Japanese Journal of Applied Physics
Vol. 43, No. 7A, 2004, pp. L930–L933

Time-Resolved Photoluminescence Study of Si/β-$FeSi_2$/Si Structures Grown by Molecular Beam Epitaxy

Takashi SUEMASU[1,2], Motoki TAKAUJI[1], Cheng LI[2], Yoshinori OZAWA[1], Masao ICHIDA[3] and Fumio HASEGAWA[1,2]

[1]*Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennohdai, Tsukuba, Ibaraki 305-8573, Japan*
[2]*Center for Tsukuba Advanced Research Alliance, University of Tsukuba, 1-1-1 Tennohdai, Tsukuba, Ibaraki 305-8573, Japan*
[3]*Department of Physics, Konan University, Kobe, Hyogo 658-8501, Japan*

(Received April 21, 2004; accepted May 16, 2004; published June 18, 2004)

Si/β-$FeSi_2$ particles/Si(001) and Si/β-$FeSi_2$ film/Si(111) structures were grown by reactive deposition epitaxy (RDE) and by molecular beam epitaxy (MBE), and time-resolved photoluminescence (PL) was measured from 8 K to 150 K. Both samples exhibited the same PL peak wavelength of 1.54 μm at low temperatures, but the PL decay time of 1.54 μm emission was different, showing that the luminescence originated from different sources. A short decay time ($\tau \sim 10$ ns) was found to be dominant for the Si/β-$FeSi_2$ particles/Si(001) at low temperatures. In contrast, the decay curve of the Si/β-$FeSi_2$ film/Si(111) was well fitted by assuming a two-component model, with a short decay time ($\tau \sim 10$ ns) and a long decay time ($\tau \sim 100$ ns). [DOI: 10.1143/JJAP.43.L930]

KEYWORDS: time-resolved photoluminescence, decay time, β-$FeSi_2$

Semiconducting iron disilicide (β-$FeSi_2$) is a promising material for silicon-based optoelectronics.[1)] Several groups have already reported room temperature (RT) electroluminescence (EL) at around 1.6 μm.[2–7)] The luminescence is considered to originate from recombination in the β-$FeSi_2$, however, the origin of the luminescence remains a topic of debate. It has also been suggested that the luminescence may be related to defects in the Si. The characteristic *D*1 line in particular corresponds to the 1.54 μm emission line for β-$FeSi_2$ at low temperatures.[8–11)] Thus, a simple continuous wave (cw) photoluminescence (PL) measurement alone cannot distinguish the luminescence of β-$FeSi_2$ from a *D*1 line. Leong *et al.* identified the 1.54 μm PL recombination in β-$FeSi_2$ precipitates by comparing the PL intensity obtained by excitation below and above the Si band edge.[12)] Katsumata *et al.* and Maeda and Terai adopted the temperature- and excitation-power-dependent PL measurements to identify 1.54 μm PL of β-$FeSi_2$.[13,14)]

Time-resolved PL measurement, as compared to cw PL measurement, is considered to be one of the most powerful methods for investigating the intrinsic optical properties of β-$FeSi_2$. There have been only four reports discussing the decay time of the 1.54 μm PL line of β-$FeSi_2$.[15–18)] Spinella *et al.* conducted detailed PL and transmission electron microscopy (TEM) investigations and reported that the 1.54 μm PL was attributed to unstrained β-$FeSi_2$ precipitates.[15)] The long decay time of 60 μs at 17 K in the samples was considered to be due to an indirect recombination in the β-$FeSi_2$ precipitates. A detailed discussion on the origin and nature of the 1.54 μm PL was previously reported.[19)] On the other hand, Schuller *et al.* reported that the 1.54 μm PL decay curve measured at 10 K was well fitted to two decay times (4 μs and 17 μs), and described that these long decay times are considered to represent the dislocation-related PL from damaged Si.[16,17)] These decay times were measured on the β-$FeSi_2$ precipitates in Si matrices formed by ion beam synthesis (IBS). It should be noted that IBS often induces a number of simultaneous defects in the Si matrices, and therefore it is necessary to handle these PL decay times very carefully. An alternative deposition technique for Si/β-$FeSi_2$ particles/Si(001) structures by reactive deposition epitaxy (RDE) and by molecular beam epitaxy (MBE) has been developed.[20–22)] These RDE- and MBE-grown samples are considered to suffer less damage than those formed by IBS, and therefore are considered suitable for time-resolved PL measurements. However, time-resolved PL measurements have not yet been performed on β-$FeSi_2$ grown by RDE and by MBE. Very recently, Chu *et al.* has reported a fast PL decay time of subnanosecond in β-$FeSi_2$ films formed by magnetron sputtering technique.[18)] However, we cannot rule out the possibility that the measured PL decay time is dominated by a nonradiative recombination process because the evaluation of temperature dependence of PL decay time was lacking.

