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Article

Preparation, Chemical Composition, and Optical Properties of (β–Ga_2O_3 Composite Thin Films)/(GaS_xSe_{1-x} Lamellar Solid Solutions) Nanostructures

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Abstract: GaS_xSe_{1-x} solid solutions are layered semiconductors with a band gap between 2.0 and 2.6 eV. Their single crystals are formed by planar packings of S/Se-Ga-Ga-S/Se type, with weak polarization bonds between them, which allows obtaining, by splitting, plan-parallel lamellae with atomically smooth surfaces. By heat treatment in a normal or water vapor-enriched atmosphere, their plates are covered with a layer consisting of β–Ga_2O_3 nanowires/nanoribbons. In this work, the elemental and chemical composition, surface morphology, as well as optical, photoluminescent, and photoelectric properties of β–Ga_2O_3 layer formed on GaS_xSe_{1-x} ($0 \leq x \leq 1$) solid solutions (as substrate) are studied. The correlation is made between the composition (x) of the primary material, technological preparation conditions of the oxide-semiconducting layer, and the optical, photoelectric, and photoluminescent properties of β–Ga_2O_3 (nanosized layers)/GaS_xSe_{1-x} structures. From the analysis of the fundamental absorption edge, photoluminescence, and photoconductivity, the character of the optical transitions and the optical band gap in the range of 4.5–4.8 eV were determined, as well as the mechanisms behind blue-green photoluminescence and photoconductivity in the fundamental absorption band region. The photoluminescence bands in the blue-green region are characteristic of β–Ga_2O_3 nanowires/nanolamellae structures. The photoconductivity of β–Ga_2O_3 structures on GaS_xSe_{1-x} solid solution substrate is determined by their strong fundamental absorption. As synthesized structures hold promise for potential applications in UV receivers, UV-C sources, gas sensors, as well as photocatalytic decomposition of water and organic pollutants.

Keywords: chalcogenides; solid solutions; Gallium(III) trioxide; thin films; single crystals; optical properties; photoluminescence; photosensitivity

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1. Introduction

Gallium oxide (Ga_2O_3) is an ultra-wide band gap emerging semiconductor material, showing a well-marked polymorphism [1,2]. Currently, there are six confirmed Ga_2O_3 polymorphs with different crystal structures and crystallization temperatures: α–Ga_2O_3 with rhomboidal lattice, β–Ga_2O_3—monoclinic, γ–Ga_2O_3—cubic defective spinel-type structure, δ–Ga_2O_3—cubic, ε–Ga_2O_3—orthorhombic, and k–Ga_2O_3 polytype, also with orthorhombic lattice [3–6]. In particular, *k*–polytype was identified in β–Ga_2O_3 layers subjected to energetic ion bombardment [6–8].

At temperatures greater than 870 °C, the α, γ, δ, and ε phases change to monoclinic β–Ga_2O_3, with stable structure and physical properties through the whole temperature range up to the melting point [7–9].

The β–Ga_2O_3 is an *n*-type semiconductor with an ultra-wide energy band gap (4.9 eV), displaying considerable application prospects in ultraviolet (UV) optoelectronics [10–13] and high-performance electronic devices [14–16]. Recent studies have demonstrated that micro- and nanostructured β–Ga_2O_3 and related nanocomposites are promising materials for gas sensing applications and photocatalytic degradation of hazardous organic pollutants. In [17], the β–Ga_2O_3/Al_2O_3 nanocomposite was synthesized by a hydrothermal method with further calcination of $Al(NO_3)_3{\cdot}9H_2O$ and $Ga(NO_3)_3{\cdot}xH_2O$ compositions. The tested response of this composite material on exposure to NO_x (about 100 ppm concentration) was ~58%. Nanocomposites with nanostructured oxide semiconductors β–Ga_2O_3, SnO, or β–Ga_2O_3/reduced graphene oxide (rGO) exhibit high sensitivity to molecular gases (O_2, H_2), along with flammable and toxic chemical compounds, such as H_2S, CO_2, NH_3, etc. [18,19].

The critical breakdown electric field of semiconductor material is an important physical parameter, determining the technical and application characteristics of electronic devices (diodes, transistors, switches, etc.). For gallium oxide, a critical field with the value f 8 MV/cm was reported in [20]. By doping β–Ga_2O_3 with Zn, it can be increased up to 13.2 MV/cm. High values of the breakdown field determine the technical power parameters of field effect transistors [21,22].

The application area of wide-gap semiconductors is enlarged with the transition from bulk single crystals to micro- and nanocrystals. Several manufacturing technologies of β–Ga_2O_3 nanoformations (nanowires, nanoribbons, nanoparticles) are known. An extensive review of these methods is presented in the recent paper [23]. Nanostructured β–Ga_2O_3 was obtained in [24,25] by heat treatment of GaN powder and plates in nitrogen (N) flow at temperatures in the range of 850–1100 °C. In [24], obtaining, through the same technological procedures, micrometer-sized GaN grains coated with β–Ga_2O_3 micro- and nanoformations was reported.

Group-III monochalcogenides, MX (M = Ga, In, X = S, Se, Te), belonging to the class of lamellar III-VI semiconductors, are quasi-two-dimensional (2D) materials that exhibit unusual physical properties (high mobility of electric charge carriers, wide photoresponse bands, marked anisotropy of electrical, and optical properties, etc.). The GaSe and GaS, together with their solid solutions (GaS_xSe_{1-x}), are typical and outstanding representatives of this class of materials.

The GaSe plates, kept for a long time in a normal atmosphere, are covered with a layer composed of gallium oxides [26]. The oxidation process of GaS and GaSe lamellae at high temperatures was studied in several works [27–30]. By conducting a heat treatment of GaS plates in argon flow at temperatures in the range of 700–900 °C, microsheets of β–Ga_2O_3 are formed on their surface [29]. Additionally, β–Ga_2O_3 nanowires and nanoribbons were obtained by high-temperature (≥900 °C) heat treatment of GaSe plates in argon/airflow [30,31].

Under certain technological conditions, β–Ga_2O_3/Ga_2Se_3 and β–Ga_2O_3/Ga_2S_3 nanocomposites can be obtained, which are prospective materials for expanding the application area of Beta–Gallium Oxide.

Depending on the arrangement of elementary Se(S)-Ga-Ga-Se(S) planar packings with respect to each other, four polytypes (α, β, γ, and ε) of GaSe single crystals were distinguished. The GaSe and GaS single crystals obtained by the Bridgman technique correspond to the ε and β phases, respectively. The layered compounds GaS and GaSe are known to form a continuous series of GaS_xSe_{1-x} ($0 \leq x \leq 1$) solid solutions. In Refs. [32,33], appealing to X-ray diffraction (XRD) analysis and Raman spectroscopy, it was demonstrated that ε and β phases predominate in $0 \leq x \leq 0.01$ and $0.5 \leq x \leq 1$ composition, respectively, while the γ phase is characteristic for single crystals with the composition $0.05 \leq x \leq 0.40$.

In this work, the chemical and elemental composition, surface morphology, light absorption in the region of the fundamental absorption edge, photoluminescence (PL), and photoconductivity of the layer formed by the heat treatment of single crystalline GaS_xSe_{1-x} solid solutions in a water vapor-rich atmosphere (AVH_2O) at a temperature of 900 °C are studied.

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