Contents lists available at ScienceDirect





journal homepage: www.journals.elsevier.com/materials-today-advances/



Emerging horizons in phase-change materials for non-volatile memory

Check for updates

Yan Chen^a, Yuqiao Le^a, Lei Chen^b, Haisong Liu^a, Tangyou Sun^a, Xingpeng Liu^a, Fabi Zhang^a, Haiou Li^a, XinXin Hu^c, Ying Peng^{a,*}, Chengyan Liu^{d,**}, Min Hong^{b,***}

^a Guangxi Key Laboratory of Precision Navigation Technology and Application, Guilin University of Electronic Technology, Guilin, 541004, China

^b Centre for Future Materials and School of Engineering, University of Southern Queensland, Springfield Central, Queensland, 4300, Australia

^c Department of Energy and Power Engineering, Hunan University of Humanities, Science and Technology, Loudi, 41700, China

^d Shaanxi Normal University, Xi an, 710119, China

ARTICLE INFO

Keywords: Phase change memory Resistance drift Aging Crystallization kinetics β-relaxation Fragile-to-strong

ABSTRACT

Phase Change Memory (PCM) technology, particularly utilizing GeTe-based materials, has emerged as a compelling alternative to traditional nonvolatile memory systems, offering significant advancements in speed, scalability, and endurance. This review provides an in-depth analysis of recent developments in GeTe PCM, focusing on the challenges and breakthroughs that have characterized its evolution. Key aspects of resistance drift, material aging, and crystallization dynamics were explored, with special attention to the fundamental mechanisms, including β -relaxation and the Fragile-to-Strong Transition in supercooled liquids, along with Peierls distortions. Innovations in material engineering, such as alloying and nanoconfinement, have been discussed for their roles in enhancing device performance and reliability. Furthermore, this review examines the impact of advanced fabrication techniques and novel device architectures on the practical applications of PCM. By integrating these technological advancements with a theoretical understanding of the material properties, this review highlights the transformative potential of GeTe PCM across various applications, setting the stage for future research directions aimed at fully realizing the capabilities of this promising memory technology.

1. Introduction

Phase Change Memory (PCM) technology has not only achieved remarkable success in the commercial market since Intel and Micron introduced 3D XPoint technology in 2015, but has also significantly advanced the development of storage technology [1–3]. 3D XPoint technology has successfully narrowed the performance gap between traditional DRAM and NAND flash memory through its non-volatile and DRAM-like fast access capabilities and has been widely used in data centers and high-end storage solutions that require high density and high endurance [4–6].

In recent years, the application of PCM technology in cutting-edge fields, such as artificial intelligence, quantum computing, and the Internet of Things, has gradually unfolded, providing new ideas for highperformance computing and efficient storage. In the field of artificial intelligence and machine learning, PCM's fast read/write capability and non-volatility make it an ideal choice for neural network weight storage

[7,8]. This not only avoids the problem of data reloading in traditional memories but also significantly reduces the energy consumption during training [9–11]. PCM maintains the neural network weights even after a power failure, which brings higher efficiency to machine learning tasks that involve frequent data reads and writes, enabling faster training of AI models [12,13]. In quantum computing, PCM's non-volatility and long-term stability are very favorable for storing quantum bit states [14-16]. Quantum computing requires a high level of storage technology that can access data quickly and reliably, and the characteristics of PCMs fulfill this need, helping quantum systems to remain stable [17-20]. PCMs are also able to maintain data invariance under temperature-controlled environments, further enhancing storage management for quantum computing [21]. The fast response and low-power requirements of IoT devices also match the characteristics of PCM technology [19,22]. IoT devices usually require persistent data storage to ensure that data is not lost under intermittent power supply conditions, and the low-power characteristics of PCM enable it to hold data for

https://doi.org/10.1016/j.mtadv.2025.100571

Received 10 December 2024; Received in revised form 9 February 2025; Accepted 21 February 2025 Available online 26 February 2025 2590-0498 /@ 2025 The Authors, Published by Elsevier Ltd. This is an open access article under the CC BY

2590-0498/© 2025 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/).

^{*} Corresponding author.

^{**} Corresponding author.

^{***} Corresponding author.

E-mail addresses: pengying@guet.edu.cn (Y. Peng), chengyanliu@guet.edu.cn (C. Liu), min.hong@unisq.edu.au (M. Hong).

long periods when the device has limited power. PCM's fast-access capability of PCMs also enables them to perform well in data-intensive IoT applications, helping to improve the device's ability to respond quickly to changes in the environment and to realize real-time data processing and efficient storage management in application scenarios, such as smart cities and industrial monitoring [23,24]. application scenarios to realize real-time data processing and efficient storage management.

In addition, PCM technology is an important advancement in brainlike computing due to its non-volatility and high-speed read/write capability, making it an ideal material for simulating synaptic connections in the human brain [25]. PCM allows the storage of multiple resistive states on the same device, simulating changes in the synaptic strength of neurons, which is crucial for implementing brain-like learning mechanisms. The ability of this material to change conductance rapidly supports fast computation without the loss of information, making it particularly important in artificial intelligence and deep learning applications. The latest research utilizes a gradient doping technique to achieve finer conductance control in multilayer PCMs, enabling devices to achieve secondary and quaternary storage states through different electrical pulse operations, greatly improving their functionality and efficiency in neuromorphic computing [26]. This technique not only demonstrates its efficiency in complex pattern recognition tasks by achieving learning accuracies of up to 94.81 %, but also highlights the potential for modeling synaptic plasticity in brain-like computing architectures [26]. However, despite showing great promise for applications, PCM technology still faces challenges in practice such as resistive drift, which may affect the long-term stability and computational accuracy of the device. Future research will need to address these challenges by developing new materials, optimizing device structures, and improving programming methods and circuit design to further enhance the efficacy of PCM for high-performance computing and smart device storage.

This review discusses the physical origins of reliability optimization in PCM using a multiscale analytical framework. Section 1 identifies Joule heating-driven phase transitions and resistive drift/aging as core challenges through macroscopic electro-thermal-structural coupling. Section 2 introduces the collective relaxation model, challenging the conventional isolated defect paradigm by redefining resistive drift as a cooperative atomic process governed by systemic energy minimization. Section 3 reveals aging in amorphous GeTe toward an ideal glassy state, elucidating its structural evolution as a metastable intermediary between amorphous and crystalline phases. Section 4 describes the dual roles of β -relaxation—its competition with α -relaxation governs resistive drift reversal, while its coupling with the fragile-to-strong (F-S) transition unveils the cross-temperature regulation of phase stability in GeTe-GeSe composites via dynamic heterogeneity. Section 5 translates these insights into nanoscale engineering strategies for thermal-electronic decoupling and low-power operation. This review establishes a unified theoretical framework for PCM reliability and proposes a pathway for designing high-performance memory through controlled structural dynamics.

2. Dynamic interactions and performance challenges of PCMs

PCM technology utilizes the large difference in resistance between the crystalline and amorphous states of a material to enable data storage. In the amorphous state, PCM exhibits high resistance, while in the crystalline state, the resistance is lowered by a factor of several thousand, with high resistance representing a logic "0" and low resistance representing a logic "1". This significant difference in resistance makes it easy to distinguish between the read currents in the two states.

Fig. 1 illustrates a typical PCM cell structure consisting of a bottom electrode, a layer of PCM, and a top electrode. Applying a voltage across these electrodes generates a current that flows through the PCM, causing Joule heating, which in turn changes the phase of the material. The SET process of the PCM consists of transforming the material from an amorphous state to a crystalline state using short, high-amplitude pulses, while the RESET process consists of transforming the material from a crystalline state to an amorphous state using longer, lower amplitude pulses, which is manifested by the fact that the longer pulses and the lower amplitude pulses cause melting, followed by rapid quenching to



Fig. 1. A diagram illustrating the interrelationship between electrical, thermal, and structural dynamics in PCM devices. The diagram also highlights the challenges faced by PCM within this system and outlines the corresponding optimization strategies.

an amorphous state [27,28]. During this reversible transformation, the PCM is profoundly affected by the interplay of electrical, thermal, and structural dynamics as shown in Fig. 1 [29]. The electrical system controls the power (*IV*) by applying voltage (*V*) and current (*I*), which directly affects the heating process of the material. The current through the PCM generates Joule heat, which significantly increases the temperature (*T*) of the material and initiates phase transitions. The performance of the electrical system affects the read/write speed of the PCM and the heating efficiency of the material and has a direct impact on the energy consumption. Efficient management of the electrical system ensures optimal power delivery to the material, which facilitates control of the precise temperatures required for phase transitions [29].

The thermal system receives power from the electrical system and regulates the temperature distribution of the material through heat transfer. Higher temperatures typically accelerate the phase change process, facilitating the transition of the material from an amorphous to a crystalline state, but may lead to a reduction in the long-term stability of the crystalline phase. Structural dynamics involves changes in the internal structure of a material under different temperature and time conditions. The thickness of the amorphous region (u_a) and the relaxation state (Σ) are key parameters that determine the electrical resistance properties and phase transition efficiency of the material [16,30,31]. Over time, relaxation processes in the structural dynamics can lead to resistance drift and aging effects, posing challenges to the long-term stability of data storage. The effect of the electrical, thermal, and structural dynamic systems acting as an interacting dynamic process within a PCM device can impact its performance. The thermal system receives this energy and regulates the temperature of the material to ensure a stable phase transition. The structural dynamics adjust the thickness and relaxation state of the amorphous regions according to temperature and time, thus affecting the resistance [29]. These dynamic interactions ensure fast and stable switching between crystalline and amorphous states, leading to efficient storage.

Based on the understanding of these dynamic processes, we can identify the challenges facing current PCM technology. The phenomenon of resistive drift, one of the main problems in PCM, is mainly related to structural relaxation within the material. Structural relaxation involves a subtle rearrangement of the internal structure of the material, which allows the material to reach a more stable thermodynamic state; however, this small change in the local arrangement of the internal atoms affects the electronic band structure and hence the resistive properties of the material [32,33]. In addition, the crystallization process of PCM is a thermally activated process involving the nucleation and growth of microcrystals in an amorphous matrix. These processes are controlled by kinetic parameters such as the nucleation rate of the material, the growth rate, and the thermal stability of the amorphous phase. The combination of these factors leads to the phenomena of resistance drift and aging in PCMs, which are key factors that must be considered when optimizing PCM performance.

