English
Research Express@NCKU Volume 6 Issue 5 - November 7, 2008
[http://research.ncku.edu.tw/re/articles/e/20081107/5.html]

Recrystallization of epitaxial GaN under indentation

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Applied Physics Letters 92, 143114 (2008)

Suji Nakamura's hard work found GaN as potential candidate for the blue light source around 365 nm for which he received the prestigious Millennium Technology Prize in 2006. Blue diode laser have the huge market for printing (1200 dpi having ~17 μm spot size with 1 mm depth of field using 6 mm optics), high density DVD memories and white light based (using diode laser based RGB) compact display devices. However, high dislocation density and related issues in GaN are one among few of the major hindrances for the application as blue laser[1]. Thus, removal of stress is one of the prime objectives for opto-electronic device applications in GaN. Growth of epitaxial (epi-) GaN film with low dislocation density either by pre-grooving the buffer layer [2] or by lateral epitaxial overgrowth (LEO) [3] is basically to reduce residual strain in the system and help crystallization process. A remarkably low threading dislocation (TD) density ~10^7 cm^{-2} is reported in epi-GaN film adopting the LEO technique. A combination of biaxial and hydrostatic stresses originating form dislocation related extended defects and point defects, respectively, are reported generally in epi-GaN.

While the above said techniques [2,3] are well reported for the growth of epi-GaN with reduced dislocation density, we have reported [4] the recrystallization of epi-GaN under indentation with various loads and loading-unloading rates. Micro-Raman spectra along with area mapping, for the spectral region of interest, are studied using 632.8 nm excitation of He-Ne laser.

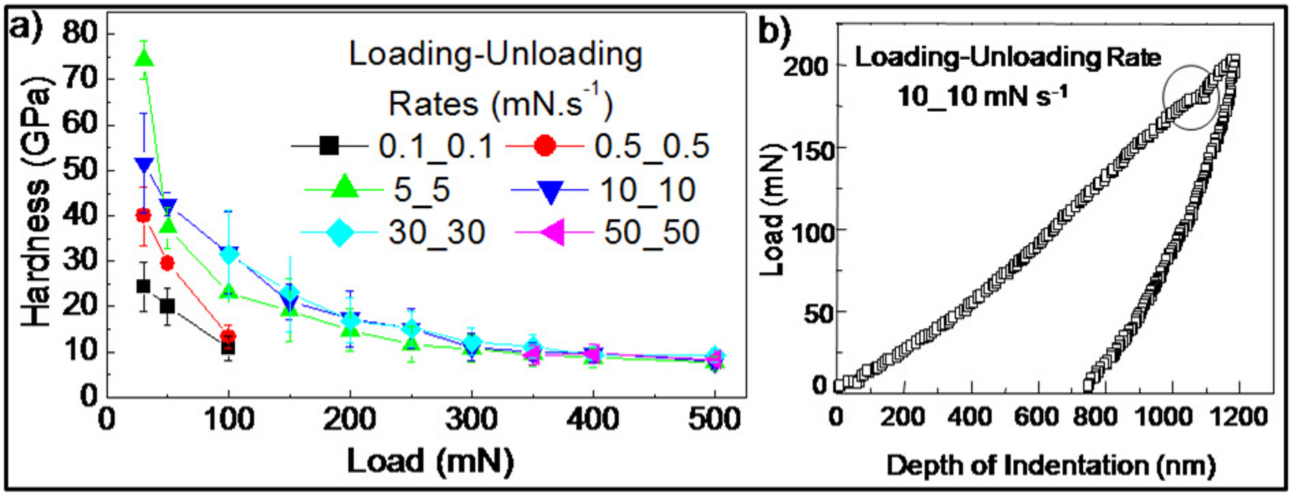

Fig. 1 a) Measured values of hardness of epi-GaN(0001) with load. Loading-unloading rate is also varied for the detailed studies. b) Typical loading-unloading lines at a fixed loading-unloading rate of 10 mN.s^{-1} The encircled regions are called as 'pop-in' burst.

A hardness value equivalent to bulk GaN ~10 GPa is achieved (Fig. 1a) at higher loads (> 100 GPa) or at lower loading rate (≤ 0.5 mN s^{-1}). This is in accordance with the conventional wisdom of materials that at low loads, where defect formation is mainly close to the surface, increasing number of extended defects (dislocations) with increasing loading rate enhances the strength (hardness) of the material. Formation of extended defects can be explained with geometrically necessary dislocation model, which in turn explains the depth dependent hardness variation for the crystalline material. Typical loading-unloading curve is shown (Fig. 1b) with 'pop-in' burst in the loading line (encircled). The mechanism responsible for the 'pop-in' burst appears to be associated with the nucleation and movement of dislocation sources including lattice atoms for the possible recrystallization process [5].

