High quality factor 1-D Er^{3+}-activated dielectric microcavity fabricated by RF-sputtering

Sreeramulu Valligatla,[1,2,3] Alessandro Chiasera,[1,*] Stefano Varas,[1] Nicola Bazzanella,[4] D. Narayana Rao,[3] Giancarlo C. Righini,[5,6] and Maurizio Ferrari[1]

[1]*CNR-IFN CSMFO Lab, Via alla Cascata 56/C, Povo, 38123 Trento, Italy.*
[2]*Università di Trento,Dipartimento di Fisica, via Sommarive 14, Povo, 38123 Trento,Italy.*
[3]*School of Physics, University of Hyderabad, Hyderabad 500046, India.*
[4]*Università di Trento, Dipartimento di Fisica, IdEA lab., Via Sommarive 14, Povo, 38123 Trento, Italy.*
[5]*Centro Fermi, Piazza del Viminale 1, 00184 Roma, Italy.*
[6]*CNR-IFAC, via Madonna del Piano 10, 50019 Sesto Fiorentino, Firenze, Italy.*
**achiaser@science.unitn.it*

Abstract: Rare earth-activated 1-D photonic crystals were fabricated by RF-sputtering technique. The cavity is constituted by an Er^{3+}-doped SiO_2 active layer inserted between two Bragg reflectors consisting of ten pairs of SiO_2/TiO_2 layers. Scanning electron microscopy is employed to put in evidence the quality of the sample, the homogeneities of the layers thickness and the good adhesion among them. Near infrared transmittance and variable angle reflectance spectra confirm the presence of a stop band from 1500 nm to 2000 nm with a cavity resonance centered at 1749 nm at 0° and a quality factor of 890. The influence of the cavity on the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission band of Er^{3+} ion is also demonstrated.

OCIS codes: (310.1860) Deposition and fabrication; (140.3945) Microcavities; (050.5298) Photonic crystals; (160.5690) Rare-earth-doped materials; (160.4760) Optical properties; (250.5230) Photoluminescence.

References and links

1. A. Chiappini, A. Chiasera, S. Berneschi, C. Armellini, A. Carpentiero, M. Mazzola, E. Moser, S. Varas, G. C. Righini, and M. Ferrari, "Sol-gel-derived photonic structures: fabrication, assessment, and application," J. Sol-Gel Sci. Technol. **60**(3), 408–425 (2011).
2. T. Yoshie, L. Tang, and S.-Y. Su, "Optical microcavity: sensing down to single molecules and atoms," Sensors (Basel Switzerland) **11**(2), 1972–1991 (2011).
3. V. E. Ferry, A. Polman, and H. A. Atwater, "Modeling light trapping in nanostructured solar cells," ACS Nano **5**(12), 10055–10064 (2011).
4. M. Ferrari and G. C. Righini, *Physics and Chemistry of Rare-Earth Ions Doped Glasses* (Trans Tech Publishers, 2008), Chap 3.
5. C. M. Johnson, P. J. Reece, and G. J. Conibeer, "Slow-light-enhanced upconversion for photovoltaic applications in one-dimensional photonic crystals," Opt. Lett. **36**(20), 3990–3992 (2011).
6. M. Clara Gonçalves, L. M. Fortes, R. M. Almeida, A. Chiasera, A. Chiappini, M. Ferrari, and S. Bhaktha, "Photoluminescence in Er^{3+}/Yb^{3+} -doped silica-titania inverse opal structures," J. Sol-Gel Sci. Technol. **55**, 52–58 (2010).
7. G. C. Righini, Y. Dumeige, P. Féron, M. Ferrari, G. Nunzi Conti, D. Ristic, and S. Soria, "Whispering gallery mode microresonators: fundamentals and applications," Riv. Nuovo Cim. **34**, 435–488 (2011).
8. H. Rigneault, C. Amra, S. Robert, C. Begon, F. Lamarque, B. Jacquier, P. Moretti, A. M. Jurdyc, and A. Belarouci, "Spontaneous emission into planar multi-dielectric microcavities: theoretical and experimental analysis of rare earth ion radiations," Opt. Mater. **11**(2-3), 167–180 (1999).
9. A. Chiasera, R. Belli, S. N. B. Bhaktha, A. Chiappini, M. Ferrari, Y. Jestin, E. Moser, G. C. Righini, and C. Tosello, "High quality factor Er^{3+}-activated dielectric microcavity fabricated by RF-sputtering," Appl. Phys. Lett. **89**(17), 171910 (2006).
10. Y. Li, L. M. Fortes, A. Chiappini, M. Ferrari, and R. M. Almeida, "High quality factor Er-doped Fabry-Perot microcavities by sol-gel processing," J. Phys. D Appl. Phys. **42**(20), 205104 (2009).
11. J. Jasieniak, C. Sada, A. Chiasera, M. Ferrari, A. Martucci, and P. Mulvaney, "Sol-gel based vertical optical microcavities with quantum dot defect layers," Adv. Funct. Mater. **18**(23), 3772–3779 (2008).

