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Qualitative improvement in the lasing performance of $PbGa_2S_4:Dy^{3+}$ crystals through Na^+ doping

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Abstract. **We have studied the lasing properties of lead thiogallate crystals codoped with dysprosium and sodium ions. Sodium doping has been shown to have an advantageous effect on the optical and lasing properties of lead thiogallate crystals. At a pump pulse duration of 1.5 ms, output pulse energies up to 15 mJ have been obtained at a slope efficiency of up to 4%. Cascade lasing on two Dy^{3+} transitions, at 4.3, 4.5, 4.65, 5.2, 5.3, and 5.4 μm, has been demonstrated.**

Keywords: *lead thiogallate crystals, dysprosium ions, sodium ions, codoping, Dy^{3+} cascade lasing.*

Dysprosium-doped lead thiogallate ($PbGa_2S_4:Dy^{3+}$) crystals are potentially attractive as gain media for mid-IR lasers [1 – 4]. Because of the large difference in ionic radius between Pb^{2+} and Dy^{3+} (1.29 and 1.03 Å in eightfold coordination), doping with even 1 at % dysprosium considerably degrades the optical homogeneity of the crystals and impairs the reproducibility of their lasing properties [3]. We believe that Na^+ doping of $PbGa_2S_4:Dy^{3+}$ might mitigate the effect of the large difference in ionic radius between Pb^{2+} and Dy^{3+} and assist in charge compensation. Recall that the ionic radius of Na^+ in eightfold coordination is 1.16 Å. As pointed out previously [3], doping with Na^+ enables the dysprosium concentration to be increased. This paper examines the effect of Na^+ doping on the spectroscopic and lasing properties of $PbGa_2S_4:Dy^{3+}$ crystals.

The lasing properties of $PbGa_2S_4:Dy^{3+}:Na^+$ (PGS : Dy : Na) crystals were studied under pumping by the 1.318-μm free-running Nd : YAG laser line at a pulse repetition rate of 3 Hz. The pump laser incorporated two pump cavities with active elements 6.3×100 mm in dimensions. To suppress lasing on the main neodymium transition at 1.064 μm, the elements were mounted in a V-shaped cavity with a deflecting dichroic mirror having the highest reflectance at the lasing wavelength (1.318 μm) and the highest transmittance at the competing wavelength (1.064 μm). Each element was pumped by a flashlamp with a 90-mm spark gap and separate power supply, with controlled discharge pulse duration. To obtain lasing only at 1.318 μm and suppress the 1.34-μm lasing line, the laser cavity contained in addition a selective mirror with high transmittance at 1.318 μm and high reflectance at 1.34 μm.

The cavity of the doped lead thiogallate laser was formed by two mirrors. The pump beam was coupled into the active element through a flat dichroic cavity input mirror having the highest transmittance at 1.318 μm and the highest reflectance (~ 0.998) in the range 4 – 5.5 μm. The output coupler had a 500-mm radius of curvature and was located ~ 40 mm from the input mirror. To optimise the output energy, we used output couplers of different reflectances. The pump beam was focused into the active element by a lens with a focal length of 100 mm. To prevent breakdown of the active element and cavity mirrors, the lens was mounted so that its focus was beyond the cavity of the PGS : Dy laser.

The output energy of the PGS:Dy laser was measured with a Coherent EPM-2000 power meter. Laser emission spectra were obtained using an MDR-23 grating monochromator (150 lines per millimetre), liquid-nitrogen-cooled indium antimonide detector and computer-interfaced Tektronix TDS-3052 digital oscilloscope.

Figure 1 plots the output pulse energy against absorbed pump energy for the PGS : Dy : Na laser at different reflectances of the output coupler. The highest pulse energy (5 mJ) and slope efficiency ($\eta_{dif} = 3.6\,\%$) were achieved at a 4.3-μm reflectance of the output coupler $R_1 = 85\,\%$. Note that, for two mirrors with identical 4.3-μm reflectances ($R_2 = R_3 = 90\,\%$), we obtained the same η_{dif} but markedly different lasing thresholds because the mirrors differed in 5.4-μm reflectance.

