A comparison of impurity-free and ion-implantation-induced intermixing of InGaAs/InP quantum wells

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(Received 20 March 2000; accepted for publication 9 August 2000)

We have compared the time integrated photoluminescence (PL) and the time resolved PL of several lattice matched InGaAs/InP quantum wells intermixed either by ion implantation or an impurity-free method. We have found that the carrier capture rates into quantum wells and carrier relaxation from the wells depend on the type of intermixing used. Our results indicate that the carrier lifetimes are significantly longer in samples intermixed by the impurity-free methods, while the carrier collection efficiency of the quantum wells is more efficient in samples intermixed by ion implantation.

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INTRODUCTION

Quantum-well intermixing (QWI) is a well known technique which provides a simple yet elegant method for monolithically integrating optoelectronic devices, or to enhance the performance of other discrete devices, such as diode lasers, infrared detectors, etc.¹ The technique is based on the modification of the quantum well shape by partially interdiffusing the constituent atoms at the quantum well-barrier interface. A range of techniques have been described in the literature which can be used to generate intermixing, including impurity induced interdiffusion,² impurity-free disordering using various dielectric cap layers,³ ion implantation,⁴ laser assisted disordering,⁵ focused ion-beam induced intermixing,⁶ etc. These various methods can all be arranged into two broad groups: those that create defects/impurities at the quantum well region, such as methods relying on ion implantation or impurity diffusion, and those that are impurity free. Each technique has its advantages and drawbacks, and for a given application, individual research groups tend to concentrate on one or another particular technique.

Typically, quantum-well intermixing is quantified by measuring the shift of the photoluminescence (PL) peak from its original (unintermixed) position. The magnitude of this shift is a measure of the change in the quantum well band-gap energy caused by intermixing. In this publication, however, we show that this single parameter, the energy shift, does not and cannot completely describe the intermixing process. By comparing an impurity-free and implantation-induced intermixing techniques we show that while the PL energy shifts achieved by both techniques were nearly identical in the materials investigated in this study, the carrier dynamics in the two types of samples were considerably different. We found that the carrier capture rates into the quantum wells and the carrier lifetimes in the wells were all significantly influenced by the type of intermixing used, even though the time integrated PL behavior was similar.

EXPERIMENT

Each In_{0.53}Ga_{0.47}As/InP sample contained three lattice matched single QWs, grown by metalorganic vapor phase epitaxy on InP substrate, and consisted of a 5 nm (QW3), a 3.5 nm (QW2), and a 2.5 nm (QW1) wide quantum wells sandwiched between 25 nm thick InP barriers followed by a 25 nm thick InP capping layer. The narrowest quantum well, QW1, was closest to the surface of the sample. Two types of intermixing were used in these experiments: samples were either intermixed using proton implantation (H⁺ ion energy 20 keV, ion dose $5 \times 10^{15} \text{ cm}^{-2}$), or intermixing was achieved using the impurity-free vacancy disordering method based on a 65 nm thick SiO_x cap layer deposited at room temperature using a plasma enhanced chemical vapor deposition system. Then half of each sample was covered during the intermixing process to keep it as an unintermixed reference sample. All samples were annealed in a rapid thermal annealer at 800 °C for 30 s.

The time integrated (cw) PL measurements were acquired using the second harmonic of the diode pumped Nd-YAG laser emitting at 532 nm. The emitted light was collected in the backscattering geometry and was analyzed by a 0.25 m monochromator and detected by a liquid nitrogen cooled Ge detector followed by a lock-in amplifier. The time resolved PL measurements were carried out using a femto-second self-mode-locked Ti:sapphire laser ($\lambda = 780 \text{ nm}$, 85 MHz, 80 fs) and a 3 mm thick β barium borate (BBO) crystal. The temporal resolution of the complete setup was approximately 200 fs. The sample temperature was $T = 15 \text{ K}$. Further details about the time resolved PL experiments can be found in a previous publication.⁷

RESULTS

The cw PL spectra of the various samples are shown in Fig. 1 where we display the PL spectra of the as-grown and that of the intermixed samples. As can be seen from the figure, the emission lines have shifted to higher energies as a result of the intermixing process. However, the intermixing

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FIG. 1. Photoluminescence spectra of the as-grown and intermixed InGaAs/InP quantum well samples measured at $T = 15$ K.

