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Fabrication of p-Si/β-$FeSi_2$/n-Si Double-Heterostructure Light-Emitting Diode by Molecular Beam Epitaxy

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We have fabricated p-Si/β-$FeSi_2$ film/n-Si double-heterstructure (DH) light-emitting diodes (LEDs) on Si(111) substrates by molecular beam epitaxy (MBE). It was found that both the thickness of an undoped Si overlayer and subsequent annealing temperature were key parameters in preventing the aggregation of the β-$FeSi_2$ film and the successful formation of a Si/β-$FeSi_2$ film/Si DH LED on Si(111). A 1.6 μm electroluminescence (EL) was realized at room temperature (RT) at a current density higher than 78 A/cm^2 by increasing the thickness of the β-$FeSi_2$ active region from 90 nm to 250 nm, suggesting that there exist numerous defects at Si/β-$FeSi_2$ heterointerfaces. [DOI: 10.1143/JJAP.44.2483]

KEYWORDS: β-$FeSi_2$, electroluminescence, double-heterostructure, light-emitting diode

1. Introduction

Semiconducting iron disilicide (β-$FeSi_2$) has been attracting much attention as a candidate for a Si-based light emitter with a wavelength (~1.5 μm) relevant to optical fiber communication.[1] A 1.6 μm electroluminescence (EL) at room temperature (RT) has already been realized from β-$FeSi_2$ particles embedded in Si *p-n* diodes formed on Si(001) by ion beam synthesis (IBS)[2–4] and molecular beam epitaxy (MBE).[5,6] In an effort to fabricate efficient light-emitting diodes (LEDs), it is necessary to form a Si/β-$FeSi_2$ film/Si double-heterostructures (DH) by embedding a continuous β-$FeSi_2$ film rather than particles in Si. However, there have been no reports to date on the formation of Si/β-$FeSi_2$ film/Si DH LED by IBS or MBE. In the case of IBS, the implantation of high Fe doses is required to form a β-$FeSi_2$ continuous film in Si, but it introduces numerous defects. Thus, low doses of Fe are implanted to form β-$FeSi_2$ precipitates in LEDs prepared by IBS.[2–4] In the case of MBE, a β-$FeSi_2$ epitaxial film grown on Si(001) exhibits a strong tendency to form islands when annealed at a high temperature to improve crystalline quality.[7,8] High-temperature annealing is believed to be inevitable to improve crystalline quality and luminescence intensity of β-$FeSi_2$.[9] Therefore, β-$FeSi_2$ particles have been used as an active region in LEDs grown by MBE.[5,6] There have been, however, some reports showing that smooth β-$FeSi_2$ films can be grown on Si(111), even at high temperatures, in spite of the large lattice mismatch (~5%) between the two materials.[10,11] Chu *et al.* reported EL from a Si/β-$FeSi_2$ film/Si DH on n-Si(111) substrates.[12] This is the only one report thus far for Si/β-$FeSi_2$ film/Si DH LEDs. They formed the β-$FeSi_2$ film by an rf magnetron-sputtering technique,[13] and this film was covered with a p-type Si overlayer by chemical vapor deposition (CVD) to form the DH. The EL intensity was, however, just comparable to that of Si at RT, indicating that the EL intensity of β-$FeSi_2$ is weak.

The purpose of this work is to realize RT EL from p-Si/β-$FeSi_2$ film/n-Si DH formed on Si(111) substrates by MBE in which the EL intensity of β-$FeSi_2$ is much stronger than that of Si.

