

Array of Tiny Quantum Cascade Lasers Provides Tunable Mid-IR Output

The microchip has advantages over a conventional FTIR spectrometer.

by Benjamin G. Lee, Dr. Mikhail A. Belkin and Dr. Federico Capasso, Harvard University

Unlike the near-infrared and visible spectral ranges, where diode lasers provide compact and reliable sources, the mid-infrared range — 3 to 15 μm — lacks good lasers. A breakthrough in this area occurred with the demonstration of the quantum cascade laser in 1994.¹ Currently, quantum cascade lasers are the only semiconductor lasers that can operate at room temperature at 3 to 15 μm .^{2,3} This spectral range comprises the “molecular absorption fingerprint” region, so named because all molecules can be uniquely identified by vibrational absorption lines in that region.

Quantum cascade lasers are similar to traditional diode lasers in external appearance and can be produced in the same foundries that produce diode lasers for the telecom industry.⁴ Their operation principle is, however, fundamentally different. In a diode laser, the light is generated through the recombination of electrons and holes, and the wavelength of the emitted light is determined by the semiconductor’s bandgap (Figure 1a).

In a quantum cascade laser, the laser transition occurs between electron levels in the conduction band of a semiconductor superlattice (Figure 1b). By changing the thicknesses of the quantum wells and barriers in the superlattice, one can engineer the energy levels to produce a laser transition at the desired wavelength. The term “cascade” comes from the fact that the energy levels are arranged in a cascading way, so that electrons making the lasing transition in one stage are reused in the next stage.

Because quantum cascade lasers can operate in the molecular fingerprint region, the potential range of applications for sensors based on them is huge. In particular, chemical sensing with these lasers can be applied to medical diagnostics, such as breath analysis, and to pollution monitoring, to environmental sensing of the greenhouse gases responsible for global warming, and to remote detection of toxic chemicals and explosives.⁵

The active region of a quantum cascade laser can be designed to provide broadband laser gain, for example, at wavelengths everywhere between 8.2 and 10.4 μm .⁶ Most spectroscopic and sensing applications, however, require a narrowband, single-mode laser output. To achieve single-mode lasing with broadband gain, quantum cascade lasers are processed as distributed feedback lasers, which have built-in Bragg gratings for mode selection, or the quantum cascade lasers are incorporated within an external cavity with a rotating diffraction grating that provides tunable single-mode operation. External cavity versions are broadly tunable but complex to build, requiring careful alignment, high-quality antireflection coatings, and a piezoelectric controller to vary the cavity length. Distributed

