

NONRADIATIVE RELAXATION AND LASER ACTION
IN TUNABLE SOLID STATE LASER CRYSTALS
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by

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Abstract

Room-temperature pulsed laser action has been obtained in chromium-activated forsterite ($\text{Cr:Mg}_2\text{SiO}_4$) for both 532-nm and 1064-nm pumping. Free running laser emission in both cases is centered at 1235 nm, and has bandwidth of ~ 30 nm. Slope efficiency as high as 22% has been measured. Using different sets of output mirrors and a single birefringent plate as the intracavity wavelength-selecting element tunability over the 1167-1268 nm spectral range has been demonstrated. Continuous-wave laser operation at room temperature has been obtained for 1064-nm pumping from a cw Nd:YAG laser. The output power slope efficiency is 6.8%. The gain cross section is estimated to be 1.1×10^{-19} cm^2 . Spectroscopic studies suggest that the laser action is due to a 'center' other than the trivalent chromium (Cr^{3+}), presumably the tetravalent chromium (Cr^{4+}) in a tetrahedrally coordinated site.

I. INTRODUCTION

Recently, we have reported room-temperature pulsed laser action in chromium-doped forsterite ($\text{Cr:Mg}_2\text{SiO}_4$) for both 532-nm and 1064-nm excitation. Laser emission is continuously tunable over the 1167-1268 nm spectral range, with a potential for extending the tuning range beyond 1300 nm. This wavelength range is of great importance in optical communication and eye-safe ranging. The large bandwidth of the laser emission promises generation of ultrashort pulses through mode-locked operation. Large crystals of chromium-doped forsterite can be grown by Czochralski method, which means that this crystal has a potential to be used as an amplifier medium in the near infrared. Furthermore, the successful cw laser operation of Cr forsterite has the practical implication that various types of laser and amplifier designs are possible.

On the other hand, spectroscopy of chromium-doped forsterite is very unusual and intriguing. It is the first chromium-activated laser crystal, to our knowledge, where the tetravalent chromium ion (Cr^{4+}) in a tetrahedral site is presumably responsible for laser action in the near infrared.

In this paper we review the lasing and spectroscopic properties of this new and important tunable solid-state laser crystal.

II. ACCOMPLISHMENTS

CRYSTAL PROPERTIES AND SPECTROSCOPY

Forsterite is a member of the olivine family of crystals. A unit cell of forsterite has four formula units in an orthorhombic structure of the space group Pbnm. The unit cell's dimensions are: $a=4.76\text{\AA}$, $b=10.22\text{\AA}$, and $c=5.99\text{\AA}$. The Cr^{3+} ion substitutes for the Mg^{2+} ion in two distinct octahedrally coordinated sites: one (M1) with inversion symmetry (C_i) and the other (M2) with mirror symmetry (C_s). The occupation ratio of the two sites by the Cr^{3+} ion is M1:M2=3:2.

Both the single crystals of $\text{Cr:Mg}_2\text{SiO}_4$ used for spectroscopic and laser experiments were grown by the Czochralski method at the Electronic Materials Research Laboratory of the Mitsui Mining and Smelting Co., Ltd., Japan. The first crystal, referred to as sample 1 hereafter, is a $9\text{mm}\times 9\text{mm}\times 4.5\text{mm}$ rectangular parallelepiped with the three mutually orthogonal axes oriented along the b , c , and a crystallographic axes of the crystal. It contains 0.04 at. % of Cr ions which is equivalent to a chromium ion concentration of 6.9×10^{18} ions/ cm^3 . The second crystal (sample 2) is a $6\text{mm}\times 6\text{mm}\times 30\text{mm}$ rectangular parallelepiped with the three mutually orthogonal axes oriented along the a , b , c , crystallographic axes of the crystal. It contains 2.8×10^{18} chromium ions per cm^3 . The $6\text{mm}\times 6\text{mm}$ faces of the crystal were broad-band anti-reflection coated, such that the reflectivity over the 1050-1250 nm spectral range is less than 0.5%.

The room-temperature fluorescence and absorption spectra of $\text{Cr:Mg}_2\text{SiO}_4$ taken with sample 1 for $E\parallel b$ axis are shown in Fig. 1.

