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Scalable Routes to Single and Entangled Photon Pair Sources - Tailored InAs/InP Quantum Dots in Photonic Crystal Microcavities.

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ABSTRACT

Optoelectronic devices based on single, self-assembled semiconductor quantum dots are attractive for applications in secure optical communications, quantum computation and sensing. In this paper we show how it is possible to dictate the nucleation site of individual InAs/InP quantum dots using a directed self-assembly process, to control the electronic structure of the nucleated dots and also how to control their coupling to the optical field by locating them within the high field region of a photonic crystal nanocavity. For application within fiber networks, these quantum dots are targeted to emit in the spectral region around 1550 nm.

Keywords: quantum dots, photonic crystals, selective-area epitaxy

1. INTRODUCTION

Efficient sources of non-classical light, single photons and entangled photon pairs etc. are required for applications in quantum information processing and quantum key distribution as well as for fundamental experiments that test the foundations of quantum mechanics.¹⁻³ Many successful single photon sources are based upon the concept of a single, two-level system coupled to the optical density of states of a photonic cavity. It has been recognized since the work of Purcell in the 1940s⁴ that the characteristic decay rate of an excited dipole emitter is not an immutable property of the emitter, but is a function of its local electromagnetic environment or equivalently the local density of optical states. In a closed cavity environment, the Purcell factor, F_p , giving the ratio of the spontaneous emission rate within the cavity to the rate within vacuum, can be expressed as⁵

$$\frac{\tau}{\tau_c} = \frac{3Q(\lambda_c/n)^3}{4\pi^2 V_{eff}} \frac{\delta\omega_c^2}{4(\omega_e - \omega_c)^2 + \delta\omega_c^2} \frac{|\mathbf{d} \cdot \mathbf{f}(\mathbf{r}_e)|^2}{|\mathbf{d}|^2} \quad (1)$$

where Q is the cavity quality factor, V_{eff} the effective optical mode volume, λ_c the wavelength and n the refractive index. The second term in this expression, involving the emitter frequency, ω_e , and the cavity mode frequency, ω_c , highlights the need to spectrally match the emitter frequency to the cavity mode frequency if one wishes to take advantage of any increased decay rate that might be afforded by an increased optical density of states at the emitter, whilst the third term involving the emitter's transition dipole moment, $\mathbf{d}$, and the cavity mode's electric field distribution, $\mathbf{f}(\mathbf{r}_e)$, highlights the requirement to locate the emitter at a field antinode with an appropriate polarization.

In a solid state environment, self-assembled semiconductor quantum dots⁶ are viewed as promising single photon emitters, since their discrete, atom-like density of states can be tuned to various parts of the optical spectrum through choice of composition, size and material environment. In addition, such dots can be embedded within high dielectric contrast nanocavities such as the planar membrane cavity⁷ shown in Fig. 1. In such structures, a periodic lattice of etched through holes in the membrane provides strong dielectric contrast and a complete photonic bandgap for lateral propagation, while total internal reflection provides confinement in the vertical direction. A single missing hole in such structures is sufficient to provide a localized photonic mode that can be spatially and spectrally matched to a carefully positioned single quantum dot. The quality factor and far

Figure 1. Schematic illustration of an InP planar photonic crystal nanocavity membrane containing a single InAs/InP quantum dot.

field emission pattern, as represented by the cone of emission in Fig. 1, of these structures can be tailored by carefully choosing the size and location of the air holes surrounding the optical defect.

In practice it is extremely difficult to control the location of individual self-assembled quantum dots in a manner that provides sufficient accuracy for alignment with highly localized optical modes, whilst still maintaining the high optical quality of the dot. In this paper we discuss how this alignment can be achieved using a directed self-assembly technique based upon the control of surface diffusing indium species during the chemical beam epitaxial growth of InAs/InP quantum dots⁸⁻¹⁰ and we give examples of how this technique can be used to embed quantum dot emitters within planar photonic crystal nanocavity membranes.

2. DIRECTED SELF-ASSEMBLY

Pyramidal nanotemplates, such as that shown in Fig. 2, are prepared in ultra high vacuum by initiating the chemical beam epitaxy of InP on an (001) oriented InP wafer that is patterned with openings in an otherwise continuous SiO₂ mask. Under these circumstances, InP growth only occurs within the oxide openings, on regions of exposed InP substrate. For square windows, with edges aligned along the $\langle 100 \rangle$ crystal directions, square based pyramidal InP nanotemplates are produced with sloping $\{011\}$ side facets and an (001) top surface that can be used for quantum dot positioning. As a function of increasing deposition time, or equivalently quantity of InP deposited, the height of the InP nanotemplate increases, whilst the (001) top surface area available for quantum dot nucleation diminishes.

