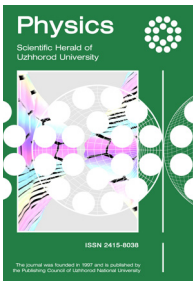
Scientific Herald of Uzhhorod University
Series "Physics"

Journal homepage: https://physics.uz.ua/en
Issue 47, 7-20
Received: 20.03.2020. Revised: 10.05.2020. Accepted: 15.08.2020



УДК 004.08; 004.85; 539.219.1; 548.213.2; 548.53
PACS 65.6+a; 73.61-r; 78.30-e; 78.66 Jq
DOI: 10.24144/2415-8038.2020.47.7-20

Investigation of the Chalcogenide Materials that Are Used for Manufacture of the Random Access Memory Devices on the Basis of Phase Transitions of these Materials

V.K. Kyrylenko[1*], M.O. Durkot[1], M.M. Pop[1,2], R.P. Pisak[1], V.M. Rubish[1]

[1]Institute for Information Recording, NAS of Ukraine
88000, 4 Zamkovi Skhody Str., Uzhgorod, Ukraine

[2]Uzhgorod National University
88000, 54 Voloshyn Str., Uzhgorod, Ukraine

Abstract

Purpose. Thin films of $Sb_xSe(Te)_{100-x}$ (provided that $x \geq 50$) are suitable for manufacture of the PRAM devices that work on the basis of the following reversible phase transition: "amorphous phase ↔ crystalline phase". This paper presents results of investigations of the temperature dependences between the electrical resistance and optical transmission coefficient in the course of changes in the conditions of thermocycling and heating conditions.

Methods. Crystallisation of thin films of $Sb_xSe(Te)_{100-x}$ (provided that $x \geq 50$) was investigated with the help of the method, which ensures simultaneous measurements of the electrical resistance and optical transmission coefficient within one and the same sample. Investigations of the $R(T)$ and $\Theta(T)$ dependences were performed on the planar structures "chromium layer – $Sb_xSe(Te)_{100-x}$ film" within the range of temperatures from 293 K up to 493°K in the continuous mode within the range of the heating rates (q) from 3°K/min. up to 6°K/min.; in the mode of thermal cycling, as well as in the heating mode on the condition of the relevant stops of the temperature sweep at the selected stabilisation temperatures (T_{st}) for a certain duration of the temperature stabilisation (t_{st}).

Results. Nonisothermal crystallisation of the amorphous films of $Sb_xSe(Te)_{100-x}$ ($x \geq 50$) is accompanied by a sharp decrease in the values of the electrical resistance and optical transmission coefficient. The transition parameters (starting temperature T_{ph} of the transition from the amorphous state into the crystalline one; temperature range of transition ΔT_{ph}; changes in the electrical resistance ΔR and optical transmission coefficient $\Delta\Theta$) depend on the chemical composition of films, as well as on the heating rate. As concerns the materials under investigation, value of the ΔT is within the range from 4°K up to 18°K, while value of the ΔR has been changed by 2 or 3 orders of magnitude. As the heating rate increases, T_{ph} and ΔT_{ph} values shift to the higher temperatures. Investigations have shown that crystallisation of the $Sb_xSe(Te)_{100-x}$ amorphous films (partial or complete crystallisation) under certain conditions can occur at the temperatures below T_{ph}. The percentage share of the crystallised volume of films and values of R, Θ, ΔR, and $\Delta\Theta$ depend on the temperature parameters and time-based parameters (for example, T_{st}, t_{st}) of the heating process.

Conclusions. It is established that $Sb_xSe(Te)_{100-x}$ ($x \geq 50$) amorphous films crystallise under certain conditions of heating. The phase transition from the amorphous state to the crystalline one is accompanied by a sharp decrease in the electrical resistance and optical transmission coefficient. The crystallisation processes and phase transition parameters depend on the chemical composition of films and on the heating conditions. The results testify that $Sb_xSe(Te)_{100-x}$ films can be used as the materials for manufacture of the temperature detectors and PRAM devices

Keywords: chalcogenide materials, amorphous films, phase transitions, crystallisation, memory cells, temperature detectors

Suggested Citation:
Kyrylenko VK, Durkot MO, Pop MM, Pisak RP, Rubish VM. Investigation of the chalcogenide materials that are used for manufacture of the random access memory devices on the basis of phase transitions of these materials. *Scientific Herald of Uzhhorod University. Series "Physics"*. 2020;(47):7-20.

*Corresponding author

Introduction

Development of the information technologies requires to develop new media and new recording methods, which would have high photoelectric sensitivity and thermosensitivity and which would ensure: high information recording density, as well as high information recording and reading speed; reversibility of the recording process and increase in quantity of the re-recording cycles; low consumption of electric power in the course of the information recording, erasing, and reading; storing of great volumes of data; long-term storing of digital information. Efficient method for solving these problems (that is, search of new photosensitive and thermosensitive structures and transition to the nanoscale sizes of the information component) will provide possibility to decrease electric power consumption in substantial degree.

The amorphous films of the chalcogenide vitreous semiconductors (CVS) are prospective materials for development of such structures, because of their internal constitution and properties can be easily changed in the following conditions: change of chemical composition; exposure to light from the spectral region of the self-absorption edge; influence of temperature, electrical field, and mechanical loads. Application of the amorphous chalcogenide films makes it possible to perform recording of the relief (three-dimensional) images having thickness of components at the level from 0.3 up to 0.8 micrometres (μm) due to irradiation at the wavelength λ=405 nanometres [1-3]. Such technology is of practical interest for development of the systems, which are intended for recording optical disks (in the formats of CD, DVD, and BD), as well as for development of the holographic diffraction gratings [5-7].

As concerns optical methods of formation of the nanoscale structures, the most practical interest is connected with the method that is based on utilization of the near optical field of nanoscale particles of the noble/precious metals, which are integrated with a thin chalcogenide film [2; 7-9].

The optomechanical method is the prospective method of formation of the nanoscale structures in the amorphous chalcogenide films. This method is based on the photoplastic effect. Generation of the nanoscale components (being from 30 up to 70 nanometres in diameter) within the zone of recording occurs in the course of the simultaneous exposure and indentation of the film surface [3; 10].

The most efficient methods are the methods of formation of the nanoscale structures with the help of the light pulse or electrical pulse within the layer of the chalcogenide material (with phase transformations), in accordance with which the following reaction is implemented: "amorphous phase ↔ crystalline phase". Recording of information on the thin layers is induced by crystallisation (amorphisation) of the nanoscale section of the chalcogenide film, and this process results in the changes of the film structure, as well as changes in optical (for example, coefficient of reflection) and electrical (resistance) characteristics of this film.

For the most part, the amorphous chalcogenide films of the ternary system Ge-Sb-Te (first of all, $Ge_2Sb_2Te_5$ and $GeSb_2Te_4$) [6; 10-17] are used in order to manufacture media of information, within which the phase reversible transitions "amorphous phase ↔ crystalline phase" (CD-RW, DVD RW, DVD–RAM, BD–RW, PCM or PRAM) are realised. As it was shown in the course of investigations [18-26], films of the multicomponent systems (In-Sb-Te, In-Sb-Se, Ga-Sb-Te, and Ag-In-Sb-Te) are the most prospective materials for manufacture of the disk media, which ensure re-recording of information, as well as for manufacture of the non-volatile memory devices (phase change memory devices, PCM, or parameter random access memory devices, PRAM). Over the last few years, active measures are taken in order to develop PRAM devices on the basis of the nominally clear chalcogenide films of various binary systems (Sb-Se, Sb-Te, and Ge-Te;

these films are always doped with various components) [27-30].

Phase transformations "amorphous phase-crystalline phase" in the chalcogenide films of binary systems (Sb-Se and Sb-Te) have been investigated already [31; 32]. In these cases, transition from the amorphous state into the crystalline one has been investigated with the help of the combined method, which makes it possible to ensure simultaneous measurements of electrical resistance (R) and optical transmission coefficient (Θ) within one and the same planar sample of the following kind: "metal layer – chalcogenide film" [31]. Semi-transparent conducting layers of nickel and chromium have been used as contacts in the course of these investigations [31; 32].

