Co

Divalent Uranium and Cobalt Saturable Absorber Q-Switches at 1.5 μm

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Abstract

Saturable absorber Q-switching of the 1.5 μm Er:glass laser has been obtained using slowly-relaxing divalent uranium ions in Ca, Sr, and Ba:F_2, and fast-relaxing divalent cobalt ions in $Y_3Sc_2Ga_3O_{12}$ (YSGG) and $Y_3Al_5O_{12}$ (YAG). Pulsewidths as short as 20 nanoseconds have been demonstrated without optical damage in both cases. Spectroscopic measurements, as well as Q-switched laser results are discussed for all of these materials.

Spectroscopic Measurements - U^{2+}

We previously reported on passive Q-switching of the Er:glass laser using U^{4+}:SrF_2, [1]. Our identification of the tetravalent uranium ion as the responsible agent for the saturable absorption at 1.54 μm was in accordance with several early publications on the spectroscopy of uranium-doped di-fluorides, [2.3]. According to Hargreaves, however, this should have been reported as U^{2+}:SrF_2, [4,5,6].

Fig. 1 compares the room temperature absorption spectra measurements for two U:CaF_2 rods used in our Q-switch experiments, with the U^{2+}:CaF_2 absorption spectrum from ref. [6]. Although the latter is at low temperature, several peaks are seen to match those of our crystals (including the one near 1.5 μm). The presence of U^{3+} was also detected, [7].

The broad absorption band of U^{2+} near 1.5 μm, in the di-fluoride crystals, peaks close to the Er:glass laser wavelength (see Fig. 2). Luminescence of our uranium-doped crystals, when excited at 1.53 μm using a free-running Er:glass laser, peaked at about 2.5 - 2.6 μm. This fluorescence corresponds to the $^5I_5$ to $^5I_4$ transition of U^{2+} (see Fig. 3). The lifetime of the metastable state $^5I_5$ was determined by pumping at 1.543 μm using a Raman-shifted Nd:YAG laser with a 14 ns pulse. The fluorescence was measured using an InAs detector with a germanium crystal filter to block the pump light. The observed fluorescence began immediately (< 50 ns) following the 1.543 μm excitation pulse and consisted of a single exponential component out to at least three e^{-1} lifetimes. The measured room temperature lifetimes are given in Table 1. The lifetimes measured for crystal temperatures from about 300 to 400 K are plotted in Fig. 4.

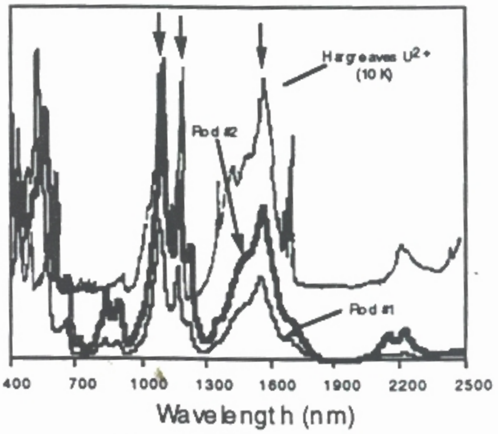

Figure 1. Comparison of U^{2+}:CaF_2 spectrum from ref. [6] to that measured for U:CaF_2 rods.

The U^{2+} absorption cross-sections were determined by bleaching the crystals using the same Raman-shifted Nd:YAG laser. Since the fluorescence lifetimes are long compared with the bleaching pulse duration, the Frantz-Nodvik equation, [8], was used to analyze the results. The cross-section σ was, for each of the di-fluoride crystals, determined from the saturation parameter ($F_{sat} \equiv h\nu/\sigma$) which best fit the experimental transmittance data. The measured absorption cross sections are given in Table 1.

Both the U:SrF_2 and U:CaF_2 crystals had damage thresholds greater than the maximum fluence used in the 1.543 μm saturation measurements (i.e., > 4 J/cm^2), however, the U:BaF_2 crystal damaged at only

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1.5 - 2 J/cm^2. The poorer surface and internal qualities of the U:BaF_2 crystal used in these experiments may have contributed to its reduced resistance to optical damage.

