Abstract—The electroluminescence (EL) wavelength tailoring of silicon-rich silicon nitride (SiN_x)-based light-emitting devices (LEDs) is achieved by the modulation of the dimensions of Ag nanoparticles. Two EL peaks are observed in our SiN_x-based LEDs, both of which are red shifted with the increasing sizes of Ag nanoparticles. A reasonable explanation on this shift is proposed from the calculation of the local electric field surrounding Ag nanoparticles based on a simple model. This red shift of the two EL peaks as well as the evolution of their relative intensities is mainly originated from its weakening electric field enhancement with the increase of the size of Ag nanoparticles. Our work provides an alternative approach toward the fabrication of SiN_x-based LEDs with tunable EL wavelengths.

Index Terms—Electroluminescence (EL), full-color display, light-emitting devices (LEDs), local electrical field, localized surface plasmons (LSPs), silicon-rich silicon nitride (SiN_x), wavelength tailoring.

I. INTRODUCTION

O VER the last decades, lots of attention have been paid on amorphous silicon-rich silicon nitride (SiN_x) films due to their potential applications on silicon-based light sources with promising luminescence efficiency as well as flat panel display with wide tunable electroluminescence (EL) wavelength [1]–[14]. Various methods on tailoring the EL wavelength to achieve full-color display have been proposed. Generally, this tailoring is achieved by the modulation of the concentration of silicon excess as well as the average sizes of silicon nanocrystals in SiN_x films [1]–[5], [15]. It means that SiN_x films with different compositions should be prepared. By simply changing the bias condition, Garrido et al. achieved the blue–green to near-IR switching EL based on a bilayer structure of silicon-rich silicon oxide/SiN_x [10]. Furthermore, the tailoring of EL wavelength in SiN_x-based light-emitting devices (LEDs) can be achieved by the modulation of applied voltages or injected currents [6], [14]. Nevertheless, the EL intensity will be also changed together with its wavelength and it is difficult for us to achieve the modulation of EL wavelength and its intensity independently.

In this study, silver (Ag) nanoparticles with different sizes were deposited onto SiN_x luminescence layers. The EL wavelength tailoring of SiN_x-based LEDs is achieved by the tuning of dipolar resonance peaks of Ag nanoparticles. The independent modulation of EL wavelength and its intensity may be achieved easily by the adjustment of the sizes of Ag nanoparticles and the injected currents, respectively.

II. EXPERIMENTAL SETUP

SiN_x films with the thickness of about 50 nm were deposited on the p-Si substrates by a plasma enhanced chemical vapor deposition (PECVD) system, where the detailed fabrication parameters have been described in our previous paper [12]. For the investigation of the influences of localized surface plasmons (LSPs, the collective oscillations of excited free electrons confined to the surrounding of metal nanoparticles) [12], [16] on the EL of SiN_x-based LEDs, Ag nanoparticles with various dimensions were introduced after the deposition of SiN_x films. Ag nanoparticles deposited by magnetron sputtering were formed via a rapid thermal annealing process in Ar atmosphere at 500 °C for 60 s. The fabrication details of Ag nanoparticles can also be found in our previous paper [12], where the sputtering time of Ag is employed to label the samples, e.g., Ag20 refers to the sputtering time of 20 s.

The scanning electron microscopy (SEM) was employed for the characterization of the dimension and morphology of Ag nanoparticles. A HITACHI U-4100 spectrophotometer was used for the measurement of the transmission spectra of Ag nanoparticles. For the fabrication of SiN_x-based LEDs, indium tin oxide (ITO) and aluminum were deposited as the top transparent electrode and rear metal contact, respectively. The EL signals of the samples were recorded by an Acton SpectraPro-2500i monochromator coupled to a photomultiplier tube at room temperature.

III. RESULTS AND DISCUSSION

Fig. 1 represents the transmission spectra of Ag nanoparticles with different sputtering time, which are normalized at the maximum values. The SEM images are also provided for the determination of the average size of Ag nanoparticles, as shown in the
As have been investigated, when the energy of excitons is close to the electron vibration energy of LSPs, the radiative recombination rates will be enhanced significantly via the Purcell effect [12], [18]. The luminescence at this resonance energy can also be improved due to its enhanced radiative recombination rates. Based on this fact, we anticipate that the luminescence wavelength of SiN$_x$-based LEDs may be tuned at a reasonable range via the modulation of the wavelength of dipolar resonance. Consequently, EL spectra of SiN$_x$-based LEDs with different sized Ag nanoparticles were measured under the same injected current (∼30 mA), as shown in Fig. 2(a)–(d). In our previous investigation [12], [19], the EL performance of SiN$_x$-based LEDs is improved significantly by the addition of Ag nanoparticles, and the optimized external quantum efficiency is achieved by placing the Ag nanoparticles of ∼100 nm in diameter onto the luminescence layer (Ag40). As can be observed in Fig. 2, two peaks can be recognized, which are especially distinct for the devices of Ag20 and Ag40. We attribute the peak with shorter wavelength (P1, labeled as the blue line) to the recombination of the electrons confined at the K center and the holes located at the band tail formed by $N^-$ [14]. The one with the longer wavelength (P2, labeled as the green line) is originated from the recombination of the electrons located at the conduction band tail and the holes confined in the center of $\equiv S_i^0$ [14]. From the Gauss fittings of these two peaks, we get that both the peaks are red shifted with the increasing sizes of Ag nanoparticles, which are consistent with the red shift of their dipolar resonance peaks. This modulation of EL peaks with the increasing dimension of Ag nanoparticles is also available under different injected currents, as shown in Fig. 3(a) and (b). A little larger amount of red shift of EL peak positions is achieved for lower injected current. The role of injected current on the modulation of EL peaks is also checked, as shown in Fig. 3(c) and (d). Both P1 and P2 are blue shifted with the increase of injected current due to the improvement of carrier injection. Carriers can be injected into a deep level along their corresponding band tails and make the...
EL peaks blue shift. Obviously, the modulation of EL peaks by the size of Ag nanoparticles is much more significant than that by injected current. It is also noteworthy that P2 is dominated for the devices with smaller sized Ag nanoparticles, especially for Ag20, by comparing the integrated EL intensity of P1 to that of P2, as shown in Fig. 2(a). With the increase of the size of Ag nanoparticles further, P1 becomes more distinct and dominant, as shown in Fig. 2(b)–(d). Interestingly, the red shift of P1 may not result from the shift of the dipolar resonance peak due to the large inconsistence between them (labeled as the vertical dash line), as shown in Fig. 2. This inconsistence still exists for P2, especially for the devices of Ag60 and Ag80, which indicates further that there is little contribution of the red shift of the dipolar resonance peak on the shift of the EL peaks in SiN_x-based LEDs.

