ISSN: 2349-7793 Impact Factor: 6.876., Volume: 18 Issue: 01 in January 2024

PREPARATION OF THIN SEMICONDUCTOR FILMS AND THEIR ELECTRICAL PROPERTIES

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Abstract. Using the SILD (layer-by-layer ion deposition) method, thin semiconductor films of tin oxosulfide with a thickness from 100 nm to 1 μm were obtained by varying the number of layering cycles from 10 to 100. The extreme nature of the influence of the number of layering cycles on the thickness, chemical composition and structural features of the resulting films was recorded. It has been shown that films with a thickness of about 1 micron consist of particles ranging in size from 70 nm to 300 nm.

Keywords: semiconductor films, SILD, tin oxosulfide, structure, elemental composition, deposition rate, electrical resistance.

INTRODUCTION

The creation of modern micro and nanoelectronic devices often involves fulfilling a number of specific requirements for functional film elements and structures, including the complex chemical composition of the films being formed, the specified morphology of particles or the spatial structure of film objects. Ordered micro- and nano-sized films and heterostructures are used to create high-electron mobility transistors, lasers, photovoltaic cells, LEDs and other electronic devices. The improvement of these devices is largely related to the search for promising materials and technologies for their production in a given structural and chemical state.

MATERIALS AND METHODS

Today, along with fairly well-studied tin oxides, tin(II) sulfides can be classified as promising low-cost materials for micro- and nanoelectronics [1–4]. SnS monosulfide is characterized by a high optical absorption coefficient in the visible and near infrared regions of the spectrum at the level of 104–105 cm–1 and an increased band gap. Depending on the structural and chemical state of the material, it is 1.1–1.8 eV [5], which is close to the value of Eg for Si and GaAs. This opens up the possibility of using

Tin(II) sulfide as a functional material for film and heterostructure optoelectronic converters, including thin-film solar cells [6–7], instead of the more toxic traditional cadmium sulfide and telluride. Interesting proposals for economical solar cells are also heterojunctions based on oxide and sulfide films of the same metal, for example copper or tin [2], or different metals, for example tin sulfide and zinc oxide [3]. In addition, mixed oxide-sulfide films of the composition $M_{ex}S_yO_z$ are being studied quite actively [4]. The ability to vary the ratio of oxide and sulfide components in these materials makes it possible to control important semiconductor parameters such as electrical resistance, band gap, and type of conductivity, since they differ significantly. For example, the narrower gap S_nS is characterized by p type, and the wider gap S_nO_2 is characterized by n type conductivity [5].

RESULTS AND DISCUSSION

To study the processes of layering of thin films of tin(II) sulfide or oxosulfide, a series of experiments were carried out, during which the number of SILD processing cycles was increased from 10 to 100. Analysis of SEM images of the surface and chips of films obtained after heat treatment showed that in all experiments, a continuous coating is formed on the surface of the

ISSN 2349-7793 (online), Published by INTERNATIONAL JOURNAL OF RESEARCH IN COMMERCE, IT, ENGINEERING AND SOCIAL SCIENCES.,
under Volume: 18 Issue: 01 in January-2024
https://www.gejournal.net/index.php/IJRCIESS

substrate. Images of fractures of the resulting structures (Fig. 1) indicate noticeable morphological and topological differences between the formed layers. In particular, low porosity layers with the size of spherical structural elements on the order of 20–30 nm were observed in thin films formed with a relatively small number of processing cycles (10–30, Fig. 1a) or, conversely, with a large number of them (80 –100, Fig. 1, d). The formation of faceted crystallites with sizes greater than 70 nm was detected in the near-surface layer of films with a thickness of at least 100 nm (Fig. 1, b, c), obtained as a result of 40–70 processing cycles. Such films have increased porosity and roughness. Films of the greatest thickness are formed by repeating processing cycles 70 times (Fig. 1, c). In this case, the layer thickness is about 1 μm. The average size of the spherical particles forming the structural matrix, as in the case of thin films, does not exceed 20–30 nm, and the maximum size of faceted crystallites reaches 300 nm, which exceeds the thickness of the low-porosity matrix part of the film.