The purpose of this work is to measure the decay time of the 1.54 μm PL from Si/β-$FeSi_2$ particles/Si(001) and Si/β-$FeSi_2$ film/Si(111) structures grown by RDE and by MBE over a wide temperature range and to discuss the origin of the luminescence. The PL decay time was found to differ for the two samples, indicating that the origin of the luminescence is different.

Si/β-$FeSi_2$ particles/Si and Si/β-$FeSi_2$ film/Si structures were grown on epitaxial n^+-Si(20 μm)/Czochralski-n^+-Si(001) and floating-zone Si(111) substrates, respectively, using an ion-pumped MBE system equipped with Si and Fe electron gun evaporation sources. The growth procedure for the Si/β-$FeSi_2$ particles/Si(001) structure was described in previous reports.[20–22)] First, a 10-nm-thick [100]-oriented β-$FeSi_2$ epitaxial layer was grown on Si(001) by RDE at 470°C, followed by *in situ* 850°C annealing for 30 min. Consequently, a 0.4-μm-thick undoped MBE-Si overlayer was grown at 500°C to embed the β-$FeSi_2$ in the Si. The grown wafer was annealed in Ar at 900°C for 14 h. The β-$FeSi_2$ subsequently aggregated into particles in Si, preserving its [100] orientation to the substrate. For the Si/β-$FeSi_2$ film/Si(111) structure, a 20-nm-thick [110]/[101]-oriented β-$FeSi_2$ template layer was grown epitaxially on Si(111) by RDE at 650°C, followed by a 70-nm-thick epitaxial β-$FeSi_2$ film grown by MBE at 750°C. Next, a 0.7-μm-thick undoped MBE-Si overlayer was grown on the β-$FeSi_2$ at 500°C. Finally, the wafer was annealed in Ar at 800°C for 14 h. The detailed growth procedure for this sample will be reported elsewhere.

Steady-state PL measurement was performed by the

standard lock-in technique using a He-Cd laser (442 nm) and a liquid-nitrogen-cooled InP/InGaAs photomultiplier (PMT) (R5509-72, Hamamatsu Photonics, Japan). The time-resolved PL was measured from 8 K to 150 K using a time-correlated single-photon counting setup. The R5509-72 PMT served as a detector. A mode-locked Ti:sapphire laser was used as the excitation source. The excitation wavelength, power and repetition rate were 783 nm, 4 mW and 0.8 MHz, respectively. Cross sections of the samples were observed by TEM or scanning electron microscopy (SEM).

Figures 1(a) and 1(b) show cross-sectional TEM and SEM images of samples fabricated on Si(001) and Si(111) substrates, respectively. The β-$FeSi_2$ formed on Si(001) aggregated into particles. In contrast, the β-$FeSi_2$ formed on Si(111) remained as a continuous film.