Indeed, the wavelength 5.4 μm corresponds to a peak in the luminescence spectrum of dysprosium in lead thiogallate crystals due to the $^6H_{9/2} - {}^6H_{11/2}$ transition. Pumping at 1.318 μm excites the dysprosium ion to the $^6H_{9/2}$ level, with subsequent decay to the $^6H_{11/2}$ upper level of the 4.3-μm laser transition. It follows from an earlier analysis [1] that the $^6H_{11/2}$ upper laser level is populated mainly through nonradiative relaxation from the $^6H_{9/2}$ level. The fraction of radiative transitions from the $^6H_{9/2}$ to the $^6H_{11/2}$ level is just 12 %. Lasing on the $^6H_{9/2} - {}^6H_{11/2}$ transition considerably accelerates the filling of the $^6H_{11/2}$ level. This suggests that cascade lasing in PGS : Dy : Na crystals should lead to an increase in 4-μm lasing efficiency.

Also shown in Fig. 1 is the output pulse energy against absorbed pump energy for a PGS : Dy crystal of the same

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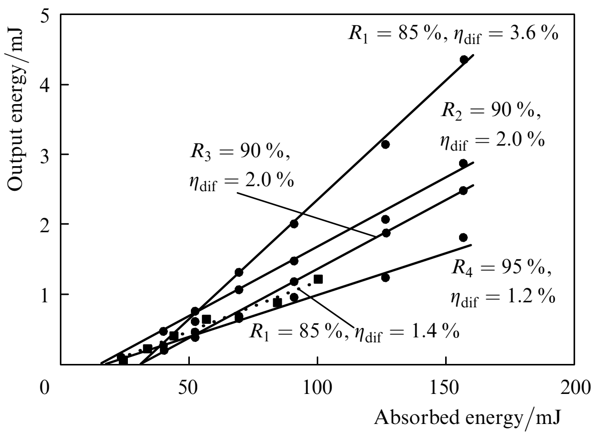

Figure 1. Output pulse energy as a function of absorbed pump energy for the $PbGa_2S_4:Dy^{3+}:Na^+$ laser at different reflectances of the output coupler. The dotted line represents the data for a Na^+-free $PbGa_2S_4:Dy^{3+}$ crystal.

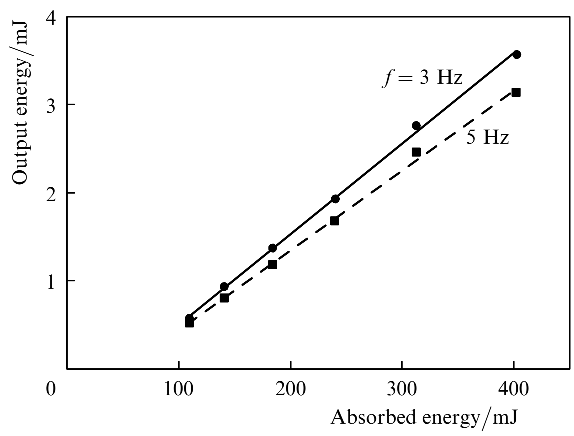

Figure 3. Output pulse energy as a function of absorbed pump energy for the $PbGa_2S_4:Dy^{3+}:Na^+$ laser at two pump pulse repetition frequencies.

length as the PGS : Dy : Na crystal. Without Na^+, the maximum absorbed pump energy is substantially lower, because of the lower dysprosium concentration, and, all other factors being the same, the slope efficiency is lower by more than a factor of 2 (1.4 %).

The emission spectrum of the PGS : Dy : Na laser with a reflectance of the output coupler $R_1 = 85\,\%$ is shown in Fig. 2 together with the luminescence spectrum of dysprosium ions in a lead thiogallate crystal. The dysprosium lasing spectrum contains peaks at 4.3, 4.5 and 4.65 μm, due to the $^6H_{11/2} - {}^6H_{13/2}$ transition. In addition, the spectrum contains lines of the higher lying laser transition $^6H_{9/2} - {}^6H_{11/2}$, with peak-emission wavelengths of 5.2, 5.3 and 5.4 μm, which points to a significant effect of this transition on the $^6H_{11/2}$ level population and to dysprosium cascade lasing in lead thiogallate pumped at 1.318 μm (to the $^6H_{9/2}$ level).

