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SPRING COLLEGE IN CONDENSED MATTER ON: PHYSICS OF LOW-DIMENSIONAL SEMICONDUCTOR STRUCTURES

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INTERFACES, QUANTUM WELLS AND SUPERLATTICES

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MOLECULAR BEAM EPITAXY

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ABSTRACT

Molecular beam epitaxy (MBE) is a sophisticated method of film growth capable of providing the device engineer with any desired structure. This flexibility has emerged from a thorough understanding of the fundamental factors controlling growth and dopant incorporation obtained using modulated molecular beam spectroscopy, reflection high energy electron diffraction and Monte-Carlo studies. In this article I will concentrate on the growth and interface properties of heterojunctions, multi-quantum well and superlattice structures and their use in devices such as high electron mobility transistors and lasers.

1. INTRODUCTION

The technique which has become known as molecular beam epitaxy (MBE) is, at its simplest, a refined form of vacuum evaporation. The molecular beams are produced by evaporation or sublimation from heated liquids or solids usually contained in pyrolytic boron nitride crucibles. The flux produced is thus determined by the vapour pressure of the element or compound at elevated temperatures. At the pressures used in the MBE equipment collision-free beams from the various sources interact chemically on the substrate to give an epitaxially related film. Ultra-high vacuum (UHV) techniques are used to reduce the pressure of gases from the ambient background and thus improve the purity of the layers. More recently gas sources mounted outside the equipment have been employed in what has become known variously as gas source MBE (GS-MBE), chemical beam epitaxy (CBE) or metalorganic MBE (MOMBE).

MBE began as a basic study of the chemical reactions occurring on surfaces during the growth of III-V compounds but quickly evolved into a practical method for the growth of high purity materials. The ability to start and stop the molecular beams in less than the time taken to grow a single atomic or molecular layer has led to the ability to produce complex multilayer structures. The use of UHV technology has enabled the physical and chemical properties of the films to be measured in-situ using reflection high energy electron diffraction (RHEED) and Auger electron spectroscopy (AES) studies. Modulated molecular beam spectrometry (MMBS) was developed to study the chemical processes involved and the dynamics of film growth have been investigated using RHEED. This led to the discovery of the so-called RHEED oscillation technique which can measure the growth rate in-situ, a unique feature of MBE.

In this article I will review the fundamental aspects of MBE using mostly examples from the best understood system AlGaAs. I will also discuss the techniques required to grow high purity samples such as multi-quantum-well (MQW) structures, superlattices (SLs) and high mobility two-dimensional electron gas structures (2DEGs). I will finally illustrate the practical uses of MBE for the preparation of two types of devices based on the new physical principles encountered in two-dimensional systems, namely the high electron mobility transistor (HEMT) and the short wavelength MQW or SL laser. Particular emphasis will be given in this paper to the nature of interfaces since they form the basis of the quantum well and superlattice structures.

It is appropriate to point out that the techniques developed for III-V compounds have also been applied to a whole range of other materials including elemental semiconductors[1], II-VI semiconductors, insulators and metals but these are outside the scope of the present article. Several excellent reviews have been published which deal in more depth with particular aspects of MBE for III-V compounds[2-4] and its application for the growth of superlattices[5], polar on non-polar materials[6], the concept and applications of delta doping[7], and finally devices based on GaAs and AlGaAs[8-10].

2. FUNDAMENTAL ASPECTS OF MBE

Several types of study have contributed to our present understanding of the processes controlling the growth of films and dopant incorporation in MBE: surface chemical processes were investigated using MMBS methods[11], the dynamics of film growth have been examined using the RHEED technique[12] and Monte-Carlo (M-C) simulations of growth have added to our knowledge of the factors influencing growth and interface roughness[13]. In addition thermodynamic calculations have shown fundamental limitations involved in using certain dopants and the factors governing the incorporation of unwanted impurities[14].

2.1 Growth of Binary Compounds

Modulated molecular beam studies: In conventional MBE group III elements such as Al, Ga and In are always supplied as the monomer by evaporation from the liquid. Over most of the temperature range used in MBE the group III elements have a unity sticking coefficient and therefore the growth rate and alloy composition are simply determined by relative supply rates to the surface. At high temperatures however, as Arthur first showed for Ga[15], the group III element with the higher vapour pressure will have a finite lifetime on the surface leading to a reduction in growth rate and a change in alloy composition (this point will be discussed in more detail below in relation to the growth of alloy films).