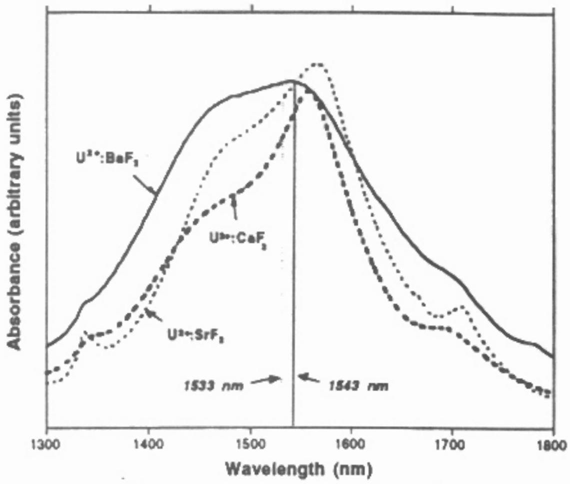


Figure 2. Room temperature absorption spectra of U^{2+}:CaF_2, U^{2+}:SrF_2, and U^{2+}:BaF_2 near 1.5 µm.

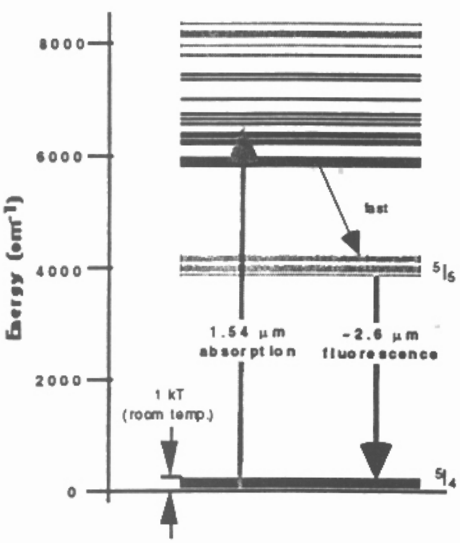


Figure 3. U^{2+}:CaF_2 energy levels, [5].

Table 1. Spectroscopic parameters of uranium-doped Q-switch materials.

Material	α_0 at 1533nm (cm^{-1})	300 K lifetime (µsec)	Cross-section (x 10^{-20}, cm^2)
U:BaF_2	0.58	43	5
U:CaF_2	1.26	5	7
U:SrF_2	1.05	25	7

Q-Switched Laser Results - U^{2+}

All three uranium-doped crystals (supplied by Optovac, Inc.) were evaluated as Q-switches in an Er:glass laser resonator. A Kigre 3 x 50 mm (QE-7S) Er:glass rod was used for the SrF_2 and BaF_2 Q-switches. The rod was flashlamp-pumped in a Kigre pump head. The U:CaF_2 Q-switch was used with a 4 x 76 mm (QE-7S) Kigre rod in an unoptimized pump cavity designed for a much larger rod. The resonator cavities consisted of two flat mirrors with physical lengths of 10 cm for the U:SrF_2 and U:BaF_2, and 15.5 cm for the U:CaF_2. The resonator output mirror reflectivity was 80% for the SrF_2 Q-switch, and 94% for CaF_2 and BaF_2. The Q-switching results are summarized in Table 2.

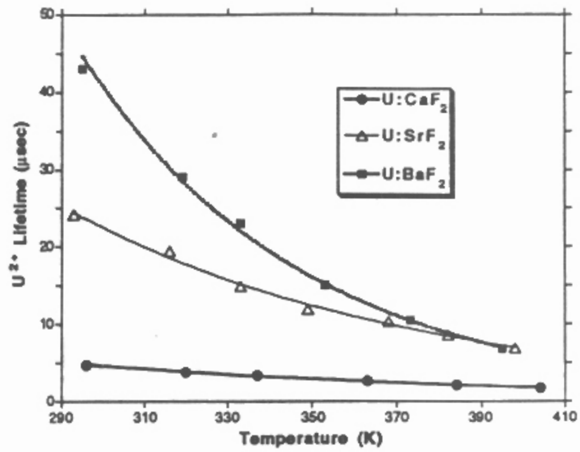


Figure 4. U^{2+}:CaF_2, SrF_2 and BaF_2 fluorescence lifetime as a function of temperature.

