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13. ABSTRACT (Maximum 200 words)  The overall goal of DARPA PWASSP is the development of innovative Quantum Cascade (QC) lasers that are spectrally, spatially and temporal agile, and to deliver them for applications in sensing and signal processing. Significant advances in several fields closely aligned with these goals have been made, namely in the development of faster, more spectrally agile, and generally novel QC lasers, as outlined in the following in more detail.				
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## **Summary of accomplishments (Cumulative):**

The overall goal of DARPA PWASSP is the development of innovative Quantum Cascade (QC) lasers that are spectrally, spatially and temporal agile, and to deliver them for applications in sensing and signal processing. Significant advances in several fields closely aligned with these goals have been made, namely in the development of faster, more spectrally agile, and generally novel QC lasers, as outlined in the following in more detail.

### **High-speed modulated, high frequency and short pulse QC lasers**

QC lasers, being based on intersubband transitions in semiconductor quantum wells, are characterized by ultrafast (picosecond) carrier lifetimes. An important consequence of this unique property is the expected absence of relaxation oscillations in the transient response of these devices. This predicted absence of relaxation oscillations has indeed been experimentally verified by measuring the modulation response of several 8- $\mu\text{m}$ -QC lasers, properly processed and packaged for high-speed operation, up to 10 GHz.

Picosecond self-mode-locked pulses from mid-infrared QC lasers, at wavelengths within the important molecular fingerprint region, have been developed. These devices are based on intersubband electron transitions in semiconductor nanostructures, which are characterized by some of the largest optical nonlinearities observed in nature and by picosecond relaxation lifetimes. The results are interpreted with a model in which one of these nonlinearities, the intensity-dependent refractive index of the lasing transition, creates a nonlinear waveguide where the optical losses decrease with increasing intensity. This favors the generation of ultrashort pulses, because of their larger instantaneous intensity relative to continuous-wave emission.

Active mode locking of a high-speed 8  $\mu\text{m}$  QC laser in a monolithic configuration is demonstrated, at a repetition rate of 11.6 GHz. Evidence of mode locking is obtained from the measured optical spectra and corresponding interferograms, as well as from the power spectra of the photocurrent detected with a fast quantum-well infrared photodetector. An estimate for the pulse width of approximately 5 ps is inferred from the experimental results. Mode-locked operation is observed up to a maximum temperature of over 120 K.

The temporal performance of mode-locked QC lasers is predominantly dependent on the shape and extent of the optical gain spectrum, which is generally narrow in QC lasers, unless it is designed to be broad.

Such broadband QC lasers were first optimized for continuous wave operation by iteratively improving and “flattening” their gain spectra. The improved quantum designs display a gain ripple of only about  $4\text{ cm}^{-1}$  over more than a 0.5- $\mu\text{m}$  spectral range. Simultaneous continuous wave emission at several wavelengths spanning the range between 6.7 and 7.4  $\mu\text{m}$  has been achieved in the temperature interval from 20 to 77 K.

These improved, broadband QC lasers were then employed to demonstrate improved high-speed short-pulse operation using active and passive mode-locking schemes. Active mode locking in broadband QC lasers with a repetition rate of about 14.3 GHz has been achieved through the modulation of the laser bias current. At low drive currents, active mode locking in broadband QC lasers resembles mode locking in single-wavelength QC lasers, while at high drive currents, the mode locking properties are governed by the broad spectral gain of these lasers. At high bias currents, the active modulation excites Fabry–Perot modes across the entire gain spectrum from 6.7 to 7.4  $\mu\text{m}$ , with clear evidence of mode locking. The spectral width of the optical gain in the broadband QC lasers exceeds 2 THz and indicates the potential for generating subpicosecond pulses.

Using intracavity optical nonlinearities in QC lasers, the pulse width of mode locked QC lasers was then measured as 12 ps; these were narrow, single-wavelength gain spectra QC lasers with homogeneously broadened gain spectra. The onset of self-mode locking in QC lasers with built-in optical nonlinearity results in a significant increase of the SHG signal. The value of 12 ps is in good agreement with the pulse duration deduced from the optical spectral width.

