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Physics

The Role of Nitride Materials for Biological Applications (Biosensors)

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Original Research Article

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Abstract: Nitride materials are very good candidates in various fields of electronics, optoelectronics (transistors, diodes…) and military technology (radars, communications, and others). Using HEMT (high electron mobilit y transistor) based heterostructures (AlGaN / GaN), it is possible to produce biological, chemical and medical sensors. The principle of operation relies on the properties of high mobility of two-dimensional electron gas (2DEG) and high saturation velocity. The 2DEG channel in AlGaN HEMT is very close to the surface (gate), rendering it very sensitive to the absorption of different analytes like (glucose, protein, DNA, bacteria). In this work, we focus on the application of AlGaN HEMT biosensors. We show the dependence of (2DEG density on multiple parameters), and we attempt to use this principle to detect very low glucose concentrations.
Keywords: Semiconductor III-N, AlGaN/GaN heterostructures, GOx, sensor, 2DEG.

INTRODUCTION

The nitrides of group III elements are formed by the combination of a group III element (Ga, In, Al, B) and group V nitrogen (N), according to Mendeleev's periodic table of elements (Fig 1). Ternary and quaternary alloys are also defined as nitrides, particularly GaN, AlN, InN and AlGaN, InGaN. These are materials widely used in the world of optoelectronics because they have a relatively high band gap compared to other semiconductors (especially in the case of GaN and AlN), which can generate light in the visible spectrum and ultraviolet (Fig 2). Therefore, they open a wide range of possibilities for optical, electronics and biological… applications.

The wurtzite nitrides of group III elements are chemically stable semiconductors with high spontaneous internal and piezoelectric polarization, rendering these materials very suitable for the production of robust and highly sensitive sensors for the detection of ions, gases and analytes [1].

In such devices the strong discontinuity of polarization at the AlGaN/GaN interface results in a two-dimensional electron gas (2DEG) forming the conducting channel of a transistor with high electron mobility (HEMT). The conductivity of the channel is modulated by changes in the potential outside the HEMT. This effect is used in metallized gate devices to detect charge loads and molecules adsorbed on the exposed area of the gate [2,3]. Consequently, AlGaN/GaN HEMT devices are used as detectors of biochemical and biological processes [4].

Fig-1: Extract of the periodic table of elements

The density of two dimensional electron gas (2DEG) may ultimately be detected by electrical measurements. Thus, many biosensors were carried out successfully and applied by using the conduction channel (2DEG) of AlGaN/GaN as detection element and in our work we use this principle of operation for the detection of minute glucose concentrations [5-6].

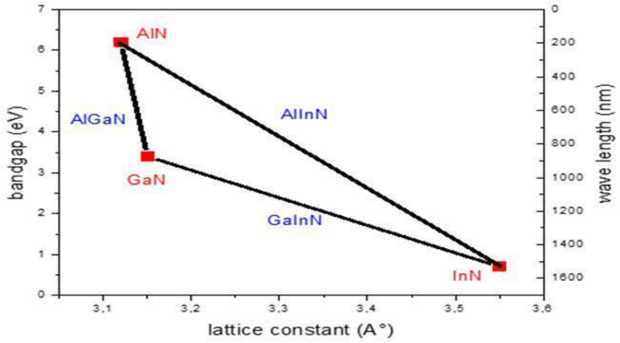

Fig-2: Bandgap of hexagonal InN, GaN, AlN and their alloys versus lattice constant and wave length

Biosensors of glucose were largely studied and developed since Clark and Lyons first proposed the concept of glucose enzyme electrodes in 1962 [15]. Three generations of glucose sensors have been developed based on electron transfer in the electrode by the mediator of electron transfer between oxygen and glucose in the enzyme glucose oxidase (GOx). Because of the importance of glucose immobilization, many studies have focused on glucose immobilization techniques using carbon nanotubes and gold particles.

