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RAMAN SCATTERING IN MBE GaInAs-InP QUANTUM WELLS

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Abstract
The phonon modes in GaInAs-InP heterostructures were studied by means of Raman scattering. The mode frequencies were found to occur at the bulk values and to be well-defined even for layers of 25Å. The phonon intensity ratios were found to depend very strongly on the incident laser power and we attribute this to screening of the modes by photoexcited carriers.

Introduction
Raman scattering studies have been made of two MBE-grown Ga0.47In0.53As-InP quantum well structures: a single and a multiple quantum well. These structures are relevant for optoelectronic device applications and are therefore of current interest. Triple crystal x-ray work on the single quantum well is reported elsewhere [1] and yielded a value of 230Å for the well thickness and 680Å for the cap layer thickness (the nominal figures given by the growth time were 160Å and 500Å respectively). The multiple quantum well consisted of 50 periods of 25Å well and 100Å barrier with a 1000Å cap layer (nominal values). We studied the samples in standard backscattering geometry using the 4880Å (2.54eV) line of an Argon ion laser and the 4825Å (2.57eV) line of a Krypton ion laser. These lines lie close to the \(E_g\) (2.62eV) resonance of the GaInAs and thus allow an enhanced signal from the buried layer. They also provide a reasonable penetration of the sample estimated at about 700Å in InP and about 200Å in GaInAs [2].

The scattered light was analysed by a 1m double spectrometer and detected by a cooled photomultiplier tube operating in photon counting mode. The etch used to remove the InP was 3:1 \(\text{H}_2\text{SO}_4:\text{HCl}\) and that for the GaInAs was 80% 3:1 \(\text{H}_2\text{SO}_4:\text{H}_2\text{O}\) and 20% \(\text{H}_2\text{O}_2\).

There is still uncertainty about the nature of the phonon modes in GaInAs. Kakimoto and Katoda [3] showed clear evidence for two-mode behaviour across the composition range of the alloy and this is endorsed by Landa et al [4]. Pearsall et al [5] demonstrated a “mixed-mode” behaviour, however, with one clear GaAs-like LO mode, one clear InAs-like TO mode and two indeterminate modes. In our bulk GaInAs sample we observe two strong modes (see Fig. 1a) and we identify the dominant mode as the GaAs-like LO phonon at 265\(cm^{-1}\) and the second mode as the InAs-like LO phonon at 232\(cm^{-1}\), after Kakimoto and Katoda [3]. Both peaks obey the LO selection rule for backscattering: i.e. allowed in the \(z(x,y)z\) configuration and forbidden in the \(z(x,x)z\) configuration. Although in both configurations scattering from TO phonons is forbidden, it is usually observed in the Raman spectra of most III-V alloys.

Results and Discussion
The Raman spectrum of the single quantum well was markedly different from that of the bulk; both LO phonon modes had been quenched to the extent that they had become indistinguishable from the forbidden TO modes (see Fig. 1b). This quench was a consistent feature of the spectra from the single quantum well and persisted when we varied the frequency of the exciting light. Initially, the quench was thought to be due to a mechanism similar to that found in the magnetophotonon experiments of Portal et al [6], where the authors found that as the electron confinement increased, so the electrons penetrated further into the barrier layers and interacted with the InP LO phonons rather than the GaAs-like LO phonons. This
mechanism does not explain the total quench of the \( L_0 \) modes, nor does it explain why this should occur for such a large well width. In order to investigate the influence of the InP barriers, we selectively etched off the InP cap layer. The spectrum of the etched sample is shown in Fig. 1c. After removal of the cap, the phonon modes of the alloy layer resembled those of the bulk material with both the \( L_0 \) modes occurring at the bulk frequencies. The complete removal of the quench suggested that the barriers did not play the major role in causing the quench and hence an explanation other than penetration of the barriers must be sought.

The effect of the incident laser power on the intensity of the GaAs-like \( L_0 \) phonon mode is shown in Fig. 2a. This suggested that the laser powers used in the experiment might be sufficiently high so as to create a photoexcited electron hole plasma (EHP) within the well which would screen the \( L_0 \) phonons. This was confirmed by monitoring both the half width at half maximum and the integrated intensity of the photoluminescence (PL) emission from the well as a function of incident laser power (see Figs. 2b and 2c). For the Raman intensity and the PL half width, the onset of a saturation behaviour occurred with a power of between 50 and 80mW incident on the sample. The Raman spectra from the single quantum well were recorded within this saturation regime, thus strongly suggesting a screening effect of the \( L_0 \) mode. This saturation is not evident in the plot of the PL integrated intensity as a function of incident power and we conclude that the carrier density within the excited volume increases to a saturation value after which the plasma expands so as to keep a constant density.

