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Piezo-phototronic effect in InGaN/GaN semi-floating micro-disk LED arrays

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ABSTRACT

With enhanced light output efficiencies, InGaN/GaN micro-disk LED has received intensive attentions recently. Combining isotropic and anisotropic dry etching processes, an innovative semi-floating InGaN/GaN micro-disk LED array is fabricated, which shows remarkable light intensity enhancement up to 150% compared to the broad-base LED. A systematic study of micro-spectrums and Poisson-Schrodinger coupling self-consistent calculation reveal that there is non-uniform residual stress distribution on the micro-disk LED. Along micro-disk center to micro-disk edge, as the Si substrate is etched off, the in-plane tensile stress in the GaN layer reduces, while the compressive stress in InGaN layer increases gradually. This gradient stress distribution has caused a non-uniform piezo-phototronic effect in the micro-disk LED, which in turn results in a maximum wavelength shift of 16 meV for the light emitted along micro-disk center to micro-disk edge. This study not only opens research of flexo-optoelectronic effect, e.g. non-uniform piezo-phototronic effect, in complex micro/nano optoelectronic/electronic devices, but also provides important guidance for the significant enhancement of light emission efficiency in micro-disk LEDs.

Development of high-efficiency GaN-based LEDs has attracted huge attention due to its wide applications in solid state lighting and full color displays [1,2]. In spite of the high internal quantum efficiency (70%) for blue LEDs, the external quantum efficiency is still not ideal due to the low light extraction efficiency [3–5]. An obvious improvement in extraction efficiency has already been proved in the generation of micro-disk LEDs, including micro-ring LED, micro-pillar LED [6], micro-pyramid LED and micro-rod LED [7–10]. More importantly, these micro-disk LED arrays have great potential in many applications, from head-wearing displays, head-up displays to viewfinders and camcorders [11,12]. Furthermore, it is highly competitive to develop high-light extraction Si-substrate InGaN/GaN micro-disk LED array since it can get fully compatible with silicon-substrate integrated circuit [13]. However, in conventional micro-InGaN epitaxy-up devices, Si substrate absorbs light emitted downward which causes optical loss in the Si based LED [14]. Therefore, it is essential to develop novel micro-disk LED structures to reduce the reabsorption of Si substrates and thus enhance the light extraction efficiency.

When growing InGaN/GaN based-LED structures on hetero-substrates such as Si, sapphire and SiC, large mismatch between

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nitrides films and epitaxial substrates results in complex stress distribution in the nitrides-based LEDs. The residual stress will induce piezoelectric field, which has a great influence on the light emission of LED, thus study of stress is an important issue in the field of micro-disk LEDs. Piezo-phototronic effect is then introduced to reveal the stress distribution and optical transition of the InGaN/GaN micro-disk LEDs since it is a three-way coupling effect between semiconductor properties, piezoelectric polarization, and optical excitation found in wurtzite structured III-nitride semiconductors [15–17]. Piezo-phototronic effect was first found in a single ZnO micro-/nanowire photodetector in 2010 [18–21]. Soon after that, this effect was introduced to Si/ZnO nanowire heterostructure to tune light emission [22]. Very recently, researchers demonstrated that some planar InGaN/GaN quantum well based optoelectronic devices could also be modulated effectively by the piezo-phototronic effect [23–25]. However, all of the above studies are focused on uniform piezo-phototronic effects in simple piezoelectric materials and devices, little research has been done on the non-uniform piezo-phototronic in complicated micro/nano optoelectronic/electronic devices so far [26,27].

Here, we succeed to combine isotropic and anisotropic dry etching processes to partly lift off the Si substrate and get a semi-floating InGaN/GaN/Si micro-disk LED array. Compared to broad-base LED, the semi-floating micro-disk LEDs show an enhanced light intensity up to 150% since this semi-floating micro structure simultaneously weakens the Si substrate to absorb the downward light and increases the lateral active emission area. For the first time, we demonstrate experimentally and theoretically the non-uniform piezo-phototronic effect in the InGaN/GaN micro-disk LEDs, which would cause the shift of the emitted light wavelength, providing potential application in multi-wavelength micro-displays. This research not only provides effective method to improve the performance of light emission in micro-disk LEDs, but also opens up study of non-uniform piezo-phototronic effect, which plays an important role in modulating and improving performance of complex micro/nano optoelectronic/electronic devices.