Figure 3 plots the output pulse energy against absorbed pump energy at two pump pulse repetition rates. As seen, increasing the pump pulse repetition rate slightly reduces the slope efficiency of the laser, presumably because of the heating of the PGS : Dy : Na crystal, which had no cooling system.

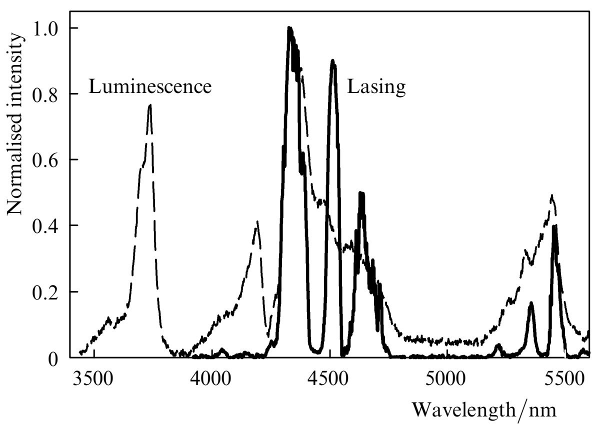

Figure 2. Emission spectrum of the $PbGa_2S_4:Dy^{3+}:Na^+$ laser with a 85 % reflectance of the output coupler and luminescence spectrum of dysprosium ions in a lead thiogallate crystal.

To examine the effect of pump pulse duration on the lasing properties of PGS : Dy : Na crystals, the duration of discharge pulses that pumped the Nd : YAG crystals was varied from 250 to 1000 μs. Figure 4 shows the output energy versus pump pulse energy for the PGS : Dy : Na laser at two discharge pulse durations. Note that, as distinct from that in Fig. 1, the abscissa here represents the total energy incident on the crystal rather than the absorbed pump energy. It follows from these data that the slope efficiency increases with an increase in pump pulse duration from 250 to 1000 μs, which is due to the better match between the pump pulse duration and the lifetime of the $^6H_{11/2}$ upper laser level (2 ms). The pump and laser pulse shapes at a discharge pulse duration of 1 ms are shown in Fig. 5. The Nd : YAG and PGS : Dy : Na laser pulses are seen to be identical in width: ~ 800 μs FWHM.

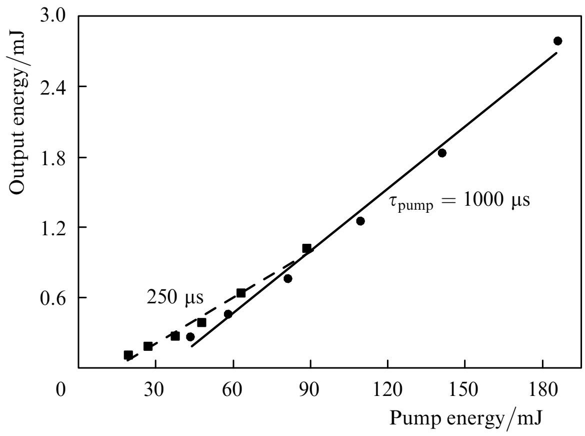

Figure 4. Output energy as a function of incident pump energy for the $PbGa_2S_4:Dy^{3+}:Na^+$ laser at two discharge pulse durations, τ_{pump}, in the lamp that pumped the Nd : YAG laser.

Figure 6 plots the measured output energy against absorbed pump energy for the PGS : Dy : Na laser at a discharge pulse duration of 1.5 ms. Under such conditions, we obtained the highest output pulse energy, 15 mJ, at a slope efficiency of 4 %, which is well above the level reached previously [1 – 4]. Taking into account the difference between the absorbed and output photon energies, we

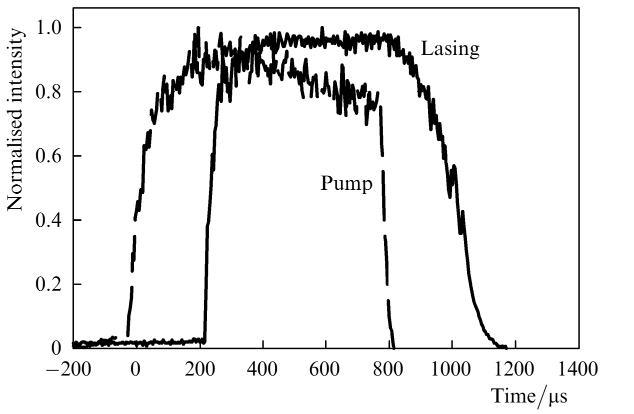

Figure 5. Oscilloscope traces of the pump laser and lead thiogallate laser pulses at a discharge pulse duration of 1 ms.

find that the quantum efficiency of the lasing process was $\sim 6\,\%$.

