Revised: 2022-02-25 Accepted: 2022-03-07 © 2022 The Author(s). Published by Trans Tech Publications Ltd, Switzerland. Online: 2022-05-31

# **Enhanced Resonant Raman Scattering of GaN Functional Layers Using** Al Thin Films - A Versatile Tool for Multilayer Structure Analysis

Submitted: 2021-10-14

Alina Muravitskaya<sup>1,2,a</sup>, Anna Rumyantseva<sup>1,b</sup>, Atse Julien Eric Ndohi<sup>3,c</sup>, Camille Sonneville<sup>3,d</sup>, Dominique Planson<sup>3,e</sup>, Pierre-Michel Adam<sup>1,f</sup>, Sylvain Potiron<sup>2,9</sup> and Mihai Lazar<sup>1,2,\*</sup>

<sup>1</sup>Laboratory Light, Nanomaterials & Nanotechnologies, CNRS ERL 7004, University of Technology of Troyes, 12 rue Marie Curie, 10004 Troyes, France

<sup>2</sup>Laboratoire de Recherche en Nanosciences, Université de Reims Champagne Ardenne, Moulin de la Housse, 51687 Reims, France

> <sup>3</sup>AMPERE, UMR 5005 CNRS, INSA Lyon, Villeurbanne, F-69621, France

<sup>a</sup>alina.muravitskaya@gmail.com, <sup>b</sup>anna.rumyantseva@utt.fr, <sup>c</sup>atse-julien-eric.ndohi@insa-lyon.fr, dcamille.sonneville@insa-lyon.fr, edominique.planson@insa-lyon.fr, fpierre\_michel.adam@utt.fr, gsylvain.potiron@univ-reims.fr, \*mihai.lazar@utt.fr (corresponding author)

Keywords: SERS, GaN, SiC, plasmonic nanoparticles, power device, Raman scattering, nanostructures, thin layers

Abstract. In order to selectively analyze active thin layers close to surface in power devices structures, Raman scattering is necessary with UV excitation. However, the Raman spectra of GaN are usually affected by the direct bandgap photoluminescence of the material, which interferes with the Raman measurements and decreases the quality and resolution of the Raman spectra. In this work, we demonstrate experimentally that nanostructured aluminum films deposed on GaN epitaxial layers decrease the influence of the photoluminescence on the resonant Raman spectra and increase its overall spatial resolution under UV illumination.

### Introduction

The resonant Raman scattering is a well-established non destructive method for analyzing the surface properties of the GaN determining the crystalline quality, stress, doping, free carrier concentration for process and growth monitoring [1,2]. In GaN power devices one needs to selectively analyze thin layers in the surface (active layers as emitters, channels, junction termination extensions created by ion implantation, metal-contact interfaces) with thickness that does not exceed micrometer at most and can go to ~100 nm and even less [3]. To gain the correct values, the influence of underlayers present in the vertical device (hetero)structure (substrate, buffer, drift layers, etc.) should be eliminated. Thus, for GaN thin layer analyses, UV excitation is preferred to the classical visible or IR excitation wavelengths, since it allows to examine a surface layer of about 40 nm thick (under 325 nm excitation) [4]. However, as GaN has a direct bandgap, strong photoluminescence (PL) contribution can interfere with the measurements and decrease the quality and resolution of the obtained Raman spectra under UV excitation.

One of the approaches to decrease the PL background and increase the Raman signal is to place plasmonic nanoparticles on the top of a layer of interest, which results in a quenching of the PL due to the high non-radiative transfer of the excitation energy to metal, and in the same time, Raman scattering benefits from the local field enhancement and increase in radiative decay rates near resonant plasmonic nanoparticles. In this work, we propose to use structured Al films to diminish the influence of the PL on the resonant Raman spectra of GaN and to increase its overall resolution.

### Methods

GaN thin epilayers (between 500 nm and  $1\mu m$  thick MOCVD) on sapphire substrates from CNRS-CRHEA with a silicon doping concentration in the range of  $10^{16}$  to  $10^{17}$  cm<sup>-3</sup> were covered with Al nanoparticles and Al thin films, which were deposited by means of the vacuum evaporation technique (e-beam PlassysMU400). The samples are presented in Figure 1. The Al nanoparticles film was obtained for a thickness of the deposition of 6 nm. The Al thin film was obtained after 20 nm deposition. In both cases samples were heated up to  $400^{\circ}$ C during the deposition. The Al nanoparticles film was formed during a dedicated e-beam evaporation step only on one of the GaN epilayers presented in Figure 1.

