

Myths and truths about optical phase change materials: A perspective

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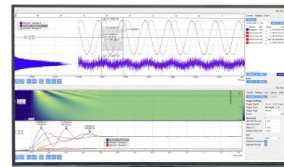
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ABSTRACT

Uniquely furnishing giant and nonvolatile modulation of optical properties and chalcogenide phase change materials (PCMs) have emerged as a promising material to transform integrated photonics and free-space optics alike. The surge of interest in these materials warrants a thorough understanding of their characteristics specifically in the context of photonic applications. This article seeks to clarify some commonly held misconceptions about PCMs and offer a perspective on new research frontiers in the field.

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INTRODUCTION

Chalcogenide phase change materials (PCMs) exemplified by $\text{Ge}_2\text{Sb}_2\text{Te}_5$ (GST-225) represent a class of materials whose electronic and optical characteristics change drastically when undergoing an amorphous-crystalline phase transition. This singular attribute underpins their commercial adoption in the nonvolatile electronic data storage, for example, Intel's Optane[™] memories. Inspired by this success, photonics naturally represents the next realm where PCMs can make an impact. Indeed, the past few years have witnessed rapid expansion of research explorations in PCM-based photonics with diverse applications spanning optical switching,^{1–8} photonic memory,⁹ optical computing,^{10–14} active metamaterial/metasurface,^{15–25} reflective display,^{26,27} and thermal camouflage.^{28,29} These optical device implementations, however, present unique challenges and requirements often distinctively different from those accepted for electronic memories. It is therefore instructive to elucidate some common confusions about these materials with respect to photonic applications, which constitutes the focus of this perspective. In the end, we will also offer our perspective on the key technical challenges that define the path forward toward making a practical impact with optical PCMs and emulating their success paradigm in the memory domain.

OPTICAL CONSTANTS ARE NOT CONSTANT

Optical constants, aka refractive indices of materials, are essential inputs for photonic designers and are used to lay out the blueprints of

functional devices. However, there is a twist when it comes to PCMs: their optical constants are not constant but rather critically dependent on processing conditions. This is evidenced by the considerable spread in literature values of PCM optical constants even for well-studied compositions such as GST-225^{30–35} (Fig. 1).

The discrepancy of amorphous phase properties shown in Fig. 1(a) can be attributed to the different preparation methods in addition to stoichiometry fluctuations—a universal trait of amorphous materials.³⁶ For instance, it is well established that as-deposited amorphous PCM films and re-amorphized films exhibit different characteristics.³⁷ Minor composition deviations in the films from the bulk target material may have also contributed to the variation.

The property changes of PCM films upon heat treatment (via furnace annealing, laser excitation, or electrothermal heating) sensitively depend on processing conditions, resulting in much larger spread of the crystalline phase optical constants [Fig. 1(b)]. On one hand, intermediate states comprising mixtures of amorphous and crystalline phases can be formed by controlling the temperature or duration of the heat treatment. Notably, there are two distinctive forms of intermediate states. When heating of PCM is non-uniform, for example, in laser switching where absorption is localized in the lossy crystalline regions, a well-defined boundary between the two phases emerges and crystallization proceeds in the form of phase boundary movement.³⁸ Spatially segregated two phase regions can

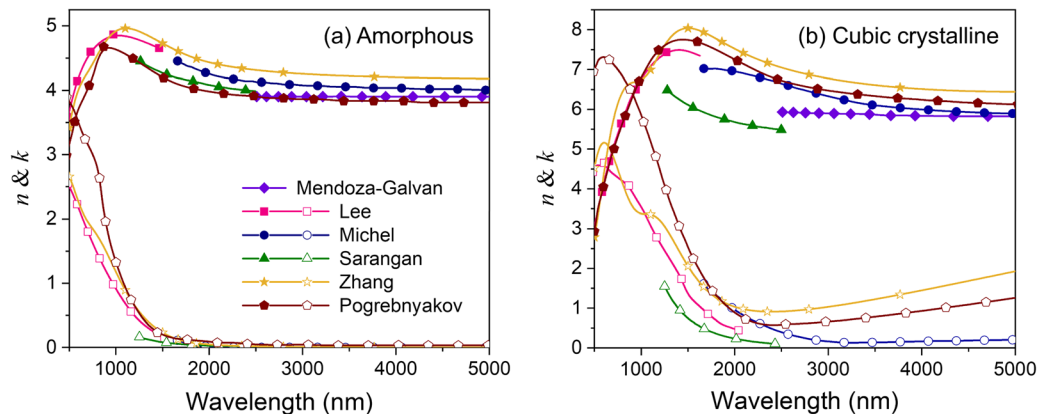


FIG. 1. A compilation of literature reports on optical constants of GST-225 in its (a) amorphous and (b) cubic crystalline states. The filled and open symbols denote the real (n) and imaginary (k) parts of the refractive indices, respectively.

