

Optical pumping of IV-VI semiconductor multiple quantum well materials using a GaSb-based laser with emission at $\lambda=2.5 \mu\text{m}$

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(Received 30 August 2004; accepted 30 November 2004; published online 10 February 2005)

Room-temperature photoluminescence (PL) measurements of IV-VI semiconductor multiple quantum well (MQW) structures using diode laser optical pumping at two different excitation wavelengths, 2.5 and 0.91 μm , are described. Active region temperatures during continuous-wave optical pumping were determined from blueshifts in PL emission energies. Temperatures were between 22.7 and 29.5 °C lower for 2.5- μm pumping as compared to 0.91- μm pumping at the same power level of 100 mW. Heating effects are also shown to be smaller for MQW samples with more narrow PbSe wells. © 2005 American Institute of Physics. [DOI: 10.1063/1.1851601]

Optical pumping of narrow-band-gap semiconductor materials has recently been employed as an effective way to demonstrate midinfrared laser emission.¹⁻³ Advantages over electrical pumping include more rapid demonstration of device concepts without also needing to develop *p-n* junctions and low-resistance electrical contacts. Most prior work using diode lasers as a pumping source has employed wavelengths in the 1- μm spectral range, but this causes excessive heating of mid-IR active region materials because there is as much as a 1 eV of excess energy above what is needed to excite electron-hole pairs. Pulsed operation with duty cycles of much less than 1% would thus be necessary to avoid excessive heating of the narrow-band-gap material. Important mid-IR laser applications such as molecular spectroscopy^{4,5} require continuous-wave (cw) operation, so this excess heating problem needs to be solved if optically pumped mid-IR lasers are to become commercially viable. A possible solution is to use GaSb-based lasers with emission at wavelengths longer than 2 μm so that electron-hole pairs are generated in the mid-IR material without also generating a large number of phonons. This article describes the demonstration of more efficient optical pumping using a GaSb-based laser with emission at $\lambda=2.5 \mu\text{m}$ (500 meV) to excite mid-IR photoluminescence (PL) from IV-VI semiconductor multiple quantum well (MQW) structures. Results are compared with the PL spectra obtained with pumping at $\lambda=0.91 \mu\text{m}$ (1.37 eV). Active region temperatures during cw optical pumping, determined from blueshifts in PL emission energies, were between 22.7 and 29.5 °C lower for 2.5- μm pumping as compared to 0.91- μm pumping at the same power level of 100 mW. The effects of different MQW struc-

tures on active region heat dissipation are also analyzed.

The IV-VI MQW samples used in this study were grown on freshly cleaved $1 \times 1 \text{ cm}^2$ BaF₂ (111) substrates by molecular-beam epitaxy (MBE) using an Intevac GEN II modular system. All samples had a buffer layer structure consisting of BaF₂ (100–400 nm thick) grown at 400–500 °C followed by a Pb_{0.93}Sr_{0.07}Se layer (3.3–4.4 μm thick) grown with a 3% Sr-to-PbSe flux ratio at 360–390 °C. Four different 40-period MQW structures were then grown at 360–390 °C with PbSe quantum well and Pb_{0.93}Sr_{0.07}Se barrier layer thicknesses in the range of 6.7–43.0 nm and 47.5–57.5 nm, respectively. A final PbSe cap layer (22–120 nm thick) was grown on top of the MQW structure to prevent oxidation of the Sr in the barrier layer material. All thicknesses were determined using calculated growth rates obtained from previously grown single-layer thickness measurements, and all IV-VI layers were grown with a 10% Se-to-PbSe flux ratio provided by a valved cracker effusion cell to keep the growth surface under Se-rich conditions. Room-temperature band gaps of the PbSe well and Pb_{0.93}Sr_{0.07}Se barrier layers are 278 and 451 meV, respectively.⁶

PL measurements were performed using cw diode laser pumping at two different wavelengths, 0.91 and 2.5 μm , and a modular Fourier transform infrared (FTIR) spectrometer (MIR8000, Oriel, Stratford, CT) equipped with a liquid-nitrogen-cooled photoconductive HgCdTe detector with a 13- μm cutoff wavelength. The output of the InGaAs-based fiber-coupled $\lambda=0.91\text{-}\mu\text{m}$ pump laser (#LT2000-2W, Laser-tel, Tucson, AZ) was delivered normally to the MQW sample surface by a 12-in.-long silica glass fiber producing a spot size of about 1 mm in diameter for a fiber-to-sample distance of about 1.6 mm. Fiber-delivered laser power was measured with a hand-held power meter (#3803, New Focus, San Jose, CA) for a range of injection currents between 325 and 500 mA, giving a range of powers (densities) between

