Nanoindentation-Induced Pop-In Effects in GaN Thin Films

Sheng-Rui Jian and Jenh-Yih Juang

***Abstract*—The nanoindentation-induced pop-in phenomena in GaN thin film are investigated using Berkovich indenters. The formation of dislocation rosettes revealed by cathodoluminescence (CL) spectroscopy is found to closely relate with the pop-in effect displayed in depth-sensitive measurements. Namely, the CL images of the indented spots show well-defined rosette structures consistent with the hexagonal symmetry of GaN, indicating that the distribution of deformation-induced extended defects/dislocations may dramatically affect the CL emission. The use of CL thus may provide an alternative means for studying the near-surface plasticity in other semiconductor thin films, as well.**

***Index Terms*—Cathodoluminescence (CL), GaN thin films, nanoindentation, pop-in.**

I. Introduction

NANOINDENTATION has been widely used as an important tool to study the mechanical characteristics of materials [1], including the measurement of hardness and elastic modulus [2]–[5], creep resistance [6] and fracture behaviors [7], [8]. The difference between the elastic and plastic deformation behaviors enables the analysis of the mechanical responses. The initial segment of the load–displacement curve resulting from nanoindentation is usually the manifestation of the elastic behavior, whereas the onset of plastic deformation is generally associated with a displacement discontinuity, i.e., pop-in event, during the loading process [9], [10]. Analysis of each part of the curves provides prominent information about the mechanical behaviors of the material under investigation.

The elastic behavior can be analyzed by the elastic contact deformation relationship given by the Hertzian elastic contact model [11], as follows:

$$P = \frac{4}{3} E_r \left(Rh^3\right)^{1/2}. \quad (1)$$

Here, $P, R,$ and h are representing the applied indentation load, the radius of indenter tip, and the corresponding indentation depth, respectively. The reduced elastic modulus E_r is given by

$$E_r = \left(\frac{1 - v_{\text{film}}^2}{E_{\text{film}}} + \frac{1 - v_{\text{tip}}^2}{E_{\text{tip}}}\right)^{-1} \quad (2)$$

where v is Poisson's ratio, E is Young's modulus, and the subscripts "tip" and "film" indicate the indenter tip and indented films, respectively. The deviation of the experimental data from the Hertzian relationship is usually attributed to the onset of plastic deformation occurring when the load applied to the material exceeds certain values [12].

In previous works, single or multiple pop-in events were observed in GaN thin films during nanoindentation [10], [13]–[16]. However, since the onset of nanoscale plasticity and mechanical properties are strongly influenced by factors, such as crystal orientation [17], [18], applied indentation loads [6], [13]–[15], the tip radius of indenter [10], [13]–[15] and temperature [16] during nanoindentation measurements, the interplays between the observed single or multiple pop-in events and the onset of micro/nanoscale plasticity for GaN thin films are not completely understood. Therefore, it is not surprising to see that there have been some confusions in understanding the onset of micro/nanoscale plasticity and the plastic deformation mechanisms of GaN thin films were misinterpreted in some cases.

In this study, the pop-in behavior and mechanical characteristics of GaN thin films derived from nanoindentation with a Berkovich indenter are discussed in conjunction with the observations of cathodoluminescence (CL) spectroscopy. The correlation between the observed Berkovich nanoindentation-induced rosettes on GaN thin films and the number of dislocation loops formed in the pop-in event was further analyzed with the aids of the classical dislocation theory [19], [20]. The results shed some light on the understanding of the nanoscale deformation behaviors of GaN thin films.