Table 2. Q-switch results.

Q-Switch Matl.	Thickness (mm)	Internal Transmit. (%)	Output energy (mJ)	Measured pulsewidth (ns)	Threshold (J)
U:BaF_2	1	94	1	306	16
U:SrF_2	1	90	11	45	15
U:CaF_2	4	60	3	21	86

All of the Q-switches were used uncoated in the laser experiments. Even better performance is anticipated for anti-reflective (AR) coated Q-switches. The CaF_2 and BaF_2 Q-switches were aligned for normal incidence, but the SrF_2 Q-switch was placed at Brewster's angle. The measured wavelength of the Q-switched laser, 1.533 µm, was very close to the peak fluorescence of Er:glass.

The shortest pulsewidth obtained without damage with the U:SrF_2 Q-switch was about 40 nsec FWHM. However, ≈20 ns pulses were produced with U:CaF_2 (Fig. 5) without any evidence of damage. Short Q-switch pulses were not attempted with U:BaF_2. It is uncertain at this point whether the damage problems observed with U:SrF_2 and U:BaF_2 are intrinsic with these materials.

We modeled the Q-switch operation using saturable absorber rate equations, [9]. Very good agreement with experiment was obtained using the spectroscopic parameters in Table 1.

Spectroscopic Measurements - Co^{2+}

Co^{2+} has the electronic structure [Ar core] $3d^7$. Broad absorption bands are a result of the strong interaction of the crystal field with the outermost 3d electron shell. Co^{2+}:YSGG and Co^{2+}:YAG crystals possess a very broad absorption feature near 1.5 µm (Fig. 6). The room temperature spectra for both crystals are very similar to that of Co^{2+}:GSGG, [10] (Fig. 7), except that the Co:YAG spectrum is shifted toward shorter wavelengths due to a higher crystal field strength. The first three excited energy levels for a $3d^7$ ion in tedrahedral symmetry are shown in Fig. 8, [10].

The Co:YSGG and Co:YAG crystals used in this work were grown along the <111> direction, using the standard Czochralski method. The cobalt concentration was 2% at. wt., and silicon was added to the melt for charge compensation.

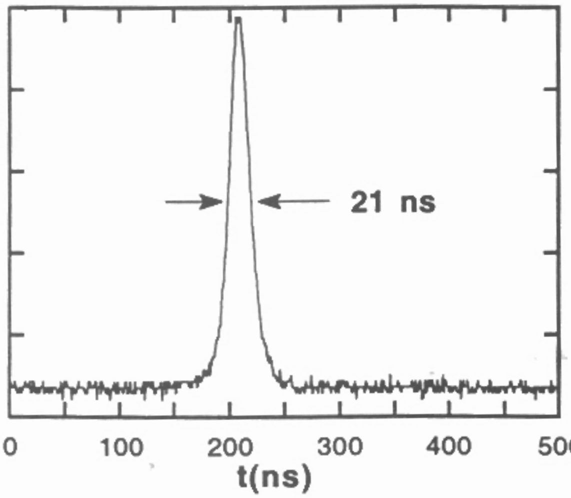


Figure 5. Experimental Er:glass pulse obtained with the U:CaF_2 Q-switch.

