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Fluorescent SiC for white light-emitting diodes

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Abstract: The strong photoluminescence from f-SiC was achieved after the optimization of the B and N concentrations. Surface nanostructures were successfully applied to enhance the extraction efficiency. f-SiC is a promising wavelength convertor for white LEDs.

OCIS codes: (230.3670) Light-emitting diodes; (160.2540) Fluorescent and luminescent materials

1. Introduction

Light-emitting diodes (LEDs) have attracted renewed interest in the past decades with the appearance of the world's first efficient **blue**-emitting GaN LED, for the reason that it could make all-solid-state lighting for large-scale energy saving. Compared with the traditional incandescent and fluorescent light sources, LEDs have longer lifetime, higher overall efficiency, as well as better technical functionality for many new lighting applications. Driven by energy saving and further CO₂ emission reduction, high brightness LEDs, as a GREEN light source, represents a multi-billion market that is predicted to grow to \$12 billion by 2013 globally.

Currently, most commercial white LEDs are made by mixing yellow color, converted by the phosphors from blue, and the rest blue color from the blue-emitting diodes. But phosphors degraded much faster than the LED chips, leading to the white LEDs turning blue over time. Recently, fluorescent SiC has also been approved as an ideal wavelength converter for white LEDs because it has better color rendering ability and long life time [1].

2. Experiments

The boron (B) and nitrogen (N) co-doped 6H-SiC epilayers were grown on 6H-SiC (0001) substrates having 1.4 degree off-orientation in the $\langle 11\bar{2}0 \rangle$ direction by the fast sublimation growth process [2] at temperature of 1725 °C. The growth process is driven by a temperature gradient created between the source, in a form of polycrystalline SiC plate, and the substrate. B was introduced into the epilayers by co-doping from the source and N incorporation was controlled by adjusting N₂ gas pressure during the growth.

The 6H-SiC epilayer codoped with B and N has strong fluorescence, so called fluorescent SiC(f-SiC), and its photoluminescence (PL) measurements were realized by using an Olympus reflected fluorescence system microscope, a 377 nm diode laser as excitation source (focused by a 20X objective), and an Instrument System CAS 140B spectrometer at room temperature.

To enhance the extraction efficiency of the f-SiC, cone-shaped antireflection nanostructures (ARS) are fabricated on the surface of f-SiC epilayers. A positive resist layer was first spin-coated on the 6H-SiC sample and then prebaked. The designed mask pattern was transferred to the resist coating by applying the e-beam lithography. After the development, a hard mask layer was deposited on the patterned resist with a subsequent lift-off process. The dot-shaped pattern was then obtained on the hard mask. The dry etching process using SF₆ and O₂ precursors was carried out in the reactive-ion etching system.

The surface reflectance of the bare and ARS SiC samples were measured from 390 to 785 nm by a goniometer system at a measured angle of 6 degrees, where 0 degree is the direction normal to the sample surface. The angle-resolved photoluminescence spectra were also acquired by the same goniometer system. The 377 nm diode laser was used as the excitation source which was normal to the sample backside and the detected emission angle varied from 16 to 80 degrees.

The scanning electron microscope (SEM) image was acquired by field-emission SEM Zeiss.

3. Results and discussions

High quality single crystalline 6H-SiC epilayers co-doped with B and N have been demonstrated after the characterization of transmission electron microscopy, lifetime etc.[2]. To investigate the optimized dopant concentrations of N and B, five samples with varied N and B concentrations were studied. The atomic dopant concentrations measured by secondary ion mass spectrometry (SIMS) are listed in Table 1.

Table 1. Dopant concentrations and normalized PL peak intensities of the samples

Sample	B concentration [cm ⁻³]	N concentration [cm ⁻³]	PL peak intensity [Normalized to sample d]
a	8.0×10^{18}	4.0×10^{16}	0.0 %
b	6.9×10^{18}	3.2×10^{18}	6.6 %
c	6.9×10^{18}	6.0×10^{18}	8.3 %
d	4.4×10^{18}	9.0×10^{18}	100 %
e	5.2×10^{18}	9.2×10^{18}	77.1 %

Fig. 1. Measured PL spectra of B-N doped 6H-SiC samples (inset: zoom-in for sample a, b, and c), same peak wavelength at 587 nm and FWHM of 120 nm were observed in all the spectra.

