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Ultrafast dynamics of Ni^+-irradiated and annealed GaInAs/InP multiple quantum wells

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Abstract

Carrier dynamics and optical activity of 10 MeV Ni^+-irradiated and annealed GaInAs/InP quantum wells (QWs) have been studied using a time-resolved femtosecond up-conversion method. Quantitative results are obtained for a carrier capture time and a carrier lifetime (decay time) as a function of implantation dose. The carrier capture time decreases with the dose, while it is little affected by rapid thermal annealing (RTA). The capture time is 8 ps for the as-grown QW and 2 ps for the irradiated QW at the highest ion dose of 50×10^{10} ions cm^{-2}. The carrier lifetime in the as-grown QW is long, 1.19 ns, but is only 3.7 ps after ion irradiation. RTA reduces the density of irradiation-induced defects and improves the optical activity. Other than the implantation dose, the lifetime also depends on the ion implantation energy (varied from 6 to 30 MeV at a fixed dose of 5×10^{10} ions cm^{-2}): the higher the implantation energy, the longer the carrier lifetime. This is because the nuclear deposited energy decreases in the QW region with an increase in implantation energy. Hence, for an efficient defect production low-energy implantation (<10 MeV for Ni^+) is preferred. The results obtained are compared with compressively strained GaInAs/GaAs QWs samples exposed to a similar dose and annealing conditions.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

GaInAs/InP quantum well (QW) heterostructures grown on InP substrates are used in opto-electronic devices operating at the 1.3–1.5 μm wavelengths. The QWs act as photo-active layers, for example, in ultra-fast semiconductor saturable absorbers (SESAMs) [1–3] which are non-linear mirrors capable of generating mode-locked optical pulses in a femtosecond regime when integrated with solid state lasers or fibre lasers [4]. Because the as-grown QWs are of high structural quality they exhibit long carrier lifetimes, often in the nanosecond time scale, which is far too long for ultra-short-pulse applications [1–4]. The recovery time should be reduced to sub-picosecond levels for ultrafast operations.

Ion irradiation (implantation) incorporates defects in the form of traps and recombination centres in semiconductors. These defect sites reduce the carrier decay time (lifetime) down to sub-picosecond levels. Heavy ions, such as Ni^+, create predominantly clusters of defects [5–9], which exhibit larger capture cross-sections and better thermal stability than do most of the isolated point-like defects created by light-ion irradiation [5–9].

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In the present paper, we investigate the effects of Ni^+-irradiation and rapid thermal annealing (RTA) on the carrier capture and decay times in GaInAs/InP QWs at the QW photoluminescence (PL) wavelength of 1.5 μm.

2. Experimental methods

A large number of epitaxial heterostructures were grown for this work using solid-source molecular beam epitaxy (MBE). Each sample consisted of an InP buffer layer of 100 nm thickness, deposited onto an *n*-type InP substrate cut along a 100 crystal plane, followed by the growth of seven $In_{0.576}Ga_{0.424}As$ / InP QWs (compressive strain $\approx 0.3\%$ in units of lattice mismatch $\Delta a/a$), each 6 nm in thickness and separated by 8 nm thick InP quantum barriers. So many QWs are often needed for the proper operation of a SESAM when used as an end-mirror of a fibre laser. The whole structure was capped with a 100 nm InP layer; all layers were nominally undoped.

The samples were irradiated with 10 MeV $^{59}Ni^+$ ions at doses between 1×10^{10} and 50×10^{10} ions cm^{-2}. Experiments were also done on varying ion implantation (kinetic) energies between 6 and 30 MeV to explore in detail the evolution of lifetime. Rapid thermal annealing (RTA) for all samples was performed at 610 °C for 60 s.

Time-resolved PL was measured using a femtosecond up-conversion method; the details are given elsewhere [10]. A self-mode-locked titanium sapphire laser, pumped with an argon ion laser, was employed as a source of excitation pulses (λ = 820 nm, pulse width = 50 fs, repetition rate = 90 MHz, excitation power = 20–30 mW). The time resolution of the instrument was 100 fs. The optical energy density at the sample surface was about 0.3 mJ cm^{-2}.