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

12. L. Persano, P. D. Carro, E. Mele, R. Cingolani, D. Pisignano, M. Zavelani-Rossi, S. Longhi, and G. Lanzani, "Monolithic polymer microcavity lasers with on-top evaporated dielectric mirrors," Appl. Phys. Lett. **88**(12), 121110 (2006).
13. S. F. Chichibu, T. Ohmori, N. Shibata, and T. Koyama, "Dielectric SiO_2/ZrO_2 distributed Bragg reflectors for ZnO microcavities prepared by the reactive helicon-wave-excited-plasma sputtering method," Appl. Phys. Lett. **88**(16), 161914 (2006).
14. Y. Li and R. M. Almeida, "Photoluminescence from a Tb-doped photonic crystal microcavity for white light generation," J. Phys. D **43**(45), 455101 (2010).
15. G. Ma, J. Shen, Z. Zhang, Z. Hua, and S. H. Tang, "Ultrafast all-optical switching in one-dimensional photonic crystal with two defects," Opt. Express **14**(2), 858–865 (2006).
16. Y. G. Boucher, A. Chiasera, M. Ferrari, and G. C. Righini, "Photoluminescence spectra of an optically pumped erbium-doped micro-cavity with SiO_2/TiO_2 distributed Bragg reflectors," J. Lumin. **129**(12), 1989–1993 (2009).
17. A. Wajid, "On the accuracy of the quartz-crystal microbalance (QCM) in thin-film depositions," Sens. Actuators A Phys. **63**(1), 41–46 (1997).
18. S. Boyadzhiev, V. Georgieval, and M. Rassovska, "Characterization of reactive sputtered TiO2 thin films for gas sensor applications," J. Phys. Conf. Ser. **253,** 012040 (2010).
19. S. J. L. Ribeiro, Y. Messaddeq, R. R. Gonçalves, M. Ferrari, M. Montagna, and M. A. Aegerter, "Low optical loss planar waveguides prepared by an organic-inorganic hybrid system," Appl. Phys. Lett. **77**(22), 3502–3504 (2000).

1. Introduction

The recent developments of optically confined structures have opened new possibilities in the field of both basic and applied physics, in a large area covering information communication, health and biology, energy, and environment monitoring technologies [1–3]. On this scenario, rare earth-activated glasses have become one of the key materials in photonic systems because of their relevance for the development of optical amplifiers [4] and effort was directed to develop appropriate material systems and configurations to exploit at the best the luminescence properties of rare earth ions like Er^{3+} [5, 6]. The last decade has seen a remarkable increase in the experimental efforts to control and enhance emission properties of emitters by tailoring the dielectric surrounding of the source [1, 5–7]. With this aim, several approaches, using nanocomposite materials or specific geometries, such as planar interfaces, photonic crystals, solid state planar microcavities, dielectric nanospheres, and spherical microresonators, have been proposed. Among these systems, planar microcavity resonators, also called one-dimensional (1-D) photonic crystals, are the simplest photonic band-gap (PBG) device exploitable to manage the spectroscopic properties of luminescent species such as rare earth ions [8–10], and quantum dots [11].