3. Resistance drift issues in PCM

3.1. Mechanisms and modeling of resistance drift

Recent studies have revealed the complex physical processes behind resistance drift by linking it to changes in structural relaxation (SR) and conductive activation energy, providing an important framework for understanding the phenomenon of changes in electrical resistance values of PCMs during long-term use [34–37].

The core equation related to resistance drift is

$$R(t) = R_0 \left(\frac{t}{t_0}\right)^{\nu} \tag{1}$$

where R(t) is the resistance at time t, R_0 is the initial resistance at the reference time t_0 , t is the elapsed time, t_0 is the reference time, and v is

the resistance drift exponent. This indicates that the resistance exhibits a power-law growth over time.

Additionally, the introduction of the Meyer-Neldel (MN) rule offers a powerful tool for analyzing the behavior of resistance drift, particularly through the equation of

$$\nu = \frac{\Delta E_{\rm C}}{\Delta E_{\rm SR}} \frac{T_{\rm A} \left(1 - \frac{T_{\rm R}}{T_{\rm C}}\right)}{T_{\rm R} \left(1 - \frac{T_{\rm A}}{T_{\rm SR}}\right)} \tag{2}$$

where $\Delta E_{\rm C}$ is the activation energy for the crystalline state, $\Delta E_{\rm SR}$ is the activation energy for structural relaxation, $T_{\rm A}$ is the annealing temperature, $T_{\rm R}$ is the reference temperature, usually room temperature, $T_{\rm C}$ is the characteristic temperature of the crystalline state, $T_{\rm SR}$ is the characteristic temperature of structural relaxation [38]. From the formula, we can see that the main factors affecting the resistance drift index are temperature, the activation energy of the material, and the characteristic temperature [39,40]. These factors work together to determine the change in the resistance drift index.

These equations not only reflect the time dependence of resistance drift but also highlight how internal physical processes—specifically changes in activation energy and temperature effects—influence resistance drift behavior. Moreover, the impact of distortions within amorphous materials on resistance drift cannot be overlooked. Distortions, inherent to the amorphous state, indirectly affect resistance by influencing the material's local electronic states and transport properties [41, 42].

During the process of structural relaxation, internal distortions, and atomic arrangements undergo dynamic adjustments. These adjustments impact not only the conductive activation energy but also the activation energy of structural relaxation itself [34,35]. Thus, the interplay between distortions and structural relaxation becomes key to understanding and controlling resistance drift. By integrating the MN rule's comprehensive interpretation of changes in activation energy and its entropy effects, we can achieve a more thorough understanding of the mechanisms of resistance drift in amorphous materials.

Based on our understanding of the mechanisms underlying resistive drift, we will further explore relaxation models of resistive drift and delve into how these models can help us understand and predict the behavior of resistive drift.

3.2. Collective relaxation and resistance drift in PCM

In amorphous GeTe, the phenomenon of resistance drift is closely related to the relaxation of the internal structure of the material. Specifically, this relaxation involves changes in specific defect structures composed of closely connected Ge atoms [41,43]. Among these defect structures, at least one Ge atom is coordinated differently from the ideal trivalent coordination (i.e., bonded only to Te atoms), as illustrated in Fig. 2a. The presence of these non-ideal coordination introduces localized electronic states within the bandgap, which affect the electrical transport properties of the material [44]. Over time, these defect structures gradually transform into lower-energy configurations characterized by chemically ordered trivalent topologies through a series of collective rearrangement events [43,45]. This process is accompanied by the elimination of stretched bonds within the amorphous network. These changes result in the annihilation of the localized electronic states in the bandgap, thereby increasing the resistance of the material. The resistance drift phenomenon in amorphous GeTe, driven by specific structural defects, underscores the role of these defects in causing the annihilation of localized electronic states in the bandgap through collective rearrangement events, leading to an increase in resistance [37, 46,47]. This process reveals the complexity of relaxation dynamics in amorphous materials, particularly in phase-change memory devices.

In describing this complex relaxation behavior, traditional Gibbs and



Fig. 2. (a). Structural relaxation of amorphous GeTe[43]. [][43][](b). The unstable amorphous state from melt-quenching relaxes to a more stable configuration [43]. Panels (a) and (b) are adapted with permission from Ref. 140, Wiley-VCH. (c). Model fitting - Cooperative relaxation model: Collective fitting of the time evolution of GST Vth at different temperatures [48]. (d). Model fitting - Gibbs model: The time evolution of GST threshold voltage fitted to the Gibbs model with three different shapes of activation energy spectra [48].Panels (c) and (d) are adapted with permission from Ref. 144, Wiley-VCH. (e) Details the temperature distribution of PCM chips from array-level drift experiments at varying environmental temperatures [37]. (f) Presents the cumulative resistance distributions in four instances, with experimental data shown as dashed lines and simulations as solid lines [37]. Reproduced with permission.

collective relaxation models provide different explanations and predictions. The Gibbs model, based on the activation energy spectrum of defect states, assumes that defects in the glassy state relax according to the activation energy distribution. In this model, defect states relax independently over time, each following its relaxation process. While this model can describe relaxation behavior over certain time scales, it shows limitations when addressing the broad and flat activation energy spectrum observed in amorphous GeTe [49,50]. In contrast, the collective relaxation model focuses on the rearrangement processes of the overall structure. This model hypothesizes that atoms or molecules within the material evolve towards lower energy states through collective rearrangement events [37,50]. In the collective relaxation model, the driving force for relaxation is provided by the energy difference (Δ_b) between two adjacent non-relaxed states [37]. This implies that as the system approaches its ideal glassy state, each relaxation step faces a higher energy barrier (E_b). Since the system's energy is already at a low level, further reducing it becomes increasingly challenging. The relaxation event rates for the equilibrium state n-direction and the non-equilibrium n + direction are given by

$$\frac{\mathrm{d}\Sigma}{\mathrm{d}t} = \Delta_{\Sigma} (n^+ - n^-) \tag{3}$$

 Σ represents the system's current state's distance or discrepancy from the ideal glassy state, which can be understood as a measure of the system's non-equilibrium level or the degree of relaxation. If each relaxation event changes Σ by a quantity $\Delta\Sigma$, then the relationships can be derived as

$$n^{-} = \nu_0 \exp\left(-\frac{E_{\rm b}}{k_{\rm B}T}\right) \tag{4}$$

$$n^{+} = \nu_{0} \exp\left(-\frac{E_{\rm b} + \Delta_{\rm b}}{k_{\rm B}T}\right) \tag{5}$$

where v_0 is the attempt frequency for relaxation, and E_b is the energy barrier for subsequent relaxation events [37]. Initially, when the

amorphous state is formed, the system is in a non-equilibrium state far from the ideal glassy state, defined as $\Sigma(0) = \Sigma_0$, the initial level of non-equilibrium. As time progresses, the system undergoes a series of relaxation events that gradually reduce the non-equilibrium level, i.e., the Σ value decreases, and the system relaxes towards the ideal glassy state. During this process, E_b changes as the state of the system changes.

As shown in Fig. 2b, when the system approaches the ideal glassy state (i.e., $\Sigma \rightarrow 0$), suggesting that the system is close to equilibrium, E_b increases to the maximum value (E_s). When the system reaches equilibrium ($\Sigma = 0$), the driving force (Δ_b) becomes zero, indicating that no extra energy discrepancy is available to drive further relaxation. In this model, a linear relationship exists between E_b and Σ , which is the basis for deriving the logarithmic time dependency of the relaxation behavior at a constant temperature. Accordingly, the evolution expressions for Σ at constant temperature are given by

$$\Sigma(t) = -\frac{k_{\rm B}T}{E_{\rm s}} \log\left(\frac{t+\tau_0}{\tau_1}\right)$$
(6)

$$\tau_1 = \left(\frac{k_{\rm B}T}{\nu_0 \Delta_{\Sigma} E_{\rm s}}\right) e^{\left(\frac{E_{\rm s}}{k_{\rm B}T}\right)} \tag{7}$$

$$\tau_0 = \tau_1 e^{\left(-\frac{\Sigma_0 E_s}{k_B T}\right)}$$
(8)

where τ_0 can be interpreted as the latency period for the transition to logarithmic decay and τ_1 is the time to reach equilibrium ($\Sigma \rightarrow 0$). Equation (6) clearly shows that the internal stresses in the material decrease with time after logarithmic decay [37]. This relaxation is influenced by the temperature (k_BT) and relaxation energy barrier (E_s), highlighting the thermally activated nature of the process. Parameters τ_0 and τ_1 adjust the timing and rate of this relaxation, respectively.

The collective relaxation model is widely used to simulate the relaxation behavior of various amorphous materials, including the temporal evolution of the threshold voltage (ΔV_{th}) [48]. Comparisons in

Fig. 2c and d reveal that the collective relaxation model can accurately fit the temporal changes in the threshold voltage at all temperatures (100 K, 150 K, 200 K, 250 K, 300 K), demonstrating good temperature dependence without requiring complex assumptions about defect distributions [48]. In contrast, while the Gibbs model offers flexibility, it requires numerous assumptions, increasing complexity. Fig. 2c illustrates that the collective relaxation model effectively captures the logarithmic increase of threshold voltage over time, whereas Fig. 2d highlights that the Gibbs model requires assumptions about different defect energy distributions, leading to suboptimal fitting performance.

In practical applications, resistance drift affects the reliability of multi-level data storage in PCM devices. By simulating 1000 devices and measuring 4000 PCM cells, the study shows that the simulated resistance distribution closely matches the experimental data (Fig. 2e). Furthermore, the model successfully captures key features of resistance drift, such as accelerated drift rates and the widening of resistance distributions (Fig. 2f). These findings validate the model's accuracy in predicting resistance drift behavior in PCM devices [48].

