Ni\textsuperscript{+}-irradiated InGaAs/GaAs quantum wells: picosecond carrier dynamics

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Abstract. Room-temperature carrier dynamics as functions of heavy-ion implantation and subsequent thermal annealing were investigated for technologically important InGaAs/GaAs quantum wells (QWs) by means of a time-resolved up-conversion method. Sub-picosecond lifetimes were achieved at 10 MeV Ni\textsuperscript{+} doses of (20–50) \times 10\textsuperscript{10} ions cm\textsuperscript{-2}. The decay rates reached a maximum at the highest irradiation dose, yielding the shortest lifetime of the confined QW states of 600 fs. A simple theoretical model is proposed for the photodynamics of the carriers. The relaxation rate depended on the irradiation dose according to a power law of 1.2, while the irradiated and subsequently annealed samples exhibited a power law of 0.35. The results are qualitatively interpreted.
1. Introduction

Among various semiconductor quantum well (QW) heterostructures for high-speed optoelectronic applications, compressively strained InGaAs/GaAs QWs are the key building blocks of devices [1]. In particular, performance characteristics of InGaAs QWs for optical switches and semiconductor saturable absorber mirrors depend on QW recovery time and optical nonlinearity properties [2]–[4]. The main techniques used to achieve short carrier lifetimes include low-temperature (LT) epitaxial growth [5] or LT-growth in conjunction with beryllium doping [6, 7] or proton [8] and heavy-ion irradiation [9, 10]. The LT-growth of GaAs, and to a lesser extent that of InGaAs, is a very efficient way of shortening the carrier lifetimes via incorporation of antisite arsenic (As) point defects acting as carrier traps. Beryllium doping of LT-InGaAs further improves the recovery time and nonlinear properties [6, 7]. A recent work by Baker et al [5] shows that LT-grown InGaAs/GaAs is a promising heterostructure for ultrafast photoconductive devices, namely, sub-500 fs carrier lifetimes and high-electrical resistivity can be realized even without beryllium doping. On the downside, LT-growth parameters are not well-controlled. Light-ion (proton) irradiation also reduces the recovery time down to a sub-picosecond level [8]. Proton irradiation seems to be as effective as heavy-ion irradiation [11], but it preferentially creates point defects, but not many defect complexes. Since the point defects usually have low-activation energies, they may be thermally unstable at room temperature [12]. Irradiation with heavy ions offers the advantage of higher implantation energies, which create more stable defects [12]. Another advantage of heavy-ion bombardment at high kinetic energies, in particular, is the creation of clusters of point defects yielding a larger number of carrier states that open new recombination channels for ultrafast operations [12]. Heavy-ion implantation has proved to be a viable alternative to LT-growth, and is closely controlled. A significant advantage of ion irradiation over LT-growth is the fact that once an epi-wafer is grown, the physical properties of QWs can be tailored by a choice of the energy and dose of implantation and annealing conditions.

We have investigated carrier decay and rise rates in heavy-ion (Ni+) implanted and post-implantation-annealed InGaAs/GaAs QWs. A photoluminescence (PL) up-conversion method was used to study the dynamics of photo-excited carriers with a sub-picosecond resolution to provide accurate information about the carrier relaxation and capture processes. By correlating irradiation-induced defects with the relaxation rates, quantitative results of dose-dependent decay rates were obtained.

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2. Experimental procedure

The samples were grown by molecular beam epitaxy. They consisted of five compressively strained QWs made of 6 nm thick In$_{0.29}$Ga$_{0.71}$As/17 nm GaAs heterostructures, which were deposited onto a 200 nm GaAs buffer layer on a GaAs (0 0 1) substrate and capped with a thin GaAs layer. PL peak emission was at $\lambda \approx 1090$ nm. Two sets of samples were studied: one set was irradiated, the other subsequently heat-treated by rapid thermal annealing (RTA) for 60 s at 610°C. Both series of samples were irradiated with 10 MeV $^{59}$Ni$^+$ ions at doses ranging from $1 \times 10^{10}$ to $50 \times 10^{10}$ ions cm$^{-2}$.

