Short wavelength (5.36–1.85 μm) nonlinear spectroscopy of coupled InGaAs/AlAs intersubband quantum wells

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We report short wavelength second-harmonic generation (SHG) spectroscopy of asymmetric coupled $In_{0.6}Ga_{0.4}As$/AlAs quantum wells (QWs). The QW is designed to show maximum second-order nonlinear susceptibility $\chi^{(2)}$ for SHG of 4 and 2 μm wavelengths by single and double resonance effects, respectively. SHG spectroscopy across the midinfrared is measured using both a CO_2 and a free electron laser as pumps. The $\chi^{(2)}$ of the QW is extracted from interference of the second-harmonic fields from the QW and GaAs substrate, determined by the azimuthal dependence of the SHG power. We measure $\chi^{(2)}$ of the QW for harmonic wavelengths between 5.36 and 1.85 μm. This is the shortest wavelength SHG to date by any QW intersubband interaction. Good agreement of experiment with theory for the dispersion of $\chi^{(2)}$ for both singly and doubly resonant conversion is observed throughout the midinfrared. © *1994 American Institute of Physics.*

Near-infrared nonlinear interactions in the 1.55–2 μm wavelength range have recently been a subject of interest for frequency conversion and electro-optic switching. Intersubband transitions in quantum wells (QWs) present strong nonlinear susceptibilities, but these transitions have been limited until recently to the far and midinfrared.[1–4] Recent publications[5–7] have shown that short wavelength intersubband transitions in InGaAs/AlGaAs QWs can be achieved where room-temperature diode lasers are available.[8] We have previously reported intersubband absorption, second-harmonic generation (SHG), and difference frequency generation near 2 μm in InGaAs/AlAs QWs grown on GaAs.[7,9,10] In this letter, detailed spectroscopy of the quadratic nonlinear optical susceptibility $\chi^{(2)}$ of an InGaAs/AlAs coupled QW obtained by measuring SHG across the midinfrared is described. The experimental QW $\chi^{(2)}$ spectrum is explained by a perturbative model. While describing the $\chi^{(2)}$ for SHG, this model will be useful for predicting the $\chi^{(2)}$ for any interaction involving light in the characterized wavelength range.[11] For example, the $\chi^{(2)}$ for difference frequency generation from near infrared to midinfrared can be predicted with accuracy if the $\chi^{(2)}$ dispersion for SHG from midinfrared to near infrared is known.[10]

The asymmetric coupled $In_{0.6}Ga_{0.4}As$/AlAs intersubband QW structure used for this study was resonant at 4 and 2 μm and was described in detail in a previous letter.[9] This QW was optimized for doubly resonant SHG of 2 μm light and singly resonant SHG of 4 μm light. A single band effective mass model with nonparabolicity included was used to calculate the QW subband energies and dipole moments.[7] The absorption spectrum for the sample showed resonances at 4.1 and 2.1 μm for the 1–2 and 1–3 transitions, respectively. A dipole moment $z_{13}=4.2$ Å, effective doping $\sigma_{\mathrm{eff}}=3.0\times10^{12}$ cm^{-2}, and half-width at half-maximum linewidths, assuming Lorentzian line shapes, $\Gamma_{12}=41$ meV and $\Gamma_{13}=67$ meV were extracted from the measured intersubband absorption spectrum by assuming a dipole moment $z_{12}=9.7$ Å from theory. The dipole moments $z_{22}-z_{11}=21.1$ Å, $z_{33}-z_{11}=1.57$ Å, and $z_{23}=11$ Å were calculated from theory.[7] With these theoretical and experimental parameters, the $\chi^{(2)}$ of the QW was calculated using a perturbative model by treating the QW as a three-level system assuming Lorentzian line shapes.[11] Both resonant and nonresonant terms were included. The dominant term for double resonance was proportional to $z_{12}z_{23}z_{13}$, while the dominant term for single resonance was proportional to $(z_{11}-z_{22})(z_{12})$.[2] This calculated $\chi^{(2)}$ magnitude is shown in Fig. 3. The double resonance peak of the QW $\chi^{(2)}$ was at a harmonic wavelength of 2.25 μm with a magnitude of 12 nm/V, and the single resonance peak was at 4.1 μm with a magnitude of 20 nm/V.

