Influence of ionizing radiation on crystal structure and electrophysical properties of $Pb_{1-X}Mn_X$ Te(Se) thin films

M.A.Mehrabova, I.R.Nuriyev

Abstract— In the given paper the obtaining of $Pb_{1-x}Mn_xSe$ (x=0.01) and $Pb_{1-x}Mn_xTe$ (x=0.04) epitaxial nanofilms and influense of γ-irradiation and accelerated electrons on their crystal structure and electrophysical properties have been investigated. $Pb_{1-x}Mn_xTe(Se)$ nanofilms have been grown on freshly broken and polished substrates of BaF_2 in a vacuum 10^{-4}Pa in УВН-71 П3 vacuum assembly by the molecular beams condensation method. In the present report are given investigation results of the structure, morphology of a surface and physical properties of $Pb_{1-x}Mn_xSe$ (x=0.02) nanofilms grown by the molecular beams condensation method. It has been defined, that after the γ-irradiation doses 25 kGy the parameters of nanofilms become the worst. It was established that the samples became more photosenstive after irradiation. By increasing of Mn concentration maximum of photoconductivity shifted towards short wave range. The energy gap of samples increase by increasing of Mn concentration.

Keywords— semimagnetic semiconductor, epitaxial nanofilms, crystal structure, morphology, substrates, γ-irradiation, photosenstive, photoconductivity. energy gap

I. Introduction

The aim of this work were obtaining and investigation of $Pb_{1-x}Mn_xTe(Se)$ SMS epitaxial nanofilms, to obtain their optimum and perfect samples with high sensitivity and radiation resistance properties for spintronic and IR electronics.

Spintronics has been attracting attention because of its potential applicability to new functional devices combining transport and magnetic properties. Spintronic devices came into action after the discovery of powerful effect called "giant magnetoresistance (GMR)" in 1988 by French and German physicists. It results from subtle electron-spin effects in ultra-thin 'multilayers' of magnetic materials, which cause huge changes in their electrical resistance when a magnetic field is applied.

Then it was obtained giant Faraday rotation in semimagnetic semiconductors (SMS). When certain materials are subjected to magnetic fields, they rotate the plane of polarization of light traveling through them. This effect, known as Faraday rotation, is important technologically because it can be used to construct optical isolators, unidirectional optical amplifiers, and other nonreciprocal devices. To be practical, however, most devices of this type require materials with very large Faraday rotations. In 1978, it was discovered that SMS display an extremely large Faraday rotation. In 90's authors theoretically investigated Faraday rotation in SMS thin films and defined, that the Faraday rotation increases by increasing of film thikness. At that time there was no large-scale technological need for such materials. Magnetic semiconductors, in which spin- and charge-dependent properties of electrons coexist, are now the most important topics of investigation in the field of new functional semiconductor devices.

In last 20 years SMS of lead chalcogenides ($Pb_{1-x}Mn_xS$, $Pb_{1-x}Mn_xSe$, $Pb_{1-x}Mn_xTe$) have been subject of intensive experimental and theoretical researches [1-6]. They attracted attention because of free carrier induced magnetism. Unlike II-VI SMS, these materials can be grown with higher concentration of free band carriers. Their magnetic properties can be controlled by modifying the carrier concentration through control of native defects.

In these SMS lead (Pb) atoms are partly replaced by uncompensated magnetic momentum manganese (Mn) transition element atoms. As a result of Mn ions' introduction in a lattice of lead chalcogenides compounds. In these SMS the lattice constant insignificiantly decreases and so the width of the band gap sharply increases. In this case the energy spectrum of charge carriers in magnetic field extraordinarily changes. In result, occur opportunities to make devices controled by magnetic field and temperature on the basis of these structures.

At present, significant development of semiconducting infrared (IR) devices' market is observed. This tendency, first of all, is associated with the application of new materials in IR technology devices. Achievements of semiconducting material science, observed in recent years, have played an important role in the accelerated development of modern technology and instrument engineering. The research and establishment of

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scientific basis of new perspective materials' technology are one of the major tasks of semiconducting material science [7].

In connection with wide application of the indicated semiconductors $A^{IV}B^{VI}$ in optoelectronic devices these materials are of large scientific interest and draw the investigators attention. They are used in manufacturing of different infrared (IR) engineering instruments [8]. There have been developed a number of methods for obtaining structural-perfect uniform epitaxial films of these materials with predetermined thickness, composition and concentration of charge carriers [9-12].