Theoretical studies

HEMT

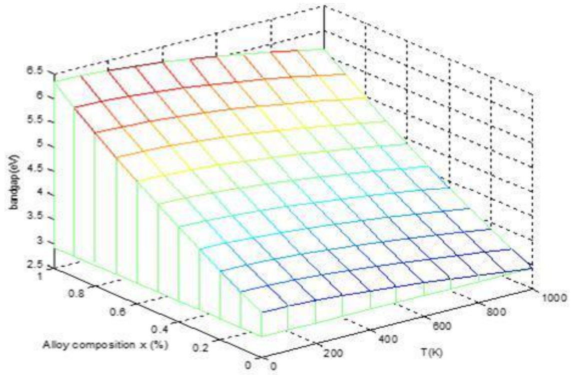

Fig-3: bandgap of $Al_xGa_{1-x}N$ versus Al content x and temperature

AlGaN/ GaN High Electron Mobility Transistors (HEMT) have become recently an interesting topic of research because of their advantages, including a wide bandgap. In Figure 3, we show the energy of conduction band for AlGaN in relation to temperature (T) and concentration of Al(x). The breakdown field of this alloy is high, favoring its application in high power, high frequency and high voltage devices [7-8].

HEMT (fig 4) are the components having known today the greatest evolution. In the HEMT, current transfer is achieved by the formation of electrons (2DEG) of the heterojunction that will play the role of a channel.

The heterojunction allows separation of the ionized doped donor layer from free electrons, the electrons are then trapped in a channel (2DEG), and they can reach high mobility atoms. The name HEMT (High Electron Mobility Transistor) comes from this property.

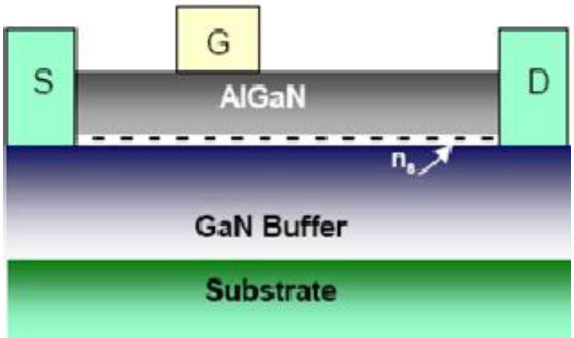

Fig-4: Schematic HEMT AlGaN / GaN device

Biosensors of glucose were largely studied and developed since Clark and Lyon proposed the first time the electrode of enzymes of glucose [15]. Three generations of glucose sensors have been developed and based on the electron transfer in the electrode by the mediator between oxygen and glucose oxidase GOx. Because of the importance of the immobilization of glucose, many studies have focused on the techniques of the immobilization of glucose with carbon nanotubes and gold particles.

2DEG transport properties

Two dimensional electron gas (2DEG) is also referred to as quantum well and it is formed at the heterointerface of wideband gap material, when it is grown over narrow band gap material. This quantum well is formed at the heterointerface due to the conduction band discontinuity and its density was studied by the solution of Poisson's equation [9]. Growth of AlGaN material above GaN provides a bias field of the spontaneous polarization and piezoelectric polarization [10].

$$\frac{d}{dz}D(z) = \frac{d}{dz}\left(-\varepsilon(z)\frac{d}{dz}V(z) + P(z)\right) = e(P(z) - n(z) + N_D^+ - N_A^-) \quad (1)$$

Where
D (z) : electric displacement
ε(z) is the position dependent dielectric constant;
$V(z)$ is the electrostatic potential
P(z) is the total polarization including spontaneous and piezoelectric polarization;
n(z) , *p(z)* are the electron and hole carrier concentrations respectively;
N^+_D ,N^-_A : the ionized donor and acceptor concentrations respectively. [9]
And the total polarization is then given by [11]

$$\sigma(x) = [P_{sp}(GaN) + P_{pz}(GaN)] - [P_{sp}(Al_xGa_{1-x}N) + P_{pz}(Al_xGa_{1-x}N)] \quad (2)$$