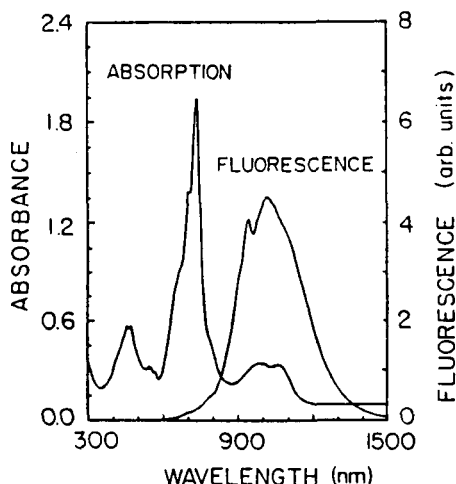


Fig. 1. Absorption and fluorescence spectra of $\text{Cr:Mg}_2\text{SiO}_4$ at room temperature. Both the spectra were taken for $E\parallel b$ axis and excitation along a axis. The thickness of the sample along a axis is 4.5 mm.

The room-temperature fluorescence is a broad band covering the wavelength range from 680-1400 nm. The absorption spectrum is characterized by two broad bands centered at 740 nm and 460 nm, attributed to the ${}^4A_2 \rightarrow {}^4T_2$ and ${}^4A_2 \rightarrow {}^4T_1$ transitions, respectively, of the Cr^{3+} ion. The broad, weak absorption band spanning the 850-1200 nm range is similar to the one observed in chromium-doped GSGG⁸ and is attributed to transitions between states in another 'center', presumably the tetravalent Cr^{4+} ion in a tetrahedral site. This

absorption band overlaps a significant portion of Cr:Mg₂SiO₄ emission, and inhibits laser action in that region.

The absorption and fluorescence spectra of the 'center' in the near infrared spectral region are shown in Fig. 2. The room-temperature absorption spectrum is a double-humped band covering the 850-1200 nm wavelength range. The room-temperature fluorescence spectrum, excited by 1064-nm radiation from a cw Nd:YAG laser extends from 1000-1400 nm and peaks at 1140 nm. At liquid nitrogen temperature both the spectra show a sharp zero-phonon line at 1093 nm followed by elaborately structured sidebands. The fluorescence lifetime is 15 μ s at room-temperature and 20 μ s at liquid nitrogen temperature.

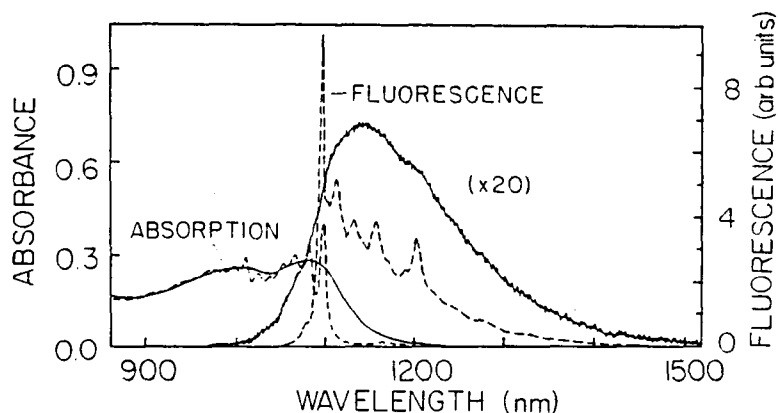


Fig. 2. Near infrared absorption and fluorescence spectra of Cr:Mg₂SiO₄ for 1064-nm excitation at room temperature (solid line) and liquid-nitrogen temperature (broken line) for E||b axis.

PULSED LASER ACTION

The first laser experiments were conducted with sample 1 in a stable cavity. Details of the cavity arrangement have been described elsewhere. The fundamental and the second harmonic emissions from a Q-switched Nd:YAG laser operating at a 10-Hz repetition rate were used for excitation of the near-infrared and the visible bands, respectively. Pulsed laser action was observed for both the 1064-nm and the 532-nm pumping at or above the respective thresholds of 1.25 mJ and 1.37 mJ of absorbed energy. The amplitude and duration of the Cr:Mg₂SiO₄ laser pulse, as well as its delay with respect to the pump pulse, varied with the pulse-to-pulse energy fluctuation of the pump pulses. However, for similar level of excitation and within the time resolution of the experiment, there was no appreciable difference in the delay between the pump pulse and the output laser pulse for the two pump wavelengths. The spectra of the free-running Cr:forsterite laser for both 1064-nm and 532-nm pumping peaked at 1235 nm and had FWHM of 30 nm and 27 nm, respectively. These facts clearly indicate that the same 'center' is active in laser action for both the 532-nm and 1064-nm excitations. For 532-nm pumping there is a fast transfer of excitation from the levels directly pumped to the lasing level. In case of 1064-nm pumping, the lasing level is directly populated. The output power slope efficiencies were 1.8%

for 1064-nm pumping and 1.4% for 532-nm pumping, indicating high losses in the cavity.

