Light emitting from the self-interstitial clusters buried in the Si^+ self-ion implanted Si films

Lingxi OuYang, Chong Wang ✉, *Mengwei Zhou, Jie Yang, Yu Yang*

School of Material Science and Engineering, Institute of Optoelectronic Information Materials, Yunnan University, Kunming 650091, People's Republic of China

✉ *E-mail: cwang@ynu.edu.cn*

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The photoluminescence (PL) properties corresponding to different types of the interstitial clusters (or defects) in the silicon ion (Si^+) self-implanted Si have been well reviewed. Given a brief of the conjectural origin, defect type, annealing temperature of the W (1218 nm), X (1193 nm) peak, R (1376 nm), and D bands concluded the present application of Si^+ self-ion-implantation. The challenges for application of light-emitting Si and application prospects are also discussed.

1. Introduction: Photoelectron-microelectronics integration is a novel research area in the semiconductor industry. One of the limiting factors in this technology is that the optical transition between silicon (Si) bandgaps requires assistance from a phonon, because of the indirect nature of this bandgap. The period of radiation recombination in Si is always in the order of a millisecond, which limits the applications of Si in optoelectronics industry [1]. There are a variety of methods that have been tried by scientists to improve the luminous efficiency of Si materials including ion-implantation (II). This is a mature process and, as such, has become the preferred method to improve the luminescence properties in Si.

Impurities and defects have been introduced into the substrate by II; these impurities or defect states form an impurity-defect recombination centre in the forbidden bandgap of Si. Electrons and holes recombine into excitons in those recombination centres making most of these recombination centres non-radiation recombination centres. However, some defect-energy levels still exist in the material and are continuously distributed in a wide range in the *k* space producing come sub-gap levels in forbidden bandgap of Si. This provides various choices for carriers in optical transitions. Si^+ self-II (SII) is a flexibly modifier approach to improve the luminous efficiency of Si materials. In contrast to the implantation of boron (B), aluminium (Al), hydrogen, iron even erbium (rare Earth), SII does not introduce hetero-ions, it generates a defect luminescence centre (DLC) which includes interstitial or vacancy clusters. Although there are some vacancy cluster processes with high thermal stability [2], the SII process usually generates interstitial clusters (ICs) (with only a few vacancies) and, in theory, this process can also attain high-density DLC [3, 4].

Currently, the light-emitting diode (LED) produced by II has been developed in succession [5–7]. Homewood *et al*. implanted B^+ into an Si substrate and produced an LED with an external quantum efficiency in the order of $\sim 10^{-3}$. The dislocations were introduced by implantation gradually formed into interstitials that were generated by closed-loop-dislocations after high-temperature annealing, so that the carried dislocations were bound in these loop-dislocations, which decreased the non-radiation recombination [6]. In 2007, using rapid thermal annealing (RTA) after implanting Si^+ in Si substrate, Bao *et al.* prepared a low-temperature high light emission efficiency LED based on the W peak. Although the luminescence was merely emitted by defects during implantation, the external efficiency exceeded $\sim 10^{-3}$ [5]. Recently, complementary metal–oxide–semiconductor compatible, integrated Si^+ was implanted in Si wave guide photodetectors that were demonstrated and characterised for use in the mid-infrared (IR), the maximum responsivity reached to $\sim$10 mA/W with a 5 V reverse-bias [8]. These effort proved that the SII process with no introduction of hetero-ions is promising for large-scale optoelectronic integrated circuits.

For many of the defects introduced by SII, the numerous non-radiation recombination centres greatly lower the luminous efficiency in the process of implantation, injection dose, energy, annealing temperature etc. which play key roles in the quantity, distribution, and varieties of radiation recombination in Si^+ implantation samples. Many questions remain such as: for the various emitting centres, which type of defect is the origin of the luminescence? How do defects evolve before and after II? How do different annealing temperatures affect the luminescence? This Letter attempts to provide a brief summary of ICs and its evolution in Si^+ SII samples.

