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SELF REPORT

1. Personal data:

   Rafał Jakieła

   [redacted]

2. Education and Degrees

Ph. D. in Physics	2005	Institute of Physics, Polish Academy of Sciences Specialization: Solid State Physics Thesis topic: Diffusion Mechanisms in A3B5 semiconductors Ph. D. advisor: prof. dr hab. Adam Barcz
M. Sc. in Physics	1999	College of Sciences in Warsaw Specialization: Physical Measuring Apparatus Thesis topic: Silicon implantation into GaAs M. Sc. degree advisor: prof. dr hab. Adam Barcz Bachelor degree1997 College of Sciences in Warsaw Specialization: Physics, Mathematics and Chemistry with a Specialty in Electronic Measuring Equipment

[redacted]

3. Information on Previous Employment

01/07/2005 – now	Specialist, Institute of Physics, Polish Academy of Sciences Studies of semiconductor structures and materials using the SIMS method
01/09/2006 – now	IT administrator, Primary School 389 in Warsaw
30/09/1998 – 31/06/2005	Ph. D. student, Institute of Physics, Polish Academy of Sciences
01/11/1998 – 31/12/2015	Assistant, Institute of Electronic Materials Technology Studies of semiconductor grown by MOCVD epitaxy.
01/05/1998 – 31/12/1998	Programmer, Computer Service Matrix sp. z o.o.

4. Description of the achievements, set out in art. 219 para 1 point 2 of the Act

Subject of publications: ***The role of atmospheric elements in the semiconductors***

H1 **R. Jakiela**, E. Dumiszewska, P. Caban, A. Stonert, A. Turos, A. Barcz – *Oxygen diffusion into GaN from oxygen implanted GaN or Al_2O_3* – Phys. Status Solidi C 8 (2011) 1513
(IF = 0, cit. 7)
My contribution to this publication consisted of the idea and preparation of whole experiment, preparing of MOCVD grown sample for implantation and annealing, performing of all SIMS measurements as well as processing the obtained results. I estimate my contribution to the 70%.

H2 A. Barcz, M. Kozubal, **R. Jakieła**, J. Ratajczak, J. Dyczewski, K. Gołaszewska, T. Wojciechowski, G. K. Celler – *Diffusion and impurity segregation in hydrogen-implanted silicon carbide* – Journal of Applied Physics 115 (2014) 223710
(IF = 2.183, cit. 11)
My contribution to this publication consisted of performing of all SIMS measurements, processing the obtained results and participation in editing the manuscript. I estimate my contribution to the 40%.

H3 A. Barcz, **R. Jakieła**, M. Kozubal, J. Dyczewski, G.K. Celler – *Incorporation of oxygen in SiC implanted with hydrogen* – Nuclear Instruments and Methods in Physics Research B 365 (2015) 146
(IF = 1.389, cit. 2)
My contribution to this publication consisted of performing of all SIMS measurements, processing the obtained results and participation in editing the manuscript. I estimate my contribution to the 40%

H4 R. Czernecki, E. Grzanka, **R. Jakiela**, S. Grzanka, C. Skierbiszewski, H. Turski, P. Perlin, T. Suski, K. Donimirski, M. Leszczynski – *Hydrogen diffusion in GaN:Mg and GaN:Si* – Journal of Alloys and Compounds 747 (2018) 354
(IF = 4.175, cit. 5)

My contribution to this publication consisted of performing of all SIMS measurements, processing the obtained results and participation in editing the manuscript. I estimate my contribution to the 40%

H5 **R. Jakiela**, A. Barcz, J. Sarnecki, G. K. Celler – *Ultrahigh sensitivity SIMS analysis of oxygen in silicon* – Surface and Interface Analysis 50 (2018) 729
(IF = 1.319, cit. 3)
The above article is a summary of over 10-years research on oxygen diffusion in silicon. My contribution to this publication consisted of carrying out all SIMS measurements, writing a FTCS diffusion simulation program (Forward Time Center Space), comparing the SIMS results with the results obtained from the simulation and co-editing the manuscript. I estimate my contribution to the 80%

H6 **R. Jakieła** – *The role of atmospheric elements in the wide band-gap semiconductors* – Acta Physica Polonica A 136(6) (2019) 916
(IF = 0.545, cit. 0)
The above article is a summary of my over 20-years' experience in the measurement of dopants and impurities in wide-band semiconductors, i.e. GaN, ZnO and SiC, in which atmospheric elements, i.e. H, C, N and O, played a key role. The article is divided into 4 chapters (one for each element) including a summary of the current state of art on the properties of the element in the given semiconductor and original results aimed at showing the measurement methodology of a given element by the SIMS method with an emphasis on its detectability. The idea, measurement, writing and editing the whole manuscript is 100% of my contribution.

Total number of citations, excluding autocitation - **23**.
Statements of co-authors of publications, confirming them individual contributions to the paper are attached in attachment 5.

5. Scientific achievement being the subject of habilitation

5.1 Introduction

Since the beginning of the establishment of the Secondary Ion Mass Spectrometry (SIMS) laboratory at the Institute of Physics Polish Academy of Sciences, I have participated in the development and improvement of this complex analytical method and its applications for semiconductor studying. As a result of developing special procedures and spectrometer configuration, it was possible to obtain detection sensitivity for elements such as H, C, N, O, especially demanding due to their prevalence - at a level unattainable for most laboratories in the world [**H5**]. This fact somehow determined the main scope of my research and, as a consequence, the subject of my habilitation thesis.

All chemical processes taking place in the Earth's atmosphere are inevitably exposed to contamination with its gaseous components. Therefore, the main elements of the Earth's atmosphere, i.e. hydrogen (H), carbon (C), nitrogen (N) and oxygen (O), can be included in matter either as the main components or as trace contaminants. The same applies to semiconductor materials where they are either the main component of some semiconductors like GaN, ZnO, SiC, or intentional dopant like C in GaN, or N in ZnO and SiC. They present a common source of contamination in processes occurring at atmospheric pressure, but also those carried out under high vacuum, by interacting with residual gases. For many semiconductor material growth processes, these elements are inert gases, but very often have a dominant effect on their electrical properties. They can introduce donor or acceptor carriers into the material, or cause passivation of carriers in a semiconductors.

On the other hand, the widespread occurrence of atmospheric gases imposes a limit of detection when measuring their concentration in semiconductors, depending on the degree of their adsorption on the surface of the sample. Most quantitative analytical methods are not well adapted to measuring the content of these elements in solids, among others due to the high contribution of the environment to the useful signal. The secondary ion mass spectrometry method, operating in ultra-high vacuum and using the ionization facility of atmospheric elements, allows, as one of the few methods, to measure their concentration in materials at a ppm level or lower. It also gives the opportunity to determine their depth distribution into the sample. This allows studying the properties of such dopants in semiconductors, i.e. their diffusion and segregation coefficients, or formation energies related to the solubility of a given dopant in a semiconductor.

5.2 Oxygen properties in gallium nitride [**H1**]

Oxygen is the second abundant atmospheric element and constitutes over 20% of the Earth's atmosphere, and moreover ~ 46% of the Earth's crust and ~ 61% of the human body. It is also the most reactive element contained in the atmosphere, easily forming chemical compounds with the majority of elements. The high oxygen reactivity is due to its electron configuration. The two unpaired electrons of the O_2 molecule make it highly susceptible to bond formation.
Oxygen occurs in two allotropic forms: (oxygen – O_2 and ozone – O_3), both are excellent oxidants. Usually, oxygen is present in the oxidation state –2 in the O^{2-} form, but it can also form other ions, e.g. O_2^{2-} or O_2^{-}. For various possible oxidation states, many molecular compounds can be formed when another element reacts with oxygen.
In material engineering, oxygen is an issue mainly in III-V semiconductors, where in most cases act as a donor. A high oxygen content occurs in semiconductors containing elements very easily reacting with oxygen, i.e. Al in AlGaN and AlGaAs or Mn in MnGaAs and MnGaN.
The oxygen in GaN very easily substitutes the nitrogen site creating donor state O_N, due to the similar size of atoms. Therefore, nominally undoped GaN exhibits an n-type. The first report on the growth of GaN crystals stated that it is defects that are responsible for the high electron conductivity of the material, mainly the nitrogen vacancy V_N. This hypothesis has been questioned by Seifert [1], showing more likely oxygen contamination. By removing oxygen from ammonia used for crystal growth and using Mg_3N_2, significant reduction of electron concentration in GaN crystals was achieved. More comprehensive research on the oxygen in the GaN layers obtained by the MOCVD method was performed by Chung and Gershenzon [2]. The increase of carrier concentration along with an increase of oxygen concentration was observed and the energy level of O_N donor amounting to 78 meV was determined by means of luminescence measurements. In the oxygen-implanted GaN the ionization energy (~29 meV) of oxygen dopant by means of electrical measurements was determined [3]. Moreover, the low activation efficiency of O_N was observed. From the redistribution of oxygen implanted into GaN, the upper limit of the oxygen diffusion coefficient at the level of 2.7×10^{-13} cm^2/s at the temperature of 1125°C was determined. Low diffusion coefficient of oxygen was confirmed in SiO_2/GaN samples [4].
The oxygen concentration in GaN depends on the conditions of material growth [5]. MBE or MOCVD growth technology allows to obtain clean enough samples that the level of oxygen concentration is below the detection limit of the SIMS method (<10^{16} at/cm^3) [**H6**]. Equally

