2005 International Conference on Indium Phosphide and Related Materials

STIMULATED ELECTRONIC ANTI-STOKES RAMAN EMISSION IN QUANTUM CASCADE LASERS

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Abstract

We report the first observation of stimulated electronic Anti-Stokes Raman emission in Quantum Cascade lasers. The pump laser is monolithically integrated with the Anti-Stokes nonlinear region in a 2-stack active waveguide core.

I. Introduction

Quantum Cascade (QC) lasers (1) are semiconductor injection lasers based on optical and electronic intersubband transitions. A QC laser typically comprises several tens of cascaded active regions and injectors, each of them containing about 8 to 10 wells and barriers. As opposed to conventional diode lasers, in which material properties such as the bandgap energy play a crucial role, the intersubband transitions in QC lasers are mostly determined by well and barrier thicknesses and the external applied electric field. These parameters can be easily modified to obtain the desired performance.

It is well known that intersubband transitions in asymmetric coupled quantum wells can display giant nonlinear optical susceptibilities (2-4). With QC lasers, it is possible to selectively replace active regions/injectors with nonlinear elements or to integrate nonlinear transitions directly into the active regions themselves, thus allowing for efficient intra-cavity wave mixing (5). This was recently demonstrated for sum frequency, second harmonic and third harmonic generation (6-8).

The monolithic integration of resonant optical nonlinearities based on intersubband transitions and QC lasers has several advantages. QC lasers can provide high optical power densities thus being very effective pump sources and, at the same time, good overlap between pump modes and the nonlinear region is obtained.

Keeping in mind the multitude of known nonlinear phenomena, we are just beginning to scratch the surface of the potential arising from the combination of nonlinear optics and QC lasers. In this context, Stimulated Raman Scattering has been one subject of research. To date, only Stokes Raman lasing has been achieved; both using externally pumped asymmetric QW structures (9) and with QC lasers where the nonlinearity occurs in the active region (10).

Here, we report on –to our knowledge- the first observation of Stimulated electronic Anti-Stokes (AS) Raman emission in a QC laser and discuss the feasibility of AS lasing. Implementing nonlinear effects based on Raman transitions has the added advantage that no phase matching is required.

II. Experiments and results

QC lasers have been designed comprising two monolithically integrated stacks of active regions and injectors, one stack being the pump laser and the other stack forming the nonlinear region (Fig. 1). Intersubband transitions in the nonlinear region were designed so as to optimize the optical dipole matrix elements between levels 2,3 and 4, z_{43} and z_{42}, respectively, thus enhancing the relevant third order nonlinearity for AS emission. Electrons in level 3 of the nonlinear region would then undergo AS Raman scattering stimulated by the pump light, emitting photons with energy $h\nu_{AS} = \Delta E_{42}+\delta$, where ΔE_{42} is the energy separation between levels 4 and 2 and δ is the detuning, defined as $\delta = h\nu_{pump} - \Delta E_{43}$. Fast depletion of the lower level (2) is obtained by tailoring ΔE_{21} close to the phonon resonance.

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Stimulated AS emission has been observed with samples of three different designs. However, due to space limitations, only results from two of them, wafers D3015 and D2924, are displayed here.

A. Anti-Stokes Raman Design with Positive Detuning

In wafer D3015, the barrier/well widths in a period of the nonlinear stack, henceforth given in nanometers, are 4.5 / **3.1** / 2.9 / **3.1** / 2.5 / **3.2** / 3.0 / **3.3** / 1.5 / **3.7** / 1.5 / **4.2** / 1.5 / **4.8** / 1.5 / **2.7**, where the wells are shown in bold face, and the barriers in plain face; the underlined layers are doped to 2×10^{17} cm^{-3}. The design is shown in Figs. 1 and 2. The pump laser is a conventional 20-stage QC stack emitting at λ~8.3 µm. The lasers are processed as conventional, deep-etched ridge waveguide lasers, with stripe widths ranging from 6 to 20 µm. The lasers are cleaved to 2 - 3 mm length and the facets are left uncoated. Fig. 3 shows the spectrum of the pump light for a peak current of 3 A, while Fig. 4 displays the light output and voltage versus current characteristics.

