

Feature Article

Relaxation and recombination in InAs quantum dots

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1 Introduction

Over the last 30 years the development of the sophisticated semiconductor growth techniques of molecular beam epitaxy (MBE) and metal organic vapor phase epitaxy (MOVPE) has resulted [1] in the growth of a range of low dimensional structures that have revealed new physical phenomena as well as being exploited by the creation of new classes of electronic and optoelectronic devices. In particular using MBE and MOVPE [1] it is possible to grow multi layer structures whose interfaces are very sharp and where the constituent layers can be as thin as a few monolayers. Initially quantum well structures made up from layers of GaAs/Al_xGa_{1-x}As were studied intensively. The underlying reason for this is that the lattice parameters of GaAs and Al_xGa_{1-x}As are virtually the same over the entire range of composition x thus no untoward effects due to strain, such as dislocation formation, occur. GaAs/Al_xGa_{1-x}As quantum wells were incorporated in the first generation of optoelectronic devices that relied on quantum effects for their operation and also have proved to be the model system for the study of the physics of quantum wells.

Of course, the fundamental consequence of quantum confinement is that by choice of the confinement layer thickness we can, to an extent, choose the energy of photons that will be emitted when electrons in the lowest confined state recombine with holes in the lowest confined state. This degree of freedom had immediate impact in the field of semiconductor lasers whereby it was possible to produce devices in which the active region was GaAs that could emit light at wavelengths shorter than 880 nm [2]. Quantum well structures have also been exploited in other devices, for example in modulators using the Quantum Confined Stark Effect [3] and solar cells [4].

The physics of low dimensional structures is usually studied using electrical or optical measurement techniques, in this article we will concentrate on the latter. Of fundamental importance in quantum well structures is the strength of the Coulomb interaction [5] between the photo-excited electrons and holes which opened up the study of the basic physics two dimensional excitons. A good example of this is the study of the low temperature exciton recombination dynamics taken under conditions of non resonant

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excitation [6]. In this work it was reported that the measured exciton decay time decreased with decreasing quantum well thickness, this was attributed to the effect of the increased overlap of the electron and hole wavefunctions due to the spatial confinement. This type of study involving the study of the dynamics of recombination has proved to be particularly fruitful, in particular temperature [7] and excitation energy [8] dependent studies led to increased understanding of the processes governing the dynamics of excitons in two dimensional systems.

In general, at low temperature and under non resonant excitation conditions, the exciton radiative recombination times in the majority of quantum well structures are in the nanosecond or sub-nanosecond time regime. This statement needs to be qualified in that it applies to type I quantum well structures, i.e. where the lowest energy electron and hole wavefunctions are confined to the same layer of material. In the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ materials system it is possible to choose the compositions and layer thicknesses to create a type II quantum well system [9], i.e. where the lowest energy electron and hole wavefunctions are confined in the different layers of material. Probably the most widely studied type II quantum well system is made up of layers of GaAs whose thickness is <3.5 nm and the barrier layers are AlAs [10]. This results in the lowest energy confined electron state being associated with the X minima in the AlAs and lowest energy confined hole state being the $n = 1$ heavy hole state in the GaAs. The resulting spatial separation of the electron–hole wavefunctions intrinsically leads to radiative lifetimes that can approach microseconds (depending on the particular configuration of the quantum well system) [11]. As well as opening up the study of the dynamics of spatially separated electrons and holes type II GaAs/AlAs quantum well system proved to be a model system for study of inter layer scattering of electrons [12].

So in summary of this very brief overview the fabrication of quantum well structures opened up a myriad of opportunities for the study of new physical phenomena as well enabling the creation of devices with improved and novel functionality. It was anticipated that further technological and scientific developments would occur if carriers could be confined in all three dimensions, i.e. quantum dots. In particular it was envisaged that the threshold current density of quantum dot lasers would exhibit reduced sensitivity to temperature [13] and could serve in systems that require the controllable emission of single photons [14].

2 Quantum dots

For many years the fabrication of high quality quantum dots proved to be problematic, the most widely employed technique involved nanolithography of quantum well structures [15]. Quantum dots fabricated in this way suffered from a range of problems, perhaps most significant of these was the incorporation of defects. These difficulties were overcome by the exploitation of the Stranski–Krastanov coherent island growth mode which enabled the fabrication of self assembled quantum dots. Although, first described in the 1930s the technique was introduced in 1990 for the growth of coherent semiconductor islands [16]. During the heteroepitaxy of a semiconductor with a lattice constant significantly different from that of the substrate, e.g. in the case of InAs on GaAs strong strain fields occur due to the large lattice mismatch of 7.2%. This leads to relaxation of the InAs after deposition of a few monolayers (ML) and dislocations are formed. If layers are grown before this relaxation has occurred small strained islands form which are free of defects. These islands appear spontaneously after deposition of a critical thickness of 1–2 MLs of InAs. The driving force of this phase transition is the reduction of the free energy however kinetic effects also play an important role. When equilibrium is reached quantum dots are formed residing on a thin wetting layer of 1–2 MLs of InAs as shown by the cross sectional Transmission Electron Microscopy (TEM) image in Fig. 1.

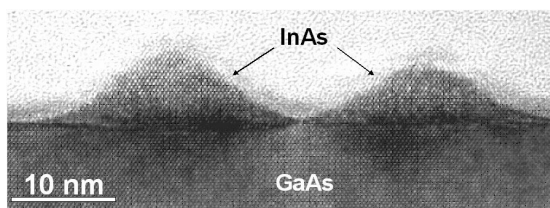


Fig. 1 High resolution cross sectional TEM image in the $\langle 110 \rangle$ direction of two neighbouring InAs quantum dots on a GaAs surface.

Using this technique has enabled the growth of quantum dots in different material systems such as Si/Ge [17], InP/GaAs [18], GaSb/GaAs [19], CdSe/ZnSe [20] and GaN/AlN [21]. Nevertheless the most widely studied system has been InAs/GaAs which, much like the GaAs/Al_xGa_{1-x}As quantum well system has become the model for the study of the basic physics of semiconductor quantum dots.

2.1 Low temperature optical properties of InAs/GaAs quantum dots

InAs/GaAs quantum dots formed by the technique described above usually have heights in the range 3–12 nm and base widths in the range 15–30 nm although it is possible, to an extent, to control these parameters by variations in the growth conditions [21]. In Fig. 2 are shown a series of photoluminescence spectra taken at $T = 6$ K using an InAs/GaAs structure with a dot density of $\sim 10^{11}$ cm⁻² with different excitation power densities and the excitation photon energy greater than the GaAs barrier bandgap.

