The Impact of the Discreteness of Low-fluence Ion Beam Processing on the Spatial Architecture of GaN Nanostructures Fabricated by Surface Charge Lithography

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We show that the descrete nature of ion beam processing used as a component in the approach of surface charge lithography leads to spatial modulation of the edges of the GaN nanostructures such as nanobelts and nanoperforated membranes. According to the performed Monte Carlo simulations, the modulation of the nanostructure edges is caused by the stochastic spatial distribution of the radiation defects generated by the impacting ions and related recoils. The obtained results pave the way for direct visualization of the networks of radiation defects induced by individual ions impacting a solid-state material.

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Gallium nitride is a wide-band-gap semiconductor compound (E_g = 3.4 eV at 300 K) widely used in optoelectronics for the production of UV LEDs and lasers. The compound is promising for applications in high-power / high-temperature electronics and many investigations have been carried out in this regard. Note that the material exhibits pronounced radiation hardness that can be considerably strengthened by nanostructuring [1]. Recently [2, 3] we reported the fabrication of suspended GaN membranes with the thicknesses down to 1 nm by using a modified version of the so-called Surface Charge Lithography (SCL) developed by us previously [4, 5]. SCL is based on direct ion-beam-writing of surface negative charge with subsequent photoelectrochemical (PEC) etching of the GaN sample [3–5]. In this Letter, we show for the first time that the discreteness of ion beam processing used as a component of SCL has a considerable impact on the spatial nanoarchitecture of narrow or nanoperforated GaN membranes, in particular it modulates the edges of the nanostructures which reflects the spatial distribution of the radiation defects generated by the impacting ions and related recoils.

The wurtzite *n*-GaN layers were grown by low pressure MOCVD on (0001) *c*-plane sapphire substrates. A buffer layer of 25 nm thick GaN was first grown at 510 °C. Subsequently a 3 μm thick *n*-GaN layer was grown at 1100°C. The concentration of free electrons was of the order of 10^{17} cm^{-3}, while the density of threading dislocations was in the range of (10^9–10^{10}) cm^{-2}. Arrays of parallel lines 10 nm in width were directly written on the sample surface by 30 keV Ga^+ ions provided by a focused-ion beam (FIB) system with a fluence of approx. $3x10^{12}$ cm^{-2}. At the same time selected areas of the sample surface were treated in automated patterning mode by the focused ion beam resulting in gentle ion-beam-processing of circular-like areas constituting a periodic network of ion-implanted negatively charged surface regions and merging with each other at the edges. The dose of the ions in the treated areas was lower than in the previous case. According to the concept of SCL [2–5], processing of the sample surface by low-energy ions creates deep acceptors that trap electrons and form a shield of negative charge that protects the material against PEC dissolution. Monte Carlo simulations predict the main projected range of 30 keV Ga^+ ions in an amorphous GaN matrix to be about 14 nm [5]. In crystalline GaN the range may be enhanced due to ion-channelling effects. PEC etching was carried out at 300 K in a stirred 0.1 mol aqueous solution of KOH for periods up to 1.5 h under in-situ UV illumination provided by focusing the radiation of a 350 W Hg lamp to a spot of 5 mm in diameter on the sample surface. The sample morphology was studied using a Zeiss ultra plus scanning electron microscopes (SEM).

A JEOL 7001F field emission SEM equipped with a Gatan XiCLone cathodoluminescence (CL) microanalysis system was used for comparative morphological and CL characterization. The monochromatic microcathodoluminescence (μ-CL) images were collected using a Peltier cooled Hamamatsu R943-02 photomultiplier tube.

