Near thresholdless laser operation at room temperature: supplementary material

I. PRIETO^{1,2}, J. M. LLORENS¹, L. E. MUÑOZ-CAMÚÑEZ¹, A. G. TABOADA^{1,2}, J. CANET-FERRER³, J. M. RIPALDA¹, C. ROBLES¹, G. MUÑOZ-MATUTANO^{3,4}, J. P. MARTÍNEZ-PASTOR³, AND P. A. POSTIGO^{1,*}

¹IMM–Instituto de Microelectrónica de Madrid (CNM–CSIC), Isaac Newton 8, PTM E–28760, Tres Cantos, Madrid, Spain

²Labor für Festkörperphysik HPF F2, Schafmattstrasse 16, 8093 Zürich, Switzerland

³UMDO (Unidad asociada al CSIC), P.O. Box 22085, E–46071 Valencia, Spain

⁴Optics and Quantum Communications Group, ITEAM, UPV, Valencia, Spain

*Corresponding author: pabloaitor.postigo@imm.cnm.csic.es

Published 16 January 2015

This document provides supplementary information to "Near thresholdless laser operation at room temperature," <http://dx.doi.org/10.1364/optica.2.000066>. © 2015 Optical Society of America
<http://dx.doi.org/10.1364/optica.2.000066.s001>

APPENDIX A: DESIGN OF THE L9 PHOTONIC CRYSTAL MICROCAVITY

There are several types of PCMs that present high Q/V_{eff} ratio, although they often require an accurate positioning and a size tuning of specific holes within the lattice [1–8] which also makes them very sensitive to fabrication imperfections. The use of elongated line-defect PCMs of the type L_n , where n stands for the number of holes suppressed to create the defect, has been shown to provide a good compromise for the Q/V_{eff} ratio.[9] We have designed L9-PCMs with lattice constant $a = 350$ nm, slab thickness $d = 190$ nm and 11 rows of neighbouring holes ($N = 11$). We have carried out simulations by the finite difference time domain (FDTD) method [10] that have allowed us to evaluate the quality factor (Q), the spectral position (λ), the effective volume (V_{eff}) and the effective refractive index (n_{eff}) of the fundamental mode of the L9-PCM. Further details on the simulation of similar structures can be found elsewhere.[11] Figure S1 shows the results of the simulations varying the size of the holes of the PCM from $r/a = 0.20$ to 0.38 in steps of 0.01 . The PCMs present an increasing Q as r/a increases, with a maximum Q for $r/a = 0.35$; for higher r/a -values the performance of the cavity is dramatically degraded and at $r/a = 0.38$ the Q -value is only several hundreds (Figure S1a). Figure S1b and S1c show the variation with r/a of the spectral position of the fundamental mode (λ) and the effective mode volume (V_{eff}), respectively. V_{eff} is related to the degree of confinement of the

photonic mode and is defined as:[12]

$$V_{\text{eff}} = \frac{\int_{V_{\text{total}}} \epsilon E^2 d^3r}{\max(\epsilon E^2)} \quad (\text{S1})$$

where ϵ and E^2 are the permittivity of the material and the electric field intensity of the mode, respectively, over the entire simulation window (V_{total}). As Q increases V_{eff} decreases and both drastically decrease for $r/a > 0.35$. At very high r/a -values, the mode is not confined anymore, i.e., V_{eff} increases, which corresponds to a strong decrease in Q . The effective refractive index (n_{eff}) or, alternatively, the effective permittivity (ϵ_{eff}) of the mode are defined as:[13]

$$\epsilon_{\text{eff}} = n_{\text{eff}}^2 = \frac{\int_{V_{\text{total}}} n^2 E^2 d^3r}{\int_{V_{\text{total}}} E^2 d^3r} \quad (\text{S2})$$

Figure S1d shows the variation of n_{eff} with r/a : n_{eff} is relatively constant ($n_{\text{eff}} \simeq 2.75$) for a wide range of r/a -values, although it decreases strongly for $r/a > 0.35$. This is an evidence of the smaller confinement within the microcavity for very high r/a values, which dramatically degrades Q . Our calculations did not consider the influence of both the imperfections introduced during fabrication and the absorption losses of the active region, which produce the main deviations from experimental measurements. We have experimentally determined that the highest Q -factors are obtained for the devices with r/a between 0.26 and 0.31 (not shown). For the device presented in

the manuscript, the calculated parameters are: $r/a = 0.30$, $Q = 440200$, $\lambda = 1290.6$ nm, $V_{\text{eff}} = 1.43 (\lambda/n_{\text{eff}})^3$ and $n_{\text{eff}} = 2.78$.

