Radiation Physics and Chemistry 83 (2013) 42–47

Contents lists available at SciVerse ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

Optical properties of electron beam and γ-ray irradiated InGaAs/GaAs quantum well and quantum dot structures

A. Aierken [a,*], Q. Guo [b], T. Huhtio [a], M. Sopanen [a], Ch.F. He [b], Y.D. Li [b], L. Wen [b], D.Y. Ren [b]

[a] Department of Micro and Nanosciences, Aalto University, P.O. Box 13500, FI-00076 Aalto, Finland
[b] Xinjiang Technical Institute of Physics & Chemistry, Chinese Academy of Sciences, Urumqi 830011, PR China

HIGHLIGHTS

► Electron beam and γ-ray irradiation degrade the optical properties of InGaAs based quantum structures.
► 1-MeV electron beam makes more damage than γ-ray.
► QD structure is more resistant than QW and bulk structure upon to the irradiation.

ARTICLE INFO

Article history:
Received 13 July 2011
Accepted 18 September 2012
Available online 26 September 2012

Keywords:
Irradiation
Electron beam
γ-Ray
Quantum structure
Photoluminescence

ABSTRACT

The effects of electron beam and γ-ray irradiation on the optical properties of InGaAs/GaAs quantum dot (QD), quantum well (QW), and bulk structures, which were grown by metal organic vapor phase epitaxy, have been investigated. Optical properties of all the structures were degraded by both kinds of irradiation. Electron beam irradiation caused a larger reduction in the photoluminescence (PL) intensity and carrier lifetime of the samples than γ-ray irradiation. Also, red-shift of the PL peak was observed in almost all the irradiated QD and QW structures. Comparing the different structures, the QD structure showed the best radiation resistance.

1. Introduction

InGaAs/GaAs quantum well (QW) and quantum dot (QD) structures have gathered much research interest due to their tunable spectral window and subsequent huge application potential in optoelectronic devices, such as diode lasers (Chen et al., 1990; Huffaker et al., 1998), photodetectors (Tidrow et al., 1997; Xu et al., 1998), and solar cells (Yang and Yamaguchi, 2000; Aroutiounian et al., 2001). However, when these devices are operated in harsh environment like space, radiation damage by electron, proton and heavy ions can lead to operational failure or the reduction in the device lifetime. The effects induced in nanostructured materials by ion and electron irradiation have been investigated intensively (Krasheninnikov and Nordlund, 2010). The displacement damage by irradiation is usually the main concern for optoelectronic devices (Johnston, 2000). It was reported that the maximum power output of InGaAs solar cell degraded by 55% and 70% after 1 MeV electron and 3 MeV proton irradiation, respectively (Yamaguchi, 1995; Dharmarasu et al., 2001). Therefore, it becomes an inevitable task to evaluate the performance of optoelectronic devices in radiation environment before using them in any space application. Although numerous efforts (Guffarth et al., 2003; Huang et al., 2003; Erchak et al., 2001; Piva et al., 2000; Ribbat et al., 2001; Chen et al., 1996; Walters et al., 2000) have been made to improve the radiation hardness of optoelectronic materials and devices, studying the effects of radiation on basic optoelectronic quantum structures is still essential for understanding the overall performance of the devices in radiation environment.

In this paper, we investigate the optical properties of $In_{0.2}Ga_{0.8}As/GaAs$ single quantum well and $In_{0.5}Ga_{0.5}As/GaAs$ quantum dot structures irradiated by electron beam and Co-60 γ-ray radiation. The effects of various energies and flux densities of the electron beam have also been studied. Low temperature photoluminescence (PL) and time-resolved photoluminescence (TRPL) have been applied for characterization of the optical properties of the samples.

2. Experimental details

QW and QD samples were grown on semi-insulating GaAs (100) substrates in a horizontal metal organic vapor phase epitaxy

* Corresponding author. Fax: +358 9 470 25008
E-mail address: abuduwayiti.aierken@aalto.fi (A. Aierken).

