Investigations on Ultrafast Electrical Switching Dynamics of In-Sb-Te Phase Change Memory Devices and Their Suitability for Multi-bit Data Storage Applications

Synopsis of the thesis
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by

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1. INRODUCTION

From ancient times to the present, there have been several techniques used by mankind to store the information such as cave paintings, papers, magnetic tapes, optical storage products (CDs, DVDs, and BDs) and recently electronic memory products. There is a radical shift in the way of storage of information over several decades. In today's modern era, portable electronic products such as mobile phone, tablet, digital camera etc. are becoming an essential part of daily life. As a result, there is a vital need of non-volatile memory (NVM) having large data storage capacity with faster data transfer rates. Ideally, such requirements could be fulfilled by '*universal memory'* device that combines superior properties of various memory products, including high speed and non-volatility [1, 2]. Silicon based NVMs, such as conventional Flash memory has extensively been used in various electronic devices. Nevertheless, owing to the limitation of physical scaling of Flash memories continued scaling becomes challenging [3]. Therefore, there is a growing interest to identify suitable memory technologies meeting the requirements of high performance NVM for future electronics.

Among various emerging memory technologies, phase change memory (PCM) has already been successfully demonstrated their capabilities for high-speed, high density, non-volatile memory [1-6]. PCM technology exploits the rapid and reversible switching between high-resistance (*logic '0'*) amorphous and low-resistance (*logic '1'*) crystalline phase using nanosecond electrical pulses as shown in Fig. 1.

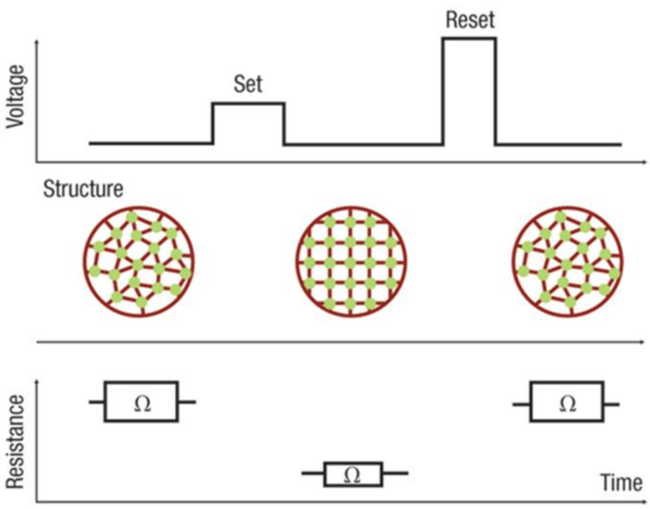

Fig. 1 *Principle of Phase change memory technology [1]*

Furthermore, owing to the increased demand for high-density memory, PCM is being explored for multi-bit data storage, with the capability of allowing more than single bit information stored into unit memory cell. Among most of the phase change (PC) materials, $Ge_2Sb_2Te_5$ has extensively been studied due to its promising features including ultrafast crystallization/re-amorphization in nanosecond timescale and high degree of scalability with low programming current [2, 4]. However, $Ge_2Sb_2Te_5$-based PC materials offer relative poor thermal stability characterized by a low crystallization temperature (~160 ºC).

In the recent past, search of novel PC materials with improved properties has received much interest to enhance the performance characteristics of PCM devices [7, 8]. The ternary compound In_3SbTe_2 (IST) lies in the InSb-InTe pseudo-binary line shows remarkably higher crystallization temperature (~ 290 ºC) [9]. Also, optical properties of IST material have already been exploited for the optical data storage. Hence, a systematic understanding on ultrafast electrical switching dynamics and multi-bit storage capabilities of IST material are important towards the development of high-speed, high-density PCM for multi-bit data storage applications.

