Spectroscopic and lasing characteristics of samarium doped glass fibre

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Abstract: The fluorescence spectra of trivalent samarium doped glass fibres are described. In silica glass Sm$^{3+}$ has a narrow fluorescence of 2.2 nm f.w.h.m. at a wavelength of 650 nm. The influence of fluorescence line narrowing and large external electric fields on this line is reported. Visible laser emission is obtained at this wavelength when the fibre is pumped in a Fabry Perot cavity. The performance of the laser in continuous, Q-switched and self mode-locked operation is described. The basic theory of self-mode-locking is presented.

1 Introduction

We achieved visible laser operation in a glass host for the first time with samarium$^{3+}$ doped silica optical fibre [1]. Optical fibres are ideal hosts for rare-earth lasers because high intensities (10$^{11}$ W/m$^2$) may be confined for long interaction lengths of many metres in a single mode waveguide. Optical pumping of low dopant concentration bulk glass lasers is difficult because of the small absorption, but a single mode optical fibre enables efficient longitudinal pumping of a glass in which the dopant concentration can be less than 0.01 wt.%. Continuous room temperature operation of some rare earth lasers has only been achieved in an optical fibre geometry because of the efficient pumping and cooling [2].

Samarium doped glass is well known as a cladding for Nd-glass laser rods owing to its high absorption at 1064 nm but low absorption between 500 nm and 900 nm. Reports of laser action in samarium to date have been limited to Sm$^{2+}$ in crystal hosts [3]. Kazakov [4] has reported observation of induced emission of Sm$^{3+}$ ions in TbF$_3$ crystals at 77 K under pulsed excitation, but until our work this was the only report of laser action in Sm$^{3+}$. We show here that laser action of Sm$^{3+}$ in glass is only possible with low concentrations which require a fibre geometry for successful operation. This laser may be operated continuously, Q-switched or self mode-locked.

Our spectroscopic study of samarium doped silica in low concentrations (0.1 wt.%) reveals a very narrow transition from the $^4$G$_{5/2}$ level which has a linewidth of 2.2 nm. We use this narrow spectral line as a spectroscopic tool to determine the influence of temperature, electric field, host glass and excitation wavelength on the samarium in glass.

2 Fabrication of Samarium doped fibre

Optical fibres were fabricated by the solution doping technique [5]. A variety of samarium doped fibres with different host glasses were fabricated. The silica fibres were fabricated by MCVD and doped with GeO$_2$, Al$_2$O$_3$ or P$_2$O$_5$ to produce fibres with numerical apertures of 0.18-0.24.

Fibres were also fabricated from both commerical multicomponent Sm$^{3+}$ glasses and phosphate glass PK50 which we doped with Sm$^{3+}$. These fibres were made using the rod in tube technique. The composition of the undoped PK50 glass is 70 wt.% P$_2$O$_5$, 12 wt.% K$_2$O, 10 wt.% Al$_2$O$_3$, 3 wt.% B$_2$O$_3$ and 5 wt.% CaO. The most convenient pump band is from ground to the $^4$I$_{15/2}$ level which has a strong absorption of the 488 nm line from an argon-ion laser. Unfortunately this corresponds to a two photon absorption of germanium defect centres in GeO$_2$ doped fibres. The effect of this colour centre excitation is to increase the absorption of the fibre from the uv to the red part of the spectrum [6]. As a result, samarium fibre lasers, containing germanium, experience an increase in the lasing threshold pump power after periods of pumping at 488 nm. To overcome this problem, fibres were made without germania, having either pure silica core and B$_2$O$_3$ doped depressed index cladding or cores doped with several percent Al$_2$O$_3$.

3 Energy levels of Samarium in silica

The energy level diagram, shown in Fig. 1, is produced from data for Sm$^{3+}$ in LaCl$_3$ from Dieka [7] with the levels adjusted to correlate with peaks in the absorption spectrum of a typical fibre, which is also shown. There is a good correlation between the measured absorption peaks and the expected position of the energy levels above 10,000 cm$^{-1}$. Samarium, unlike most rare earths in glass, has a particularly narrow linewidth of 2.2 nm f.w.h.m. for fluorescence and absorption from the $^4$G$_{5/2}$ metastable level. The Gaussian shape of this line indicates that there is only likely to be one Stark level active.

