The use of SiC/Si(111) hybrid substrate for MBE growth of GaN nanowires

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Abstract. This work demonstrates the possibility of using a silicon substrate with nanoscale buffer layer of silicon carbide for growth of GaN nanowires by molecular epitaxy on. Morphological and optical properties of the grown arrays are studied. It is shown that the integral intensity of the photoluminescence of such structures is more than 2 times higher than the best NWs GaN structures without buffer layer of silicon carbide.

1. Introduction
The wide bandgap nanoheterostructures based on GaN are of great interest for creating electronic [1] and optoelectronic [2] devices. High mechanical strength combined with chemical resistance and high (2500°C [3]) melting temperature ensures the stability of the devices. Lack of substrates of gallium nitride encourages researchers to seek suitable substrates for the growth of such structures. The use of sapphire substrates leads to the generation of high dislocation density [4] due to the significant (13%) lattice mismatch. Substrates of silicon carbide, the most suitable in terms of matching the lattice parameters can’t be widely used because of the small size and high cost. On the other hand, the successful attempts of growing GaN layers on silicon [5] are very promising, as silicon is a rather cheap material having acceptable thermal conductivity. Moreover, Si substrates can be of a large size and enables the integration of optoelectronic devices based on gallium nitride with the silicon micro- and nanoelectronics base. However, lattice mismatch of Si (111) and GaN (0001) is 17%, and the difference in thermal expansion coefficients is 33% resulting in a high density of different defects suffer its characteristics in created devices. It is known that the optoelectronic GaN based devices can operate for a long time without degrading despite the high density of the linear defect. The dislocation density in the light-emitting diodes based on gallium nitride may exceed the maximum dislocation density in the GaAs based devices to five orders of [6]. The fact that, although dislocations and are recombination centers in GaN, the diffusion length of minority carriers is less than the distance between dislocations (less than 250 nm) [7]. However, to extend the lifetime of GaN based optoelectronic devices an increase their perfection is highly desirable.
In this work to increase the optical quality of the material, we use the nanometer (about 50 ÷ 100 nm) SiC buffer layer which was grown on Si substrate. If we combine two factors (i) the small lattice mismatch between GaN and SiC(111) (only 3%) [2] together with (ii) the nanowires (NWs) growth, the resulting structure can be better in a view of the quality of the structure [8]. Finally, controlled synthesis of GaN NWs allows one to control their physical properties, including the level of doping for both n- and p-type [9], and to create on their basis LED [9] and UV lasers [10].

The aim of this work is to demonstrate the possibility to grow GaN nanowires on the SiC buffer layer on a top of silicon substrates and to compare the optical properties with the GaN NWs grown on silicon without a buffer layer.

2. Experiments
Formation of the buffer layer of silicon carbide on Si (111) substrate was performed by the method described in [11-13]. This method of silicon carbide on silicon films producing is different from all currently existing methods and technologies for growing single crystals, films and nanostructures. Briefly, the method is based on the substitution of silicon atoms on carbon ones on a silicon substrate. At the first stage carbon atom is embedded into interstitial site of silicon. Then nearby silicon atom is removed forming a silicon vacancy. As a result, at the surface of the silicon dilatation dipoles are created (stable complexes consisting of dilatation centers — the silicon vacancy and a carbon atom in interstitial site). These two centers are elastically interacting each other in crystal of cubic symmetry. The rate of the chemical reaction is maximal in the direction along attraction of dilatation dipoles (i.e. along the (111) direction of the silicon substrates). Orientation of the film depends on the crystal structure of the original Si matrix, not only the surface of the substrate, as typically used in conventional film growth techniques. The temperature and pressure of the gas are selected to nucleate the silicon carbide and pores. Formation of the elastic dipoles (carbon atom — silicon vacancy) provides a high quality SiC film.

Growth experiments were carried out using Riber Compact 12 MBE setup equipped with the effusion Ga cell and the nitrogen source. Firstly, the substrate was transferred into the growth chamber, and the substrate temperature was set at 950°C for further cleaning. Then, the substrate temperature was lowered to 600°C and was opened gallium source for 20 seconds to form nanoscale islands on surface to further NWs growth. Finally, and the substrate temperature was raised up to the growth temperature setting at 800°C. Then nitrogen source plasma was ignited and nitrogen flow and the gallium source were opened simultaneously. The growth rate for Ga was set at 0.01 ML/s (according to the previous calibrations). Total growth time of each GaN NWs samples was equal to 16 hours.