low oxygen concentration was also achieved in crystals grown by hydride vapor phase epitaxy (HVPE), which makes it possible to obtain layers of even hundreds of microns thick [6, 7].
Bulk crystals growth methods such as high-pressure method [8], ammonothermal method [9] or more and more popular *Na-flux* method, unfortunately not yet achieved such crystal purity in terms of oxygen content [10, 11]. The greatest purity in this respect was achieved in *Na-flux* method where the oxygen concentration has been reduced to a level of ~3×10^{16} at/cm^3. In the ammonothermal and high-pressure methods, the oxygen concentration exhibits values respectively >10^{18} at/cm^3 and >10^{19} at/cm^3.
In the paper [**H1**] I focused on the high oxygen content in the GaN layer adjacent to the sapphire substrate, often observed in measurements of GaN layers deposited on Al_2O_3 by various epitaxial methods. The aim of the study was to check if the extent oxygen that builds up in the semiconductor during the growth of GaN layers is oxygen from sapphire or extent oxygen adsorbed on the substrate is the result of improper substrate preparation for the epitaxy process. For this reason, I wrote a research project to the **SPIRIT** consortium and I was able to visit the **Helmholtz-Zentrum Dresden-Rossendorf Ev** laboratory in order to prepare appropriate specimens. The experiment consisted of implanting oxygen and argon into sapphire substrates as well as oxygen into GaN layers. GaN epitaxy by the MOCVD method, under standard growth conditions for this material at 1150ºC was then performed. In order to increase the detectability of the tested element, ^{18}O oxygen isotope with 0.2% abundance was used for implantation, which allows for reduction of the detection limit of oxygen in GaN by three orders of magnitude. Then, oxygen depth profiles were determined to study the oxygen diffusion from three different sources: an oxygen-implanted GaN substrate, an oxygen-implanted sapphire substrate, or a sapphire substrate in which oxygen should be released by radiation damage.
Test results showed that oxygen diffusion into the deposited GaN layer occurs only when oxygen was implanted into the Al_2O_3 substrate (Fig. 1a). In the case of oxygen implanted into GaN and oxygen released from sapphire through radiation damage caused by Ar implantation, the diffusion into the overgrown GaN layer was not observed (Rys. 1 b, c).

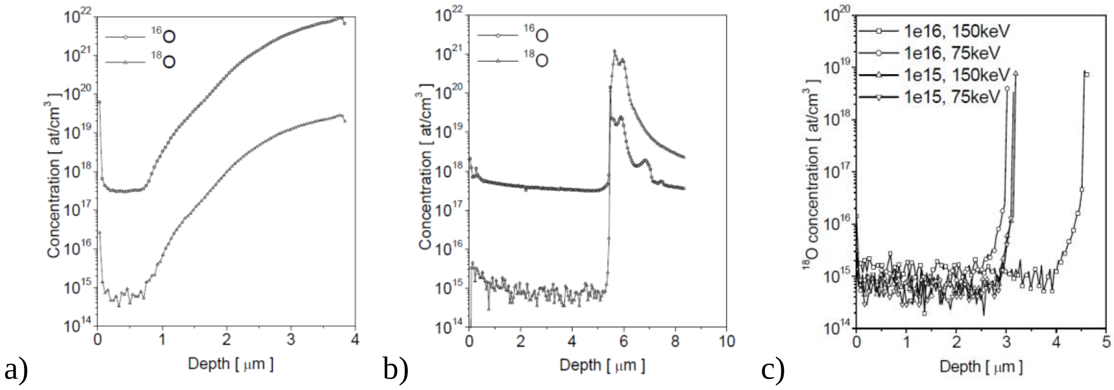

Fig 1. Oxygen depth profiles in MOCVD GaN layers deposited on: a) O-implanted saphire, b) O-implanted GaN, c) Ar-implanted saphire. [**H1**]

The experiment showed that unbound residual oxygen atoms from the GaN / sapphire interface or from the sapphire surface diffuse and build into the GaN layer during the epitaxy process. At the same time, the oxygen implanted into GaN is strongly bound to the GaN crystal lattice, most likely substituting nitrogen vacancies and has no possibility of diffusion and incorporation into the growing epitaxial layer. Similarly, the sapphire subjected to ion bombardment was also excluded as a potential source of oxygen trapped in the GaN layer.

5.3 Hydrogen properties in silicon carbide [**H2**] and [**H3**]

Hydrogen is one of the most widespread elements on Earth. In the Earth's atmosphere, in the largest amount occurs bonded with oxygen as water, but its large reservoir is also hydrocarbons being the main component of all fossil fuels. Hydrogen is also common impurity in many semiconductors, mainly due to the presence in the atmosphere, but also as constituents of precursors used for the epitaxy of all semiconductors. Despite the simple structure (only one proton and one electron) hydrogen exhibits very complex behavior. Its ability to bind to broken or weak covalent bonds found in localized or extended defect centers often results in shifting defect energy levels from or into the energy gap. This results in passivation or compensation of both shallow and deep defects, but also the creation of extended defects, which have a large impact on the electrical and optical properties of semiconductor materials. Hydrogen properties are well known in more traditional semiconductors, such as Si and GaAs, while to a lesser extend in wide bandgap semiconductors, such as gallium nitride GaN, zinc oxide ZnO or silicon carbide SiC.

Hydrogen, due to its size, in the crystal lattice of the semiconductor usually occurs in the interstitial position where can act in three electrical states: H^O, H^+ and H^-. As a result of Coulomb attraction, H+ (proton) builds into places with high electron density, hence interacts preferably with anions, whereas H^- (a proton with 2 electrons) builds into places with low electron density, i.e. connects to cations. Charge of the hydrogen atom in the semiconductor is determined by Fermi's energy level, hence H^+ is the preferred state in the p-type whereas H^- in the n-type material. The amphoteric properties of hydrogen atom make it able to compensate both electron and acceptor conductivity.

Chris van de Walle showed [12] that the H^O state is generally unstable in semiconductors, and the value of the transition energy level between H^+/H^- is pined relative to the vacuum level. Thus, the properties of hydrogen in a given semiconductor depend on the position of the semiconductor energy gap relative to the vacuum level, in the result on such parameters as electron affinity and ionization potential Fig.2

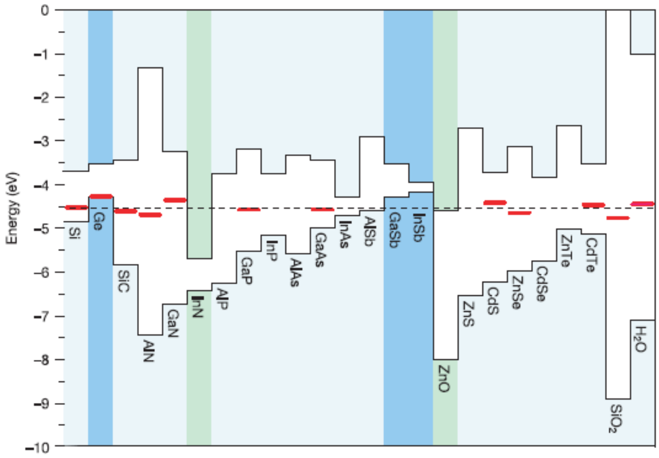

Fig. 2 Band line-ups and position of the (+/–) level for a range of semiconductors and insulators. For each material, the lower line indicates the position of the VBM, the upper line the position of the CBM, and the thick red line the position of the hydrogen (+/–) level with respect to the VBM.

Therefore, depending on the location of the energy gap relative to the vacuum level in a given semiconductor, hydrogen may exhibit acceptor properties as in Ge or GaSb, or donor – as in InN or ZnO.

SiC silicon carbide is an excellent material for the production of electronic components operating at high temperatures, as well as achieving high power and high breakdown voltage. Due to the wide bandgap of 3 eV for 6H politype to 3.2 eV for 4H politype, typical leakage currents in devices based on this material are order of magnitude lower than for silicon. In addition, SiC is the only two-component semiconductor that can be thermally oxidized to produce uniform SiO_2 oxide. High electrical stability is undoubtedly an advantage, but it imposes severe requirements when it comes to technological processes such as doping. Due to the low diffusion coefficients of the dopants, they must be introduced in the implantation process and then activated by heating at high temperatures. The use of high temperatures very often causes the build-up of impurities, e.g. hydrogen. Hydrogen is also a component of gas precursors used for SiC epitaxial growth, i.e. silane (SiH_4) or propane (C_3H_8), as well as dopants, e.g. diborane (B_2H_6).

The high mobility of hydrogen in SiC at temperatures of above 1000°C [**H2**] was demonstrated and the difference in the diffusion coefficient depending on the electrical properties of the semiconductor was confirmed. In the p-type material, where hydrogen is present as a proton, the diffusion range at the temperature 1000^{O}C during 1h annealing is ~0.5 microns higher than in n-type material. Fig 3. Both depth profiles exhibit a characteristic drop in concentration, indicating diffusion governed by concentration dependent diffusion coefficient.

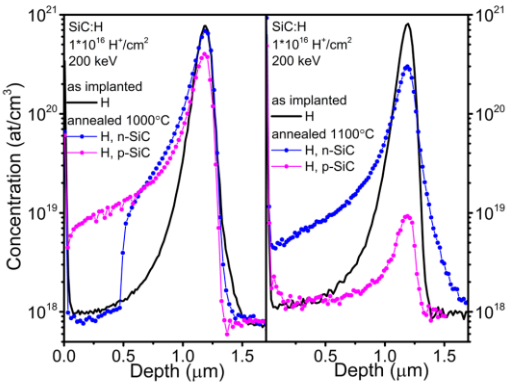

Fig. 3 Hydrogen depth profiles in *n*- and *p*-type SiC crystal, as implanted and annealed at the temperature of 1000 and 1100°C during 1h under Ar pressure. [**H2**]

Such characteristic profiles appear in the case of various dopants in several crystalline materials and indicate that the atom diffusion is determined by an additional factor reducing the atom mobility [13, 14, 15, 16]. Influencing factors can be level of dopant trapping defects resulting from the Fermi level, the charge state of defects affecting they mobility – which may also result from the Fermi energy level or as in the case SiC, the complexes of dopants with diffusing atoms. Then the hydrogen diffusion proceeds as follows: hydrogen atoms diffusing into subsequent layers of the semiconductor, first passivates the dopants becoming less mobile, then saturating all defects the excess of atoms diffuse deeper and the process repeats. The characteristic drop of the hydrogen depth profile occurs at the H concentration level equal to both the acceptor and donor concentration. The diffusion coefficient is higher for boron-doped material, indicative of lower H-B binding energy compared to the H-N bond formed in the nitrogen doped material. Our results also show that the diffusion of hydrogen in SiC take place with the contribution of point defects produced by post-implantation damage, because the diffusion process was observed only towards the area of high concentration of defects. Interestingly, defects arising from the implantation process also cause oxygen segregation.