also be observed in growth-dominant PCMs even in the absence of a temperature gradient.³⁹ Alternatively, when uniform heating is applied to a nucleation-dominant PCM, the intermediate states are formed during controlled crystallization and involve crystalline precipitates uniformly dispersed throughout an amorphous matrix.⁴⁰ Therefore, the two scenarios yield different spatial distributions of phases and hence refractive index.

On the other hand, the optical constants of the crystalline phase are not constant and continue to evolve during heat treatment even after the phase transition is fully complete, a phenomenon resulting from vacancy ordering.^{41,42} Crystalline PCMs formed at low annealing temperatures contain a large amount of atomic vacancies randomly distributed in the material. The structural disorder induces Anderson localization of electronic carriers, which lowers free carrier absorption (FCA). Upon further heat treatment, these defects spontaneously group into ordered vacancy planes, and the FCA rises accordingly due to increased carrier mobility.

The dependence of optical constants on heat treatment conditions poses a technical challenge on both optical and thermal designs of PCM devices. Spatial variation of optical constants induced by temperature differentials should be mitigated via thermal engineering or properly accounted for in optical modeling. The dependence also provides a unique opportunity to fine-tune a PCM's optical responses without chemically modifying its composition. This is illustrated in Fig. 2 that plots the real and imaginary parts of the refractive index of $\text{Ge}_2\text{Sb}_2\text{Se}_4\text{Te}_1$ (GSST) at $2\mu\text{m}$ wavelength with different annealing temperatures. Below 360°C , there is a significant change of the real refractive index accompanied by a relatively minor increase in optical loss with elevating annealing temperatures. This is associated with progressive transition from the amorphous to the crystalline phase. Above 360°C , the index change is minimal, but the absorption increase becomes more pronounced likely due to the vacancy ordering mechanism. The result indicates a method to *independently* tune the refractive index and absorption of the PCM, a highly coveted capability for many active photonics applications.

A DIFFERENT WAY TO SWITCH

Electrical pulsing is often a preferred option for PCM switching: unlike furnace annealing, it is capable of reversible switching of PCMs;

and compared to laser switching, it can be readily integrated with nanophotonic devices to enable chip-scale reconfigurable optical platforms. Additionally, the heat supply and distribution are independent of the phase composition in the PCM in electrical switching, while optical heating relies on optical absorption so the temperature profile is highly variable depending on the PCM's condition. In phase-change random access memories (PRAM), the phase transition is triggered by Joule heating from electrical currents directly passing through the PCM. While the same scheme is frequently proposed for electrical switching of PCMs in optical devices, an important limitation arises due to a phenomenon known as “filamentation”⁴³ (a name derived from its similarity to resistive switching in oxides⁴⁴): when electrical current runs through the PCM to induce crystallization, a thin wire of PCM that initially crystallizes forms an electrical current pathway with significantly higher conductivity than the surrounding amorphous matrix, hampering uniform crystallization throughout the material. Filamentation occurs only when the PCM size exceeds a few hundred nanometers. That is why filamentation has not been an issue for today's deeply scaled PRAM cells. However, in photonic applications

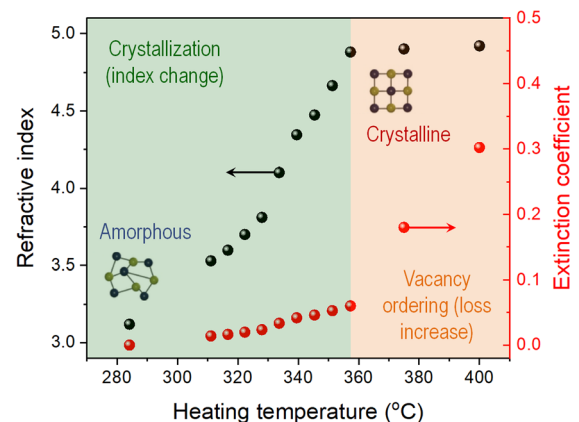


FIG. 2. Measured optical constants of $\text{Ge}_2\text{Sb}_2\text{Se}_4\text{Te}_1$ (GSST) as a function of transient heat treatment condition.

