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Omnidirectional luminescence enhancement of fluorescent SiC via pseudoperiodic antireflective subwavelength structures

Yiyu Ou,[1,*] Valdas Jokubavicius,[2] Rositza Yakimova,[2] Mikael Syväjärvi,[2] and Haiyan Ou[1]

[1]*Department of Photonics Engineering, Technical University of Denmark, Lyngby DK-2800, Denmark*

[2]*Department of Physics, Chemistry and Biology, Linköping University, Linköping SE-58183, Sweden*

**Corresponding author: yiyo@fotonik.dtu.dk*

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In the present work, an approach of fabricating pseudoperiodic antireflective subwavelength structures (ARS) on fluorescent SiC by using self-assembled etch mask is demonstrated. By applying the pseudoperiodic (ARS), the average surface reflectance at 6° incidence over the spectral range of 390–785 nm is dramatically suppressed from 20.5% to 1.62%, and the hydrophobic surface with a large contact angle of 98° is also achieved. The angle-resolved photoluminescence study presents a considerable omnidirectional luminescence enhancement with an integral intensity enhancement of 66.3% and a fairly preserved spatial emission pattern.

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White light-emitting diodes (LEDs) consisting of a nitride-based blue LED chip and wavelength converter material are very promising candidates for the general lighting applications as energy-saving and environment friendly light sources. High-efficiency nitride-based LEDs are extensively reported [1–3] and phosphors are the most common wavelength converters with high conversion efficiency [4–7]. Recently, donor–acceptor doped fluorescent SiC has been proven as a highly efficient wavelength converter material much superior to the phosphors in terms of high color rendering index (CRI) value and long lifetime [8,9]. The donor–acceptor pair (DAP) band luminescence from nitrogen (N)-boron (B) doped 6H-SiC presents a warm, white color. Combined with the DAP luminescences from the nitrogen–aluminium doped 6H-SiC, pure white light with CRI larger than 90 could be produced [10,11]. Furthermore, SiC is a well-established substrate material for nitride growth and has excellent thermal conductivity.

The light extraction efficiency of the semiconductor LED is usually low due to the internal reflection loss arising from the large refractive index difference between the semiconductor and air interfaces. Traditionally, a single-layer quarter-wavelength thin-film antireflection coating can be applied to enhance the lighting for a specific wavelength at very low level. Broadband application can be achieved by applying a stack of antireflection coatings with appropriate design [12,13]. However, it is usually limited by the availability of materials with appropriate refractive indices and thermal expansion coefficients. Antireflective subwavelength structures (ARS) have been proved as an ideal approach to enhance the light transmittance over a broad spectral bandwidth [14–19]. In addition, several recent works to increase the light extraction efficiency in GaN-based LEDs had been demonstrated by using photonic crystals with large index contrast [20], and self-assembled colloidal microlens arrays [3,21,22]. In the present work, applying pseudoperiodic ARS on the fluorescent SiC to enhance the light extraction efficiency over the entire visible spectral range has been studied.

To fabricate periodic ARS, time-consuming e-beam lithography process is usually indispensable. Here we present a simple approach to fabricate the pseudoperiodic cone-shaped ARS on N-B doped fluorescent 6H-SiC by using self-assembled etch mask. The fabrication process steps are schematically illustrated in Fig. 1 together with the corresponding scanning electron microscope (SEM) figures. First, a 10 nm Au film layer is deposited on the fluorescent SiC by using e-beam evaporation (Alcatel thin film deposition system). After treated by rapid thermal processing (Jipelec RTP) at 350 °C for 5 min in N_2 ambient, the thin Au layer turns into discontinuous nano-islands with half sphere-like shape. Their average size and density are well controlled by adjusting the annealing condition as well as the Au layer