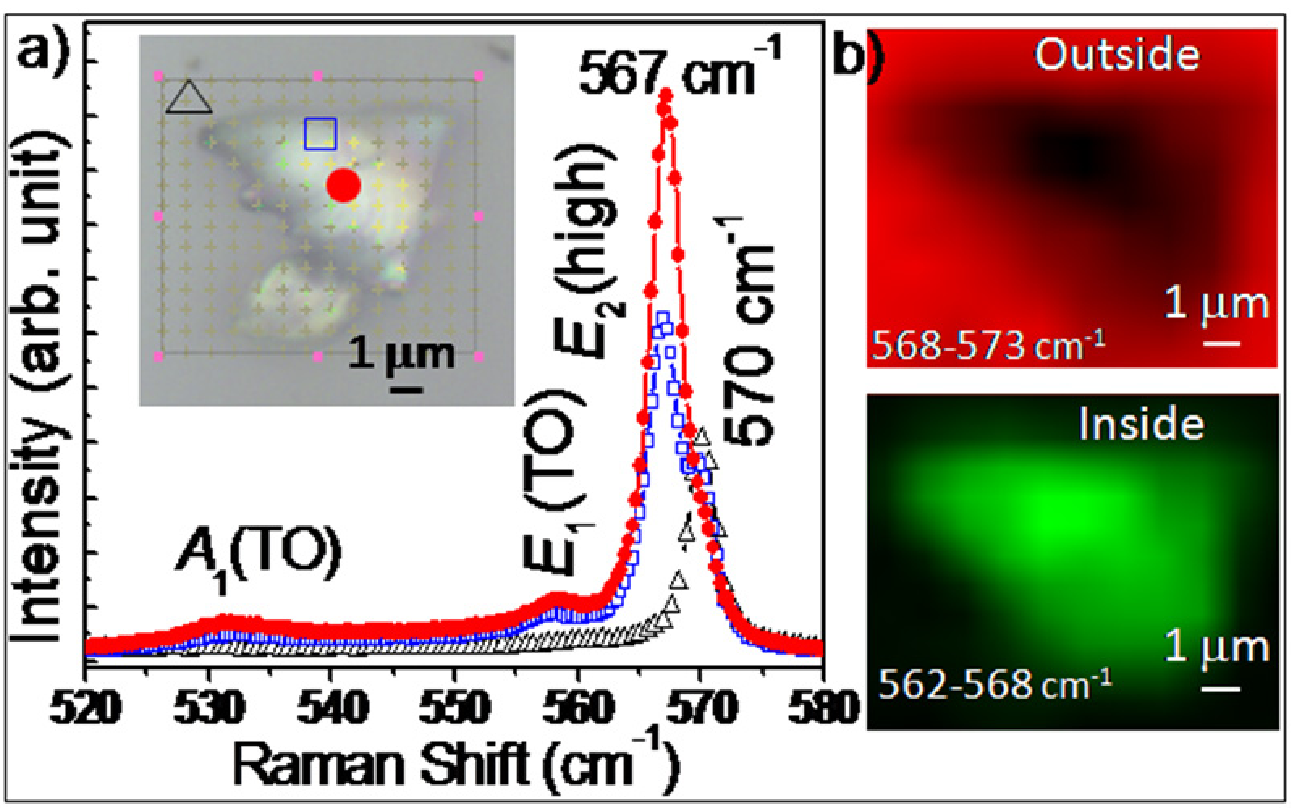

Fig. 2. a) Micro-Raman spectra for epi-GaN outside and different regions inside the indentation spot. Inset shows corresponding optical image of the indentation spot. b) Area mappings of outside and inside of the indentation spot, using different spectral regions indicated in the picture.

The structural transformation is studied close to the indented region using micro-Raman spectroscopy (Fig. 2). E_2(high) mode at 570 cm^{-1} is measured outside the indented region and the value resembles the reported value of epi-GaN on sapphire substrate. Inset shows the spots measured outside and insides of the indentation mark recorded in the optical microscope attached to the spectrometer. Micro-Raman measurements inside the indented region show redshift of phonon mode gradually to 567 cm^{-1} from interface region (edge of the spot) to the center of indented spot. This value is close to the calculated and measured value for E_2 (high) phonon of bulk GaN and its nanostructure under stress free conditions [6]. Double peaks are observed for the interface region close to the edge of the indentation (Fig. 2a), showing contributions from both the stressed outside region and stress free inside region the indented region. Raman area mapping (Fig. 2b) using spectral part of 568-573 cm^{-1} shows red region lying outside the indented region and 562-568 cm^{-1} shows green region lying inside the indented region. It clearly shows that the 567 cm^{-1} and 570 cm^{-1} peaks originates from the stress free inside region and stressed outside region of the sample, respectively. Two additional peaks at 531 cm^{-1} and 559 cm^{-1} (Fig. 2a) corresponding to A_1(TO) and E_1(TO) modes, respectively, are also observed in the indented region. According to selection rule in wurtzite crystal of GaN, TO phonon modes are forbidden in the backscattering geometry for the (0001) oriented planes [6]. However, small misorientations of crystallites in the indented region allow phonon modes corresponding to other crystalline orientations.

