InGaAs/AlInAs quantum cascade laser sources based on intra-cavity second harmonic generation emitting in 2.6-3.6 micron range


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ABSTRACT

We discuss the design and performance of quantum cascade laser sources based on intra-cavity second harmonic generation operating in at wavelengths shorter than 3.7 μm. A passive heterostructure tailored for giant optical nonlinearity is integrated on top of an active region and patterned for quasi-phasematching. We demonstrate operation of λ≈3.6 μm, λ≈3.0 μm, and λ≈2.6 μm devices based on lattice-matched and strain-compensated InGaAs/AlInAs/InP materials. Threshold current densities of typical devices with nonlinear sections are only 10-20% higher than that of the reference lasers without the nonlinear section. Our best devices have threshold current density of 2.2 kA/cm² and provide approximately 35 μW of second-harmonic output at 2.95 μm at room temperature. The second-harmonic conversion efficiency is approximately 100 μW/W². Up to two orders of magnitude higher conversion efficiencies are expected in fully-optimized devices. Keywords: quantum cascade lasers, second harmonic generation, short wavelength, room temperature, intersubband, giant nonlinear susceptibility, quasi-phase matching

1. INTRODUCTION

InGaAs/AlInAs/InP quantum cascade lasers (QCL) have recently been developed into reliable high-power sources that operate continuous-wave (CW) at room-temperature (RT) in the spectral range 3.7-12 μm [1]. Their growth and fabrication process is compatible with telecommunication diode lasers production lines which makes manufacturing cost efficient. The spectral range 2.5-12 μm is called ‘molecular fingerprint region’; it contains a large number of molecular absorption lines and is highly important for chemical sensing. Widely-tunable QCL sources have been developed to address spectroscopic needs in this region. Examples include an external-cavity QCL tunable between 7.6 μm and 11.4 μm reported in Ref. [2] and a QCL source based on an array of distributed-feedback (DFB) devices with frequency output variable between 8 μm and 9.8 μm [3]. A 2.5-3.7 μm portion of the ‘molecular fingerprint region’ contains a number of absorption lines important for chemical sensing and spectroscopy. Ideally, one would want to have a device that can provide spectral coverage of the whole ‘molecular fingerprint region’. However, the operation of InGaAs/AlInAs/InP QCLs at 2.5-3.7 μm spectral range suffers from inter-valley scattering, even when highly strained heterostructures are used [4]. Diode lasers [5] and interband cascade lasers [6] can operate CW at λ=2.5-3.7 μm but they cannot operate at RT at longer wavelengths. In addition, their growth and fabrication process is not compatible with that of telecommunication diode lasers. Heterostructures based on InAs/AlSb [7] and GaInAs/AlAsSb/InP [8] materials may be used to extend the operation of QCLs to the wavelengths shorter than 3.7 μm, but these devices are yet to demonstrate CW operation at RT and their fabrication is also not compatible with that of telecommunication diode lasers. We note that most spectroscopic application require a narrowband CW laser source with only about a milliwatt of output power.
Intra-cavity second-harmonic generation (SHG) has long been suggested as a mean to extend the spectral range of QCLs to shorter wavelengths [9-14]. However, none of these lasers operated at RT. Here we discuss an approach [15] for producing short-wavelength, high-performance QCL sources based on intra-cavity SHG. Our method allows extending the spectral range of RT CW InGaAs/AlInAs/InP QCLs to \(\lambda=2.5-3.7\) \(\mu\text{m}\). We believe that these devices may provide a cost-effective solution for spectroscopic applications in the 2.5-3.7 \(\mu\text{m}\) range, and may lead to broadband QCL chips with spectral output tunable in the whole 2.5-10 \(\mu\text{m}\) spectral range and beyond (using both SHG light and fundamental light). We discuss the performance of our proof-of-principle QCLs operating at \(\lambda=2.6, 3.0\) and 3.6 \(\mu\text{m}\).

