Temperature Dependence of Photoluminescence Spectra from Crystalline Silicon

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Photoluminescence (PL) spectra from lightly boron (B) doped p−Si(100) under 488.0 nm Ar+ ion laser excitation over the temperature range of 22 K–290 K is presented. Change of PL peak height (maximum intensity), peak position, peak area (areal intensity), full-width-at-half-maximum (FWHM: peak width) were determined as a function of temperature. PL intensity was sharply decreased with temperature increase in the temperature range of 22 K – 170 K and then slowly increased again in the temperature range of 170 K–290 K. Phonon replicas of a relatively sharp band-to-band (or band edge (BE)) PL peak were clearly measured at low temperatures (<90 K). PL spectra became broader and the phonon replicas were barely distinguishable as the temperature was increased. The envelope of the main PL peak and the broadening of peak width with the Si temperature were qualitatively in good agreement with the Maxell-Boltzmann probability distribution function. The direction of peak position shift with Si temperature change was also in good agreement with the temperature dependence of the Si bandgap. All measured PL spectra were curve fitted using combinations of modified Gaussian function(s) and standard Gaussian function(s). A simplified curve fitting method for broad PL spectra, consisting of the BE peak and band tail peak, using an exponentially modified Gaussian (EMG or ExGaussian) function and a number of standard Gaussian functions, was proposed from a practical usage point of view. Radiative recombination processes in Si and potential industrial applications of the PL characterization technique were discussed.

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For the characterization of electrical behaviors of Si, resistivity, carrier concentration, mobility and Hall effect measurements were routinely inspected during wafer fabrication and incoming quality control at the wafer fabs. A number of noncontact characterization techniques, such as microwave detected photodcoconductance decay (µ-PCD)1,2 and inductively coupled quasi-steady-state photoconductance (QSSPC),3,4 have been used for in-line monitoring of contamination and process induced modulation of electrical behaviors of Si. Steady state PCD measurement techniques5 enabled by improved laser technology were recently introduced.

With the introduction of photoluminescence (PL) imaging and spectrum analysis, a noncontact, purely optical technique is now available for spatially resolved electrical characterization.6,7 Qualitative raw room temperature photoluminescence (RTPL) imaging and spectrum analysis of various types of Si wafers, such as; single crystalline Si wafers,7 epitaxial Si wafers,8 bonded single crystalline Si wafers,9 implanted single crystalline Si wafers,10–12 plasma processed single crystalline Si,13–16 single crystalline Si on oxide (SOI) wafers,17 Si implanted oxide (SIMOX) wafers18 and microcrystalline-Si (mc-Si) wafers sliced from brick,6,19 has been examined. Correlation has been demonstrated between RTPL intensity and various characteristics of Si wafers, such as, distribution of defects, contamination, implant damage, plasma damage, interface quality, impurities, bulk lifetime etc.

For industrial applications, the ease of measurement without special sample preparation is important. This is especially true for in-line monitoring applications in semiconductor device manufacturers. PL measurements at room temperature (RT) are preferred. However, the RTPL spectra from Si are generally very broad and it is difficult to identify the origin of variations in RTPL spectra. In this paper, temperature dependence of PL spectra from lightly boron (B) doped p−Si(100) was measured in the temperature range of 22 K–290 K and the effect of temperature on the change of PL spectra was investigated. Practical PL spectra curve fitting methods were tested.

Experimental

A lightly B doped p-type Si wafer (double side polished, 775 µm thick, 300 mm diameter) with a resistivity of ∼10 Ω·cm (equivalent to a B concentration of 1.3 × 1019 cm−3), which is typically used in device manufacturing, was chosen for this study. A small sample (10 mm × 10 mm in size) was cut out from the center of the wafer. No special sample treatment was done.

