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Film thicknesses influence on the interfacial thermal resistances within Ge-rich $\text{Ge}_2\text{Sb}_2\text{Te}_5/\text{Ge}_2\text{Sb}_2\text{Te}_5$ multi-layers

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Phase change memories (PCRAM) are often made of chalcogenide alloys in the form of multilayer systems (MLS). One of the most used alloys are $\text{Ge}_2\text{Sb}_2\text{Te}_5$ and Ge-rich $\text{Ge}_2\text{Sb}_2\text{Te}_5$. This article reports on the thermal characterization of very thin (<5 nm) Ge-rich $\text{Ge}_2\text{Sb}_2\text{Te}_5/\text{Ge}_2\text{Sb}_2\text{Te}_5$ MLS by Modulated Photothermal Radiometry (MPTR). The MPTR method allows for the investigation of such samples by determining, with an inverse method, the total thermal resistance of the stack deposited on the substrate. With the measurement of the total thermal resistance it is possible to determine the thermal conductivity of the deposit and the interfacial thermal resistances between layers. The interfacial thermal resistance between Ge-rich $\text{Ge}_2\text{Sb}_2\text{Te}_5/\text{Ge}_2\text{Sb}_2\text{Te}_5$ was characterized, which is an important parameter to reduce the energy cost of the PCRAM functioning. It was also possible to highlight a decrease in interfaces quality inside the MLS after the beginning of the phase transition around 250°C.

I. INTRODUCTION

In the field of non volatile memories, phase change materials (PCM) are widely studied for their use in the phase-change RAM or PCRAM [1–4]. The chalcogenide alloys involved in those devices are made at least from one chalcogenide element such as . Se, Te, S. The most characterized associations of chalcogenide elements are based on In-Sb-Te and Ge-Sb-Te systems. The electrical resistivity R_e of these alloys varies along decades depending on their crystalline state, which make them great candidates for non-volatile memory devices. In the amorphous state, the electrical resistivity is high and the material behaves as an insulator whereas, in the crystalline state, the electrical resistivity is significantly lower. This change of properties allows to link a bit 1 or 0 or even intermediate [4, 5] to the electrical state. It is also important to measure the thermal properties of PCM for their use in PCRAM [6–8]. Knowing precisely the thermal properties as a function of the temperature and depending on the crystalline state is mandatory to determine the electrical power required for the phase change. The SET programming of the PCRAM is responsible for the transition from the insulating state to the conductive state whereas the RESET programming of the PCRAM is responsible for the reverse change. The PCRAM devices have been scaled down for the past few years to the point of reaching the nanometer scale [6, 7]. As the transitions duration for the phase change and the energy consumption are reduced with the technological improvements, the alloy implementation went from the so-called “mushroom” programming volume [9, 10] to nanowires [11–13] with a few nanometers diameter or even PCM superlattices leading to the inter-facial phase change memory technology [14]. Moreover, other PCRAM configurations have been explored by industrials, notably by Samsung,

IBM, Intel/Micron (XPoint) and Hynix/Ovonyx [15–17]. The low scale of the PCRAM makes necessary to control the cross-talks effects with neighboring cells [18, 19]. It is shown that the thermal boundary resistance (TBR) between PCM micro volume and neighboring materials such as the metal electrodes and the dielectrics has an influence of the same order as the thermal conductivity for the heat transfer within the device [18, 20–22]. The measurement of the thermal resistance or conductance of these nanostructured materials can hardly be achieved due to their low scale.

The most common chalcogenide alloy in the framework of PCRAM is $\text{Ge}_2\text{Sb}_2\text{Te}_5$ (GST) mainly due to its fast transformation rate and high cyclability [9]. In today’s PCRAM applications one of the most limiting parameter of GST is its low crystallization temperature (150°C) guaranteeing data retention up to 10 years at 75°C only. Thus it is not appropriate for applications where operations at temperatures higher than the crystallization temperature of the GST are realized. This limitation motivates new research works in order to engineer new materials to improve the PCRAM performance. The two main focus points are the improvement of the stability of the amorphous phase to increase data retention performances, and the reduction of current needed for the RESET operation. Indeed, by reducing the current, the programming energy is reduced and therefore the cross-talks risks lowered. One of the way to tune the GST properties is by doping it with carbon [23, 24], nitrogen [25, 26], silicon [27, 28] and/or Ge atoms (GeGST) [29, 30]. The GeGST or Ge-rich GST has a higher crystallization temperature than GST which ensures a better thermal stability of the reset state. Another benefit of the Ge-rich GST is its higher electrical resistivity compared to the GST [31] which means that the current needed for the Joule effect during the reset cycle is lower for the GeGST compared to the current needed for the GST. However, studies show that after annealing at high temperature (>450°C) multiple phase segregation took place. Indeed,

