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Photoluminescence evolution in self-ion-implanted and annealed silicon*

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Si^+ ion-implanted silicon wafers are annealed at different temperatures from room temperature to 950 °C and then characterized by using the photoluminescence (PL) technique at different recorded temperatures (RETs). Plentiful optical features are observed and identified clearly in these PL curves. The PL spectra of these samples annealed in different temperature ranges are correspondingly dominated by different emission peaks. Several characteristic features, such as an R line, S bands, a W line, the phonon-assistant W^{TA} and Si^{TO} peaks, can be detected in the PL spectra of samples annealed at different temperatures. For the samples annealed at 800 °C, emission peaks from the dislocations bounded at the deep energy levels of the forbidden band, such as D_1 and D_2 bands, can be observed at a temperature as high as 280 K. These data strongly indicate that a severe transformation of defect structures could be manipulated by the annealing and recorded temperatures. The deactivation energies of the main optical features are extracted from the PL data at different temperatures.

Keywords: photoluminescence, silicon, self-ion-implanted, defects

PACC: 4255P, 6170T, 8140T

1. Introduction

Silicon is the dominant material for semiconductor electronics and electronic applications. Since silicon is a kind of indirect band-gap semiconductor in nature, radiation recombination from direct band-to-band transitions is rare, and it is manifested by a large radiation recombination lifetime on the order of milliseconds.[1] Currently, however, the luminescence from silicon is being researched extensively because this approach is very promising for silicon-based optical-electrical integrated devices.[2−5]

Ion implantation is a standard process for introducing dopants and other impurities into silicon crystals. Recently, novel physics and material optical properties in silicon have been demonstrated by employing this technique and its subsequent annealing process. This promotes the ion-implanted silicon to be a hot research area again.[5−7] Ion implantation or irradiation of silicon produces intense and sharp photoluminescence in the formation of the W band with a zero-phonon line at 1.018 eV.[6] Davies *et al* mapped out the conditions when the PL intensity is expected to be approximately proportional to the concentration of centres in silicon, induced by low dose ion implantation.[7] The transition from small interstitial clusters to extended {311} defects has also been identified clearly, and the evolution of implantation-induced defect structures has been well discussed in recent work.[8] Giri *et al* reported on the optical emission from Si^+ implanted silicon with a relatively low energy (1.2 MeV) and dose.[9] Homewood *et al* illuminated the effects of dislocation loops on the suppression of thermal quenching of luminescence.[10] Interstitial-type defects have been observed beyond the projected ion range in high energy ion-implanted and annealed silicon.[11] Most recently, Bao *et al* presented a novel approach to enhancing the emission intensity of the W-line in silicon and demonstrated a good sub-band-gap light emitting diode based on the introduction of point defects.[12] Thus, several questions subsequently arise. For example, which defects control the light emission from silicon and where are they located? Although considerable efforts have been made to answer these questions, those studies focused primarily on the description of central lines in implanted sil-

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icon with a high implanting energy (MeV) and low dose ions.[6−11,13−15] To our knowledge, little work has been done on the PL evolution of defects with temperature in silicon, especially those emissions produced by self-ion implantation.

In the present work, with a view to understanding the origin of radiation recombination from different defects, an investigation into PL evolution with annealing temperature (AT) and recorded temperature (RET) is conducted in self-implanted Si samples. The PL features of the implanted samples annealed at different temperatures are identified clearly and deactivation energies of the main PL lines (bands) are obtained. PL emission from the samples is observed at 280 K. It indicates that room temperature luminescence from silicon wafers can also be achieved by self-ion implantation.

2. Experimental details

The base materials used in this investigation were p-type (100) silicon wafers (boron doped, $\rho \approx 15\ \Omega \cdot \mathrm{cm}$). At room temperature, these wafers were implanted with a Si^+ ion dose of $3 \times 10^{14}\ \mathrm{cm}^{-2}$ and an implanting energy (E_{imp}) of 300 keV. The radius of the corresponding projected ion range (R_{p}) is about 450 nm. All the implantation experiments were conducted with an ion beam at an incident angle of 7° with respect to the normal of the Si wafer to avoid channelling effects. After completing the ion implantation, all the wafers were cut into several pieces, then they were annealed in N_2 atmosphere for 30 min at different temperatures ranging from room temperature to 950 °C in temperature steps of 50 °C.

