Near field imaging of a plasmon photonic crystal patterned on the facet of a quantum cascade laser

Dibyendu Dey, Ryan. M. Gelfand, John Kohoutek, Alireza Bonakdar and Hooman Mohseni[1]

Department of Electrical Engineering and Computer Science

Northwestern University, Evanston, Illinois, USA

ABSTRACT

Planar photonic crystal (PPC) has recently attracted much attention as a promising platform for the realization of compact nanocavity devices. Our proposed photonic crystal (PC) structure consists of a periodic hole array with a point defect at the center. The device has been integrated on the facet of a quantum cascade laser working in the mid-infrared region of optical spectrum. Finite-difference time domain (FDTD) simulations have been performed to optimize the design structure. Simulations showed that with a periodicity of the holes (Λ) between 1.3um and 1.4um, the near field enhancement at the center of the cavity on the same level as the top metal surface can be as high as 10 times the incident electric field. The radius of the hole and center cavity radius are 0.45 and 0.2 times Λ. The structure was simulated at experimentally measured operating wavelength (λ=5.98um) of our device. During fabrication, we used a buffer SiO_2 layer thickness of 100nm followed by metal-dielectric-metal structure with layer thicknesses of Au – SiO_2 –Au (100/20/ 100 nm). Next, the MDM photonic crystal design was fabricated on the MDM coated facet of the QCL using focused ion beam (FIB) milling. The integrated device has been tested using an apertureless mid-infrared near field scanning optical microscopy (a-NSOM). The measurement set-up is based on an inverted microscope coupled with a commercially available Atomic Forced Microscopy (AFM). Using this technique, we could simultaneously measure the topography and NSOM image of the photonic crystal integrated QCL. It showed that the combination of high quality factor and extremely low mode volume of the PC design can squeeze the optical mode within a nanometric spot size ~ 450nm. The experimental results is a proof of concept, although we believe, further optimization and improvisation with different PC designs can lead squeezing the optical mode into a much smaller volume. Such integrated device are capable of focusing radiant infrared light down to nanometer length scale and strongly enhance the near field intensity which can be extremely useful in molecular sensing.

Key words: Bio-sensing, Focused ion beam (FIB), Near-field scanning optical microscopy(NSOM), Planar photonic crystal (PPC), Plasmonics, Quantum cascade laser (QCL), Surface plasmon resonance (SPR).

1. INTRODUCTION

Photonic crystals[1] have received much attention for their ability to control light and in particular for their ability to squeeze light[2] in a cavity. One dimensional photonic crystal are already in commercially use in the form of

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[1] hmohseni@ece.northwestern.edu, 1-847-491-5412; www.bisol.northwestern.edu

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thin film optics[3]. Two and three dimensional photonic crystals are of great interests for both fundamental and applied research. Two dimensional photonic crystals have been commercially used in making photonic crystal fiber[4] and have been used as a platform for realization of compact and efficient nanocavities[5] and lasers[6] .

An array of periodically arranged holes on the order of the wavelength forming an artificial crystal has a photonic band structure so that there are states of allowed modes and a bandgap of forbidden modes[7]. The geometry can be designed so that a point defect (one removed hole) built into the crystal will trap modes found in the bandgap. This trapped mode will spend a much longer time in the cavity because every time it tries to escape it will be reflected back, thus building up energy. The quality or Q-factor is the measure of how much energy is stored inside to the amount of energy that leaks out. Photonic crystals can have a Q factor in the millions, but can only squeeze the wavelength around the diffraction limit[8]. For molecular sensing[9], especially on the order of a single molecule, the light has to be squeezed much further; ideally on the order of the molecule, nanometers. Also, many important bio-molecules, such as proteins and pharmaceuticals, have their natural resonances in the THz which for optical excitation is in the mid-infrared (2 - 20 microns), and the challenge of interaction between these two becomes untenable. The only ways to accomplish this is to first couple the optical mode into a plasmonic mode and then squeeze the plasmonic mode down to the volume needed[10]. Our simulations have shown that this possible by using an metal-dielectric-metal (MDM) sandwich to squeeze the mode in the electric field direction and a photonic crystal cavity defect to squeeze the mode in the planar direction. Furthermore the effective index of the MDM or gap mode is quite high and so it is very slow, in this way the mode is not only squeezed by many orders of magnitude but also lasts more time in the cavity. In this way we can attain an electromagnetic mode that resonates in the THz but can be trapped to a volume on the order of a single molecule.