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Sb, Ge and Te atoms are scattered through PCM in an inhomogeneous way [32, 33]. This undesirable phenomenon is not reported as completely detrimental. In fact, the memory cell after a stable segregation show an improvement in endurance [34], which illustrates the interest for the use of the Ge-rich GST. In order to extend the advantages of the tuned PCM, multi-layers composed of them started to be implemented in memory cells [20, 35–37]. Having multiple layers of PCM will slow down heat diffusion since the phonons will face obstacles at the interfaces, thus reducing the cost of heating the PCM and also improving heat confinement. Using multi-layers PCM also allows for an increase of temperature crystallization. PCM layers are indeed under greater stress while heating, leading to a higher activation energy and more linear grain shapes. The difference in electrical resistivity between GeGST, GST and GeGST(10nm)/GST(10nm) multi-layers is measured in the literature [31] and shows that the resistivity contrast between the amorphous and crystalline phase is usually much larger for the multi-layer structure than for the mono-layer structure. However, using multi-layer PCRAM means that interfacial thermal resistances between layers need to be well characterized. It is important to determine the value of the said resistances but also to study their dynamics while heating. A decrease in the interfaces quality while heating can have detrimental effects on the functioning.

In this work we implement the Modulated Photothermal Radiometry (MPTR) configuration within the infrared domain (IR) for thermal characterization of GeGST/GST multilayer systems (MLS). This method has been fully detailed in the literature [38] and several applications to PCM have been reported [39, 40]. In the present work, interfacial thermal resistances between GeGST and GST values are given for the amorphous phase. It is also shown that the quality of the interfaces starts to decrease as soon as the crystallization happens. Such characterization requires experimental work since simulating interfaces integrity over the phase transition can be very complex. However, concerning the value of the interfacial thermal resistance between GeGST and GST in the amorphous state, the experimental work realized in this paper is in good concordance with some theoretical models.

II. EXPERIMENTAL CONDITIONS

A. Experimental setup

The schematic view of the setup is presented in Fig. 1 and is explained in details in reference [41]. The temperature of the sample is driven by commercially available oven working under argon as inert gas. Samples are characterized from room temperature to 400°C, then cooled again to room temperature. The thermal perturbation seen by the sample is generated by a laser of 1064 nm wavelength and 1.7 W maximum power. The laser spot

radius is 0.8 mm. An acousto-optic modulator enables the modulation of the laser using the squared signal coming from a function generator. The thermal response is then measured by an infrared HgCdTe detector with an infrared sensitive element of 0.5 mm in diameter. A lock-in amplifier was used to measure the amplitude and the phase lag ϕ between the reference and the detector output according to the frequency f . The reference signal used for the lock-in detector is the TTL signal issued from the function generator. In order to avoid the phase lag induced by the modulator and the measurement chain, a calibration was realized on an already well characterized bulk semi-infinite sample. The calibration function is found as: $\phi_{real} = \phi_{exp} + 7.49e^{-4}f - 0.14$ for frequencies ranging from 100 Hz to 10 kHz. In addition, the real temperature of the sample at a given controlled temperature of the furnace has been calibrated using an infrared camera. The temperature T of the sample according to that of the furnace is found as: $T = 0.8685T_{oven} + 12.24$.

B. Sample preparation

The GeGST/GST MLS have been fabricated by magnetron sputtering, using a mixture of argon and nitrogen as sputtering gas. The samples are made in 4 variations with different unit layer thickness (e_{GeGST} and e_{GST}) repeated N times but resulting in the same total film thickness: $e_{MLS} = (e_{GeGST} + e_{GST})N \approx 100$ nm. The stacks compositions are reported in Table I. The configuration for MPTR setup is presented in Fig. 2. The GeGST composition is obtained using co-sputtering from two stoichiometric targets, namely, Ge and $Ge_2Sb_2Te_5$. The Ge-rich composition used is referred as “GST + Ge 45%” where 45% is the nominal fraction of the Germanium added to the $Ge_2Sb_2Te_5$ reference alloy [29]. The GeGST/GST layers have been deposited onto 200 mm in diameter silicon wafers (750 μ m in thickness) covered by a 300 nm thick Si_3N_4 (SiN) passivation layer. The sputtering gas used (mixture of Ar and N_2) was chosen to ensure a nominal N doping of 7% of the GeGST film. Nitrogen addition inside the layer is proven to enable a finer grain structure in the films and also to increase the crystallization temperature [42–45]. The GeGST/GST MLS are deposited in the amorphous phase for both constituents. Transmission electron microscopy (TEM) and Energy-dispersive X-ray spectroscopy (EDX) images have been realized on the different configurations to control the quality of the interfaces after deposition. The TEM and TEM EDS analysis for a (GST 1.5nm/GST+Ge45% 1.5nm)x15 configuration are provided in Fig. 3. It is shown that interfaces inside the MLS are well defined before annealing. Following the deposition of the GeGST/GST multi-layers, all samples were covered by sputtering with a 100 nm thick platinum film in order to obtain a optical to thermal transducer for photothermal measurements and to avoid possible oxidation and evaporation of the MLS at high temperature. A