2. MOTIVATION OF THIS THESIS

Phase change memory (PCM) technology is a promising candidate for future non-volatile data storage due to its excellent properties such as high speed, non-volatility, better endurance, low power programming, and good scaling characteristics. These attributes make PCM technology a strong contender for "*universal memory*", which could possibly replace all data storage devices from random access memories (RAMs) to hard-disc drives (HDDs). However, some of the critical issues, which are mentioned below, need to be addressed towards successful commercialization of PCM technology.

A. The speed of crystallization is governed by a combined effect of threshold-switching from amorphous-off (*a-off*) state to amorphous-on (*a-on*) state, followed by a crystallization (*set*) state made by Joule heating in the conducting state of PC material. Therefore, these two key factors must be discerned together when high-speed crystallization of PC material is addressed. The speed of threshold-switching is essentially dictated by transient parameters such as delay time, i.e., the time elapsed prior to initiation of the switching event. Despite efforts to understand the dependence of delay time on applied voltage pulses, it has so far only been realized in the order of a few ns. Hence, it is a challenge to realize faster crystallization made by ns electrical pulses, as it is primarily limited by the dependence on ultrafast threshold switching transient characteristics of PCM devices. Owing to these facts, the speed of crystallization is slower compared to that of re-amorphization, which is the main drawback keeping us from achieving picosecond (ps) programming characteristics of the PCM devices. Therefore, a systematic understanding of time-resolved electrical pulse measurements of threshold-switching dynamics and *set* process of the PCM devices together in ps timescale is essential.

B. In recent years, tremendous technological efforts have been devoted to improve the programming characteristics, structural stability, reliability and performance of PCM devices. However, achieving an ultrafast, yet low power threshold switching and crystallization using time-resolved measurements is a key challenge. Weak electric field induced switching offers an extremely fast crystallization through incubation-assisted thermal pre-structural ordering, owing to faster nucleation and growth process as manifested by an ultrafast *set* process [2]. However, such measurements lack the evidence of time-resolved data of current-voltage characteristics, which could possibly unveil a systematic enhancement over threshold switching and the crystallization speed.

C. Multi-bit data storage technique secures much attraction for improving the density of non-volatile PCM, which is highly desirable for the low-cost high-density memory devices. In multi-bit PCM, memory cells contain intermediate resistance levels between amorphous and crystalline phases to store more numbers of logical bits. These intermediate resistance levels can be obtained by controlling the crystalline or amorphous volume between the two electrodes. It is important to note that electrical current plays a crucial role in the memory operation. Moreover, suitable increment of device current could be precisely program PCM devices into high contrast multiple resistance levels consisting of various partially crystalline states. Therefore, a wide range of programming margin via. current controlled promotion of crystallization would be highly helpful for multi-bit data storage.

D. Furthermore, multi-bit PCM device requires an appropriate memory element, which can be modulated to various thermally stable resistance levels between amorphous and crystalline states by employing suitable electrical pulses. Resistance drift also affects the stability of these resistance levels. Due to aforementioned reasons, search of novel phase materials is received significant interest. Therefore, structural and physical properties of IST material have been investigated as a better memory element.

3. OBJECTIVES AND THE SCOPE OF THE PRESENT THESIS WORK

The critical concerns mentioned above necessitate a systematic investigation that drove this thesis to orient with the following major objectives:

I. To investigate the sub-nanosecond threshold-switching dynamics and *set* process of IST phase-change memory devices.

II. To understand the effect of a weak electric field on the ultrafast electrical switching dynamics of IST phase-change memory devices.

III. To explore the current induced multiple resistance levels for the development of multi-bit PCM applications.

IV. To study the local structure and physical properties of IST material using temperature dependent resistivity and *in situ* XRD measurement.