2. Interstitial and its evolution correspond to photoluminescence: Spectra in photoluminescence (PL) generally result from the various defects in SII. To obtain good performance from an optoelectronic device, the luminescence mechanisms of various interstitials need to be explored to find the most stable intrinsic interstitial with highest intensity that is easy to make. Therefore, the following discussion will focus on the W, X, R, D1, and D2 centres in SII samples.

2.1. W band and X band: The peak located at the 1218 nm in the PL spectrum of the SII sample is the well-known W line. Hayama and Giri found that the local vibrational mode of the W band is 70.0 meV, which indicates that the 1244 nm spectrum in PL is a phonon replica of the W band [9, 10], as is shown in Fig. 1. On the basis of the experimental results from many studies, researchers have found that the W band can be detected in various II samples, even in a proton implantation sample with electron or neutron irradiation [10–12]. Thus, some reports have indicated that the origin of the W band is the intrinsic defect of diffusion point defect resulting from primary cascade damage [13, 14]. After thermal treatment, these point defects aggregate to ICs. Using molecular-dynamics simulations, Richie and Gharaibeh found that the three-interstitial (I3) with the lowest energy is a particularly stable configuration [15, 16]. Coomer and Carvalho found that the W band may originate from the I3 by utilising the first-principles local-density-functional (LDF) [17, 18]. However, in variation from the early simulation studies, Nakamura found that the ICs in the sample implanted with protons using electron irradiation was smaller than those

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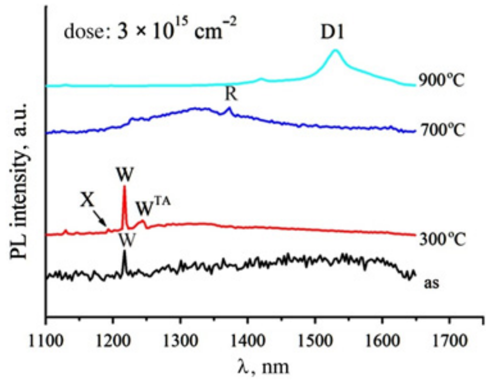

Fig. 1 *PL spectra of the Si samples annealed for 30 min at temperatures from 300 to 900°C, spectra at the bottom were taken from samples before annealing, the spectral signals were recorded at 7 K, samples were implanted with Si^+ doses $3 \times 10^{15}/cm^{-2}$ at 300 keV in p-type Si wafers*

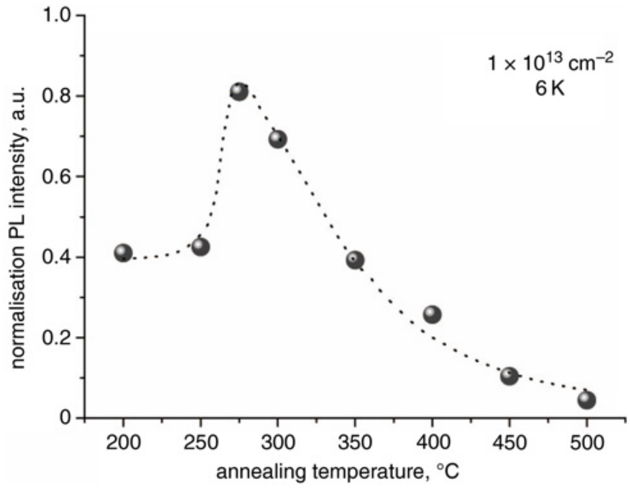

Fig. 2 *Normalisation PL intensity of SOI samples annealed at different temperatures in 30 min, samples were implanted Si^+ with dose of $1 \times 10^{15}/cm^{-2}$, 300 keV, recorded at 6 K*

introduced by II and using the PL measurements and calculations, Nakamura indicated that the [111] split mono-interstitial (I1) and the [111] split triple (ST) di-interstitial were the probable origin of the W centre [12]. The I3 of high mobility [19] also indirectly indicates that I3 may not be the ideal choice for the formation of the W centre because of its instability.