APPENDIX B: POWER THRESHOLD

Figure S2a shows the integrated power emitted by the fundamental mode versus the excitation power (LL-curve) at room temperature (RT). The curve presents a rather smooth transition within the ASE region which is typical from high- β lasers.[14] We have performed a linear fit of the LL-curve for high excitation powers. A threshold of $P_{\text{th}} = 4.1 \pm 0.2$ μW is obtained by extrapolation of the red line to zero output.[3, 15] That value corresponds to the total irradiated power. The estimation of the actual excitation power absorbed by the slab is obtained by considering the reflectivity of the GaAs surface ($\sim 33\%$), the slab thickness (190 nm) and the absorption coefficient of GaAs for the excitation wavelength of 785 nm (10^4 cm^{-1}). Therefore, the absorbed pump power is a $\sim 21\%$ of the total incident power [16] and the effective threshold pump power is 860 nW. Figure S2b shows the spectrum of the fundamental mode at the lowest excitation ($P_{\text{exc}} = 370$ nW) with a $Q = 7400$. We should notice that Q is referred to empty cavities, where no material gain or loss mechanisms are considered. For a lasing system, the stimulated emission affects the linewidth and the term Q is thus no longer valid. In general, a fraction of stimulated emission is always expected at powers well below threshold.[17] Therefore, we have considered Q of the fundamental mode as that measured at the lowest excitation power, and we have used $\lambda/\Delta\lambda$ as an effective Q altered by gain and loss mechanisms. For $P_{\text{exc}} = 10.14$ μW , well above the threshold, an intense emission from the mode is observed with $\lambda/\Delta\lambda = 12100$. A set of 30 working devices were fabricated and measured. All of the measured devices with emission inside the PL (around half) showed laser emission with similar features to the sample shown, that showed the best β .

APPENDIX C: RATE EQUATION ANALYSIS

We have fitted the LL-curve to a rate equations model:[18]

$$\frac{dN}{dt} = R_p - \frac{N}{\tau_r} - \frac{N}{\tau_{\text{nr}}} - g \frac{c}{n_{\text{eff}}} (N - N_{\text{tr}}) P \quad (\text{S3})$$

$$\frac{dP}{dt} = \Gamma g \frac{c}{n_{\text{eff}}} (N - N_{\text{tr}}) P + \Gamma \beta \frac{N}{\tau_r} - \frac{P}{\tau_c} \quad (\text{S4})$$

where N and P are the carrier and photon densities within the microcavity, R_p is the pumping rate, g is the differential gain coefficient and N_{tr} is the transparency carrier density. A cavity photon lifetime $\tau_c = 6.14$ ps is obtained from the linewidth of the mode below the threshold. The confinement factor $\Gamma = 0.0018$ accounts for the overlap between the PCM mode with the QDs (see Appendix D). The radiative and non radiative recombination lifetimes are $\tau_r = 1.20$ ns and $\tau_{\text{nr}} = 0.24$ ns, respectively (see Appendix E). The Purcell effect [19–21] is not expected to modify the effective radiative recombination rate in our system, mainly due to the large linewidth broadening of the emitters at RT.[22] Therefore we have excluded this effect from our rate equations model. We have used β , g and N_{tr} as fitting parameters and obtained $\beta = 0.85$, $g = 1.495 \times 10^{-11} \text{ cm}^2$ and $N_{\text{tr}} = 1.0 \times 10^{14} \text{ cm}^{-3}$. Using those values we can estimate the pumping threshold:[18]

$$R_{p,\text{th}} = (1 - \beta) \frac{N_{\text{th}}}{\tau_r} + \frac{N_{\text{th}}}{\tau_{\text{nr}}} \quad (\text{S5})$$