Oxide-based dielectric materials are particularly suitable for fabricating PBG structures because they have wide transparency from the ultraviolet to the near infrared (NIR). Furthermore, oxide-based dielectric materials have good resistance to temperature, corrosion and radiation as well [12–14]. Various techniques have been employed to fabricate Fabry-Pérot dielectric microcavities where reproducible deposition of thin dielectric layers, that constitute distributed Bragg reflectors (DBRs), is mandatory to achieve a high quality factor (Q). Processes like ion implanting [8], sol–gel [10, 11], electron-beam evaporation [15], and sputtering [9, 13] can be successfully employed for the fabrication of microcavities based on oxide dielectric materials. However, to reach high Q factor using dielectric material, where the refractive index difference between the different materials is not so high as for the semiconductor, the real time control of the deposition process is mandatory to allow a precise tailor of the deposition rate and obtain an enough good uniformity in thickness. Moreover, the increasing of the interest in shifting the rejected wavelengths in different zone ranging from visible to near infrared [5, 14], requires an accurate design of the structures [16] and the definition of flexible experimental protocol capable to adapt itself to different materials and spectral range. A possible way to monitor the thicknesses of the processed film during the deposition procedure is represented by the quartz-crystal microbalance (QCM) which could be used for monitoring the growth-rate in physical vapor deposition and sputtering processes [17]. Sputtering methods are widely used in industrial process because high quality films can be obtained at low temperature substrates [18]. We have also demonstrated as the RF

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

sputtering is a suitable technique for fabrication of dielectric microcavities and it is a cheap and versatile technique to deposit alternating layers of different materials with controlled refractive index and thickness [9]. With these advantages, as well as the possibility to incorporating QCM, RF-sputtering process is an extremely appropriate candidate to fabricate high quality and homogeneous 1-D photonic crystals.

In this article, we report on a reproducible fabrication protocol based on RF-sputtering technique and on optical, spectroscopic and morphological characterization of the realized high quality Er^{3+}-activated dielectric microcavity consisting of alternating silica and titania films. A radical improvement of the fabrication and diagnostic methodologies reported in [9] is presented, leading to novel quantitative and qualitative results in terms of quality factor, morphology, and extended spectroscopic functionalities of the microcavity.

2. Experimental

Erbium doped dielectric microcavity is composed of alternating high and low refractive index dielectric layers, SiO_2 and TiO_2 respectively, with an optical thickness of $\lambda/4$ (λ = 1.749 μm) [8, 9]. SiO_2/TiO_2 one dimensional photonic crystals with SiO_2 doped Er^{3+} defect layer was prepared by multi target RF sputtering technique [9] using silicon and silica substrates. The substrates were cleaned inside the RF sputtering deposition chamber by heating at 120°C for 30' just before the deposition procedure. Sputtering deposition of the films was performed by sputtering alternatingly a 15x5 cm^2 titania target and a 15x5 cm^2 silica target. For the defect layer a 15x5 cm^2 silica target, on which metallic erbium pieces were placed, was employed. The deposition time, necessary to reach the appropriate thickness of the Bragg mirror layers, was 2 h 30 min for titania target and 1 h 20 min for silica target. The deposition time necessary to reach the appropriate thickness of the silica defect layer, to obtain cavity resonance centered at 1.749 μm, was 2 h 40 min. The residual pressure, before the deposition, was about $1.1x10^{-6}$ mbar. During the deposition process, the substrates were not heated and the temperature of the sample holder during the deposition was 30 °C. The sputtering occurred with an Ar gas pressure of $5.4x10^{-3}$ mbar; the applied RF power was 150 W and 130 W and the reflected powers 0 W for silica and titania targets, respectively. Particular attention was paid to the reproducibility of the single layers. To monitor the thickness of the layers during the deposition, two quartz microbalances Veeco instruments thickness monitor model QM 311, faced on the two targets were employed. Thickness monitor was calibrated for the two kinds of materials by a long deposition process (24h of deposition) and by directly measuring the thickness of the deposited layer by an m-line apparatus [9, 19]. The final resolution on the effective thickness obtained by this quartz microbalance is about 4 Å.