The X line which is located at 1193 nm in Fig. 1 can also be attributed to a light emission centre induced by intrinsic defects [20]. In addition to the W centre, the X centre was also induced by ICs [10]. Its origin was assumed to be the four-interstitial (I4) B3 electron paramagnetic resonance centre [21, 22], which corresponds to the first-principles computational methods of the electrical properties of this interstitial calculated consequence as reported by Coomer [21]. Cooper's experimental results of the infrared absorption peaks in 530 cm^{-1}, 550 cm^{-1} and PL spectra also confirmed this conclusion. Carvalho *et al.* reached the same conclusion through LDF calculations, where they confirmed that the I4 is highly symmetrical. When a light source of 1.03 eV was used for illumination, I4 produced a triplet excited state, implying that the I4 interstitial had a deep donor level [17]. The I4 compacted interstitial is also believed to possess high stability and low mobility [23]. According to the experimental results of Giri *et al.* [24], the W centre reaches its maximum intensity after annealing at 300°C and as the annealing temperature was elevated to 450°C, the intensity of the X centre increased and the W band was annihilated. By controlling the annealing temperature, Yang *et al.* [25] found that X band can only be detected at a relatively high annealing temperature rather than the W band, which indicated that the X centre was more stable than the W centre, as a result of Ostwald Ripening [12, 26]. These authors also considered that the ICs of the X centre were also larger. However, the X centre does not simultaneously form with the W centre, which implied that X centre evolved from the W centre. According to these conclusions, Nakamura indicated that the [111] split I1 and the [111] ST di-interstitial were the probable origins of the W centre [12]. Giri [24] also suggest that the [111] ST di-interstitial was a more appropriate source of the W centre. After thermal annealing, these interstitials can easily combine and form an I4 ICs, which maybe the origin of X centre. However, in a recent study by our group, as shown in Fig. 1, the PL spectra of the samples with the implanted dose of $3 \times 10^{15}/cm^{-2}$ were found to have a distinct peak at 1376 nm in the non-annealed curve. The luminescence of the W line was detected whether the sample was annealed or not and the luminous efficiency reached a high level in the sample annealed at low temperature (about 250°C) as shown in Fig. 2. This suggested that defects or interstitials introduced during the early stage of implantation can also be radiation recombination centres. According to former literature reports and our experiment results [5, 27, 28], it is believed that the simple I1 formed by implantation (or irradiation) could be attributed as the origin of the W line.

Although the origin of the W line is still under investigation, research must continue on an Si^+ SII LED device based on the W line, because of its sharp luminescence peak and stable luminescence properties. However, there are some barriers that hinder the practical application of the Si^+ SII LED device based on the W line. For example, the device based on the W line can only work at low temperature (∼70 K) [5] and the external quantum efficiency is only 10% in a gallium arsenide device.

2.2. R band: In Fig. 1, it is apparent that as the annealing temperature is increased to nearly 700°C, the PL spectrum of the SII sample with a dose of $3 \times 10^{15}/cm^{-2}$ was dominated by a peak located in the 1376 nm [24, 29]. Using deep level transient spectroscopy (DLTS) and transmission electron microscope (TEM) analysis, it was found that the R band (1376 nm) was attributed to a luminescence defect centre that extended from the [110] split interstitial to the [311] rod-like defect [29–33]. By measuring the electrical and optical properties of this material, Libertino *et al.* [29] found that from a point defect to a small interstitial extension of the [311] defect, each aggregation was not simply followed by Ostwald ripening. A low annealing temperature (300–500°C) caused the formation of I-type point-like defects. Annealing at intermediate temperatures (550–650°C) produced the I-clusters. The high annealing temperatures will cause a transition from I-clusters to defects with a dose of at least $1 \times 10^{13}/cm^{-2}$. Coffa [34] found that only when the dose was beyond $1 \times 10^{13}/cm^{-2}$ and the annealing temperature was higher than 600°C could the [311] defect be generated. So, Coffa indicated that this [311] defect was responsible for the peak located in 1376 nm which was composed the 'building block' of the [100] interstitial. This stable configuration was found to be compact for small clusters, elongated for medium clusters and planar for large clusters [35]. Goss *et al.* [36] utilised DLTS and found that the [311] defect was the origin of the R band. Through PL measurements of the Si substrate implantation with copper ions and subsequently the proton irradiation, Nakamura claimed that the [311] defect was formed by the [100] extended split interstitial [11]. So, most of the research results appear to suggest that the [311] defects are composed of the [100] interstitial that could be attributed as the origin of the R band. Yang *et al.* [25, 37] found that the full width at half height full width at half maximum (FWHM) of R band is about 6 meV, Giri [24] also found a ∼10 meV FWHM of