The use of higher implant doses leads to the formation of an irreversible, well-defined layer of microcavities, voids and other extended defects containing large amounts of agglomerated hydrogen. At high temperatures, such a layer tends to exfoliation, i.e. separation from the substrate. In addition, a high oxygen content was observed in the defective layer, the concentration of which drastically exceeded the level usually observed in bulk silicon carbide.

The results of research on the origin of these excess amounts of oxygen was presented in the next publication [**H3**]. The aim of this work was to clarify the source of oxygen incorporation into 4H-SiC - a semiconductor in which both solubility and diffusivity of oxygen are believed to be extremely low. For this purpose, hydrogen and deuterium have been implanted into silicon carbide at the energy range of 200 - 1000 keV to a doses of 10^{16} – 10^{17} at/cm^2. The use of deuterium instead of hydrogen allow better H detection by the SIMS method and higher energy density deposited at the end of the ion penetration path where the most of damage is formed. For comparison, silicon substrates obtained by Czochralski method and FZ (floating zone) were also implanted in the same way. Similarly to the first publication [**H2**], SIMS studies showed that heating of such a damaged layer in both Si or SiC samples, causes agglomeration of many impurities. Oxygen was detected in both thereby produced silicon materials. In the case of SiC, high oxygen concentrations were observed between the substrate and the implanted layer after heating at 1150ºC under both steam and pure argon atmosphere Fig. 4a and 4b.

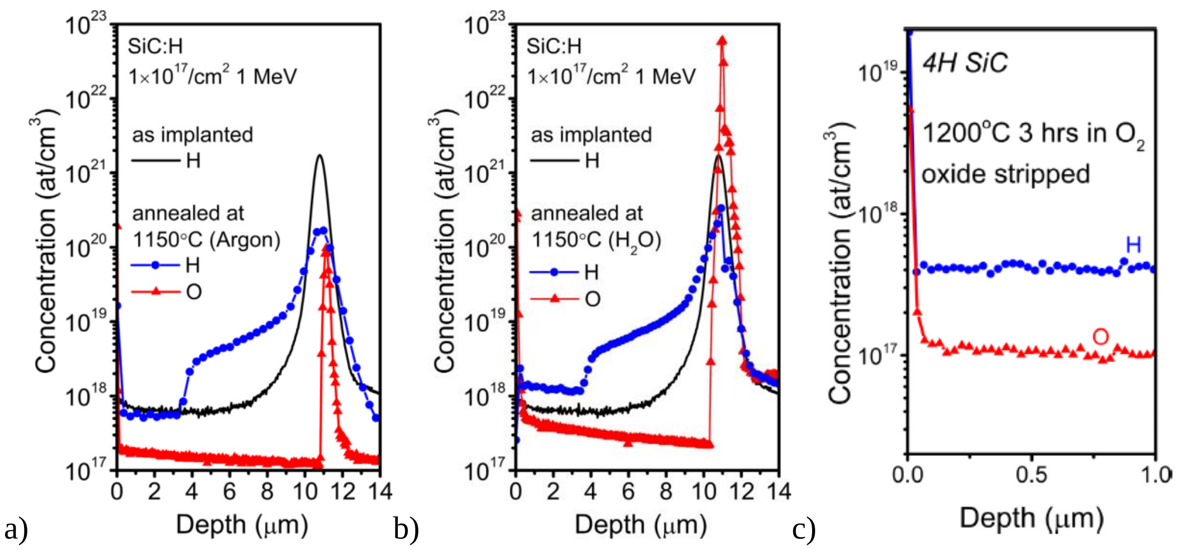

Fig. 4 Hydrogen and oxygen depth profiles in the H-implanted SiC at the energy of 1MeV to a dose $10^{17}/cm^2$ annealed at 1150ºC: a) Ar ambient pressure, b) H_2O ambient pressure, and c) pure SiC crystal annealed at 1200ºC under H_2O ambient pressure. [**H3**]

In the case of annealing of the SiC crystal substrate under the water vapor atmosphere, incorporation of oxygen or hydrogen was not observed Fig. 4c. In addition, the following observations were made. Heating of implanted silicon samples in the air or in argon at the temperature of 725ºC caused complete outdiffusion of deuterium and the appearance of hydrogen in the radiation-defective layer. Further heating of Si samples at higher temperatures resulted in a decrease of hydrogen and an increase of oxygen content.

In the case of H-implanted SiC, hydrogen did not completely outdiffuse from the semiconductor even at the temperature of 1150ºC. At the same time, SiC layer was completely exfoliated under conditions:

- H-implanted material at the energy of 400keV dose 8×10^{16} /cm^2 annealed at 1050ºC
- D-implanted material at the energy of 600keV dose 1.2×10^{17} /cm^2 annealed at 950ºC

Very interesting results were received in sample implanted with the highest energy atoms, i.e. 1 MeV. Similarly to the experiment described in the paper [**H2**], when heating at the highest temperature of 1150ºC, hydrogen diffusion to the surface with a characteristic drop in concentration indicating concentration-dependent diffusion coefficient was observed Fig. 4a and 4b. At the same time, annealing did not bring measurable hydrogen diffusion into the deeper layers of the sample. The most interesting and surprising feature is the oxygen peak coinciding with the projected range of H^+ ions. It is worth mention that the oxygen layer was also observed in the sample annealed in nominally pure argon Fig. 4b.

Comparing the amount of oxygen found in the cavity band from sample annealed in Ar ([O] = 10^{15} /cm^2) Fig. 4a, to the maximum O concentration in SiC inferred from Fig. 4c, it follows that the number of agglomerated oxygen atoms exceeds the quantity of O atoms contained in the entire 300 micrometer-thick wafer. This circumstance strongly suggests that the source of oxygen should be the annealing ambient rather than the semiconductor bulk. Such conclusion is further supported by a much higher gettering efficiency when using water vapor as an annealing ambient relative to argon. At this point one may question why oxygen is being found also in samples annealed in pure argon. Our experience (confirmed by similar observations in other laboratories) is that air-tightness of conventional quartz tube furnaces is never perfect, hence, some contamination with air or water vapor, cannot be avoided. As a result, thin surface oxides inevitably grow on both Si and SiC surfaces irrespective of the purity of gases released to the furnace. However, because the low solubility, the supply of oxygen from the surface oxide to the gettering layer cannot account for the magnitude of the observed effect.

We postulated that the only path for oxygen agglomeration is migration of gaseous O_2 or H_2O from the edge of the sample through the porous layer. The exact nature of such lateral diffusion remains to be further explored. There is no doubt, however, that the circumstance favoring this before unseen process is the considerable porosity of the buried layer, with voids and cavities visible in the TEM image.

5.4 Hydrogen properties in gallium nitride [**H4**]

The increase of interest in research on hydrogen in gallium nitride occurred when fabrication of GaN based p-n junction was attempted. Unfortunately, the magnesium doped GaN layers were highly resistive despite very high dopant concentration. The hydrogen passivation of magnesium, which forms inactive complexes with a dopant was showed by Nakamura [17]. The annealing of the layers at suitable temperature results in the breaking of Mg-H bonds and thus activation of magnesium. Interestingly, subsequent experiments on hydrogen in GaN at low temperatures showed a complete lack of hydrogen atom mobility up to 900° C in GaN as well as InN or AlN [18]. It would follow that when heating GaN crystals at high temperatures, hydrogen bonds with magnesium are broken, but atoms are not removed from the material. Subsequent studies have shown, that the acceptor conductivity in the GaN layers grown by MBE can be extinguished by introducing hydrogen into the material. [19].

The variation of hydrogen mobility in materials with different Fermi levels been explained from the *first principles* calculations [20]. Hydrogen as a proton H^+ prefers the position close to the nitrogen, forming a bond similar to that found in NH_3 ammonia. Among the possible positions of both atoms, the nitrogen antibonding site is the energetically most stable. Interestingly, this is a different position than in Si or GaAs, due to the polarity of gallium nitride. The activation energy for migration of bound hydrogen as 0.7 eV was calculated. For neutral hydrogen H^O, much smaller energy differences were found according to different sites. The energetically most stable site for H^O is the Ga antibonding site. For a proton with two electrons H^-, the Ga antibonding site is energetically most stable, as well as in the tetrahedral interstitial site, where the distance between hydrogen and the neighboring Ga atoms is maximized, and the charge density of the bulk crystal has a global minimum. In this position, the energy barrier for hydrogen migration is as much as 3.4 eV. The charge state of hydrogen in GaN affects its diffusion coefficient. In an n-type material (eg GaN: Si) this parameter is much lower than in a p-type material (e.g. GaN: Mg), which was shown by calculations from the *first principles* [21,22] and confirmed experimentally in the hydrogen implanted [23], plasma diffused [24, 25] or epitaxial [26] GaN layers.

During my collaboration with the Epitaxy Lab at ITME, one of the first tasks was to develop a method for removing hydrogen from GaN:Mg layers grown by the MOCVD method. Thus, the heat treatment of a series of GaN samples at various temperatures and atmospheres was carried out. The concentration of hydrogen in the heated layers was determined by the SIMS method. The paper published in Optica Applicata [27] summarize the results of our studies. SIMS measurements showed that annealing at 900°C for 15 min in N_2 atmosphere allows removed hydrogen to the background level of 10^{18} at/cm^3.

Simultaneous SIMS depth profiling of hydrogen and magnesium allowed me to develop a methodology for Mg measurement in GaN using negative ions, which is a non-standard method for electropositive element. I have shown that the Mg profile can be obtained by measuring the $MgGaN^-$ ion cluster, obtaining Mg detection limit in GaN at the concentration level of 10^{17} at/cm^3.

