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An AlGaAs–GaAs quantum cascade laser operating with a thermoelectric cooler for spectroscopy of NH₃

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Abstract

A new design of AlGaAs–GaAs quantum cascade laser (QCL) operating at a wavelength around 11.2 μm is reported. An improved injection region and increased barrier height in the active region have enabled high temperature operation with reduced temperature dependence of the threshold current. The laser has been employed at temperatures up to 244 K (accessible with a thermoelectric cooler) for spectroscopy of gas phase NH₃, using the injected current pulse to linearly tune a single mode of its multi-mode Fabry–Perot output through an absorption line of the gas.

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Since their inception in 1994 by Faist et al. [1], quantum cascade lasers (QCLs) have been under continuous development, principally using InGaAs–InAlAs multi-layer structures lattice matched to InP and resulting in room temperature operation [2]. In 1998, Sirtori et al. [3] reported a new QCL design based on AlGaAs–GaAs. An advantage of this material system is that the GaAs substrate is mechanically more robust than InP, thereby making the manufacture of devices easier. Although AlGaAs–GaAs QCLs have performed

well at low temperature [4–6], room temperature operation has been more difficult to achieve.

Subsequently two ways of improving the high temperature operation have been developed: (i) shifting the laser operation to longer wavelength [7], and (ii) increasing the aluminium content of the barrier region. In the first method, the levels involved in the transition are located deeper in the wells. Longer wavelength operation results from improved electron confinement, but at the expense of an increase in free carrier absorption leading to a higher value of the threshold current. In the second approach, a higher Al compositional fraction in the barrier increases the band offset between GaAs and AlGaAs, again improving the electron confinement. Based on these principles, Page et al. [8] have demonstrated recently a device

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emitting at $\lambda \sim 9 \mu\text{m}$ with a working temperature up to 300 K, using $\text{Al}_{0.45}\text{Ga}_{0.55}\text{As}$ for the barrier.

Previously, AlGaAs-GaAs QCLs have been employed for gas spectroscopy [9], where the laser has been operated at cryogenic temperatures. In this paper an improved AlGaAs-GaAs QCL design is reported that allows the laser to operate at 244 K. Spectroscopy can therefore be carried out using a compact thermo-electrically cooled laser package.

The QCL described here is designed to emit light in the range 10.5–11.5 μm for gas spectroscopy. The Al compositional fraction x of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ in the first barrier of the active region is 0.40, while subsequent barriers have $x = 0.33$. This combination leads to a deeper confinement of the levels and an increase of the band offset. The details of the active region are shown in Fig. 1. It employs three coupled quantum well in the radiative region, in which lasing occurs between the $n = 3$ and $n = 2$ levels when the electric field across the structure reaches 48 kV cm^{-1} . The energy gap between these levels, E_{32} , is estimated to be 118 meV, corresponding to $\lambda \sim 10.5 \mu\text{m}$. The energy gap between the $n = 3$ level and the bottom of the continuum is estimated to be 51.5 meV, sufficient

to reduce the thermal leakage of electrons to the continuum. The bottom of the injector is just 6 meV below the $n = 3$ level.

The QCL structure described here was grown by molecular beam epitaxy (MBE) on an n^+ -GaAs substrate doped to $3 \times 10^{18} \text{ cm}^{-3}$ with Si, and contained a 40 period core. The waveguide is similar to that proposed by Sirtori et al. [3]. Since $\text{Al}_{0.9}\text{Ga}_{0.1}\text{As}$ is used for cladding layers, this QCL is less efficient than similar ones using the plasmon effect [10]; however, the optical confinement is less dependent on wavelength.

The lasers were processed by deep wet etching in a $\text{HCl-H}_2\text{O}_2$ solution to form ridges 20 μm wide and 1.5 mm long. The contacts (Ti/Pd/Au on top and Au-Ge/Ni/Au on the back) were deposited by evaporation. The laser chips were mounted epilayer-up on gold plated copper mounts using a layer of indium. The mounts were fitted into a specially designed thermoelectric cooler box, stabilised to $\pm 0.01 \text{ K}$, allowing substrate temperatures to reach 233 K. The lasers were driven by current pulses ranging in width from 4 to 200 ns with a maximum amplitude of 20 A, and at maximum pulse repetition frequency (PRF) of 100 kHz. The transmission line was matched to 50Ω . A continuously scanning, high resolution Fourier transform spectrometer (Bomem DA-3 FTS) was used for spectral characterisation of the devices. It relies on a method described in detail recently [11], in which the QCL appears as a quasi-continuous-wave light source. The FTS has a maximum effective resolution of 0.005 cm^{-1} (150 MHz), and is fitted with a 2–20 μm mercury cadmium telluride (MCT) photoconductive detector.