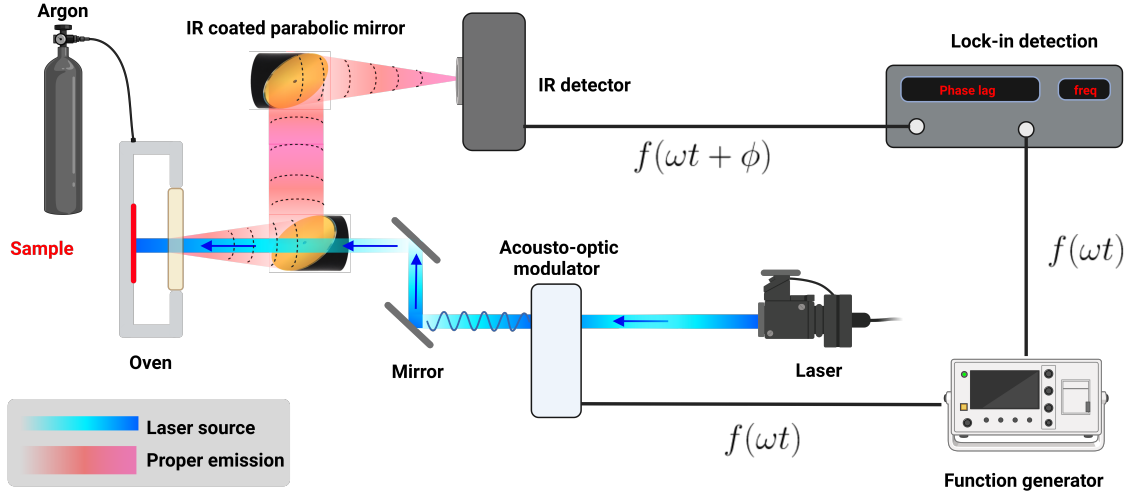


Figure 1: Schematic description of the MPTR experimental setup. The sample is put inside a furnace that permits reaching up to 1200°C under an inert gas.

10 nm thick TiN layer is deposited on the MLS to improve the Pt adhesion.

III. MATHEMATICAL DETAILS AND MODELING

The periodic temperature variation $\Delta T(\omega)$ at the surface of the sample being small enough, it is assumed that the measured proper emission seen by the IR detector is linearly proportional to $\Delta T(\omega)$. Under the assumption of 1D heat transfer within the sample, with no heat loss and a semi-infinite medium behavior for the substrate, it is established a simple relationship between the temperature change $\Delta T(\omega)$ and the heat flux $\varphi_0(\omega)$ at a given pulsation $\omega = 2\pi f$ (f is the frequency) as [46, 47]:

$$\frac{\Delta T(\omega)}{\varphi_0(\omega)} = Z_{Si}(\omega) + RTH \quad (1)$$

$$RTH = N \left(\frac{e_{GeGST}}{k_{GeGST}} + \frac{e_{GST}}{k_{GST}} \right) + (2N - 1)TBR_{GeGST/GST} + \frac{e_{SiN}}{k_{SiN}} + TCR \quad (3)$$

With:

$$TCR = TBR_{Pt/TiN} + TBR_{TiN/GeGST} + TBR_{SiN/Si} + TBR_{GST/SiN} \quad (4)$$

where e_i and k_i denote respectively the thickness and the

thermal conductivity of layer i , N is the number of repe-

where $Z_{Si}(\omega) = 1/(E_{Si}\sqrt{j\omega})$, with $E_{Si} = \sqrt{(\rho Cp)_{Si}k_{Si}}$ (ρ is the density, Cp is the specific heat and k_{Si} is the thermal conductivity of the silicon wafer and $j^2 = -1$), is the thermal impedance of the wafer, and RTH is the thermal resistance of the (Pt/TiN/MLS/SiN) stack. The thermal conductivity of the silicon wafer is reported in Table II and $(\rho Cp)_{Si} = -2.64T^2 + 2.11e^3T + 1.57e^6$ J/m³/K. The phase lag $\phi(\omega)$ is calculated from (1) as :

$$\phi(\omega) = \tan^{-1} \left(\frac{-\sqrt{\frac{\alpha_{Si}}{2\omega k_{Si}^2}}}{\sqrt{\frac{\alpha_{Si}}{2\omega k_{Si}^2}} + RTH} \right) \quad (2)$$

where $\alpha_{Si} = k_{Si}/(\rho Cp)_{Si}$ is the thermal diffusivity. The Levenberg-Mardquardt [48–50] minimization algorithm is used to minimize the gap between the experimental data and the model, allowing the identification of the total thermal resistance of the stack RTH . Assuming the Pt and TiN layers fully thermalized at each pulsation ω , the thermal resistance RTH of the stack is expressed as:

