High quality factor GaAs-based photonic crystal microcavities by epitaxial re-growth

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Abstract: We investigate L7 photonic crystal microcavities (PCMs) fabricated by epitaxial re-growth of GaAs pre-patterned substrates, containing InAs quantum dots. The resulting PCMs show hexagonal shaped nano-holes due to the development of preferential crystallographic facets during the re-growth step. Through a careful control of the fabrication processes, we demonstrate that the photonic modes are preserved throughout the process. The quality factor ($Q$) of the photonic modes in the re-grown PCMs strongly depends on the relative orientation between photonic lattice and crystallographic directions. The optical modes of the re-grown PCMs preserve the linear polarization and, for the most favorable orientation, a 36% of the $Q$ measured in PCMs fabricated by the conventional procedure is observed, exhibiting values up to ~6000. The results aim to the future integration of site-controlled QDs with high-$Q$ PCMs for quantum photonics and quantum integrated circuits.

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References and links


1. Introduction

Photonic crystal microcavities (PCMs) with embedded quantum dots (QDs) have been shown as excellent test bed systems for experiments in the field of cavity quantum electrodynamics (c-QED) [1,2] that may open doors to efficient quantum photonic devices. Single quantum dots embedded in a PCM become efficient quantum emitters which might be used for the generation of single-photons [3–5], entangled photon pairs [6], ultra-low threshold lasing [2], polariton lasing [2,7] or to explore new strong coupling phenomena [8–10]. Most c-QED applications require of a large optical quality factor ($Q$), a small electromagnetic mode volume ($V$), and a large coupling between the QD emission and the PCM mode [11]. Therefore, an accurate positioning of the QD within the PCM while maintaining a high $Q/V$ ratio is a critical requirement not easily attainable with self-assembled QD growth methods [12]. Several procedures have been proposed for the fabrication of site-controlled QDs [13,14] coupled to PCMs [15,16]. Among them, local oxidation lithography by atomic force microscopy (LOL-AFM) is a powerful technique for the patterning of GaAs substrates which is compatible with the epitaxial growth of site-controlled InAs QDs. LOL-AFM allows the positioning of high quality QDs in any place of a wafer in a deterministic way [17], and therefore is very promising for quantum photonic applications [15,16]. However, since the QD has to be embedded in the PCM slab underneath the surface, the development of a special re-growth procedure is needed to complete the photonic structure after the LOL-AFM step. The epitaxial re-growth of photonic structures [18] has been realized in distributed feedback (DFB) lasers and photonic crystal lasers by metal-organic chemical vapor deposition (MOCVD) [19,20] and photonic crystal surface emitting lasers (PCSELs) by metal-organic vapor phase epitaxy (MOVPE) [21]. More recently, PCMs have been fabricated by re-growth processes over patterned GaN wafers by nitride-MOCVD [22,23] showing photonic modes with $Q$ up to 2400 at a wavelength of 383 nm [24]. In addition, cavity modes with $Q = 1800$ at 426 nm have been shown in PCMs obtained by re-growth of pre-patterned L3-PCMs on slabs of AlN material with embedded GaN/AlN QDs [25].

In this work we demonstrate that PCMs fabricated by epitaxial re-growth of a previously patterned GaAs substrate exhibit optical modes with $Q$ factors comparable to those obtained by standard lithography and etching procedures. The studied PCMs contain self-assembled InAs QDs with emission wavelengths in the range 890-1000 nm and therefore, the developed method paves the way for the use of one or several LOL-AFM deterministically site-controlled QDs integrated with PCMs with emission in the 980 nm telecom window. We have modeled the optical properties of the re-grown PCMs, by means of finite difference time domain (FDTD) simulations, to evaluate the impact of the re-growth procedure, obtaining an explanation for the high $Q$-values found.
2. Design and fabrication