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http://dx.doi.org/10.1016/j.radphyschem.2012.09.022

(MOVPE) reactor at atmospheric pressure. Trimethylindium (TMIn), trimethylgallium (TMGa), tertiarybutylarsine (TBAs), and tertiarybutylphosphine (TBP) were used as precursors for indium, gallium, arsenic, and phosphorus, respectively. The structures of the QW and the QD samples are schematically illustrated in Fig. 1(a) and (b). The $In_{0.2}Ga_{0.8}$ As QW structure was grown at 650°C and V/III ratios used for the GaAs and the InGaAs layers were 27 and 23, respectively. In the QD structure, both $In_{0.5}Ga_{0.5}$ As islands and the GaAs capping layer were grown at 550°C but a V/III ratio of 10 and 18 was used for the islands and the capping layer, respectively. Also, a 150-nm-thick GaAs/GaAs epilayer (Fig. 1(c)) and a 150-nm-thick $In_{0.5}Ga_{0.5}As$/InP lattice-matched epilayer (Fig. 1(d)) were grown for reference.

After MOVPE growth, each sample was cut into eight pieces. Seven pieces were used for different types of irradiation experiments, while one piece was kept as a reference. The different irradiation types used in this work are summarized in Table 1 and labels A–G are used to identify the irradiation types in the figures and in the text. Both electron and γ-ray irradiation were carried out at room temperature. Electron beam irradiation was carried out by an ELV-8 vertical electron accelerator. Two kinds of electron beams were chosen, 1.1 MeV electron beam with a flux density of 2.6×10^{11} cm^{-2} s^{-1} and 1.8 MeV electron beam with a flux density of 5.0×10^{10} cm^{-2} s^{-1}. A Co-60 source was applied for γ-ray irradiation with a dose rate of 1.22 Gy(Si)/s. Gray (Gy) is the unit of absorbed radiation dose of ionizing radiation, and is defined as the absorption of one joule of ionizing radiation by one kilogram of matter. For semiconductor materials, the fluence of the beam is calibrated for absorption in silicon and, therefore, the unit Gy(Si) is normally used. The electron flux density and γ-ray dose rate were measured by using Faraday cup beam integration and Fricke dosimetry system, respectively.

Low-temperature (10 K) continuous-wave PL measurements were conducted by utilizing a diode-pumped frequency-doubled Nd:YVO4 laser emitting at 532 nm for excitation. A liquid-nitrogen-cooled germanium detector and standard lock-in techniques were used to record the PL spectra. The low-temperature TRPL measurements were performed by exciting the samples with 150 fs pulses at 780 nm from a mode locked Ti:sapphire laser and by detecting the signal using a Peltier-cooled micro-channel plate multiplier and time-correlated single photon counting electronics.

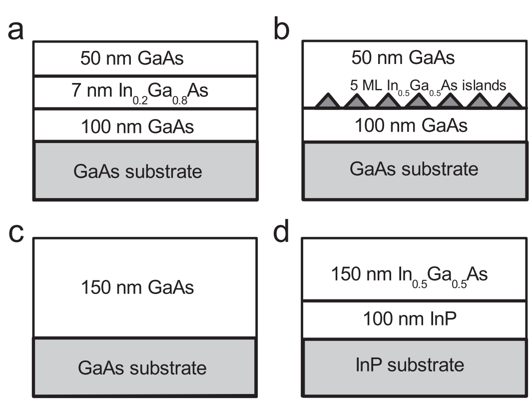

Fig. 1. Schematic diagrams of the sample structures: (a) $In_{0.2}Ga_{0.8}As$/GaAs quantum well, (b) $In_{0.5}Ga_{0.5}As$/GaAs quantum dot, (c) GaAs/GaAs epilayer, and (d) $In_{0.5}Ga_{0.5}As$/InP epilayer.

Table 1
Different irradiation types used in this article and their corresponding labels.

Irradiation type	Dose rate e: cm^{-2} s^{-1} γ: Gy(Si)/s	Total dose e: cm^{-2} γ: Gy(Si)	Label
1.1 MeV electron	2.6 × 1011	1 × 1015	A
1.1 MeV electron	2.6 × 1011	2 × 1015	B
1.8 MeV electron	5.0 × 1010	5 × 1014	C
1.8 MeV electron	5.0 × 1010	1 × 1015	D
1.8 MeV electron	5.0 × 1010	2 × 1015	E
γ-Ray	1.22	5.4 × 105	F
γ-Ray	1.22	1.1 × 106	G

