Excitation power density and temperature dependence of photoluminescence study on electron-irradiated GaAs middle cell of GaInP/GaAs/Ge triple-junction solar cell

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Abstract. Excitation power density (2.603-9.196 W/cm²) and temperature (10-300 K) dependence of photoluminescence (PL) measurements have been presented and discussed in detail for the carrier recombination mechanism and quenching of the GaAs middle cell in 1.0 MeV electron-irradiated GaInP/GaAs/Ge triple-junction solar cell. When the excitation power density increases, the PL intensity increases. The dependence of PL intensity on excitation power density is observed to be linear below 40 K, and becomes increasingly superlinear with increasing temperature and eventually quadratic at 300 K. The observed phenomenon revealed a competing mechanism between radiative and non-radiative recombination process of photogenerated carriers. However, with the increase of temperature, the PL intensity decreases, and strong thermal quenching phenomenon of PL intensity can be observed above 40 K. By analyzing with multiple centre model, it is due to the presence of thermally activated non-radiative recombination centres named E4 electron trap with an activation energy of ~0.76 eV and named E5 electron trap with an activation energy of ~0.96 eV.

Keyword: GaInP/GaAs/Ge; Nonradiative Recombination Centers; Carrier Recombination Mechanism; Thermal Quenching; Activation Energy

1. Introduction
Multi-junction solar cell, especially GaInP/GaAs/Ge triple-junction (3J) solar cell, have been used for space application widely because of its high photoelectric conversion efficiency [1]. However, in space application, performance of solar cells was degraded because they were explored to a hostile space irradiation environment composed of various charged energetic particles including electrons [2]. And the existence of defects which induced by charged energetic particles irradiation and acted as non-radiative recombination centers is the primary reason of space solar cells’ performance degradation. These defects deduce the minority-carrier diffusion length and lifetime by capturing free carriers, as a result, the performance of solar cell declined [3]. In addition, among the three subcells of electron-irradiated 3J solar cell, the GaAs middle cell’s performance degradation is the most serious [4,5], so to understand the performance degradation of electron-irradiated 3J solar cell profoundly, it is necessary to investigate the carrier recombination mechanism through defects and identify the main defects causing the performance degradation in electron-irradiated GaAs middle cell. Therefore, in this paper, the carrier recombination mechanism through defects has been analyzed by the excitation power density...
dependent photoluminescence (PL) measurements, and the main defects affected the performances of electron-irradiated GaAs middle cell have been identified via temperature dependent photoluminescence measurements.

2. Experiments
The GaAs middle cell of GaInP/GaAs/Ge 3J solar cells investigated here was grown by the metal organic chemical vapor deposition (MOCVD) technique. The detail structure of the GaInP/GaAs/Ge 3J solar cells is displayed in reference [6]. The 3J solar cells were irradiated by 1.0 MeV electron at room temperature, with the fluence of 5×10^{14} cm^{-2}. The current-limited subcell is GaAs middle cell in GaInP/GaAs/Ge 3J solar cell after 1.0 MeV electron irradiation with this fluence [7].

The temperature and excitation power density dependent PL measurements were carried out by an adjustable solid-state laser emitting at ~ 730 nm (1.70 eV), which energy is above the GaAs band gap for any temperature. The maximum power density of this laser is ~ 10 W/cm^2. The luminescence signal of the GaAs middle cell was dispersed by a monochromator and detected by a Si photodetector. And then the luminescence signal detected was measured by a lock-in amplifier and transferred to a computer for data processing. The excitation power density (I_{ex}) was varied from 2.603 to 9.196 W/cm^2. A closed-cycle cryogenic refrigerator equipped with a digital thermometer controller was employed for the temperature range of 10 K to 300 K, and the temperature fluctuations (not more than 0.1 K) can be neglected.

Figure 1. (a) excitation power density dependent PL spectra of electron-irradiated GaAs middle cell at T=150 K. (b) PL spectra of electron-irradiated GaAs middle cell with the temperature range of 10-300 K at I_{ex}=8.729 W/cm^2.