3. Results
After the growth, the morphology of grown nanowires was studied by scanning electron microscopy (SEM) SUPRA 25 Zeiss, and their optical properties were investigated by room temperature photoluminescence (PL) and Raman techniques.

Figure 1 shows typical SEM images of GaN nanowires grown on SiC/Si combined substrate. It is seen that on the substrate Si/SiC (111) GaN NWs formed mainly in [111] direction and their average height is about 1.6 mkm. It should be noted that grown structures have high surface density ~ 2•10^10 cm^-2, that’s why some of the nanowires became merge. From SEM image (figure 1,a) it is clear that some NWs are inhomogeneous by their diameter - it increases to the top of NWs, possibly due to the rather high substrate temperature and/or relatively low N/Ga fluxes ratio.
Figure 1. SEM images of GaN NWs grown on SiC/Si: (a) tilted by 20 cross section view; (b) top-view.

Figure 2,a shows a comparison of PL spectra of the grown sample and the most successful of GaN NWs sample on silicon measured at room temperature. Maxima of both spectra are coincide in wavelength and conform to experimentally measured radiation value GaN [14]. At the same time, the integral PL intensity of GaN NWs grown on SiC buffer layer more than 2 times higher than the intensity of the best structures of grown on silicon at the same growth conditions.

Figure 2,b shows a typical Raman spectrum of the sample GaN NWs grown on SiC/Si. Despite of the small thickness of the SiC, cubic SiC related-line is clearly visible, along with the lines of the hexagonal GaN and Si. It is worth noting the extremely small width of GaN lines (1.5 cm$^{-1}$), indicating a high structural perfection GaN NWs.

Figure 2. (a) the typical PL spectra of grown samples and the most successful GaN structures grown on silicon; (b) the typical Raman spectrum of grown on SiC/Si GaN NWs.
4. Conclusions
In this paper we demonstrate the possibility of the growth of GaN nanowires by molecular beam epitaxy method on silicon with nanoscale silicon carbide buffer layer for the first time. It was found that the intensity of the photoluminescence spectrum of the GaN NWs on SiC/Si(111) substrate integrally more than 2 times higher than that of the best structures of GaN NWs without a buffer layer of silicon carbide. The extremely small width of GaN lines on Raman spectrum indicates a high crystallographic quality GaN NWs grown on SiC/Si open a new route of the use of these structures in future optoelectronic applications.

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References
[1] Pearton S J and Ren F 2006 Adv. Mater. 12 1571
[2] Nakamura S and Fasol G 1997 New York: Springer-Verlag 368
[3] Ioffe data archive http://www.ioffe.ru/SVA/NSM.
[4] Oliver R A, Kappers M J and McAleese C 2008 J. Mater. Sci. Mater. Electron. 19 208
[5] Aksyanov I G, Bessolov V N, Zhilyaev Y V, Kompan M E, Konenko E V, Kukushkin S A, Osipov A V, Rodin S N., Feoktistov N A, Sharofdinov S and Shcheglov M P 2008 Technical Physics Letters 34 479
[6] Cherns D, Young W T, Saunders M A, Ponce F A and Nakamura S 1997 Microscopy of Semiconducting Materials 157 187
[7] Rosner S J, Carr S E, Ludowise M J, Girolami G and Erikson H I 1997 Appl. Phys. Lett. 70 420
[8] Dubrovskii V G, Cirlin G E and Ustinov V M Semiconductors 43 1539
[9] Zhong Z, Qian F, Wang D and Charles M. Lieber 2003 Nano Letters 3 343
[10] Choi H J, Johnson J C and He R 2003 J. Phys. Chem. B. 107 8721
[11] Kukushkin S A and Osipov A V 2008 Physics Of The Solid State 50 1238
[12] Kukushkin S A, Osipov A V, Feoktistov N A 2014 Physics Of The Solid State 56 1507
[13] Kukushkin S A and Osipov A V 2014 J. of Phys. D: Appl. Phys. 47 313001
[14] Tchernycheva M, Sartel C, Cirlin G E, Travers L, Patriarche G, Harmand J-C, Dang Le Si, Renard J, Gayral B, Nevou L and Julien F 2007 Nanotechnology 18 385306