For the time integrated spectra shown in Fig. 2 the excitation spot was ~ 3 mm diameter, thus the width of the spectra represent the variation in the energies of the confined electron and hole states of the quantum dots due to variations in the dimensions and chemical compositions of the dots within the excitation volume. The excitation power densities used were such that the range of average carrier densities varied from less than one electron–hole pair per dot to greater than one electron–hole pair per dot. The spectrum recorded at the lowest excitation density (Fig. 2(c)) has a single feature, with a peak energy of 1.131 eV, which is due to recombination involving carriers in the lowest energy confined electron and hole states (designated as ground state recombination). As the excitation density is increased, features at higher energy appear in the spectra at 1.205, 1.277 and 1.345 eV. These peaks have been attributed previously [22] to recombination involving excited states of the InAs/GaAs quantum dots. The emergence of the recombination due to the higher lying transitions is due to state blocking effects, caused by Pauli exclusion, when the lower states are fully occupied. In Fig. 3 are shown photoluminescence decay curves of the ground state recombination for three different excitation power densities equivalent to those used for the spectroscopy shown in Fig. 2. As the excitation density is increased a progressive change occurs in the form of the decay curves. For the lowest excitation density (Fig. 3(c)) the rise of the photoluminescence intensity is rapid, and then slows, reaching a peak after ~ 800 ps. As the excitation density is increased the rise of the photoluminescence intensity becomes more rapid (Fig. 3(a), (b)) and also for the highest excitation density a plateau occurs at the top of the decay curve after the maximum intensity has been reached.

The observed plateau is a particular characteristic of the zero dimensional character of the quantum dots. In the simplest picture the plateau occurs because of state blocking when, on average, the quantum

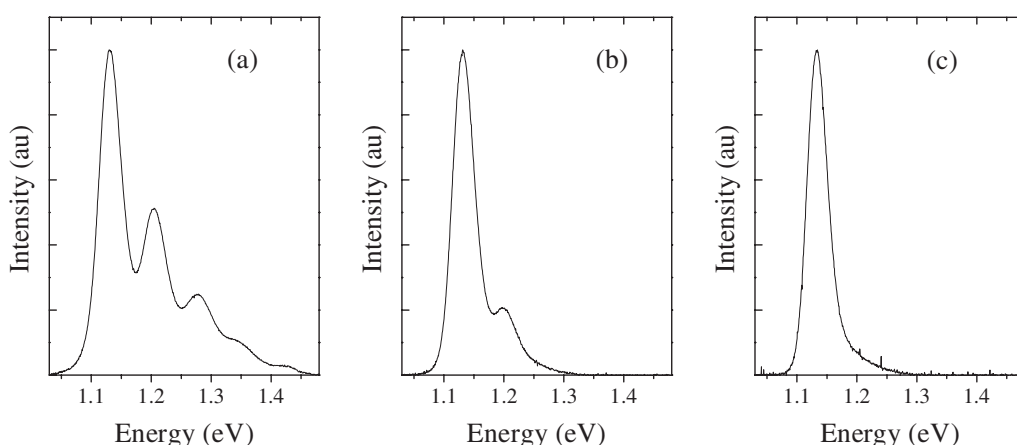


Fig. 2 Time integrated photoluminescence spectra for excitation photon densities per pulse of (a) 2×10^{14} cm⁻², (b) 8×10^{12} cm⁻² and (c) 3×10^9 cm⁻².

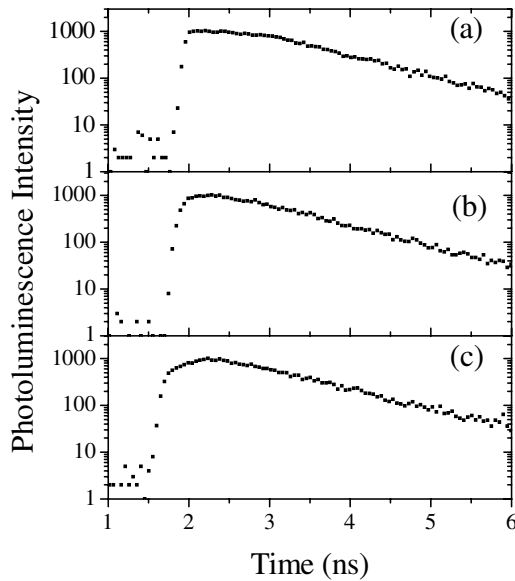


Fig. 3 Photoluminescence time decays measured at the peak of the ground state emission for excitation photon densities per pulse of (a) $2 \times 10^{14} \text{ cm}^{-2}$, (b) $8 \times 10^{12} \text{ cm}^{-2}$ and (c) $3 \times 10^9 \text{ cm}^{-2}$.

dots are occupied by more than one electron–hole pair. In this case if a ground state electron–hole pair recombines there is a finite probability that the ground states can be filled by carriers that are in higher lying states having been prevented from relaxing to the ground states. If this occurs the photoluminescence intensity from a significant subset of the quantum dots in any excitation cycle remains constant after the excitation has been switched off leading to the observed plateau. This behavior has been analyzed and quantified using either rate equations [23, 24] or Monte Carlo simulations [25]. In all cases the photoluminescence decays with a time constant ~ 1 ns, indicative of the strong spatial overlap of the electron–hole wavefunctions.

Significant modifications occur to the spectra if the excitation photon energy is tuned to below the GaAs bandgap and in particular to lie within the inhomogeneously broadened spectrum. So called resonant excitation can reveal two types of features, firstly phonon assisted absorption [22, 26–28] and secondly features [29–31] due to the resonant excitation of a sub-set of the excited states of the quantum dots which then leads to an enhancement of the intensity of the ground state photoluminescence of the resonantly excited sub-set. This latter process is of particular relevance to the discussion to be presented on the optical properties of InAs quantum dots with AlAs barriers.

Along with the study of quantum dot ensembles the study of the spectroscopy of single quantum dots is proving to be particularly fruitful. Such studies are usually made in samples where the quantum dot density is relatively low and the individual quantum dots are isolated by the use of a sub-micron mesa structure. Using this technique the inhomogeneous broadening due to variations in size and composition of the dots is removed from the emission spectra and the photoluminescence spectra usually consist of very sharp lines due to recombination of, for example, excitons, multiple exciton complexes and charged excitons depending on the experimental conditions [32]. If InAs quantum dots are to be useful in device structures such as lasers or sources for single photon emission they must maintain their radiative efficiency at higher temperatures, in particular room temperature.

