band of the host crystal. If the color center produced following this photoionization is also absorptive at the 308-nm pump wavelength, then the competition between the photobleaching process and the creation mechanism can mask the simple quadratic intensity dependence.

If the more permanent color centers involve fluorine vacancies, it may be possible to reduce the absorptive losses by

improved crystal growth and treatment. For transient centers related to the presence of the cerium ions themselves,⁶ there is much less of an opportunity to reduce the deleterious role of these centers.

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