1359
submitted to Journal of Luminescence 8/96
accepted 12/96

Version Date: 13 August 1996

Spectroscopy of potential mid-infrared laser transitions in gallium lanthanum sulphide glass

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Abstract

This paper presents experimental results on the mid-infrared emission of rare-earth doped gallium lanthanum sulphide glasses and fibres. Measured fluorescence includes the 3.4 μm transition ($^1G_4 \rightarrow {}^3F_4$) in Pr^{3+}-doped Ga:La:S fibre, the 3.6 μm transition ($^4F_{9/2} \rightarrow {}^4I_{9/2}$) in Er^{3+}-doped Ga:La:S fibre, and the 4.3 μm transition ($^6H_{11/2} \rightarrow {}^6H_{13/2}$) in Dy^{3+}-doped Ga:La:S bulk glass. Fibre lasers with these emission wavelengths would be of interest for gas sensing applications.

Keywords: glass, fibre, laser, infrared

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Main Text

Diode-pumped rare-earth doped solid-state lasers in bulk or fibre form would offer a compact and efficient alternative to the either relatively weak or very complex existing mid-infrared sources such as thermal emitters, gas lasers and OPOs. A prerequisite for the rare-earth host material is a low phonon energy leading to low non-radiative decay rates and therefore higher quantum efficiencies for mid-infrared transitions. The longest laser wavelengths for solid-state crystal and fibre lasers obtained today are 7.24 μm in a Pr^{3+}-doped $LaCl_3$ crystal and 3.95 μm in a Ho^{3+}-doped ZBLAN fibre [1,2]. Both materials are hygroscopic and difficult to handle. Although the fluorozirconate glass has the more advantageous fibre geometry, the high phonon energy of more than 500 cm^{-1} results in much lower quantum efficiencies for mid-infrared transitions than in the $LaCl_3$ crystal (210 cm^{-1}) and a multiphonon absorption edge that starts at about 3 μm causing absorption losses in the mid-infrared wavelengths region.

Our approach towards mid-infrared laser sources is based on the stable, non-toxic and non-hygroscopic chalcogenide glass gallium lanthanum sulphide (Ga:La:S) which can be pulled into fibres [3]. Bulk glasses and fibres with the molar composition $70Ga_2S_3$:$30La_2S_3$ doped with different rare-earth ions were investigated with regard to mid-infrared emission.

The low phonon energy (425 cm^{-1}) of the glass lattice causes a wide infrared transmission extending beyond 8 μm with a loss minimum around 5 μm [3,4]. It also reduces the multiphonon decay rate of rare-earth energy levels compared to fluorozirconate and silica glasses. These properties in conjunction with higher radiative decay rates due to the large refractive index (2.4) and the high covalency of the glass bondings open the possibility for laser transitions between closely spaced energy levels.

Fig. 1 and 2 show fluorescence spectra of Pr^{3+}-, Er^{3+}-, and Dy^{3+}-doped Ga:La:S glass measured with a 300 mm monochromator and a liquid nitrogen cooled InSb detector. The fluorescence was collected from the end of the fibres and from the side of the bulk samples. The spectra were corrected using a black body source.

The 500 ppm wt. Pr^{3+}-doped fibre with a length of 2.05 m and a diameter of 170 μm was pumped with 4 W of a Nd:YAG laser at 1064 nm. The pump power was absorbed by the long wavelength tail of the $^3H_4 \rightarrow {}^1G_4$ absorption. The 3.4 μm emission from the $^1G_4$ level to the next lower lying level $^3F_4$ is shown in Fig. 1.a. Additional transitions from the $^1G_4$ level were measured at 2.9 μm, 2.1 μm, 1.85 μm and 1.3 μm.

The 24400 ppm wt. Er^{3+}-doped fibre had a length of 8.6 cm and a diameter of 270 μm. A DCM dye laser tuned to 670 nm with an output power of 70 mW was used to excite the Er^{3+} ions into the $^4F_{9/2}$ level. Emission from the $^4F_{9/2}$ level was measured at 3.6 μm ($^4F_{9/2} \rightarrow {}^4I_{9/2}$) as shown in Fig. 1.b. and at 2.0 μm ($^4F_{9/2} \rightarrow {}^4I_{11/2}$). Laser action an the $^4F_{9/2} \rightarrow {}^4I_{9/2}$ transition was reported in ZBLAN fibre [5]. We predict that this transition should have a quantum efficiency about four times higher in Ga:La:S glass.