$$N_{\text{th}} = \frac{1}{\tau_c \Gamma \frac{c}{n_{\text{eff}}} g} + N_{\text{tr}} \quad (\text{S6})$$

where $R_{p,\text{th}}$ and N_{th} are the pumping rate and the carrier density, respectively, evaluated at threshold in the steady state. We have obtained $R_{p,\text{th}} = 884.1$ nW and $N_{\text{th}} = 11.1 \times 10^{14} \text{ cm}^{-3}$, which are consistent with the values extracted from the linear fitting to experimental data.

APPENDIX D: CONFINEMENT FACTOR Γ

The confinement factor (Γ) accounts for the matching between the active region and the optical mode. The conventional definition of Γ assumes that the entire active region is spectrally matched to the optical mode.[18] Nevertheless, that statement is no longer valid when the active region consists of a set of QDs, because the inhomogeneous broadening of the QDs is much larger than the linewidth of the cavity mode. Therefore, we have defined Γ as:

$$\Gamma = \xi \Gamma_{\text{spatial}} \quad (\text{S7})$$

where ξ is the spectral matching factor and Γ_{spatial} is given by:[18]

$$\Gamma_{\text{spatial}} = \frac{\int_{\text{active}} n_a^2 E^2 d^3r}{\int_{\text{cavity}} n^2 E^2 d^3r} \quad (\text{S8})$$

where E^2 is the electric field intensity of the cavity mode and n_a and n are the refractive indexes of the active region and the host material, respectively.

Spectral matching: ξ

To determine ξ it is necessary a comparative analysis of the emission properties of the ensemble of QDs with respect to the cavity mode. The analysis (not shown) of the PL spectrum of the QD ensemble at RT reveals the presence of a family of QDs centered in 1280 nm with an inhomogeneous linewidth of 23 meV.[23] Considering a homogeneous broadening of the QDs at RT of 10 meV [24] only those QDs within a spectral window of 10 meV around the fundamental mode are expected to be spectrally matched to the cavity mode. The analysis, similar to that of Ref. [25] shows that 60% of the QDs are spectrally matched to the mode at RT, i.e., $\xi = 0.6$.

Spatial matching: Γ_{spatial}

To evaluate Γ_{spatial} we performed FDTD simulations varying the total number of QDs within the cavity defect (N) in random positions. The spatial distribution of the L9-fundamental mode profile is presented in Figure S3. We assumed that every QD is independent on the others and that the final value results from the sum of each of the individual contributions of the QDs. We modeled each QD as a 3D box with the same size than a single FDTD cell in X and Y axes and twice the size of the FDTD cell in Z axis. The dimensions in X, Y and Z axes are $L_{\text{QD},x} = 17.5$ nm, $L_{\text{QD},y} = 15.1$ nm, and $L_{\text{QD},z} = 9.5$ nm. Those dimensions agree well with experimental and theoretical estimations.[26] We used a statistical analysis to evaluate the impact of the random character of the positions of the QDs on Γ . One thousand random configurations for the QD positions were realized for each of the systems containing $N = 1, 3, 5, 10, 100, 200, 300$ or 1000 QDs within the defect. The value of E^2 was evaluated in the position of every QD.