For a thickness h_o of InP deposited on a planar substrate, the pyramidal nanotemplate height h and top width w are given by Eqs. 2-5, where we have assumed that a fraction β of the indium species impinging on the $\{011\}$ sidewalls is transferred to the (001) apex, whilst the remaining fraction $(1 - \beta)$ is desorbed from the surface. In Eqs. 2-5, w_o represents the initial oxide window width and θ is the angle of the $\{011\}$ sidewalls to the horizontal. Fig. 3(a) shows the calculated variation of the pyramid top width as a function of the quantity of deposited InP for a 500 nm wide oxide window and a sidewall capture fraction of 90%. As the pyramidal template nears completion, one can see that the rate at which the (001) top width diminishes is increasing rapidly. To locate a single InAs quantum dot at the apex of the pyramidal template, growth is continued until the top width is slightly larger than the width of a typical dot, approximately 30-40 nm, at which point one stops growth of InP and initiates growth of InAs. Deposited indium adatoms migrate away from the $\{011\}$ sidewalls and accumulate on the (001) top surface, where a single InAs quantum dot is nucleated. Under appropriate conditions, the quantum dot that is nucleated at the pyramid apex will conform to the shape of the apex itself. This behavior has two consequences; on the one hand it suggests that the electronic properties of individual dots can be tuned by controlling their size and shape, but on the other hand, considering the behavior indicated in Fig. 3(a), it suggests that the size and shape of the pyramidal template must be tightly controlled if one wishes to achieve reproducible quantum dot behavior.

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Figure 2. Schematic illustration (top) and scanning electron micrograph (bottom) of a single InAs/InP quantum dot nucleated at the apex of a pyramidal InP nanotemplate.

$$h = \frac{w_o \tan \theta}{2} + \frac{(4R)^{1/3}}{4\beta} - \frac{(1-\beta)w_o^2 \tan^2 \theta}{(4R)^{1/3}} \quad (2)$$

$$R = \beta^{2/3} w_o^2 \tan^2 \theta [\beta^{1/2} (w_o \tan \theta (2\beta - 3) + 6h_o) + F^{1/2}] \quad (3)$$

$$F = w_o^2 \tan^2 \theta (4 - 3\beta) + 12h_o w_o \beta \tan \theta (2\beta - 3) + 36\beta h_o^2 \quad (4)$$

$$w = w_o - \frac{2h}{\tan \theta} \quad (5)$$

In a single growth run, one is able to produce single dots on a number of single pyramidal templates where each dot is tuned to a different emission energy. As can be seen from Eqs. 2-5, for the same planar quantity of InP deposited, h_o , the final top width of the pyramidal template will vary if the initial oxide window width is varied. This behavior is shown in Fig. 3(b) for a set of individual pyramidal templates where the initial width is varied between 470 nm and 490 nm. As a function of increasing template width, the emission energy decreases,¹¹ at a rate of approximately 4.2 meV/nm and, for these growth conditions, produces emission at 800 meV, $\lambda = 1550$ nm, for a template base width of 485 nm. Clearly then, by choosing the base width and growth conditions appropriately, it is possible to tune the emission wavelength of individual, site-selected InAs/InP quantum dots close to the wavelengths suitable for fiber communications and close to the mode frequency for the photonic crystal cavities discussed below.

Figure 3. (a) Pyramid top width, w , as a function of planar InP thickness, h_o , for an initial oxide window width of 500 nm and a sidewall capture fraction, β , of 0.9. (b) Dependence of the emission energy of single, site-selected InAs/InP quantum dots on the pyramidal nanotemplate base width.