The 9900 ppm wt. Dy^{3+}-doped bulk sample was pumped with a Ti:sapphire laser at 815 nm to excite the closely spaced $^6F_{3/2}$, $^6F_{5/2}$ levels. The metastable $^6H_{11/2}$ levels is populated via non-radiative decay. The 4.3 μm emission ($^6H_{11/2} \rightarrow {}^6H_{13/2}$) is shown in Fig. 3. The uncorrected spectrum (a) shows clearly the fundamental absorption of carbon dioxide present in air. Further infrared transitions were measured at 1.76 μm and 2.83 μm.

Table 1 contains the emission cross sections calculated from the results of Judd-Ofelt calculations and the Füchtbauer-Ladenburg equation and the measured lifetime of the upper levels in lightly doped samples for each transition. The lifetimes do not correspond to the

energy gaps clearly indicating that the decay rates in this efficient low phonon material are dominated by the radiative decay.

In conclusion we present the spectroscopic data of three mid-infrared transitions in Pr^{3+}-, Er^{3+}-, and Dy^{3+}-doped Ga:La:S glass. The recent report of fibre fabrication and laser action in this glass system show the potential for mid-infrared fibre lasers that would find application in gas sensing [3,4].

Acknowledgements

Starting materials were supplied by Merck Ltd. The authors thank Prof. H. Rutt for providing equipment and for helpful discussions.

References

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Tables

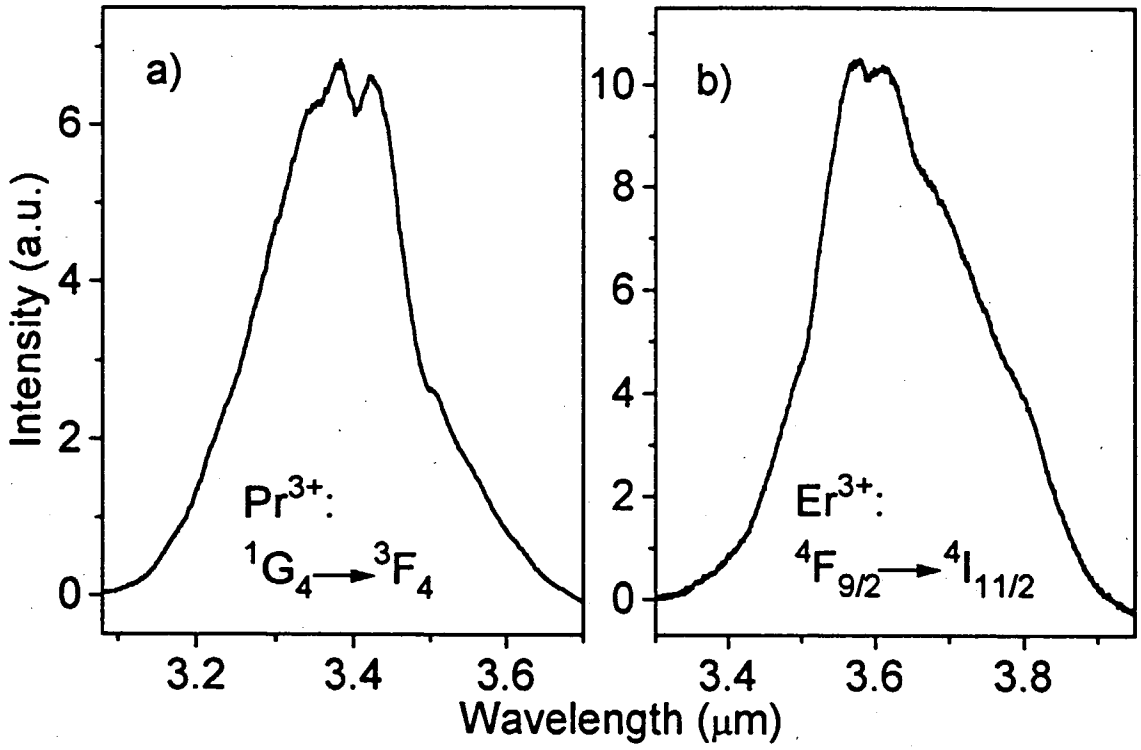
Table 1. Emission wavelength, peak cross section, and upper level lifetime of mid-infrared transitions in Pr^{3+}-, Er^{3+}-, and Dy^{3+}-doped Ga:La:S glass