Figures 1a and 1b are SEM images illustrating

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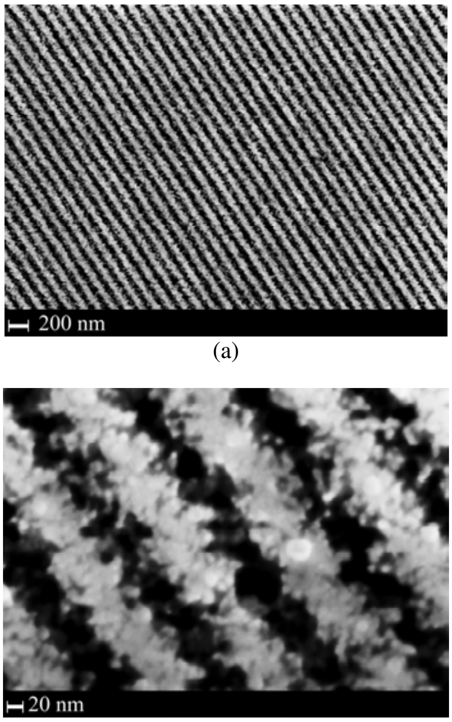
an array of GaN “nanobelts” fabricated by employing SCL. Interestingly, the nanobelts exhibit many protrusions along the edges. To throw light upon this phenomenon, one should take into account the discrete nature of the ion beam processing applied. This feature will be discussed below.



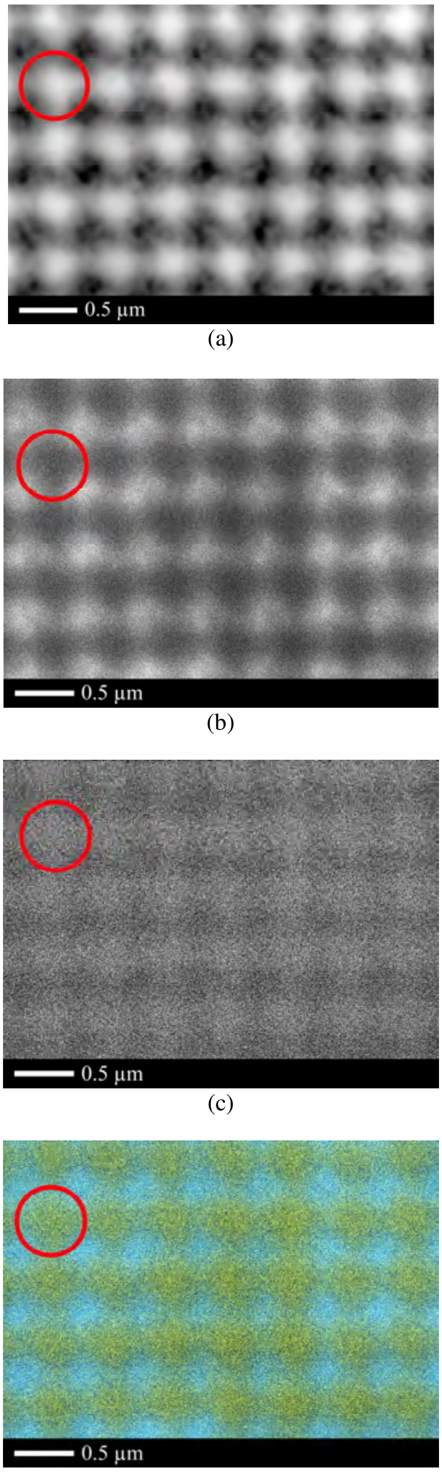
(a)

(b)

Fig. 1. SEM images taken at different magnifications from arrays of GaN nanobelts.

Figure 2a illustrates the morphology of the GaN surface area treated by FIB in automated patterning mode with subsequent PEC etching. Exploration by SEM revealed the formation of an ultrathin membrane with ordered nanopores that is supported on a network of whiskers representing threading dislocations [6]. The membrane is nanoperforated as a result of bridging between neighbouring circular-like areas patterned by FIB. Figures 2b,c,d show monochromatic and color composite cathodoluminescence images taken from the same portion of the membrane. Characteristic GaN CL is observed. As expected, the ultra-thin membrane exhibits mainly very low intensity yellow luminescence at ~2.25 eV [6], while the more intense UV luminescence at ~3.4 eV comes from the underlying regions of PEC-etched GaN. An interesting feature is the irregular nature of the shape of the membrane holes, see Fig. 2a.

Analysis of the SEM image presented in Fig. 1b suggests that the irregular edges of the nanobelt are not “white noise” with respect to spatial frequencies, but are evidence of structural roughness or irregularities of the nanobelt. An autocorrelation analysis confirms this conclusion. We suggest that these protrusions are due to the effect of the interaction of individual impacting Ga^+ ions with the crystalline matrix of GaN.