In accordance with results of these investigations, it was established that crystallisation of the amorphous films of systems of Sb-Se and Sb-Te is accompanied by a sharp decrease in R *and values*. At the same time, change of resistance (ΔR) was equal to several orders of magnitude. Temperature interval of the phase transition ΔT_{ph} from the amorphous state into the crystalline one for the samples, which have been investigated in the course of [31; 32], was within the following ranges: from 3°K up to 8°K (system of Sb-Se) and from 5°K up to 15°K (system of Sb-Te). In the course of increase of the transition heating rate (q), ΔT_{ph}, as well as the temperature of the transition from the amorphous state into the crystalline one (T_{ph}), shifts into the section of the higher temperatures. Parameters of the phase transition (T_{ph}, ΔT_{ph}, ΔR, $\Delta\Theta$) depend on chemical composition of films, on the transition heating rate, on the thermal prehistory of relevant samples, as well as on the heat treatment conditions. For example, results of [31] (investigation of the following samples: "layer of chromium film — $Sb_{65}Se_{35}$") have demonstrated that in the course of the transition "freshly-prepared sample – aged sample – annealed sample", ΔR value increases almost by two orders of magnitude.

This paper presents results of investigations of the temperature dependences between electrical resistance and optical transmission coefficient [of the amorphous films of $Sb_xSe(Te)_{100-x}$ ($x \geq 50$)] in the course of changes in the conditions of thermocycling as well as in the heating conditions.

Techniques of the Experiments

Investigations of temperature dependencies of the electrical resistance and optical transmission coefficient of the thin chalcogenide films of the "stibium-selenium" system and "stibium-tellurium" system have been performed with the help of the combined method [31], which makes it possible to perform simultaneous measurements of these parameters at one and the same planar sample of the following kind: "metal layer-chalcogenide film".

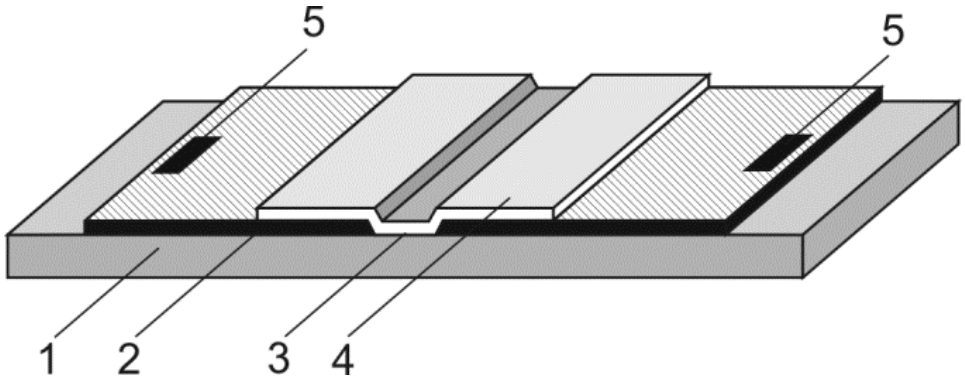


Figure 1. Structure of the planar sample: 1 – glass substrate; 2 – semi-transparent metal film; 3 – gap in the metal film; 4 – film of the material under investigation; 5 – contacts that are made of the indium-gallium paste

Figure 1 presents structure of the planar sample. In the course of these investigations, the chromium film (being 50-80 nanometres in thickness) has been used as the semi-transparent conduct-ing layers. Width of the gap in the films of the chromium, which was obtained with the help of the scribing method, was equal to approximately 5 μm. Later on, thin films of $Sb_xSe(Te)_{100-x}$ (being 50-100 nanometres in thickness) have been applied upon the region of the gap with the help of the method of vacuum thermal evaporation of the polycrystalline alloys of relevant compositions from the quasi-closed effusion cells.

The polycrystalline alloys of $Sb_xSe(Te)_{100-x}$ ($50 \le x \le 75$) have been manufactured with the help of the direct synthesis from the primitive components in the vacuumised (0.01 Pa) quartz ves-sels/vials. Mass of loads was equal to 15 grammes. Temperature of homogenisation T_h of the liquid melt has been individually selected for each chemical composition taking into account state-transition diagrams of the Sb-Se system and Sb-Te system. As concerns the materials under investi-gation, T_h of the liquid melt has not been exceeded the melting temperature by more than 50°K-80°K. Duration of homogenisation of the liquid melts is equal to 48 hours. The liquid melts have been regularly mixed. Cooling of the liquid melts has been performed in the mode of the de-energised oven.

Measurements of dependencies $R(T)$ and $\Theta(T)$ have been performed within the range of temperatures from 293°K up to 493°K within the range of the heating rates from 3°K/min. up to 6°K/min. Accuracy of temperature measurements was equal to ± 0.5°K. In the course of investigations of the $\Theta(T)$ dependencies the emitting diode with $\lambda = 880$ nanometres has been used as the source of irradiation.

Results and Discussion

As concerns all the samples under investigation, Figures 2, 3, and 4 present conventional temperature dependencies of R and Θ values of the following freshly-prepared samples: "layer of chromium – film $Sb_{65}Se_{35}$" and "layer of chromium – film $Sb_{50}Te_{50}$". These dependencies were obtained within the range of the heating rates from 3°K/min. up to 6°K/min. It is worth to note that as concerns the amorphous state, the samples with the films of the "Sb-Te" system have the lesser resistance values as compared with the samples with the films of the "Sb-Se" system.

The presented data (Figures 2, 3, and 4) demonstrate that crystallisation of films is accompanied by a sharp decrease in the values of R and Θ. Temperature interval of the transition from the amorphous state into the crystalline one is within the range from 4°K up to 18°K. In the course of increase of the transition heating rate this interval shifts into the section of the higher temperatures. In the course of increase of q values into the section of the higher temperatures, temperature of the phase transition is shifted as well. It is worth to note that in such conditions of the experiment, the "amorphous phase-crystalline phase" transition is not reversible one. In the course of cooling, values of the electrical resistance and optical transmission coefficient of the crystallised films (both the "Sb-Se" system, and the "Sb-Te" system) continue to stay practically unchanged.

From a practical point of view, it is important to take into account the information on the time stability and thermal stability of the materials under investigation. In view of this, the following investigations have been performed: investigations of the thermocycling influence upon the electrical resistance and optical transmission coefficient of the "layer of chromium – chalcogenide film" planar samples, as well as investigations of the $R(T)$ and $\Theta(T)$ dependencies on the condition of the relevant stops of the time-base sweep at the selected temperatures.

Figures 5, 6, and 7 present conventional (for all the samples under investigation) temperature dependencies of R and Θ values of the planar samples "layer of chromium — film $Sb_{65}Se_{35}$" and

"layer of chromium – film $Sb_{50}Te_{50}$" in the course of thermocycling (heating ↔ cooling). In this case, heating of the samples has been performed at the predetermined value of the transition-heating rate, while cooling has been performed in the mode of the de-energised heating component. It is obvious that at the temperatures, which are substantially lower than T_{ph} (for example, 66°C at the dependencies of $R(T)$ and 63°C at the dependencies of $\Theta(T)$, changes in the temperature dependencies of R and Θ values are practically absent (Fig. 4). At the higher temperatures and at the stepped change of temperature (in the course of each subsequent cycle, the heating temperature has been increased by a certain value), hysteresis effects (Fig. 5, 6) are obvious at the dependencies of $R(T)$ and $\Theta(T)$. These effects are the most obvious and prominent at the temperature dependencies of the optical transmission coefficient. Decrease (stepped) in the electrical resistance and optical transmission coefficient evidences that there exists partial crystallisation of the chalcogenide films, as well as that change in the Θ value is essential. Similar behaviour of the $\Theta(T)$ dependencies in the course of thermocycling was also found and established in the course of investigations of the amorphous films of selenium [33]. In accordance with analysis of the obtained results, it is possible to state that that specific share of the crystallised volume of films depends on the modes of the heat treatment. This fact evidences that within a certain temperature interval films of the "Sb-Se" system and "Sb-Te" system can be used as the indicative materials in order to record thermal fields.