4. Aging effects on resistive drift in amorphous GeTe

4.1. Structural evolution in aging GeTe

In amorphous GeTe, the aging process leads to significant changes in the ratio of isopolar Ge-Ge bonds and tetrahedrally coordinated Ge (Ge^{T}) , which is manifested in the gradual evolution of the material towards a more ordered and lower energy structure (Fig. 3a). This evolution has multiple effects on the physical properties of the materials [51–55]. The aging-induced changes were systematically characterized

through photothermal deflection spectroscopy (PDS) and impedance spectroscopy measurements. The results of the PDS show that the band gap of amorphous GeTe significantly increases during the aging process (Fig. 3b), which helps to enhance the material's optoelectronic performance. The increase in band gap means that it is more difficult for the material to excite a charge, thus improving the stability of the material to some extent, which is important for the application of GeTe in high-stability optical storage. Meanwhile, the impedance spectroscopy results show that the resistivity of amorphous GeTe increases about five times compared to that before aging, leading to a significant decrease in the electrical conductivity of the material (Fig. 3c). This increase in resistivity is mainly due to the decrease in the number of free carriers caused by the breakage of homopolar Ge-Ge bonds and the gradual transformation of tetrahedral Ge^{T} atoms into trihedral $Ge(Ge^{III})$ atoms [39]. In addition, the reduction of tetrahedral Ge^T atoms also disrupts the conductive paths in the material, which further reduces the overall electrical conductivity. During the aging of amorphous GeTe, a significant increase in the Peierls distortion is also observed [53,39,56,57]. As aging progresses, the tetrahedrally coordinated Ge^T atoms gradually decrease and shift to the trihedrally coordinated Ge^{III}. This process makes the local chemical environment of the Ge atoms more asymmetric and reduces the coordination number of Ge, which in turn reduces the local atomic stress and makes the atoms more susceptible to rearrangement. This change in coordination number leads to an uneven distribution of bond lengths and the gradual formation of alternating long and short bond structures, i.e. the Peierls twist [53,58].

Simultaneously, the Te atoms gradually change from twice-coordinated (Te^{II}) to thrice-coordinated (Te^{III}). This change further exacerbates the inhomogeneity of the local structure of the material,



Fig. 3. (a) Different atomic configurations of amorphous GeTe during aging Te, tetrahedral Ge, and defective octahedral Ge atoms are presented as blue, red, and orange spheres, respectively [53]. (b) Relationship between the optical band gap of relaxed amorphous GeTe and the fraction of tetrahedral Ge atoms (Ge^T) in the structure. The reduction in the number of tetrahedrally coordinated germanium atoms leads to an increase in the optical bandgap [53]. (c) Resistivity of deposited amorphous GeTe [53]. (d). ALTBC diagram of Ge^H and Ge^{III} [53]. Panels (a)–(d) are reproduced with permission. Copyright 2015, Springer Nature Limited. (e) The crystallization mechanism of GeTe [59]. Reproduced under terms of the CC-BY license. Copyright 2024, Simon Wintersteller, published by Springer Nature Limited. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

leading to a more distorted lattice. Thus, the overall structure of the material evolves towards a more distorted and ordered state. Fig. 3d shows the bond-length distributions of Ge^H and Ge^{III}. As aging proceeds, it can be seen that the high-energy defect states of the Ge^H motifs gradually decrease, whereas the trihedrally coordinated Ge^{III} becomes more prominent in the aged glassy state, with the appearance of two distinct peaks of long and short bonds. These distributions indicate that intensification of the Peierls twist occurs during aging, indicating that the material is gradually transitioning from a high-energy disordered state to a low-energy ordered state. In addition, this structural rearrangement process is accompanied by the gradual disappearance of high-energy defect states (e.g., Ge^H), resulting in the formation of more stable Ge^{III} and Te^{III} coordination structures, which further strengthens the Peierls torsion network and leads to an increase in the band gap of the material [53,60-62]. However, the details of how this increased Peierls twisting affects the resistivity and the conductivity mechanism still need to be revealed by further studies.

4.2. The role of the ideal glassy state in aging GeTe

A recent study on the role of the ideal glassy state in the aging process of GeTe presents a new perspective in aging studies. The ideal glassy state is a state between a completely disordered amorphous state and a completely ordered crystalline state (Fig. 3e), which is characterized by partial ordering, but not to the extent of complete crystallization [52, 59]. It has been shown that the existence of an ideal glassy state helps to maintain the stable structure of GeTe during the aging process of GeTe and reduces the drastic phase transition, thus delaying the resistance drift and improving the aging resistance of the material [52,59].

In amorphous GeTe, the Ge chains are formed by Ge-Ge homopolar and Ge-Te coordination bonds, forming a complex disordered network. The Ge atoms have a variable number of coordinations in the chains, usually triple or quadruple-coordinated, depending on their specific positions in the chains [45,52,40,59,63]. Quadruple-coordinated Ge atoms, which are usually connected to one Ge atom and three Te atoms, are located at the junctions or less branching regions of the chain. Triple-coordinated Ge atoms, on the other hand, may be fully coordinated to Te atoms or coordinated to one Ge and two Te atoms, and these Ge atoms are commonly found at the ends or turning points of chains with a structure similar to that of the chains of unsaturated hydrocarbons in organic chemistry.

Te atoms play a key bridging role in these chains, and are usually present in a doubly coordinated form, connecting neighboring Ge atoms to form stable Ge-Te-Ge bridge states. This bridge state structure is important for reducing atomic stresses within the material, as it provides a mechanism to stabilize the Ge chains by reducing local inhomogeneities. These Te bridges help maintain the integrity of the Ge chains during aging or transition to a more ordered state of the material, preventing excessive chain breakage and facilitating a smooth transition from disorder to order. During the aging process, the partial disorder of Te atoms is gradually transformed into a more ordered Te sublattice. This ordering process provides the material with a more stable lowenergy structure, which supports the gradual breakage and recoordination of the Ge chains. With the gradual ordering of the Te sublattice, the local environment of the material tends to stabilize, prompting the Ge chains, which were originally connected by Ge-Ge homopolar bonds, to start breaking. The formation of Te-Ge-Te bridge states during this breaking process provides new coordination opportunities for Ge atoms, which promotes the rearrangement of atoms within the material and leads to a gradual transition to an ideal glassy state, which may eventually evolve into a more stable crystalline structure.

This ideal glassy state acts as a buffer state during the aging process, effectively mitigating the internal stresses due to structural rearrangement, to maintain stability and avoid drastic phase transitions, thus improving the aging resistance. This structural preordering and rearrangement guided by the Te sublattice is a key intermediate step in the amorphous-to-crystalline transition, ensuring the functional reliability and fast response of phase-transformed materials. In nanoparticles, the presence of an ideal glassy state is more pronounced. This is because, at the nanoscale, the material structure is affected by surface effects and size constraints, resulting in a slower and more stable formation of Ge chains and Te sublattices. The behavior of Te sublattices and Ge chains in nanoparticles results in the presence of an ideal glassy state for a longer period, which further improves the material's aging resistance. This gradual evolution of nanoparticles is more pronounced compared to bulk materials, effectively mitigating the increase in resistivity during aging and enhancing the stability of data storage [59, 64-66]. On this basis, it is reasonable to assume that the intensification of Peierls distortion may be linked to the preordering of the Te sublattice. The Peierls distortion manifests itself as an alternating change in the bond lengths in the material, which may be exacerbated by the reordering of Ge atoms during the partial ordering of the Te sublattice, thus further affecting the electrical conductivity and the overall stability of the material. The gradual evolution toward the ideal glassy state, mediated by Te sublattice preordering and Ge chain reorganization, establishes a structural foundation for mitigating aging-induced resistivity drift. This metastable intermediate phase acts as a kinetic buffer that redistributes atomic stress during structural relaxation, enabling gradual rather than abrupt property changes. Crucially, the nanoconfinement-induced persistence of this state highlights its critical role in designing aging-resistant PCM materials through controlled structural dynamics.

5. Crystallization kinetics in PCM

In PCM, analyzing the three critical temperatures— T_g , T_c , and $T_{\rm m}$ —helps to understand the complexity of crystallization dynamics [67]. limitations of conventional methods such as dielectric spectroscopy and dynamic mechanical analysis. T_c represents the temperature at which the material begins to crystallize, influenced by nucleation and growth mechanisms, as well as the stability of the crystal structure. $T_{\rm m}$ defines the temperature at which the material transitions from solid to liquid, determined by interatomic bonding strength and crystal structure. T_{g} is linked to the dynamics of the supercooled liquid, revealing the unique behavior in this temperature range. In the study of supercooled liquid dynamics, β -relaxation and the fragility-strength (F-S) relationship are key aspects. β -relaxation reflects the local dynamic behavior of materials near $T_{\rm g}$, impacting crystallization, stability, and physical properties. The F-S relationship describes the changes in the viscosity and fragility of materials near T_g , influencing the crystallization behavior and overall performance. In the next section, we will explore the behavior of β -relaxation in PCM and its connection to the F-S relationship.

5.1. β -relaxation

Although β -relaxation in the PCM has been demonstrated, accurate measurement remains challenging due to the limitations of conventional methods such as dielectric spectroscopy and dynamic mechanical analysis. The temperature-dependent loss modulus (*E''*) of GeTe (Fig. 4a and b) clearly shows β -relaxation. Fig. 4c shows that at high temperatures, α - and β -relaxation occur at similar time scales, suggesting that the atoms are moving in concert. As the temperature approaches the glass transition temperature (*T*_g), α -relaxation slows down and becomes viscous, whereas β -relaxation maintains faster-localized motions, allowing structural adjustments to be made even as large-scale atomic motions slow down [59]. This is consistent with the anomalous resistivity behavior seen in the Ge₃Sb₆Te₅ annealing experiments (Fig. 4d), where the annealing process usually induces a resistive drift leading to a gradual increase in resistivity. However, a reversal of the resistivity drift was initially observed in the annealing experiments at



Fig. 4. (a) Diagram of the experimental setup for powder mechanical spectroscopy [73]. (b) Loss modulus E'' of GeTe, with the shaded area representing the excess wing [73]. (c) illustrates the behavior of α -relaxation and β -relaxation in PCM and non-PCM covalent glasses [73]. Panels (a) \sim (c)Reproduced with permission. Copyright 2020, AAAS. (d) Resistivity drift of Ge₃Sb₆Te₅ during isothermal holding at 160 °C [68]. Reproduced with permission. Copyright 2022, Wiley-VCH.

