Mode tuning of quantum cascade lasers through optical processing of chalcogenide glass claddings

Shanshan Song,[a)] Scott S. Howard, Zhijun Liu, Afusat O. Dirisu, Claire F. Gmachl, and Craig B. Arnold[b)]
Department of Electrical Engineering and Princeton Institute for the Science and Technology of Materials, Princeton University, New Jersey 08544

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In this letter, we demonstrate a method of tuning quantum cascade (QC) lasers by modifying the optical properties of an overlying cladding material. An amorphous chalcogenide cladding layer is deposited through a low temperature, solvent-casting technique that is compatible with current QC laser fabrication and operation. Above band gap illumination ($\lambda < 530$ nm) of this cladding causes a permanent change in its index of refraction leading to a change of the modal refractive index and a corresponding modal shift in the laser. Combined with deep-etched distributed Bragg gratings, a tuning of over 30 nm is obtained at an operating wavelength of 7.9 μm for constant current and temperature. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2236296]

Interest in quantum cascade (QC) lasers has continued to grow since their first experimental demonstration in the mid 1990s.[1] Due to their versatility and high performance, QC lasers have become a central component in the development of mid-infrared sensing applications. Several key advantages distinguish QC lasers from other traditional semiconductor lasers, such as a greater flexibility in output spectrum covering much of the mid-infrared and THz (3–30, 60–200 μm) range.[2,3] High power and room temperature operation make QC lasers a promising choice for trace-gas detection in the mid-infrared where the spectroscopic "fingerprints" of most atmospheric trace gas are found.

The nature of gas-sensing applications requires single-mode operation and good tunability. One way to achieve single-mode QC lasers is to integrate distributed feedback (DFB) or distributed Bragg reflectors (DBRs);[4] DFB or DBR gratings select certain mode(s) close to the Bragg wavelength

$$\lambda_B = 2n_{\mathrm{eff}}(T)\Lambda_B, \qquad (1)$$

where Λ_B is the Bragg grating period and n_{eff} is the effective refractive index of the waveguide mode. Since the effective refractive index is a function of temperature, a QC-DFB laser can be tuned either by directly changing the heat sink temperature or by adding a dc current ramp to the laser drive current to dissipate heat in the laser. A tuning rate from 0.4 to 0.65 nm/K at around 8 μm has been reported.[4] Typically, the laser would be coarse tuned through the heat sink temperature to close to the spectral features of interest, and then fine tuned with current. However, for significant tuning (>10 nm) this method of tuning, especially the coarse tuning, requires the operating temperature of the laser to cover a range up to 100 K, which impairs the compactness, portability, and power output of the lasers. In this letter we report on an approach to enable a spectral shift of room temperature QC lasers that does not require the change of the laser temperature. We achieve this by adding chalcogenide glass claddings to DBR gratings and using the photorefractive properties of the chalcogenide materials to shift the output.

Chalcogenide glasses are amorphous compounds containing sulfur, selenium, or tellurium, and are known to be low-loss mid-infrared materials.[5] A wide range of photoinduced phenomena have been discussed, including photodarkening, photobleaching, and photodissolution of metals into the chalcogenide glass.[6] Recently, attention has focused on their high optical nonlinearity,[7] photoinduced index contrast, and excellent infrared transparency for optical applications such as all-optical switches[8] and laser-written waveguides.[9] In this letter, we examine arsenic sulfide (As_2S_3) as a cladding material for QC lasers. Photodarkening of As_2S_3 features an increase of the refractive index by ~1.5% using above band gap illumination.[10] Controlled illumination of the cladding material enables us to shift the spectrum of QC lasers by several wave numbers, as a result of manipulating the effective refractive index of a DBR grating.

The DBR gratings on the QC lasers are fabricated by a dual-beam focused ion beam system Strata DB-235 and an example is shown in Fig. 1. The DBR gratings are located near the back facet of the laser ridge; the gratings are about 300 μm long and have a periodicity of 1.22 μm, a duty cycle of about 50%, and a depth of approximately 1.8 μm. The emission wavelengths of lasers processed in this fashion

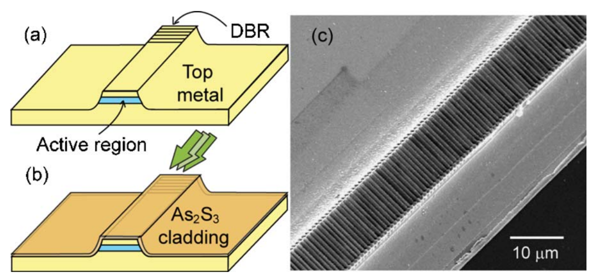

FIG. 1. (Color online) Structure of the gratings and cladding: (a) Schematic of a QC laser with DBR gratings fabricated into back facet mirror on top; (b) QC laser with As_2S_3 cladding, the refractive index of which can be modified by above band gap illumination (arrows); (c) scanning electron microscope image of the DBR grating fabricated by focused ion beam milling.

