Design and fabrication of photonic crystal resonators for single-mode and vertical surface emission from strain-compensated quantum cascade lasers operating at 4.32 μm

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Photon crystal resonators with the C4v symmetry were designed and fabricated on quantum cascade lasers with a strain-compensated multiple quantum well to achieve single-mode and vertical surface emission at 4.32 μm. Their fabrication accuracy was confirmed by high-resolution reflection spectroscopy. The maximum output power was 10 mW at 77 K. A far-field pattern with a small divergence angle below 1 degree was observed. Its main peak had a donut shape, which was attributed to the spatial symmetry of the resonance mode of the photonic crystal.

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Quantum cascade lasers (QCLs) are regarded as the most promising candidates for the future light sources for various types of gas sensing and analysis, because their emission spectral range covers 3 to 8 μm, where vibrational absorption peaks of important gas molecules are included. In addition to their role as an alternative to conventional thermal light sources, QCLs are expected to enable extremely sensitive analyses at the ppb level based on the multi-pass technique, and the detection of toxic chemical species such as volcanic gasses from distant places. For these applications, a good beam profile with a small divergence angle and a high output power are required.

However, the conventional QCLs of the edge-emission type suffer from large divergence angles, relatively low beam quality and relatively low output power. Although the surface-emission technique known as vertical cavity surface emitting laser (VCSEL), which has been developed for semiconductor lasers of the interband transition type, generally resolves these problems, it is not applicable to QCLs because the light emission from their active region composed of multiple quantum wells (MQW), which is caused by intersubband transitions, is TM (transverse magnetic) polarized. For this reason, another technique known as photonic crystal surface emitting laser (PCSEL) has been applied to QCLs, since the photonic crystal (PC) can materialize laser resonators with a big output aperture and a sufficiently large quality factor for both transverse electric (TE) and TM polarizations. The effective area of the active region of the QCL, which is limited by the size of the PC, can be of a centimeter order, so we can also expect a high output power.

The performance of the QCL with the PC structure, which we hereafter call PC-QCL, specifically depends on the resonance frequency and the Q factor of the PC resonator, the spatial overlap of the resonance mode and the active region, and the extraction efficiency of the laser light. In addition, the resonance mode of the PC should be that of the point of the first Brillouin zone, since the vertical emission can be achieved only for this case. To meet all these requirements, we need a precise design and fabrication of the PC in addition to the high-quality fabrication of the QCL device.

In this paper, we report on the design and fabrication of PC resonators together with the fabrication of the QCL device with a strain-compensated MQW operating at 4.32 μm. We confirmed the fabrication accuracy of the PC by high-resolution reflection spectroscopy and achieved a single-mode and vertical surface emission by the PC-QCL. Although PC-QCLs operating at 8.5 and 8.75 μm have already been reported, this study is the first report, as far as the authors know, on the single-mode vertical-emission PC-QCL operating in the 4 μm region, for which the careful strain compensation of the MQW is required to achieve a higher intersubband transition frequency.

Figure 1 is a schematic illustration of our PC-QCL device. The refractive index, n, of each layer taking into consideration the imaginary part due to impurity doping, which was obtained by the data base integrated in the QCL simulator, Erwin Jr, is also shown in Fig. 1. The main parts of the PC-QCL are MQW and PC regions sandwiched by two InP cladding layers. The 5.0 μm thick lower cladding layer and the MQW were successively grown on a S-doped InP substrate by solid-source molecular beam epitaxy (MBE). For the fabrication of the layer, a 1.0 μm thick InGaAs layer was grown by MBE and etched by electron beam lithography to form a square array of circular InGaAs pillars (n = 3.401 + 6.28 × 10−14 i). Then the gap between adjacent pillars was filled with InP (n = 3.074 + 1.98 × 10−3 i) by metal-organic vapor phase epitaxy (MOVPE) to improve heat conduction. The 3.0 μm thick upper cladding layer was also fabricated by MOVPE. The details of the fabrication of the MQW and PC regions are as follows.