II. Experimental Details

The GaN thin films used in this study were grown on (0 0 0 1)-sapphire substrates by using metal–organic chemical vapor deposition (MOCVD) method with an average thickness of about 2 μm. The detailed growth procedures of GaN thin films can be found elsewhere [13]. Nanoindentation tests were performed on an MTS NanoXP Nanoindenter system (MTS Cooperation, Nano Instruments Innovation Center, TN, USA) with a diamond pyramid-shaped Berkovich-type indenter tip. The radius of curvature of the tip is 50 nm. A Berkovich indenter was employed with its area shape being calibrated using fused silica. The frame stiffness and thermal drift were corrected for

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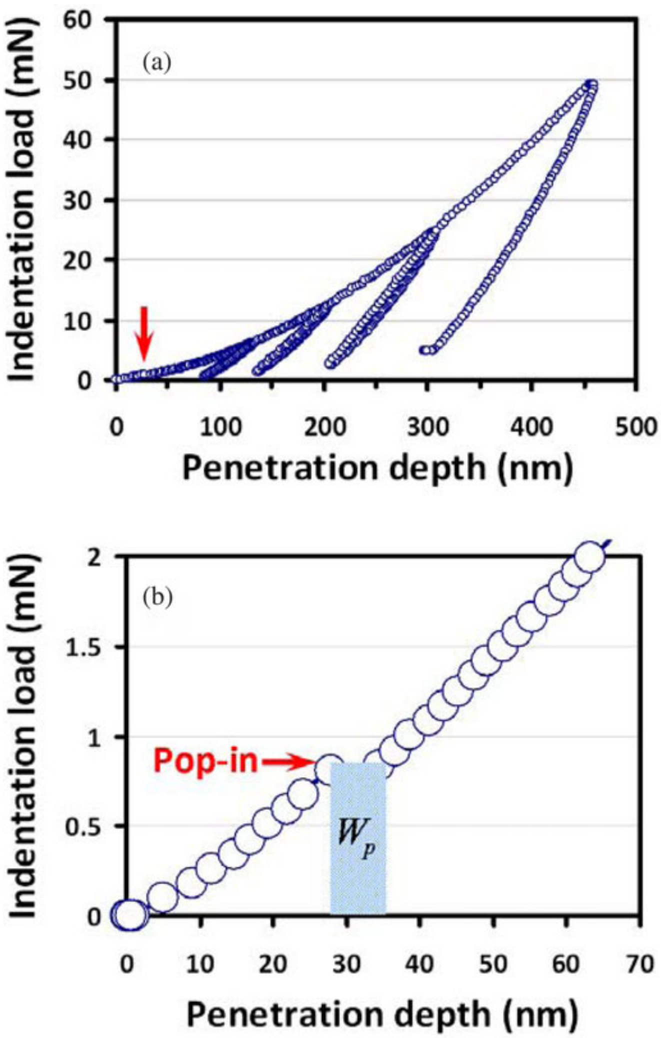

Fig. 1. (a) Load–displacement curve measured by Berkovich indenter on GaN thin films at the indentation load of 50 mN, and (b) corresponding pop-in event (see the arrow) from (a) is zoomed in, where the plastic strain work is denoted as, W_p (critical loading times the displacement).

all measurements. The cyclic nanoindentation tests were performed in a sequence described as followings. We first apply the indentation loading to the set maximum load and unloading by 90%. Then, reload the indenter to the maximum load and unload it by 90% again. At this stage, the indenter was kept on hold for 10 s at 10% of the maximum load for thermal drift correction. After that, the indenter was completely unloaded and, thus, finished a cyclic nanoindentation test for a certain targeted maximum load. Fig. 1(a) displays a typical load–displacement curve comprising four cyclic tests at the maximum loads of 6, 12, 25, and 50 mN, respectively. The thermal drift was kept below $\pm$0.05 nm/s for all indentations considered in this study. The same loading/unloading rate of 10 mN/s was used. At least ten indents were performed and the nanoindentations were sufficiently spaced to prevent from mutual interactions. The room temperature CL measurement was performed using a Gata monocle equipped on the JEOL JSM-7000F field-emission SEM. A 20-keV electron beam energy level was selected to excite the sample.