1. Results and discussion

An innovative process is induced to fabricate the semi-floating micro-disk LEDs. The anisotropic etching is carried out to produce deep silicon trenches and the isotropic etching step is then performed to free the structure laterally [28,29]. Compared to some wet etching or dry etching process, this process has advantages such as accurately controlling, well-aligned shape, easily-integrated into Si-based circuit, together with low contamination from environmental and solution. The top-view SEM image of the InGaN/GaN micro-disk LED array (Fig. 1a) shows that the size of each micro-disk is 200 μm and the adjacent

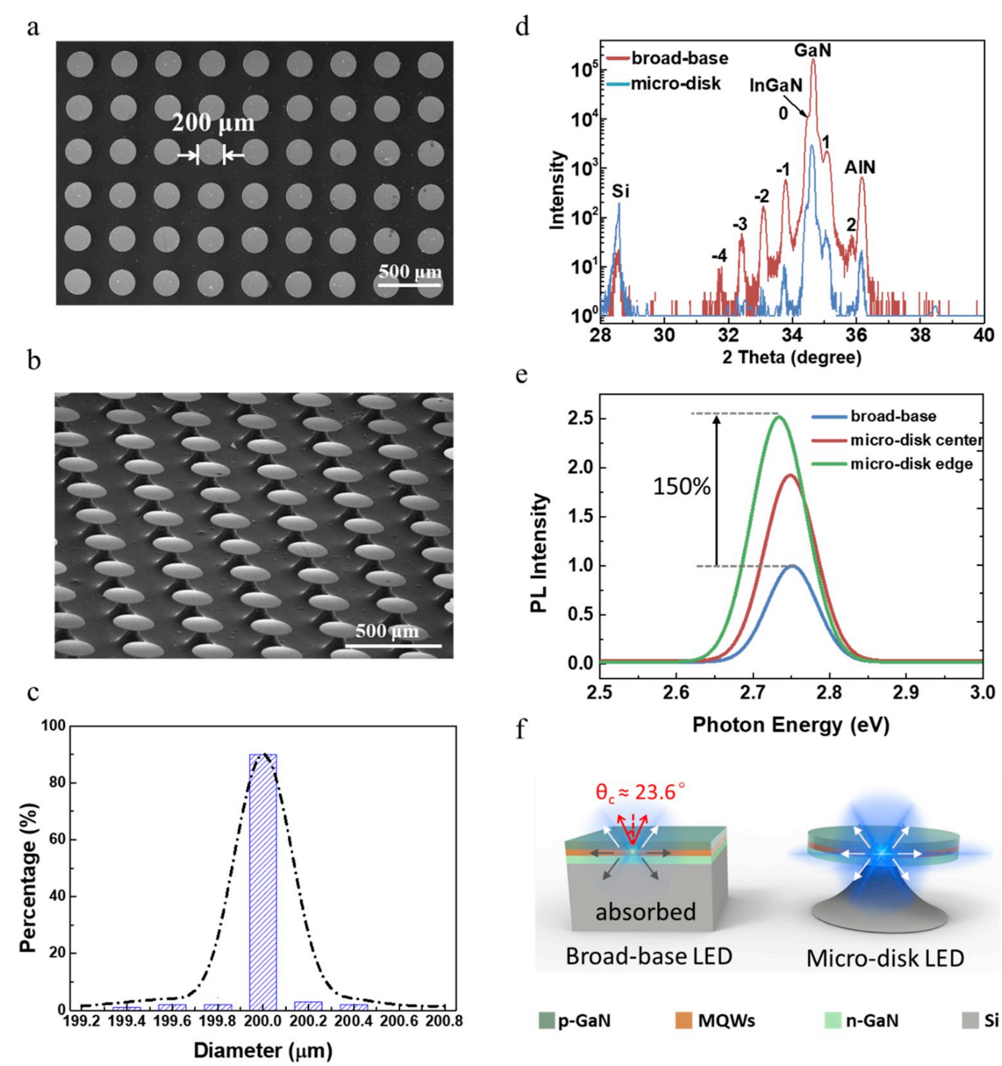

Fig. 1. (a), (b) Planar and titled SEM photographs of InGaN/GaN micro-disk LED array. (c) Statistical distribution of diameters of the micro-disk LEDs. (d) XRD ω-2θ curve for InGaN/GaN micro-disk LED array. (e) PL spectra for micro-disk LED and broad-base LED. (f) Schematic diagram of the effective light emission region for broad-base LED and micro-disk LED.