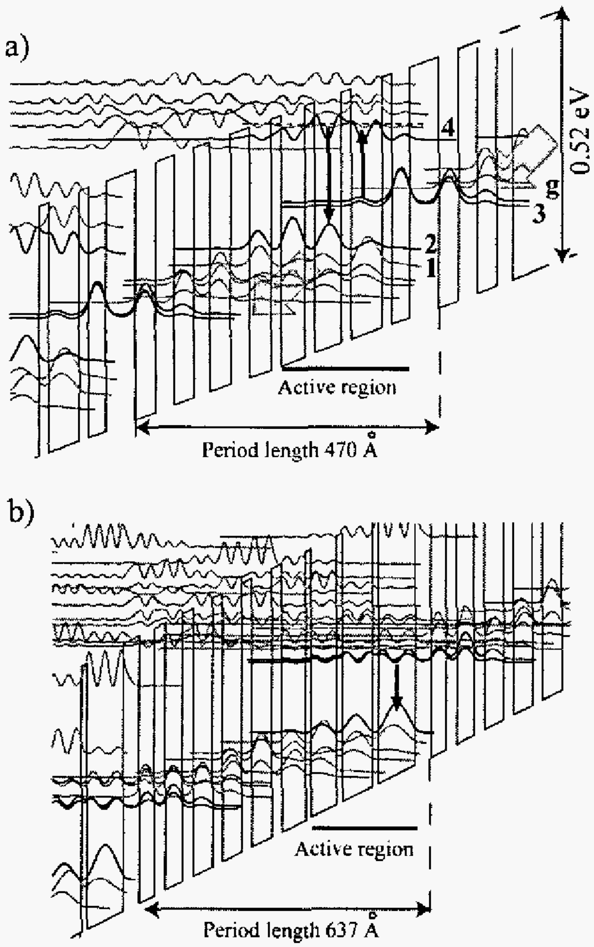

Fig. 1. Conduction band diagram of the (a) nonlinear and (b) pump regions in wafer D3015. The InGaAs wells and AlInAs barriers are grown lattice matched to InP substrate. The moduli squared of the relevant wave-functions are also shown. In (a), the black arrows indicate the Raman transition, while the grey arrows show the electron transport through the injector regions. In (b), the black arrow indicates the laser transition.

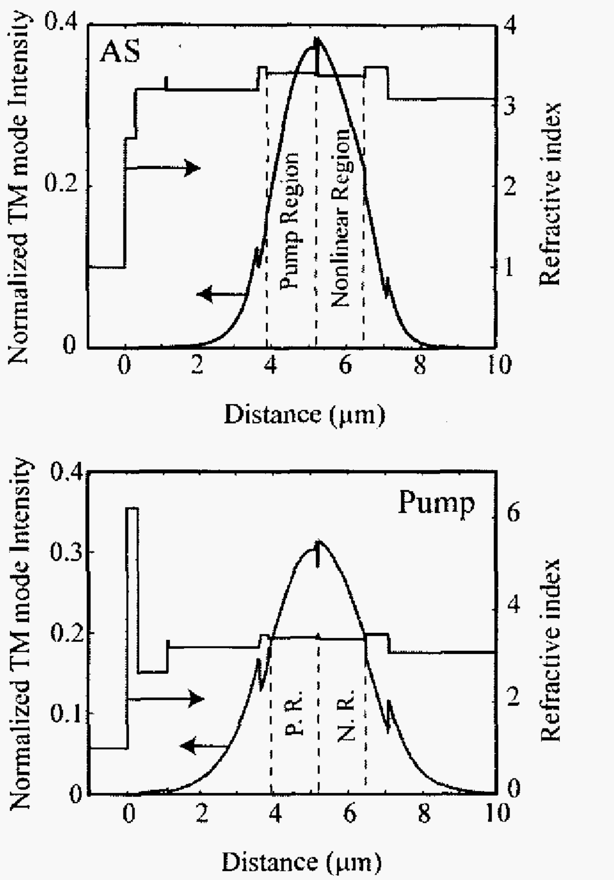

Fig. 2. Normalized intensity mode profile and profile of the real part of the refractive index in the (reverse) growth direction for both Anti-Stokes and pump fundamental modes in D3015.

