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# Growth and Characterization of Si-Based Light-Emitting Diode with $\beta$-FeSi$_2$-Particles/Si Multilayered Active Region by Molecular Beam Epitaxy

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We fabricated single-, double- and triple-layered $\beta$-FeSi$_2$-particles structure on Si(001) substrates by reactive deposition epitaxy (RDE) for $\beta$-FeSi$_2$ and by molecular beam epitaxy (MBE) for Si, and realized electroluminescence (EL) at 310 K. Photoluminescence (PL) measurements revealed that the 77 K PL intensity of $\beta$-FeSi$_2$ increased almost proportionally with the number of $\beta$-FeSi$_2$-particles/Si layers. It was also found that the multilayered structure enhanced the EL intensity of $\beta$-FeSi$_2$ particularly at low temperatures. [DOI: 10.1143/JJAP.44.3951]

KEYWORDS: $\beta$-FeSi$_2$, LED, electroluminescence, reactive deposition epitaxy

## 1. Introduction

Semiconducting iron disilicide ($\beta$-FeSi$_2$) has been attracting significant interest as a Si-based light emitter with a wavelength (~1.5 µm) suitable for optical fiber communications.[1)] In 1997, Leong *et al.* reported low-temperature electroluminescence (EL) from $\beta$-FeSi$_2$ precipitates embedded in a Si *p*–*n* junction by ion-beam synthesis (IBS).[2)] However, room-temperature (RT) EL has been difficult to achieve due to the presence of a large number of induced defects. We therefore adopted reactive deposition epitaxy (RDE; deposition of Fe on hot Si) instead of IBS to form $\beta$-FeSi$_2$, and developed a formation technique for single-crystalline $\beta$-FeSi$_2$ particles embedded in Si by molecular beam epitaxy (MBE), which enabled us to realize the first RT 1.6 µm EL.[3)] There have been several reports thus far on RT EL at approximately 1.6 µm,[4–8)] and further efforts have been made to enhance the luminescence intensity of $\beta$-FeSi$_2$.[9,10)] One way to enhance the EL intensity of $\beta$-FeSi$_2$ is to embed a $\beta$-FeSi$_2$ continuous film in Si as an active region instead of $\beta$-FeSi$_2$ particles, and to form Si/$\beta$-FeSi$_2$ film/Si double heterostructures (DH). Very recently, a Si/$\beta$-FeSi$_2$ film/Si DH was realized on Si(111) substrates,[11–13)] but there has been no report on the formation of Si/$\beta$-FeSi$_2$ film/Si DH on Si(001). Epitaxial $\beta$-FeSi$_2$ film on Si(001) is found to easily aggregate into isolated islands when it is annealed at a high temperature for improving the crystalline quality or embedded in Si by MBE at high temperatures.[14)] It is therefore difficult to embed a continuous $\beta$-FeSi$_2$ film in Si and to realize Si/$\beta$-FeSi$_2$ film/Si DH LEDs on Si(001). Another way of enhancing the luminescence intensity of $\beta$-FeSi$_2$ is by increasing the volume of $\beta$-FeSi$_2$ particles embedded in Si. We have been trying to enhance the EL intensity of $\beta$-FeSi$_2$ by increasing the volume of $\beta$-FeSi$_2$ embedded in Si through the increase of the size of the $\beta$-FeSi$_2$ particles.[15)] However, it was found to merely increase the defect densities in the surrounding Si. We therefore developed a formation technique for double- and triple-layered $\beta$-FeSi$_2$-particles/Si structures for the purpose of increasing the volume of $\beta$-FeSi$_2$ in Si with $\beta$-FeSi$_2$ particle size kept constant.

In this paper, we report that a multiple $\beta$-FeSi$_2$-particles/Si layered structure enhances the PL (photoluminescence) and EL obtained from *p*-Si/$\beta$-FeSi$_2$-particles/*n*-Si LEDs compared with a single-layered structure.