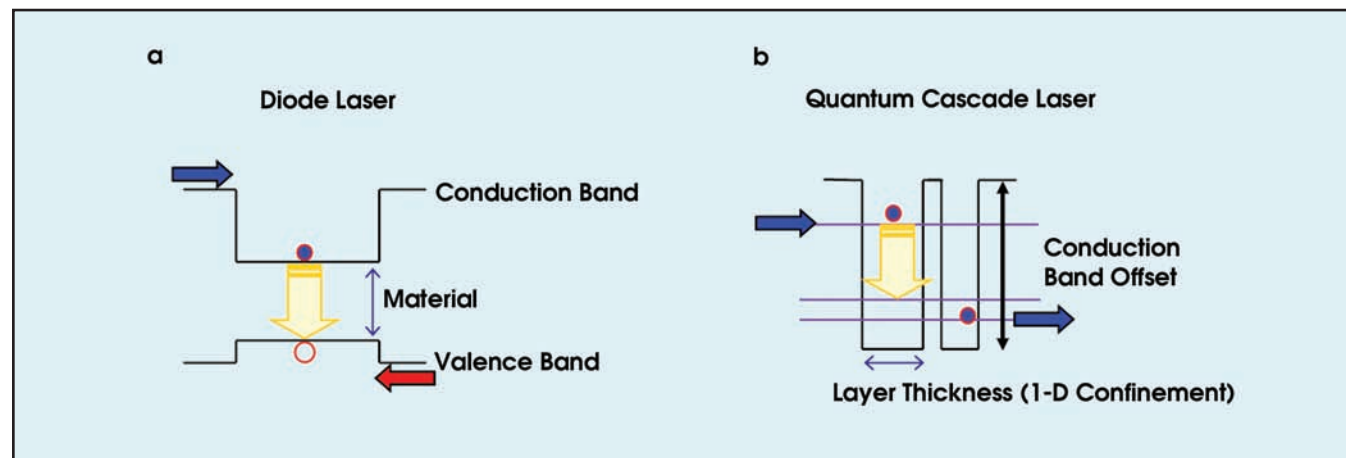


Figure 1. A diode laser (a) generates light from the recombination of electrons and holes across the semiconductor’s bandgap, and the material’s bandgap determines the wavelength of emission. In a quantum cascade laser (b), light is generated from an energy transition in the conduction band of a semiconductor superlattice. By changing the thicknesses of the quantum wells and barriers in the superlattice, we can change the wavelength of light emitted.

feedback versions are very compact — a few millimeters in length — and easily can be microfabricated in large quantities, but their tunability is limited to temperature tuning of ~ 0.5 nm/K.

We have developed a single-mode quantum cascade laser source that combines the advantages of external cavity and distributed feedback devices — it is broadly tunable and can be readily made using standard microfabrication techniques.

Our device is based on an array of distributed feedback quantum cascade lasers monolithically fabricated on the same chip.⁷ A fabricated device with 32 lasers on a single chip is shown in Figure 2. Each individual laser in the array is designed to emit at a different wavelength, and the emission wavelength of each individual laser can be temperature-tuned in a small range so that it can cover the entire region between the emission wavelengths of its neighbors. Together, all the lasers in the array can cover a broad range of wavelengths; in the case of our fabricated devices, the 32 lasers cover a range from 8.73 to 9.43 μm . Thus, a single device can be used to target all the molecular absorption lines in that range for sensing.

Our quantum cascade laser array was fabricated using standard microfabrication techniques similar to those used for fabricating diode lasers for the telecom industry. Bragg gratings, with periods ranging from 1.365 to 1.484 μm , were defined for all 32 lasers in the array at the same time by optical lithography, and all the lasers were processed in a single process run. After processing is completed, we have a “laser array chip” that is 4×5 mm, which then is connected to a custom-designed electronics controller. The controller can power the lasers, tune their emission wavelengths and interface with a laptop/desktop computer via a USB serial port. All 32 lasers were individually connected to separate channels on the controller, allowing us to address each laser — to adjust their frequencies with temperature tuning and to fire them with arbitrary timing.

Array performance

We operated the laser array at room temperature in pulsed mode with 40-ns pulses at a repetition rate of 100 kHz. We first demonstrated individual addressing and firing of the lasers in arbitrary order using the controller. All 32 lasers in the array operated in single mode with more than 20-dB suppression of other, unwanted modes. The lasing wavelengths of the lasers in the array spanned from 8.73 to 9.43 μm with regular spacings of ~ 22 nm between the emission wavelengths of adjacent lasers.

We further demonstrated that the gap between the nominal emission wavelengths of two adjacent lasers can be filled by temperature tuning, providing continuous spectral coverage. Temperature tuning in the laser can be accomplished by heating it locally — sending a DC bias current through the laser — or by changing the temperature of the whole array — heating and cooling the system’s heat sink. We used the latter method, heating the lasers from