III. Results and Discussion

Typical load–displacement curve from GaN thin films loaded to an indentation load of 50 mN is shown in Fig. 1(a). Similar to many previous studies [10], [14], [15], the results indicate a slight pop-in event of yield response for GaN thin films, which has been commonly attributed to the sudden nucleation of dislocations propagating along the slip systems lying on the $\{0\ 0\ 0\ 1\}$ basal planes and the $\{1\ 0\ \bar{1}\ 1\}$ pyramidal planes. As shown in Fig. 1(b), a single pop-in is observed at the critical indentation load ($P_{\rm cr}$) of $\sim$0.8 mN. The initial yielding is related to the onset of plasticity since the deformation behavior prior to yield excursion is elastic. The elastic deformation behavior does not involve any dislocation motion until the applied stress approaches the theoretical shear strength of the materials. Beyond this point, homogeneous nucleation of dislocations or activation of pre-existing dislocation sources starts to emerge and is followed by subsequent glide and multiplication events [21]. The tip radius of our Berkovich-type indenter is only $\sim$50 nm; therefore, the pop-in effect should have been dominated by homogeneous dislocation nucleation [22]. Under such circumstances, it can be assumed that the applied shear stress required to trigger the dislocation nucleation is the maximum shear stress $\tau_{\rm max}$ beneath the indenter during the pop-in and can be described by

$$\tau_{\rm max} = 0.18\left(\frac{P_{\rm cr}E_r^2}{R^2}\right)^{1/3}. \tag{3}$$

From the classical dislocation theory [19], [20], the formation free energy F of a circular dislocation loop with radius r is determined by the line energy of the newly formed loop $\gamma_{\rm dis}$ and the work ($\sim\tau_{\rm max}\,b$ per loop area) to extend it, as following:

$$F = \gamma_{\rm dis}2\pi r - \tau_{\rm max}b\pi r^2. \tag{4}$$

Taking into account the Hertzian contact stress [23] during the indentation, the dislocation line energy for a circular loop is obtained as

$$\gamma_{\rm dis} = \frac{Gb^2}{8\pi}\frac{2-v_{\rm film}}{1-v_{\rm film}}\left[\ln\left(\frac{4r}{r_{\rm core}}\right)-2\right]. \tag{5}$$

Here, the value of $\tau_{\rm max}$ is about 6.4 GPa, $v_{\rm film}$ is Poisson's ratio (=0.25) [13], G is the shear modulus ($=E_{\rm film}/2(1+v_{\rm film})\approx$ 120 GPa), and b is the Burgers vector (=0.319 nm) [24] of the GaN thin film.

As expressed in (4), nucleation of a dislocation loop involves two prominent terms, namely the line energy of the dislocation loop and the work-done to extend the dislocation loop in the expense of reducing the stress energy. This is similar to the nucleation process of a spherical new phase of radius, r, within the parent phase, wherein the surface energy associated with the newly formed interface between the nucleus and the parent phase and the free energy reduction resulting from forming new phase are the two relevant energy terms. In both cases the free energy has a maximum at a critical radius r_c which gives $dF/dr = 0$ and the formation free energy has a critical maximum value at r_c, which has to be at least of the order of the thermal energy. Since the thermal energy is very small compared to the two energy terms on the right-hand side of (4), the $F(r_c) \sim 0$ is thus assumed as an additional condition for calculating r_c. These considerations lead to

$$r_c = \frac{2\gamma_{\text{dis}}}{b\tau_{\max}} \quad \text{and} \quad r_c = \frac{e^3}{4} r_{\text{core}} \approx 5 r_{\text{core}}. \tag{6}$$

