Tunable mid-infrared generation by difference frequency mixing of diode laser wavelengths in intersubband InGaAs/AlAs quantum wells

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We demonstrate difference frequency generation (DFG) of 8.66–11.34 μm wavelength light in intersubband InGaAs/AlAs quantum wells by mixing of 1.92 μm±25 nm and 2.39 μm±39 nm. The peak DFG second order nonlinear susceptibility $\chi^{(2)}$ is measured to be 12±1 nm/V, more than 65 times that of GaAs, at a difference frequency output wavelength of 9.50 μm. The intersubband absorption for the 1–2 and 1–3 transitions is measured to be 9.3 and 2.1 μm, respectively. Second harmonic generation (SHG) of 4.76, 5.12, and 5.36 μm light with a CO_2 laser is observed with a peak SHG $\chi^{(2)}$ of 52±3 nm/V. Good agreement of experiment with theory is found for both the linear and nonlinear optical properties. This demonstration of mid-infrared DFG opens the possibility for monolithic diode laser pumps and compact waveguide frequency converters as tunable midinfrared sources. *© 1995 American Institute of Physics.*

Compact mid-infrared sources have many potential applications from pollution monitoring and intelligent process controls to laser radar and noninvasive medical diagnosis. Recent developments in long wavelength diode lasers have pushed room temperature continuous operation to 2.3 μm using GaSb based materials.[1,2] However, room temperature operation of diode lasers of wavelengths much longer than 4 μm may not be possible due to Auger recombination.[3,4] In particular, the 8–12 μm atmospheric window may not be attainable at all with such lasers. An alternative approach is to use frequency conversion of near-infrared diode lasers to generate mid-infrared light. Intersubband quantum wells (QWs) are promising materials for such frequency conversion processes. Intersubband QWs have been demonstrated to have nonlinear susceptibilities several orders of magnitude larger than bulk materials.[5–8] Another important advantage of using QWs is the possibility of fabricating monolithic diode laser pumps and quasi-phasematched QW frequency converters as compact mid-infrared sources. Previously, these intersubband transitions had been limited to the far and mid-infrared, but recent advances in strained QW materials have allowed intersubband nonlinearities to reach near-infrared wavelengths[9–12] so that diode lasers could be used to pump the frequency conversion processes.

In this letter, we use the high indium content InGaAs/AlAs QW system which yields short wavelength intersubband transitions due to the large available conduction band offsets. First, the linear spectroscopy of the intersubband transitions will be discussed, followed by second harmonic generation (SHG) characterization of the QW second order nonlinear susceptibility $\chi^{(2)}$. Finally difference frequency generation (DFG) is demonstrated by mixing of near-infrared wavelengths of light to cover most of the 8–12 μm band.

The QW sample used for these studies was designed for mixing of wavelengths around 2 μm to generate 10 μm light. The sample was composed of 300 coupled QWs separated by 50 Å AlAs barriers. The coupled QWs consisted of $In_{0.5}Ga_{0.5}As$ wells of 9 and 7 monolayers in width separated by a 3 monolayer AlAs barrier. One monolayer of GaAs was added to either side of the coupled QW for interface smoothing.[13] The QWs were doped uniformly across the well regions at 5.0×10^{18} cm^{-3} for a sheet charge density of 2.3×10^{12} cm^{-2} per QW. The sample was grown on a (100) semi-insulating GaAs substrate by molecular beam epitaxy (MBE) in a Varian Gen II system with As_2 at a substrate temperature of 360 °C (calibrated by band edge absorption). A linearly graded InGaAs buffer with a final indium composition of 30% was used for strain compensation.[13,14]

The intersubband transition energies were calculated using a single band effective mass model with nonparabolicity included.[12] The transitions were predicted to be $E_{12}=107$ meV (11.6 μm) and $E_{13}=546$ meV (2.27 μm) with dipole moments of $z_{12}=12.3$ Å, $z_{13}=-5.8$ A, and $z_{23}=7.5$ Å. A Fourier transform infrared radiation (FTIR) spectrometer was used to measured the absorption from the sample. The measurement was performed with the sample mounted at Brewster's angle to TM polarized light. The measured absorption spectrum is shown in Fig. 1. Absorption peaks of $E_{12}=133$ meV (9.3 μm) and $E_{13}=591$ meV (2.10 μm) were observed. This E_{13} is one of the largest 1–3 transition energies to date.[9] The half-width at half-maximum (HWHM) transition linewidths were $\Gamma_{12}=11.5$ meV and $\Gamma_{13}=41.6$ meV. Assuming $z_{12}=12.3$ Å from theory, an effective sheet charge density of $\sigma_{\mathrm{eff}}=4.7\times10^{11}$ cm^{-2} per double QW was extracted.[15]

The second order nonlinear susceptibility $\chi^{(2)}$ of the QWs was calculated using a simple perturbative model[16] with linewidths and effective carrier concentration taken from the absorption measurement and dipole moments from theory.[9.17] For SHG, the calculated $\chi^{(2)}$ is plotted in Fig. 2. The two peaks in the QW $\chi^{(2)}$ at 9.55 and 4.3 μm correspond to singly resonant conditions, where the pump photon energy matches E_{12} and the second harmonic photon energy