4. MAJOR CONTRIBUTION

4.1 Thin film device fabrication and experimental techniques

Multi-target DC/RF magnetron-sputtering system has been used for thin film deposition including top/bottom electrodes and phase change material in all the experiments. For electrical characterization, device is fabricated in crossbar type structure using mechanical masks where phase change material is deposited in sandwich geometry between top electrode (TE) and bottom electrode (BE). We have used a custom-designed programmable electrical tester (PET) for capturing the ultrafast switching dynamics of IST cells. DC current measurement setup is used to identify the current induced electrical switching in PCM cells. X-ray diffraction (XRD), X-ray Reflectometry (XRR) and Scanning Electron Microscopy (SEM) were employed. Details investigations on crystallization temperature and resistance contrast between amorphous and crystalline phase have been done using custom-built temperature dependent sheet resistance measurement. Furthermore, *in situ* XRD at beamline (BL-12) on Indus-2 synchrotron source (*Raja Ramanna Centre for Advanced Technology, Indore*) provides the direct evidence on the structural transformation from as-deposited amorphous to crystalline IST material.

4.2 Development of temperature-dependent resistance measurement setup

In-situ resistance measurement setup using van der Pauw (VDP) method is developed to identify the resistance of thin films from few Ω upto 10 GΩ under controlled temperatures starting from room temperature to 600 °C. This setup mainly includes the sample holder, tubular furnace for heating, and custom-built switching network board (SNB) with necessary electronic accessories. SNB is designed for making different connections according to the VDP geometry. Importantly, SNB in connection with low noise shielded coaxial cable reduces the effect of leakage current as well as the capacitance in the circuit thereby enhancing the accuracy of measurement. Furthermore, the setup has been used to investigate the *in situ* resistance change of several phase change materials including $Ge_2Sb_2Te_5$, $Ag_5In_5Sb_{60}Te_{30}$ and In_3SbTe_2 materials from as-deposited amorphous to crystalline phase transitions. The setup is fully automated through LabVIEW and after initiating the program, the complete experiment is carried out automatically along with plotting of the data in real time. Moreover, the developed setup could be also useful in various scientific and industrial applications with different samples including semiconductors, metals, and insulators.

4.3 Sub-nanosecond electrical switching dynamics and *set* process of In_3SbTe_2 device

The dependence of transient switching parameters such as delay time, t_d, and switching time, t_s, on the applied voltage pulses of the IST devices at the picosecond-timescale was demonstrated. The time-resolved ultrafast switching characteristics of several IST cells were investigated by varying the pulse amplitude, V_A from 1.8 V to 2.4 V having pulse parameters of rise/fall time (t_r / t_f) of 1 ns and plateau time (t_p) of 100 ns. Small value of t_d of 300 ps is observed for a V_A of 2.1 V and remains as a constant value of t_d upto a V_A of 2.4 V. In order to investigate the ultrafast *set* process of IST cells, a voltage pulse with a very short pulse-width (FWHM) of 1.5 ns, amplitude of 2.3 V, t_r and t_f of 1 ns was chosen. Figure 2 displays the V_A, the response of device current, I_D and the change of dynamic resistances corresponding to switching from the amorphous to the poly-crystalline state of IST devices. It is noteworthy to mention here that I_D rapidly increases from *a-off* to a conducting state after a finite t_d of 300 ps, validating present experimental findings of the dependence of t_d on V_A for very short pulses having pulse-width of 1.5 ns (FWHM). Rapid transition of dynamic resistances from high resistance (10 MΩ) amorphous state to a low resistance (10 kΩ) state is shown in Fig. 2(c). The permanent change in resistance from amorphous to a low resistance poly-crystalline state (10 kΩ) made by an ultrafast *set* pulse is confirmed by means of a subsequent read pulse (with an amplitude of 0.2 V, pulse-width of 100 ns). Therefore, this result reveals a strikingly fast *set* operation of IST devices within 1.5 ns, wherein a change in the dynamic resistance is obtained well within 550 ps, which includes t_d of 300 ps and t_s of 250 ps.