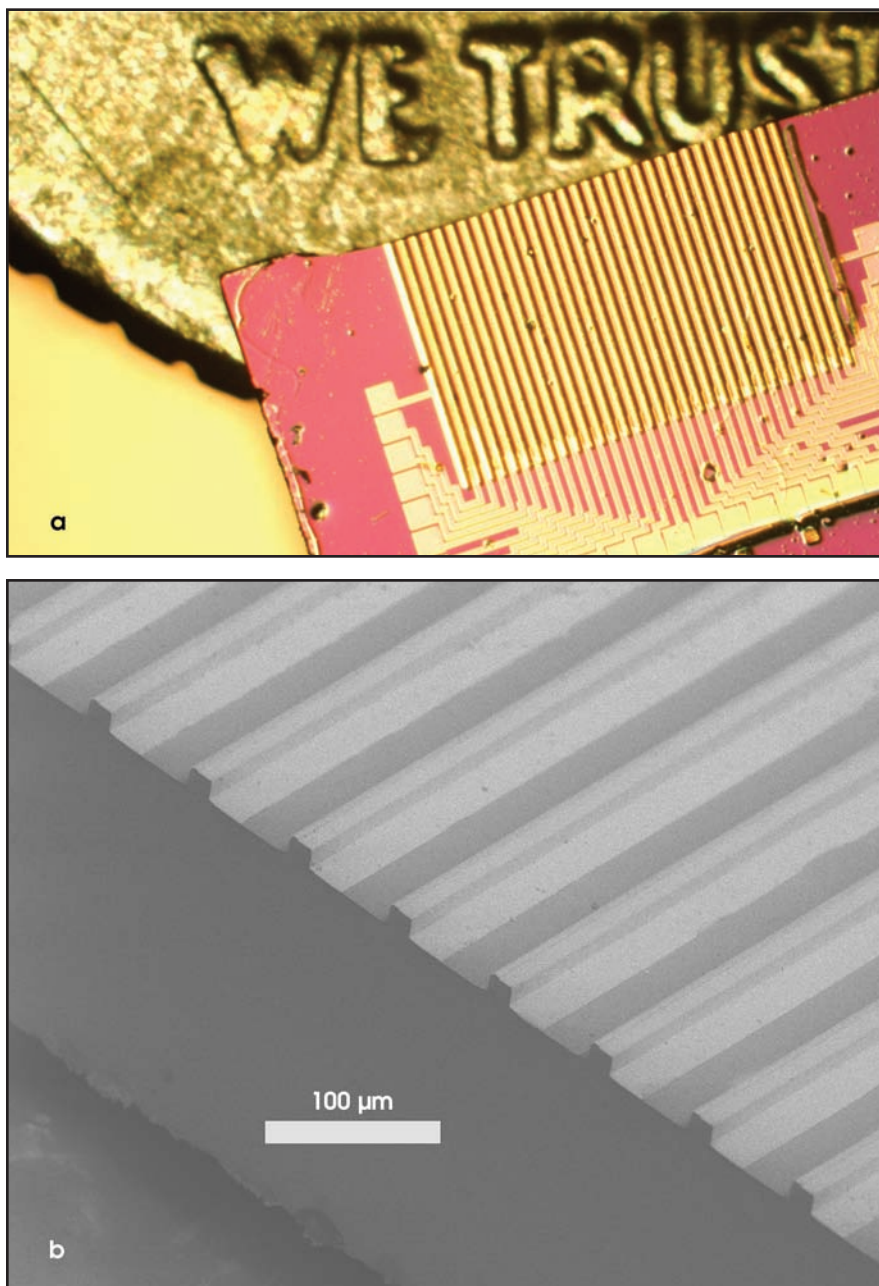


Figure 2. A fabricated quantum cascade laser array (a) with 32 lasers on a single chip was placed on top of a dime for size comparison. The entire chip is less than 4×5 mm in size. An SEM image (b) shows individual laser ridges (diagonal stripes) in the array. Laser light is emitted from the end facets of the ridges (shown facing).

300 to 390 K. The emission wavelengths were tuned continuously, with each individual laser's output wavelength changing by ~50 nm over the full temperature range. Thus, with temperature tuning, our array can emit any wavelength in the range of 8.73 to 9.43 μm .

A typical dependence of the power output versus pump current for lasers in the array is shown in Figure 5. The power output of our lasers is more than sufficient for spectroscopic applications.

Absorption spectroscopy

After calibration of the laser emission wavelengths, our quantum cascade laser array can be used for a variety of spectroscopic applications. We demonstrated its use for infrared absorption spectroscopy of liquids.

Our setup consisted of the quantum cascade laser source, a transparent BaF_2 fluid cell — with a 23.6- μm thick fluid chamber — containing the analyte and a mercury cadmium telluride liquid nitrogen cooled detector (Figure 6, inset). A single lens, 12 mm in diameter and with a 12-mm focal length, was used to image the 2.5-mm-wide laser array onto the 0.25×0.25 -mm active area of the detector. To take a spectrum, the lasers were fired sequentially, and the intensities of the transmitted beams were recovered from the detector using a gated integrator. After taking the background and sample spectra, we obtained the absorption spectrum using a frequency table with data for each laser in the array (Figure 6). It took approximately 5 s to obtain each spectrum, and with further improvements, the measurement could be done substantially faster. Our results compare favorably with spectra obtained using a conventional Fourier transform infrared (FTIR) spectrometer.