The crystalline structure and physical properties of these films are much determined by substrates parameters. It is desirable that the maximum passible coincidence of parameters of lattice, coefficients of substrate thermal expansion and film to be sputtered. The use as substrates of monocrystalline plates of the indicated compounds or solid solutions allows achieving the full coincidence of all parameters. On the other hand, the epitaxial films and structures obtained on insulating dielectric substrates are of great practical interest.

Development of new types of optoelectronic devices operating in IR region of the spectrum and resistant against effect of ionizing radiation of a various type is the important task facing by a modern science and engineering. So the characteristics of many materials nowadays using in engineering become considerably worse under the effect of radiation as a result of transformations occurring in them.

From this point of view $A^{IV}B^{VI}$ type compounds and solid solutions on their basis are of special interest. So, in these solid solutions as a result of introduction of Mn ions in a lattice of lead chalcogenides a lattice constant decreases though it is unsignificant. In results the band gap is increase and in structures made on the basis of these substances an opportunity to control their properties by magnetic field and temperature occurs and it is possible to use them for manufacturing radiation-proof optoelectronic devices.

There is a little number of works devoted to preparation, study and application of $Pb_{1-x}Mn_xTe(Se,S)$ epitaxial films, however we didn't find any publications devoted to the influence of irradiation on photoelectrical, electrophysical and optical properties of $Pb_{1-x}Mn_xTe(Se,S)$ thin films.

Although there is a lot of literary information on studying SMS nanolayers, the influence of ionizing radiation on physical parameters of these materials has not been studied to this day.

Development of new generation radiation resistant spintronic devices operating in IR region of spectrum is the important task of modern science and engineering, as the characteristics of materials, using in engineering nowadays are considerably worse under the ionizing radiation. As a result transformations occur in them [13].

In order to study the preparation possibilities of radiation-resistant devices on the base of $Pb_{1-x}Mn_xTe(Se)$ thin films and to improve their parameters, we have researched the obtaining technology of $Pb_{1-x}Mn_xTe(Se)$ thin films (thickness d=10-50nm), their crystal structure, surface morphology and ionizing radiation influence on these properties.

Thus, the aim of the research project:

- ❖ Obtaining of optimum and perfect samples of $Pb_{1-x}Mn_xTe$ (Se,S) nanofilms,
- ❖ Investigation of ionizing radiation influence on photoelectrical, electrophysical and optical properties of $Pb_{1-x}Mn_xTe$ (Se,S) nanofilms to study their use potentials in spintronics.

II. Obtaining Technology

$Pb_{1-x}Mn_xSe$ (x=0.01) and $Pb_{1-x}Mn_xTe$ (x=0.04) nanofilms have been grown on freshly broken and polished plates substrates of BaF_2 in a vacuum 10^{-4}Pa in УВН-71 П3 vacuum assembly by the molecular beams condensation (MBC) method [14-21] . As a substrate it has been used natural layers of BaF_2 monocrystals, cut of accordingly on its plane (111). The choice of BaF_2 as a substrate is due to that it has cubic structure of with the parameter of elementary unit of 6.19A°. It is dielectric, has good mechanical strength and chemically inert. The obtained epitaxial nanofilms was grown along the surface paralel to substrate.

More crystal perfect film ($W_{1/2}$=80÷100") of 10-50nm thickness was obtained at condensation rates $\upsilon_к$=8÷9Å/sec and substrate temperature T_s=612÷645K. It was established, that epitaxial growth occurs at film temperature T_f=462÷511K.

On the basis of developed regime there have been obtained high ohmic epitaxial films $Pb_{1-x}Mn_xTe(Se)$ of n- and p-type conductivity with concentration $n(77K)=5\cdot10^{15}\div1.6\cdot10^{16}cm^{-3}$, charge carriers mobility $\mu(77K)=2.7\div3\cdot10^4 cm^2/B\cdot sec$ and lattice constant a= 6.11÷6.05A° (Fig.1)