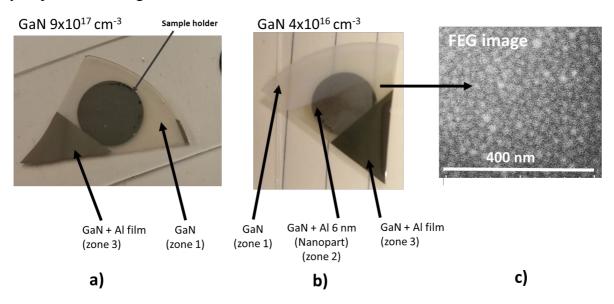


Fig. 1. Optical photos of the GaN epilayers partially covered with Al nanoparticles and films (a and b) and a scanning-electron microscopy image of the area covered with Al nanoparticles (c)

From the area covered with the Al nanoparticle a FEG-SEM (Field emission Scanning Electron Microscope) image is also presented in the right side of Figure 1. On these parts of the samples, photoluminescence (PL) and  $\mu$ Raman measurements were done under UV ( $\lambda_{ex}$ =325 nm) illumination and in back-scattering geometry with LabRAM spectrometer of Horiba Scientific (scheme in Fig. 2). Three Raman modes are allowed in GaN layers in the backscattering configuration [5]: 732 cm<sup>-1</sup> –  $A_1(LO)$ , 568 cm<sup>-1</sup> –  $E_2^{high}$  and 140 cm<sup>-1</sup> -  $E_2^{low}$ 

In order to compare and analyze PL and  $\mu$ Raman signals similar commercial 4H-SiC samples (substrate with n-type epilayers of 10  $\mu$ m thickness and  $5\times10^{15}$ cm<sup>-3</sup> doping) have been prepared with the same experimental Al deposition setup.

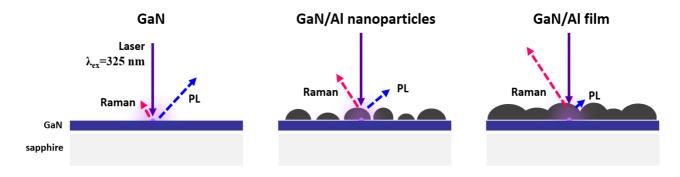


Fig. 2. PL and Raman signals obtained from different GaN epilayers surfaces covered or not with Al nanoparticles and films, schematically highlighting the SERS effect. The length of the arrows corresponds to the relative intensity.

### **Results and Discussion**

As shown in figure 3a, the deposition of the Al on top of the GaN resulted in a drastic fall of the photoluminescence intensity. In the case of the Al nanoparticles film the PL has decreased 10 times, while for the Al thin film it has decreased 100 times.

Then we observed in figure 3b the revealing of the three Raman modes in the spectra:  $732 \text{ cm}^{-1} - A_1(\text{LO})$ ,  $568 \text{ cm}^{-1} - E_2^{\text{high}}$  and  $140 \text{ cm}^{-1} - E_2^{\text{low}}$ . A schematic presentation of this SERS (surface-enhanced Raman spectroscopy) effect is highlighted in Figure 2 and measurements are presented in Figure 3.

Important to note that we do not observe the contribution of the sapphire (GaN epilayer substrate in our samples) modes, which often change the shape of the spectrum of GaN. Indeed the light penetration depth in GaN is less than the epilayer thickness.

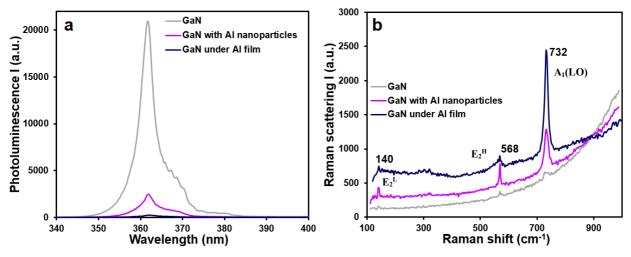


Fig. 3. Photoluminescence (a) and Raman scattering spectra (b) ( $\lambda_{ex}$ =325 nm) of the GaN epilayers covered with layer of Al nanoparticles and covered with thin Al film layer.