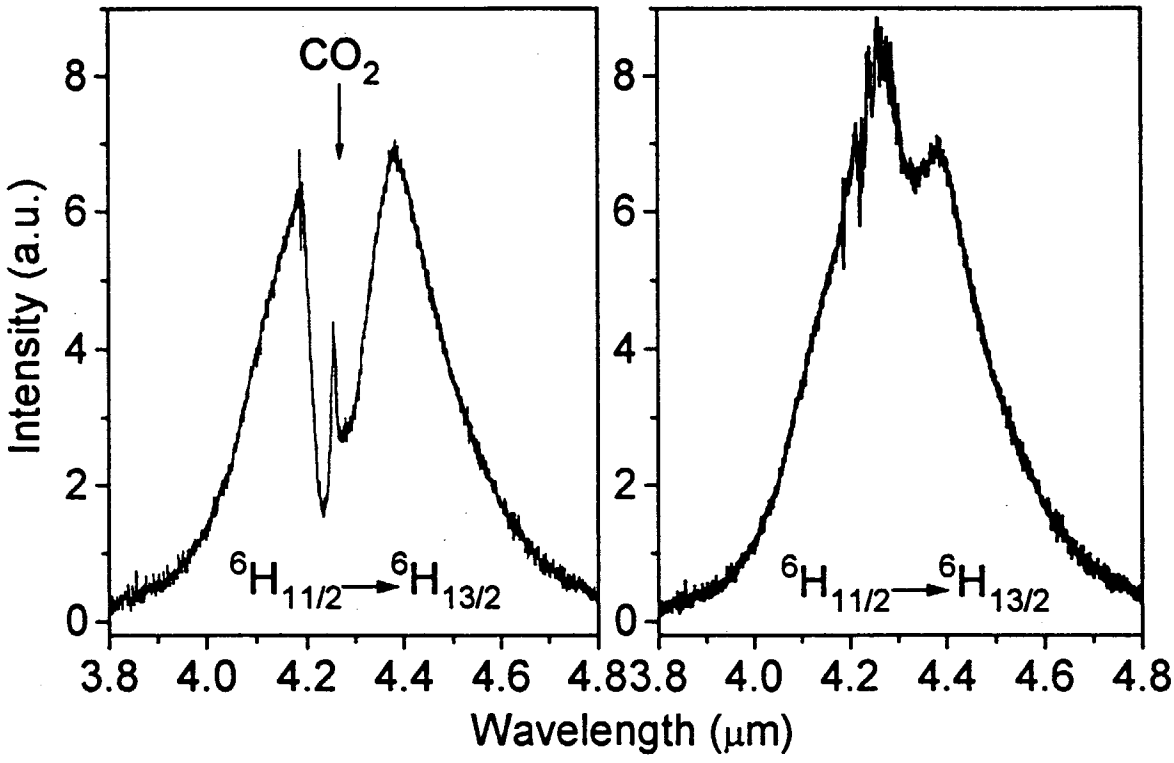
Rare-earth Transition	λ_{peak} (nm)	σ_{em} ($10^{-20}cm^2$)	τ_{upper} (μs)
Pr^{3+} $^1G_4 \rightarrow {}^3F_4$	3425	0.86	295
Er^{3+} $^4F_{9/2} \rightarrow {}^4I_{9/2}$	3620	0.43	100
Dy^{3+} $^6H_{11/2} \rightarrow {}^6H_{13/2}$	4270	1.17	1200

Figure Captions

Fig. 1. Fluorescence spectra of a) Pr^{3+}-doped Ga:La:S fibre and b) Er^{3+}-doped Ga:La:S fibre

Fig. 2. Uncorrected (a) and corrected (b) fluorescence spectrum of Dy^{3+}-doped Ga:La:S glass


a)
b)
Intensity (a.u.)
Wavelength (μm)
Pr3+:
1G4 → 3F4
Er3+:
4F9/2 → 4I11/2
0
2
4
6
8
10
3.2
3.4
3.6
3.8


CO2
Intensity (a.u.)
0
2
4
6
8
6H11/2→6H13/2
6H11/2→6H13/2
3.8
4.0
4.2
4.4
4.6
4.8
Wavelength (μm)