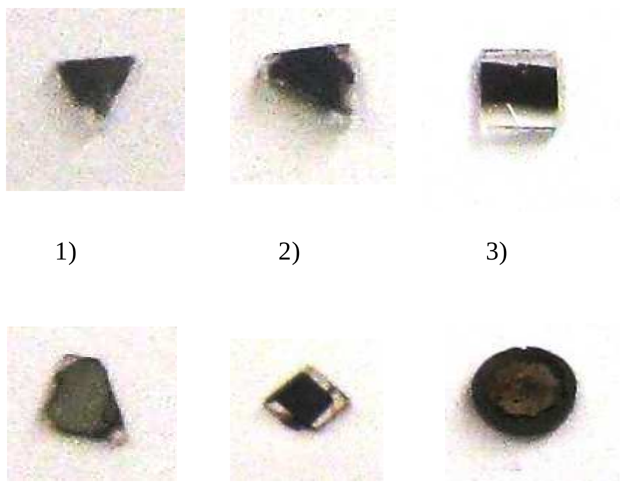
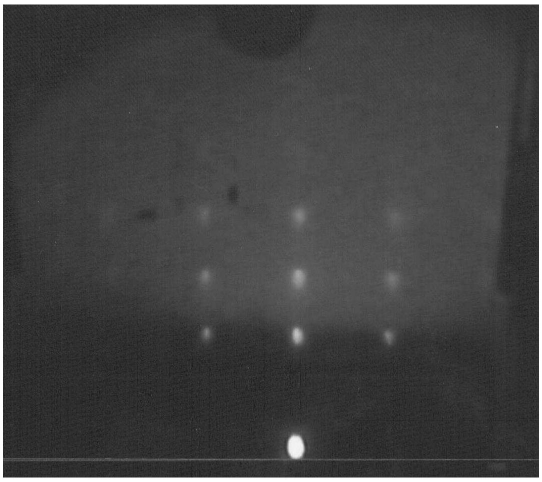


Fig.1 Samples of nanofilms: 1) n-type $Pb_{1-x}Mn_xSe$ (x=0.01, d=10nm), 2) p-type $Pb_{1-x}Mn_xSe$ (x=0,01, d=10nm), 3) $Pb_{1-x}Mn_xSe$ (x=0.04)4) $Pb_{1-x}Mn_xTe$ (x=0.04, d=50nm), 5) $Pb_{1-x}Mn_xTe$ (x=0.04, d=50nm), 6) $Pb_{1-x}Mn_xTe$ (x=0.05, d=10nm)

With the aim of preparing films of more perfect structure and with required values of electrophysical parameters the additional compensating source of Te (Se) vapours has been used during growth.

The $Pb_{1-x}Mn_xTe(Se)$ nanofilms with different types of conductivity have been obtained by changing of temperature and by using an additional compensating Se (selenium) vapor source during the growth.

We have been studied crystal structure and surface morphology of $Pb_{1-x}Mn_xTe(Se)$ nanofilms. Crystal perfection of the films has been studied by electron diffraction, X-ray diffraction methods (Fig.2), and its surface morphology by the method of atomic-force microscope (AFM) (Fig.3). Lattice constants of nanofilms have been calculated from X-ray diffraction curves. It has been determined three-dimensional topography of the sample, studied by AFM and its surface roughness (Fig.3).



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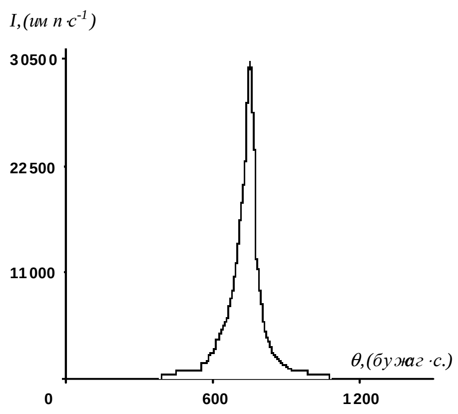

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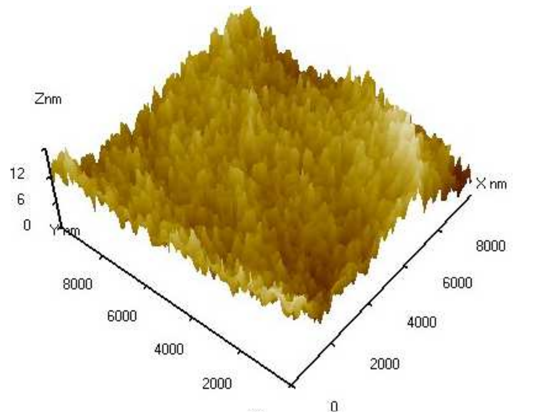
Fig.2 Electron diffraction (a) and X-ray diffraction (b) of $Pb_{1-x}Mn_xSe$ (x=0,01) epitaxial films

We have defined, that after the γ-irradiation doses 25 kGy the parameters of nanofilms become the worst.