The next challenge is finding a strong enough source and in the mid-infrared region of optical spectrum and quantum cascade laser (QCL) is considered to be right solution for it. QCL - a semiconductor intersubband laser first demonstrated in 1994[11]. QCL has already shown path breaking performance in term of wall-plug efficiency[12] and currently considered as one of the most efficient source in the mid-infrared (MWIR) region of optical spectrum[13]. Moreover MOCVD - considered to be comparatively cheaper epitaxial growth technology, has reached quite a maturity in recent years and currently being used for QCL growth[14]. Thus QCL is getting much cost-effective and it opens up the possibility to use QCL based bio-sensors commercially available.

By patterning the facet of our QCL devices we show that an integratable lab on a chip single molecule detector is possible in future. The lasers emit a very narrowband light ~ 6 microns which gets squeezed by the plasmonic photonic crystal designed onto the front facet. The light will interact with a molecule or protein placed inside the cavity defect, thus modulating the signal, which can then be measured as it passes back through the laser and out the back facet. We have designed and built an aperturless near field scanning optical microscopy[15] setup to first test the optical properties of our devices. A scattering probe AFM tip is used to map the optical intensity in the near field with the eventual goal to be to remove the AFM and use a molecule in the cavity to modulate the near field.

We hope that this device is the first step towards a fully integratable molecular detection scheme, with future plans to include integrating the molecular transport mechanism with micro and nanofluidics, and the detector built directly onto the back facet. But at least by showing the near field map we have demonstrated interaction geometry suitable to molecule detection fabricated on the facet of a room temperature operating QCL.

2. SIMULATIONS

We investigated the photonic crystal integrated plasmonic QCL design using commercially available 3d finite difference time domain software, LUMERICAL. In order to suppress unphysical reflections, the Perfectly Matched Layers (PML) is used at the boundaries of the calculation region. All material data used in the simulation, other than the laser region, is from ref 16. The refractive index of the laser material is chosen to be 3.2, which is the weighted average of the refractive index of the active region,$In_{0.44}Al_{0.56}As/In_{0.6}Ga_{0.4}As$.

The simulated structure has a 100nm buffer silicon dioxide followed Au-SiO_2 – Au (100/20/100 nm) MDM layers perforated with the photonic crystal design. The side view of the basic geometry is shown Figure 1(b). The photonic crystal design is illuminated with a broad band source with a wavelength range between 4-9um. The polarization of light is TM. The electric field enhancement normalized with respect to the incident field is recorded at the *point A* shown in Figure 2 (b). The monitor at *point A* is over the cavity defect on the same level as the top metal surface. The period of holes of photonic crystal (Λ) is varied from 1.2, 1.3, 1.4, 1.5 to 1.6 um. The plot between near field intensity enhancement at *point A* over the wavelength range is plotted for the different period in Figure 1 (a). The device has operating lasing wavelength of 5.98 um, which is the experimentally found operating wavelength for photonic crystal integrated QCL as shown in Figure 2 (b). Thus with the fixed wavelength, the design period is chosen between 1.4 and 1.5 um to achieve a maximum field enhancement of a factor of 10. We used a multilayer structure instead of single metal, so that sandwiched dielectric can help increasing the transverse coupling between surface plasmons. We have recently got a considerable increase in electric field enhancement for MDM design compared to a single metal for nanorod antenna. The results will be presented on SPIE optics and Photonics, San Diego[17].

