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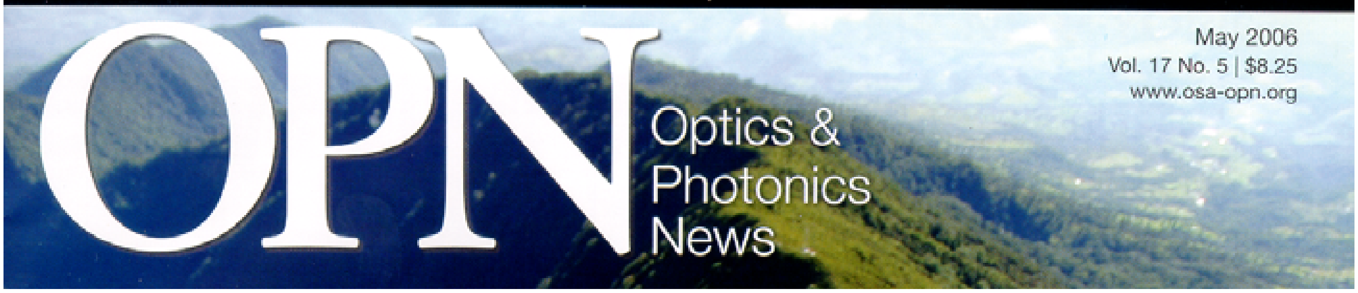


Continuous-Wave, Room-Temperature

Quantum Cascade LASERS

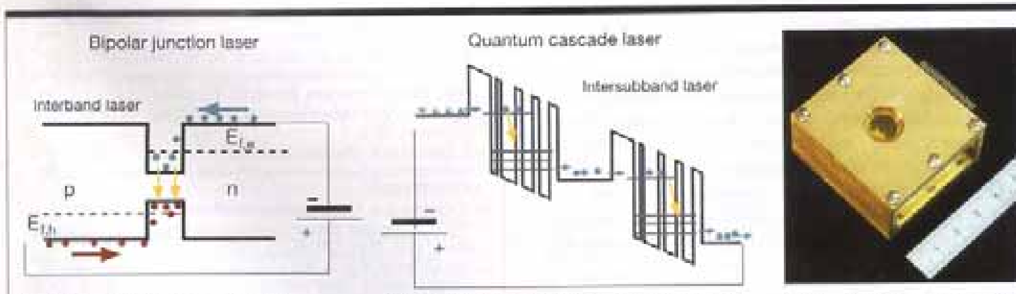
Jerome Faist

Quantum cascade lasers are now capable of continuous-wave, room-temperature, single-frequency operation with large powers. This progress may be translated into a number of applications, including medical breath analysis and environmental monitoring.

A nose can be a remarkably sensitive instrument, as any dog owner can attest. The nasal cavity is lined with cells equipped to detect tiny amounts of substances and send that information to the brain. But noses have their limits: They cannot pick up scents remotely; they are susceptible to cross-sensitivities; and they cannot discern the isotopic content of a chemical.

Chemical sensing based on infrared spectroscopy, on the other hand, can be both very sensitive and selective. This technique takes advantage of the fact that molecules behave like small mechanical oscillators with well-defined resonance frequencies that are defined not only by their chemistry but also by their isotope content.

