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Enhancement of Photoluminescence from Cu-Doped β-$FeSi_2$/Si Heterostructures

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We have investigated photoluminescence (PL) behaviors of the Cu-doped β-$FeSi_2$ thin film/Si heterostructure. Pronounced enhancement of an intrinsic A band and an impurity-related C band emissions has been observed in all the Cu-doped samples. The photo-carrier injection (PCI)-PL measurements have revealed that the PL enhancement is attributed to dynamic process of migration of holes where a repeated trap process of holes can be controlled by Cu-doping.

1. Introduction

β-$FeSi_2$/Si heterostructures may be one of the promising morphologies for light emission at the telecom wavelength [1]. Doping of impurity such as Al, Cu, C into β-$FeSi_2$ has sometime bring enhancement of photoluminescence (PL) efficiency [2-4]. Especially Cu doping was reported to be effective for enhancement of the PL intensity because of reduction of a nonradiative recombination centers (NRC) at the interface [3].
However, there have been no reports on impurity doping into ion-beam synthesized (IBS) β-$FeSi_2$ thin film/Si heterostructures expect for Al doping. In the case of Al doping, evident effect for PL enhancement has been reported by Terai and Maeda [2]. This is because a density of Si vacancy in the β-$FeSi_2$ lattice may be sufficiently decreased by Al doping. In this study, we have investigated PL behaviors of IBS Cu-doped β-thin film/Si nanostructures.

2. Experiments

The β-$FeSi_2$ thin films with the thickness of 60 nm was fabricated on a Si substrate by ion-beam synthesis (IBS) processes [1]. In the IBS, triple ion implantation of mass separated $^{56}Fe^{+}$ into n-type Si(100) with 500 μm thickness was carried out at 100, 80 and 50 keV. The each ion dose was $3.3\text{x}10^{16}$ ions/cm^2. After the triple ion implantation, the sample was annealed at 800 ^{o}C for 2 h to make the β-$FeSi_2$ polycrystalline thin films on the Si(100) substrate. Cu doping was carried out by a diffusion method. Cu films evaporated on the β-$FeSi_2$ thin films, then annealed at 800 ^{o}C for activation of Cu atomic diffusion into the deep site of the β-$FeSi_2$ films.
Photoluminescence (PL) was excited with a 641 nm-wavelength laser diode (LD) and the PL spectrum was detected with a monochromator (Jobin-Yvon HR 320) and a Ge PIN photodiode (Edinburgh Instrument).
Photo-injection PL (PCI-PL) was able to be observed when the sample back-side (Si substrate side) was irradiated with the LD. In the PCI-PL measurements, we measured the intensity and phase shift from the reference signal of irradiated laser in order to get information of transport of minority carrier (in this study, holes) and its efficiency of injection into β-$FeSi_2$ films where a radiative recombination of electron and hole takes place.

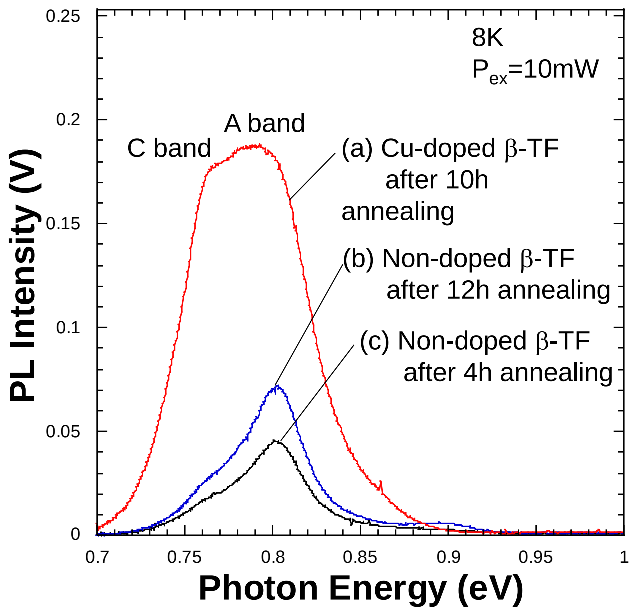

Fig. 1. Photoluminescence (PL) spectra of (a) Cu-doped β-$FeSi_2$ thin film (TF) after annealing for 10 h and PL spectra of non-doped β-$FeSi_2$ thin film after annealing for (b) 12 h and (c) 4h. The intrinsic emission from β-$FeSi_2$ thin film (A band) and impurity or defect-related emission (C band) were clearly observed in (b) and (c). The very broad band PL spectrum observed in (a) indicates that Cu-doping is effective to enhancement of both the A and C bands emissions.

