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Electrical switching and *in situ* Raman scattering studies on the set-reset processes in Ge–Te–Si glass

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Bulk $Ge_{15}Te_{83}Si_2$ glass has been found to exhibit memory-type switching for 1 mA current with a threshold electric field of 7.3 kV/cm. The electrical *set* and *reset* processes have been achieved with triangular and rectangular pulses, respectively, of 1 mA amplitude. *In situ* Raman scattering studies indicate that the degree of disorder in $Ge_{15}Te_{83}Si_2$ glass is reduced from *off* to *set* state. 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. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2770770]

Phase change memories (PCMs) based on chalcogenide glasses are being considered recently as a possible replacement for conventional nonvolatile random access memories (NVRAMs). The main advantages of PCM are their direct write/overwrite capability, lower voltages of operation, write/erase cycles, easiness to integrate with logic, etc.[1,2] The basic principle of operation of a chalcogenide PCM is the irreversible transition (memory type) exhibited by the glass from a semiconducting *off* state to a conducting *on* state induced by an electric field.[3,4]

Electrical switching in chalcogenide glasses occurs when the field injected charge carriers fill the charged defect states in the sample.[5] In glasses which are easily devitrifiable, the Joule heating in the current carrying path can result in the formation of a conducting crystalline channel, which leads to memory switching.[6]

The memory switches may be *reset* back to the *off* state by the application of a current/light pulse, during which local melting of the conducting channel and re-solidification into amorphous state take place.[3] Generally, the *reset* process is accomplished by applying a sharp current pulse having a higher magnitude compared to the *set* pulse.[7,8]

Efforts are being made recently to understand the *set-reset* processes and to optimize the electrical parameters such as amplitude, pulse-width, etc., which are important from the viewpoint of the stable performance of a memory device.[9–12] Attempts are also being made to reduce the *reset* current by modifying the device structure or by doping with other elements.[13]

In this letter, we report electrical switching studies on $Ge_{15}Te_{83}Si_2$ glass. *In situ* Raman scattering studies have also been undertaken during the *set* and *reset* operations to elucidate the local structural transformation during the *set-reset* operations.

Bulk $Ge_{15}Te_{83}Si_2$ glass has been prepared by the conventional vacuum-sealed melt quenching method.

Electrical switching studies and the electrical *set* and *reset* processes have been carried out using a Keithley 2410c dc source meter.

A gap-cell arrangement is used for *in situ* electrical switching under Raman scattering, with a typical sample size of about $3\times 3\times 0.4$ mm^3 and the electrode (gold) width of ~0.3 mm. The electrical connections from the sample to the source meter have been done with copper wires bonded to the electrodes using silver paste.

The confocal micro-Raman studies on $Ge_{15}Te_{83}Si_2$ glass have been carried out in backscattering geometry using DILOR-XY instrument equipped with a liquid nitrogen-cooled charge coupled device detector. The samples are illuminated by the 514.5 nm line of an argon ion laser (Coherent Innova 300) focused using a 50× objective at a spot of glassy sample in between the gold electrodes. All the data have been recorded using 2 mW of laser power and for 600 s of acquisition. The spectral resolution is 0.8 cm^{-1}.

The current-voltage characteristics and electrical switching behavior of $Ge_{15}Te_{83}Si_2$ glass (0.15 mm thickness) are shown in Fig. 1. It is found to exhibit memory type of switching for 1 mA input current pulse at a threshold electric field of E_{th}=7.3 kV/cm (corresponding I_{th}=50 μA).

The inset in Fig. 1 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-type switching 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

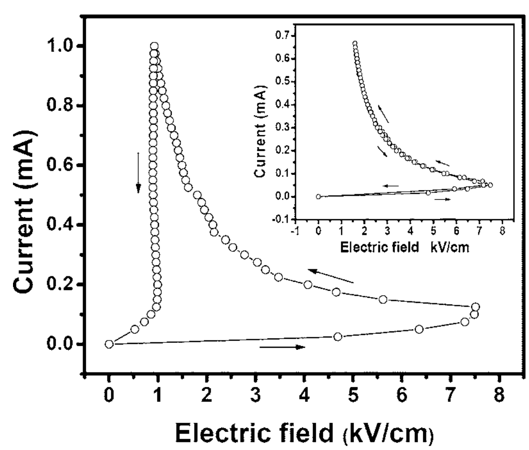

FIG. 1. *I-V* characteristics of $Ge_{15}Te_{83}Si_2$ glass with 1 mA input current showing memory switching. The inset shows the *I-V* characteristics of the sample with 0.7 mA input current (threshold behavior).