The PL spectra were recorded by using a Syner JY spectrometer (Tria 550). In the PL measurements, samples were mounted on the cold-tip of a drip-feed liquid helium cryostat. The temperatures of the samples in the optical experiments varied from 4.7 to 300 K. An argon-ion laser with a wavelength of 460 nm was used as an irradiation source. The spot diameter and the exciting power of the laser beam were ∼2.5 mm and 17 mW, respectively. Light emission from the samples was dispersed by a monochromator and detected by using a Ge detector. The standard lock-in technique was used to improve the ratio of luminescence signal to noise.

3. Results and discussion

Figure 1 shows typical PL spectra of the as-implanted sample and those samples annealed at different temperatures for 30 min. The PL spectra fit the Gaussian line model well.[16] It is worth noticing that the PL spectra of all the as-implanted samples and the samples with AT lower than 200 °C feature a wide photon energy range of 0.765–1.050 eV (1620–1180 nm). The centre of this broad range is near 0.805 meV (1540 nm), which is determined by the Gaussian line fitting. This broad feature is a typical PL emission from the disordered regions of the unknown structure (probably amorphous) which are located in the top layer (at a depth of ∼500 nm from the surface) and usually produced by ion implantation.

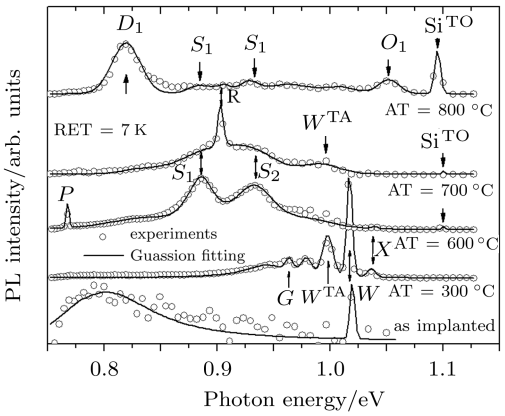


Fig.1. PL spectra of the as-implanted Si wafers, those at different annealing temperatures (AT) and those without any annealing process (as-implanted) separately. A time of 30 min is chosen in the annealing process. The recorded temperature (RET) in these PL experiments is 7 K. All the Si samples are implanted with a Si^+ ion dose of $3{\times}10^{14}\ \mathrm{cm}^{-2}$ and an E_{imp} of 300 keV.

A strong and sharp W line is observed at 1.017 eV (1219 nm) in the PL spectra of those samples annealed at low temperatures ($\leq$ 350 °C). In fact, the PL spectra are dominated by the W line and its phonon replica W^{TA} at 0.998 eV (1244 nm), as in the typical PL line of the 300 °C annealed sample shown in Fig.1. The full width at half height (FWHM) of the W line is in a range of 4–6 meV at 7 K, but that of the W^{TA} line is about 12 meV on average. The peak at 1.040 eV (1193 nm), which is labelled "X", was also observed in the PL spectra of those samples annealed at low temperatures. This feature is believed to originate from the bigger interstitial clusters. Similar optical structures were also demonstrated in Refs.[17] and [18].

The FWHM of the X line is about 10 meV in this AT range on average. The PL feature at 0.978 eV (1268 nm) is indistinct yet, and it is proposed that this emission peak is probably related to the interstitial complexes.[19] The peak located at 0.967 eV (1282 nm) could be attributed to the G line. Usually, a G-type centre is formed when one interstitial carbon atom is trapped at a substitutional carbon atom in silicon (C_iC_s).[12] This type of centre can be annihilated by annealing the silicon samples at a higher temperature for a long time.

When AT increases to 600 °C, great changes take place in the PL spectra. The PL line of the sample annealed at 600 °C mainly exhibits two broad peaks at 0.887 eV (1398 nm) and 0.936 eV (1325 nm), which are labelled S_1 and S_2, respectively. The S_1 and S_2 emission peaks can be ascribed to exciton recombination occurring in the strained regions which surround the small interstitial clusters embedded in the Si matrix.[8] The FWHMs of the S_1 and S_2 features are 38 and 43 meV, respectively. The peak at 0.767 eV (1616 nm) is ascribed to the P line, which is associated with a certain C centre perturbed by a second oxygen atom.[20,21]

For the samples annealed at 700 and 750 °C, it is interesting to note that a sharp emission peak labelled 'R' is observed between the S_1 and S_2 features with an FWHM of 6 meV at 7 K. It is proposed that the recombination of {311} rod-like defects should be responsible for this optical feature.[8,9] Although changes in feature of the PL spectra with the AT are continuous, there is no sharp critical temperature again. The W, S_1, S_2, and Si^{TO} features also can be observed in the same PL spectra of the sample annealed at 700 °C. This is very different from that reported in Refs.[7] and [8], in which only one or two of those features mentioned above can be detected.