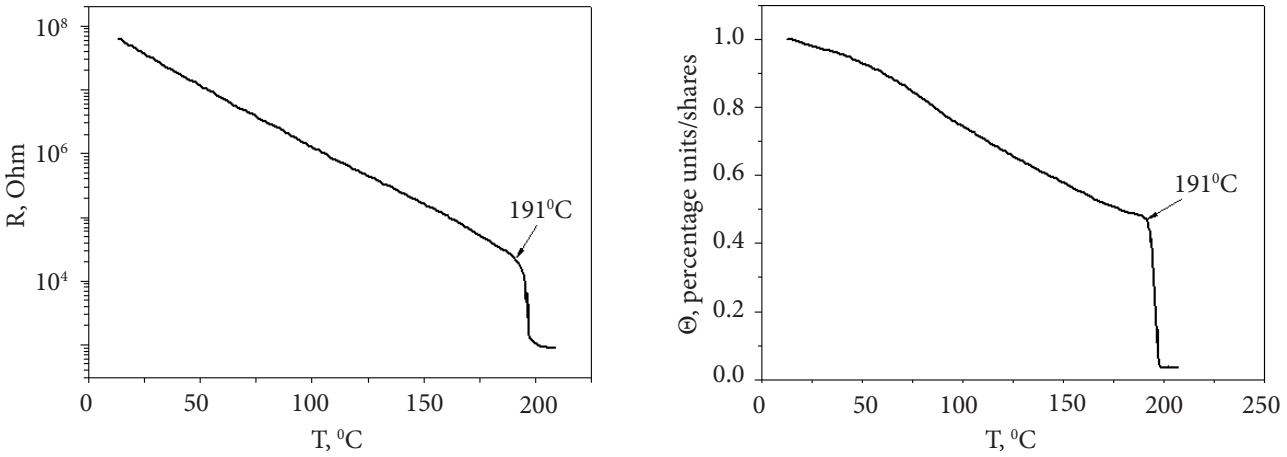


Figure 2. Temperature dependencies of $R(T)$ and $\Theta(T)$ of the "layer of chromium – film $Sb_{65}Se_{35}$" freshly-prepared sample at q=3°K/min

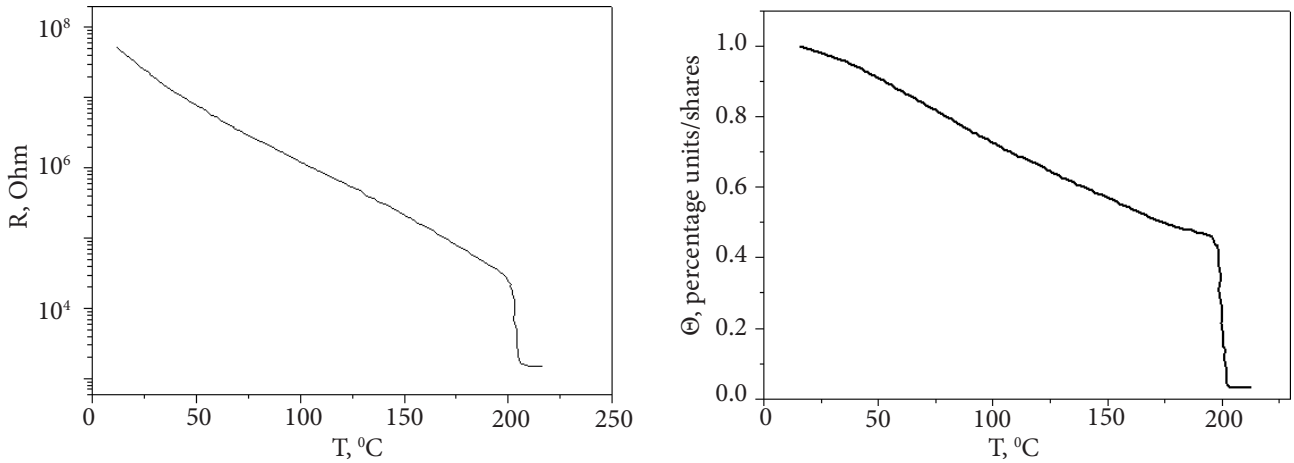


Figure 3. Temperature dependencies of $R(T)$ and $\Theta(T)$ of the "layer of chromium – film $Sb_{65}Se_{35}$" freshly-prepared sample at q=6°K/min

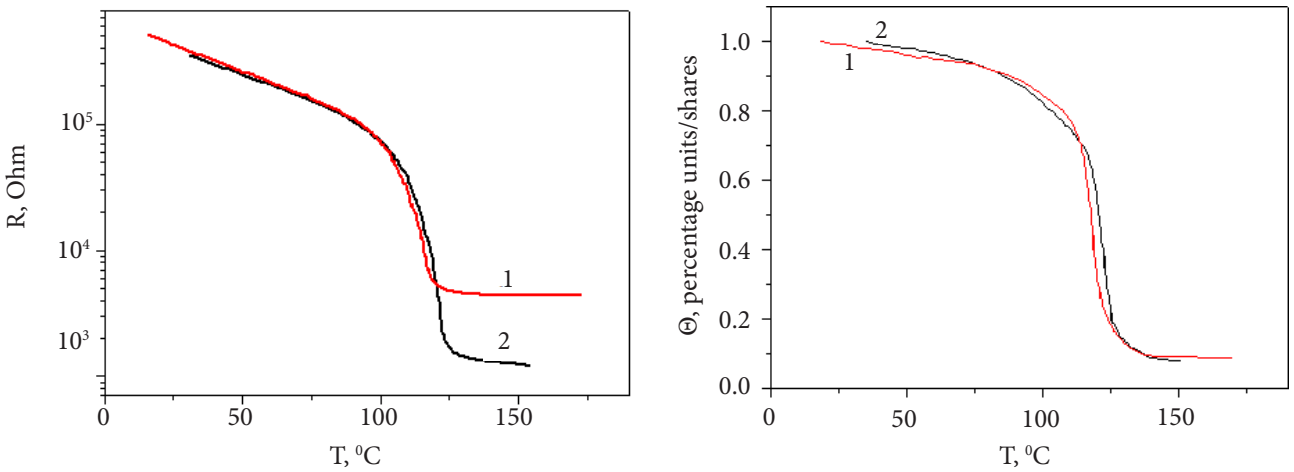


Figure 4. Temperature dependencies of $R(T)$ and $\Theta(T)$ of the "layer of chromium – film $Sb_{50}Te_{50}$" freshly-prepared sample at q=3°K/min. (1) and 6°K/min. (2)

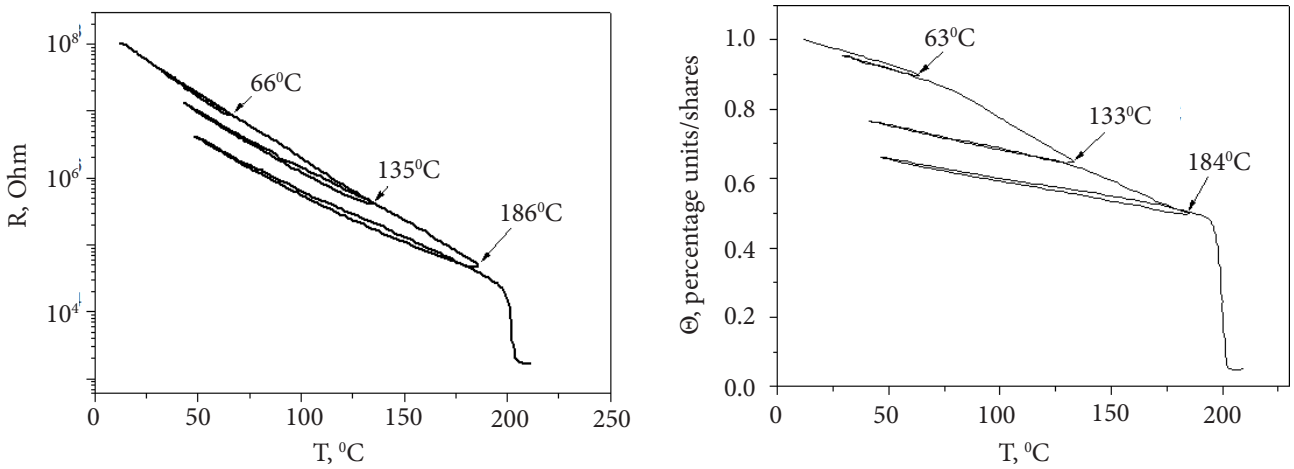


Figure 5. Changes in the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{65}Se_{35}$" sample in the course of thermocycling (q=6°K/min.) (see explanations in the text)