The frequency resolution of our quantum cascade laser source is determined by the lasers' linewidths, which were measured to be ~0.1 nm in pulsed operation. This resolution is an order of magnitude higher than that provided by a standard "bench-top" commercial FTIR spectrometer and is roughly comparable to the linewidth of gas absorption features at atmospheric pressure. With careful stabilization of the current source and the device temperature, a quantum cascade laser operating in the continuous wave regime can achieve linewidth smaller than 0.001 nm.⁸ We intend to pursue this in our future work.

When comparing quantum cascade laser-based sensors with FTIR spectrometers, it is useful to consider factors besides resolution, such as spectral range, brightness, size, portability and cost.

Although the spectral bandwidth provided by a broadband quantum cascade laser is smaller than that of an FTIR spectrometer, this limitation does not impede most sensing applications. Most of the important absorption lines occur in the 8- to 12- μm spectral range, and quantum cascade lasers with broadband gain that covers most of this

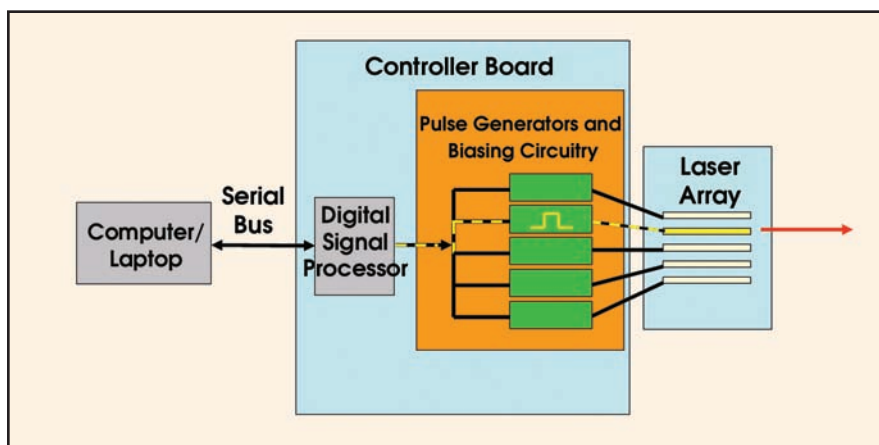


Figure 3. The schematic of the entire quantum cascade laser source shows the laser array being controlled and powered by custom-built electronics. The dotted line shows the routing of a current pulse to fire a specific laser. The electronics can be interfaced with a laptop computer for programming of the laser firing and data collection.