3. Results and discussion

3.1. Irradiation effects on the QW structure

First, a Monte-Carlo simulation of electron beam-sample interactions was carried out on the $In_{0.2}Ga_{0.8}As$/GaAs QW structure (Fig. 1(a)) by the simulation software CASINO V2.42 (Drouin et al., 2007). Fig. 2(a) shows the simulated 1 MeV electron beam trajectories in the whole sample thickness, from which it can be seen that most of the electrons penetrate the sample and the rest are mainly stopped in the substrate bulk layer. Fig. 2(b) shows the electron trajectories in the QW active area, in which the electron beam goes straight through. Therefore, it can be concluded that 1 MeV electron beam irradiation can make displacement damage not only in the QW active layer but also in the barrier, buffer and substrate layers as well. The damage in the substrate might be even more severe.

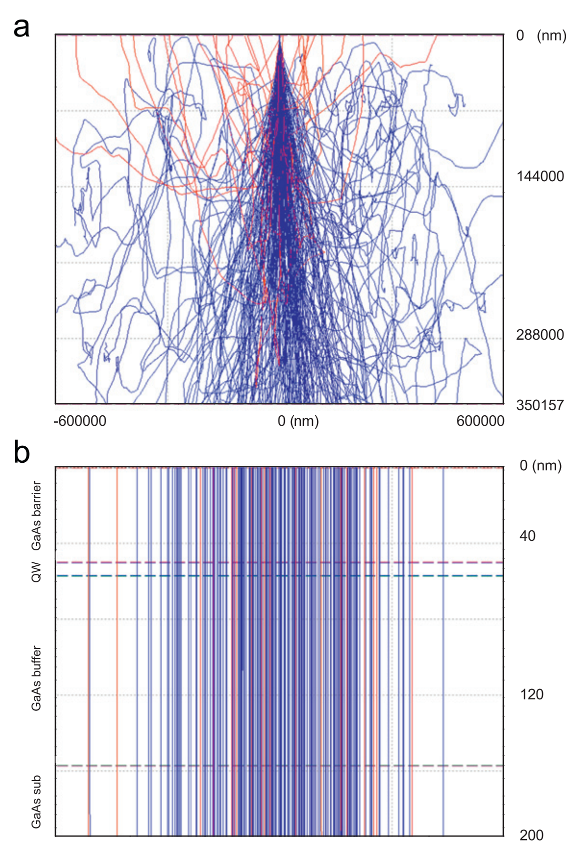

Fig. 2. Computer simulation showing the electron trajectories of 1 MeV electron beam in the $In_{0.2}Ga_{0.8}As$/GaAs QW sample (a) in the whole sample thickness, and (b) within 200 nm from the surface. Electron beam direction is from top to bottom. The blue lines represent forward electrons and the red lines represent back scattered electrons. The figures show the trajectories of 200 electrons entering in an area with a radius of 500 nm. The thickness of the substrate was set at 350 μm. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The key parameters from the PL and TRPL measurements of various irradiated QWs are summarized in Table 2. The low temperature (10 K) PL spectra of irradiated (types B, E and G) and non-irradiated QWs are shown in Fig. 3. PL peak from the non-irradiated sample was observed at 1.326 eV ($\lambda_e = 935$ nm) with a full width at half maximum (FWHM) value of 9 meV. Compared to the reference non-irradiated sample, the maximum PL intensities of the irradiated QWs decreased more than 90% when the total electron fluence reached 2×10^{15} cm^{-2}, regardless of the electron energy and flux density. Likewise, γ-ray irradiation reduced the maximum PL intensity to 50% (5.4×10^5 Gy) and 20% (1.1×10^6 Gy) of its initial value. Moreover, in all the irradiated samples except sample A, a red-shift of the PL peak position with respect to that of the non-irradiated sample was observed. The red-shift increases with increasing irradiation dose and the highest red-shift, 19 meV, was observed in G-type irradiated sample. No changes in the FWHM were observed in any of the irradiated samples.