We have fabricated PCMs of the L7-type, which consists of a set of seven missing holes along the ΓK direction of an array of circular holes with triangular symmetry [26,27]. The separation of the holes (with radius r) is given by the lattice constant a, which corresponds to the distance between two neighboring holes along the ΓK direction. For such a triangular lattice, the air-filling factor (FF) is given by the ratio of the volume of one hole and that of a slab unit cell, \[ FF = \frac{2\pi \left( r \right)^2}{\sqrt{3} a}. \] The L7 PCM presents two important advantages: on one side, its elongated shape is useful for the positioning of nanostructures like quantum wires [28,29] or the use of several QDs sharing an optical mode [16]. On the other side, it provides a good Q/V ratio without the need of tuning the holes that surround the cavity, which increases its robustness to fabrication imperfections. The L7 PCMs have been fabricated in samples from two different wafers, named hereafter, W-I and W-II (Fig. 1). Both W-I and W-II wafers are grown by molecular beam epitaxy (MBE) and consist of a GaAs based active slab on top of an Al0.7Ga0.3As sacrificial layer 1 μm thick underneath. A high-density self-assembled InAs QDs (SAQD) was grown 70 nm over the Al0.7Ga0.3As/GaAs interface in both W-I and W-II. The main difference between W-I and W-II is the active GaAs slab thickness, 140 nm for W-I and 115 nm for W-II.

For the fabrication of the PCMs in the first wafer (W-I) we have used a standard procedure, based on electron beam lithography (EBL) and plasma ion etching. On top of the W-I wafer, a layer of ~80 nm of SiOx was deposited by plasma enhanced chemical vapor deposition (PECVD) at 300 °C as a hard mask. A ~360 nm thick layer of ZEP-520A was spun coated over the SiOx for the EBL patterning (30 kV). After EBL and a developing process, the patterns were transferred to the SiOx layer by CHF3/N2 reactive ion beam etching (RIBE) that provides excellent opening of the nanoholes in the SiOx [27]. Reactive ion etching (ICP-RIE) with a BCl3:N2 mixture was used to transfer the pattern to the GaAs active slab, which results in vertical and smooth holes [30,31]. Finally, the photonic crystal membrane is released by diluted HF wet etching of the Al0.7Ga0.3As sacrificial layer under the structures. We will refer to those PCMs as standard L7. In the W-II wafer, L7 PCMs were fabricated using the same procedure as in W-I followed by an epitaxial re-growth step to complete the slab thickness to 140 nm. After the membrane release and prior to the re-growth step, a careful cleaning process is performed in order to remove resist residuals and contaminants from the previous fabrication process. Once inside the MBE chamber, the native oxide is removed by exposure of the GaAs surface to atomic H that preserves the flatness of the surface [32] between the holes. For that, atomic hydrogen is supplied at substrate temperature of T3 = 450 °C during 30 minutes. The re-growth continues with the deposition of a GaAs layer by atomic layer molecular beam epitaxy (ALMBE) [33] at T3 = 450 °C to complete the 70 nm of GaAs over the SAQD layer. Two sets of L7-PCMs were fabricated in both W-I and W-II with the ΓK direction of the PCM either perpendicular (L7⊥) or parallel (L7∥) to the GaAs [110] crystallographic direction. Four lattice constants (a = 250, 260, 270 and 280 nm)
for the PCMs were fabricated to cover the luminescence of the QD emitters. For a given $a$, different $r/a$-values ranging 0.26-0.33 were practiced for a fine-tuning of the PCM optical modes with the QD emission. Figure 2 shows images taken by scanning electron microscopy (SEM) on the PCMs in W-I [Figs. 2(a) and 2(b)] and in W-II [Figs. 2(c)–2(f)].

The fabrication process in W-I results in holes with circular shape, vertical and smooth [30,31]. In W-II, the re-growth step induces a change from the initial circular shape of the holes towards an elongated hexagonal shape, due to the development of new crystallographic facets [34]. The long axis of the hexagon is aligned along the [110] crystalline direction. This direction corresponds to the intersection of $B$-type facets (As terminated) with the (001) surface plane; the short axis of the hexagon is aligned along [1–10] that corresponds to the intersection of $A$-type facets (Ga terminated) with the (001) surface plane. A lateral flux of Ga atoms towards $B$-type facets and away from $A$-type facets during the re-growth step can explain the observed hexagonal shape. Lateral flux of Ga atoms has been previously reported [34,35] and a similar evolution has been described during the initial overgrowth of nanoholes fabricated by EBL and dry etching [36] or by LOL-AFM and HF selective oxide etching on GaAs substrates [37]. The evolution of the shape of the holes may affect the photonic properties of the PCMs, so our task is to determine the influence of this effect in the photonic performance of the PCMs, verifying if photonic modes are still present and, in that case, evaluating the optical quality of the PCMs through the measurement of their $Q$.