where the PCM size is of wavelength scale, Joule heating must rely on an external resistive heater rather than the PCM itself to circumvent filamentation. Reversible electrical switching of PCMs in photonic devices has been accomplished with micro-heaters made of metals,^{23,30,40} doped silicon,^{45–47} transparent conducting oxides,³ and graphene.⁴⁸ The only exception to the rule is a plasmonic nanogap device, where the PCM volume is minimized via sub-diffraction-limited plasmonic light confinement.⁴⁹ The PCM can also be reduced to subwavelength-scale structures as the optical mode is only sensitive to their collective optical properties.^{5,14} In this way, each PCM element in a subwavelength-scale structure is independently switched by electro-thermal heating with a much smaller volume, and thus the switching performance in terms of speed and repeatability is enhanced while effectively suppressing filamentation.

We also note that the statistical nature of nucleation and filamentation processes in nucleation-dominant PCMs may be turned into a feature in a different context, e.g., for generating physical unclonable functions.⁵⁰

THE THICKNESS BARRIER OF SPEED

Reversible switching of PCMs also imposes a limit on the size of PCM elements—a constraint often unknowingly neglected in the device design, particularly in active metasurfaces based on PCMs.

To amorphize the PCM, the material is heated to above its liquidus temperature and rapidly quenched to “freeze” the liquid-like amorphous state. In order to ensure that crystallization is kinetically suppressed, the cooling rate must exceed a threshold value known as the critical cooling rate.³⁶ The attainable cooling rate in a thick material becomes limited by thermal conduction since it takes time for heat to escape from its interior to the surface. As a result, for each PCM, there exists a maximum thickness beyond which complete reversible switching proves difficult. For GST-225, its fast crystallization kinetics implies a critical cooling rate of the order of 10^9 °C/s,⁵¹ corresponding to a maximum reversible switching thickness of approximately 150 nm. Uncoincidentally, reversible switching of GST-225 in photonic devices has only been realized in thin films with sub-100-nm thickness. This thickness limit is generally not a concern for integrated photonic devices, where the light-PCM interaction can be boosted simply by increasing path length along a waveguide. In free-space optical devices exemplified by optical metasurfaces, thin film filters and

diffractive optical elements, however, the small thickness bounds the tuning range of optical phase delay, thereby severely compromising the device performance. The thickness limit can be relieved by exploring PCMs with slower crystallization kinetics or equivalently improved amorphous phase stability, which however introduces tradeoffs in speed and energy consumption.

Here, we propose an alternative solution to overcome the thickness limit without compromising the crystallization speed. The concept makes use of growth-dominant PCMs such as Ag-In-Sb-Te, which are characterized by sluggish homogeneous nucleation of crystalline “seeds” inside the material and exceptionally high crystal growth rates when such a seed is already present.⁵² The design involves a tadpole-shaped PCM block, where the tip of the block’s tail locates outside the active switching zone and thus always remains in a previously prepared crystalline state (Fig. 3). The crystalline tip allows the PCM block to sidestep the nucleation period enabling swift crystallization. During re-amorphization, the tail of the PCM block is rapidly quenched to sever the crystal growth path, thereby avoiding crystallization in the main PCM element despite its slow cooling.

FASTER OR NOT FASTER, THAT IS THE QUESTION

Developing PCMs with ever faster speed has been a Holy Grail for the phase change memory community, as expedited crystallization kinetics leads to memories with enhanced write speed.⁵³ Nonetheless, the game rules must be redefined in photonics. The energy dissipation overhead required for switching large-volume PCMs in optical devices means that PCMs are hardly competitive for ultrafast optical modulators, where electrooptic and electroabsorption materials offer far superior performances. Moreover, the limited cycle lifetime of PCMs likely disqualifies them from applications, where constant fast switching is demanded over an extended period of time. For free-space optics, switching speed of PCM is further limited by the thickness barrier discussed in “The Thickness Barrier of Speed” section. In integrated photonics, the thermal decay time in photonic structures defines the ultimate switching speed limit. Doped Si heater designs in standard foundry-compatible silicon-on-insulator (SOI) platforms offer a thermal decay time of 1–10 μ s,^{54,55} limiting the practically attainable PCM switching speed to the same level. SiN, another broadly employed integrated photonics material platform, comes with metal heaters with even longer thermal decay time.