2. Experimental

An ion-pumped MBE system equipped with electron-beam evaporation sources for Fe and Si was used. The p-Si/β-$FeSi_2$ film/n-Si DH LEDs were fabricated on an n-type epitaxial Si($\rho = 0.1$–$1\,\Omega\cdot$cm)/Czochralski n$^+$-Si(111) ($\rho = 0.004\,\Omega\cdot$cm) substrate as follows. A 20-nm-thick [110]/[101]-oriented β-$FeSi_2$ epitaxial layer was grown by reactive deposition epitaxy (RDE; Fe deposition on hot Si) at 650°C as a first step, and this layer was used as a template. Next, a 70-nm-thick [110]/[101]-oriented undoped β-$FeSi_2$ layer was epitaxially grown by MBE (codeposition of Si and Fe under stoichiometric conditions) at 750°C. To control the stoichiometry of the films, Fe deposition rate was controlled by an electron impact emission spectroscopy (EIES) sensor by maintaining the input power of the electron-beam gun for Si constant. Next, an undoped MBE-Si overlayer was grown at 500°C. The growth procedure was described in our previous report.[14] For the fabrication of LEDs, a 200-nm-thick boron-doped p$^+$-Si capping layer (p–1.0×10^{18} cm^{-3}) was grown at 700°C. To improve crystal quality, all the wafers were annealed at 800°C or 900°C in Ar for 14 h. High-temperature annealing is necessary to obtain a luminescence of β-$FeSi_2$ in Si/β-$FeSi_2$ Si(111) as in the case of Si/β-$FeSi_2$/Si(001).[14] The mesa structure of 1.5 × 1.5 mm^2 was formed by wet chemical etching. An Al finger-type contact was formed on the p$^+$-Si layer and sintered at 450°C for 20 min. The backsurface contact was formed using AuSb. Samples were prepared as summarized in Table I.

The crystal quality of the grown layers was characterized by X-ray diffraction (XRD), and the cross-sectional profile

Table I. Sample preparation: Growth temperature and thickness of β-$FeSi_2$, undoped and *p*-type-doped Si layers are listed. Annealing conditions are also specified.

Sample	β-$FeSi_2$	Si overlayer	p$^+$-Si	Annealing
A	90 nm	500°C (300 nm)	no	900°C/14 h
B	90 nm	500°C (300 nm)	no	800°C/14 h
C	90 nm	500°C (900 nm)	no	900°C/14 h
D	90 nm	500°C (900 nm)	no	800°C/14 h
E	10 nm	500°C (300 nm)	no	900°C/14 h
F	90 nm	500°C (500 nm)	700°C (200 nm)	800°C/14 h
G	250 nm	500°C (500 nm)	700°C (200 nm)	800°C/14 h

was observed by scanning electron microscopy (SEM). EL spectra were measured at 77 K and RT under 200 Hz-pulsed current biasing of 50% duty cycle. Luminescence was dispersed by a 25-cm focal-length grating monochromator, and detected phase sensitively by a liquid nitrogen cooled InP/InGaAs photomultiplier (Hamamatsu Photonics R5509-72).

3. Results and Discussion

To fabricate a Si/β-$FeSi_2$ film/Si DH, the Si overlayer was grown on the β-$FeSi_2$ film by MBE at a low temperature of 500°C, due to the strong tendency to form Si islands when grown at higher temperatures, such as 750°C. SEM showed that both the thickness of the Si overlayer and the annealing temperature are important parameters in preventing the aggregation of β-$FeSi_2$ and the successful formation of the Si/β-$FeSi_2$ film/Si DH on Si(111). Figure 1(a) shows the cross-sectional SEM image of sample A, obtained after annealing the Si(300 nm)/β-$FeSi_2$ film/Si DH at 900°C for 14 h. The white parts correspond to β-$FeSi_2$. As can be seen, the β-$FeSi_2$ moved up to the surface. The same result was obtained for sample B, obtained after annealing the same DH at 800°C for 14 h. These results suggest that a 300-nm-thick Si overlayer is not sufficient to prevent the aggregation of a 90-nm-thick β-$FeSi_2$ film. In an early study, the aggregation of β-$FeSi_2$ was avoided by capping it with a thick SiO_2 layer.[15] We, therefore, supposed that the aggregation of β-$FeSi_2$ can be prevented by capping a thicker Si layer, because it is considered to be more stable than a thin layer against the transformation of the β-$FeSi_2$ underlayer during high-temperature annealing. Thus, the thickness of the Si overlayer was increased from 300 nm to 900 nm in samples C and D. However, even if the Si overlayer was thick (900 nm), the β-$FeSi_2$ was found to aggregate into particles in Si as shown in Fig. 1(b) when the annealing temperature was 900°C. The β-$FeSi_2$ continuous film was successfully embedded in Si as shown in Fig. 1(c) for sample D, obtained after annealing at 800°C. The same result was obtained when the Si overlayer was 700 nm in thickness. On the basis of these results, a 700-nm-thick Si overlayer including the p-type Si layer was grown on the β-$FeSi_2$, followed by 800°C annealing for the fabrication of LEDs. The aggregation of β-$FeSi_2$ was considered to decrease β-$FeSi_2$/Si interface energy due to the lattice mismatch between the two materials with decreasing contact area. However, further studies will be required to fully understand the mechanism underlying this aggregation. For comparison, a typical example of Si/β-$FeSi_2$ particles/Si structure formed on Si(001),[8] sample E, is shown in Fig. 1(d).