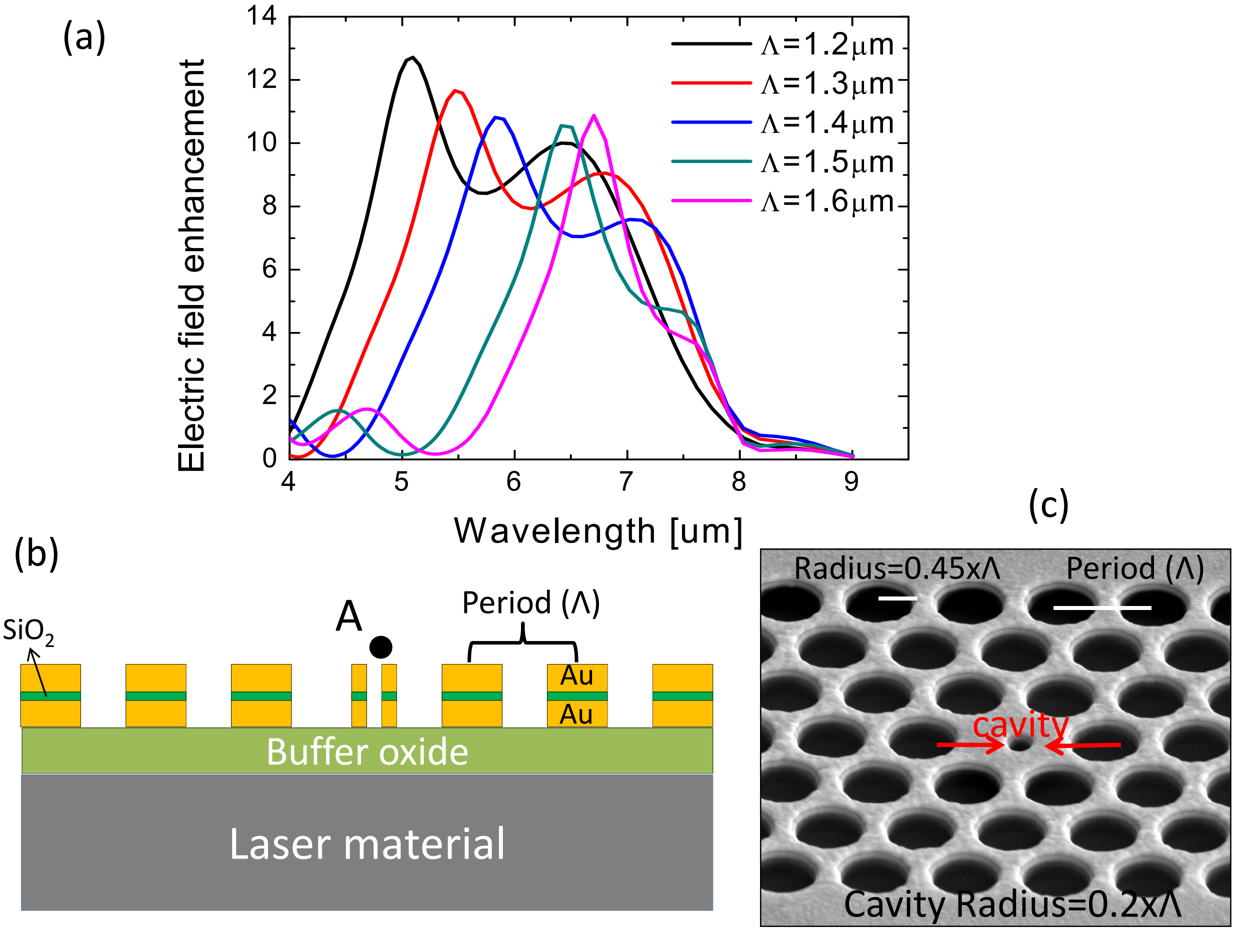

Figure 1 - (a) Simulated near field enhancement vs wavelength at different periods of the photonic crystal. The radius of the hole and cavity are 0.45 and 0.2 times the period. (b) Schematic diagram of the simulation design structure. The *point A* represents the position of field monitor. (c) SEM image of the fabricated photonic crystal with a cavity at the center. The period of the structure is ~ 1.35um.

2. FABRICATIONS

We fabricated a photonic crystal structure on an edge-emitting quantum cascade laser operating at room temperature. The core design of the QCL is based on $In_{0.44}Al_{0.56}As/In_{0.6}Ga_{0.4}As$ with a core design as outlined in a previous letter[18]. To prevent electrical shorting with the MDM antenna, an insulating buffer layer of 100nm SiO_2 is deposited on the facet of QCL using ebeam evaporation. The $SiO_2/Au/SiO_2/Au$ (200/75nm/20nm/75nm) MDM layers were then deposited on the buffer silicon dioxide film.

After that, the optimized photonic crystal structure was defined on the multilayer film using Focused Ion beam (Helios FEI). Precision milling was performed to keep all parameters matched with the resonant condition found from the simulations (Section 2). Gallium ion beam at high voltage (30keV) and low current (48pA) was used to achieve a high precision of milling. Figure 3 shows the scanning electron micrograph image of the fabricated photonic crystal (PC) integrated QCL. The device was electrically tested in pulsed mode with 1% duty cycle (100ns, 100 KHz) and the threshold current was found to be 2.28A (without pattern) and 1.65A (with pattern). Other than reduction in threshold current (due to enhanced reflectivity from the coated facet), the fabricated device didn't have any serious effect on the operation of the laser.