In conventional MBE As can be supplied either as the tetramer As_4 by sublimation from the solid or as the dimer As_2, by evaporating from GaAs. As_2 can also be obtained by dissociation of As_4 using a two-zone "cracker" furnace with the front end operating at about 900°C.

The first studies by Arthur[15-16] and the later work by Foxon and Joyce [17] show that As_2 is dissociatively chemisorbed on single Ga adatoms as seen in Fig. 1. The sticking coefficient of As is therefore proportional to the Ga supply reaching the surface and any excess As is lost by re-evaporation. It follows that stoichiometric GaAs is always obtained provided an excess As flux is supplied. At low temperatures, As_2 molecules can associate on the surface to form As_4 which is subsequently desorbed. This is the only direct evidence for the surface migration of As species.

For most layers grown by MBE, As_4, from elemental As has been used to avoid impurities associated with dopants in GaAs or contamination from the hot zone of the cracker. The chemical reactions involved with the tetramer are more complex, as shown in Fig 2. Pairs of As_4 molecules interact on adjacent Ga sites with the excess As atoms being desorbed[18]. The main experimental observations that led to this model are that the maximum sticking coefficient of As_4 is 0.5 even when a large excess of Ga is supplied to the surface and in addition the second order dependence of the As_4 desorption rate on the adsorption rate at low As coverages.

The behaviour with As_4 is in complete contrast to that of As_2 where a unity sticking coefficient can be achieved on a Ga rich surface. The lifetime of the tetramer is also much greater than the dimer on the GaAs surface. This in turn implies that there is a much higher population density in the mobile precursor state for equivalent fluxes of As. The different behaviour of the two As species may be expected to influence the properties of films grown under otherwise identical conditions[19].

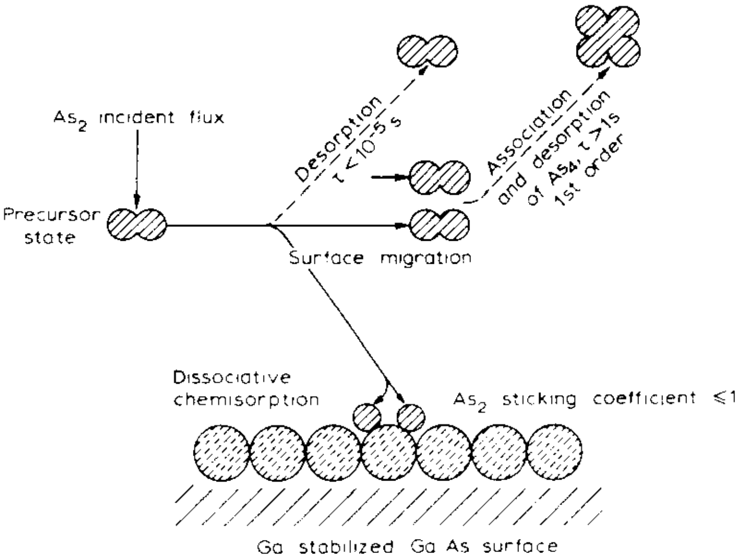

Fig. 1. The model proposed for the growth of GaAs from Ga and As_2.

This has been confirmed by a number of observations; studies of the electronic structure of the GaAs surfaces grown with As_2 and As_4[20], the lower concentration of deep levels in films grown with the dimer[21] and the improved minority carrier properties of AlGaAs-GaAs double heterostructures obtained with As_2[22]. In this last study both the GaAs and the AlGaAs-GaAs interfaces were improved when the dimer was used. At high substrate temperatures it is possible that As_4 may be decomposed to form As_2 on the surface but there is no direct evidence for this suggestion. The fact that in a two-zone cracker furnace a temperature of 900°C is required for complete dissociation suggests that this will not occur on the GaAs surface at 700°C unless there is some catalytic process involving Ga taking place. In general, films grown with the dimer are better than those grown with the tetramer especially at low substrate temperatures.

<u>Reflection high energy electron diffraction studies</u>: RHEED studies during growth by MBE showed a variety of surface structures depending upon the temperature and relative fluxes of Ga and As used[23]. Subsequent work established that the crystal surface during growth is disordered in a complex manner[24-26]. The study of the dynamics of film growth resulted from the observation of oscillations in the intensity of electrons specularly reflected from the surface with a period corresponding to the time taken to deposit a single monolayer of material, a complete layer of Ga+As atoms[27]. The experimental arrangement used for such studies is shown in Fig. 3 and typical examples for GaAs and AlAs are shown in Fig. 4. The damped oscillations in the intensity of various features of the GaAs pattern can be observed over a wide range of temperatures (500-750°C) and Ga fluxes[28]. On stopping growth the intensities of the various features return gradually to their original values in a time determined mainly by the substrate temperature.