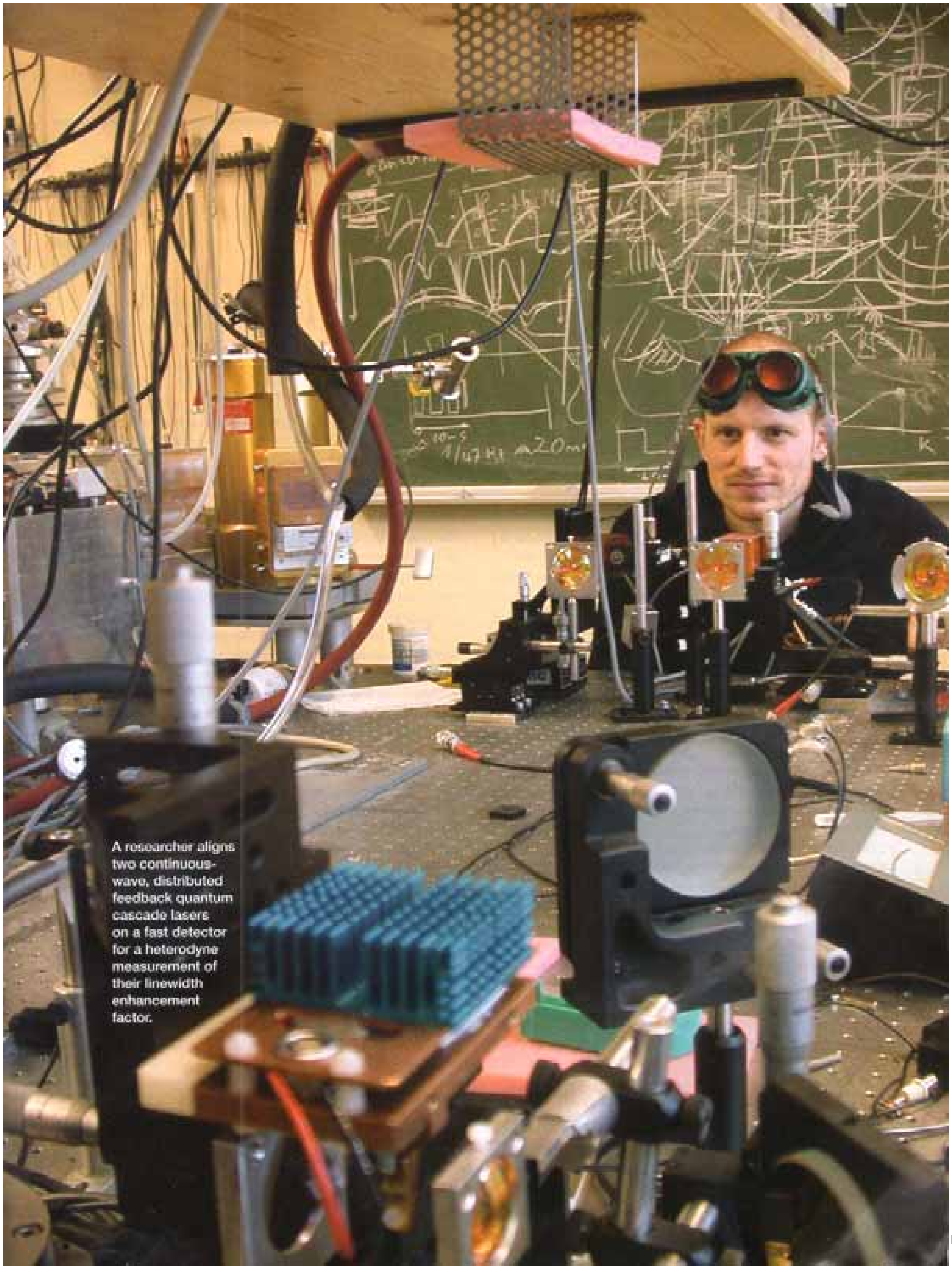
A little more than a decade ago, the concept of widespread chemical sensing using an optical method seemed impossible. The right optical source required for the task simply did not exist. Or, to be more accurate, the available source—the lead-salt laser—was flawed in a number of ways: It required cryogenic cooling, tended to suffer from poor reliability and was difficult to manufacture in large volume. Today, these problems have been largely addressed by the quantum cascade laser. Rather than generating light using a radiative recombination between conduction band electrons and valence band holes across a forbidden bandgap, this device uses transitions between subbands created by quantum confinement in a semiconductor heterostructure.



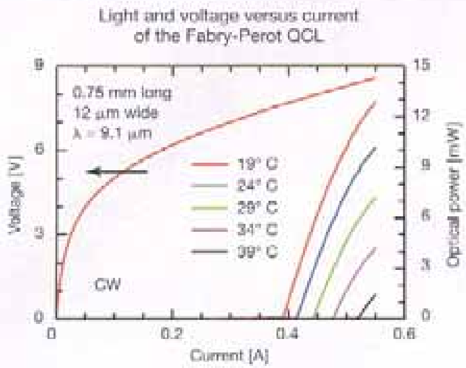
Light is emitted by recombination of electrons and holes.

Electrons flow through the structure, emitting photons between confined states. One electron may therefore emit a large number of photons.

Laser mounted in a laboratory Peltier-cooled enclosure. The ruler unit is centimeters.

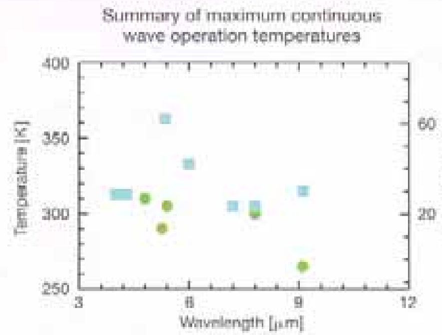


A researcher aligns two continuous-wave, distributed feedback quantum cascade lasers on a fast detector for a heterodyne measurement of their linewidth enhancement factor.



Data from quantum cascade lasers operating in continuous wave at 9.1 μm.

Adapted from the data presented in Beck et al.



From the data published in the literature for Fabry-Perot lasers (squares) and distributed feedback quantum cascade lasers (full discs).