Time-resolved photoluminescence (TRPL) was measured using the fs up-conversion method, as described elsewhere [13]. A fs self-mode-locked titanium sapphire laser, pumped with an argon ion laser, was employed as a source of excitation pulses. The excitation wavelength was 840 nm, the pulse width 50 fs, the repetition rate 90 MHz, the time resolution 100 fs (FWHM), and the excitation power 20–30 mW. The energy density of excitation was roughly 0.3 mJ cm$^{-2}$.

3. Results and discussion

3.1. Photoluminescence dynamics

One of the subjects of primary interest for this study is the rate of carrier relaxation at confined QW states. To this end, figure 1 shows the PL decays of a few selected samples. PL rises in a few ps after excitation and then decays at the rate that depends on irradiation dose and annealing conditions. The as-grown sample shows the longest emission lifetime. Irradiation of the sample by Ni ions incorporates additional defects, thereby increasing the relaxation rate gradually via recombination of the carriers at these trapping centres. Annealing recovers the relaxation rate to considerable extent but not completely. At this point it is important to notice that for the entire...
Figure 2. TRPL from the samples exposed to different Ni\(^+\) ion doses (indicated in \(10^{10}\) ions cm\(^{-2}\)) in a shorter timescale. The dots represent experimental data; the solid lines are bi-exponential fits. The initial slopes of PL intensity curves remain almost independent of irradiation doses.

dose regime studied here, the PL decays are fairly straight lines in semi-logarithmic plot, which indicates that the carrier relaxation in these samples is well-described by the first-order reaction. Figure 2 presents a shorter timescale where the formation of the PL can be seen clearly. In a few ps timescale, PL intensity rises from zero to its maximum value indicating a rise in the population of the confined QW states. Since the samples were excited by photons with energy above the confined energy levels of the QW, the photo-excited carriers were generated in the continuum of their corresponding bands (with a population of about \(10^{19}\) carriers cm\(^{-2}\)). These photogenerated carriers thermalize in a very short time of tens of fs to the bottom of their respective bands. Upon thermalization, they are still located in the continuum states so that no emission due to confined QW states can be seen at this point. After the thermalization process (which is very short and thus can be neglected), two processes can contribute to the formation (rise) of the PL: (i) direct carrier trapping and (ii) the diffusion of carrier to the location of the QW followed by subsequent carrier trapping by the QW. The observed exponential nature of the PL formation in figure 2 shows that the time-limiting step in filling the confined states is not diffusion, but trapping (capture). In other words, the carriers involved in the PL emission are those generated relatively near the QW, leaving little room for diffusion. Since the hole capture is a much faster process, mainly due to the large hole effective mass, the limiting factor of the observed PL dynamics throughout the present study is mainly related to the electron dynamics [14].

3.2. Photoluminescence analysis framework

In view of the data analysis, especially at higher doses, we consider a simple theoretical model for the photodynamics of the carrier. The results obtained are interpreted by fitting the experimental data to this kinetic model.
From the theoretical consideration it is essential to account for the two processes: carrier capture by the QW from continuum levels \((N_{CB} \rightarrow N_{QW})\) and carrier recombination at QW \((N_{QW} \rightarrow \cdots)\). The relaxation dynamic can be presented by the following scheme:

\[
N_{CB} \xrightarrow{k_c} N_{QW} \xrightarrow{k_q} \ldots,
\]

where \(N_{CB}\) is the population of the continuum states (conduction band), \(N_{QW}\) is the population of the confined QW states, \(k_c\) is the rate of carrier capture by the QW (confined states), and \(k_q\) is the relaxation rate of the carriers at confined QW states. One can add an additional term of carrier trapping and relaxation rate at the continuum levels, which avoids the QW; however this will decrease only the efficiency of the PL but will not affect the dynamics of the QW PL and will not be considered here for the sake of simplicity.