2.2 Temperature dependent optical properties of InAs/GaAs quantum dots

Studies of the temperature dependence of the optical properties of an ensemble of quantum dots can reveal information on the following. Firstly the temperature dependence of the photoluminescence intensity is a useful way of monitoring the emergence of any thermally activated non radiative recombination processes. Secondly by monitoring such properties as the photoluminescence line width and peak energy

information can be obtained on how the carriers may be redistributed amongst the quantum dot ensemble. These two aspects can be interrelated in that if the carriers are redistributed by a thermally activated process(es) then the carriers can be lost non-radiatively during this cycle. In particular it has been found [32–41] that the peak energy (E_p) can decrease more rapidly as a function of temperature than might be expected if the change was governed solely by the change in the bandgap of InAs. Furthermore it has been observed that the full width at half maximum height (FWHM) of the spectrum can also decrease as the temperature is raised up to some critical temperature followed by an increase in the FWHM at higher temperatures. Linked to this behavior the photoluminescence intensity remains essentially constant in the low temperature regime but decreases as the temperature is increased further. It should be stressed that this behavior is not universal [42] and depends on the particular structure being studied. In fact the parameter that determines whether this behavior is seen is the value of the peak energy at low temperature which in turn reflects the depth of the carrier confinement.

In Fig. 4 are shown the results of measurements taken with low excitation density of the integrated photoluminescence intensity, relative peak position and FWHM from two quantum dot structures (sample numbers 617 and 592) with values of E_p at low temperature of 1.105 and 1.23 eV. These two samples show the contrasting behavior described above. It should be noted that the data for E_p have been corrected to reflect only changes due to relaxation and thermalisation so the expected change in the bandgap of InAs has been removed using Varshni's equation incorporating the constants provided by Vurgaftman et al. [43]. As can be seen in Fig. 4 the change in E_p for sample 592 is much less pronounced than that for sample 617. The FWHM for 592 remains relatively unchanged with increasing temperature whereas the FWHM for 617 decreases then increases with a minimum occurring around 140 K. The integrated intensity from sample 592 decreases by only about a factor of 2 on increasing the temperature from 10 to 200 K, whereas, the integrated intensity of 617 remains relatively constant up to 70 K but then falls much more rapidly having decreased by well over an order of magnitude at 200 K.

Qualitatively these changes in the optical properties of sample 617 can be explained as follows. As the temperature is increased the carriers that are in less well confined quantum dots, i.e. those in the smaller quantum dots that contribute to the high energy part of the photoluminescence spectrum, are thermally emitted to the barrier and/or wetting layer and subsequently retrapped or lost through non radiative or radiative recombination in the barrier layers. Thus as the temperature is increased the linewidth is reduced and the peak energy gradually shifts to lower energies until the temperature is sufficiently high that the carriers are thermally distributed across the quantum dot states. As for sample 592 the average confinement depth is much greater as reflected in much lower value of E_p meaning that any thermally

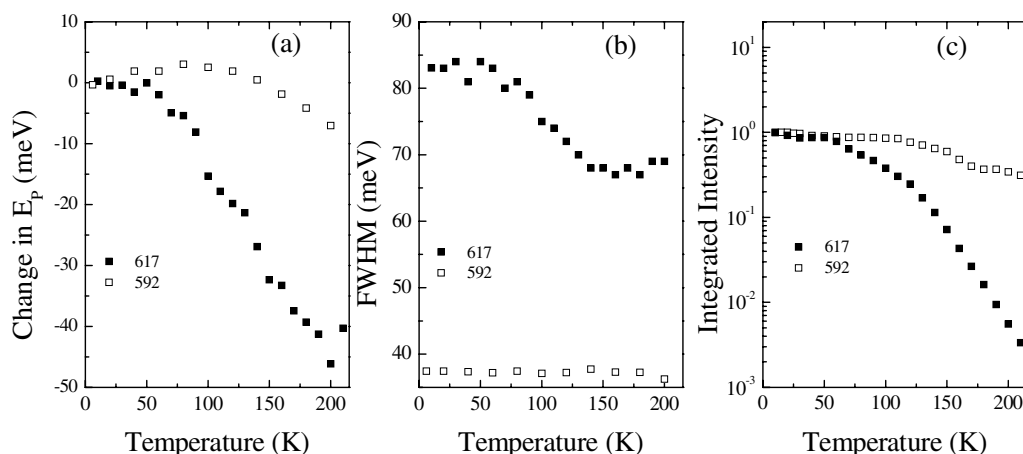


Fig. 4 Low excitation intensity experimental data for (a) the relative photoluminescence peak energy, (b) the FWHM of the spectra and (c) the integrated photoluminescence intensity for the samples 592 and 617.

activated carrier transfer processes are much less effective over the temperature range studied. Theoretical descriptions of this behavior are usually achieved using a set of rate equations which describe the kinetics of the carrier exchange between the quantum dots and the barrier via the wetting layer [44, 45]. In the past it has been assumed that the carriers behave as correlated electron–hole pairs (excitons) during the thermally stimulated exchange processes [46, 47]. Recently a much more comprehensive theoretical model [48] was described in which the consequences of carrier exchange involving independent electrons and holes was compared with that involving excitons. We do not propose to describe this work in detail here but below we note the main elements of the theory and show the contrast between the predictions of the theoretical results for the independent electron–hole and exciton models compared with the experimental data. The main elements of the model as follows:

- a system of quantum dots in a semiconductor matrix where electron–hole pairs are generated continuously in the barriers,
- these carriers can either recombine or be captured by the quantum dots,
- the quantum dots have only one confined electron and hole states and the spectral distribution function for these states was assumed to be Gaussian,
- carriers captured into the quantum dots can then either be thermally activated into the barrier layer or recombine radiatively.

The time evolution of the carrier concentrations in the barrier and the quantum dots were then described by a set of coupled rate equations. In contrast to previous theoretical treatments this model includes the following:

- A clear distinction was made between carrier relaxation in the form of independent carriers or excitons.
- The redistribution of carriers in the quantum dots occurs via the GaAs continuum states.
- The energy differences between the quantum dot states and the matrix were not used as fitting parameters but were determined as a function of dot size [49].
- When the photoluminescence spectra were calculated the anticorrelation of the confinement energies of the electron and hole states was taken into account. This means that the smaller quantum dots have smaller confinement energies for both electrons and holes.