160 °C, where the resistivity decreased, but then stopped decreasing and instead increased to a higher value than before the reversal. This inversion phenomenon is thought to be related to the inherent "cross-over effect" in glasses, which arises from the dynamic competition between α - and β -relaxation when their characteristic time scales approach each other near the glass transition regime. For example, the "rejuve-nation" effect in the fast relaxation region is similar to the local tuning of

the β -relaxation, while the slow relaxation region may be related to the global rearrangement of the α -relaxation. This crossover effect reflects the dynamic synergy between β -relaxation and α -relaxation, and the annealing process may have suppressed the local rearrangement of β -relaxation and further affected α -relaxation, which triggered the reversal of resistivity drift and its subsequent behavior [68–70]. Over time, the resistivity stabilizes, showing that these dynamic processes



Fig. 5. (a). Comparison of the excess wing area ratio between PCM (blue) and non-PCM (orange) [73]. Panel (a) is reproduced with permission. Copyright 2020, AAAS. (b) Normalized loss modulus E["] of $Ge_{15}Sb_{85}$ annealed at 458 K for 3 and 6 h, showing β -relaxation, measured by powder mechanical spectroscopy [71]. (c) Crystallization probability as a function of pulse duration at ~40 mW constant power [71]. (d) Kissinger plots for crystallization of $Ge_{15}Sb_{85}$, annealed for 3 and 6 h, using DSC and flash DSC at different heating rates [71]. (e) *S*(*q*) spectra during isothermal annealing at ~458 K for 6 h [71]. (f) Dielectric function of as-deposited and annealed $Ge_{15}Sb_{85}$ thin films [71]. Panels (b) ~ (f) are reproduced under the terms of the CC-BY license. Copyright 2022, Yudong Cheng, published by Springer Nature Limited. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

eventually reach a new equilibrium state. These interactions have a significant impact on the performance and efficiency of PCM materials, especially in terms of improving data retention and optimizing the reliability of write/erase cycles. Non-PCM materials such as GeSe show significant differences in β-relaxation compared to phase change storage materials. For example, GeSe shows very weak β -relaxation (Fig. 5a), which leads to less efficient local atomic motions, thus slowing down the crystallization kinetics and affecting the material's performance during the phase transition. In contrast, PCM materials such as Ge15Sb85 exhibit different properties during thermal annealing. In $Ge_{15}Sb_{85}$, β -relaxation was significantly suppressed after thermal annealing, especially after 6 h of annealing, and the excess wing area was significantly smaller than that after 3 h of annealing (Fig. 5b). Although thermal annealing usually promotes crystallization, in this sample, T_p increased while the crystallization rate slowed down (Fig. 5c and d), suggesting that thermal annealing may affect the crystallization kinetics by inhibiting β-relaxation. In situ monitoring showed that after 6 h of annealing, the Ge₁₅Sb₈₅ sample remained amorphous but with an increase in local structural order (Fig. 5e), which may be related to the enhancement of Peierls torsion. The Fourier transform infrared spectroscopy (FTIR) results show that the imaginary part of the dielectric constant decreases significantly after annealing (Fig. 5f), indicating that the local p-bonding structural distortion increases, which supports the Peierls distortion enhancement [46,71,72]. Although annealing can suppress β -relaxation, β -relaxation is not completely suppressed due to microstructural inhomogeneity and local atomic activation.

5.2. The relationship between β -relaxation and fragility-strength transition

Early studies have found that water under high pressure undergoes a liquid-liquid transition, i.e., a transition between low-density liquid water (LDL) and high-density liquid water (HDL) [74,75]. Similar transitions have been observed in some organic liquids and PCMs, which

indicate significant changes in structure and viscosity under different conditions.

The temperature dependence of viscosity is usually described by the Arrhenius equation, which shows stable applicability in materials such as silica and germanium dioxide GeO₂. However, near the glass transition temperature, some liquids exhibit non-Arrhenius behavior, and liquids can be classified as strong or brittle based on the degree to which the viscosity deviates from the equation [76,77]. While the fragile-to-strong (F-S) behavior of metallic glasses has been intensively studied, PCMs face more challenges due to the complex structural and energetic changes during the phase transition, where both the rate and stability of the phase transition are affected by the F-S behavior [78–80].

In the $(GeTe)_x(GeSe)_{100-x}$ system, researchers explored the relationship between the F-S transition and β-relaxation by adjusting the ratio of fast-crystallizing, brittle GeTe to thermally stable, strong GeSe. Despite β-relaxation properties [78], GeTe alone does not exhibit F-S transition behavior. However, when the GeTe content decreases below 85 %, the brittleness change (f) is consistent with the W_{ex} change, especially the positive correlation between F-S and β -relaxation at GeTe contents between 40 % and 70 % (Fig. 6c and d), where (GeTe)₅₈(GeSe)₄₂ exhibits a clear F-S transition, and Raman spectra show that a plateau exists in this range, suggesting that the GeTe content has a high F-S transition behavior (Fig. 6a). The presence of a plateau suggests a strong structural competition between GeTe and GeSe (Fig. 6b), and this apparent transition is driven by the dynamic heterogeneity of the amorphous structure, in which the restricted motion of GeSe and the activated motion of GeTe decouple the β and α -relaxations above T_{g} (Fig. 6e). This decoupling affects the viscosity and creates distinct F-S dynamic properties [78].

Although there is no direct evidence for a relationship between Peierls twist and F-S, previous studies have linked Peierls twist to β -relaxation. Brittle GeTe and strong GeSe provide an opportunity to explore this potential relationship. Although traditional PCMs (e.g., GeTe, AIST, and GST) do not show a direct link between β -relaxation,



Fig. 6. (a) Viscosity and viscosity activation energy variation for the (GeTe)₅₈(GeSe)₄₂ mixture with higher GeTe content (58 %) [78]. (b) Structural ratio of GeTe and GeSe in (GeTe)_x(GeSe)_{100-x} films [78].(c) Relationship between GeTe content and the F-S magnitude *f* (left vertical axis) and the excess wing ratio W_{ex} (right vertical axis) in (GeTe)_x(GeSe)_{100-x} films [78]. Panels (a) ~ (c) are reproduced with permission Copyright 2023, Elsevier. (d) The change in excess wing ratio with increasing GeSe content [73]. Reproduced with permission, Copyright 2020, AAAS. (e) The relation between F-S transition and β-relaxation. [78]Reproduced with permission. Copyright 2023, Elsevier.

and F-S, combining phase change materials with non-PCMs opens new avenues of investigation and has the potential to reveal hidden correlations between different materials.

6. Optimization strategies for PCM

Multiple optimization methods have been developed to improve the performance of PCMs. Improvements in programming algorithms, including precise current control and timing techniques, reduce energy consumption during write and erase processes while increasing speed and reliability. Advanced thermal management technologies use thermal isolation materials to enhance efficiency [81,82]. Additionally, 3D integration technology boosts storage density through vertical stacking of units, optimizing space utilization and reducing energy consumption [83].

Enhancing PCM performance requires a multi-scale approach, integrating nanostructure design and chemical composition tuning to regulate atomic dynamics and energy transport. Nanostructure designincluding nanoscale confinement, heterostructures, and superlattices—progressively enhances phase stability by limiting atomic diffusion, optimizing interface effects, and leveraging quantum confinement. These strategies not only address distinct kinetic challenges but also exhibit synergistic effects, such as heterointerfaces reinforcing superlattice stability.

Chemical tuning (e.g., doping) modifies defect states and phasetransition kinetics by adjusting electronic structures and bonding characteristics. The dual approach of physical confinement and chemical regulation ensures structural stability while optimizing electrical and thermal properties. This framework translates theoretical insights into practical design strategies, providing a pathway for high-density, lowpower PCM devices.

6.1. Nanostructure design

6.1.1. Nano-confinement

In studies of ultrathin antimony films, researchers reduced the thickness of the films to approximately 5 nm to obtain a stable amorphous antimony phase [84]. T_c increased with decreasing film thickness, as did the crystallization activation energy (E_a) , indicating that the amorphous phase was more thermally stable. All three amorphous antimony films transitioned rapidly to the crystalline phase when T_c was reached, as evidenced by a sharp decrease in the resistance of the thin layer (Fig. 7c) [84]. By conducting aging experiments on amorphous antimony films of different thicknesses, it was found that the 3 nm and 4 nm thick films remained amorphous for a longer period and showed stronger resistance to recrystallization, whereas the 5 nm thick films were more prone to crystallization. This suggests that thinner antimony films have better structural stability (Fig. 7e). However, thinner films (especially 3 nm antimony films) exhibit more pronounced resistance drift [84]. It was shown that deeply annealed antimony film samples and rapidly quenched devices exhibit ultra-low drift performance, whereas sputter-deposited films exhibit higher drift indices during the initial ~ 2 h aging phase. On the one hand, this difference may be due to the different initial microstructures of the two types of amorphous antimony, leading to different subsequent relaxation paths. To investigate this phenomenon, the researchers simulated amorphous antimony films in two different states, representing the less-equilibrium (LE) and more-equilibrium (ME) structures, respectively, by molecular dynamics (Fig. 7a and b), and found that there are differences in their relaxation behaviors and drift properties after annealing [84]. The LE state exhibits a stronger electronic localization, especially at the Sb/SiO₂ interface, which strengthens covalent bonding, stabilizes the disordered structure, and prevents self-ordering. The disordered structure prevents spontaneous recrystallization. On the other hand, thinner antimony films



Fig. 7. (a) Amorphous LE model in silica glass after annealing at 300 K for 87 ps, showing the average ELF value along the Sb layer's thickness [85]. (b) Amorphous ME model and average ELF value along the Sb thickness [85]. (c) Temperature dependence of sheet resistance for Sb films (3, 4, and 5 nm) encapsulated by SiO₂ [85]. (d) 10-year data retention of thin Sb samples [85]. (e) Sheet resistance over time for as-sputtered Sb samples at room temperature [85]. Panels (a) \sim (e) are reproduced under terms of the CC-BY license, Copyright 2023, Bin Chen, published by Wiley-VCH. (f) The resistance measurements for PCM arrays with GeTe layers of 3, 10, and 100 nm thickness over time at 85 °C [86]. Reproduced with permission. Copyright 2020, AIP.

exhibit higher initial resistances and faster phase transitions due to more pronounced interfacial effects and Peierls distortions [84]. These differences explain why the performance of different antimony films in terms of resistance drift can vary.

Upon comparing the ultrathin antimony films, we found that the GeTe nanoscale films also exhibit significant physical property changes as the thickness is reduced to the nanoscale. Notably, the thermal stability and resistive drift properties of GeTe films in the amorphous state improve significantly with film thickness reduction. The slower transition of GeTe to the crystalline phase upon reaching its crystallization temperature compared to antimony thin films is mainly attributed to the higher local ordering and stability provided by the added tetrahedral structures in GeTe, which effectively retard the crystallization process and allow GeTe to remain amorphous over a wider temperature range. In terms of resistive drift, GeTe ultrathin films exhibit extremely small resistive drift, which is closely related to the stability and orderliness of their internal structure [86]. In GeTe ultrathin films, the highly ordered tetrahedral structure helps to reduce the localization of electrons, which not only suppresses the extent of resistive drift but also enhances the thermal stability of the material [86]. The stability and order of this structure provide better resistive stability, and GeTe exhibits lower performance degradation in sustained use compared to antimony films.

