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Editorial

Metal oxides for electronics and the SPQEO journal

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Abstract. This article discusses the main trends in the physics and preparation of metal oxides and summarizes the results of research published by SPQEO in this area over the past decade. The main metal oxides studied include ZnO, $Zn_{1-x}Cd_xO$, $Zn_{1-x}Co_xO$, $Mg_xZn_{1-x}O$, ZnO:Mn, VO_2, $ZrO_2–Y_2O_3$, TiO_2, WO_3, Gd_2O_3, Er_2O_3, $WO_3–CaO–SiO_2–B_2O_3$: Tb^{3+}, Dy_2O_3, NiO, Fe_xO_y, Ga_2O_3, Al_2O_3, ITO, Ag_2O and graphene oxide. These oxides were obtained by the following methods: sintering in air or in a stream of various gases, magnetron sputtering, atomic layer deposition, explosive evaporation, sol-gel, spin coating, spray pyrolysis, rapid thermal annealing, green synthesis from plant solutions, melt quenching, rapid thermal annealing, self-ignition, ion-plasma co-sputtering, vacuum sputtering, reactive ion beam sputtering, and the Hammer method. The electrical and optical properties of the studied oxides are illustrated.

Keywords: SPQEO journal, semiconductor physics, metal oxide, methods of obtaining metal oxides, electrical characterization, optical characterization.

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1. Metal oxides are a new type of semiconductor materials

Metal oxides (MO), as chemical materials with sufficiently wide bandgaps, were initially used as insulators. Subsequently, they were increasingly used as semiconductor materials, e.g. [1-8]. In fact, the first wave of publications on metal oxides appeared in the 1980s and 1990s. Thus, in [1, 2], the current understanding of the geometrical, chemisorption and electronic properties of metal oxide surfaces was rethought. The paper [1] has shown the relationship between the surface properties of single crystal oxides and the geometry, ligand coordination and defect structure of certain sites. The paper discusses the correlation between the geometry of the oxide surface and the structure of the crystalline body. The peculiarities of the electronic structure of ideal and defective surfaces were highlighted. In addition, modern findings on chemisorption of individual atoms and molecules on single-crystal oxide surfaces and electron- and photon-stimulated desorption of oxides were considered.

One of the key properties of MO surfaces is their ability to be modified by electron beam and scanning probe tips [4]. Such modifications may produce O vacancies or different regions of reconfiguration, which can in principle serve as anchors or templatesfor molecular or metallic interconnections. O. Whu [5] investigated the origin of unique physical and chemical properties of oxide/oxide interfaces. He demonstrated the physicochemical properties of a thin or ultrathin oxide films deposited on another oxide bulk or film at the atomic scale. Based on the analysis of the literature, it was shown that atomically pure interfaces between oxides have anomalous electronic and magnetic properties.

The rapid evolutions in synthesizing MOs and application of them in electronic devices was summarized in [6]. It examined and evaluated the latest trends and progress in synthesizing, designing and building MO-based electronic devices, including thin-film transistors, field-effect transistors, diodes, and photodetectors.

The properties of surface metal oxides surfaces at the atomic scale were examined in [7]. This contribution has discussed recent results of applying the surface science method, where systems are investigated under idealized conditions. Such experiments directly relate to first-principles calculations and provide insights into mechanisms and processes at a level that cannot be achieved in any other way. The review has shown recent developments with a main emphasis on metal oxides, a versatile and extremely useful class of materials.

Book [8] has collected the latest achievements in MO science for rechargeable batteries energy applications, molybdenum disulfide (MoS_2) and its nanocomposites as high-performance electrode material for supercapacitors; manganese dioxide (MnO_2): a high-performance energy material for electrochemical energy storage applications; conductive oxides role in flexible electronic device applications, indium-free alternative transparent conducting electrodes: an overview and recent developments; thin film metal oxides for displays and other optoelectronic applications; zinc oxide as a multifunctional material: from biomedical applications to energy conversion and electrochemical sensing; metal oxide- and sulfide-based gas sensors: recent trends and development; contribution of metal nanomaterials in algal biofuel production; fabrication of capacitive humidity sensors with nanostructured metal oxide thin films; multiferroic properties of rare-earth doped $BiFeO_3$ and its spintronic applications.

Zinc oxide is one of the MOs that has attracted much attention due to its possible applications in UV emitters, spin-functional devices, gas sensors, transparent electronics and surface acoustic wave devices [9]. It also indicates that the integration of ZnO with other wide bandgap ceramic semiconductors, such as AlInGaN systems, is of particular interest.

Zinc oxide can be produced by various methods: hydrothermal growth of ZnO [10], magnetron sputtering [11], sol-gel technique [12], atomic layer deposition [13], MOCVD [14], spin-coating [15] and others.

The application of metal oxides in electronics is extremely wide: from indium tin oxide electrodes to functional devices [16-19]. Since typical organic semiconductors have no charge carriers of their own, all charge in the device must be injected from the electrode/organic interface, whose energy structure determines the performance of the device. The energy barrier at the interface is closely related to the working function of the electrode. Therefore, various types of thin-film metal oxides can be used as buffer layers to modify the working function of the electrodes [16].