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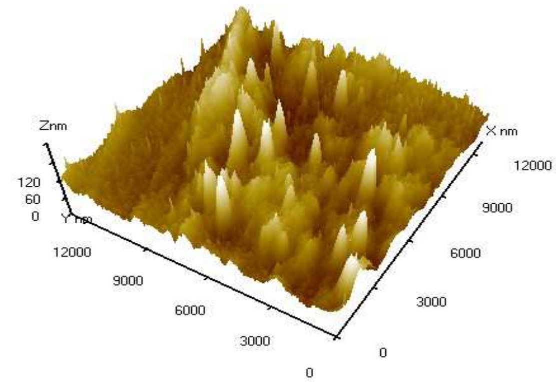

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Fig.3 The view of surface layer of $Pb_{1-x}Mn_xSe$ (x=0.01) epitaxial nanofilms in atomic-force microscope: 1. before γ-irradiation, 2. after γ-irradiation, doze energy was 25kGy

Fig.4 Samples are irradiated in ^{60}Co Isotopic Radiation Source

It has been investigated the influence of electron rays and γ –irradiation on $Pb_{1-x}Mn_xSe$ SMS epitaxial nanofilms. The samples were irradiated at room temperature in ^{60}Co Isotopic Radiation Source (in γ-irradiation doses till 25kGy) (Fig.4). The influence of ionizing radiation on crystal structure of $Pb_{1-x}Mn_xSe$ SMS nanofilms have been investigated.

The samples were irradiated in Electron Linear Amplifier ELA-6 (electron flux $\phi=5\cdot10^{17}cm^{-2}$ energy E=5MeV) (Fig.5). The influence of ionizing radiation on photoelectrical, electrophysical and optical properties of $Pb_{1-x}Mn_xTe(Se,S)$ SMS nanofilms have been investigated.

Fig.5 "Electron Accelerator" Electron Linear Amplifier ELA-6

III. Experiments

We have studied:

- The influence of electron rays on photoconductivity of $Pb_{1-x}Mn_xTe(Se)$ epitaxial films ,
- Mn concentration dependence on $Pb_{1-x}Mn_xTe$ epitaxial films,
- As a result of increase in Mn composition, the width of band gap increases.

The experimental data, obtained in the course of study of p-type crystals, don't allow predicting with enough reliability the nature of n-type alloys parameters change in irradiation depending on ratio of rates of generation of defects of donor and acceptor nature. The irradiation of n-type crystal can lead to both n-p-conversion of conductivity type ($dN_d/d\phi<dN_a/d\phi$) and to the increase of electrons concentration in conductivity zone up to the stabilization of Fermi level at energy level of donor type defect ($dN_d/d\phi>dN_a/d\phi$). Besides, the points on energy position of radiation level of donor type and nature of rebuilding of energy spectrum of irradiated alloys in variation of tin content in alloy remain uncertain.

Therefore one of tasks of the present work was the study of effect of electrons rays on electrophysical properties of unalloyed n-$Pb_{1-x}Mn_xTe$ (x=0.04) thin films for determination of charge carriers energy spectrum and parameters of these materials.

On the basis of developed regime there have been obtained high ohmic epitaxial films $Pb_{1-x}Mn_xTe$ of n- and p-type conductivity with concentration $n(77K)=4\cdot10^{15}\div1.5\cdot10^{16}cm^{-3}$ and charge carriers mobility $\mu(77K)=2.5\div3\cdot10^4 cm^2/B\cdot sec$.

The films with different types of conductivity have been obtained by changing temperature of basic $Pb_{1-x}Mn_xTe$ and compensating source of Te. It has been established that under the above mentioned conditions the epitaxial films are photosensitive at the temperature of liquid nitrogen (77K) (Fig.6). As is seen from Fig.7 the maximum of spectrum of films $Pb_{1-x}Mn_xTe$ (x=0.04) photoconductivity is shifted towards the shorter wakes in comparison with similar spectra for the other compositions of the given solid solution ($0\le x\le0.04$) carried out. This fact explained by increasing sample thickness by increasing of energy gap of the studied samples. From Fig.7 is seen that after irradiation, the samples become more photosensitive.