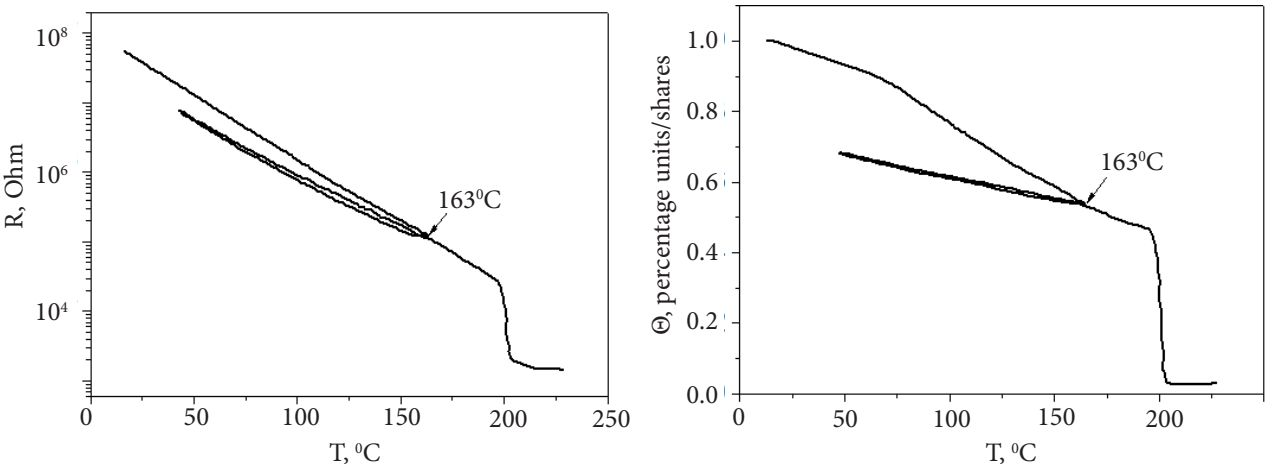


Figure 5. Changes in the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{65}Se_{35}$" sample in the course of thermocycling (q=6°K/min.) (see explanations in the text)

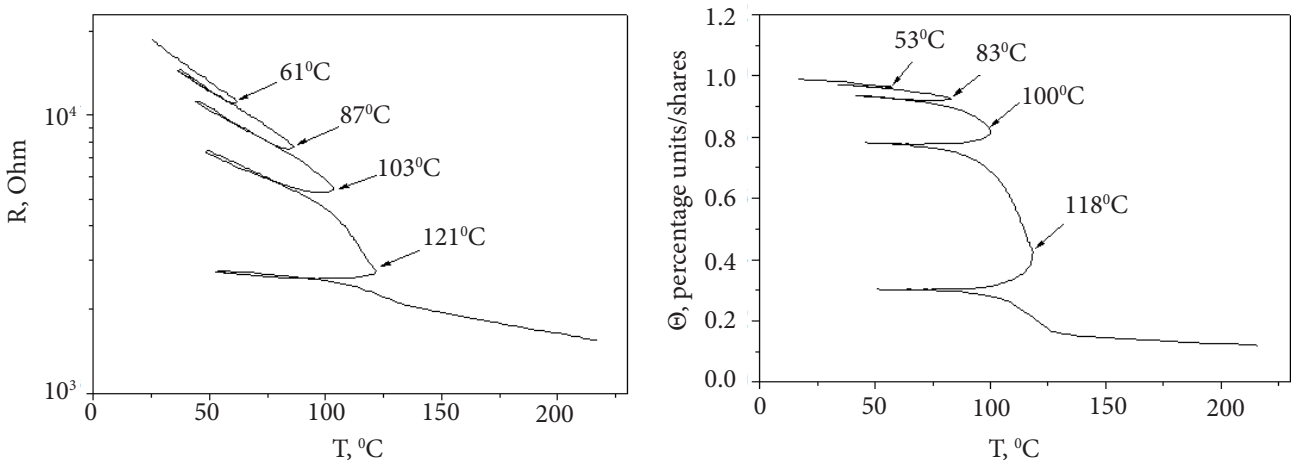


Figure 7. Changes in the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{50}Te_{50}$" sample in the course of thermocycling (q=6°K/min.) (see explanations in the text)

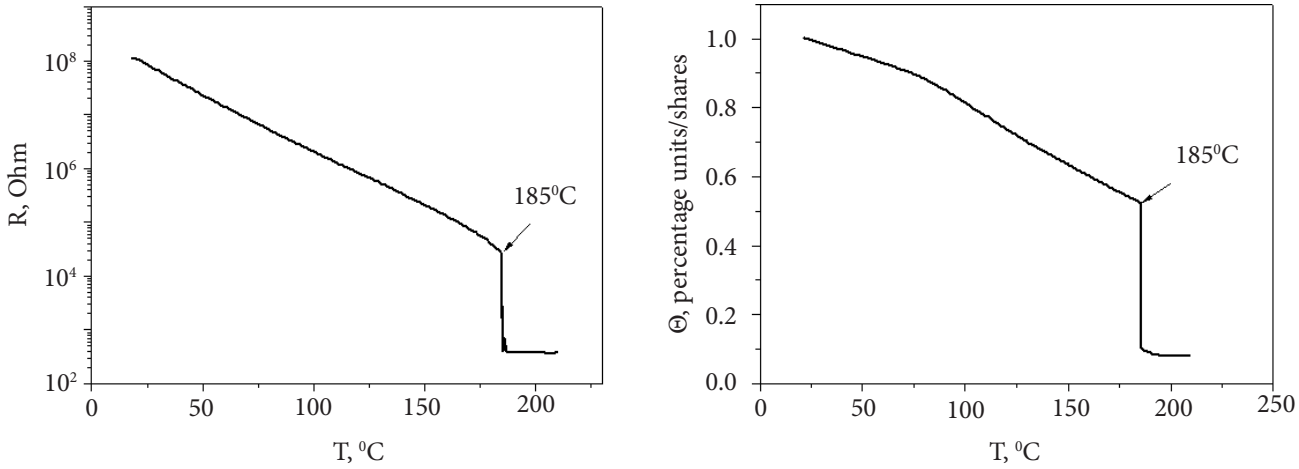


Figure 8. Dependencies of the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{55}Se_{45}$" sample (q=6°K/min.) in the case of the stop of the temperature sweep by 20 minutes at T=185°C

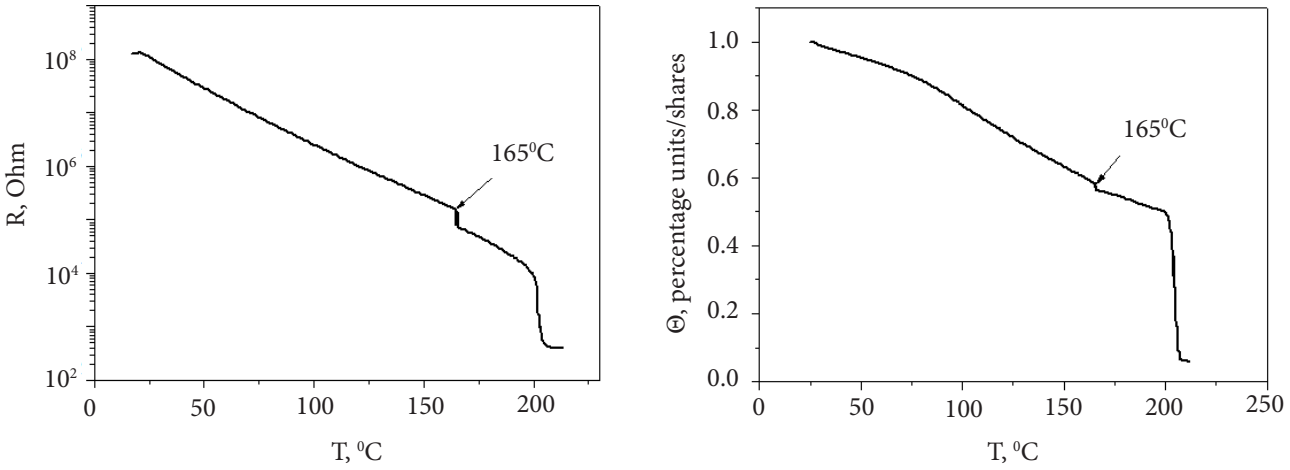


Figure 9. Dependencies of the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{55}Se_{45}$" sample (q=6°K/min.) in the case of the stop of the temperature sweep by 30 minutes at T=165°C