micro-disk spacing is 150 μm. In Fig. 1b, the tilted cross-sectional SEM image of the micro-disk LED array shows that the deeply undercut GaN-based micro-disk structure has large air gap and long suspending distance (~80 μm). With up to 90% etching accuracy (Fig. 1c) and precisely controlling shape, complex laser arrays or flexible micro/nano devices may be prepared using this method. From ω-2θ curve of GaN (0002) plane shown in Fig. 1d, it is noted that many high order satellite peaks arising from the periodicity of the InGaN/GaN MQWs (multi-quantum wells), indicating that there still remains a fine quantum well/barrier periodic structure in the InGaN/GaN micro-disk LED after substrate etching [30].

In the traditional micro-pillar LED and micro-ring LED, the increase of the emitted-light intensity are only 39% and 70% relative to broad-base LED [8,31]. While in the micro-disk LED, the enhancement reaches up to 150% as shown in Fig. 1e. The mechanism of enhanced light emission intensity in micro-disk LED has been demonstrated in Fig. 1f. In broad-base LED, most of light emitted from the LED active region will be trapped in the GaN layer since it has a high refractive index [32]. In the micro-disk LED, by contrast, the effective light-emitting area is increased mainly due to the following two points. Like other micro-disk LEDs, due to the micron size, most laterally transmitted photons can avoid the reabsorption and reach the sidewalls successfully [33]. More advantageous than conventional micro-disk LEDs, the semi-floating micro-disk LED can effectively weaken the Si substrate to absorb the downward light since the Si substrate has been etched off deeply [14,34]. In addition, the strains also play an important role in micro LEDs. The piezo-phototronic effect has been proved to effectively modulate the light emission (internal quantum efficiency) [22–25] and this study has also revealed a non-uniform piezo-phototronic effect in the semi-floating micro-disk LED. Chen & Shen et al. proved that the strain relaxation makes a great contribution to enhance the IQE in micro LEDs, which clarified the significant issue of high-performance micro-LEDs well [6]. This semi-floating InGaN/GaN micro-disk LEDs provide reliable technical support for improving the illumination of micro-LEDs furthermore.

To characterize the piezo-phototronic modulations caused by remove of Si substrate, micro-PL spectroscopy and micro-Raman scattering are used to investigate the residual stress in InGaN/GaN micro-disk LEDs. As shown in Fig. 2a, a total of fifteen test points are taken along the radius of micro-disk LED. Point 1 to point 5 and point 11 to point 15 (the spacing of adjacent points is 13 μm) are evenly distributed in the edge of the micro-disk LED, where there is no Si substrate underneath. Point 7 to point 9 (the spacing of adjacent points is 11 μm) are evenly distributed in the central region of the micro-disk LED, where there is Si substrate underneath. Point 6 and point 10 are distributed at the junction with/without the Si substrate. Fig. 2b is PL spectrums measured from these test points, while introducing the spectrum of broad-base LED as a contrast. A vertical structured light path is used in order to increase the spectral resolution. Comparing with the energy peak (2.756 eV) of broad-base LED, all of the energy peaks on the micro-disk LED red-shift towards the low-energy. Fig. 2d shows the distribution of PL peaks as a function of test point position which helps to analyze the trend of peak changes. Along the vertical radius of the micro-disk, from micro-disk center to the junction, the energy peak at point 6 drops significantly by 12 meV compared to point 8. When the laser source is swept to micro-disk edge, the energy is almost constant and maintained at around 2.728 eV, which has small decrease relative to the junction point. The test points localized at the horizontal radius of the micro-disk LED show the same variation trend. The different optical performance between junction point 6 and point 10 is reasonable since their properties depend on whether there is Si residue underneath. Currently, the full-color display based on GaN LED is mainly achieved through hybrid devices, a blue LED is combined with red or green phosphors, or packaging three independent RGB LEDs into one device [35]. These traditional techniques have problems such as high cost, large area, low efficiency, low reliability … etc. [36] The multi-emission micro-disk LED provides another potential and attractive method to realize high-resolution full-color display on a single device.