The compositional analysis was performed using energy dispersive spectroscopy (EDS), employing a Oxford mod. INCA PentaFETx3 apparatus. EDS measurement was employed in particular to quantify the Erbium content in the active layer. Scanning electron microscopy (SEM) was used to analyze the morphology of the multi-layer films and thickness of the each layer. The cross section of the microcavity was analyzed by a FEG mod. JEOL JSM-7001F apparatus at 15 kV after covering the films with a 20 nm gold layer. The transmittance and variable angle reflectance spectra in the NIR region of the sample was obtained by using a double beam Varian-Cary spectrophotometer with a resolution of 0.1 nm. Photoluminescence (PL) spectroscopy was performed using the 514.5 nm line of an Ar^+ ion laser as excitation source. The luminescence was dispersed by a 320 mm single-grating monochromator with a resolution of 1 nm. The light was detected by using a Hamamatsu photomultiplier tube and standard lock-in technique. More details about the experimental setup can be found in [9].

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

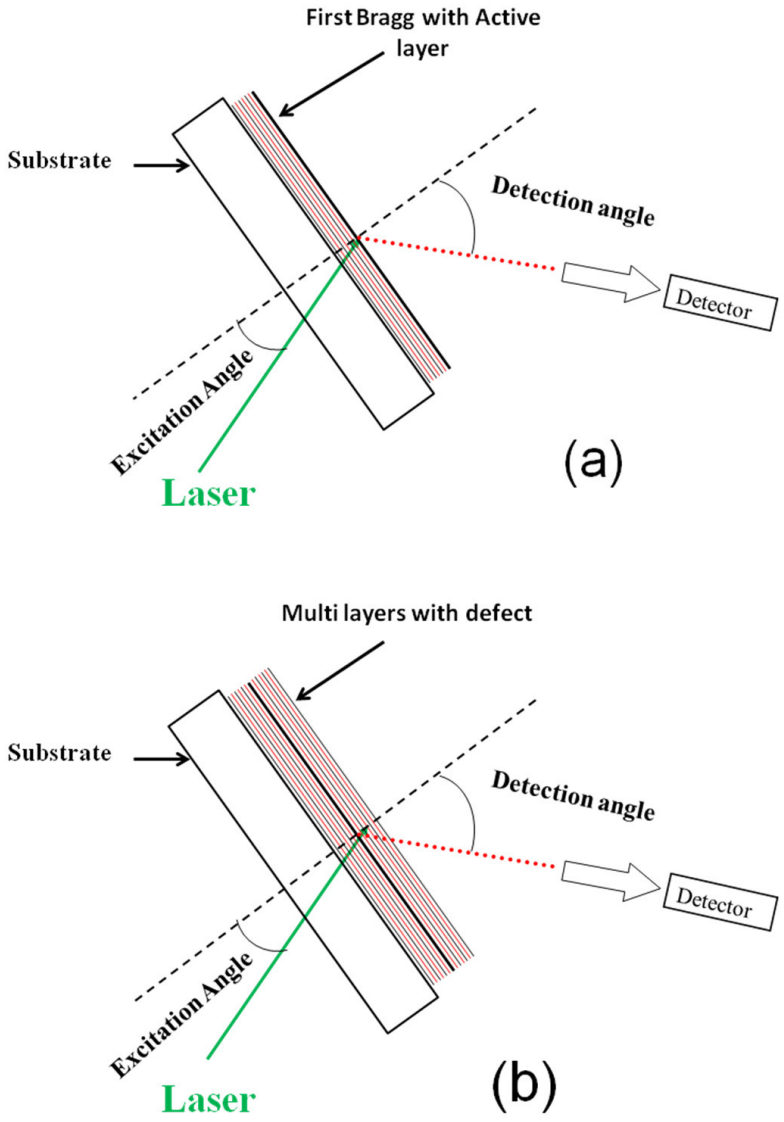

Fig. 1. Schematics of the excitation and detection geometries employed for a reliable assessment of the influence of the cavity on the 1.5 μm emission band of Er^{3+} ion. Figure 1(a): configuration employed for the Er^{3+}-activated reference sample: Figure 1(b): configuration employed for the Er^{3+}-activated 1-D microcavity.