For the MQW region, we followed the design by Evans et al. It consists of 30 stages of strain-compensated In0.669Ga0.331As/Al0.638In0.362As quantum wells/barriers with a residual strain of around 1%. We slightly modified...
the thickness of the semiconductor layers from their original structure, which was designed for an emission at 4.7 μm, to meet the characteristic absorption around 4.3 μm by CO₂ molecules, the key chemical species of global warming. The total thickness of the MQW was 1.6 μm and its averaged refractive index was 3.263 + 1.20 × 10⁻⁴ i. Figure 2(a) shows the X-ray diffraction profile of the epi-wafer thus fabricated, where the measured profile is compared with a simulation by LEPTOS. Because of their good agreement, we can conclude that we achieved the strain compensation of the MQW layers as we designed.

To materialize the resonance wavelength of 4.3 μm, the PC structure was designed using the finite element method (FEM) with the commercial software COMSOL. We adopted the circular pillar structure so that the PC had the C₄ᵥ symmetry, which offers polarization selection rules for reflection peaks to make the mode assignment easy. The height and the radius of the InGaAs pillar were fixed at 0.8 μm and 0.551 μm, respectively, while the lattice constant of the PC, which we denote by a, was varied to adjust the resonance wavelength. Figure 2(b) is a cross-sectional SEM image of our specimen, by which we confirmed the fabrication of the periodic PC structure and the absence of unintended defects such as voids.

We calculated the eigenfrequency and eigenfunction on the Γ point of the first Brillouin zone by imposing the periodic boundary condition on the surface of the PC unit cell in the lateral directions and the perfectly matched layer absorbing boundary condition in the vertical direction. As we show in the following, we achieved the target resonance wavelength with a around 1.38 μm.

Table I shows the wavelength, Q factor and overlap factor of TM-like modes on the Γ point for a = 1.380 μm whose eigenfrequency lies in the gain spectrum of the QCL. In this table, E, A₂ and B₂ denote the spatial symmetry of their magnetic field, and are irreducible representations of the C₄ᵥ point group. The Q factor includes the contributions from the radiation loss through the substrate and the Joule loss in the semiconductor layers and the upper metallic electrode.

The overlap factor is defined by the ratio of the electric field intensity distributed in the MQW region to the total intensity. Because the overlap factor is considerably large (>0.56) for all modes, these TM-like modes are well localized in the MQW region and we may conclude that their coupling to the electronic transition is sufficiently strong. Because the B₂ mode has the largest Q factor and the largest overlap factor, we may expect that the laser action takes place with this mode. Because only the E mode can radiate into the direction normal to the PC surface, the lasing with the B₂ mode may result in an oblique emission. This point will be discussed later.

We examined the accuracy of the PC fabrication by observing their angle-resolved reflection spectra with our home-made high-resolution setup integrated in a commercial FT-IR (JASCO 6800). We measured the reflection spectra of the PC-QCL specimens from the top side before filling the air gap in the PC with InP, since the eigenmodes in the PC can be accessed most efficiently in this geometry.

Figure 2(c) shows an example of the reflection spectrum, which was measured for the PC-QCL with a = 1.390 μm from the normal direction. An angle resolution of 0.3 deg was achieved by carefully preparing an unfocused incident light beam. The black arrows show the frequencies of TE- and TM-like modes of the E symmetry on the Γ point calculated by FEM, which are 2245 cm⁻¹ and 2253 cm⁻¹, respectively. The measured peak frequencies are 2241 and 2255 cm⁻¹. Their discrepancy was less than 0.2%. So, we can conclude that the accuracy of the PC fabrication was pretty good. The symmetry assignment of the observed peaks was made by observing the angle-resolved reflection. For more details of the measurement, please see Ref. 31.

After the air gap in the PC region was buried with InP and the upper cladding layer was formed, a 0.1 μm thick InGaAs contact layer was grown by MOVPE. Then, the laser mesa, whose areal size was approximately 500 μm × 500 μm, was formed by dry-etching from the contact layer to the surface of the lower cladding layer. The sidewalls and top edges of the laser mesa were covered with 200 nm thick SiO₂ for electric
insulation. The epi-side (top side) of the PC-QCL was completely covered with a Ni/Au electrode. On the rear surface of the InP substrate, the emission aperture was surrounded by a 100 μm wide Ni/Au electrode. Although Ni caused a considerable amount of light absorption, which was disadvantageous to lasing, we used it to improve the adhesion between the Au and InGaAs layers. The PC-QCL device was mounted with its epi-side down on a CuW heat sink by indium solder.