An analysis of the relationship between the geometric thickness of the film and the number of SILD deposition cycles was carried out using SEM images of cleavages of the resulting structures (see Fig. 1). When processing the experimental data, we took into account the presence of an electrically conductive ITO layer about 40 nm thick on the glass plate (see Fig. 1a). At the same time, gravimetric and spectrophotometric measurements were carried out, which made it possible to judge the amount of deposited material.

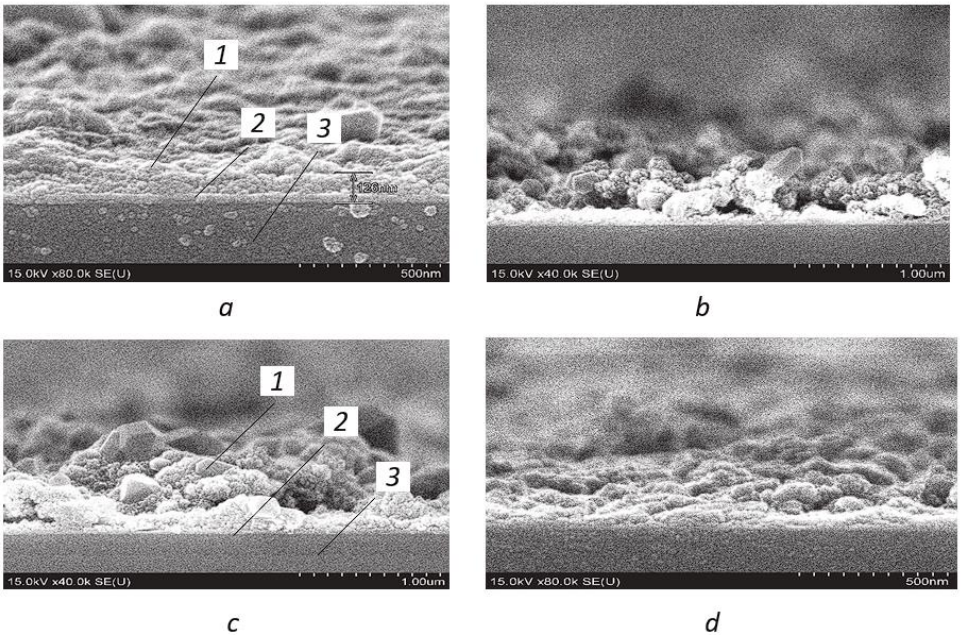

Fig. 1. SEM images of $S_nO_xS_y$/ITO/glass structures with the number of processing cycles: a – 30; b – 50; c – 70; d – 90; 1 – $S_nO_xS_y$ film; 2 – ITO film; 3 – glass

Various methods have been used to establish the extreme nature of the dependence of film thickness, as well as their optical transmittance and mass, on the number of deposition cycles (Fig. 2). The extremum point of all these dependencies corresponds to the case of 70 processing cycles, in which the geometric thickness (0.9 μm) according to electron microscopy, specific weight gain according to gravimetry (0.52 mg/cm2) and optical transmittance at a wavelength of 600 nm (33 %) according to spectrophotometry indicate the formation of the thickest, most massive and low-transparent film. The most dramatic changes in the characteristics of the resulting films occur at 50–80 layering cycles. It seems that a slight increase in the thickness and mass of the film with an increase in the number of layering cycles from 10 to 40 (Fig. 2, curves 2, 3) may be associated with kinetic