In order to make a simple estimate of the saturation density of the photoexcited EHP, we used the assumptions that each absorbed photon creates one electron-hole pair, that these decay to the bottom of the conduction band within 10 picoseconds and then remain there for 10 nanoseconds before recombining. This yields a value of \( 2.4 \times 10^{10} \) electron-hole pairs/Watt. From the absorption lengths in GaInAs and InP [2] we were able to determine where the carriers would be created. For the 57mWatts of our experiment, this yields a density of \( 1.9 \times 10^{17} \) cm\(^{-3}\) in the InP cap layer and a 2D density of \( 1.3 \times 10^{12} \) cm\(^{-2}\) in the well. This density is certainly of the correct order of magnitude to populate the well and screen the phonons [8], and about ten times that determined by Schubnikov–de Haas measurements.

![Raman scattering spectra](image-url)
The effect on the surface depletion width of such an electron concentration was estimated using the expressions given by Schubert et al. [9]. Making an order of magnitude assumption about the surface potential (1 eV), the depletion width was calculated to be about 500Å in the InP cap layer. This puts the quantum well beyond the surface depletion width thus suggesting that it could be sufficiently populated to screen the LO phonon scattering.

Screening of the LO phonon by free carriers in bulk semiconductors is a well-understood phenomenon [see, eg., 10]; with increased carrier density, the plasma frequency increases, first approaching and then becoming greater than the phonon frequency. This results in an anticrossing behaviour and yields the standard $w^+$ and $w^-$ modes. In two dimensions (2D), this effect is different. The reduced dimensionality yields slab-like plasma oscillations of the 2D electron gas (2DEG) in the plane of the layer. In order to couple to these, one must have a component of scattering wavevector, $q$, parallel to the 2D layer [11]. This is not the case for the backscattering geometry of our experiment. Since the electron energy levels perpendicular to the plane of the layer are quantised, we would expect to couple to intersubband excitations which have the parallel component of $q$ approximately equal to zero. Previous experimental studies [12, 13], have investigated this effect in GaAs-AlGaAs heterostructures, where it was demonstrated that the collective intersubband excitations coupled strongly to the LO phonon modes. We suggest that it is this coupling that causes the quench of the LO modes in our Raman spectra.

Our experimental parameters are inappropriate to observe this electronic scattering since our experiments were all done at ambient temperature. Liquid helium temperatures are needed to ensure that the quantised well energy levels are free from thermal broadening which tends to smear out any structure. This constraint would also apply to the 2D plasma oscillations [11]. Low temperature work is in progress.

In the multiple quantum well sample, the thick cap layer of InP (1000Å) was removed together with the first well leaving 100Å of InP on the sample surface. The spectrum observed is shown in fig 3a. The InP barrier was subsequently etched off and the spectrum observed is shown in fig 3b. It is clear that these spectra do not significantly differ in either mode frequency or intensity ratio. Moreover, the phonon modes of the alloy layers have the same frequencies as those in the bulk; there is no shift of the modes towards the spectrum of the quaternary alloy, GaInAsP, thus suggesting that the layers are well-defined and the interfaces are sharp. There is also no evidence of a GaAs-like LO mode quench which is further evidence that the InP barrier layers do not play a significant role in this process.

It is worth noting that a 2cm$^{-1}$ shift to lower frequencies of the InP LO phonon mode is observed.
Fig. 3
Raman scattering spectra of a Ga$_{0.47}$In$_{0.53}$As-InP multiple quantum well sample with 50 periods of 25Å well and 100Å barrier. (a) 100Å InP barrier on surface. (b) 25Å GaInAs well on surface. These spectra do not significantly differ in either mode frequency or intensity ratio.

in scattering from a cap or barrier layer with respect to a buffer layer or bulk sample. We have looked at various possible explanations for this shift including strain effects induced by variations in alloy composition, As contamination of cap and barrier layers and the formation of GaInAsP interface layers. Each of these possible mechanisms would, however, also give rise to other, much stronger, effects than the shift of the InP LO mode and we do not observe any of these other effects. The origin of the shift of the InP LO mode is not yet clear and is under further investigation.

Conclusions
We have observed well-defined phonon frequencies from a GaInAs single and multiple quantum well and these occur at the bulk GaInAs values. There is no evidence of graded interface regions even in 25Å layers. The phonon intensity ratios strongly depend on the free carrier concentration with high laser powers generating a sufficiently dense EHP to screen the dominant LO phonon mode in the well.

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