The resulting confinement factor: Γ

The value of Γ was calculated from Eq. (S7). Since more than 70% of the cavity mode is localized within the cavity defect, we have restricted the calculation to the cavity defect. Figure S4a shows Γ for each of the 1000 random configurations. Each configuration has N QDs inside the cavity defect. The large dispersion of the results for configurations with a small amount of QDs (aprox. $N < 100$) is attributed to the strong role of the position of the QDs with respect to the mode profile distribution. As the number of QDs increases, Γ is less sensitive to the random distribution of the QDs. The maximum value of Γ ($\Gamma = 1$) is achieved when every spatial position where the mode profile is non-zero is occupied by a QD that is also spectrally tuned to the mode. That is expected to occur when the amount of QDs in a single layer within the defect is as large as $\sim 10^5$, i.e., the averaged QD density on the wafer is $\sim 10^{13} \text{ cm}^{-2}$. Nevertheless the cavity defect can only host a maximum number of QDs $N_{\text{max}} \simeq 8000$ which corresponds to a QD density of $3.8 \times 10^7 \text{ cm}^{-2}$. Therefore the maximum value of the confinement factor is $\Gamma_{\text{max}} = 0.07$. On the other side, Γ equals 0 when the active region is either spatially located in the nodes and/or is not spectrally tuned to the mode. In particular, for the determination of Γ in our case, it is necessary to evaluate the number of QDs lying within the defect. We have performed transmission electron microscopy (TEM) and atomic force microscopy (AFM) analysis (not shown) to determine an averaged density of QDs of 10^{10} cm^{-2} . [27] Therefore, we have estimated the total number of QDs lying within the cavity defect to be approximately 210 QDs. We note that, for a total amount of 210 QDs, the value of Γ strongly converges to $\Gamma = 0.0018 \pm 0.0002$, even though the random character of the spatial and spectral distributions of the QDs lying within the cavity defect. In general, the random distribution of the emitters might degrade the performance of the final device, provided that Γ might adopt a large dispersion depending on the specific distribution of the emitters within the cavity. Instead, for the configuration here presented, Γ shows only small deviations ($\simeq 10\%$) which plays an important role in the observation of the emission characteristics of the devices, that systematically exhibited lasing. We believe that the combination of the spatial and spectral properties of the emitters with the optical properties of the L9-cavity provides such a systematic performance.

APPENDIX E: RADIATIVE (τ_R) AND NON RADIATIVE (τ_{NR}) LIFETIMES

We have determined the radiative (τ_r) and the non radiative (τ_{nr}) lifetimes from time resolved photoluminescence (TRPL) experiments (Figure S5) outside of the cavities for a wide range of temperatures from 10 K to RT. Whereas at low temperatures the total decay time (τ) can be related to radiative processes, as the system reaches high temperatures the dynamics is rather governed by the non radiative mechanisms. At temperatures below 180 K, the τ -values are within 1.20–1.6 ns and at T close to RT $\tau = 0.2$ ns. Assuming that τ_r does not present strong variations with T and

$$\frac{1}{\tau} = \frac{1}{\tau_r} + \frac{1}{\tau_{nr}} \quad (\text{S9})$$

we have obtained $\tau_{nr} = 0.24$ ns using $\tau_r = 1.20$ ns.