Since, we can neglect the diffusion of the carriers during the capture process, the relaxation of the continuum states is given by a simple first-order equation

\[
\frac{dN_{CB}}{dt} = -k_c N_{CB},
\]

which has a trivial solution

\[
N_{CB}(t) = N_0 e^{-k_c t}.
\]

Relaxation of the carriers at confined QW states is mainly due to carrier trapping and this is also described by a first-order equation

\[
\frac{dN_{QW}}{dt} = k_c N_{CB} - k_q N_{QW}.
\]

Solution of the above equation yields

\[
N_{QW}(t) = \frac{A k_c}{k_q - k_c} (e^{-k_{q} t} - e^{-k_{c} t}),
\]

where \(A\) is a constant determined by the initial population at continuum level. The PL intensity is proportional to the population of confined QW states; therefore the PL emission time profile should follow equation (5), i.e.

\[
I(t) = \frac{c k_c}{k_q - k_c} (e^{-k_{q} t} - e^{-k_{c} t}),
\]

where \(c\) is a constant. One can notice that for an as-grown sample \(k_c \gg k_q\), thus \(k_q - k_c\) is negative and the sum of exponents in the brackets is also negative (at \(t > 0\)), giving together positive \(I(t)\) at \(t > 0\), and zero intensity at \(t = 0\). The irradiation by Ni⁺ ions increases the relaxation rate \(k_q\) of the carriers at QW, but it should have little effect on the carrier capture rate, \(k_c\), at least at reasonably low doses. Therefore, the initial slope of the PL rise should be insensitive to Ni⁺ irradiation, since \(dI(t)/dt = ck_c\). This is indeed the case for PL curves shown in figure 2: the signal decay rates and amplitudes are changing significantly as a function of the dose, while the slope of initial PL rise...
Figure 3. Dose dependence of the formation ($k_1$) and relaxation rate ($k_2$), as obtained from the bi-exponential fit equation (7) of the TRPL data. The lines give extrapolations of the actual QW rise (---) and decay (----) rate trends.

remains virtually the same. Thus the capture rate, $k_c$, is not affected by the irradiation as strongly as $k_q$.

From the viewpoint of the analysis of the measured PL data, equation (6) is a sum of two exponent terms:

$$I(t) = a_1 e^{-k_1t} + a_2 e^{-k_2t},$$

where $a_1 = -a_2$, and $a_1 > 0$ if $k_1 < k_2$ and $a_1 < 0$ if $k_1 > k_2$, to keep $I(t) > 0$ at $t > 0$. Therefore, a standard bi-exponent fitting was used to analyse the data. For all the goodness-of-fit data, $\chi^2$ (weighted mean square deviation) was better than 1.2 indicating that equation (7) models the decay reasonably well. Another indication of the applicability of the model was a good match between the pre-exponential factors, which were $a_1 = -a_2$ as expected with 5% accuracy.

Pairs of rates obtained from the bi-exponential fit equation (7) for the series of samples subjected to different Ni+ irradiation doses are presented in figure 3, where the fitted rates are ordered so that $k_1 > k_2$ (thus $a_1 < 0$). Phenomenologically, this ordering means that $k_1$ corresponds to the rise of the PL and $k_2$ to the decay, respectively. For as-grown samples and samples at low irradiation doses, $k_c > k_q$, thus $k_c = k_1$ and $k_q = k_2$. However, at higher irradiation doses, the rate of the QW carrier’s relaxation increases and at the point B, in figure 3, the relaxation rate approaches the capture rate, $k_q \sim k_c$. Further increase in the dose should lead to $k_q > k_c$, which gives another assignment $k_q = k_1$ and $k_c = k_2$. (Already at point B we can expect $k_q > k_c$.)

To verify the latter statement we can examine the maximum amplitude of the PL, which also depends on the relation between $k_c$ and $k_q$. From equation (6), $dI(t)/dt = 0$, one can calculate the delay time for the theoretical maximum PL intensity

$$t_{max} = \frac{\ln(k_c/k_q)}{k_c - k_q}. \quad (8)$$

4 Strictly speaking, at $k_c = k_q$ the solution of equation (4) is no longer exponential. It is $N_{QW} \sim t e^{-k_1t}$.

