(65)

LARGE ENERGY INTERSUBBAND TRANSITIONS IN HIGH INDIUM CONTENT InGaAs / AlGaAs QUANTUM WELLS

H. C. Chui, E. L. Martinet, M. M. Fejer, and J. S. Harris, Jr.
Solid State Laboratory
Stanford University
Stanford, CA 94305-4055
U.S.A.

ABSTRACT. Large energy intersubband transitions are necessary for extending intersubband applications to the near-infrared (≤ 2μm wavelength) where compact diode laser based sources are available. By growing high indium content InGaAs / AlGaAs quantum wells (QWs) on GaAs substrates with linearly graded InGaAs buffers, we have demonstrated peak intersubband absorption energies as high as 580meV (2.1μm wavelength). We have also demonstrated intersubband absorption at 580meV in asymmetric coupled InGaAs / AlAs QWs. These are the largest bound-to-bound intersubband transition energies to date. Experimental studies of buffer and QW growth parameters for optimized intersubband absorption have been performed. The well width dependence of intersubband transition energies in both $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ QWs and $In_{0.5}Ga_{0.5}As$ / AlAs QWs have been measured and theoretically modelled.

1. Introduction

Intersubband transitions have been used for applications such as photodetectors, modulators, and nonlinear optics.[1,2,3] These intersubband applications, however, are restricted in wavelength of operation by the range of intersubband transition energies available. Intersubband transition energies in GaAs / AlGaAs QWs and InGaAs / InAlAs QWs lattice-matched to InP have been typically limited to less than 300meV (≥ 4.1μm wavelength), and only recently have intersubband energies of greater than 400meV (≤ 3.1μm) been achieved.[4,5] By using InGaAs and AlGaAs for the well and barrier materials, respectively, QWs can have large conduction band offsets ΔE_c and thus, large intersubband transition energies. However, these InGaAs / AlGaAs QWs are highly strained and are difficult to grow. Lord, et. al.[6] demonstrated that by the initial growth of a linearly graded InGaAs buffer, high quality $In_{0.5}Ga_{0.5}As$ / AlGaAs QWs could be grown on a GaAs substrate by molecular beam epitaxy (MBE). This graded buffer acts as an effective substrate with a lattice constant corresponding to the InGaAs at the top of the buffer. By grading the buffer up to a lattice constant intermediate between the QW barrier and well materials, the wells and barriers can be strain balanced. Using this growth technique, we had previously demonstrated intersubband transitions in InGaAs / AlGaAs QWs with indium compositions of up to 50%.[7] We present here the following: (1) MBE growth studies performed on $In_{0.5}Ga_{0.5}As$ / AlGaAs QWs for optimization of intersubband absorption, (2) measurements of the well width dependence of intersubband absorption in both $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ QWs and $In_{0.5}Ga_{0.5}As$ / AlAs QWs, (3) a theoretical model for predicting the intersubband energies, and (4) results on large energy intersubband transitions in both square and asymmetric coupled QWs.

H. C. Liu et al. (eds.), Quantum Well Intersubband Transition Physics and Devices, 251–259.

2. Sample Description

The layer structure for the QW samples is shown in Figure 1. All of the InGaAs / AlGaAs QW samples were grown by molecular beam epitaxy (MBE) in a Varian Gen-II system on semi-insulating (SI) GaAs substrates. These QWs were grown atop linearly graded InGaAs buffers which were graded at a rate of 16% indium composition per µm from GaAs up to a final buffer indium composition, usually near the average indium concentration of the QWs. In addition, all of the QW samples were uniformly silicon doped n-type over the well regions only. The indium concentrations of the InGaAs wells and buffers were verified to be within 3% indium composition of the target values by high resolution x-ray diffraction (HRXRD) measurements on thick, relaxed InGaAs samples grown with the same conditions and in the same run as the QW samples. The intersubband absorption spectra from these samples were measured using a Fourier transform infrared spectrometer (FTIR) with the samples mounted at Brewster's angle to the linearly polarized light.

