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Influence of in-situ plasma treatment during PE-ALD of GaN on growth rate and morphology

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Abstract. In this work, the plasma-enhanced atomic layer deposition (PE-ALD) technique, including continuous hydrogen plasma, was studied for GaN growth. Also, the use of plasma at the nitrogen step only as well as argon plasma surface activation were explored. The structural properties of GaN layers grown on Si substrates at different conditions were studied by atomic force microscopy (AFM). It was shown that in-situ Ar plasma treatment during the PE-ALD process of GaN growth leads to improvement of the surface roughness as well as an increase in growth rate. On the contrary, the use of hydrogen plasma during the process leads to a drastic increase in surface roughness due to parasitic deposition.

Keywords: gallium nitride, plasma treatment, atomic layer deposition

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Материалы конференции
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Влияние плазменной обработки in situ при атомно-слоевом осаждении GaN на скорость роста и морфологию пленки

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Аннотация. В данной работе для выращивания GaN был использован метод плазменного атомно-слоевого осаждения (PE-ALD) с непрерывной водородной плазмой. Также было исследовано использование плазмы только на этапе осаждения монослоя азота, а также активация поверхности аргоновой плазмой. Методом атомно-силовой микроскопии (АСМ) исследованы структурные свойства слоев GaN, выращенных на подложках Si в различных условиях. Было показано, что плазменная обработка Ar in situ в процессе PE-ALD роста GaN приводит к улучшению шероховатости поверхности, а также к увеличению скорости роста. Напротив, использование водородной плазмы в процессе приводит к резкому увеличению шероховатости поверхности за счет паразитных осаждений.

Ключевые слова: нитрид галлия, плазменная обработка, атомно-слоевое осаждение

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Introduction

The efficiency of solar cells based on monocrystalline Si has already approached the theoretical limit. Further development of photoelectric conversion of solar radiation requires increasing efficiency. The efficiency of solar energy conversion is limited by losses, some of which are of a fundamental nature. The strongest limitation on the maximum achievable efficiency of single-junction solar cells comes from carrier the thermalization losses, where excess photon energy transferred to the generated charge carriers is transferred to thermal vibrations of the lattice. The most effective approach to reducing losses due to thermalization of charge carriers consists of the formation of tandem (multijunction) solar cells.

GaP-based diluted nitride is a potential material for tandem III-V/Si solar cells [1]. However, a low-temperature mass production technology is more preferable for its successful photovoltaic application.

Among a wide variety of semiconductor growth methods, atomic layer deposition (ALD) has several advantages, such as high-quality uniform deposition on large areas, conformal layer growth on textured surfaces with a high aspect ratio. Due to these advantages, the layers obtained by this method are currently used in the fields of nanoelectronics, photonics, and photovoltaics [2, 3]. ALD can be used to create thin layers of material over large areas with high throughput, which is one of the main advantages for photovoltaic applications.

To avoid the problem of nitrogen incorporation control, a sub-monolayer digital alloy (SDA) approach to PE-ALD process was successfully applied to improve the control of nitrogen incorporation into GaPN [4]. Using a combination of a sequence of layers in the form of short-period superlattices (digital alloys), GaN/GaP allowed for precise control of the band gap with compensation of elastic stresses arising from the difference in the lattice constant. However, the growth rate and morphology of the GaN layers should be precisely controlled during the growth of the short-period superlattices.

Recently, the use of in-situ annealing in argon plasma was shown to improve the crystalline quality at the epitaxial level of AlN [5] and GaP [6] layers obtained by PE-ALD. Here we explore the influence on the growth rate and morphology of GaN layers using PE-ALD at temperatures below 400 °C using in-situ annealing in hydrogen and argon plasma.

Materials and Methods

To form the required structures, Oxford Instruments Plasmalab 100 PECVD with capacitive coupled RF (13.56 MHz) plasma equipment with the ability to control the precursor flow rate, heater table temperature, and capacitance-coupled plasma RF power was used. The system was evacuated with a BOC Edwards iH600 dry vacuum pump to a minimum pressure of <1 mTorr. Immediately before the beginning of the deposition process, the c-Si substrates were treated in a 10% HF/H_2O solution to remove the natural oxide and hydrogenate the surface.