To improve the laser performance by reducing the losses anti-reflection-coated sample 2 was used, and care was taken to overlap the pump beam and cavity mode accurately. The sample was longitudinally pumped by the fundamental 1064-nm, 10-ns pulses from a Q-switched Nd:YAG laser in a cavity similar to that used earlier. An output power slope efficiency of 22% was obtained using an output coupler having 87% reflectivity over the lasing region.

TUNABLE OPERATION OF FORSTERITE LASER

Tunable operation of Cr:forsterite laser has been obtained over the 1167-1268 nm spectral range. A single birefringent crystalline quartz plate was inserted in the cavity at Brewster's angle with respect to the cavity axis as the intracavity wavelength-selective element. Smooth tuning over the 1167-1268 nm spectral range was obtained by rotating the tilted plate about an axis perpendicular to its surface and the result is displayed in Fig. 3.

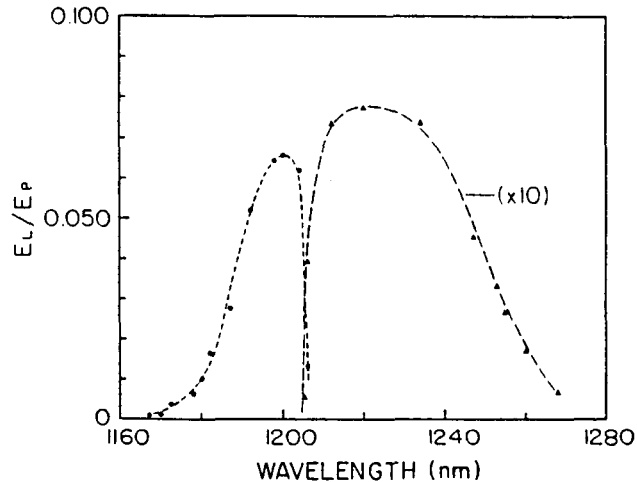


Fig. 3. The ratio of Cr:forsterite laser output (E_L) to the absorbed pump energy (E_p) as a function of wavelength. The curve to the right was taken with an output coupler having 98% reflectivity for 1200-1300 nm range, and one to the left with an output coupler with reflectivity that varied from 99% at 1150 nm to 87% at 1200 nm.

At the peak of the tuning curve at 1220 nm, the output laser energy is ~ 7 μ J/pulse for an absorbed pump energy of 0.9 mJ/pulse. Similar tunable operation has been obtained for 532-nm pumping as well.

CW LASER OPERATION

To obtain cw laser action in Cr:forsterite, sample 2 was placed at the center of a nearly concentric cavity formed by two 5-cm radius mirrors. The output mirror had ~ 1% transmission for the 1175-1250 nm range. The 1064-nm radiation from a cw Nd:YAG laser was focused

by a 75-mm focal length lens to longitudinally pump the sample along the 30-mm path length. The pump beam propagated along the c axis and was linearly polarized along the b axis of the crystal. The pump beam was chopped at a duty factor of 9:1 to reduce heating effects. When the pump beam was not chopped, the Cr:forsterite laser operated at 40% reduced power, indicating the effect due to local heating.

Quasi-cw laser operation was readily obtained for pumping above the lasing threshold of 1.25 W of absorbed power. The spectrum of the free-running cw Cr:forsterite laser peaks at 1244 nm and has a bandwidth of 12 nm.

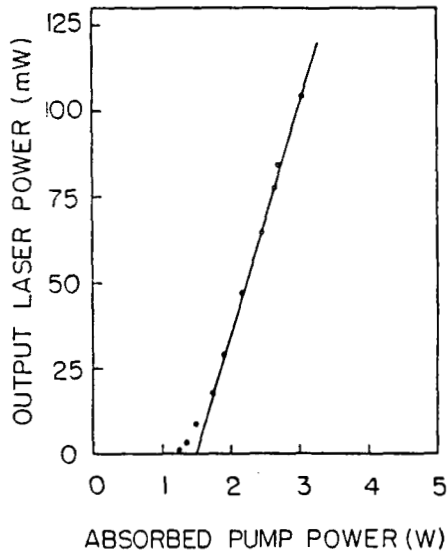


Fig. 4. Output power of the cw Cr:forsterite laser as a function of absorbed pump power.

