One-dimensional photonic Crystal for the 1.3-1.5 μm region

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ABSTRACT

Multilayer of PbTe quantum dots embedded in SiO2 were fabricated by alternatively use of Plasma Enhanced Chemical Vapor Deposition and Laser Ablation techniques. The optimal growing parameters for both the SiO_2 films and the PbTe quantum dots were obtained. The refractive index and optical absorption of the sample were studied. Multilayer X-ray diffraction patterns were used to estimate the nanoparticles diameter. Morphological properties of the nanostructured material were studied using Transmission Electron Microscopy. Both absorption spectra and X-ray diffraction patterns reveled the nanoparticles are 6-8 nm in diameter, consequently appropriate for developing optical devices in the infra red region. Finally the multilayer was grown inside a Fabry Perot cavity. The transmittance of the one-dimensional photonic crystal was measured.

Keywords: pulsed laser deposition, PECVD, Quantum Dots, Optical Device

1. INTRODUCTION

The study of semiconductor Quantum Dots (QD) embedded in glasses [1] has attracted great attention because their non-linear optical properties[2], making them promising materials for the development of integrated "all-optical" devices. However, their use has in many cases been hindered bay the lack of appropriate depositions techniques with the capability to control the dimensions, shape, and size distribution of the QDs.

In this work we report the fabrication of multilayers of SiO_2/PbTe QDs using alternatively pulsed-laser deposition (PLD) to grow the semiconductor nanoparticles and Plasma Enhanced Chemical Vapor Deposition (PECVD) to grow the dielectric host.

PbTe was chosen because its QDs absorption band can be controlled by its size to fall in the spectral window of interest for optical communications (1.3-1.5 μm). This, together with the QD high optical nonlinearity, makes this material an excellent candidate for development of optical devices.

PLD is a thin film deposition technique that has high potential for development of nanostructured materials. Once fixed the target-substrate distance, vacuum chamber pressure, and laser energy density, the QD size depends only on the amount of ablated material i.e. the number of laser pulses used to grow each QD layer. It means, that controlling the number of laser pulses one is able to control the QD size.

In order to produce QDs with absorption band in the infrared region, it is necessary to grow those nanoparticles with dimension from 6.5 to 8nm in diameter according to reports form Tudury et al[3]. In previous works we reported the fabrication of nanostructured material in form of SiO_2/PbTe QD multilayer. The growing parameter used in the cited work, required about 1400 to 1800 laser pulses to grow nanoparticles with the desired size this yielding a broadening in the size distribution of the nanoparticles due basically to coalescence[4]. At that time we pointed out that for the fabrication of an efficient optical device, it was necessary to obtain bigger nanoparticles with a narrower size distribution. For the used technique it means obtaining bigger nanoparticles by using a lower number of laser pulses[5,6].

On the other hand, if the device is designed to operate in the infrared region, the dielectric host should no exhibit absorption in this region. FTIR absorption spectra of the SiO_2 layers fabricated showed an absorption peak around 3400cm^{-1} related to H_2O. Consequently it was a second feature to be improved in the nanostructured material we have previous fabricated.

In the present work we propose changes in the growing parameters for both the semiconductor QDs and the dielectric matrix, in order to improve the QDs size distribution and diminish the H_2O absorption band present in the dielectric host.

Our proposal to grow nanoparticles with the desired sizes and a narrow size distribution was to use higher background pressures. Under these growing conditions QDs are formed at the earlier stages of the plasma formation and can gradually increase their seizes during the flight to the substrate.

For the glass matrix, we studied the influence of growing parameters like RF power, distance between the RF electrodes and the total pressure in the quality of the SiO_2 films.

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2. EXPERIMENTAL

Multilayers showing absorption bands in the 1.3-1.5 μm region were fabricated using PbTe quantum dots embedded in a dielectric host (SiO_2). The quantum dots were grown by pulsed laser deposition (PLD) of a PbTe target using the second harmonic of a Q-Switched Quantel Nd:YAG laser under high purity argon atmosphere. The optimal laser fluence and target-substrate distance used for the QDs fabrication were 20 J/cm^2 and 7 cm, respectively.

