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INITIAL STUDY OF GaN THIN FILMS FOR PHOTOCATHODES PREPARED BY MAGNETRON SPUTTERING ON COPPER SUBSTRATES

C. Wang, M. Vogel†, X. Jiang, University of Siegen, IfWT, Siegen, Germany
J. Schaber, P. Murcek, R. Xiang, HZDR, Dresden, Germany

Abstract

On the path for high brightness electron beams, Gallium Nitride (GaN) is one promising candidate for a photocathode material. In this contribution, we report on the continuation of the study to optimize the crystallization quality and crystallography of Mg doped GaN samples on copper substrates which are synthesized by RF magnetron sputtering.

SEM and XRD results show that the pretreatment methods and the sputtering conditions (temperature, sputtering power and partial pressure of the reactive gas) can both affect the morphology and crystal quality of GaN films. The initial QE measurements of these samples are done in our newly build in-situ QE measurement system and first results of QE analyses done at Helmholtz-Zentrum Dresden-Rossendorf (HZDR) are presented in a dedicated contribution.

INTRODUCTION

High performance photocathodes are required as high brightness electron sources for particle accelerators, free-electron lasers (FEL), and synchrotron- and THz radiation sources. GaN based photocathodes could be a possible solution because of its high quantum efficiency (QE), high thermal stability and low dark current [1-3]. GaN is a III-V semiconductor material with a wide band gap of 3.4 eV. By means of a Cs/O activation process, a negative electron affinity (NEA) at the surface can be produced and more than 10% QE can be achieved [4-6]. The typical preparation methods for GaN thin films are metalorganic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE), which require high reaction temperatures (> 1000 °C) for epitaxial growth of single crystal GaN on dielectric substrates such as sapphire [6]. In this contribution, we continue to study the feasibility of polycrystalline GaN films as a photocathode material by using rf magnetron-sputtering on copper. The use of metallic instead of dielectric substrates bears some important advantages such as thermal stability, no electron depletion and clean installation.

For a photocathode, it is necessary to produce p-type GaN by Mg doping to form the downward band bending around the contact surface which can produce an effective NEA surface and increase the escape rate of excited electrons [5]. But due to the growth method of GaN, donor impurities such as N and O vacancies are "unintentionally" formed leading to n-type doping with a background carrier concentration of $n\times10^{18}$-10^{19}/cm^3 for RF magnetron-sputtering [7-9].

† michael.vogel@uni-siegen.de

The Mg ionization energy in GaN is about 223 meV [10], i.e. only a few percent of Mg acceptors can be activated and form hole carriers. This also indicates, that a heavy doping level is necessary to prepare a p-type GaN:Mg. However, a high Mg concentration will also increase lattice defects, which results in a shorter electron diffusion length. Consequently, Mg concentration control is essential.

In this contribution, we deposited p-type GaN:Mg films on OFHC copper substrates by means of rf magnetron-sputtering. The main influencing factors that affect the morphology and crystal quality of the GaN films are studied, and initial QE measurements are performed.

EXPERIMENT

After the pretreatment (electropolishing [11], hydrogen plasma etching or in-situ ion-gun cleaning) of the polycrystalline copper substrates, Mg doped GaN films are deposited by rf magnetron-sputtering. Three of four sputtering cathodes are used in the coating system. Two of them are high purity GaN (6 N) and the other one is Mg (10 at.%) - GaN (4 N). The Mg content is modified by controlling the sputtering power of all tree targets, with the total power of 100 W, sputtering time of 60 min and sputtering temperature of 600 °C. The sputtering atmosphere is a mixture of N_2 (70%) and Ar (30%) with a total gas flow of 100 sccm. The total pressure is maintained at 8.0×10^{-3} mbar. The base pressure of the system is in the range of 10^{-8} mbar.

After deposition, the sample is transferred to the quantum efficiency (QE) measurement system in vacuum. Here the sample is tilted by 60° to be perpendicular to the incident light. Then the sample is activated by Cs utilizing a dispenser (SAES GETTERS S.p.A., Italy) filled with Caesium chromate (Cs_2CrO_4) which is reduced to Cs during heat up to temperatures above 550 °C. The photoelectrons are collected by a ring anode which sits 20 mm away from the sample. The system has been reported in a previous contribution [12]. For the activation and initial measurement, the bias voltage of the anode is set to 150 V and the background pressure increases to about 1.5×10^{-7} mbar due to the Cs evaporation. In addition, a GaN sample was measured after surface cleaning and heat treatment at 350 °C in ultra-high vacuum in a measurement system at HZDR.