REFERENCES

1. M. Nomura, N. Kumagai, S. Iwamoto, Y. Ota, and Y. Arakawa, "Photonic crystal nanocavity laser with a single quantum dot gain," *Opt. Express* **17**, 15975–15982 (2009).
2. Y. Akahane, T. Asano, B. S. Song, and S. Noda, "High-Q photonic nanocavity in a two-dimensional photonic crystal," *Nature*, **425**, 944–947 (2003).
3. S. Strauf, K. Hennessy, M. T. Rakher, Y. S. Choi, A. Badolato, L. C. Andreani, E. L. Hu, P. M. Petroff, and D. Bouwmeester, "Self-tuned quantum dot gain in photonic crystal lasers," *Phys. Rev. Lett.* **96**, 127404 (2006).
4. K. Hennessy K, A. Badolato, M. Winger, D. Gerace, M. Atatüre, S. Gulde, S. Fält, E. L. Hu, and A. Imamoglu, "Quantum nature of a strongly coupled single quantum dot-cavity system", *Nature* **445**, 896–899 (2007).
5. B. S. Song, S. Noda, T. Asano, and Y. Akahane, "Ultra-high-Q photonic double-heterostructure nanocavity," *Nat. Mater.* **4**, 207–210 (2005).
6. E. Kuramochi, M. Notomi, M. Mitsugi, A. Shinya, T. Tanabe, and T. Watanabe, "Ultrahigh-Q photonic crystal nanocavities realized by the local width modulation of a line defect," *Appl. Phys. Lett.* **88**, 041112 (2006).
7. S. Frédéric, D. Dalacu, J. Lapointe, P. J. Poole, G. C. Aers, and R. L. Williams, "Experimental demonstration of high quality factor, x-dipole modes in InAs/InP quantum dot photonic crystal microcavity membranes," *Appl. Phys. Lett.* **89**, 091115 (2006).
8. M. Shirane, S. Kono, J. Ushida, S. Ohkouchi, N. Ikeda, Y. Sugimoto, and A. Tomita, "Mode identification of high-quality-factor single-defect nanocavities in quantum dot-embedded photonic crystals," *J. Appl. Phys.* **101**, 073107 (2007).
9. S. H. Kim, G. H. Kim, S. K. Kim, H. G. Park, Y. H. Lee, and S. B. Kim "Characteristics of a stick waveguide resonator in a two-dimensional photonic crystal slab," *J. Appl. Phys.* **95**, 411–416 (2004).
10. FDTD Solutions. Lumerical Solutions, Inc., Vancouver, BC, Canada.
11. J. M. Llorens, I. Prieto, L. E. Muñoz-Camúñez, and P. A. Postigo, "Analysis of the strong coupling regime of a quantum well in a photonic crystal microcavity and its polarization dependence studied by the finite-difference time-domain method," *J. Opt. Soc. Am. B* **30**, 1222–1231 (2013).
12. R. Coccioli, M. Boroditsky, K. W. Kim, Y. R. Samii, and E. Yablonovitch, "Smallest possible electromagnetic mode volume in a dielectric cavity," *Optoelectronics, IEE Proceedings* **145**, 391–397 (1998).
13. K. Hennessy, "Photonic Crystal Nanocavities Positioned and Tuned for Cavity-QED," Ph.D. Thesis, University of California Santa Barbara, 2006.
14. G. Björk, A. Karlsson A., and Y. Yamamoto, "On the linewidth of microcavity lasers," *Appl. Phys. Lett.* **60**, 304 (1992).
15. K. Srinivasan, P. E. Barclay, O. Painter, J. Chen, A. Y. Cho, and G. Gmachl, "Experimental demonstration of a high quality factor photonic crystal microcavity," *Appl. Phys. Lett.* **83**, 1915–1917 (2003).
16. Atlasov K., "Light Control and Microcavity Lasers Based on Quantum Wires Integrated in Photonic-Crystal Cavities," Ph.D. Thesis, École Polytechnique Fédérale de Lausanne, 2009.
17. J. Canet-Ferrer, I. Prieto, G. Muñoz-Matutano, L. J.

- Martínez, L. E. Muñoz-Camúñez, J. M. Llorens, D. Fuster, B. Alén, Y. González, L. González, P. A. Postigo, and J. P. Martínez-Pastor, "Excitation power dependence of the purcell effect in photonic crystal microcavity lasers with quantum wires," *Appl. Phys. Lett.* **102**, 201105 (2013).
18. L. A. Coldren, S. W. Corzine, and M. Mashanovitch, "Diode lasers and photonic integrated circuits," Wiley, Hoboken, N.J., 2012.
 19. T. Baba, and D. Sano, "Low-threshold lasing and purcell effect in microdisk lasers at room temperature," *IEEE J. Sel. Top. Quantum Electron.* **9**, 1340–1346 (2003).
 20. J. Canet-Ferrer, L. J. Martínez, I. Prieto, B. Alén, G. Muñoz-Matutano, D. Fuster, Y. González, M. L. Dotor, L. González, P. A. Postigo, and J. P. Martínez-Pastor, "Purcell effect in photonic crystal microcavities embedding InAs/InP quantum wires," *Opt. Express* **20**, 7901–7914 (2012).
 21. T. Suhr, N. Gregersen, M. Lorke, and J. Mørk, "Modulation response of quantum dot nanolight emitting diodes exploiting purcell enhanced spontaneous emission," *Appl. Phys. Lett.* **98**, 211109 (2011).
 22. H. Y. Ryu and M. Notomi, "Enhancement of spontaneous emission from the resonant modes of a photonic crystal slab single-defect cavity," *Opt. Lett.* **28**, 2390–2392 (2003).
 23. A. G. Taboada, A. M. Sánchez, A. M. Beltrán, M. Bozkurt, D. Alonso-Álvarez, B. Alén, A. Rivera, J. M. Ripalda, J. M. Llorens, J. Martín-Sánchez, Y. González, J. M. Ulloa, J. M. García, S. I. Molina, and P. M. Koenraad, "Structural and optical changes induced by incorporation of antimony into InAs/GaAs(001) quantum dots," *Phys. Rev. B* **82**, 235316 (2010).
 24. M. Bayer, and A. Forchel, "Temperature dependence of the exciton homogeneous linewidth in $\text{In}_{0.60}\text{Ga}_{0.40}\text{As}/\text{GaAs}$ self-assembled quantum dots," *Phys. Rev. B* **65**, 041308 (2002).
 25. M. Nomura, S. Iwamoto, K. Watanabe, N. Kumagai, Y. Nakata, S. Ishida, and Y. Arakawa, "Room temperature continuous-wave lasing in photonic crystal nanocavity," *Opt Express* **14**, 6308–6315 (2006).
 26. A. G. Taboada, J. M. Llorens, D. Alonso-Álvarez, B. Alén, A. Rivera, Y. González, and J. M. Ripalda, "Effect of Sb incorporation on the electronic structure of InAs quantum dots," *Phys. Rev. B* **88**, 085308 (2013).
 27. I. Prieto, L. E. Muñoz-Camúñez, A. G. Taboada, C. Robles, J. M. Ripalda, and P. A. Postigo, "Fabrication of high quality factor GaAs/InAsSb photonic crystal microcavities by inductively coupled plasma etching and fast wet etching," *J. Vac. Sci. Technol. B* **32**, 011204 (2013).