The above results demonstrate that, from center to edge of the micro-disk, the energy peak shows a decreasing trend (a red shift) with a maximum difference of 16 meV. The red-shift of the PL spectrum from center to edge indicates that the residual compressive strain in InGaN layer gradually increases after remove of Si substrate. Furthermore, this gradual stress distribution leads to the non-uniform polarization field in the InGaN/GaN quantum wells, thus resulting in a varying band to band transition in InGaN quantum well. In complex optoelectronic devices, when there is a non-uniform internal stress distribution, the piezoelectric effect also works as non-uniform, which can sensitively modulate the optical performance under different internal stress states. The non-uniform piezoelectric effect could provide a theoretical basis for the application of complex heterojunction micro/nano optoelectronic devices in flexible or curved environment.

Due to the large mismatch in lattice constants and thermal expansion coefficients between Si and GaN film, as well as between the InGaN and GaN layer, the epitaxial layer experiences a high level of in-plane stress [37]. The residual stress in GaN layer can be evaluated from the shift of E_{2h}-high phonon mode since it is sensitive to the stress state of the GaN film [38]. The E_2-high peak has been reported to appear at 567.5 cm^{-1} for a 400 μm-thick freestanding bulk GaN material. Fig. 3a shows micro-Raman spectra measured from fifteen test points on the micro-disk LED, while introducing the spectrum of the broad-base LED as a contrast. Comparing with the Raman shift of 566.19 cm^{-1} for the broad-base LED, all of the E_{2h}- mode peaks shift towards the larger

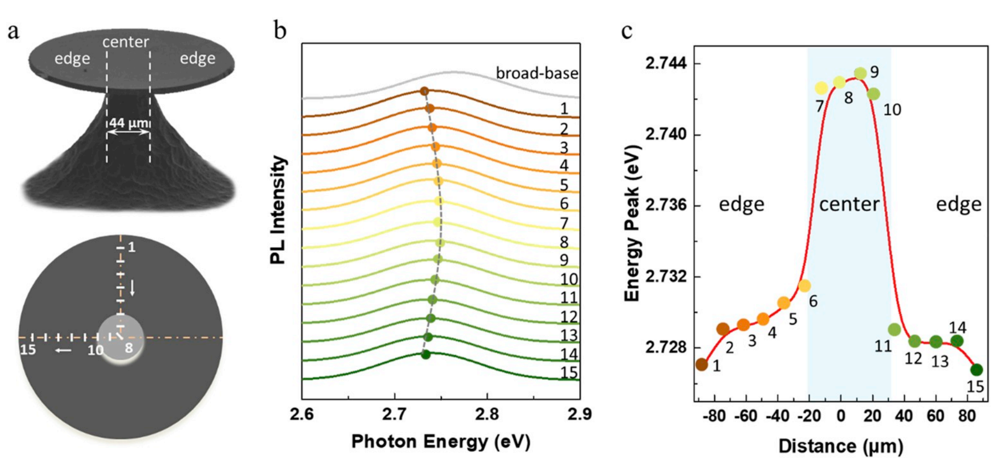

Fig. 2. (a) Cross-sectional SEM image of a single micro-disk LED and fifteen test points along the radius on the micro-disk LED. (b) PL spectrums of point 1 to point 15 on the micro-disk LED and PL spectrum of broad-base LED. (c) Distribution of PL peaks as a function of test point position.