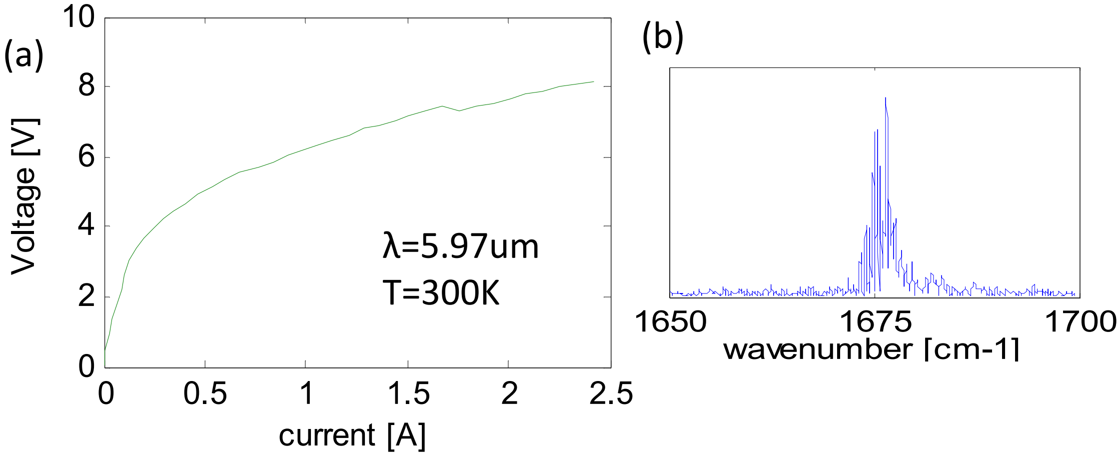

Figure 2 - (a) I-V characteristics of QCL integrated with photonic crystal. (b) Emission spectrum at 300K for QCL integrated photonic crystal.

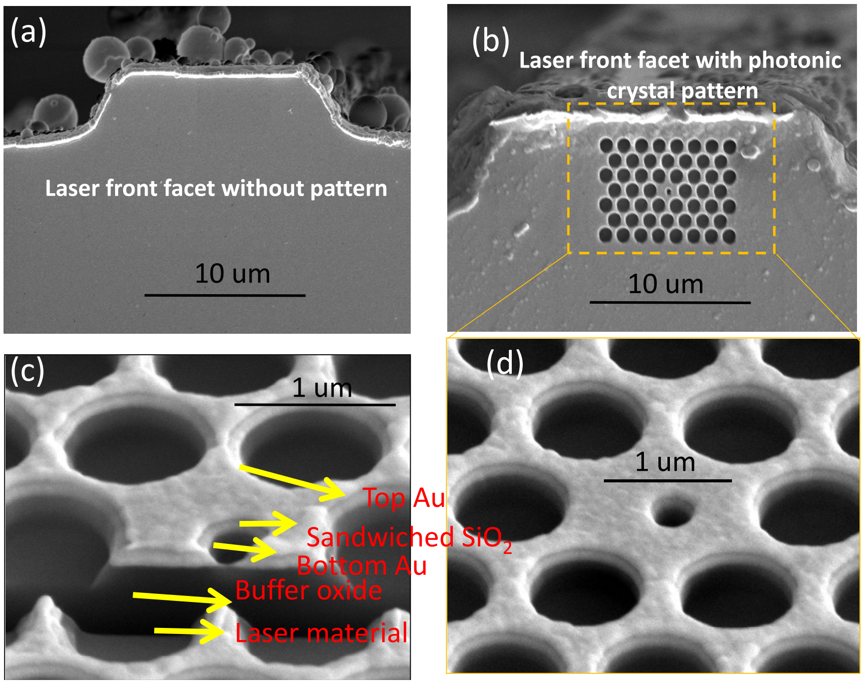

Figure 3 - (a) Laser front facet with buffer coated buffer oxide (100nm) and MDM material (Au-SiO$_2$ - Au). (b) Laser facet after fabrication of the photonic crystal structure using Focused ion beam milling. (c) The cross-section of the MDM layers structure. (d) Top view of the photonic crystal structure showing the cavity defect which has a radius ~ 270nm.