PL spectra were measured using 488.0 nm laser radiation from a 24 mW Ar+ laser. The laser beam was focused on the sample. The diameter of the focused laser beam at the sample surface was ∼44 µm. The excitation power density at the sample surface was ∼3 kW/cm2. The laser beam was passed through a 488.0 nm interference filter (laser line filter) to excite the sample. The PL signal was passed through a Shamrock SR-163 (Andor Technology) 163 nm monochromator with a grating blazed at 1200 nm. A four-stage thermoelectric (TE) cooled InGaAs detector array (DU490-A-1.7, Andor Technology) with a pixel width of 500 µm was used as a detector. The detector was operated at −60 °C during measurements. The sample was mounted on the cold finger of a closed-cycle liquid helium (He) cryostat. The temperature of the cold finger was measured and controlled at predetermined set points in the range of 22 K–290 K. We verified that the 488.0 nm excitation laser beam irradiation did not significantly heat the sample by comparing spectra measured at a given temperature with variable laser power.

Results and Discussion

Temperature dependence of PL spectra.— PL spectra measured in the wavelength range of 1040 – 1275 nm were plotted in both linear and semilogarithmic scales in Figs. 1a and 1b. To make peak identification easier, the photon energy converted from wavelength was used as the first X-axis and the corresponding wavelength was indicated as the second X-axis, on top of the graph.

A seen in Fig. 1a, the PL intensity was the strongest at the lowest temperature of 22 K and sharply dropped with temperature increase up to 170 K. The PL intensity slowly increased up to 290 K. The maximum PL intensity changed 27 times in the temperature range of 22 K–290 K (maximum at 290 K and minimum at 170 K). Spectral distribution also showed significant change with temperature. However, it is difficult to see the details in the superimposed linear scale plot. The semilogarithmic plot in Fig. 1b shows a clear picture of the change of spectral distribution with temperature. For PL spectra measured at or below 90 K, several distinctive peaks, with certain intervals, are evident. The main peaks are positioned near 1.097 eV and the effect of temperature on the change of PL spectra was investigated. Practical PL spectra curve fitting methods were tested.

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the temperature decreases from RT. The bandgap of Si at low temperature is expected to be larger than that for RT. The peak near 1.137 eV (≈1090 nm) is obviously above the bandgap. The other two peaks at 1.037 eV and 1.077 eV (≈1195 nm and ≈1151 nm) and one hump at 1.057 eV (≈1173 nm) are located below the main peak. All peaks and humps are separated by multiples of 20 meV. The peak (at 1.137 eV), above the bandgap, is 40 meV higher than the main peak (at 1.097 eV).

The PL signal is dominated by indirect band-to-band transitions, which are phonon-assisted recombinations of electron-hole (e-h) pairs. Above the bandgap energy, photon emission is accompanied by phonon absorption. Below the bandgap energy, photon emission is accompanied by phonon emission. The peaks and humps in the spectra that correspond to the emission of photons with absorption and emission of variable numbers of phonons are indicated in Fig. 1b. Four types of phonons (TA = 18.3 meV, LA = 57.7 meV, TO = 90.5 meV and LO = 122.4 meV) in Si are involved in the indirect absorption, with temperatures of 212 K, 670 K, 1050 K and 1420 K, respectively. Different values of phonon energies are also reported from vacuum grown Si in a lattice absorption study in the temperature range of 20 K – 365 K. Table I summarized the Si associated phonon energies and temperatures from absorption study by MacFarlane et al. and Johnson. Since the spacing between peaks are multiples of 20 meV, the peaks and humps are denoted as TA phonon replicas associated with the indirect band-to-band recombination of e-h pairs.

Figure 1. Temperature dependence of PL spectra from p-Si (100) wafer in the temperature range of 22 K–290 K: (a) linear scale plot and (b) semilogarithmic scale plot (488 nm Ar+ laser excitation).

Table I. Si associated phonon energies and corresponding temperatures from absorption study.20,21

|   | TA   | LA   | TO   | LO   |
|---|------|------|------|------|
| Si | 18.3 | 57.7 | 90.5 | 122.4 |
| 15.8 | 71.5 | 41.4 | 694  | 1420 |

The sensitivity and low noise measurement capability of our experimental set up made the PL spectrum measurements of approximately five orders of magnitude into the spectral range possible. The emission of photons accompanied by the simultaneous emission (the phonon replicas) of up to four phonons was measured without special sample preparation.