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## 2. Experimental

$\beta$-FeSi$_2$-particles/Si multilayered structures for PL and EL measurements were fabricated as follows. First, 10-nm-thick [100]-oriented $\beta$-FeSi$_2$ epilayers were grown on $n^+$-Si(001) substrates by RDE at 470°C.[16,17)] The sample was then annealed *in situ* at 850°C for 1 h to improve the crystal quality of the $\beta$-FeSi$_2$ film. The $\beta$-FeSi$_2$ film agglomerated into islands during this process, due probably to the lattice mismatch (1–2%) between the two materials. Consequently, a 0.3-µm-thick undoped Si layer was grown by MBE at 500°C.[4)] For a double-layered structure, another 10-nm-thick [100]-oriented $\beta$-FeSi$_2$ epilayer was grown on the undoped Si layer by RDE at 470°C, followed by annealing at 850°C and a subsequent overgrowth of a 0.3-µm-thick undoped Si layer at 500°C. This process was repeated for a triple-layered structure. For EL measurements, a boron-doped $p^+$-Si cap layer with a doping concentration of approximately $1.0 \times 10^{18}$ cm$^{-3}$ was grown at 700°C on top of the undoped Si layer[18)] for the single- and double-layered structures. Unfortunately, we did not prepare triple-layered LEDs due to a problem with the growth chamber. All the samples for the PL and EL measurements were finally annealed at 900°C for 14 h in Ar atmosphere to further improve their crystal quality, resulting in $\beta$-FeSi$_2$ particles embedded in the Si matrix.[4)] A mesa structure of $1.5 \times 1.5$ mm$^2$ was made by wet chemical etching. An Al finger-type contact was formed on the $p^+$-Si layer by standard photolithography and sintered at 450°C for 20 min. The backside contact was made of AuSb. The crystallinity of the grown films was measured by X-ray diffraction (XRD). PL was measured using a He–Cd laser (442 nm). The EL spectra were measured with 200 Hz pulsed current biasing with a 50% duty cycle. The device was packaged in a metal can and mounted on a copper holder in a closed-cycle He cryostat. Luminescence was dispersed by a 25-cm-focal-length grating monochromator, and detected phase-sensitively using a liquid-nitrogen-cooled InP/InGaAs photomultiplier (Hamamatsu Photonics R5509-72).

## 3. Results and Discussion

Cross-sectional SEM images of single-, double- and triple-layered $\beta$-FeSi$_2$-particles/Si structures without the $p^+$-Si capping layer are shown in Figs. 1(a)–1(c), respectively. The white parts in these figures are $\beta$-FeSi$_2$ particles. We can see a clear layered structure of $\beta$-FeSi$_2$ particles in

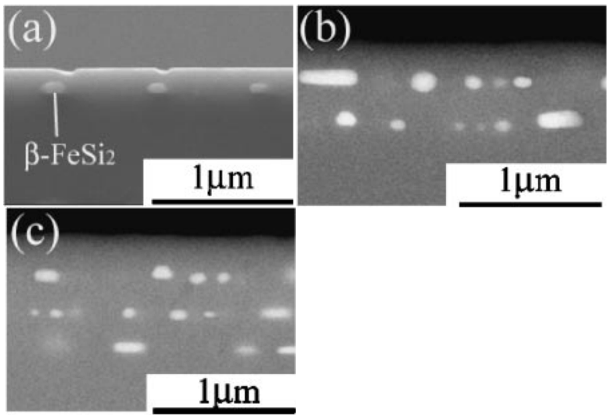


Fig. 1. SEM cross sections of (a) single-, (b) double- and (c) triple-layered $\beta$-$FeSi_2$-particles/Si structures for PL measurements. The $p^+$-Si capping layers were not grown.

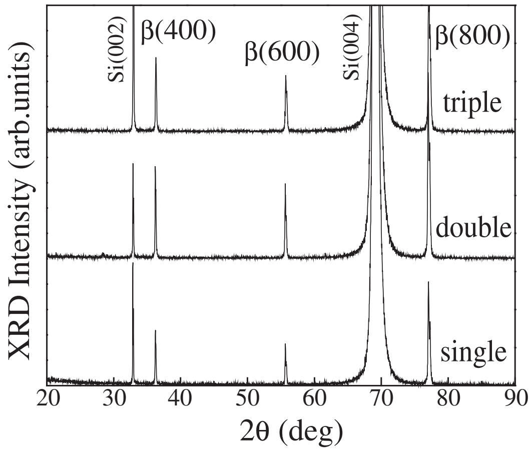


Fig. 2. $\theta$–$2\theta$ XRD curves for single-, double- and triple-layered $\beta$-$FeSi_2$-particles/Si structures for PL measurements.