The irradiation effects were also observed clearly in the TRPL measurements. The low temperature TRPL transients of the irradiated samples (types A, E and F) and the non-irradiated reference sample, taken at the wavelength of the maximum continuous-wave PL intensity, are shown in Fig. 4. The background signal has been subtracted from the data. The PL decay time or carrier lifetime, τ, was determined by using a first-order exponential fit

$$y(t) = A\exp(-t/\tau) + C \quad (1)$$

as indicated in the figure. From Table 2, it can be seen that in all the irradiated QW samples, the carrier lifetime decreases with increasing total dose of irradiation. The carrier lifetimes of both samples with an electron irradiation fluence of 2×10^{15} cm^{-2} (types B and E) were reduced from the initial value of 1.52 ns to about 0.35 ns. This degradation trend, which is independent of the energy and flux density of the electron beam, is the same as observed in the PL measurements. In the γ-ray irradiated samples F and G, the carrier lifetimes decreased to 0.55 ns and 0.38 ns, respectively.

Table 2
Key parameters from the PL and TRPL measurements of various irradiated QW samples. The maximum PL intensity of the non-irradiated QW has been normalized to 1.

Irradiation type	PL intensity	PL red-shift (meV)	τ (ns)
Non-irradiated	1	0	1.52
A	0.092	−8	0.44
B	0.026	2	0.36
C	0.253	12	0.45
D	0.079	10	0.48
E	0.026	17	0.35
F	0.515	8	0.55
G	0.216	19	0.38

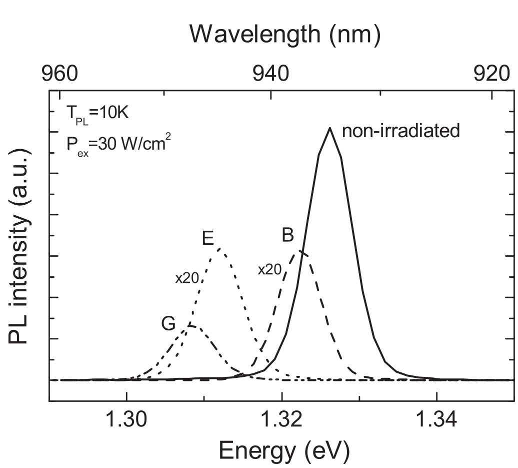

Fig. 3. Low-temperature PL spectra of $In_{0.2}Ga_{0.8}As/GaAs$ QW samples irradiated by electron beam (types B and E) and by γ-ray (type G) radiation.

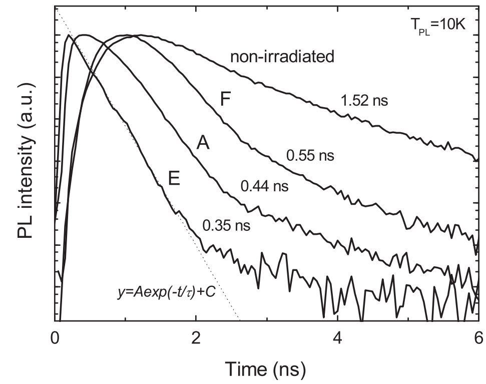

Fig. 4. Low-temperature (10 K) TRPL transients of $In_{0.2}Ga_{0.8}As/GaAs$ QW samples irradiated by electron beam (types A and E) and γ-ray (type F) irradiation.

The degradation of the optical properties of the QW is a direct consequence of the displacement damage caused by irradiation. Vacancy and interstitial defects, which act as non-radiative recombination centers (Johnston, 2000; Guffarth et al., 2003), are introduced by irradiation into the well and barrier layers. When the electrons and holes are excited by the pump beam, part of the carriers will be captured by the irradiation-induced defects before they reach the quantum well, and, consequently, the PL intensity is reduced. Introduction of this non-radiative recombination channel also decreases the carrier lifetime. This explains the PL and TRPL measurement results presented above.

Regarding the red-shift of the PL peak positions, two factors should be taken into account: the uniformity of the QW before irradiation and the structural change in the QW induced by the irradiation. According to our MOVPE growth experiments, the variation of the layer thickness and composition is less than 5%. Based on this assumption, the theoretically calculated values of QW PL peak shift caused by 5% variation of the indium concentration and the well thickness of a 7-nm-thick $In_{0.2}Ga_{0.8}As/GaAs$ are $\pm$ 7 meV and $\pm$ 5 meV, respectively. In order to check the sample uniformity, a similar QW sample was grown separately and the investigation of PL peak energy across the whole sample gave a variation of $\pm$10 meV. The observed PL peak shifts from the irradiated samples, however, increased with increasing irradiation dose. Fig. 5 shows the QW PL peak energies and their error bars for all the samples. In the non-irradiated reference sample, the standard error is $\pm$1.6 meV, which is the smallest one among all the samples. Based on these results, we assume that these red-shifts of the QW PL peak listed in Table 2 are caused by the combined effect of the sample non-uniformity and the irradiation.

