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Photoluminescence Enhancement of $\beta$-FeSi$_2$ Nanocrystals Controlled by Transport of Holes in Cu-Doped N-Type Si Substrates

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We have investigated PL behavior of $\beta$-FeSi$_2$ nanocrystals controlled by transport of holes in Cu-doped n-type Si substrates. PL enhancement was observed and PCI-PL measurements revealed that PL enhancement was attributed to a transport process of holes with a larger time constant in Cu-doped n-Si substrate in which an interval trap process is controlled by Cu doping.

1. Introduction

Beta ($\beta$)-FeSi$_2$ nanocrystals ($\beta$-NCs) embedded in Si crystals ($\beta$-NCs/Si nano-composite phase) may be best for IR light emitters because of their strong light emission in the telecom wavelength region. In the nano-composite in Si, we can not expect the valence band offset between $\beta$-NCs and Si enough to make sufficient confinement of holes in $\beta$-NCs near room temperature. Replacing Si with SiO$_2$ has contributed to make both band offsets larger and thermal quenching (damping) of light emission smaller [1]. However, such a replacement may be unfavorable since Si substrates embedding $\beta$-NCs is a crucial material for considerable applications. Doping of impurity such as Al, Cu, C into $\beta$-FeSi$_2$ has sometime bring enhancement of photoluminescence (PL) efficiency [2-5]. Especially Cu doping was effective for enhancement of PL, and there may be two different mechanisms in the PL enhancement. One is increase of density of states at the Cu$^+$ accepter level and other is formation of trap centers for holes in a Si phase. Former mechanism contributed to enhancement of the C band emission, and later one to decrease of holes transport from $\beta$-NCs to n-type Si by the interval trap process, which results increase of holes remained in $\beta$-NCs and increase of radiative recombination rate at interband (A band emission). This finding taught us that Cu doping into n-type Si is a key point for enhancement of the A band emission. In this study, taking above discussion into account, we have investigated PL behavior of $\beta$-NCs controlled by holes transport in Cu-doped n-type Si substrates.

2. Experiments

The nano-composite phase with $\beta$-NCs and Si was fabricated by ion-beam synthesis (IBS) processes [6]. In the IBS, ion implantation of mass separated $^{56}$Fe$^+$ into n-type Si(100) with 500 $\mu$m thickness was carried out at 200 keV. The ion dose was $10^{17}$ ions/cm$^2$. After the implantation, the sample was annealed at 800 $^\circ$C for 8 h to make the nano-composite phase on Si(100) substrates. Cu doping was carried out by a diffusion method. Cu films evaporated on back-side of n-type Si(100) substrates and
annealed at 800 °C for activation of Cu diffusion. Photoluminescence (PL) of nano-composite phase was excited with a 641 nm wavelength laser diode (LD) and PL spectrum detected with a monochromator (Jobin-Yvon HR 320) and a Ge PIN photodiode (Edinburgh Instrument). Photocarrier-injection PL (PCI-PL) was excited when sample back-side (Si substrate) 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 minority carrier transport and injection into β-NCs which may play as a radiative well for electron-hole pairs.

3. Results and Discussion

Figure 1 shows PL spectra for cases of (a) the β-NCs/Si nano-composite phase on Cu-doped n-type Si substrate, (b) the Cu-doped β-NCs/Si nano-composite phase, and for comparison (c) the non-doped β-NCs/Si nano-composite of both A and C bands PL than Cu-doping direct into the nano-composite phase. This finding is of importance for thinking the PL enhancement mechanism. We know that Cu atomic diffusion in Si is very activated and fast, and can estimate that Cu atoms doped from a back-side of Si substrate can diffuse all over the area. This situation has been confirmed by Rutherford backscattering spectrometry (RBS). Next we investigated transport of minority carriers, in this study it is a positive hole, in such a Cu doped Si substrate. For this purpose, a Photocarrier-injection PL (PCI-PL) measurement is a unique method and consists of the following physical mechanisms; (1) generation of electrons and holes at the back-side of Si substrate by photo-excitation, (2) formation of minority (hole) profiles inside of n-Si, (3) transport of holes with trap processes into the surface layer (nano-composite phase), (4) radiative recombination at β-NCs resulting PCI-PL. The frequency dependence of the PCI intensity and phase shift from excitation gives origin of the emission site, time constants (τ) and a ratio of possible radiative processes.

![Fig. 1. PL spectra measured at 8 K for cases of Cu doping into (a) n-Si substrates and (b) the β-NCs/Si nano-composite phase, and for (c) the non-doped composite phase.](image-url)
Figure 2 (a) shows PL spectrum and the corresponding PCI-PL spectrum. We have observed two peaks corresponding to the A and C band PL peaks. This means that the PCI-PL process above described from process (1) to (4) takes place. Figure 2 (b) shows a phase shift ($\Delta \theta$) spectrum of PCI-PL signals. We have observed a large change of $\Delta \theta$ in which PL from $\beta$-NCs is observed. The phase shift in PL spectrum has not been observed, and in Figure 2 (b) the $\Delta \theta$ in PCI-PL for non-doped sample was smaller than that for Cu-doped sample. This fact implies that the $\Delta \theta$ in PCI-PL is originated from hole’s transport from the back-side to $\beta$-NCs near the surface, and that in the Cu-doped n-Si hole’s transport has a larger time constant than in non-doped samples.

In order to obtain the time constant ($\tau$) of the PCI-PL, frequency dependence of $\Delta \theta$ was measured and analyzed by the following equation,

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

where $\Delta \theta(0)$ is an off-set shift. We found that there are two time constants $\tau_1 = 110$-$130 \mu s$ and $\tau_2 = \sim 20 \mu s$ which are slightly dependent upon anneal time for diffusion of Cu atoms into the Si substrate. In the non-doped sample, a single $\tau = \sim 22 \mu s$ was obtained. These results indicate that doping of Cu atoms into n-Si substrates makes hole’s transport with larger $\tau_2$ which may be attributed to an interval trap process of holes. We think that PL enhancement observed in Fig.1 comes from such an interval trap process of holes in Cu-doped n-type Si substrates because this dynamic situation brings reduction of number of holes going out from $\beta$-NCs and adapts to increase of the radiative recombination rate in $\beta$-NCs.

4. Conclusions

We have investigated PL behavior of $\beta$-FeSi$_2$ nanocrystals controlled by hole’s transport in Cu-doped n-type Si substrates and observed PL enhancement. PCI-PL measurements reveal that PL enhancement is attributed to a transport process of hole with a larger time constant in Cu-doped n-Si substrate in which an interval trap process is controlled by Cu doping.
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