Here, r_{core} is the cut-off radius of the dislocation core, which has been directly revealed as an eight-fold ring with diameter in the order of 0.4–0.5 nm for threading dislocations in GaN [25]. Therefore, the number of loops formed can be estimated from the work-done associated with the pop-in event. From the shaded area depicted in Fig. 1(b), this work is estimated to be $\sim 5.24 \times 10^{-12}$ Nm, implying that $\sim 3 \times 10^5$ dislocation loops with critical diameter might have been formed during the pop-in event. This number is low and is consistent with the scenario of homogeneous dislocation nucleation-induced pop-in, instead of activation of pre-existing dislocations [12]. When the total dissipation energy, namely the area between the loading and unloading curves shown in Fig. 1(a), is taken as the energy to generate dislocations with critical radius, dislocation loops as high as $\sim 4 \times 10^8$ may be formed within a load–unload cycle. Although it is not realistic to assume that all the dissipated indentation energy was entirely transferred to generate dislocation loops, the estimation has, nevertheless, provided an upper limit for the number of dislocation loops with critical radius in the initial state. Subsequent to their formation, the embryonic loops grow further and coalesce under the action of the applied indentation load. The unloading may give rise to the relaxation of dislocation loops, which eventually stabilizes into the dislocation rosette patterns shown later.

Fig. 2 shows two typical CL images of near-gap emission obtained from the pristine and the indented area with the maximum indentation load of 50 mN on GaN thin film, respectively. The Berkovich nanoindentation-induced "rosette" pattern characteristically reflecting the hexagonal symmetry of GaN is clearly observed in the indented area, whereas in the pristine region it is essentially featureless in the CL image. It is noted that, depending on the indenter shapes, the nanoindentation-induced CL patterns on GaN thin films can be very different [10], [18]. Nevertheless, the symmetrical CL patterns that resulted from various indenters do reflect intimately the nanoindentation-induced dislocation activities [18]. As can be seen in Fig. 2, the CL image from the indented region appear to display two well separated zones: 1) a strongly perturbed central zone, appearing as a very dark zone presumably containing extremely dense damage distribution, and 2) the six secondary arms, nearly parallel to the crystal axes separated by an angle of $\sim 60°$, which is indicative of certain active defect propagation systems for the hexagonal symmetry.

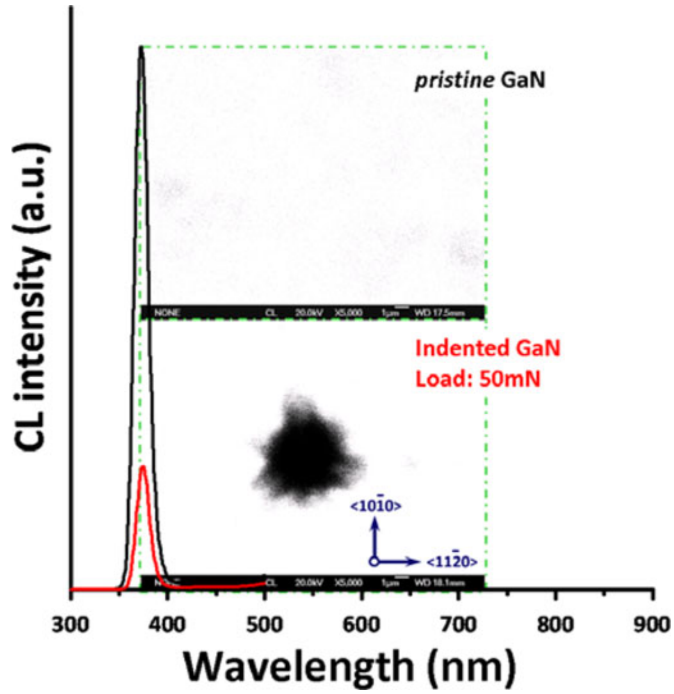

Fig. 2. Room temperature CL images and CL spectra of the pristine GaN surface and indented area (at the indentation loading of 50 mN). CL imaging conditions: electron beam energy = 20 keV, CL wavelength = 366 nm.