QW PL peak shifts by electron, proton and γ-ray irradiation have been reported previously (Li et al., 2005; Fu et al., 1999; Borkovska et al., 2009). Variation of the indium concentration and the well width of an electron beam irradiated InGaN QW structure was studied by transmission electron microscopy (Li et al., 2005). It was concluded that the indium concentration of the InGaN QW increases and the well width decreases with increasing irradiation dose. The reason for this change is contributed to the vacancy-enhanced diffusion of indium atoms due to the displacement of nitrogen atoms. The strain relaxation (Borkovska et al., 2009; Toda et al., 1998) caused by the irradiation induced defects lowers the band edge, and, consequently, decreases the ground state energy level of the QW. Hence, a red-shift in the

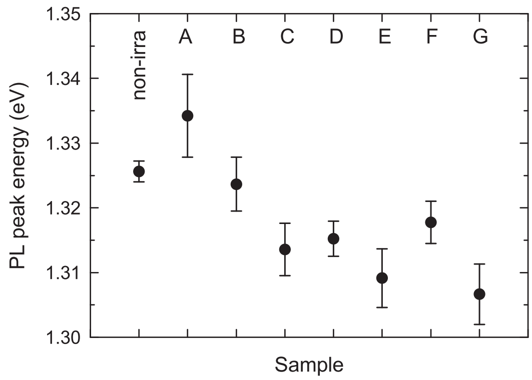

Fig. 5. Mean values and standard errors for low temperature (10 K) PL peak energies of all the $In_{0.2}Ga_{0.8}As/GaAs$ QW samples. PL was measured from five different positions in each sample.

emission wavelength is obtained. On the other hand, interdiffusion of group III elements and consequent change in the QW shape caused by the irradiation increases the ground state energy level of the QW and results in a blue-shift of the PL peak of a 1.5 MeV proton irradiated InGaAs/GaAs multi-QW structure (Fu et al., 1999).

In our study, we observed a slight blue-shift of the PL peak in type A irradiated sample. In the other irradiated samples, the combined effect caused a red-shift. However, reasons behind the peak shift upon irradiation are not so clear. A further study of more uniform QW samples with different characterization methods, such as TEM, would give more information on the PL peak shift caused by irradiation.

3.2. *Irradiation effects on the QD structure*

Fig. 6(a) shows the low temperature PL spectra of the non-irradiated and three irradiated (types A, B and G) QD samples. Three clear luminescence peaks are observed in all the samples. The peaks labeled QD0 and QD1 are related to the ground state and the first excited state of the QDs, respectively. The peak labeled WL corresponds to the InGaAs wetting layer. The normalized QD0 PL intensities and red-shifts of all the QD samples are also listed in the inset of Fig. 6. PL intensity of each energy state decreases with increasing dose in both electron beam and γ-ray irradiation.

From Figs. 6(a) and 7, it can be clearly seen that the observed reduction in the QD PL intensity by electron beam irradiation is much smaller compared to that of the QW structure. In A- and B-type irradiated samples, the QD0 PL intensity was reduced to 77% and 35% of its initial value, respectively. For the QW samples, the PL intensity decreased by to 9.2% and 2.6% with the same types of irradiation. The PL intensity dropped about 50% in F-type γ-ray irradiated QW and QD samples, whereas a slightly larger reduction was observed in the QW sample (80%) compared to the QD sample (60%) upon G-type γ-ray irradiation.

As the inset of Fig. 6(a) shows, PL peak red-shift was also observed in all the irradiated QD samples. Only in types A and B (1.1 MeV electron beam) irradiated samples, the PL shift increased (5 meV and 7 meV) with increasing irradiation dose. However, in types C, D, E (1.8 MeV electron) and F, G (γ-ray), the biggest red-shift was observed in the sample with the smallest irradiation dose and the amount of PL shift decreased with increasing irradiation dose. Also, the QD1 and WL peaks exhibited almost the same amount of red-shift and the same behavior upon irradiation as observed for the QD0 state, i.e., the whole PL spectrum is red-shifted by irradiation.

