Conference materials
UDC 535.417, 544.015.4, 536.331, 538.958, 538.975
DOI: https://doi.org/10.18721/JPM.153.342

Investigation of crystallinity degree for $Ge_2Sb_2Te_5$ films by reflection and transmission photometry

N. M. Tolkach [1]✉, N. V. Vishnyakov [2], V. G. Litvinov [2], A. O. Yakubov [1], E. S. Trofimov [2], A. A. Sherchenkov [1]

[1] National Research University of Electronic Technology (MIET), Zelenograd, Moscow, Russia;

[2] Ryazan State Radio Engineering University named after V. F. Utkin, Ryazan, Russia

✉ n.m.tolkach@gmail.com

Abstract. To assess the crystallinity degree for $Ge_2Sb_2Te_5$ (GST) films the method of reflection and transmission photometry and the approximation of an effective medium of the Brueggemann type were used. It was found that the changes in the energy and duration of the laser pulse lead to the change in the ratio of crystalline and amorphous fractions and change in crystallinity degree. It was found that an increase of crystalline fraction leads to an increase of refractive index and extinction coefficient at telecommunication wavelength of 1550 nm, which also leads to changes of reflectivity and transmissivity of the GST film and can be used to perform modulation and switching of signals.

Keywords: crystallinity degree, photometry, $Ge_2Sb_2Te_5$, phase change material, phase transition, phase state, amorphous, crystalline

Funding: The reported study was funded by RFBR according to the research project No. 19-37-60023 “A study of optical switching processes in alternating structures based on crystal-amorphous materials of Ge-Sb-Te system for optical switches”.

Citation: Tolkach N. M., Vishnyakov N. V., Litvinov V. G., Yakubov A. O., Trofimov E.S., Sherchenkov A. A., Investigation of crystallinity degree for $Ge_2Sb_2Te_5$ films by reflection and transmission photometry. St. Petersburg State Polytechnical University Journal. Physics and Mathematics, 15 (3.3) (2022) 219–222. DOI: https://doi.org/10.18721/JPM.153.342

Материалы конференции
УДК 535.417, 544.015.4, 536.331, 538.958, 538.975
DOI: https://doi.org/10.18721/JPM.153.342

Исследование степени кристалличности пленок $Ge_2Sb_2Te_5$ методами фотометрии отражения и пропускания

Н. М. Толкач [1]✉, Н. В. Вишняков [2], В. Г. Литвинов [2], А. О. Якубов [1], Е. С. Трофимов [2], А. А. Шерченков [2]

[1] Национальный исследовательский университет «МИЭТ», Зеленоград, Москва, Россия;

[2] Рязанский государственный радиотехнический университет им. В. Ф. Уткина, Рязань, Россия

✉ n.m.tolkach@gmail.com

Аннотация. Для оценки степени кристалличности $Ge_2Sb_2Te_5$ (GST) в данной работе мы использовали метод фотометрии отражения и пропускания и приближение эффективной среды типа Брюйггемана. Установлено, что изменение энергии и длительности лазерного импульса приводит к изменению соотношения кристаллической и аморфной фракций, что наблюдается по изменению степени кристалличности. Также установлено, что увеличение доли кристаллитов приводит к увеличению показателя преломления и коэффициента экстинкции для телекоммуникационной длины волны 1550 нм, что также приводит к изменению отражательной и пропускательной способностей пленки GST и будет использовано для осуществления модуляции и переключения сигналов.

Ключевые слова: степень кристалличности, фотометрия, $Ge_2Sb_2Te_5$, материал с фазовым переходом, фазовый переход, фазовое состояние, аморфный, кристаллический

Финансирование: Работа выполнена при финансовой поддержке РФФИ в рамках научно-исследовательского проекта № 19-37-60023.

Ссылка при цитировании: Толкач Н. М., Вишняков Н. В., Литвинов В. Г., Якубов А. О., Трофимов Е. С., Шерченков А. А. Исследование степени кристалличности пленок $Ge_2Sb_2Te_5$ методами фотометрии отражения и пропускания // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2022. Т. 15. № 3.3. С. 219–222. DOI: https://doi.org/10.18721/JPM.153.342

Introduction

Semiconductor materials with first kind phase transition (Phase Change Material – PCM), in particular, chalcogenide glassy semiconductors, have found their application in non-volatile radiation-resistant memory elements for creating dynamic storage devices. Currently, PCM material $Ge_2Sb_2Te_5$ (GST) is actively used for this purpose. GST has a fast and low-energy phase transition, which can be excited by both an electric and a laser pulse [1–3]. In addition to the main phase states (amorphous and crystalline) of GST, there can be additional intermediate states due to the presence of nanosized fractions of the material in both these states. Controlling the ratio of the amorphous and crystalline fractional composition (crystallinity degree) of GST opens up wide possibilities in fine-tuning its optical parameters, but at the same time, it requires studying the processes and peculiarities that occur when laser radiation is applied to the GST material. Therefore, the aim of the work is to investigate in the GST thin films the amorphous and crystalline fraction ratio, which is characterized by the crystallinity degree.