The glass matrix was fabricated by PECVD using tetramethoxysilane (TMOS) as precursor. The RF power was supplied by a RF-150 TOKYO HI-Power operating at 13.56 MHz and coupled to the RF electrodes through a matching box. The distance between the RF electrodes, the RF power and the TMOS partial pressure were varied in order to obtain the optimal conditions to fabricate de SiO_2 films. The TMOS was kept at 30^o C to assure a constant material evaporation rate. The chamber was kept constantly in argon and silicon-alkoxide TMOS atmosphere during the growth of both the PbTe QDs and SiO_2 layers. The best growing parameter were obtained for films with the lowest H_2O content. For an RF power of 40 W, the optimal distance between RF electrodes and partial TMOS pressure were 6.5 cm and 0.01 mbar, respectively.

The total background pressure was kept constant at 0.2 mbar (0.19 for Argon and 0.01 for TMOS). The alternating growth of the samples was achieved with a computer controlled interface using a LabView code.

High Resolution Transmission Electron Microscopy (HRTEM) measurements of the multilayers were performed using a 300kV Jeol JEM 3010 microscope with 1.7 Å point-to-point resolution. For HRTEM measurements a Si(111) wafer was used as substrate. Samples were grown at room temperature.

For optical absorption measurements a BK7 Corning glass was used as substrate. Measurements were done on a 160 bilayers (2.5 μm thick SiO2/PbTe QDs) structure.

The structural analysis of the samples were carried out by means of X-ray diffraction at room temperature (T ≈ 300K) using θ:2θ scans in Philips (PW 11700) diffractometer system with Cu Kα radiation (λ=1.54056 Å) and a graphite monochromator for diffracted beam. The measurements were obtained using 0.02^o step size and 2s/step. The diffractograms were analyzed by means of the Scherrer's method.

Refractive index measurements of the multilayer were carried out with a METRICON prism coupler system. The transmittance of the produced photonic crystal was measured using a Perking Elmer (Lambda 9) spectrophotometer operating in the 300-3000 nm region.

3. RESULTS AND DISCUSSIONS

3.1 SiO_2 glass matrix fabrication and characterization

SiO_2 films were fabricated using different RF powers in order to determine the optimal growing parameters to produce the dielectric host, where the QDs will be embedded. FTIR spectra of the fabricated glass samples (Figure 1) show peaks associated to H_2O, CH_2,CH_3, Si-OH and Si-H. An increasing in the RF power produced a diminution of the H_2O content in the film and consequently an increasing in its refractive index.

Studies also revealed that the increasing in the refractive index was also associated with an increasing in the OH^- content. On the other hand the OH^- content is related to an increasing of the plasma dissociation degree due to the increasing of the RF power. Si-OH bondings could generate energy levels in the gap of the dielectric matrix which could increase the refractive index.

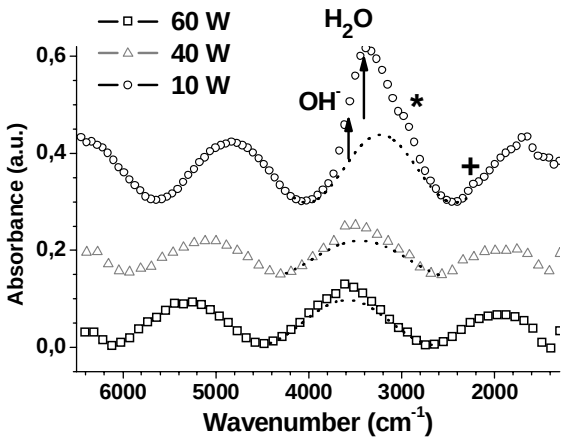

Figure 1. FTIR spectra of the fabricated glass samples in the 6500-1300 cm^{-1} region for different RF power (10W; 40 W; 60 W)). Doted lines are the simulation of the interference due to the multilayer structure. Two arrows indicate the position of the absorptions peaks related to H_2O and OH^-. (*****) represent the peak corresponding to CH_2 and CH_3 vibrations modes and (+)represent the peak corresponding to Si-H vibrations mode.

3.2- SiO2/PbTe multilayer characterization

The structural properties of the samples were studied using X-ray diffractometry. Figure 2 shows two diffractograms of the fabricated multilayer. Diffraction peaks corresponding to the (200) and (111) direction of the PbTe fcc structure can be seen in the diffractograms. According to Scherrer's method the QD diameter (Φ) can be calculated trough the relation: $\Phi = \frac{k * \lambda}{\beta * cos(\theta)}$, where $0.9 < k < 1$, λ is the wavelength, θ is the peak position and β is the peak FWHM. Using the diffraction peak located at $2\theta = 27.58^o$ ($\beta = 2.973^o$) corresponding to the (200) PbTe diffraction was possible to estimate the QDs size to 7.9 nm.