The lasing performance was examined at 77 K. Figures 3(a) and 3(b) show the light-current-voltage (LIV) characteristics and the emission spectrum of our PC-QCL with \(a = 1.38 \mu m\). To measure the output power, we placed a thermal power meter (Ophir, model 3 A thermal sensor) just in front of the PC-QCL, which was driven by pulsed currents with a pulse width of 300 ns and a duty of 1.5%. The threshold current was 1.9 A and the threshold current density was 760 A cm\(^{-2}\). The voltage at the lasing threshold was

### Table I. Characteristics of the TM-like modes on the \(\Gamma\) point for the PC-QCL specimen with \(a = 1.380 \mu m\).

| Mode | Wavelength (\(\mu m\)) | Q factor | Overlap factor |
|------|------------------------|----------|---------------|
| E    | 4.4504                 | 2778     | 0.527         |
| A\(_2\) | 4.4408               | 3072     | 0.602         |
| B\(_2\) | 4.4318               | 3307     | 0.636         |

**Fig. 2.** (Color online) (a) X-ray diffraction profile of the epi-wafer fabricated on the InP substrate by MBE (b) SEM image of the PC-QCL. (c) Normal-incidence reflection spectrum of the PC-QCL specimen \((a = 1.39 \mu m)\) without the InP capping layer or the Ni/Au electrode.
14.5 V and the series resistance was 2 Ω. The maximum output power was 10 mW. The output power can be increased by thinning the substrate. In this study, we used a 650 μm thick InP substrate doped with sulfur ($3 \times 10^{18}$ cm$^{-3}$), whose optical absorption was approximately 70% at 4.32 μm. By thinning the InP substrate down to 200 μm, for example, we can decrease the absorption and increase the output power by two times.

On the other hand, the emission spectrum was measured with an FT-IR spectrometer (Thermo Scientific, Nicolet 8700) with a resolution of 0.5 cm$^{-1}$. As shown in Fig. 3(b), the emission spectrum had a sharp single peak located at 4.32 μm, which was somewhat shorter than the designed PC resonance frequency of 4.4318 μm. There was a discrepancy of 2.6% between the designed resonance wavelength (4.4318 μm) and the observed lasing wavelength (4.32 μm). This discrepancy was mainly caused by the decrease in the InGaAs pillar radius during the pretreatment of the InGaAs surface with a hydrogen gas before the regrowth of the InP layer by MOVPE, which decreased the volume fraction of InGaAs in the PC region to reduce its averaged refractive index and increase its resonance frequency.

Figure 4(a) is the far-field pattern (FFP) of the emission intensity measured with a lens-less mid-IR camera (NEC, IRV-T0831) located 26.8 mm away from the output aperture of the PC-QCL, which clearly shows a vertical surface emission with a small divergence angle of less than 1 deg. There were some side lobes in the FFP, but their intensity was much smaller than the central peak. Together with the clear threshold and the sharp single emission peak in Fig. 3, we can conclude that we achieved the single-mode surface emission with our PC-QCL. We also observed a decrease in the lasing wavelength with decreasing lattice constant of the PC, which agreed with the behavior of the resonance frequency calculated by FEM, so we can also conclude that the vertical laser emission took place with a resonance mode on the Γ point in the first Brillouin zone.

The central peak of the FFP had a donut shape, which was clearly observed in the magnified three-dimensional picture shown in Fig. 4(b). The peak emission angle was tilted by approximately 0.25 deg from the normal direction. This feature can most probably be attributed to the spatial symmetry of the PC resonance mode.

As we have already mentioned, there were three eigenmodes on the Γ point in the frequency range of the gain spectrum (see Table I). The B$_2$ mode had the largest quality factor and the largest overlap factor, so we may assume that the lasing took place with this mode. Because the exact vertical emission is forbidden for the B$_2$ mode by symmetry, the emission through the output aperture cannot be observed if the PC structure is perfectly periodic. However, the actual PC has finite lateral dimensions, so its in-plane wavevector has an ambiguity $\Delta k$, which is determined by the size of the PC, $L$, as $\Delta k = \pi/L$. Then, the wavevector of the B$_2$ mode on the Γ point is not exactly

![Fig. 3.](a) LIV characteristics of the PC-QCL specimen and (b) its emission spectrum at 77 K.)

