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Broadband, intensity dependent absorption in tetravalent chromium-doped crystals

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Cr^{4+} -doped Y_2SiO_5 and $\text{Ca}_2\text{Al}_2\text{SiO}_7$ have been investigated as broadband, solid state saturable absorbers. Saturation properties in the near infrared as well as a demonstration of tunable, passive Q switching of a flash-lamp pumped Cr:LiSAF laser are discussed.

Despite the broad absorption bands of tetravalent chromium in crystals, previous studies of the dependence of the absorption on the intensity of the radiation field have been limited to the Nd:YAG laser wavelength.¹⁻⁶ The widening use of tunable lasers in the 800–1200 nm region (i.e., Ti:sapphire, Alexandrite, Cr:LiSAF, and Cr:emerald), warrants investigation of intensity dependent absorption throughout the Cr^{4+} bands. In this work we use a Cr^{3+} :LiSAF laser having a lasing spectrum within the absorption bands of Cr^{4+} : Y_2SiO_5 (Cr:YSO) and Cr^{4+} : $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (Cr:CAS) to study broadband intensity dependent absorption. We also demonstrate the generation of tunable light with pulse duration of a few tens of nanoseconds by using a Cr^{4+} -based, saturable absorber Q switch with the Cr^{3+} :LiSAF laser.

Many applications of solid state lasers require reliable, simple, and compact laser systems. When pulses with nanosecond duration are required, the method chosen to switch the Q of the laser cavity greatly influences the size and complexity of the system. Electro-optical devices require high external voltage supplies and polarizing optics while acousto-optical switches need high frequency electronic drivers. The passive Q switch is both compact and simple, consisting only of a bleachable absorber introduced in the optical resonant cavity. Although several types of liquid organic dye solutions have been used for a long time as passive Q switches⁷ these materials suffer from poor chemical stability and inadequate thermal properties. To overcome these problems flow systems have been added to the liquid dye solution but at the expense of simplicity and compactness. On the other hand solid state saturable absorbers may be stable and, depending on the host material, may have acceptable thermal properties. As a result, research efforts have been directed toward their development. Different approaches were reported including the use of transparent plastic materials impregnated with organic dyes,⁸ F_2^- color centers in lithium fluoride,⁹⁻¹¹ and more recently, tetravalent chromium in such crystalline hosts¹⁻⁶ as YAG, Mg_2SiO_4 , GSGG, YSGG, GSAG, and GIGG. The dye impregnated plastics suffer the same drawbacks as do liquid dye solutions. Color centers Q switches suffer from fading of the optical centers^{9,10} and so have limited applicability. On the other hand Cr^{4+} -doped crys-

tals, recently reported as potentially useful passive Q switches for the Nd:YAG laser at 1.06 μm , carry the advantages of a much higher damage threshold as compared to plastic absorbers, high chemical and optical stability, and good thermal properties.

The crystals we chose for this study contain only Cr^{4+} as the hosts structures contain no octahedral sites capable of incorporating Cr^{3+} . Further, these crystals require no codoping for the sake of charge compensation as the Cr^{4+} ions substitute for Si^{4+} in tetrahedrally coordinated sites. These properties contrast with the Cr-doped materials studied so far,¹⁻⁶ in which either charge compensation is required for both Cr^{3+} and Cr^{4+} are present. The samples used in this work were Czochralski grown single crystals prepared at the University of Central Florida. Detailed spectroscopic studies of Cr:YSO and Cr:CAS have been reported elsewhere.¹²⁻¹⁴

Low intensity polarized absorption spectra of both the Cr:YSO and the Cr:CAS samples used in this work are shown in Fig. 1. In both crystals, the absorption bands feature a very strong dependence on the polarization of the incoming radiation relative to the crystal orientation. Absorption through the Cr:LiSAF-laser tuning range (~ 770 –

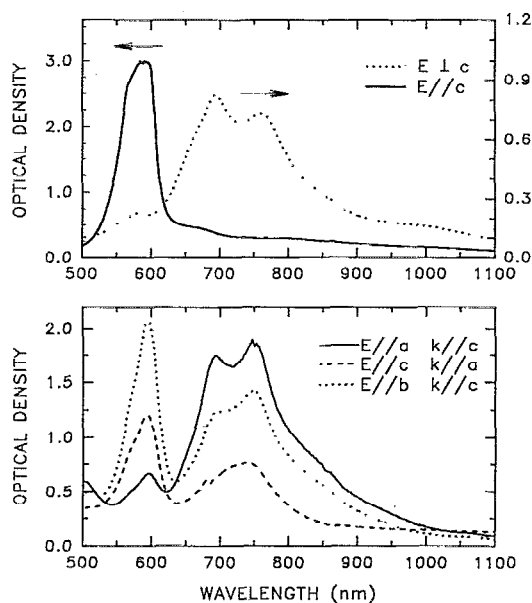


FIG. 1. Polarized absorption spectra of Cr^{4+} : $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (top) and Cr^{4+} : Y_2SiO_5 (bottom).