thermal conductivity of layer i , N is the number of repe-

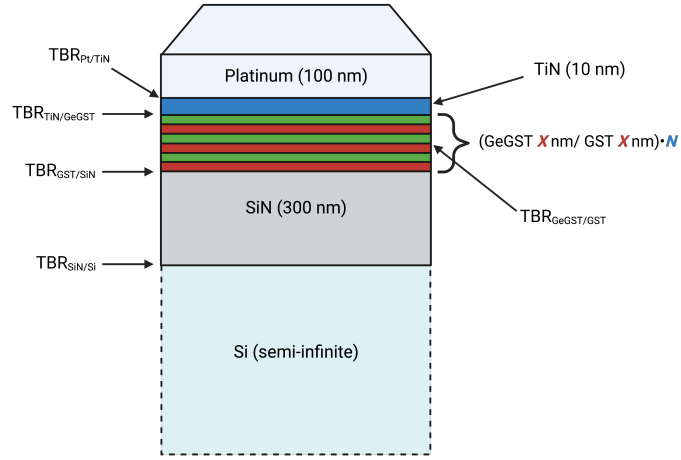


Figure 2: Schematic view of the sample composition. The GeGST/GST MLS configurations are reported in Table I. A Pt film is deposited to act as an optical to thermal transducer. The TiN layer improves the adhesion of Pt. The stack involves several interfacial thermal resistances. The sum of those thermal boundary resistances is depicted as $TCR = TBR_{Pt/TiN} + TBR_{TiN/GeGST} + TBR_{GST/SiN} + TBR_{SiN/Si}$. X denotes the thicknesses of the PCM layers and N denotes the number of times the GeGST/GST unit cell is repeated to make the stack.

Sample label	Deposit constitution	Deposit thickness
P05	(GST 10nm/GST+Ge45% 10nm)x5	100 nm
P10	(GST 5nm/GST+Ge45% 5nm)x10	100 nm
P17	(GST 3nm/GST+Ge45% 3nm)x17	102 nm
P33	(GST 1.5nm/GST+Ge45% 1.5nm)x33	99 nm

Table I: The four GeGST/GST MLS configurations with unit layer thickness of 10, 5, 3, 1.5 nm. The total thickness for the four configurations is 100 nm

	$k(T(^{\circ}C))$ (W/m/K)	ref.
SiN	$4.66e^{-9}T^3 - 5.80e^{-6}T^2 + 2.84e^{-3}T + 1.15$	[51]
Si	$5.23e^{-4}T^2 - 0.451T + 144.3$	[Unpublished]
GeGST	0.29	[31]
GST	0.18	[31]

Table II: Parameters required for the calculation of RTH in the amorphous phase from relation (3). References are also indicated for each parameter.

tions of GeGST/GST unit cell in the MLS and $TBR_{i/j}$ denotes the thermal boundary resistance between contiguous layers i and j . The required parameters for the calculation of RTH from relation (3) are reported in Table II.

It is possible to compute a theoretical value of the interfacial thermal resistance between layers using the diffuse mismatch model (DMM) [52]. The expression of the TBR between materials 1 and 2 from the DMM is:

$$TBR_{1 \rightarrow 2} = \left(\sum_j \frac{k_B^4 T^3}{8 \pi^2 \hbar^3 V_j^2} \alpha_{1 \rightarrow 2} \int_0^{\Theta_{D,1}/T} \frac{x^4 e^x}{(e^x - 1)^2} dx \right)^{-1}, \quad j = (L, T, T) \quad (5)$$

In this equation, \hbar is the reduced Planck constant, k_B is the Boltzmann constant and V_j is the velocity of the phonons in the different directions ($j=(L,T,T)$, L is the longitudinal direction and T are the two transversal ones). The phonon transmission coefficient α is defined

as:

$$\alpha_{1 \rightarrow 2} = \frac{v_{L,2}^{-2} + 2v_{T,2}^{-2}}{v_{L,1}^{-2} + 2v_{T,1}^{-2} + v_{L,2}^{-2} + 2v_{T,2}^{-2}} \quad (6)$$

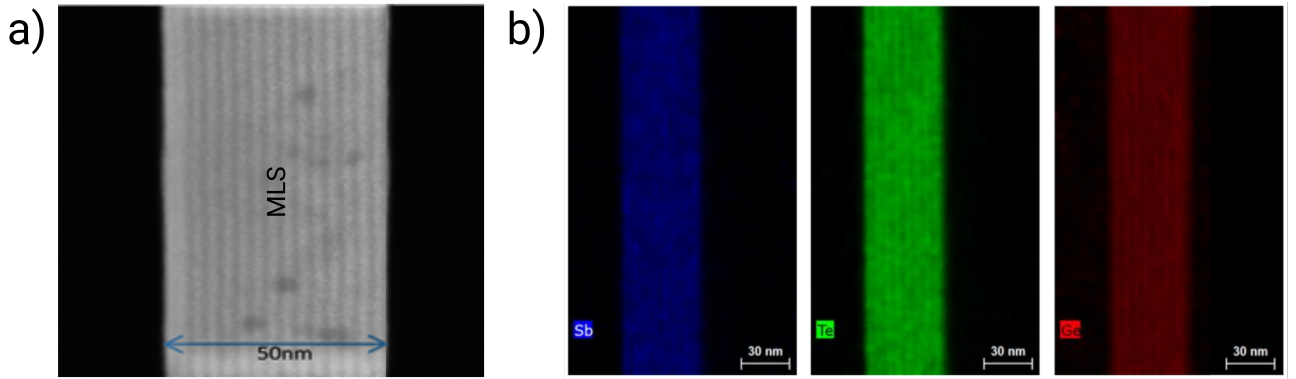


Figure 3: a) Transmission electron microscopy (TEM) image of a (GST 1.5nm/GST+Ge45% 1.5nm)x15 stack configuration sample. b) TEM EDS analysis of a (GST 1.5nm/GST+Ge45% 1.5nm)x15 stack configuration sample.