3. Results and discussion
The excitation power density dependent PL spectra of GaAs middle cell after radiation with 1.0 MeV electron with the fluence of 5×10^{14} cm^{-2} at 150 K were exhibited in Figure 1 (a). As shown in figure 1 (a), the PL intensities increase with increasing excitation power densities. This is because more carriers are activated from localized regions and recombined through radiative recombination channels. According to figure 1 (a), the PL peak energy contributed to band to band transition of carriers is ~1.421 eV with the excitation power density of 2.603 W/cm^2, and when the excitation power density increases from 2.603 W/cm^2 to 4.428 W/cm^2, 5.970 W/cm^2, 7.031 W/cm^2, 7.753 W/cm^2, 8.290 W/cm^2, 8.729
W/cm$^2$, and 9.196 W/cm$^2$, the PL peak energy decreases to 1.419 eV, 1.418 eV, 1.416 eV, 1.415 eV, 1.411 eV, 1.410 eV, and 1.407 eV, respectively. Apparently, a slight redshift phenomenon of PL peak energy with increasing excitation power density was observed, which can be illuminated by the heating effect of excitation laser. When the excitation power density increases, the temperature of the excitation regions of GaAs middle cell will increase, correspondingly. In addition, according to Varshni’s research [8], the band gap of GaAs material will shrink with the increase of temperature, which causes the redshift of PL peak. Figure 1 (b) shows the PL spectra of electron-irradiated GaAs middle cell with the temperature range of 10 K-300 K and a constant excitation power density of 8.729 W/cm$^2$. According to figure 1 (b), the PL intensities decrease with the increase of temperature. Examination of figure 1 (b) shows that the 10 K spectrum is determined by a sharp emission peak centered at ~1.423 eV, and with increasing temperature from 10 K to 40 K, 100 K, 150 K, 220 K, 270 K, and 300 K, the PL peak energy decreases to 1.418 eV, 1.413 eV, 1.408 eV, 1.402 eV, 1.397 eV, and 1.389 eV, respectively. With the increase of temperature, an obvious redshift phenomenon of the PL peak was observed. The PL peak redshift because of the increase of temperature is dominated by the band gap shrinkage following the Vanish law [8].

![Figure 2](image_url)  
**Figure 2.** Log-Log plots of integrated PL intensities versus excitation power densities of electron-irradiated GaAs middle cell at different temperatures. The solid lines are best fits for the experimental data to equation (2).

A comprehensive study of the relationship between integrated PL intensities ($I_{PL}$) and excitation power densities with the temperature range of 10 K-300 K have been made to further investigate the dependence of integrated PL intensities on the excitation power densities. Figure 2 presents the Log-Log plots of integrated PL intensities versus excitation power densities of electron-irradiated GaAs middle cell at different temperatures. From figure 2, at different temperatures, integrated PL intensities and excitation power densities show different dependence. In fact, the integrated PL intensities are directly related to the photogenerated carriers which participate in the radiative recombination process. For a given level of monochrome laser power, the $I_{PL}$ is proportional to the radiative recombination rate $R(T)$ of electrons and holes via band-band transitions by [9,10]:

$$I_{PL} \propto R(T) = B(T)N_eN_h$$  

where $B(T)$ represents radiative recombination coefficient, and with the increase of temperature, the values of $B(T)$ were found to decrease [9], $T$ is the sample temperature. And $N_e$ and $N_h$ are electron and hole concentrations.
In addition, the $I_{PL}$ is also related to the excitation power density $I_{ex}$, and the relationship between $I_{PL}$ and $I_{ex}$ can be written as [11,12]

$$I_{PL} = \beta \cdot I_{ex}^z$$

(2)

where $\beta$ and $z$ are unknowns, and the value of exponent, $z$, is related to the dominant carrier recombination process. The solid lines in figure 2 are best fits for the measured data using equation (2) to extract the value of slope $z$ in Log-Log plot. As shown in figure 2, at low temperature range (lower than 70 K), the value of $z$ is close to 1, when temperature is higher than 70 K, the values of $z$ increase with increasing temperature, and considering the experimental errors, $z$ is equal to 2 at 300 K.

We consider a competing mechanism between radiative and non-radiative recombination of photogenerated carriers to explain the superlinear dependence between integrated PL intensities and excitation power densities. Under stable excitation and neglecting the dopant-related electrons, the relationship between excitation power density and photogenerated carrier concentration can be given by [13]:

$$\frac{\alpha I_{ex}}{E_0} = \left( G + \frac{GD}{R+D} \right) (N_eN_h) + \frac{N_e}{\tau_e}$$