The cw output power of the Cr:forsterite laser as a function of the absorbed pump power is displayed in Fig. 4. The measured slope efficiency is 6.8%. The experimentally obtained values of the absorbed pump power at the threshold and the slope efficiency together with known mirror reflectivities have been used to estimate a number of key laser parameters. The round-trip cavity loss is estimated to be 12.7%. The effective emission cross section is $\sim 1.1 \times 10^{-19} \text{cm}^2$, and threshold inversion density is $2 \times 10^{17} \text{cm}^{-3}$. Important laser and spectroscopic properties of Cr:forsterite are summarized in Table 1.

TABLE 1 Spectroscopic and laser properties of Cr:forsterite

Property	Value
Major pump bands	850-1200 nm, 600-850 nm, and 350-550 nm
Fluorescence band	680-1400 nm
Room temperature fluorescence lifetime	15 μ s
Cr-ion concentration	$\sim 7 \times 10^{18}$ ions/cm ³
Lasing wavelength (center)	1235 nm (pulsed) 1244 nm (cw)
Spectral bandwidth	~ 30 nm (pulsed) ~ 12 nm (cw)
Slope efficiency	22% (pulsed) 6.8% (cw)
Tuning range	1167-1263 nm
Effective emission cross section	1.1×10^{-19} cm ²

III Publications and Presentations

The presentations of research findings in international conferences and papers published during this reporting period are listed below:

1. "Spectroscopic Properties of Trivalent Chromium-Doped Forsterite: A New Potential Tunable Laser Crystal", V. Petričević, S.K. Gayen, R.R. Alfano, and K. Moriya, Topical Meeting on Tunable Solid State Laser Technical Digest Series, Vol. 20, (Optical Society of America, Washington, D.C. 1987) pp. PD8-1-PD8-4. Presented at the Topical Meeting on Tunable Solid State Lasers, 26-28 October, 1987, Williamsburg, Virginia.
2. "Laser Action in Chromium-Doped Forsterite", V. Petričević, S.K. Gayen, R.R. Alfano, K. Yamagishi, and K. Moriya. Presented at the International Conference on Lasers '87, 7-11 December, 1987, Lake Tahoe, Nevada.
3. "Laser Action in Chromium-Doped Forsterite", V. Petričević, S.K. Gayen, R.R. Alfano, K. Yamagishi, H. Anzai and Y. Yamaguchi, Appl. Phys. Lett. 52, 1040 (1988).
4. "Chromium-Doped Forsterite: A New Tunable Solid State Laser Crystal", V. Petričević, S.K. Gayen and R.R. Alfano, Photonics spectra 22, (3), 95 (1988).
5. S. K. Gayen, V. Petričević, R. R. Alfano, K. Yamagishi, and K. Moriya, "Spectroscopic Properties of $\text{Cr}^{3+}:\text{Mg}_2\text{SiO}_4$: A New Tunable Solid-State Laser Crystal", Bull. Am. Phys. Soc. **33**, 810 (1988). Presented at the March Meeting of the American Physical Society, 21-25 March, 1988, New Orleans, Louisiana.
6. V. Petričević, S. K. Gayen, R. R. Alfano, K. Yamagishi, and K. Moriya, "Room-Temperature Pulsed Laser Action in $\text{Cr}^{3+}:\text{Mg}_2\text{SiO}_4$ ", Bull. Am. Phys. Soc. **33**, 811 (1988). Presented at the March Meeting of the American Physical Society, 21-25 March, 1988, New Orleans, Louisiana.
7. V. Petricevic, S. K. Gayen and R. R. Alfano, "Laser Action in Chromium-Activated Forsterite for Near-Infrared Excitation", Appl. Optics **27**, 4126 (1988).
8. V. Petricevic, S. K. Gayen and R. R. Alfano, "Lasing and Spectroscopic Properties of Chromium-Activated Forsterite", Bull. Am. Phys. Soc. **33**, 1626 (1988). Presented at the Fourth International Laser Science Conference (ILS-IV), October 2-6, 1988, Atlanta, Georgia.
9. V. Petricevic, S. K. Gayen and R. R. Alfano, "Laser Action in Chromium-Activated Forsterite for Near-Infrared Excitation: Is Cr^{4+} the Lasing Ion?", Appl. Phys. Lett. **53**, 2590 (1988).

10. V. Petricevic, S. K. Gayen and R. R. Alfano, "Continuous-Wave Laser Operation of Chromium-Doped Forsterite", Optics Lett. (submitted).
11. V. Petricevic, S. K. Gayen and R. R. Alfano, "Near-Infrared Tunable Operation of Chromium-Doped Forsterite Laser", Applied Optics (submitted).
12. V. Petricevic, S. K. Gayen and R. R. Alfano, "1167-1345 nm Tunable Laser Operation of Chromium-Doped Forsterite", IEEE Photonics Technol. Lett. (submitted).