More interesting features are observed in the PL spectrum of the sample annealed at 800 °C. As shown in Fig.1, a strong and broad peak D_1 is observed at 0.816 eV (1518 nm); probably this emission process is associated with the deep energy levels D_1,[22] and it can be detected until RET is as high as 280 K, at which the peak D_1 undergoes a red shift of 32 meV to 0.784 eV (1580 nm) in comparison with the peak position recorded at 7 K. The bound-exciton TO assistant emission peak Si^{TO} is observed at 1.101 eV (1126 nm) at low RETs. In fact, a very weak Si^{TO} peak has also been detected in the PL of samples annealed at 600 and 700 °C. It is likely that the intensity of Si^{TO} peak increases with the increase of AT, and further implies that the irradiated damage to the Si wafer, induced by self-implantation, is repaired remarkably and continuously in the AT range of 600–900 °C. The broad peak O_1 with an FWHM of 22 meV at 1.052 eV (1179 nm) is probably produced by heating Czochralski (CZ) silicon at high temperature;[12,23] it can also be attributed to a kind of defect-induced emission.

Despite the importance of FWHM in studying the optical properties of silicon, there has been little work on the mechanisms by which the line width of photoluminescence is generated. For example, there are little data on the FWHM of the luminescence. That luminescent centres exist in materials is an intrinsic character, so the FWHM is strongly dependent on the measuring method and techniques, such as the sensitivity and resolving power of the detector, and the wavelength and power of the exciting light. In general, the FWHM obtained by using a grating monochromator is much larger than by using a Fourier transform spectrometer. The results here are in agreement with the FWHM data of PL spectra reported in Refs.[8], [9] and [24], even though those authors did not emphasize it.

Figure 2 shows the temperature evolution of the PL spectra of the sample annealed at 400 °C. All the PL spectra exhibit a broad background in a range between 0.950 eV (1305 nm) and 0.75 eV (1653 nm), which is probably contributed by the collective emission of several damaged structures localized at different energy levels. These structures are usually produced by implanting particles into the matrix materials and cannot be destroyed even in our annealing environment at 500 °C for 30 min. However, well-resolved and shape optical peaks can be observed simultaneously in the high photon energy range ($>$ 0.950 eV). The PL spectrum at 7 K shows mainly W and X emissions, and very weak S_1 and S_2 peaks. Increasing RET to 40 K, the W line dominates the broad background. Further increasing RET to 80 K, the high energy luminescence is changed very weakly, but the intensity of the Si^{TO} feature increases with the increase of RET, which indicates the contribution of the TO phonon to the indirect transition. The broad band is always strong in the RET range; essentially, this complex luminescence may originate from a vacancy and often occurs in a low concentration region of small vacancy clusters in the near-surface.[19]

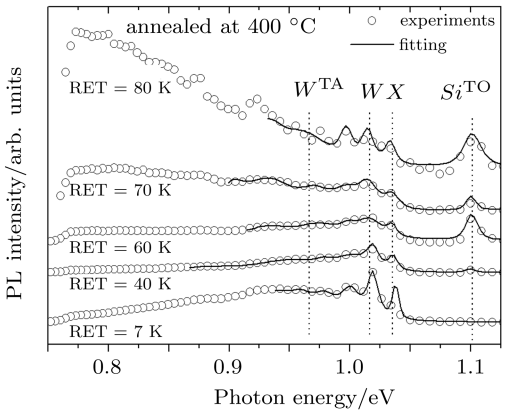


Fig.2. PL evolution of the p-Si sample self-implanted and then annealed at 400 °C with temperature. The Si^+ ion-implanting dose, energy, and subsequent annealing time are 3×10^{14} cm^{-2}, 300 keV, and 30 min, respectively.

PL curves for the sample annealed at 700 °C at different RETs are shown in Fig.3. All the PL spectra are basically dominated by the R line at 0.903 eV (1372 nm). At lower RETs (< 40 K), the apparent S_1 and S_2 bands and the weak G line at 1.000 eV (1240 nm) can be easily identified even in the absence of Gaussian line fitting. With the increase of RET, the intensities of the S_1 and S_2 peaks decreases continuously. Furthermore, the G line emission cannot be detected any more when RET is up to 60 K. This indicates that most of the excitons from the S and G bands have been excited to the extended {311} defects and undergone a non-radiative combination process, except for those related to the R line.