(3)

where $E_0$ is the photo energy of excitation laser; and $\alpha$ is the absorption coefficient of GaAs materials for photo energy of excitation laser; $G, D,$ and $R$ are the electron-hole-pair generation rate, exciton dissociation rate, and recombination rate, respectively; $\tau_e$ is non-radiative recombination lifetime of electron; $N_e$ and $N_h$ have the same physical means as mentioned above. The first item in Equation (3) is to describe the radiative recombination process and the second item is to describe the non-radiative recombination process. For a stationary temperature, the $I_{PL}$ is proportional to $N_eN_h$ from equation (1). And according to equation (3), if the radiative recombination mechanism dominates the recombination process, the excitation power density $I_{ex}$ is proportional to $N_eN_h$, so $I_{PL} \propto I_{ex}$. In this case, the value of $z$ equal to 1, that is the $z$ value of 1 corresponding to radiative recombination process. The is the case observed in figure 2 for temperature below 70 K. On the contrary, if the non-radiative recombination mechanism governs the carrier recombination process, $I_{ex}$ is proportional to $N_e$, and $I_{PL}$ is proportional to $N_eN_h$, so $I_{PL} \propto I_{ex}^2$, that is the $z$ value of 2 corresponding to non-radiative recombination process. The case of quadratic dependence between integrated PL intensities and excitation power densities can be observed at 300 K considering the experimental errors. If the main recombination process includes both non-radiative and radiative recombination, $z$ has a value between 1 and 2. Therefore, for given experimental temperature range, we can draw the conclusion that the relationship between integrated PL intensity and excitation power density changes from linear dependence to quadratic dependence with increasing temperature, and the carrier recombination mechanism changes from radiative recombination to non-radiative recombination with increasing temperature. And from figure 1 (b), the thermal quenching of PL spectra has been observed, and we can consider that the reason of the PL thermal quenching is the non-radiative recombination centers activated with increasing temperature.

To further determine the defects acted as non-radiative recombination centres, a comprehensive study of the temperature dependent PL with excitation power densities range of 2.602-9.196 W/cm$^2$ have been made. Figure 3 displays the $I_{PL}$ of electron-irradiated GaAs middle cell as an inverse function of temperatures with excitation power densities of 2.602, 7.031, and 8.729 W/cm$^2$. The data for each excitation power density have been normalized to show the same $I_{PL}$ at 10 K, respectively. It is obvious that the $I_{PL}$ drops with increasing temperature and the quenching behavior can definitely slow down as the excitation power density increases. Three thermal quenching process can be seen in figure 3. At temperature below 70 K, a slight thermal quenching process can be observed, at the temperature range from 70 K to 220 K, a stronger PL quenching can be observed, and the most intense PL quenching occurs at temperature above 220 K.

Furthermore, an important formula which used for analyzing the thermal quenching phenomenon of PL for multiple centre model have been proposed [14,15], as followed:
\[
\frac{I_{PL}(T)}{I_{PL}(0)} = \frac{1}{1 + \sum_{j=1}^{m} C_j \exp \left( -\frac{E_j}{k_BT} \right)}
\]  

where \(I_{PL}(T)/I_{PL}(0)\) represents the normalized integrated PL intensity. \(C_j\) is the proportional parameter of the non-radiative recombination process \(j\), and \(E_j\) denotes thermal activation energy of non-radiative recombination process \(j\). \(m\) means the number of non-radiative recombination process. \(k_b\) represents the Boltzmann constant and \(T\) has the same physical means as mentioned above. As shown in figure 3, three different thermal quenching process has been found, we assume that this is because of the participation of three non-radiative recombination centres in thermal quenching process in the observed temperature range, and the value of \(m\) in equation (4) should be 3. In this case, the equation (4) can be written as

\[
\frac{I_{PL}(T)}{I_{PL}(0)} = \frac{1}{1 + C_1 \exp \left( \frac{-E_1}{k_BT} \right) + C_2 \exp \left( \frac{-E_2}{k_BT} \right) + C_3 \exp \left( \frac{-E_3}{k_BT} \right)}.
\]

These solid lines in figure 3 are the best fits for experimental data to equation (5), and these fitting results are listed in table 1. The PL thermal quenching phenomenon occurs when the defect’s ionization energy equals to its activation energy, according to the multiple centre mechanism of semiconductor PL quenching [16,17]. Thus, the energy levels of these non-radiative defects are 0.017 eV below the conduction band, 0.76 eV below conduction band, and 0.96 eV below the conduction band. From table 1, a same activation energy at different excitation power intensities can be found, this is because the thermally activated mechanism of nonradiative recombination centers should not depend on the excitation power [18].