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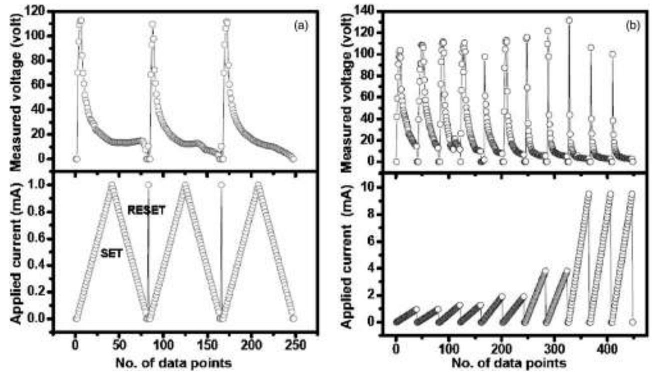

FIG. 2. (a) *set*/*reset* operations in $Ge_{15}Te_{83}Si_2$ glass carried out using a triangular (*set*) and a rectangular (*reset*) pulse of 1 mA amplitude. (b) Electrical switching behavior of $Ge_{15}Te_{83}Si_2$ glass obtained with a saw-tooth current pulse.

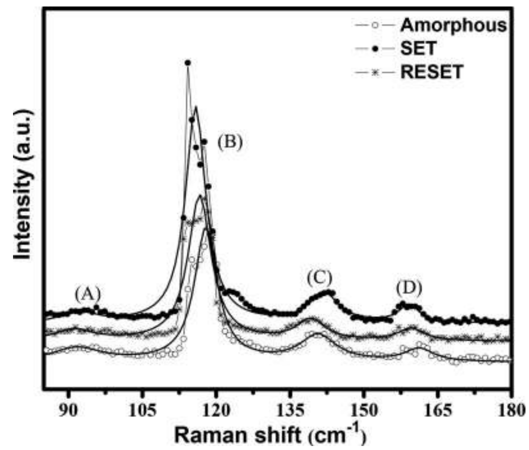

FIG. 3. *In situ* Raman spectrum of $Ge_{15}Te_{83}Si_2$ sample in (a) amorphous, (b) *set*, and (c) *reset* states.

I_{th}=50 μA (E_{th}=7.3 kV/cm), the sample does not get latched to the *on* state at currents lower than 0.7 mA of input current. Upon increasing the current in the *on* state beyond 1 mA, the Joule heating in the current carrying path and the consequent increased mobility of atoms lead to local structural rearrangements and crystallization, which result in the *set* state.[5,8]

The most interesting aspect of the electrical switching in chalcogenide glasses is its behavior during the ramping down of the input current. In memory switching samples, the slow cooling occurring during this process maintains the crystalline region formed (*set* state). In these cases, it is necessary to electrically *reset* the memory state by applying a short duration current pulse, usually of higher magnitude than that required for setting the device in the *on* state.

It is interesting to note from Fig. 2(a) that the *set* and the *reset* processes in $Ge_{15}Te_{83}Si_2$ sample are achieved by applying 1 mA triangular pulse (for *set* process) and short width rectangular pulse (for *reset* process), respectively, for a sample thickness of 0.15 mm. During the ramp-up process of the *set* operation, the electrical switching from the *off* to *on* state occurs at about 50 μA current and any further increase in the *on* state current (>0.7 mA) leads to the phase change in the conducting path and the *set* state is reached. As mentioned earlier, during the ramp down of triangular current pulse, the *set* state is retained in $Ge_{15}Te_{83}Si_2$ sample; the electrical resistivity in the *off* state and the *set* state differs by about three orders of magnitude.

The *reset* process in $Ge_{15}Te_{83}Si_2$ glass is accomplished by applying a current pulse of 1 mA magnitude and 10 ms width of rectangular pulse, which heats up the crystallized conducting channel rapidly; this causes the local melting of the conducting channel and its subsequent solidification into amorphous state. Usually, the resetting pulse is of higher magnitude compared to the setting pulse.[7] However, our present study shows that current pulses of the same magnitude (with different wave forms) can perform both *set* and *reset* processes in the $Ge_{15}Te_{83}Si_2$ glass.