3. EXPERIMENTAL SET-UP

The resolution of Fourier transform infrared microcopy (FTIR) can't exceed more than a few micrometer due to diffraction limitation. In contrast, apertureless near-field scanning optical microscopy (a-NSOM) offers spatial resolution up to few tens of nanometers and independent of any wavelength. The experimental set-up has been illustrated in Figure 4. The tip of an atomic force microscope (AFM) scans parallel to the surface normal of the photonic crystal in tapping mode. Local electromagnetic field from PC structure is scattered by the AFM tip. Part of it gets transmitted through the laser cavity and collected by a mercury-cadmium telluride (MCT) detector after beam collimation.

The scattered signal possesses local field information. However, there is also scattering from the cantilever which produces a huge background noise. This background has been overcome by modulating the distance between the probe and the sample. At a small distance between the probe and sample, the apex signal gets hugely enhanced. There is also a contribution to the modulated signal from many other scattering centers along the probe shaft. But the signal contribution of the more distant ones can be effectively suppressed due to their rapid fading at larger distance from the sample[19,20]. Thus modulating the signal using AFM in tapping mode and then demodulating using a lock-in amplifier can give an excellent background suppressed near-field signal.

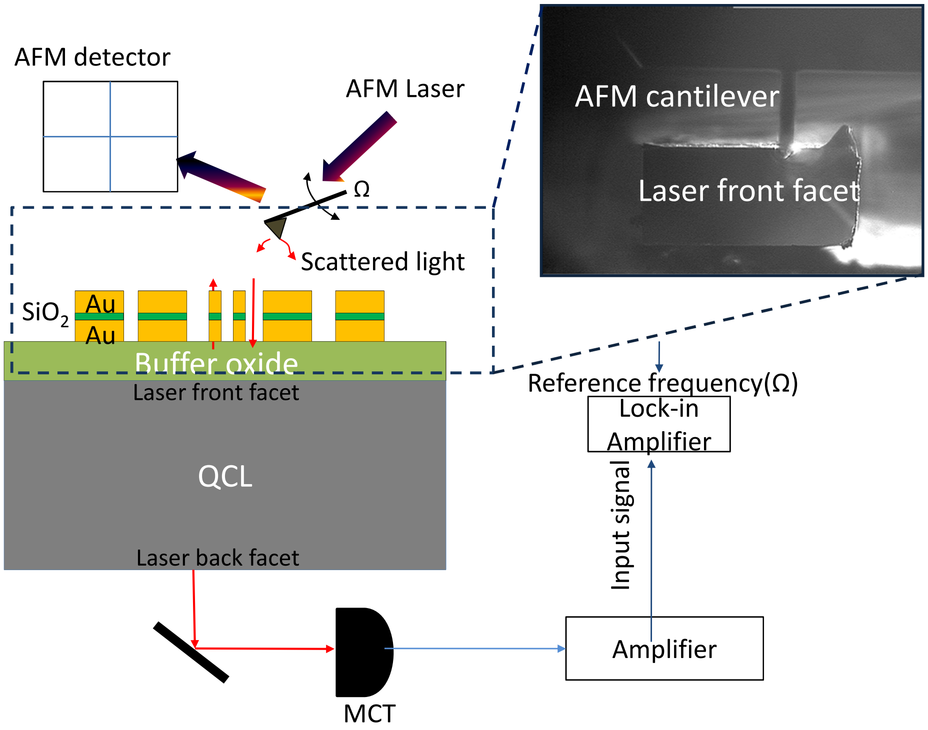

Figure 4 - Apertureless near-field optical microscope (a-NSOM) to probe near field intensity of photonic crystal integrated QCL.

Due to selection rule for intersubband transition, QCL emits TM polarized light[21], where direction of the electric field is parallel to the growth direction. The photonic crystal structure is symmetric thus incident laser polarization isn't required to be coincided with any particular axis of the design. There is also a perturbation effect on the antenna near-field due to the presence of probing tip, but it can be suppressed by using dielectric tip instead of metal tip as been suggested in ref 22.

4. MEASUREMENT

The squeezed optical mode generated by the device generates SPs to the AFM tip, which in turn radiates a signal detected by infrared detector to generate NSOM image. In Figure 4 (b) and (c), the topography and near field image of Au-SiO_2- Au (100/20/100 nm) photonic crystal are shown. The near-field image of the device was recorded with high pixel resolution and with a reduced scan speed of 0.1 lines/s. The image distortions, as seen in Figure 6 (a) - (b), are due to small drifts in sample position occurring over the long acquisition time for NSOM measurement.