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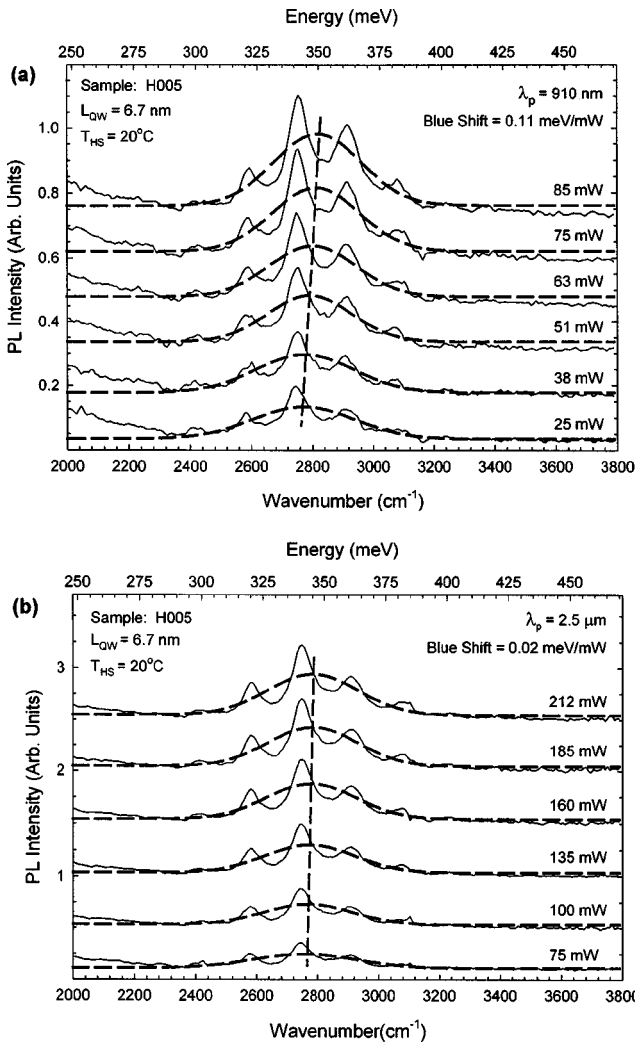


FIG. 1. PL spectra with Gaussian fits from a 40-period PbSe/Pb_{0.93}Sr_{0.07}Se MQW sample (#H005) grown by MBE on BaF₂ (111) with 6.7-nm-thick PbSe wells using different diode laser pumping powers at (a) $\lambda = 0.91$ μm and at (b) $\lambda = 2.5$ μm . Both tests were performed with a constant heat sink temperature of 20°C .

25 mW (3.18 W/cm²) and 85 mW (10.8 W/cm²). The output of the bare-faceted GaSb-based $\lambda = 2.5$ - μm laser, the fabrication of which is described in Ref. 7, was delivered to the MQW sample by placing the exposed laser facet about 1.6 mm from the epilayer surface producing an elliptical spot covering a 2×2.4 mm² area. Optical power was measured using a thermal power meter (#PM3, Molectron, Portland, OR) for injection currents between 600 and 2500 mA, giving a range of powers (densities) between 75 mW (1.97 W/cm²) and 212 mW (5.58 W/cm²). Mid-IR PL emission from the MQW sample was collected with a 2-in.-diameter gold-coated off-axis parabolic mirror (OAPM) from the backside of the transparent BaF₂ substrate, then passed through the FTIR interferometer and focused by a second OAPM onto the HgCdTe detector. The MQW samples were attached to a copper heat sink, which was stabilized at 20°C with a single-stage thermoelectric cooling module and thermistor mounted adjacent to the sample, using an Al₂O₃ adhesive interface pad (Melcor, NJ) between the BaF₂ and the copper. The lateral distance between the illuminated area of the