3. Growth Optimization

Growth studies of $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ QWs with 40Å wells were performed to minimize the intersubband absorption linewidth and maximize the integrated absorption fraction (IAF).[8] The optimal substrate temperature during the growth of the QWs was found to be between 350°C and 400°C for both As_2 and As_4 species. At higher temperatures, the absorption linewidths were broader, indicative of poor interface quality,[9] while at lower temperatures the IAF and measured Hall sheet charge density dropped, indicative of an increased trap density and a reduced material quality. By adding 1 monolayer GaAs smoothing layers at the QW interfaces and ramping the substrate temperature to 600°C for growth of the AlGaAs barriers, we observed that the linewidths and IAFs were significantly improved. However, the added temperature ramps and growth interrupts greatly increased the growth time.

The final indium composition y_b of the $In_yGa_{1-y}As$ buffer was also determined to be a critical growth parameter; FTIR results from samples with varying y_b are shown in Figure 2. The linewidth monotonically decreased with increasing y_b up to $y_b = 0.3$. For samples grown with $y_b = 0.4$ and 0.5, the wafer had an extremely rough surface and was optically very lossy, and the intersubband absorption was weak, indicative of relaxed material. Thus, the optimal effective substrate lattice constant corresponded to that of an $In_{0.3}Ga_{0.7}As$ buffer; if a GaAs substrate or an InP substrate (corresponding to an $In_{0.53}Ga_{0.47}As$ buffer) was used without a graded InGaAs buffer, significantly poorer QW material quality would result. This optimal $y_b = 0.3$ is larger than the average indium concentration of the multi-quantum well, $y_{avg} = 0.17$, but lower than the indium composition of the InGaAs well, $y_{well} = 0.5$. That is, the optimum buffer composition appears to minimize the strain in the well without exceeding a critical thickness due to strain for an individual barrier layer or due to cumulative strain from the MQW structure. With different QW geometries, the optimal y_b is likely to be different; however, optimal material quality should have y_b higher than y_{avg} but lower than y_{well}. For instance, if a thinner barrier was used, a higher y_{avg} would result, and a $y_b > 0.3$ would likely be optimal.[6]

Using near optimized growth conditions (As_2, $y_b = 0.3$, substrate temperature = 400°C), we were able to obtain full-width at half-maximum (FWHM) linewidths of 34meV for 1 to 2 transition energies of 350meV. This linewidth corresponds to a relative linewidth (FWHM divided by the transition energy) of less than 10%; similar relative linewidths were observed in well doped GaAs / AlGaAs QWs. This FWHM is also comparable to that obtained in InGaAs / InAlAs QWs grown on InP with similar transition energies (29meV for unstrained and 60meV for strained).[10]

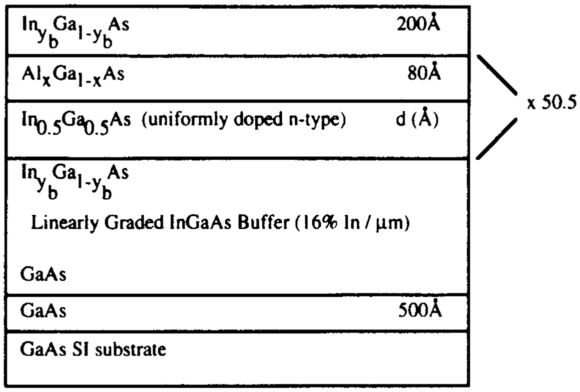

Figure 1. Layer diagram of quantum well structures.

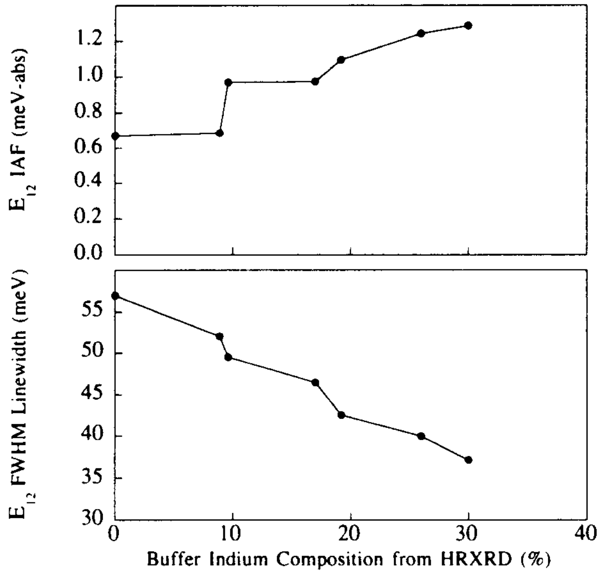

Figure 2. Dependence of intersubband absorption linewidth and IAF on final buffer indium composition y_b as determined by HRXRD. For samples with y_b = 0.4 or 0.5, the QWs were relaxed, and the intersubband absorption was extremely weak.

