行政院國家科學委員會補助專題研究計畫成果報告

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氮化鎵量子結構之模型及處理（計畫名稱）

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計畫主持人：彭隆瀚　副教授
共同主持人：

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共同主持人：--
計畫參與人員：研究生 廖清賢，徐崑庭，王耀興

一、中文摘要

本報告研究以光化學方式在氮化鎵上升呈自然氧化膜，並研究其對於光電性質之影響。

關鍵詞：氮化鎵，氧化膜

Abstract

Abstract *---- We investigate the photo-enhanced wet oxidation effects on the optical properties of GaN/InGaN quantum wells. Oxidation of GaN is found to take place in phosphorus acid solutions in a pH range from 3 to 4, and results in a maximum oxidation rate of 224 nm/h. The spectral red shift and more than 50% enhancement on the photo-luminescence intensity on the oxidized InGaN QW are attributed the oxide induced stress and surface passivation effects.*

Index Terms ----GaN, surface passivation, oxidation

I. INTRODUCTION

Bandgap III-V nitrides have been long recognized as a material system of outstanding opto-electronic properties for short wavelength applications. Rapid development in the nitride technology has brought in a plethora of research activity ranging from material investigation to device fabrication. Due to the lack of efficient surface passivation techniques, common practice has pursued the use of silicon oxide (SiO_x) or silicon nitride (Si_xN_y) as the dielectric coating in the processing of GaN-based devices. However, it has been recently noted the deposited SiO_x layer can result in a significant degradation of the photo-luminescence (PL) intensity due to the incorporation of oxygen as non-radiative recombination centers in GaN. [1] It is therefore desirable to develop alternative surface passivation techniques that could be immune from such problems.

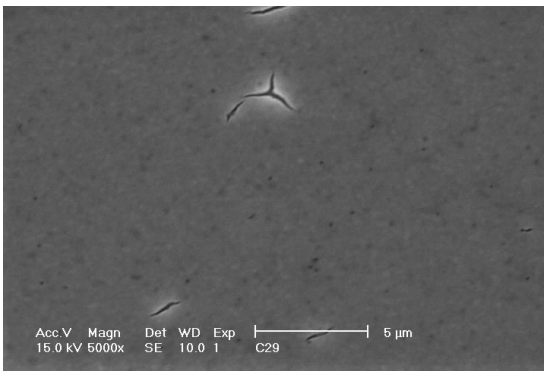
We investigate the photo-enhanced-oxidation process on GaN and its impact on the optoelectronic properties.

Keywords: GaN, wet oxidation

To date, direct thermal oxidation of GaN can only occur with a rather slow oxidation rate of 20 nm/hr at temperature above 900 °C. [2] There are, however, concerns on the high temperature treatment due to the surface degradation issues. In this work, we investigate a *room* temperature photo-oxidation process of GaN and the passivation effects on InGaN quantum wells (QW). We thereby are able to reveal a reaction-rate limited oxidation process and observe enhancement in the PL response on InGaN QWs for the first time. We attribute the latter to the surface passivation effects with the formation of good thin film quality of gallium oxide.

The employed photo-chemistry process in GaN is that the ultraviolet (UV)-excited hot carriers at the GaN/electrolyte interface have excess energy to access the H^+/H_2 and OH^-/O_2 redox levels in water and enhance the oxidative dissolution of GaN.[3] In the experiments, the galvanic cell was formed by immersing a GaN working electrode and a platinum (Pt) counter electrode in the electrolyte and was illuminated with a 254 nm mercury line source of 10 mW/cm^2. The experiments were carried out in aqueous H_3PO_4 solutions at room temperature with *no* bias applied between the GaN sample and the Pt counter electrode.