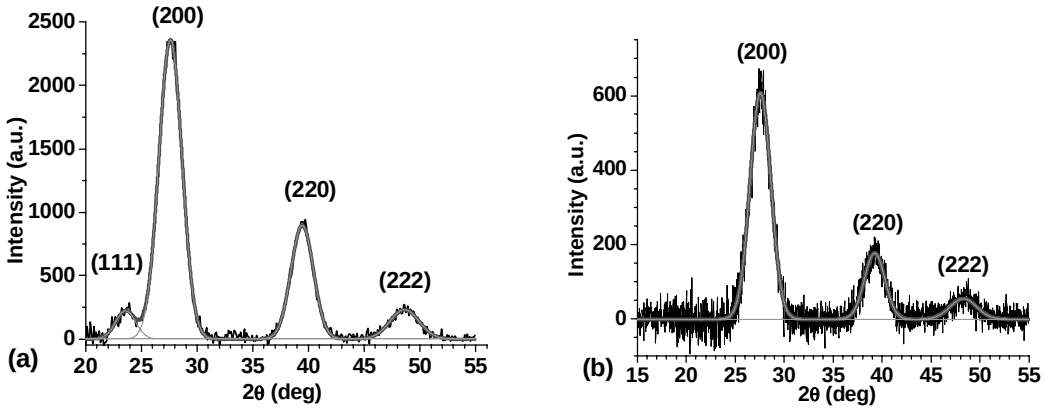

Figure 2. X-ray diffractograms of the SiO2/PbTe multilayer. Measurements were carried out both in (a) grazing incidence (1.75^o) and (b) θ:2θ mode.

Figure 3 shows the absorptions spectra of one fabricated sample. The absorption measurements were carried out at three different sample regions P, Q and R (Figure 3 (b)) where the QDs have different sizes according to the different depositions rates obtained in PLD when one moves from position P toward position R. The absorption measurements were performed at 100-160 bilayers SiO_2/PbTe QDs structure. Even with this number of bilayers the absorption was too

weak to be detected with a single pass through the film; therefore we used a total internal reflection multipass arrangement for this measurement as shown in Figure 3(c). Absorption bands shift to lower wavelength when measurements were moved from position P to position R due to quantum confinement effects of the PbTe nanoparticles.

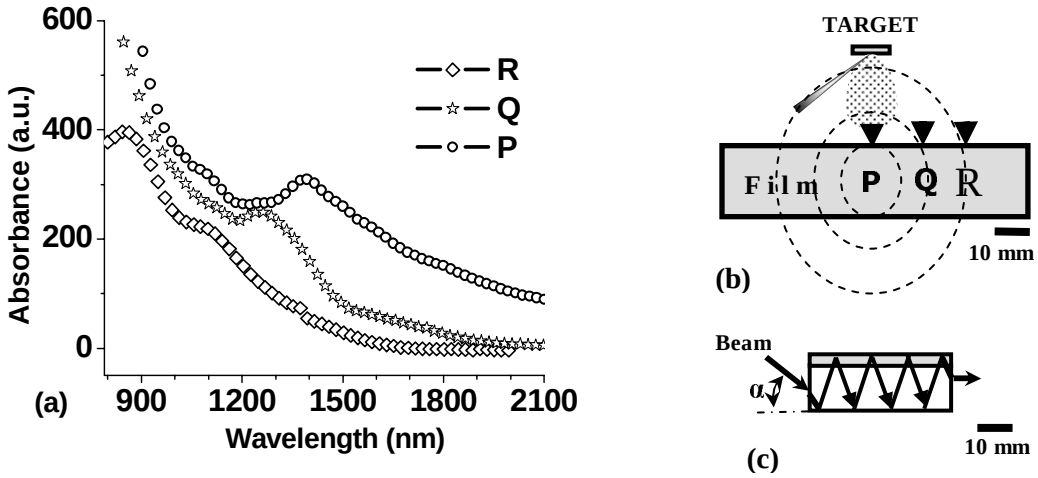

Figure 3. (a) Absorption spectra of a SiO_2/PbTe quantum dot multilayer grown by alternately PLD of PbTe nanoparticles and PECVD of SiO_2. The measurements were carried out at three different sample regions P, Q and R (b), where the quantum dots have different sizes. Dashed rings are the isothickness contours corresponding to the in-plane distribution of the amount of PbTe deposited. (c) Schematic representation of the multiple pass total internal reflection geometry used for these measurements.