The spectrum of Fig. 2 shows the mode structure of a Fabry-Perot AlGaAs-GaAs QCL at 236 K using a PRF of 100 kHz and pulse duration of 9 ns with 12 A amplitude. The Gaussian-type envelope of the longitudinal mode structure is more than 20 wave-numbers ($880\text{--}900 \text{ cm}^{-1}$) with an emission wavelength centred at 890 cm^{-1} (11.2 μm). The spectral data indicate that the mode spacing is slightly less than 1 cm^{-1} which, for a cavity length of 1.5 mm gives an effective refractive index of $n \sim 3.75$. The value for the effective linewidth or “wavelength-up chirp” of each mode cannot be retrieved as it is limited by instrument

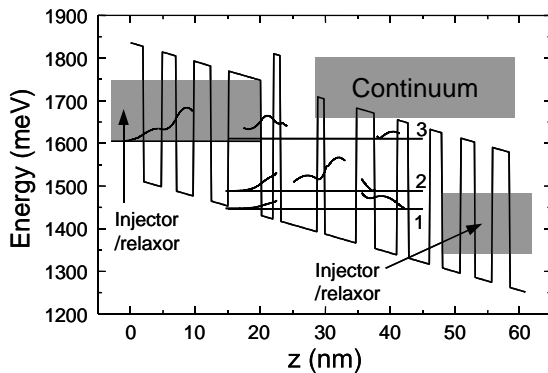


Fig. 1. Portion of the conduction band profile under an electric field of 48 kV cm^{-1} . The moduli square of the wave functions are shown. The laser transition occurs between the $n = 3$ and $n = 2$ levels. The sequence of the layers, starting from the injection barrier, is: **5.1/1.9/1.1/6/1.1/4.9/2.8/3.6/1.7/3.2/2/2.8/2.2/2.7/2.6/2.7** (units of nm). The bold and italic characters represent the barriers containing 33% and 40% aluminium, respectively. The underlined terms correspond to the 8×10^{17} silicon-doped layers.

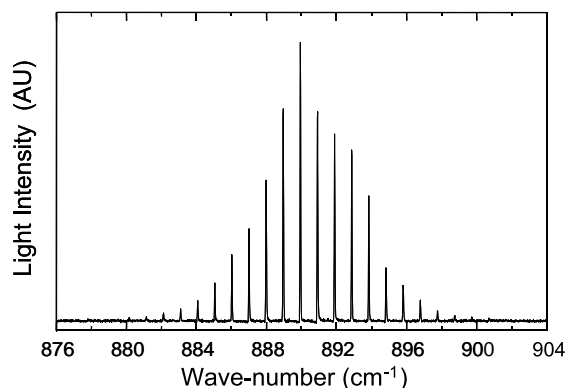


Fig. 2. Emission spectra of the AlGaAs–GaAs quantum cascade laser recorded with a Fourier transform infrared spectrometer using a resolution of 0.005 cm^{-1} . The base temperature of the laser was 243 K.

resolution that was set to 0.02 cm^{-1} . However, using the maximum resolution setting on the FTS (0.005 cm^{-1}) and a 4 ns pulse duration, the FWHM of a mode was found to be approximately 0.015 cm^{-1} .

Fig. 3 shows light–current (L – I) curves at various temperatures in the range 20–260 K. For low temperature characterisation, a helium flow cryostat capable of reaching 10 K was used. The lasers were driven with 100 ns current pulses at a PRF of 5 kHz. Above threshold, the devices display a linear L – I until high currents where thermal roll-over is approached.

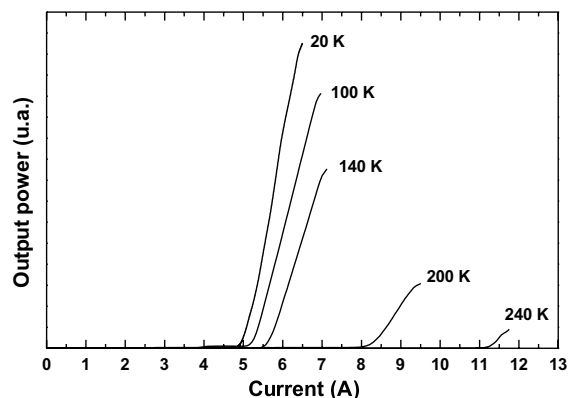


Fig. 3. The light–current (L – I) curves at several temperatures for the AlGaAs–GaAs quantum cascade laser.