The effectiveness of a dielectric microcavity is determined by its Q factor, in particular for active systems, where the luminescence features are crucial. To allow a direct comparison of the Er^{3+} luminescence intensity between the photonic crystal and the single active layer a reference sample was fabricated. The reference sample was deposited on SiO_2 substrate during the same deposition run for the fabrication of the photonic crystal. It was therefore obtained employing the same fabrication protocol and targets used for the photonic crystal and so the layers that constitute the samples exhibit the same thickness and composition. Thus, in the reference sample the second Bragg reflector that should be placed above the active layer, is not present. In this way the luminescence from the reference sample can be emitted without any cavity effect but the excitation laser light that will come from the substrate through the Bragg mirror toward the active layer will be affected by the presence of the multilayer

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

structure in the same way for the both photonic crystal and reference samples. As a result the total amount of excitation laser light that reaches the active layer is the same for both the samples, as far the experimental conditions allow. Photoluminescence measurements were performed paying particular attention to the excitation and detection angles and the experimental condition adopted for the photonic crystal and reference sample was, as far as possible, the same. Figure 1(a) and 1(b) show the schematics of the excitation and detection geometries employed for the Er^{3+}-activated reference sample and the 1-D cavity, respectively.

3. Results and discussions

The cavity is constituted by an Er^{3+}-doped SiO_2 active layer inserted between two Bragg reflectors, each one consisting of ten pairs of SiO_2/TiO_2 layers. SEM image of the cross section of the 1-D photonic crystal is shown in the Fig. 2(a) and 2(b).