2. DEVICE DESIGN

The details of our devices design are shown in Fig. 1(a). Devices are grown on InP substrates using either lattice-matched or strain-compensated InGaAs/AlInAs heterostructures. A 400-600nm-thick nonlinear layer (NL) is grown epitaxial on top of a QCL active region. The NL contains multiple repetitions of a multi-quantum-well structure designed to have giant resonant optical nonlinearity associated with intersubband transitions [16]. A portion of the NL is then patterned into a ~50%-duty-cycle grating for quasi-phase-matching (QPM) [12,17] for SHG between TM00 modes. QPM allows us to generate SHG output in TM 00 mode, which is important for practical applications. The separation of an active region and a NL allows us to optimize the two elements independently.

The NL is passive and increases optical losses in the laser cavity. However, the losses can be reduced to manageable values if we remove most of the NL to leave only a small section (200-400 \(\mu\text{m}\) long in the case of our devices) near the output facet as shown in Fig. 1(a). The upper waveguide cladding is then overgrown. We note that these processing steps are similar to those used for fabricating distributed feedback QCLs [18]. Devices are then processed into ridge-waveguide QCLs with 10-12 \(\mu\text{m}\)-wide ridges following standard procedure for.

To understand the expected device performance, we consider the laser structure designed for 3.6 \(\mu\text{m}\) SHG emission [15]. The laser is grown on an InP substrate, n-doped to 1×10\(^{17}\) cm\(^{-3}\). First a 1.6-\(\mu\text{m}\)-thick active region, consisting 30 repetitions of a 2-phonon-resonance QCL structure [19], is grown; then a 400-nm-thick NL is grown on top of the active region. The NL consists of 24 repetitions of the structure depicted in Fig. 1(b). After NL patterning, the top waveguide cladding layers (3.5\(\mu\text{m}\) of InP doped to 5×10\(^{16}\) cm\(^{-3}\), followed by 0.5\(\mu\text{m}\) of InP doped to 3×10\(^{18}\) cm\(^{-3}\)) are re-grown. Devices are processed into 10\(\mu\text{m}\)-wide and 3mm-long ridge lasers. The intensity loss, \(\alpha_{NL}\) and SHG nonlinearity (\(\chi^{(2)}\)) for TM-polarized light in the NL can be calculated using well-known expressions for intersubband absorption and optical nonlinearity [20]. Using the transition dipole moments shown in Fig. 1(b), the doping level in the NL, and assuming a typical transition linewidth of 20meV full width at half maximum (FWHM), we obtain \(\alpha_{NL}(\omega)=630\text{cm}^{-1}\) and \(\alpha_{NL}(2\omega)=230\text{cm}^{-1}\) for fundamental (\(\lambda=7.2\mu\text{m}\)) and SHG (\(\lambda=3.6\mu\text{m}\)) light, respectively, and \(\chi^{(2)}=2.2\times10^{-4} \text{pm/V}\). The loss

![Fig. 1. (a) Schematic of the devices design and processing steps. Top panel shows a longitudinal cross section a QCL structure without top waveguide cladding. The nonlinear layer (shown in black) is positioned on top of the active region. The bottom panel shows a longitudinal cross section of the device with part of the nonlinear section removed, patterned for quasi-phasmorphching of the SHG process, and then top waveguide cladding regrown. (b) Conduction band diagram of one period of a nonlinear structure for 7.2\(\mu\text{m}\) to 3.6\(\mu\text{m}\) frequency conversion. The layer sequence (in nm) is 9.0/2.0/1.1/4.6 where AlInAs barriers are shown in bold and InGaAs wells are shown in regular font. The center 3 nm of a 9 nm barrier is n-doped to 3×10\(^{17}\) cm\(^{-3}\). Shown are energy levels, transition energies, and transition dipole moments.](image_url)
for the TM$_{00}$ mode in the waveguide with the nonlinear grating, $\alpha_{\text{TM00GR}}$ is calculated to be $\alpha_{\text{TM00GR}}(\omega)=40\text{cm}^{-1}$ and $\alpha_{\text{TM00GR}}(2\omega)=13\text{cm}^{-1}$ for fundamental and SHG frequencies, respectively. TM$_{00}$ modal loss at $\lambda=7.2\mu\text{m}$ in the laser waveguide without nonlinearity, $\alpha_{\text{TM00LAS}}(\omega)$, is calculated to be $9.6\text{cm}^{-1}$. The total round-trip distributed loss ($\alpha_{\text{tot}}$) for a TM$_{00}$ laser mode at $\lambda=7.2\mu\text{m}$ is then given as

