Continuous Wave Mid-Infrared IV–VI Vertical Cavity Surface Emitting Lasers

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Continuous wave emission of mid-infrared IV-VI vertical-cavity surface-emitting laser structures with PbSe as active medium is demonstrated for the 6 to 8 µm wavelength region. The lasers are based on ultra-high finesse microcavity structures formed by high reflectivity EuSe/PbEuSe Bragg mirrors. Optically pumped cw laser emission is observed up to temperatures of 120 K. We achieved internal threshold pump intensities of down to 25 W/cm², which is two orders of magnitude smaller than reported so far. The line width of the laser emission is only 18 µeV (0.9 nm) with a strong narrowing as compared to the line width of the cavity resonance. Continuous wave output powers are up to 4.8 mW.

Introduction

Narrow bandwidth coherent mid-infrared emitters are extremely useful for ultrahighsensitive chemical gas analysis and atmospheric pollution monitoring [1] because most strong absorption lines of molecular gases are found in the mid-infrared spectral region. Due to their nearly symmetric conduction and valence bands and their very small non-raditative Auger recombination rates [2], the IV-VI semiconductors or lead salt compounds are very well suited for such applications. Therefore, edge emitting IV-VI semiconductor laser diodes have long been used for MIR gas spectroscopy and detection with operation temperatures up to 223 K in continuous wave (cw) [3] and above room temperature in pulsed mode [4]. As compared to edge emitting lasers, vertical cavity surface emitting lasers (VCSELs) offer several advantages such as very small beam divergences, single mode operation, and the possibility of monolithic integration and potentially lower threshold pump powers. For IV-VI lasers, another important advantage of VCSELs is that they can be grown on readily available BaF2 substrates, which have a much higher thermal conductivity and mechanical hardness than the usual high-cost IV-VI substrates. Since their first demonstration [5], [6], the performance of optically pumped IV-VI semiconductor vertical-cavity surface-emitting lasers (VCSELs) has been continuously improved [7], and pulsed laser emission has been obtained up to 65°C [8]. In contrast, cw operation of mid-infrared VCSELs at higher temperatures has not been achieved so far, and the longest wavelength for cwoperating mid-infrared VCSELs reported has been 2.9 µm for a type-II antimonide active region [9]. In the present work, we report the fabrication and operation characteristics of cw PbSe VCSEL samples for long-wavelength emission between 6.7 and 8 µm.

Structure and Design

As our previous photoluminescence studies have shown that PbSe exhibits superior luminescence efficiency as compared to PbTe [10], Se-based compounds were used

for all VCSEL layers. The microcavity structures were designed by matrix transfer calculations [5] – [8]. Due to the strong temperature dependence of the band gap, IV-VI VCSELs have to be tailored for a certain operation temperature [7], [8]. As PbSe emits at 6.5 µm or longer only at cryogenic temperature, the lasers were designed for an operation temperature of around 90 and 4 K. Thus, the VCSEL structure is formed by two high-reflectivity Bragg mirrors consisting of five $\lambda/4$ EuSe/Pb_{0.94}Eu_{0.06}Se layer pairs. Due to the high refractive index contrast of 50% between these mirror materials, only five layer pairs are required to obtain a reflectivity well above 99%.

The 2 λ thick cavity region between the mirrors consists of a 2.2 µm Pb_{0.94}Eu_{0.06}Se buffer and a 1.1 µm PbSe active region. The cavity design ensures that neither the PbSe laser emission nor the 5.3 µm pump laser is absorbed in the mirror layers, and that the pump wavelength does not coincide with the mirror stop band. The complete laser structure was grown by molecular beam epitaxy onto (111) BaF₂ substrates at a substrate temperature of 250 °C. Two cw-VCSELs were fabricated for different operation temperatures and emission wavelengths. The first one was designed for operation at liquid He temperature, and thus, the cavity resonance wavelength was set to the 7.9 µm PbSe band gap emission at 4 K. The second VCSEL was designed for 85 K operation with a corresponding PbSe emission at about 6.7 µm. A cross sectional electron microscopy image of this VCSEL is shown in the insert of Fig. 1 (b).