4. Well Width Dependence of Intersubband Absorption

The dependence of the intersubband absorption on well width for both $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ and $In_{0.5}Ga_{0.5}As$ / AlAs QWs was also determined. These samples each had 50 QWs and were doped at a sheet charge density per QW of $2.8x10^{12}$ cm^{-2} and $4.0x10^{12}$ cm^{-2} for the $Al_{0.45}Ga_{0.55}As$ and AlAs barrier samples, respectively. The samples were grown at a substrate temperature of 400°C with As_4 for the $Al_{0.45}Ga_{0.55}As$ samples and with As_2 for the AlAs samples. The final buffer indium composition was targeted to be near the average indium concentration of the QWs for the AlGaAs samples and slightly higher than the average for the AlAs samples. As determined by our growth parameter studies, using a final buffer indium composition slightly greater than the average lattice constant of the quantum wells should help improve the QW material quality. The intersubband absorption peak energies for these QWs are plotted in Figure 3. The 1 to 2 transition energies for the narrow AlAs QWs are among the largest observed to date for intersubband transitions. For the $Al_{0.45}Ga_{0.55}As$ barrier samples, the 23Å well had a 1 to 2 transition energy of 384meV, lower than that for the 30Å well. This indicates that the second energy level was near the top of the well so that narrowing the well width from 30Å to 23Å caused the ground state subband energy to increase more rapidly than the second subband energy.

The FWHM linewidths and IAFs for the $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ and $In_{0.5}Ga_{0.5}As$ / AlAs QWs samples are shown in Figure 4. The measured IAFs are approximately half of the theoretically calculated values (model is described in Section 5) for all of the samples except the narrowest well samples for both series for which the IAF dropped significantly below 50% of the theoretical value. The IAFs are calculated by using dipole moments obtained from our theoretical model and by using the targeted dopant concentration for the carrier concentration.[11] We have observed this 50% experimental to theoretical IAF ratio, even for GaAs / AlGaAs QW samples. The reduced IAFs in the narrowest wells may be due in part to the high dopant concentrations in these narrow wells required to achieve the same sheet charge density as the wider wells. The linewidths follow a generally increasing trend with narrower well width. Many linewidth broadening mechanisms contribute to this well width dependent linewidth, including increased impurity and interface scattering with decreased well widths and increased transition energies. The broad linewidth for the narrowest well may be due to Γ-X scattering in the AlAs barrier, since according to our model, the second energy level is above the AlAs X-valley.

5. Theoretical Modelling

The transition energies and dipole moments for these quantum wells were modelled with a single band effective mass model, and band nonparabolicity was taken into account by using an energy dependent effective mass m*(E) given by[12,13]

$$m^*(E) = \frac{m_0^*}{1 + \frac{\alpha}{E_g}\left(E - E_{c0}\right)} \tag{1}$$

where E is the electron energy, E_{c0} is the conduction band edge energy, α is the nonparabolicity factor,[14] and m_0^* is the effective mass at the bottom of the conduction band.[15] For energies less than 400meV above the conduction band edge, the nonparabolic band parameterization given by Equation 1 is used. However, for energies of more than 400meV above the band edge, the fourth order approximation to the band dispersion (energy E versus wavenumber k) becomes inaccurate for $In_{0.5}Ga_{0.5}As$ and in fact, approaches a maximum in E vs. k so that the effective mass

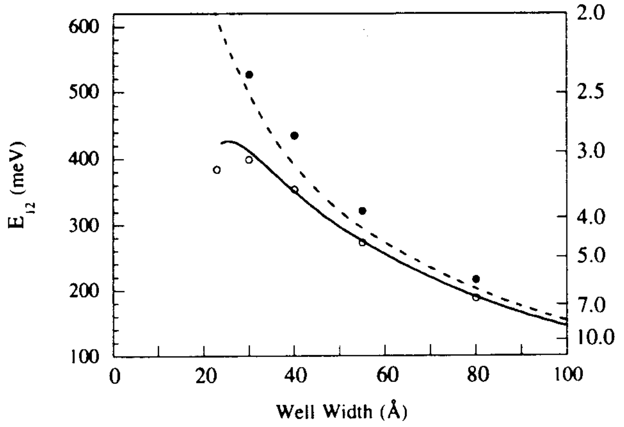

Figure 3. Well width dependence of 1 to 2 intersubband absorption peak energies for $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ (hollow circles) and $In_{0.5}Ga_{0.5}As$ / AlAs QWs (filled circles). Solid and dashed lines are calculated energies corresponding to the $Al_{0.45}Ga_{0.55}As$ and AlAs QW samples, respectively.