In the experiment with hydrogen diffusion in GaN layers with different types of conductivity described in the article [**H4**] we showed the dependence of the diffusion coefficient on the type of doping. However, unlike previous works, the experiment was conducted on homoepitaxial GaN layers with low (10^7 /cm^2) dislocation density. It allowed observing diffusion in the bulk crystal and reducing the effect of hydrogen diffusion through dislocations.

For this purpose, 4 different layers with dislocation density and conductivity type shown in Table 1 were annealed in the MOCVD reactor at a temperature of 1020ºC under the H_2-NH_3 atmosphere for 20 min.

Sample	Dislocation density (1/cm^2)	Conductivity type and carrier concentration (1/cm^3)
bulk crystal	10^7	n = 6.1×10^{18}
GaN:Mg layer (MBE)	10^7	p = 3.5×10^{17}
GaN:Si layer - template	5×10^8	n = 3.6×10^{18}
GaN:Mg layer (MOCVD)	5×10^8	p = 7.2×10^{17}

Table 1. Samples used in the experiment

The SIMS was the key method used to study the hydrogen depth profiles. The main difficulty in SIMS measurement was achieving low hydrogen detection limit in GaN:Mg layers 0.5 micron and GaN:Si layers 1.5 micron thick. For such thin GaN layers, the primary beam current and the setting of the secondary beam optics must be adequate to achieve proper detection (low hydrogen background) at suitable depth resolution. In my measurement hydrogen detection at the level of 2×10^{17} at/cm^3 was achieved, which is the value only a one order of magnitude higher compared to that achieved in the bulk GaN [**H6**], where the higher primary beam current densities can be used.

We showed a high hydrogen diffusion coefficient in *p*-type (Fig. 5a and 5b), and no hydrogen diffusion in *n*-type gallium nitride layers (Fig. 5c and 5d).

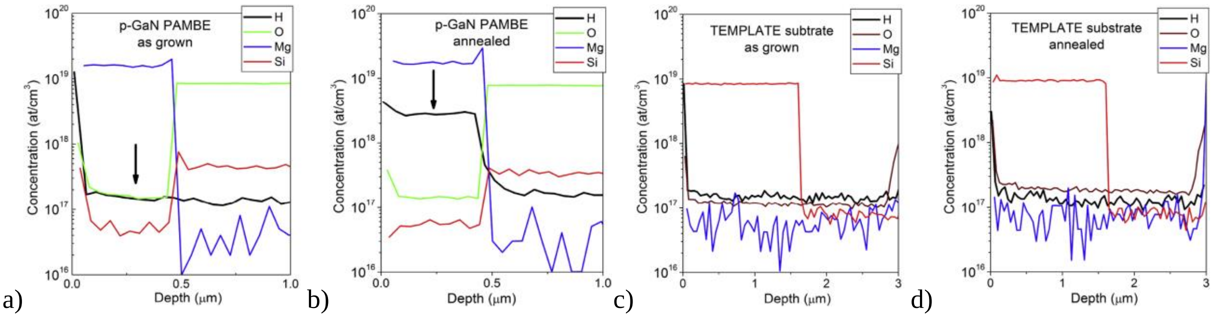

Fig. 5 SIMS depth profiles in: a) as deposited GaN:Mg layer, b) annealed GaN:Mg layer, c) as deposited GaN:Si layer, d) annealed GaN:Si layer. [**H4**]

Luminescence measurements carried out on the samples showed the greatest effects in the GaN: Mg layer obtained by the MBE method. In the layer, which the SIMS measurement did not show the presence of hydrogen before annealing and a high hydrogen concentration after annealing (Fig. 5a and 5b), a decrease and shift of the "blue" luminescence and an increase in the "yellow" luminescence compared to the original sample were observed. The effect most likely resulted from the passivation of the Mg dopant by hydrogen through the formation of Mg-H complexes (reduction of "blue luminescence"), and the formation of hydrogen complexes with the V_{Ga}-H gallium vacancy ("yellow luminescence").

5.5 Oxygen properties in silicon [**H5**]

The above work was devoted to the subject probably best studied by our group, namely the study of thermodynamic properties of oxygen in silicon using the SIMS method, based on its solubility and diffusion. The first results of these studies were published in the paper [28] before my Ph.D. degree. Publication [**H5**], which was submitted last year to the journal entitled *Surface and Interface Analysis*, is a summary of our years-long research in this field, conducted mainly on silicon detectors developed for the **European Hadron Collider of the European Organization for Nuclear Research CERN.**

In silicon, similarly to SiC, stable planar SiO_2 films serve as insulators, masks, and gate barriers in metal-oxide semiconductor transistors. Czochralski-grown Si wafers usually contain oxygen in quantities 7–10×10^{17} at/cm^3. At room temperature, oxygen-supersaturated Si is stable as the oxygen atoms occupy predominantly bond-centered interstitial sites in the lattice [29]. At elevated temperatures, diffusion of oxygen leads to its aggregation into SiO_x precipitates and to the migration of atomic oxygen toward the external boundaries of the sample [30]. If the oxygen content is sufficiently low, as in the float zone (FZ) silicon, it is possible to indiffuse oxygen into the semiconductor from thermally grown or deposited SiO_2 film [31].

To determine principal quantities characterizing the behavior of oxygen in Si, diffusivity (D) and solid solubility (C_s) have been subject to numerous investigations employing a wide spectrum of methods. However, SIMS delivers combined information on the depth profile of both isolated and agglomerated species. Heating of supersaturated silicon leads to outdiffusion of the interstitial O atoms and their segregation to the surface oxide, leaving a so-called "denuded zone" in the semiconductor.

The resultant surface concentration, $C_{surf}(out)$, has commonly been taken as the solid solubility of oxygen C_s at a given temperature [32, 33, 34]. However, we have shown that a correct measure of C_s is the surface concentration $C_{surf}(in)$ that is being set upon indiffusion of oxygen into an FZ Si [28]. This is because atomic diffusion is an equilibrium process and, applying sufficiently rapid quenching to room temperature, the precipitation can be effectively suppressed. The outdiffusion profile cannot provide reliable information on the solid solubility of oxygen because the atomic diffusion is accompanied by aggregation into immobile SiO_2 precipitates that cannot be removed by diffusion. As a consequence, $C_{surf}(out)$, determined from SIMS data, always appears higher than $C_{surf}(in)$. Regarding the diffusion coefficient of oxygen in Si, determination of which requires only relative values of the concentration profile, our findings do not deviate appreciably from the previously established data [35]. Our corrected formula for C_s is:

$$C_S = 9{,}1 \times 10^{22} e^{\left(\frac{-1{,}57eV}{kT}\right)}$$

Our considerations on the measurement of oxygen in silicon by the SIMS method should be started with the detection limit of this element. Fig. 6 summarizes the measured oxygen concentration in different types of silicon that we have dealt with:

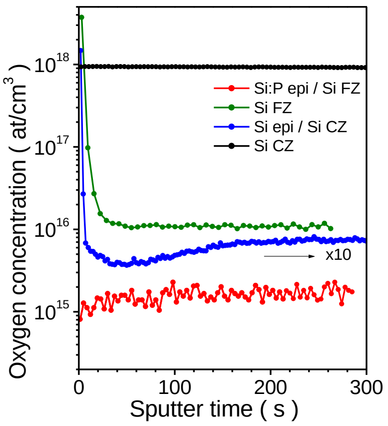

Fig. 6 Oxygen concentration levels measured in Si CZ, Si FZ, and in epitaxial Si grown on either CZ or FZ silicon. [**H5**]

The values of 10^{18} at/cm^3 and 10^{16} at/cm^3 are considered typical for Si CZ and Si FZ, respectively.

Our record low O signal of 2×10^{15} at/cm^3 was obtained for silicon grown by atmospheric pressure CVD, on Si FZ substrate. This value coincides with the detection limit of oxygen in Si predicted by Gnaser [36] through extrapolation of the experimental data to the ideal vacuum in the analysis chamber. The plot shown was taken after several minutes of uninterrupted sputter etching with Cs beam at the energy of 14.5 keV. It should be noted that this result was achieved by using a chromate cesium container in the ion source, yielding a beam of 600 nA current, ie, twice higher than the current of a carbonate cesium source that we routinely use. A series of tests with different raster sizes indicated that the detected O signal originates from the O-containing particles that adsorb on the analyzed surface and not from sample bulk. These particles are likely to be resputtered by multiply scattered primary Cs ions. One such test is shown in the Fig. 7.

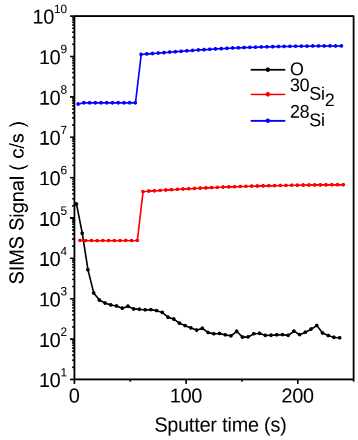

Fig. 7. Raw data of SIMS depth profile of oxygen in Si. A step in Si profiles is caused by increase of primary beam current density (raster change) [**H5**]

At first, the surface was raster-scanned over 200 µm × 200 µm area, then the raster was reduced to 50 µm × 50 µm while keeping the probing area unchanged. The O signal does not follow the signals from silicon, which means that the source of "measured" oxygen is located outside the sample bulk. In our system, the conversion factor from SIMS signal to oxygen concentration *relative sensitivity factor* (RSF) amount 10^{19}. Than oxygen concentration is calculated as follows:

$$C_O = 10^{19} \frac{I_{^{16}O}}{I_{^{30}Si_2}} (at/cm^3)$$

where: C_O – oxygen concentration, $I_{^{16}O}$ – oxygen signal, $I_{^{30}Si_2}$ – silicon signal.