3. Optical characterization

We have performed optical characterization by confocal microscopy at 4 K to measure the micro-photoluminescence ($\mu$PL) emitted from the PCMs. The excitation laser CW light at 785 nm was delivered through a single mode optical fiber to the microscope and focused onto the PCM within a diffraction limited optical spot. The light emitted by the sample was collected through a different single mode optical fiber, dispersed by a 750 mm focal length spectrometer and detected with a cooled Silicon Charge Coupled Device. The spectral...
linewidth of the resonances ($\Delta\lambda$) determines the $Q$-values ($Q = \lambda/\Delta\lambda$). We focus in the $Q$ of the fundamental mode (FM) since it provides the best $Q/V$ ratio [26]. Figure 3 shows the PL spectra of a representative set of L7-PCMs, one corresponding to a standard L7 in W-I [Fig. 3(a)] and the others to the re-grown L7-PCMs, i.e. L7⊥ [Fig. 3(b)] and L7∥ [Fig. 3(c)] in W-II.

![Micro - photoluminescence (μPL) spectra corresponding to a set of L7 photonic crystal microcavities (PCMs); (a) standard L7, (b) L7⊥ and (c) L7∥. Quality factor ($Q$) and spectral position ($\lambda$) of the fundamental mode (FM) are presented. Insets show the polarization diagrams for each of the observed L7-PCM modes.](image)

Both the L7⊥ and L7∥ re-grown PCMs present spectral features that resemble very well those observed in standard L7. This is already indicative of the good optical performance of the re-grown PCMs, so we will refer as photonic modes the observed features in the re-grown cavities. The results obtained from a statistical study of the spectral position of the observed modes in every set of PCMs, show a blueshift of 26 ± 6 nm for the FM of the L7⊥ and of 45 ± 13 nm in the L7∥ configurations with respect to the standard L7. We attribute the observed blueshifts to the two following reasons, that will be supported by simulations: 1) the cleaning process previous to the epitaxial re-growth that results in an isotropic removal of the GaAs oxide, which produces a slight enlargement of the radius of the holes and a decrease of the slab thickness [38], 2) the evolution of the circular shape of the holes to hexagonal during the re-growth step, which also enlarges the effective size of the holes. The standard L7 PCMs exhibit $Q$-values for the FM ranging from ∼5000 to ∼1000. In particular, Fig. 3(a) shows the photonic structure of a standard L7 PCM with $Q = 9333$. Figures 3(b) and 3(c) show $Q = 4169$ for L7⊥ and $Q = 1749$ for L7∥. The statistical analysis shows that in the re-grown L7 PCMs the $Q$ is preserved a 36% for L7⊥ and a 15% for L7∥ with respect to the standard L7 PCMs. It is found that the optical performance observed for the re-grown PCMs depends on the relative orientation between the crystallographic and the photonic crystal directions. This result could be explained as follows: in the L7⊥, the distance between first neighboring holes along [110] is $\sqrt{3}$ times longer than in L7∥. Since growth leads to nanoholes elongated along [110] direction, neighboring holes first collapse when ΓK is aligned along the [110] direction (L7∥). Therefore, a more robust optical performance (i.e., higher $Q$) is expected for L7⊥ than for L7∥. To further confirm the photonic modal structure of the PL spectra, we have analyzed the polarization properties of the observed resonances of the PCMs in W-I and W-II. It is well known that the Ln-type PCMs should present optical modes linearly polarized [26,27]. The
insets in Fig. 3 show the polarization polar plots for the first three PCM modes. These plots clearly show that the first three modes present linear polarization for both standard [Fig. 3(a)] and re-grown L7 [Figs. 3(b) and 3(c)] PCMs. Similar behavior results from the analysis of every set of PCMs, confirming the photonic modal structure of re-grown PCMs.

