Investigations on Ge-Te-Si glasses and their suitability for phase change memory applications

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INTRODUCTION

The development of newer phase change materials for NVRAM applications is mainly based on synthesizing newer glass compositions and investigating the SET and RESET processes by applying current/voltage pulses of different waveform/amplitude and duration.[1,2] The understanding of the SET-RESET processes and optimization of the input electrical parameters such as pulse amplitude, source resistance, pulse width, etc., are very important for selecting a glass for phase change memory applications.[3,4] Usually, a higher current pulse is required for resetting the memory state and reduction in RESET current is one of the most crucial parameter for developing high-density (lower programming volume) Phase Change Random Access Memories (PCRAM).[1,5,6] Many attempts have been made to reduce the RESET current by modifying the device structure or by doping with other elements to reduce the melting temperature and eventually to reduce the programming volume.[7]

In the present work, investigations have been undertaken on the electrical switching behavior and thermal parameters of $Ge_{15}Te_{85-x}Si_x$ glasses, over a wide range of compositions ($2 \leq x \leq 12$). Efforts have also been made to understand the composition dependence of switching voltage (V_T) and thermal parameters such as T_g, T_{c1}, non-reversing heat flow (ΔH^{NR}) etc., of $Ge_{15}Te_{85-x}Si_x$ glasses and to identify the composition range in the $Ge_{15}Te_{85-x}Si_x$ system suitable for Phase change Memory (PCM) applications.

Further, detailed electrical switching analysis and electrical SET-RESET operations are performed on the identified new phase change material, namely $Ge_{15}Te_{83}Si_2$ glass. Also, In-situ Raman scattering experiments are performed on $Ge_{15}Te_{83}Si_2$ sample, during the electrical SET-RESET processes, in order to elucidate the local structural changes during the SET and RESET operations. HRTEM studies are carried out on switched samples to probe the structural transformations during the SET operation.

EXPERIMENTAL DETAILS

Bulk $Ge_{15}Te_{85-x}Si_x$ glasses ($2 \leq x \leq 12$) have been prepared by melt quenching technique. The electrical switching, SET and RESET operations have been performed using a programmable dc source-meter (Keithley 2410^c) controlled using LabVIEW6.1. Alternating Differential Scanning Calorimetric (ADSC) studies have been carried out in the temperature range 70-375 ºC, at a scan rate of 3 °Cmin^{-1}. The glass transition temperature (T_g) is deduced from the reversing heat flow curves and the crystallization temperature (T_c) from the non-reversing curves. The enthalpy change obtained from the non reversible heat flow curve (ΔH^{NR}) is used as a measure of relaxation enthalpy during glass transition. The typical error in the ADSC measurements is within ± 2 °C for T_g, T_c and ± 0.05 J/g for ΔH^{NR}.

A gap-cell arrangement, with a channel width of ~0.3 mm and with gold coated electrodes, has been used for in-situ Raman scattering under electrical switching on $Ge_{15}Te_{83}Si_2$ samples of dimensions approximately 3 x 3 x 0.4 mm. The confocal micro-Raman studies have been carried out in back scattering geometry using DILOR-XY instrument equipped with a liquid nitrogen-cooled CCD detector. The samples have been illuminated by the 514.5 nm line of an argon ion laser (COHERENT INNOVA 300) focused at a spot of glassy sample in between

the gold electrodes. All the data are recorded using ~2 mW of laser power and for about 600 sec of accumulation. The spectral resolution is 0.8 cm^{-1}. The Raman spectra have been acquired in three different stages, namely as-quenched, after switching (SET process) and re-amorphized (RESET process). The in-situ electrical SET-RESET processes are performed on the surface of the sample whereas remaining electrical measurements have been undertaken across the sample. The sample behavior is found to be similar in both the cases. To probe the structural details, a FEI TECNAI High Resolution Transmission Electron Microscope (300 kV) is used.

RESULTS AND DISCUSSION

Electrical switching studies

The electrical switching behavior of $Ge_{15}Te_{83}Si_2$, $Ge_{15}Te_{82}Si_3$, $Ge_{15}Te_{78}Si_7$ and $Ge_{15}Te_{77}Si_8$ glasses representing $Ge_{15}Te_{85-x}Si_x$ glass system, is shown in figure1.