The morphology and texture of GaN films were measured by SEM and XRD. Samples prepared by different pretreatment methods and the same sputtering conditions are analyzed for comparison.

RESULTS AND DISCUSSION

As shown in Fig. 1, the measured GaN film thickness is about 350 nm. Also shown is the surface morphology of GaN films grown on copper substrates prepared by

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Table 1: Crystal Analyses of GaN Films with Different Mg Contents and Pretreatment Methods

Pretreatment	Mg (at.%)	Peak Position (10-11) 2θ (°)	FWHM (°)	Grain Size (nm)
Electropolishing	0	36.71	0.3550	24.62
	4	36.64	0.3224	27.10
Hydrogen plasma cleaning	0	36.70	0.3622	24.12
	4	36.63	0.3395	25.73

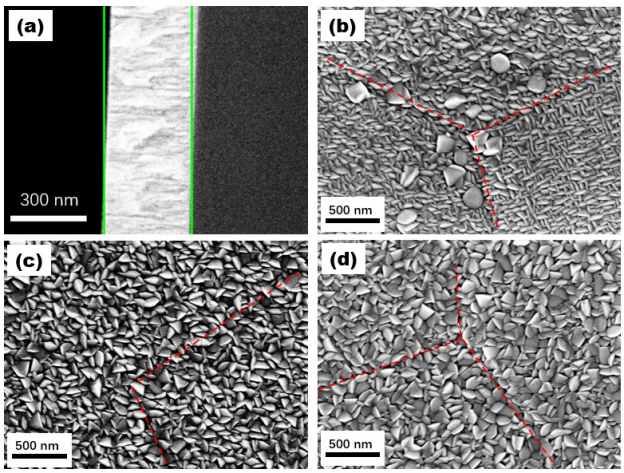

Figure 1: SEM images of GaN samples of (a) a cross sectional view on Si, and surface view of GaN on copper pretreated by (b) hydrogen plasma cleaning, (c) electropolishing, (d) ion-gun cleaning.

different pretreatment methods. The red lines in the SEM picture represent the grain boundary of the underlaying copper substrate. The results show that the GaN film has different arrangements on the substrate pretreated by hydrogen plasma (Fig. 1 b). The lower corner on the right shows the typical ordered morphology of a heteroepitaxial grown thin film. More in-depth studies are in progress. Through electropolishing and in-situ ion-gun cleaning pretreatment, GaN films grown on different copper crystals exhibit uniform morphologies.

As shown in Fig. 2 and Table 1, all GaN films exhibit a wurtzite structure with no particular texture and a predominant refraction peak at $(10\bar{1}1)$. In Fig. 2 the $(10\bar{1}1)$ peak positions of undoped GaN films are 36.71° and 36.70° for the upper blue line and lower black line, respectively. This is close to the reference powder pattern of 36.73° [13]. With 4% Mg content, the peak position shifts to the left-hand side (36.64° and 36.63°) indicating a larger lattice parameter. The substitution of larger Mg^{2+} ions [r (Mg^{2+}) = 0.72 Å] for smaller Ga^{3+} ions [r (Ga^{3+}) = 0.62 Å] in GaN may cause this lattice expansion, and result in peak shifts [14]. In addition to the peak shift, the decrease of the (0002) peak is also obvious, indicating that the texture of GaN film has changed. The crystallite size of all tested samples is around 25 nm, which is calculated by the Scherrer equation (K = 0.94).

Figure 3 shows the initial in-situ QE test results of a GaN sample with a Mg content of 0.5 at.%. The blue curve is the power of the monochromatic light source measured by a photodiode. The experiment was carried out with a Xenon light source, which results in a very low power output of light with short wavelength under 400 nm. The red curve represents the measured photocurrent. The QE curve increases rapidly around 300 nm (4.1 eV) and reaches its peak at 230 nm (5.4 eV).

In Fig. 4 the QE measurement result of a sample with 3.5 at.% Mg content measured at HZDR is shown. The maximum photocurrent measured at 310 nm ultraviolet light with 300 V bias voltage in 8×10^{-10} mbar is about 14 nA, and the calculated QE is 0.043 %.