Silica glass has approximately a tetrahedral symmetry. From this and a knowledge of the energy levels involved we can predict that there should be three possible Stark levels of both the $^4$G$_{5/2}$ metastable level and the $^4$H$_{11/2}$ level.
ground state, but only one is seen. This observation may be caused by a variation between the symmetry of the host glass and that seen by the samarium ions.

4 Influence of host glass

The host glass is seen to strongly influence the fluorescence spectra of samarium shown in Figs. 2–6. In all cases, the fibre was weakly pumped with 488 nm to prevent distortion of the spectra by amplification. Si/Ge fibres have a narrow main transition between \( ^4G_{5/2} \) level and the \( ^4H \) levels with only one Stark component dominating, but there is evidence of weak components either side. These weak components are smeared out into a continuum which decreases in intensity with temperature owing to a reduction in the thermal population of the upper Stark components. The lifetime of the peak fluorescence in silica fibres was measured as 1.5 ms. Fibres containing phosphorus have the fluorescence spread more evenly among the Stark components than Si/Ge fibres which leads to a reduction in the peak cross-

sections. This is probably because of the P=O bonds in phosphorus glass. A similar effect has been observed by Ainslie et al. [13] in Nd\(^{3+}\) doped silica glass with increasing P\(_2\)O\(_5\) concentration.

![Fluorescence spectra of samarium in germania doped silica](image1)

**Fig. 3** Fluorescence spectra of samarium in germania doped silica

![Fluorescence spectra of samarium in phosphorous/germania doped silica](image2)

**Fig. 4** Fluorescence spectra of samarium in phosphorous/germania doped silica

The most significant change in fluorescence with host glass occurs in fibres doped with phosphorus and aluminium (Fig. 5). Strong fluorescence from the \( ^4H_{11/2} \) level is seen for the first time in these glasses. The peak at 680 nm has a cross-section of \( 2.5 \times 10^{-21} \) \( \text{cm}^{-1} \), determined by Fuchtbauer-Ladenburg analysis, which is larger than that at 650 nm in this fibre. Aluminium codoping has been shown [4] to give rise to Sm\(^{2+}\) ions in the glass host which have fluorescence peaks at 684 nm and 725 nm. Further work is necessary to determine the contribution from Sm\(^{2+}\) and Sm\(^{3+}\) in this codoped glass,

**Fig. 5** Fluorescence spectra of samarium in phosphorus-alumina doped silica
but the wide spread of fluorescence from 560 nm to 750 nm in these glasses suggests that it may be possible to achieve laser action over this range in a correctly designed host glass with efficient pumping.

For comparison we also include our results from 1 wt.% Sm$^{3+}$ doped PK50 phosphorus glass. In this glass the largest cross-section is at 600 nm (Fig. 6). This is

![Image](image_url)

**Fig. 6** Fluorescence spectra of samarium in PK50 glass fibres

weaker than the 650 nm peak cross-section in silica glass owing to a broader fluorescence spectrum in this glass type. The lifetime of 1.9 ms is the longest that we have observed in any samarium doped glass. Samarium doped PK50 may provide a way of making short visible fibre lasers. The long life time and short length will be particularly suitable for high power Q-switching. Fibre fabricated from commercial Sm$^{3+}$ doped glasses with 5 and 10% concentrations have lifetimes of less than 100 μs at 650 nm, which is probably a result of concentration quenching.

We have, to date, only achieved laser action in silica-germania and silica-phosphorus germania glasses. This is because of the large cross-section of 6.4 x 10$^{-24}$m$^2$ at the 650 nm fluorescence peak.