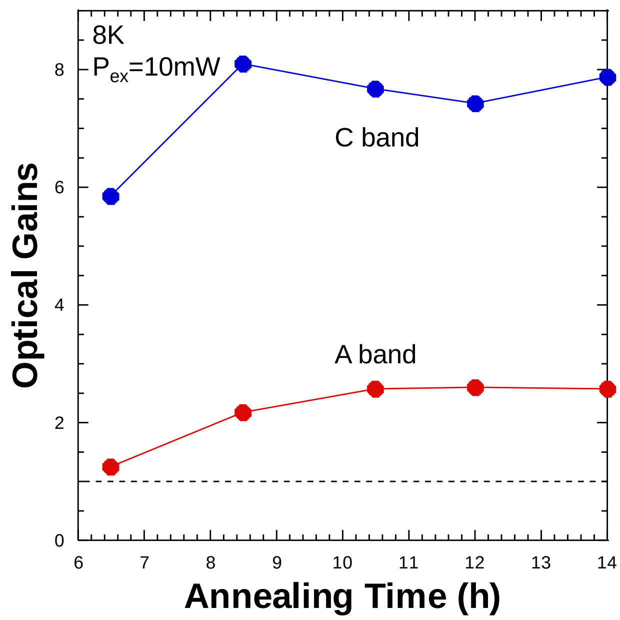

Fig. 2. Optical gains (ratio of intensity enhanced) for the A and C bands emissions as a function of annealing duration (time). Each peak intensities of the A and C bands were obtained by analysis of spectral peak separation.

3. Results and Discussion

Figure 1 shows PL spectra of (a) Cu-doped β-$FeSi_2$ thin film (TF) after annealing for 10 h and PL spectra of non-doped β-$FeSi_2$ thin film after annealing for (b) 12 h and (c) 4h. We observed that the very broad band emission consisting of the intrinsic emission from β-$FeSi_2$ thin film (A band) and impurity-related emission (C band) and pronounced enhancement of the both A and C bands emissions was caused by Cu-doping. Especially, the increase of the C band intensity was larger than that of the A band intensity. The C band peak energy was observed at 0.75 eV which was shifted by 0.02 eV from the usual 0.77eV observed in the non-doped samples in (b) and (c) of Fig.1.
Figure 2 shows optical gains (OG) for the A and C bands emissions as a function of annealing duration. The OG was defined by a ratio of PL intensities between Cu-doped and non-doped samples annealed at the same duration. The maximum OGs for the A band and C band emissions reached about 2.5 and 8 times, respectively. It was characteristic that both OGs of the A and C band emissions indicated a saturation behavior after 8 h annealing. Depth profiles of Cu atoms diffusing from the surface of β-$FeSi_2$ thin films were analyzed by Rutherford backscattering spectrometry (RBS). The RBS measurements suggested that Cu concentration in the β-$FeSi_2$ thin films is less than 1 at.% because of fast diffusion of Cu atoms though the β-$FeSi_2$ grain boundary. This result may suggest that solubility limit of Cu atoms in β-$FeSi_2$ dominates the PL behaviors observed in Figs. 1 and 2. The OG for the C band emission was larger than that for the A band emission. This is because Cu atoms can replace with Fe atoms in the β-$FeSi_2$ lattice and increase density of Cu^+ acceptor levels. The C band emission has been thought to be originated from radiative transitions between the conduction or valence bands and the defect- or impurity related levels. In the thin film, the C band enhancement is more pronounced than in that the nanocrystals or small precipitates. The very broad band emission shown in Fig.1 may be valuable for application to a light source for wavelength multiplex communication. The present maximum PL intensity of the Cu-doped thin film with the thickness of ~60 nm is by one fourth smaller than that of β-$FeSi_2$ nanocrystals in Si.
Moreover we investigated migration of minority carriers, in this study it is a positive hole, in such a Cu doped Si substrates. This purpose can be realized by Photo-Carrier Injection Photoluminescence (PCI-PL) measurements which consists of the following physical processes; (1) generation of electrons and holes at the back-side by LD excitation, (2) generation of hole profiles in n-type Si, (3) migration of holes with repeated trap processes to the thin film/Si heterointerface, (4) radiative recombination inside β-thin films resulting to PCI-PL. Frequency dependence of phase shift ($\Delta\theta$) from excitation signals gives a delay response with time constants (τ).

