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Prediction of the Properties of Annealed InAs/GaAs Quantum Dot Samples Assuming Conservation of Indium Atoms

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ABSTRACT

Annealing is known to induce In/Ga interdiffusion and intermixing in InAs/GaAs quantum dots. It results in a blueshift of the emission energy and a decrease in the intersublevel spacing. We claim that the conservation of Indium within each dot leads to a very tight link between these two effects. To illustrate this, we use a simple 3D harmonic oscillator model to describe the energy levels in the dots. We can then derive analytical expressions linking the optical properties, the structural parameters and the Indium content of each dot. Indium conservation then leads to an almost linear dependence between the emission energy and the intersublevel spacing. These predictions fit very well with published results. We furthermore show that the same arguments can be used to understand changes in the inhomogeneous broadening upon annealing. Annealing is often cited as a method of tuning the energy emission or the intersublevel spacing in quantum dot devices. By linking theoretically the properties of annealed samples to the as-grown sample, annealing a sample can also be a useful probe to determine its structural properties.

INTRODUCTION

Much progress has been made over the last few years in controlling of the growth of self-assembled Quantum Dots **(**QDs**)**. A careful choice of growth parameters enables good control of the density, size and shape and consequently the optical properties. One of the main issues is also to improve the uniformity of the QDs to achieve narrow inhomogeneous broadening. Post-growth Rapid Thermal Annealing **(**RTA**)** has also proved to be an effective way to change the properties of the QDs. It has been shown by several groups that this treatment results in a blueshift of the interband transition energies. There is also a concomitant decrease in the interlevel spacing and a narrowing of the photoluminescence **(**PL**)** inhomogeneous broadening [1-5]. The anneals are thought to induce indium/gallium interdiffusion within each dot, which results in size and compositional changes. In previous articles, the emphasis was put on the possibility of using RTA to tune the energy levels to a desired value. In this paper we present a simple model which allows us to understand the changes in the electronic structure of InAs QDs arising from annealing treatments. We show that the observed changes in the optical properties are a direct consequence of the conservation of Indium atoms within a dot during annealing.

EXPERIMENTAL DETAILS

The sample was grown using conventional solid source MBE but with the InAs layer deposited at a low-growth rate. This mode of growth [6] results in emission at 1.3 μm at room temperature **(**1.2 μm at 10K**)**. PL spectra were obtained at 10K using an Argon ion laser and

recorded with a 0.5 m grating monochromator and cooled Ge diode using standard lock-in techniques. Under high laser excitation, the spectrum of the as-grown sample exhibits ground state **(**GS**)** emission at 1.047 eV and emission from at least 3 excited states. The spacing between the transitions is constant at around 68 meV. The Full Width at Half Maximum **(**FWHM**)** of the GS is 24 meV. Several pieces of this sample were capped with 100 nm of SiO_2 and subjected to 10 s anneals in an RTA at temperatures ranging from 650ºC to 780ºC. After annealing all the samples exhibit emission from several excited states with a constant intersublevel spacing. Figure 1 shows the emission obtained at low excitation for all the samples. The inset shows the emission from two of the annealed samples at high excitation and demonstrates clear level filling indicating that the dot characteristics are retained after annealing. The symbols in Figure 2 plot the intersublevel spacing for different ground state emission energies for all the samples studied here and others derived by annealing the same as-grown sample. The plot is almost linear suggesting a strong correlation between size and indium concentration in the dots.

RESULTS

In order to use the argument of conservation of indium, we need to relate the indium content of a dot to its structural and optical characteristics. The main structural parameters are the size, shape, composition and strain within the dot. It is relatively easy to link the indium content to these parameters, however, calculating the optical characteristics from the structural ones is a more complicated problem. Several studies have attempted to do this using more or less complicated models for pyramidal [7-10] or lens-shaped [11] dots. These models require a large