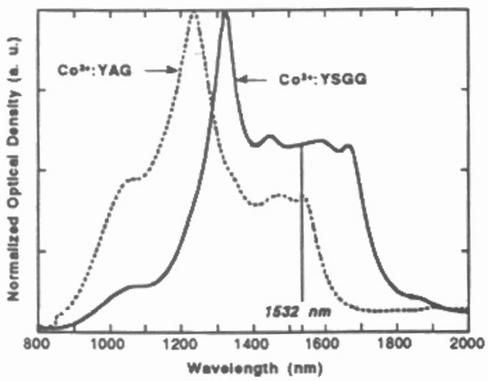


Figure 6. Measured room temperature Co^{2+}:YSGG and Co^{2+}:YAG absorption spectra near 1.5 µm.

An estimate of the absorption relaxation lifetime was made for Co:YSGG using a pump-probe method. The crystal was pumped with a 10 ns laser pulse at 1.543 µm. The pumped region was simultaneously probed with a 0.633 µm HeNe laser (Fig. 8). The 0.633 µm transmittance was monitored using a photomultiplier tube (PMT), with a Schott KG3 glass filter to block the pump light. Both the 1.543 µm pulse (measured with a fast InGaAs detector), and the PMT signal (negative-going) are shown in Fig. 9. The PMT signal was delayed electronically by about 40 ns.

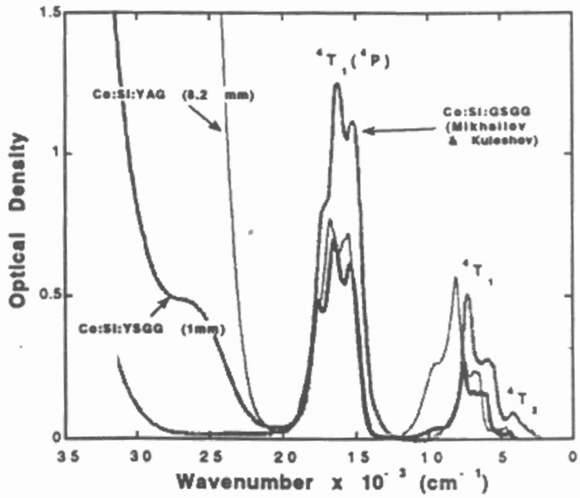


Figure 7. Comparison of Co:YSGG and Co:YAG absorption spectra to published Co:GSGG spectrum, [10].

The 1.543 µm pump, by removing electrons from the ground-state, alters the transmittance at 0.633 µm due to a reduction in the population difference between the $^4A_2$ and $^4T_1(^4P)$ states (Fig. 8). The decay time of the 0.633 µm transmittance disturbance, should therefore be equal to the relaxation lifetime of ions from the $^4T_1$ level. The fact that the 0.633 µm transmittance pulse (Fig. 9) has nearly the same shape and duration as the 1.54 µm pump pulse, indicates that the lifetime of the $^4T_1$ relaxation is fast compared to the duration of the bleaching pulse (<< 10 ns).

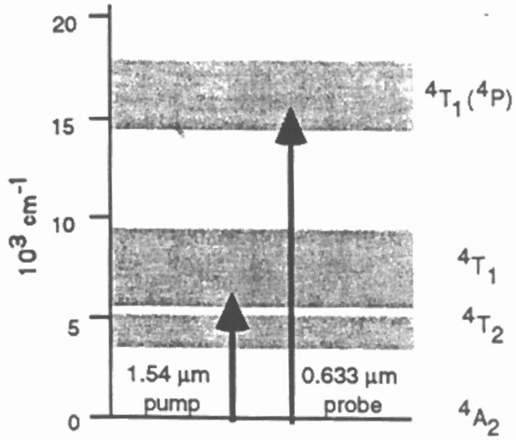


Figure 8. Energy levels for Co^{2+} in tetrahedral sites, [10].