Fig. 1: (a) Reflectivity of the 7.9 μm cw-VCSEL, and (b) transmission of the 6.7 μm cw-VCSEL, both measured at 300 K. The solid lines correspond to the theoretical spectra calculated using the transfer matrix method. The insert in (a) shows the transmission around the central cavity mode at 60 K on an enlarged scale, and the insert in (b) the cross-sectional scanning electron microscopy image of the VCSEL structure. Right hand side: (c) Emission spectra of the 7.9 μm VCSEL at 2 K in cw (dots) and pulsed mode (triangles), both at 400 mW pump power. (d) Emission of the 6.7 μm VCSEL at 85 K at a pump power of 95 mW (■) above threshold and 43 mW (□) below threshold.

Optical Properties

Figure 1 (a) shows the reflectivity spectrum of the 7.9 µm VCSEL measured at 300 K. At this temperature, the spectrum exhibits three narrow cavity resonances of $m = 3^{rd}$, 4^{th} , and 5^{th} order at 133, 163, and 190 meV (9.33, 7.61, and 6.53 $\mu\text{m}),$ respectively. The measured line width of the 3rd resonance mode is 0.25 meV, whereas that of the central 4th order mode is only 0.10 meV, demonstrating a high cavity finesse of about 400, taking into account the order of the mode. The calculated reflectivity spectrum, shown by the solid line in Fig. 1 (a), is in very good agreement with the experimental data. Owing to the decrease of the energy band gap of PbSe from 279 meV at 300 K to 146 meV at 2 K, at low temperature the higher energy cavity resonances are damped by the interband PbSe absorption. Thus, at 60 K (see insert of Fig. 1 (a)), the 4th order cavity resonance is broadened from 0.10 meV at room temperature to 0.13 meV. At even lower temperatures, the 4th order resonance completely disappears because this wavelength is completely absorbed by the PbSe active region. Comparing the 300 K and 60 K spectra one can see that the spectral position of the resonances shifts to lower energies at a rate of 22 µeV/K to 158 meV or 7.8 µm at 60 L due to the increasing refractive index of the cavity material. For the room temperature transmission spectrum of the 6.7 µm VCSEL (Fig. 1 (b)), again, three cavity resonance peaks are observed, but now at energies of 155, 190 and 223 meV or λ = 8.0, 6.53, and 5.23 µm, respectively. The line width of the central 4th order cavity mode is only 0.17 meV or 7 nm, indicating a cavity finesse of about 300. As the temperature is lowered, again the resonance positions are red-shifted due to the increasing refractive index and below 100 K the central cavity mode becomes damped due to the PbSe absorption.

Laser Emission

Both VCSELs were optically pumped with a cw-CO laser emitting at a 5.28 µm wavelength (235 meV). The pump beam was focused onto the sample surface to a spot size of about 200 µm and the emitted light was detected with an HgCdTe detector through a grating spectrometer using the lock-in technique. The total emitted power was measured with a calibrated detector using an InSb long pass filter. Figure 1 (c) shows the emitted laser line for cw as well as pulsed excitation with 400 mW pump power in both cases. At 2 K, the stimulated emission of the 4th order resonance of the 7.9 µm VCSEL is found at 156.51 meV, *i.e.*, λ = 7.92 µm. This represents the longest emission wavelength of all VCSELs reported to date. For cw operation, an extremely narrow line width of only 12 µeV (0.6 nm) is observed, as found from the Lorentzian line fits shown as solid lines in Fig. 1 (c). This line width is 10 times smaller than the 130 µeV width found for the unpumped cavity resonance in the 60 K transmission measurements of Fig. 1 (a); and it is also much narrower than that observed for pulsed mode emission that is shown by the solid triangles in Fig. 1 (c). At 400 mW peak power excitation, the FWHM of the pulsed laser emission is 45 µeV (2.3 nm), but increases up to 100 µeV when the pump power is increased. This is still below the width of the unpumped cavity and is attributed to dynamical broadening effects. For cw-excitation, the measured line width is exactly the resolution of the spectrometer setup. Thus, the true cw line width must be actually much smaller than 0.6 nm.