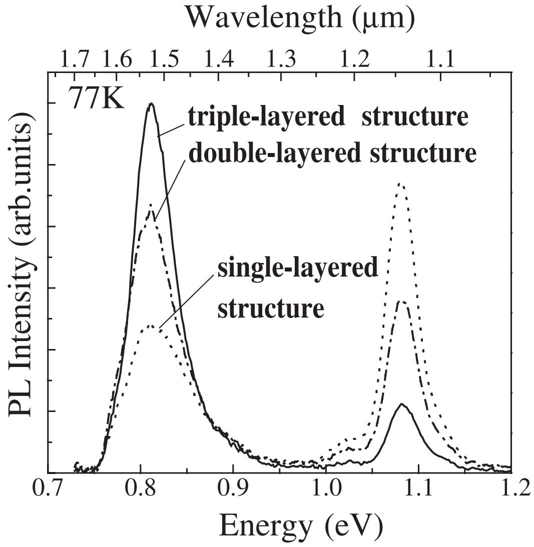


Fig. 3. PL spectra measured at 77 K for single-, double- and triple-layered $\beta$-$FeSi_2$-particles/Si structures.

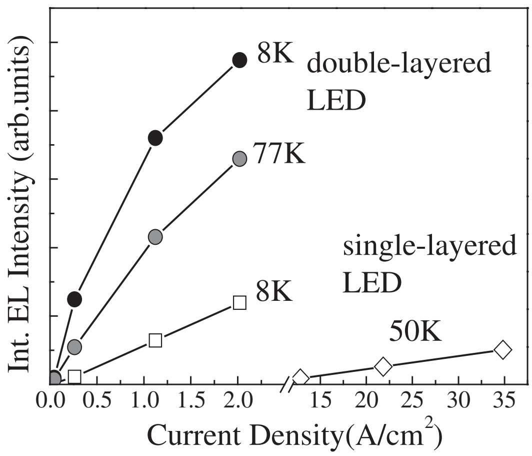


Fig. 4. Integrated EL intensity as a function of biasing current density for single- and double-layered LEDs measured at various temperatures.

these samples. In the $\theta$–$2\theta$ XRD curves shown in Fig. 2, only [100]-oriented diffraction peaks of $\beta$-$FeSi_2$ were observed, indicating that the epitaxial relationship between $\beta$-$FeSi_2$ and Si was preserved even after aggregation and MBE-Si overgrowth.

Figure 3 shows PL spectra measured at 77 K for these three samples. The PL measurements revealed that the 1.53 μm PL intensity increased almost proportionally with the number of $\beta$-$FeSi_2$-particles/Si layered structures. The thermal processing repeated for the fabrication of double- and triple-layered structures did not induce dislocation-related characteristic *D*-line emissions.[19–21] This result indicates that the number of electron–hole pairs which recombine radiatively in $\beta$-$FeSi_2$ is increased by increasing the number of $\beta$-$FeSi_2$ particles embedded in Si.

Next, we compare the EL characteristics of the double-layered LED with those of the single-layered one. Both samples showed clear rectifying properties of Si *p*–*n* junctions in their current–voltage (*I*–*V*) characteristics. Figure 4 shows the dependence of integrated EL intensity on the injected current density *J* for the single- and double-layered LEDs measured at various temperatures. The integrated EL intensity of $\beta$-$FeSi_2$ increased linearly with *J* at low temperatures, and the EL intensity was much more enhanced in the double-layered LED. Figure 5 shows the temperature dependence of integrated EL intensity for the single- and double-layered LEDs under $J = 2\,A/cm^2$. For reference, EL spectra obtained for the single-layered LED at various temperatures are shown in the inset. The EL of $\beta$-$FeSi_2$ is dominant and the EL from other sources was negligible. Similar EL spectra were also observed for the double-layered LED. The EL intensity of the double-layered LED was much higher than that of the single-layered LED at low temperatures. This result indicates that the EL intensity was greatly enhanced by doubling the $\beta$-$FeSi_2$-particles layers than the PL intensity. This difference is probably caused by the difference in the spatial distributions of carriers between the cases of light excitation and current injection. In the case of current injection, it is important to design the doping concentrations of *p*- and *n*-type Si so that the *pn* product becomes maximum at the $\beta$-$FeSi_2$ precipitates. Thus, it is considered that the *pn* product in the upper $\beta$-$FeSi_2$ particles is much higher than that in the lower ones due to the heavy doping of the $n^+$-type Si(001) substrate. On the other hand, there is not significant difference in the *pn*