Fig. S1. Calculation of the optical parameters of the L9 photonic crystal microcavity. **a)** Quality factor (Q). **b)** Spectral position (λ). **c)** Effective volume (V_{eff}). **d)** Effective refractive index (n_{eff}). The calculation has been made for the fundamental mode with pitch and slab thickness $a = 350$ nm and $d = 190$ nm, respectively.

Fig. S2. Optical properties of the L9 photonic crystal microcavity laser at room temperature. **a)** Light in – light out curve of the laser (black dots). The red line shows the linear fit used to obtain an estimated total power threshold $P_{\text{th}} = 4.1 \pm 0.2 \mu\text{W}$. **b)** Photoluminescence (PL) of the fundamental cavity mode (FM) measured (black dots) at the lowest excitation power $P_{\text{exc}} = 370$ nW. The best lorentzian fit (red line) provides $Q = 7400$. **c)** PL of the FM measured (black dots) at high excitation power $P_{\text{exc}} = 10.14 \mu\text{W}$. The best lorentzian fit (red line) provides $\lambda/\Delta\lambda = 12100$.

Fig. S3. Spatial distribution of the electric field ($|E|^2$) of the fundamental mode of the L9 photonic crystal microcavity. **top**, Planar view (XY) in the mid-slab. **middle**, XZ cross section through the center of the cavity. **bottom**, YZ cross section through the center of the cavity. The calculation has been made for pitch, radius and slab thickness $a = 350$ nm, $r = 105$ nm and $d = 190$ nm, respectively.

Fig. S4. Determination of the confinement factor for the fundamental mode of the L9 photonic crystal microcavity with self-assembled QDs. **a**) Value of Γ for different configurations with N QDs ($N = 1, 3, 5, 10, 100, 200, 300, 1000$) in random positions. The green dashed line sets the maximum value of Γ ($\Gamma_{\max} = 0.07$). **b**) Statistical analysis of Γ from data in **a**). The dashed line is shown for guidance purposes. The green line sets the maximum number of QDs that the cavity defect can host ($N_{\max} \simeq 8000$). The calculation has been made for L9-PCMs with pitch, radius and slab thickness $a = 350$ nm, $r = 105$ nm and $d = 190$ nm, respectively. Error bars account for the statistical deviation of the data.

Fig. S5. Determination of the radiative and non radiative lifetimes from the total decay time. The plot shows the total decay time (τ) measured by time-resolved photoluminescence (TRPL) as a function of the temperature for an ensemble of the InAsSb QDs outside of the photonic crystal structures.