3. PHOTONIC CRYSTAL CAVITIES

To incorporate single pyramidal templates within a photonic crystal membrane as shown schematically in Fig. 4(a), it is necessary to modify the growth procedure that was discussed above for growing site-selected individual dots. Samples intended for subsequent cavity fabrication are grown on substrates in which an InAlAs sacrificial layer, approximately $1\mu\text{m}$ thick, is included below a thin surface InP layer. InAlAs is chosen as a sacrificial layer because it produces virtually no optical emission in the wavelength region around 1550 nm, so no interference is expected with the very weak optical signature from a single dot emitting at 1550 nm and sitting on top of a large thickness of InAlAs. This feature means that it is possible to characterize individual quantum dots in emission immediately prior to cavity fabrication, before the InAlAs sacrificial layer has been removed, and to then spectrally match the cavity resonance to the quantum dot emission. To begin fabrication, the starting structure is masked with SiO_2 and square windows for pyramidal template growth are opened up in exactly the way described in Section II above, except that these windows are now accurately aligned with respect to electron beam alignment marks that are etched into the substrate. Single dot nucleation is produced in the manner described in section II and the aligned dots are capped with a small amount of InP, to complete the pyramidal template, before the whole structure is removed from the growth chamber. At this stage the dots are already completely encapsulated within an InP matrix and consequently protected from the environment. The samples are now re-patterned to open up large windows in the SiO_2 , centered on the existing pyramids, before being re-introduced into the growth chamber for further epitaxial InP growth. In this second growth step, InP is again grown only on the regions of exposed semiconductor, within the large SiO_2 windows. Growth of InP is continued until the pyramidal template is buried within a planarised membrane whose thickness is appropriate for photonic crystal fabrication, typically 200-300 nm and whose lateral dimension, set by the SiO_2 window dimension, is large enough, typically 50–100 μm , to accommodate enough rings of etched photonic crystal holes that the optical loss from the cavity is dominated by vertical loss and not by leakage in the plane. Fabrication of the photonic crystal structure is achieved by depositing a layer of SiO_2 and then a layer of electron beam resist across the planarised dot structure. The electron beam resist is exposed (Fig. 4(d)) using the alignment marks already etched into the substrate and this is used to etch a hole pattern into the oxide layer. The patterned oxide layer is then used as a mask to transfer the desired hole pattern into the InP membrane, which is dry etched using a CH_4 , Cl_2 , H_2 chemistry into the underlying InAlAs layer. The InAlAs layer is now removed by simple wet chemistry to leave the suspended InP photonic crystal nanocavity membrane with the single pyramidal InP template and single InAs quantum dot precisely located at the optical defect position. An SEM image of a completed structure,

Figure 4. (a) Schematic illustration of a single InP pyramidal nanotemplate aligned within the optical defect of a photonic crystal membrane. (b) Schematic illustration of the hole pattern used to produce high Q , small mode volume cavities in InP membranes. Additional holes are added to the basic single missing hole design along the x -axis and moved towards the cavity centre, while nearest neighbor radial holes are shrunk and moved outward. (c, d) Scanning electron micrograph images of a single InP pyramidal nanotemplate aligned within the optical defect of a photonic crystal membrane. In (d), the photonic crystal cavity is produced only in the electron beam resist, so that alignment to the pyramidal template can be demonstrated.

cleaved through the cavity center is shown in Fig. 4(c).

The photonic crystal cavities targeted in the work discussed here are single missing hole, hexagonal symmetry structures in which the modes of interest are predominantly TE-like modes that couple strongly to the in-plane transition dipole of a single quantum dot. The simplest structure, where the optical defect is produced by removing a single hole, and no further modifications are present, produces two energetically degenerate dipole modes with modest Q values in the range from 100 to 1,000. Removal of the dipole mode degeneracy and large increases in Q , accompanied by small mode volumes of the order of $0.5(\lambda/n)^3$, can be achieved using designs such as that shown in Fig. 4(b),¹² where additional holes are added along the x -axis and moved toward the cavity center by a distance δx , while the off-axis holes in the first ring are reduced in size and moved radially outward by a distance δr . We have produced such structures with Q values in excess of 30,000 and with Gaussian emission modes normal to the plane, ideally suited for coupling into single mode fiber. However, the fabrication of such structures is difficult to optimize, since accuracies of the order of 10 nm are required for the shifts δx , δr if the highest Q values are to be realized.

Once fabricated, the cavity mode energy and quantum dot transition energy are invariably mismatched, however carefully the cavity fabrication is performed. In consequence, it is desirable to be able to tune the mode energy of the cavities after they are fabricated. This is achieved by repetitive oxidation and wet chemical stripping of the cavities.¹³ The oxidation process consumes a small quantity of InP, approximately 6-7 Å per cycle, from the top and bottom surfaces of the membrane and also from the inner surface of the etched holes. The process is self-limiting in that once a certain thickness of oxide is produced, the oxidation stops. The increase in

Figure 5. (a) Luminescence from a photonic crystal nanocavity incorporating a single, site-selected InAs/InP quantum dot after successive oxidation cycles, 28-39. The cavity mode (CM) can be seen coming in at long wavelength and tuning to shorter wavelength with successive oxidation steps. The quantum dot (QD) and cavity mode are brought into resonance at etch step 38. Traces are systematically offset along the wavelength axis for clarity. (b) Cavity mode energy and integrated luminescence intensity as a function of dot-cavity detuning.

hole radius and change in thickness of the InP membrane after every oxidation cycle produces an increase in the cavity mode energy and allows one to match the mode frequency to the quantum dot transition frequency. This can be seen in Fig. 5, which shows luminescence from a cavity incorporating a spatially aligned single InAs/InP quantum dot emitting close to 1475 nm. After successive oxidation cycles the cavity mode (CM) is observed to tune to shorter wavelength, coming into resonance with the quantum dot emission after 38 oxidation cycles. On resonance, the luminescence collected from the quantum dot increases substantially, in agreement with the predictions of Eq. 1 above.