Band-to-band transition mechanism.— For indirect semiconductors such as Si and Ge, optical transitions require a change in both energy and momentum. A two-step process is required because the photon cannot provide a change in momentum.22–24 Momentum is conserved via a phonon interaction as illustrated in Fig. 2a. Schematic energy band diagrams and density distributions of electrons in the

Figure 2. Schematic illustrations of (a) radiative recombination processes in Si, (b) energy band diagram and (c) density distribution of electrons in conduction band and holes in valence band.
Conduction band (CB) and holes in the valence band (VB) are shown in Figs. 2b and 2c, respectively. To complete the transition, a phonon is either emitted or absorbed. In optical excitation of indirect semiconductors, photons and phonons are required simultaneously for the generation of an electron in the conduction band and a hole in the valence band by changing both energy and momentum.22,25,26

During optical excitation, photon absorption simultaneously requires either phonon absorption or photon emission. Photon absorption with either phonon absorption or photon emission can also accompany stimulated photon emission with either phonon absorption or photon emission. During radiative recombination between electrons in CB and holes in VB, spontaneous photon emission also requires either phonon emission or phonon absorption for momentum change. Wavelength and photon energy of emitted photons are determined by the bandgap (Eg) and the energy of associated phonon. When a phonon is absorbed during photon emission, the photon energy will exceed the bandgap. When phonon(s) are emitted with a photon, the wavelength and photon energy will be reduced by the sum of emitted phonons from the bandgap energy (Eg).

At very low temperatures, the phonon density is very small. Therefore, the absorption constant for a transition with phonon absorption is also small. The number of phonons for absorption during optical excitation is very small at lower temperatures. Phonon absorption during photon emission is expected to be negligible. However, the emission of photons by electrons which are already at a high-energy state is very probable.22 Figure 2a shows the possible paths for optical excitation and emission with phonon absorption and emission. The emission of photons by electrons in the conduction band with phonon absorption and emission is shown in Fig. 1b. PL spectra with phonon replicas, can be explained by the illustration of the radiative recombination process in Si (Fig. 2a).

As the temperature is increased, the bandgap narrows and the energy distribution broadens. Thus, the main peak of the PL spectra broadens and small peaks and hump(s) smear each other. Identification of peaks becomes difficult.22,25,26

Normalized PL spectra.— Temperature dependence of normalized PL spectra from a p–Si (100) wafer and normalized Maxwell-Boltzmann probability distribution27 in the temperature range of 22 K–290 K are plotted in Figs. 3a and 3b, respectively. The peak positions of the Maxwell-Boltzmann probability distribution were manually adjusted to the measured PL spectra for comparison. For PL intensity as a function of photon energy, the following relationship was assumed.

\[ I_{PL} \propto \sqrt{E - E_g(T)} \exp \left( \frac{E - E_g(T)}{k_B T} \right) - Adjustment \]  

where \( E \) and \( E_g(T) \) are photon energy and the bandgap at temperature \( T \) in K, \( k_B \) and \( T \) are Boltzmann constant \( (k_B = 1.38065 \times 10^{-23} \text{ J/K}) \) and temperature in K.

The shape of PL spectra and normalized probability distributions seemed to be similar. The temperature dependence of normalized probability distribution also seemed to show similar trends with the PL spectra. As seen in Figs. 1a and 1b there are many phonon replicas in the PL spectra. Since the normalized probability distribution does not consider phonon replicas, the shapes are generally sharper and narrower than the measured PL spectra. However, the normalized probability distribution curve provided a general idea of PL spectra at different temperatures and the effect of temperature on PL spectra.

The temperature dependence of the gap for many semiconductor materials has been fitted by the Varshni’s empirical relation2: \n
\[ E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta} \]  

where \( E_g(0) \) is the energy gap at 0 K and \( \alpha \) and \( \beta \) are constants. \( E_g(0) \), \( \alpha \) and \( \beta \) for Si are 1.166 eV, 4.73 × 10^{-4} and 636, respectively. According to the equation, the \( E_g \) changes from 1.166 eV at 22 K to 1.123 eV at 290 K. \( E_g \) becomes approximately 43 meV wider as it cools down to 22 K. The change in bandgap is on the order of phonon energy and the absorption and/or emission of a combination of phonon(s) is accompanied in a photon emission (e-h pair radiative recombination), it is difficult to predict accurate PL spectra. However, it is possible to analyze the PL spectra and gain additional information on materials.