IV. RESULTS AND DISCUSSION

A. Stack thermal resistance R_{TH}

The samples, listed in Table I, were heated from 50°C up to 400°C with a heating rate of 100°C/min with a short 30 seconds plateau to stabilize the temperature before each acquisition. Each temperature measurement was realized at two frequencies, 2551 Hz and 3182 Hz, which led to a measurement time of 5 minutes per temperature. Those two frequencies were chosen to have a heat penetration depth greater than the stack thickness and to still have the wafer acting as a semi infinite medium. The R_{TH} value is identified after subtracting the SiN contribution. Results are reported in Fig. 4 according to the annealing temperature. Previous results for 100 nm thick GeGST [31] and GST [41] are also reported in Fig. 4. Two R_{TH} transitions are observed in Fig. 4. The first one, around 150°C, is rather sharp and is assigned to the well known amorphous to fcc crystalline transition within the GST layers of the MLS. A second smoother and less marked R_{TH} drop is observed around 300°C. The latter is related to the delayed amorphous (am) to fcc crystalline transition within the GeGST layers of the MLS. In between those two R_{TH} drops, the R_{TH} decreases smoothly as T increases. This behavior may be tentatively related to the ongoing diffusion of atomic Ge, which lessens/spreads over T the expected T -lag of the am -to- fcc transition within Ge-doped layers and hinders the fcc -to- hcp transition in the GST layers of the MLS. The results demonstrate also that, in the amorphous phase, increasing the number of interfaces in the MLS, while keeping its thickness constant, leads to increase R_{TH} (at the low temperature range). It is a consistent observation that adding more interfaces leads to increase the total thermal resistance. It is also possible to note a delay (denoted η) in the first transition temperature with the increase of the number of interfaces. However after the first phase transition (above at 250°C), a similar value of R_{TH} it is obtained for each

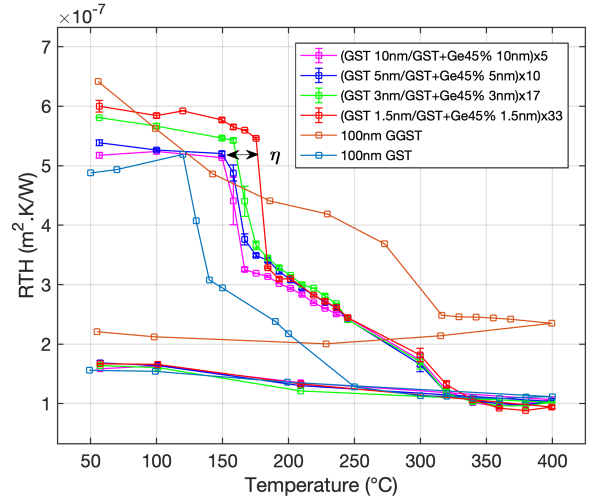


Figure 4: Total thermal resistance of the stack deposited on the silicon wafer for samples P05, P10, P17, P33 and two reference samples. The heating rate is 100°C/min, each temperature measurement required 5 minutes. The samples were in the amorphous phase before heating. After cooling, the samples are in a stable crystalline phase. The SiN contribution to the total thermal resistance R_{TH} was subtracted for MLS and GGST. Regarding the GST, the contribution of the SiO₂ was also subtracted. The difference between $R_{TH_{GGST}}$ and thermal resistances from the MLS $R_{TH_{MLS}}$ for the crystalline phase is denoted as ΔR_{TH} . The shift in crystallization temperature from amorphous to fcc is denoted η .

MLS. Moreover, the values of R_{TH} for the MLS are the same as the 100 nm thick GST. This phenomenon can be explained by the loss of interface integrity from both the crystallization process and species inter-diffusion at high temperature. The degradation of interface quality subsequent to heating has been shown already in the literature [53].