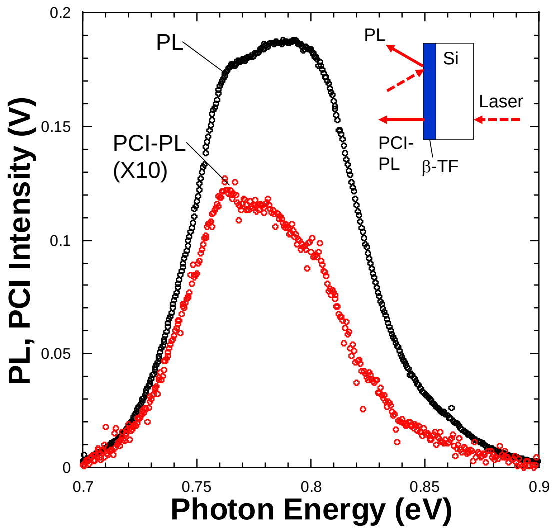

Fig. 3. PL and PCI-PL spectra for Cu-doped β-$FeSi_2$ thin film/Si (see the inset). The PCI-PL spectrum includes information of migration of minority carrier (hole) with a repeated trap process and radiative recombination efficiency of electrons and holes at the

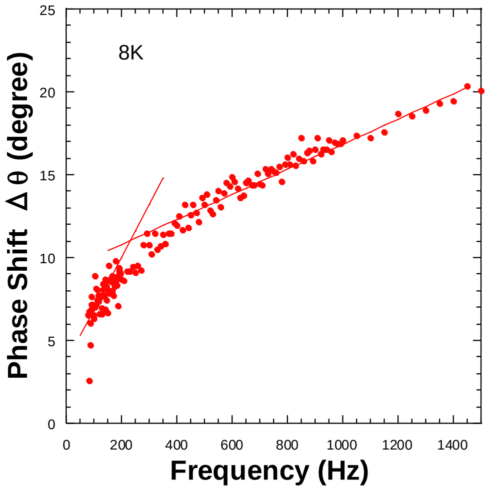

Fig. 4. Excitation frequency dependence of phase shift $\Delta\theta$ of PCI-PL signals at the A band energy of 0.803 eV. The $\Delta\theta$ at 0.803 eV was calculated by a difference from the phase of background signals. We found two processes with a large different time constant corresponding to the line slope. We obtained two time constants of τ_1=88 μs and τ_2=21 μs which are longer than τ_1=27 μs and τ_2=10 μs found in the non-doped samples. This time constant obtained in PCI-PL measurements shows a migration behavior of positive holes from the back-side to the β-$FeSi_2$ thin film at the surface side.

Figure 3 shows PL and the PCI-PL spectra of the Cu-doped thin film/Si heterostructure. A broad PCI-PL spectrum consisting of two PCI-PL peaks corresponding to the A and C band emissions was observed. This PCI-PL measurements indicate that photocarrier injection can be succeeded through the β-$FeSi_2$ thin film/n-Si heterojunction as well as in a β-$FeSi_2$ nanocrystal/Si composite phase. The time constant τ can be obtained from measurements of the frequency dependence of $\Delta\theta$ was analyzed by the following equation [5]. Actually the following relation can be seen in Fig. 4.

$$\Delta\theta(\deg) = 360\tau(s)\cdot f(Hz) + \Delta\theta(0),$$

where $\Delta\theta(0)$ is an off-set shift. In the Cu-doped samples, we obtained two time constants of τ_1=88 μs and τ_2=21 μs which are longer than τ_1=27 μs and τ_2=10 μs found in the non-doped samples. Such a longer time constant of τ_1=88 μs may be originated from a repeated trap process of positive holes during migration in n-type Si substrates as well as that observed in the β-$FeSi_2$ nanocrystal/Si composite phase [5]. We think that PL enhancement observed in Fig.1 is attributed to a repeated trap process of holes in the β-$FeSi_2$ thin film/Si heterostructures because this dynamic process can contribute to reduction of number of holes going out from β-$FeSi_2$ thin films and adapts to increase of the radiative recombination rate of electrons and holes at the thin film.

4. Conclusion

We have observed PL enhancement in the Cu-doped thin film/Si heterostructure. The PCI-PL measurements have revealed that the PL enhancement is attributed to dynamic process of migration of holes where a repeated trap process of holes can be controlled by Cu-doping.

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