To characterize the $\chi^{(2)}$ of this QW sample, SHG measurements were performed using a free electron laser (FEL) tuned to wavelengths between 3.71 and 6.66 μm, and a Q-switched CO_2 laser was used for wavelengths between 9.51 and 10.72 μm. The experimental setup was described in detail in previous papers.[9,12] The QW sample was placed in a rotation stage which allowed rotation in angle ϕ about the sample normal [001], where ϕ is the angle between the [100] axis and the intersection of the plane of incidence with the surface of the wafer. The pump beam was then incident on the sample at an angle $\theta=45°$. TM polarization was selected for both the input and collection side.

Since the QWs are grown on a GaAs substrate, the measured SHG signal from the sample is a coherent superposition of the second-harmonic (SH) fields from the QW and bulk. Assuming the same bulk $\chi^{(2)}$ from the epilayer as from

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the GaAs substrate, the normalized SHG signal is given by[13]

$$\frac{P_{2\omega}}{P_{\omega}^{2}}\propto\left|A\chi^{(2)}_{\mathrm{GaAs}}\cos(2\phi)l_c\,\sin(\zeta)e^{i\zeta}+Bl_{\mathrm{MQW}}\chi^{(2)}_{\mathrm{QW}}\right|^{2},\tag{1}$$

where P_{ω} and $P_{2\omega}$ are the fundamental and harmonic powers, l_{MQW} is the thickness of the QW epilayer, l_c is the coherence length in bulk GaAs, and 2ζ is the phase mismatch between the pump and SH fields. A and B are geometry-dependent factors. Since the tensor elements of the susceptibilities from the bulk, $\chi^{(2)}_{\mathrm{GaAs}}$, and the QW, $\chi^{(2)}_{\mathrm{QW}}$, are different, sample rotation and polarization selection can be used to extract the complex $\chi^{(2)}_{\mathrm{QW}}\cdot\chi^{(2)}_{\mathrm{GaAs}}$, as calculated from Miller's rule, is weakly dispersive and real with a value of 0.18 nm/V over the measured wavelength range.[11] The dependence on ϕ of the bulk SH field arises from (xyz) tensor elements. The $\chi^{(2)}_{\mathrm{QW}}$, however, is a (zzz) tensor element, so that the SH field from the QW is independent of ϕ. As a result, the overall SHG power follows a $|\cos(2\phi)+q^*|^2$ dependence, where q^* is a complex constant that is determined by Eq. (1). The $\chi^{(2)}_{\mathrm{QW}}$ is then determined relative to the bulk $\chi^{(2)}_{\mathrm{GaAs}}$ by fitting the measured SHG power versus angle ϕ (ϕ scan) with a free fitting parameter q^*.[9,12]

The SH field from the bulk is governed by the phase mismatch between the pump and SH fields, 2ζ, where

$$\zeta\cong\frac{\pi}{2}\,\frac{L}{l_c\,\cos\,\theta_{\mathrm{int}}}=\nu[\pi],\tag{2}$$

L is the total thickness of the sample, θ_{int} is the refracted angle inside the sample, and ν is the optical thickness in coherence lengths. Note that the coherence length varies from ~22 to ~106 μm for pump wavelengths from 3.71 to 10.72 μm, respectively. Since the measured sample thickness ($L=401\pm1$ μm) is several coherence lengths long, ζ varies rapidly with λ. If ζ is known with sufficient accuracy, the $\chi^{(2)}_{\mathrm{QW}}$ can be extracted from the parameter q^*. The coherence length can be calculated from published values of the refractive indices.[14] However, the uncertainty in the published indices results in insufficiently accurate values of ζ.