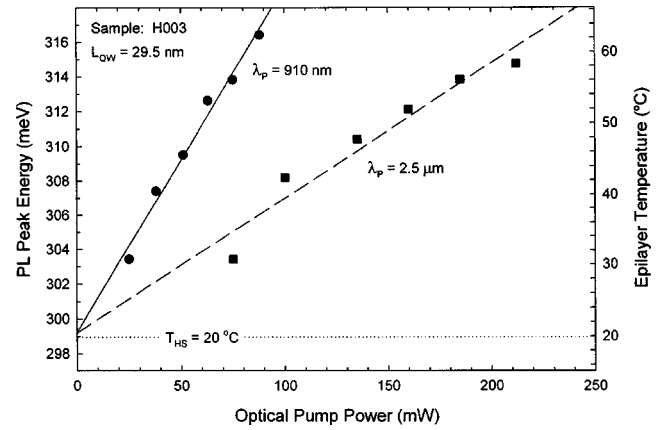


FIG. 2. Gaussian-fitted PL peak energies as a function of incident optical pump power for $\lambda = 0.91$ - and $\lambda = 2.5$ - μm pumping from a 40-period PbSe/Pb_{0.93}Sr_{0.07}Se MQW sample (#H003) with 29.57-nm-thick PbSe wells. The 20°C heat sink serves as a reference temperature for calculation of epilayer temperature shown on the right-hand scale.

MQW sample and the copper heat sink was about 3 mm.

Figure 1 shows measured PL spectra obtained from a sample with a PbSe well thickness of 6.7 nm (#H005) using optical pumping at (a) $\lambda = 0.91$ μm and at (b) $\lambda = 2.5$ μm . Gaussian fits are used to obtain peak PL energies for each pumping level since Fabry–Perot fringes originating from optical resonance in the 5.4- μm -thick epitaxial layer dominate each measured spectrum. Blueshifts in the peak PL energies occur as pumping power increases because of an increase in the band-gap energy caused by optical heating of the epilayer. Figure 2 shows PL peak energy versus incident optical pump power (note that absorbed power is lower by about 50% due to reflection losses) at 0.91- and 2.5- μm pumping wavelengths for another sample with a PbSe well thickness of 29.5 nm (#H003). The right-hand scale shows calculated epilayer temperatures obtained using the blueshift data and the temperature tuning coefficient of 0.41 meV/K for PbSe.⁸ Linear extrapolation to zero pumping power for both wavelengths gives the same PL energy of 299 meV, which represents the interband normal valley transition energy for an “unheated” epilayer that is at the same temperature as the 20°C heat sink. This extrapolated “zero-heating” PL energy agrees fairly well with the 290-meV value expected for a 29.5-nm PbSe well width MQW sample at 20°C based on $n = 1$ normal valley interband transition energies obtained from differential transmission spectroscopy (DTS) experiments where minimal epilayer heating occurs during measurement.⁹ Good convergence of PL energy at zero pumping power to the unheated 20°C interband transition energy for the two different pumping wavelengths confirms a linear heating effect for both pumping configurations where total power delivered to the epilayer is the relevant parameter rather than power density. This suggests rapid lateral heat spreading in the epilayer since the power densities differed by about a factor of 2.

Table I summarizes the data collected from four MQW samples at a 20°C heat sink with different PbSe well widths. Extrapolated PL energy at zero pumping power yielded values in agreement with expected $n = 1$ normal valley interband transition energies for all samples except

TABLE I. PL energy data for four PbSe/PbSrSe MQW samples with different PbSe well widths and calculated epilayer temperatures, from linear fits to the PL blueshift data, for $\lambda=0.91\text{-}\mu\text{m}$ and $\lambda=2.5\text{-}\mu\text{m}$ diode laser pumping.

Sample	PbSe well width (nm)	PL energy at 0 mW (meV)	Pump laser wavelength (μm)	PL energy at 100 mW (meV)	Blueshift 0–100 mW (meV)	Shift rate (meV/W)	Epilayer temp at 100 mW ($^{\circ}\text{C}$)	ΔT at 100 mW ($^{\circ}\text{C}$)
H005	6.7	340.7	0.91	351.9	11.2	110	47.3	22.7
			2.5	342.6	1.9	20	24.6	
H002	16.5	332.5	0.91	346.5	14.0	140	54.1	24.1
			2.5	336.6	4.1	40	30.0	
H003	29.5	299.0	0.91	318.9	19.9	200	68.5	29.5
			2.5	306.8	7.8	80	39.0	
H004	43.0	294.0	0.91	310.3	16.3	160	59.8	26.9
			2.5	299.3	5.3	50	32.9	