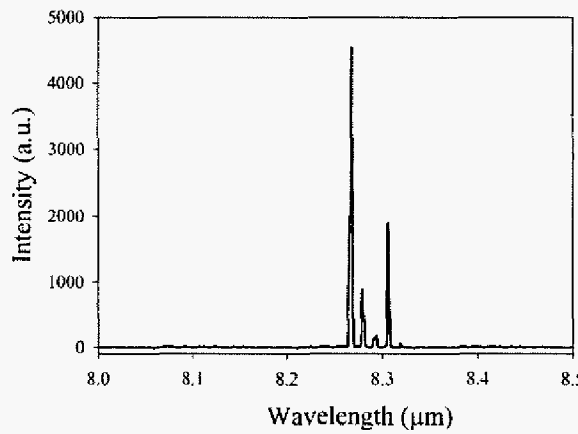

Fig. 3. Spectrum showing the pump laser emission in D3015 for 3A peak current. The measurements have been taken at cryogenic (liquid nitrogen) temperature and in pulsed current mode using a Nicolet 860 FTIR.

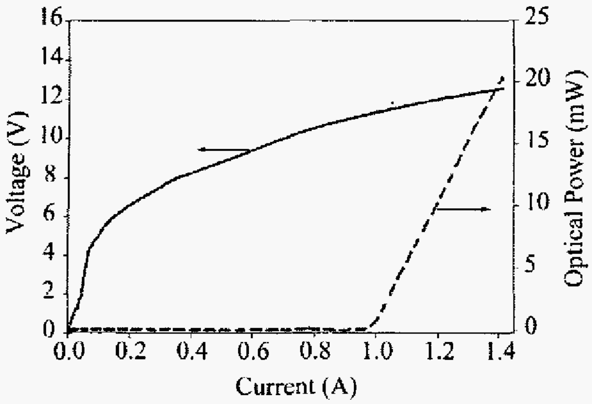

Fig. 4. Light output and voltage versus current characteristics of the laser corresponding to wafer D3015. This measurement was carried out at liquid nitrogen temperatures.

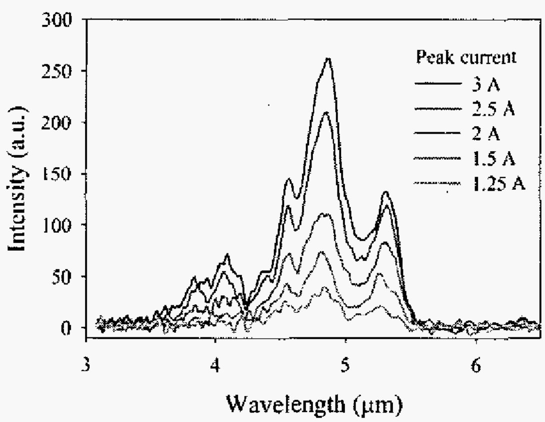

Fig. 5. Short wavelength emission spectra of D3015 above laser threshold. Peaks attributed to stimulated Anti-Stokes emission (4.8 μm) and incoherent upconversion (5.3 μm) can be distinguished. The dip observed around 4.3 μm is characteristic of CO_2 absorption.

Fig. 5 shows the short wavelength luminescence spectra measured for different peak currents, all above pump laser threshold (1 A). These measurements were carried out at liquid nitrogen temperatures in pulsed mode (100 ns current pulses), using a Nicolet 860 Fourier Transform Infrared spectrometer and a cooled InSb detector. The main peak at 4.8 μm can be attributed to Stimulated AS Raman Scattering ($h\nu_{pump}$=150 meV and ΔE_{32}=110 meV -from luminescence measurements below threshold-, giving an expected λ_{AS}=4.7 μm). When pumping above resonance (δ >0), as is the case for D3015, and concomitantly into the higher excited states, then electrons excited by pump photons can scatter directly into the upper level (4) and spontaneously decay into level 2. This incoherent upconversion process is the cause for the peak observed at 5.3 μm (ΔE_{42}=225 meV by design, corresponding to 5.5 μm, also taking into account the sharp wavelength cut-off ~5.4 μm of the InSb detector used in this measurement).

In these measurements, no narrowing of the AS luminescence peak evidencing gain has been observed so far. However, we believe that further work in optimization of the nonlinear region and improving the pump efficiency and pump laser power may finally lead to achieving electronic Anti-Stokes Raman lasing.