[a)]Author to whom correspondence should be addressed; electronic mail: shanshan@princeton.edu
[b)]Also at: Department of Mechanical and Aerospace Engineering, Princeton University.

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are ~7.9 μm. The quantum design and operating characterization of the QC lasers used in this study is described elsewhere.[11]

The chalcogenide claddings are produced by grinding bulk As_2S_3 (typically 99.9% pure) into a fine powder using a ceramic mortar and pestle; 2.50±0.03 g of the powder is combined with 10±0.1 ml of butylamine and allowed to dissolve for four days.[12] An ultrasonic homogenizer is used to expedite this dissolving process. The solution is then centrifuged at 3000 rpm for 3 minute to remove any suspended particulates or impurities. Exposure of solution or samples to ambient light and atmospheric moisture is kept to a minimum throughout the preparation procedure.

We apply 30 μl of solution onto a bar of 5–10 lasers already attached to the submount and with the lasers wire bonded. The QC laser sample is then baked in a vacuum oven for 7 h at 45 °C allowing the butylamine to evaporate. The As_2S_3 film deposited in this fashion is proved to be amorphous by X-ray diffraction. Differential scanning calorimeter measurement gives the glass transition temperature T_g=185 °C. Optical microscopy reveals that this method is able to produce a complete and even coating on QC lasers. A comparison of the characteristics of the original and coated laser shows that the deposition and baking process do not adversely affect the operation of the laser.

The laser emission spectra are measured with a Nicolet Fourier transform infrared spectrometer using a cooled mercury cadmium telluride detector and a resolution of 0.125 cm^{-1}. The QC lasers are operated in pulsed mode at room temperature with a pulse length of 100 ns and a repetition rate of 80 kHz. Above band gap illumination is achieved with a UV black light using a standard F4T5-BLB bulb with a main wavelength of 368 nm. Below band gap illumination with a 50 mW rated 650 nm continuous wave diode laser has also been used as a control measurement.

For the DBR structures, the Bragg condition gives

$$\lambda_B = 2(n_{\mathrm{eff}\,1}d_1 + n_{\mathrm{eff}\,2}d_2), \tag{2}$$

where n_{eff1} and n_{eff2} are the effective refractive indices of the unetched and etched part of laser cavity, d_1 and d_2 are the widths of the unetched and etched part in one period ($d_1+d_2=\Lambda_B$), and d_1/Λ_B is the duty cycle. A change of the effective refractive index in the Bragg grating leads to a shift of the peak reflection wavelength and hence the preferred laser modes. The relationship between the change in effective refractive index Δn_{eff2} and the shift of the emission wave number $\Delta\nu$ can be derived from Eq. (2) and expressed as

$$\frac{\Delta\nu}{\nu} = -\frac{d_2}{d_1+d_2}\frac{\Delta n_{\mathrm{eff}\,2}}{n_{\mathrm{eff}\,2}}, \tag{3}$$

where n_{eff2} is affected both by filling the grooves of the DBR grating with cladding material and by optically modifying the cladding itself. Figure 2 shows this phenomenon. As expected, there is a shift in the spectrum to lower wave numbers corresponding to higher effective refractive index, described by Eq. (3). The emission peak shifted −3.80 and −1.31 cm^{-1}, i.e., by 0.30% and 0.10%, by cladding and illumination, respectively. Additional above band gap illumination shows no effect on the output and the peak positions are stabilized thereafter.