In Figs. 5 and 6 we show the predictions of this model for the optical properties of the samples that we studied experimentally for both the independent electron–hole and exciton models. Let us first discuss the comparison of theory and experiment for sample 617 (Fig. 5) where the most radical changes in the optical properties are observed. For all the observed properties the predictions of the independent elec-

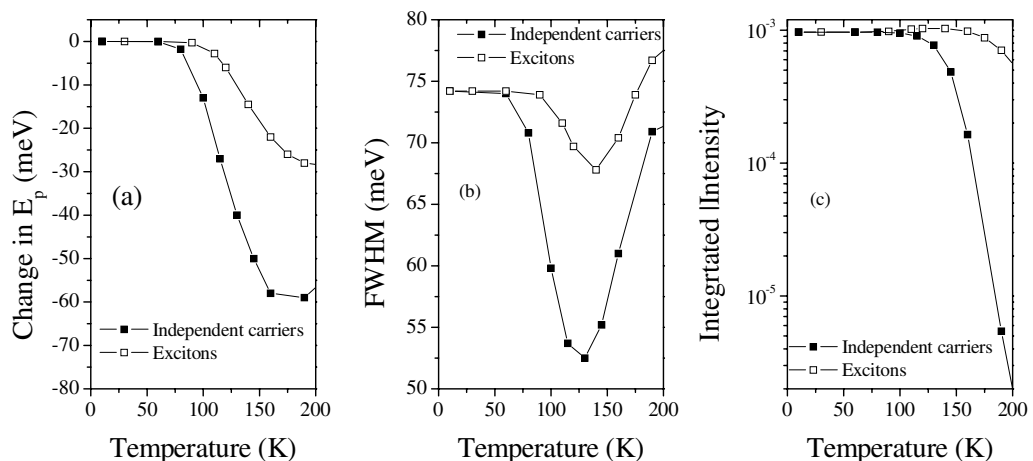


Fig. 5 Predictions of (a) the relative photoluminescence peak energy, (b) the FWHM of the spectra and (c) the integrated photoluminescence intensity for the sample 617. The open and filled symbols correspond to the excitonic and non excitonic models respectively.

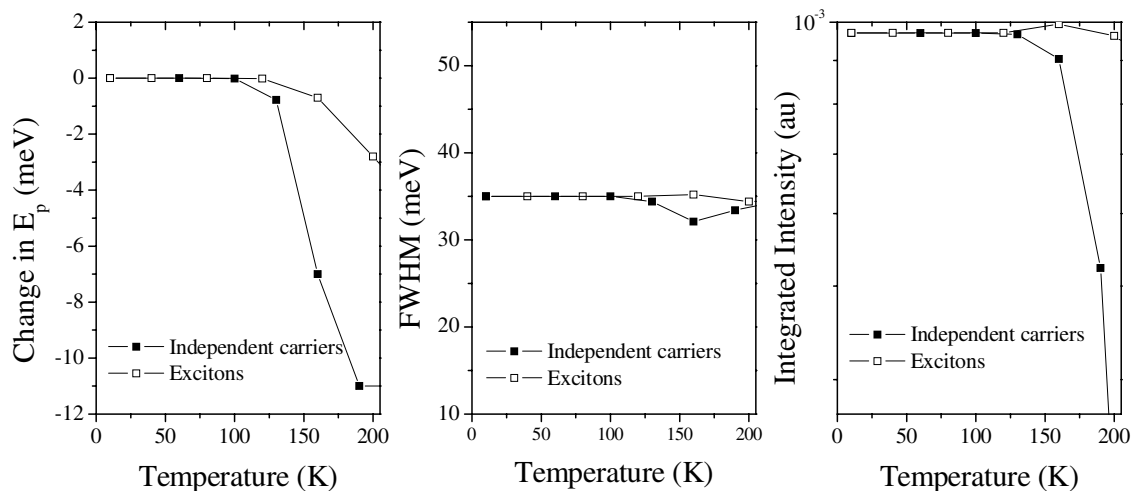


Fig. 6 Predictions of (a) the relative photoluminescence peak energy, (b) the FWHM of the spectra and (c) the integrated photoluminescence intensity for the sample 592. The open and filled symbols correspond to the excitonic and non excitonic models respectively.

tron–hole model are in far better agreement with the experimental data than the exciton model. Particularly striking is the prediction that the fall off in integrated intensity with increasing temperature is much less for the excitonic model than the free electron–hole model. This is because in the exciton description the recombination partner for an electron or hole is always available, whereas in the case of the independent electron–hole model the probability of finding a recombination partner decreases with increasing temperature thus increasing the chance of non radiative recombination.

As for sample 592 (Fig. 6) again the agreement between the predictions of the independent electron–hole model and the experimental data are much better than the exciton model.

Of course the changes in the optical properties are much less severe, both theoretically and experimentally, which is due to the much greater carrier confinement in this sample. To an extent the agreement (or not) between the theory and experiment was governed by the choice some of the input parameters. In the original work [48] the conclusions concerning the dominance of the free carrier escape were further supported by comparison between theory and experiment at higher injection levels which removed the qualification above concerning the choice of input parameters.

As we have already said the latter description is in much better agreement with the experimental data as might be expected because the Coulomb binding energy in this type of quantum dot is relatively small, i.e. a few meV. In systems where this energy might be expected to be much larger, e.g. nitride materials, this exciton effect may play a role in determining the room temperature photoluminescence efficiency.

The results of the study described above clearly suggest that because of the dominance of the independent electron–hole mechanism to achieve high optical efficiency at room temperature it should be advantageous to use as wide a bandgap barrier material as possible.

2.3 Optical properties of InAs/AlAs quantum dots

As just stated to minimize the effects of thermionic emission it should be advantageous to use barrier material with as large a bandgap as possible. The natural choice would therefore seem to be quantum dots surrounded by AlAs barriers. The use of AlAs barriers also brings the added advantage of shifting the peak of the emission from the GaAs quantum dots into the spectral region around 780 nm where they could be used in devices used for optical reading. With these ideas in mind we have undertaken a study of the basic optical properties of InAs/AlAs quantum dots, in particular we have investigated in detail the

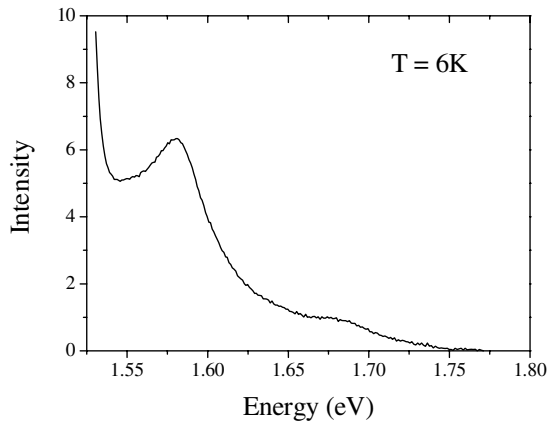


Fig. 7 Time integrated photoluminescence spectrum from InAs/AlAs quantum dot structure recorded at $T = 6$ K.

recombination dynamics. Depending on the growth conditions the peak energy can lie in the range 1.5–1.7 eV. An example of the photoluminescence spectrum measured at $T = 6$ K is shown in Fig. 7. The emission from the quantum dots is the broad feature that peaks at 1.583 eV, the sharply rising feature at the low energy limit of the spectrum is due to emission from the underlying GaAs buffer layer. Not only is the emission wavelength moved to much higher photon energy when compared with InAs/GaAs quantum dots but a significant change occurs in the recombination dynamics as illustrated in Fig. 8.