To reduce this uncertainty in ζ, we looked for wavelengths at which the bulk contribution to SHG power vanished. At these wavelengths, the optical path length in the sample was exactly an even integer multiple of the coherence lengths so that $\sin(\zeta)=0$. One such zero crossing is shown in Fig. 1. These ϕ scans were taken with the sample oriented at $\theta=45°$ and with FEL pump wavelength of 3.84, 3.85, and 3.86 μm. The 3.84 μm scan had larger peaks at 180° and 360° while the 3.86 μm scan had larger peaks at 90° and 270°. That is, the ϕ scans at 3.84 and 3.86 μm had opposite asymmetry. This change in asymmetry was due to a sign reversal of the bulk contribution to the SHG power, which is proportional to $\sin(\zeta)$, when the wavelength was changed from 3.84 to 3.86 μm. Thus, at a 3.85 μm wavelength, the optical path length through the sample was exactly an even number of coherence lengths long so that N was an integer. Similar nulls in the GaAs contribution were observed at 4.04 and 5.72 μm and are plotted in Fig. 2. From the l_c calculated from Sellmeier refractive index data,[14] Eq. (2) yielded $\nu=8.10$, 7.07, and 3.15 at 3.85, 4.04, and 5.72 μm, respectively. Identifying these nulls as $\nu=8$, 7, and 3, respectively, Eq. (2) yields an effective sample thickness of $L_{\mathrm{eff}}=377.2\pm0.5$ μm. The discrepancy with the actual sample thickness $L=401\pm1$ μm is presumably due to the inaccuracy of the published refractive indices in the 2–50 μm wavelength range, which were measured at 105 K.[14] Equation (2)

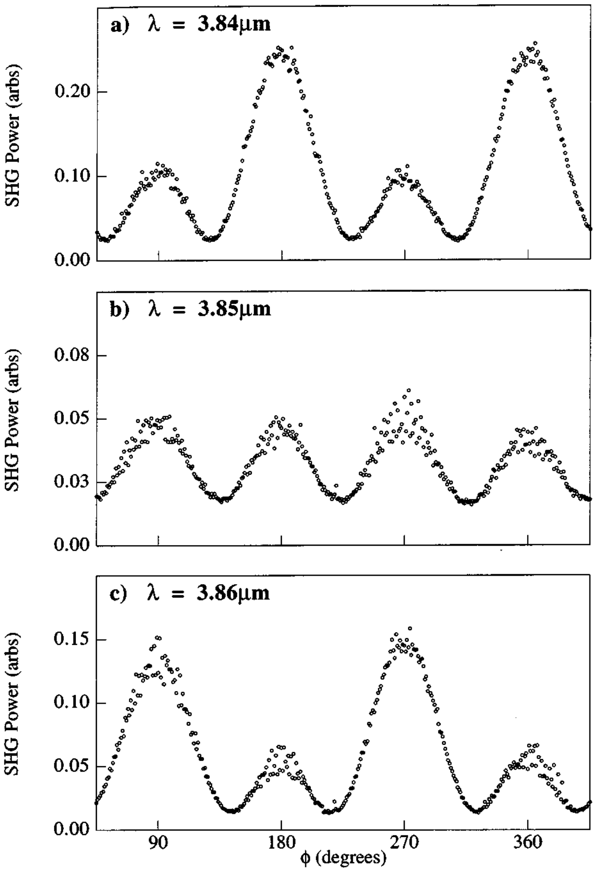

FIG. 1. SHG power vs azimuthal angle ϕ of the sample at pump wavelengths of 3.84 μm (top), 3.85 μm (middle), and 3.86 μm (bottom) showing a null in the GaAs contribution at 3.85 μm.

