Chalcogenide Glass Microsphere Laser

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Abstract: Laser action has been demonstrated in chalcogenide glass microsphere. A sub millimeter neodymium-doped gallium lanthanum sulphide glass sphere was pumped at 808 nm with a laser diode and single and multimode laser action demonstrated at wavelengths between 1075 and 1086 nm. The gallium lanthanum sulphide family of glass offer higher thermal stability compared to other chalcogenide glasses, and this, along with an optimized Q-factor for the microcavity allowed laser action to be achieved. When varying the pump power, changes in the output spectrum suggest nonlinear and/or thermal effects have a strong effect on laser action.

OCIS codes: (140.4780) Optical resonators; (230.5750) Resonators; (160.2750) Glass and other amorphous materials; (140.3530) Lasers, neodymium

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1. Introduction

Spherical particles made from optically transparent materials have the ability to trap light internally by continuous reflections at the surface, effectively forming an optical cavity. The modes supported by such a structure, known as whispering gallery modes, have the ability to confine and store optical energy and as a result there is widespread interest in their application. Microspheres on the order of tens to hundreds of microns in diameter are easily fabricated, either individually or in large quantities by a variety of methods [1,7-11,18-19]. Spheres of this diameter range are compatible with optical fiber and integrated optical circuits and in recent years, microspheres have emerged as an important component in a wide range of photonic devices and find application in signal processing [2], cavity quantum electrodynamics [3] and nonlinear optics [4].

A whispering gallery mode travels around a sphere with virtually no loss apart from the absorption and scattering with the dielectric material and as a result microspheres offer high-Q cavities with very low mode volume. These optical properties lend themselves naturally to laser action and to date, laser oscillation has been demonstrated in spheres formed from a variety of materials including dye-doped polymers [5], crystals [6] and a range of glasses [8-11]. These lasers are characterized by very low thresholds, high pump efficiency and very narrow emission linewidth. Glass spheres in particular offer some of the highest Q-factors, with reports of silica microspheres offering Q-factors as high as 10^9, possibly the highest of any optical resonator [7]. To date, laser action has been reported for microspheres made of silica [8], phosphate [9], fluoride [10] and tellurite [11] glasses, in most cases doped with a rare earth ion. In addition, Spillane reported a Raman laser based on a silica microsphere formed on the tip of a standard optical fiber [4].

In this paper we report on the laser performance of a rare earth doped chalcogenide glass microsphere. Chalcogenide glasses are interesting materials for laser production because of their low phonon energy and infrared transparency. These properties allow fluorescence with higher efficiencies and at longer wavelengths than in other rare earth doped glass [12] and could result in a new generation of solid state mid infrared lasers. There has been considerable work on chalcogenide glasses based on gallium lanthanum sulphide (GLS) as a laser and amplifying medium [12-15]. Compared to other chalcogenides GLS lends itself to active applications because of its excellent rare earth solubility [16]. Gallium and rare earth sulphide together are excellent glass formers and form an environmentally stable, yellow/orange glass with transmission from 0.5 to 10 microns [17]. The refractive index of these glasses is approximately 2.5 at 500 nm wavelength; however the exact index will depend on the dopant and ratio of gallium to rare earth. Fluorescent properties of rare earth doped GLS glasses have been extensively studied by Schweizer [12] who identified 27 transitions between 2 and 5 microns, 7 of which had never previously been seen in a glass host. Table 1 compares the spectroscopic properties of a series of Nd^{3+}-doped glasses. As can be seen, the emission cross sections are larger and the radiative lifetimes shorter than those reported for other glass hosts making this material a good candidate for laser applications [16].

Table 1. Spectroscopic properties of selected Nd^{3+}- doped glasses.

Glass Host	*Wavelength (nm)*	*Stimulated emission cross section (pm^2)*	*Radiative lifetime (μs)*
Silicate	1057 – 1088	0.9 – 3.6	170 – 1090
Phosphate	1052 – 1057	2.0 – 4.8	280 – 530
Fluorzirconate	1049	2.9 – 3.0	430 – 450
Fluorphosphate	1049 – 1056	2.2 – 4.3	310 - 570
GLS	1075 - 1077	7.9	100

[a]adapted from Ceramic Bulletin **69** 1977-1984 (1990)

Thermal analysis of GLS glasses reveals a glass transition temperature of approximately 520^{o}C while crystallization behavior reveals a single exothermic peak indicating the crystallation of a single phase at approximately 740^{o}C. Again these thermal properties will vary slightly with composition. Noteworthy is the relatively high characteristic temperatures, approximately 200^{o}C higher than more well known arsenic or germanium based chalcogenides. Further details on the properties of these and other chalcogenide glasses are summarized in reference [17].