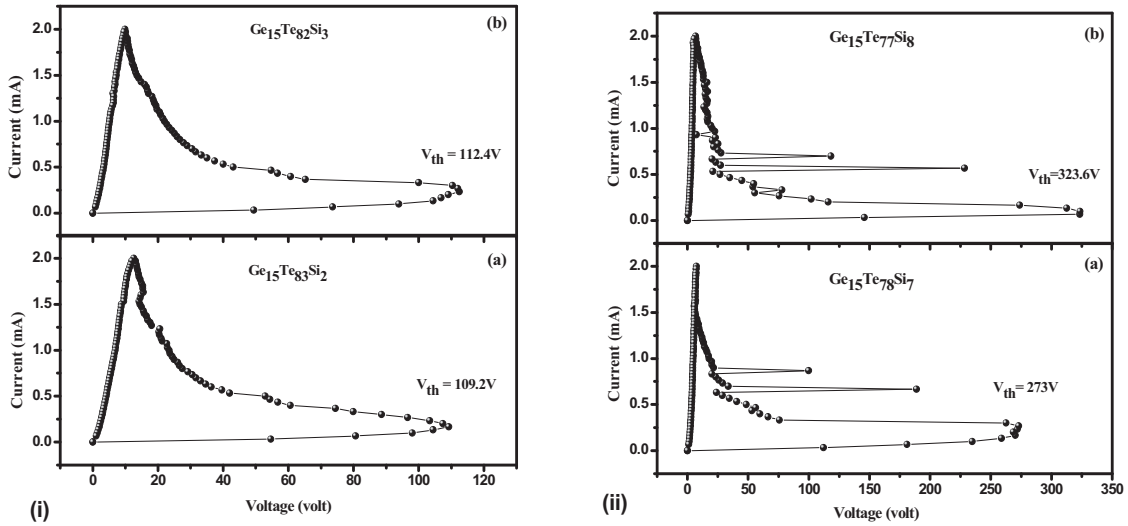

Figure 1 *i) I-V characteristics of $Ge_{15}Te_{83}Si_2$ and $Ge_{15}Te_{82}Si_3$ glasses, representing the composition range $x \leq 5$. ii) I-V characteristics of representative $Ge_{15}Te_{78}Si_7$ and $Ge_{15}Te_{77}Si_8$ glasses representing the composition range $x > 5$.*

It is seen from the I-V characteristics that the $Ge_{15}Te_{85-x}Si_x$ glasses exhibit memory type electrical switching and the threshold voltages are in the range 100-600 V. It is interesting to note that the $Ge_{15}Te_{85-x}Si_x$ glasses exhibit two kinds of memory switching behavior; a normal switching is seen in samples with $x \leq 5$ as shown in figure 1(i). However, instability is seen in glasses with $x > 5$, during the transition to the ON state above the threshold voltage V_T as shown in figure 1(ii). This behavior seems to be connected with network rigidity/connectivity of the glasses and is discussed in more detail, later.

Figure 2 shows the variation of switching voltages (V_T) with composition/average coordination number (<r>) for the $Ge_{15}Te_{85-x}Si_x$ system of glasses. The average coordination number (<r>) has been estimated using the coordination numbers of 4 for Ge/Si and 2 for Te, confirming with Mott's (8-N) rule.[8,9] It can be seen from figure 2 that the V_T of $Ge_{15}Te_{85-x}Si_x$ glasses increase with the addition of Si content (x) upto a composition x = 5 (<r> = 2.40). A change in slope is seen at x = 5, above which V_T increases more sharply with further increase in silicon content. The inset in figure 2 shows the variation of the starting electrical resistance of $Ge_{15}Te_{85-x}Si_x$ samples (measured across the electrodes), with composition. It can be seen that

composition dependence of V_T and the electrical resistances of the sample are very similar. It is also interesting to note here that the electrical resistance of the $Ge_{15}Te_{85-x}Si_x$ samples exhibit an abrupt increase at the composition x = 5 (<r> = 2.40).