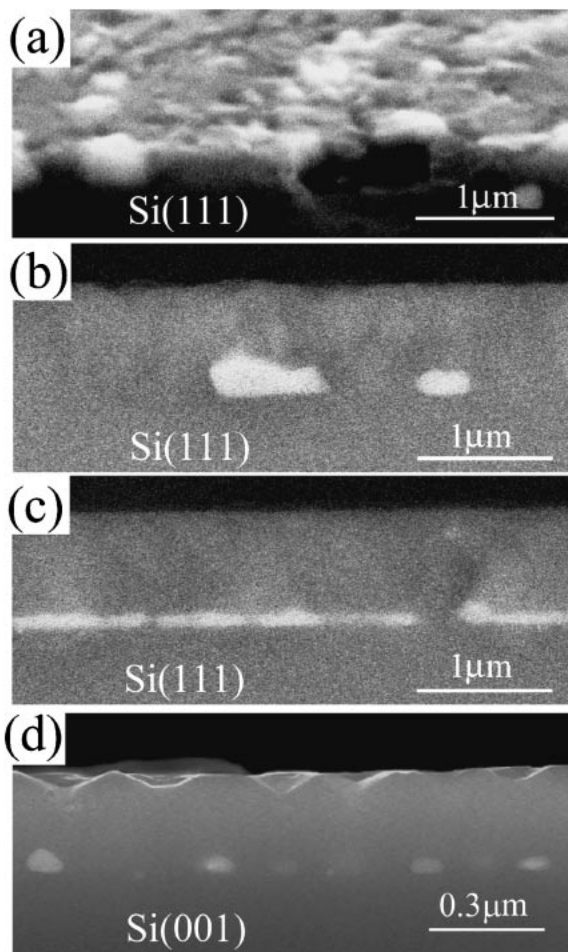

Fig. 1. Cross-sectional SEM images of Si/β-$FeSi_2$/Si structure on Si(111) of (a) sample A, obtained after annealing at 900°C. The thickness of the Si overlayer is 300 nm. (b) and (c) are SEM images of samples C and D, obtained after annealing at 900°C and 800°C, respectively. The thickness of the Si overlayer is 900 nm. (d) is a SEM images of sample E, a typical example of Si/β-$FeSi_2$ particles/Si structure formed on Si(001), obtained after annealing at 900°C.[8]

Figure 2 shows the θ–2θ XRD curves of sample F, the DH LED structure. The β-$FeSi_2$(220) and/or (202) diffraction peak is dominant and there are no other peaks except for that of Si and a weak β-$FeSi_2$(040) and/or (004) peak, indicating that the highly [110]/[101] oriented β-$FeSi_2$ film was embedded in the Si matrix with its epitaxial relationship to the substrate preserved. It is difficult to distinguish the diffraction peaks of β-$FeSi_2$(220) and β-$FeSi_2$(040) from those of β-$FeSi_2$(202) and β-$FeSi_2$(004), respectively, due to the small differences in the orthorhombic b and c of β-$FeSi_2$. Two arrangements of the β-$FeSi_2$ domains, that is β-$FeSi_2$[110]//Si[111] and β-$FeSi_2$[101]//Si[111], are therefore considered to coexist in the samples.