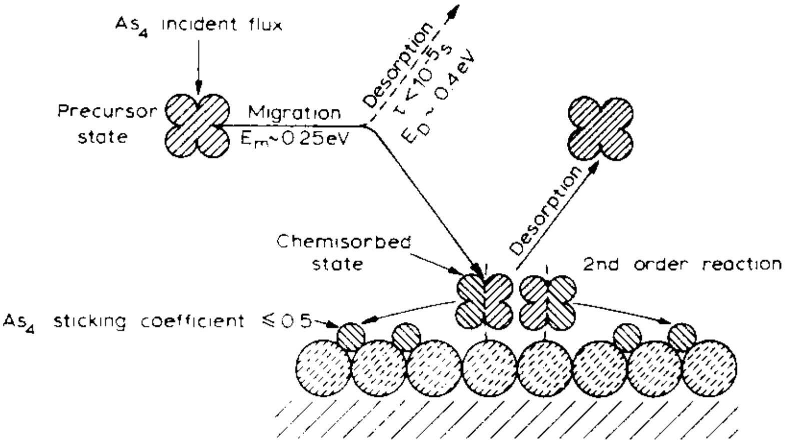

Fig. 2. The model proposed for the growth of GaAs from Ga and As_4.

This observation has been explained using a simple model based on a single scattering optical analogue where changes in specular beam intensity are equated with corresponding variations in surface roughness on an atomic scale. This picture is qualitatively correct since the wavelength of the elecrons (0.1Å) is small compared to the step height of 2.83Å. In this interpretation the damping is thought to arise from growth of the second monolayer starting before the first monolayer is completed. The recovery after growth stops suggests that the step density on the surface can be reduced i.e. the surface is smoothing[28-30]. For this process to occur it is essential for Ga to diffuse to the existing step edges and the growth of GaAs from predeposited Ga on supplying an As_2 flux to the surface suggests that this may be the case[30]. Direct evidence for the Ga being mobile on the surface came from studies of growth on vicinal samples[31], as shown in Fig. 5. At low temperatures, where the Ga diffusion length is smaller than the distance between steps on the off-cut surface, conventional RHEED oscillations are observed. At high temperatures, however, the diffusion length is long enough for step edge growth to dominate and the oscillations disappear. From such data the activation energy for the diffusion of Ga on the surface has been deduced and the figures for other group III elements estimated.

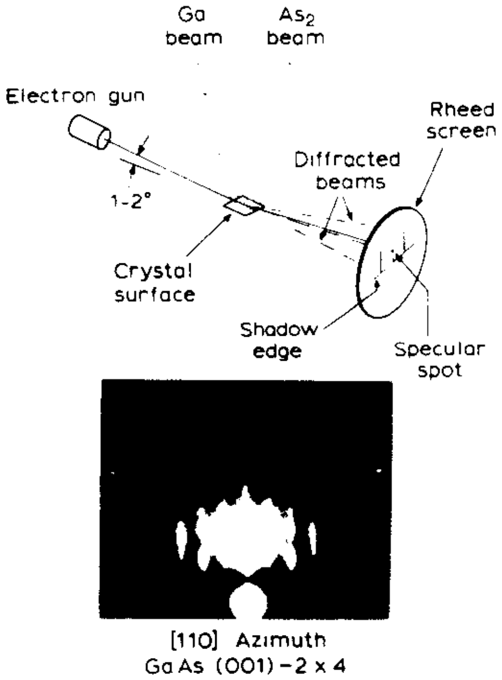

Fig. 3. The experimental arrangement used for the study of RHEED data during MBE growth.

This simple picture of the origin and significance of RHEED oscillations and the recovery of intensity after growth suggests strongly that the growth front exists over several monolayers and that interrupting growth may lead to smoother interfaces. This last point will be discussed in more detail below.