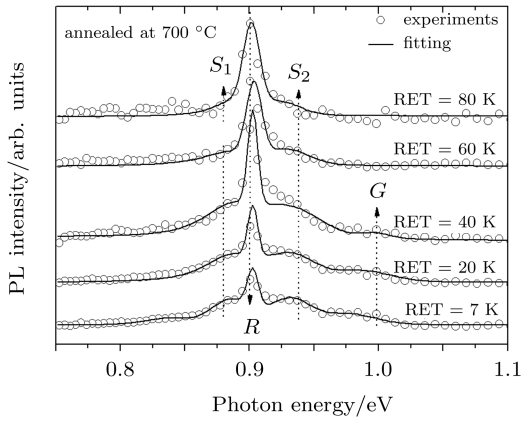


Fig.3. PL evolution of the p-Si sample self-implanted and then annealed at 700 °C with temperature. Except for the AT, the implantation and the annealing conditions of the sample are the same as those described in the captions of Figs.1 and 2.

After the Si^+ ion-implanted sample has been annealed at 900 °C, the concentration of the defects is reduced apparently. As a result, a very strong D_1 emission can be observed in a very broad temperature range (7–280 K), as shown by the typical PL spectra in Fig.4. This is very promising for optoelectronic applications in fabricating low loss fibres operating at $\sim$ 1.55 μm for telecommunications. It indicates that the emission characteristic of the silicon matrix materials can be tailored as an efficient light emitter operating in a near-infrared band through the self-ion-implanting process. Additionally, another weak feature D_2 related to the dislocations has also been detected in the whole RET range. This peak exhibits an average FWHM of 21 meV and a red-shift of 53 meV which is larger than that of the D_1 peak (32 meV). It indicates that the dislocations related to the D_2 band are more sensitive to temperature than those related to the D_1 band. Similar to those samples annealed at other temperatures, the S_1 feature of the sample annealed at 900 °C cannot be detected in the PL curve recorded at 160 K as shown in Fig.4.

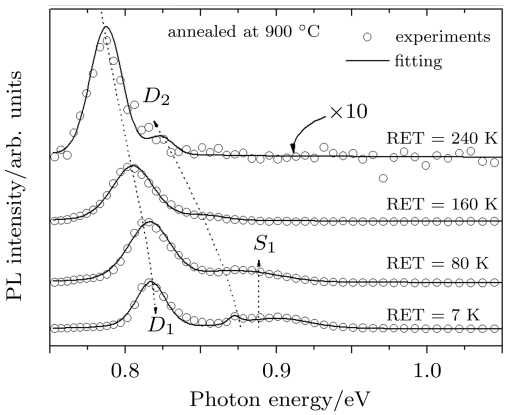


Fig.4. PL evolution of the p-Si sample self-implanted and then annealed at 900 °C with temperature. Except for the annealing temperature, the implantation and the annealing conditions of the sample are the same as those described in the captions of Figs.1 and 2.

All the characteristic peaks, such as W line, R line, S_1 band, S_2 band, and D band, were measured in a temperature range from 7 to 280 K. The PL peaks from the D_1 band, W line, R line, S_1, and S_2 bands disappear at about 280, 110, 160, 130, and 110 K, respectively. It is assumed that the PL intensity can be expressed approximatively in the exponential function,[12] i.e. $I(T) = I(I_0)\exp(E/k_BT)$. Then, one can obtain the deactivation energies of the D_1 band, W line, R line, S_1 band, and S_2 band to be 186, 54, 26, 34, and 37 meV, respectively. Although these data are not sufficiently accurate and the behaviour changes a little from sample to sample, the

very high deactivation energy of some features implies great potential applications in high temperature optoelectronic devices.

Some experimental techniques, such as PL spectroscopy, electron paramagnetic resonance (EPR), positron annihilation signal (PAS), and deep level transient spectroscopy (DLTS), are utilized to study the formation, migration and evolution of the interstitial clusters and vacancies in silicon. The I_2- and I_4-clusters in silicon are also identified by using the electron spin resonance (ESR) technique. In fact, the single vacancy works as a fast-diffusing species. Depending on its charge state, the vacancy can turn into a mobile particle at temperatures above 150 K, and be trapped by impurities such as dopants, group-IV species, interstitial oxygen or other vacancies.[25] Stable vacancy centres at room temperature are divacancies,[26] and at higher temperatures they are converted into multi-vacancy centres (V_3 and V_4, even V_5 and V_6), which were also assigned by the EPR and other optical measurements.[27−29]