Data from Northwestern University, Harvard University, Alpes Lasers and the University of Neuchâtel.

The quantum cascade laser seemed an unlikely contender at first. Although it had the advantages of being manufacturable using InP-based or GaAs-based III-V materials, which are widely used in optoelectronics, and of being able to cover a wide frequency range using the same heterostructure material, the upper state lifetime was limited by the optical phonon emission; it was a fraction of a picosecond at room temperature—which would seem to limit its range of operation to cryogenic temperatures.

The solution to this problem was not to find a way to increase the upper state lifetime, but rather to mitigate its negative effects. A first step was to use new active region architectures to maintain a population inversion even at high temperatures and under strong optical fields. Architectures had to be found that could maintain a very short lower state lifetime, and therefore a very high electron extraction efficiency, even at high temperatures.

To accomplish this, Hofstetter and colleagues introduced two states separated each by an optical phonon energy below the lower state of the laser transition. This double resonance strongly reduced the lower state population, as was shown quantitatively in transport simulations. Another solution was to replace the ladder of two states by a miniband (see Faist et al., 2001). These so-called two-phonon or bound-to-continuum approaches are the basis of most of today's high-performance devices.

A second step was to reduce the waveguide losses by fabricating buried heterostructures. In a buried heterostructure device, the active region is literally "buried" by a non-conducting layer of InP, creating a low-loss lateral waveguide. These innovations led to the first demonstration of continuous wave operation at room temperature at the wavelength of 9 μm in 2002 (see Beck et al., 2002).

In order to reach shorter wavelengths, one must first prevent carrier leakage above the barriers and therefore find a material with a larger band discontinuity than lattice-matched InGaAs/AlInAs. One very successful approach was to use strain-compensated InGaAs/AlInAs, where the compressive strain in the InGaAs is compensated by the tensile strain in the AlInAs. This allows for an increase of the band discontinuity from about 520 meV to more than 710 meV in 1 percent compressive/tensile strained material (see Hofstetter et al., 2001 and Faist et al., 1998).

In 2003, Yu and colleagues also achieved a room-temperature continuous wave at shorter wavelengths and very large output powers by reducing the doping in the active region and ridge width. Most promising is the achievement of a maximum operating temperature of 90° C by Evans and colleagues at Northwestern University as well as Yu et al.'s development of a continuous wave at a wavelength of 4 μm, which was described in a 2006 issue of *Applied Physics Letters*.

To achieve the single frequency required by chemical sensing applications, one must use a technology very similar to the one used in telecommunication lasers, in which a grating is etched in the active region. These so-called distributed feedback

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quantum cascade lasers (DFB-QCLs) were first developed in Bell Laboratories by Gmachl and coworkers. They can now operate in single mode in continuous wave at room temperature with an optical power slightly below that achieved by Fabry-Perot devices.

One can obtain up to 50 mW of single mode optical power —more than enough for the vast majority of sensing applications. The frequency range of operation (about 1900 cm^{-1}) is well adapted to NO spectroscopy. Frequency tuning is achieved by varying the active region temperature. At this wavelength, the tuning coefficient is 0.12 cm^{-1}/K, mostly due to the change of refractive index with temperature. This translates into a tuning range of about 6 cm^{-1} if the temperature of the Peltier is changed directly between -30° C and 20° C, or about 2 cm^{-1} if the current is changed at a fixed operating temperature.

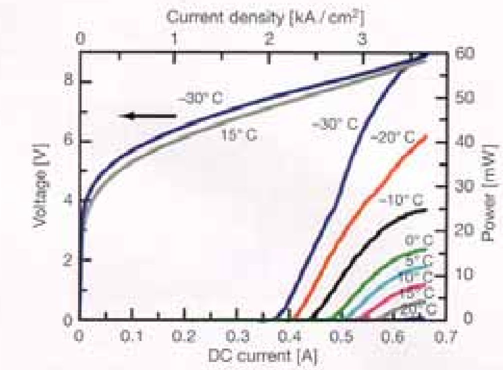
Because of their narrow linewidth (a few megahertz) and large optical powers, continuous wave DFB-QCLs are extremely well adapted to gas spectroscopy applications. The potential of this technology was recognized very early by spectroscopists who quickly adapted the sensing techniques used with other lasers sources to the quantum cascade laser. Scientists have demonstrated chemical sensors using cavity ring-down spectroscopy (Kosterev et al.), integrated cavity output spectroscopy (Bakhirkin et al.), long multipass cells (Nelson et al.), photoacoustic detection (Nagele et al.) and Faraday detection (see Ganser et al.). In some cases, sensitivities were reached in the range of a few part per billion.

In these systems, the advantages of the quantum cascade laser are its very low amplitude noise and high optical power, its long-term stability and its ability to be frequency modulated up to about 100 MHz. An example of such a chemical sensing experiment is shown in the figure on the right.