Assuming hydrostatic stress alone inside the indentation region, a stress of <1 GPa is required for the 3 cm^{-1} shift (Fig. 2a) of E_2(high) peak position.. It is concluded from the reported [7] pressure coefficient of Raman peak of -3.55 cm^{-1} GPa^{-1} in GaN for hydrostatic stress alone. The distribution of pressure under the indenter given by[8]

$$p(r) = \frac{E}{2(1-\nu^2)} \frac{\cosh^{-1}(a/r)}{\tan \Psi}, 0 \le r \le a \quad \text{.................................} (1)$$

where E is Young’s modulus, v is Poisson’s ratio (0.22 for epi-GaN), a is the contact radius, and r is the radial coordinate in the surface. In the Berkovich indenter (Ψ ~ 65.3•) with experimentally observed reduced modulus of 100 GPa, the pressure at the central region can be calculated as 10 GPa (at the boundary) to 55 GPa (exactly at the centre) using Eqn.1. Thus the estimated pressure requirement of <1 GPa for the observed Raman shift can be always provided in this technique.

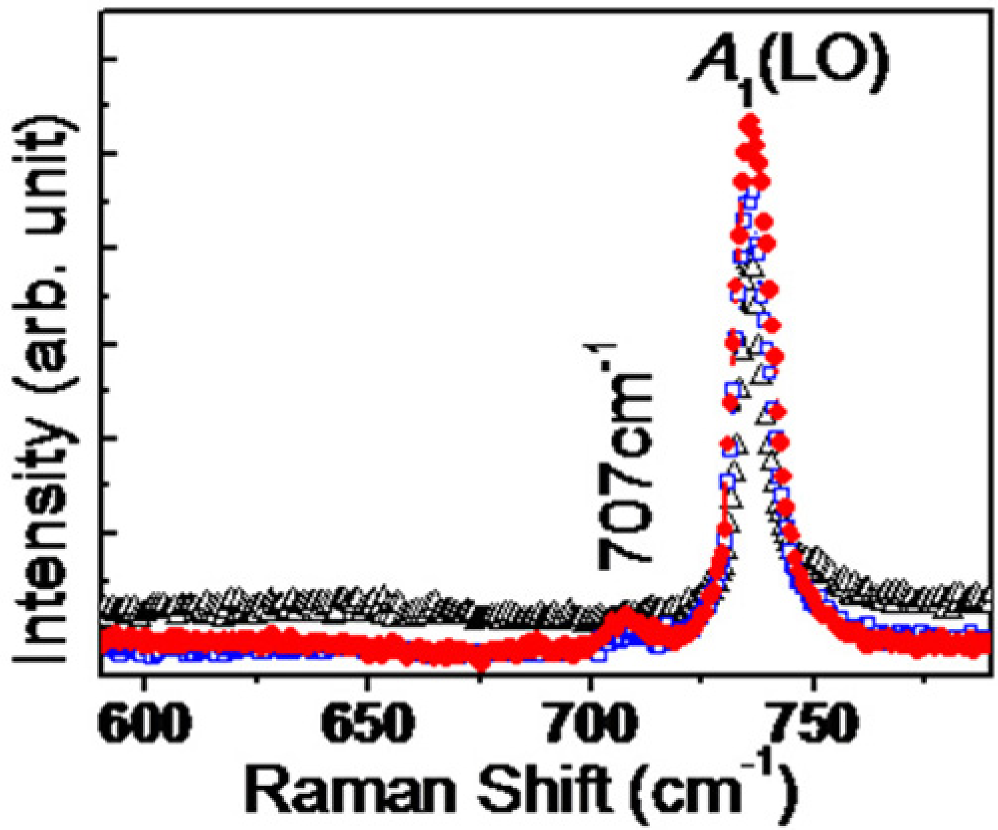

Fig. 3. Micro-Raman spectra for epi-GaN outside and different regions inside the indentation spot as shown in the inset of Fig. 2a.