Down-conversion of the detector signal by heterodyning with an RF signal allows the direct observation of the pulsed laser emission in the time domain and reveals a stable train of pulses characteristic of mode-locked lasers, when homogeneously broadened QC lasers were brought to mode-locking. Mode-locking on multiple wavelengths from inhomogeneously broadened QC lasers, indicated multiple pulse trains, as inferred from the spectral characteristics of broadband mode-locked QC lasers as well as from their heterodyne beats.

### **Spectrally agile QC lasers**

Spectral agility of QC lasers includes their ability to be developed for an extremely wide wavelength range including the far-infrared and THz wavelength regime, their ability to be designed for multi-wavelength and broadband operation, as well as their spectral purity and tuning characteristics.

QC lasers operating at 19, 21, and 24  $\mu\text{m}$  have been developed. Pulsed operation was obtained up to 140 K and with a peak power of a few milliwatts at cryogenic temperatures. Laser action originates from interminiband transitions in “chirped” superlattice active regions. The waveguides are based on surface-plasmon modes confined at a metal–semiconductor interface. The wavelengths were chosen in order to avoid major phonon absorption bands, which are particularly strong at energies just above the reststrahlen band. A 21.5- $\mu\text{m}$ -wavelength laser based on a two-sided interface-plasmon waveguide was also developed.

Continuous wave laser action has been achieved in a superlattice QC device operating on surface plasmon waveguide modes. The emission wavelength of  $\sim 19 \mu\text{m}$  is the longest ever reported for continuous wave III-V semiconductor lasers.

Intersubband electroluminescence is reported in a quantum-cascade structure based on asymmetric superlattice active regions and designed for emission in the THz range  $\sim 80 \mu\text{m}$ . Comparison with a structure based on a “vertical transition” in a single quantum well shows an increased full width at half maximum  $\sim 2.8$  vs.  $0.9$  meV of the emission line. In both cases the dependence of the optical power on the injected current is linear or sublinear and remains in the pW range.

A QC laser with a heterogeneous cascade containing two substacks previously optimized to emit at  $5.2 \mu\text{m}$  and  $8.0 \mu\text{m}$  wavelengths, respectively, is presented. The low-temperature performance of the two-wavelength laser is comparable to the respective homogeneous stack lasers, indicating no penalty from the heterogeneity of the cascade. Each substack is apportioned the optimum fraction of the applied bias. This demonstrates the general applicability of this scheme. In addition, an etch-stop layer inserted between the two substacks allowed fabrication of a “tap” into the cascade. The latter was used to selectively manipulate the laser threshold of one substack, turning the  $8.0 \mu\text{m}$  laser on and off while the adjacent  $5.2 \mu\text{m}$  QC laser was operating undisturbed.

This development of two-wavelength operation was further developed into the concept of inhomogeneously broadened, ultra-broadband QC lasers. The fundamental mechanism behind laser action leads in general only to narrowband, single-wavelength emission. Several approaches for achieving spectrally broadband laser action have been put forward, such as enhancing the optical feedback in the wings of the gain spectrum, multi-peaked gain spectra, and the most favored technique at present, ultrashort pulse excitation. Each of these approaches has drawbacks, such as a complex external laser cavity configuration, a non-flat optical gain envelope function, or an inability to operate in continuous mode, respectively. Here a monolithic, mid-infrared “supercontinuum” semiconductor laser has been developed by adopting a QC configuration, where a number of dissimilar intersubband optical transitions are made to cooperate in order to provide broadband optical gain from  $5$  to  $8 \mu\text{m}$  wavelength. Laser action with a Fabry-Perot spectrum covering all wavelengths from  $6$  to  $8 \mu\text{m}$  simultaneously is demonstrated with this approach. Lasers that emit light over such an extremely wide wavelength range are of interest for applications as varied as terabit optical data communications or ultra-precision metrology and spectroscopy.

Measurements performed at Pacific Northwest National lab (Dr. John Schultz et al.) determined the intrinsic frequency fluctuations of two single-mode QC distributed-feedback lasers operating continuously at a wavelength of  $8.5 \mu\text{m}$ . A Doppler-limited rovibrational resonance of nitrous oxide was used to transform the frequency noise into measurable intensity fluctuations. The QC lasers, along with recently improved current controllers, exhibit a free-running frequency stability of  $150$  kHz over a  $15$ -ms time interval.