Figure 4 shows HRTEM images of two multilayer fabricated during the experiences reported in this work. Samples were prepared using the conventional sample preparation procedure. The ~25 nm amorphous layers between substrate and the first QD layer could be generated by the high power RF applied to substrate before the deposition of the first QD layer. This procedure was always used to improve the multilayer adherence to susbtrate.

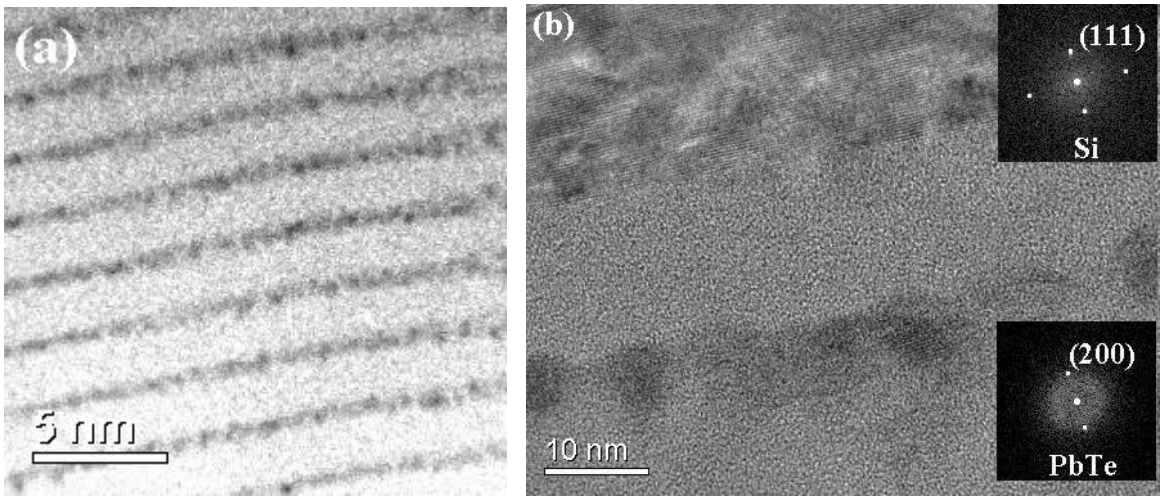

Figure 4 (a) cross-section image of a SiO_2/PbTe QDs structure fabricated in this work. Black layers correspond to PbTe nanoparticles separated by ~ 4 nm SiO_2 layers. For the complete structure 100 bilayers were deposited onto a Si(111) substrate. (b) High resolution image of a second multilayer fabricated during the experiences. In the HRTEM image can be appreciated the Si substrate (top) and the first QD layer (bottom). Insets showing the (111) diffraction for the Si substrate and the (200) diffraction for a PbTe nanoparticle. This micrograph shows ~ 6 nm thick QD layers.

3.3 Fabrication of the One-Dimensional photonic Crystal

Figure 5 shows the schematic diagram of the photonic crystal fabricated in this work. First a dielectric stack (Bragg mirror) was deposited by electron beam onto a BK7 Corning glass substrate. SiO_2 (n=1.46) and TiO_2 (n=2.35) were chosen as the low and high refractive index quarter wave layers, respectively. Secondly a SiO_2/PbTe multilayer was deposited onto the dielectric stack. The multilayer was fabricated according to the procedure described in the previous item, with PbTe QDs absorption around 1400 nm. The multilayer thickness was set to a half-wave. Finally, a second dielectric stack, equal to the firs one, was deposited onto the multilayer.

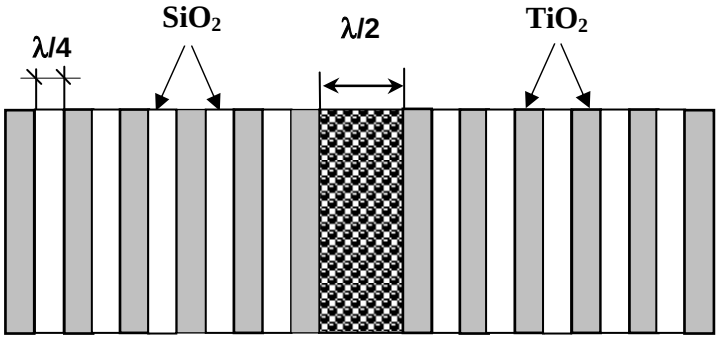

Figure 5. Schematic drawing of the one dimensional photonic crystal fabricated in this work. The gray and white strips represent the TiO_2 and SiO_2 quarter-wave layers, respectively. In the center of the Fabry Perot Etalon a SiO2/PbTe QDs multilayer (λ/2-thick) was deposited.