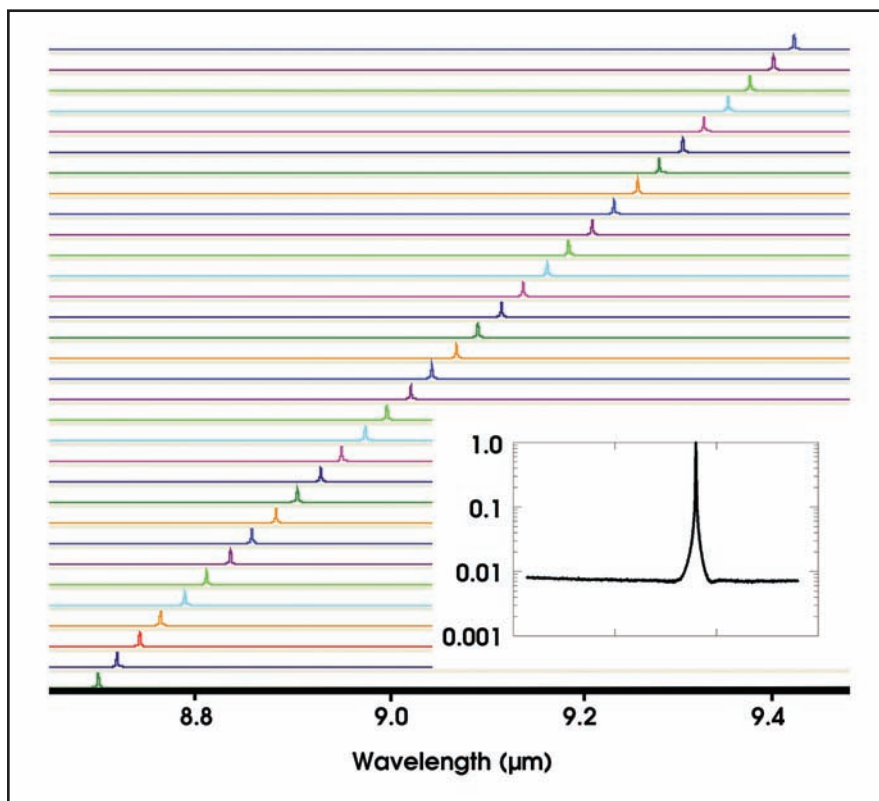


Figure 4. Spectra of the 32 single-mode distributed-feedback lasers in the array are shown. The laser wavelengths are spaced ~22 nm apart and span the range from 8.73 to 9.43 μm . The inset shows the spectrum of a representative laser in the array on a log scale, showing that unwanted side modes are suppressed by more than 20 dB.

spectral range already have been demonstrated. In addition, the chemicals to be monitored usually are known, and a quantum cascade laser can be designed to have laser gain at the wavelengths where those chemicals absorb light. The much higher brightness of quantum cascade lasers as compared with the thermal sources (glow bars) used in FTIR spectrometers should lead to substantial improvements in sensitivity.

In terms of size and portability, there is no question that quantum cascade laser-based sensors would be much more compact and portable than FTIR spectrometers. Because the lasers can be produced in the same foundries that produce diode lasers for the telecom industry, their cost potentially could drop to the levels of laser diodes. Thus, quantum cascade laser-based sensors could be quite inexpensive, particularly when compared with FTIR spectrometers, which contain sophisticated mechanical and optomechanical components. With all the benefits of quantum cascade laser-based sensors, we hope that they will find numerous applications for mid-infrared chemical sensing and analysis.

□

Acknowledgments

The authors would like to thank Ross Audet and Jim MacArthur for assistance in the design of the micro-electronic controller; David Bour, Scott Corzine, Gloria Höfler, Doug Oakley, David Chapman and Antonio Napoleone for the materials growth; and Tom Tague, Laurent Diehl, Christian Pflügl and Jérôme Faist for valuable technical discussions. Funding support was provided by the DARPA Optofluidics Center under grant number HR0011-04-1-0032. Devices were fabricated in the Center for Nanoscale Systems at Harvard University, which is a member of the National Nanotechnology Infrastructure Network.

Meet the authors

Benjamin G. Lee is a doctoral candidate in applied physics in the School of Engineering and Applied Sciences at Harvard University; e-mail: bglee@fas.harvard.edu.

Mikhail A. Belkin is a research associate in the School of Engineering and Applied Sciences at Harvard University; e-mail: mbelkin@seas.harvard.edu.

Federico Capasso is the Robert L. Wallace professor of applied physics and Vinton Hayes senior research fellow in electrical engineering, in the School of Engineering and Applied Sciences at Harvard University; e-mail: capasso@seas.harvard.edu.

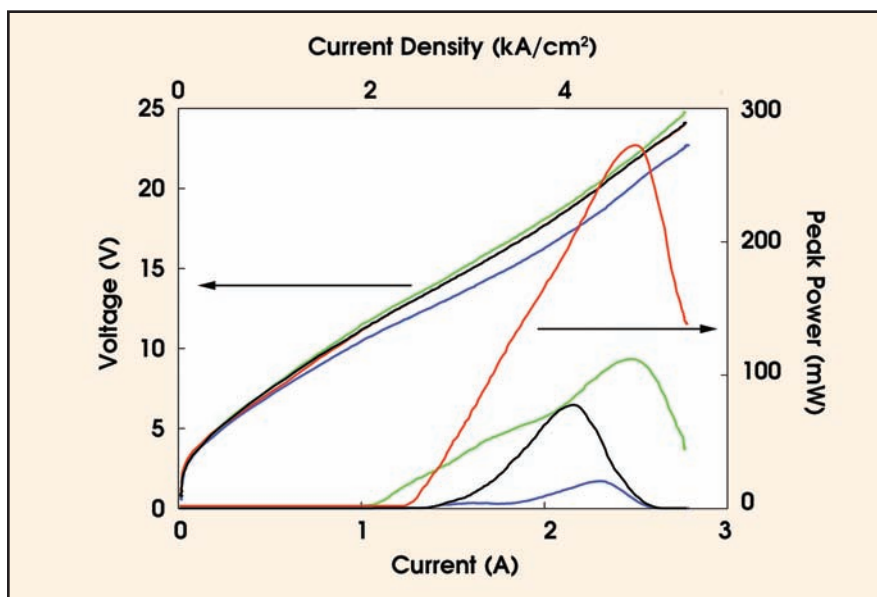


Figure 5. A plot of the voltage (left axis) and the light output (right axis) from several representative lasers in the array is shown as a function of the laser pump current. Operated pulsed, the lasers have tens to hundreds of milliwatts in peak light intensity. Although there is significant variability in output power among various lasers in the array, even the weakest lasers have more than sufficient power for spectroscopic applications.