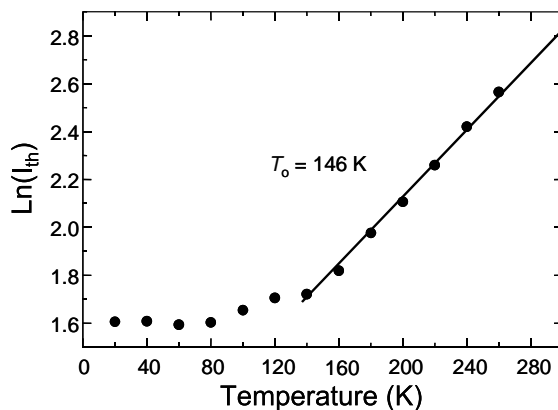


Fig. 4. Natural logarithm of threshold current ($\ln(I_{th})$) vs temperature.

Fig. 4 shows the temperature dependence of the threshold current. The calculated value of T_0 within the temperature range 140–260 K is 146 K. This high T_0 indicates that the temperature dependence of the threshold current is low and therefore high temperature operation is possible. Indeed, QCL operation up to 276 K has been observed for devices mounted epilayer-down.

The AlGaAs–GaAs QCL described above, operating as a multi-longitudinal mode Fabry–Perot laser at temperatures around 244 K, was employed as a source for gas spectroscopy of ammonia (NH_3) at wavelengths around $11.2\text{ }\mu\text{m}$. The method employed takes advantage of a wavelength chirp during the applied current pulse to tune the laser through an absorption line of the ammonia (NH_3) gas.

A continuously scanning high resolution FTS (Bomem DA-3), equipped with a removable 20 cm path-length absorption cell, was used for spectral characterisation of the QCL and to record transmission spectra of NH_3 . The experimental set-up is shown in Fig. 5. The QCL was driven by current pulses with a repetition frequency and pulse width ranging from 40 kHz to 1 MHz and 4 to 60 ns, respectively. This frequency range was chosen to be at least twice that of the FTS maximum sampling frequency, and the latter was further decreased to 2 kHz by a digital filter. The amplitude and shape of the current pulses applied to the devices were monitored with a Rogowski coil. The MCT detector and a digital oscilloscope were used

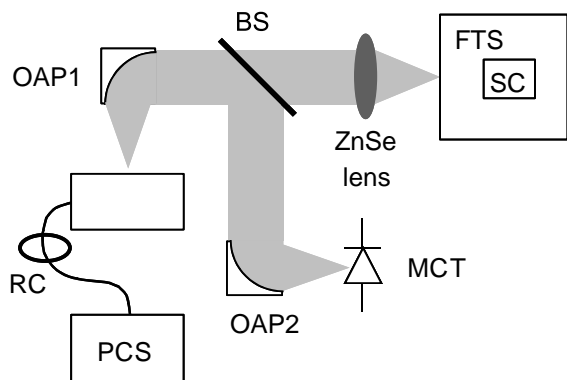


Fig. 5. Schematic of the experimental arrangement: QCL, quantum cascade laser; RC, Rogowski coil; PCS, pulsed current source; BS, beam splitter; MCT, mercury cadmium telluride detector to monitor QCL output; FTS, Fourier transform spectrometer; OAP, off-axis parabolic; and SC, sample chamber (20 cm path absorption cell).

to record the temporal response of the light pulses generated by the QCL.

A sample gas cell of ammonia (NH_3) was introduced in one chamber of the FTS and transmission spectra were then recorded with a broadband source (globar) at a resolution of 0.005 cm^{-1} . The emission from the QCL was then recorded with a resolution of 0.005 cm^{-1} . At a laser base temperature of 243 K, it was noted that one of the longitudinal modes fell between a set of absorption lines at approximately 892 cm^{-1} . The laser wavelength was then tuned to lower wavelength by decreasing the temperature to 237 K so that the applied wavelength-up chirp covered a spectral micro-window from 891.9 to 892.3 cm^{-1} ($\sim 0.4 \text{ cm}^{-1}$). The top-hat shaped current pulses were set to 12.8 A with 60 ns duration and a PRF of 40 kHz. The rate of change of wave-number as a function of pulse duration was determined to be $-6.6 \times 10^{-3} \text{ cm}^{-1}/\text{ns}$ (200 MHz/ns).

