Luminescence of SiC films grown by a vapor phase reaction

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Abstract. SiC films grown by vapor phase reaction consist of multicrystalline grains. The size of grains depends on growth parameters, such as temperature and the relative concentration of the Si and C components. A small fraction of large grains demonstrate very efficient luminescence. Several techniques have been used to study the origin of the luminescence. Cathodoluminescence and photoluminescence revealed flat regions as sources of emission. Diffraction of back scattered electrons and X-ray analysis suggests that efficient emission is observed for grains with a small fraction of the rhombohedral phase. On the basis of the experimental data we assumed that the layers of rhombohedral phase or the interface between these layers and the host material are the origin of the luminescence.

1. Introduction
SiC, as a wide band gap semiconductor with unique properties, has attracted considerable attention as a material for high temperature electronics and short wavelength optical applications. In electronics great success has been achieved in the field of high power transistors and hostile-environment devices [1]. However due to its indirect energy gap, SiC has a significant drawback in optical device applications: light emission is a phonon mediated process with a low probability. To overcome this difficulty, two approaches can be used: reducing the non-radiative channels or making the radiative process much faster. One of the solutions for the second approach is to exploit the quantum confinement effects [2] to increase the radiative probability. As an alternative to the epitaxial growth of quantum well structures, defect engineering looks rather promising for this purpose. This work is devoted to an analysis of efficient radiation arising from individual grains of SiC films.

2. Experimental method
Samples were grown by a vapor phase reaction. This technique has the advantage of being cheap and producing a potential to use different types of substrates such as Si, Al\textsubscript{2}O\textsubscript{3} and graphite. A vertical graphite crucible was employed as a reactor. Si was placed at the bottom of the reactor while natural gas was discharged from the top. The pressure inside the reactor was held in the range of 2–10 mbar. The reactor was placed into a resistance heating furnace and heated to 1600°C. Decomposition of hydrocarbons started in the upper hot part of the reactor. Due to the opposite gradients of Si and hydrocarbons, their relative concentration along the height of the reactor was height dependent.

The morphology of the surface was analyzed by optical and scanning electron microscopy (SEM). X-ray diffraction (XRD) and diffraction of back scattered electrons (EBSD) were used for structure analysis. Photoluminescence (PL) was measured using 325 nm excitation of a He-Cd laser with a
conventional phase sensitive registration. The photomultiplier F79 was used as a detector. Cathodoluminescence (CL) was measured by SEM.

3. Results and discussion

Figure 1 shows an example of SEM images for films grown on graphite. The size of grains and film morphology strongly depend on the growth parameters. Figure 1 illustrates the influence of relative Si concentration on morphology. Large grains were created in the zone of high Si concentration (left image); quite a different structure was formed at a lower Si concentration (right image). For the sake of clarity we will call these films type 1 and type 2. Typically large grains are well shaped, have an arbitrary orientation and their dimensions can reach several hundred microns. In the case of a low Si concentration we have small rounded grains. Their dimensions are about 1 micron and the particles consist of smaller granules.

![SEM images of SiC films illustrating the influence of the relative relation of Si concentration and C vapors. The left and right films were grown with the excess of Si and C, respectively.](image)

It turned out that the PL properties of these types of films are quite different. While type 2 films demonstrate a weak but uniform broad luminescence band with a maximum around 400 nm, the luminescence of type 1 films is strongly non-uniform. Most of the grains show the same weak luminescence as type 2 films with an additional band in the red part of the spectrum, but there is a limited number of grains with a very efficient luminescence. We will concentrate on type-1 grains with efficient luminescence.

Analysis of PL by optical microscopy revealed that even within a single grain the emission is non-uniform (figure 2). There are some small luminescent parts and the light emitted from them penetrates into the grain, is scattered by the imperfections on the surface and in the bulk giving an illusion that the whole grain glows.

Figure 3 shows a spectral distribution of PL for bright grains of type 1. The spectra are normalized to the spectral sensitivity. Generally emission consists of two bands, blue and yellow. The intensity of the blue band grows slightly with decrease of temperature, but remains unresolved. The yellow part of the luminescence is most active at low temperature and disappears completely at 150K. Therefore grains with different visual appearance have the same spectral features but with different relative intensity.
Cathodoluminescence, measured in the monochrome mode, clearly shows (figure 4) that there is a sharp boundary between dark and white parts of the grain. The white lines on the SEM image show an approximate location of the luminescent part. Its shape replicates some features of the grain relief. It appears that the last layer grown gives the origin of luminescence.

In the case of a more complicated lamellar structure shown in figure 5, several active layers are seen simultaneously. Taking into account that the grain surface morphology is defined by the growth direction we can assume that the luminescent parts have a flat shape oriented perpendicular to the growth direction. So far as a distance of the electron penetration into the sample is about 1 micron, we cannot say definitely whether the active planes are located at the surface or inside the grain. The dependence of integrated CL intensity on acceleration voltage (figure 6) implies that the active layer is located under the surface. Here the growth of CL intensity can be explained by the contribution of the surface recombination, which affects the initial increase of CL intensity with increasing penetration.

**Figure 2.** Photoluminescence excited by 325 nm radiation of HeCd laser. The excitation spot is shown by the circle.

**Figure 3.** PL spectra of grains with different visual appearance of the glow. Solid line for blue grains, dashed for yellow grains.

**Figure 4.** SEM image and CL of the same part of film.
Further decrease of CL at high voltage implies that electrons passed through the active layer due to a deeper penetration of electrons.

**Figure 5.** SEM image and CL of grain with lamellar structure.

**Figure 6.** Normalized dependence of CL integrated intensity upon accelerating voltage.

The two possible reasons can be suggested to explain the difference of particular grains from the remaining material: it can be either local segregation of some impurity or some structural features. We checked the composition of the material in dark and light places. No significant difference was found. On the other hand, the comparison of diffraction of backscattered electrons for dark and luminescent grain segments clearly shows the structural difference of these parts (figure 7).

**Figure 7.** Diffraction of backscattered electrons measured in dark and light regions of the grain. The left part is SEM and CL images of the grain. The particular spots marked as 1 and 2 in the CL image for EBSD analysis are shown in the figure on the right.

Therefore we can make a conclusion that the PL and CL active parts are layers of different polytypes, or the interface between the host material and the different polytype. From the data of figure 7 we cannot say unambiguously what structure corresponds to the two patterns, but we can
definitely state that the origin of the intense luminescence is caused by the different structure of the small layer regions.

Figure 8. XRD patterns of dark grain (left image) and grain with efficient luminescence (right image). The additional reflections of a rhombohedral phase are shown by arrows.

To elucidate the nature of luminescent layers, we studied dark grains and grains with luminescent fragments separately by the XRD technique. It was found that dark grains have a pure hexagonal structure while PL active grains have an additional small fracture of the rhombohedral phase. Figure 8 illustrates the two XRD images for different types of grains, which clearly shows additional reflections for luminescent grains, corresponding to the rhombohedral phase. Taking into account the above consideration, we can infer that thin interlayers of the rhombohedral phase created during the growth of some grains are the origin of luminescence. It should be noted that the possibility of creating a quantum well like structure due to different polytypes in SiC was suggested by Iwata et al [3]. One possible mechanism of creation of a different polytype layers is the motion of partial dislocations in adjacent basal planes [4]. Identification of the growth conditions responsible for creation of these interlayer fragments is yet to be investigated.

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