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Mid-infrared guided photoluminescence from integrated Pr^{3+}-doped selenide ridge waveguides

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ABSTRACT

Praseodymium-doped selenide thin films are deposited by radio frequency magnetron sputtering on thermally oxidized silicon wafers and undoped selenide layers. Ridge waveguides are then processed using photolithography and dry etching techniques. Under optical pumping at 1.55 μm, broadband guided mid-infrared photoluminescence is recorded for the first time for wavelengths above 4 μm from rare earth-doped integrated chalcogenides waveguides. Optical design confirmed that these active waveguides allow single-mode optical propagation at a wavelength of 4.70 μm.

1. Introduction

There is a growing interest for broadband light sources and amplifiers operating in the mid-infrared (mid-IR) and especially in the 3–5 μm spectral window. Supercontinuum generation in the mid-IR may benefit from high power laser emitting around 4 μm [1,2]. Furthermore, since many chemical molecules such as hydrocarbons, carbon dioxide and carbon monoxide display fundamental vibrational absorption lines in this spectral region, mid-IR light sources may also find applications in medical diagnostics, chemical sensing [3] or environmental monitoring [4].

Several options exist for mid-IR light sources devoted to sensing applications amongst which synchrotron radiation, global or optical parametric oscillator [5]. Mid-IR photonics advances have been driven by the development of room-temperature quantum cascade lasers (QCL) and interband cascade lasers which have extended the operating range of tunable semiconductor lasers towards mid-IR [6]. Single DFB QCL provide discrete laser sources spanning the entire mid-IR but have limited tuning range whereas external cavities QCL are widely tunable but can suffer from mechanical vibrations which could impede their use in harsh environments. Mid-IR lasing above 2.3 μm has also been achieved using transition metal-doped II-VI compounds [7] and rare earth (Ho^{3+}, Er^{3+}, Dy^{3+})-doped fluoride fibers [8]. Supercontinuum generation spanning from 1.4 to 13.3 μm has been recently obtained based on nonlinear effects in chalcogenide step-index fibers [2]. However, to sustain the steadily increasing demand for more compact and cost-effective sensing, on-chip integration of a broadband mid-IR light source is required.

Rare earth (RE)-doped chalcogenides glasses are promising candidates to implement mid-IR lasers and amplifiers. Compared to oxide or fluoride glasses, their lower phonon energies enable broad mid-IR transparency and long-wavelength radiative transitions that are generally quenched due to large multi-phonon relaxation rates between RE energy levels separated by a small energy gap [9]. These glasses exhibit also large refractive indices that will allow

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dense photonic integration [10] and enhance the local field factor and therefore RE ion absorption and emission cross-sections [11]. Mid-IR emission from several RE ions, including Dy^{3+}, Tm^{3+}, Tb^{3+}, Ho^{3+} or Er^{3+} have been demonstrated in both bulk glass and chalcogenides fibers [12–15]. Lately, photoluminescence (PL) at 2.8 μm from Er^{3+}-doped sulphide films and integrated waveguides was also reported [16,17]. Numerous experimental studies have also been focused on mid-IR (3.5–5.5 μm) spectroscopic properties of Pr^{3+} ions in various glass hosts (AsGeGaSe [18,19], AsGeInSe [20], BaGaGeInSe [21], or GaGeSbS(Se) [22,23]). Praseodymium ion, whose simplified energy level structure is reported in Fig. 1(a), has been shown to display broad emission bands in the mid-IR and to allow convenient pumping around 1.5 and 2 μm by semiconductor diodes and Tm^{3+}/Er^{3+}-based fibers or solid state lasers. Numerical investigations of mid-IR PL from Pr^{3+}-doped chalcogenide-based fibers [24,25] and integrated micro-disks [26] also highlighted the potential of these materials to implement mid-IR lasers or amplifiers.

Ge-Sb-S(Se) glasses have good thermal and chemical stability, while adding Ga helps increasing RE solubility and dispersion [27,28]. Substituting sulphur atoms by selenium ones in the Ga-Ge-Sb-S(Se) system produces films with extended transmission towards the mid-IR. These selenide films exhibit lower non-radiative multiphonon transition rates due to lower phonon energies (150-300 cm^{-1} and 300-450 cm^{-1} for selenides and sulphides, respectively [29]) and higher refractive indices compared to sulphide films. Despite lower deposition rates, sputtering technique may provide improved morphology, topography and better respect of target composition than pulsed laser deposition or thermal evaporation techniques in case of multi-element composition.

In this paper, Pr^{3+}-doped selenide straight ridge waveguides are fabricated by radio frequency (RF) magnetron sputtering with subsequent patterning using photolithographic and dry etching techniques. Optical simulations are performed to design single-mode ridge waveguides at the mid-IR propagation wavelength of 4.7 μm. Photoluminescence experiments enabled the first recording of broadband mid-IR emission above 4 μm from single-mode RE^{3+}-doped integrated chalcogenide waveguides.