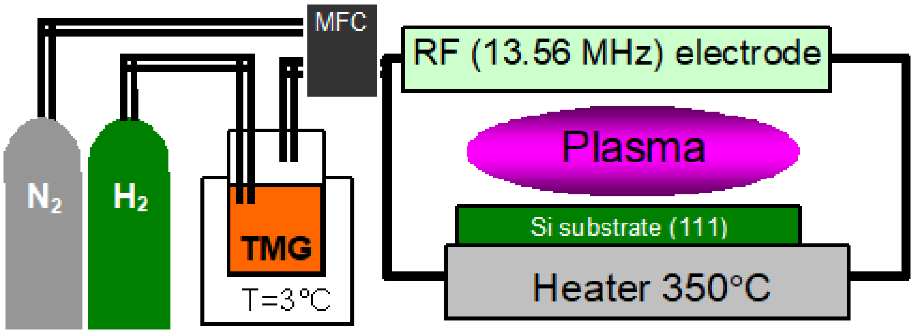

Fig. 1. Schematic representation of deposition chamber

Deposition of GaN layers was carried out at 350 °C on crystalline p-type Si substrates (111). A metalorganic precursor of gallium, trimethylgallium ($Ga(CH_3)_3$, TMG) 8% diluted in hydrogen (H_2) was entered into the chamber using a bubbling system; H_2 was used as a gas carrier with chamber pressure at 350 mTorr (see Fig. 1).

To realize the PE-ALD process, the Ga monolayer was followed by the N_2 plasma step, leading to the formation of the GaN sub-monolayer. The schematic presentation of the PE-ALD process without and with Ar in-situ plasma treatment (Fig. 2, *a*, *b*). After exposure to each precursor, the chamber was purged with argon to avoid mixing precursors and parasitic CVD growth. Continuous hydrogen plasma mode with RF power increase was also used to guarantee ignition and hold of discharge.

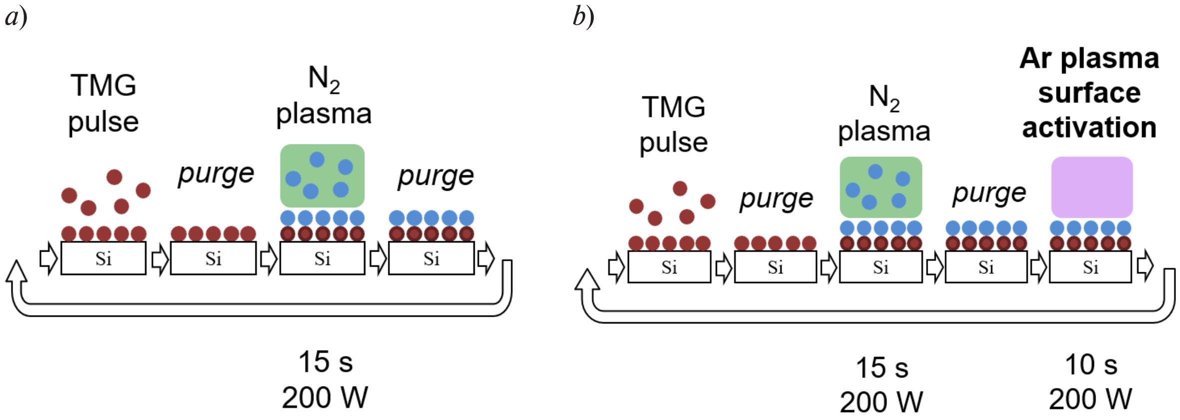

Fig. 2. PE-ALD realization of GaN growth (*a*) with Ar in-situ plasma treatment (*b*)

To study structural properties and surface morphology, an image of the GaN layer deposited on a silicon substrate was obtained using scanning electron microscopy (SEM) from a Zeiss Supra 25 set-up. Furthermore, film surface roughness was studied by atomic force microscopy (AFM) using the BioScope Catalyst Bruker setup.

Results and Discussion

An SEM image of GaN film grown by PE-ALD with continuous H_2 plasma is presented in Fig. 3. The GaN layer grown by PE-ALD with continuous hydrogen plasma has a rough surface and a film thickness of 40 nm.