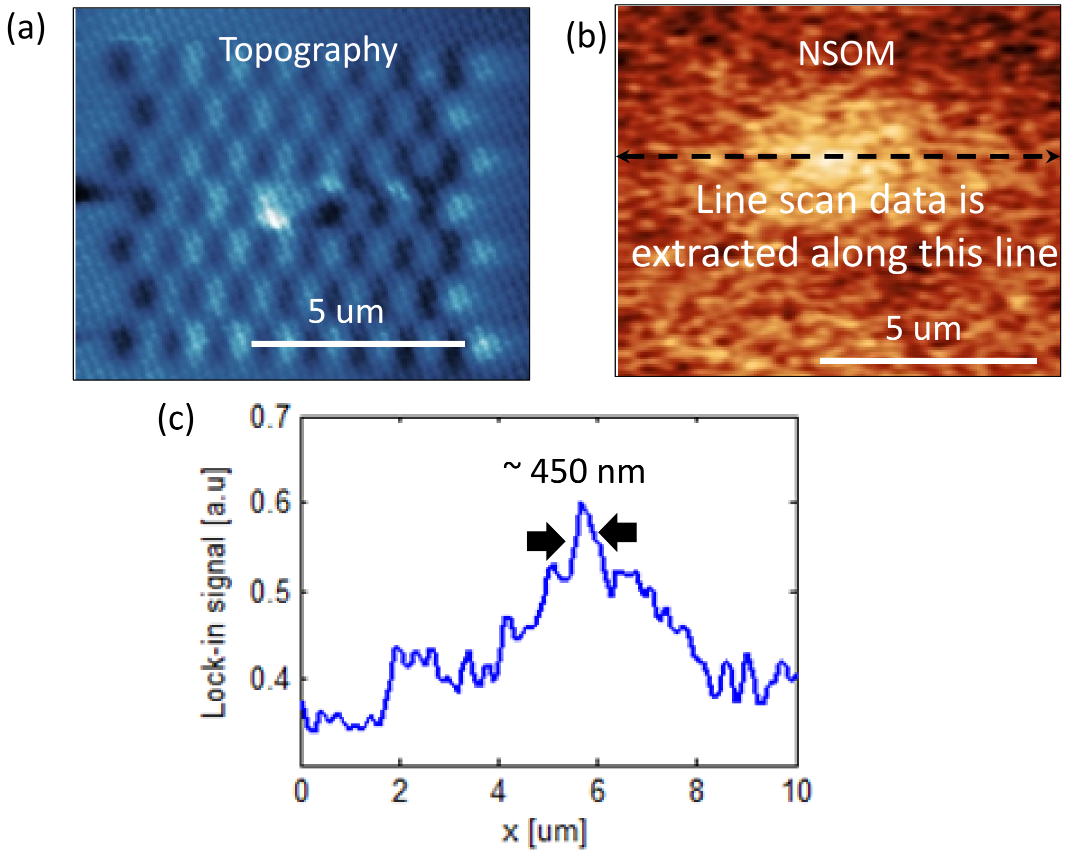

Figure 5 – (a) – (b) Topography and NSOM images of the photonic crystal integrated QCL. (b) Line scan of the NSOM image along the while cross-sectional line shown in (b). The optical mode gets squeezed and the FWHM is ~ 600nm.

The optical mode gets squeezed within a very small volume region. The line scan of the NSOM image shows that the full width at half maximum (FWHM) for the squeezed mode is ~ 450nm. This is 10 times smaller than the operating lasing wavelength. Tuning the silicon dioxide buffer thickness can squeeze the optical mode even further. We are currently working on many different photonic crystal structures and it has been discussed in the section 6.

5. FUTURE WORK

As been shown from the design and experiment, MDM photonic crystal integrated on an mid-infrared room temperature working quantum cascade laser can squeeze the optical mode within a volume 10 times smaller than the operating wavelength. The confinement volume can be reduced further by improvisation of the design. Currently we are working on various different photonic crystal designs as shown in Figure 6.