Compared to visible non resonant Raman spectra (e.g at 532 nm) in which the  $E_2(H)$  peak is the more intense [1], in our UV Raman spectra enhanced by Al, the  $A_1(LO)$  peak dominates the resonant Raman spectra. This effect is more visible with the Al film than with Al nanoparticles. More over  $A_1(LO)$  peak is more intense compared to the PL background in the case of the Al thin film, than for the Al nanoparticles film. However, for the layers with Al nanoparticles film, the lines width is sharper and the  $E_2(H)$  peak is more pronounced. Al nanoparticles film seems more appropriate to be used in the fine UV Raman analysis of GaN epilayer characteristics as mechanical stress and defects, by considering the resolution obtained for the  $E_2H$  peak. For doping,  $A_1(LO)$  peak could be considered using Al films.

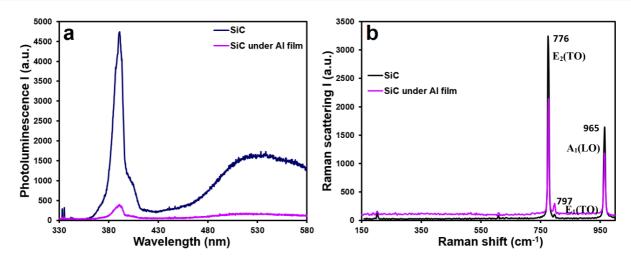


Fig. 4. Photoluminescence (a) and Raman scattering spectra (b) ( $\lambda_{ex}$ =325 nm) of the SiC epilayers covered with thin Al film.

For both films the absorption of the GaN emission by Al layers and consequent fall of the PL play an important role in the Raman lines appearing. Also, the plasmonic enhancement of the Raman scattering may occur, especially as Al sustains localized surface plasmon resonances in the UV region [5, 7].

4H-SiC epilayers and substrates have been also tested without and with Al layer deposited during the same run with the GaN samples. Even though the PL signal was reduced 10 times (Figure 4a) the enhancement of the Raman scattering was not observed (Figure 4b). We note that SiC compared to GaN has an indirect bandgap, and then the PL signal is not so high. Therefore, doping check or other stress and defects identification could be done by direct UV Raman measurements without Al nanoparticles or nanolayers on the surface.

#### **Conclusions**

As for SiC layers  $\mu$ Raman can be utilized on GaN layers as a versatile and nondestructive tool to investigate critical layers as those close to two-dimensional electron gas in AlGaN/GaN heterostructures. We show that Al thin layers deposited on top of the GaN epitaxial layers may improve the quality and intensity of the resonant Raman scattering of the GaN functional layers and therefore provide a way to probe doping, defects and various kinds of stress in thin layers (less than  $1\mu m$ ) of actual structures of power devices. In this case, a thin Al layer is needed to increase Raman scattering signal and decrease the photoluminescence. This metallic layer can be kept or removed after test.

## Acknowledgments

The authors acknowledge our colleagues from CRHEA-CNRS for the GaN/sapphire samples. This work has been made within the framework of the Graduate School NANO-PHOT (École Universitaire de Recherche, contract ANR-18-EURE-0013). The authors acknowledge the project RETOR.

### References

- [1] Hiroshi Harima 2002 J. Phys.: Condens. Matter 14 R967
- [2] M. Kuball, Surface and Interface Analysis. 31, 19 (2001) 987-999.
- [3] Richard Denny, Abirami. M, Kanimozhi G, Ronit Hara Yandra, Journal of Critical Reviews, Vol 7, Issue 12, 2020, 456-467
- [4] J.F. Muth, J.H. Lee, I.K. Shmagin, R.M. Kolbas, H. C. Casey Jr., B. P. Keller, U. K. Mishra, S.P. DenBaars, Appl. Phys. Lett. 71, 18 (1997) 2572.
- [5] Z. C. Feng, W. Wang, S. J. Chua, P. X. Zhang, K. P.J. Williams, G. D. Pitt, JRS. 32, 10 (2001) 840-846.
- [6] M. W. Knight, N. S. King, L. Liu, H. O. Everitt, P. Nordlander, N. J. Halas, ACS Nano. 8 (2013) 834–840.
- [7] A. Muravitskaya, A. Gokarna, A. Movsesyan, S. Kostcheev, A. Rumyantseva, C. Couteau, G. Lerondel, A.-L. Baudrion, S. Gaponenko, and P.-M. Adam. Nanoscale. 12, 11 (2020) 6394-6402.