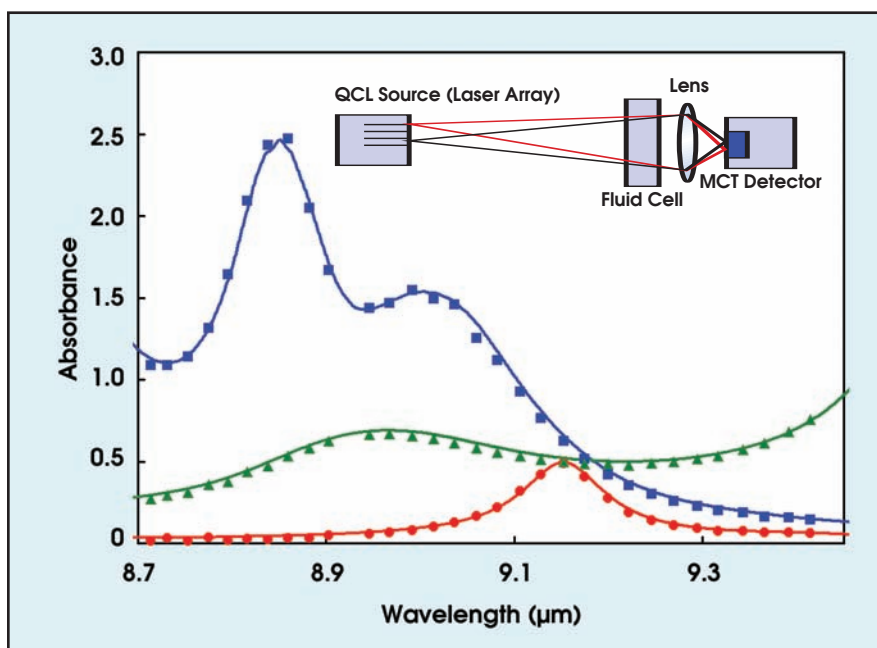


Figure 6. Absorption spectra of isopropanol (squares), methanol (triangles) and acetone (circles) were obtained with a quantum cascade laser (QCL) source and a Fourier transform infrared spectrometer. The inset shows the experimental setup for infrared absorption spectroscopy using the quantum cascade laser source. MCT = mercury cadmium telluride.

1. J. Faist et al (April 22, 1994). Quantum cascade laser. *SCIENCE*, pp. 553-556.
2. F. Capasso et al (May 2002). Quantum cascade lasers. *PHYSICS TODAY*, p. 34.
3. C. Gmachl et al (November/December 2000). New frontiers in quantum cascade lasers and applications. *IEEE JOUR SELECTED TOPICS IN QUANT ELEC*, pp. 931-947.

4. L. Diehl et al (May 17, 2006). High-power quantum cascade lasers grown by low-pressure metal organic vapor-phase epitaxy operating in continuous wave above 400 K. *APPL PHYS LETT*, Vol. 88, 201115.
5. A.A. Kosterev and F.K. Tittel (June 2002). Chemical sensors based on quantum cascade lasers. *IEEE JOURN QUANT ELECT*, pp. 582-591.
6. R. Maulini et al (May 17, 2006). External cavity quantum-cascade laser tunable from 8.2 to 10.4 μm using a gain element with a heterogeneous cascade. *APPL PHYS LETT*, Vol. 88, 201113.
7. B.G. Lee et al (Dec. 3, 2007). Widely tunable single-mode quantum cascade laser source for mid-infrared spectroscopy, *APPL PHYS LETT*, Vol. 91, 231101.
8. R.M. Williams et al (1999). Kilohertz linewidth from frequency-stabilized mid-infrared quantum cascade lasers. *OPT LETT*, Vol. 24, pp. 1844-1846.