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# Mid-infrared second-harmonic generation in chalcogenide photonic crystal fiber



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## 1. Introduction

Mid-infrared (mid-IR) light sources have been among the most intensively developed photonic devices in recent years [1–3]. The sources in this range are useful for many important applications, such as spectroscopy, medical treatment, environmental protection, remote sensing, and military applications [4–6]. Various technologies, including those that use antimony compound bandgap semiconductor materials, IR crystal nonlinear frequency conversion technology, optical parametric oscillator technology, and rare-earth-doped low phonon energy glass fiber pumped by solid-state laser, were developed to exploit the mid-IR region effectively [7–9].

Optical frequency conversion by second-harmonic generation (SHG) is a simple method used to extend the spectral range of laser sources and all-optical wavelength multiplexing [10,11]. Compared with nonlinear crystals, optical fibers exhibit several excellent properties, such as low cost, long interaction length, high transparency, and high optical damage threshold. SHG fibers have received considerable attention since first being observed in GeO<sub>2</sub>-SiO<sub>2</sub> glass fiber in the 1980s [12–14].

Over the past decades, various poling techniques have been successfully used to induce SHG in glass fibers. These techniques include the thermal/electric poling [15–18], electron beam irradiation [19–22], laser-induction [14,23], and so on. In terms of the generation mechanism of the SHG effect in glasses, scientists propose different views for different poling methods, such that no unified understanding on the matter exists. Among the various

# ABSTRACT

This study theoretically investigated a feasible modal phase-matching second-harmonic generation (SHG) in chalcogenide photonic crystal fiber. SHG is a technique that enables the acquisition of the mid-infrared resource at 5.3  $\mu$ m with the use of a 10.6  $\mu$ m commercial laser. The phase-matching condition between two low-order modes can be realized simply by modifying the air hole size and lattice pitch in the designed photonic crystal fiber. Numerical results showed that the confinement loss of the second-harmonic wave was 4.15  $\times$  10<sup>-4</sup> dB/m at 5.3  $\mu$ m, and the effective phase-matched fiber length reached as long as 6.382 m.

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poling methods, the thermal/electric poling generating nonlinearity is considered by many researchers mainly because of two mechanisms that are periodic at a microscopic scale. The first mechanism is related to the reorientations of polar bonds or hyperpolarizable entities during poling treatment; such reorientations induce macroscopic second-order nonlinearity [16]. The second mechanism is related to the migration of ionic species under the applied electric field. Such migration facilitates the generation of a permanent electric field, E<sub>dc</sub>. The coupling of E<sub>dc</sub> and the third-order susceptibility ( $\chi^{(3)}$ ) of the glass produce an effective  $\chi^{(2)}$  through equation  $\chi^{(2)} = \chi^{(3)}E_{dc}$ . Thus, glasses with large  $\chi^{(3)}$  are conducive for achieving good second-order nonlinearities.

The nonlinear coefficient  $n_2$  of chalcogenide glasses (ChGs) are between 2 and  $20 \times 10^{-18}$  m<sup>2</sup>/W, which is 100–1000 times larger than that of silica glasses [24,25]. This condition implies that ChGs have great potential for high SHG. The generation of second-order nonlinearities in ChGs is of great interest for their interesting properties, including large IR spectral range of transparency (up to 10 or 20 µm depending on the composition), low phonon energy, high linear and nonlinear refractive indices, and the possibility to perform optical fibers. Guignard et al. [16,26] observed SHG in ChGs by thermal poling, and they obtained a second-order susceptibility  $\chi^{(2)}$  as great as (8.0 ± 0.5) pm/V when Ge<sub>25</sub>Sb<sub>10</sub>S<sub>65</sub> glass was poled at 170 °C under (4.0–5.0) kV for 30 min. Therefore, ChG fibers present great potential for the optical frequency conversion in the IR spectral region.

