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Laser performance of $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$

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Laser action in Cr^{4+} doped yttrium-orthosilicate $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ cooled to 77 K was demonstrated for the first time with 1064 and 532 nm pumping from a pulsed Nd:YAG laser and broad band (@ 840 nm) pumping from a Q-switched Cr:LiSAF laser.

In this letter we present, for the first time, the laser properties of a pure Cr^{4+} system, namely $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$. We observed two interesting characteristics in this new laser: (1) the free running output spectrum depends on the pump wavelength, and (2) the output energy indicates some form of saturation at higher absorbed energies.

The discovery of laser action in the near-infrared (NIR) 1.2 μm band in $\text{Cr}:\text{Mg}_2\text{SiO}_4$ (Forsterite)¹ and $\text{Ca}_2\text{Cr}:\text{Y}_3\text{Al}_5\text{O}_{12}$,² pumped with 1064 nm radiation, indicated Cr^{4+} as the active ion^{2,3} responsible for lasing. However, existence of octahedral sites which accommodate Cr^{3+} ions in these crystals complicates a conclusive analysis of the role that Cr^{4+} plays in the laser action. In order to be able to address this problem, efforts have been underway for quite some time to search for new host materials for Cr^{4+} containing no octahedral site capable of accommodating Cr^{3+} ions.⁴

High quality $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ single crystals were grown by the Czochralski technique. The Cr-concentration in the melt was 2 at. % of the Si ions. The space group of Y_2SiO_5 is C_{2h}^6 (C2/c).⁵ The dimensions of the monoclinic cell are $a=10.410$ Å, $b=6.721$ Å, $c=12.490$ Å, $\beta=109^\circ39'$.⁶ Its melting point is 2070 °C. Cr^{4+} substitutes for Si^{4+} in a slightly distorted tetrahedral site. There is an octahedral symmetry at the Y^{3+} site, but size incompatibility of Cr^{3+} precludes it from entering that site.

The 77 K absorption and fluorescence spectra of the $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ sample used for the laser experiment are shown in Fig. 1. The characteristics of the fluorescence in the NIR depend on pump wavelength. The spectroscopy of $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ is discussed in more detail elsewhere.^{7,8}

The laser experiment with $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ is sketched in Fig. 2. It is a conventional longitudinal laser pumped laser cavity with a flat high reflector and a curved out coupler. The crystal used for the experiment was a parallelepiped with dimensions $5.4 \times 6.65 \times 12.5$ mm³. The end faces of the crystal as well as the fused silica windows of the liquid nitrogen dewar used in this experiment were antireflection coated between 1200 and 1300 nm.

The laser performance characteristics of the $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ crystal at 77 K are summarized in Table I.

However lasing at temperature up to 257 K was demonstrated.

Figure 3 is a plot of the single pulse output energy from the $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ laser versus the absorbed energy for different pump wavelengths. With 1064 nm pumping (pulse duration 15 ns, beam diameter ~ 400 μm), the slope efficiency of the $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ laser decreased drastically as the absorbed energy was increased above 1.0 mJ, showing a strong saturation behavior in the laser output. Similar saturation behavior is observed for 532 nm pumping (pulse duration 15 ns, beam diameter ~ 400 μm) when the absorbed energy is higher than 3.0 mJ. When pumped with 840 nm (pulse duration 35 ns, beam diameter ~ 400 μm), on the other hand, the slope efficiency changes discontinuously to a smaller value at about 2.5 mJ of absorbed energy and the crystal continues to lase at this efficiency up to absorbed energy of about 7.5 mJ. The cause of this saturation behavior is yet to be determined.

The free running spectra due to 1064 nm pumping is distinct from that due to 532 nm pumping. The broadband pumping at 840 nm produces roughly the same output free-running laser spectrum (Fig. 4) as that due to 532 nm pumping. The pump wavelength dependence of the $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ laser indicates the existence of two types of emission in the NIR resulting from different excitations. The exact mechanisms involved in these emissions are not presently understood but are the subject of detailed spectroscopic studies in progress.

The near-infrared broadband luminescence and the laser action due to the 1064 nm excitation is attributed to the

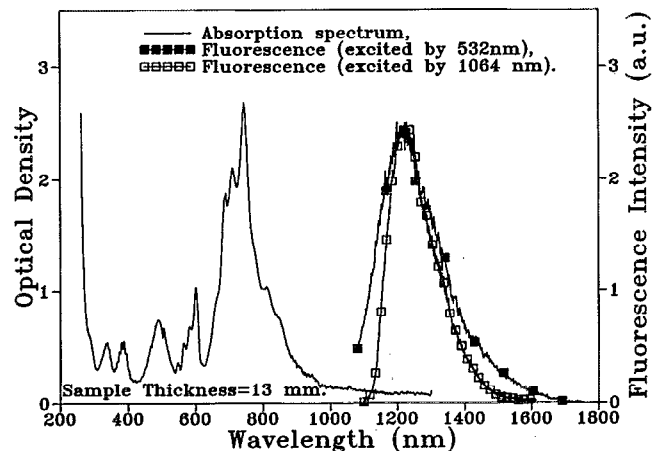


FIG. 1. Absorption and fluorescence spectra of $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ at 77 K.