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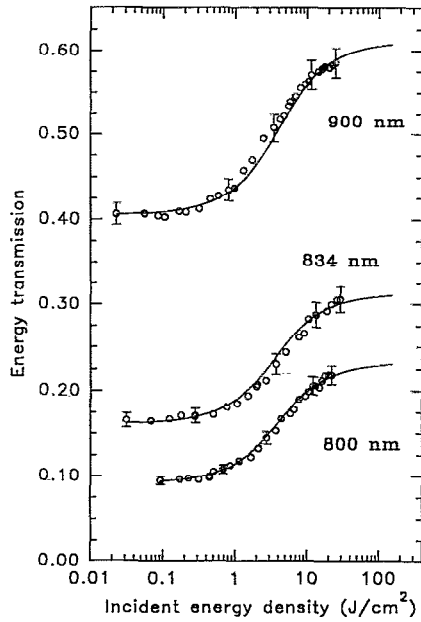


FIG. 2. Energy transmission for $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ vs incident energy density (light polarization was parallel to the a axis and propagation direction was along the c axis). The error bars shown represent estimated experimental errors in determining transmission. At 800 and 900 nm the pulse duration was 48 ns and at 834 nm it was 40 ns.

970 nm) is attributed to the ${}^3\text{A}_2\text{-}{}^3\text{T}_2$ transition of tetravalent chromium in the tetrahedral site.

Saturable absorption measurements were performed by using an electro-optically Q -switched $\text{Cr}:\text{LiSAF}$ laser as the source. This pump beam was focused with a 15 cm lens into a 125 μm pinhole capable of handling high energy densities. Neutral densities filters were used to change the intensity of the incoming beam and also to limit the energy incident at the detector. By placing the sample to be studied immediately after the pinhole we could estimate the beam area as about the area of a pinhole. The intensity dependent absorptions of a 12.3 mm long, antireflection coated $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ sample at three wavelengths are presented in Fig. 2.

A four level scheme with excited state absorption as shown in Fig. 3 was suggested by Eilers *et al.*¹⁵ to explain

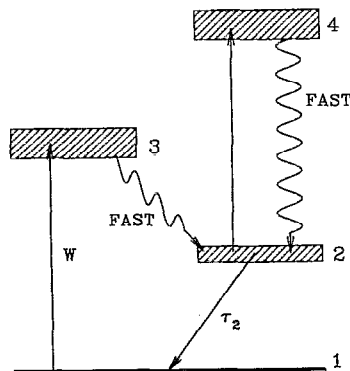


FIG. 3. Energy level scheme for a four level saturable absorber with excited state absorption at the excitation wavelength.

TABLE I. Absorption cross sections for $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ extracted from experimental data in Fig. 2.

Wavelength (nm)	σ_{gs} (cm^2)	σ_{es} (cm^2)
900	$(1.5 \pm 0.2) \times 10^{-19}$	$(0.80 \pm 0.2) \times 10^{-19}$
834	$(3.0 \pm 0.3) \times 10^{-19}$	$(1.9 \times 0.3) \times 10^{-19}$
800	$(3.9 \pm 0.4) \times 10^{-19}$	$(2.4 \pm 0.4) \times 10^{-19}$

saturable absorption in $\text{Cr}:\text{Ca}:\text{Y}_3\text{Al}_5\text{O}_{12}$ at 1.06 μm . We can apply this model to the systems studied here but as discussed below, other models may explain the properties we observe. The rate equations for the system in Fig. 3 are given by

$$\frac{\partial n_1}{\partial t} = -Wn_1 + n_2/\tau_2, \quad (1a)$$

$$\frac{\partial n_2}{\partial t} = Wn_1 - n_2/\tau_2, \quad (1b)$$

$$\frac{\partial I_\nu}{\partial z} = -(an_1 + \beta n_2)I_\nu, \quad (1c)$$

where τ_2 is the effective lifetime of level 2, n_1 and n_2 are the fraction of ions in levels 1 and 2, respectively, I_ν is the laser power density at frequency ν , and t and z denote the temporal and longitudinal coordinates. $W = \sigma_{gs}I_\nu/h\nu$, where h is the Planck's constant and σ_{gs} the ground state absorption cross section, $\alpha = \sigma_{gs}N$ and $\beta = \sigma_{es}N$, where N is the total number of ions/ cm^3 and σ_{es} is the excited state absorption cross section. For this model, the population of levels 3 and 4 are neglected as the nonradiative rates for those levels are considered much faster than the other rates involved, therefore $n_1 + n_2 \cong 1$.