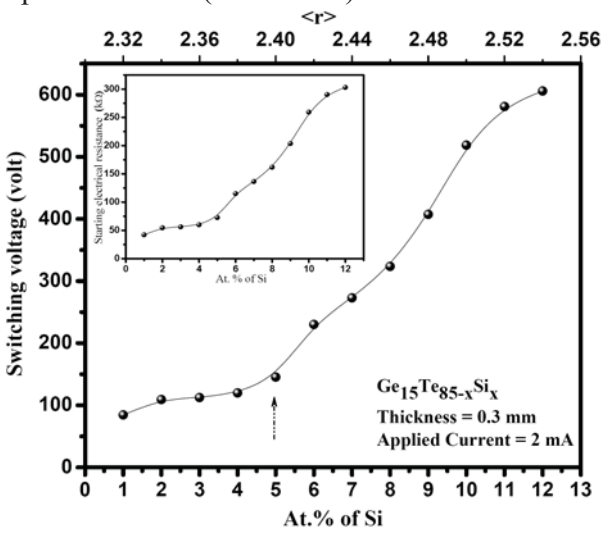

Figure 2: *The composition dependence of V_T of $Ge_{15}Te_{85-x}Si_x$ glasses. The inset shows the variation of electrical resistance (initial) of the samples with composition.*

As pointed out by several authors the composition dependence of switching voltages of chalcogenide glasses is determined by different factors such as the resistivity of the additive element, the network connectivity and rigidity percolation, etc.[10-12] In general, the switching voltages are found to decrease with the addition of more metallic additives. The earlier studies on a variety of glasses show that the switching voltages increase with an increase in network connectivity and rigidity and a sharp change in slope (lower to higher) is seen in the composition dependence of switching voltages at the rigidity percolation threshold of the system.[13-17]

In the present $Ge_{15}Te_{85-x}Si_x$ glassy system, an increase in switching voltage with an increase in silicon content is expected, based on both the metallicity factor as well as rigidity percolation. The composition x = 5 (<r> = 2.40) at which a slope change is seen in the composition dependence of switching voltages corresponds to the rigidity percolation threshold of the Ge-Te-Si glasses. In this context, it is interesting to note that bulk Ge-Te glasses exhibit the rigidity percolation at an average coordination number <r> = 2.40.[17] It is found that the addition of four fold coordinated silicon atoms to the Ge-Te system is not altering the rigidity percolation threshold of the system.

ADSC Studies

Figure 3i shows the composition dependence of glass transition temperature (T_g) and the first crystallization temperature (T_{c1}). It can be seen from figure 3i that the T_g of $Ge_{15}Te_{85-x}Si_x$ glasses increases almost linearly with the addition of Si in the entire composition tie-line $2 \leq x \leq 12$. The magnitude of the glass transition temperature and its variation with composition are closely connected to the network connectivity and its evolution with composition. The stiffness of a covalently bonded random network and consequently the glass transition temperature increases progressively with an increase in the average coordination number and network connectivity.[18]