![Figure 3. Arrhenius plots of temperature-dependent integrated PL intensities of electron-irradiated GaAs middle cell with excitation power densities of 2.603, 7.031, and 8.729 W/cm². The solid lines are the best fits for experimental data to equation (5).](image)

**Table 1.** Values of the parameters in equation (5) obtained from the fits to experimental data of electron-irradiated GaAs middle cell at fixed excitation power densities.

| Cell | \(I_{ex}\) (W/cm²) | \(C_1\) | \(E_1\) (eV) | \(C_2\) | \(E_2\) (eV) | \(C_3\) | \(E_3\) (eV) |
|------|-------------------|--------|--------------|--------|--------------|--------|--------------|
| GaAs | 8.729             | 2.531  | 0.0169       | 9.891×10^{11} | 0.760  | 3.132×10^{15} | 0.960  |
|      | 7.031             | 4.260  | 0.0170       | 1.062×10^{12} | 0.761  | 7.651×10^{15} | 0.962  |
|      | 2.603             | 12.500 | 0.0170       | 1.336×10^{12} | 0.762  | 6.597×10^{16} | 0.963  |
For the non-radiative recombination defect with an energy level of 0.017 eV below the conduction band, which is very near to the conduction band. In fact, this non-radiative recombination centre should be ignored because it is not a momentous role which affect the performance of solar cell, as non-radiative defects located near the middle of the gap were considered as the efficient defects deduce the solar cells’ performance [19].

For the non-radiative recombination defect with an energy level of 0.96 eV below the conduction band, it has been measured by deep level transient spectroscopy (DLTS) measurements [20], which is directly sensitive to recombination centers, and named E5 electron defect and labeled Ec-0.96 eV, and E5 electron trap has been characterized with introduction rate of 0.1 cm⁻¹ for 1.0 MeV electron irradiation and electron capture section of 1.9×10⁻¹² cm². J C Bourgoin et al [19] calculated the electron capture section of this defect by analyzing the degradation of open-circuit voltage and short-circuit current, and determined that the non-radiative defect in GaAs middle cell after radiation with 1.0 MeV electron is the E5 electron defect. And in our previous work, the room-temperature PL spectra of electron-irradiated GaAs middle cell after injection-enhanced annealing have been analyzed, and found that the E5 electron defect plays an important role for controlling the performance of GaAs middle cell [21]. Thus, the E5 (Ec-0.96 eV) electron defect determined by temperature dependent PL measurements is an efficient non-radiative defect in the electron-irradiated GaAs middle cell.

For the non-radiative recombination defect with an energy level of 0.76 eV below the conduction band, this defect also has been investigated by DLTS measurements [20] and named E4 electron trap labeled Ec-0.76 eV, and this electron trap has been characterized with introduction rate of 0.08 cm⁻¹ for 1.0 MeV electron irradiation and electron capture section of 3.1×10⁻¹⁴ cm². Due to this defect near to the middle of band gap of the GaAs middle cell, whose band gap is about 1.42 eV and because of its high electron capture section, although it is lower than that of E5 electron trap but it is higher than other defects measured by DLTS measurements from reference [20]. We conclude that E4 (Ec-0.76 eV) electron trap is also the efficient non-radiative defect in the electron-irradiated GaAs middle cell.

In summary, a competing mechanism between radiative and non-radiative recombination process was proposed to interpret the superlinear dependence between integrated PL intensities and excitation power densities. The reason of degradation of integrated PL intensity with temperature increasing is that the activated non-radiative defects capture photogenerated carriers in electron irradiated GaAs middle cell. And a three centers model of PL thermal quenching is proposed and used to fit experiment data of the integrated PL intensity with temperature increasing. Furthermore, three non-radiative defects in electron irradiated GaAs middle cell were obtained. By analyzing the energy level of the three defects and comparing the electron capture section, the two efficient non-radiative defects (E4 electron trap (Ec-0.76 eV) and E5 electron trap (Ec-0.96 eV)) controlling the performance of GaAs middle cell were determined.

4. Conclusions
The GaAs middle cell in GaInP/GaAs/Ge 3J solar cell after irradiation by 1.0 MeV electron have been analyzed by excitation power density dependent PL measurements and temperature dependent PL measurements. The relationship between integrated PL intensities and excitation power densities varies from linear to square, and the PL thermal quenching was explained quantitatively by competition mechanism between radiative and non-radiative recombination process. Furthermore, by fitting the relationship between integrated PL intensities and temperature and analyzing the energy levels of non-radiative defects, defects influenced the performance of electron irradiated GaAs middle cell mainly are E4 electron trap (Ec-0.76 eV) and E5 electron trap (Ec-0.96 eV). These results are helpful to understand the performance degradation induced by energetic particles irradiation in GaAs middle cell and provide some references for designing solar cells with outstanding irradiation resistance in space applications.

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