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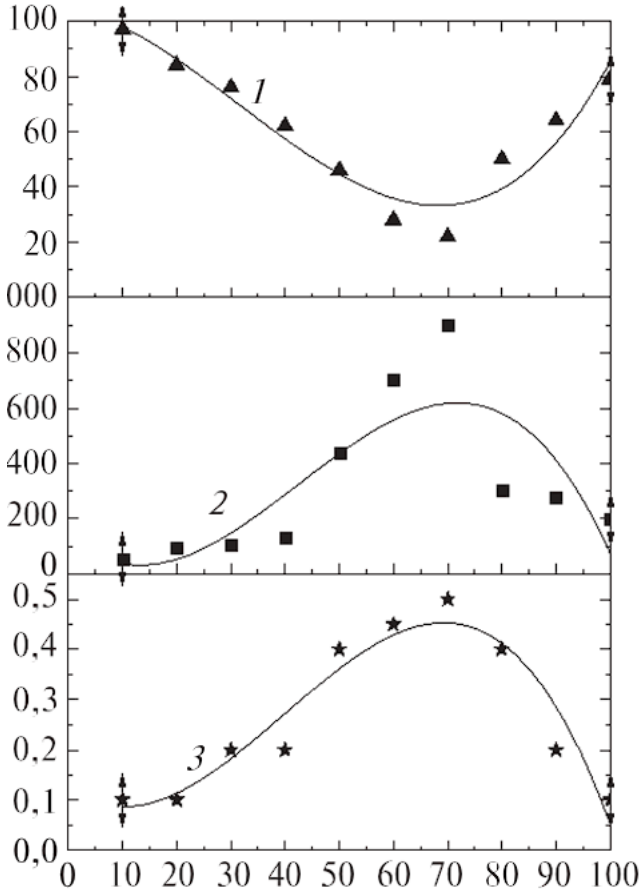
inhibition of the stage of formation of a dense matrix sublayer, when filling and compaction of transition layers occurs on the surface film-substrate interface. A significant increase in the thickness and mass of the film during 50–70 fold processing of the substrate, on the one hand, may be due to the facilitation of the processes of film deposition on a chemically related surface. On the other hand, in these cases it is possible to implement a combined ion-colloid mechanism of layering, which is favored by a rough surface, as well as an increase in the operating time of precursor solutions prone to hydrolysis.



Rice. 2. Dependence of the characteristics of $S_nO_xS_y$ films on the number of processing cycles:

1 – optical transmittance, %;

2 – thickness, nm; 3 – mass, mg/cm2

From a practical point of view, an important indicator of the formation of functional layers is the deposition rate. In this work, the relative layering speed (nm/cycle), normalized to the number of processing cycles, was assessed, as well as the absolute linear speed (nm/min), which was calculated taking into account the full duration of the layering process, including washing operations. The obtained values of the relative deposition rate of 2–13 nm/cycle (Table 1) in the case of films of increased thickness significantly exceed the theoretical values of the order of 0.3–1 nm/cycle, corresponding to the mechanism of monolayer ion deposition [2]. This is most likely due to the implementation of a mixed mechanism of ion-colloid deposition of particles from precursor solutions. This result in practice makes it possible to vary the conditions for the formation of functional layers to achieve a given technological task, including either slow ion layering of nanosized low porosity

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coatings or fast ion colloidal layering of submicron layers of increased porosity and roughness. The absolute values of the deposition rate at the level of 5 nm/min are comparable to the rate of vacuum deposition processes.

Table 1

Physicochemical parameters of SnOxSy films

Number of processing cycles	Average thickness, nm	Deposition rate		Settlement composition	Surface electrical resistance, Ohm
		relative, nm/cycle	absolute nm/min		
10	35	3,5	1,7	$SnO_{1,02}S_{0,41}$	220
20	40	2,0	1,0	$SnO_{0,59}S_{0,20}$	230
30	85	2,8	1,4	$SnO_{0,62}S_{0,10}$	244
40	130	3,2	1,6	$SnO_{0,60}S_{0,12}$	260
50	410	8,2	4,1	$SnO_{0,88}S_{0,08}$	360
60	700	11,7	5,8	$SnO_{0,81}S_{0,08}$	355
70	900	12,9	6,4	$SnO_{0,85}S_{0,08}$	362
80	310	3,9	1,9	$SnO_{0,87}S_{0,06}$	320
90	290	3,2	1,6	$SnO_{1,47}S_{0,16}$	300
100	180	1,8	0,9	$SnO_{1,84}S_{0,18}$	288

CONCLUSION

The features of the formation of nano- and micro-sized SnOxSy films by the SILD method have been studied. An extreme dependence of the thickness of the functional coating on the number of layering cycles was recorded when the number of processing cycles varied in the range of 10–100. Films of reduced thickness (up to 200 nm) have a denser and more uniform structure with a grain size of 20–30 nm, in contrast to porous layers up to 1 μm thick, on the surface of which crystallites with a size of 70–300 nm are formed.

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