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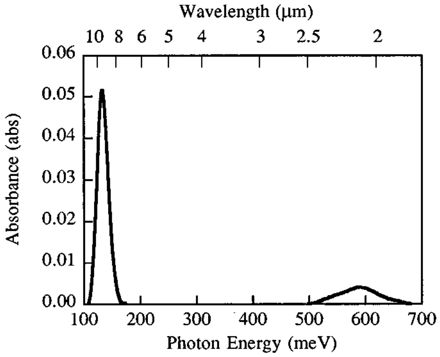

FIG. 1. Intersubband absorption spectrum of the asymmetric coupled QW sample showing peaks at 9.3 and 2.1 μm corresponding to the 1–2 and 1–3 transitions.

matches E_{13}, respectively. An interferometric technique was used to measure the QW $\chi^{(2)}$.[9,17] The SHG from the sample is a superposition of the fields from the GaAs substrate and QW. The $\chi^{(2)}$ tensor elements for the GaAs (xyz) and QW (zzz) are different so that polarization selection can be used to extract only the QW contribution. By selecting only TM polarizations, this extraction can be performed by measuring the SHG from the sample versus azimuthal angle ϕ of the sample about its normal (ϕ scans). With only a GaAs contribution, the SHG power is fourfold symmetric about the (001) normal to the sample. By adding a QW contribution, the SHG power becomes only twofold symmetric.[9]

The SHG measurements were performed with a CO_2 laser tuned to pump wavelengths of 9.51, 10.23, and 10.72 μm. The CO_2 laser used for the experiment generated ~300 ns pulses with peak powers of ~1 kW. The pump beam was focused down to a ~100 μm diameter spot on the sample. A multilayer dielectric filter was placed before the sample to block light at wavelengths shorter than ~8 μm. An InSb detector was used to measure the SHG power, and ZnSe polarizers were used to select only the TM polarization. A fraction of the pump was focused onto a $AgGaSe_2$ crystal as a reference SHG signal. Normalization of the SHG signal from the sample to fluctuations in the pump power were then eliminated by ratioing the SHG signal to the reference SHG power. Although we did not calibrate the setup for absolute power measurements, we estimate that the peak SHG power from the sample was on the order of a few milliwatts. With this setup, ϕ scans were taken at an incidence angle of ~45°. The parameters for extracting the QW $\chi^{(2)}$ were sample thickness of 384±1 μm, total MQW thickness of 3.3 μm, and bulk coherence lengths of 108.9±1, 107.5±1, and 105.6±1 μm at 9.51, 10.23, and 10.72 μm, respectively. The coherence lengths were calculated from published refractive index data.[18] The resulting measured QW $\chi^{(2)}$ values for SHG are shown in Fig. 2. The maximum QW $\chi^{(2)}$ of 52±3 nm/V, 280 times the bulk GaAs $\chi^{(2)}$ of 180 pm/V,[19] was measured at a pump wavelength of 9.51 μm. The error bars are absolute errors in the measured values resulting from inaccuracies in the sample thickness and coherence lengths. Good agreement of these measured values to theory is observed with slight deviations arising from inaccuracies in the coherence lengths, sample thickness, or transition line shapes.

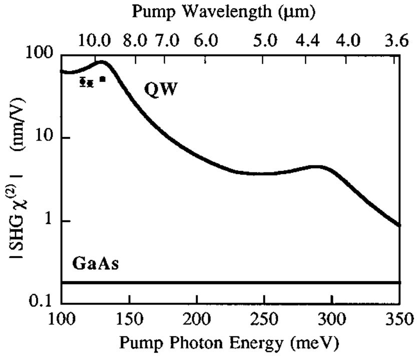

FIG. 2. Measured (points) and theoretical (solid curve) SHG $\chi^{(2)}$.

DFG measurements were performed using the same interferometric technique. The two pump beams for the DFG experiment were taken from an optical parametric oscillator (OPO). This OPO consisted of two walk-off compensated KTP crystals pumped by a Nd:YAG laser at a wavelength of 1.064 μm. Thus, the OPO signal and idler output beams have photon energies which sum to a 1.064 μm photon energy. The OPO used for this experiment generated pulses of ~10 ns duration with peak powers of several hundred kilowatts. Tuning of the OPO was accomplished by adjusting the angle of the KTP crystals, and a spectrometer with an InSb detector was used to accurately determine both of the pump wavelengths. Two 1.064 μm high reflectors were used to eliminate any transmitted 1.064 μm light, and a rutile prism polarizer was used to select the TM polarization for both beams to the sample. Glass filters were placed before the sample to eliminate any light at wavelengths longer than ~3 μm. A HgCdTe detector with a Ge window was used to measure the DFG signal. Several multilayer dielectric filters were used and verified to eliminate any radiation at wavelengths shorter than ~8 μm while the HgCdTe detector response ensured that radiation at wavelengths longer than ~12 μm was not detected. A wire grid polarizer was used to select only the TM polarization of the DFG output.