The 1.543 µm absorption saturation was measured for both Co:YSGG and Co:YAG using the same experimental set-up as with the uranium-doped crystals, [1]. The 1.543 µm beam was in this case aligned along the <111> direction. For a fast-relaxing absorber, with negligible excited-state absorption, the saturated transmittance T is given by, [11],

$$\ln\left(\frac{T}{T_0}\right) = \left(\frac{I_0}{I_s}\right)(1 - T) \qquad (1)$$

where I_0 is the incident 1.543 μm intensity, I_s is the saturation intensity ($\equiv h\nu/\sigma\tau$), τ is the relaxation lifetime, and T_0 is the small-signal transmittance. The solid curves in Figs. 10 and 11 were obtained by adjusting the I_s parameter in (1) for the best fit of the experimental data. Average values of 180 MW/cm^2 (Co:YSGG), and 140 MW/cm^2 (Co:YAG) were measured for I_s (Table 3). With the Co:YAG sample, damage occurred at about 180 MW/cm^2, so only transmittance measurements below the damage threshold were considered in determining the saturation intensity (Fig. 11). Damage was not observed in the Co:YSGG crystal, even up to the maximum intensity tested (570 MW/cm^2).

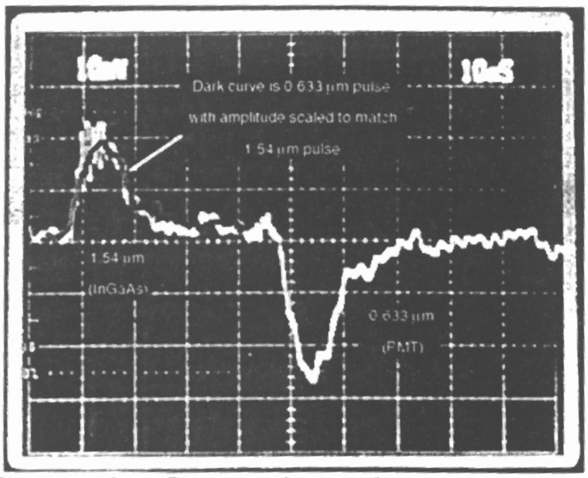


Figure 9. Pump-probe relaxation lifetime measurement for Co^{2+}:YSGG, (10 ns per division).

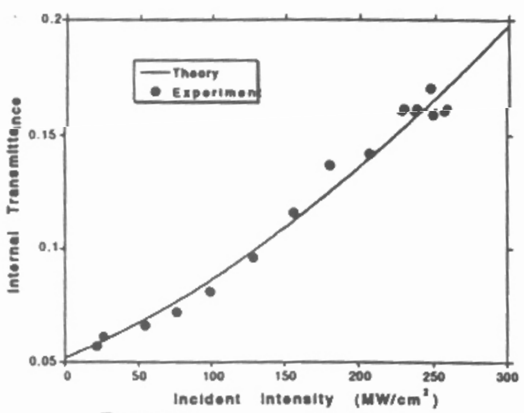


Figure 10. Transmittance saturation measurement for Co^{2+}:YSGG.

Q-Switched Laser Results - Co^{2+}

Q-switching of the Er:glass laser was achieved with both Co:YSGG and Co:YAG using intracavity focusing, [12]. The pulse (4mJ, 20ns) shown in Fig. 12 was obtained using a Co:YSGG Q-switch, with a focusing parameter (A_g/A_a) of approximately 17, where A_g and A_a are the beam cross-sectional areas in the Er:glass rod and Q-switch, respectively. The same Kigre Er:glass pump head was used as with the U:CaF$_2$ Q-switch. Threshold, with the Q-switch inserted in the resonator cavity, was approximately 40 J, and the threshold for the free-running laser (i.e. after removal of the Q-switch) was 27 J.

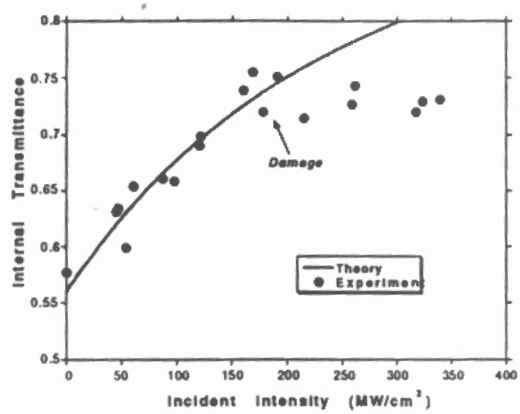


Figure 11. Transmittance saturation measurement for Co^{2+}:YAG.