2. Experimental

Praseodymium-doped (5000 ppm Pr^{3+}) and undoped selenide glass targets with respective nominal compositions of $Ga_5Ge_{20}Sb_{10}Se_{65}$ and $Ge_{28.1}Sb_{6.3}Se_{65.6}$ were prepared by means of conventional melt and quenching method. High purity elements (3N for Pr_2S_3, 5–7N for Se, Sb, Ge and Ga) were weighted in a dry glove box, introduced in a fused silica ampoule and pumped under high vacuum. To ensure melt homogenization, sealed tubes were heated in a rocking furnace at 850 °C for 12 h. After quenching in water (cooling rate of 100 °C s^{-1}), the glass rods were annealed below the glass transition temperature [30]. The rods were finally cut and polished to prepare 3-mm thick glass targets with diameter of 2". Praseodymium-doped films were deposited by RF-magnetron sputtering on both thermally oxidized (4-μm thick SiO_2) 2″ silicon wafer and undoped selenide layer (5-μm thick $Ge_{28.1}Sb_{6.3}Se_{65.6}$) sputtered on 2″ silicon wafer with a thickness of 1.7 μm and 2 μm, respectively. The deposition parameters (argon pressure and flow, RF power and target-to-substrate distance) were selected in relation with morphology, topography and growth rate of the films. Under optimized conditions, a deposition rate around 25 nm min^{-1} and a RMS roughness of 0.47 nm measured by Atomic Force Microscopy (AFM, Nanoscope III) were achieved. The morphology and chemical composition of the films were analyzed using a scanning electron microscope coupled to an energy-dispersive X-Ray system (JEOL JSM 6400-Oxford Link Inca) showing a close correspondence to that of the glass targets (4.8% Ga, 21.6% Ge, 10.8% Sb, 62.8% Se for $Ga_5Ge_{20}Sb_{10}Se_{65}$ and 28.3% Ge, 6.8% Sb, 64.9% Se for $Ge_{28.1}Sb_{6.3}Se_{65.6}$ [31]). Linear refractive indices of chalcogenide thin films were obtained from the analysis of variable angle spectroscopic ellipsometry (VASE) data measured using two ellipsometers: a rotating analyzer ellipsometer measuring in UV–Vis–NIR (300–2300 nm) and a rotating compensator ellipsometer working in mid-IR (1.7–30 μm) (J.A. Woollam Co., Inc., Lincoln, NE, USA) [30,32].

Ridge waveguides of different widths (ranging from 1.5 to 10 μm) were then fabricated using a classical i-line photolithographic process followed by a dry etching procedure. The positive photoresist (Microposit S1805) was exposed through a Cr mask using a MJB4 Suss Microtech mask aligner and subsequently dissolved in a MIF319 developer. The pattern was then transferred to the doped selenide film by a fluorine-based inductively coupled plasma-reactive ion etching (ICP-RIE) process (Corial 200IL). An etching rate of 165 nm min^{-1} was measured by in situ reflectrometry and later confirmed by SEM imaging of the chalcogenide films (Fig. 1(b) and (c)).

Guided PL experiments were performed at room temperature on 5-mm long ridge waveguides of different widths using a continuous 1.55 μm laser in co-propagative configuration. The pump laser was amplified using an erbium-doped fiber amplifier and coupled to the waveguides using a microlensed fiber. A fiber variable attenuator allows power dependent PL measurements. Mid-IR emission from Pr^{3+} was collected from the waveguide output facet using a ZnSe microscope objective and focused on the entrance slit of a monochromator (Horiba iHR320) equipped with a

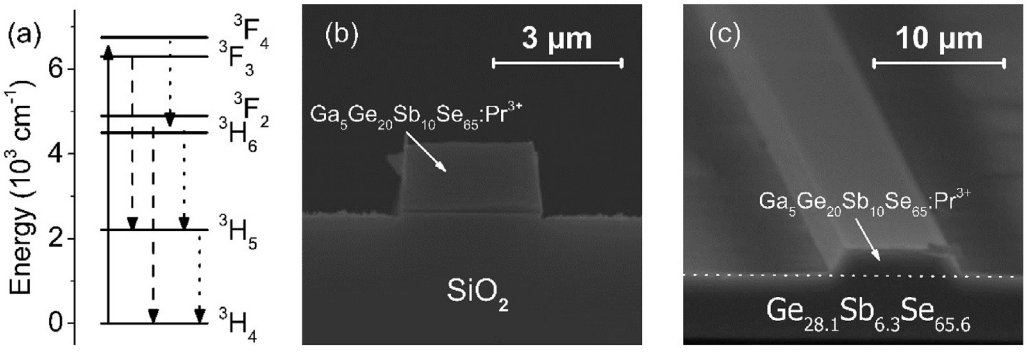

Fig. 1. (a) Simplified low energy part of the energy level diagram of Pr^{3+} ions with excitation and emission transitions investigated. Emission between 2-2.8 μm and 3.5–5.5 μm are represented by dashed and dotted lines, respectively. Scanning electron microscope image of the processed waveguide after ICP-RIE of Pr^{3+}-doped $Ga_5Ge_{20}Sb_{10}Se_{65}$ ridge waveguides on (b) SiO_2 or (c) $Ge_{28.1}Sb_{6.3}Se_{65.6}$.