The intensity of the oscillations for specularly reflected electrons depends strongly upon both the incidence angle and azimuth of the electron beam. Lent and Cohen[32] explained the variation with incidence angle using a simple kinematic model; electrons scattered from adjacent

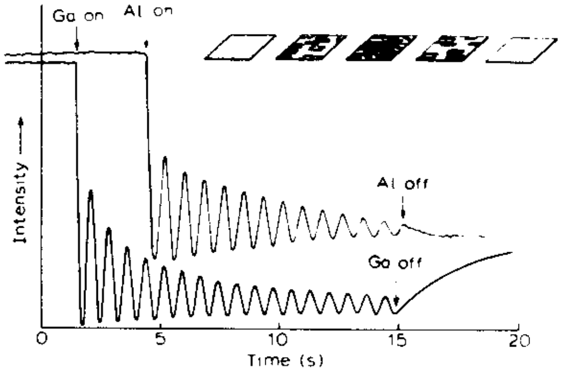

Fig. 4. Typical RHEED oscillations for GaAs and AlAs using As_4 at a substrate temperature of about 600°C. The intensity of electrons specularly reflected from the surface in a [100] azimuth from a (001) oriented monitor slice are measured as a function of time at the centre of rotation. This ensures accurate determination of the true growth rate for actual samples grown subsequently under the same conditions.

terraces in a two level system give rise to constructive or destructive interference. When the beams from neighbouring terraces are out of phase ("off-Bragg") the intensity of the specular beam will depend more strongly upon the step density on the surface than it will when the beams

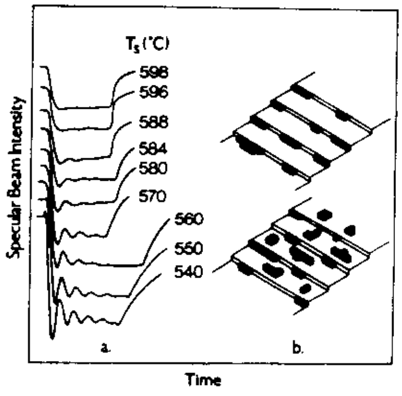

Fig. 5. RHEED intensity measurements on a vicinal plane sample showing oscillations at low temperature corresponding to growth at islands nucleated on the terrace. At higher temperature where the diffusion length of the group III element is longer oscillations disappear because growth takes place at step edges.

are in phase ("on-Bragg"). Whilst this model cannot explain the observed azimuthal dependence or the dynamical nature of many of the effects observed[33] it does nevertheless appear that under appropriate conditions and at small angles of incidence, the specular beam intensity can give a measure of the step density on the surface.

Monte Carlo studies: The first M-C studies of film growth[12,34] seemed to confirm the models proposed on the basis of a kinematical treatment of the RHEED data. The early work of Singh and Madhukar[34] suggested that the growth front would exist over a number of monolayers (ML) with the transition region getting broader with increasing temperature (3ML at 600K to 7ML at 960K).

Later, using a kinematic approximation based on the M-C predictions of the growth front morphology, the expected RHEED intensity oscillations were calculated with impressive agreement[35]. This led subsequently to a number of important suggestions. First, that the difference between the growth of AlGaAs on GaAs, the so-called "normal" interface, and the growth of GaAs on AlGaAs, the "inverted" interface, is due fundamentally to a lower diffusion length of Al on the surface compared to Ga at the same temperature[36,37]. The morphology of the growth front was shown to depend upon the mean number of sites "visited" by the cation (Ga or Al) before becoming incorporated into the lattice. Increasing the substrate temperature to improve the diffusivity, particularly of Al, should result in a smoother interface, contrary to the earlier work[34] and slowing the growth rate should improve matters. Other important results to emerge from these studies include: the observation that an optimum set of growth conditions can be found, depending primarily on substrate temperature and arsenic flux, which will result in good inverted interfaces[38]; the idea that thin layers containing only the more mobile cations will improve the morphology of thick alloy films[39]; and most crucially that interrupting the growth to allow the surface to smooth might improve interface morphology[40]. The factors controlling the roughness of interfaces will be discussed in more detail below following a discussion of processes occurring at high temperatures in both binary compounds and alloys.

All of the calculations discussed above take as a basis the kinetic data obtained using modulated molecular beam measurements and the models have attempted to include all of the processes known to occur during growth. This led Ghaisas and Madhukar to propose their configuration dependent reactive incorporation (CDRI) model in which, depending upon the model parameters they chose, the build up of As coverage is delayed with respect to Ga, the reaction limited incorporation (RLI) situation[35].