Upon comparing single-atom Sb and GeTe ultrathin films in PCM devices, it can be seen that there is a significant difference in performance between the two materials. The monatomic Sb ultrathin film does not contain the element Te, so it is likely to be more stable in the

amorphous phase, which may cause fewer conductivity disturbances in GeTe through the formation of valence alternating pairs (VAPs) [86]. This difference means that resistance drift and aging phenomena may be lower in amorphous Sb, which can improve the reliability and stability of the PCM. GeTe ultrathin films, on the other hand, despite exhibiting excellent thermal stability and low resistive drift at nanoscale thicknesses, their complex chemical structure-especially the presence of Te-may lead to the risk of conductivity disturbances in long-term applications, as evidenced by the ease with which lone pairs of electrons in the chain of Te atoms can form localized charged defect states. The role of these defect states in the conductance can change over time, potentially affecting the material's performance.

These differences highlight the influence of a material's internal structure on its electronic performance at the nanoscale and emphasize the need to consider film thickness, internal structural ordering, and the microstructural properties of the material when designing nextgeneration, high-performance amorphous phase memory devices. Through a deeper understanding and exploitation of these factors, the performance of PCM devices can be further optimized to meet the demands of more demanding applications.

6.1.2. Heterostructure

Recently, researchers optimized the performance of PCM devices by designing mushroom-shaped programming regions and a multilayer phase change heterostructure (PCH) (Fig. 8a and b). The structure consists of alternating nanoscale-constrained material (CM) layers and



Fig. 8. (a) and (b) Schematic representations of PCM and PCH cells [44]. (c) PCH devices show stable resistance levels in both RESET and SET states, unlike GST devices [44]. (d) Relationship between SET speed and voltage bias for PCH and GST devices of the same geometry [44]. (e) AIMD simulation of the TiTe₂/Sb₂Te₃ PCH model at high temperatures, with Ti, Sb, and Te atoms represented as red, yellow, and blue spheres, respectively [44]. Panels (a) \sim (e) are reproduced with permission Copyright 2019, AAAS. (f) Structural diagram of Sb₂Te₃/GeTe superlattices (SLs) [87]. (g) In-plane thermal conductivity of 60 nm thick Sb₂Te₃/GeTe SL films at room temperature, varying with period size and thickness ratio [87]. Panels (f) and (g) are reproduced under the terms of the CC-BY license. Copyright 2021, Heungdong Kwon, published by ACS. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

phase change material (PCM) layers, with $TiTe_2$ as the CM and Sb_2Te_3 as the PCM, with layer thicknesses of about 3 nm and 5 nm, respectively. The cycle life of PCH devices is improved by several orders of magnitude compared to conventional GST devices (Fig. 8c), which is attributed to the effective design of the confinement layer that limits atomic migration and reduces compositional and structural variations, resulting in improved data storage accuracy [44].

The TiTe₂ layer acts as a robust separator layer that not only isolates the neighboring Sb₂Te₃ sublayers but also inhibits chemical interactions between them and ensures structural stability during high-temperature crystallization. The layer also serves as a thermal barrier to concentrate thermal energy and reduce energy consumption, and its excellent electrical conductivity promotes a more uniform current transfer between the Sb₂Te₃ sublayers, which improves the device performance and reliability. The TiTe₂ layer significantly improves the crystallization rate of the Sb₂Te₃ (Fig. 8d), which may stem from the interfacial interactions to provide additional nucleation sites and accelerate the crystallization process. In addition, the PCH structure effectively reduces the resistive drift, and the lack of Ge and tetrahedral defects in the amorphous Sb₂Te₃ eliminates the driving force for structural relaxation. The TiTe₂ layer significantly slows down the diffusion of Sb and Te atoms and maintains a good atomic arrangement (Fig. 8e). A recent study has shown that different confining material layers (CM layers) have a significant effect on the performance of PCM devices with phase change heterostructure (PCH). Such as NiTe2 and MoTe2, two different CM layers, these new materials exhibit their unique advantages and limitations compared to the conventional TiTe₂ [88].

TiTe₂ can maintain structural stability during high-temperature crystallization due to its high thermal stability and good chemical stability, which is crucial for long-term data storage reliability. In contrast, NiTe₂'s high conductivity allows it to realize phase transition at lower voltages, showing potential in low-power operation, however, this may also lead to thermal buildup at high current densities, which can affect the long-term stability of the device [89]. MoTe₂, on the other hand, shows high thermal stability thanks to its high melting point and excellent thermal barrier effect, which allows it to sustain its performance over long periods and at high temperatures and is less susceptible to thermal degradation. In addition, the robust atomic structure of MoTe₂ effectively limits atomic migration during high-temperature operation, thereby enhancing device durability and stability [89]. The chemical stability and interfacial compatibility of each material also play a decisive role in the performance of the CM layer. Good compatibility reduces chemical reactions at the interface, keeps the interface clear, and improves the response speed and repeatability of the device. Combining these properties, MoTe₂ shows better performance, especially in applications requiring high stability and long-term reliability, while NiTe2 is suitable for specific applications due to its fast switching and low energy consumption [89]. And TiTe2's stability and durability make it ideal for long-term and high-reliability storage applications [89].

6.1.3. Superlattices

Superlattices, such as Sb₂Te₃/GeTe multilayers (Fig. 8f), transcend conventional heterostructures by integrating periodic interfaces to manipulate thermal and electronic properties at the quantum level. In these architectures, thermal conductivity is modulated by interface density and layer thickness. Increasing the Sb₂Te₃/GeTe thickness ratio (e.g., 4:1) enhances interfacial phonon scattering, reducing in-plane thermal conductivity by 40 % (Fig. 8g). This reduction arises from the mismatch in vibrational modes between Sb₂Te₃'s layered structure and GeTe's tetrahedral order, which disrupts phonon propagation. Concurrently, ultrathin superlattice layers (<2 nm) minimize electron scattering, achieving vertical resistivity values as low as one-fifth of bulk materials [90]. The tetrahedral order in GeTe layers and the layered structure of Sb₂Te₃ collaboratively suppress electron localization in the amorphous phase, mitigating resistance drift while lowering operational energy consumption.

The quantum confinement effects in superlattices further enhance performance. For instance, in Sb₂Te₃/GeTe superlattices, the confinement of charge carriers within GeTe layers alters the band structure, reducing the density of mid-gap states that contribute to resistance drift. This effect is amplified in thinner layers, where quantum confinement dominates over bulk-like behavior. However, practical challenges such as atomic layer intermixing and stacking faults during fabrication demand atomic-level precision. Techniques like atomic layer deposition (ALD) enable the synthesis of near-perfect interfaces, but defects such as Te vacancies or Sb-Ge antisite defects can still degrade performance. Recent advances in in-situ TEM characterization have provided insights into interface dynamics, revealing that interfacial strain fields in superlattices can stabilize metastable phases and delay crystallization [91].

By harmonizing nanoconfinement and interfacial engineering, superlattices exemplify the pinnacle of hierarchical design—where static confinement and dynamic quantum effects coalesce to decouple thermal and electrical transport. For example, in a GeTe/Sb₂Te₃ superlattice, the GeTe layers act as quantum wells for electrons, while the Sb₂Te₃ layers serve as phonon-blocking barriers. This decoupling enables independent optimization of thermal and electrical properties, a feat unattainable in homogeneous materials.

6.2. Defect engineering

Defect engineering involves the deliberate introduction, manipulation, and control of atomic-level defects in the crystalline or amorphous structure of a material [92]. These defects may include vacancies, gaps, and substituted atoms, which affect the electronic, thermal, and structural properties of the material. By managing these defects, researchers can tailor phase transition properties, improve thermal conductivity, and modulate the energy barriers of switches, leading to more efficient and reliable PCM devices.

Recent studies have shown that the properties of PCMs are significantly improved by doping GeTe nanowires with Bi to modulate the defects. TEM images show that the pristine GeTe nanowires are almost defect-free, whereas GBT8 nanowires doped with 8 % Bi are significantly disordered (Fig. 9a and b). The increase in the concentration of Bi leads to a widening of the lattice spacing, which is accompanied by an increase in the lattice strain, which not only induces a structural disorder but also affects the phase-transition behavior of the material. This strain not only causes structural disorder but also affects the phase transition behavior and electron transport properties of the material. The bismuthdoped samples show higher resistivity at low temperatures, indicating an enhanced carrier localization effect, which lowers the energy barrier for the transition from the crystalline to the amorphous state and improves the energy conversion efficiency of the phase transition process (Fig. 9c). The GBT8 nanowire device reaches an intermediate resistive state at ~0.9 V and a highly resistive RESET state at ~1.2 V (Fig. 9d and e). By controlling the electrical pulse parameters, different degrees of defect accumulation can be achieved, resulting in various stable resistive states (Fig. 9f). This sequential switching (from the SET state to the intermediate state to the RESET state) demonstrates a key advantage over quenched systems, where the state transitions are more difficult to precisely control and often require the entire system to return to the ground state to reach the desired state.

In addition to doping to introduce defects, ion irradiation, and ion implantation are commonly used methods. High-energy helium ion irradiation creates defects in GeTe nanowires, strengthening carrierlattice interactions. This enhancement increases energy exchange efficiency while reducing heat loss. As a result, the current density required for amorphization decreases. By adjusting the ion dose, the type and density of defects can be precisely controlled. Devices irradiated with a specific dose have significantly lower current and power densities during the phase transition compared to conventional melt quenching



Fig. 9. (a) HRTEM images of pristine GeTe nanowires and (b) 8 % Bi-doped (GBT8) nanowires, showing increased disorder and lattice strain with Bi doping. Green arrows indicate the nanowire growth direction [93]. (c) Average programming current density for various dual-doping percentages, with error bars representing RESET current density variation [93]. (d) Crystalline-to-amorphous transitions in GBT8 nanowires as a function of programming voltage and pulse width [93]. (e) Transition after 2 cycles of 10 ns pulses in GBT8 nanowires [93]. (f) Schematic of defect-based amorphization in doped GeTe nanowires [93]. Panels (a) ~ (f) are reproduced under the terms of the CC-BY license. Copyright 2020, Gaurav Modi, published by ACS. (g) Scaling behavior of switching in defect-induced GeTe nanowire devices [94]. Reproduced under terms of the CC-BY license. Copyright 2016, Pavan Nukala, published by Springer Nature Limited. (h) Schematic diagram of ion implantation [95].Reproduced with permission. Copyright 2019, Trans Tech Publications. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(Fig. 9g), supporting polymorphic storage. Another method is ion implantation, where defects are introduced by argon ions implanted into the Sb_2Te_3 film (Fig. 9h), which reduces the energy demand during the phase transition and improves the phase transition efficiency. Tests show that RESET energy is reduced by about 20 % compared with unimplanted material, power consumption is also reduced, and the switching speed is increased to 100 ns, which enhances the stability of data storage.