In Fig. 6, a high resolution (0.005 cm^{-1}) transmission spectrum of NH_3 recorded with a black body source (trace (a)) is compared with the molecular absorption of ammonia seen via the dip in one line of the multi-mode emission spectrum of the QCL (trace (b), no gas; and trace (c), absorption of gas phase NH_3 detected). Fig. 7 shows an expanded view of the wave-number region in which absorption is observed. Note that the partial

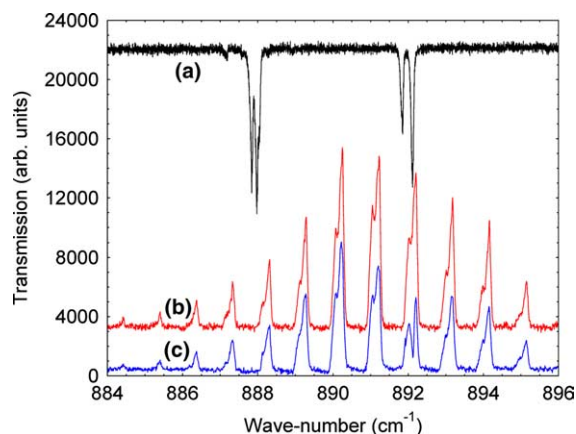


Fig. 6. A comparison of the transmission spectrum of NH_3 recorded using a black body source (trace (a)) and the emission from a multi-longitudinal mode QCL without (trace (b)) and with NH_3 at a pressure of 5 torr in the 20 cm sample cell located in the FTS (trace (c)). Absorption by the NH_3 causes a strong dip in one QCL mode around 892 cm^{-1} . The QCL was cooled to 237 K and driven by current pulses of 40 kHz PRF, 60 ns duration and 12.8 A amplitude. All spectra were recorded at a resolution of 0.005 cm^{-1} . Traces (a) and (b) have been displaced vertically for clarity.

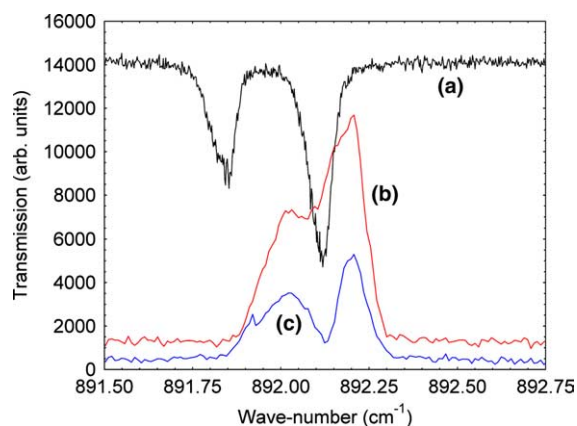


Fig. 7. Expanded version of Fig. 6 showing detail of the mode dip resulting from strong absorption by NH_3 at approximately 892.15 cm^{-1} . The traces have not been displaced vertically.

splitting of virtually all the longitudinal modes of the QCL (trace (b)) is associated with a tendency of the lasers to emit with more than one transverse mode, rather than being an absorption effect. Although ammonia has a sparse spectrum, comprising widely spaced absorption lines throughout

most of its 10 μm band system, the utilisation of a multi-mode Fabry–Perot laser maximises the possibility of coincidence between the output lines of the QCL and the molecular absorption lines of this light molecule.

An AlGaAs–GaAs quantum cascade laser design has been developed for higher temperature operation making it a much more convenient source for spectroscopy. The laser has been employed for spectroscopy of gas phase NH_3 at wavelengths close to 11.2 μm . It is shown that a high resolution, continuously scanning Fourier transform infrared spectrometer may be used as a very sensitive probe of the spectral/time dependence of the output of pulsed QCLs. This is made possible by the use of pulsed light sources with high repetition rate, obviating the need for a time-resolved Fourier transform instrument usually limited to much lower resolution. The high resolution FTS allows the spectral behaviour of pulsed QCLs to be studied over a much broader spectral region compared to other methods. As a result, it is possible to find, select and spectrally adjust (tune) a suitable longitudinal mode for absorption by the NH_3 line of interest. The linear heat-induced wavelength-up chirp of a pulsed QCL has also been characterised and this intrinsic spectral behaviour has been used for absorption spectroscopy of NH_3 gas. The work presented here shows that a relatively simple design of Fabry–Perot AlGaAs–GaAs QCL can be employed for spectroscopy at close to room temperature. Furthermore, although a FTS has been used in the experiments described here to select a single mode, it would also be possible to select a single mode with an etalon. A simple spectrometer based on the Fabry–Perot quantum cascade laser and an etalon has the advantage of many selectable modes and therefore increases the probability of achieving an overlap with the absorption line of a par-

ticular gas, an important consideration for molecules such as NH_3 with a sparse spectrum of lines.

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