The predicted small linewidth enhancement factor of QC lasers was measured in outside collaboration (Prof. Shun-Lien Chuang at UIUC) and confirmed to be extremely small; the measured values were  $\sim -0.5$  without corrections for thermal shifts, and  $\pm 0.2$  with corrections for thermal effects. The latter is the generally accepted definition of the linewidth enhancement factor, and within the measurement error is practically zero. The small linewidth enhancement is an essential result for any application that requires modulating the QC laser output. The linewidth enhancement factor was deduced from Hakki-Paoli measurements, which also yielded the gain and the group effective index.

Recently, the spectral agility of QC lasers has been significantly enhanced by expanding into the field of nonlinear QC lasers. Using intracavity optical nonlinearities, emission frequencies can be doubled (as in second harmonic generation (SHG)) or otherwise manipulated.

An efficient intracavity nonlinear interaction of laser modes in a specially adapted QC laser has been developed. A two-wavelength QC laser structure emitting at wavelengths of  $7.1$  and  $9.5 \mu\text{m}$  included cascaded resonant optical intersubband transitions in an intracavity configuration leading to resonantly enhanced sum-frequency and second-harmonic generation at wavelengths of  $4.1$ ,  $3.6$ , and  $4.7 \mu\text{m}$ , respectively. Laser peak optical powers of  $60$  and  $80$

mW resulted in 30 nW of sum-frequency signal and 10–15 nW of second-harmonic signal, both in good agreement with theoretical calculations in the first demonstration of this new approach.

An about a 100-fold improvement of SHG in a QC laser with integrated optical nonlinearity was subsequently obtained by including phase-matching considerations in the design of the deep-etched ridge waveguide. The waveguide layer structure was optimized to minimize the phase mismatch of the zero-order mode of the fundamental light with the second-order transverse mode of the second-harmonic light. Exact phase matching is made possible by the faster decrease of the modal refractive index of the fundamental light with decreasing ridge width relative to the refractive index of the second-harmonic light. Up to 240  $\mu\text{W}$  of the second-harmonic power and a nonlinear power conversion efficiency of up to 36  $\text{mW}/\text{W}^2$  were achieved.

These improved SHG QC lasers, which recently allowed 2  $\mu\text{W}$  of nonlinear light output, employed similarly recently developed improved nonlinear quantum design. Nonlinear optical cascades of resonantly coupled intersubband transitions with giant second-order nonlinearities were integrated with each QC-laser active region. QC lasers with three-coupled quantum-well (QW) active regions showed up to 2  $\mu\text{W}$  of SHG light at 3.75  $\mu\text{m}$  wavelength at a fundamental peak power and wavelength of 1 W and 7.5  $\mu\text{m}$ , respectively. These lasers resulted in an external linear-to-nonlinear conversion efficiency of up to 1  $\mu\text{W}/\text{W}^2$ . An improved 2-QW active region design at fundamental and SHG wavelengths of 9.1 and 4.55  $\mu\text{m}$ , respectively, resulted in a 100-fold improved external linear-to-nonlinear power conversion efficiency, i.e. up to 100  $\mu\text{W}/\text{W}^2$ . Full theoretical treatment of nonlinear light generation in QC lasers is given, and excellent agreement with the experimental results is obtained.

These devices were also used to test the spectral and temperature properties of the nonlinear light output. SHG has been measured from 10 up to 250 K heat sink temperature, with about 1  $\mu\text{W}$  of nonlinear power at 10 K, and about 50 nW at 250 K. Single-mode and tunable SHG at 3.5  $\mu\text{m}$  wavelength has been measured from single-mode QC distributed feedback lasers operating at the fundamental pump wavelength of 7.0  $\mu\text{m}$ . Thermal tuning results in a tuning rate for the SHG emission of  $\sim 0.2$  nm/K for temperatures above  $\sim 100$  K.

Aside from SHG, also third harmonic generation was investigated. An InGaAs/AlInAs QC laser based on a three-well diagonal transition active region with an integrated third-order nonlinear oscillator has been designed. The device displayed pump radiation at  $\lambda \sim 11.1$   $\mu\text{m}$  and third order nonlinear light generation at  $\lambda \sim 3.7$   $\mu\text{m}$  as well as second harmonic generation at  $\lambda \sim 5.4$   $\mu\text{m}$ .