Several characteristic PL peaks due to I-clusters are observed in both n-type and p-type Si. It was proposed by Commer *et al*[30] that the zero phonon line (X band) at 1.040 eV in the PL spectra and the infrared absorption peaks at 530 and 550 cm^{-1} should be attributed to the defects formed from the tetra-interstitial I_4-clusters. This identification was confirmed in a subsequent study.[31] The non-phonon-assistant W line at 1.018 eV probably originates from self-interstitial aggregates, and its emission mechanism is closely related to the radiation combination of the I_3-clusters.[32] This kind of defect is formed in most types of irradiated Si and is not connected with any impurities. Studies on the stress show that the symmetry of the W-line is trigonal, and the magnitude of the stress splitting of this PL line suggests that an interstitial centre is bonded into the lattice with rather strong bonds.[33]

The PL measurements indicate that the X centre forms at the expense of the W centre. However, the X centre does not form simultaneously with the W centre even at elevated temperatures due to the higher formation-temperature of the X centre. Therefore, the former is generally more stable than the latter. The number of interstitials in the X centre is proposed to be larger than that of the W centre, according to the concept of ripening of the clusters with the increase of AT.[34,35] It can be concluded that the X centre defect must evolve from a possible combination of the W centre defects. As a result, the W centre may be composed of a half unit of the X centre or I_4–cluster, and the I_2-cluster is most likely a candidate for the basic structure of the W centre. In fact, the I_2-cluster can easily be combined and form an I_4–cluster on applying thermal energy. Therefore, it is proposed in this work that the $\langle 111 \rangle$ split triple (ST) di-interstitial defects are just the defects which should be responsible for the emission of the W-line.

The S_1 and S_2 peaks show average FWHMs of ∼38 and ∼43 meV, respectively. The two FWHM values are quite a lot larger than those of W and X lines. The large FWHMs of the S_1 and S_2 bands indicate that related defects have spread in the activation energy and are likely to form an extended structure. The presence of lattice strain and the extended structure of the corresponding I-clusters could be used to interpret the broad S_1 and S_2 emission-bands corresponding to a lattice-strain of about 2%.[9] According to the proposition given by Kim *et al*,[36] the stable defect structure evolves from a compact to a chain-like then to a rod-like {311} defect with the increase of interstitial number. Giri *et al*[9] attributed broad S_1 and S_2 to the I-chains which form in an intermediate temperature range (400–600 °C). For the sample annealed at 700 °C, the R feature at 0.903 eV results from the recombination of excitons at the {311} rod-like defects. The FWHW (6 meV) of the R line is of the same order as those observed in the W and X bands. Therefore, the formation of the {311} defects involves the release of strain from I-chains or extended I-clusters. These defects induce the production of a well-defined energy level in the forbidden gap of the silicon. This usually leads the corresponding PL peak to be very sharp.

The D_1 peak was observed not only in the ion-implanted silicon, but also in the plastically deformed and then annealed silicon by using a continuous wave laser.[37] It was confirmed by Sauer *et al* that the D_1 and D_2 bands were similar in nature and corresponded to bound-to-bound transitions.[22] In fact, the D_1 and D_2 emissions originate from deformation-induced point defects located in a strain region of the curved dislocation. For low dislocation densities ($< 10^6$ cm^{-2}), the D_1 line becomes much narrower and exhibits a double fine structure.

4. Conclusions

The photoluminescence evolution of Si^+ ion-implanted and annealed silicon wafers with recorded temperature and annealed temperature is investigated. For the self-implanted Si samples with annealing temperature below 750 °C, all the PL spectra exhibit an underlying broad band between 1180 nm (1051 meV) and 1620 nm (765 meV), in which these visible peaks are ascribed to the W line, R line, S bands, X line, etc, respectively. As for the sample annealed at lower temperatures ($\leq$ 200 °C), the broad band in the PL spectra is concentrated at 1.550 μm. For those samples with annealing temperatures in the range 250–350 °C, the W line becomes the main feature of the PL curves. For those samples annealed between 400 and 650 °C, the S_1 and S_2 bands dominate the PL spectra. For those samples annealed between 700 and 750 °C, the dominant feature is an R line at 0.903 eV. For those samples annealed at 800 °C, the PL spectra mainly show emissions originating from the deep energy levels, such as the D_1 and D_2 bands, which can be observed until the recorded temperature reaches as high as 280 K. With the increase of annealing temperature and the evolution of defects from bigger interstitial clusters, to small interstitial clusters, to rod-like defects, and to deep energy levels, the dominant emission peaks in the PL spectra change from the X line, to the S band, to the R line to the D band correspondingly. It is indicated that the subrogation of the characteristic PL peak in the implanted Si wafers reflects the defect evolution induced by different annealing temperatures in nature.

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