$P_{sp}(GaN), P_{sp}(Al_xGa_{1-x}N)$:Spontaneous polarization of GaN and AlGaN, respectively
$P_{pz}(GaN), P_{pz}(Al_xGa_{1-x}N)$: Piezoelectric polarizations of AlGaN and GaN respectively

2DEG density of a HEMT of AlGaN / GaN can be written as

$$n_s(x) = \frac{+\sigma(x)}{e} - \left(\frac{\varepsilon_0\varepsilon(x)}{de^2}\right)[e\phi_b(x) + E_F(x) - \Delta E_c(x)] \quad (3)$$

where n_s is the 2DEG density shown in figure 8.
ΔE_c:the conduction band offset at the AlGaN/GaN interface.
d : the width of the AlGaN
ε_0 : the permittivity of the vacuum is equal to 854.10^{-14} ($F.cm^{-1}$) and $\varepsilon(x)$ that of AlGaN
ϕ_b : The height of the barrier of AlGaN
The final expression for σ is

$$\sigma(x) = P_{sp}(GaN) - P_{sp}(Al_xGa_{1-x}N) - 2\left(\frac{a(0)-a(x)}{a(x)}\right) * (e_{31}(x) - \frac{e_{33}(x)c_{13}(x)}{c_{33}(x)} \quad (4)$$

c_{13}, c_{33}: elastic constants:
e_{31}, e_{33} : piezoelectric constants

EXPERIMENTAL STUDY

Sensor Fabrication

The HEMT (fig 5) structure consists of a 3 µm thick undoped GaN buffer, 30 Å thick $Al_{0.3}Ga_{0.7}N$ spacer, and 220-Å-thick silicon-doped $Al_{0.3}Ga_{0.7}N$ cap layer. Epilayers are grown by metal organic chemical vapor deposition on thick GaN buffers on sapphire substrates.

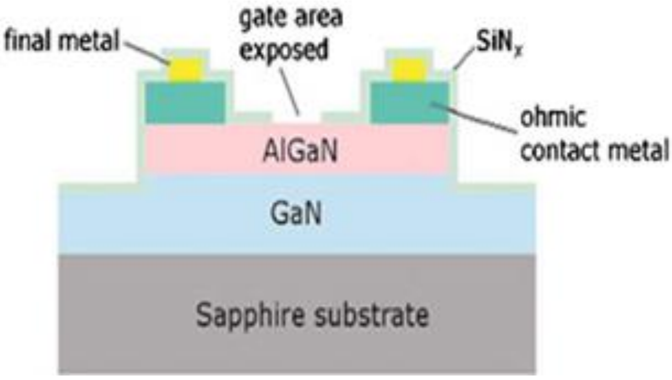

Fig-5: schematic of gateless HEMT used as a sensor for biological molecule introduced to the gate region

ZnO Nanorod Growth for Glucose Sensing. The glucose sensor was achieved with selective-area ZnO nanorod growth on the gate area of the glucose sensing HEMT. By incorporating the nanorods on the HEMT gate sensing area, the total sensing area and sensitivity could increase significantly. ZnO nanocrystals, prepared with the Pacholski method [12] were coated on the gate area of the glucose sensor. The ZnO nanorods were subsequently grown in a solution of 20 mM zinc acetate hexahydrate $[Zn(NO_3)_2 \cdot {}_6H_2O]$ and 20 mM hexamethylene triamine ($C_6H_{12}N_4$). Subsequently, the sample was removed from solution, rinsed thoroughly with acetone, followed by deionized water to remove any residual salts, and air dried at room temperature. A schematic device cross sectional view of the HEMT sensor is shown in Figure 6 [13].