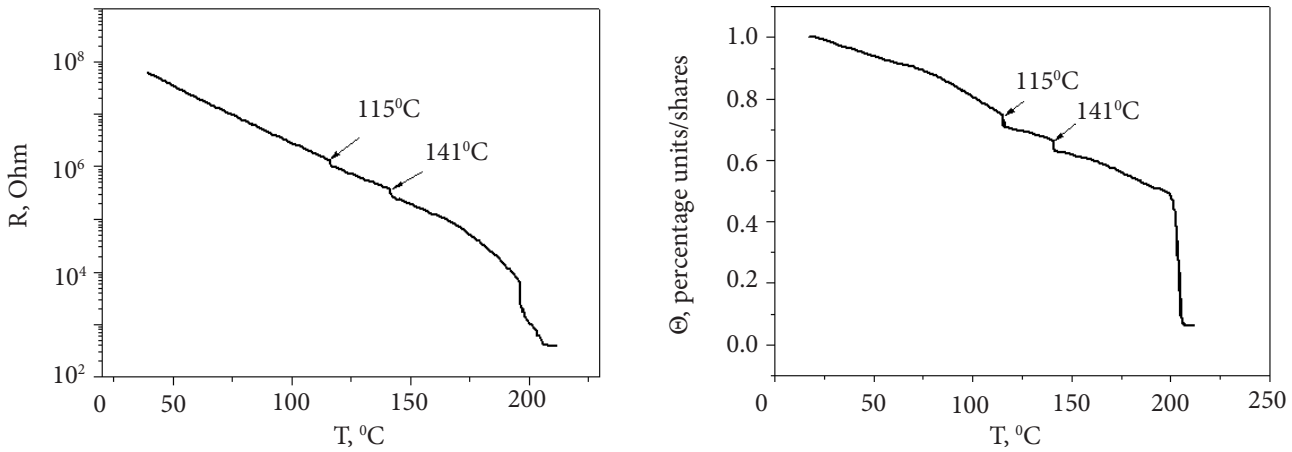


Figure 10. Dependencies of the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{55}Se_{45}$" sample (q=6°K/min.) in the case of the stops of the time-base sweep by 30 minutes at the temperatures 115°C and 140°C

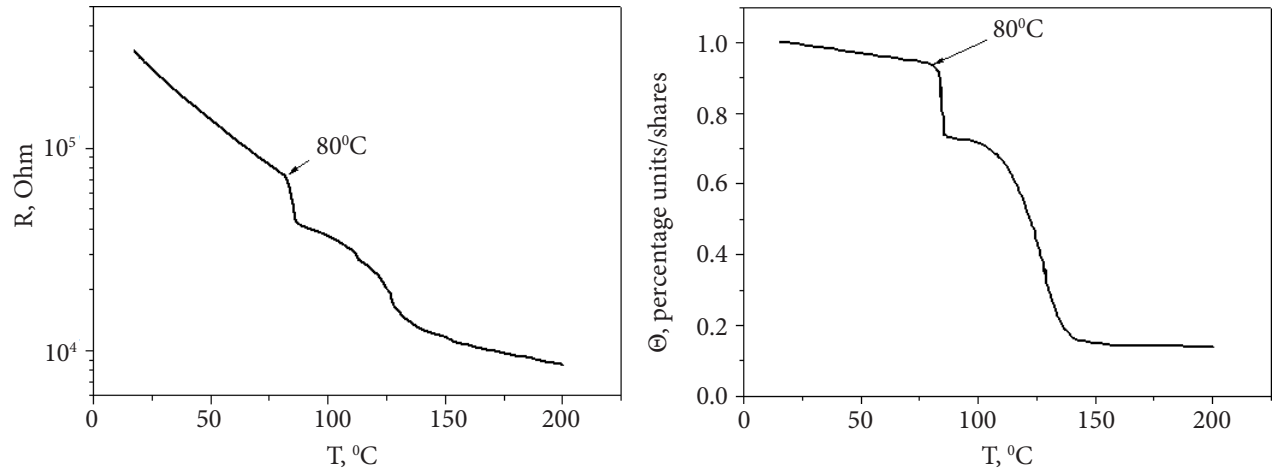


Figure 11. Dependencies of the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{50}Te_{50}$" sample (q=3°K/min.) in the case of the stop of the temperature sweep by 60 minutes at T=80°C

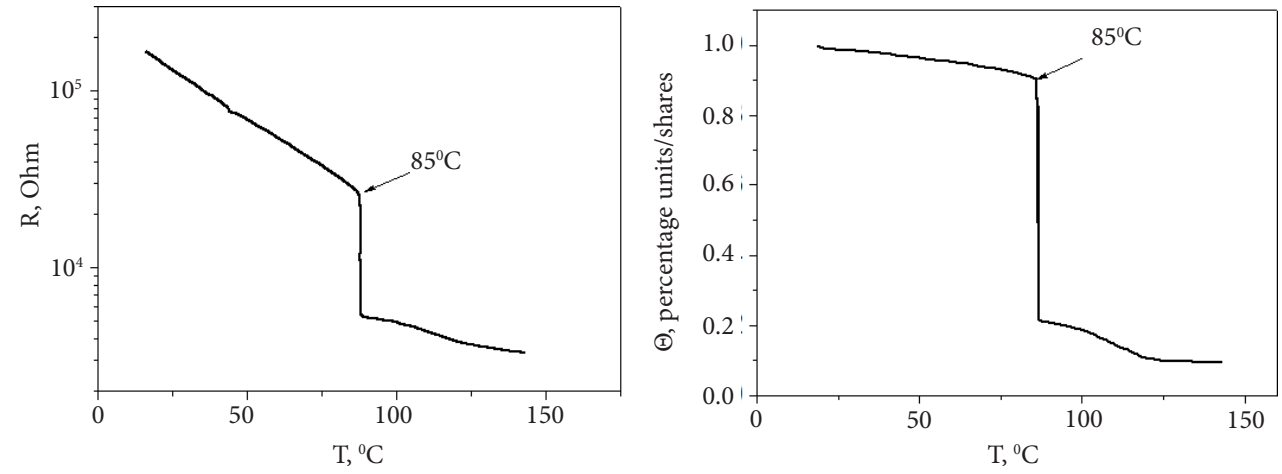


Figure 12. Dependencies of the $R(T)$ and $\Theta(T)$ values of the "layer of chromium – film $Sb_{50}Te_{50}$" sample (q=3°K/min.) in the case of the stop of the temperature sweep by 45 minutes at T=85°C

Interesting results were obtained in the course of investigations of the temperature dependencies of R and Θ values of the planar "layer of chromium – chalcogenide film" structures at the change in the conditions of the samples heating. Figures 8, 9, 10, 11, and 12 present dependencies of the $R(T)$ and $\Theta(T)$ values of the "film of chromium – film of $Sb_{55}Se_{45}$" and "layer of chromium – film of $Sb_{50}Te_{50}$" samples within the range of the heating rates from 3°K/min. up to 6°K/min. On the condition of the relevant stops of the time-base sweep for a certain period of time. In such mode of heating at the selected temperatures (T_{st}), temperature control valve has been switched into the mode of the temperature stabilization for a certain period of time (t_{st}).

As it is evident from the Figure 8, a stop of the heating process at the temperatures, which are close to the temperature of transition of the chalcogenide film into the crystalline state T_{ph}, has no practical influence upon the form of the temperature dependencies of R and Θ. Crystallisation begins at the predetermined selected temperature (for example, at 185°C in the Fig. 8), and this process is accompanied by decrease in the electrical resistance and optical transmission coefficient. In the course of comparison of these dependencies of $R(T)$ and $\Theta(T)$ with the temperature dependencies of R and Θ values of the freshly-prepared samples, it is possible to note that crystallisation has occurred within the entire volume of the chalcogenide film. As concerns stops of the temperature sweep at the temperatures, which are lower than the Tph (Fig. 9, 10), somewhat another patterns are observed in these situations. In the case of a stop of the heating process at the certain temperature (for example, at 165°C as it can be seen in Fig. 9), and in the case of subsequent switching the temperature control valve into the mode of the temperature stabilisation during a certain period of time, R and Θ values decrease.