3. Results and discussion

3.1. Carrier lifetime and optical activity of the QWs

Photo-excitation of the sample with intense pulses creates hot carriers in the InP barriers and the GaInAs QWs. The hot carriers quickly cool to lattice temperature [11, 12], followed by their transfer from the InP barriers into the QWs either by diffusion (if the barriers are thick) or direct quantum capture. The wave function of a carrier occupying the discrete energy level of a thin QW spreads out upto a certain distance into the surrounding barrier layers. If the barrier is thinner than this distance, the wave functions in the barrier and the QW overlap each other, which means that carrier transfer from the barrier into the QW occurs quantum mechanically. If the barrier is much thicker than this critical length, the diffusion process will dominate the carrier transfer. According to [13–15], for multiple QW structures this critical length of barrier is 20 nm. Therefore, with the InP barriers of only 8 nm thickness for the samples studied in this work, the carriers are likely captured by the direct quantum capture (no diffusion), which is a very fast process compared with diffusion. After the transport of carriers to the QW via the quantum capture process, the carriers start accumulating in the QWs. Once the carrier population reaches its maximum, the depletion (decay) of the QWs takes place with time that depends upon the amount of non-radiative

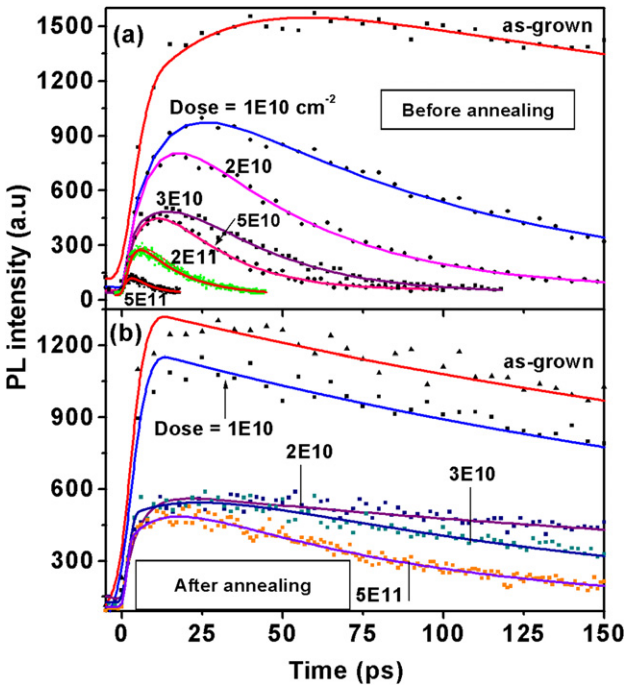

Figure 1. The time evolution of PL after the photo-excitation of GaInAs/InP QWs at different 10 MeV Ni^+ doses: (*a*) before annealing and (*b*) after annealing at 610 °C for 60 s.

traps/defects present in the sample. At room temperature, the carriers recombine non-radiatively at these defects' sites mainly via thermal activation of the carrier out of the well [16].

The experimental time-resolved PL profile can be fit to a theoretical intensity profile $I_{PL}(t) = c[\exp(t/\tau_{decay}) - \exp(t/\tau_{rise})]$, where c is a constant, τ_{rise} is a single exponential rise time and τ_{decay} is a single exponential decay time (figure 1). The rise time is represented by the slope of the leading section of $I_{PL}(t)$; the decay time is represented by a descending section of $I_{PL}(t)$.The decay of carriers follows the first order reaction (monomolecular recombination) according to the differential rate equation $dn/dt = -kn$, where k is the decay rate and n is the carrier concentration in the QW. Therefore, the decay is an exponential function $[n(t) = n_0 \exp(-kt)]$. The decay time for monomolecular recombination is independent of excitation intensity, and this is the case in our experiments.

As seen in figure 1, Ni^+-irradiation has a tremendous effect on PL dynamics. The as-grown sample has the lifetime of 1.19 ns, but ion irradiation at the dose $(\phi)1 \times 10^{10}$ cm^{-2} reduces the lifetime to 92 ps. We may compare these values with those obtained for compressively strained GaInAs/GaAs QWs, which were exposed to similar Ni^+ doses and annealing conditions [17]. The as-grown GaInAs/GaAs QWs exhibited the lifetime of 460 ps—three times shorter than 1.19 ns for the GaInAs/InP QWs. This difference may be attributed to high compressive strain present in the GaInAs/GaAs QWs, which likely contain more defects than do the weakly strained GaInAs/InP QWs.