7. Conclusion and outlook

This review systematically explores advancements in phase-change memory materials and analyzes their multi-scale characteristics, from macroscopic device behavior to atomic-level mechanisms. It proposes optimization strategies for next-generation non-volatile memory technologies. The study challenges conventional defect models by introducing the collective relaxation model, attributing resistance drift to cooperative atomic motions in the amorphous phase, offering a unified framework for understanding dynamic relaxation processes. The discovery of the ideal glassy state redefines the aging pathway of amorphous materials, revealing the role in suppressing abrupt phase transitions and delaying structural relaxation. Proposed strategies such as nanoscale confinement, superlattice engineering, and defect modulation have enhanced thermal stability and electrical reliability. For instance, superlattice structures reduce thermal conductivity and resistance drift through interface phonon scattering and quantum confinement effects, demonstrating the potential of atomic-scale engineering to improve device performance.

Despite these advances, PCM materials continue to face several challenges. First, phase separation at elevated temperatures or during prolonged operation limits long-term stability. Second, interface stress and atomic interdiffusion may compromise the reliability of multilayer structures. Furthermore, the high manufacturing costs and scalability limitations of current fabrication techniques impede large-scale deployment. The microscopic mechanisms underlying β -relaxation and the fragile-to-strong (F-S) transition remain elusive, necessitating further investigation through in-situ characterization and multi-scale simulations.

In considering future directions, several key areas could drive further advancements in phase-change materials. First, multi-element alloy optimization strategies should be investigated to develop material systems with enhanced thermal stability and reduced power consumption, thereby extending device lifespan and energy efficiency. Second, establishing an "atomic-to-device" integrated optimization platform will facilitate a seamless transition from dynamic relaxation mechanisms to device-scale architecture, enhancing memory density and endurance. Third, the application of PCM materials should be expanded into emerging fields, such as neuromorphic computing and reconfigurable optoelectronic devices, where their multi-state tunability can enable more efficient information processing. Finally, the development of environmentally sustainable fabrication technologies will promote the advancement of PCM materials towards low-power, eco-friendly solutions, fostering broader adoption in next-generation information technologies.

CRediT authorship contribution statement

Yan Chen: Writing – original draft, Visualization, Formal analysis. Yuqiao Le: Writing – original draft, Visualization. Lei Chen: Writing – review & editing, Visualization. Haisong Liu: Visualization, Validation. Tangyou Sun: Writing – review & editing, Resources. Xingpeng Liu: Writing – review & editing, Visualization. Fabi Zhang: Writing – review & editing, Visualization. Haiou Li: Writing – review & editing, Visualization. XinXin Hu: Writing – review & editing. Ying Peng: Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition. Chengyan Liu: Writing – review & editing, Supervision, Project administration. Min Hong: Writing – review & editing, Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was financially supported by the National Key Research and Development Program of China (No. 2022YFE0119100), the National Natural Science Foundation of China (Nos. 52061009, U21A2054, 52262022), Guangxi Science and Technology Planning Project (No. AD21220056 and AD24010060).

Data availability

Data will be made available on request.

References

- [1] F. Pellizzer, A. Redaelli, 8 3DXpoint fundamentals, in: A. Redaelli, F. Pellizzer (Eds.), Semicond. Mem. Syst., Woodhead Publishing, 2022, pp. 253–276, https:// doi.org/10.1016/B978-0-12-820758-1.00005-4.
- [2] M. Zabihi, S. Resch, H. Cilasun, Z.I. Chowdhury, Z. Zhao, U.R. Karpuzcu, J.-P. Wang, S.S. Sapatnekar, Exploring the feasibility of using 3-D XPoint as an inmemory computing accelerator, IEEE J. Explor. Solid-State Comput. Dev. Circ. 7 (2021) 88–96, https://doi.org/10.1109/JXCDC.2021.3112238.
- [3] J. Guo, H. Wang, W. Jing, H. Li, Y. Du, Z. Song, B. Chen, Subsystem under 3Dstorage class memory on a chip, Comput. Electr. Eng. 74 (2019) 47–58, https://doi. org/10.1016/j.compeleceng.2019.01.009.
- [4] J. Yang, B. Li, D. Lilja, Exploring performance characteristics of the optane 3D xpoint storage technology, ACM Trans. Model. Perform. Eval. Comput. Syst. 5 (2020) 1–28, https://doi.org/10.1145/3372783.
- [5] A. Bafna, Intel's Optane Memory: Revolutionising Data Storage with 3D XPoint Technology. https://doi.org/10.13140/RG.2.2.35906.80320, 2019.
- [6] K. Bourzac, Has Intel created a universal memory technology? IEEE Spectr. 54 (2017) 9–10, https://doi.org/10.1109/MSPEC.2017.7906883 [News].
- [7] G. Molas, E. Nowak, Advances in emerging memory technologies: from data storage to artificial intelligence, Appl. Sci. 11 (2021) 11254, https://doi.org/ 10.3390/app112311254.
- [8] V. Egloff, Exploration and conception of computing architectures of type computing in-memory based on emerging non volatile memories, phdthesis, Aix-Marseille Université, 2022. https://hal.science/tel-04055973. (Accessed 8 November 2024).
- [9] H. Zhang, G. Chen, B.C. Ooi, K.-L. Tan, M. Zhang, In-memory big data management and processing: a survey, IEEE Trans. Knowl. Data Eng. 27 (2015) 1920–1948, https://doi.org/10.1109/TKDE.2015.2427795.
- [10] A. Beloglazov, J. Abawajy, R. Buyya, Energy-aware resource allocation heuristics for efficient management of data centers for Cloud computing, Future Gener. Comput. Syst. 28 (2012) 755–768, https://doi.org/10.1016/j.future.2011.04.017.
- [11] A. Beloglazov, R. Buyya, Y.C. Lee, A. Zomaya, Chapter 3 a taxonomy and survey of energy-efficient data centers and cloud computing systems, in: M.V. Zelkowitz (Ed.), Adv. Comput., Elsevier, 2011, pp. 47–111, https://doi.org/10.1016/B978-0-12-385512-1.00003-7.
- [12] A. Arif, V. Reddy, K. Srividya, U. Mallampalli, Utilizing multilayer perceptron for machine learning diagnosis in phase change material-based thermal management systems, Heat Transf. 53 (2024) 4922–4947, https://doi.org/10.1002/htj.23163.
- [13] T.-H. Nguyen, M. Imran, J. Choi, J.-S. Yang, HYDRA: a hybrid resistance drift resilient architecture for phase change memory-based neural network accelerators, IEEE Trans. Comput. 73 (2024) 2123–2135, https://doi.org/10.1109/ TC.2024.3404096.