B. Interfaces integrity

In-situ Raman scattering analysis from room temperature to 450°C was performed using an INVIA Renishaw confocal spectrometer coupled to a Leica optical microscope. A 785 nm monochromatic laser excitation was used. The temperature of the samples was controlled the same way as the MPTR experiment. The Raman analysis was also conducted under inert argon atmosphere with a 100°C/min heating rate with a short 30 seconds plateau to stabilize the temperature before each spectral acquisition. A low level laser power density was used in order to avoid unwanted additional heating by the incident radiation. The spectrometer is fitted with an edge filter cutting the black-scattered light at $\sim 100 \text{ cm}^{-1}$. The spectral resolution of the Stokes-side Raman spectra was $\sim 1 \text{ cm}^{-1}$. The results for the (GST 5nm/GST+Ge45% 5nm) $\times 5$ stack are reported in Fig. 5. SEM micrographs of this particular stack after annealing at 410°C were also realized and reported in Fig. 6. At room temperature the main feature of the Raman spectra of the amorphous stack is a large band spreading over the 100-190 cm^{-1} spectral region. This large band, mainly composed of two overlapping bands peaking around 125 cm^{-1} and 150 cm^{-1} is comparable to the expected Raman signature of the amorphous as-deposited $\text{Ge}_2\text{Sb}_2\text{Te}_5$ (GST) alloy. At higher frequencies, the $\nu_3(F2)$ antisymmetric stretching mode of tetrahedra in amorphous GeTe, peaking at 220 cm^{-1} , most likely contributed to the broad bulge in the 190 – 300 cm^{-1} region associated with a-Ge in tetrahedral sites within the *N*-doped GeGST sublayers of the MLS [54, 55]. The Raman spectra remained essentially unchanged from room temperature to 130°C. Upon reaching 160°C, the relative intensity of the band at $\sim 150 \text{ cm}^{-1}$ decreased markedly, and shifted towards higher frequencies at around 160 cm^{-1} . The band at $\sim 125 \text{ cm}^{-1}$ shifted towards lower frequencies at about 115-120 cm^{-1} . This evolution can be explained by the transition from amorphous to metastable *fcc* crystalline state within the GST sublayers of the MLS. Regarding the significant drop of the band originally peaking at 150 cm^{-1} , building on the work found in the literature [56], we may argue that the decreasing of this band could be associated with the appearance of a transient GeTe phase within the GeGST. However, it is important to acknowledge that this transient phase would usually appear above 300°C. At 330°C, the Raman spectrum does not evolve significantly with respect to 160°C. At this high temperature, the lowered intensity of the band assigned to Sb-Te vibrations can be explained by the appearance of the aforementioned transient GeTe phase nucleating in the GeGST sublayers [56]. On the other hand, at such a high temperature, one would expect that the GST sublayers that switched to the *fcc* state at lower temperature (160°C) now undergo the transition towards the hexagonal-closed-packed (*hcp*) lattice. Yet, the *fcc*-to-*hcp* transition was essentially not observed. In Fig. 5 (330°C), we merely see two discrete shoulders at 110

and 170 cm^{-1} , which may barely suggest this transition to *hcp*. For reference, the inset in Fig. 5 provides the non ambiguous Raman signature of the GST *hcp* crystalline lattice, much more sharply than the discrete observed shoulders at 330°C. In the current case study, we observe instead a more pronounced signature of the *fcc* transformation, reflected by the increasing relative intensity of the band peaking at about 160 cm^{-1} . This behavior may reflect the onset of the thermally delayed *fcc* transformation within the GeGST amorphous sublayers. According to the literature [56], the delayed thermal crystallization of cubic GST from amorphous GeGST precursors is a three-fold process whereby a transient GeTe phase first appears, which triggers the nucleation and the growth of cubic Ge, cubic GST forming last. Unfortunately, the low signal-to-noise ratio of the Raman spectrum acquired at 330°C did not allow to confirm the presence of cubic Ge around 300 cm^{-1} . At 450°C, the crystallization of *fcc* GST from the GeGST sublayers of the MLS is clearly confirmed by the two broad bands peaking at about 120 cm^{-1} and 160 cm^{-1} . At this high temperature, cubic GST and cubic Ge do coexist [29, 56–58]. The absence of the cubic-to-hexagonal transition at high temperature in GeGST thin films has been reported in the literature [57]. Thus, an excess of a-Ge with respect to the 225 stoichiometry of GST would prevent the latter transition. As the *hcp* crystal system is here not observed, despite the presence of GST sublayers, we speculated that atomic Ge may migrate from GeGST sublayers into GST sublayers after the amorphous to *fcc* transition around 160°C in the latter sublayers. However, the thermal resistance of the MLS being the same as the GST mono-layer one could be a sign of a *fcc* to *hcp* transition happening during cooling. The *fcc* phase being highly metastable and the oven following temperature set-points during cooling, such transition could have happened. The scanning electron micrograph (SEM) of the MLS annealed at high temperature reported in Fig. 6 seem to state only partial mixing. Indeed, some interfaces may be damaged into interphases in the annealed samples, but the sample is still closer to a multi-layered system than a mono-layered system. This may support the partial mixing (diffusion of atomic Ge) between GST and GeGST sublayers.

The in-situ Raman scattering analysis suggests that the germanium migrated in whole the MLS. This migration explains the similitude on the *RTH* values for MLS and the 100 nm thick GST stack. The phase change allowed the atomic structure of the GeGST/GST multi-layers to relax itself and reach its minimum of potential energy by distributing the germanium atoms in all the MLS. Thus, hindering the effect of the doping. It is then possible to consider the crystallization dynamics of the GeGST/GST MLS similar to a pure GST stack even though the GST has been doped with Ge atoms. The difference in thermal resistances ΔRTH illustrated in Fig. 4 is a testimony of the doping being hindered and the MLS being similar to pure GST. Since at high temperatures *TCR* will most likely decrease, the

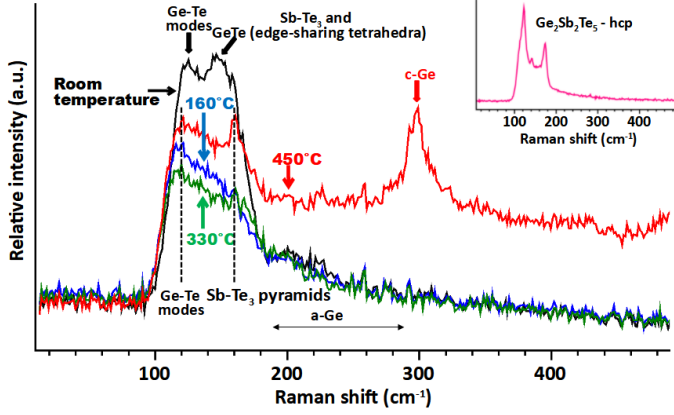


Figure 5: In-situ Raman analysis of a the (GST 5nm/GST+Ge45% 5nm)x5 stack sample. The room temperature spectrum (black), the 160°C spectrum (blue spectrum), the 330°C spectrum (green spectrum) and the 450°C spectrum (red spectrum) are reported on the figure along with a $\text{Ge}_2\text{Sb}_2\text{Te}_5$ hcp reference spectrum.