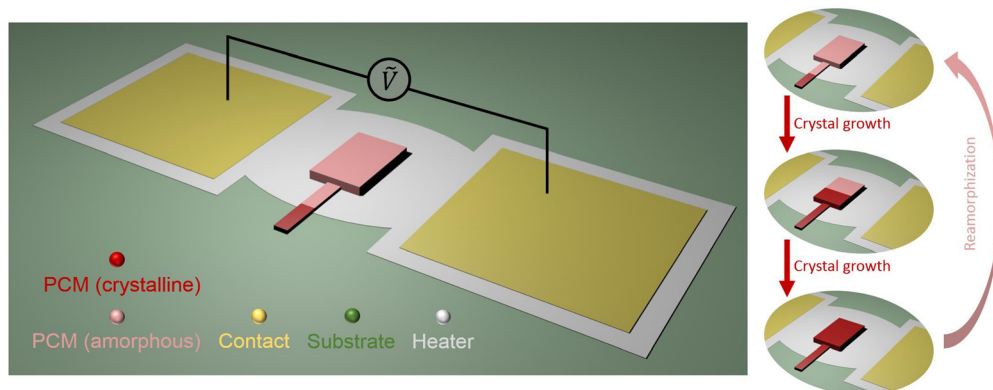


FIG. 3. Schematic configuration to enable fully reversible switching of large-volume PCMs without compromising switching speed.

To further enhance the speed of electrothermally switched PCM devices, we optimized the doping profile of a 220-nm SOI waveguide-integrated heater [Fig. 4(a)] to minimize the thermal decay time and driving voltage while maintaining a moderately low heater-induced insertion loss of 0.042 dB. The loss value is quoted for a 6 μm long heater to impart π phase shift to the waveguide mode and only includes FCA in doped Si without considering absorption or scattering due to the PCM. Figure 4(b) plots the simulated time-domain temperature change inside the PCM when the heater is driven by two types of “amorphization” pulses, 4.5 V for 1 μs and 8 V for 25 ns (with 10 ns rising and trailing edges in both cases). While both pulses attain

similar peak temperature values enough to melt the PCM, the thermal decay times (defined as the time for the temperature rise to decay to $1/e$ of its peak value) are very different, yielding 160 ns for the 4.5 V pulse and 35 ns for the 8 V pulse. The difference can be explained by the volume of the heating zone as shown in Fig. 4(c). The short 8 V pulse minimizes heat diffusion and leads to a much smaller effective thermal mass. The high voltage and large current (15.5 mA for the 8 V pulse) however may pose a challenge for integration with electronic drivers. It is also possible to further reduce the thermal decay time through lowering the heater thermal mass, for example, by using graphene instead of Si as the heater material,^{48,56} which nevertheless comes at the expense of more complicated and nonstandard processing. The modeling results suggest that PCM compositions with a time-to-crystallization below tens of nanoseconds may find limited applications in photonics since they are difficult to electrically switch in standard photonics platforms.

THE CHALLENGES THAT LIE AHEAD

Before PCMs can make a sustained impact on photonic applications, there are still key technical challenges that must be addressed. Cycle lifetime or endurance of PCMs in photonic devices remains to be validated or improved. In electronic memories, PCM endurance and failure mechanisms have been extensively characterized. In photonic devices, while endurance $> 1.5 \times 10^8$ has been measured in the PCM Sb_7Te_3 ,⁵⁷ similar levels of endurance performance have not been validated in other PCMs including GST-225 and emerging low-loss optical PCMs such as GSST, Sb_2S_3 , and Sb_2Se_3 .^{58,59} In light of the distinctive electrothermal switching configuration for photonic applications, we have generalized a set of material and device design rules for enhancing switching endurance:⁶⁰ (1) single-phase system, (2) minimal volume change during phase transition, (3) temperature uniformity, and (4) encapsulation design. Understanding of failure modes specific to photonic devices and implementation of the design rules to boost device lifetime will be imperative to facilitating adoption of PCMs in many practical applications.

