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## Broadband transparent optical phase change materials

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**Abstract:** We report a new group of optical phase change materials Ge-Sb-Se-Te (GSST) with low loss from telecom bands to LWIR. We further demonstrated GSST-integrated SiN photonics with significantly improved switching performance over conventional GST alloys.

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Optical phase change materials (O-PCMs) are a unique class of materials which exhibit extraordinarily large optical property change (e.g. index change  $\Delta n > 1$ ) when undergoing a solid-state phase transition. These materials, exemplified by Mott insulators such as VO<sub>2</sub> and chalcogenide alloys (ChAs) such as Ge-Sb-Te (GST) compounds, have been exploited for a plethora of emerging optical applications including optical switching, photonic memories, reconfigurable metasurface, and non-volatile display [1-5]. These traditional phase change materials, however, generally suffer from large optical losses even in their dielectric states. For instance, the archetypal ChA phase change material GST is optically absorbing at the telecommunication bands due to its small bandgap and the resulting interband absorption, whereas its crystalline form is plagued by high free carrier absorption (FCA) in the mid-wave and long-wave infrared (LWIR) (Fig. 1a). The large optical losses fundamentally limit the performance of photonic devices based on traditional O-PCMs. We define the optical figure-of-merit (FOM) for O-PCMs as: FOM =  $\Delta n/k$ , where  $\Delta n$  is the index change upon phase transition and k denotes the extinction coefficient. It can be directly shown that this FOM dictates the attainable insertion loss and contrast ratio of tunable optical devices based on O-PCMs [6].



Figure 1: (a) Optical absorption of the classical phase change alloy Ge<sub>2</sub>Sb<sub>2</sub>Tes fitted to show combined contributions from interband transition, Urbach tail, and FCA based on Drude models; (b, c) optical properties of GSST alloys in their (b) amorphous and (c) crystalline states; (d) optical properties of amorphous (dashed lines) and crystalline (solid lines) Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>4</sub>Te<sub>1</sub> from the visible range to LWIR; (e, f) TEM image and SAED patterns of crystallized (e) Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>4</sub>Te<sub>1</sub> and (f) Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>5</sub>.

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Here we report the synthesis, characterization and device integration of a new class of O-PCMs, Ge-Sb-Se-Te (GSST) alloys. A series of GSST thin films with the compositions of Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>x</sub>Te<sub>5-x</sub> (x = 1, 2, 3, 4, and 5) were prepared using thermal evaporation. We experimentally validated that Se substitution of Te results in an increase in optical band gap, enabling low loss operation in the technologically important 1310 nm and 1550 nm telecommunication bands (Figs. 1b and 1c). Meanwhile, the GSST materials claim reduced free carrier concentrations and mobility compared to GST as revealed by Hall measurements, which effectively suppresses FCA in the infrared. As an example, Fig. 1d shows that the Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>4</sub>Te<sub>1</sub> (GSS4T1) material features broadband transparency covering 1 micron to the LWIR. The large index contrast between the amorphous and crystalline states ( $\Delta n = 1.8$ ) and the low optical loss of the GSS4T1 alloy lead to an exceptionally large FOM of > 900 at 1550 nm wavelength, over 20 times larger than that of the classical Ge<sub>2</sub>Sb<sub>2</sub>Te<sub>5</sub> (GST 225) phase change alloy. Figures 1e and 1f present high-resolution TEM images and selected area electron diffraction (SAED) patterns (insets) of the GSS4T1 and Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>5</sub> alloys in their crystalline form. Unlike GST which exhibits an intermediate cubic phase during transition, GSS4T1 directly crystallizes into a hexagonal structure. On the other hand, the Ge<sub>2</sub>Sb<sub>2</sub>Se<sub>5</sub> alloy forms an orthorhombic structure which is not reported in the classical GST group.



Figure 2: (a) SEM images of fabricated ring resonator devices; (b, c) transmission spectra of (b) GST-based device and (c) GSS4T1-based device.

To demonstrate the enhanced optical performance of the new O-PCMs, we deposited and patterned via lift-off GSST and GST patterns on single-mode SiN waveguide resonator devices fabricated following standard CMOS process protocols detailed elsewhere [7]. Figures 2b and 2c plot the transmittance spectra of SiN micro-ring devices integrated with the classical GST 225 phase change material and the GSS4T1 alloy when they are switched from amorphous to crystalline state. The device integrated with the GSS4T1 material claims a large on/off contrast ratio of 41 dB and an insertion loss of 0.2 dB, both of which represent significant improvements compared to state-of-the-art GST-based devices [1, 8]. Reversible switching of the GSS4T1 material using 4 ns laser pulses was also experimentally validated. Our theoretical model shows that improved cavity Q-factor due to reduced parasitic optical loss from GSST and enhanced switching contrast benefiting from the large FOM account for the performance boost.

In sum, we have demonstrated a new group of optical phase change materials Ge-Sb-Se-Te featuring broadband optical transparency covering the telecom bands up to LWIR. We anticipate that the materials will open up numerous emerging applications based on non-volatile photonic reconfiguration capitalizing on the enhanced optical performance of the materials compared to conventional phase change alloys.

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