Supplementary Materials

Experimental procedure: Bulk chalcogenide glasses of $(GeSe_2\text{-}3As_2Se_3)_{1-x}PbSe_x$ with 20 mol% and 40 mol% PbSe were prepared by conventional melt-quenching technique. All glasses were prepared using high purity (99.999%) raw materials from Alfa-Aesar: Se, Ge, Sb, and Pb. These elements were weighed and batched in a nitrogen purged MBraun Labmaster 130 glove box. The weighed batches were loaded into cleaned fused quartz tubes and sealed under vacuum using a methane-oxygen torch to form sealed ampoules. The batches were melted in a rocking furnace overnight at a melting temperature of 850 °C. The furnace was then cooled to the quench temperature of 650 °C, prior to removal from the furnace for quenching using compressed air. To minimize the quench-related stress, glasses were annealed at 177 °C for 2 h and cooled to room temperature. The prepared glass rods were removed from the ampoules and cut into slices of thickness ~3 mm using a slow speed saw and subsequently polished on both sides using a polishing pad with 0.05 μm Al_2O_3 slurry. Film chalcogenide glasses of 20 μm thickness of $(GeSe_2\text{-}3As_2Se_3)_{1-x}PbSe_x$ with 40 mol% PbSe were deposited by co-evaporation of target materials at room temperature, using 20 Å/s of $GeSe_2\text{-}3As_2Se_3$ and 8.5 Å/s of PbSe at a growth pressure of 8×10^{-7} torr in the Kurt J. Lesker evaporator. Following the deposition processes, the films underwent a stress-relieving anneal for 2 h at 145 °C under atmospheric pressure with N_2 carrier gas in the furnace CEE 4500. Chalcogenide glasses of 20 μm thickness film of $(GeSe_2\text{-}3As_2Se_3)_{1-x}PbSe_x$ with 40 mol% PbSe were deposited onto 2″ Schott IRG 24 substrate by thermal evaporation [1] of $GeSe_2\text{-}3As_2Se_3$ and PbSe in a custom-designed thermal evaporator (112 Evaporator-Sputter Station from PVD Systems Inc.). The bulk target materials were ground to powder and loaded into two Tantalum baffled source boats, respectively. The film was deposited at a base pressure of 8×10^{-7} Torr and the heating power of the two sources were separately controlled to maintain the deposition rate of 20 Å/s for $GeSe_2\text{-}3As_2Se_3$ and 8.5 Å/s for PbSe. The substrate was maintained at around room temperature throughout the deposition process. Following the deposition processes, the films underwent a stress-relieving anneal for 2 h at 145 °C under atmospheric pressure with N_2 carrier gas in the furnace CEE 4500. To determine the morphology and chemical composition of the glasses, cross-sectional transmission electron microscopy (TEM) specimens were prepared by focused ion beam-assisted milling followed by lift-out processing in a FEI Helios Nanolab 660 focused ion beam. The specimens were mounted on Cu grids and ion polished to ~50 nm thickness for electron transparency. TEM imaging was carried out using a Talos F200X at 200 kV. X-ray energy dispersive spectroscopy (XEDS) data were collected using scanning transmission electron microscopy (STEM) in the Talos F200X at 80 kV to assess any chemical segregation within the glasses. Transmission spectra of the glasses were measured using both a ThermoFisher Nicolet iS5 FTIR spectrometer and a CARY 500 UV-VIS spectrophotometer to cover the full spectral range of the material. Linear refractive indices of the glasses were measured at wavelengths from 2 μm to 10 μm and angles from 69° to 75°, using J.A. Woollam IR-VASE including a Fourier transform infrared interferometer with a rotating compensator ellipsometer where IR light with an 8 mm beam spot was emitted by a Globar. Signals were collected by a deuterated triglycine sulfate detector. Nonlinear refractive indices and multiphoton absorption coefficients of the glasses were measured at a wavelength of 4.515 μm, using closed and open z-scan measurements where ultrashort pulses with a temporal width of 150 fs and a repetition rate of 1 kHz were used. Samples were scanned through the focal point of a plano-convex spherical CaF_2 lens, where an intensity variation with respect to its spatial location exists. The closed Z-scan measurement utilizes an aperture in front of a detector whereas the open Z-scan measurement does not.

1. Hu, J.; Tarasov, V.; Agarwal, A.; Kimerling, L.; Carlie, N.; Petit, L.; Richardson, K. Fabrication and testing of planar chalcogenide waveguide integrated microfluidic sensor. *Opt. Express* **2007**, *15*, 2307–2314.