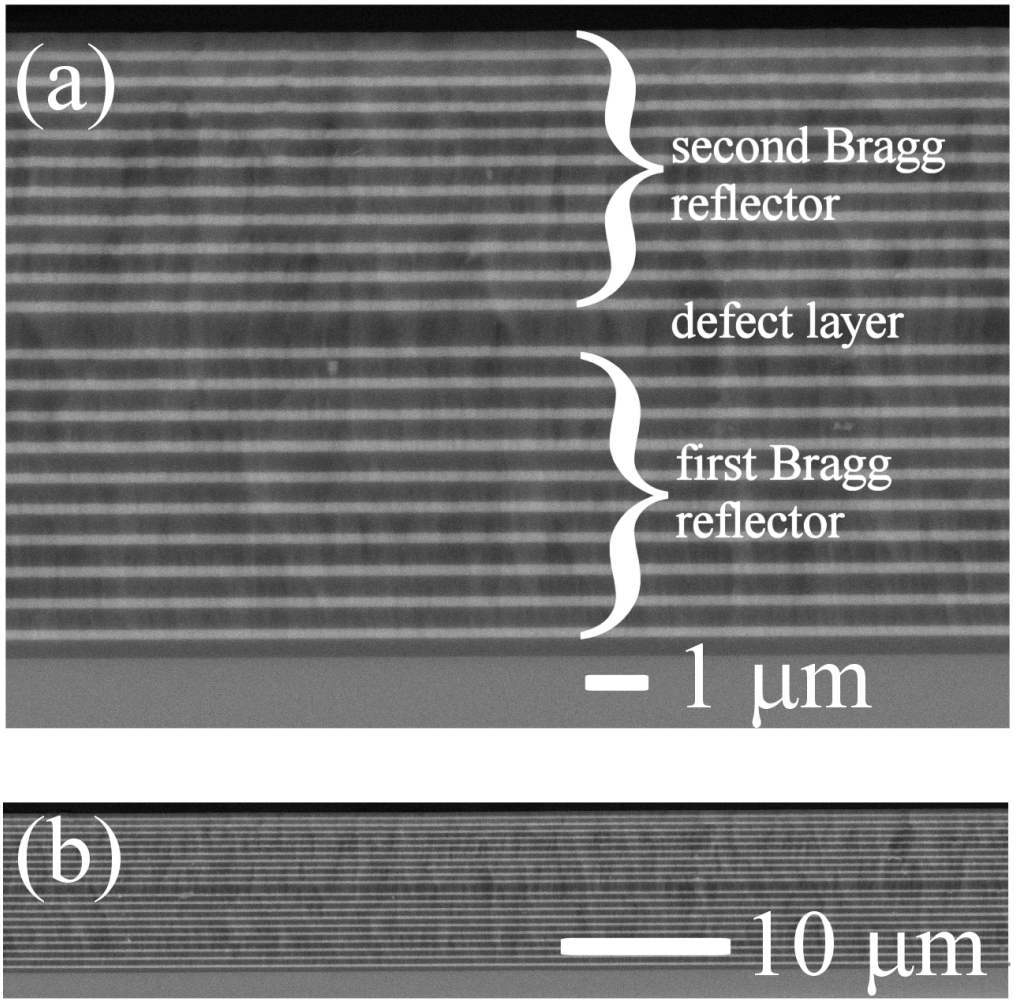

Fig. 2. SEM micrograph of the 1-D microcavity cross section. The bright and the dark areas correspond to TiO_2 and SiO_2 layers, respectively. The substrate is located on the bottom of the images and the air on the top. (a): image for a section of the sample of about 16 μm in length. (b): image for a section of the sample of about 60 μm in length.

The dark regions correspond to the SiO_2 layer and the bright regions correspond to the TiO_2 layer. The substrate is located at the bottom of the images and the air on the top. It is possible to identify the defect layer and the two Bragg reflectors. The SEM analysis allow to

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

measure the thickness for the Bragg mirrors of 320 ± 5 and 170 ± 5 nm for the silica and titania layers, respectively, and a thickness of 625 ± 5 nm for the SiO_2 Er^{3+}-doped defect layer. From Fig. 2(b) it is possible to note how the deposition technique allows to have a good uniformity of the thickness as well as a perfect adhesion of the films on extent of the order of tens of micrometers in length. EDS measurements indicate that the Erbium content in the active layer is about 0.6 ± 0.1 mol%.

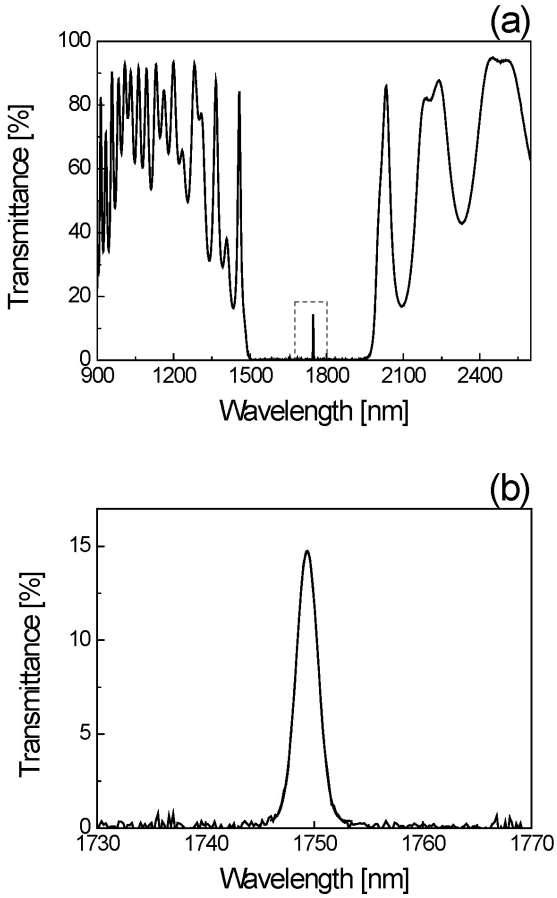

Fig. 3. (a) Transmittance spectrum of the cavity with two Bragg reflectors, each one consisting of ten pairs of SiO_2/TiO_2 layers, in the region between 1000 and 2600 nm. The stop band range from 1490 to 1980 nm. The cavity resonance corresponds to the sharp maximum centered at 1.749 μm. The incident light is unpolarized. (b) High resolved (resolution 0.1 nm) transmission measurement of the cavity resonance. The line width of the resonance is 1.97 nm that correspond to a quality *Q* factor of 890. The spectral region shown in (b) is evidenced by the dashed box in (a).