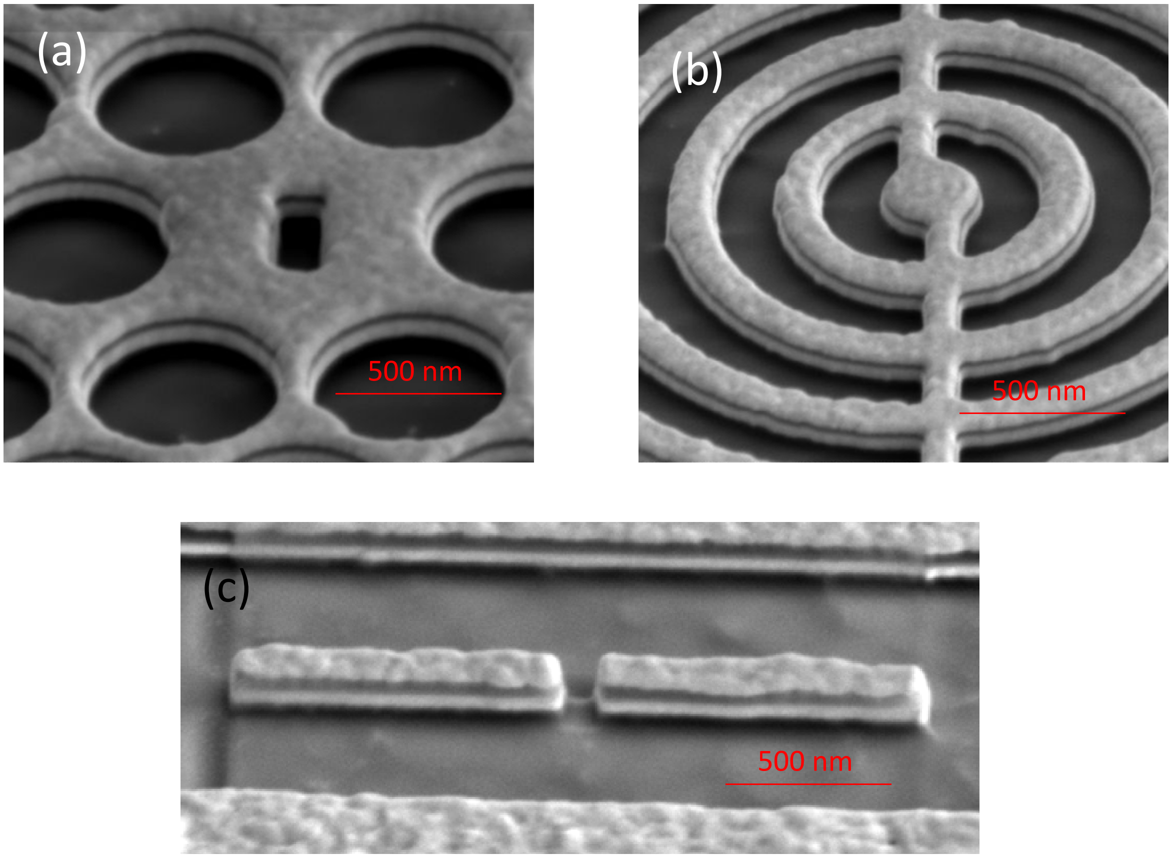

Figure 6 - (a) Photonic crystal structure with a slit instead of a hole defect at the center (b) An MDM bull's eye design fabricated on the facet of QCL. (c) An MDM coupled nanorod antenna fabricated on the facet of QCL.

6. DISCUSSION

We have successfully integrated a photonic crystal structure on the facet of a quantum cascade laser working in the mid-infrared region of optical spectrum. The integrated photonic crystal didn’t affect the operation of laser substantially other than reduction of threshold voltage due increase in reflectivity. Finite-difference time domain (FDTD) simulations have that within a periodicity (Λ) between 1.3um and 1.4um, where the radius of the hole and cavity radius are 0.45 and 0.2 times Λ, the near field enhancement at the center of the cavity on the same level as the top metal surface can be as high as 10 times the incident field. MDM photonic crystal has been fabricated on the facet of QCL using focused ion beam (FIB). The integrated device has been tested using an apertureless mid-infrared near field scanning optical microscopy (a-NSOM). The measurement set-up is based on an inverted microscope coupled with a commercially available Atomic Forced Microscopy (AFM). Using this technique, we could simultaneously measured topography and NSOM image of the photonic crystal while the QCL is in operation. It showed that the combination of high quality factor and extremely low mode volume of the PC design can squeeze the optical mode within a spot size of diameter 450nm. We believe such squeezed spot size can be extremely useful for bio-sensing applications.