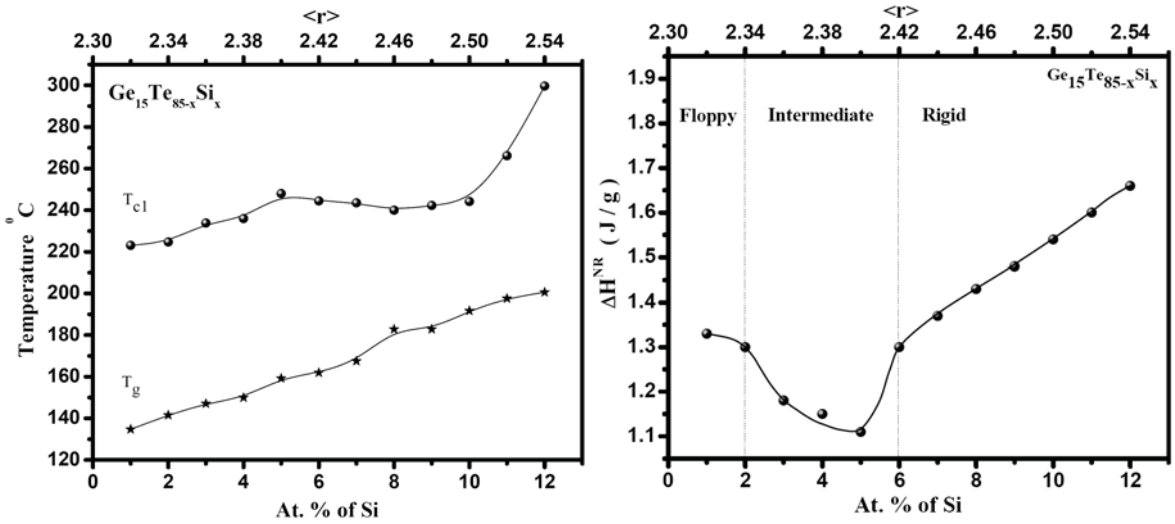

Figure 2: *(i) The composition dependence of T_g and T_{c1} of $Ge_{15}Te_{85-x}Si_x$ glasses.(ii) The composition dependence of ΔH^{NR} of $Ge_{15}Te_{85-x}Si_x$ glasses.*

In the present $Ge_{15}Te_{85-x}Si_x$ system, the $Ge_{15}Te_{85}$ base glass with an average coordination number <r> = 2.3 is intrinsically elastically floppy.[10,11] The progressive addition of tetrahedrally coordinated silicon results in a continuous increase in average coordination number, network connectivity and rigidity. This results in a near linear increase in T_g with silicon content. Further, there is no saturation or decrease in glass transition temperature, which implies that there is no phase separation or fragmentation of the network in the entire composition range ($2 \leq x \leq 12$).[18] It is also seen from figure 3i that the first crystallization temperature (T_{c1}) exhibits an increase with silicon content upto a composition x = 5 (<r> = 2.4). T_{c1} remains almost a constant in the composition range $5 < x \leq 10$ ($2.4 < <r> \leq 2.50$) and it is found to increase comparatively more sharply with silicon content thereafter. It is also interesting to note here that the presence of an extended stiffness transition and a thermally reversing window in $Ge_{15}Te_{85-x}Si_x$ glasses (in the range $2.34 \leq <r> \leq 2.42$) has been exhibited in the composition dependence of ΔH^{NR} (figure 3ii). However, the variation with composition of T_{c1} show only the signature of the mean-field threshold at <r> = 2.4.

On the basis of electrical switching and thermal characteristics of $Ge_{15}Te_{85-x}Si_x$ glasses, the $Ge_{15}Te_{83}Si_2$ glass is found to have better performance and hence considered for detailed investigations on the SET and RESET processes and their structural origin.[19-22]

The I-V characteristics and electrical switching behavior of the $Ge_{15}Te_{83}Si_2$ glass is shown in Figure 4, which shows that the sample exhibits electrical switching at a switching voltage (V_{th}) of 110±2 volts (I_{th}~50 μA). It is interesting to note that the sample latches onto the memory state for an input current pulse of 1 mA. However, the inset in figure 4 shows the I-V characteristics of the same sample at lower input current, which indicates that the $Ge_{15}Te_{83}Si_2$ glass shows a threshold behavior for 0.7 mA input current pulse. Though the electrical switching in $Ge_{15}Te_{83}Si_2$ glass takes place at a threshold current I_{th} = 50 μA, the sample does not get latched to the ON state at currents lower than 0.7 mA of input current. It is clear from the above that ON state current upto 0.7 mA do not considerably alter the state of the sample in the switched region and upon increasing the current upto 1 mA, the Joule heating induces the phase transformation of the material in the conducting channel, leading to the SET state.