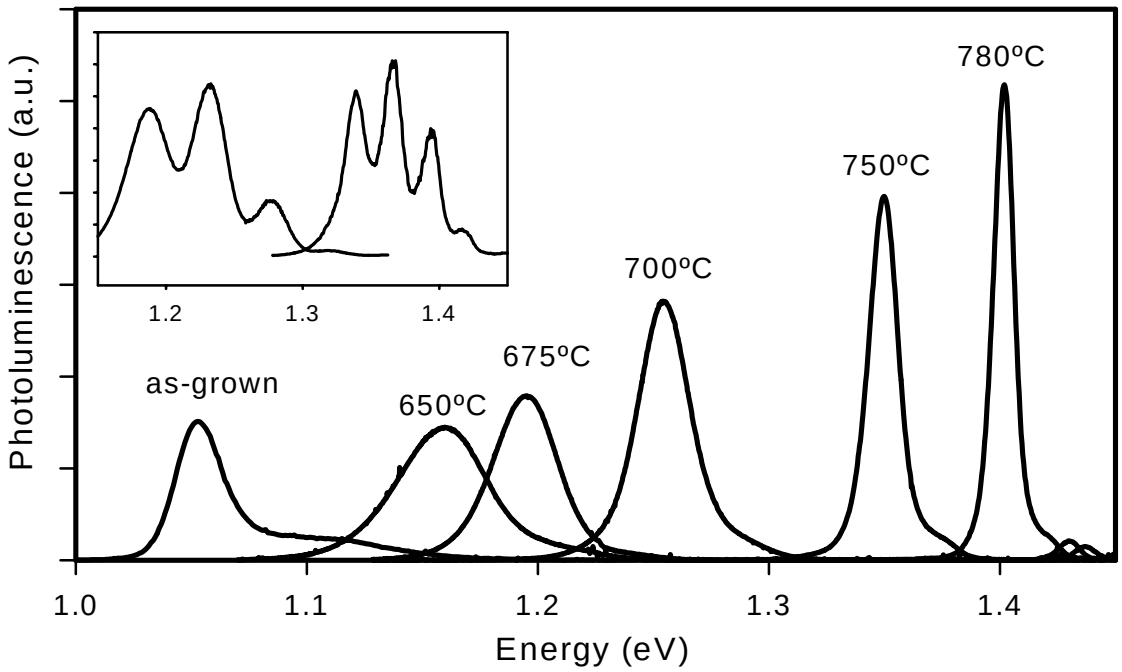

Figure 1: *PL spectra at 10K for as-grown and annealed samples under low excitation. The inset shows the high excitation spectra for samples annealed at 675ºC and 750ºC to demonstrate the presence of the excited states.*

number of parameters many of which are not very well known. It is this reason that has prompted us to adopt a much simpler model in order to emphasize the physical ideas of our indium conservation argument. It may be argued that the model is in some ways less realistic, but as we shall see, it allows us to understand the main features of the dots using only a few parameters. Moreover, most of the calculations can then be performed analytically and the predictions fit well with experiments. We believe applying a more complicated model would not change the conclusions at this stage.

Presentation of the model:

Based on the observation of equally spaced optical transitions, we have opted for a one particle model in a 3D Harmonic Oscillator Potential, which is our starting point for the description of the dot. The potential at each point only depends on the indium composition and the strain within each dot. For simplicity, we have only considered two cases: unstrained dots or fully strained dots. The difference is then only seen in the relation between the band gap and indium composition and in the value of the effective mass of the electrons [12]. To achieve a parabolic potential, the indium concentration must then increase gradually from the barrier to the center of the dot. This is not a contentious issue as several studies have already presented