Following the moment of switching on the process of heating, form of the dependencies $R(T)$ and $\Theta(T)$ is the same as before the stop of the temperature sweep. It is worth to note that the stepped nature of the temperature dependencies of the electric resistance and the optical transmission coefficient in the case of changes in the conditions of heating, is also observed at the temperatures, which are substantially lower than T_{ph} (Fig. 10). The more clear and prominent dependencies of $R(T)$ and $\Theta(T)$ were obtained in the mode of heating with stops of the time-base sweep for the planar samples, which were made on the basis of the antimony tellurides (stibium tellurides, see Figures 11 and 12). In the case of stop of the heating process and subsequent temperature stabilisation at the values of $T<T_{ph}$ during 60 minutes, substantial decrease of the R and Θ values is observed in the same manner as it was observed for the samples, which were made on the basis of the films of the antimony selenides. In this case (as it can be seen from Figures 11 and 12), forms of thermal and time-based dependencies of the electrical resistance and optical transmission coefficient are determined (for the most part) by the value of the temperature, at the moment of achievement of which the temperature control valve is switched into the mode of the temperature stabilisation. Comparison of these Figures demonstrates that changes in the values of $\Delta R(T)$ and $\Delta\Theta(T)$ in the case, which is presented in Fig. 12 (a stop of the heating process at 85°C during 45 minutes), are substantially higher as compared with the case, which is presented in Fig. 11 (a stop of the heating process at 80°C during 60 minutes). The obtained results let us make the following conclusion: it is possible to control the crystallisation process through changing thermal and time-based modes of the heating process.

Conclusions

It was established that the thin films of $Sb_xSe(Te)_{100-x}$ (provided that x ≥ 50) are crystallised at the certain conditions of the heating process. The phase transition from the amorphous

state into the crystalline one is accompanied by a sharp decrease in the values of the electrical resistance and optical transmission coefficient. Parameters of the transition (T_{ph}, ΔT_{ph}, ΔR, $\Delta\Theta$, percentage share of the crystallised volume) depend on the chemical compositions of relevant films, as well as on the conditions of heating (q, T_{st}, t_{st}) and thermocycling, that is, it is possible to control the crystallisation process through changing thermal and time-based modes of processing these films. The obtained results confirm possibility of application of the $Sb_xSe(Te)_{100-x}$ (provided that $x \geq 50$) films as the threshold detectors of the temperature and of the materials that are used for manufacture of the parameter random access memory devices (PRAM devices).

References

[1] Kryuchin AA, Kostyukevich SA. Analysis of methods of forming the nanoscale relief structures on the surface of storage media. *Data Recording, Storage & Processing*. 2010;12(1):3-11.
[2] Petrov VV, Kryuchin AA, Kunitsky YuA, Rubis VM, Lapchuk AS, Kostyukevich SO. [Methods of nanolithography]. Kyiv: Naukova dumka; 2015. 262 p. Ukrainian.
[3] Petrov VV, Litvin PM, Trunov ML, Kryuchin AA, Belyak EV, Rubish VM, et al. Methods of forming of the nanoscale structures on the films of the chalcogenide vitreous semiconductors. *Data Recording, Storage & Processing*. 2016;18(1):3-13.
[4] Petrov VV, Kryuchin AA, Shanoilo SM, Kraets VG, Kossko IO, Beliak EV, et al. [Superdense optical recording of information]. Kyiv: Institute of the Information Recording Problems of the National Academy Sciences of Ukraine (NASU); 2009. 283 p. Ukrainian.
[5] Neilsona JR, Kovalskiy A, Vlček M, Jain H, Miller F. Fabrication of nano-gratings in arsenic sulphide films. *J. Non-Cryst. Solids*. 2007;353(13-15):1427-30.
[6] Petrov VV, Kryuchin AA, Kostiukevich SO, Rubish VM. Inorganic photolithography. Kyiv: Institute of the Metal Physics of the NAS of Ukraine; 2007. 195 p.
[7] Korotun AV, Koval AO, Kryuchin AA, Rubish VM, Petrov VV, Titov IM. [Nanoscale photonic technologies. modern state and prospects]. Uzhhorod: Individual entrepreneur Sabov A.M.; 2019. 482 p. Ukrainian.
[8] Trunov ML, Lytvyn PM, Nagy PM, Oberemok OS, Durkot MO, Tarnaii AA, et al. Light-induced mass transport in amorphous chalcogenides/gold nanoparticles composites. *SPQEO*. 2013;16(4):354-61.
[9] Trunov ML, Lytvyn PM, Nagy PM, Csik A, Rubish VM, Kökényesi S. Light-induced mass transport in amorphous chalcogenides: Toward surface plasmon-assisted nanolithography and near-field nanoimaging. *Phys. Stat. Solidi B*. 2014;251(7):1354-62.
[10] Kryuchyn AA, Petrov VV, Rubish VM, Trunov ML, Lytvyn PM, Kostyukevich SA. Formation of nanoscale structures on chalcogenide films. *Phys. Stat. Solidi B*. 2017;255(6):1700405.
[11] Kozyukhin SA, Sherchenkov A.A. Prospects of use of the chalcogenide alloys in the phase memory elements. *Appendix to the "Vestnik RGRTU" Magazine*. 2009;(4):81-7.
[12] Terao M, Morikawa T, Ohta T. Electrical phase-change memory: Fundamentals and state of the art. *Jap. J. Appl. Phys.* 2009;(48):080001.
[13] Tsendina KD, editor. [Electronic phenomena in the chalcogenide vitreous semiconductors]. St. Petersburg: Nauka; 1996. 486 p. Russsian.
[14] Kado H, Tonda T. Nanometer-scale recording on chalcogenide films with an atomic force microscope. *Appl. Phys. Lett.* 1995;(66):2961-62.
[15] Yamada N. Origin, secret, and application of the ideal phase-change material GeSbTe. *Phys. Stat. Solidi B.* 2012;249(10):1837-42.
[16] Wüttig M, Yamada N. Phase-change materials for rewriteable data storage. *Nat. Mater.* 2007;(6):824-32.
[17] Ohta T. Phase-change optical memory promotes the DVD optical disk. *J. Optoelectron. Adv. Mater.* 2001;3(3):609-26.

[18] Matsunaga T, Umetani Y, Yamada N. Structural study of a Ag3.4In3.7Sb76.4Te16.5 quadruple compound utilized for phase-change optical disks. *Phys. Rev. B.* 2001;(64):184116.
[19] Kolobov AV, Tominaga J. Chalcogenide glasses in optical recording: Recent progress. *J. Optoelectron. Adv. Mater.* 2002;4(3):679-86.
[20] Rubish VM, Shtets PP, Rubish VV, Semak DG, Tsizh BR. Optical media for information recording based on amorphous layers of Sb– Se–In system. *J. Optoelectron. Adv. Mater.* 2003;5(5):1193-97.
[21] Song WD, Shi LP, Miao S, Chong TC. Phase change behaviors of Sn-doped Ge-Sb-Te material. *Appl. Phys. Lett.* 2007;(90):091904.
[22] Cheng HY, Kao KF, Lee CM, Chin TS. Characteristics of Ga-Sb-Te films for phase-change memory. *IEEE Transactions on Magnetics.* 2007;43(2):927-29.
[23] Kim YT, Kim S-I. Comparison of thermal stabilities between Ge-Sb-Te and In-Sb-Te phase change materials. *Appl. Phys. Lett.* 2013;(103):121906.
[24] Kim ET, Lee JY, Kim YT. Investigation of electrical characteristics of the In3Sb1Te2 ternary alloy for application in phase-change memory. *Phys. Stat. Solidi RRL.* 2009;4(3):103-5.
[25] Petrov VV, Kryuchin AA, Rubish VM. [Materials of the prospective optoelectronic devices]. Kyiv: Naukova dumka; 2015. 335 p. Russian.
[26] Bilovol V. Exploring the applicability of the amorphous films of the Is-Sb-Te system as the phase change materials. *J. Non-Cryst. Solids*. 2016;(447):315-21.
[27] Yoon S-M, Lee N-Y, Ryu S-O, Choi K-J, Park Y-S, Lee S-Y, et al. Sb-Se-based phase-change memory device with lower power and higher speed operations. *IEEE Electron Device Lett.* 2006;27(6):445-47.
[28] Wang F, Zhang T, Song Z, Liu C, Wu L, Liu B, et al. Temperature influence on electrical properties of Sb-Te phase-change material. *Jap. J. Appl. Phys.* 2008;47(2):843-46.
[29] Kozyukhin SA, Sherchenkov AA, Babich AV. Phase separation in chalcogenide semiconductors of the Ge-Te system upon thermal cycling. *Journal Proceedings of Universities. Electronics*. 2013;100(2):3-8.
[30] Rossa JA, Rodriguez OC, Ureña MA, Bilovol V, Fontana M. Switching effect properties on Sn-doped $Sb_{70}Te_{30}$ thin films. In: Amorphous and Nanostructured Chalcogenides; 2019 June-July 30-4; Chisinau. P. 24-25.
[31] Kyrylenko VK, Marjan VM, Durkot MO, Rubish VM. Investigation of the amorphous chalcogenide materials of the memory elements based on the phase transitions. *Data Recording, Storage & Processing*. 2014;16(2):7-13.
[32] Rubish VM, Kyrylenko VK, Durkot MO, Maryan VM, Tarnaj AA, Gorina OV. Phase transition in the Sb_xTe_{100-x} amorphous films. *Scientific Herald of the Uzhhorod University. Series Physics.* 2016;(39):44-50.
[33] Gasinets SM, Gorina OV, Kozusenok AV, Regan MYu, Rubish VM, Popovich KO, et al. Influence of the thermal prehistory upon processes of the non-isothermal crystallization of the vitreous and amorphous selenium. Nanosystems, Nanomaterials, Nanotechnologies. 2010;8(3):521-33.