Normalized PL spectra measured at 8 K for these samples are shown in Fig. 2. Both samples have a peak wavelength of 1.54 μm, but the PL intensity of the Si/β-$FeSi_2$ film/Si(111) sample was approximately four times stronger than that of the Si/β-$FeSi_2$ particles/Si(001) sample. In an effort to conduct a detailed investigation of the 1.54 μm PL, the time-resolved PL measurement was performed first at 80 K. The wavelength resolution was limited by the slit opening of the monochromator, and was approximately 15 nm. The PL decay curves of the 1.54 μm emission obtained from the Si/β-$FeSi_2$ particles/Si(001) and the Si/β-$FeSi_2$ film/Si(111) samples are shown in Figs. 3(a) and 3(b), respectively. The dotted line in Fig. 3(a) represents the system response. The time resolution of the system was approximately 1 ns. The decay curve obtained from the Si/β-$FeSi_2$ particles/Si(001) was fitted relatively well to one exponential decay curve with a decay time of 15 ns. However, a double-logarithmic plot of this decay curve (not shown) suggested the possible inclusion of other components with longer decay time. Unfortunately, the signal intensity is too weak to allow a detailed investigation of these longer decay components. At present, it is possible to say from Fig. 3(a) that the fast decay component is dominant in the Si/β-$FeSi_2$ particles/Si(001) sample although we cannot conclude that the decay curve is composed of a single fast exponential component. In contrast, the decay curve obtained from the Si/β-$FeSi_2$ film/Si sample shown in Fig. 3(b) cannot be fitted to one exponential decay curve. In order to estimate the decay time of this sample, the decay curve was fitted using eq. (1), which is based on the assumption that it is composed of the sum of two exponentials, as discussed by Schuller *et al.*[16,17]

$$I(t) = I_1 \exp\left(-\frac{t}{\tau_1}\right) + I_2 \exp\left(-\frac{t}{\tau_2}\right) \quad (1)$$

Here, I_1 and I_2 are the PL intensities of the components with decay times τ_1 and τ_2, respectively. The fitting curve shifted downwards, as seen in the figure. Furthermore, it was in good agreement with the experimental curve when τ_1 and τ_2 were 16 ns and 104 ns, respectively. The ratio of the PL intensity I_1 to I_2 was 1.6. This finding indicates that the 1.54 μm PL originated from two sources. We cannot rule out other components with different decay times, but it can be said at least that the two components are dominant. It is important to note that the short decay time of this sample was nearly identical to that of the Si/β-$FeSi_2$ particles/Si(001) sample.

Next, in order to obtain a decay time at various temperatures, time-resolved PL was measured from 5 K to 150 K. The temperature dependence of PL decay curves of the emission from the Si/β-$FeSi_2$ film/Si(111) sample is shown in Fig. 4. Solid lines are the fits using a two-component model. The PL decay curves can be well described by two discrete single decay times. In contrast, PL decay curves for the Si/β-$FeSi_2$ particles/Si(001) sample were fitted almost

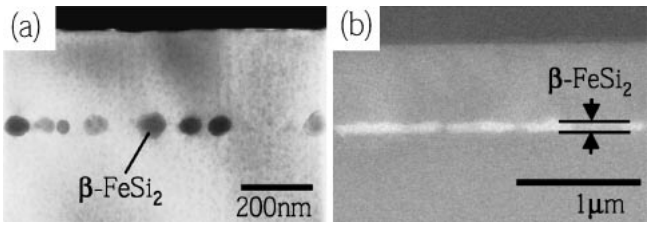

Fig. 1. Cross-sectional (a) TEM and (b) SEM images obtained for the Si/β-$FeSi_2$ particles/Si(001) and Si/β-$FeSi_2$ film/Si(111) structures, respectively.

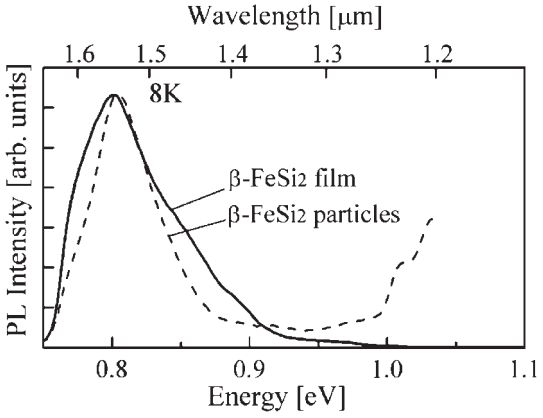

Fig. 2. Normalized PL spectra measured at 8 K for the Si/β-$FeSi_2$ particles/(001)Si (dotted line) and Si/β-$FeSi_2$ film/(111)Si (solid line) structures.