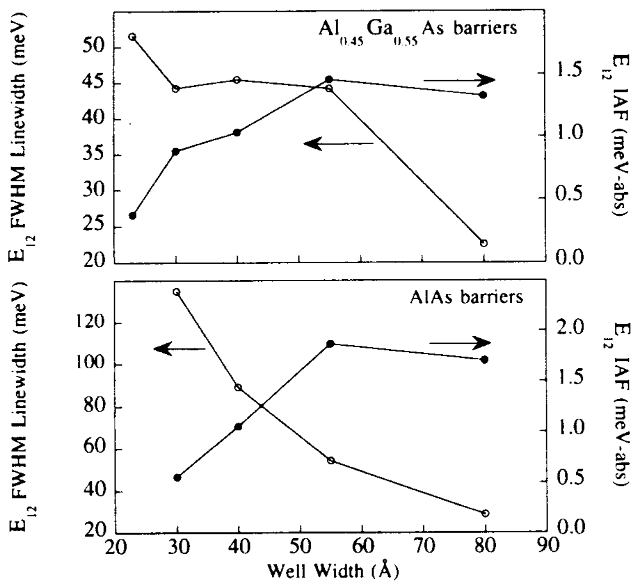

Figure. 4. Well width dependence of 1 to 2 intersubband absorption FWHM linewidth (hollow circles) and IAF (filled circles) for $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ (top graph) and $In_{0.5}Ga_{0.5}As$ / AlAs QWs (bottom graph).

becomes infinite. In order to resolve this problem, we noted that the E vs. k curves for InAs and GaAs, calculated using a pseudopotential method,[16] are nearly linear for energies high above the band edge around the Γ-point. Thus, we assumed a linearized E vs. k for energies of more than 400meV above the conduction band edge with the E vs. k curve and slope matched at 400meV. The calculated energies using this method for both the $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ and $In_{0.5}Ga_{0.5}As$ / AlAs QWs are also plotted in Figure 3. Good agreement between the calculated and measured energies is observed even for energies of more than 750meV above the conduction band edge. This energy dependent effective mass formulation may be a good guide for intersubband and hot electron transistor modelling where electron energies are extremely high above the conduction band edge.

6. Large Energy Intersubband Absorption Results

To obtain even larger transition energies, we have investigated samples with higher indium concentrations. Three $In_{0.6}Ga_{0.4}As$ / $Al_xGa_{1-x}As$ QW samples with 50 QWs and aluminum compositions of x = 0.45, 0.67, and 1.0 were grown with a final buffer indium composition of 0.3 using As_2 at a substrate temperature of 375°C. The samples were uniformly Si doped in the well regions at 5.7×10^{12} cm^{-2}/QW. The intersubband absorption spectra for these three samples are shown in Figure 5. The AlAs barrier sample has a peak transition energy of 580meV (2.1μm wavelength). To our knowledge, this is the largest bound-to-bound intersubband transition energy reported to date. The relative linewidths for these samples are 14%, 17%, and 25% for the x = 0.45, 0.67, and 1.0 samples, respectively.

Asymmetric coupled double $In_{0.6}Ga_{0.4}As$ / $Al_xGa_{1-x}As$ QWs with large transition energies were also grown for future nonlinear optics experiments.[17] The double QW structure is shown in the insert of Figure 6. 200 double QWs were grown with Si doping in the well regions at a sheet charge density of 3.0×10^{12} cm^{-2} per double QW. The QWs were grown atop a graded InGaAs buffer with a final buffer indium composition of 0.3 using As_4 at a substrate temperature of 375°C. The intersubband absorption spectrum for the QWs is shown in Figure 6. Two absorption peaks are seen at 295meV (4.2μm wavelength) and 580meV (2.1μm) with relative linewidths of 24% and 18% corresponding to the E_{12} and E_{13} transitions, respectively. This E_{13} = 580meV is the largest 1 to 3 intersubband transition to date. This coupled QW sample should exhibit a strong second order nonlinear susceptibility for 4μm to 2μm wavelength second harmonic generation since it is doubly resonant.