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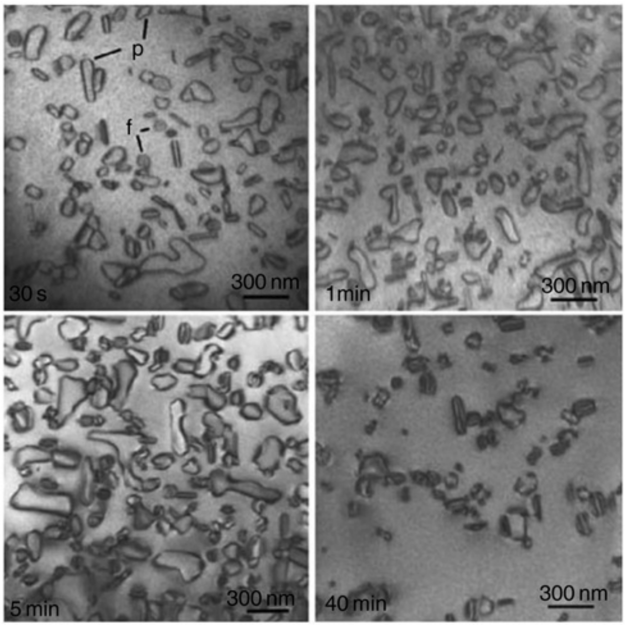

Fig. 3 *Plan-view bright-field TEM images of Si samples annealed for different times, taken near [001] Si, examples of perfect loops are marked as p and faulted as f [39]*

the R band, which was on the same order of the W band and the X band, implying that the formation of the [311] defect was associated with an I chain-like interstitial or the release of strain in the I-cluster. The R defect centre was distributed in the range of 700 nm [38] below the substrate surface and the R peak was quenched at the recorded temperature (RET) of 160 K. According to Ostwald Ripening [26], as the annealing temperature is increased, a small I1 interstitial gradually grows and migrates to surface becoming more stable.

The loop-dislocation composed of the [311] rod-like defect is advantageous for fabricating optoelectronic devices based on the R band in SII Si materials. In Fig. 3, the sample was implanted with B^+ at a dose of $1 \times 10^{15}/cm^{-2}$ and an energy of 30 keV, annealed for 30 s, 1 min, 5 min, and 40 min. There are perfect (p) and faulted (f) loops marked in Fig. 2, where p represents the clear closed loops; otherwise, they are marked as f. The size and density of both types of loops increased as the annealing time was increased from 30 s to 5 min; for annealing times longer than 5 min, the loops became smaller. The high density, small size loop-dislocations will bind carriers and reduce the phonon scattering. With RTA for 1–5 min at 950°C, the Si samples with B^+ implantation could be used to fabricate LED devices with a dislocation density of $1 \times 10^9/cm^2$ and an external quantum efficiency of up to $\sim 10^{-3}$ [39].