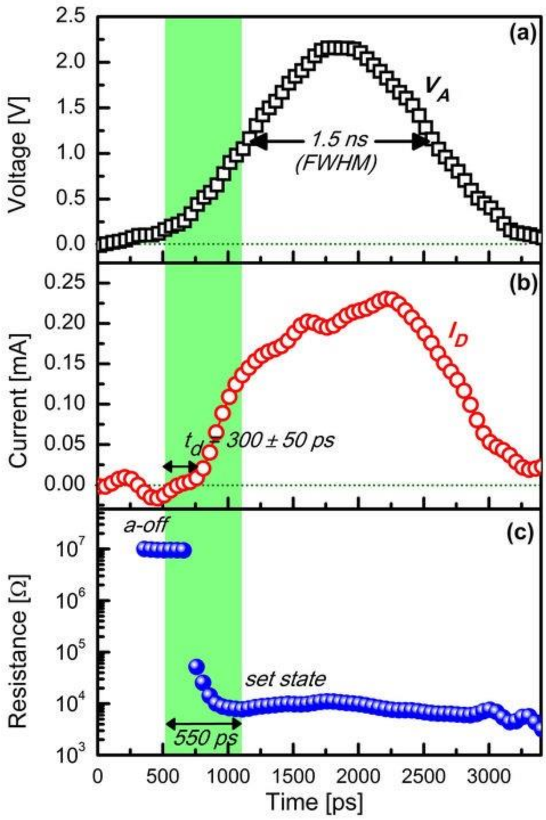

Fig. 2 *Ultrafast set operation of IST device using voltage pulse with a pulse-width of 1.5 ns (FWHM) and* t_r*,* t_f *of 1 ns*

4.4 A weak electric field-assisted ultrafast switching characteristics of In_3SbTe_2 device

Prefixing a weak electric field (incubation) might enhance the crystallization speed via pre-structural ordering and thereby achieving faster programming of phase change memory (PCM) devices. We employed a weak electric field, equivalent to a constant small voltage (that is incubation voltage, V_i of 0.3 V) to the V_A (main pulse) for a systematic understanding of voltage-dependent rapid threshold switching characteristics and crystallization (*set*) process of IST PCM devices. In order to understand the effects of incubation voltage on the dependency of transient parameters including t_d, and t_s, we used various V_A by systematically varying the pulse amplitudes from 1.0 V to 2.3 V having the pulse parameters t_r, and t_f of 1 ns and t_p of 100 ns for both cases (i.e. with and without V_i). Fig. 3 displays the device current measured for varying V_A from 1.0 V to 2.3 V for both cases such that V_i = 0 V (as marked in black line) and V_i = 0.3 V (as marked in grey line). It is very interesting to note that IST devices exhibit switching from a significantly lower V_A of 1.4 V under incubation field, below which the device remains in the high resistance amorphous off (*a-off)* state. On the other hand, in the case of no incubation field, the device exhibits switching at a higher V_A of

1.7 V. Furthermore, it is noteworthy to mention here that incubation-assisted switching characteristics unravel a strikingly small value of t_d of 300 ± 50 ps for a lower V_A of 1.7 V (i.e. 0.82 times of V_A compared to without incubation). This finding corroborates that the incubation-assisted switching offers approximately one order smaller value of t_d , revealing a remarkable threshold switching speed of 300 ± 50 ps of IST devices, which is about one order faster than that of without incubation voltage.

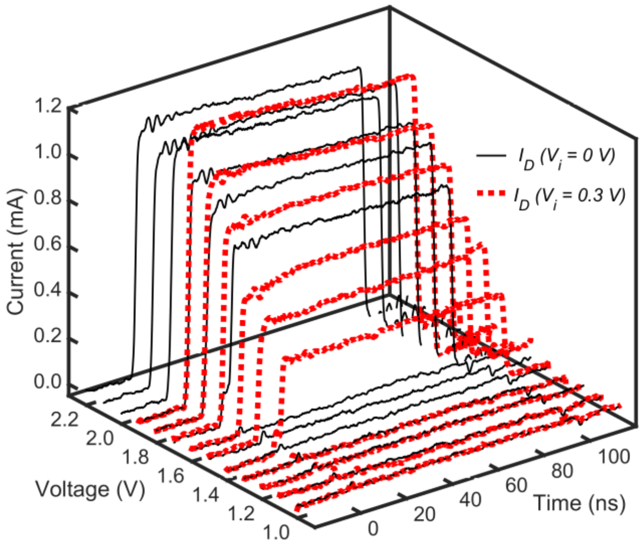

Fig. 3 *Time-resolved measurement of the device current for applied voltage pulses ranging from 1.0 to 2.2 V for both cases such as without incubation voltage (black color, solid lines) and also with incubation voltage (red color, dashed lines).*