Their corresponding PL spectra are shown in Fig. 1. All samples show PL emission peak at 587nm and FWHM of 120nm. As N concentration increases from sample a to sample d, the emission intensity increases as well. Sample d has the strongest PL emission. Then the emission intensity drops as the N concentration increases further. The detailed physical analysis of the relationship between the emission intensity and dopant concentration could be found [3].

Bare and ARS samples are compared in terms of reflectance, spatial emission distribution and angle-resolved PL and the results are shown in Fig. 2. An oblique-view SEM image of the ARS pattern is shown in the inset of Fig. 2(a). The cone-shaped ARS structure has base diameter of 250 nm, pitch of 350 nm, and height of 1.4 μm . The reflectance spectra of bare sample and ARS structure are shown in Fig. 2(a). It is seen that the surface reflection is effectively suppressed by applying the SiC ARS. The average reflectance over the measured spectral range decreased from 20.5 % to 1.0 % and the minimum reflectance close to 0 was observed at around 400 nm for the ARS SiC sample. Fig. 2(b) shows the spatial emission patterns for both samples. It can be seen that the luminescence intensity for ARS sample increased at all emission angles with a well preserved spatial emission pattern. It is also found that the luminescence intensity was enhanced by more than 91 % in a very large emission angle range (up to 70 degree). From Fig. 2(c), it is seen that the luminescence intensity of the bare SiC decreases together with a blue shift of the peak wavelength as the emission angle increases from 20 to 70 degree, which could be attributed to the Fabry-Pérot microcavity interference effect. In Fig. 2(d), the peak wavelength remains the same which is due to the elimination of the Fabry-Pérot microcavity interference effect by introducing the ARS on the SiC surface.

To reduce the cost of ARS fabrication, self-assembled nanostructured metal as etching mask was applied instead of the expensive e-beam lithography. The pseudo-periodic cone-shaped nanostructures are shown in the inset of Fig. 3. They have almost the same average height, pitch and base diameter as the periodic nanostructures. The same good anti-reflectance have been achieved, shown in Fig. 3. The surface reflectance was reduced from 20.5% of the bare SiC to 1.7% of ARS SiC.

Fig. 2: (a) Surface reflectance (inset: SEM figure of fabricated ARS on SiC); (b) spatial emission pattern; angle-resolved photoluminescence of (c) bare and (d) ARS SiC samples.

Fig. 3: Surface reflectance of bare SiC and pseudo-periodic nanostructured ARS SiC (inset: SEM figure of pseudo-periodic ARS on SiC);

4. Conclusion

Optimized N and B concentration for f-SiC has been found. Cone-shaped ARS structures have been fabricated and demonstrated that it is an effective way to suppress the surface reflection for the fluorescent SiC sample in the whole visible spectral range. These promising results pave the way for f-SiC as a wavelength converter for white LEDs.

5. Acknowledgements

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References

- [1] S. Kamiyama, S. Kamiyama, T. Maeda, Y. Nakamura, et al., "Extremely high quantum efficiency of donor-acceptor-pair emission in N- and B-doped 6H-SiC", *J. Appl. Phys.* **99**, 093108 (2006).
- [2] M. Syväjärvi, J. Müller, J. W. Sun, V. Grivickas, Y. Ou, V. Jokubavicius, P. Hens, M. Kaisr, K. Ariyawong, K. Gulbinas, P. Hens, R. Liljedahl, M. K. Linnarsson, S. Kamiyama, P. Wellmann, E. Spiecker, H. Ou, "Fluorescent SiC as a new material for white LEDs" *Physica Scripta* **2012**, 014002 (2012).
- [3] Y. Ou, V. Jokubavicius, S. Kamiyama, C. Liu, R. W. Berg, M. Linnarsson, R. Yakimova, M. Syväjärvi, H. Ou, "Donor-acceptor-pair emission characterization in N-B doped fluorescent SiC", *Opt. Mater. Express* **1**, 1439-1446 (2011).