$$\alpha_{\text{tot}}(\omega)=\alpha_m+\alpha_{\text{TM00GR}}(\omega)+\alpha_{\text{TM00LAS}}(\omega)\alpha_{\text{tot}}(2\omega)=17.7\text{cm}^{-1}.$$  

where $\alpha_m$ is the mirror loss, $L_{\text{GR}}$ is the length of the nonlinear grating section, and $L_{\text{LAS}}$ is the length of the laser section without optical nonlinearity. For a TM$_{00}$ mode in a 3mm-long device with 400$\mu$m nonlinear section we obtain $\alpha_m=4\text{cm}^{-1}$ and $\alpha_{\text{tot}}(\omega)=17.7\text{cm}^{-1}$. In comparison, the same calculation gives $\alpha_{\text{tot}}(\omega)=13.6\text{cm}^{-1}$ for a device of the same length without any nonlinearity. Thus, with proper design, the addition of the nonlinear section is only expected to increase the total loss in our devices by ~30%.

The SHG power output is given as [9,17,21]

$$W(2\omega)=\frac{(2\omega)^2}{8E_0c^3 n(2\omega)n(\omega)n(\omega)} \left| \frac{\chi^{(2)}}{\pi} \right|^2 \frac{(W(\omega))^2}{S_{\text{eff}}^2} \times I_{\text{eff}}^2,$$  

where $W(\omega)$ is the pump power at the entrance of the nonlinear section, $W(2\omega)$ is the SHG power at the end of the nonlinear section, $n(\omega)$ is the refractive index at frequency $\omega$, $|\chi^{(2)}/\pi|$ is the effective nonlinearity for the QPM process [17], and $S_{\text{eff}}$ is the effective area of the beam’s overlap with the nonlinear region [3,14]. We estimate $S_{\text{eff}}$ to be approximately $5\times10^3\ \mu\text{m}^2$ in our devices. The parameter $L_{\text{eff}}$ is the effective nonlinear interaction length, which includes the effect of optical absorption of both fundamental and SHG waves, and is given as:

$$I_{\text{eff}}^2 = \frac{e^{-\alpha_{\omega} L_{\text{eff}}^2} \left( e^{-\frac{(\alpha_{\omega}-\alpha_{\omega_2}) L_{\text{eff}}}{2}} - 1 \right)^2}{\left( \alpha_{\omega} - \alpha_{\omega_2}/2 \right)^2},$$  

where $\alpha_{\omega}$ and $\alpha_{\omega_2}$ are the net modal intensity losses for the fundamental and SHG waves in the section of the device with the nonlinear grating, and $L_{\text{GR}}$ is the length of the nonlinear section. Equation 3 assumes a perfect first-order QPM [15] of the SHG process. We can estimate $L_{\text{eff}}$ by taking $\alpha_{\omega}=\alpha_{\text{TM00GR}}(\omega)-g_{\text{TM00}}(\omega)=22\text{cm}^{-1}$, where $g_{\text{TM00}}(\omega)=\alpha_{\text{tot}}(\omega)=17.7\text{cm}^{-1}$ (cf. Eq. (1)) is the modal gain, and $\alpha_{\omega_2}=\alpha_{\text{TM00GR}}(2\omega)=13\text{cm}^{-1}$. We then obtain $L_{\text{eff}}=230\mu\text{m}$ for a nonlinear grating length $L=400\mu\text{m}$ and the SHG internal conversion efficiency $\eta_{\text{int}}=W(2\omega)/(W(\omega))^2=2.3\text{mW/W}^2$ for 3mm-long devices with 400$\mu$m-long nonlinear sections. We may also define the ‘external conversion efficiency’ ($\eta_{\text{ext}}$) for the same device that links SHG power output through the front facet with fundamental power output through the front facet:

$$\eta_{\text{ext}} = \eta_{\text{int}} \frac{T_\omega}{T_{\omega_2} e^{-2\alpha_{\omega_2} L_{\text{eff}}}} \approx 19 \text{ mW/W}^2,$$  

where $T_\omega=0.7$ is power transmission through the front facet for fundamental and SHG light. Given typical Watt-level power output of modern QCLs [1], we may expect to generate several milliwatts of more of SHG output in our devices, when QPM and the nonlinear layer structure are fully optimized.

### 3. PROOF-OF-PRINCIPLE DEVICES PERFORMANCE

The results obtained with three of our proof-of-principle device designs are presented below. The power-current (L-I) characteristics for fundamental output for a $\lambda=3.6\mu\text{m}$ SHG device at room temperature and 80L are shown in Fig. 2(a). The laser was 3mm long, had a high reflectivity coating on the back facet, and had a 400$\mu$m-long nonlinear grating section (the longest available in our processing run) near the front facet. Also shown in Fig. 2(a) are the current-voltage (I-V) characteristic of the same device and the L-I curve for a 3mm-long reference laser without a nonlinear layer. Fig. 2(b) shows L-I's for SHG light for the best-performing 3mm-long devices with 400$\mu$m-long nonlinear section at room temperature and 80K. Fundamental power measurements were performed with a thermopile detector, SGH power measurements were performed with a calibrated InSb detector using optical filters to reject fundamental light. The data was corrected for an approximately 40% collection efficiency of our optical setup and the filters transmission. The data in Fig. 1(a) indicates that RT threshold current density for a reference laser was nearly identical to that of the device with a 400$\mu$m-long nonlinear section. This indicates that the nonlinear section adds less than 2 $\text{cm}^{-1}$ of optical loss to $\alpha_{\text{tot}}$ cf. Eq. (4). This number is smaller than the 4 $\text{cm}^{-1}$ estimated theoretically, which is likely because the energy levels in the
nonlinear section is not in exact resonance with the fundamental frequency. RT emission spectra for fundamental and SHG light of our devices are shown in Fig. 2(c). The spectra were taken using a Fourier-transform infrared spectrometer in rapid-scan mode. Using the fundamental power output of the devices without the nonlinear section we estimate the internal conversion efficiency of our device with the nonlinear section to be approximately 0.1W/W² at RT. This value is over a factor of 20 smaller than 2.3mW/W² estimated theoretically. This is most likely due to the fact that the energy levels in the NL are not fully resonant with fundamental and SHG transitions and thus the value of $\chi^{(2)}$ in the nonlinear section is significantly reduced. We note that if the actual energy position of level 3 is 30meV below the calculated value, the optical nonlinearity is reduced by a factor of 3.5 and the conversion efficiency is reduced by a factor of 12. Careful adjustment of the NL design for a given active region design is needed to produce devices with optimal SHG conversion efficiency.

SHG conversion efficiency is highly dependent on the QPM grating period in the NL. This is shown in Fig. 3, where we tested SHG conversion efficiencies of devices with QPM grating period in the range 24 μm to 37 μm. Devices with 31-μm-grating period displayed the highest SHG conversion efficiency. This is in close agreement with our theoretical estimates that predict the phase mismatch ($\Delta k=2k_{\omega}k_{2\omega}$, where $k_{\omega}$ and $k_{2\omega}$ are the propagation constants for fundamental and SHG modes) between TM$_{00}$ modes of 2100 cm$^{-1}$ (The QPM grating period is then $\Lambda=2\pi/\Delta k=30\mu m$ [17]).