The NIR transmission spectrum is shown in Fig. 3(a) and 3(b). The transmittance spectra were measured at zero degree of incident angle. In Fig. 3(a) it is possible to note as the spectral reflectance range, i.e. the stop band, ranges from 1490 to 1980 nm. A sharp peak in the transmittance spectrum appears at 1749 nm. It corresponds to the cavity resonance wavelength related to the half wave layer inserted between the two Bragg mirrors. The transmittance spectrum in Fig. 3(b), obtained with a resolution of 0.1 nm, shows the sharp resonance line. The full width at half maximum (FWHM), calculated by a fit with a Gaussian curve, is 1.97 nm, corresponding to a quality factor of the cavity, *Q*, of about 890.

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

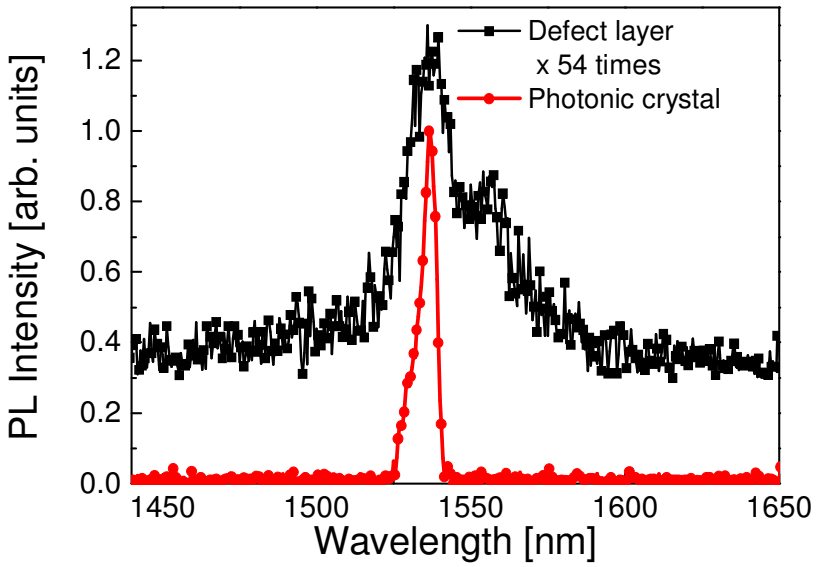

Fig. 4. $^4I_{13/2} \rightarrow {}^4I_{15/2}$ photoluminescence spectra of the cavity activated by Er^{3+} ion in 1-D photonic crystal (—●—) and of the single Er^{3+}-doped SiO_2 active layer with first Bragg mirror (—■—). The light is recorded at 50° from the normal on the samples upon excitation at 514.5 nm.

Figure 4 shows the luminescence spectra relative to the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of the Er^{3+} ions obtained for the 1-D photonic crystal and for the reference sample composed by the single Er^{3+}-doped SiO_2 active layer, with only one Bragg mirror.

The luminescence from the cavity and from the Er^{3+} - doped single SiO_2 layer with one Bragg reflector was detected at 50° from the normal on the samples, with a solid angle of 10^{-1} sr. Both the cavity and the Er^{3+}-doped single SiO_2 layer with first Bragg reflector were excited with the 514.5 nm line of an Ar^+ ion laser with an excitation power of 180 mW. The erbium emission from the reference sample is centered at 1538 nm with a FWHM of 29 nm and exhibits the characteristic shape of Er^{3+} ion in silica glass [1]. The position of the cavity resonance is strongly dependent on the detection angle. For a detection angle of 50°, the cavity resonance corresponds to the maximum of the erbium PL of the Er^{3+} -doped SiO_2 active layer with the first Bragg. The peak luminescence intensity of Er^{3+} ions is enhanced by a factor 56, in respect to that detected for the reference at the corresponding wavelength. The Er^{3+} $^4I_{13/2} \rightarrow {}^4I_{15/2}$ PL line shape is strongly narrowed by the cavity and exhibits a FWHM of 5 ± 0.5 nm (Fig. 4). The Er^{3+} emission is enhanced when the wavelength corresponds to the cavity resonant mode and weakened for the other emission wavelengths.

To put in evidence the effect of the detection angle on the emission feature of the photonic crystal it is possible to repeat the luminescence measurements with different detection configuration.