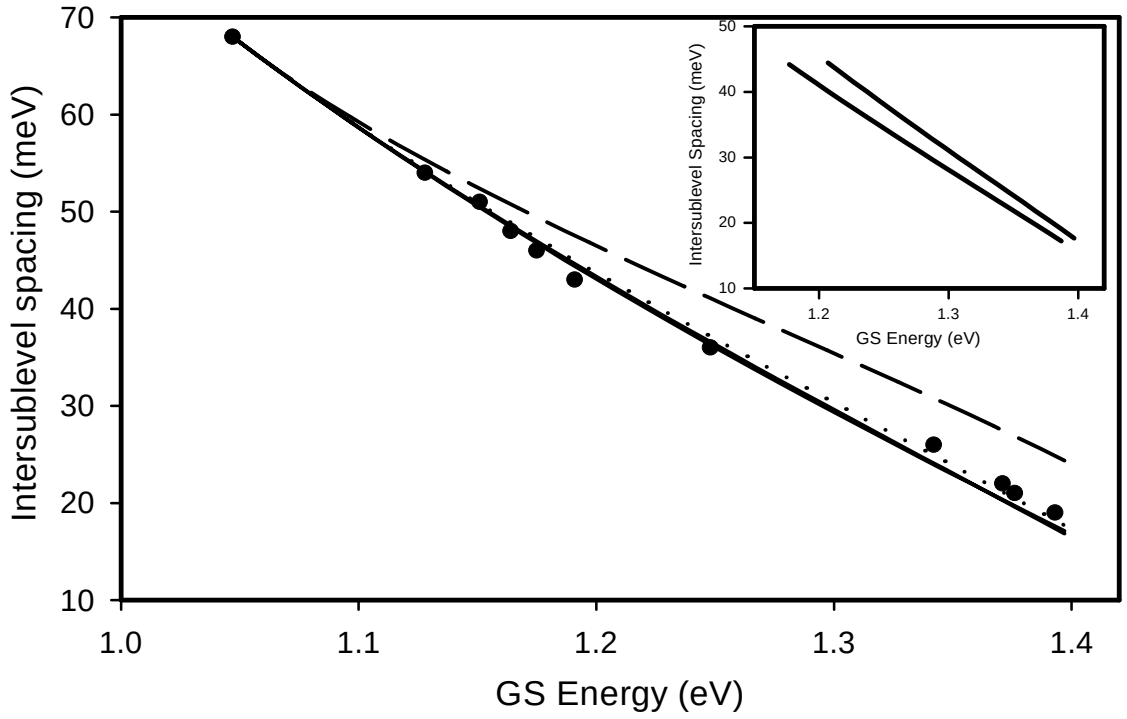

Figure 2: *Plot of the intersublevel spacing as a function of GS energy for as-grown and annealed samples. The symbols represent the experimental results obtained by annealing pieces of the same as-grown sample at different temperatures. The lines show the theoretical prediction assuming indium conservation and: constant radius (dashed),* d_z/d_x^2 = constant *(dotted), constant aspect ratio or constant height (solid lines, nearly indistinguishable). The dots are assumed to be unstrained. The inset shows the predictions for two dots on either side of the dot distribution showing the narrowing of the FWHM.*

evidence of non uniform composition and alloying within the dots [13,14]. The description of the details of the model will be the subject of another paper, and we will therefore concentrate on its predictions. The main feature is that we can calculate all the parameters of the dots (for example, radius d_x, height $2d_z$, composition at the center c_0, ground state energy E_0, intersublevel spacing δ, indium content N_{In}, etc…) using only three independent parameters. All the other parameters can be evaluated by calculating the electronic levels from the confinement potential, relating the potential to indium concentration, calculating the indium content from the concentration profile, etc… For example, when the relation between band gap E_g and indium concentration c (%) is linearised ($E_g(c) = a - b\,c$), the number of indium atoms per dot is:

$$N_{In} = \frac{32\pi {d_x}^2 d_z}{15 b \Omega_L} V_b \qquad (1)$$

where V_b is the barrier height ($V_b = a - E_0$), $\Omega_L = {d_L}^3$ is the volume of an elementary cell of the lattice, a is the barrier material band gap (a=1.5 eV), b=1.30 eV in the unstrained case and b=0.74 eV in the fully strained case. Knowing the density of dots, D, this number can be expressed in terms of monolayer (ML) of InAs:

$$N_{ML} = \frac{16}{15 b} D\pi {d_x}^2 \frac{d_z}{d_L} V_b \qquad (2)$$

For the as-grown sample, we can measure two parameters: GS energy E_0=1.047 eV and intersublevel spacing δ=68 meV. Therefore, we have only one remaining free parameter which can be varied to describe the sample. We have chosen to take the aspect ratio κ as our free parameter because this choice simplifies the calculations. The results presented next are virtually the same whatever value we choose for κ within a reasonable range ($4<\kappa<10$). We will therefore present the results for κ=6.7. We choose this value because the calculated properties of the as-grown sample are then very close to the structural measurements made by AFM. We derive a radius of 24 nm with a height of 7.2 nm. In the unstrained case, the indium concentration at the center is 70% and the total indium content in a dot is equivalent to 1.24 ML. In the strained case, these numbers are changed to 100% and 2.09 ML. These numbers seem consistent, although the total indium content may be overestimated. A more sophisticated model would probably give a more realistic absolute value. Here, we intend to apply an indium conservation argument and the absolute value is not so important.

Consequences of Indium conservation on the properties of the annealed samples:

Several studies have suggested that the effect of the annealing is to trigger interdiffusion of In/Ga atoms in the dots. This results in an increase in the size and a decrease in the indium composition at the center. The fact that the states remain equally spaced after annealing indicates that the structures retain their dot-like characteristics and may be described by the same model. The main argument we use is that the indium atoms remain part of the same dot. If there was transfer of indium between dots we would expect them to coalesce and form a quasi-quantum well, which is inconsistent with the observation of level filling (see inset Figure 1).