2.3. D band: Among the detected peaks of SII in the PL spectrum, the D1 band was the only luminescence centre that could be detected with an RET of ~280 K [25], as shown in Fig. 4. As the annealing temperature was increased, the D1 band underwent a redshift. Plastic deformation and direct Si bonding were used to fabricate the room-temperature LED [40–42], the D1 band that was obtained showed the great potential of the fabricated device. In the previous effort, through the simulated calculations, most researchers have considered that the defects related to dislocation could be attributed to the origin of the D1, D2 bands [43, 44]. Giri's experiments on Al^+ implanted samples examined using measurements that included cross-sectional TEM, optical microscopy, and PL, indicated that the D1, D2 bands originated from dislocation, instead of oxidation-induced stacking faults [45]. Suezawa reported that the D1, D2 bands could be associated with the geometric kinks and believed that D1–D4 bands could be attributed to the transitions between shallow levels (Ei = 4–7 meV) and deep levels (Eg–Ei-hn)

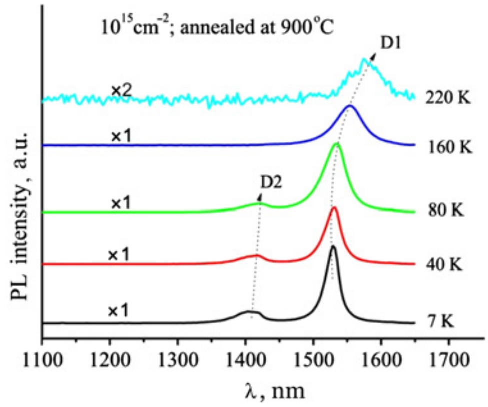

Fig. 4 *PL spectra of the D1 band measured at different temperatures. D1 band sample implanted Si^+ to dose of $1 \times 10^{15}/cm^{-2}$ then annealed at 900°C for 30 min*

in the forbidden gap [46, 47]. Yet another viewpoint was suggested where the D1, D2 bands resulted from point defect deformations caused by the dislocation strain region [48]. Our reported studies on Si-on-insulator (SOI) samples implanted by Si^+ SI subsequently reactive ion etching (RIE) showed the phenomenon where the intensity of D3 peak in the PL is stronger than D1, D2 bands and only the D3 peak could be detected in the sample annealed at 200° C. So, the origin of the D3 peak appeared to be different from the D1, D2 bands. In the RIE experiments, we found that the D1, D2 bands were quenched before the region of loop-dislocation had been etched. Hence, we believed that the origin of D1, D2 bands was related to point defects among the dislocations [38].

In the mid-1990s, application research of the D bands was conducted. The wavelength of the D1, D2 was close to the optical fibre communication band. It had great potential for application in photoelectron-microelectronics integrated circuits. In addition, the RET reaching to ~280 K, nearly room temperature, all of the excellent characteristics of the D band drive kept researchers digging. At present, the LED based on the D band is effective only at 77 K. So, determining the proper dose and annealing temperature to increase luminous efficiency, external quantum efficiency, working temperature is now the primary target for future research in this area.

3. Summary and outlook: This Letter attempted to provide a brief summary of ICs and their evolution in Si^+ SII samples. In summary, in the case of the W, X, R, D1, D2 bands, with the elevation of annealing temperature of the Si^+ SII samples, the luminescence centre evolved from small interstitial through large interstitial to dislocation and the stability increased continuously. The maximum detection temperature could be elevated from 110 to 280 K. In addition, the excitation wavelength of the Si^+ SII samples also increased. For each PL peaks, various formation mechanisms invited modulation in different ways, but the origins of the W, R, and D bands is still controversial. Understanding the luminescence mechanism is important for further advancement of the art, but enhancing the efficiency of radiation recombination, the device working temperature and light emission efficiency are also great challenges for the SII Si materials. Methods to accomplish these goals need to be developed. For example, increasing the thickness of the active layer by repeated ion implanting, but the implantation energy will decrease with each implantation. Therefore, two or three Si^+ enriched layers can be formed after uniform annealing, and a saturated optical interstitial would be generated deep in the active layer. On the basis of the Purcell effect, the emission of the material could be modified by the surrounding environment due to the change of the LD of states. If the shapes, size, distribution of optical nanocavity are modulated in the implantation samples, the interstitials may couple, so the spontaneous emission intensity would increase by the optical resonance generated by these optical nanocavities. These approaches will be attempted in the future to improve the emission performance of the Si^+ SII and studies will target these luminescence centres of the SII defects to provide new concepts for producing high-efficiency full-Si optoelectronic devices.

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