Thus, the result presented in the Fig. 7 indicates that silicon epitaxial layers have an extremely low concentration of oxygen (<10^{15} at/cm^3), while oxygen adsorption during such a sensitive measurement have to be monitored and minimized as much as possible. It is noteworthy that there is no confirmation in the literature for such a low oxygen content in Si. Most epitaxial structures are produced on a silicon substrate obtained by the Czochralski method with an oxygen concentration above 10^{18} at/cm^3, which diffuses into the layer when growing at 1100°C. Achieving a low detection limit requires a high current density of the primary beam so that the flux of oxygen secondary ions sputtered from the material prevails over adsorption rate of the oxygen-containing species. However, while the adsorption rate of residual gases may be assumed constant, the resputtering caused by the (back) scattered incident Cs ions will scale with the primary current. Also, the removal speed cannot be excessive when fine features in the concentration profile have to be distinguished. Distribution of oxygen implanted into the epi-Si was measured with the sputtering rate of ~20 nm/s (Fig. 8). The oxygen implantation parameters were energy 210 keV and a dose of 5×10^{14} /cm^2. Here, the background level, below 10^{16} at/cm^3, could have been lowered by using higher current but at the expense of the profile definition.

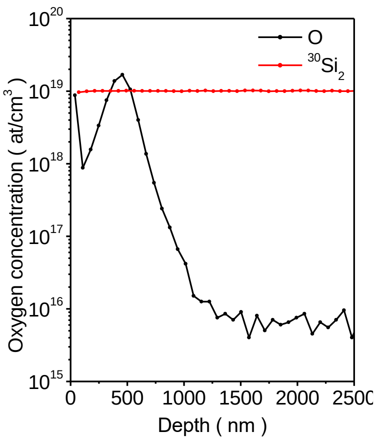

Fig. 8. Depth profile of oxygen-implanted epi-Si. Dose 5×10^{14} at/cm^2, energy 210 keV [**H5**]

Application of the conditions described above allowed studying the oxygen kinetics in Si crystals. On Fig. 9 the oxygen profiles in Si as received under different annealing condition are shown.

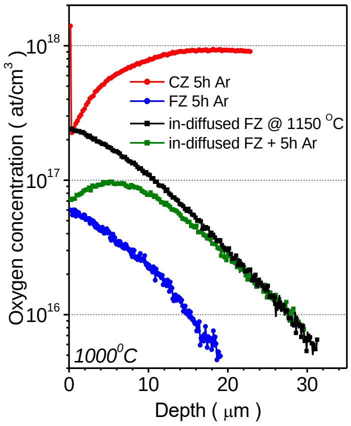

Fig. 9. Oxygen profiles: - outdiffusion form CZ Si (red), - diffusion into FZ Si (blue), - diffusion into FZ Si at temp. 1150°C (black), - after additional annealing the previous sample (green) [**H5**]

The most important result is the difference in surface concentrations of oxygen in FZ (low O concentration) and Czochralski (high O concentration) silicon crystals after annealing at 1000°C. Oxygen diffuses into the Si FZ, while it outdiffuses from the Si CZ. The surface concentrations of oxygen are not identical despite equal heating conditions. It is different when oxygen is diffused at a higher temperature (black profile) and then annealed it in a lower temperature (green) profile. Then the surface concentration of oxygen becomes comparable under the same annealing conditions (green and blue profile).

A common observation is that heating of Si CZ crystal containing ~10^{18} at/cm^3 results in outdiffusion of oxygen to the oxide with surface concentration pinned at a value depending on the solid solubility and degree of precipitation for a given temperature. Theoretically, however, if Si with atomically clean, oxide-free surface could be subjected to heat treatment in an inert gas, a zero oxygen concentration at the surface should be observed.

One method to realize such artificial situation is to heat the Si wafer in vacuum. We have employed a vacuum chamber used for evaporation of metals. Oxygen profiles in Si crystal annealed under such condition are presented on Fig. 10.

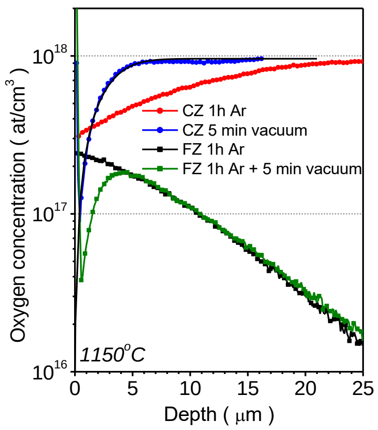

Fig. 10. Oxygen diffusion profiles into Si FZ (black) and outdiffusion form Si CZ (red) as annealed at 1150°C, effect of 5 min. annealing in vacuum at 1150°C for CZ Si (blue) and FZ Si (green) [**H5**]

Oxygen profiles for both bare CZ and previously indiffused FZ Si at 1150°C exhibit a deep depletion near the surface, with surface concentrations ~10× lower than the corresponding values for annealing in argon. This result shows, that the native oxide was removed by sublimation and a clean Si surface was exposed to vacuum. In such case, oxygen freely outdiffuses from the crystal.

For some applications, there is a need to monitor the oxygen distribution over large depths up to the entire thickness of the wafer. Such is a case of indiffusion of oxygen from both sides into an FZ Si for enhanced radiation hardness of charged particle detectors. In this situation, when the depth of the eroded crater becomes comparable with its lateral size, substantial redeposition of material on the crater bottom and resputtering from the crater walls make the resultant profile unreliable, as shown in Fig. 11. To get correct values, beveling of the sample is necessary, followed by step-by-step measurements. Enlarging the crater to dimensions typically used for "nonatmospheric" species would improve the depth resolution but degrade the detectability limit (not to mention the unacceptable increase of the time of analysis).

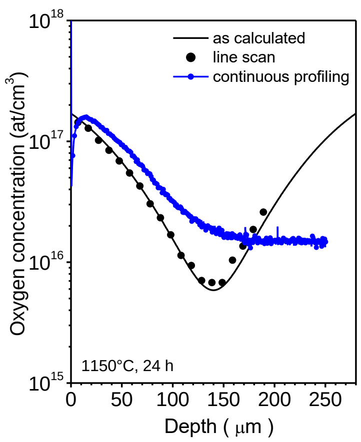

Fig. 11. Comparison of different results concerning deep O diffusion into FZ Si: (1) calculated numerically with $D=2\times10^{-10}$ cm^2/s, $C_O=1.7\times10^{17}$ at/cm^3, (2) line scan on a beveled surface with a background correction (3) dynamic SIMS profile.

5.6 Properties the atmospheric elements in wide-bandgap semiconductors (**H6)**

The above publication is a review and summary of years-long research related to the detection and profiling of atmospheric elements in wide-band semiconductors such as GaN, ZnO and SiC performed in our laboratory. The publication contains four sections on the properties of particular atmospheric elements, i.e. H, C, N and O. The first chapter concerns the properties of hydrogen in GaN, ZnO and SiC, the second chapter describes the properties of carbon in GaN and ZnO, the third chapter concerns the properties of nitrogen in ZnO and SiC, while the fourth chapter describes the properties of oxygen in GaN and SiC. All chapters also contain the methodology of SIMS measurements for mentioned elements in the relevant materials, developed based on my own measurement experience. For this purpose, standards of studied elements in the defined semiconductors were prepared as the implanted samples with a specific dose D and implantation energy E:

a) Hydrogen in:
   - GaN – D=1e16 cm^{-2}, E=25 keV,
   - ZnO – D=1e16 cm^{-2}, E=250 keV,
   - SiC – D=1e16 cm^{-2}, E=150 keV
b) Carbin in:
   - GaN – D=1e16 cm^{-2}, E=350 keV,
   - ZnO – D=1e16 cm^{-2}, E=500 keV,
c) Nitorgen in:
   - ZnO – D=8.6e15 cm^{-2}, E=500 keV,
   - SiC – D=2e16 cm^{-2}, E=100 keV,
d) Oxygen in:
   - GaN – D=1e16 cm^{-2}, E=400 keV,
   - SiC – D=5e15 cm^{-2}, E=100 keV.

All standards were measured by the SIMS method under the following measurement conditions:

a) O_2^+ beam at 8keV, positive secondary ions, primary beam current 800 nA, sputtered area R=200×200 μm,
b) Cs^+ beam at 5.5keV, positive secondary ions, primary beam current 200 nA, sputtered area R=200×200 μm,
c) Cs^+ beam at 14.5keV, negative secondary ions, primary beam current 200 nA, sputtered area R=200×200 μm,

equal for all elements. Comparison of depth profiles of atmospheric elements for individual measurement conditions has allowed to determine the best SIMS measurement methodology. Although all atmospheric elements provide the best detectability as negative ions, a methodology for measuring these elements as positive ions was developed. The results described in the paper [**H6**] present the circumstances and possibilities of atmospheric elements detection as positive secondary ions. This is particularly applicable when the simultaneous study of atmospheric elements with electropositive elements is crucial for understanding the processes occurring in semiconductor material. Examples of such measurements published in peer-reviewed journals are collected in references to this work.

Particular importance was placed on the detection of nitrogen, which of all atmospheric elements has the lowest electron affinity and the highest ionization energy. These parameters are extremely unfavorable for the SIMS measurement, compared to the other elements from the Periodic Table. In the publication [**H6**] I described the methodology needed to achieve adequate nitrogen detection (detection of the lowest concentrations) by measuring its clusters with other elements, i.e. N-Si and N-C in silicon carbide or N-O in zinc oxide.

The work also contains the result of SIMS measurement showing the lowest detection limit of atmospheric elements in gallium nitride obtained by the Halide Vapor Phase Epitaxy method Appropriate methodology allowed to achieve the hydrogen detection at the level of 8×10^{15} at/cm^3, carbon – 5×10^{15} at/cm^3 and oxygen – 10^{16} at/cm^3 (Fig. 12)

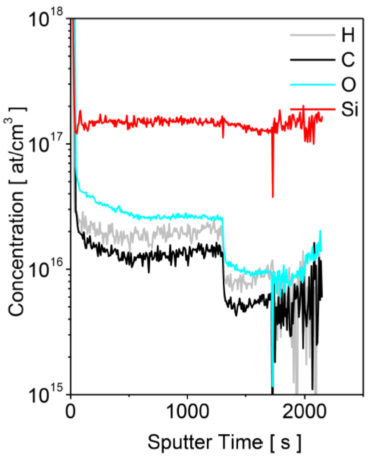

Fig. 12. Depth profiles of H, C, O and Si in GaN bulk crystals [**H6**]

Characteristic drops on the SIMS profiles of particular ambient elements result from testing the analytical methodology by increasing the density of the primary beam current with appropriate settings of the secondary beam optics.