The first reported chalcogenide lasers by Scheiwzer [14] and later Mairaj [15] exploited optical fibre and optical waveguide cavities formed in GLS glass. With the achievement of GLS microspheres by Elliott in 2007 [18] we set out to demonstrate laser action from an Nd^{3+} doped GLS sphere.

2. Experimental

GLS microsphere fabrication is described in detail in reference [19]. In our experiments, GLS glass with a composition of 70 mol% gallium sulphide and 30 mol% lanthanum sulphide, doped with 1.5 mol% neodymium sulphide was used. Microspheres were collected, sorted while suspended in methanol and stored in vials of methanol until ready to be characterized (Fig. 1.).

Typical microsphere diameters range from 30 to 300 micron, though spheres as small as 1-5 microns have been selected and individually manipulated. A typical fabrication run would yield several hundred spheres of good quality and close to the target diameter. From this collection, the best spheres were selected by inspection through an optical microscope. Among these, several representative spheres had their quality quantified through Q-factor measurements, as described in [18].

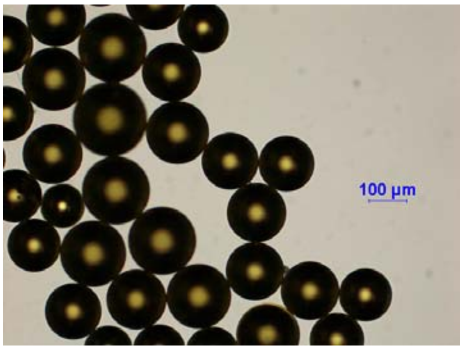

Fig. 1. A selection of Nd^{3+}-doped GLS microspheres as observed under an optical microscope.

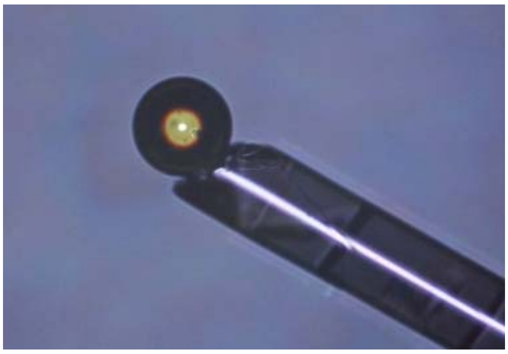

Fig 2. Photograph of microsphere attached to the angle cleaved end of a graded index fibre.

There are a variety of methods of coupling to and from a microsphere and these are described in references [8-11, 20]. We found that free space pumping of the sphere was most convenient in our initial trials to achieve laser action, though this had the disadvantage that only the incident light on the sphere could be accurately measured and the true absorbed power and hence laser thresholds were not obtained. Using an optical fiber to collect the modal pattern allowed the collection of low level of light from a fluorescing or lasing sphere, which resulted in a much lower noise spectrum.

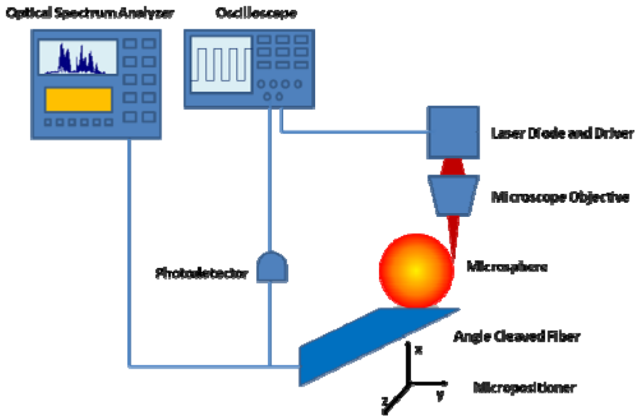

Fig. 3. Schematic of the experimental apparatus used to optically pump the microsphere and detect emission.

A graded index fiber (GIF625) was cleaved at an angle and a pre-selected Nd:GLS microsphere typically 100µm in diameter was glued to its face with superglue (Cyanoacrylate), see Fig 2. The fibre and sphere were mounted on a positioning stage and pumped with an 808nm laser diode. The laser diode was pulsed at 2.18k Hz using a signal generator and the light from it was focused on to the microsphere using a 40x objective. The fiber served two purposes, first to allow precise positioning of the sphere relative to the laser pump source and second to collect a portion of the light confined at the surface of the sphere as a whispering gallery mode. The light coupled into the fibre was split using a conventional fibre splitter with one part going to an optical spectrum analyser (Yokogawa AQ6370), while the other was passed through an 850nm long pass filter (Thorlabs FEL0850) to a high speed detector (New Focus 2053). The output from the detector was then passed to an oscilloscope (Tecktronix TDS 2022). This configuration is shown schematically in Fig. 3.