As seen in figure 1, PL intensity drops significantly upon ion irradiation, while the carrier recombination rate increases. At the highest ϕ of 50×10^{10} ions cm^{-2}, the lifetime is 3.7 ps. For comparison, the shortest lifetime in the GaInAs/GaAs QWs is 0.62 ps at $\phi = 50 \times 10^{10}$ ions cm^{-2} [17].

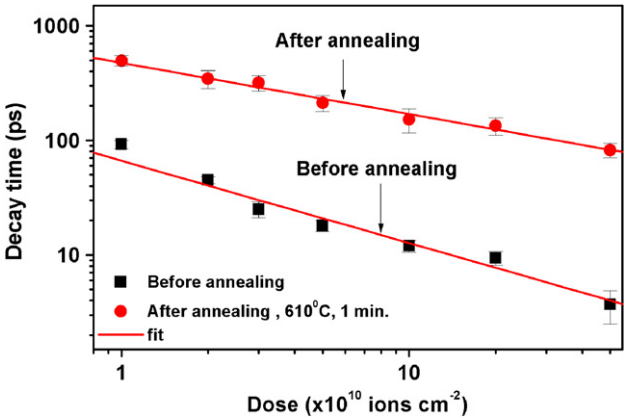

Figure 2. The decay times of irradiated and subsequently annealed GaInAs/InP QWs as a function of irradiation dose.

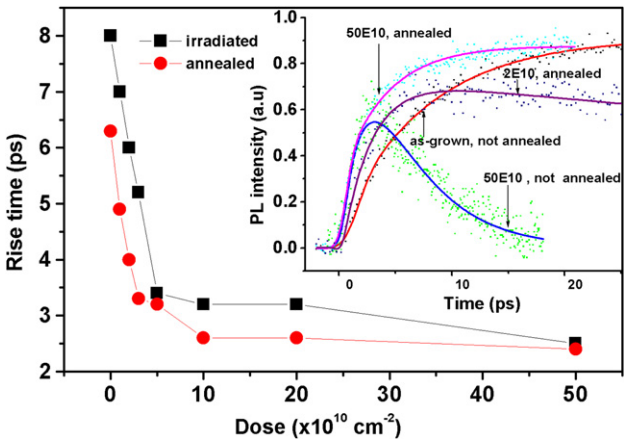

Figure 3. The PL rise time (≈ carrier capture time) for irradiated and annealed samples as a function of irradiation dose. The inset highlights the experimental PL profiles for a few selected samples.

The decay time for the irradiated and post-irradiation annealed GaInAs/InP QWs is shown in figure 2. Annealing of an irradiated sample ($\phi = 1 \times 10^{10}$ ions cm^{-2}) recovers the lifetime from 92 to 500 ps (which is almost the same (one half) as the lifetime of the as-grown QW). Annealing of the QW irradiated at $\phi = 50 \times 10^{10}$ ions cm^{-2} lengthens the lifetime from 3.7 to 82 ps. For comparison, the carrier lifetime in the as-grown GaInAs/GaAs QWs and in the QWs with $\phi = 50 \times 10^{10}$ ions cm^{-2} increases upon annealing from 460 ps to 530 ps and from 0.62 ps to 109 ps, respectively.