The emission spectra of the 6.7 μ m VCSEL at 85 K are shown in Fig. 1 (d). At a low pump power of 43 mW, a weak emission at 6.685 μ m with an 11 nm FWHM is found (open squares in Fig. 1 (d)), which corresponds to the expected wavelength and line width of the 4th order cavity resonance at 85 K. Therefore, this signal is attributed to spontaneous emission, filtered out by the cavity mode. At a doubled excitation power of 95 mW (filled squares in Fig. 1 (d)), the emitted intensity increases drastically by a factor of 5000 and it is a factor of 20 narrower. The line width as determined by a Gaussian line fit (solid line) is only 0.6 nm or 16 μ eV, again limited only by the spectrometer



resolution (denoted by -||- in the figure). Thus, above threshold we observe a very strong spectral narrowing not only with respect to the width of the passive cavity mode, but also compared to the spontaneous emission signal.

Fig. 2: (a) Cw output power of the 7.9 μm VCSEL plotted as a function of pump power at 2 K: (o) spontaneous emission, (●) laser emission. The 25 mW laser threshold is determined from the inflection point. Inset: pulsed laser output versus excitation power. (b) Cw output power of the same VCSEL at 130 mW pump power plotted versus operation temperature. (c) Cw output intensity of the 6.7 μm VCSEL plotted versus pump power at temperatures between 60 and 110 K: full symbols: stimulated emission, open symbols: spontaneous emission. Insert output intensity versus pump power at 85 K on a linear scale, with the thresholds indicated by arrows. (d) Tuning characteristics of the 6.7 μm VCSEL: emission energy and emission wavelength plotted versus operation temperature. This yields a linear tuning coefficient of +0.058 meV/K or -2.1 nm/K. The insert shows the measured angular emission characteristics of the laser (dots). The solid line represents the Gaussian line fit to the data points.

Figure 2 (a) shows the cw power emitted by the 7.9 μ m VCSEL as a function of excitation power. For weak excitation, only spontaneous emission (open circles) is observed. Above threshold, the emitted power rapidly increases (full circles). As can be seen by the inflection point of the S-shaped curve of the double logarithmic plot in Fig. 2 (a), the external laser threshold is 25 mW. Since the pump laser was focused to a diameter of 200 μ m, this corresponds to an internal threshold of 25 W/cm², where the 63 % reflectivity of the laser structure at the pump energy is taken into account. At an excitation power of 1.2 W, the cw power emitted through one Bragg mirror is 4.8 mW. This is the highest observed from any cw VCSEL in the mid-infrared. For higher excitation powers, pulsed mode excitation was used in order to prevent thermal damaging of the sample. The inset of Fig. 2 (a) shows the emitted pulse peak power as a function of pump power. For 500 W pump pulses, a peak power of 23 W is emitted from one side of the VCSEL. Due to the symmetric cavity design, the total emitted power is twice the power measured from one side. Therefore, the conversion efficiencies for cw and pulsed operation at maximum output powers are 0.8 % and 9 %, respectively. The maximum pulsed mode efficiency is 20 %, at 100 W pump power. Below 100 W, the slope efficiency determined by linear fits is 24 % whereas for CW operation a value of 1% was found.