3. Results and discussion

The microspheres used in these experiments were fabricated in a typical run, which produced a large number of spheres which were sorted to a collection of spheres predominately 90 to 105 microns in diameter. Q-factors of representative spheres were measured as described in [18] and values were typically in the range 10^4 – 10^5 although spheres outside this range were also obtained. Poorer quality spheres were easily identified under an optical microscope and could suffer damage during fabrication and sorting. The highest quality spheres were difficult to identify it is conceivable that higher Q-factors than those measured were obtained and that the handling of spheres during characterization effected their quality.

Of the many rare-earth ions that can provide fluorescence, neodymium provides an ideal four-level laser system in the $^4F_{3/2}$ - $^4I_{11/2}$ transition which can be conveniently pumped with a laser diode around 810 nm. The fluorescence spectrum for this transition, obtained from a 1.5 mol% Nd^{3+}-doped GLS microsphere pumped below threshold is shown in fig 4. During initial laser trials, a number of spheres were tested. On occasion spheres would demonstrate rapid thermal decomposition and essentially “explode”, leaving no trace. We now believe these spheres represented those with the highest Q-factors amongst those we fabricated and the energy storage was sufficiently high to rapidly and catastrophically melt the sphere. Others spheres on the other hand, could withstand pump powers as high as 220 mW at 808 nm, which is the power limit of our pump source, but not demonstrate modal structure seen in Fig. 4. This preliminary screening helped identify those spheres with optimum Q’s for lasing. In the results which follow, we present and discuss the results obtained from a single sphere with the diameter and doping concentration described above.

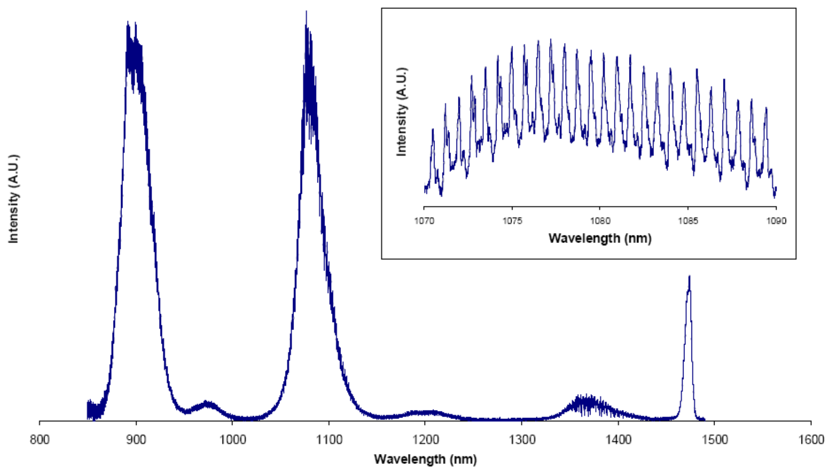

Fig 4. Fluorescence spectrum from a GLS microsphere doped with 1.5 mol% Nd_2S_3. The inset shows the region between 1070 and 1090 nm, the region of interest in our laser experiments and the clear spectrum of whispering gallery modes supported by the sphere.

Using the apparatus described above it was found that a Nd:GLS microsphere could achieve laser action. A threshold was observed at 82mW of incident pump power for a 1082 nm laser line after which sharp spectral peak with a full width at half maximum of <0.05nm emerged. As the pump power was increased the laser wavelength moved to longer wavelength (red-shifted) and above 100mW incident pump power the microsphere laser became multimode (see Fig. 5.). As the pump power increased further the laser became increasingly multimode and continued to red-shift. The Nd:GLS fluorescence spectrum has two peaks (~1077 and 1082 nm), but they are not usually very distinct. However they are exaggerated by laser action and show up as two distinct regions of laser action.

Between threshold and the onset of multimode action, the power output increased linearly with a slope efficiency of 1.6×10^{-5} %, but when the laser became multimode there was no consistent pattern to the power output for the laser peaks. However the position of the laser peaks did move linearly with the increasing pump power and this shift was the same for all modes. When the incident pump power was increased from 82 mW to 220 mW, the maximum pump power we used, the total wavelength shift was 2.5nm. It was also noted that the chop frequency at which the laser was pumped had an effect on the spectral content, but this has not yet been studied.