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

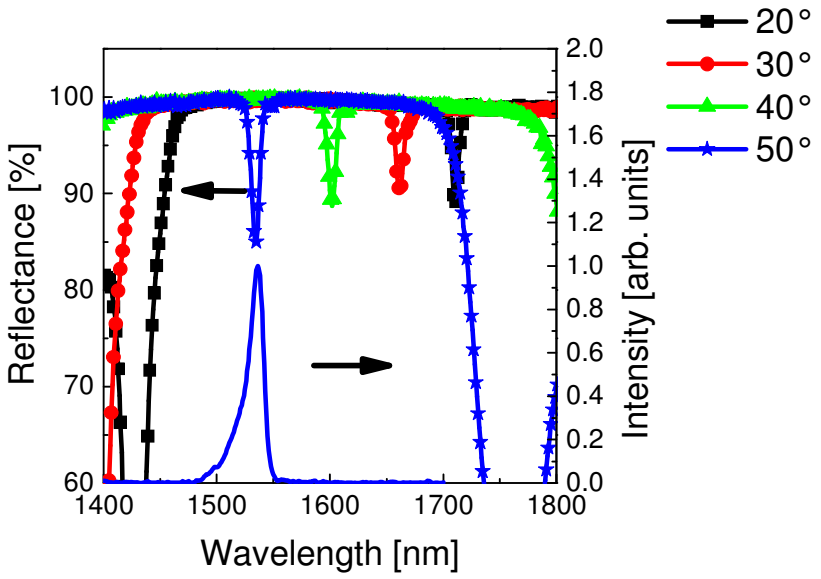

Fig. 5. Angle dependence reflectance spectra and luminescence spectrum obtained detecting the luminescence at 50° from the normal incidence.

In Fig. 5 are reported the angle dependent reflectance spectra obtained at 20°, 30° 40° and 50°, respectively, and the luminescence spectrum obtained detecting the luminescence at 50° from the normal incidence. The position of the cavity resonance shifts with the detection angle and when the resonance corresponds to the wavelength of the emission band of the Er^{3+} ions the emission peak emitted from the sample is aligned with the minimum of the reflectance.

4. Conclusions

A valuable protocol based on RF-sputtering technique was developed for the fabrication of Er^{3+} activated 1-D photonic crystals. The cavity is constituted by an Er^{3+}-doped SiO_2 active layer inserted between the two Bragg reflectors consisting of ten pairs of SiO_2/TiO_2 layers. SEM microscopy was used to measure directly the thickness of the layers and put in evidence the homogeneities of the layers and their good adhesion. NIR transmittance and variable angle reflectance spectra confirm that the presence of a stop band from 1500 nm to 2000 nm with a cavity resonance centered at 1749 nm at 0°, with a FWHM 1.97 nm, corresponding to a quality factor Q of about 890. The effect of the cavity on the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission band is demonstrated by the narrowing of the emission band as well as by the enhancement of the Er^{3+} photoluminescence PL intensity. The latter effect is verified by the observation that the peak luminescence intensity from the photonic crystal is 56 time higher than that of the reference sample under the same excitation condition. In conclusion, three important outcomes characterize this work: 1) fabrication protocol leading to a specific control of compositional, optical, and geometrical parameters allowing high Q factor and luminescence enhancement; 2) extent of the stop band and the resonance to longer wavelengths leading to potential new functionalities; 3) the reference sample fabrication and the employed geometry allow a reliable assessment of the influence of the cavity and we believe this method will be largely employed in the community being the errors on luminescence enhancement measurement, due to the different configurations, drastically reduced.

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012

Acknowledgment

This research was performed in the framework of the India-Trento Program for Advanced Research ITPAR Phase II research project and of the tutorial project "La ricerca come Mestiere/ la tecnologia nei mestieri" coordinated by the Bruno Kessler Foundation and driven by Dr. Micaela Vettori. AC and MF acknowledge the impressive scientific motivation of the students Alessio Gerola, Luca Dalbosco, and Lorenzo Nicoletti tutored by Prof, Gianfranco Festi (ITI Marconi Rovereto) and Prof. Silvia Filosi (Liceo Rosmini Rovereto).

#172721 - $15.00 USD Received 17 Jul 2012; revised 7 Aug 2012; accepted 9 Aug 2012; published 31 Aug 2012