In Fig. 8 is shown a photoluminescence decay curve of the quantum dot emission, the non exponential decay occurs on a time scale of many thousands of nanoseconds. Initially two explanations were put forward to explain this particular observation. The first suggestion [50] drew parallels with the situation in GaAs/AlAs quantum wells where the recombination can occur over a similar time scale (type II structure).

As already discussed the long time scale for recombination in GaAs/AlAs type II quantum wells is because the optically injected electrons and holes become spatially separated with the electrons scattering to confined states associated with the X minima in the AlAs. The first striking observation of this work was the appearance of a series of sharp lines, as shown in Fig. 9, in the low temperature ($T = 6$ K) photoluminescence spectra as the excitation energy was changed.

So it was proposed that the X electron states played a role in the recombination from InAs/AlAs quantum dots by allowing the electrons that were photo excited in the InAs quantum dots to scatter to adjacent quantum dots that were not occupied by holes. Thus the subsequent recombination that occurred

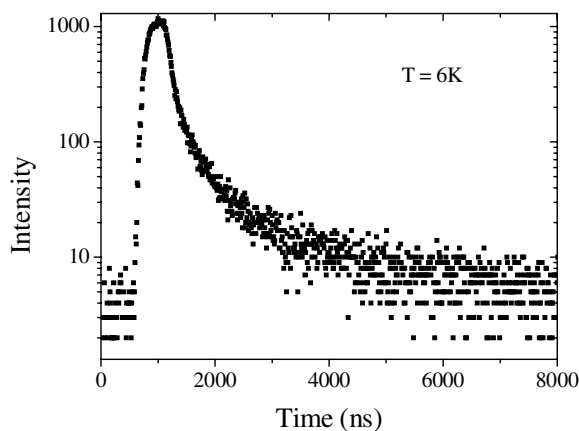


Fig. 8 Photoluminescence decay curve of recombination from InAs/AlAs quantum dot structure measured at $T = 6$ K.

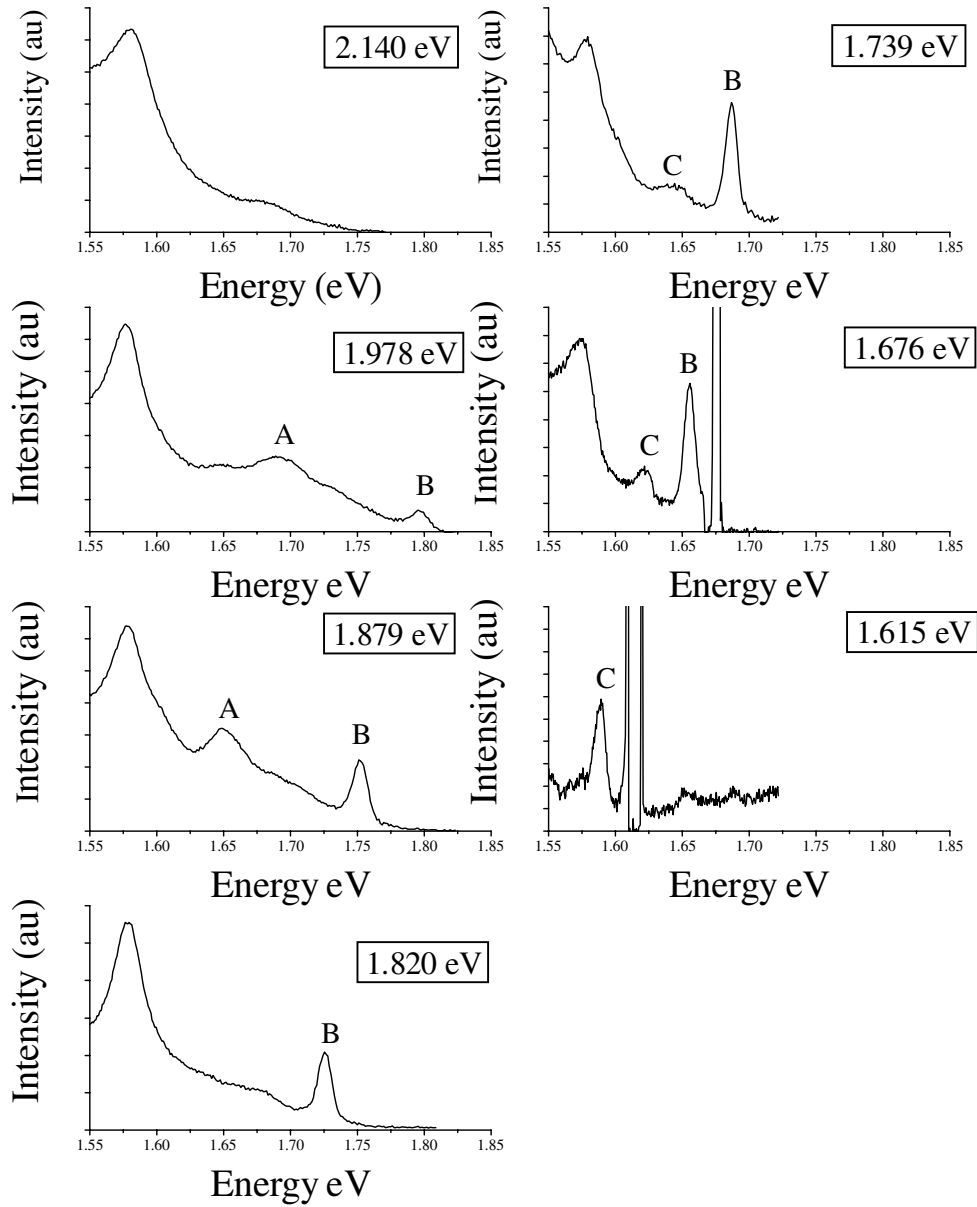


Fig. 9 Low temperature ($T = 6$ K) photoluminescence spectra measured with the indicated excitation photon energies.

involved spatially separated electrons and holes that occupied different quantum dots. An alternative explanation was put forward by Shamirzaev et al. [51] who explained the long-lived nature of the decays as being due to the quantum dot ground states being triplet in nature. To an extent this argument has been supported by theoretical estimates [52] of the exchange splitting in small InAs/AlAs quantum dots. More recently a modification of the model described in the work by Dawson et al. [50] has been put forward [53] to explain the long timescales for the recombination. This work involved more detailed low temperature photoluminescence decay time and spectroscopy measurements.