Minimizing the switching voltage and current of PCM devices is another important direction. The high voltage and current required for amorphization of PCMs (8 V and 15.5 mA in the example shown in Fig. 3) are not compatible with most CMOS transistor drivers and result in a large transient power consumption. The issue can be mitigated by (1) identifying PCMs with lower melting points without compromising other desirable attributes of the material, (2) designing devices with strong light confinement inside the PCM and without introducing excessive parasitic optical losses such that the PCM volume can be downscaled, and (3) enhancing heat localization within the PCM to effectively lower thermal mass. As illustrated in the SOI heater design example above, the multiscale, multi-physics nature of PCM-enabled active photonic devices further warrants a holistic coupled electro-thermo-optical co-design framework for device engineering and optimization.

Finally, integration into the standard photonics foundry process represents an essential technical milestone for PCMs. Foundry process integration is not only the practical route toward scalable manufacturing of PCM devices, but also eases access to PCM components for the entire photonics community. Notably, PCMs are readily poised for CMOS backend integration with their non-epitaxial nature and low processing temperatures, evidenced by their seamless integration in the 3D

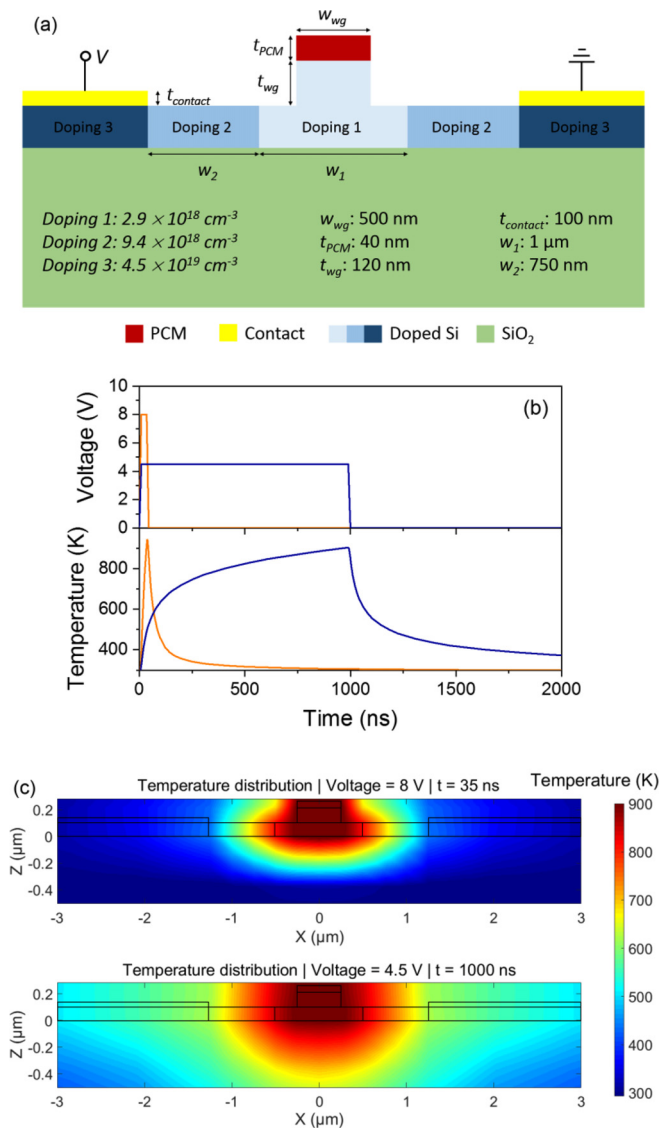


FIG. 4. (a) Electrothermal heater design for PCM switching; (b) time-domain response of the heater (bottom panel) to two types of voltage pulses (top panel); and (c) transverse temperature profiles corresponding to the peak temperature points at the end of the voltage pulses.

XPoint memory architecture. We foresee that achieving this milestone will significantly expedite PCMs' integration into large switching matrices and open up emerging applications such as arbitrary wavefront synthesis, energy-efficient optical switching and routing, and quantum optical networks as well as scalable neuromorphic computing.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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