4.5 Extremely high resistance contrast of In_3SbTe_2 device for multi-bit data storage

Current controlled promotion of crystallization offers stable multi-level resistances after threshold switching process that are being exploited for multi-bit phase change memory applications. We have demonstrated ultra-high contrast multi-level resistance characteristics of the IST device by means of a systematically varying current controlled crystallization process for a wide range of currents starting from few μA to 40 μA and the corresponding resistance levels ranging from 10 MΩ to 10 kΩ respectively as shown in Fig. 4. Current driven I-V characteristics demonstrate the high resistance sub-threshold region followed by threshold switching and subsequent multiple resistance levels. Conduction in amorphous phase and stability of multiple resistance levels have been analyzed using trap-limited sub-threshold electrical transport model based on the analytical solution [10]. The effective crystalline thicknesses are also calculated from the aforementioned model upon systematic increment in the current. Furthermore, IST devices show large resistance ratio $R_{highest}/R_{lowest}$

which is calculated to be about 570 and is much higher as compared to the earlier reported values for other PCM devices. Current induced successive curtailment in the device resistance of IST device demonstrates eight resistance states which is highly suitable for multi-bit PCM devices.

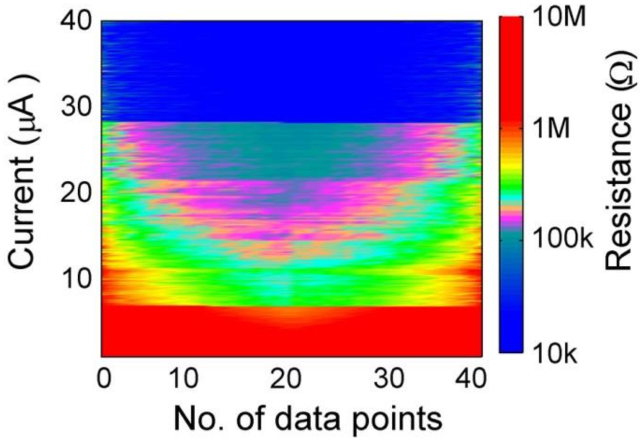

Fig. 4 *Device resistance upon systematic increment of I_a starting from 500 nA to 40 μA with increment of 100 nA.*

4.6 Structural transformation and physical properties of In_3SbTe_2 material

Search for novel phase-change materials (PCMs) has intrigued extensively for the development of high performance memory devices. The novel ternary alloy IST shows some of the promising attributes including high thermal stability for next-generation non-volatile phase change memory. Structural transformation and crystallization kinetics upon annealing of IST have been investigated by means of temperature-dependent sheet resistivity measurement and *in situ* XRD. Upon increasing the temperature a minimal change in resistivity is observed at around 300 °C, which can be attributed to the formation of crystalline InSb and InTe phases confirmed by *in situ* XRD as shown in Fig. 5. Interestingly, emergence of crystalline In_3SbTe_2 (rocksalt structure) phase is also seen at 300 °C and thereafter the peak intensity of In_3SbTe_2 phase increases. A large change in resistivity is identified at around 410 °C revealing complete crystallization. Moreover, the resistivity measurement upon heating upto various annealing temperatures and subsequent cooling displayed multiple resistive states with large resistivity contrast (more than six orders). XRR study reveals a density increase and consequent thickness decrease upon annealing. The calculated activation energies of crystallization using Kissinger's method at 300 °C and 410 °C are 5.3 eV and 5.7 eV respectively. Our experimental results indicate that In_3SbTe_2 with large resistivity contrast and more thermal stability is suitable as an active storage element for future multi-bit phase-change memory devices.