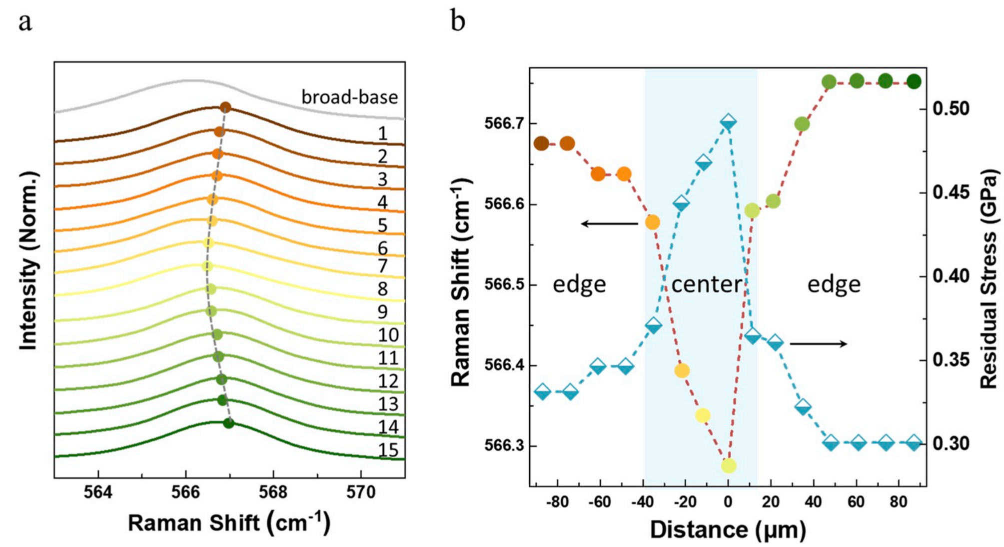

Fig. 3. (a) Raman spectrums of point 1 to point 15 on the micro-disk LED and Raman spectrum of broad-base LED. (b) Distribution of Raman peaks and residual stress in GaN layer as a function of test point position.

wavenumber. According to the principle of stress measurement by Raman spectroscopy, GaN film of the micro-disk LED suffering tensile stress since the observed phonon frequencies is lower than the strain-free value ω_0. Fig. 3c shows the distribution of E_{2h} peak positions and in-plane stress as a function of test point position. It can be seen that the peaks of test points distribute symmetrically about the central point. Along micro-disk center to micro-disk edge, from point 8 to point 1, the value of E_{2h} increases from 566.273 cm^{-1} to 566.675 cm^{-1}, that is 0.402 cm^{-1} increase, owing to the release of Si substrate. And point 8 to point 15 also has the same situation, with E_2-high value increasing from 566.273 cm^{-1} to 566.750 cm^{-1} (0.477 cm^{-1} increase). The in-plane tensile stress can be estimated from the relation $\Delta\omega = a_{GaN}\sigma_{xx}$, where $\Delta\omega$ is the shift of phonon frequency with respect to that of stress free GaN, a_{GaN} is the Raman stress coefficient, and σ_{xx} is the stress [39]. Taking a_{GaN} as $-2.49\ cm^{-1}\ GPa^{-1}$, the residual stress of each point is calculated as shown in Fig. 3c [40]. It demonstrates that from micro-disk center to micro-disk edge, with the peeling of Si substrate, the residual stress decreases gradually with a maximum stress difference of 0.2 GPa.

The lattice mismatch in InGaN/GaN hetero-structure obtained from Raman and PL results is visually shown in Fig. 4. At room temperature, the lattice constants of Si (111), GaN (002) and $In_{0.15}Ga_{0.85}N$ (002) is 3.84 Å, 3.189 Å, and 3.24285 Å, respectively. Therefore, GaN layer grown on Si is subjected to in-plane tensile stress while the InGaN layer grown on GaN layer is compressive in plane as shown in Fig. 4a [41,42].