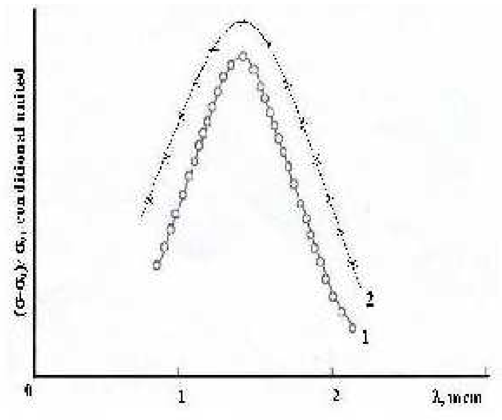

Fig.6 Spectrum of photoconductivity of $Pb_{1-x}Mn_xTe$ (x=0.04) films at temperature 77K. 1. before irradiation, 2. after irradiation.

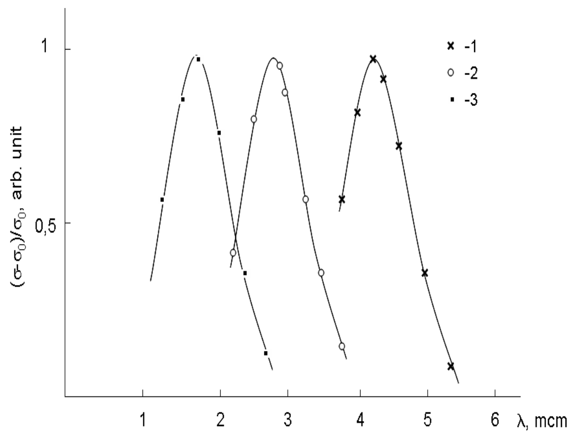

Fig.7 Spectrum of photo sensitivity of $Pb_{1-x}Mn_xTe$ thin films: 1. x=0.02, 2. x=0.04, 3. x=0.05

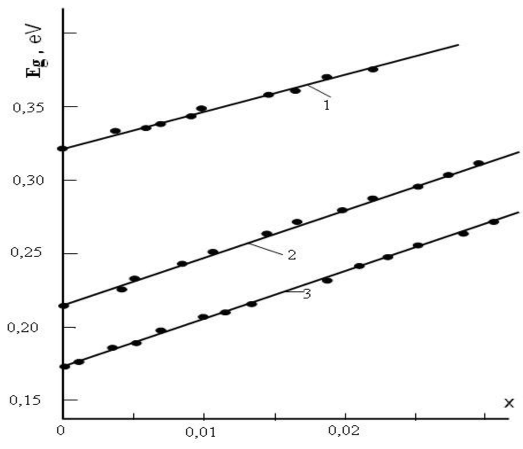

Fig.8 Dependence of band gap E_g on $Pb_{1-x}Mn_xS$(1), $Pb_{1-x}Mn_xTe$ (2), $Pb_{1-x}Mn_xSe$ (3) thin films' composition : T - 77 K.

The starting samples were irradiated at room temperature in electron linear accelerator of ELU-6 (E=5MeV, $\phi=5\cdot10^{17}cm^{-2}$). For each sample prior and after irradiation the temperature dependence of specific resistance has been studied (Fig 8).

The temperature dependence of specific resistance for all studied samples at first decreases slowly and then increases at temperature 77K. The more significant changes being characteristic for samples with the least starting concentration of electrons. The nature of dependences ρ (1/T) of the samples with high starting concentration of electrons doesn't change. Within temperature region close to room one, the activation section appears connected with own ionization of charge carriers.

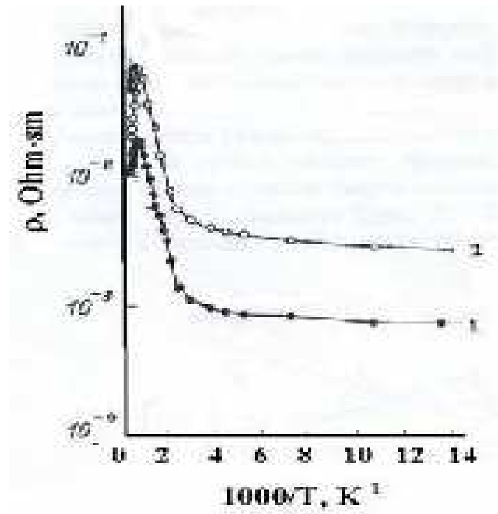

Fig.8 Temperature dependence of specific resistance of electrons irradiated ($\phi=5\cdot10^{17}$ cm^2) sample 1. Before irradiation, 2. after irradiation.