The SET process in a Phase Change Material consists of three sequential stages. In the first stage, the voltage across the sample increases monotonously until it reaches the threshold

value; at the threshold voltage, the material undergoes a transformation from a high resistance state to a low resistance state and a conducting path is established. In the second phase, the preservation of the low resistance state, established during switching, takes place. The process at this stage is reversible as no permanent change takes place in the conducting path during this phase. The subsequent increase in current flow leads to the third phase during which the amorphous to crystalline phase change occurs in the conducting channel. The crystallization of the material in the conducting channel is likely to commence by a nucleation mechanism. The nucleation of crystallites may trigger an enhanced electrical conduction which can lead to more Joule heating due to which the crystallites may grow rapidly to replace an amorphous matrix.[20]

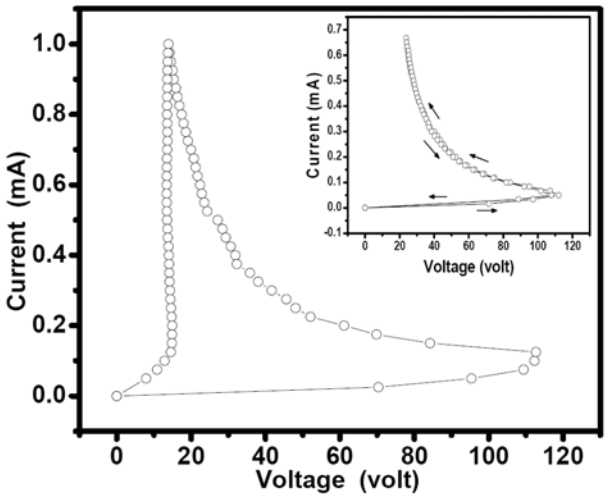

Figure 4: *I-V characteristics of $Ge_{15}Te_{83}Si_2$ glass with 1mA input current. The inset in figure shows the I-V characteristics of the same sample with 0.7 mA input current.*

In the present experiments, the SET/RESET processes in $Ge_{15}Te_{83}Si_2$ glass are achieved by applying 1 mA triangular/1 mA rectangle pulse of 10 msec width current pulses respectively (figure 5i). The electrical switching occurs during ramp-up, at ~50 μA; The SET process is accomplished operation on further current flow in the ON state, between 0.7 mA - 1 mA (I_P).

It is interesting to note from figure 5i that the current is reduced gradually while ramping down the current during the SET operation, leading to a controlled slow cooling of the material. This maintains the crystallized conducting channel (SET state). Further, the RESET operation is accomplished by applying a short duration rectangle pulse of 10 msec width, having the same magnitude as that of SET current (I_P), which causes the local melting of the conducting crystalline channel. The quick dissipation of heat from the molten conducting channel to the surrounding matrix results in the re-amorphization of the region (RESET process). Generally, the current required to RESET the chalcogenide glassy switch is much larger than the SET current.[3] However, in the present $Ge_{15}Te_{83}Si_2$ glass, it is found that the SET and RESET processes can be achieved using the current pulse of same magnitude, but with different type of waveforms.[20]

In order to understand the influence of the current waveform on the switching behavior, experiments have been carried out by applying saw-tooth current pulses of different amplitudes (1-10 mA) to the $Ge_{15}Te_{83}Si_2$ glass (figure 5ii). It is interesting to note that the sample exhibits threshold (reversible) switching with saw-tooth excitation upto 10 mA, while a memory behavior is seen with triangular pulses of 1 mA magnitude. As we know, the ramp-down is abrupt in a saw-tooth pulse, whereas the ramp-up remains the same in both. The threshold behavior seen in the experiment using the saw-tooth pulse means that the abrupt ramp-down results in a fast

cooling of the material leading to the re-amorphization of the switched region and hence the threshold behavior. Further, the threshold behavior of the sample is retained even if the magnitude of the saw-tooth input current pulse is increased to 10 mA (Figure 5ii). This indicates that the $Ge_{15}Te_{83}Si_2$ glass is easily resettable, and the sharp ramp-down with the saw-tooth current pulse results in self-Resetting of the sample.[20]