4. Numerical calculations

In order to evaluate the impact of the change of the hole shape on the optical properties, we have performed three dimensional FDTD (3D-FDTD) simulations [39] of the re-grown PCMs. We focus on the determination of the $Q$ and $\lambda$ of the FM of the standard and the re-grown L7 PCMs. We consider a dipole source with a narrow bandwidth for the excitation of the FM and a point monitor located in the antinode of the FM profile registers the time evolution of the mode. The Fast Fourier Transform (FFT) of the time evolution of the electric field amplitude determines the spectral position of the mode ($\lambda$). The $Q$ is obtained by analyzing the signal decay with the time. Further information related to FDTD simulations can be found in Ref [40]. We have modeled the re-grown hole as a vertical circular cylinder with radius $r_b$ on the bottom part that corresponds to the pre-pattern. The top part of the hole affected by the re-growth step is modeled as a truncated cone with a circular bottom base, with radius $r_b$, that evolves to an elliptical shape in the surface, with short and long axes, $r_e$ and $r_E$, respectively [Fig. 4(a) inset]. We have taken the $r_e$ and $r_E$ values from AFM measurements and the $r_b$ value from SEM images (circular base of standard L7 PCMs). In general, a reduction of the symmetry of the point lattice may affect the photonic performance of PCMs [41–43]. To evaluate the impact of the loss of symmetry due to the relative orientation of the holes with the crystallographic directions we calculate the $Q$-values of the re-grown PCMs while changing the angle ($\theta$) between $\Gamma K$ and the [110] crystallographic direction. Figure 4(a) shows the variation of $Q$ with $\theta$ for a PCM with air filling factor, $FF = 0.31$. Compared to those measured in the experiments, the values of $Q$ obtained from simulations are larger mainly due to surface scattering processes, which for simplicity are not considered [44]; therefore, those results are not intended to match the measured quantities, but the analysis of the simulations is key to understand the experimental results. From the simulations, we obtain the maximum $Q$-values for $\theta = 90^\circ$ (L7⊥) and for $\theta = 0$ (L7||). Extended calculations for different $FF$-values (not shown) reveal that $\theta = 0$ and $\theta = 90^\circ$ are the most robust configurations (i.e., higher $Q$-values). Figure 4(b) shows the evolution of $Q$ with the $FF$ for the two optimum orientations ($\theta = 0$, 90°) and for the standard L7. We obtain a similar trend for the three types of PCMs. Figure 4(b) shows that the main impact of the change in the hole shape is a slight decrease of $Q$ for $FF$ below ~0.35; especially in the case of L7⊥, $Q$-values are largely preserved for small $FF$-values. Therefore, the L7⊥ configuration is expected to be more robust to the evolution of the hole shape during the re-growth step, as our experiments had shown (Fig. 3).
Fig. 4. Finite difference time domain (FDTD) simulations of standard and re-grown L7-photonic crystal microcavities (PCMs); (a) variation of the quality factor \( Q \) of the fundamental mode (FM) for the standard L7 with an air filling factor, \( FF = 0.31 \) and for the re-grown PCMs defined by the angle \( \theta \) between \( \Gamma K \) and the [110] crystallographic direction; the insets describe the model for the holes after the re-growth step and the planar views of the PCMs for \( \theta = 0, 45^\circ, 90^\circ \) and a schematics of the model for the hole shape; (b) variation of \( Q \) with \( FF \) for standard L7, \( L7\perp \) (\( \theta = 0^\circ \)) and \( L7\parallel \) (\( \theta = 90^\circ \)); (c) evolution of the spectral positions of the FM of the standard and re-grown PCMs for different values of \( r/a \) where \( r \) is the hole radius for standard PCMs and the starting hole radius for the re-grown PCMs. Solid lines represent a guide to the eye.

Figure 4(c) shows the calculated spectral positions of the FMs for different \( r/a \)-values. For the re-grown PCMs we have taken into account the decrease in the slab thickness and the enlargement of the radius \( r \) due to the cleaning process. As \( r/a \) increases, the modes of the standard L7 PCMs experience a blueshift. The same trend is obtained for the re-grown (both \( L7\perp \) and \( L7\parallel \)) PCMs. The relative blueshifts between the standard and re-grown PCMs are within 20-30 nm for the entire considered spectral window (910-1020 nm) in good agreement with the experimental values.

5. Conclusion

In summary, we have fabricated GaAs-based L7-PCMs using nano-patterned photonic crystal templates and epitaxial re-growth. The circular shape of the holes in standard L7 PCMs evolves to a hexagonal shape in the re-grown L7 PCMs. Optical characterization performed in standard and re-grown L7 PCM shows that the modal photonic structure is preserved after the re-growth step including its linear polarization. A blueshift for the fundamental mode in the re-grown structures with respect to the standard PCM has been measured and attributed to a larger effective \( FF \). The fundamental mode of re-grown L7 PCMs present high \( Q \)-values (up to \(~6000\)) and in average maintains a 36% of its \( Q \) for \( L7\perp \) PCMs and a 15% for \( L7\parallel \) PCMs with respect to those in standard L7 PCMs. FDTD simulations predict that \( Q \)-values can be similar to standard cavities for a broad range of \( r/a \) values and show a better performance for re-grown PCMs with \( \Gamma K \) perpendicular to [110] in agreement with experiments. Overall, our result supports the use of epitaxial re-growth methods to obtain PCMs coupled to site controlled QDs.

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