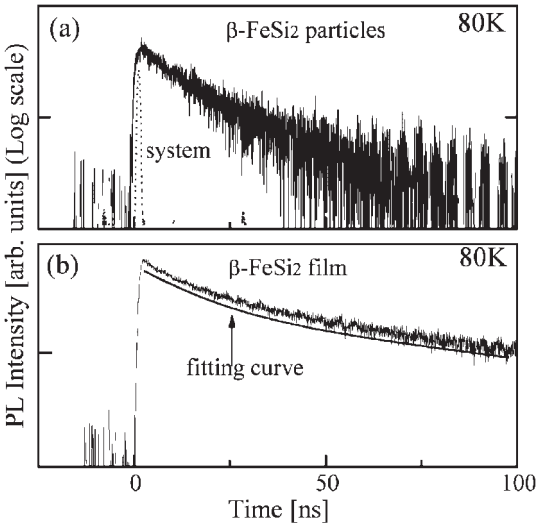

Fig. 3. Time-resolved PL decay curves of the 1.54 μm emission from (a) the Si/β-$FeSi_2$ particles/Si(001), and (b) the Si/β-$FeSi_2$ film/Si(111) measured at 80 K. The dotted line represents the system response.

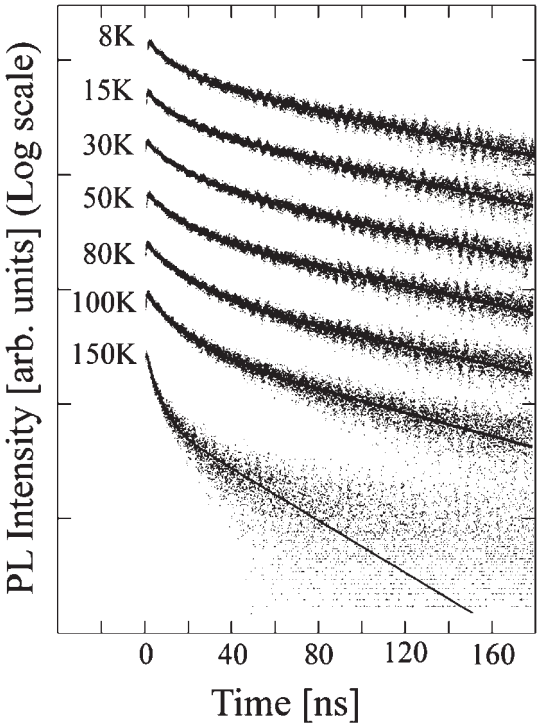

Fig. 4. Temperature dependence of PL decay curves of the 1.54 μm emission from the Si/β-$FeSi_2$ film/Si(111) sample. Solid lines are the fits using a two-component model.

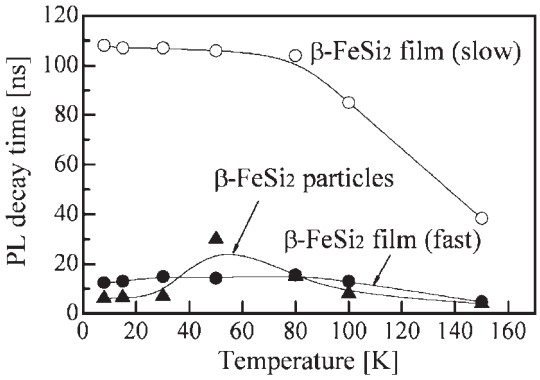

Fig. 5. PL decay time versus temperature obtained from the decay curves.

to one exponential decay component as in the case of the PL decay curve obtained at 80 K. Thus we may say that a fast decay component is dominant in this sample.