Materials and Methods

To estimate the crystallinity degree of GST films in this work, we used the method of reflection and transmission photometry and the approximation of an effective medium of the Brueggemann type. Such approximation represents a medium in the form of amorphous matrix with growing quantity of spherical crystallites. Fraction of crystals in the matrix (crystallinity degree) is equal to [4]:

$$C = (\bar{n}^2 - \bar{n}_a^2)\,(2\bar{n}^2 + \bar{n}_c^2)\ /\ (3\bar{n}(\bar{n}_c^2 - \bar{n}_a^2)), \tag{1}$$

where $\bar{n}$, $\bar{n}_a$ and $\bar{n}_c$ are the complex refractive index of GST film in the current, amorphous and crystalline state respectively. The complex refractive index $\bar{n}$ is found from the model expressions determined by the Airy formulas for reflectivity $\Re$ and transmissivity $\Im$ of the GST structure taking into account processes of multiple re-reflection of electromagnetic waves in the film and their interference.

System of nonlinear equations is solved to search the complex refractive index:

$$\begin{cases} f_1 = \Re_e - \Re(n,k) = 0 \\ f_2 = \Im_e - \Im(n,k) = 0 \end{cases}, \tag{2}$$

where n and k are a refractive index and an extinction ratio of the researched medium; $\Re_e$ and $\Im_e$ are experimental data on reflectivity and transmissivity; $\Re(n, k)$ and $\Im(n, k)$ – model expressions for reflectivity and transmissivity of the thin film structure taking into account processes of multiple re-reflection of electromagnetic waves in the film and their interference, are determined according to the Airy and Fresnel formulas [5].

Solution of simultaneous equations (2) is performed numerically according to the nonlinear system optimization method. This solution is the global minimum of the objective function of $F = (f_1 / \Delta f_1)^2 + (f_2 / \Delta f_2)^2$. Here Δf_1 and Δf_2 are scale coefficients of f_1 and f_2 function calculated as the average of experimental and model data ($\Delta f_1 = (\Re_e + \Re(n, k)) / 2$, $\Delta f_2 = (\Im_e + \Im(n, k)) / 2$).

More generally, the objective function is the following:

$$F(\Re_e, \Im_e, n, k) = ((\Re_e - \Re(n,k)) \;/\; (\Re_e + \Re(n,k))^2 + ((\Im_e - \Im(n,k)) \;/\; (\Im_e + \Im(n,k))^2. \quad (3)$$

The GST thin film with the thickness of 24 nm were deposited by magnetron sputtering. The measurement setup based on the NTEGRA Spectra (NT-MDT SI, Russia) was used and allowed to carry out investigations using methods of Raman spectroscopy, atomic force microscopy, optical microscopy, photometry, laser modification.

Laser modification of the GST film (see square regions in Figure 1ab) has been performed by radiation with the 405 nm wavelength, with the focused spot diameter of about 1 μm and with various duration values (from 4 ns to 1 microseconds), repetition intervals T (from 20 ns to 1 microseconds) and radiant exposures (from 0.02 nJ/μm^2 to 20 nJ/μm^2) of the pulse.

Photometry was used to measure the reflectivity and transmissivity of the GST film at 405, 532, 638 and 1550 nm wavelengths. Reflection and transmission photometry maps (see Fig. 1) obtained by scanning the GST film with the focused laser with a wavelength of 532 nm and a power of 5.3 μW (energy exposure 1.44 nJ/μm^2).