- [14] S. Raoux, R. Shelby, J. Jordan-Sweet, B. Munoz, M. Salinga, Y.-C. Chen, Y.-H. Shih, E.-K. Lai, M.-H. Lee, Phase change materials and their application to random access memory technology, Microelectron. Eng. 85 (2008) 2330–2333, https://doi.org/ 10.1016/j.mee.2008.08.004.
- [15] M. Le Gallo, Phase-Change Memory: Device Physics and Application to Non-von Neumann Computing, ETH Zurich, 2017, https://doi.org/10.3929/ethz-b-000250342, Doctoral Thesis.
- [16] G.W. Burr, M.J. Breitwisch, M. Franceschini, D. Garetto, K. Gopalakrishnan, B. Jackson, B. Kurdi, C. Lam, L.A. Lastras, A. Padilla, B. Rajendran, S. Raoux, R. S. Shenoy, Phase change memory technology, J. Vac. Sci. Technol. B Nanotechnol. Microelectron. Mater. Process. Meas. Phenom. 28 (2010) 223–262, https://doi. org/10.1116/1.3301579.
- [17] Z. Lv, Y. Wang, J. Chen, J. Wang, Y. Zhou, S.-T. Han, Semiconductor quantum dots for memories and neuromorphic computing systems, Chem. Rev. 120 (2020) 3941–4006, https://doi.org/10.1021/acs.chemrev.9b00730.
- [18] N. Thirumalaivasan, S. Gopi, K. Karthik, S. Nangan, K. Kanagaraj, S. Rajendran, Nano-PCM materials: bridging the gap in energy storage under fluctuating environmental conditions, Process Saf. Environ. Prot. 189 (2024) 1003–1021, https://doi.org/10.1016/j.psep.2024.06.079.
- [19] A. Chen, A review of emerging non-volatile memory (NVM) technologies and applications, Solid State Electron. 125 (2016) 25–38, https://doi.org/10.1016/j. sse.2016.07.006.
- [20] C.L. Philip Chen, C.-Y. Zhang, Data-intensive applications, challenges, techniques and technologies: a survey on Big Data, Inf. Sci. 275 (2014) 314–347, https://doi. org/10.1016/j.ins.2014.01.015.
- [21] A. Arshad, M. Jabbal, Y. Yan, J. Darkwa, The micro-/nano-PCMs for thermal energy storage systems: a state of art review, Int. J. Energy Res. 43 (2019), https:// doi.org/10.1002/er.4550.
- [22] J. Curry, A. Louri, A. Karanth, R. Bunescu, PCM Enabled Low-Power Photonic Accelerator for Inference and Training on Edge Devices, IEEE Computer Society, 2024, pp. 600–607, https://doi.org/10.1109/IPDPSW63119.2024.00118.
- [23] B. Jahannia, J. Ye, S. Altaleb, C. Patil, E. Heidari, V.J. Sorger, N. Peserico, H. Dalir, Optical memory in IoT devices: an energy-efficient approach using phase change materials, in: AI Opt. Data Sci. V, SPIE, 2024, pp. 11–24, https://doi.org/10.1117/ 12.3003184.
- [24] H. Nakamura, T. Nakada, S. Miwa, Normally-off computing project: challenges and opportunities, in: 2014 19th Asia S. Pac. Des. Autom. Conf. ASP-DAC, 2014, pp. 1–5, https://doi.org/10.1109/ASPDAC.2014.6742850.
- [25] Energy-efficient memcapacitor devices for neuromorphic computing | Nat. Electr., (n.d.). https://www.nature.com/articles/s41928-021-00649-y?fromPaywallRec =true (accessed November 12, 2024).
- [26] L. Wang, G. Ma, S. Yan, X. Cheng, X. Miao, Reconfigurable multilevel storage and neuromorphic computing based on multilayer phase-change memory, ACS Appl. Mater. Interfaces 16 (2024) 54829–54836, https://doi.org/10.1021/ acsami.4c11087.
- [27] N. Ciocchini, M. Laudato, M. Boniardi, E. Varesi, P. Fantini, A.L. Lacaita, D. Ielmini, Bipolar switching in chalcogenide phase change memory, Sci. Rep. 6 (2016) 29162, https://doi.org/10.1038/srep29162.
- [28] S. Abdollahramezani, O. Hemmatyar, H. Taghinejad, A. Krasnok, Y. Kiarashinejad, M. Zandehshahvar, A. Alù, A. Adibi, Tunable nanophotonics enabled by chalcogenide phase-change materials, Nanophotonics 9 (2020) 1189–1241, https://doi.org/10.1515/nanoph-2020-0039.
- [29] M.L. Gallo, A. Sebastian, An overview of phase-change memory device physics, J. Phys. Appl. Phys. 53 (2020) 213002, https://doi.org/10.1088/1361-6463/ ab7794.
- [30] N.H.A. ElHassan, Development of Phase Change Memory Cell Electrical Circuit Model for Non-volatile Multistate Memory Device, (n.d.).
- [31] T. Titirsha, MdM.H. Shuvo, S. Islam, Reliability Analysis of Phase Change Memory-Based Neuromorphic Circuits, 2024, https://doi.org/10.1109/ ISVLSI61997.2024.00144.
- [32] D. Ielmini, D. Sharma, S. Lavizzari, A. Lacaita, Reliability impact of chalcogenidestructure relaxation in phase-change memory (PCM) cells—Part I: experimental study, electron devices, IEEE Trans. On 56 (2009) 1070–1077, https://doi.org/ 10.1109/TED.2009.2016397.
- [33] Z. Li, Y. Gong, A. Xu, J. Zhao, Q. Li, L. Dong, C. Xiong, M. Jiang, Relaxationinduced significant room-temperature dielectric pulsing effects, Adv. Funct. Mater. 33 (2023) 2301009, https://doi.org/10.1002/adfm.202301009.
- [34] W. Zhang, E. Ma, Unveiling the structural origin to control resistance drift in phasechange memory materials, Mater. Today 41 (2020) 156–176, https://doi.org/ 10.1016/j.mattod.2020.07.016.
- [35] M. Boniardi, D. Ielmini, S. Lavizzari, A. Lacaita, A. Redaelli, A. Pirovano, Statistics of resistance drift due to structural relaxation in phase-change memory arrays, Electr. Dev. IEEE Trans. On 57 (2010) 2690–2696, https://doi.org/10.1109/ TED.2010.2058771.
- [36] D. Ielmini, S. Lavizzari, D. Sharma, A.L. Lacaita, Physical interpretation, modeling and impact on phase change memory (PCM) reliability of resistance drift due to chalcogenide structural relaxation, 2008, pp. 939–942, https://doi.org/10.1109/ IEDM.2007.4419107.
- [37] M. Gallo, D. Krebs, F. Zipoli, M. Salinga, A. Sebastian, Collective structural relaxation in phase-change memory devices, Adv. Electron. Mater. 4 (2018) 1700627, https://doi.org/10.1002/aelm.201700627.
- [38] C. Ahn, B. Lee, R.G.D. Jeyasingh, M. Asheghi, F. Hurkx, K.E. Goodson, H.-S. P. Wong, Effect of resistance drift on the activation energy for crystallization in phase change memory, Jpn. J. Appl. Phys. 51 (2012) 02BD06, https://doi.org/ 10.1143/JJAP.51.02BD06.

- [39] J.Y. Raty, V. Godlevsky, Ph Ghosez, C. Bichara, J.P. Gaspard, J.R. Chelikowsky, Evidence of a reentrant Peierls distortion in liquid GeTe, Phys. Rev. Lett. 85 (2000) 1950–1953, https://doi.org/10.1103/PhysRevLett.85.1950.
- [40] G.C. Sosso, J. Colombo, J. Behler, E. Del Gado, M. Bernasconi, Dynamical heterogeneity in the supercooled liquid state of the phase change material GeTe, J. Phys. Chem. B 118 (2014) 13621–13628, https://doi.org/10.1021/jp507361f.
- [41] P. Noe, C. Sabbione, N. Castellani, G. Veux, G. Navarro, V. Sousa, F. Hippert, F. D'Acapito, Structural change with the resistance drift phenomenon in amorphous GeTe phase change materials' thin films, J. Phys. Appl. Phys. 49 (2015) 035305, https://doi.org/10.1088/0022-3727/49/3/035305.
- [42] M. Rütten, M. Kaes, A. Albert, M. Wuttig, M. Salinga, Relation between bandgap and resistance drift in amorphous phase change materials, Sci. Rep. 5 (2015) 17362, https://doi.org/10.1038/srep17362.
- [43] F. Zipoli, D. Krebs, A. Curioni, Structural origin of resistance drift in amorphous GeTe, Phys. Rev. B 93 (2016), https://doi.org/10.1103/PhysRevB.93.115201.
- [44] K. Ding, J. Wang, Y. Zhou, H. Tian, L. Lu, R. Mazzarello, C. Jia, W. Zhang, F. Rao, E. Ma, Phase-change heterostructure enables ultralow noise and drift for memory operation, Science 366 (2019) 210–215, https://doi.org/10.1126/science. aav0291.
- [45] H. Li, J. Robertson, A unified mid-gap defect model for amorphous GeTe phase change material, Appl. Phys. Lett. 116 (2020) 052103, https://doi.org/10.1063/ 1.5138998.
- [46] N. Amini, J. Pries, Y. Cheng, C. Persch, M. Wuttig, M. Stolpe, S. Wei, Thermodynamics and kinetics of glassy and liquid phase-change materials, Mater. Sci. Semicond. Process. 135 (2021) 106094, https://doi.org/10.1016/j. mssp.2021.106094.
- [47] B. Kersting, Quest for a solution to drift in phase change memory devices, 2024, https://doi.org/10.48550/arXiv.2401.09462.
- [48] B. Kersting, S.G. Sarwat, M.L. Gallo, K. Brew, S. Walfort, N. Saulnier, M. Salinga, A. Sebastian, Measurement of onset of structural relaxation in melt-quenched phase change materials, 2021. http://arxiv.org/abs/2106.06270. (Accessed 7 July 2024).
- [49] YuK. Tovbin, Second law of thermodynamics, Gibbs' thermodynamics, and relaxation times of thermodynamic parameters, Russ. J. Phys. Chem. A 95 (2021) 637–658, https://doi.org/10.1134/S0036024421020266.
- [50] Y. Nishikawa, L. Berthier, Collective relaxation dynamics in a three-dimensional lattice glass model, 2024, https://doi.org/10.48550/arXiv.2307.08110.
- [51] J.-Y. Raty, Aging in phase change materials: getting insight from simulation, Phys. Status Solidi RRL - Rapid Res. Lett. 13 (2019) 1800590, https://doi.org/10.1002/ pssr.201800590.
- [52] S. Gabardi, S. Caravati, G. Sosso, J. Behler, M. Bernasconi, Microscopic origin of resistance drift in the amorphous state of the phase-change compound GeTe, Phys. Rev. B 92 (2015), https://doi.org/10.1103/PhysRevB.92.054201.
- [53] J.Y. Raty, W. Zhang, J. Luckas, C. Chen, R. Mazzarello, C. Bichara, M. Wuttig, Aging mechanisms in amorphous phase-change materials, Nat. Commun. 6 (2015) 7467, https://doi.org/10.1038/ncomms8467.
- [54] S. Gabardi, E. Baldi, E. Bosoni, D. Campi, S. Caravati, G. Sosso, J. Behler, M. Bernasconi, Atomistic simulations of the crystallization and aging of GeTe nanowires, J. Phys. Chem. C 121 (2017), https://doi.org/10.1021/acs. jpcc.7b09862.
- [55] A.C. Galca, F. Sava, I.D. Simandan, C. Bucur, V. Dumitru, C. Porosnicu, C. Mihai, A. Velea, Structural and optical properties of optimized amorphous GeTe films for memory applications, J. Non-Cryst. Solids 499 (2018) 1–7, https://doi.org/ 10.1016/j.jnoncrysol.2018.07.007.
- [56] T. Fujita, Y. Chen, Y. Kono, S. Takahashi, H. Kasai, D. Campi, M. Bernasconi, K. Ohara, H. Yumoto, T. Koyama, H. Yamazaki, Y. Senba, H. Ohashi, I. Inoue, Y. Hayashi, M. Yabashi, E. Nishibori, R. Mazzarello, S. Wei, Pressure-induced reversal of Peierls-like distortions elicits the polyamorphic transition in GeTe and GeSe, Nat. Commun. 14 (2023) 7851, https://doi.org/10.1038/s41467-023-43457-v.
- [57] J.-P. Gaspard, Structure of covalently bonded materials: from the Peierls distortion to Phase-Change Materials, C. R. Phys. 17 (2016) 389–405, https://doi.org/ 10.1016/j.crhy.2015.12.009.
- [58] K. Ghosh, A. Kusiak, P. Noé, M.-C. Cyrille, J.-L. Battaglia, Thermal conductivity of amorphous and crystalline GeTe thin film at high temperature: experimental and theoretical study, Phys. Rev. B 101 (2020) 214305, https://doi.org/10.1103/ PhysRevB.101.214305.
- [59] S. Wintersteller, O. Yarema, D. Kumaar, F.M. Schenk, O.V. Safonova, P.M. Abdala, V. Wood, M. Yarema, Unravelling the amorphous structure and crystallization mechanism of GeTe phase change memory materials, Nat. Commun. 15 (2024) 1011, https://doi.org/10.1038/s41467-024-45327-7.
- [60] K. Konstantinou, F.C. Mocanu, J. Akola, S.R. Elliott, Electric-field-induced annihilation of localized gap defect states in amorphous phase-change memory materials, Acta Mater. 223 (2022) 117465, https://doi.org/10.1016/j. actamat.2021.117465.
- [61] D. Lee, K. Lee, D. Yoo, W. Jeong, S. Han, Crystallization of amorphous GeTe simulated by neural network potential addressing medium-range order, Comput. Mater. Sci. 181 (2020) 109725, https://doi.org/10.1016/j. commatsci.2020.109725.
- [62] R. Mantovan, R. Fallica, A. Mokhles Gerami, T.E. Mølholt, C. Wiemer, M. Longo, H. P. Gunnlaugsson, K. Johnston, H. Masenda, D. Naidoo, M. Ncube, K. Bharuth-Ram, M. Fanciulli, H.P. Gislason, G. Langouche, S. Ólafsson, G. Weyer, Atomic-scale study of the amorphous-to-crystalline phase transition mechanism in GeTe thin films, Sci. Rep. 7 (2017) 8234, https://doi.org/10.1038/s41598-017-08275-5.
- [63] H. Weber, M. Schumacher, P. Jóvári, Y. Tsuchiya, W. Skrotzki, R. Mazzarello, I. Kaban, Experimental and ab initio molecular dynamics study of the structure and

physical properties of liquid GeTe, Phys. Rev. B 96 (2017), https://doi.org/ 10.1103/PhysRevB.96.054204.