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Figure 4. Dependence of normalized relative PL maximum intensity on $k_q/k_c$. The solid curve represents a theoretical intensity maximum $I(t = t_{\text{max}})$ obtained from equation (6). The filled squares are the experimental data points for doses $(0–10) \times 10^{10}$ ions m$^{-2}$, fitted by equation (7) on the assumption that $k_c > k_q$ (i.e., $k_c = k_1$ and $k_q = k_2$). The experimental data points obtained at the last two highest doses, $20 \times 10^{10}$ and $50 \times 10^{10}$ ions cm$^{-2}$ are plotted on the theoretical curve considering the two possibilities: (a) $k_c > k_q$ (i.e., $k_c = k_1$ and $k_q = k_2$), shown as empty squares and (b) $k_c < k_q$ (i.e., $k_c = k_2$ and $k_q = k_1$), shown as empty circles. The case $k_c < k_q$ fits better to the theoretical curve at the last two highest doses than the other one.

This value can be substituted in equation (6) to calculate the maximum PL amplitude. The dependence of the relative maximum PL intensity on the rate $k_q$ as obtained from equations (6) and (8), and from the experimental data are presented in figure 4. Clearly, the assignment $k_q > k_c$ for the last two points fits better to intensity dependence. Based on this assignment, the straight lines in figure 2 show the trends of $k_c$ and $k_q$ dependence on the irradiation dose, which are quite linear in double logarithmic scale.

With increasing dose, the experimental data systematically deviate from the theoretical curve (figure 4). The deviation is due to the fact that recombination of carriers through defects at high doses is much faster than at low doses and starts competing with the trapping rate, which is not included in the relaxation model (equation (1)) and, therefore, not accounted for in the theoretical curve.

The relaxation time (460 ps) for an as-grown QW only slightly increases (up to 530 ps) upon annealing, which is an indication of good crystallographic structure of the samples grown by MBE, but for the QW irradiated with $50 \times 10^{10}$ Ni$^+$ ions cm$^{-2}$ the relaxation time becomes extremely short, 600 fs. In this case, annealing has a tremendous effect on relaxation time, which is now 109 ps. Hence annealing recovers a large number of defects in a selective way, leaving some defects unaffected.

Figure 5 shows that the carrier capture time is not much influenced by ion irradiation. For the annealed samples, we have $k_c > k_q$ at all doses; i.e., $k_1 = k_c$ and $k_2 = k_q$, which is opposite to the case $k_c < k_q$ at high irradiation dose before annealing (figure 4). Numerical values for
the relaxation and capture rates for irradiated and irradiated-annealed samples are compiled in table 1.

### 3.3. Relaxation rate as a function of ion dose

Judging from the dependence of the decay time on ion dose one can estimate the trapping rates of carriers in the QW. Carriers recombine radiatively or non-radiatively. Non-radiative recombination occurs via carrier capture at defects and dominates the recombination mechanism at room temperature [15]. Because the PL peak intensity depends linearly on the excitation power, the carrier relaxation rate \( (k_q) \) extracted from our up-conversion PL measurements is the first-order non-radiative recombination, which is expressed by the Shockley–Read–Hall equation

\[
k_q = v_{th} \sigma N_t.
\]  

### Table 1. PL formation \((k_1)\) and relaxation \((k_2)\) rates, for irradiated and post-annealed samples, and the carrier lifetime \((\tau)\) in the QW. See the text on correlation between \(\tau\) and \(k_1, k_2\) at doses \(20 \times 10^{10}\) and \(50 \times 10^{10}\) ions cm\(^{-2}\), respectively.

| Dose \((\times 10^{10}\) ions cm\(^{-2}\)) | Irradiated Irradiated Irradiated–annealed Irradiated–annealed Irradiated QW lifetimes |
|---|---|---|---|---|---|
|  | \(k_1\) (ns\(^{-1}\)) | \(k_2\) (ns\(^{-1}\)) | \(k_1\) (ns\(^{-1}\)) | \(k_2\) (ns\(^{-1}\)) | \(\tau\) (ps) |
| 0 | 175 | 2.2 | 212 | 1.9 | 460±20 |
| 1 | 217 | 16 | 238 | 3.7 | 62±3 |
| 2 | 200 | 23 | 227 | 4.4 | 43±2 |
| 5 | 330 | 90 | 303 | 5.3 | 11±1 |
| 10 | 400 | 270 | 222 | 5.9 | 3.7±0.3 |
| 20 | 500 | 416 | 250 | 6.9 | 2.0±0.2 |
| 50 | 1600 | 500 | 227 | 9.2 | 0.6±0.1 |