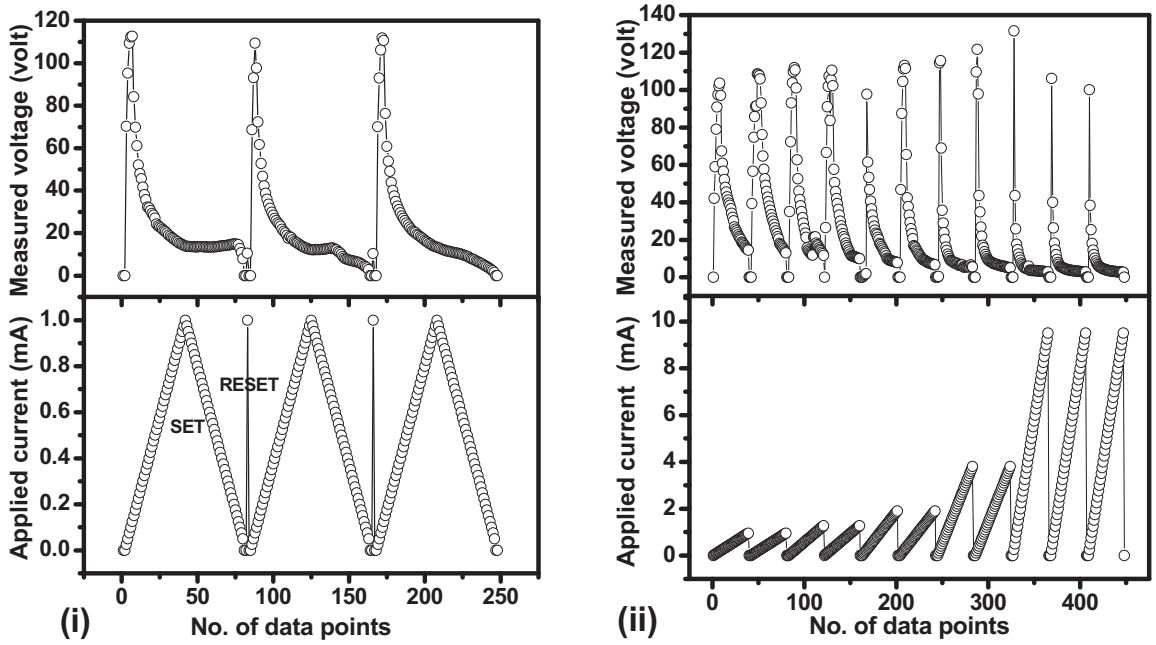

Figure 5: *(i) The SET/RESET processes in $Ge_{15}Te_{83}Si_2$ glass achieved with 1 mA of triangular/ rectangle pulse of 10 msec width respectively. (ii) Electrical switching behavior of $Ge_{15}Te_{83}Si_2$ glass with saw-tooth current pulses.*

In-situ Raman scattering studies

Figure 6 shows the Raman spectra of $Ge_{15}Te_{83}Si_2$ glass obtained during various stages, namely as-quenched and after the SET and RESET processes. Here, the Raman spectra have been recorded in three sequential stages. At first stage, the Raman spectra of amorphous sample is obtained by focusing at a spot in the chalcogenide glass in between the gold electrodes. In second step, the spectrum has been acquired by focusing the laser beam at a spot in the conducting crystalline channel formed during the SET process. In third step, the spectrum is obtained after resetting the conducting channel by passing a short width rectangular current pulse. The nature of three different phases of sample namely, Amorphous, SET and RESET states has been confirmed by measuring the electrical resistance between electrodes (approximately 0.3 MΩ for amorphous, 56 Ω for SET and 0.3 MΩ for RESET states respectively).[20]

In Figure 6, three main bands are seen in the Raman spectra of the amorphous, SET and the RESET states, in the frequency range of 50-250 cm^{-1}. Approximate wave number ranges of the band positions in all three states are as follows: band B ~ 115.8-117.9 cm^{-1}; band C ~ 139.2-141.8 cm^{-1}; band D ~ 159.2-161.2 cm^{-1} and a weak hump, band A ~ 91.7-92.1 cm^{-1}. The details of the line shape fitting parameters are given in Table 1.