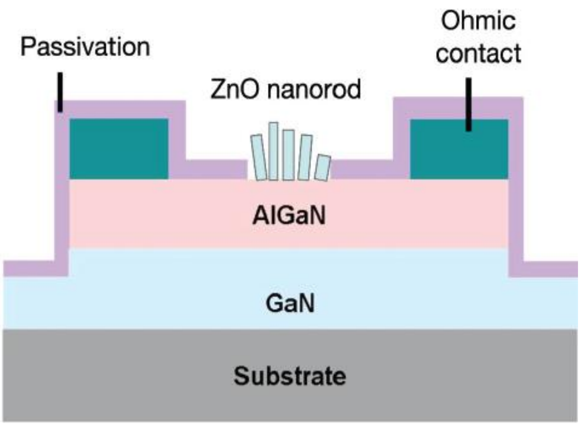

Fig-6: Schematic of ZnO nanorod gated AlGaN/GaN HEMT

Preparation of the medium to be detected

The GO*x* solution was prepared with concentration of 10 mg/ml in 10 mM phosphate buffer saline (*p*H value of 7.4., Sigma Aldrich). After fabricating the device, 5 µl GO*x* (~100 U/mg, Sigma Aldrich) solution was introduced to the surface of the HEMT. The HEMT was kept at 4°C in the solution for 48 h for GO*x* immobilization on the ZnO nanorod arrays followed by an extensively washing to remove the unimmobilized GO*x*. The HEMT device was kept in the incubator for 30 min to make the enzyme active around 37°C. The target glucose was applied to the device through a syringe autopipette (2–20 µl). The current-voltage characteristics were measured using an Agilent 4156C parameter analyzer with the gate region exposed [15].

RESULTS AND DISCUSSION

We were interested in the realization of sensors for the detection of glucose. The microstructure of ZnO nanorod-gated AlGaN/GaN HEMT biosensors for electrical detection of glucose, as revealed by scanning electron microscopy is shown in Figure 7. Glucose was specifically recognized by GOx anchored in the gate area (ZnO) (immobilized GOx has a hugeh affinity for glucose) due to electrostatic interactions between positively charged ZnO nanorods and negatively charged GOx. We investigated a wide range of concentrations from 0.5 nM to 125 µM and obtained an experimental limit of detection of 0.5 nM, which is the lowest value reported to date [15].

2DEG density, as reflected in the drain current of HEMT sensors, is increasingly used in amperometric measurements of glucose concentration. The change in current density in the operational zone of GOx / ZnO is related to the concentration of glucose. The gate polarization electrode is connected to the gate of the AlGaN / GaN HEMT via a saline bridge, so that the control channel (2DEG) of AlGaN/ GaN HEMT detects changes in induced spontaneous and piezoelectric polarization.

Detection of glucose is due to the reaction between glucose and the electrode GOx / ZnO, which leads to a transfer of electrons to the ZnO nanorods. Therefore, the distribution of ZnO nanorods plays a very important role in setting the response time of the HEMT, the detection sensor [14].

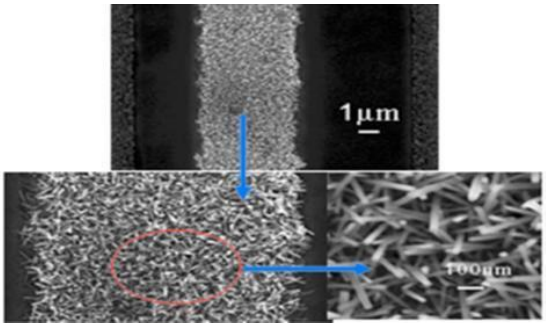

Fig-7: Scanning electron microscopy image of ZnO nanorod gated AlGaN/GaN HEMT [14]