Using the preceding parameters to describe the as-grown sample, we can now predict some properties of the annealed samples. First each annealed sample is characterized by its GS energy E_0. To compare the predictions with the observations, we will use the model to predict the intersublevel spacing δ as a function of E_0. The conservation of indium after annealing will impose a relation between the parameters of the annealed samples, reducing the number of free parameters to 2. To obtain δ as a function of E_0, we therefore need to make some assumptions about our free parameter κ and its behavior during annealing. It is not really known whether indium interdiffuses preferentially along the growth direction, or in the lateral directions, or equally in both. To span these possibilities, we tried 4 assumptions:

- Constant height: d_z = constant
- Constant radius: d_x = constant
- Constant aspect ratio: κ = constant
- Intermediate case: d_z/d_x^2 = constant (diffusion favored in the lateral direction)

The last two cases are probably more realistic than the first two extreme cases. The lines in Figure 2 show the predicted intersublevel spacing after annealing for each of the four cases for unstrained dots. The results obtained for strained dots are nearly identical in terms of the optical properties and only the predicted concentrations of indium are different. Apart from the case of constant radius, which is probably the least realistic, we see that the three other cases lead to nearly identical predictions. Moreover, these predictions fit very well with the experimental data. This shows that the indium conservation condition alone dictates a very strong link between the intersublevel spacing and the GS energy. We believe that other models should lead to the same conclusion. The slight differences between theory and experiment observed for high annealing temperature can be explained by a gradual loss of indium from the dots towards the wetting layer. Because different hypotheses lead to the same results, it is not possible to deduce from this analysis whether strain has a significant effect or not, and whether the annealing occurs at constant aspect ratio, constant height, or a combination of the two. However, trends and even quantitative predictions can be made concerning the evolution of the radius d_x, the height $2d_z$, and the concentration at the center c_0. To illustrate this, these three predicted characteristics are plotted in Figure 3 as a function of GS energy for three different assumptions. We see that the lateral size of the dots increases, due to diffusion of indium, and can be almost doubled for the strongest anneal. Meanwhile, the concentration at the center decreases from 70% to 20% **(**unstrained**)** or from 100% to 30% **(**strained**)**. This shows that there is a transfer of indium atoms away from the center of the dots due to diffusion. These numbers can also be used to study the actual physical processes of intermixing and their temperature dependence.

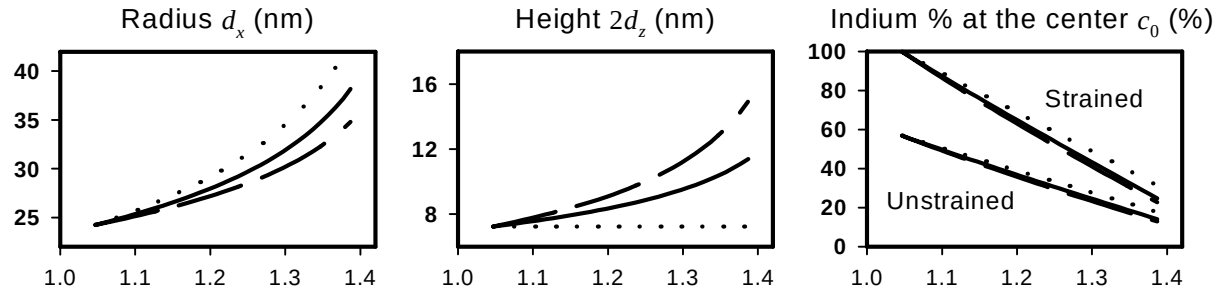

Figure 3: *Prediction of the structural properties of the annealed samples assuming: a constant height (dotted lines), a constant aspect ratio (solid lines) or* d_z/d_x^2 = constant *(dashed lines).*

Finally, these arguments can also be used to study the inhomogeneous broadening of annealed samples. For samples annealed at temperatures higher than 675ºC, we observe that the FWHM of the GS is equal to the FWHM of the excited states. This shows that the intersublevel spacing in these samples is uniform throughout the distribution. Starting with two dots having different GS energy on either side of the average GS energy of the sample annealed at 675ºC, we can predict the evolution of the optical properties for samples annealed at higher temperature. The predicted curves are shown in the inset of Figure 2. We see on this plot that the decrease of the FWHM for high temperature annealing is also a consequence of indium conservation. Looking in more detail at the predicted characteristics, we can show that this decrease is mainly due to the decrease of the barrier height (induced by the blueshift). The fluctuations have less effect on the broadening for a shallow potential than for a deeper potential. A more detailed study of the FWHM in annealed samples will be presented elsewhere.

CONCLUSION

Using simple arguments of indium conservation, we have shown that the effects of annealing are largely predictable. The observed linear dependence of the ground state energy with the intersublevel spacing is a direct consequence of Indium conservation within each dot during annealing. We used a very simple model, which gives good qualitative and even quantitative results. These arguments and the predictions obtained for the dot characteristics can also help to understand the physical process of indium diffusion and intermixing.

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