Figure 6a shows the Raman spectrum of the $Ge_{15}Te_{83}Si_2$ glass, which exhibits three distinct peaks around 117.9 (B), 140.7 (C) and 161.2 cm^{-1} (D) respectively. The spectrum also has a weak hump around 91.7 cm^{-1}(A). The peak B can be attributed to the A_1 mode and peaks A and C to the E_{TO} modes of crystalline Te-Te chain.[23]

In the present study, the composition of the base glass ($Ge_{15}Te_{85}$) is well below the critical composition, $Ge_{33}Te_{77}$, defined by the Chemically Ordered Covalent Network (COCRN) model. This model presumes that the Te atoms are arranged as one-dimensional chains in between which the Ge atoms are present as crosslinks.[24] Thus, the structural network in the base glass is primarily decided by Te-Te chains, which are interlinked by Ge-Te bonds. It is also known that amorphous Te crystallizes at 10 °C and is unstable at room temperature.[25] Therefore; the Te-atom chains in $Ge_{15}Te_{83}Si_2$ glass are likely to have a certain degree of order. This conjecture is consistent with the observation that the Raman peaks (A), (B) and (C) in $Ge_{15}Te_{83}Si_2$ glass correspond to modes of crystalline Te.

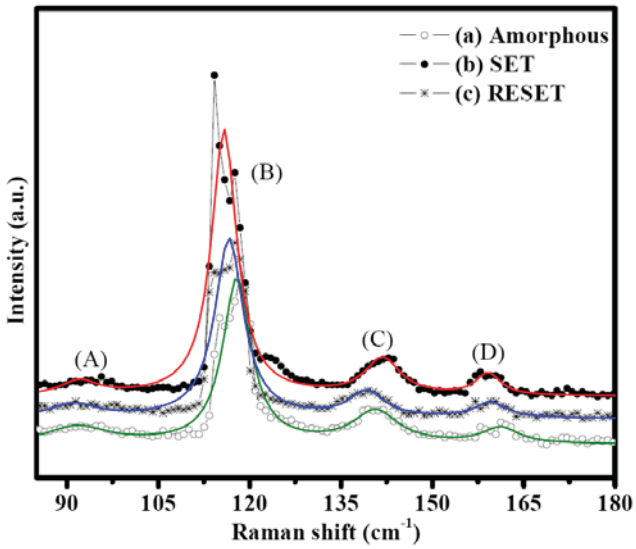

Figure 6: *In-situ Raman scattering study on (a) amorphous sample (b) electrically switched (SET) and (c) re-amorphized (RESET) states.*

Parameters		Amorphous	SET	RESET
Peak A	Position	91.7 ± 1.5	92.1 ± 2.7	91 ± 1.9
	Linewidth	12.6 ± 5	7.3 ± 8.4	9.8 ± 6.5
Peak B	Position	117.9 ± 0.1	115.8 ± 0.1	116.6 ± 0.1
	Linewidth	5.6 ± 0.3	5 ± 0.3	5.6 ± 0.3
Peak C	Position	140.7 ± 0.6	141.8 ± 1	139.2 ± 0.8
	Linewidth	8.9 ± 1.8	7.1 ± 3.1	7.1 ± 2.5
Peak D	Position	161.2 ± 1.1	159.2 ± 1.4	159.6 ± 1.3
	Linewidth	7.2 ± 3.2	4.8 ± 4.2	6.4 ± 3.9

Table 1: *Line shape fitting parameters of the Raman spectra of $Ge_{15}Te_{83}Si_2$ in amorphous, SET and RESET conditions.*

It is also interesting to note from Raman studies on Si-Te glasses that it exhibits a peak at 138 cm^{-1} attributed to the tetrahedral $SiTe_{4/2}$ units, which shows a blue shift towards 141 cm^{-1} during thermal annealing.[26] Hence, the peak at 140.7 cm^{-1}(C) may be attributed to the Te chain as well as the vibrational motions in $SiTe_{4/2}$ face-sharing tetrahedra. Further, the peak D can be assigned to the symmetric stretching mode of the edge-sharing $GeTe_4$ tetrahedra.[24]

The Raman spectrum of the $Ge_{15}Te_{83}Si_2$ sample, acquired by focusing the beam on the switched region after the SET process is shown in Fig. 6b. It is interesting to note that there are no drastic changes in the Raman Spectra of the $Ge_{15}Te_{83}Si_2$ sample during the SET transition: While peak (A) remains almost unaffected, peaks (C) and (D) exhibit marginal blue- and red-shifts (about one wave number each), respectively. However, the peak (B) at 117.9 cm^{-1} becomes more intense during the SET operation. It can be observed from the present in-situ Raman studies that the local structure remains mostly unaltered during the SET process, while the degree of disorder is reduced in the Te- atom chains present in the glassy $Ge_{15}Te_{83}Si_2$ system keeping the original glassy network intact. This behavior is in contrast to other GeTe systems.[24] In this case, there is no amorphous-crystal phase transition as a whole during the SET process. Rather, a structural rearrangement takes place which leaves the system in a more ordered state during switching, though the electrical properties are changed by a large extent.

