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# Enhancing extraction efficiency of mid-infrared fluorescence in chalcogenide glass via photonic crystal



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#### ARTICLE INFO

#### ABSTRACT

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#### 1. Introduction

A 2-5 µm midinfrared (mid-IR) light source covers the important atmospheric windows and molecular fingerprints of numerous gases, liquids, and solids; thus, it exhibits potential applications in laser communications, biomedical sensing, and environmental monitoring [1]. The use of rare earth (RE)-doped chalcogenide glasses is a suitable way to obtain mid-IR fluorescence. These glasses offer favorable properties for mid-IR emission, such as high refractive indices, high absorption and emission cross sections, and generally low phonon energies for efficient radiative processes of doped RE ions (REIs). Furthermore, chalcogenide glasses are chemically durable in liquid water and open atmosphere [2]. They are also mechanically robust. Various REIs, such as  $Er^{3+}$ ,  $Dy^{3+}$ ,  $Ho^{3+}$ ,  $Py^{3+}$ , and  $Tm^{3+}$ , have been introduced into chalcogenide glasses, and a mid-IR fluorescence covering from 1.5  $\mu$ m to 4.9  $\mu$ m has been successfully obtained [3–8]. However, chalcogenide glasses have not yet been developed as mid-IR REIdoped glass fiber lasers because of the relatively high losses and low doping concentration. Moreover, mid-IR fiber lasers have not been available for use beyond 3 µm wavelength thus far, even if the fluorescence emission in this range is very weak. Pursuing effective ways to enhance IR emission in REI-doped chalcogenide glasses is a challenge. Various techniques and skills have been proposed to enhance fluorescence emission, including the use of microcrystalline [9], doping metal particles, halogen [3,10,11], or

The use of rare earth-doped chalcogenide glass is an attractive method to develop mid-infrared sources. In this work,  $Er^{3+}$ -doped chalcogenide glass is prepared, and photonic crystal (PC) pattern is designed to improve the extraction efficiency of light emission from the sample surface. The finite difference time domain simulation shows that the light extraction efficiency from the sample surface can be 1.62 times stronger than that from the sample without PC structure by introducing a simple two-dimensional (2D) PC structure into glass samples. This improvement was the result of the efficient light diffraction on the surface because of the integrated 2D PC. Results in this work offer a potential in developing midinfrared light sources.

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more than one kind of REI in a glass [12,13]. These methods are mainly based on crystal field effects, plasmon resonances, or energy level modification. In addition to these methods, fluorescence emission can be further enhanced by improving the extraction efficiency of the emission from a sample surface. Chalcogenide glasses are known to have high refractive index [14], which is beneficial to photonic devices. However, given a high refractive index, only a small fraction of the photons generated inside the bulk glass can escape because of the total internal reflection at the interface. A rough estimation of a single interface for the photons generated inside a GaN LED device leads to an extraction efficiency of approximately  $\eta \sim 1/4n^2 \sim 4\%$  [15]. The trapped energy can be released by destroying the internal reflection via surface roughening [16,17], surface plasmon resonance [18], or integration of two-dimensional (2D) photonic crystal (PC) patterns [19-22]. Thus, using 2D PC patterns on the device surface may be a fascinating technique to enhance the light emission from REI-doped chalcogenide glass because of its simplicity and effectiveness on light outcoupling.

PC is an artificial optical material in which refractive indices are distributed periodically [23]. The two main features of a PC are photon localization and photonic band gap [24,25]. Although, PC has been initially proposed to inhibit spontaneous emission, researchers in the subsequent studies determined that it not only can control light propagation but can be also used to enhance spontaneous emissions. Previous studies showed that a photonic band gap can compress the photon modes within the band gap, and the excitation of the luminescent materials at band edges or defect modes can be greatly enhanced [26]. On the contrary, the periodically modulated PC structures can significantly reduce the

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reflected energy at the surface of a sample surface, which cannot be ignored for materials with high refractive indices, such as chalcogenide glasses. In addition to direct emission, fluorescence can now use Bragg scattering on the structure, thereby greatly reducing the amount of light trapped by the total internal reflection compared with that in an unpatterned substrate. Light extraction efficiency can be enhanced by 50% with a simple submicrometer-sized PC structure in OLEDs [27–31]. However, reports on improving mid-IR light extraction efficiency by fabricating PC arrays on REI-doped chalcogenide glasses is rare.

In this work, we performed a theoretical study of the relationship between the extraction efficiency of the mid-IR fluorescence from chalcogenide glasses via PCs and their lattice structures. An Er<sup>3+</sup>-doped Ga<sub>5</sub>Ge<sub>20</sub>Sb<sub>10</sub>S<sub>65</sub> glass was selected as the basal material, and the PC structures were introduced into the sample to improve the extraction efficiency of mid-IR fluorescence. A chalcogenide glass was fabricated with melt-quenching route method. Moreover, the refractive index, absorption spectrum, and emission spectra were measured, which exhibited good fluorescence emission properties with the peak at approximately 2.74 µm. Given the measured optical parameters, the PC structures were proposed to enhance the extraction efficiency of fluorescence emission from the sample. The extraction efficiency from the sample with different lattice constants ( $\Lambda$ ) and air hole radius (r) was investigated with finite difference time domain (FDTD) method [32,33]. The extraction efficiency of mid-IR fluorescence from the sample surface can be 1.62 times higher than that from the sample without PC structures by optimizing the designed structure. The fluorescence emission of the sample also exhibits high directionality.