### **New QC laser designs**

In this area several different directions were explored and new inventions and developments achieved and reported. The various areas can roughly be divided into three major sub-fields, namely new quantum designs of QC lasers, new characterization methods for QC lasers, and new resonator designs.

A quantum-cascade laser using a double quantum-well graded superlattice (SL) as the active region has been developed. Each SL period consists of two strongly coupled quantum wells resulting in the splitting of the lowest miniband into two minibands. These two minibands can be designed to be flat and to contain delocalized, spatially symmetric wavefunctions under an applied electric field which in turn leads to a high optical dipole for the interminiband transition. In addition, the new design allows independent control of the energy levels of the lowest two minibands, their width and the splitting separating them, enhancing design flexibility. Using a cascade design of 55 pairs of alternated active regions and injectors, pulsed laser action is achieved at  $\sim 11.6$   $\mu\text{m}$ .

An “injectorless” QC laser has been developed based on these new double-quantum well SL designs. The prior requirement of using injector regions to transport electrons from the lower laser level and other low-lying energy levels of one active region to the upper laser level of the next electron-downstream active region was eliminated by using an appropriately designed double-quantum-well “chirped” superlattice active region. The major advantage of the “injectorless” QC laser is the close packing of the active regions and the concomitant large optical confinement factor. Using a cascade of 75 consecutive active regions, a laser was designed and demonstrated for emission at  $\sim 11.5$   $\mu\text{m}$ .

A dual-wavelength QC laser with an interdigitated cascade has been presented. Aside from providing two-wavelength operation at 8.0 and 9.5  $\mu\text{m}$  wavelength, this laser design was used to test the role of extrinsic carriers in the injectors. An interdigitated cascade was grown with undoped injectors bridging 9.5 and 8.0  $\mu\text{m}$  active regions, but doped injectors bridging 8.0 and 9.5  $\mu\text{m}$  active regions. Clear laser action on both wavelengths demonstrates that doping of all injector regions is not a firm requirement for QC lasers. Comparison with a conventionally doped interdigitated cascade QC laser shows a threshold reduction by a factor of approximately 2 for the laser based on the active regions preceded by the undoped injector. This can be understood from the absence or strong reduction of impurity scattering related to the dopant ions.

Aside from new designs in the InGaAs/AlInAs on InP material system, also new active region designs in new material systems were explored. The first Al-free InP/InGaAs QC structures, with InP and InGaAs as barrier and well materials, respectively, were demonstrated. Electroluminescence emission at  $\sim 12 \mu\text{m}$  from two different structures was observed and compared in light of their characteristic different band structure designs.

A technique has been developed which allows the observation of intersubband spontaneous emission in unipolar QC lasers above threshold. The technique consists of cleaving the laser stripe in the direction perpendicular to its facets. This does not negatively affect the operation of the lasers thanks to their unipolar nature. The new method has been applied to superlattice QC lasers with various active region designs. The saturation of the luminescence intensity at the laser transition is directly measured, and a bottleneck effect for transitions separated from the lasing one by less than one optical phonon is confirmed. This technique helps in the optimization of QC laser design.

The local temperature of QC lasers operating in continuous wave mode is reported. This information is extracted from the thermal shift of the band-to-band photoluminescence peaks in the AlInAs and InP cladding layers of QC laser facets using a high-resolution microprobe setup. Interpolation by means of a two-dimensional heat diffusion model allows obtaining the temperature profile and the thermal conductivity in the waveguide core. Comparison between substrate and epilayer-side mounted lasers shows the superior thermal dissipation capability of the latter, and explains their better performance with respect to threshold current and maximum operating temperature.

The nonequilibrium optical phonons population associated with electron transport in QC lasers has been measured and reported. The phonon occupation number was measured in the range 75–280 K by using a combination of microprobe photoluminescence and Stokes/anti-Stokes Raman spectroscopy. The excess phonon population is observed to decrease as the lattice temperature increases. From the nonequilibrium phonon population, interface phonon lifetimes of 5 ps at 75 K and 2 ps at 280 K were extracted.