In the experiments described above, end pumping was used. It was of interest to study the performance of a side-pumped PGS : Dy : Na laser. To produce an appropriate pump beam profile, we used cylindrical focusing lenses and beam expansion to a diameter of $\sim$ 10 mm. Figure 7 plots the output energy against incident pump pulse energy for a side-pumped PGS : Dy : Na laser at two focal lengths of the cylindrical lens. As would be expected, the shorter focus lens increases the pump power density and lowers the lasing threshold. Moreover, at low pump energies the slope efficiency increases, whereas at high energies absorption saturation leads to bleaching of the pump channel and a reduction in absorbed energy.

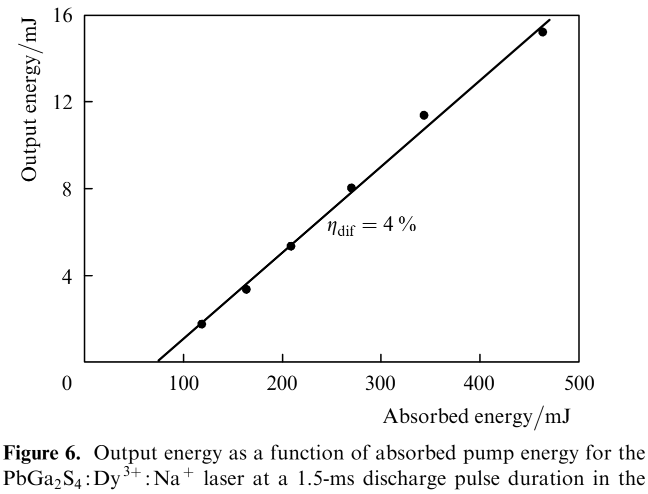

Figure 6. Output energy as a function of absorbed pump energy for the $PbGa_2S_4 : Dy^{3+} : Na^+$ laser at a 1.5-ms discharge pulse duration in the lamp that pumped the Nd : YAG laser.

Thus, we prepared more optically homogeneous $PbGa_2S_4 : Dy^{3+} : Na^+$ crystals with an increased Dy^{3+} concentration. The high optical quality of the crystals allows higher pump energies to be used, without optical breakdown of the active element. Using active elements produced from the crystals, an output pulse energy of 15 mJ in the mid-IR has been obtained for the first time at a slope efficiency of 4 %. Cascade lasing on the dysprosium $^6H_{9/2} - {}^6H_{11/2}$ (5.2, 5.3 and 5.4 μm) and $^6H_{11/2} - {}^6H_{13/2}$ (4.65, 4.5 and 4.3 μm) transitions has been demonstrated.

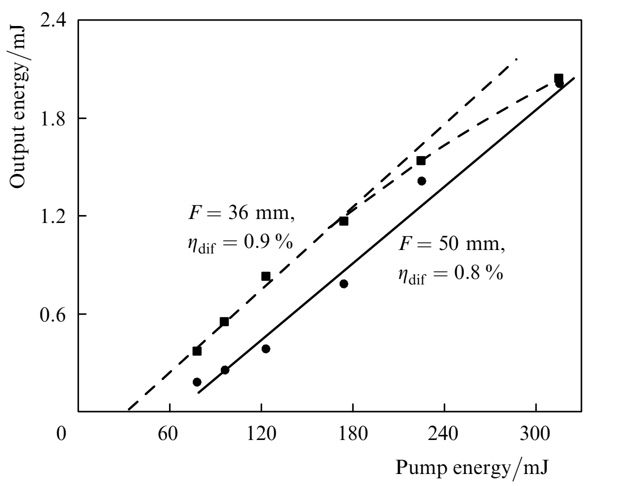

Figure 7. Output energy against incident pump pulse energy for the $PbGa_2S_4 : Dy^{3+} : Na^+$ laser at two focal lengths F of the cylindrical focusing lens.

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