Compared with conventional fibers, photonic crystal fibers (PCF) exhibit a variety of unique properties, such as flexibility in dispersion design [27], high nonlinear coefficient [28], and low confinement loss (CL) [29]. In particular, the modal phase-matching condition of PCF can be fulfilled simply by tuning the

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fiber structure. Monro et al. [30] successfully accomplished SHG by modifying the air holes size and lattice pitch of their holey fibers.

In our work,  $Ge_{25}Sb_{10}S_{65}$  glass was especially chosen and systematically studied. This glass is a non-arsenic environmentfriendly glass material, which possesses excellent optical properties and good thermal stability for infrared applications [6,26,31– 36]. Researchers in PERFOS and University of Rennes [31,35] have successfully drawn the PCF in this glass, the loss of 3 dB/m at 1.55 µm and 4.5 dB/m at 3.39 µm was obtained.

In this paper, a feasible modal phase-matching SHG was examined in the designed chalcogenide PCF. By optimizing the fiber structure, the phase-matching condition between the two low-order modes was realized in the designed PCF and an efficient mid-IR light resource at  $5.3 \,\mu m$  could be obtained with a 10.6  $\mu m$  laser.

#### 2. Glass synthesis and properties

A Ge<sub>25</sub>Sb<sub>10</sub>S<sub>65</sub> ChG, from which high second-order susceptibility can be obtained after thermal poling, was particularly selected for this work. This ChG glass was prepared by conducting a meltquenching method in silica ampoules under vacuum [22]. The starting materials used were distilled in different tubes, and sulfur was purified through multiple distillations to eliminate water and carbon, which are undesirable for transmitting glasses. After purification, the required amounts of the different elements were placed in the same silica ampoules under vacuum. The ampoules were then melted using a rocking furnace. Subsequently, the ampoules were quenched in ice water to enable glass formation and to avoid any crystallization. Finally, the vitreous sample was annealed at a temperature slightly below the glass transition temperature before being slowly cooled to room temperature.

The transparency spectrum of the prepared glass was measured with a Fourier transform IR spectroscopy spectrophotometer. Fig. 1 displays the transmission curve. The figure reveals that the transmittance of the sample in the range 1 to 10  $\mu$ m is over 70%, except for some absorption bands. This finding confirms the excellent optical quality of photonic devices. The thermal stability of the fabricated glass was then measured. The thermal properties of the glass were studied by performing differential scanning calorimetry (DSC) at 10 °C/min, as shown in Fig. 1(b). The results of the experiment indicated that the glass transition temperature

 $(T_g=294 \ ^{\circ}C)$  could inhibit the reuse of the glass for a temperature range over 200  $\ ^{\circ}C$ . No exothermic peak associated with crystallization up to 450  $\ ^{\circ}C$  was observed, indicating that the fabricated glass is thermally and dynamically stable toward devitrification. A glass with  $\Delta T=T_g-T_x$  (where  $T_x$  is the temperature of crystallization onset) higher than 150  $\ ^{\circ}C$  is suitable for fiber drawing. Therefore, the fabricated  $Ge_{25}Sb_{10}S_{65}$  glass is suitable for PCF applications.

# 3. Design of ChG PCF

For multi-cladding air hole PCF, the mode field is mainly limited to the three innermost rings of the fiber, and every additional ring can significantly reduce the PCF loss [37], but only when the complexity of fiber fabrication and the computation time of the procedures are considered. A PCF with four air hole rings is proposed in this work. The cross-sectional diagram of the designed ChG PCF is shown in Fig. 2, where  $\Lambda$  is the lattice pitch and d is the diameter of air hole size.

### 4. Verification of SHG

An efficient SHG in chalcogenide PCF can be achieved by satisfying the phase-matching condition between two low-order modes. In this research, the guided modes were first analyzed at a wavelength of 10.6  $\mu$ m for the designed PCF with  $\Lambda$ =3  $\mu$ m and  $d/\Lambda$ =0.8. In this work, plane wave expansion (PWE) method







Fig. 1. Transmittance curve (a) and DSC curve (b) of glass Ge<sub>25</sub>Sb<sub>10</sub>S<sub>65</sub>

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