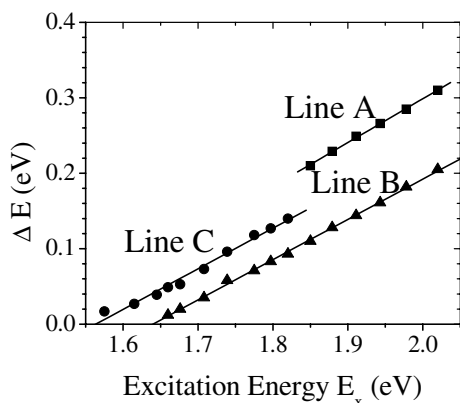


Fig. 10 Plots of the difference in the peak energies of lines A, B and C and the excitation photon energy (ΔE) and the absolute value of the excitation photon energy E_x .

For the highest excitation photon energy (2.140 eV) used the spectrum has a well defined peak at 1.583 eV and a distinct shoulder at 1.645 eV. As the photon energy is progressively reduced (for the sake of clarity not all the spectra recorded are shown here) the spectra change radically. For the 1.978 eV excitation two extra distinct features emerge, labeled A and B. As the excitation energy is further reduced A and B move to lower energy, until eventually line A disappears around 1.64 eV. For further reductions in excitation energy below 1.820 eV another line, labeled C emerges. A particular characteristic of A, B, and C are that as the excitation energy is reduced the energy difference between A, B and C and the excitation energy also reduces such that in the cases of B and C the excitation and emission energies finally become coincident. This overall behavior is illustrated in Fig. 10 where the difference in the excitation energy (ΔE) and the peak emission energy for the lines A, B and C is plotted against the excitation energy (E_x). The gradients of the linear fits to the data shown in Fig. 10 are equal within the range 0.55 ± 0.03 . Along with the appearance of these relatively sharp line spectra we also note that for excitation photon energies below 1.615 eV the broad photoluminescence feature at 1.583 eV, which dominates the spectra at high excitation photon energies, disappears.

To try to understand the nature of the sharp lines the decay of the photoluminescence at a particular energy was measured when the sharp line was present at that particular detection energy and when it was not. This was achieved by varying the excitation energy. Experimentally it was found that the decay curves were essentially unchanged suggesting that when a sharp line is present at a particular detection energy there is no radical change in the nature of the emission process but rather the excitation energy used has led to an enhancement of the emission intensity at that energy. This enhancement is as was observed [29–31] in the InAs/GaAs quantum dot system when the excitation energy was resonant with the excited states of a subset of the quantum dots resulting in increased emission from the ground states of the particular subset. In such a process the photo excited electron and hole remain within the same quantum dot. We can rule out at least part of that process in the situation considered here because the photoluminescence decays occur over a much longer time scale than nanoseconds.

The results described above demanded that modifications needed to be made to the previous work [50] in which it was proposed that the long recombination times were due the spatial separation of electrons and holes into different quantum dots via scattering involving the X states of the AIs. If this model was wholly correct the sharp line spectra could only be explained by resonant excitation of a subset of the quantum dots followed by recombination of the electron–hole pair within the same quantum dot. If this was the case then the observed decay times of the sharp lines would be on the nanosecond time scale, which they clearly are not.

Thus the following modification was proposed. As pointed out by Williamson et al. [54] for the InP/GaP quantum dot system the local strain field in the GaP caused by the InP quantum dots can lead to localized states associated with the GaP X minima. The depth of the localisation is influenced by the size of the quantum dots and also the degree of lattice mismatch between the quantum dots and the barrier

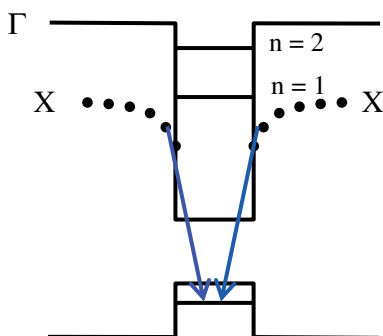


Fig. 11 (online colour at: www.pss-b.com) Schematic diagram of the recombination process involving electrons at the localised AlAs X minima (dotted line) and holes in the InAs. The Γ conduction band edges and the confined states in the InAs quantum dots are indicated by the black continuous lines.

materials. Thus this idea was extended to InAs/AlAs quantum dots, bearing in mind that the separation of the InAs quantum dots can be as small as 1 nm this strain effect could lead a continuous distribution of localized X states where the precise energy of any specific state is determined by the size and composition of the adjacent InAs quantum dots. Thus because of the variation in the sizes and the chemical composition of the quantum dots one could anticipate a wide range of localised X like states in the barrier material. Thus it was proposed that the emission above 1.675 eV is due to the recombination of electrons localised in X related states located in the AlAs with holes in the InAs quantum dots as shown schematically in Fig. 11.

This view then leads to a natural explanation for the lines A and B as being due to the resonant excitation of electrons to either the $n = 1$ or the $n = 2$ states in the InAs quantum dots and the subsequent enhancement of the emission involving the adjacent localized X states. Also this model can be used to explain why the recombination dynamics occur over a relatively long time scale and are strongly influenced by the energy difference between the excitation and recombination states. The strong decrease of the recombination rates of A and B with decreasing energy difference between the excitation and recombination states was ascribed as being due to the strong spatial mixing of the Γ and X electron wavefunctions. Furthermore, the non exponential character of the decay of the quantum dot emission was explained by the fact that at any particular detection energy emission was monitored where the degree of mixing varied.

According to the previous model [50] the 1.583 emission band is due to recombination of electrons and holes being localized in spatially separated InAs quantum dots. The spatial separation of the electrons and holes is possible as long as the Γ states within the dot are at higher or equal energy as the lowest X states in the barrier (note: whose position depends on the dot size as well due to the strain fields involved). As the excitation photon energy decreases below the X states (in the largest dots) the route for electrons to leave the quantum dots where they have been excited originally is removed and only fast recombination within the quantum dots, i.e. line C remains.