Figure 3 shows the current–voltage (I–V) characteristics of sample F measured at RT. Rectifying characteristics are clearly observed. The solid lines show the slopes with an ideality factor $\eta = 1$ and 2. Figure 4 shows EL spectra as a function of injected current density J at 77 K. The integrated

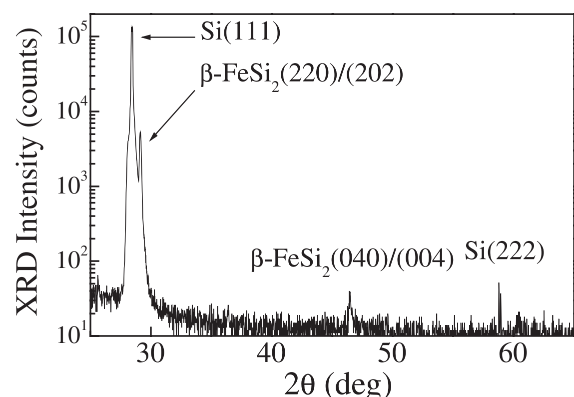

Fig. 2. θ–2θ XRD curves of sample F, p-Si/β-$FeSi_2$ film/n-Si DH LED.

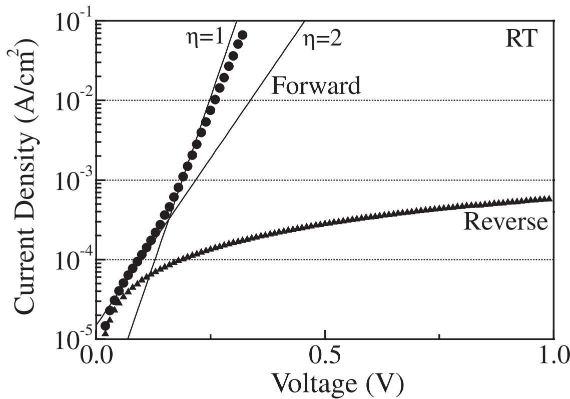

Fig. 3. Current–voltage characteristics of sample F measured at RT.

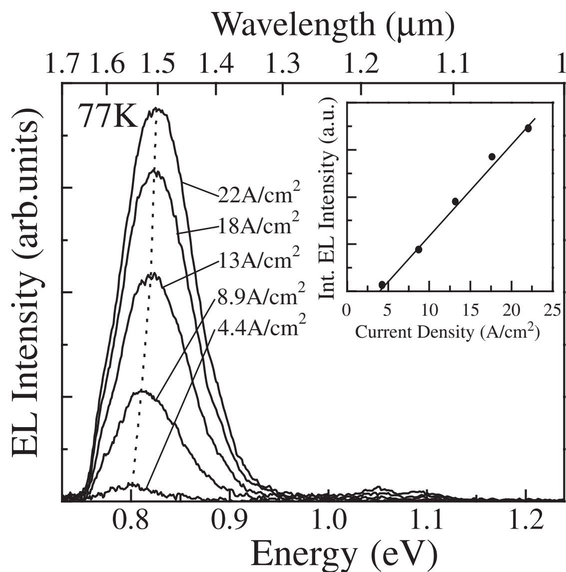

Fig. 4. EL spectra of sample F measured at 77 K. The inset shows the dependence of integrated EL intensity on current density.