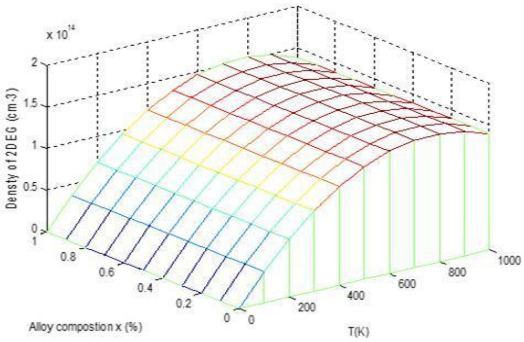

Fig-8: Variation of 2DEG density as a fonction Al content x and temperature T

Figure 9 shows a real-time glucose detection experiment in PBS buffer solution using the drain current change with a constant bias of 250 mV. No current change could be detected upon addition of buffer solution after approximately 200 s, showing the specificity and stability of the device. By sharp contrast, the current change showed a rapid response in less than 5 s when glucose was added to the surface. So far, glucose detection using ZnO nanorod with immobilized GOx is based on electrochemical measurement. Since a reference Pt electrode is required in the solution. The current density is measured with a fixed potential applied between nanomaterials and the reference electrode as shown in Figure10.

Glucose sensing was achieved via a change in the drain current of HEMT induced by a change of the surface charge of the ZnO nanorods and the detection signal was amplified by the HEMT. Although the response of the HEMT based sensor is similar to that of electrochemical based sensors, a far lower detection limit of 0.5 nM was achieved for the HEMT based sensor due to this amplification effect.

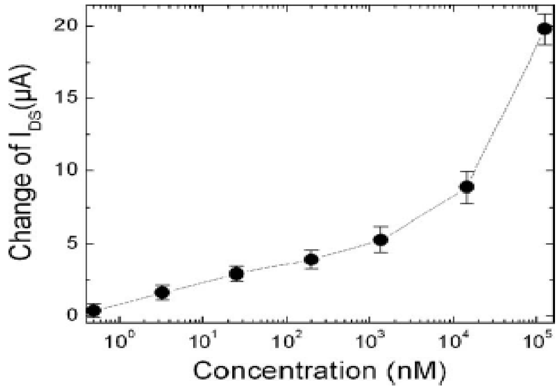

Fig-9: Plot of change of drain current as a function of glucose concentrations from 500 pM to 125 μM in 10 mM phosphate buffer saline with a pHvalue of 7.4 [15]

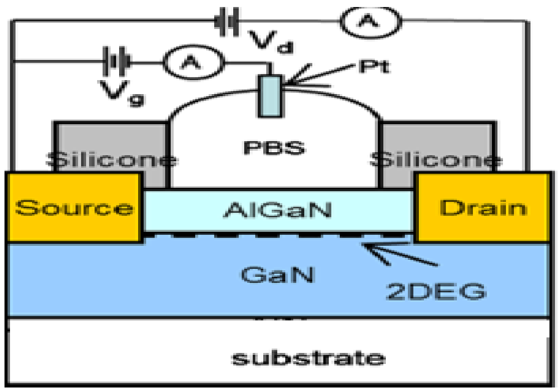

Fig-10: The cross-sectional view and device configurationof AlGaN/GaN HFET biosensors [16]

The glucose sensing was measured through the drain current of HEMT with a change of the charges on the ZnO nanorods and the detection signal was amplified through the HEMT. Although the response of the HEMT based sensor is similar to electrochemical based sensor, much lower detection limit of 0.5 n*M* was achieved for the HEMT based sensor due to this amplification effect.

CONCLUSION

In this work, we demonstrated that the HEMTs AlGaN / GaN show a strong dependence with 2DEGs caused by spontaneous and piezoelectric polarization on changes in electrostatic boundary conditions of the free surface above the 2DEG due to the presence of analytes such as glucose, protein,DNA, This principle gives characteristics particular to the heterostructures of AlGaN/GaN allowing these sensors to operate with an excellent chemical and mechanical stability also a high sensitivity of glucose detection which is the element selected in our study. What makes nitride HEMTs excellent candidates for biological sensor and other applications in the medical field.

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