Figure 5 shows the obtained PL decay time versus temperature plots. As seen in the figure, the PL emission of the Si/β-$FeSi_2$ film/Si(111) sample exhibited two different decay times over a wide temperature range. The short decay times were of the same order as those obtained from the Si/β-$FeSi_2$ particles/Si(001) sample, and are approximately three orders of magnitude smaller than those previously reported on IBS β-$FeSi_2$.[15–17] The origin of these short decay times is considered to be due to recombination in β-$FeSi_2$. The β-$FeSi_2$ particles embedded in Si at 500°C were found to be under tensile strain in the *a*-axis direction[5] and thus we suppose that they have a direct band gap nature. As far as the β-$FeSi_2$ film embedded in Si is concerned, we have no definite information for discussing

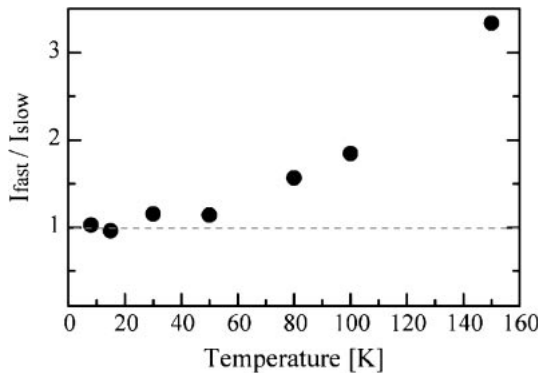

Fig. 6. Temperature dependence of the PL intensity ratio of a fast component to a slow one.

strain. The short decay time increased with temperature at low temperatures for both samples and then decreased above a critical temperature (~60 K). This decay time behavior was reported to be typical for excitons,[23,24] indicating that a nonradiative recombination process was negligible at the lowest temperature measured. On the other hand, the long decay time obtained from the Si/β-$FeSi_2$ film/Si(111) sample is considered to be due to the *D*1 luminescence. This is because the *D*1 line is another main origin of the 1.54 μm PL. In addition, the long decay time obtained was comparable to the reported *D*1 decay time.[25] The decay time was observed to decrease with increasing temperature, indicating that the nonradiative recombination rate increases.

Figure 6 shows the temperature dependence of the PL intensity ratio of the fast component (I_{fast}) to the slow component (I_{slow}) of the Si/β-$FeSi_2$ film/Si(111) sample. The ratio (I_{fast}/I_{slow}) was close to one at low temperatures, but it increased with temperature. This increase was considered to be due to the *D*1 line being more rapidly quenched. These findings suggest that the PL of β-$FeSi_2$ dominates at higher temperatures in the Si/β-$FeSi_2$ film/Si(111) sample.

In summary, the decay time of the 1.54 μm PL from Si/β-$FeSi_2$ particles/Si(001) and Si/β-$FeSi_2$ film/Si(111) structures grown by RDE and by MBE using time-resolved PL measurements has been investigated. The PL decay time of 1.54 μm emission was different between the two samples, showing that the luminescence originated from different sources. The origin of the 1.54 μm PL of the Si/β-$FeSi_2$ particles/Si(001) was considered mainly to be the carrier recombination in β-$FeSi_2$, which is characterized by a short decay time. The obtained decay time was approximately three orders of magnitude shorter than that previously reported in the case of β-$FeSi_2$ formed by IBS. In contrast, other luminescence such as the *D*1 line was considered to contribute to the 1.54 μm PL from the Si/β-$FeSi_2$ film/Si(111) structure.

This work was supported in part by Grants-in-Aid for Scientific Research (B)(12450137 and 12555084) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan, the TARA project of the University of Tsukuba and the Industrial Technology Research Grant Program in 2002 from the New Energy and Industrial Technology Development Organization (NEDO) of Japan. The authors express their sincere thanks

to Dr. T. Koyano (Cryogenics Center of the University of Tsukuba) for his help in SEM observation.

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