PL spectra curve fitting.— For simplicity, the PL spectra were fitted with an exponentially modified Gaussian (ExGaussian) function and a number of standard Gaussian functions. The main peak was fitted using the ExGaussian function and the rest of the peaks were fitted using the standard Gaussian function as follows:

\[ y = k_0 \exp \left( -\frac{(x - k_6/\kappa)^2}{2\kappa^2} \right) + k_3 \times \exp \left( -\frac{(x - k_6/\kappa)^2}{2\kappa^2} \right) \]  

where \( k_0 \) is baseline intensity and \( k_1 \sim k_6 \) are fitting coefficient. The \( x \) is photon energy in eV.

Figure 4 shows a curve fitting summary of 488.0 nm excited PL spectra in the temperature range of 22 K–290 K. Measured PL spectra, deconvoluted peaks, curve fitted spectra and fitting residuals are plotted for each PL spectra measured at different temperatures. All PL spectra were fairly well fitted. The ExGaussian curves have captured characteristics of the main PL peak from the band-to-band transition including temperature dependence of the main PL peak width.

Temperature dependence of (a) peak height (intensity maximum), (b) center wavelength, (c) peak area (areal intensity) and (d) FWHM of the main band-to-band PL peak (ExGaussian fitted peak) from
Figure 4. Curve fitting summary of 488.0 nm excited PL spectra in the temperature range of 22 K–290 K. Measured spectra, deconvoluted peaks, curve fitted spectra and residuals are shown for each PL spectra measured at different temperatures in the same temperature range.
p-Si(100) wafer in the temperature range of 22 K–290 K under 488 nm excitation were plotted in Fig. 5. The peak height of the ExGaussian fitted main peak decreased with the sample temperature. The peak wavelength was shifted from 1.099 eV (~1129 nm) at 22 K to 1.078 eV (~1150 nm) at 290 K. As the temperature was increased, the peak area started to decrease and reached a minimum at 170 K then increased significantly near room temperature. The peak width (FWHM: full-width-at-half-maximum) increased from 6.4 nm at 22 K to 202.6 nm at 290 K. Simplified curve fitting using an ExGaussian function with a number of standard Gaussian functions has captured all the important characteristics of PL spectra. This simplified curve fitting is found to be very convenient and useful for broad PL spectra measured at higher temperature toward in-line monitoring of surface, interface and/or bulk Si quality in semiconductor or solar cell applications.

**Discussions.**— PL characterization can reveal a lot of insights into Si properties related to electrical properties since the PL signal is the result of e-h pair recombination. Photo-generated (photo-excited) e-h pairs must travel within the CB and VB by density gradients and coulomb repulsion induced diffusion among the same type of carriers before radiative recombination to emit photons during PL. The photo-generated carriers have to go through the paths where carriers will travel in semiconductor devices. Electrical characterization is obtained without making electrodes or physical contacts. Temperature varying PL measurements can provide more information than PL measurements at RT. It can be very powerful for off line material analysis and failure analysis of devices. However, the temperature varying PL measurement technique is impractical for industrial in-line monitoring applications. Temperature cycling of large diameter Si wafers is impractical. For in-line monitoring applications, PL measurement at RT is strongly desired. The use of RTPL intensity measurements and spectrum measurement in various steps during Si wafer processing have been reported for characterizing epitaxial layer quality, possible contamination, dielectric/Si interface quality, passivation quality, degree of implant activation, degree of process induced damage and spatial distribution of electrical properties. RTPL intensity imaging showed the correlation between PL intensity and the bulk lifetime of multicrystal-Si (mc-Si) bricks for solar cell applications. Full spectrum analyses showed clear correlation between the FWHM of normalized RTPL signal and bulk lifetime. Narrow RTPL signals are measured from portions of mc-Si bricks with longer lifetime. Broad RTPL signals are measured from the portions of mc-Si bricks with shorter lifetime. Spectral intensity ratio of RTPL measured using different types of imaging devices with different spectral responses enabled bulk lifetime imaging, doping imaging and interstitial iron (Fe) imaging for mc-Si bricks. RTPL measurements are very convenient and powerful for characterizing solar cell grade Si materials with large lateral Si property variations. A purely optical technique is now available for spatially resolved electrical characterization by introduction of luminescence imaging. Numerous qualitative and quantitative methods based on PL and electroluminescence (EL) imaging have recently been developed for solar cell applications. These luminescence imaging-based, noncontact electrical characterization techniques are widely adapted in characterizing Si bricks for solar cell applications prior to wafer slicing. They are based on photoconductance measurement principles and spatially resolved data (maps), typically generated by raster scanning of the Si brick surface.