5 Line width measurements

The narrow fluorescence line enables examination of broadening mechanisms of the samarium ions in glass. Nonresonant fluorescence line narrowing was used to determine the inhomogeneous line width. Pumping with a range of wavelengths available from an argon ion laser accesses different host sites. For clarity only the two extreme fluorescence spectra corresponding to pump wavelengths of 488 nm and 496.5 nm are shown in Fig. 7. The maximum line shift was measured as 0.35 nm (8 cm$^{-1}$). Assuming that there is negligible cross site relaxation we take this to be the material’s inhomogeneous linewidth. This is much narrower than reported in lanthanum aluminium silicate glass [10] in which the inhomogeneous line width was measured as 240 cm$^{-1}$. The residual linewidth of 70 cm$^{-1}$ may be attributed to homogeneous broadening. This homogeneous line width is typical of rare earth doped glasses at 20°C. On cooling to 77 K a small linewidth reduction would be expected but no change was observed in samarium doped silica.

The narrow inhomogeneous linewidth indicates that the optically active levels in Sm$^{3+}$ in silica are well shielded from the lattice. This leads to a larger cross-section and hence the realisation of laser action in Sm$^{3+}$ doped silica glass. Further evidence of shielding is observed when electric fields are applied to a samarium doped fibre during fluorescence measurements. The fluorescence spectra was measured in zero field and in an electric field of 150 kV/mm across the fibre core by using a fibre incorporating metal electrodes [11] to obtain such high fields. No change in the width or position of the fluorescence spectra was observed within the measurement resolution of 1 Å.

6 Continuous laser operation

A narrow fluorescence spectra and long lifetime provide the basis for a laser material. We reported the first observation of continuous simulated emission in samarium doped glass [1]. The laser is formed into a Fabry Perot cavity by butting dielectric mirrors to each cleaved end of the fibre. One mirror is 99% reflecting at the lasing wavelength and nonreflecting at the pump wavelength of 488 nm. Output mirrors ranging from 97% reflectivity to 4%, owing to Fresnel reflection at the fibre end, have been used to obtain lasing.

Laser output versus pump power is shown in Fig. 8. An efficiency of 12.7% was obtained with an output coupler of 60% reflectivity. The gain is estimated at 0.02 dB/mW/m in a fibre with a dopant concentration of 250 ppm and a core area of 7 x 10$^{-12}$ m$^2$. Laser operation is restricted by fibre absorption and scatter loss at the laser wavelength. In the unpumped state the loss at 650 nm is 50 dB/km, but this increased in germania doped fibres owing to pumping [6].

7 Q-switched operation

The long lifetime (1.6 ms) of samarium in silica glass provides a large energy storage capacity in the laser cavity.
in the cavity between the fibre and the output mirror. The low dopant concentrations used here require fibre lengths of 2 m for efficient operation. A Q-switched pulse from a 2 m long fibre laser with an output mirror of 60% reflectivity is shown in Fig. 9. The long tail is partly because of the cavity decay time and may be reduced by slicing the end of the pulse with fast turn off of the AO modulator. Peak powers of 10 W have been achieved with pulse widths of 300 ns. This is below expected power levels owing to intracavity loss in the fibre. To fully utilise the energy storage capacity of samarium doped glass fibres for high power pulses the dopant concentration must be increased to 10,000 parts in 10⁶ without increase in loss at 650 nm or decrease in lifetime.

![Graph showing Q-switched pulse from a samarium fibre laser](image)

**Fig. 9** 10 W Q-switched pulse from a samarium fibre laser

8 Self mode-locking

In Q-switched operation a high intensity is maintained in the fibre for several metres of interaction length. This leads to a break up of the pulse into narrow mode-locked pulses as shown in Fig. 10. A nonlinear index change is produced by the Q-switched pulse within the laser cavity. The phase change per round trip of the cavity is given by

\[
\Delta \phi = \frac{4 \pi n_2 l}{\lambda A} P
\]

where \( l \) is the cavity length taken as 8 m, \( n_2 \) is the nonlinear index of fused silica, \( P \) is the intracavity pulse power taken as 2 W and \( A \) is the mode spot area taken as \( 7 \times 10^{-12} \) m². The resultant phase shift is 0.9\( \pi \) which is sufficient to couple the modes together and produce self mode locking. This phase shift was only possible when the intercavity intensity is increased by Q-switching.