(a)

(b)

(c)

(d)

Fig. 2. SEM image taken from a portion of the nanoperforated GaN membrane (a), and comparison of monochromatic (b – 3.4 eV; c – 2.25 eV) μ-CL images with two-color (blue – 3.4 eV; yellow – 2.25 eV) composite image (c). The red circles mark the same position in the GaN membrane.

For the low dose fluences used one should take into account both the stochastic distribution of the impacting Ga ions and the fact that each impacting ion generates many radiation defects by recoils.

Figure 3a and 3b show the results of SRIM simulations [7] of the trajectories (no recoils) of 5 and 500 ions in GaN matrix, while Figures 3c and 3d illustrates ion trajectories accompanied by recoils projected on the XY plane. Distribution of gallium ions and related recoils projected on the transverse YZ plane is presented in Figures 3e and 3f. It is clear that recoil cascades define the spatial distribution of point defects caused by each impacting ion, while overlapping of defect clouds related to different Ga^+ ions may be related to the spatial nanoarchitecture of the nanobelts emerging during PEC etching. Bridging between circular areas and formation of holes with stochastic shapes is also the result of overlapping of clouds of radiation defects generated by recoils, see Figures 2a, 3e, 3f.

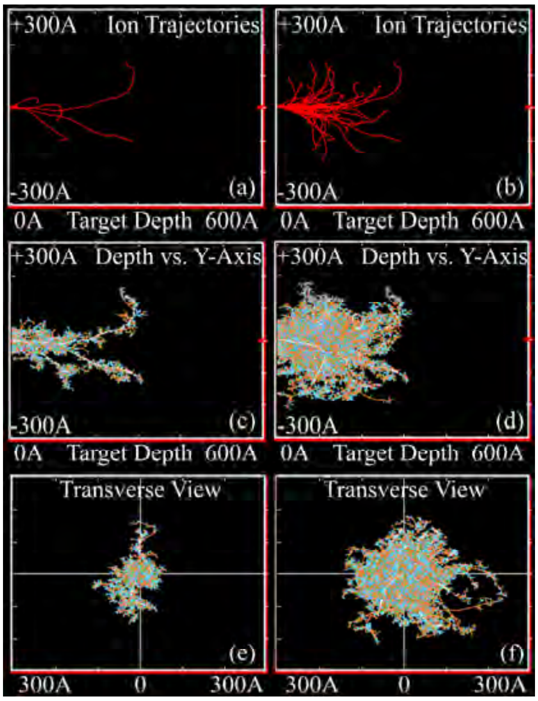


Fig. 3. The results of SRIM simulations: the trajectories (no recoils) of 5 (a) and 500 ions (b) in an amorphous GaN matrix; trajectories of 5 (c) and 500 ions (d) accompanied by recoils projected on the XY plane; distribution of 5 (e) and 500 Ga^+ ions (f) and related recoils projected on the transverse YZ plane.

Thus, photoelectrochemical etching of GaN is extremely sensitive to the crystalline quality of the material. Generation in a controlled fashion of negatively charged radiation defects by focused ion beam can be used for the purpose of material mesostructuring and nanostructuring. Fabrication of ultra-thin membranes, however, requires use of low-fluence ion beam processing which assures transparency of the membrane to UV irradiation necessary for PEC etching in depth. The discreteness nature of the ion beam treatment, especially under low-fluence conditions, proves to have a considerable impact on the spatial architecture of the membranes involved, in particular in regard to narrow belts and nanoperforated membranes. The obtained results show that the architecture of nanobelts and nanoperforated membranes reflects the stochastic distribution of radiation defects generated by impacting ions and related recoils. Our approach of SCL followed by PEC etching can be applied to enable direct visualization of the networks of radiation defects induced by individual ions impacting a solid-state material.

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Реферат

Показано, что дискретная природа ионно-лучевой обработки, использованной в методе литографии поверхностного заряда, приводит к пространственной модуляции краев наноструктур GaN, таких как нанopемни и наноперфорированные мембранны. Согласно расчетов по методу Монте-Карло, модуляция краев наноструктур вызвана стохастическим пространственным распределением радиационных дефектов. Полученные результаты указывают на путь визуализации сети радиационных дефектов, генерируемыми отдельными имплантируемыми ионами.