Список використаної літератури

[1] Крючин АА, Костюкевич СА. Анализ методов получения наноразмерных рельефных структур на поверхности носителей информации. *Реєстрація, зберігання і обробка даних*. 2010;12(1):3–11.
[2] Петров ВВ, Крючин АА, Куницький ЮА, Рубіш ВМ, Лапчук АС, Костюкевич СО. Методи нанолітографії. Київ: Наукова думка; 2015. 262 с.
[3] Петров ВВ, Литвин ПМ, Трунов МЛ, Крючин АА, Беляк ЄВ, Рубіш ВМ, та ін. Методи формування нанорозмірних структур на плівках халькогенідних склоподібних напівпровідників. *Реєстрація, зберігання і обробка даних.* 2016;18(1):3–13.
[4] Петров ВВ, Крючин АА, Шанойло СМ, Краєць ВГ, Коско ІО, Беляк ЄВ, та ін. Надщільний оптичний запис інформації. Київ: ІПРІ НАНУ; 2009. 283 с.

[5] Neilsona JR, Kovalskiy A, Vlček M, Jain H, Miller F. Fabrication of nano-gratings in arsenic sulphide films. *J. Non-Cryst. Solids.* 2007;353(13-15):1427–30.
[6] Крючин АА, Костюкевич СО, Рубіш ВМ. Неорганічна фотолітографія. Київ: ІМФ НАНУ; 2007. 195 с.
[7] Коротун АВ, Коваль АО, Крючин АА, Рубіш ВМ, Петров ВВ, Тітов ІМ. Нанофотонні технології. Сучасний стан і перспективи. Ужгород: ФОП Сабов А.М.; 2019. 482 с.
[8] Trunov ML, Lytvyn PM, Nagy PM, Oberemok OS, Durkot MO, Tarnaii AA, et al. Light-induced mass transport in amorphous chalcogenides/gold nanoparticles composites. *SPQEO*. 2013;16(4):354–61.
[9] Trunov ML, Lytvyn PM, Nagy PM, Csik A, Rubish VM, Kökényesi S. Light-induced mass transport in amorphous chalcogenides: Toward surface plasmon-assisted nanolithography and near-field nanoimaging. *Phys. Stat. Solidi B*. 2014;251(7):1354–62.
[10] Kryuchyn AA, Petrov VV, Rubish VM, Trunov ML, Lytvyn PM, Kostyukevich SA. Formation of nanoscale structures on chalcogenide films. *Phys. Stat. Solidi B*. 2017;255(6):1700405.
[11] Козюхин СА, Шерченков АА. Перспективы применения халькогенидных сплавов в элементах фазовой памяти. *Приложение к журналу «Вестник РГРТУ»*. 2009;(4):81–7.
[12] Terao M, Morikawa T, Ohta T. Electrical phase-change memory: Fundamentals and state of the art. *Jap. J. Appl. Phys*. 2009;(48):080001.
[13] Цэндина КД, редактор. Электронные явления в халькогенидных стеклообразных полупроводниках. Санкт-Петербург: Наука; 1996. 486 с.
[14] Kado H, Tonda T. Nanometer-scale recording on chalcogenide films with an atomic force microscope. *Appl. Phys. Lett.* 1995;(66):2961–62.
[15] Yamada N. Origin, secret, and application of the ideal phase-change material GeSbTe. *Phys. Stat. Solidi B.* 2012;249(10):1837–42.
[16] Wüttig M, Yamada N. Phase-change materials for rewriteable data storage. *Nat. Mater.* 2007;(6):824–32.
[17] Ohta T. Phase-change optical memory promotes the DVD optical disk. *J. Optoelectron. Adv. Mater.* 2001;3(3):609–26.
[18] Matsunaga T, Umetani Y, Yamada N. Structural study of a Ag3.4In3.7Sb76.4Te16.5 quadruple compound utilized for phase-change optical disks. *Phys. Rev. B.* 2001;(64):184116.
[19] Kolobov AV, Tominaga J. Chalcogenide glasses in optical recording: Recent progress. *J. Optoelectron. Adv. Mater.* 2002;4(3):679–86.
[20] Rubish VM, Shtets PP, Rubish VV, Semak DG, Tsizh BR. Optical media for information recording based on amorphous layers of Sb– Se–In system. *J. Optoelectron. Adv. Mater.* 2003;5(5):1193–97.
[21] Song WD, Shi LP, Miao S, Chong TC. Phase change behaviors of Sn-doped Ge-Sb-Te material. *Appl. Phys. Lett.* 2007;(90):091904.
[22] Cheng HY, Kao KF, Lee CM, Chin TS. Characteristics of Ga-Sb-Te films for phase-change memory. *IEEE Transactions on Magnetics.* 2007;43(2):927–29.
[23] Kim YT, Kim S-I. Comparison of thermal stabilities between Ge-Sb-Te and In-Sb-Te phase change materials. *Appl. Phys. Lett.* 2013;(103):121906.
[24] Kim ET, Lee JY, Kim YT. Investigation of electrical characteristics of the In3Sb1Te2 ternary alloy for application in phase-change memory. *Phys. Stat. Solidi RRL.* 2009;4(3):103–5.
[25] Петров ВВ, Крючин АА, Рубиш ВМ. Материалы перспективных оптоэлектронных устройств. Київ: Наукова думка; 2012. 336 с.
[26] Bilovol V. Exploring the applicability of amorphous films of system Is-Sb-Te as phase change materials. *J. Non-Crystalline Solids.* 2016;(447):315–21.
[27] Yoon S-M, Lee N-Y, Ryu S-O, Choi K-J, Park Y-S, Lee S-Y, et al. Sb-Se-based phase-change memory device with lower power and higher speed operations. *IEEE Electron Device Lett.* 2006;27(6):445–47.
[28] Wang F, Zhang T, Song Z, Liu C, Wu L, Liu B, et al. Temperature influence on electrical properties of Sb-Te phase-change material. *Jap. J. Appl. Phys.* 2008;47(2):843–46.