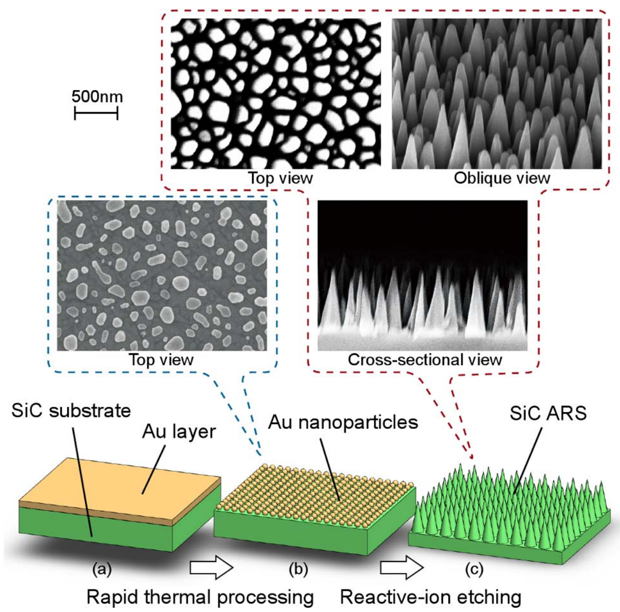

Fig. 1. (Color online) Schematic illustrations of the pseudoperiodic SiC ARS fabrication process steps: (a) Au film deposition, (b) rapid thermal processing, and (c) RIE and corresponding SEM figures.

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thickness. The pseudoperiodic cone-shaped ARS with good uniformity are formed on the fluorescent SiC surface by using the Au nano-islands as a mask layer by applying reactive-ion etching (RIE) with SF_6 and O_2 gases mixture (4∶1) for 15 min. The residual Au are removed by using iodine-based solution ($KI:I_2:H_2O$–100 g:25 g:500 ml) and the sample is then characterized by SEM (Carl Zeiss SMT GmbH). It is found that the pseudoperiodic ARS has a mean pitch of approximately 115 to 230 nm and the structure height varies from 400 to 850 nm.

The antireflection usually depends on the dimension of the ARS. From the previous work [11], it was found that structure height is the crucial factor to affect the antireflection ability for cone-shaped ARS. It is found that at least 400 nm high structures are usually required to achieve fairly good antireflection performance. Hence, RIE conditions used in the process are well optimized and pseudoperiodic ARS with an average height larger than 400 nm are obtained by using the following conditions: process pressure of 30 mT, RF power of 100 W, gases flow rates of 24 sccm SF_6 and 6 sccm O_2, and process time of 15 min.

The surface reflectance has been measured by using a calibrated goniometer system (Instrument Systems GON360) at near-normal incidence of 6° over a wavelength range of 390–785 nm, which covers the entire visible spectral range (typically from 390 to 750 nm). The reflectance spectra are shown in Fig. 2(a), where the average surface reflectance is significantly suppressed from 20.5% to 1.62% by a factor of 11.6 after introducing the ARS. The reflectance at the luminescence peak (576 nm) is lower than 2% and the minimum value of 0.05% is obtained at 405 nm. Although the reflectance starts to increase at longer wavelength, the value through the whole measured spectral range is below 4%. The antireflection performance of pseudoperiodic SiC ARS is quite comparable to the values of other reported periodic ARS [15,16]. From the inset photograph of Fig. 2(a), it is also seen that the fluorescent SiC surface turns from shiny light green color to dark green–black color after introducing the ARS on the surface. Figures 2(b) and 2(c) show the water contact angle measurements realized by using a drop shape analyzer (Krüss DSA 100S). The fluorescent SiC surface turns from hydrophilic with a contact angle of 49° to hydrophobic with a contact angle of 98° after introducing the pseudoperiodic ARS, which is an appreciated merit for LED applications especially used at low temperature and in humid environment.

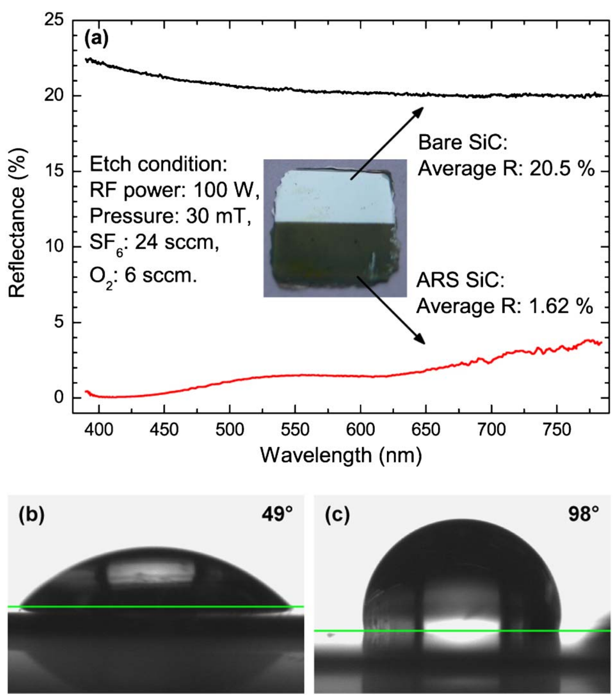

Fig. 2. (Color online) (a) Surface reflectance of the bare and ARS SiC samples measured at near-normal incidence of 6°, inset: photograph of the bare (top half) and ARS SiC (bottom half). Water droplet contact angle measurements on (b) bare SiC (49°) and (c) ARS SiC (98°).