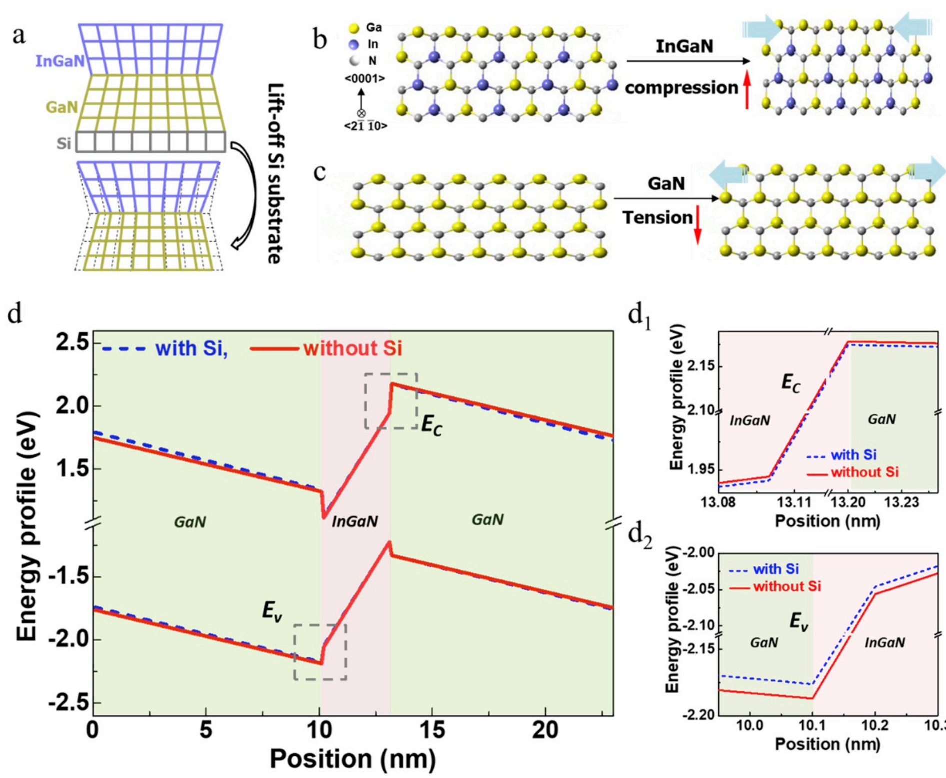

Fig. 4. (a) Schematic diagram of lattice mismatch in the InGaN/GaN epitaxial layer before and after Si substrate is etched off. (b) Schematic diagram of InGaN atomic structure before and after the Si substrate is lifted off. (c) Schematic diagram of GaN atomic structure before and after the Si substrate is lifted off. (d) InGaN/GaN quantum well energy band structures obtained from Poisson-Schrodinger coupling self-consistent calculations. (d_1) and (d_2) Enlarged conduction band and valence band at the interface between InGaN well and GaN barrier.

Spatial Raman spectrums have demonstrated that the stress state of InGaN/GaN epitaxial layer will be changed with the remove of Si substrate. From micro-disk center to micro-disk edge, the decreasing in-plane tensile stress in GaN layer causes the degree of atomic stretching in GaN ($2\overline{1}\overline{1}0$) weakened (Fig. 4c). Thus the degree of atomic compression in InGaN ($2\overline{1}\overline{1}0$) is correspondingly intensified (Fig. 4d) to accommodate this change in GaN layer. The above results indicate that the smaller the in-plane tensile stress in GaN layer, the greater the in-plane compressive stress in InGaN layer [14]. PL and Raman results further show that from center to edge of the micro-disk, maximum illuminating wavelength red shifts by 16 meV and Raman peak shifts by 0.48 cm^{-1} as the stress changes by 0.2 GPa. These phenomena are mainly related to the modulated piezoelectric fields and electron/hole quantum levels in the InGaN/GaN micro-disk LEDs caused by the gradient residual stress due to the remove of Si substrate. Therefore, the non-uniform piezo-phototronic effect plays a significant role for modifying the energy profiles and internal piezoelectric fields in InGaN/GaN micro-disk LEDs [43].

We have demonstrated experimentally the non-uniform piezo-phototronic effect caused by the gradient stress distribution in the micro-disk LED. In order to theoretically conform the non-uniform piezo-phototronic effect in the InGaN/GaN MQWs micro-disk LED, a self-consistent numerical model is established to calculate the energy band structure in an InGaN/GaN quantum well. The calculated energy band profiles of the InGaN/GaN heterostructure with Si substrate and without Si substrate are shown in Fig. 4d, and the detailed band diagrams at each interface are shown in Fig. 4d_1 and 4d_2. Obviously, as the Si substrate is etched off, with the larger piezoelectric field in InGaN/GaN quantum wells, positive polarization charges at $+c$ side increases while negative polarization charges at $-c$ side increase. As a result, E_V of the GaN/InGaN heterojunction interface bends downward and E_C of the InGaN/GaN heterojunction interface bends upward, which results in the band structure of quantum wells more inclined. Furthermore, this tilt-intensified energy band structure will lead to a reduction in the difference in ground state energy levels between electrons and holes, thus resulting in a red shift in the emission wavelength like Fig. 2d. Fig. 5 shows the PL peak energy as a function of test point position on the micro-disk LED obtained from the Poisson-Schrodinger coupling self-consistent calculation. The simulation shows that the energy peak decreases from micro-disk center to micro-disk edge with the lift off of Si substrate, which agrees well with the experimental result. The maximum energy difference between center and edge of micro-disk is 13.7 meV, which also matches with the PL results in Fig. 2. Thus, we experimentally and theoretically demonstrate the non-uniform piezo-phototronic effect can effectively modulate the optical performance of the InGaN/GaN micro-disk LED arrays with gradient internal stress distribution.