7. ACKOWLEDGEMENT

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[1] J.D. Joannopoulos, S. G. Johnson, J. N. Winn and R. D. Meade. “*Photonic Crystals : Molding the flow of light*” Second edition, Princeton University Press, NJ (2008)

[2] H. Kosaka, T. Kawashima, A. Tomita, T. Sato and S. Kawakami. “*Photonic crystal spot-size converter*” App. Phys. Lett. **76**, 268 (2000)

[3] I. Moreno, J. J. Araiza and M. A. Alejo. “*Thin film spatial filters*” Opt. Lett. vol **30**, pp. 914-916 (2005)

[4] C. Martelli, J. Canning, .t Gibson, and S. Huntington, "*Bend loss in structured optical fibres*," Opt. Exp. **15**, 17639-17644 (2007)

[5] T. Yoshie, J. Vučković, A. Scherer, H. Chen, and D. Deppe. “*High quality two-dimensional photonic crystal slab cavities*” Appl. Phys. Lett. **79**, 4289 (2001)

[6] O. Painter, R. K. Lee, A. Scherer, A. Yariv, J. D. O'Brien, P. D. Dapkus and I. Kim. “*Two dimensional photonic band-gap defect mode laser ,*” Science **284**, 1819 (1999)

[7] R. M. Gelfand, L. Bruderer and H. Mohseni. “*Nanocavity plasmonic device for ultrabroadband single molecule sensing*” Opt. Lett. **34**, 1087 (2009)

[8] B. Song, S. Noda, T. Asano, Y. Akahane. “*Ultra-high–Q photonic double-heterostructure nanocavity*,” Nat. Mater. **4**, 207 (2005)

[9] F. Vollmer and S. Arnold. “Whispering*–gallery–mode biosensing: label-free detection down to single molecules*,” Nature Methods **5**, 591 (2008)

[10] M. Dragoman, D. Dragoman, “*Plasmonics: Applications to nanoscale terahertz and optical devices*, “Prog. in Quantum Electron. **32**, 1 (2008)

[11] J. Faist**,** F. Capasso , D. L. Sivco , C. Sirtori , A. L. Hutchinson , and A.Y. Cho.”*Quantum Cascade Laser*” Science **264**, 553 (1994)

[12] Y.Bai, S. Slivken , S. Kuboya , S. R. Darvish and M. Razeghi. “*Quantum cascade laser that emits more light than heat,*”Nature Photonics **4**, 99 (2010)

[13] P. Q. Liu, A. J. Hoffman, M. D. Escarra, K. J. Franz, J. B. Khurgin, Y. Dikmelik, X. Wang, J.-Y. Fan and C. F. Gmachl. “*High power efficient quantum cascade*,” Nat. Photonics **4**, No. 98 (2010)

[14] Y Bai, S Slivken, SR Darvish, M Razeghi. “Room *temperature continuous wave operation of quantum cascade laser with 12.5% wall-plug efficiency*,” Applied Physics Letter **93**, 021103 (2008)

[15] J. Kim and K.-B. Song. “*Recent progress of nano-technology with NSOM*,” Micron*,* **38**, 409 (2006)

[16] E. D. Palik. “*Handbook of Optical constants of solids* ”. Academic Press, NY (1985)

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[17] D.Dey, R. M. Gelfand, J. Kohoutek, A. Bonakder and H. Mohseni. "*Quantum cascade laser integrated with metal-dielectric-metal antenna*" to be appeared in SPIE proceeding (2010)

[18] L. Diehl, D. Bour, S. Corzine, J. Zhu, G. Hofler, M. Loncar, M. Troccoli and F. Capasso. "High-temperature continous wave operation of strain-balanced quantum cascade lasers grown by metal organic vapor-phase epitaxy," *Appl. Phys. Lett.* 83, 3245-48 , Aug. 2003

[19] B. Knoll and F. Keilmann. "*Enhanced dielectric contrast in scattering-type scanning near-field optical microscopy*". Optics Communication **182**, 321 (2000)

[20] R. Hillenbrand, B. Knoll and F. Keilmann."*Pure optical contrast in scattering-type scanning near-field microscopy*" Journal of Microscopy **202**, 77-83 (2000)

[21] H. C. Liu. "*Intersubband transition in Quantum wells: Physics and device application II*" Academic Press (1999)

[22] R. Rang, A. C. Jones, F. Zhou, Z.Li, B. J. Wiley, Y. Xia and M. B. Raschke. "*Optical Near0field mapping of plasmonic nanoprisms*" Nano Letter **8**, 3357-3363 (2008)