7. Conclusions

We have demonstrated intersubband transitions in high indium content InGaAs / AlGaAs QWs grown on GaAs using a linearly graded InGaAs buffer. Growth studies were performed to optimize growth conditions for minimum intersubband absorption linewidth with maximum IAF. The well width dependence of intersubband absorption for both $In_{0.5}Ga_{0.5}As$ / $Al_{0.45}Ga_{0.55}As$ and $In_{0.5}Ga_{0.5}As$ / AlAs QWs was determined, and good agreement of the intersubband energies to a simple model was obtained. A peak 1 to 2 intersubband transition energy as high as 580meV (2.1μm wavelength) was observed in $In_{0.6}Ga_{0.4}As$ / AlAs QWs. A E_{13} also at 580meV (2.1μm) was observed in asymmetric coupled $In_{0.6}Ga_{0.4}As$ / AlAs QWs. These are the largest bound-to-bound intersubband transition energies to date. The large transition energies observed in these InGaAs / AlGaAs QWs should be useful for short wavelength intersubband applications in the near-infrared (≤ 2μm wavelength) where compact diode laser based sources are available.

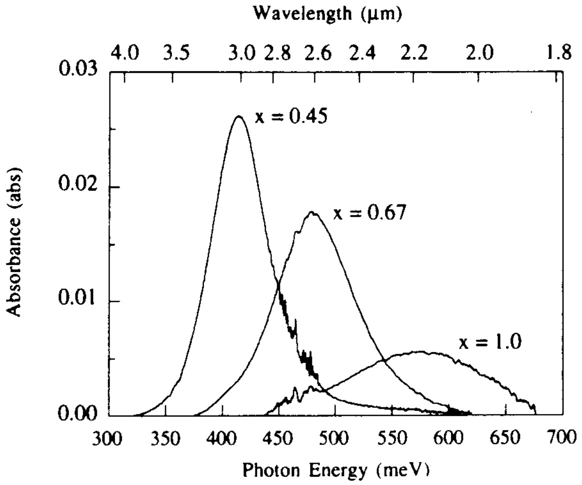

Figure. 5. Intersubband absorption spectra for $In_{0.6}Ga_{0.4}As$ / $Al_xGa_{1-x}As$ QWs with 30Å wells for x = 0.45, 0.67, and 1.0.

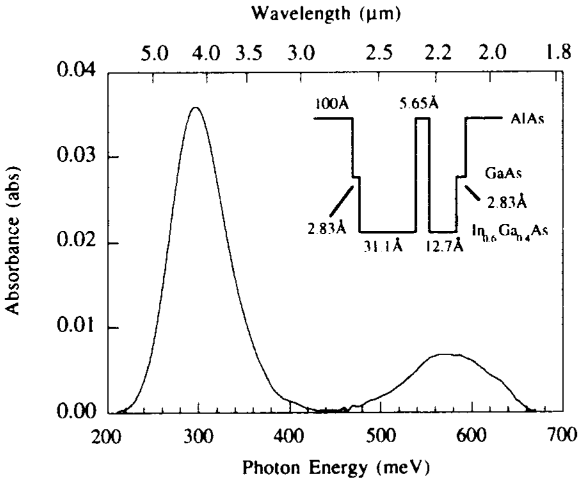

Figure. 6. Intersubband absorption spectra for doubly resonant asymmetric coupled $In_{0.6}Ga_{0.4}As$ / AlAs QWs with conduction band diagram (insert).

Acknowledgements

H. C. Chui acknowledges fellowship support from the Office of Naval Research (ONR), and E. L. Martinet, from ONR and Lockheed. This work was supported by ONR under contract numbers N00014-91-J-0170 and N00014-92-J-1903 and by ARPA under contract number N00014-90-J-4056. FTIR measurements were performed on a Bruker FTIR at the Stanford Free Electron Laser facility.