B. Anti-Stokes Raman Design with Negative Detuning

In wafer D2924, the sample with negative detuning, the pump is a conventional 30-period QC-laser emitting at λ~10.5 μm. The layer thicknesses of one period of the nonlinear stack of D2924 are 4.0/ **2.6**/ 2.9/ **2.9**/ 2.6/ **2.7**/ 2.3/ **2.7**/ 2.0/ **2.7**/ 1.7/ **3.8**/ 2.5/ **4.2**/ 1.8/ **7.1**, following the same notation as above in A. The doping concentration in the underlined wells/barriers is 2×10^{17} cm^{-3}. The corresponding conduction band diagram is shown in Fig. 6.

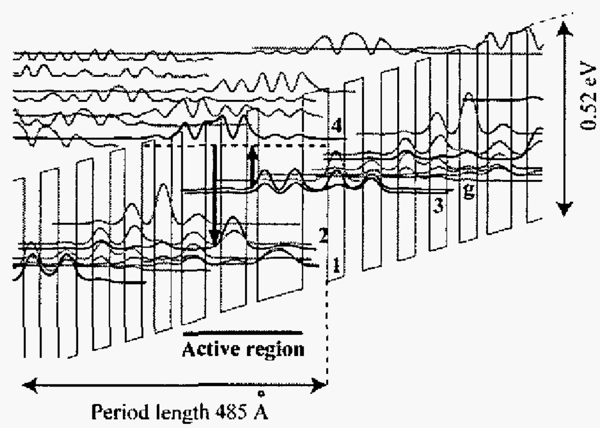

Fig. 6. Conduction band diagram of the nonlinear region in D2924. In a similar fashion to Fig. 1, the moduli squared of the relevant wave-functions are also plotted. The AS Raman transition is again indicated by black arrows.

The short wavelength luminescence spectra were measured at 7 K for different peak currents (1.8, 2.0, 2.25 and 2.5 A) above the pump laser threshold (1.1 A), and are displayed in Fig. 7. The main feature is a peak at around 4.5 μm (276 meV), which is attributed to Anti-Stokes Raman scattering ($h\nu_{pump}$=118 meV and ΔE_{32}=155 meV -from luminescence measurements below threshold-, giving an expected λ_{AS}=4.5 μm).

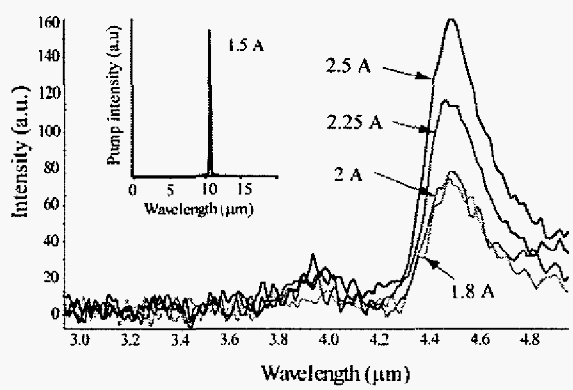

Fig. 7. Short wavelength luminescence spectra, above threshold, of D2924. The main peak at 4.5 μm is attributed to stimulated Anti-Stokes emission. The inset shows the pump spectrum at ~90 K for a peak current of 1.5 A.

In the nonlinear region of D2924 several transitions from level 4 to lower energy states other than 2 present large optical dipole matrix elements. As a result, it is possible to have AS emission at various wavelengths. This is the case of the state immediately above 2 and 145 meV below 3 by design, which would give AS emission at around 4.7 μm, thus explaining the peak's asymmetry. On the other hand, the small peak observed at approximately 3.9 μm (318 meV) can be explained by AS emission involving the pair of anticrossed states labeled as 1, (202 meV and 207 meV below state 3 by design, respectively).

The spectra in Fig. 7 show no trace of incoherent upconversion. In this case, we are pumping the nonlinear region well below resonance, with a detuning $\delta = h\nu_{pump} - \Delta E_{43} =$ (118-137) meV, which makes the real transfer of electrons to state 4 less likely.

In our measurements on D2924, no narrowing of the AS emission peak possibly indicating gain has been observed so far. However, we believe that further optimization of the nonlinear region, especially in the light of an optimized (smaller) detuning δ, and an increase in the pump power should finally lead to achieving electronic Anti-Stokes Raman lasing.

Acknowledgments

This work was supported by the MRSEC Grant at Princeton University, NSF DMR-0213706. A.G.I wishes to thank EPSRC for financial support. A.B. acknowledges support from TAMU TITF Initiative and the ONR.

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