Fig. 3. (a) Dependence of $\eta_{ext}$ on grating period for 3mm-long lasers with 170μm-long nonlinear section. For a given laser, the conversion efficiency varies as a function of pump current because of changes in the laser mode structure and emission wavelength. Plotted are the average values of $\eta_{ext}$ for each laser with error bars indicating the range in which $\eta_{ext}$ varies with current.
Using ~1% strain-compensated InGaAs/AlInAs/InP heterostructures one could extend the spectral range of InGaAs/AlInAs SHG QCLs to wavelengths as short as 2.5 μm. To demonstrate that, we present our results obtained with two proof-of-principle devices designed for SHG emission at λ = 2.95 μm and λ = 2.6 μm.

Figure 4(a) shows the LI characteristics of fundamental and SHG emission from a device designed for SHG emission at λ = 3.0 μm. Also shown there is the LI characteristic of the device without the nonlinear section. The laser is based on an In₀.₆₇Ga₀.₃₃As/Al₀.₅₇In₀.₄₃As heterostructure with 2.3-μm-thick active region and 460-nm-thick NL based on a bandstructure design similar to that shown in Fig. 1(b). The lasers were 3-mm-long with 400-μm-long nonlinear sections patterned for QPM. Devices operated at RT with threshold current density of 2.2 kA/cm² and produced over 35 μW of SHG power output. Low threshold current density indicates that the lasers may be operated CW at RT when processed into buried heterostructures. RT emission spectra of a typical device are shown in Fig. 4(b). Threshold current density of devices with the nonlinear section was similar to that of devices without the nonlinear section although threshold current density increase of ~30% was expected for devices with the nonlinear section. Similarly to the λ = 3.6 μm devices discussed above, this indicates that additional adjustment of the NL structure may be needed to make χ(2) fully resonant for the SHG process. More details of the device design and performance will be presented elsewhere [22].

![Fig. 4](image)

Fig. 4. (a) RT LI characteristics for fundamental output from a λ≈2.95μm SHG device with and without a nonlinear section (left axis, solid and dashed lines, respectively) and the LI characteristic for SHG power output (solid line, right axis). Devices were operated in pulsed mode with 100 kHz 50 ns pulses (b) Spectrum of RT SHG output from the device with the nonlinear section in (a). Inset: spectrum of the fundamental output from the same device.
Figure 5 shows the emission spectra of a $\lambda=2.6\mu$m SHG QCL with NL based on In$_{0.65}$Ga$_{0.35}$As/Al$_{0.55}$In$_{0.45}$As heterostructure. The laser provided over 1mW of SHG output at 80K and approximately 10$\mu$W of SHG output at RT. The laser consisted of an approximately 1.5-$\mu$m-thick active region and an approximately 800-nm-thick nonlinear layer based on a bandstructure design similar to that shown in Fig. 1(b). Details of the device design and performance will be presented elsewhere [23].

4. CONCLUSION AND OUTLOOK

In conclusion, we discussed a new design architecture for short-wavelength InGaAs/AlInAs QCL sources based on intracavity SHG and demonstrated a operation of proof-of-principle devices with emission wavelengths in the range 2.6-3.6$\mu$m. Our best devices provide over 35$\mu$W of SHG output at $\lambda\approx2.95\mu$m at RT with threshold current density of only 2.2kA/cm$^2$. Our theoretical and experimental results indicate that RT CW operation of the proposed SHG QCLs is possible with proper device thermal packaging. The SHG power output of these devices is expected to improve to provide conversions efficiency of over 2mW/W$^2$. Given multi-Watt-level power output of current state-of-the-art QCLs, SHG QCLs presented in this work may produce over 10s of mW of output in the spectral range 2.5-3.7$\mu$m, when fully optimized.

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