By fitting the low temperature TRPL transient curves of the QD samples, almost the same carrier lifetimes as for the non-irradiated sample were obtained from all the irradiated samples

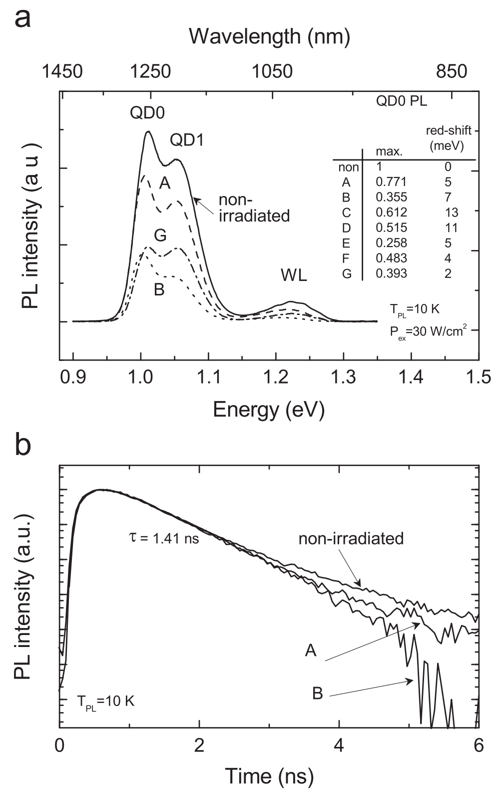

Fig. 6. (a) Low-temperature (10 K) PL spectra of $In_{0.5}Ga_{0.5}As/GaAs$ QD samples irradiated by electron beam (A and B types) and γ-ray (type G) radiation. The inset shows the relative maximum PL intensity and PL peak shift of various irradiated QDs at QD0 energy state. The maximum PL intensity of the non-irradiated QD has been normalized to 1. (b) Low-temperature TRPL transients of non-irradiated and electron beam (types A and B) irradiated QD samples.

(Fig. 6(b)). The carrier lifetime of the QD samples was unaffected by all types of irradiation and remained at 1.42 ns before and after irradiation.

From the PL and TRPL measurement results, it can be concluded that the QD structure has much better radiation resistance compared to the QW structure with respect to the irradiation types used in this study. The same carrier lifetime for irradiated and non-irradiated QD samples probably indicates that the carriers in the QD energy levels are not affected by the irradiation induced non-radiative recombination centers. The reduced PL intensity, however, shows that a reduced number of carriers reaches the QDs due to the large amount of irradiation induced recombination centers in the barrier layers. As for the PL red-shift, it also might be the combined effect of non-uniformity of the island size and the irradiation.

3.3. *Comparison of irradiation effects on bulk, QW and QD structures*

For reference, thick GaAs and InGaAs epilayers were exposed to the same irradiation types as the QW and QD structures and their post-irradiation PL measurement results were compared to those of the QW and QD structures. Although the mechanism of luminescence in thick bulk layers and QWs and QDs is different, we present the following results for comparing the overall PL degradation in different structures upon the same irradiation dosage.

The irradiation damage of electron beam and γ-ray irradiation on a GaAs bulk layer has been studied intensively (Yamaguchi and Amano, 1983; Summers et al., 1993, 1995; Xapsos et al., 1994). The dependence of displacement damage by electron beam