[29] Kozyukhin SA, Sherchenkov AA, Babich AV. Phase separation in chalcogenide semiconductors of the Ge-Te system upon thermal cycling. *Journal Proceedings of Universities. Electronics.* 2013;100(2):3–8.
[30] Rossa JA, Rodriguez OC, Ureña MA, Bilovol V, Fontana M. Switching effect properties on Sn-doped Sb70Te30 thin films. In: Amorphous and Nanostructured Chalcogenides; 2019 June-July 30–4; Chisinau. P. 24–25.
[31] Кириленко ВК, Мар'ян ВМ, Дуркот МО, Рубіш ВМ. Дослідження аморфних халькогенідних матеріалів елементів пам'яті на основі фазових переходів. *Реєстрація, зберігання і обробка даних*. 2014;16(2):7–13.
[32] Рубіш ВМ, Кириленко ВК, Дуркот МО, Мар'ян ВМ, Тарнай АА, Горіна ОВ. Фазові перетворення в аморфних плівках SbxTe100-x. *Науковий вісник Ужгородського університету. Серія «Фізика».* 2016;(39):44–50.
[33] Гасинець СМ, Горіна ОВ, Козусенок ОВ, Риган МЮ, Рубіш ВМ, Попович КО, та ін. Вплив термічної передісторії на процеси неізотермічної кристалізації склоподібного та аморфного селену. *Наносистеми, наноматеріали, нанотехнології*. 2010;8(3):521–33.

Дослідження халькогенідних матеріалів для пристроїв пам'яті з довільним доступом на базі фазових переходів

В.К. Кириленко[1], М.О. Дуркот[1], М.М. Поп[1,2], Р.П. Пісак[1], В.М. Рубіш[1]

Ужгородська лабораторія матеріалів оптоелектроніки та фотоніки Інституту проблем реєстрації інформації НАН України
88000, вул. Замкові сходи, 4, м. Ужгород, Україна

Ужгородський національний університет
88000, вул. Волошина, 54, м. Ужгород, Україна

Анотація

Мета. Тонкі плівки $Sb_xSe(Te)_{100-x}$ (за умови, що $x \geq 50$) придатні для виробництва пристроїв PRAM, які працюють на основі такого фазового реверсивного переходу: «аморфна фаза ↔ кристалічна фаза». У цій статті представлені результати дослідження температурних залежностей між електричним опором та коефіцієнтом оптичного пропускання при зміні умов термоциклювання та нагрівання.

Методи. Кристалізацію тонких плівок $Sb_xSe(Te)_{100-x}$ (за умови, що $x \geq 50$) досліджували за допомогою методу, який забезпечує одночасне вимірювання електричного опору та коефіцієнта оптичного пропускання в одному і тому ж зразку. Вимірювання залежностей $R(T)$ та $\Theta(T)$ проводилися на площинних структурах «шар хрому – $Sb_xSe(Te)_{100-x}$ плівка» в діапазоні температур від 293 К до 493 К у безперервному режимі в межах швидкості нагріву (q) від 3 К/хв. до 6 К/хв.; у режимі термоциклюванні, а також у режимі нагріву за умови відповідних зупинок температурної розгортки при обраних температурах стабілізації (T_{cm}) протягом певної тривалості стабілізації температури (t_{cm}).

Результати. Неізотермічна кристалізація аморфних плівок $Sb_xSe(Te)_{100-x}$ ($x \geq 50$) супроводжується різким зменшенням значень електричного опору та коефіцієнта оптичного пропускання. Параметри переходу (початкова температура $T_{ф}$ переходу з аморфного стану в кристалічний; діапазон температур переходу $\Delta T_{ф}$; зміна електричного опору ΔR та коефіцієнта оптичного пропускання $\Delta\Theta$) залежать також від хімічного складу плівок та умов нагрівання. Що стосується досліджуваних матеріалів, значення ΔT знаходиться в діапазоні від 4 °К до 18 °К, тоді як значення ΔR було змінено на 2 або 3 порядки. Зі збільшенням швидкості нагрівання значення $T_{ф}$ і $\Delta T_{ф}$ зсуваються в ділянку вищих температур. Дослідження показали, що кристалізація аморфних плівок $Sb_xSe(Te)_{100-x}$ (часткова або повна кристалізація) за певних умов може відбуватися при температурах нижче $T_{ф}$. Процентна частка кристалізованого об'єму плівок та значення R, Θ, ΔR та $\Delta\Theta$ залежать від температурних параметрів і параметрів, що залежать від часу (наприклад, T_{cm}, t_{cm}) процесу нагрівання.

Висновки. Встановлено, що аморфні плівки $Sb_xSe(Te)_{100-x}$ ($x \geq 50$) кристалізуються за певних умов нагрівання. Фазовий перехід з аморфного стану в кристалічний супроводжується різким зниженням електричного опору та коефіцієнта оптичного пропускання. Процеси кристалізації та параметри фазового переходу залежать від хімічного складу плівок та умов нагрівання. Результати свідчать, що плівки $Sb_xSe(Te)_{100-x}$ можуть використовуватися як матеріали для виготовлення температурних датчиків і пристроїв PRAM

Ключові слова: халькогенідні матеріали, аморфні плівки, фазові переходи, кристалізація, комірки пам'яті, детектори температури

Исследование халькогенидных материалов для устройств памяти с произвольным доступом на основе фазовых переходов

В.К. Кириленко[1] , М.А. Дуркот[1] , М.М. Поп[1,2], Р.П. Писак[1] , В.М. Рубиш[1]

Ужгородская лаборатория материалов оптоэлектроники и фотоники Института проблем регистрации информации НАН Украины
88000, ул. Замковые сходы, 4, Ужгород, Украина

Ужгородский национальный университет
88000, ул. Волошина, 54, г. Ужгород, Украина

Аннотация

Цель. Тонкие пленки $Sb_xSe(Te)_{100-x}$ (при условии, что $x \geq 50$) пригодны для производства устройств PRAM, которые работают на основе такого фазового реверсивного перехода: «аморфная фаза ↔ кристаллическая фаза». В этой статье представлены результаты исследования температурных зависимостей между электрическим сопротивлением и коэффициентом оптического пропускания при изменении условий термоциклирования и нагрева.

Методы. Кристаллизацию тонких пленок $Sb_xSe(Te)_{100-x}$ (при условии, что $x \geq 50$) исследовали с помощью метода, который обеспечивает одновременное измерение электрического сопротивления и коэффициента оптического пропускания в одном и том же образце. Измерения зависимостей $R(T)$ и $\Theta(T)$ проводились на плоскостных структурах «слой хрома – $Sb_xSe(Te)_{100-x}$ пленка» в диапазоне температур от 293 К до 493 К в непрерывном режиме в пределах скорости нагрева (q) от 3 К / мин. до 6 К / мин .; в режиме термоциклирования, а также в режиме нагрева при соответствующих остановок температурной развертки при выбранных температурах стабилизации (T_{cm}) в течение определенной продолжительности стабилизации температуры (t_{cm}).

Результаты. Неизотермическая кристаллизация аморфных пленок $Sb_xSe(Te)_{100-x}$ ($x \geq 50$) сопровождается резким уменьшением значений сопротивления и коэффициента оптического пропускания. Параметры перехода (начальная температура $T_ф$ перехода из аморфного состояния в кристаллическое, диапазон температур перехода $\Delta T_ф$, изменение электрического сопротивления ΔR и коэффициента оптического пропускания $\Delta\Theta$) зависят также от химического состава пленок и условий нагрева. Что касается исследуемых материалов, значение ΔT находится в диапазоне от 4 °К до 18 °К, тогда как значение ΔR было изменено на 2 или 3 порядка. С увеличением скорости нагрева значение $T_ф$ и $\Delta T_ф$ сдвигаются в область более высоких температур. Исследования показали, что кристаллизация аморфных пленок $Sb_xSe(Te)_{100-x}$ (частичная или полная кристаллизация) при определенных условиях может происходить при температурах ниже $T_ф$. Процентная доля кристаллизованного объема пленок и значения R, Θ, ΔR и $\Delta\Theta$ зависят от температурных параметров и параметров, зависящих от времени (например, T_{cm}, t_{cm}) процесса нагрева.

Выводы. Установлено, что аморфные пленки $Sb_xSe(Te)_{100-x}$ ($x \geq 50$) кристаллизуются при определенных условиях нагрева. Фазовый переход из аморфного состояния в кристаллическое сопровождается резким снижением сопротивления и коэффициента оптического пропускания. Процессы кристаллизации и параметры фазового перехода зависят от химического состава пленок и условий нагрева. Результаты свидетельствуют, что пленки $Sb_xSe(Te)_{100-x}$ могут использоваться в качестве материалов для изготовления температурных датчиков и устройств PRAM

Ключевые слова: халькогенидные материалы, аморфные пленки, фазовые превращения, кристаллизация, ячейки памяти, датчики температуры