The group refractive index dispersion in ultra-broad-band QC lasers has been determined using Fabry–Pérot spectra obtained by operating the lasers in continuous wave mode below threshold. In the wavelength range of 5–8  $\mu\text{m}$ , the global change of the group refractive index is as small as  $\sim 8.2 \times 10^{-3} \mu\text{m}^{-1}$ . Using the method of Hakki and Paoli, the subthreshold gain of the lasers has furthermore been measured as a function of wavelength and current. At the wavelength of best performance, 7.4  $\mu\text{m}$ , a modal gain coefficient of  $16 \text{ cm} \times \text{kA}^{-1}$  at threshold and a waveguide loss of  $18 \text{ cm}^{-1}$  have been estimated.

The facet temperature profile and the thermal resistance of operating QC lasers have been assessed using a microprobe band-to-band photoluminescence technique. Substrate-side and epilayer-side-mounted QCLs based on GaInAs/AlInAs/InP and GaAs/AlGaAs material systems have been compared. The dependence of the thermal resistance on the continuous wave or pulsed injection conditions and its correlation with the output power have been studied. These results were used as inputs for a two-dimensional heat-diffusion model which gives the heat fluxes and the thermal conductivity of the active regions, in order to design QC lasers with improved thermal properties.

Several new device geometries incorporating new waveguide designs and layouts have been developed. The first QC laser amplifier has been demonstrated. It was used to obtain high power single-mode emission at 7.4  $\mu\text{m}$  from a QC distributed feedback laser, together with enhanced beam quality. Laser and amplifier are directly coupled in a master oscillator power amplifier configuration. Peak optical powers of 0.5 W at 80 K have been obtained. Ninety percent of the total power is thereby emitted within a divergence of  $20^\circ$  in the lateral direction. The device showed single mode operation with a side mode suppression ratio of 30 dB in the temperature range from 10 to 280 K. This allowed tuning of the emission wavelength in the range from 7.36 to 7.46  $\mu\text{m}$ . The estimated peak amplifier gain is 6.4 and 4.9 dB at 80 and 300 K, respectively, and the cavity losses are  $12.5$  and  $22 \text{ cm}^{-1}$  at the corresponding temperatures.

QC lasers with double metal-semiconductor waveguide resonators have been developed for operating wavelengths of 19, 21, and 24  $\mu\text{m}$ . The waveguides are based on surface-plasmon modes confined at the metal-semiconductor interfaces on both sides of the active region/injector stack and are not restricted by a cutoff wavelength for the TM polarized intersubband radiation. The double metal-semiconductor resonator devices are fabricated using an epilayer transfer process. Optical confinement factors close to 1 are obtained, with low waveguide losses. The performance of the devices is compared with that of QC lasers based on single-sided surface-plasmon waveguides. The concept of QC laser with double metal-semiconductor waveguide is of course applicable to a much wider wavelength range.

Two-wavelength QC distributed feedback (QC-DFB) lasers based on a heterogeneous-cascade two-wavelength active waveguide core and a multi-sectioned cavity featuring two different Bragg gratings have been demonstrated.

Optimized lasers display single mode emission at 5.0 and 7.5  $\mu\text{m}$  simultaneously and a tunability on both modes equal to single-wavelength QC-DFB lasers.

Finally, the last year also saw the important development of the first Photonic Crystal QC laser. Photonic and electronic band structure engineering was combined to create a surface-emitting QC microcavity laser. A high-index contrast two-dimensional photonic crystal was used to form a micro-resonator that simultaneously provides feedback for laser action and diffracts light vertically from the surface of the semiconductor surface. A top metallic contact allows electrical current injection and provides vertical optical confinement through a bound surface plasmon wave. The miniaturization and tailorable emission properties of this design are potentially important for sensing applications, while electrical pumping can allow new studies of photonic crystal and surface plasmon structures in nonlinear and near-field optics. As a preamble to the demonstration of the QC photonic crystal laser several essential photonic crystal manufacturing techniques were developed and demonstrated.

The developments of DARPA PWASSP have directly or indirectly led to a bibliography that includes more than 60 articles in reviewed international scientific journals, more than 100 contributed or invited presentations at national and international workshops, conferences, and meetings, and several patents.

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