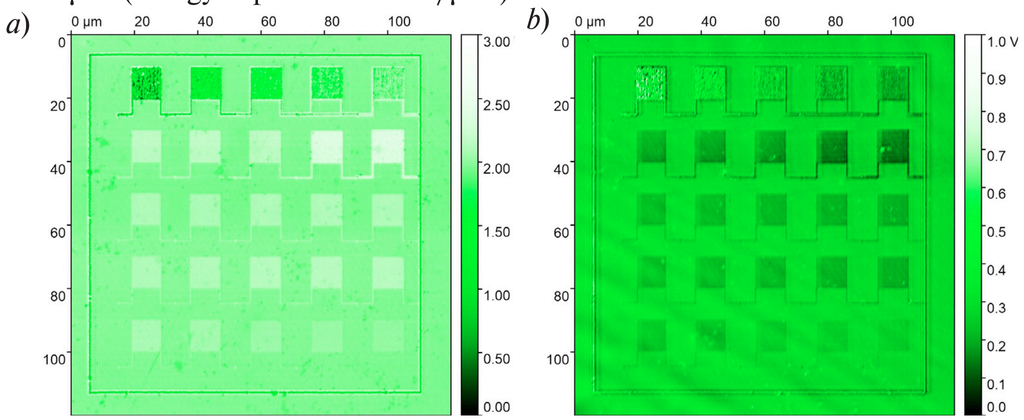

Fig. 1. Reflection (a) and transmission (b) photometry maps

Results and Discussion

Crystallinity degree of the local film area was determined using the obtained values of the complex refractive index at the wavelengths of 405, 638, and 1550 nm. As can be seen from Fig. 2, a, when films are exposed to laser pulses with wavelength of 405 nm at energy exposure of pulse from 2.92 to 3.26 nJ/μm^2, the crystallinity degree changes from 1 to 0.

The evaluation of crystallinity degree showed that amorphization of the film is observed when exposed to laser pulses with wavelength of 405 nm at energy exposure of pulse from 3.26 to 3.37 nJ/μm^2, with pulse durations up to 12 ns (see Fig. 2, b).

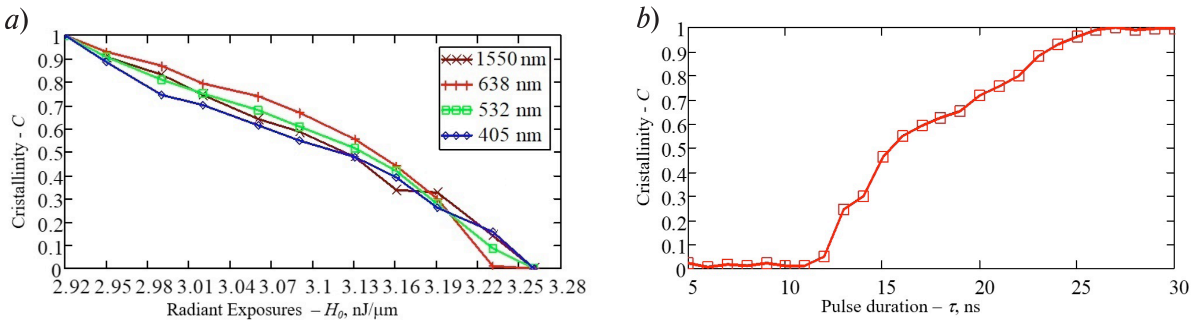

Fig. 2. Changes in the crystallinity degree of the local region in the GST film: depending on the energy exposure of the 405 nm laser pulse (a); depending on the laser pulse duration (b)

Partial crystallization of amorphous GST at these energy exposures is observed at the pulse durations from 12 to 25 ns, while full crystallization is observed at the pulse durations from 25 to 78 ns. An increase in the proportion of crystalline fractions leads to an increase of refractive index and extinction coefficient at telecommunication wavelength of 1550 nm, which also leads to changes of reflectivity and transmissivity of the GST film and can be used to perform multilevel recording, and amplitude modulation or routing of signals.

Conclusion

Thus, it was found that a change in the energy and duration of a laser pulse leads to a change in the crystallinity degree and, so, to a change in the ratio of crystalline and amorphous fractions.

Acknowledgments

The study was performed with the equipment of Region Centre of probe microscopy of Ryazan State Radio Engineering University named after V. F. Utkin.

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THE AUTHORS

TOLKACH Nikita M.
n.m.tolkach@gmail.com
ORCID: 0000-0001-9183-7348

VISHNYAKOV Nikolay V.
rcpm-rgrtu@yandex.ru
ORCID: 0000-0002-1270-3446

LITVINOV Vladimir G.
vglit@yandex.ru
ORCID: 0000-0001-6122-8525

YAKUBOV Alexey O.
alexsey007@mail.ru
ORCID: 0000-0003-0530-7186

TROFIMOV Egor S.
egor_trofimov.ru@mail.ru
ORCID: 0000-0002-1516-9154

SHERCHENKOV Alexey A.
aa_sherchenkov@rambler.ru
ORCID: 0000-0002-5596-4363

Received 15.08.2022. Approved after reviewing 24.08.2022. Accepted 15.09.2022.