3.2. Carrier capture time

Figure 3 illustrates PL rise profiles created by photo-excitation and affected by irradiation/annealing. The exponential rise profiles with steep slopes characteristic of fast dynamics indicate that the carriers reach the QWs predominantly *via* quantum capture. Therefore, the quantum capture process dominates the filling of the QWs so that the carrier capture time and PL rise time become approximately the same (no diffusion, very short thermalization time). Figure 3 also reveals that τ_{rise} drops steeply at small irradiation doses ($0 < \phi < 5 \times 10^{10}$ ions cm^{-2}) and then it decreases slowly at higher doses. In fact, ion irradiation at $\phi = 50 \times 10^{10}$ ions cm^{-2} shortens the capture time from 8 ps (as-grown) to 2.4 ps (irradiated). This decrease in capture time with dose can be explained as follows. The as-grown sample contains only the native defects but as the dose is increased additional defects/vacancies are likely to be created at the QW interfaces. These defects act as trapping and recombination centres which accelerate the carrier trapping rate into the QWs. The annealed samples exhibit a similar trend, as ϕ is varied. The capture times after RTA remain approximately the same as they were before annealing, between about 2 and 8 ps—no dramatic change at all. Since the RTA has substantial effects on the decay time (figure 2) of the irradiated samples but not on the capture time (figure 3), it suggests that different kinds of defects are responsible for the observed changes in capture and decay times. We have no clear explanation for this very weak dependence of the capture time on the RTA treatment. It looks to us that some defects are responsible for the change in the capture time of the carriers while the majority of defects play no significant role in the rate of filling the QWs.

3.3. Dose dependence of carrier relaxation

Next, we calculate the power dependence of the carrier relaxation (decay) rate on ϕ. The QW decay rate (k) is the first order non-radiative recombination, which can be expressed by the Shockley–Read–Hall equation $1/\tau_{decay} = k = v_{th}\sigma N_t$. Here v_{th} is a thermal velocity, and σ and N_t are the capture cross-section and the density of non-radiative centres, respectively. At room temperature, v_{th} and σ may be assumed constant for the as-grown (σ_0) and irradiated samples (σ_d). N_t can be written as a sum of the native defect density N_0 and the irradiation-induced defect density N_d. Then k can be given in terms of the relaxation rate constant k_0 for the as-grown sample and the relaxation rate constant k_d for the irradiated samples, i.e. ($k = k_0 + k_d$). If N_d varies with ϕ as $N_d = \gamma\phi^\alpha$. then k/k_0 can be written as a function of ϕ [17]:

$$\frac{k}{k_0} - 1 = \frac{v_{th}\sigma_d\gamma}{k_0}\phi^\alpha = C\phi^\alpha.$$

When $(k/k_0 - 1)$ is plotted against ϕ (logarithmic scale), the slopes (α) of the straight lines corresponding to the irradiated samples and irradiated-annealed samples establish the dependence of decay rate on ϕ. As seen in figure 4, fairly straight lines are obtained for irradiated alone and irradiated-annealed samples in double logarithmic scale. Since k/k_0 varies between 13 and 320, therefore $(k/k_0 - 1) \cong k/k_0$. According to figure 4, α is 0.78 ± 0.06(or $k = 1/\tau_{decay} \propto \phi^0.8$) for the irradiated samples and 0.57 ± 0.04 (or $k = 1/\tau_{decay} \propto \phi^{0.8}$) for the annealed samples. The slope of 0.8 (≈ 1) for the irradiated samples signifies that the lifetime is inversely proportional to ϕ. The GaInAs/GaAs QWs exhibit the corresponding values of 1.2 and 0.35 [17]. A lower value of α (0.8) for Ni^+-irradiated GaInAs/InP samples signifies that the QWs are depleted rather slowly in these samples as compared with the compressively strained GaInAs/GaAs QWs ($\alpha = 1.2$). On the other hand, compared with the value α (0.35) for annealed GaInAs/GaAs samples, a higher value of α (0.6) for irradiated-annealed GaInAs/InP samples suggests a lower thermal stability of GaInAs/InP samples after the annealing. Thus, the comparison shows that ion irradiation and subsequent annealing modify the carrier lifetimes more drastically for the GaInAs/GaAs QWs than for the GaInAs/InP QWs.