It was reported that the surface topology of Si wafers strongly affects the luminescence spectra due to the change in photon escape probability of luminescence from PL spectra measurements of Si wafers with various surface finishing conditions. Double side polished Si wafer peaks around 1140 nm. Si wafers with textures on both sides showed PL spectra peaks around 1170 nm due to the enhancement of the emission of long-wavelength photons. A peak shift of about 30 nm toward the longer wavelength side was reported. Single side polished Si wafers also peaked around 1170 nm, but the enhancement of long-wavelength photon emission was not as strong as for double side textured Si wafers. We have found similar effects in bonded Si wafers with different surface finishing conditions. We also found that the wafer thickness and dielectric layers on Si wafers influence the PL spectra due to the interference between the Si wafer and/or dielectric films. When comparing RTPL spectra between Si wafers with different surface finishing (different thicknesses and/or different structures), great attention must be paid.
If a pn junction is present, photo-generated e-h pairs in the junction area (depletion layer) will be separated by the built-in potential across the junction and will be isolated from each other as they occur within a solar cell. The separated charges can cause band bending and move physical positions of e-h pair radiative recombinations. In our RTPL measurements on reverse biased pn junctions with a transparent electrode in the photo excitation side, proportional RTPL intensity increase with Si temperature change was also in good agreement with the temperature distribution function. The direction of a peak position shift with Si temperature increase, was qualitatively consistent with the temperature dependence of the Si bandgap. Radiative recombination processes in Si are temperature dependent, can be identical, can be done very easily by comparing various characteristics of PL spectra measured under the same excitation condition, such as spectral distribution, intensity, peak position and FWHM at a given measurement temperature. Few examples of RTPL applications in Si device processes can be found in previous reports. In-line monitoring of materials and devices which are expected to be identical, can be done very easily by comparing various characteristics of PL spectra measured under the same excitation condition, such as spectral distribution, intensity, peak position and FWHM at a given measurement temperature. Few examples of RTPL applications in Si device processes can be found in previous reports. 1

Summary

Temperature dependence of PL spectra from lightly boron (B) doped p-type Si (100) under 488.0 nm Ar+ ion laser excitation over the temperature range of 22 K–290 K was studied. A simplified curve fitting method for broad PL spectra, consisting of the BE peak and band tail peak, using an ExGaussian function and a number of standard Gaussian functions, was proposed from a practical usage point of view. Changes of PL peak height (maximum intensity), peak position, peak area (areal intensity), FWHM were determined as a function of temperature after curve fitting using the proposed ExGaussian function and a number of standard Gaussian functions. PL intensity was sharply decreased with temperature increase in the temperature range of 22 K – 170 K and then slowly increased again in the temperature range of 170 K–290 K. Phonon replicas of a relatively sharp band-to-band (or band edge (BE)) PL peak were clearly measured at low temperatures (<90 K). PL spectra became broader, while the phonon replicas were not easily distinguishable, as the temperature was increased due to the broadening of the main PL peak and peaks from phonon replicas. The envelope of the main PL peak and the broadening of peak width, with Si temperature increase, were qualitatively in good agreement with the Maxwell-Boltzmann probability distribution function. The direction of a peak position shift with Si temperature change was also in good agreement with the temperature dependence of the Si bandgap. Radiative recombination processes in Si and its temperature dependency were discussed. Potential industrial applications of the PL characterization technique were discussed with examples given.

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