EL intensity increased linearly with J as shown in the inset of Fig. 4. This result suggests that most of the bias current contributed to EL from the embedded β-$FeSi_2$ active layer. The EL intensity of this DH LED measured at 77 K was stronger than that of previous Si p-n LEDs with β-$FeSi_2$-particle active region by more than one order of magnitude. The EL peak energy was observed at approximately 1.55 μm under low-current injection, but it showed a clear blue shift with increasing J. This is due probably to the band-filling effect. When the bias current is sufficiently large, the ground states of β-$FeSi_2$ are occupied and thus injected carriers start to fill the higher energy levels, giving rise to EL with a higher peak energy. Our first, nonoptimized LED showed an observable but weak EL at RT. We speculated that one explanation for the weak EL at RT is nonradiative recombination due to point defects in the β-$FeSi_2$ film. It has been reported that the conduction type in undoped β-$FeSi_2$ depends on the deposited Si/Fe ratio.[16,17] Electron paramagnetic resonance (EPR) measurements of undoped n- and p-type β-$FeSi_2$ showed that the Fe and Si vacancies in β-$FeSi_2$ act as donors and acceptors, respectively.[18,19] First-principle calculations supported these results.[20] Our undoped β-$FeSi_2$ films grown by MBE tend to show p-type conductivity;[21] therefore, we suspect the existence of numerous Si vacancies in the film. High-temperature annealing is a very effective method of reducing these point defects,[22,23] and of obtaining luminescence from DH.[14] Another explanation is nonradiative recombination due to defects at the β-$FeSi_2$/Si(111) heterointerfaces. Han *et al.* have observed misfit dislocations at the β-$FeSi_2$/Si(111) heterointerface by transmission electron microscopy.[24] The formation of misfit dislocations is caused by the lattice mismatch between epitaxial β-$FeSi_2$ and Si. According to Lee and Dentai,[25] the influence of nonradiative recombination at the heterointerfaces on luminescence can be suppressed by increasing the thickness of the active layer. Thus, we attempted to fabricate LEDs having a wider β-$FeSi_2$ active layer, in which the β-$FeSi_2$ thickness was increased from 90 nm to 250 nm. Figure 5 shows EL spectra of sample G measured at RT. A 1.6 μm EL peak is clearly observed for $J > 78\,A/cm^2$. The EL intensity of β-$FeSi_2$ was stronger than that of Si by several orders of magnitude, and thus it should be noted that the EL properties obtained here were well improved compared to those of β-$FeSi_2$ DH LED reported by Chu *et al.*[12] The asymmetric EL spectra are attributed to the fact that the quantum efficiency of the detector decreases significantly for wavelengths beyond 1.6 μm. EL intensity increased superlinearly with J. This result suggests that so many defects working as nonradiative recombination centers still exist in the LED even after annealing at 800°C. When current passing through the defects saturates, bias current begins to contribute to the radiative recombination and a reasonable EL output is obtained. At present, a current density of $78\,A/cm^2$ necessary for an observable EL output is much larger than $14\,A/cm^2$ for Si/β-$FeSi_2$ particles/Si LEDs on Si(001).[5] However, the observation that the EL intensity at 77 K obtained in this work was much stronger than that of Si/β-$FeSi_2$ particles/Si LEDs shows the potential of Si/β-$FeSi_2$ film/Si DH LEDs. We believe that practical Si-based DH LEDs will be obtained in the near future by optimizing the diode structure and the doping profile.

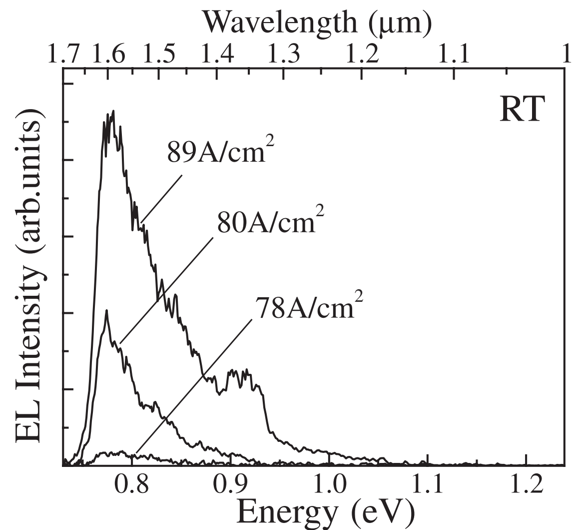

Fig. 5. EL spectra of sample G measured at RT.

4. Conclusion

We have fabricated p-Si/β-$FeSi_2$ film/n-Si DH LEDs on Si(111) substrates by MBE. Both the thickness of the Si overlayer and annealing temperature were key parameters in the formation of Si/β-$FeSi_2$ film/Si DH. A 1.6 μm EL at RT was realized for $J > 78\,A/cm^2$ by increasing the thickness of the β-$FeSi_2$ active region from 90 nm to 250 nm, suggesting that there exist numerous defects at Si/β-$FeSi_2$ heterointerfaces.