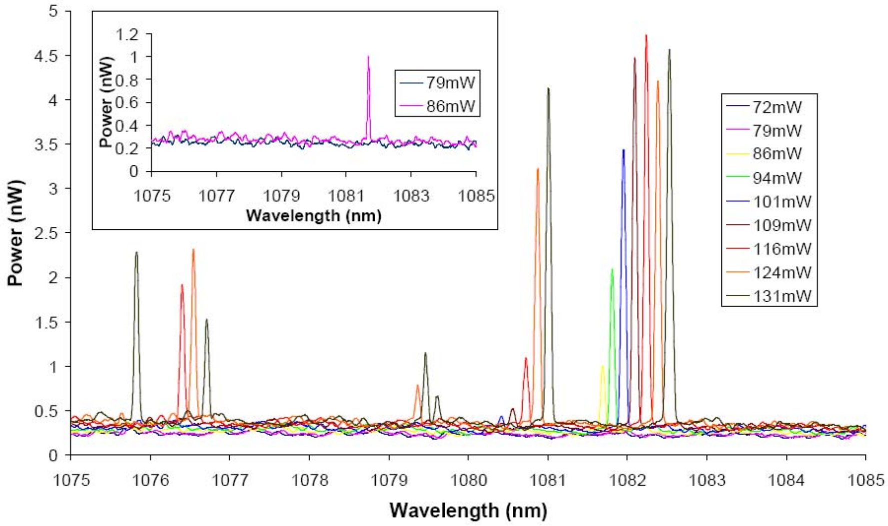

Fig. 5. Laser peaks, showing the change of mode, movement of peaks and the increase of laser output with increased pump power. Inset shows the power output from the microsphere as the laser threshold (incident power) is crossed.

Tellurite glass has a high refractive index similar to GLS and it has been used to make Nd^{3+}-doped microsphere lasers that were also pumped by free space coupling. Sasagawa et al. [21] reported an incident pump power threshold of 81mW with a coupling system that also utilized a microscope objective to couple. This is very similar to our threshold of 82mW which also coupled the pump with a microscope objective.

It has been noted in a previous study [11] that as pump power is increased the resonant peaks move continuously to longer wavelengths while new shorter wavelengths also start to laze. This has also been found to be the case here where individual modes move to longer wavelengths as pump power increases and there is a tendency for new modes to appear at shorter wavelengths, as shown in Fig. 6. There does not however appear to be any simple relationship that determines how the power is distributed amongst these modes. Peng et al ascribe the individual modal shift to thermal expansion of the sphere and thermal refractive index change, but do not make mention of third order nonlinearity. In a glass such as GLS nonlinearity is very high [18] and is likely to have some impact on laser action. However thermal and third order nonlinear effects would both cause the resonant wavelength to become longer, which makes it difficult to tell these effects apart. Regardless of which effect dominates, both are likely to play a role and this could be used to control the operation of the laser. The use of thermal control is widespread, but the use of nonlinear control is not. A form of nonlinear control has been demonstrated in chalcogenide photonic crystals [22], in which the structure was tuned with an external laser. The ability to control the wavelength of a laser could be of significant use in optical circuits, particular the low threshold, narrow line width lasers that microsphere geometries offer.

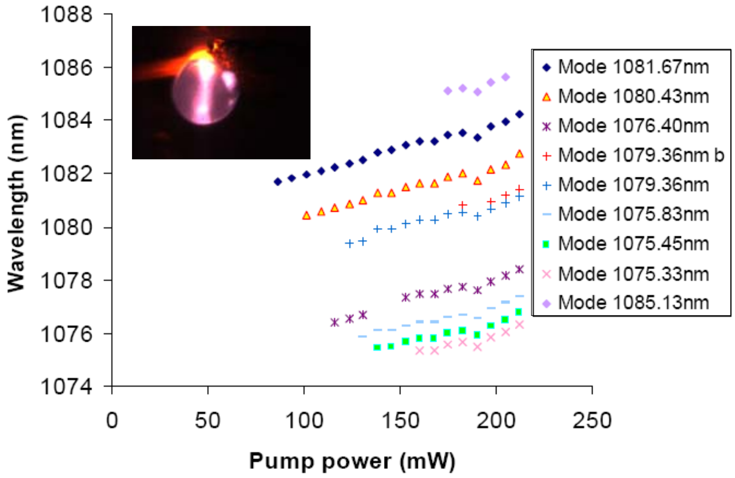

Fig. 6. Wavelength of laser peaks and their shifts as pump power increases. Laser modes move up to 2.5 nm. Inset shows a CCD camera image of a lasing sphere.

4. Concluding remarks

We have fabricated chalcogenide glass microspheres from Nd^{3+}-doped gallium lanthanum sulphide glass and demonstrated laser action at wavelengths between 1075 and 1086 nm. The spheres had Q-factors on the order of 10^4; a value which we believe is close to optimum for a chalcogenide microsphere laser. Experimental evidence suggests higher Q's could lead to thermal instability. The relatively high thermal stability of GLS glass compared to other chalcogenides allowed incident pump powers at 808 nm in excess of 200 mW. This is considerably higher than that reported for other chalcogenide microsphere [21] and this inherent thermal stability along with the excellent rare earth solubility make GLS an excellent candidate for solid state lasers, particularly in the mid-infrared. Further studies are now underway to fully characterize the laser performance and explore methods for efficient coupling and packaging of the laser device.

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