- [64] D. Acharya, O. Abou El Kheir, S. Perego, D. Campi, M. Bernasconi, Atomistic simulations of the crystallization of amorphous GeTe nanoparticles, J. Phys. Chem. C (2024), https://doi.org/10.1021/acs.jpcc.4c05157.
- [65] O. Yarema, A. Perevedentsev, V. Ovuka, P. Baade, S. Volk, V. Wood, M. Yarema, Colloidal phase-change materials: synthesis of monodisperse GeTe nanoparticles and quantification of their size-dependent crystallization, Chem. Mater. 30 (2018) 6134–6143, https://doi.org/10.1021/acs.chemmater.8b02702.
- [66] B. Chen, D. de Wal, G.H. ten Brink, G. Palasantzas, B.J. Kooi, Resolving crystallization kinetics of GeTe phase-change nanoparticles by ultrafast calorimetry, Cryst. Growth Des. 18 (2018) 1041–1046, https://doi.org/10.1021/ acs.cgd.7b01498.
- [67] W. Götze, L. Sjögren, β-relaxation in supercooled liquids, J. Non-Cryst. Solids 131–133 (1991) 161–168, https://doi.org/10.1016/0022-3093(91)90292-E.
- [68] J. Pries, C. Stenz, L. Schäfer, A. Gutsche, S. Wei, P. Lucas, M. Wuttig, Resistance drift convergence and inversion in amorphous phase change materials, Adv. Funct. Mater. 32 (2022) 2207194, https://doi.org/10.1002/adfm.202207194.
- $\label{eq:based} \begin{array}{l} \mbox{[69]} M. \mbox{ Sellitto, Crossover from β to α relaxation in cooperative facilitation dynamics, $$ Phys. Rev. Lett. 115 (2015), https://doi.org/10.1103/PhysRevLett.115.225701. \\ \end{array}$
- [70] S. Capaccioli, K.L. Ngai, N. Shinyashiki, The Johari–Goldstein β-relaxation of water, J. Phys. Chem. B 111 (2007) 8197–8209, https://doi.org/10.1021/ jp071857m.
- [71] Y. Cheng, Q. Yang, J. Wang, T. Dimitriadis, M. Schumacher, H. Zhang, M.J. Müller, N. Amini, F. Yang, A. Schoekel, J. Pries, R. Mazzarello, M. Wuttig, H.-B. Yu, S. Wei, Highly tunable β-relaxation enables the tailoring of crystallization in phase-change materials, Nat. Commun. 13 (2022) 7352, https://doi.org/10.1038/s41467-022-35005-x.
- [72] J. Moesgaard, T. Fujita, S. Wei, Unveiling the boson peaks in amorphous phasechange materials. https://doi.org/10.48550/arXiv.2404.17226, 2024.
- [73] S.-X. Peng, Y. Cheng, J. Pries, S. Wei, H.-B. Yu, M. Wuttig, Uncovering β-relaxations in amorphous phase-change materials, Sci. Adv. 6 (2020) eaay6726, https://doi. org/10.1126/sciadv.aay6726.
- [74] G.P. Johari, J. Teixeira, Thermodynamic analysis of the two-liquid model for anomalies of water, HDL-LDL fluctuations, and liquid-liquid transition, J. Phys. Chem. B 119 (2015) 14210–14220, https://doi.org/10.1021/acs.jpcb.5b06458.
- [75] G. Franzese, G. Malescio, A. Skibinsky, S.V. Buldyrev, H.E. Stanley, Generic mechanism for generating a liquid–liquid phase transition, Nature 409 (2001) 692–695, https://doi.org/10.1038/35055514.
- [76] S. Sathivel, J. Huang, W. Prinyawiwatkul, Thermal properties and applications of the Arrhenius equation for evaluating viscosity and oxidation rates of unrefined pollock oil, J. Food Eng. 84 (2008) 187–193, https://doi.org/10.1016/j. jfoodeng.2007.04.027.
- [77] V.V. Hoang, N.H. Tuan Anh, H. Zung, Diffusion in simulated liquid GeO2 under pressure, Phys. B Condens. Matter 394 (2007) 39–45, https://doi.org/10.1016/j. physb.2007.02.005.
- [78] F. Kong, N. Han, Q. Chen, L. Song, G. Wang, C. Gu, Y. Gao, J.-Q. Wang, X. Shen, Y. Chen, Understanding the link between fragile-to-strong kinetics and β-relaxation in chalcogenide glasses, Ceram. Int. 50 (2024) 8352–8359, https://doi.org/ 10.1016/j.ceramint.2023.12.169.
- [79] J. Pries, H. Weber, J. Benke-Jacob, I. Kaban, S. Wei, M. Wuttig, P. Lucas, Fragile-to-Strong transition in phase-change material Ge3Sb6Te5, Adv. Funct. Mater. 32 (2022) 2202714, https://doi.org/10.1002/adfm.202202714.
- [80] J. Orava, H. Weber, I. Kaban, A. Greer, Viscosity of liquid Ag–In–Sb–Te: evidence of a fragile-to-strong crossover, J. Chem. Phys. 144 (2016) 194503, https://doi. org/10.1063/1.4949526.
- [81] Hybrid thermal management strategy with PCM and insulation materials for pulsed-power source controller in extreme oil-well thermal environment, Appl. Therm. Eng. 214 (2022) 118864, https://doi.org/10.1016/j. applthermaleng.2022.118864.
- [82] A.R.M. Siddique, S. Mahmud, B. Van Heyst, A comprehensive review on a passive (phase change materials) and an active (thermoelectric cooler) battery thermal management system and their limitations, J. Power Sources 401 (2018) 224–237, https://doi.org/10.1016/j.jpowsour.2018.08.094.
 [83] J. Kim, A. Hong, S. Kim, K.-S. Shin, E. Song, Y. Hwang, F. Xiu, K. Galatsis, C. Chui,
- [83] J. Kim, A. Hong, S. Kim, K.-S. Shin, E. Song, Y. Hwang, F. Xiu, K. Galatsis, C. Chui, R. Candler, S. Choi, J.-T. Moon, K. Wang, A stacked memory device on logic 3D technology for ultra-high-density data storage, Nanotechnology 22 (2011) 254006, https://doi.org/10.1088/0957-4484/22/25/254006.
- [84] D. Dragoni, J. Behler, M. Bernasconi, Mechanism of amorphous phase stabilization in ultrathin films of monoatomic phase change material, Nanoscale 13 (2021) 16146–16155, https://doi.org/10.1039/D1NR03432D.
- [85] B. Chen, X.-P. Wang, F. Jiao, L. Ning, J. Huang, J. Xie, S. Zhang, X.-B. Li, F. Rao, Suppressing structural relaxation in nanoscale antimony to enable ultralow-drift phase-change memory applications, Adv. Sci. 10 (2023) 2301043, https://doi.org/ 10.1002/advs.202301043.
- [86] P. Ma, H. Tong, M. Xu, X. Cheng, X.S. Miao, Suppressed resistance drift from short range order of amorphous GeTe ultrathin films, Appl. Phys. Lett. 117 (2020) 022109, https://doi.org/10.1063/5.0009362.
- [87] H. Kwon, A.I. Khan, C. Perez, M. Asheghi, E. Pop, K.E. Goodson, Uncovering thermal and electrical properties of Sb2Te3/GeTe superlattice films, Nano Lett. (2021), https://doi.org/10.1021/acs.nanolett.1c00947.
- [88] W. Yang, N. Hur, D.-H. Lim, H. Jeong, J. Suh, Heterogeneously structured phasechange materials and memory, J. Appl. Phys. 129 (2021) 050903, https://doi.org/ 10.1063/5.0031947.
- [89] Effect of transition metal dichalcogenide based confinement layers on the performance of phase-change heterostructure memory - Kim, Small - Wiley Online

Y. Chen et al.

Library, 2023. https://onlinelibrary.wiley.com/doi/10.1002/smll.202303659. (Accessed 12 November 2024).

- [90] Z. Aksamija, I. Knezevic, Thermal conductivity of Si 1–x Ge x/Si 1– y Ge y superlattices: competition between interfacial and internal scattering, Phys. Rev. B 88 (2013) 155318, https://doi.org/10.1103/PhysRevB.88.155318.
- [91] X. Chen, Z. Zhou, Y.-H. Lin, C. Nan, Thermoelectric thin films: promising strategies and related mechanism on boosting energy conversion performance, J. Materiom. 6 (2020) 494–512, https://doi.org/10.1016/j.jmat.2020.02.008.
- [92] F. Liu, Z. Fan, Defect engineering of two-dimensional materials for advanced energy conversion and storage, Chem. Soc. Rev. 52 (2023) 1723–1772, https://doi. org/10.1039/D2CS00931E.
- [93] G. Modi, E.A. Stach, R. Agarwal, Low-power switching through disorder and carrier localization in bismuth-doped germanium telluride phase change memory nanowires, ACS Nano 14 (2020) 2162–2171, https://doi.org/10.1021/ acsnano.9b08986.
- [94] P. Nukala, C.-C. Lin, R. Composto, R. Agarwal, Ultralow-power switching via defect engineering in germanium telluride phase-change memory devices, Nat. Commun. 7 (2016) 10482, https://doi.org/10.1038/ncomms10482.
- [95] Y. Wang, K. Ren, S.N. Song, Z.T. Song, Defect engineering in antimony telluride phase-change materials, Mater. Sci. Forum 944 (2019) 607–612. https://dx.doi. org/10.4028/www.scientific.net/MSF.944.607.