For the determination of the origin of this red shift of EL in our SiN_x-based LEDs with the increasing average size of Ag nanoparticles, the local electric field at the interface between the ITO electrode and the active layer is considered. This field will be enhanced significantly by the insertion of Ag nanoparticles, from which the injection of carriers, especially for electrons, can be improved [12]. Obviously, the improvement of the injection of carriers is mainly determined by the quantity of this enhancement. Based on the band diagram we proposed earlier [14], electrons (holes) can be injected into a higher energy level along the conduction (valence) band tails when a stronger local electric field is obtained. A shift (red or blue) of P1 and P2 can be achieved conveniently by the adjustment of the local electric field via the modulation of the size and shape of Ag nanoparticles as well as the distance between them.

Consequently, the electric field surrounding Ag nanoparticles is calculated based on a simple model where a single sphere with the radius of a is employed, as shown in the inset of Fig. 4(a). By solving the Laplace equation with proper boundary conditions at \( r = a \) and at infinity [20], we can get the field outside the metal sphere as follow:

\[
E_{\text{out}} = E_0 \left[ \frac{x^2}{r^3} - \frac{3x}{r^5} (x \hat{x} + y \hat{y} + z \hat{z}) \right]
\]

where \( \alpha \) is the sphere polarizability [21]. For a metal sphere \( (\varepsilon_i) \) in a medium with the dielectric constant of \( \varepsilon_D \), the polarizability is given by

\[
\alpha = \frac{\varepsilon_i - \varepsilon_D}{\varepsilon_i + 2 \varepsilon_D} a^3.
\]

The values of \( \varepsilon_D \) and \( \varepsilon_i \) can be obtained from the ellipsometric measurement and in [22], respectively. Hence, the enhancement of the electric field surrounding Ag nanoparticles can be obtained by

\[
E_h = \frac{|E_{\text{out}}| - |E_0|}{|E_0|}.
\]

This \( E_h \) at \( r = a \) is shown in Fig. 4(a), where the magnitude is normalized by the maximum. Obviously, the local electric field surrounding Ag nanoparticles becomes more and more insignificantly with the increasing size. Furthermore, the effect of the average distance \( d \) between Ag nanoparticles on the enhancement of the local electric field is also considered, where the simple computational model is shown in the inset of Fig. 4(b). The values of \( d \) can be obtained by

\[
\eta_S = \frac{\pi a^2}{(2a + d)^2}
\]

where \( \eta_S \) stands for the surface coverage of Ag nanoparticles. The coupling between Ag nanoparticles is more distinct for smaller sizes, especially for Ag20, by which the local electric field will be enhanced further due to its rather smaller \( d \), as shown in Fig. 4(b) [23]. For these reasons, the enhancement of the local electric field at the interface between the ITO electrode and the active layer as well as the improvement of the carrier injection due to this enhanced field is much more insignificant for Ag nanoparticles with larger dimensions, from which the EL peaks red-shift, as have been speculated previously and observed in Fig. 2. The lessened field enhancement of large sized Ag nanoparticles will make the injection of electrons weaker than that of holes, as the injection barrier for holes (1.9 eV) is much lower than that for electrons (3.0 eV) [3], [14]. Consequently, P1 dominates over P2 gradually with the increasing sizes of Ag nanoparticles, as shown in Fig. 2.

**IV. Conclusion**

In summary, two red-shifted EL peaks (P1 and P2) with the increase of the dimension of Ag nanoparticles are observed. The shift of the dipolar resonance peak of Ag nanoparticles only has a little contribution on the tailoring of EL wavelength, especially for P1, which is due to the large inconsistence between their peak positions. Based on a simple model for the calculation of the local electric field surrounding Ag nanoparticles, a reasonable interpretation on this shift is proposed. We attribute the red-shift of EL peaks as well as the evolution of their relative
intensities with the increasing size of Ag nanoparticles mainly to its weakening electric field enhancement. Our work provides an alternative approach toward the fabrication of Si- or SiN\textsubscript{x}-based LEDs with EL wavelength and its intensity modulated independently. Finer tuning may be achieved by the finer modulation of the dimension of metal nanoparticles as well as its local electric field.

REFERENCES