The above tuning process is irreversible and the energy shift per cycle is large if one would like to spectrally match emitters and cavities with sub-meV linewidths. Condensation of inert gases on the photonic crystal surface¹⁴ provides dynamical tuning of the cavity mode with arbitrarily small step size dictated by volume of gas introduced into the cryostat per cycle. Fig. 6(a) shows a typical tuning curve for a cavity emitting at 1575 nm where 100 Torr of N₂ in a ~ 0.5 L volume is introduced into the cryostat per cycle. The shift is initially linear at ~ 400 μ eV per cycle, then saturates after ~ 6 nm, at which point the crystal surface is likely covered by one monolayer of nitrogen.

The process is reversible, the sample simply has to be warmed up, and the cavity shifts are to lower energy as opposed to the blue-shift from oxidation. The tuning mechanisms can therefore be used together: oxidation for rough tuning through resonance to the high energy side of the dot, and gas condensation for fine tuning back into resonance. This procedure was performed on the sample in Fig. 5 using 39 oxidation cycles to tune the cavity through resonance from lower energy and 30 N₂ condensation cycles to tune back through resonance, equivalent to tuning back to oxidation cycle 37. The spectra from the gas tuning are shown in Fig. 6(b) as a contour plot versus detuning ($\omega_c - \omega_e$) for detunings corresponding to oxidation cycles 37 to 39. Spectra such as those shown in Fig. 6(b) elucidate the complex nature of coupled quantum dot-cavity systems.¹⁵

4. CONCLUSIONS

We have demonstrated a scalable technique for the spatial and spectral matching of single InAs/InP quantum dots to the high Q, spatially localized modes of a photonic crystal nanocavity membrane. These structures are expected to find application in the areas of quantum information processing and fiber based quantum key distribution.

Figure 6. (a) Fining tuning of a cavity mode using condensation of N₂. (b) N₂ tuning of a cavity through a single InAs/InP quantum dot.

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REFERENCES

- [1] E. KNILL, R. LAFLAMME and G. MILBURN , *Nature* **409**, 46 (2001).
- [2] A. BEVERATOS, R. BROURI, T. GACOIN, A. VILLING, J.-P. POIZAT, and P. GRANGIER , *Phys. Rev. Lett.* **89**, 187901 (2002).
- [3] P. HAWRYLAK and M. KORKUSIŃSKI, *Nonlinear Optics* **29**, 329 (2002).
- [4] E. M. PURCELL, *Phys. Rev.* **69**, 681 (1946).
- [5] L. C. ANDREANI, G. PANZARINI, and J. M. GÉRARD, *Phys. Rev. B* **60**, 13276 (1999).
- [6] I. N. STRANSKI and L. VON KRASTANOW, *Akad. Wiss. Lit. Mainz Math.-Naturwiss Kl. Iib* **146**, 797 (1939).
- [7] O. J. PAINTER, A. HUSAIN, A. SCHERER, J. D. O'BRIEN, I. KIM, and P. K. DAPKUS, *J. Lightwave Technol.* **17**, 2082 (1999).
- [8] R. WILLIAMS, G. AERS, P. POOLE, J. LEFEBVRE, D. CHITHRANI, and B. LAMONTAGNE, *J. Crystal Growth* **223**, 321 (2001).
- [9] J. LEFEBVRE, P. J. POOLE, J. FRASER, G. C. AERS, D. CHITHRANI, and R. L. WILLIAMS, *J. Crystal Growth* **234**, 391 (2002).
- [10] P. J. POOLE, G. C. AERS, A. KAM, D. DALACU, S. STUDENIKIN, and R. L. WILLIAMS, *J. Crystal Growth*. **310**, 1069 (2008).
- [11] D. DALACU, M. R. REIMER, S. FRÉDÉRIC, D. KIM, J. LAPOINTE, P. J. POOLE, G. C. AERS, R. L. WILLIAMS, W. R. MCKINNON, M. KORKUSIŃSKI and P. HAWRYLAK , *Laser & Photonics Rev.* DOI: 10.1002/lpor.200810077 (2009).
- [12] S. FRÉDÉRIC, D. DALACU, J. LAPOINTE, P. J. POOLE, G. C. AERS, and R. L. WILLIAMS, *Appl. Phys. Lett.* **89**, 191115 (2006).
- [13] D. DALACU, S. FRÉDÉRIC, P. J. POOLE, G. C. AERS, and R. L. WILLIAMS, *Appl. Phys. Lett.* **87**, 151107 (2005).
- [14] S. MOSORU, J. HENDRICKSON, B. C. RICHARDS, J. SWEET, G. KHITROVA, H. M. GIBBS, T. YOSHIE, A. SCHERER, O. B. SHCHEKIN, and D. G. DEPPE, *Appl. Phys. Lett.* **87**, 151107 (2005).
- [15] S. HUGHES and P. YAO, *Opt. Exp.* **17**, 3322 (2009).