II. RESULTS



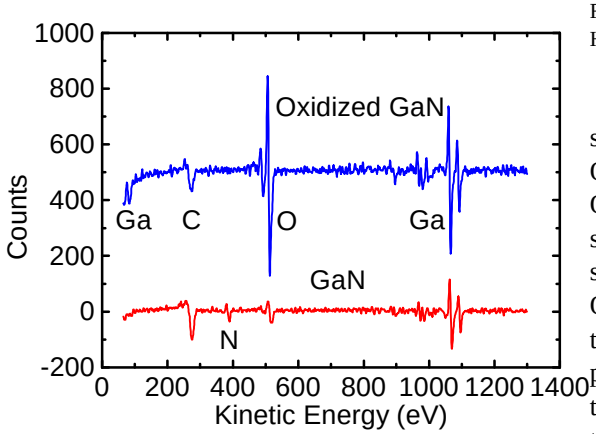


Fig. 1 (a). SEM, and (b) Auger analysis of an oxidized GaN surface

We note a transition from a transparent-looking, mirror-like oxidized surface to stress induced surface crack occurs at an oxide thickness of ~ 400 nm. Shown in Fig. 1 are (a) the SEM micrograph, and (b) the Auger spectral analysis on an oxidized GaN surface covered with 420 nm oxide layer. The N-peak is found to totally disappear (within the detection resolution) from the oxidized GaN surface with an enhancement is observed at the O-peak position. The EDX analysis reveals a normalized atomic ratio of O: Ga = 58.85% : 40.23% which suggests the composition of the oxide is likely to be Ga_2O_3.[4] This observation is supported by a subsequent dissolution of the oxide layer in a 2M potassium hydroxide (KOH) solution. Had gallium hydroxide formed in the oxidation process, it would be insoluble in the alkaline solution.[3]

Shown in Fig. 2 is the oxidation rate analysis of the photo-grown oxide in a H_3PO_4 electrolyte of pH = 3.5. Data here reveals a linear time dependence with a slope of 224 nm/hr. This indicates a reaction-rate limited process has taken place in the photo-chemical reaction. We note the UV-assisted *wet* oxidation rate of GaN at room temperature is one order of magnitude higher than that previously reported in the high temperature treatment [2]. Also shown in Fig. 2 is the pH dependence of the normalized UV-assisted oxidation rate. The observation of a peak reaction rate indicates a hydration action such that both of the solute (H_3PO_4) and the solvent (free water molecules) play an important role in the photo-oxidation process [3].

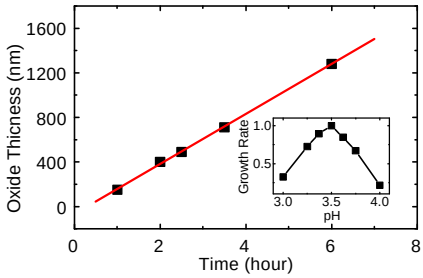
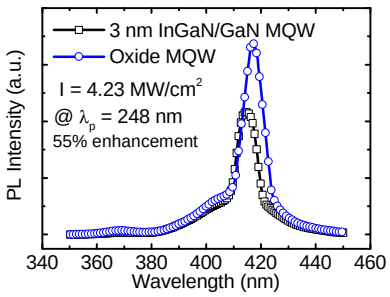


Fig. 2. Rate analysis of the photo-oxide in aqueous H_3PO_4 solutions.

Finally we shown in Fig. 3(a) the PL spectra of an oxidized 3.0 nm InGaN/GaN QW excited by a 248 nm KrF excimer laser. Our calculation in Fig. 3 (b) indicates the spectral red-shift is opposite to that of blue shift observed in the anodized GaAs/AlGaAs QW system.[5] This observation suggests the oxide stress-induced piezo-electric effect plays an important role in the optical transition energy. Since the Fresnel loss due to the optical reflection at the GaN/air interface is no more than 20%, the observation of 55% enhancement in the PL intensity is attributed to an efficient surface passivation on this InGaN/GaN material system. This process will be valuable to the application of nitride-based opto-electronic devices.



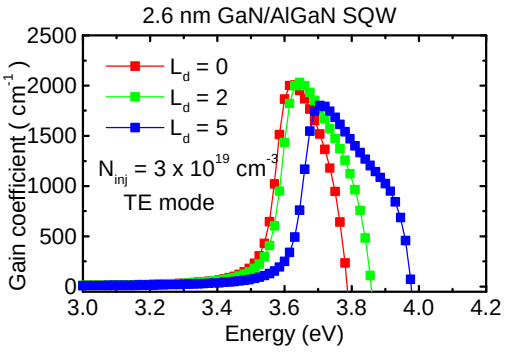


Fig. 3 (a) Measured room temperature PL response from the as-grown and photo-oxidized InGaN/GaN QW, (b) calculated optical gain spectra considering the inter-diffusion effects.

III. SUMMARY

In summary, we report a UV-enhanced, reaction-rate limited *wet* oxidation process on GaN at room temperature. A peak oxidation rate as high as 224 nm/hr can be achieved in aqueous H_3PO_4 solution at pH = 3.5. Enhancement in the PL response from the oxidized GaN/InGaN QW suggests an efficient surface passivation due to the photo-oxidation process on GaN. This research was sponsored by the NSC Grant No. 89-2215-E-002-023.

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