Recently a much simpler model has been used by Clarke and Vvedensky to simulate MBE growth. In their model a single species, randomly deposited on the surface, is allowed to migrate before incorporation into the growth front, to diffuse following emission from such growth islands and to re-evaporate from the surface[41]. The step density on the surface is calculated during the simulation for comparison with the time dependent RHEED intensity measurements. All of the features discussed above are reproduced by this simplified model including the disappearance of RHEED oscillations on vicinal surfaces at high temperature[42] and the improved interface smoothness resulting from slow or interrupted growth[43]. This agreement suggests that the conditions normally employed for MBE growth, namely using an excess As flux, correspond to the CDRI situation discussed by Ghaisas and Madhukar rather than the RLI situation in which the As is delayed with respect to the Ga coverage. It follows that the growth mode under such circumstances is determined by the behaviour of the group III species on the surface. The presence of the group V flux can however have a significant influence on the diffusion behaviour of the cations as shown both theoretically, by the M-C studies and experimentally, by Horikoshi et al[44], who demonstrated improved growth at low temperatures by alternately supplying Ga and As to the surface. The diffusion of Ga and Al is enhanced if a low As flux is used during cation deposition.

2.2 Growth of Alloy (mixed binary compounds) Films

There are two distinctly different situations in the growth of alloy films namely those in which different group III elements are used such as $In_xGa_{1-x}As$ or $Al_xGa_{1-x}As$ and mixed group V elements such as $GaAs_xP_{1-x}$ or $InGa_xSb_{1-x}$. For simplicity I will refer to these as InGaAs, AlGaAs, GaAsP and InGaSb respectively.

Alloys with mixed group III elements: The two examples of alloys of this type listed above have been studied in some detail using the same techniques applied to binary compounds. At low temperatures with an excess group V flux the situation is quite straightforward, both group III elements have a unity sticking coefficient and the growth rate is determined by the total cation flux reaching the surface whilst the composition depends on the relative concentrations of the group III elements.

For In containing alloys of this type the rate of loss of group V species from the surface at high temperatures has been shown both by MMBS and RHEED studies to be similar to that observed in InAs or InP[45]. To compensate for the loss of As_2 it is necessary therefore to use rather high group V fluxes. For AlGaAs, however, it has been suggested that the opposite behaviour is observed and that the alloy grows more As rich than GaAs at high temperatures. Evidence for this statement is based entirely on RHEED studies, but the surface reconstructions observed for AlAs are quite different from those seen in GaAs which may confuse the issue[46]. A much more comprehensive study combining RHEED with MMBS studies is required to resolve this question.

In both InGaAs and InGaP evidence for the segregation and subsequent desorption of the higher vapour pressure element, In, has been observed[45]. This produces both a change in film thickness and composition compared with growth at low temperatures using the same fluxes.

Similar segregation behaviour has been reported in Al containing alloys[47-49]. The evaporation of Ga during the growth of AlGaAs at high temperatures has been considered from a thermodynamic standpoint by Heckingbottom[50]. For a fixed As overpressure the calculated activation energy from an Arrhenius plot is about 4.6eV. This is significantly larger than the figure observed under equilibrium conditions for Ga over Ga[51] or Ga over GaAs[52-54] which is about 2.7eV. The difference arises because at low temperatures evaporation is occurring under As rich conditions whereas at higher temperatures there is a gradual transition to Ga stable growth. This implies that both the growth rate and composition of AlGaAs alloys will be altered by an increase in As overpressure and that the rate of loss of gallium for fixed temperature and As flux will depend upon the Al flux reaching the surface during growth.

RHEED oscillations corresponding to layer by layer sublimation of GaAs have been observed at high temperatures[55,56]. The GaAs re-evaporation rate depends upon the As flux reaching the surface as expected thermodynamically[50] and it was also established that a few monolayers of AlAs could totally suppress the loss of underlying GaAs. This last observation suggests that the observed segregation of Ga during the growth of AlGaAs[47-49] does not always occur.

Direct measurements of the rate of loss of Ga during the growth of GaAs and AlGaAs using RHEED oscillations suggest that the loss of Ga may occur from the mobile precursor state of the atoms on the surface and not by dissociation of the AlGaAs[57-58]. In this study no dependence upon the As flux or the Al fraction was observed as might be expected if Ga is lost before film growth. This is entirely consistent with the earlier work of Arthur[15] who observed a finite temperature dependent lifetime for

Ga. It is also reasonable since the binding energy of Ga in its mobile state (about 2.5-2.7eV) is much lower than the measured activation energies for the re-evaporation of GaAs 4.7eV[55] and 4.6eV[56] from the RHEED sublimation studies and calculated thermodynamically 4.6eV[50].