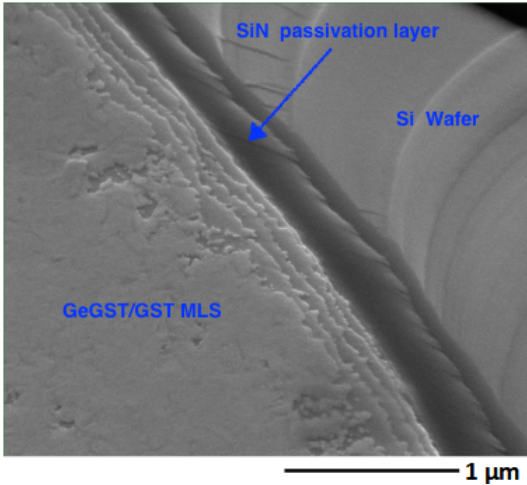


Figure 6: Scanning electron micrograph (SEM) of the (GST 5nm/GST+Ge45% 5nm)x5 stack sample annealed at 410°C. We can see the trace of 5 interphases while the original as-deposited MLS included a vertical alternation of 10 elementary sublayers. This may suggest only a partial mixing between the GST and the GGST sublayers, the interfaces are closer to interphases.

gap ΔRTH can be explained mostly by the change in thermal conductivity between GGST and GST, that is $\Delta RTH \approx e_{MLS}(1/k_{GGST} - 1/k_{GST}) + \epsilon$. The residues ϵ can be for the most part attributed to TCR between samples being slightly different or to small regions of the MLS where interphases are still closer to being interfaces coupled to doping residues.. The thermal conductivities of GGST and GST are reported in Fig. 7. A TiN adhesion layer is present between the Pt layer and the GGST layer for the samples characterized in this

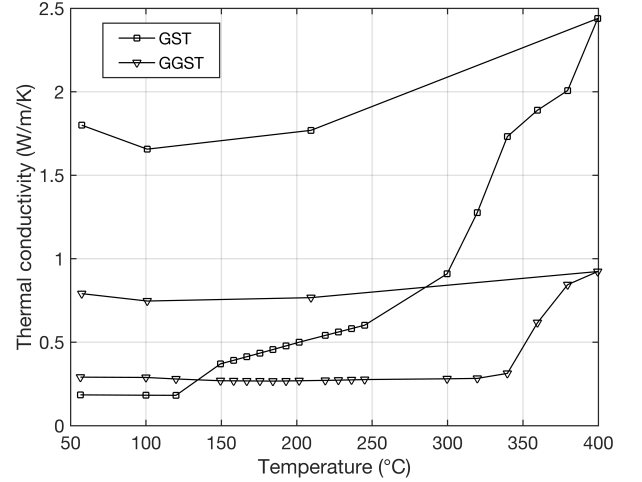


Figure 7: Thermal conductivities of GGST and GST [31].

study. The pure GGST sample was characterized with the Pt directly deposited on the GGST, thus the Pt adhesion is much worse than in the MLS. It means that $TBR_{PT/GGST} > TBR_{PT/TiN} + TBR_{TiN/GGST}$. This relation has already been confirmed for the amorphous phase by previous work [31, 59] carry on for the crystalline phase. After annealing the MLS is now comparable to a GST stack in terms of thermal properties and $TBR_{GGST/GST}$ tends to 0.

C. Thermal interface resistances characterization

Basing on equation (3), one has:

$$RT = (2N - 1)TBR_{GeGST/GST} + TCR \quad (7)$$

with: $RT = N(e_{GeGST}/k_{GeGST} + e_{GST}/k_{GST}) - e_{SiN}/k_{SiN}$. By performing a linear regression on $RT = f(2N - 1)$, it is possible to extract the value of $TBR_{GeGST/GST}$ and TCR at the different annealing temperatures. The linear regressions are reported Fig. 8. The results are shown is Fig. 9 considering the temperature range where the MLS is still in its amorphous state. Indeed, as showed in the previous section, the MLS interfaces are damaged at elevated temperature, which does not allow the calculation of a thermal resistance between GeGST and GST layers within the MLS. The average values are $TBR_{GeGST/GST} = (1.39 \pm 0.16) \cdot 10^{-9} \text{ m}^2\text{K/W}$ and $TCR = (1.72 \pm 0.06) \cdot 10^{-7} \text{ m}^2\text{K/W}$ in the amorphous state. As reported in the figure, these values are close to the theoretical values computed by the Diffuse Mismatch Model (DMM), that is $1.23 \cdot 10^{-9} \text{ m}^2\text{K/W}$ for $TBR_{GeGST/GST}$, [52] and values found in the literature [31].