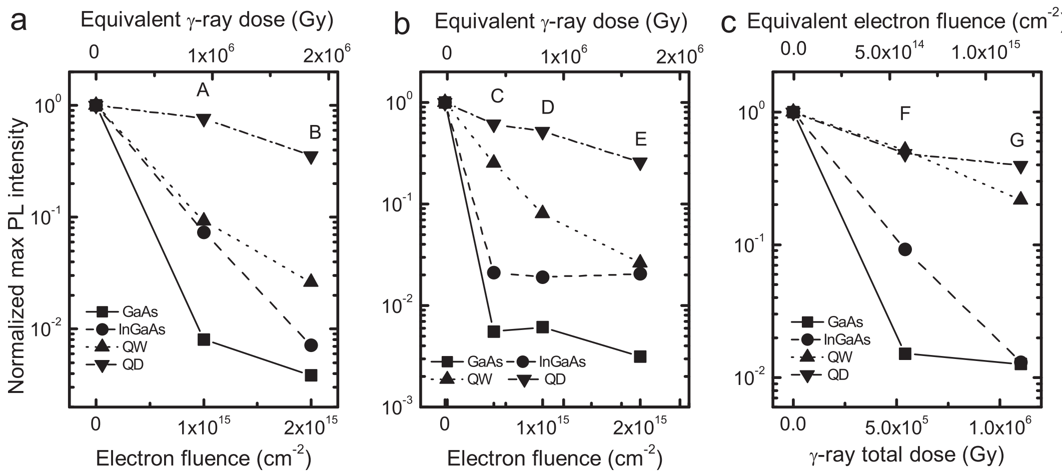

Fig. 7. Normalized PL intensities of bulk, QW and QD structures as a function of the total dose of (a) 1.1 MeV electron beam, (b) 1.8 MeV electron beam, and (c) γ-ray irradiation.

irradiation on the electron energy has been studied and the correlating damage coefficients of electron beam and γ-ray irradiation have been given by using nonionizing energy loss (NIEL) calculation (Summers et al., 1993). For GaAs, the equivalent 1 MeV electron fluence for inducing the same displacement damage by 1.0×10^6 Gy is $1.08 \times 10^{15}\,\mathrm{cm}^{-2}$.

Fig. 7 shows the PL intensities from epilayers, QWs, and QDs as functions of total dose of electron beam and γ-ray irradiation in this work. In order to compare the degradation of PL intensity, the equivalent γ-ray dose or electron fluence are also labelled in Fig. 7(a)–(c) correspondingly. It can be seen that in all the irradiated epilayers the PL intensity drops more severely than in the QW or the QD structure. After B-type electron irradiation, the maximum PL intensities of the GaAs bulk layer, the InGaAs bulk layer, the QW, and the QD structures decreased to 0.3%, 0.7%, 2.6%, and 36% of their initial values, respectively. For G-type γ-ray irradiation, the PL intensities of both the GaAs and the InGaAs epilayers dropped to 1.3% of their initial values, whereas the QW and QD PL intensities dropped to 21% and 39% of their initial values, respectively. The severe degradation of PL in the epilayers compared to the QW and the QD structure might be a direct result of the intensive displacement damage in the bulk layer induced by the irradiation. In the bulk layer, there is no confinement of the carriers and the mean diffusion length is usually in micrometer scale. On the other hand, in the QW and QD structures, carriers are confined one and three dimensions, respectively. So, the probability of recombination of the carriers by non-radiative recombination centers decreases with increasing carrier confinement dimensionality.

Additionally, in the GaAs samples, the PL peak was observed at 1.490 eV (λ_e=832 nm) and no shift of the peak position by irradiation was observed (PL spectra are not shown here). But, in the InGaAs samples, the PL peak was observed at 0.843 eV (λ_e=1471 nm) and random red/blue shift of the PL peak, in a $\pm$10 meV range, was observed in the irradiated samples.

4. Conclusion

In summary, the optical properties of electron beam and γ-ray irradiated InGaAs/GaAs QD and QW structures and epilayers have been investigated. Samples were fabricated by MOVPE and irradiated by two kinds of electron beams with different energy, 1.1 MeV and 1.8 MeV, and also by Co-60 γ-ray radiation.

All types of irradiation caused reduction in the PL intensity of the samples. The reduction increased with increasing total dose of irradiation. Electron beam irradiation, with a total fluence of $2 \times 10^{15}\,\mathrm{cm}^{-2}$, caused larger irradiation effects in all the samples compared to 1.1×10^6 Gy γ-ray irradiation. Comparing the InGaAs/GaAs QD, QW and bulk structures, QDs showed the best radiation resistance and the QW structure was better than the bulk. Carrier lifetime of the QW structure decreased with increasing irradiation total dose. But, for the QD structure, carrier lifetime was the same before and after all kinds of irradiation types. A red-shift of the PL peak position, within a scale of 0–20 meV, was observed in almost all the irradiated QW and QD structures.

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