![Fig. 4.](a) The far-field radiation pattern of the PC-QCL specimen and (b) its magnified three-dimensional view.)
equal to zero, but has a distribution up to $\Delta k$, which results in a distribution of the emission angle up to $\lambda/2L$ (rad), where $\lambda$ is the lasing wavelength. For the present case, this emission angle ambiguity is 0.247 deg, which is very close to the observed peak emission angle and supports our interpretation. However, the emission efficiency of the $E_1$ mode must be much lower than the $E$ mode. We plan to improve this point by lowering the symmetry of the PC unit cell in our future studies.

In summary, we designed and fabricated a PC resonator on a QCL with a strain-compensated InGaAs/AlInAs MQW to achieve a single-mode and vertical surface emission at 4.32 $\mu$m. The far-field beam profile was a donut shape with a peak emission angle of 0.25 deg, which was attributed to the spatial symmetry of the resonance mode on the $\Gamma$ point of the first Brillouin zone. Although the output power was 10 mW at 77 K, we should be able to achieve greater power by thinning the substrate and reducing the symmetry of the PC unit cell.

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1) J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, Science 264, 553 (1994).
2) M. Beck, D. Hofstetter, T. Aellen, J. Faist, U. Oesterle, M. Ilgerns, E. Gini, and H. Melchior, Science 295, 501 (2002).
3) S. Slivken, A. Evans, J. David, and M. Razeghi, Appl. Phys. Lett. 81, 4321 (2002).
4) A. Evans, J. S. Yu, S. Slivken, and M. Razeghi, Appl. Phys. Lett. 85, 2166 (2004).
5) F. K. Tittel, Y. Bakhirkin, A. A. Kosterev, and G. Wysocki, Rev. Laser Eng. 34, 275 (2006).
6) A. Macikawa, Y. Shiomi, M. Uchida, and T. Ikarino, Proc. SPIE 10111, 1011106 (2017).
7) K. Iga, F. Koyama, and S. Kinoshita, IEEE J. Quantum Electron. 24, 1845 (1988).
8) K. Iga, IEEE J. Sel. Top. Quantum Electron. 6, 1201 (2000).
9) K. Takaoka, M. Ishikawa, and G. Hatakoshi, IEEE J. Sel. Top. Quantum Electron. 7, 381 (2001).
10) K. Iga, Jpn. J. Appl. Phys. 57, 08PA01 (2018).
11) K. Iga, Proc. SPIE 11263, 1126302 (2020).
12) C. Sirtori, F. Capasso, J. Faist, D. L. Sivco, A. L. Hutchinson, and A. Y. Cho, Appl. Phys. Lett. 66, 4 (1995).
13) J. Faist, Quantum Cascade Lasers (Oxford University Press, Oxford, 2013).
14) M. Imada, S. Noda, A. Chutinan, T. Tokuda, M. Murata, and G. Sasaki, Appl. Phys. Lett. 75, 316 (1999).
15) S. Noda, M. Yokoyama, M. Imada, A. Chutinan, and M. Mochizuki, Science 293, 1123 (2001).
16) E. Taylor, S. Khamas, R. A. Hogg, N. Ikeda, and Y. Sugimoto, Jpn. J. Appl. Phys. 51, 02BG05 (2012).
17) K. Hirose, Y. Liang, Y. Kurosaka, A. Watanabe, T. Sugiyama, and S. Noda, Nat. Photon. 8, 406 (2014).
18) S. Noda, K. Kitamura, T. Okino, D. Yasuda, and Y. Tanaka, IEEE J. Sel. Top. Quantum Electron. 23, 4900107 (2017).
19) R. Morita, T. Inoue, M. De Zoya, K. Ishizaki, and S. Noda, Nat. Photon. 15, 311 (2021).
20) R. Colombelli, K. Srinivasan, M. Troccoli, O. Painter, C. F. Gamchi, D. M. Tennant, A. M. Sergent, D. L. Sivco, A. Y. Cho, and F. Capasso, Science 302, 1374 (2003).
21) Y. Bai, B. Gokden, S. R. Darvish, S. Slivken, and M. Razeghi, Appl. Phys. Express 14, 102003 (2021).