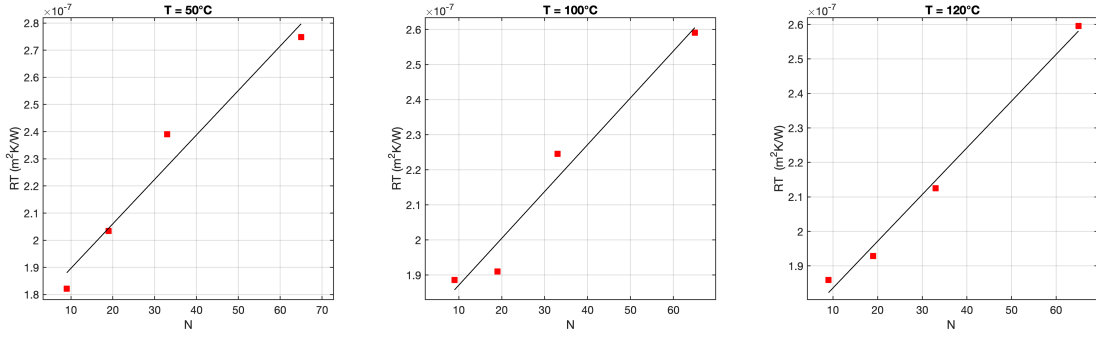


Figure 8: Linear regression for the amorphous phase based on equation (7).

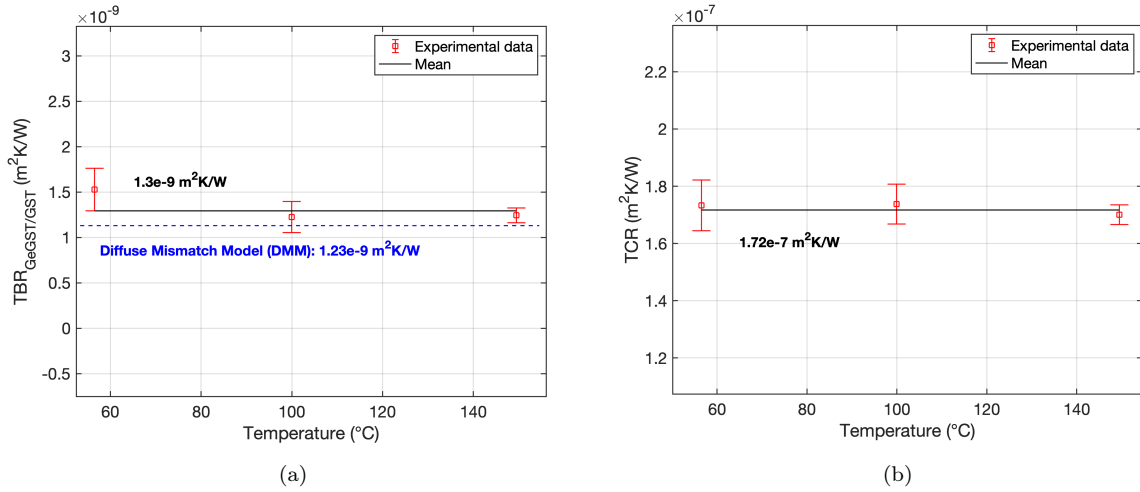


Figure 9: a) Interface thermal resistance $TBR_{\text{GeGST}/\text{GST}}$ between a GeGST and a GST layer within the MLS at 3 different temperatures where the MLS is in its amorphous state. The value is compared to theoretical value computed with the DMM. The parameters values used for the DMM computation are reported in Table III. b) Sum of the thermal interface resistances TCR clarified in equation (4) for 3 different temperatures. All the values have been computed by linear regression on equation (7) using the data displayed in Fig. 4.

Layer	Longitudinal speed (m/s)	Transversal speed (m/s)	ref.
GeGST	3600	2530	[60]
GST	2350	1350	[61]

Table III: Phonon velocities used for the theoretical computation of $TBR_{GeGST/GST}$ using the DMM.

V. CONCLUSION

In this work it was shown that increasing the number of interfaces in the phase change material multilayer increases the total thermal resistance for temperatures below 250°C. Increasing the total thermal resistance of the multilayer is a key aspect in the development of PCRAM since having a higher thermal resistance means a more efficient heating of the PCM. A temperature delay in the phase transition was found when the number of interfacial thermal resistances is increasing.

However, it was also proven that after the crystallization onset the interfaces between the layers in the GeGST/GST multilayer stack deteriorate. Thus, above 250°C the number of interfaces do not have an influence on the total thermal resistance of the stack in the crys-

talline phase. This can be explained by a deterioration of the interface quality between layers. Raman analysis showed that after the phase change, the Ge atoms started to migrate in the whole sample, hindering the effect of the doping as consequence.

Moreover, the thermal resistance of the interface between GeGST/GST layers has also been characterized. The value found is in accordance with DMM model and the literature.

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