The rosettes arms in Fig. 2 are aligned along the $\langle 1\,1\,\bar{2}\,0\rangle$ directions. As reported in our previous study [15], the CL and cross-sectional transmission electron microscopy (XTEM) studies indicated that the active slip systems in GaN films induced by the Berkovich nanoindentations are on the $\{0\,0\,0\,1\}$ basal planes and the $\{1\,0\,\bar{1}\,1\}$ pyramidal planes with the slip bands extending exclusively along the $\langle 1\,1\,\bar{2}\,0\rangle$ orientation. This is also consistent with the plane-view TEM observations reported by Jahn *et al.* [26], which confirmed that the Burgers vector of indentation-induced dislocations in c-plane GaN is $\langle 1\,1\,\bar{2}\,0\rangle$. Both are consistent with the extending directions of the rosette arms observed in Fig. 2. Furthermore, comparing with the featureless CL picture taken from the pristine region of the same sample, it is suggestive that the rosette pattern is indeed risen from indentation-induced dislocations.

Typical CL spectra for the pristine surface and the indented areas of GaN thin film are also displayed in Fig. 2. Both exhibit a room temperature near-band edge emission peak at ~ 370 nm, albeit the emission density is markedly different, presumably due to the defect-induced local luminescence quench in the indented areas. The effects of the defect-induced local luminescence quench are further evidenced by the progressive suppression of the near-gap CL emission intensity with increasing loading. An exponential decay relationship between the relative CL intensity and indentation loads are observed as shown in Fig. 3. It is evident that the data can be fitted very well with the expression: $I = C\exp(0.86 - 0.05P)$, which might be related to the density of the indentation-induced dislocations. It is noted that, although the near-band edge luminescence intensity decreases

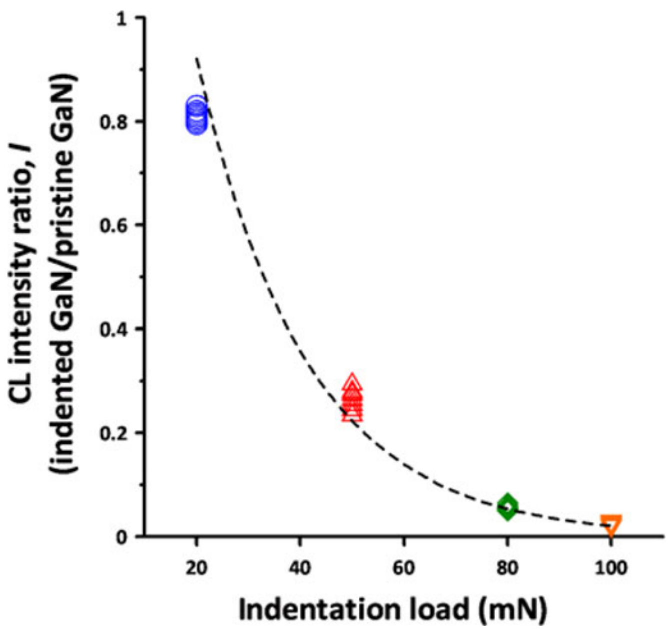

Fig. 3. Relationship of CL intensity ratio (indented GaN/pristine GaN) versus with various indentation loads.

significantly after indentations, there is no additional emission band observed, indicating that the Berkovich nanoindentation-induced dislocations merely act as non-radiative recombination centers and did not induce newly formed phases or emission bands.

IV. Conclusion

In summary, a combination of Berkovich nanoindentation and CL techniques was carried out to investigate the Berkovich nanoindentation-induced pop-in effect in c-plane GaN thin films. The number of the indentation-induced dislocation loops estimated from the work-done within the course of the pop-in event suggested that the pop-in was mainly due to homogeneous nucleation of dislocations. The CL images of Berkovich nanoindentation on GaN thin film showed a very well-defined rosette structure with the hexagonal symmetry, which consists of a strongly perturbed central dark zone and six double arms emanating from this region along the $\langle 1\ 1\ \bar{2}\ 0\rangle$ directions. Furthermore, the CL spectra recorded at room temperature showed the near band-edge luminescence intensity falling exponentially with the applied load and reduced to about 2% of that obtained in pristine area as the maximum indentation load up to 100 mN, presumably due to the increased number of induced dislocations.

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