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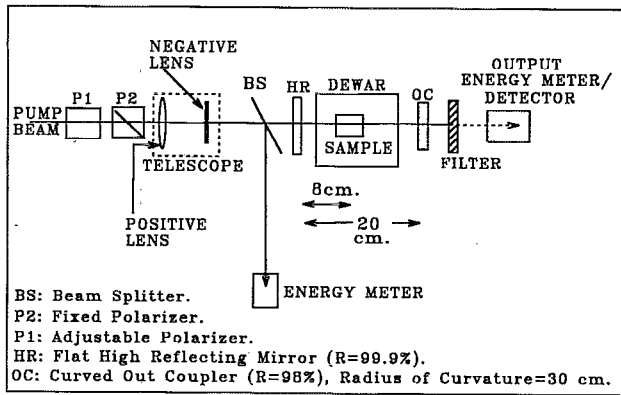


FIG. 2. Schematic diagram of the experimental arrangement for the laser experiment.

TABLE I. Laser performance characteristics of the free-running $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ laser at 77 K.

Pump wavelength (nm)	1064	532	840
Threshold energy (mJ)	0.32	1.5	0.93
Maximum slope efficiency (%)	6.3	12.4	8.2
Peak laser wavelength (nm)	1270	1225	1225
Spectral bandwidth (nm)	10	23	30

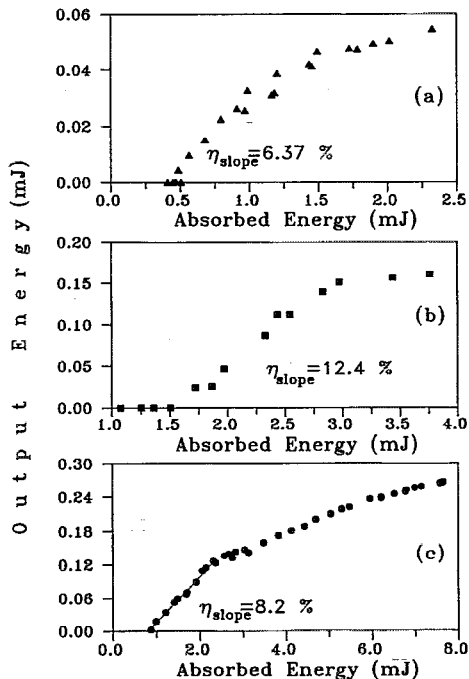


FIG. 3. Output-input curve of $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ laser at 77 K due to pumping with (a) 1064 nm and (b) 532 nm from a Nd:YAG laser, and (c) broadband @ 840 nm from a Cr:LiSAF laser.

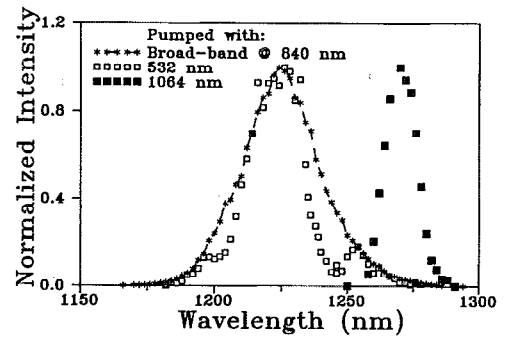


FIG. 4. Free-running $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ laser spectrum due to different pump wavelengths.

transition from the 3T_2 manifold of the Cr^{4+} ion to the 3A_2 ground state. It was observed that intensity of the near-infrared fluorescence of the Cr^{4+} ion in $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ is much weaker than in forsterite ($\text{Cr}^{4+}:\text{Mg}_2\text{SiO}_4$). This resulted in poorer laser performance of $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$ compared to forsterite and is attributed to the nearly ideal tetrahedral symmetry of the Cr^{4+} site in $\text{Cr}^{4+}:\text{Y}_2\text{SiO}_5$. The resulting symmetry selection rule forbids electric dipole radiative transitions between the 3A_2 ground state and the 3T_2 excited state in the ideal tetrahedral symmetry. The key to increasing the electric-dipole coupling between these two states is the degree of distortion of the $(\text{CrO}_4)^{4-}$ tetrahedron. As it becomes distorted the symmetry of the Cr^{4+} site is lowered, splitting the 3T_2 state into crystal field components and increasing the radiative transition probability between the ground state and one or more of the crystal field components of the 3T_2 state in the lower symmetry.⁷ Such effects are evident in forsterite in which the Cr^{4+} site has one of the lowest symmetry (C_s) in the tetrahedral group.⁹ This point is important in that it provides a guide to a systematic approach to the problem of developing more efficient Cr^{4+} doped crystals. It has been suggested that introduction of additional impurity elements having size and charge mismatch with the substituted ions in the host material may provide the necessary mechanism for distorting the $(\text{CrO}_4)^{4-}$ tetrahedron.⁷

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