is not equal for all the QWs. Impurity-free intermixing, which relies on the surface as a source of vacancies, results in larger shifts for the QWs close to the surface (QW1) than for the deeper quantum wells (QW3). Ion implantation, on the other hand, results in a flat distribution of defects for the given ion energy (close to the QWs), resulting in an approximately equal energy shift. For simplicity, we shall initially concentrate on only the quantum well closest to the sample surface (QW1) because, as can be seen in Table I, the shift of the PL peak in the case of QW1 is approximately 100 meV for both types of intermixing techniques. Not only are the PL shifts similar, but the integrated PL intensities are also comparable to the as-grown (reference) sample. From these results we could conclude that impurity-free and defect-induced intermixing produces materials with similar properties. The time resolved PL experiments, on the other hand, convey a different picture. The temporal evolution of the PL of the two types of samples was significantly different, as is shown in Fig. 2. The PL rise and decay times are quite dissimilar for the two intermixed samples, with the decay times differing by nearly an order of magnitude, and the rise times deviating by approximately a factor of two, as is given in Table II. The rise and decay times were determined using the following function to fit the data:

TABLE I. PL peak energy shift as a result of intermixing.

	QW1 energy shift (meV)	QW2 energy shift (meV)	QW3 energy shift (meV)
Ion implanted	105	110	110
Impurity free	91	43	37

FIG. 2. (a) shows the time evolution of the photoluminescence of as-grown $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ quantum wells (b) is a sample intermixed by an impurity-free method, and (c) shows the sample intermixed by proton implantation.

$$I_{\text{PL}}(t) = a(e^{-t/\tau_d} - e^{-t/\tau_r}),$$

where τ_d and τ_r are the decay- and rise-time constants, respectively. Results for the other quantum wells (QW2 and QW3) situated deeper in the material were analogous, and are also given in Table II. It is therefore clear that while the time averaged (cw) PL data of samples intermixed by various methods can be indistinguishable, the carrier dynamics in the various samples is quite different and is a function of the type of intermixing used.

The reason for the difference in the PL decay times can be explained by the presence of defects in the quantum well and or the barrier region of the implanted sample, and by their absence in the samples intermixed by the impurity-free method. Ion implantation is known to create a range of non-radiative states which act as trapping centers thereby reducing the effective lifetime of the photoexcited carriers.⁸ However, since the total (time integrated) PL of the two types of samples was found to be similar, the introduction of nonra-

TABLE II. PL rise times (τ_r) and decay times (τ_d) for the quantum wells in the as-grown and intermixed samples.

	As-grown		Impurity-free intermixed		Ion-implanted intermixed	
	τ_r (ps)	τ_d (ps)	τ_r (ps)	τ_d (ps)	τ_r (ps)	τ_d (ps)
QW1	18	879	15	1000	10	197
QW2	22	1716	21	1700	11	243
QW3	29	6000	22	5500	8	577

diative centers in the implanted samples has to be balanced by some other process which increases the PL efficiency in these materials. We have recently shown that ion-implantation-induced intermixing improves the carrier collection efficiency in quantum wells, an effect which results in shorter carrier capture times (into the wells) and is revealed by a shorter PL rise time.⁹ Using resonant and above-gap excitation we were able to separate the various components effecting the carrier capture into the quantum well. We have shown that the reduction in the carrier capture times is the consequence of the change in the quantum well potential caused by intermixing, and is not related to the introduction of nonradiative centers.⁹ Since the PL rise times in the samples intermixed by implantation (for all three quantum wells) are consistently shorter than those intermixed by the impurity-free method, we can conclude that the carrier collection efficiency in the ion-implanted samples is more efficient than in the samples intermixed by the impurity-free method. This improvement in the carrier collection can explain the similar integrated PL intensities in the two types of samples, in spite of the existence of nonradiative traps in the implanted samples. This effect is even clearer for the deeper quantum wells, QW2 and QW3, for which the impurity-free method can only produce a limited amount of intermixing for reasons that are related to the decrease of vacancy diffusion with depth. For these quantum wells, the PL rise times are significantly shorter in the case of implantation than in the case of the impurity-free intermixing.

The effect of carrier transport on the PL rise time can be neglected because the optical excitation depth coincides approximately with the thickness of the QW structure. Taking the absorption coefficient of InP from the literature¹⁰ we find

that the penetration depth of the excitation is approximately 300 nm, while the layer thickness of the QW structure is 180 nm.

In summary, we have shown that in intermixed quantum wells the carrier capture into quantum wells and carrier relaxation from the wells depends on the type of intermixing used. We found that the carrier lifetimes are significantly longer in samples intermixed by the impurity-free method, but the carrier collection of the quantum wells is more efficient in the samples intermixed by ion implantation.

ACKNOWLEDGMENT

The authors gratefully acknowledge the financial support of the Australian Research Council.

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