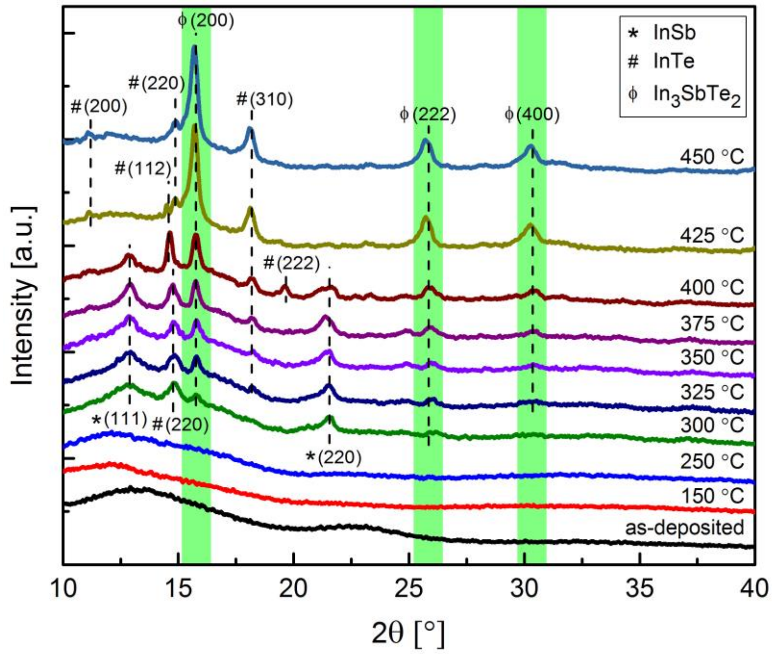

Fig. 5 Structural transformation of IST phase change material using in situ X-ray Diffraction measurement

5. CONCLUSIONS

This thesis presents the sub-nanosecond switching characteristics and *set* operation of the IST device and their suitability for multi-bit data storage for the development of high-speed, high density and non-volatile electronic memories. Our findings reveal that t_d decreases rapidly upon increasing V_A culminating in a extremely small delay time of 300 ps. Furthermore, an ultrafast *set* operation of IST devices was achieved within for a very short pulse width of 1.5 ns (FWHM) revealing a rapid phase-change from a high-resistance amorphous to a low resistance polycrystalline state. The voltage-dependent incubation-assisted threshold switching dynamics confirms that the initiation of switching process proceeds even for 0.82 times of V_A, corroborating low power switching of IST devices. Also, the incubation-driven ultrafast *set* process was achieved for V_A of a very short pulse-width of 1.5 ns and amplitude of 1.7 V, which is about 18% lower as compared to no-incubation. Furthermore, a systematic increment of programming current unfolds more than eight discrete stable resistive states with a remarkably larger programming margin (~ 570 times) between amorphous and crystalline (*set*) state revealing the multi-bit programming capabilities of IST device for high-density data storage applications. Finally, temperature-depedent resistivity and *in situ* XRD measurement corraborates excellent thermal stability and large resistance contrast of IST material evidencing their capability for multi-bit PCM applications.

6. REFERENCES

[1] M. Wuttig, *Phase-change materials-towards a universal memory?* Nat. Mater. **4**, 265 (2005).

[2] D. Loke, T. H. Lee, W. J. Wang, L. P. Shi, R. Zhao, Y. C. Yeo, T. C. Chong, and S. R. Elliot, *Breaking the Speed Limits of Phase-Change Memory*, Science **336**, 1566 (2012).

[3] M. H. R. Lankhorst, B. W. S. M. Ketelaars, and R. A. M. Wolters, *Low-cost and nanoscale non-volatile memory concept for future silicon chips*, Nat. Mater. **4**, 347 (2005).

[4] M. Wuttig, and N. Yamada, *Phase Change Materials for rewritable data storage*, Nat. Mater. **6**, 824 (2007).