The implanted samples were also used as standards for determining the relative sensitivity factors (RSF) for studied elements, enabling the quantitative determination of atomic concentration.

The bibliography of the publication [**H6**] contains nearly 50 references in which my SIMS measurements were an important contribution to understanding the results presented in the cited papers.

6. Bibliometric information

6.1 Total Number of Publications

191 including **160** after obtaining a Ph.D. degree in Physics. The papers were published in peer-reviewed journals having high international reputation, such as:

- Acta Physica Polonica – 21 articles (all after obtaining Ph.D. degree)
- Journal of Crystal Growth – 14 articles (including 13 after obtaining Ph.D. degree)
- Applied Physics Letters – 13 articles (including 10 after obtaining Ph.D. degree)
- Journal of Alloys and Compounds – 12 articles (including 11 after obtaining Ph.D. degree)
- Physical Review B – 9 articles (all after obtaining Ph.D. degree)
- Semiconductor Science and Technology – 8 articles (including 7 after obtaining Ph.D. degree)
- Journal of Physics: Condensed Matter – 4 articles (all after obtaining Ph.D. degree)
- Journal of Applied Physics – 4 articles (all after obtaining Ph.D. degree)

6.2 Total Impact Factor of publications

after the *Journal Citation Reports* (JCR) according to year of publication = **302**

6.3 Total citation number of publication

after the WoS base on 20/01/2020 = **1974** including without autocitation **1737**

6.4 Hirsch Index of publication

after the WoS base on 20/01/2020 = **22**

7. Presentation of significant scientific activity carried out at more than one university, scientific or cultural institution, especially at foreign institutions.

7.1 Scientific work before obtaining a Ph. D. degree in physics

After graduating from the Electronic Technical School in Rzeszów in 1994, I started studying at the College of Sciences in Warsaw, the Non-Public Higher School at the Polish Academy of Sciences. After passing three years of study and doing two bachelor classes entitled: a) *Microwave rotational and oscillatory particle spectroscopy* under the advisory of **prof. Z. Kisiel**, and b) *Low-temperature transport properties of PbTe layers grown by MBE method* under the advisory of **prof. G. Grabecki**, I obtained a bachelor's interdisciplinary degree in physics, chemistry and mathematics. Then I continued my master's studies in the topic of physical measuring devices. Simultaneously, I started a student internship in a newly created SIMS laboratory equipped in the new CAMECA IMS6F spectrometer under the advisory of **prof. Adam Barcz**. In addition to the improvement of SIMS measurements, I conducted research on the diffusion and activation of silicon dopant in GaAs. Master's studies were completed with presenting of the master's thesis entitled *Silicon implantation into GaAs*, under the advisory of **prof. Adam Barcz**.

After receiving a master's degree, I have begun Ph. D. studies at the Institute of Physics of the Polish Academy of Sciences in the SIMS laboratory under the supervision of **prof. Adam Barcz**. Simultaneously I started cooperation with the group of Semiconductor Epitaxial Growth at **Institute of Electronic Material Technology (IEMT)**. The aim of the cooperation was studying the GaAs-, InP- and GaN-based semiconductor structures grown by the MOCVD method. Cooperation has led to significant progress in the characterization of semiconductor structures using the SIMS method, which allowed to produce an increasingly better quality structures. It resulted in many publications regarding the properties and parameters of epitaxial growth of materials such as InP [37,38], GaN [39,40], InGaAs [41,42,43].

Cooperation with the laboratory of semiconductor epitaxy, allowed also to conduct my own research on the dopants diffusion in gallium nitride. The result of this research concerning the removing of hydrogen from the GaN:Mg layer resulting in activation of magnesium dopant, was published in the journal *Optica Applicata* **[44]**.

In order to improve the SIMS measurement technique, I completed a two-week internship at the **SIMS laboratory at the Chalmers University of Technology in Gothenburg**. During my stay, I resolved the calibration procedure allowing to determine the composition of the ternary semiconductor compound (Ga,Mn)As.

As part of my Ph.D. studies I conducted research on the dopants diffusion in semiconductor materials. One of the topic of research concerned the oxygen diffusion in silicon crystals. The results were published in the journal *Vacuum* **[45]** and *Semiconductor Science and Technology* **[46]**. However, my main interests concerned the dopants diffusion in III-V materials. The first research topic was the diffusion and activation of silicon in gallium arsenide. As part of these studies, the effect of doping, and thus the Fermi energy level, on Si diffusion in GaAs was observed. The results have been published in the journal *Vacuum* **[47]**. At the same time, I conducted the research resulting from the work on InP-based photodetectors carried out at ITME, concerned the Zn diffusion in InP. In this material, the influence of the annealing conditions on the generation of indium and phosphorus vacancies as well as the concentration-dependent diffusion coefficient were observed. The results were also published in the *Vacuum* journal **[48]**. Another research subject resulting from previous studies was the Si diffusion in gallium nitride. The main conclusion was the influence of the annealing conditions on the generation of Ga vacancies participated in silicon diffusion. The studies were presented at the ICNS-6 conference in Bremen and published in the journal *physica status solidi (c)* **[49]**. The last research topic resulting from the increased interest in dilute magnetic semiconductors was the diffusion of manganese in the gallium arsenide. As in previous studies, the influence of the annealing conditions on the diffusion coefficient was examined, and the concentration-dependent diffusion coefficient was also observed. The results have been published in the journal *Journal of Alloys and Compounds* **[50]**. Interesting research on Mn diffusion in CdTe was conducted in cooperation with **prof. Karczewski** group and have been published in the journal *Thin Solid Films* **[51]**.

As part of the cooperation between the SIMS laboratory and the laboratories of the Institute of Physics Polish Academy of Sciences, I conducted research on dopants and impurities in bulk crystals of ZnO [52,53] (ON 1.1 department under the leadership of **prof. A Mycielski**) and ZnO layers grown by ALD method [54] (ON 4.2 department under the leadership of **prof. M. Godlewski**). In addition, I have established cooperation with leading polish institutes dealing with semiconductor technologies. For the Department of Micro- and Nanotechnology of Wide Bandgap Semiconductors at **Institute of Electron Technology (IET),** under the leadership of **prof. Anna Piotrowska** I performed SIMS characterization supporting the research on ZnO layers [55, 56, 57, 58, 59] and contacts for the wide-bandgap semiconductors [60, 61, 62, 63]. For **High Pressure Research Center (HPRC), Polish Academy of Sciences** I characterized Mg-doped GaN layers grown by MBE method [64].

Also, I conducted research in cooperation with many foreign laboratories. For MBE laboratory at **Physikalisches Institut Universitat Wurzburg**, I characterized (Ga,Mn)As layers [65, 66]. Cooperation with **Lawrence Berkeley National Laboratory** in the study of hydrogen and oxygen impurities in one of the first InN layers obtained by the MBE method resulted in the publication with the largest number of citations in my bibliography (195) [67]. Improvement of SIMS and capacitance-voltage profilometer measurements for studying the diffusion and electrical properties of dopants in GaAs, InP and GaN semiconductors was the main topic of my Ph.D dissertation entitled: *Diffusion mechanisms in A3B5 semiconductors* under the advisory of **prof. A. Barcz** (June 2005) As part of my Ph. D. studies, I also wrote a software for converting raw data obtained from the SIMS apparatus to a format that facilitates their import into the Origin program. I have also written software that simulates diffusion in various thermodynamic conditions and parameters of atomic mobility. The studies for the Ph.D. thesis resulted in two more publications on diffusion of Mn in GaAs and Si in GaN.

7.2 Scientific work after obtaining a Ph. D. degree in physics

After obtaining the Ph. D. degree I continued cooperation with the laboratories of the Institute of Physics PAS as well as with other institutes dealing with semiconductor technologies, i.e. IEMT, IET and HPRC PAS.

I also expanded my cooperation with polish and foreign institutes. A great contribution to the research on doping and impurities of GaN bulk crystals brought the cooperation with Ammono company started in 2007. However, for commercial reasons, no publications were allowed. Similar cooperation was established with the **VIGO** company, for characterization the CdTe-based semiconductor structures.

I continued research on the diffusion and activation of atoms in semiconductors. Due to the increasing attractiveness of dilute magnetic semiconductors (DMS) one of the first steps of my research after Ph.D. was the characterization of Fe diffusion in gallium nitride. The results were presented as a poster on Jaszowiec 2006 and ELTE 2007 conferences.

Another subject of my study closely related to the habilitation thesis was the diffusion of oxygen in gallium nitride. As part of this research, I wrote a proposal for the project **SPIRIT (Support of Public and Industrial Research using Ion Beam Technology)** and I received the contract for research entitled *Oxygen diffusion into GaN from oxygen or argon implanted* Al_2O_3. Within this project, I passed a one-week internship at **Helmholtz-Zentrum Dresden-Rossendorf Ev** to perform oxygen implantation into GaN and Al_2O_3 crystals. The results of research on oxygen diffusion in GaN were published in the journal *Physica Status Solidi C* [68] and presented as a poster presentation on Jaszowiec 2009 and The 3rd International Symposium on Growth of III-Nitrides in Montpellier conferences.

Another research topic carried out under the statute of the Institute of Physics Polish Academy of Sciences and related to the habilitation thesis, was the diffusion of hydrogen in silicon carbide crystals. Differently doped and H-implanted SiC crystals were annealed by the rapid thermal annealing method (RTA). The dependence of the diffusion coefficient on the doping type and oxygen segregation effect was shown. The results were published in *Journal of Apllied Physics* **[69]** and *Nuclear Instruments and Methods in Physics Research B* **[70]**. The studies on As diffusion in zinc oxide crystals annealed under controlled pressure of arsenic vapors resulted in the publication on As properties in ZnO published in *Journal of Alloy and Compounds* **[71]**, and poster presentation on Jaszowiec 2013 conference.