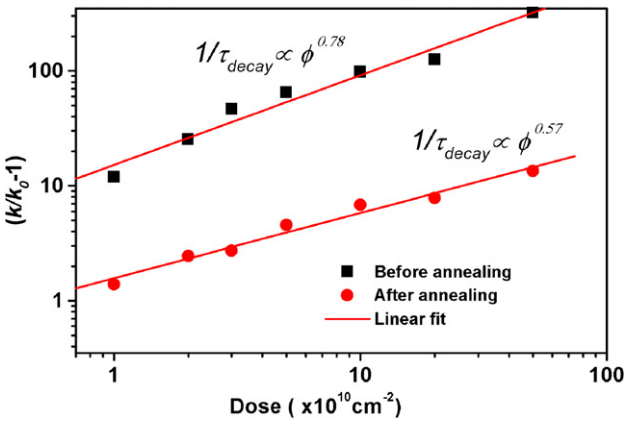

Figure 4. The dependence of the relative relaxation rate $(k/k_0 - 1)$ on the ion dose (ϕ). The solid lines are linear fit. The slope (α) for irradiated samples is 0.78 ± 0.06 and for the irradiated-annealed sample is 0.57 ± 0.04. Since $k/k_0 >> 1$ in the entire dose regime, therefore $k = 1/\tau_{\text{decay}} \propto \phi^{\alpha}$.

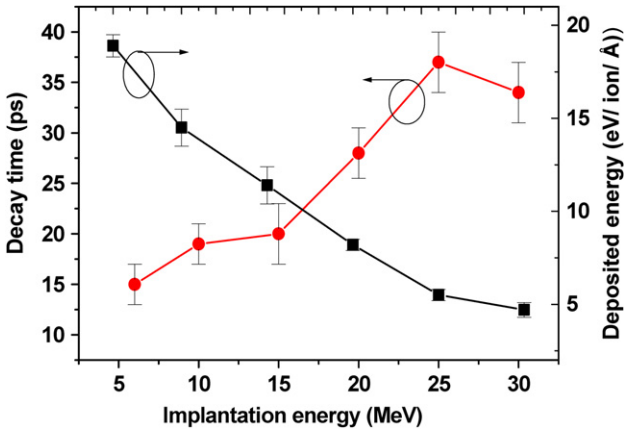

Figure 5. The decay time (●) and deposited energy (■) in the active region of GaInAs/InP QWs structure irradiated at 5×10^{10} ions cm^{-2} Ni^{+} dose, as a function of implantation energy.

3.4. Implantation energy and nuclear deposited energy

The decay time also depends on ion implantation energy. Figure 5 illustrates the decay times for the QWs irradiated at $\phi = 5 \times 10^{10}$ ions cm^{-2} for the implantation energies between 6 and 30 MeV. There is a systematic increase in τ_{decay} from 15 to 37 ps in the energy range from 6 to 25 MeV. This behaviour can be understood in terms of the nuclear deposited energy (in units of eV/ion/Å) in the QW region, since the damage produced in InP and GaAs is dominated by nuclear collision processes for ions in this range [18] (track formation becomes significant only at higher electronic deposition [19]). The nuclear deposited energy in the QW region was calculated using the SRIM2003 computer code [20] by calculating the energy transferred to recoil by the incoming ion in the active region in the depth range 100 to 200 nm. The result was averaged over the InP and InGaAs multiplayer parts (below the top 100 nm InP layer) and is given in figure 5 (right-hand Y-axis). The results show that the nuclear deposited energy in the QW region decreases with increasing implantation energy, and this trend is almost exactly opposite to the change in lifetime. Less damage is correlated with longer lifetimes. Accordingly, in order to effectively produce defects, low implantation energies (less than 10 MeV for Ni^{+}) are preferred.

4. Conclusion

Using the time-resolved up-conversion method, the dynamical and optical properties of Ni^{+}-irradiated and rapid-thermal-annealed samples consisting of multiple GaInAs/InP QWs were investigated. The carrier lifetime was shown to decrease rapidly with an increase in irradiation dose. The longest lifetime of 1.19 ns was for an as-grown sample, while the shortest lifetime was 3.7 ps for a sample irradiated at a 50×10^{10} ions cm^{-2} dose. The carrier capture process was interpreted primarily as due to carrier quantum capture, and it was dependent on the implantation dose but relatively little affected by annealing. The capture time was always between 2 and 8 ps. The nuclear deposited energy in the QW region had a maximum value at the smallest implantation energy, suggesting that low Ni^{+} implantation energies, less than 10 MeV, are preferred for efficient defect production.

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