References

1. *Intersubband Transitions in Quantum Wells*, edited by E. Rosencher, B. Vinter, and B. Levine (Plenum, New York, 1992).
2. C. G. Bethea, B. F. Levine, V. O. Shen, R. R. Abbott, and S. J. Hseih, `10-μm GaAs/AlGaAs multiquantum well scanned array infrared imaging camera', *IEEE Electron. Devices* **38**, 1118 (1991).
3. M. M. Fejer, S. J. B. Yoo, R. L. Byer, A. Harwit, and J. S. Harris, Jr., `Observation of extremely large quadratic susceptibility at 9.6-10.8 μm in electric-field-biased AlGaAs quantum wells', *Phys. Rev. Lett.* **62**, 1041 (1989).
4. L. H. Peng, J. H. Smet, T. P. E. Broekaert, C. G. Fonstad, `Transverse electric and transverse magnetic polarization active intersubband transitions in narrow InGaAs quantum wells', *Appl. Phys. Lett.* **61**, 2078 (1992).
5. H. Asai and Y. Kawamura, `2.4μm intersubband absorption in $In_{1-x}Ga_xAs/AlAs_{1-y}Sb_y$ multiple quantum wells', *Fourth International Conference on InP and Related Compounds* (1992).
6. S. M. Lord, B. Pezeshki, and J. S. Harris, Jr., `Investigation of high In content InGaAs quantum wells grown on GaAs by molecular beam epitaxy', *Electron. Lett.* **28**, 1193 (1992).
7. H. C. Chui, S. M. Lord, E. Martinet, M. M. Fejer, and J. S. Harris, Jr., `Intersubband transitions in high indium content $In_{0.5}Ga_{0.5}As/AlGaAs$ quantum wells', *Appl. Phys. Lett.* **63**, 364 (1993).
8. More details of these growth studies will be presented at the 1993 North American Conference on MBE with proceedings submitted as H. C. Chui and J. S. Harris, Jr., `Growth studies of InGaAs/AlGaAs quantum wells grown on GaAs with a linearly graded InGaAs buffer', *J. Vac. Sci. Technol. B*.
9. M. J. Ekenstedt, S. M. Wang, and T. G. Andersson, `Temperature-dependent critical layer thickness for $In_{0.36}Ga_{0.64}As/GaAs$ single quantum wells', *Appl. Phys. Lett.* **58**, 854 (1991).
10. H. Asai and Y. Kawamura, `Intersubband absorption in highly strained InGaAs/InAlAs multiquantum wells', *Appl. Phys. Lett.* **56**, 1149 (1990).
11. L. C. West and S. J. Eglash, `First observation of an extremely large dipole infrared transition within the conduction band of a GaAs quantum well', *Appl. Phys. Lett.* **46**, 1156 (1985).
12. G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (les éditions de physique, France, 1988), Chapt. 2.
13. S. J. B. Yoo, *Linear and Nonlinear Spectroscopy of Quantum Well Intersubband Transitions*, Stanford Univ. Thesis, 1991.
14. J. S. Blakemore, `Semiconducting and other major properties of gallium arsenide', *J. Appl. Phys.* **53**, R123 (1982).

15. The material parameters used for this calculation were a Γ-point band gap energy E_g of 0.755eV, 1.424eV, 1.985eV, and 3.018eV for $In_{0.5}Ga_{0.5}As$, GaAs, $Al_{0.45}Ga_{0.55}As$ and AlAs, a conduction band offset ratio $\Delta E_c/E_g$ of 0.5 for $In_{0.5}Ga_{0.5}As$ to GaAs and 0.6 for GaAs to $Al_xGa_{1-x}As$, a m_0* of 0.034 m_0, 0.098 m_0, and 0.141 m_0 for $In_{0.5}Ga_{0.5}As$, $Al_{0.45}Ga_{0.55}As$ and AlAs, respectively, and an $\alpha = -0.86$ for $In_{0.5}Ga_{0.5}As$.

16. J. R. Chelikowsky and M. L. Cohen, `Nonlocal pseudopotential calculations for the electronic structure of eleven diamond and zinc-blende semiconductors', *Phys. Rev. B* **14**, 556 (1976).

17. Initial results are presented here at the 1993 *NATO Advanced Research Workshop on Intersubband Transition Physics and Devices* by E. L. Martinet, B. J. Vartanian, G. L. Woods, H. C. Chui, J. S. Harris, Jr., M. M. Fejer, B. I. Richman, and C. A. Rella , `Applications of high indium content InGaAs/AlGaAs quantum wells in the 2-7μm regime'.