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1) M. C. Bost and J. E. Mahan: J. Appl. Phys. **58** (1985) 2696.
2) D. Leong, M. Harry, K. J. Reeson and K. P. Homewood: Nature **387** (1997) 686.
3) M. A. Lourenco, T. M. Butler, A. K. Kewell, R. M. Gwilliam, K. J. Kirkby and K. P. Homewood: Jpn. J. Appl. Phys. **40** (2001) 4041.
4) L. Martinelli, E. Grilli, M. Guzzi and M. G. Grimaldi: Appl. Phys. Lett. **83** (2003) 794.
5) T. Suemasu, Y. Negishi, K. Takakura and F. Hasegawa: Jpn. J. Appl. Phys. **39** (2000) L1013.
6) T. Suemasu, Y. Negishi, K. Takakura, F. Hasegawa and T. Chikyow: Appl. Phys. Lett. **79** (2001) 1804.
7) T. Suemasu, M. Tanaka, T. Fujii, S. Hashimoto, Y. Kumagai and F. Hasegawa: Jpn. J. Appl. Phys. **36** (1997) L1225.
8) Y. Ozawa, T. Ohtsuka, C. Li, T. Suemasu and F. Hasegawa: J. Appl. Phys. **95** (2004) 5483.
9) T. Suemasu, Y. Iikura, T. Fujii, K. Takakura, N. Hiroi and F. Hasegawa: Jpn. J. Appl. Phys. **38** (1999) L620.
10) C. Lin, L. Wang, X. Chen, L. F. Chen and L. M. Wang: Jpn. J. Appl. Phys. **37** (1998) 622.
11) T. Koga, H. Tatsuoka and H. Kuwabara: Appl. Surf. Sci. **169–170** (2001) 310.
12) S. Chu, T. Hirohada, H. Kan and T. Hiruma: Jpn. J. Appl. Phys. **43** (2004) L154.
13) S. Chu, T. Hirohada and H. Kan: Jpn. J. Appl. Phys. **41** (2002) L1200.
14) M. Takauji, N. Seki, T. Suemasu, F. Hasegawa and M. Ichida: J. Appl. Phys. **96** (2004) 2561.
15) T. Suemasu, N. Hiroi, T. Fujii, K. Takakura and F. Hasegawa: Jpn. J. Appl. Phys. **38** (1999) L878.
16) M. Komabayashi, K. Hijikata and S. Ido: Jpn. J. Appl. Phys. **29** (1990) 1118.
17) K. Takakura, T. Suemasu, Y. Ikura and F. Hasegawa: Jpn. J. Appl. Phys. **39** (2000) L 789.
18) T. Miki, Y. Matsui, Y. Teraoka, Y. Ebina, K. Matsubara and K. Kishimoto: J. Appl. Phys. **76** (1994) 2097.
19) I. Aksenov, H. Katsumata, Y. Makita, Y. Kimura, T. Shinzato and K. Sato: J. Appl. Phys. **80** (1996) 1678.
20) J. Tani and H. Kido: J. Alloys and Comd. **352** (2003) 153.
21) T. Suemasu, K. Takakura, C. Li, Y. Ozawa, Y. Kumagai and F. Hasegawa: Thin Solid Films **461** (2004) 209.
22) K. Takakura, T. Suemasu, N. Hiroi and F. Hasegawa: Jpn. J. Appl. Phys. **39** (2000) L233.
23) N. Hiroi, T. Suemasu, K. Takakura, N. Seki and F. Hasegawa: Jpn. J. Appl. Phys. **40** (2001) L1008.
24) M. Han, M. Tanaka, M. Takeguchi, Q. Zhang and K. Furuya: J. Cryst. Growth **255** (2003) 93.
25) T. P. Lee and A. D. Dentai: IEEE J. Quantum Electron. **14** (1978) 150.