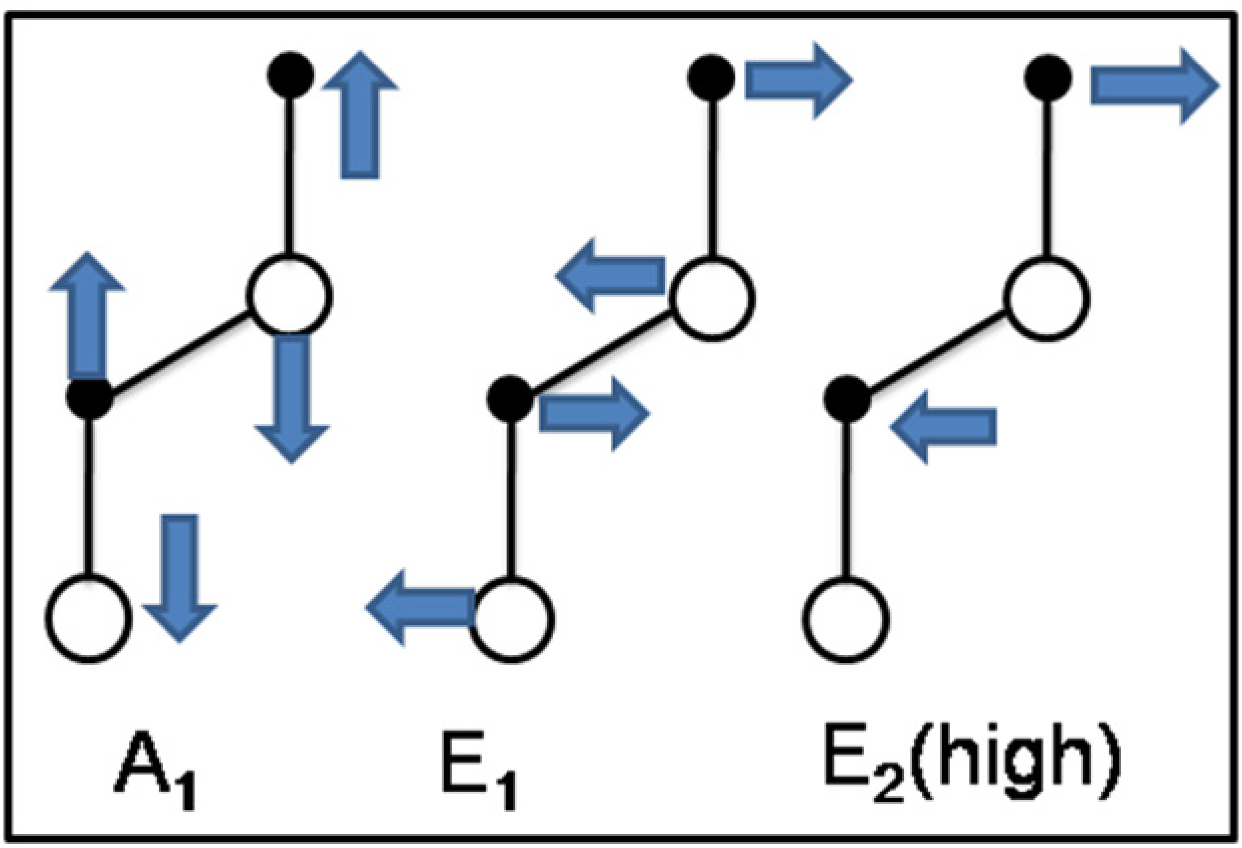

Fig. 4. Optical phonon modes in the wurtzite structure of GaN. Filled and unfilled symbols are represented by Ga and N, respectively. Arrows indicate directions of atomic vibrations.

Interestingly, peak ~736 cm^{-1} corresponding to A_1(LO) mode in the different regions close to the indentation spot (Fig. 3) show no shift in peak position. A small peak ~707 cm^{-1} peeps in the spectra collected from the regions inside the indentation. This peak can be assigned as surface optic mode of nanocrystalline GaN [9]. Raman scattering configuration for the wurtzite crystal of GaN(Fig. 4), for phonon corresponding to LO mode of A_1 symmetry is always along Z direction (normal to XY plane). Thus with no change in peak position of A_1(LO) mode, it is obvious that lattice modes normal to XY plane remain unaltered. From the evolution of E_2(high) and TO phonon modes (Fig. 2a), collected in the backscattering geometry, it seems that stress is released in the planar direction so that Raman modes in the XY plane gets modified. Thus, overall evolution of phonon modes may be due to nucleation of dislocation and release of its stress field under indentation stress to set in planar motion at the centre of indentation region by dislocation climb. Dislocation climb in the material are reported under very high hydrostatic stress due to indentation.

In conclusion, an indentation induced recrystallization is reported in epi-GaN using a Berkovich indenter. The amount of stress required for the recrystallization process is validated with the calculated value of stress in the indented volume. Dislocation stress field might have been released at the centre of indentation region with the dislocation motion under the indentation stress. A clear picture of defect dynamics, as studied in this report, will help for further understanding of the removal of dislocation in GaN leading to the possible commercialization of the material for the service of people.

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