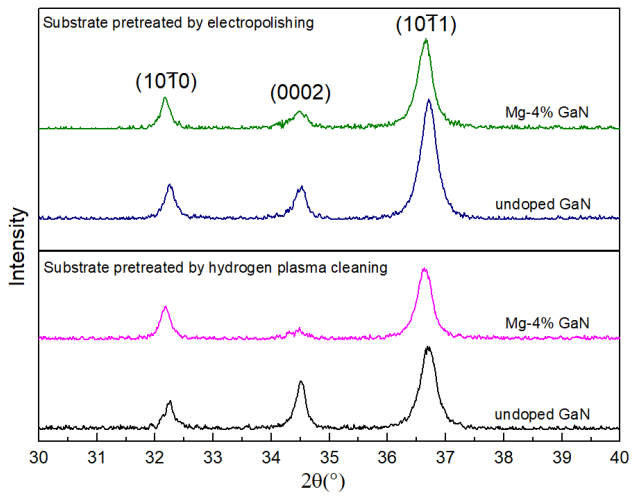

Figure 2: XRD pattern of GaN films deposited on differently prepared copper substrates.

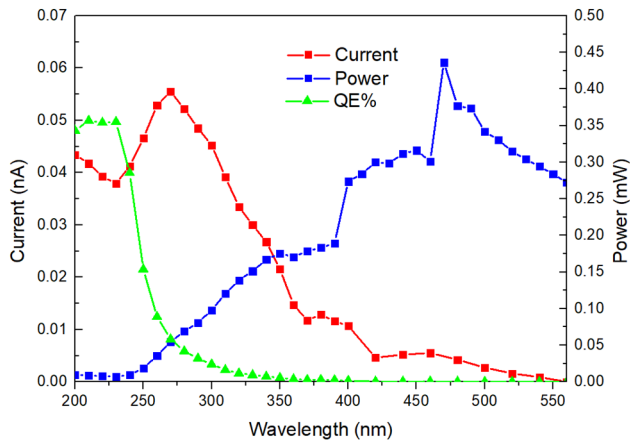

Figure 3: The QE curves of GaN photocathode film with 0.5 at.% Mg.

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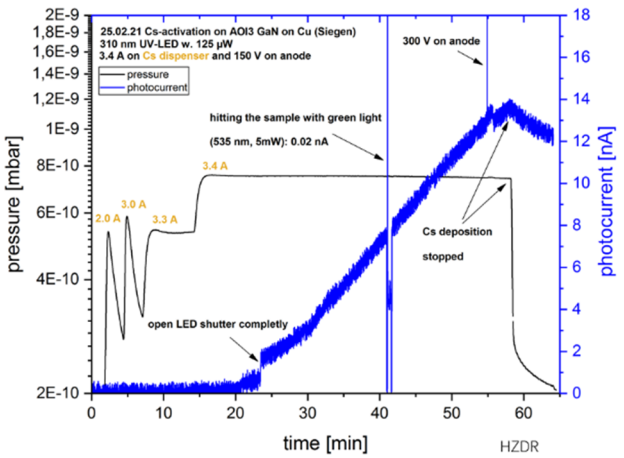

Figure 4: The photocurrent and pressure curves of GaN photocathode film with 3.5 at.% Mg, measured in HZDR.

CONCLUSIONS

Mg doped GaN films are grown on polycrystalline copper substrates by rf magnetron sputtering. The grain size of the GaN films is around 25 nm, with no particular texture. On copper substrate pretreated by hydrogen plasma cleaning the GaN films grow with different texture on different oriented copper grains. A possible heteroepitaxial growth mode found on certain Cu grain orientations will be investigated in the coming experiments. The fact that electropolished or ion-gun pretreated Cu samples exhibit a randomly oriented crystal structure of the film helps in understanding the possible impact of a certain crystallinity on the quantum efficiency. As the Mg content increases, the lattice parameters become larger and the texture changes. Finding the optimum Mg doping level is key for the coming investigations and improvements of the films' QE.

Besides the above mentioned planned investigations, the next step we will be an optimization of the in-situ QE measurement system and equip it with a deuterium light source to provide higher light power in the 200-400 nm range. A series of experiments will be conducted using the in-situ QE system to optimize the parameters of magnetron sputtering. Samples with higher QE will be sent to HZDR for further study.

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