#H002, which were too high by about 37 meV. This discrepancy could be due to an incorrect estimate for the PbSe well thickness in this sample. PL blueshifts corresponding to epilayer temperatures as high as 68.5°C are obtained for cw pumping at 100 mW. In each case there are significantly smaller PL blueshifts and epilayer heating for pumping at $\lambda=2.5\text{ }\mu\text{m}$ as compared with pumping at $\lambda=0.91\text{ }\mu\text{m}$. Heating differences range from 22.7 to 29.5°C and are larger for samples with wider PbSe wells. This temperature difference is directly associated with a smaller population of phonons created by the lower-energy photoexcited electrons (and holes) as they decay into quantized states in the PbSe wells. Longer wavelength pumping at $\lambda=2.5\text{ }\mu\text{m}$ excites valence-band electrons to only about 50 meV above the bottom of the conduction band of the $\text{Pb}_{0.93}\text{Sr}_{0.07}\text{Se}$ barrier material, whereas shorter wavelength pumping at $\lambda=0.91\text{ }\mu\text{m}$ excites electrons more than 910 meV into the conduction band of the barrier material.

The relatively smaller additional heating and lower epilayer temperatures observed for thinner PbSe well samples are probably due mostly to the quantum-defect effect—the energy difference between the laser pump energy and PL emission energy.¹⁰ The larger PL energy from the thinner well samples leaves a relatively smaller percentage of absorbed pump energy to contribute to nonradiative epilayer heating. For example, assuming the same quantum efficiencies, an additional 9% of the $\lambda=2.5\text{-}\mu\text{m}$ absorbed pump energy will not contribute to epilayer heating for a sample with 6.7-nm-thick wells as compared to a sample with 43-nm-thick wells. By contrast, only an additional 3% of the $\lambda=0.91\text{-}\mu\text{m}$ absorbed pump energy will not contribute to epilayer heating for a sample with 6.7-nm-thick wells as compared to a sample with 43-nm-thick wells. Other phenomena that can cause different heating effects include less Umklapp phonon scattering and thinner overall epilayer thicknesses for the thinner PbSe well samples, both of which allow better heat conduction normal to the MQW structure.

Results from these experiments show that use of GaSb-based diode lasers should enable the development of mid-IR lasers with cw operation at temperatures maintained with thermoelectric cooling (TEC) modules. This prediction is based on a recent demonstration of IV-VI vertical cavity sur-

face emitting lasers (VCSELs) with optical pumping laser thresholds of $10.5\text{ kW}/\text{cm}^2$ at 260 K.¹¹ Pumping was performed using a pulsed Ho:YAG (yttrium aluminum garnet) laser ($\lambda=2.098\text{ }\mu\text{m}$) with a low duty cycle (80-ns pulses, 1-Hz repetition rate), so it can be assumed that the active region quickly returns to the 260-K heat sink temperature between pulses, which can be considered the highest active region temperature at which this type of laser will operate. GaSb-based laser emission of 200 mW focused to a spot size of less than $50\text{ }\mu\text{m}$ will deliver in excess of $10\text{ kW}/\text{cm}^2$, and according to the average blueshift rate of $20\text{ meV}/\text{W}$ for $\lambda=2.5\text{ }\mu\text{m}$ pumping of sample #H005 there will be 9.8°C of active region heating for this cw lasing condition. A heat sink at about 250 K, achievable with the current TEC technology, can thus maintain a 260-K active region under mid-IR cw lasing conditions. By contrast, pumping with a $\lambda=0.91\text{-}\mu\text{m}$ diode laser, where the blueshift rate for sample #H005 is $110\text{ meV}/\text{W}$, there will be 53.7°C of active region heating. The heat sink would thus need to be below 207 K, which cannot be easily achieved with the existing TEC technology.

In conclusion, room-temperature cw PL measurements of IV-VI MQW structures allowed the determination of localized epitaxial layer temperatures for two different diode laser pumping wavelengths. It was shown that pumping at $\lambda=2.5\text{ }\mu\text{m}$ with a GaSb-based laser causes significantly less epilayer heating than pumping at $\lambda=0.91\text{ }\mu\text{m}$ with an InGaAs-based laser. Active region heating under cw pumping conditions at power-density levels high enough to achieve lasing in IV-VI VCSELs is sufficiently low to allow cooling with presently available TEC modules. It should therefore be possible to develop compact mid-IR laser sources with cw emission using this combined III-V and IV-VI semiconductor device technology.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation Grant Nos. (DMR-9802396, DMR-0080054, and EPS-0132354) and the Oklahoma Center for the Advancement of Science and Technology under Grant Nos. AR021-043 and AR03-057. SUNY authors would like to acknowl-

edge support from United States Air Force Office of Scientific Research, Grant No. F-49620-01-10108.

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