Cooperation with a **prof. Maciej Sawicki** group concerning dilute magnetic semiconductors such as (Ga,Mn)N layers grown by MBE method resulted in interesting results regarding the influence of residual oxygen on Mn diffusion in GaN. These findings have been published in a renowned *Journal of Alloy and Compounds* **[72]** and presented at 34th International Conference on Physics of Semiconductors 2018 in Monpellier and 7th International Symposium on Nitride Growth 2018 in Warsaw.

Simultaneously with the research on the atomic diffusion in semiconductors, I have improved the technique of SIMS measurements and presented the results at conferences on mass spectrometry [73, 74, 75].

Even before the Ph.D. title, I have established cooperation with the **European Organization for Nuclear Research CERN** oriented to silicon-based detector structures. Silicon detectors manufactured at CERN and our own samples were prepared to study thermodynamic properties and oxygen diffusion mechanisms in the silicon crystal. This extensive study resulted in a comprehensive work published in the journal *Surface and Interface Analysis* [76]. In this paper we report on the detection of very low oxygen concentration in silicon by SIMS method. Applying a very high primary Cs+ ion flux, prolonged presputtering, extensive vacuum chamber baking, titanium sublimation pump, and an liquid nitrogen (LN) trap, we have reached a detection limit of ~2×10^{15} at/cm^3 in chemical vapor deposition epitaxial Si films. This value appears to be at least 10 times lower than in any published or unpublished source known to the authors, including the reference sensitivities listed by the instrument manufacturer. Most likely, the key improvement that has allowed us to drive the detection limit to 10^{15} at/cm^3 is the use of an ion pump in the analysis chamber. This paper demonstrates optimized analytical conditions for the oxygen measurements in Si, as a function of depth:

- very shallow profiles are practically impossible to measure accurately because of native oxide at the surface,
- shallow – to – medium range profiles, up to ~20 μm, are the most amenable to SIMS measurements,
- medium – to – deep (~20 - 50 μm) range is required to follow interdiffusion and segregation in epitaxial layers when the oxygen – free layer is grown on a CZ Si substrate,
- extremely deep profiles, up to full thickness of the wafer, definitely necessitate beveling.

Our studies elucidated oxygen indiffusion and outdiffusion during annealing in Ar and in vacuum, noting that the results are qualitatively and quantitatively different for FZ Si as compared with CZ Si. We pointed out that using FZ Si and the indiffusion method provides the most accurate values of the equilibrium oxygen solubility concentration.

For the research in cooperation with laboratories at the **Institute of Physics PAS** the most abundant with respect to the SIMS characterization were studies of the layers grown by the ALD method in the ON4.2 division led by **prof. Marek Godlewski**. Together with scientists from this laboratory, we have published 31 papers. The majority of them concerned zinc oxide and its properties [77, 78, 79, 80, 81]. Published papers were in particular related to the defects [82, 83], dopants [84, 85, 86, 87, 88] and electronic properties [89, 90] of ZnO. The SIMS characterization of zinc oxide layers consisted mainly of the study of impurities, i.e. H or C, and dopants, i.e. N or Al. ZnO-based dilute magnetic semiconductors (DMS) performed by adding Co or Mn elements to the matrix were also widely studied [91]. The research on a such materials concerned the growth [92, 93, 94], optical [95, 96, 97], or electrical and magnetic [98, 99, 100, 101, 102] properties. Characterization by the SIMS method was to examine the amount of embedded magnetic elements, homogeneity of doping and the impurities incorporation. For this purpose, appropriate Co and Mn standards in ZnO were created, as well as the relative sensitivity factor (RSF) coefficients were determined, allowing to calculate the content of both elements with accuracy ±10%.

As part of cooperation with the ALD growth group, the research on the ZnMgO ternary semiconductor [103], HfO_2 as a diffusion barrier of ZnO/Ag junction [104] or $Ti_{1-x}Nb_xO_2$ properties grown by ALD method [105] was also started.

Equally publication-efficient is my cooperation with the Laboratory of Cryogenic and Spintronic Research (SL2) of **prof. Tomasz Dietl**, and currently under the leadership of **prof.**

Maciej Sawicki in cooperation with **Institut für Halbleiter- und Festkörperphysik Johannes Kepler University (JKU) z Linz** under the leadership of **prof. Alberta Bonanni**. Together with colleagues from SL2, I became a co-author of 13 publications, most of which were published in the renowned Physical Review B journal. As part of these studies, structural, electrical magnetic [106, 107] and optical [108] properties of GaN:Fe, (Ga,Fe)N layers grown by MOCVD, as well as the effect of Mg doping on epitaxial growth [109] were characterized. Similar studies were conducted for GaN:Mn and (Ga,Mn)N layers [110, 111, 112]. In cooperation of Institute of Physics PAS, JKU and **Institute of Solid State Physics University of Bremen** groups, magnetic layers (Ga,Mn)N grown by MBE were also studied [113, 114]. One of the papers [115] was awarded of the director's award for the best paper of Institute of Physics PAS of 2016 year. The scope of the research also included the Mg-doped GaN layers [116, 117]. A part of the research employing SIMS measurements consisted in determining the content of both the magnetic elements Fe and Mn, as well as the dopants, i.e. Mg and Si, but also the H and O impurities.

Similarly, as for the ZnO, appropriate Mn and Fe in GaN standards were developed, allowing determination of the atomic content of these elements in GaN layers up 10 at%. SIMS-characterized manganese-doped layers brought insights into the measurement artifacts associated with the Mn depth profiles, presented at the conference SIMS Europe 2016 [118].

Another scientific group from the Institute of Physics PAS, I had a close cooperation for a long time, is the ON1.1 group under the leadership of **prof. Andrzej Mycielski**. The group mainly deal with the growth of CdTe based bulk crystals. In addition to testing the purity of the elements used to the crystal growth, my part of the study also included checking the homogeneity and the stoichiometry, as well as doping level of the crystals. The scope of publications with colleagues from the ON1.1 group refers to such subject as: fabrication of contacts for semi-insulating crystals CdMnTe:V [119, 120, 121], CdMnTe and CdMgTe based X-ray detectors [122, 123], growth and annealing of CdMnTe bulk crystals [124, 125] and properties of Se doped CdTe bulk crystals [126]. The ZnO bulk crystals were also characterized [127, 128].

As part of the statutory research of the Institute of Physics PAS, I also collaborated with ON1.2 group on molecular beam epitaxy growth the CdTe-based layers and crystals under the leadership of **prof. Tomasz Story** and ON4.7 group dealing with molecular beam epitaxy of ZnO of **prof. Adrian Kozanecki**. The scope of research in cooperation with the ON1.2 group mainly involved the GeMnTe/BaF_2 layer characterization [129, 130, 131, 132]. In the ON1.2

group I worked together with **dr. hab. Ewa Przeździecka** regarding the characterization of As and Sb [133, 134, 135, 136, 137] or N [138, 139] doped ZnO layers grown by MBE method.
The continuation of research on the Mn diffusion in GaAs, started as part of my Ph. D. thesis, was conducted with a SL1.2 research group of which I am a member, headed by **prof. Krystyna Ławniczak-Jabłońska**. Within the EAGLE project, we investigated the local properties of Mn atoms in GaAs crystals [140, 141, 142].
Other studies I took part in were: behavior of GaMnSb/GaSb annealed at the elevated temperatures [143], properties of In/PbTe junction [144], properties of GaN/Si grown by MBE method [145], structure of GaN/AlGaN superlattice [146], Co diffusion in $Gd_3Ga_5O_{12}$ crystals [147] and properties of (Ga,Mn)As and (Ga,Mn)(Bi,As) layers [148, 149].
After my Ph. D. thesis, I also continued cooperation with other Polish institutes dealing with growth and crystal characterization. The largest number of projects, and thus publications, resulted from joint projects with Department of Micro- and Nanotechnology of Wide Bandgap Semiconductors led by **prof. Anna Piotrowska** and Department of Photonics under the leadership of **prof. Maciej Bugajski,** both from **Institute of Electron Technology (IET).** Research in cooperation with prof. Piotrowska group mainly relied on the characterization of ZnO layers deposited by magnetron sputtering [150, 151]. Published research results concerned: characterization of ZnO and ZnMnO layers produced by oxidation of Zn(Mn)Te [152, 153, 154], ZnO doping by N, As or Sb elements [155, 156, 157] or ZnO doping by group 1B elements [158]. Other works devoted to the electric contacts for wide band-gap semiconductor eg. Ti-Al-N [159, 160, 161] or Ni-Si [162]. The research conducted together with the Department of Photonics concerned mainly the characterization of layers and devices based on III-V semiconductors grown by the MBE method [163, 164, 165, 166, 167, 168, 169, 170]. For research groups from IET, I also characterized materials such as Si:Mn [171], GaMnAs [172] and TaO_2 [173].
After receiving the Ph.D. degree, I also continued cooperation with the **Institute of Electronic Materials Technology (IEMT)** for the characterization of semiconductor layers based on GaAs, InP, GaN and SiC grown by the MOVPE method [174]. The most fruitful cooperation was brought with **dr. Katarzyną Racką-Dzietko** concerning the characterization of bulk SiC crystals obtained by Physical Vapor Transport (PVT) method [175, 176, 177, 178, 179, 180, 181].
As for the cooperation with the **High Pressure Research Center PAS** I took part in characterization of GaN materials manufactured by either MOVPE method (**prof. Tadeusz Suski)** [182, 183, 184, 185, 186] or using a novel Hydride Vapor Phase Epitaxy (HVPE) [187,

188, 189]. Properties of GaN crystals grown by Sublimation Sandwich Method (SSM) were also studied [190, 191].