The absorber energy transmission T_E is given by

$$T_E = \frac{\int I_{out} dt}{\int I_\nu dt} = \frac{\int T(t)I_\nu(t) dt}{\int I_\nu(t) dt}, \quad (2)$$

where I_{out} is the output power density and $T(t) = I_{out}(t)/I_\nu(t)$.

For the present case in which we consider Gaussian excitation of short duration as compared to the absorber lifetime (0.59 μs for Cr^{4+} in YSO), Eqs. (1a)–(1c) can only be solved by numerical methods. By fixing the number density of absorbing ions we find the ground state absorption cross section from the experimental data. Then the excited state absorption cross section is varied at each wavelength to obtain the best fit. Our analyses showed that an absorber concentration of $0.5 \times 10^{19} \text{ cm}^{-3}$ resulted in the fitted curves shown for each wavelength in Fig. 2. Since, during crystal growth, the concentration of Cr in the melt was 2% this number density implies that the distribution coefficient of Cr^{4+} in YSO is 0.027. The values of ground and excited state absorption used to obtain the curves shown in Fig. 2 are given in Table I.

The high energy density data in Fig. 2 were taken close to the damage threshold of our sample. Since the abscissa uses a log scale the transmission at this limit may appear to have not yet reached a constant value. However the experimental uncertainty indicated by the error bars shows that saturation was complete. This claim is supported by the extension of the theoretical model to still higher intensities where the fully saturated transmission is seen in Fig. 2 to

be very nearly equal to that achieved experimentally. The fact that the absorption at high intensity does not go to zero may be caused by nonsaturating excited state absorption at the pump wavelength as in the model of Ref. 15. On the other hand, incomplete bleaching can be caused by rapid relaxation of an inhomogeneously broadened absorption feature and/or the presence of different dopant sites. Relaxation via energy transfer processes between interacting dopants in different sites could contribute to the incomplete bleaching of the absorption. Therefore, we present the model involving excited state absorption as one possible explanation for the results we observe. Experiments are in progress to determine if the saturation is homogeneous or inhomogeneous and to measure the dynamics of the recovery of the absorption. Such data will help give confidence in the model chosen to explain saturable absorption in Cr^{4+} .

The data in Fig. 2 refer to light polarization parallel to the a axis and propagation along the c axis. For polarization in the other directions, the data follow similar curves. In particular, for light polarization parallel to the b axis and propagation as before (i.e., same axis and sample length for the sake of comparison) the initial transmission for 800, 834, and 900 nm are, respectively, 0.15, 0.23, and 0.49 with transmittance at bleaching equal to 0.25, 0.35, and 0.65. This dependence upon polarization in anisotropic solid state absorbers could be advantageous in some applications, as it makes possible Q switching a polarized laser at different pump levels by rotating the absorber plate to obtain the necessary initial absorptivity.

Measurements on a 2.93 mm thick Cr:CAS sample showed intensity dependent absorption with the radiation electric field vector \mathbf{E} perpendicular to the c axis while not showing significant change in the absorption for \mathbf{E} parallel to the c axis. Further, saturation of the absorption in this material was observed in a narrower spectral range than that observed in Cr:YSO. For \mathbf{E} perpendicular to the c axis, low intensity transmittance at 800 nm was 0.36 and transmittance at bleaching was 0.46. No appreciable change in absorption was observed at 900 nm.

To evaluate the materials performance in a Q -switched laser we used a flash lamp pumped Cr:LiSAF laser and placed the antireflection coated Cr:YSO sample inside the resonator to serve as the saturable absorber. Without any intracavity wavelength-selective element we observed pulses at 880 nm (15 nm wide FWHM) which were as short as 35 ns (FWHM) and contained 4.3 mJ output with a flash lamp input of 80 J. The 42 cm long resonant cavity consisted of a flat total reflector and an 82% reflectivity, flat output coupler. Introduction of two prisms in a 56 cm long cavity allowed us to tune the Q -switch pulses from 807 up to 927 nm without changing the absorber. Single pulse operation was possible by adjusting the pump level to account for the change of the optical density of the absorber. Due to the combined effects of the lasing medium gain curve with the spectral dependence of the loss introduced by the absorber the duration of the Q -switch pulses was not constant throughout the tuning range. The pulse duration changed from 37 ns at 807 nm up to about 80 ns

at 927 nm. The extracted energy with an 82% reflectivity output coupler ranged from 0.25 mJ at the short wavelength limit up to 4.2 mJ at the long wavelength limit. No attempt was made to optimize the resonator and Q -switch parameters at any specific wavelength.

In conclusion we have demonstrated broadband, intensity dependent absorption in Cr^{4+} -doped crystalline saturable absorbers with application to the generation of tunable Q -switched pulses in Cr:LiSAF lasers. Pulses as short as 35 ns were generated with energy of 4.3 mJ in an untuned cavity resonator. Energy extraction was found to be limited by the presence of residual absorption, possibly caused by excited state absorption at the wavelength of interest.

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