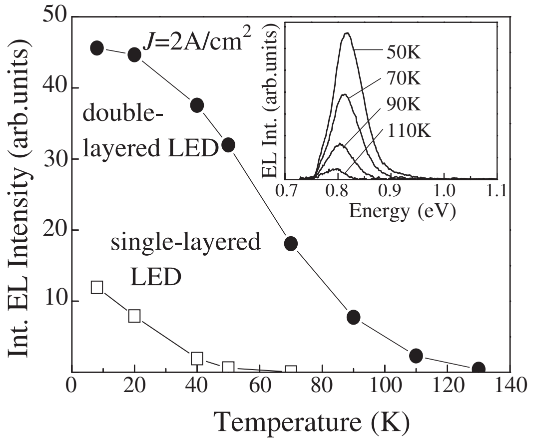


Fig. 5. Temperature dependence of integrated EL intensity for single- and double-layered LEDs when injected current density was 2 A/cm$^2$. The inset shows the EL spectra at various temperatures with $J = 2$ A/cm$^2$ for the single-layered LED.

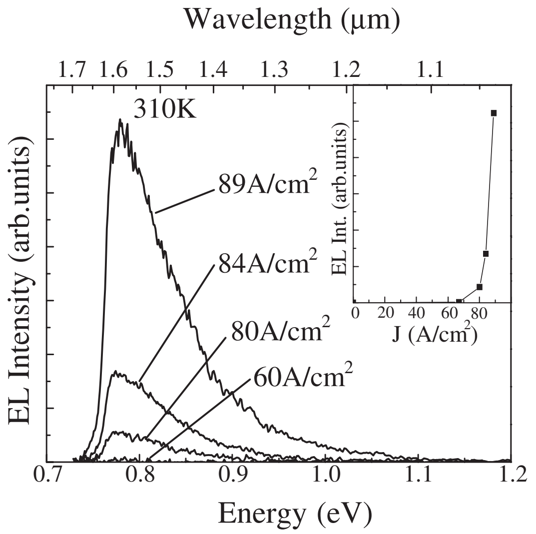


Fig. 6. EL spectra under various injected current densities measured at 310 K for the double-layered LED. The inset shows the dependence of integrated EL intensity on injected current density.

product between the upper and lower $\beta$-FeSi$_2$ precipitates in the case of the 442-nm-wavelength light excitation due to the penetration depth being approximately 1 μm in Si.

RT EL spectra were obtained for both single- and double-layered LEDs. Figure 6 shows the EL spectra as a function of injected current density at 310 K obtained for the double-layered LED. The asymmetric EL spectra are caused by the detection limit of the detector for wavelengths beyond 1.6 μm. The inset shows the dependence of EL intensity on injected current density. In both single- and double-layered LEDs, a 1.6 μm EL was observed only when $J$ was larger than 60 A/cm$^2$, and the EL intensity increased superlinearly with $J$, as observed in our previous single-layered LEDs, due probably to nonradiative recombination centers in and around the $\beta$-FeSi$_2$ precipitates.[3,4] This result suggests that numerous defect levels acting as nonradiative recombination centers still exist in the LEDs even after annealing at 900°C. When the current flowing through the defect levels saturates, the bias current begins to contribute to radiative recombination and a reasonable EL output is obtained. Unfortunately, the EL intensity of the double-layered LED was comparable to that of the single-layered LED at around RT. However, the EL enhancement observed at low temperatures indicated the potential of the multilayered structure. By optimizing the diode structure and doping profile, we believe that practical Si-based LEDs will be obtained in the near future.

## 4. Conclusion

We fabricated $\beta$-FeSi$_2$-particles/Si multilayered LED structures on Si(001) substrates by RDE for $\beta$-FeSi$_2$ and by MBE for Si, and achieved RT 1.6 μm EL from both the single- and double-layered LEDs. It was found that the PL intensity increased almost proportionally with the number of layered structures, and that the EL intensity of the double-layered LED was much more enhanced than the PL intensity, particulary at low temperatures.


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