SPQEO focuses on major trends in MO physics and applications. This article reflects some of the articles that SPQEO has published in this area over the past decade.

2. Overview of Metal Oxide Materials in SPQEO

The most popular MO articles in SPQEO are those on ZnO [20-29], $Zn_{1-x}Cd_xO$ [30], $Zn_{1-x}Co_xO$ [31] and ceramics based on $Mg_xZn_{1-x}O$ [32-35] or ZnO:Mn [36, 37]. They are followed by MO based on refractory metals VO_2 [38-41], ZrO_2–Y_2O_3 [42], TiO_2, [43-48], WO_3 [49]. There are enough articles devoted to rare earth metal oxides [49-53]. And some articles deal with NiO [54], Fe_xO_y [55], Ga_2O_3 [56], Al_2O_3 [57-60], graphene oxide [61], indium-tin oxide [62] and silver oxide [63].

3. Methods of obtaining metal oxides described in SPQEO

This section outlines the following main methods for obtaining metal oxides:

- the sintering in air or a stream of various gases [20, 32, 35, 36],
- the magnetron sputtering [21, 28, 30, 31, 38, 54],
- the atom layer deposition (ALD) [22],
- the explosive evaporation [25]
- the sol-gel [27, 29, 43],
- the spin coating [27],
- the spray pyrolysis [33],
- the rapid thermal annealing [44-46, 50, 51, 53],
- the green synthesis from plant solutions [47, 63],
- the melt quenching [49],
- the rapid thermal annealing [50, 51],
- the autoignited combustion [52],
- the ion-plasma co-sputtering [55],
- the sputtering in vacuum [58],
- the reactive ion beam sputtering [59, 60, 62],
- Hummer's method [61].

Another process is the production of metal oxide nanoparticles (NPs) [24]. ZnO/porous silica nanocomposites were successfully fabricated by three different types of synthesis techniques. In all cases, the molecular sieve SBA-16 was used as a porous matrix. The in situ growth the nanoparticles of zinc oxide within the matrix pores was done using either gaseous or liquid precursors. The ex-situ method implied growing of nanoparticles in a colloidal solution with further penetration of the ripened ZnO nanoparticles into the pores of the matrix.

4. Electrical, optical and other characterization of metal oxides

It is necessary to highlight some results obtained for MOs. For nominally undoped ZnO ceramic the self-activated orange PL band with a peak at 610 nm was separated by Gaussian deconvolution. It was established that the native defects responsible for the self-activated orange band were zinc vacancies [20]. At the same time the doped with Li-, Ag-, Cu- and Zn samples demonstrated more intense PL than the undoped ones and that used doping enabled to prepare MgZnO phosphors with bright emission in the blue-yellow spectral range [32]. The incestigation of the structural characteristics of (Mg,Zn)O ceramic samples with different magnesium content and specular IR reflection spectra have shown the following [34]: (i) the electrical conductivity extracted from the IR reflection spectra corresponds to that of hexagonal phase in a solid solution, while plasmon in cubic phase was not observed; (ii) the tendency to decrease electron concentration in the hexagonal grains of solid solution with further growth of the MgO content, which was explained by extraction of zinc interstitials, responsible for ZnO conductivity, from ZnO under formation of the MgZnO cubic phase; (iii) the decrease in electron concentration for the samples with a high MgO content ($x = 40…70$ mol.%) is due to a decrease in the Zn_i content in the hexagonal phase of solid solution exactly because of zinc extraction to form the cubic phase.

At the same time the Mn doping of ZnO ceramics have shown two effects [36]: i) drastic quenching of self-activated PL accompanied by gradual red-shift of spectral boundary of the quenching with increasing the Mn content; ii) appearance of a new emission band peaking at 645 nm that becomes dominant in the PL spectrum at

NMn = 1020 cm^{-3}. The observed effects were believed to be due to re-absorption of self-activated ZnO emission by Mn-related centers. The following recombination in excited centers was supposed to occur by both radiative and nonradiative ways, the former being responsible for 645 nm PL band. In the samples doped with $MnSO_4$, formation of Mn-related phase, namely, $ZnMn_2O_4$ spinel occurred at ZnO grain boundaries under sintering [37]. It has been ascertained that a thin layer of this substance separates adjacent ZnO grains, which provides appearance of the back-to-back Schottky barriers at grain boundaries and “varistor behavior” of current-voltage characteristics.

In [38] it has been shown that the low-temperature (200 °C) of vanadium oxide films deposition results in the VO_x amorphous film formation with a high content of V_2O_3 phase. The subsequent low-temperature annealing leads to the shift of the film thermodynamic equilibrium state in direction to the formation of VO_2 and V_2O_5 crystallization centers. An amorphous state of film with a smaller size of V_2O_3 crystallites is more thermally stable. Article [39] discusses the nanomechanical properties of polycrystalline vanadium oxide thin films with different phase compositions. Also, article [40] outlines some possible implementations to change the temperature of the metal-insulator phase transition in VO_2.