The angle-resolved photoluminescence (PL) measurement has been performed by using the same goniometer system, and a 377 nm laser beam from a diode laser has been used as the excitation source. The sample was optically excited from its backside and the emission angle-resolved PL was measured from 0 (normal to the sample front surface) to 90° in steps of 10°. The PL spectra of the bare and ARS SiC measured at 0° are shown in Fig. 3(a). Broad DAP band luminescences with a peak wavelength of 576 nm and a full width at half-maximum (FWHM) of around 110 nm are observed for both samples. The ARS SiC has a luminescence enhancement of 55% at the emission angle of 0°, which indicates a higher light extraction efficiency. This enhancement is probably due to the escape of the emitted light with an emission angle larger than the critical angle through the surface ARS.

Furthermore, the luminescence enhancement of fluorescent SiC at different emission angle after introducing the ARS is presented in Fig. 3(b) where the ARS demonstrate the omnidirectional luminescence enhancement. The enhancement increased from 55% at 0° to 186% at 90°, and the integral luminescence enhancement in the whole range is 66.3%. Spatial emission patterns of the two samples are shown in the inset of Fig. 3(b). It is seen that the spatial emission pattern is fairly preserved when the emission intensity is enhanced by introducing the pseudoperiodic ARS.

In conclusion, fabricating pseudoperiodic ARS on the fluorescent SiC is a timesaving and effective method to achieve the surface antireflection in a large spectral range and omnidirectional luminescence enhancement. The average surface reflectance at 6° incidence is significantly decreased from 20.5% to 1.62% by a factor of 11.6 over the spectral range of 390–785 nm. The hydrophobic surface with a large water contact angle of 98° is obtained. The luminescence intensity is enhanced by 55% at 0° emission angle and the enhancement further increased with a larger emission angle. The spatial emission pattern is fairly preserved from the angle-resolved PL measurement with an integral intensity enhancement of 66.3%. It is shown that the pseudoperiodic ARS have comparable antireflection performance with other reported periodic structures and can improve the light extraction efficiency of fluorescent SiC to a large extent and hence the external quantum efficiency of SiC-based white LEDs.

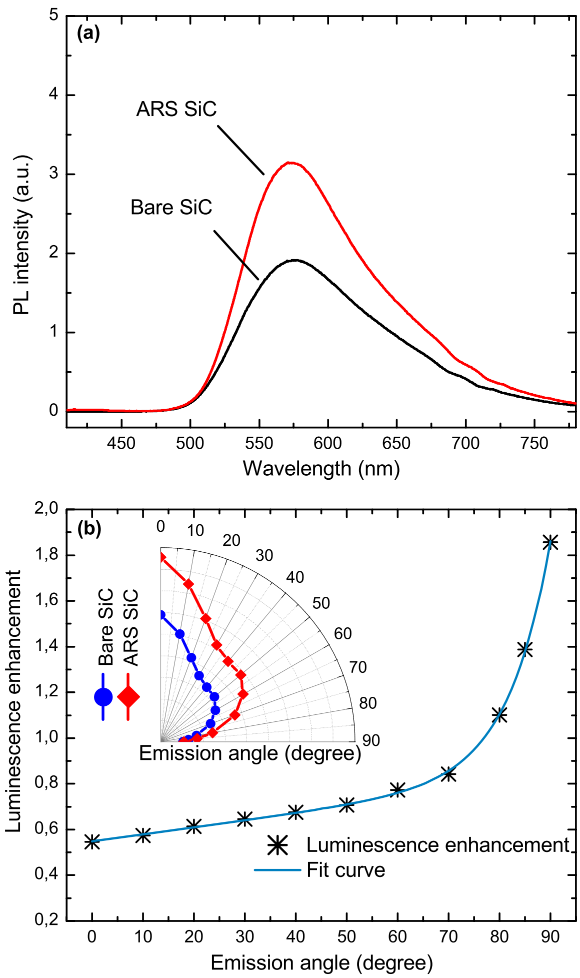

Fig. 3. (Color online) (a) PL spectra of the bare and ARS SiC measured at 0°. (b) Luminescence enhancement of the SiC at different emission angle after introducing the ARS. Inset: spatial emission patterns for both bare and ARS SiC.

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