### Figure 5. Dose dependence of \(k_1\) and \(k_2\) for the irradiated and subsequently annealed samples. The solid lines are linear approximations.
Here $v_{th}$ is a thermal velocity, and $\sigma$ and $N_i$ are the capture cross-section and the density of non-radiative centres, respectively. $N_i$ can be written as a sum of the native defect density $N_0$ and the irradiation-induced density $N_d$. Correspondingly, $k_q$ may be given in terms of the relaxation rate constant $k_0$ of an as-grown (native) sample, and the rate constant $k_d$ due to relaxation via the Ni$^{+}$ irradiation induced defects

$$ k_q = k_0 + k_d = v_{th}(\sigma_0 N_0 + \sigma_d N_d), \quad (10) $$

where $k_0 = v_{th}\sigma_0 N_0$. $N_d$ varies with implantation dose. If the relaxation rate dependence on the irradiation dose is plotted in a double logarithmic scale, a linear relationship is obtained for both the irradiated and subsequently annealed samples; i.e., $N_d$ is a power function of dose ($\phi$)

$$ N_d = \gamma \phi^\alpha. \quad (11) $$

Inserting equation (11) into equation (10) and rearranging the terms one obtains

$$ \frac{k_q}{k_0} - 1 = \frac{v_{th}\sigma_0 \gamma}{k_0} \phi^\alpha = C \phi^\alpha. \quad (12) $$

For the irradiated samples (no annealing), equation (12) gives $\alpha = 1.20 \pm 0.06$; i.e., $k \propto \phi^{1.2}$ (figure 6), which is close to the power of 1, giving the well-known result that the carrier lifetime in the QW is inversely proportional to the dose, while the density of defects $N_d$ varies linearly with the dose. Interestingly, the annealed samples exhibit a much smaller value: $\alpha = 0.35 \pm 0.02$ (figure 6), or $k \propto \phi^{0.35}$. Such a weak dependence of the decay rate on ion dose upon annealing signifies an occurrence of very different defect kinetics for the annealed samples and for the irradiated ones. According to [11, 12], and similar studies in [10, 16, 17], heavy-ion irradiation of InGaAs tends to create clusters of point defects. These findings suggest that bombarding the

Figure 6. Dependence of relative relaxation rate ($k_q/k_0 - 1$) on ion dose in a double logarithmic scale. The solid lines are linear fits.
samples with 10 MeV Ni⁺ produces plenty of clusters of (point) defects. These clusters (and some other defects) are removed by annealing and isolated point defects left behind. Qualitatively, the small value of \( \alpha (0.35) \) could be attributable to remaining point defects in the annealed QWs. This is also supported by observations in [18], according to which a change in electrical properties of ion-irradiated InGaAs after annealing is due to remaining point defects rather than remaining complexes. Further work needs to be carried out to verify the defect kinetics in these samples.

4. Conclusions

We have investigated effects of 10 MeV Ni⁺ ion irradiation and subsequent annealing on carrier lifetimes in bi-axially compressively strained InGaAs/GaAs QWs in terms of carrier relaxation and formation rates, as measured by TRPL. It is observed that the capacity of the QW to catch thermalized carriers from the continuum of the conduction band is not much affected by ion irradiation. Experimental data are analysed on the basis of a simple theoretical model for the photodynamics of the carriers. The irradiated samples exhibited about equal relaxation and formation rates in the QW, about 0.5 ps⁻¹, at the dose of \( 20 \times 10^{10} \) ions cm⁻². The highest dose, \( 50 \times 10^{10} \) ions cm⁻², is found to reduce the carrier lifetime by a factor of 10³, from 460 ps for the as-grown sample to 600 fs for the irradiated sample, without any sign of lifetime saturation, while subsequent annealing increases the lifetime by two orders of magnitude, due to a sharp reduction in defect density. The relaxation rate for the irradiated samples depends on the ion dose according to the power law of 1.2±0.06, while that for the annealed samples follows the power law of 0.35±0.02. So, a weak dose dependence of the annealed QW indicates that heat treatment carried out in this work probably removes preferentially irradiation-induced defect clusters but leaves behind a number of isolated point defects.

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