Thin nanocrystalline films of TiO_2 demonstrated E_{gi} = 3.3 eV, which coincides with the bandgap width for the anatase phase of bulk TiO_2 [43]. Addition of zirconium oxide to TiO_2 leads to a decrease in this value down to 2.9 eV, which is related with the presence of surface levels at the bottom of the conduction band. For the triple films modified with gold and silver nanoparticles, the corresponding bandgap widths are E_{gi} = 3.6 eV and E gd = 4.2 eV, respectively. Upon the restructuring of the energy spectra,especially within the bandgap, we can expect a substantial change in photogenerative and recombinational properties of TiO_2 thin films, which leads to the improvement of photocatalytic activity in the UV and visible ranges.

The comparison of TiO_2, Gd_2O_3, Er_2O_3 thin films on SiC substrates has shown [46], that Gd_2O_3/SiC exhibited the fewest defects, while Er_2O_3 and TiO_2 showed more, with Er_2O_3 being the most mismatched and roughest. The results indicate that internal strains in oxide thin films on SiC substrates can influence on surface morphology, leading to formation of defects and spatial inhomogeneity.

The electrical conductivity of the sample was shown [52] to be linear behavior over the entire temperature range. The decrease in grain and grain boundary resistances with temperature suggests a thermally activated conduction mechanism. Thermal activation has been found to be of hopping type, and the mobility of charge carriers increases with increasing temperature. The activation energies of grain and grain boundary as well as the electrical conductivity values prove application of these materials for fabrication of electrolytes in solid oxide fuel cells. In [58], the Al-Al_2O_3-p-CdTe-Mo structure was considered as an n+-p diode structure with a long base, where the current transport process is described by the ohmic relaxation drift model, which occurs under non-equilibrium carrier recombination conditions through a pair of two-level recombination complexes.

The enhancement of the substrate temperature up to 280 °C has shown in [59] to be lead to formation of $AlCuO_2$ phase, when the Al/Cu stoichiometric ratio approaches to unity.

Passivation of front illuminated surface ITO layer with formation of ITO/silicon heterojunction, unlike silicon dioxide layer passivation, leads to a significant reduction of the effective surface recombination velocity. It significantly increases the value of the internal quantum efficiency in the wavelength range from 550 to 1050 nm and, as a result, significantly increases the value of short-circuit current of solar cells [62].

Conclusion

The SPQEO Journal focuses on main trends in metal oxide physics and technology. Over the past decade, 44 articles have been published on metal oxides, including ZnO, $Zn_{1-x}Cd_xO$, $Zn_{1-x}Co_xO$, $Mg_xZn_{1-x}O$, ZnO:Mn, VO_2, ZrO_2–Y_2O_3, TiO_2, WO_3, Gd_2O_3, Er_2O_3, WO_3–CaO–SiO_2–B_2O_3: Tb^{3+}, Dy_2O_3, NiO, Fe_xO_y, Ga_2O_3, Al_2O_3, ITO, Ag_2O and graphene oxide. The preparation of metal oxides and some of their electrical and optical properties are discussed.

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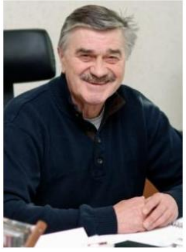

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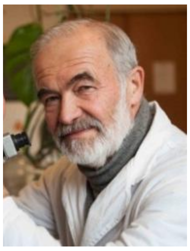

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Оксиди металів для електроніки та журнал SPQEO

О.Є. Бєляєв, З.В. Максименко та П.С. Смертенко

Анотація. У цій статті обговорюються основні тенденції у фізиці та одержанні оксидів металів, а також узагальнюються роботи, опубліковані в SPQEO в цій галузі за останнє десятиліття. Основні оксиди металів, що досліджувалися, такі: ZnO, $Zn_{1-x}Cd_xO$, $Zn_{1-x}Co_xO$, $Mg_xZn_{1-x}O$, ZnO:Mn, VO_2, $ZrO_2–Y_2O_3$, TiO_2, WO_3, Gd_2O_3, Er_2O_3, $WO_3–CaO–SiO_2–B_2O_3$: Tb^{3+}, Dy_2O_3, NiO, Fe_xO_y, Ga_2O_3, Al_2O_3, ITO, Ag_2O та оксид графену. Ці оксиди були отримані наступними методами: спікання на повітрі або в потоці різних газів, магнетронне розпилення, осадження атомних шарів, вибухове випаровування, золь-гель, спінкоутер нанесення, спрей-піроліз, швидкий термічний відпал, зелений синтез з рослинних розчинів, гартування розплаву, швидкий термічний відпал, самозапалювання, іонно-плазмове співрозпилення, розпилення у вакуумі, реактивне розпилення іонним пучком, метод Хаммера. Проілюстровано електричні та оптичні властивості досліджуваних оксидів.

Ключові слова: журнал SPQEO, фізика напівпровідників, оксид металів, методи отримання оксидів металів, електрична характеристика, оптична характеристика.