For DFG output wavelengths between 8 and 12 μm, the coherence length in GaAs is fairly short, on the order of 30 μm as calculated from refractive index data. Since the sample thickness is 384±1 μm, small inaccuracies in the coherence length cause large inaccuracies in the GaAs contribution to the DFG. This inaccuracy can be eliminated by determining the wavelength at which the GaAs contribution is null; this was accomplished by minimizing the DFG power while tuning the OPO. Using the technique, a GaAs null was measured at a DFG output wavelength of 8.86±0.05 μm. If the coherence lengths calculated from refractive index data are assumed to be correct, an effective sample length of 376.6±1.5 μm yields the correct GaAs null wavelength. On opposite sides of this GaAs null wavelength, the GaAs contribution to the DFG field has opposite signs; ϕ scans at 8.66 and 9.00 μm showing DFG power versus sample azimuthal angle are plotted in Fig. 3. The asymmetry between the 0°

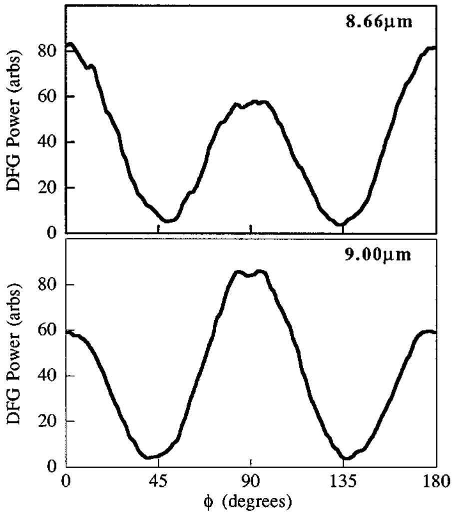

FIG. 3. DFG power vs azimuthal angle ϕ of the sample at output wavelengths of 8.66 and 9.00 μm showing opposite signs of asymmetry due to a sign reversal of the bulk GaAs contribution.

(or 180°) and 90° (or 270°) peaks arises from a QW contribution to the DFG signal. The opposite signs in the GaAs contribution for the two wavelengths result in opposite signs in the asymmetry in the peaks; at 8.66 μm, the 0° peak is higher than the 90° peak, while at 9.00 μm, the 90° peak is higher than the 0° peak.

By taking ϕ scans and using the calculated coherence lengths and effective sample length, accurate determination of the QW DFG $\chi^{(2)}$ was obtained over a wide wavelength range in the 8–12 μm window. A plot of the measured QW DFG $\chi^{(2)}$ versus DFG output wavelength is shown in Fig. 4. The error bars were determined from inaccuracy in the measured effective sample length. The theoretical DFG $\chi^{(2)}$ for this process was calculated using the same parameters (dipole moments, linewidths, and effective carrier concentration) in the same perturbative expression[16] as was used for the SHG $\chi^{(2)}$. This theoretical DFG $\chi^{(2)}$ is also plotted in Fig. 4. Excellent agreement of experiment to theory is observed. A peak DFG $\chi^{(2)}$ of 12±1 nm/V, more than 65 times that of bulk GaAs (also 180 pm/V for these wavelengths as calculated using Miller's rule),[19] at an output wavelength of 9.50 μm is measured. Tunable radiation from 8.66 to 11.34 μm was generated with large $\chi^{(2)}$'s for only slight tuning of the pump wavelengths (1.92 μm±25 nm and 2.39 μm±39 nm). This is the first demonstration of mid-infrared DFG in any QW system; far-infrared DFG by mixing of two CO_2 lasers in intersubband QWs was recently demonstrated.[20] The pump wavelengths used for our DFG coincide with recent room temperature diode laser wavelengths[1] so that integration of such a mixer with diode lasers might be possible.

In conclusion, we have demonstrated DFG of mid-infrared light by mixing of diode laser wavelengths in intersubband InGaAs/AlAs QWs. We have characterized the linear absorption and nonlinear optical properties; SHG and DFG were used to measure the $\chi^{(2)}$ of the QWs. Both measured absorption and $\chi^{(2)}$ are found to be in good agreement with theory. With large nonlinearities at diode laser wavelengths, these intersubband QWs combined with diode laser pumps should result in efficient difference frequency mixers as compact mid-infrared sources operating at room temperature.

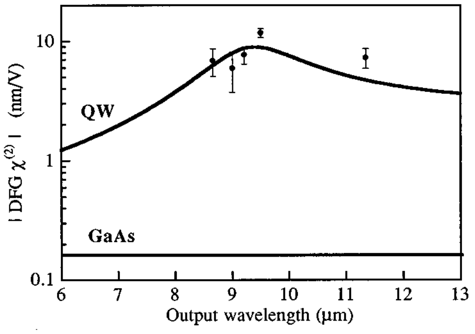

FIG. 4. Measured (points) and theoretical (solid curve) DFG $\chi^{(2)}$ for mixing of the signal and idler outputs from a 1.064 μm pumped OPO.

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