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Effect of Ge concentration on optical properties of films synthesized by vacuum-thermal evaporation of glassy Ge–Se alloys

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ABSTRACT

Four non-crystalline Ge_xSe_{100-x} ($x = 15-25$ at. %) films were obtained by vacuum-thermal deposition of glassy Ge–Se alloys. With the use of spectroscopic ellipsometry, it was determined that the films are semiconductors with indirect band gap energy $E_g = 2.0-2.07$ eV. An anomalous dispersion of the refractive index n (λ) with n_{max} at λ_{max} depending on x was revealed in the films at λ = 240–500 nm. It was established that as the concentration of Ge decreases, n_{max} increases, and λ_{max} red shifts. Based on the Wemple-DiDomenico model, the nonlinear coefficients $\chi^{(3)}$ and n_2 characterizing the synthesized films were calculated as a function of x. The highest $\chi^{(3)}$ and n_2 values are achieved in the films at $x = 15$ at. %. The results obtained can be used for the development and creation of Ge–Se optical filters with an adjustable blue absorption band edge.

1. Introduction

Non-crystalline alloys based on Ge–Se belong to chalcogenide glasses, which contain at least one of the chalcogens from group 16 of the periodic table of elements (S, Se, Te) [1,2]. They have semiconducting properties and were first synthesized in the mid-50s of the 20th century [3]. Chalcogenide glasses containing As, Ge, P, Sb, Ga, In, Tl, lanthanides or some other electropositive elements exhibit unique specific physical and chemical properties [4] and are now the subject of numerous scientific studies (see, e.g., [5–8]). It is noteworthy that the electrophysical properties of chalcogenide glasses practically do not depend on impurities, and the mobility of charge carriers in them is extremely small [3]. At the same time, the parameters of two- or multicomponent easily synthesizable glassy alloys and film structures can be varied in a wide range by smoothly changing the concentration of the constituent elements [8–14]. The specificity of the properties of the chalcogenide materials has led to their widespread application in various optical and photonic technologies [2,4,7,15–17].

Chalcogenide glasses remain stable under high-energy ion irradiation [18] and are transparent in the infrared (IR) region of electromagnetic radiation, have a wide bandgap and a high refractive index, both depending on the composition [19,20]. These materials are chemically and thermally stable [21–23]. They have high non-linear refractive indices and non-linear absorptions [24–28], and exhibit amorphization–crystallization reversibility [29,30].

Recently, much attention has been paid to the study of the optical properties and photoinduced phenomena in nontoxic Ge–Se chalcogenides and films of various compositions based on them due to their applications as solar photocells [31–33], universal photodetectors [34], optical and holographic information recording devices [5], waveguides [7,35], passive and active elements for integrated optics, optical switching, imaging devices, lenses in objectives for microscopes [23,36], radiation dosimeters [37], optical temperature sensing devices [38,39] and printable materials for optical and electronic applications [40]. The most studied of such films are $GeSe_2$ [41–43].

It has been found in [44,45] that the interaction with high-energy photons in glassy Ge–Se samples can lead to the rearrangement of atoms in structural units, as well as the dangling of weak homopolar bonds and the formation of new bonds (for example, heteropolar ones). It has been established that the transition from one metastable state of the glass structure to another can be achieved by thermal annealing or by exposing the glass to light quanta of a certain energy [46–48] and causing changes in its physical and chemical properties (density, refractive index, optical absorption coefficient, bandgap and other properties) [49,50]. The revealed features of Ge–Se-based materials make them attractive for use in the development of various optical coatings in corrective optics. For this purpose, it is necessary to have knowledge of the band gap and refractive index values of such materials

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