At the same time the experimental data, obtained [22,23] in the course of study of p-type crystals, don't allow to predict with enough reliability the nature of n-type alloys parameters change in irradiation depending on ratio of rates of generation of defects of donor and acceptor nature. The irradiation of n-type crystal can lead to both n-p-conversion of conductivity type ($dN_d/d\phi<dN_a/d\phi$) and to the increase of electrons concentration in conductivity zone up to the stabilization of Fermi level at energy level of donor type defect ($dN_d/d\phi>dN_a/d\phi$). Besides, the points on energy position of radiation level of donor type and nature of rebuilding of energy spectrum of irradiated alloys in variation of tin content in alloy remain uncertain.

IV. Results

- The energy gap of samples increase by increasing of Mn concentration
- By increasing of Mn concentration maximum of photoconductivity shifted towards the short wave range [24].
- After electron irradiation (electron flux $\phi=5\cdot10^{17}$ cm^{-2}) the samples become more photosensitive [25].
- At γ-irradiation doses 25 kGy the radiation stability of nanofilms become worse

V. Conclusion

We have defined the optimum values of physical parameters of $Pb_{1-x}Mn_xSe$ (x=0.01) and $Pb_{1-x}Mn_xTe$ (x=0.04) nanofilms and ionizing radiation dose, which give us possibility to make radiation resistant and high sensitive spintronic devices controlled by magnetic field and temperature. After the γ-irradiation doses 25 kGy the parameters of nanofilms become the worst.

In the presented work the influence of electron rays and γ-irradiation on electrophysical and properties of $Pb_{1-x}Mn_xSe$ (x=0.01) and $Pb_{1-x}Mn_xTe$ (x=0.04) epitaxial thin films have been studied. The resistance of the researched sample has been measured before and after irradiation. It has been determined that, after irradiation the samples' resistance at nitrogen temperature first gradually decreased, then sharply increased. This change is mostly observed in the samples of electrons with low concentration. This result wasn't achieved in the samples with high concentration. On the basis of the researches it has been determined that, by changing of Mn quantity in epitaxial films, maximum of photosensitivity slips towards short waves. It results with increase of Mn composition and width of the band gap. It has been defined from the experiments that, after irradiation the samples become more sensitive to light.

Thus, we have defined the optimum values of physical parameters of Pb1-xMnxTe(Se,S) nanofilms and ionizing radiation dose, which give us possibility to make radiation resistant and high sensitive spintronic devices controlled by magnetic field and temperature.

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H.R.Nuriyev became a Member of the: International Scientific and Engineering Conference on Photoelectronics and Night Vision Devices in Moscow (1992-2010), 9th International Symposium "High-purity Metallic and Semiconducting Materials" in Kharkov, Ukraine (2003), National Conferences of growth of crystals in Moscow (2004-2008), 3rd International Conference on Technical and Physical Problems in Power Engineering in Ankara, Turkey (TPE-2006), Russian Symposium on Scanning Microscopy, Probe Microscopy and analytic research Technique, Chernogolovka settlement in Moscow (2001-2011), 17th International Conference on Advanced Laser Technologies in Antalya, Turkey (2009), National Conference on application X-ray, Synchrotron radiations, Neutrons and Electrons for research of materials in Moscow (2003-2009), International Conference "Micro- and nanoelectronics 2005" in Moscow, Second International Conference on Technical and Physical Problems in Power Engineering (TPE-2004)in Tabriz, Iran (2004), Photovoltaic and Photoactive Materials-Properties, Technology and Applications, NATO Advanced Study Institute in Sozopol, Bulgaria (2001), Senior Member of the WSEAS –Intern. Conferences in Prague, Czech.(2011), Catania, Italy (2011), Montreux, Switzerland (2011), Rovaniemi, Finland (2012), Porto, Portugal (2012)

Career/Employment: Head of the "Diagnostics of Surface Epitaxial and Metal-ceramic Structures" Laboratory in the Institute of Physics of Azerbaijan National Academy of Sciences

Consultant and participant of the projects:
1. STCU Project № 3237, 01.07.2006-01.07.2008, "Make photoreceivers on the base of the epitaxial films of GaSe, GaTe, InSe lamellar semiconductors and monocrystals".
2. INTAS Project 01-0190, 01.01.2002 - 01.01.2004, "$Pb_{1-x}Mn_xTe$ epitaxial films and photosensitive homo- and heterostructures on their base".
3. ANAS project, 01.01.2010-31.12.2010, "Complex researches in the direction of making renewable energy sources with high efficiency on the base of nanostructure materials".

Publications: Number of papers in refereed journals: 200
Number of communications to scientific meetings: 43
Inventions: 6

Language Skills: Azeri, Russian, Georgian

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