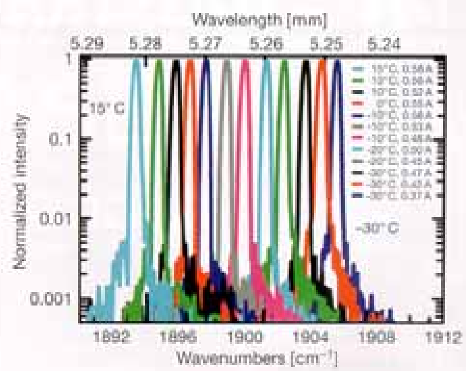
That experiment, which was conducted by Aerodyne Corporation, was a long-term monitoring of NO on the rooftop of the company premises (located close to a major highway) over 25 days of unattended operation. Researchers used a DFB-QCL that was operated in a continuous wave in conjunction with a 76 m multipass absorption cell tuned to a strong line of NO. The data demonstrate the fluctuation of the NO concentration as a function of time as traffic and wind patterns change. A sensitivity below 0.l ppb over a 1 s integration time was estimated for this experiment.

The tuning range of a DFB-QCL covers one or two absorption lines of a gas. However, some applications call for the spectroscopy of multi-component gas or solids that require a much larger tuning range. Fortunately, the intersubband transitions

Optical power versus injected current of a continuous wave DF-QCL



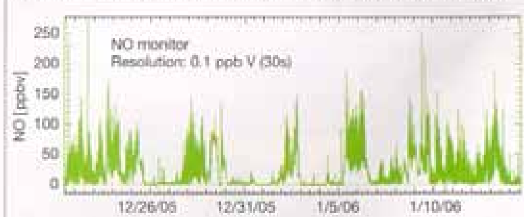
Single mode emission of the device



Emission for various temperatures and currents. The operation frequency (1900 cm^{-1}) is well adapted to spectroscopy of NO.

Data courtesy of Stephane Blaser and Lubos Hvozdara, Alpes Lasers (www.alpeslasers.ch)

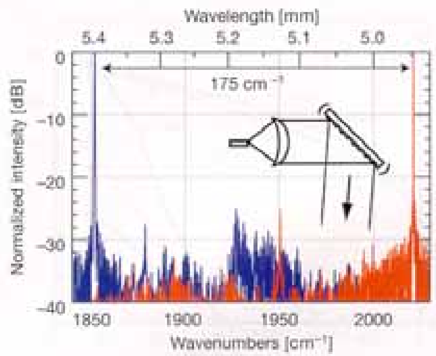
Continuous monitoring of the outside NO concentration



Monitoring using a set-up based on TEC cooled lasers and detectors by Aerodyne over more than 25 days continuously, without operator intervention. The accuracy was 0.1 ppb over 30 s.

Data courtesy of M. Zahniser, Aerodyne Research Inc. (www.aerodyne.com)

Continuous wave tuning of a quantum cascade laser over 175 cm^{-1}



The external cavity configuration is of the Littrow type, as shown in the inset. The same device was tuned over 150 cm^{-1} with a sidemode suppression ratio larger than 30 dB.

Figure adapted from Maulini et al.

can be tailored to enable the design of active regions with very large gain bandwidth.

One possibility is to have active regions tuned to different transitions coexisting in the same waveguide structure, as demonstrated in 2002 by Gmachl et al. However, the selection of the single frequency by an external cavity is easier if the broad gain is achieved using a bound-to-continuum transition that offers an essentially homogeneous broadening of the gain spectrum.

Using such an approach, Maulini and colleagues have achieved tuning of a quantum cascade laser operating in a continuous wave over a record frequency span of 175 cm^{-1}

The next important challenge is to develop systems that are expressly dedicated to the quantum cascade laser, rather than having to adapt optical systems that were originally developed for other laser sources.

around a center frequency of 1850 cm^{-1}. In this tuning range, the continuous power was larger than 10 mW. Coarse tuning is achieved by rotating the grating; fine tuning involves changing the cavity length and laser chip temperature. This narrow linewidth, broadly tunable source is especially well suited for spectroscopy. Early experiments performed by Frank Tittel at Rice University confirmed this potential by performing the spectroscopy of NO on many gas lines.

The next important challenge is to develop systems that are expressly dedicated to the quantum cascade laser, rather than having to adapt optical systems that were originally developed for other laser sources. In this way, the ultimate goal of achieving low-cost, high-sensitivity sensors could be reached. A good omen may come from the heavens: The Jet Propulsion Laboratory just chose a DFB-QCL–based sensor in the 2009 Mars Science Laboratory mission, to participate in the quest for determining whether Mars was ever habitable.

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