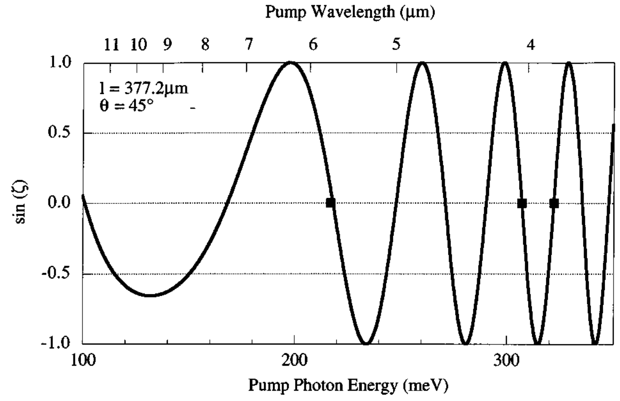

FIG. 2. Calculated bulk contribution $\sin(\zeta)$ curve vs pump photon energy and wavelength for an effective sample thickness of 377.2 μm. Measured nulls shown as squares.

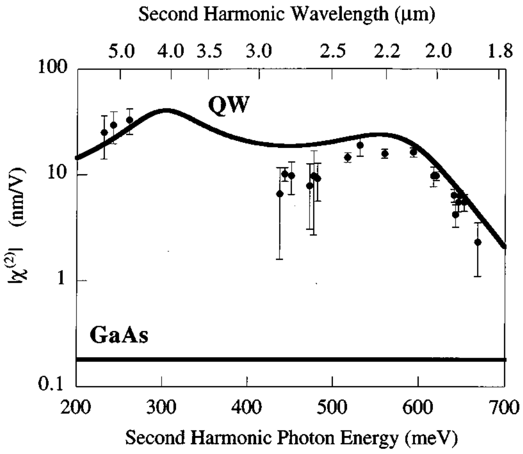

FIG. 3. Theoretical (line) and measured (points) magnitude of the coupled $\chi^{(2)}_{QW}$. Also shown is the magnitude of $\chi^{(2)}_{GaAs}$ for comparison.

with this L_{eff} is used to interpolate ζ between the accurately measured nulls as shown in Fig. 2.

ϕ scans similar to those shown in Fig. 1 were taken over a range of wavelengths with both the FEL and CO_2 lasers. By fitting each of the QW ϕ scans with a complex q^* fitting parameter, the magnitude of the QW $\chi^{(2)}$ was accurately determined. The measured $\chi^{(2)}_{QW}$ is shown in Fig. 3. The doubly resonant $\chi^{(2)}_{QW}$ peak was measured at a SH wavelength of 2.1 μm to be ~16 nm/V, ~90 times that of bulk GaAs. A $\chi^{(2)}_{QW}$ of ~2 nm/V was measured at a SH wavelength of 1.85 μm; this is the shortest wavelength intersubband nonlinearity observed to date in any QW system. The theoretically calculated $\chi^{(2)}_{QW}$ is also shown in Fig. 3, and excellent agreement between experiment and theory is observed. Slight differences between theory and measurement, especially between the two resonances, may have been due to the line shape of the transitions. Since line shapes are not perfect Lorentzians as was assumed in the model, the measured $\chi^{(2)}_{QW}$ should deviate from this theory, especially for large detuning from resonance. Inaccuracies in the measured 1–3 intersubband energy and linewidth, local field effects, and inaccuracies in the theoretical z_{23} and $z_{22}-z_{11}$ may also contribute to the inaccuracy in the theoretical estimate for the $\chi^{(2)}_{QW}$.

In conclusion, we have performed spectroscopically resolved measurements of the nonlinear susceptibility $\chi^{(2)}$ of an intersubband QW across the midinfrared spectrum with SHG pump wavelengths ranging from 10.72 to 3.71 μm. The QW $\chi^{(2)}$ was measured relative to the bulk GaAs substrate contribution. The substrate contribution was accurately determined by measuring the wavelengths at which this contribution was null. A $\chi^{(2)}$ as large as 16 nm/V, 90 times that of GaAs, was measured for SHG of 2.1 μm light. Even at wavelengths as short as 1.85 μm, well detuned from the double resonance, the $\chi^{(2)}$ was still more than 10 times that of GaAs. A perturbative model, with parameters measured by absorption spectroscopy, was found to accurately describe the QW $\chi^{(2)}$ throughout the infrared for SHG and should be useful for predicting the quadratic nonlinear susceptibility for any interactions involving light in the characterized infrared range (10.72–1.85 μm).

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