In order to understand the influence of the current wave form on the switching behavior, experiments have been carried out by applying a saw-tooth-type current pulse to the $Ge_{15}Te_{83}Si_2$ glass, as shown in Fig. 2(b). These studies reveal that the sample exhibits the threshold-type switching for saw-tooth pulses of 1 mA amplitude, whereas memory behavior is seen with triangular pulses of the same magnitude. 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 reamorphization of the switched region. It is also interesting to note that with a saw-tooth excitation wave form, the sample gets *reset* even at 10 mA *on* state current [Fig. 2(b)].

As mentioned earlier, *in situ* Raman studies have been undertaken on $Ge_{15}Te_{83}Si_2$ glassy samples, during various stages of switching (*off*, *set*, and *reset* states) (Fig. 3). The Raman spectra have been recorded in three sequential steps. In the first step, the Raman spectrum of amorphous sample is obtained by focusing at a spot on 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 described above. In third step, the spectrum is obtained after resetting the conducting channel by passing a short width rectangular current pulse. The nature of the three different phases of sample, namely, amorphous, *set*, and *reset* states has been confirmed by measuring the electrical resistance between the electrodes (~0.3 MΩ for amorphous, 50 Ω for *set*, and 0.3 MΩ for *reset* states).

It can be seen from Fig. 3 that three main bands are seen in the Raman spectra of the amorphous, the *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}; and 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 I.

Figure 3(a) shows the Raman spectrum of the $Ge_{15}Te_{83}Si_2$ glass, which exhibits three distinct peaks around 117.9 cm^{-1} (*B*), 140.7 cm^{-1} (*C*), and 161.2 cm^{-1} (*D*), respectively. The spectrum also has a weak hump around 91.7 cm^{-1} (*A*). 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.[14]

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 model. This model presumes that the Te atoms are arranged as one-dimensional chains in between which the Ge atoms are present as crosslinks.[15] 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

TABLE I. Line shape fitting parameters of the Raman spectra of $Ge_{15}Te_{83}Si_2$ in amorphous, *set*, and *reset* conditions.

Sample No.	Peak	Line shape	Amorphous	*Set*	*Reset*
I	*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
II	*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
III	*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
IV	*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

temperature.[16] 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–C in $Ge_{15}Te_{83}Si_2$ glass correspond to modes of crystalline Te.

It is also interesting to note from Raman studies[17] that Si–Te glasses exhibit a peak at 138 cm^{-1} attributed to the tetrahedral $SiTe_{4/2}$ units, which shows a blueshift toward 141 cm^{-1} during thermal annealing. 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, peak D can be assigned to the symmetric stretching mode of the edge-sharing $GeTe_4$ tetrahedra.[15]

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. 3(b). 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 redshifts (about one wave number each), respectively. However, 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 $Ge_{15}Te_{83}Si_2$ system keeping the original glassy network intact. This behavior is in contrast to other GeTe systems.[15] 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 3(c) 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 Fig. 3(c) 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* states 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. These observations are consistent with the electrical switching results, which indicate that the Ge–Te–Si sample can be *set* with a relatively lower current (1 mA for 0.15 mm thickness sample), and it is electrically easily *resettable* with the same magnitude of current and also the sample gets self-*reset* with a saw-tooth current pulse.

In summary, bulk $Ge_{15}Te_{83}Si_2$ glass has been found to exhibit electrical switching at a threshold current I_{th} $=50$ μA ($E_{th}=7.3$ kV/cm). The sample exhibits a threshold behavior at currents lower than 0.7 mA and it gets latched to the *on* state (*set* process) at higher *on* state currents ($\approx$1 mA). It is also found that in $Ge_{15}Te_{83}Si_2$ glass, current pulses of the same magnitude (with different wave forms) can perform both *set* and *reset* processes. *In situ* Raman scattering studies indicate that in $Ge_{15}Te_{83}Si_2$ glass, the Raman spectra of the sample in the glassy (*off* state), the *set* and the *reset* states are similar.

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