The temperature dependence of the 7.9 μ m VCSEL emission is shown in Fig. 2 (b) for a constant 130 mW excitation power. Up to 30 K, the spontaneous emission of the PbSe active material shifts closer to the 4th order cavity mode. Thus, the emitted intensity increases. At 30 K, the maximum of the spontaneous emission coincides with the cavity resonance, manifested in best laser performance. Above 30 K, the emitted intensity is reduced because the maximum of the spontaneous emission shifts above the cavity mode. At 57 K, the laser emission is quenched due to complete detuning of resonance and spontaneous emission.

The threshold behavior of the 6.7 µm VCSEL sample is shown in Fig. 2 (c) for different temperatures. Again, the data is plotted on a logarithmic scale to reveal also the subthreshold signals. For clarity, the measurement for 85 K is also plotted in the inset on a linear scale. The threshold pump power was evaluated for operation temperatures of 70 (triangles), 85 (squares) and 100 K (circles) to be 87, 57 and 83 mW, respectively, as indicated by the arrows at the inflection points of the logarithmic plots. Thus, the minimum threshold is at the design temperature, where the maximum of the spontaneous emission and the cavity mode position coincide. The minimum pump threshold of 57 mW at 85 K corresponds to an internal threshold power density of 67 W/cm². In the logarithmic plots one can not only see the sub-threshold signals, but also the emission at 60 and 110 K (open symbols in Fig. 2 (c)), at which only weak and much broader spontaneous emission is observed. Thus, this VCSEL operates only at temperatures between 65 and 100 K because at higher as well as lower temperatures, the PbSe emission is above or below the cavity mode. The maximum observed total cw output power at 85 K is 1.2 mW at a pump power of 230 mW, which corresponds to a conversion efficiency of 0.5 %. We have also fabricated a third VCSEL with a design wavelength of 6.5 µm. This laser shows essentially the same performance as the 6.7 µm laser, but due to the shorter wavelength it lases up to a temperature of 120 K.

For applications of the lasers for molecular spectroscopy, the tuning ability and beam divergence are of crucial importance. These are characterized in detail in Fig. 2 (d) for the 6.7 µm VCSEL. The total tuning range by temperature variation was found to be 70 nm, with a linear tuning coefficient of +0.058 meV/K or -2.1 nm/K. This large tuning range is particularly advantageous for spectroscopy and it is substantially larger as compared to that of III-V quantum cascade lasers operating in the same wavelength range with a typical tuning coefficient of 0.01 meV/K [11]. In addition, we have recently shown that the emission wavelength can also be tuned quite elegantly by applying external magnetic fields [12]. In this case, we have found that the laser emission splits into two circularly polarized lines with opposite polarization. Therefore, the lasing action involves only spin polarized electronic states, *i.e.*, these VCSELs can be viewed as a novel kind of spin laser. We have also determined the angular emission characteristic of the cw VCSELs. As demonstrated in the inset of Fig. 2 (d) for an emission angle of only 1° off the surface normal (see sketch in the inset), the emission intensity (filled squares) essentially drops to zero, and the Gaussian line fit to the emission profile (solid line) yields a half width at half maximum of only 0.5°. This narrow forward directed emission is a clear advantage of the VCSELs as compared to edge emitters. Up to now, the cw-operation temperatures were limited by the pump laser wavelength as well as the temperature dependence of the PbSe photoluminescence emission. It is therefore anticipated that already minor changes in the cavity structure and active region design as well as the use of other pump sources will soon lead to higher cw-laser operation temperatures.

Conclusion

In conclusion, lead salt vertical-cavity surface-emitting lasers offer attractive properties as coherent infrared laser sources. They feature single mode operation, emit circularly shaped parallel beams with extremely small beam divergence, and exhibit very sharp emission lines widths below 12 μ eV. Based on the use of high finesse infrared microcavity structures, cw-operation up to 120 K was demonstrated, which is expected to be significantly increased in the near future. In comparison with quantum cascade lasers, the lead salt VCSELs show a substantially larger wavelength tunability, which is of crucial importance for spectroscopy applications.

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