3 Summary

Using the sophisticated epitaxial growth technique of MBE it has been possible to grow high quality InAs quantum dot structures which has opened up the study of relaxation and recombination of carriers in quantum dots. In this article we have described recent work in two particular aspects of this field of study. Firstly, we considered the results of a detailed comparison of theory with experiment on the temperature dependence of the photoluminescence spectra and intensity of InAs/GaAs quantum dots. In the past the temperature induced change of carriers between the quantum dots had been described in terms of correlated electrons and holes (excitons). By comparing the results of a theoretical treatment in which the carrier exchange between the quantum dots was considered to involve excitons or free electrons and holes we were able to demonstrate the dominance of the former process for the InAs/GaAs quantum dot system. The theory developed is not only of importance for the InAs/GaAs system, in systems where the Coulomb interaction is much stronger the excitonic contribution to the exchange process may have im-

pact on the high temperature photoluminescence efficiency. Secondly we presented the current understanding of the optical properties of the InAs/AlAs quantum dot system. Similarly to GaAs/AlAs *quantum wells* there is a significant body of evidence that the indirect nature of the AlAs bandgap plays a significant role in determining the nature and time scale of the recombination at low temperatures. Specifically some of the spectral features have been attributed as involving electrons localised at strain induced X related minima in the AlAs with holes in the InAs quantum dots. Although it should be stressed that this conclusion is at variance with an alternative view presented by other workers who have suggested that the electrons occupy states in the InAs quantum dots. Clearly more work needs to be done to fully understand the character of the electron state. Perhaps there is again an opportunity for the technique of Optically Detected Magnetic Resonance to provide some definitive conclusions, much as was the case for type II GaAs/AlAs quantum wells [55]. Nevertheless the long recombination times observed at low temperatures suggest that the InAs/AlAs quantum dot system may not be as readily exploited in optical devices at room temperature as the time scale for radiative recombination may make such quantum dots susceptible to non-radiative recombination processes.

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References

- [1] E. O. Göbel and K. Ploog, *Prog. Quantum Electron.* **14**, 289 (1991).
- [2] K. Woodbridge, P. Blood, E. D. Fletcher, and P. J. Hulyer, *Appl. Phys. Lett.* **45**, 16 (1984).
- [3] D. A. B. Miller, D. S. Chemla, T. C. Damen, W. Weigman, T. H. Wood, and C. A. Burus, *Phys. Rev. Lett.* **53**, 2173 (1984).
- [4] K. W. J. Barnham and G. Duggan, *J. Appl. Phys.* **67**, 3490 (1990).
- [5] R. L. Greene, K. K. Bajaj, and D. E. Phelps, *Solid State Commun.* **45**, 831 (1983).
- [6] E. Göbel, H. Jung, J. Kuhl, and K. Ploog, *Phys. Rev. Lett.* **51**, 1588 (1983).
- [7] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliott, *Phys. Rev. Lett.* **59**, 2337 (1987).
- [8] B. Deveaud, F. Clérot, N. Roy, K. Satzke, B. Sermage, and D. S. Katzer, *Phys. Rev. Lett.* **67**, 2355 (1991).
- [9] P. Dawson, B. A. Wilson, C. W. Tu, and R. C. Miller, *Appl. Phys. Lett.* **48**, 541 (1986).
- [10] P. Dawson, K. J. Moore, and C. T. Foxon, *Proc. SPIE* **792**, 208 (1987).
- [11] P. Dawson, K. J. Moore, C. T. Foxon, G. W. 't Hooft, and R. P. M. van Hal, *J. Appl. Phys.* **65**, 3606 (1989).
- [12] J. Feldmann, E. O. Göbel, J. Nunnenkamp, J. Kuhl, K. Ploog, P. Dawson, and C. T. Foxon, in: *Condensed Systems of Low Dimensionality*, NATO ASI Series, Vol. 253 (Plenum Press, New York, 1991), p. 191.
J. Nunnenkamp, J. Kuhl, K. Ploog, J. Feldmann, E. O. Göbel, P. Dawson, and C. T. Foxon, in: *Ultrafast Phenomena VII*, Springer Series in Chemical Physics, Vol. 53, edited by C. B. Harris, E. P. Ippen, G. A. Monroe, and A. H. Zewail (Springer Verlag, Berlin, Heidelberg, 1990), p. 230.
- [13] Y. Arakawa and H. Sakaki, *Appl. Phys. Lett.* **40**, 939 (1982).
- [14] C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon, and Y. Yamamoto, *New J. Phys.* **6**, 89 (2004).
- [15] L. Birotheau, A. Izrael, J. Y. Marzin, R. Azoulay, V. Thierry-Mieg, and F. L. Ladan, *Appl. Phys. Lett.* **61**, 3023 (1992).
P. M. Petroff, A. C. Gossard, R. A. Logan, and W. Weigmann, *Appl. Phys. Lett.* **41**, 635 (1982).
A. Izrael, B. Sermage, J. Y. Marzin, A. Ougazzaden, R. Azoulay, and J. Erillard, *Appl. Phys. Lett.* **59**, 3577 (1991).
- [16] J. M. Moisan, F. Houzay, F. Barthe, E. André, and O. Vatel, *Appl. Phys. Lett.* **64**, 196 (1994).
- [17] S. Fukatsu, H. Sunamura, Y. Shiraki, and S. Komiyama, *Appl. Phys. Lett.* **71**, 258 (1997).
- [18] M. K. K. Nakaema, F. Iikawa, M. J. S. P. Brasil, E. Ribeiro, G. Medeiros-Ribeiro, W. Carvalho, M. Z. Maialle, and M. H. Degani, *Appl. Phys. Lett.* **81**, 2743 (2002).
- [19] E. Ribeiro, A. O. Gororov, W. Carvalho, and G. Medeiros-Ribeiro, *Phys. Rev. Lett.* **92**, 126402 (2004).
- [20] D. Hommel, K. Leonardi, H. Heinke, H. Selke, K. Ohkawa, F. Gindele, and U. Woggon, *phys. stat. sol.* (b) **202**, 835 (1997).