Figure 6c shows the Raman spectrum of the $Ge_{15}Te_{83}Si_2$ sample, acquired by focusing the beam on the switched region after the RESET process. It is clear from this figure that the Raman spectrum of the sample in the RESET state is very similar to the amorphous spectrum. The present in-situ Raman studies indicate that in Ge-Te-Si sample, the local structures in the glassy (OFF state) and RESET state are similar and they are not very different from the local structure in the SET state. This implies that the three states are close to each other in terms of local structure and the transitions between them are likely to be less energy intensive.

TEM Studies

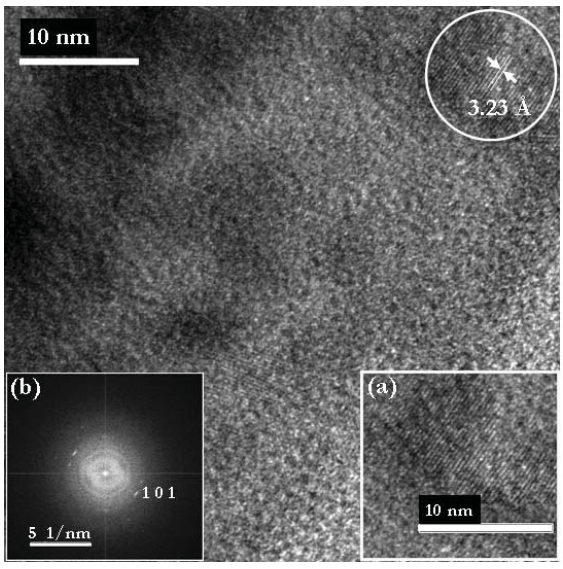

Figure 7: *HRTEM image of switched samples indicating the crystalline fringes formed during SET process (a) magnified region of crystalline fringes (b) Fast Fourier Transform (FFT) of the electron diffraction pattern.*

High Resolution Transmission Electron Microscopic (HRTEM) analysis performed on switched samples reveals crystalline fringes formed during SET operation corresponding to hexagonal c-$SiTe_2$ (with a space group of P3m1; a = 4.28 Å, c = 6.70 Å).[27]

CONCLUSIONS

It is proposed that the under constrained $Ge_{15}Te_{85-x}Si_x$ glasses with $x < 5$ are likely to be more suitable for phase change memory applications. In-situ Raman scattering studies on $Ge_{15}Te_{83}Si_2$ sample, undertaken during the SET and RESET processes indicate that the degree of disorder in the glass is reduced from OFF to SET state. It is also found that the local structure of the sample under RESET condition is similar to that in the OFF state. The Raman results are consistent with the switching results which indicate that the $Ge_{15}Te_{83}Si_2$ glass can be SET and RESET easily. Electron Microscopic studies on switched samples indicate the formation of nanometric sized particles of c-$SiTe_2$.

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Biography

Anbarasu Manivannan was born in Tamilnadu, India on 15th May 1978. He received the B.E. degree in electrical and electronics engineering from Bharathiyar University, Coimbatore, in 2000, and Ph.D. degree from the faculty of engineering, Indian Institute of Science, Bangalore, India in 2008. He also received Dr. Srinivasa Rao Krishnamurthy award for the best Ph.D. thesis 2008. His research is concerned in the field of chalcogenide glasses and phase change memories.

Dr. Anbarasu Manivannan's papers have been published in Applied Physics Letters, Journal of Applied Physics, Philosophical Magazine, Journal of non crystalline solids, Journal Physics D: Applied Physics. Further he has been selected for Alexander von Humboldt Post Doctoral Research to do research at I. Physikalisches Institut IA, RWTH Aachen University from 1st September 2009 onwards.