Calculations of the crystal spectral response were performed using the matrix method. The multilayer refractive index was assumed to be 1.49 +0.001i for the calculation. Calculated an measured transmission spectrum have a difference of about 50 nm near the resonance wavelength (1400nm). This difference can be related to a difference between the refractive index assumed for calculations (n=1.46 for SiO_2 and n=2.35 for TiO_2) and the real refractive index of the quarter–wave layers fabricated by electro beam.

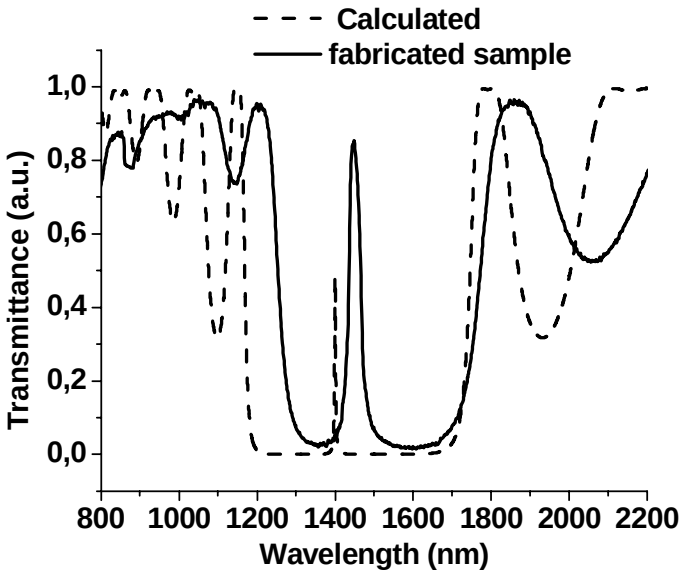

Figure 6. Measured (solid lines) and calculated (dashed lines) transmission spectrum of the fabricated photonic glass. Resonance is located at 1450 nm for the fabricated sample.

4. CONCLUSSIONS

SiO_2/PbTe QDs multilayer showing absorption in the infrared region were fabricated. The multilayer was grown inside a Fabry Perot cavity. The photonic crystal shows resonance at 1450 nm this indicating the material can be used for developing "all optical" switching device such as a saturable absorber operating in the region of interest for optical communications. Relaxation time measurements or the PbTe nanoparticles embedded in the SiO_2 matrix are performed at the moment.

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6. REFERENCES

[1] K. Tsunetomo, S. Shunsuke, T. Koyama, S. Tanaka, F. Sasaki and S. Kobayashi, "Ultrafast nonlinear optical response of CdTe microcrystallite-doped glasses fabricated by laser evaporation" Molecular Crystals and Liquid Crystals Science and Technology Section B, Nonlinear Optics, **v 13, n 1-3**, 109-126, (1995).
[2] H. Gleiter: Prog. Mater. Sci. **33**, 223 (1989)
[3] G. E. Tudury, M. V. Marquezini, L. G. Ferreira, L. C. Barbosa and C. L. Cesar, "Effect of Band Anisotropy on Electronic Structure of PbS, PbSe, and PbTe Quantum Dots", Phys. Rev. B, **62(11)**, 7357-64 (2000)
[4] E. Rodriguez, E. Jimenez, L. A. Padilha, A. A. R. Neves, G. J. Jacob, C. L. Cesar and L. C. Barbosa, "SiO2/PbTe Quantum Dots Multilayer Production and Characterization", Appl. Phys. Lett., **86** 113117-113120, (2005)
[5] E. Rodriguez, E. Jimenez, C. L. Cesar and L. C. Barbosa, "1D Photonic Band Gap Silica Doped PbTe Quantum Dot Optical Device". Glass Technology **Vol. 46 (2)**, 47-49, (2005)
[6] E. Rodriguez, E. Jimenez, A.A.R. Neves, G. J. Jacob, C. L. Cesar and L. C. Barbosa, "Fabrication and characterization of PbTe quantum dot multilayers for Photonic Fabry-Perot Devices", Physica E, **26,** 361-365, (2005)