- [21] B. Daudin, F. Widmann, G. Feullet, Y. Samson, M. Arlery, and J.-L. Rouviere, *Phys. Rev. B* **56**, R7069 (1997).
- [22] M. J. Steer, D. J. Mowbray, W. R. Tribe, M. S. Skolnick, M. D. Sturge, M. Hopkinson, A. G. Cullis, C. R. Whitehouse, and R. Murray, *Phys. Rev. B* **54**, 17738 (1996).
- [23] F. Adler, M. Geiger, A. Bauknecht, F. Scholz, H. Scheizer, M. H. Pilkuhn, B. Ohnesorge, and A. Forchel, *J. Appl. Phys.* **80**, 4019 (1996).
- [24] M. Grundmann and D. Bimberg, *Phys. Rev. B* **55**, 9740 (1997).
- [25] P. D. Buckle, P. Dawson, S. A. Hall, X. Chen, M. J. Steer, D. J. Mowbray, M. S. Skolnick, and M. Hopkinson, *J. Appl. Phys.* **86**, 2555 (1999).
- [26] M. Bissiri, G. Baldassarri Höger von Högersthal, A. S. Bhatti, M. Capizzi, A. Frova, P. Frigeri, and S. Franchi, *Phys. Rev. B* **62**, 4642 (2000).
- [27] R. Heitz, O. Stier, I. Mukhametzhanov, A. Madukar, and D. Bimberg, *Phys. Rev. B* **62**, 11017 (2000).
- [28] K. H. Schmidt, G. Medeiros-Ribeiro, M. Oestreich, P. M. Petroff, and G. H. Döhler, *Phys. Rev. B* **54**, 11346 (1996).
- [29] M. Bissiri, G. Baldassarri Höger von Högersthal, M. Capizzi, P. Frigeri, and S. Franchi, *Phys. Rev. B* **64**, 2445337 (2001).
- [30] F. Adler, M. Geiger, A. Bauknecht, D. Haase, P. Ernst, A. Dömen, F. Scholz, and H. Schweizer, *J. Appl. Phys.* **83**, 1631 (1998).
- [31] R. Heitz, M. Grundmann, N. N. Ledentsov, L. Eckey, M. Veit, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, P. S. Kop'ev, and Zh. I. Alferov, *Appl. Phys. Lett.* **68**, 361 (1996).
- [32] M. S. Skolnick and D. J. Mowbray, *Annu. Rev. Mater. Res.* **34**, 181 (2004).
- [33] S. Fafard, S. Raymond, G. Wang, R. Leon, D. Leonard, S. Charbonneau, J. L. Merz, P. M. Petroff, and J. E. Bowers, *Surf. Sci.* **361/362**, 778 (1996).
- [34] Z. Y. Xu, Z. D. Lu, X. P. Yang, Z. L. Yuan, B. Z. Zheng, J. Z. Xu, W. K. Ge, Y. Wang, J. Wang, and L. L. Chang, *Phys. Rev. B* **54**, 11528 (1996).
- [35] L. Brusaferrri, S. Sanguinetti, E. Grilli, M. Guzzi, A. Bignazzi, F. Bogani, L. Carraresi, M. Colocci, A. Bosacchi, P. Frigeri, and S. Franchi, *Appl. Phys. Lett.* **69**, 3354 (1996).
- [36] Y. T. Dai, J. C. Fan, Y. F. Chen, R. M. Lin, S. C. Lee, and H. H. Lin, *J. Appl. Phys.* **82**, 4489 (1997).
- [37] Z. Y. Xu, Z. D. Lu, Z. L. Yuan, X. P. Yang, B. Z. Zheng, J. Z. Xu, W. K. Ge, Y. Wang, J. Wang, and L. L. Chang, *Superlattices Microstruct.* **23**, 381 (1998).
- [38] S. Sanguinetti, M. Henini, M. G. Alessi, M. Capizzi, P. Frigeri, and S. Franchi, *Phys. Rev. B* **60**, 8276 (1999).
- [39] A. Polimeni, A. Patane, M. Henini, L. Eaves, and P. C. Main, *Phys. Rev. B* **59**, 5064 (1999).
- [40] W. Yang, R. R. Lowe-Webb, H. Lee, and P. C. Sercel, *Phys. Rev. B* **56**, 13314 (1997).
- [41] H. Y. Liu, B. Xu, Q. Gong, D. Ding, F. Q. Liu, Y. H. Chen, W. H. Jiang, X. L. Ye, Y. F. Li, Z. Z. Sun, J. F. Zhang, J. B. Liang, and Z. G. Wang, *J. Cryst. Growth* **210**, 451 (2000).
- [42] R. Heitz, I. Mukhameetzhano, A. Madukar, A. Hoffmann, and D. Bimberg, *J. Electron. Mater.* **28**, 520 (1999).
- [43] I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, *J. Appl. Phys.* **89**, 5815 (2001).
- [44] H. Lee, W. Yang, and P. Sercel, *Phys. Rev. B* **55**, 9757 (1997).
- [45] S. Sanguinetti, M. Henini, M. Gyassi Alessi, M. Capezzi, P. Frigeri, and S. Franchi, *Phys. Rev. B* **60**, 8276 (1999).
- [46] D. P. Popescu, P. G. Eliseev, A. Stinz, and K. J. Malloy, *Semicond. Sci. Technol.* **19**, 33 (2004).
- [47] F. V. de Sates, J. M. R. Cruz, S. W. de Silva, M. A. G. Soler, P. C. Morais, M. J. da Silva, A. A. Quivy, and J. R. Leite, *J. Appl. Phys.* **94**, 1787 (2003).
- [48] P. Dawson, O. Rubel, S. D. Baranovski, K. Pierz, P. Thomas, and E. O. Göbel, *Phys. Rev. B* **72**, 235301 (2005).
- [49] J. A. Barker and E. P. O'Reilly, *Phys. Rev. B* **61**, 13840 (2000).
- [50] P. Dawson, Z. Ma, K. Pierz, and E. O. Göbel, *Appl. Phys. Lett.* **81**, 2349 (2002).
- [51] T. S. Shamirzaev, A. M. Gilinksny, A. K. Bakarov, A. I. Toporov, D. A. Tenne, K. S. Zhuravlev, C. von Borczyskowski, and D. R. T. Zahn, *JETP Lett.* **77**, 389 (2003).
- [52] P. Offermans, P. M. Koenraad, J. H. Wolter, M. Roy, and P. A. Maksym, *Phys. Rev. B* **72**, 165332 (2005).
- [53] P. Dawson, E. O. Göbel, and K. Pierz, *J. Appl. Phys.* **97**, 013541 (2005).
- [54] A. J. Williamson, A. Franceschetti, H. Fu, L. W. Wang, and A. Zunger, *J. Electron. Mater.* **28**, 414 (1999).
- [55] H. W. Van Kesteran, E. C. Cosman, P. Dawson, K. J. Moore, and C. T. Foxon, *Phys. Rev. B* **39**, 13426 (1989).