The AFM 3D topology images of the 40 nm thick GaN samples are presented in Figure 4. The growth rate and surface roughness strongly depend on plasma treatment. Growth per cycle (GPC) and root mean square (RMS) roughness are equal to 0.095 nm/cycle and 0.49 nm, respectively, for PE-ALD without plasma, while Ar in-situ plasma treatment leads to GPC increasing to 0.185 nm/cycle and RMS decresing to 0.38 nm. Such behaviour is associated with local surface heating, which leads to the enhancment of gallium precursor mobility.

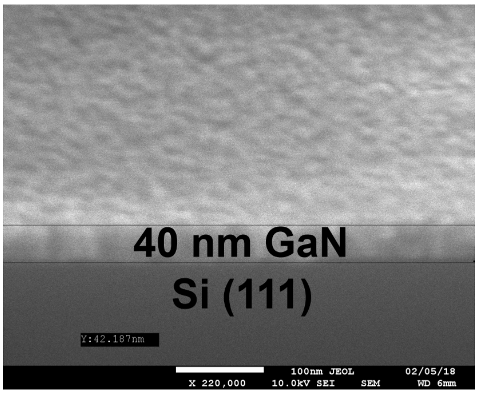

Fig. 3. SEM image of PE-ALD grown GaN film

Continuous hydrogen plasma treatment leads to GPC rising to about 0.2 nm/cycle while the surface roughness increases (Fig. 4, *c*) with an RMS of 1.19 nm. The parasitic deposition during hydrogen plasma treatment is supposed to occur in this case (all data are presented in Table 1).

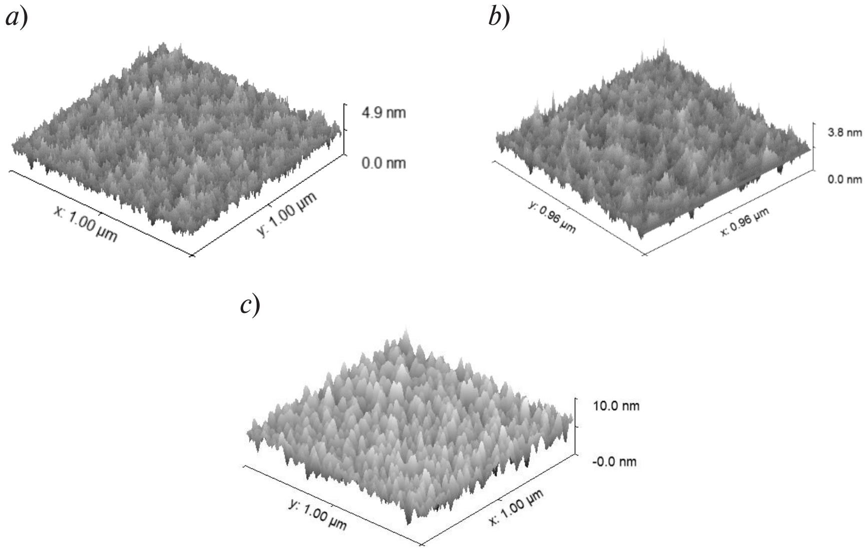

Fig. 4. AFM 3D images of 40 nm thick GaN on Si grown by PE-ALD without (*a*) and with Ar plasma treatment (b), with continuous hydrogen plasma mode (*c*)

Table 1

PE-ALD additional treatment influence on roughness and deposition rate

PE-ALD GaN 350°C	No Ar treatment	Ar plasma treatment	H_2 continuous plasma
RMS, nm	0.49	0.38	1.19
GPC, nm/cycle	0.095	0.185	0.2

Conclusion

In this study, the use of atomic layer deposition (ALD) for the growth of GaN layers on Si was investigated. The results showed that the ALD method, combined with in-situ annealing in argon and continuous hydrogen plasma mode, could affect the growth rate and morphology of GaN layers. The way to improve the surface roughness as well as increase the growth rate by using in-situ Ar plasma treatment during PE-ALD growth of GaN was demonstrated. On the contrary, usage of hydrogen plasma during the process leads to a drastic increase in surface roughness due to parasitic plasma deposition.

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