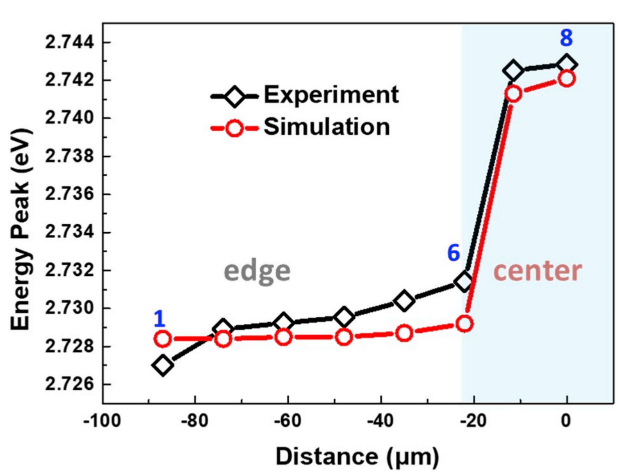

Fig. 5. Energy peaks as a function of test points position obtained from the Poisson-Schrodinger coupling self-consistent calculation. The calculation is in agreement with the experimental results.

In the literature, piezotronic effect has been studied for uniform strain in wurtzite structures. For a system that has a non-uniform strain, a gradient of strain can cause interesting flexoelectric effect [44,45]. The relationship between flexoelectric effect and piezotronic effect is analogous to definite integration and differentiation, which means that the flexoelectric effect is a total sum of results from many segmented piezotronic effects at local regions along the length of the structure [46]. In reality, one may measure the result of the flexoelectric effect, but its interpretation can be not easy. Same token applies for flexo-optoelectronic effect.

2. Conclusions

In summary, an innovative semi-floating InGaN/GaN micro-disk LED array is fabricated by combining isotropic and anisotropic dry etching processes. It has a significant intensity increase reaching up to 150% compared to broad-base LED. For the first time, we reveal non-uniform piezo-phototronic effect in complex micro optoelectronic devices through spatial micro-spectrums and Poisson-Schrodinger coupling self-consistent calculation, which caused by the gradient in-plane stress distribution with the lift-off of Si substrate. The phenomenon of wavelength shift due to the non-uniform piezoelectric effect makes the micro-disk LED expected to be used as multi-wavelength micro-displays. Our research is important not only for exploring the non-uniform piezo-phototronic effect in complex micro/nano optoelectronic devices, but also can largely improve the extraction efficiency and performance of micro-disk LEDs.

3. Methods

Structure of InGaN/GaN LED: $In_{0.15}Ga_{0.85}N$/GaN LED with a blue emission at 455 nm is grown on a 2 inch (111) silicon wafer. The bare wafer consists of a 330 nm-thick AlN buffer layer, a 600 nm-thick AlGaN buffer layer, a 400 nm-thick undoped GaN layer, a 3.4 μm-thick Si-doped n-GaN layer, a MQWs active region consisting of nine periods of 3 nm-thick $In_{0.15}Ga_{0.85}N$ well layers and 10 nm-thick GaN barrier layers, a 35 nm-thick p-AlGaN electron blocking layer, a 60 nm-thick Mg-doped p-GaN layer and a 20 nm-thick Mg-doped GaN layer.

Fabrication of InGaN/GaN micro-disk LED: The fabrication sequences are as followings: 800 nm-thick SiO_2 is first deposited in the LED bare chip to serve as a mask for subsequent dry etching of the GaN layer and Si substrate. Photolithography is then performed to define features of circular dimension (200 μm). Electron beam evaporation of Cr/Ni (50 nm/150 nm) and conventional lift-off processes provide metal mask to etch SiO_2. The following dry etch process is conducted in the ICP (Inductively Coupled Plasma) chamber at constant process parameter. The plasma etching process begins with removing SiO_2 in the atmosphere of CHF_3 (20 sccm), CF_4 (40 sccm) and Ar (10 sccm) for 5 min. The following is GaN layer, which is conducted with 30 sccm Cl_2 flow, 15 sccm BCl_3 flow and 5 sccm Ar. The anisotropic etching is then performed to realize the release of the GaN material by sacrificial etching of Si substrate through SF_6 (30 sccm), O_2 (5 sccm) and Ar (10 sccm).

Characterization of InGaN/GaN micro-disk LED: The structural and morphological properties of the pivotal micro-disks are examined by SEM (FEI/Nova NanoSEM 450). The ω-2θ curves of LED is measured by XRD. Both Raman and PL measurements are performed through a confocal micro Raman system (HORIBA/LabRAM HR Evolution) with different laser diode sources.

Declaration of competing interest

The authors declare no competing financial interest.

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