Table 3. Measured 1.543 μm saturation intensities for Co^{2+}:YSGG, YAG.

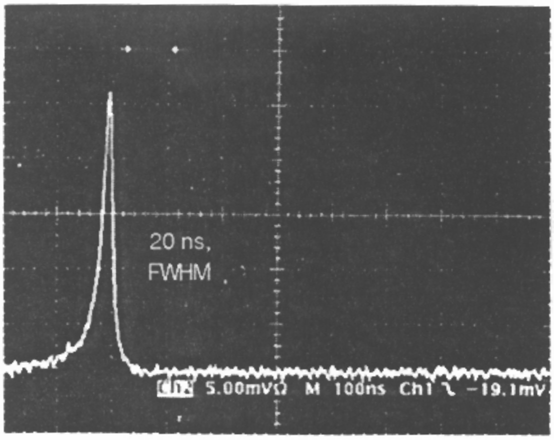
Crystal	Thick-ness (mm)	Small-signal internal transmittance at 1.543 μm	α_0 at 1.543 μm (cm^{-1})	1.543 μm saturation intensity (MW/cm^2)
Co:YSGG	8.92	0.054	3.27	180
Co:YAG	3.91	0.8	0.57	140

The 4 mJ Q-switched output energy was roughly 13% of the free-running output (i.e. with Q-switch removed) at the same flashlamp input energy. However, the Q-switch was used uncoated (at normal incidence), so the Fresnel surface reflection losses must be taken into account. For YSGG, the single-pass reflective loss is about 20%. Assuming that none of this loss appears in the output of the laser, the outcoupling efficiency (η_c) can be estimated using the following equation, [13],

$$\eta_c = \frac{T_m}{T_m + L} \qquad (2)$$

where T_m is the transmittance of the output mirror and L is the round-trip resonator cavity loss. For our laser, T_m = 6% and L = 40% (double pass losses), therefore

$\eta_C \approx 13\%$ from (2). This means that a significant increase in efficiency can be achieved using an AR-coated Q-switch.



100 ns/div.

Figure 12. Q-switched pulse using Co:YSGG.

Co:YAG was also tested as a Q-switch, but the shortest pulse that could be obtained without damage was 88 ns FWHM. The output energy in this case was about 1 mJ. The pulse shape was similar to that of Co:YSGG. Attempts to obtain shorter pulses by increasing the focusing in the crystal were thwarted by damage to the crystal. Only a single sample Co:YAG was available for these experiments, but it is believed that higher damage threshold crystals can be grown in the future.

The Q-switch results for both Co:YSGG and Co:YAG are summarized in Table 4. Computer modeling using saturable absorber Q-switch rate equations in the fast-relaxing regime agreed reasonably well with the experiments. The numerical simulations also indicate that with AR-coated crystals, a significantly smaller focusing parameter (A_g/A_a) may be used.

Table 4. Co^{2+} Q-switch results.

Q-Switch Matl.	Q-switch thickness (mm)	Int. Trans. (%)	FWHM (ns)	Output energy (mJ)	Thresh-old (J)
Co: YSGG	0.5	85	20	4	40
Co: YAG	8.1	58	88	≈1	84

Conclusions

Passive Q-switching of the Er:glass laser has been demonstrated with U^{2+} and Co^{2+}-doped crystals. The Q-switch results for $U^{2+}:CaF_2$ and Co^{2+}:YSGG are the most encouraging at this time, where 20 ns pulses have been obtained without optical damage. Work to improve the damage resistance of Co:YAG is under way. In addition, further improvements in efficiency and performance are expected with AR-coated Q-switches.

Relaxation lifetime measurements indicate that the Co^{2+}-doped garnet crystals are fast-relaxing compared with a typical Q-switch laser pulse duration, in contrast with the slowly-relaxing U^{2+} Q-switches.

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