#### 2. Theory

Fig. 1(a) shows the schematic of the chalcogenide glass samples investigated in this study. The light is assumed to be emitted from the excitons, which are embedded in the  $Er^{3+}$ -doped glasses. The PC structure is created at the surface of the sample. For the sample without a PC structure, a fraction of the light cannot escape the sample because of total internal reflection. The light, which occurs at an excessively oblique angle, is not transmitted, as shown in Fig. 1(b). The extracted light intensity from a point source in the sample glass to the air can be described with Lambertian emission equation, which is expressed as follows [15,34]:

$$I_A = \alpha I_G \cos \theta_A,\tag{1}$$

where  $I_G = \frac{P_{\text{source}}}{4\pi r^2}$  presents the light intensity in the RE-doped chalcogenide glass at a distance *r* from the source;  $\alpha = (\frac{n_A}{n_G})^2$ , where  $n_A$  and  $n_G$  are the refractive index of air and glass; angle  $\theta_A$  is the escape angle into the air from the glass, as measured from the normal to the surface.



Fig. 1. Schematic depicting the sample structure and computational model.

Snell's law can be used when  $\theta_A$  is expressed in terms of angle  $\theta_G$ , which is the angle of light incidence from the source in the glass sample to the glass/air interface. This law can be described with the following equation:

$$I_A = \alpha I_G \sqrt{1 - \sin^2 \theta_G / \alpha} \tag{2}$$

In our case, when the periodically modulated PC structures are introduced to the sample surface, high-order diffractive beams exist in addition to the original incident direction, as shown in Fig. 1(c). These beams are allowed to propagate from the sample. Thus, the transmitted energy is increased, and the amount of the light trapped by total internal reflection is greatly reduced compared with those of an unpatterned substrate. The extra emission direction  $\theta_A$  can be obtained with parallel momentum conservation law, which is expressed as follows [35,36]:

$$\frac{2n_G\pi}{\lambda}\sin\theta_G + G_{||} = \frac{2n_A\pi}{\lambda}\sin\theta_A, \ r/\Lambda$$
(3)

where  $G_{//}$  is the parallel component of any reciprocal lattice vector G. Given the contribution of the lattice vector G, a considerable number of photon modes are extracted, leading to a significant enhancement of the fluorescence emission.

#### 3. Experimental and simulation result

#### 3.1. Glass preparation

Chalcogenide glasses with the composition of  $Ga_5Ge_{20}Sb_{10}S_{65}$ doped with 1 wt%  $Er^{3+}$  were prepared via conventional meltquenching route. High-purity elements Ge, Ga, Sb, S(5N), and  $Er_2S_3$ (3N) were weighed and vacuum sealed in a silica tube under a high vacuum level ( $10^{-3}$  Pa). Subsequently, the tube was placed into a rocking furnace and melted at approximately 980° for 12 h to ensure the homogenization of mixtures. The tubes were quickly immersed in water for quenching and then annealed for several hours before they were cooled slowly down to room temperature. Finally, the specimens were cut into 2 mm disks and polished for optical characterization.

#### 3.2. Sample measurements

All of the optical tests were performed at room temperature. The refractive index was measured based on autocollimation method, with a value of 2.25 at the wavelength of 2740 nm. The absorption spectrum of the glass sample was recorded with a Perkin Elmer Lambda 950 UV-vis-NIR spectrophotometer in the wavelength range of 600-1700 nm. A tunable titanium:sapphire laser (Coherent Mira 900-D) at 980 nm with a power of 500 mW was used as the excitation source to obtain efficient fluorescence emission. The mid-IR fluorescence spectra in the range of 2600-3000 nm were measured through a computer-controlled system, which consisted of a monochromator (Zolix Omni- $\lambda$ 3015) with a lock-in amplifier (SCITEC Model420) and an InSb detector (DInSb 55-De) cooled with liquid nitrogen. The measured absorption spectra of glass samples in the wavelength region of 600-1700 nm are shown in Fig. 2(a). Four visible absorption peaks centered near 657, 806, 983, and 1535 nm were observed, which could be attributed to the  ${}^{4}I_{15/2} \rightarrow {}^{4}F_{9/2}$ ,  ${}^{4}I_{15/2} \rightarrow {}^{4}_{19/2}$ ,  ${}^{4}I_{15/2} \rightarrow {}^{4}I_{11/2}$ , and  ${}^{4}I_{15/2} \rightarrow {}^{4}I_{13/2}$  transitions, respectively. The electronic absorption edge of the host glass started from 600 nm, and the higher energy levels of the REIs were obscured. Fig. 2(b) displays the measured mid-IR (MIR) emission spectra for  $Er^{3+}$  in the glass samples when pumped at 980 nm. The Er<sup>3+</sup> fluorescence spectrum exhibits a major band with peak at 2.74 µm, which can be attributed to the

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