The measured change in the effective refractive index is significantly lower than the change of pure chalcogenide

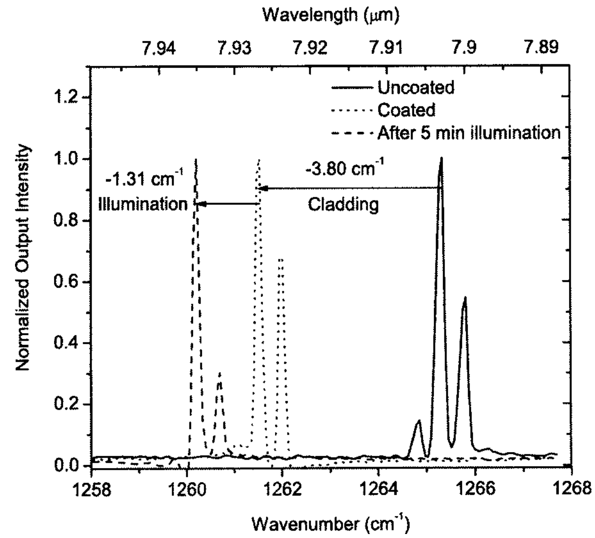

FIG. 2. Output spectra of a QC laser before and after application and modification of the As_2S_3 cladding. The emission wavelengths shift to successively lower wave numbers as indicated. Redshifts of 3.80 cm^{-1} for cladding and 1.31 cm^{-1} for illuminating the cladding are denoted with dotted and dashed lines, respectively.

where the photoinduced index change can be as high as 1.5%[10] in the visible and near infrared, because the Bragg grating overlaps only with the exponential tail of the waveguide mode. This fact limits the maximum achievable strength of the wavelength shift through the cladding. The strength of the wavelength shift can be reinforced by fabricating gratings with bigger depth or smaller duty cycle. Furthermore, since the photodarkening effect is irreversible, we can only achieve redshifting of the wavelength with the existing materials. Nevertheless, chemical methods of removing the cladding or annealing above the glass transition temperature have the potential to reset the initial refractive index of the glass and the emission wavelength of the laser.

A control experiment was conducted to verify that the output spectrum remains constant under sub-band gap illumination. The modes of the clad laser remain unchanged from the unexposed sample after 5h of laser operation and illumination at 650 nm. This finding demonstrates that the chalcogenide glass cladding can be modified by UV light but remains unaffected by subsequent mid-IR laser operation. However, since ambient light and/or sun includes above band gap energy and might affect its refractive index, exposure of the lasers to ambient light should be kept to a minimum throughout the processing and operation.

Finally, we use a one-dimensional numerical simulation based on the effective refractive indices in each Bragg mirror layer to calculate the tuning of the Bragg wavelength by the cladding, thereby we treat the semiconductor layers assuming a simple Drude model and the chalcogenide as a dielectric. The effective refractive indices are calculated by numerically solving Helmholtz equations for a slab waveguide. For the uncoated grating we find n_{eff1}=3.285 and n_{eff2}=3.273 at an emission wavelength of 7.9 μm; in the region with chalcogenide glass cladding we find n_{eff2}=3.277. Using the transfer matrix method for a Bragg mirror,[13] we calculate the reflection factor for the intensity as a function of wave number. The stop band of the Bragg mirror is presented in Fig. 3. This theoretical calculation shows the same trends as the experiments; nevertheless, we

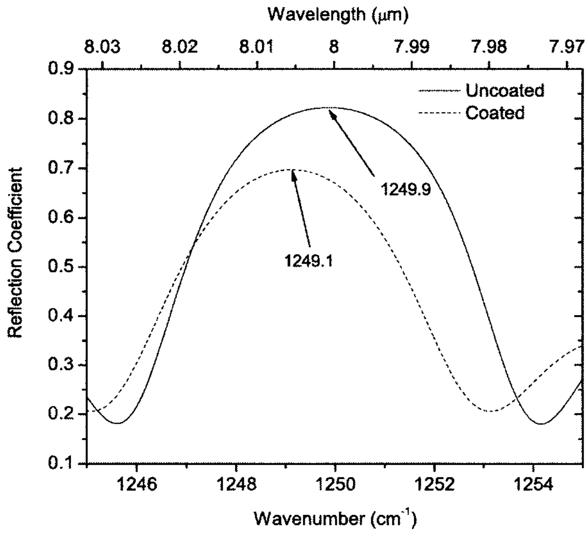

FIG. 3. Calculated reflection coefficient of the Bragg mirror as a function of wave number.

measure a stronger shift than calculated. This is understandable given the simplicity of the model used.

The precise emission wavelengths of QC lasers depend sensitively on variations or uncertainties in the fabrication process of the gratings. By combining a photorefractive As_2S_3 cladding with a DBR grating on top of QC lasers, we can achieve coarse redshift of the emission wavelengths after all other processing, and even laser testing is completed, and therefore account for process variation in these devices. A shift in wavelength of 0.3% and 0.1% is achieved by cladding and illumination by above band gap frequencies, respectively. We further show that the low temperature, wet solution process does not impede the function of the lasers, and that the laser waveguide tuning is unaffected by sub-band gap illumination.

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