For completeness the list below comprises other studies of materials together with cooperating institutions and publications:

- (Ga,Mn)N layers [192, 193, 194] – **Institute of Solid State Physics University of Bremen**,
- (Ga,Mn)As [195, 196] and (In,Fe)As [197] layers – **Helmholtz-Zentrum Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research**,
- InSe:Mn [198, 199] and ZnO [200] – **I. M. Frantsevich Institute for Problems of Materials Science National Academy of Sciences of Ukraine**,
- HgCdTe:As [201, 202, 203, 204] – **National Research Tomsk State University,**
- hydrogen diffusion in $LiNbO_3$ [205] and Mn in $YAlO_3$ [206] – **Lviv Polytechnic National University**
- (Ga,Mn)As [207, 208, 209] – **Department of Physics University of Notre Dame**,
- SnTe:In [210] and InAlAs/InAs:Mn structures [211] – **Institut fur Experimentelle und Angewandte Physik Universitat Regensburg**
- GaN:Si/Al_2O_3 [212] – **V. Lashkaryov Institute of Semiconductor Physics, National Academy of Sciences of Ukraine**.

Currently I am conducting a statutory research on Na diffusion and activation in zinc oxide crystals. First results from the innovative method of introducing sodium into ZnO crystals from aqueous NaCl solution at temperatures up to 300°C were presented at the 10th International Workshop on Zinc Oxide and Other Oxide Semiconductors conference in Warsaw [213]. While within the TECHMAGSTRATEG grant received by the Institute of High Pressure Physics PAS, I characterize Be diffusion mechanisms in GaN crystals.

8. Presentation of teaching and organizational achievements as well as achievements in popularization of science or art.

   8.1 Creative professional works:

   - Coauthor of the book: Crystal Growth: Theory, Mechanism and Morphology, Chapter 9: MBE Growth of Type-II InAs/GaSb Superlattices on GaSb Buffer – A. Jasik, I. Sankowska, K. Regiński, E. MAchowska-Podsiadło, A. Wawro, M. Wzorek, R. Kruszka, R. Jakieła, J. Kubacka-Traczyk, M. Motyka, J. Kaniewski

- Internship in the SIMS laboratory in Chalmers Institute in Goteborg, Sweden – determination of the composition of AlGaN and MnGaAs layers by the SIMS method
- Stay in Helmholtz-Zentrum Dresden – Rossendorf e.V., Germany according to project of European Community as an Integrating Activity Support of Public and Industrial Research Using Ion Beam Technology (SPIRIT) in order to carry out the research entitled *Oxygen diffusion into GaN from oxygen or argon implanted* Al_2O_3
- 6 seminars, including 5 after obtaining Ph.D. degree
- 155 conference presentation:
  - 2 coauthors of invited talk,
  - 15 oral presentations (2 presented personally), including 11 after obtaining Ph.D. degree (1 presented personally)
  - 138 poster presentations (23 presented personally), including 109 after obtaining Ph.D. degree (15 presented personally)

8.2 Managing and Participation in Domestic or International Research Projects before obtaining Ph.D. degree

- 2001 – 2002, **principal investigator** in the government project KBN 7 11TB 001 21 entitled *Si and Zn diffusion in InP* performed in the Institute of Electronic Material Technology

8.3 Managing and Participation in Domestic or International Research Projects after obtaining Ph.D. degree

- 1996 – 1999, **investigator** in the government project KBN 8T11B08210 managed by prof. A. Barcz, entitled: *Study of thermodynamic properties of GaAs*
- 2000 – 2003, **investigator** in the European Commission project IN LINE SIMS IST-01-6-2B
- 2001 – 2005, **investigator** in the government project KBN 7T08A06021 managed by prof. A. Barcz, entitled: *Oxygen transport kinetics in the SiO2/Si FZ system*
- 2008 – 2011 **investigator** in the government project MNiSW N507436834 managed by prof. A. Barcz, entitled: *Characterization of materials and SIC based structures - SIMS, DLTS, I(C)-V*
- 2009 – 2013, **investigator** in the project of ERC Advanced Grant Research – UE-27/1 managed by prof. T. Dietl, entitled: *Functionalisation of Diluted Magnetic Semiconductors* FunDMS

- 2010, **principal investigator** of contract 227012 in the project SPIRIT (Support of Public and Industrial Research using Ion Beam Technology) entitled: *Oxygen diffusion into GaN from oxygen or argon implanted* Al_2O_3
- 2012 – 2017, **investigator** in the government project P-376/M managed by prof. T. Dietl, entitled: *Quantum phase transitions in magnetic layers controlled by an external electric field*
- 2014 – 2017, **investigator** in the government NCN project P-418/O managed by prof. T. Dietl, entitled: *Tunneling electrons and Cooper's pairs in quantum structures containing a ferromagnetic insulator (Ga,Mn)N*
- 2016 – 2018, **investigator** in the government project managed by prof. Andrzej Mycielski, entitled: *Insulating crystals (Cd,Mn)Te: Exploration and description of spatial carrier transport and compensation conditions*
- 2018 – now, **investigator** in the government project TECHMATSTRATEG 1/346720/8/NCBR/2017 managed by prof. Andrzej Mycielski, entitled: *Technology for the production of materials and structures for the detection of X and gamma radiation, using low defect homogeneous crystals (Cd,Mn)Te, with high resistance to defect generation as a result of irradiation*
- 2019 – 2022, **investigator** in the government project NCN Opus 15 nr 2018/29/B/ST5/00338 managed by prof. Michał Boćkowski, entitled: A*cceptors implantation into intentional undoped GaN layers grown by the HVPE method - structures that are the basis of vertical high power transistors*

8.4 List of invited papers delivered at national or international conferences:

- E. Kamińska, E. Przeździecka, A. Piotrowska, J. Kossut, E. Dynowska, A. Barcz, **R. Jakieła**, W. Dobrowolski, I. Pasternak, P. Bogusławski – *p-type doping in ZnO - invited lecture* – XXXV International School on the Physics of Semiconducting Compounds, Jaszowiec 2006
- Andrzej Mycielski, M. Witkowska-Baran, D. Kochanowska, A. Szadkowski, B. Witkowska, W. Kaliszek, **R. Jakiela**, V. Domukhovski – *(Cd,Mn)Te as a New Material for X-ray and Gamma-ray Detectors* – MRS 2007 Fall Meeting, Boston

8.5 Other papers personally delivered at national or international conferences:

- **R. Jakieła**, A. Jasik, W. Strupiński, K. Góra, K. Kosiel, M. Wesołowski – *Zależność koncentracji wodoru w warstwach epitaksjalnych GaN:Mg od sposobu ich* *wygrzewania – charakteryzacja metodą SIMS* – IX Seminarium: Powierzchnia and Struktury Cienkowarstwowe, Szklarska Poręba 2002

- **R. Jakiela**, A. Barcz, J. Sarnecki, G. K. Celler – *Ultra-high Sensitivity SIMS Analysis of Oxygen in Silicon* – 21st International Conference on SIMS, Kraków 2017

8.6 Seminars presented:

- *Charakteryzacja struktur półprzewodnikowych metodą SIMS* – Seminarium Fizyki Ciała Stałego Wydziału Przyrodniczego WSP Rzeszów, 2000
- *SIMS ? .. seems good !* – seminar in Ammono company, 2008
- *Spektrometria Masowa Jonów Wtórnych* – Seminarium Rentgenowskie IFPAN – 2008
- *Transport atomowy w ciele stałym* (2 seminars) – Konwersatorium ITME – 2008
- *Dyfuzja powierzchniowa* – Konwersatorium ITME – 2008
- *Spektrometria Masowa Jonów Wtórnych – podstawy and zastosowanie w badaniach struktur półprzewodnikowych* – Seminarium Centrum Dydaktyczno-Naukowe Mikroelektroniki and Nanotechnologii Uniwersytetu Rzeszowskiego – 2014

8.7 Construction of a unique science equipment

- Service and maintenance of the secondary ion mass spectrometer – CAMECA IMS 6F
- Adaptation of Capacitance-Voltage profilometer BIORAD PN4300 for the wide band gap semiconductors

8.8 Organization of conferences, reviewing and editorial activities

- Reviewer of article entitled *P-type single-crystalline ZnO films obtained by (P,O) dual implantation through dynamic annealing process* for journal **Superlattices and Mictrostructures 2018**
- Reviewer of article entitled *The effect of alpha particle irradiation on electrical properties and defects of ZnO thin films prepared by sol-gel spin coating* for journal **Material Science in Semiconductor Processing 2018**
- Reviewer of article entitled *Structural and Optical Characteristics Investigations in Oxygen Ion Implanted GaN Epitaxial layers* for journal **Material Science in Semiconductor Procesing 2019**

8.9 Didactics and popularization of science

- Presentation of the SIMS laboratory on an annual *Festiwal Nauki*
- Presentation of the SIMS laboratory on an annual Dni Otwarte Instytutu Fizyki PAN
- Physics teacher in first class of high school with a biological - chemical profile
- Notes in the science section of blog page Salon24.pl:
  - *Mercury - a rare possibility of observation.*
  - *Semiconductor blue laser structure for lay people*
  - *Two-dimensional electron gas in a field effect transistor*

8.10 Patents and patent applications

- patent co-author: PL218567, obtained on 20.05.2014 – *A method for producing semiconductor wafers from CdTe* – **Rafał Jakieła**, Zbigniew Gołacki
- patent co-author: PL215095, obtained on 15.04.2013 – *The method of producing heterostructures n-(Eu,Gd)Te/p-PbTe and heterostructure to generate random numbers* – Mąkosa Andrzej, Wrotek Sylwia, Dziawa Piotr, Osinnij Victor, Taliashvili Badri, Tkaczyk Zbigniew, **Jakieła Rafał**, Figielski Tadeusz, Wosiński Tadeusz, Story Tomasz

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[7] **R. Jakiela**, A. Barcz, J. Sarnecki, G. K. Celler - *Ultra-high Sensitivity SIMS Analysis of Oxygen in Silicon* – oral presentation on SIMS 2017 conference in Kraków
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