[5] A. Sebastian, M. L. Gallo, and D. Krebs, *Crystal growth within a phase change memory cell*, Nat. Commun. **5**, 4314 (2014).

[6] P. Nukala, C.-C. Lin, R. Composto, and R. Agarwal, *Ultralow-power switching via defect engineering in germanium telluride phase-change memory devices*, Nat. Commun. **7**, 10482 (2016).

[7] J. H. Los, T. D. Kühne, S. Gabardi, and M. Bernasconi, *First-principles study of the amorphous* In_3SbTe_2 *phase change compound*, Phys. Rev. B **88**, 17420 (2013).

[8] J.-K. Ahn, K.-W. Park, H.-J. Jung, and S.-G. Yoon, *Phase-Change InSbTe Nanowires Grown in Situ at Low Temperature by Metal-Organic Chemical Vapor Deposition*, Nano Lett. **10**, 472 (2010).

[9] Y. Maeda, H. Andoh, I. Ikuta, and H. Minemura, *Reversible phase change optical data storage in InSbTe alloy films*, J. Appl. Phys. **64**, 1715 (1988).

[10] D. Ielmini, and Y. Zhang, *Analytical model for subthreshold conduction and threshold switching in chalcogenide-based memory devices*, J. Appl. Phys. **102**, 054517 (2007).

List of Publications (from the thesis work)

Journal Publications:

1. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *Sub-nanosecond threshold-switching dynamics and set process of In_3SbTe_2 phase-change memory devices*; **Appl. Phys. Lett.** 108**,** 233501 (2016); DOI: 10.1063/1.4953196; Impact factor: 3.41.
2. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *A weak electric field-assisted ultrafast electrical switching dynamics in In_3SbTe_2 phase-change memory devices*; **AIP Advances** 7, 075206 (2017); DOI: 10.1063/1.4994184; Impact factor: 1.60.
3. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *Extremely high contrast multilevel resistance states of In_3SbTe_2 device for high density non-volatile memory applications*; **Phys. Status Solidi RRL**, 1700227 (2017) (*in-press*); DOI: 10.1002/pssr.201700227; Impact factor: 3.03.
4. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *A fully automated temperature-dependent resistance measurement setup using van der Pauw method; (under review).*
5. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *Direct evidence for phase transitions in thin In_3SbTe_2 films revealed by temperature-dependent X-ray diffraction (in-preparation).*

International Conference Proceedings:

1. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *Investigation on In_3SbTe_2 phase change material for multi-bit data storage applications*; Proceedings of European Phase Change and Ovonic Symposium **(E\PCOS)**; pp. 132, 2016.

International Conferences:

1. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *A Systematic Study on Electrical Switching Characteristics of InSbTe Phase Change Material for Multi-bit Data Storage*; Materials Research Society **(MRS)** Spring Meeting; Phoenix, USA; 2016.
2. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *A weak electric field assisted ultrafast switching characteristics of InSbTe phase change memory devices*; International Conference on Nanoscience and Technology **(ICONSAT)**; IISER Pune, India; 2016.
3. **Shivendra Kumar Pandey** and Anbarasu Manivannan; *Electrical and structural properties of thin InSbTe films for multi-bit phase change random access memory applications*; Proceedings of International Workshop on Physics of Semiconductor Devices **(IWPSD)**; IISc Banglore, India; 2015.

List of Publications (outside the thesis work)

Journal Publications:

1. Smriti Sahu, **Shivendra Kumar Pandey**, Anbarasu Manivannan, Uday Prabhakarrao Deshpande, Vasant G. Sathe, Varimalla Raghavendra Reddy, and Murugavel Sevi; *Direct evidence for phase transition in thin $Ge_1Sb_4Te_7$ films using in situ UV–Vis–NIR spectroscopy and Raman scattering studies*; **Phys. Status Solidi B,** 253, 1069 (2016); DOI: 10.1002/pssb.201552803; Impact factor: 1.6.