Alternatively, this phenomena may be considered as a set of coupled nonlinear wave equations of the form

\[
\frac{dE_i}{dx} = gE_i + S_p(\omega_i) + \varepsilon_0 \chi^{(3)} \sum E(\omega_j)E(\omega_k)E(\omega_l)
\]

(2)

where \( \chi^{(3)} \) is the third order nonlinear susceptibility and \( E(\omega) \) are the fields of each longitudinal mode in the fibre. The gain is given by \( g \) and the spontaneous emission is \( S_p \). The summation is taken over all the modes, which are assumed to be equally spaced and contribute by 4-wave mixing to the generated mode. Therefore, if the generated mode has a frequency \( \omega \) the mode spacing is \( \Delta \), then for example modes with frequencies \( \omega - 3\Delta, \omega + \Delta, \omega + 2\Delta \) will contribute to the generated mode if the phases add constructively. The condition for phase matching requires that the propagation constants obey this condition

\[
k(\omega) = k(\omega - 3\Delta) + k(\omega + \Delta) + k(\omega + 2\Delta)
\]

(3)

There are similar equations for the phase matching between all the other modes. We argue that a self consistent solution is only possible if all the modes are in phase. The effect of the nonlinear coupling in eqn. 2 is to drive the phases of all the modes together. The spontaneous emission coupled into a mode will act against this, but, when operating well above threshold, this effect is small. A detailed analysis of this process will be published later.

Fig. 10 shows the Q-switched pulse envelope with a train of self mode locked pulses starting from the peak of the envelope. Measurement of the pulse widths were instrument limited to 1 ns. Sampling methods were not possible because the timing of the pulses is unstable because of amplitude fluctuations in the Q-switched pulse. These mode-locked pulses are different from the sub-pulses that are seen when a few modes are Q-switched. The modulation in the Q-switch envelope in the few moded case is a result of beating between the modes of the detector. The self-mode-locked pulses are distinguished by starting from the peak of the Q-switch envelope and they are much narrower.

9 Laser emission spectra

Further understanding of the self-mode-locking process is gained from the laser emission spectra in the cw, Q-switched and self mode-locked operation shown in Figs. 11 and 12. The mode spacing of a fibre laser with 8 m
cavity length is 12.5 MHzs, which is not resolved by the monochromator. The cw spectrum consists of a series of narrow spikes separated by 0.3 nm. This corresponds to a 0.7 mm cavity and is probably caused by an etalon effect in the laser mirrors. When the cavity is Q-switched with an acousto-optic modulator the mirror etalon effect is disrupted to produce a smooth laser emission spectrum with a line width of 1 nm (fwhm).

Fig. 12 Emission spectra of samarium fibre laser operated Q-switched and Q-switched, self mode-locked

As the laser is driven harder and the intercavity intensity increased, the laser begins to self mode-lock. The emission spectra in Fig. 12 shows that power is coupled from the centre of the lasing spectrum to the weaker modes in the wings, therefore broadening the laser spectrum. The result of coupling power to the modes in the wings is to narrow the pulses of the laser.

10 Conclusions

Samarium doped silica glass is a very unusual laser material. Its narrow linewidth and long fluorescence decay time enable laser action to be obtained in the single-mode fibre geometry. This narrow line is shown to be well shielded from the effects of temperature and external electric fields. Continuous visible emission from a glass laser has never been achieved before so this laser may find many applications. The fluorescence spectrum was seen to vary considerably with different host glass compositions from which fibres were made. This leads to the possibility of laser emission over a wide range of wavelengths from 570 nm to 750 nm. Currently the laser has to be pumped in the blue-green region of the spectrum but codoping may provide a means of pumping at longer wavelengths.

The samarium fibre laser is a useful pulsed source. When Q-switched, 10 W pulses were obtained but peak powers over 100 W are believed to be possible with optimised fibre design. Self-mode-locking provides much shorter pulses than Q-switching on its own. The samarium fibre laser maintains a high intensity throughout its long cavity length leading to nonlinear coupling of the laser modes. The 2 nm linewidth of samarium should lead to sub picosecond pulses but competing nonlinearities and fibre dispersion will frustrate this process.

11 References