At present therefore there are two conflicting views of how to predict the loss of the more volatile group III element during the growth of alloys such as AlGaAs, InGaAs and InAlAs at high temperatures. The thermodynamic arguments suggest that the rate of loss will depend upon both the As flux and the alloy fraction and this will certainly occur for a situation where the loss occurs after growth of the film. If the group III element is lost from the mobile precursor state before reacting with the group V element no strong dependence upon either the anion or less volatile cation flux is expected. Further work is required to clarify this situation. The possibility of surface segregation of the more volatile group III element is also not clear at present.

Alloys with mixed group V elements: The situation for mixed group V element alloys is quite simple when growth occurs at temperatures where there is no substantial loss of material from the surface. Under such conditions the element with the lower vapour pressure is incorporated preferentially, thus for GaAsP[59] or InAsP[59] the sticking coefficient of As is much greater than P and does not depend upon whether the dimer or tetramer is used. For GaSbAs[60] or other antimony containing species Sb has the higher sticking coefficient. At low temperatures therefore the composition of the alloy can be easily controlled by limiting the amount of the preferentially incorporated element and providing an excess of the more volatile species. For example, if the As flux, J_{As}, reaching the $GaAs_yP_{1-y}$ surface is small compared to the Ga flux, J_{Ga}, and an excess P flux is supplied, the As fraction y will be given by:

$$y = 2\, J_{As}/J_{Ga} \; .$$

This will be true for As supplied as As_2 or As_4.

At present the reason for the difference in sticking coefficients for the group V elements is not clear. It has been suggested that it may relate to the different lifetimes of the species[59-60]. This explanation must be incorrect, however, since despite the lifetime of As_2 being much shorter than that of P_4, As is nevertheless incorporated preferentially.

At higher temperatures, when rate of loss of the dimer by dissociation and re-evaporation is comparable to the supply rate composition control becomes much more difficult[60-61]. The rate of loss of the dimer does not relate in any simple way to its sticking coefficients but is determined more by the thermodynamic vapour pressure over the alloy. For GaAsP, therefore, the As fraction will decrease with increasing temperature but for InAsP the reverse will be true[61]. No simple rule can be given therefore in this situation. For Sb containing alloys similar behaviour is expected[60].

For alloys of this type there is a further complicating factor resulting from significant interdiffusion across the heterojunction[62]. Auger and SIMS (secondary ion mass spectrometry) depth profiling through layers of GaP grown on GaAs substrates suggests that the relatively abrupt interfaces observed in AlGaAs-GaAs heterojunctions may not be obtained when a change in group V element is involved. This data may have been influenced by differences in lattice constant giving rise to enhanced interdiffusion. Further work in this area is required to resolve this matter.

2.3 Interfaces in AlGaAs-GaAs Structures

The nature of the interface between GaAs and AlGaAs has been studied in great detail. Despite this, as will be clear from the discussion below, a detailed picture of structural quality of this hetero-interface is only now emerging and some of the models proposed apply, at best, to specific samples studied and should not be regarded as "typical".

If AlGaAs films are grown in the temperature range 630-690°C using As_4 with an Al fraction exceeding 33%, a surface texture is observed which will give rise to interface roughness[47,63-64]. Films grown with As_2 do not, however, exhibit this behaviour. Various causes of this roughening have been proposed including: carbon accumulated at the interface[65-66], an insufficient As surface population when using As_4[67] a reduced mobility for Al compared to Ga[64,68] and Ga surface segregation[47-49]. All of these suggestions appear reasonable and evidence for each model proposed has been presented, however, the generality of the observation lends credance to the intrinsic causes listed above. In a relatively clean growth system (see below), using As_4, we have invariably observed such surface roughness when growing thick films of AlGaAs which supports this conclusion.

More direct evidence for the quality of the interfaces in multi-quantum well (MQW) and superlattice (SL) structures has come from transmission electron microscopy, X-ray diffraction, photoluminescence (PL) and photoluminescence excitation (PLE) studies.

Petroff et al[69] studied the growth mode for GaAs-AlAs SLs as a function of temperature using a cross-section TEM technique. They found that growing at high temperatures (610°C) produced no superlattice reflections but gave rise to a diffraction pattern similar to that observed in an alloy. At lower temperatures (560°C) satellite spots corresponding to the SL period were observed with the degree of ordering decreasing with increasing temperature. The loss of ordering at higher temperatures was thought to arise from a thermodynamic roughening of the type described by Weekes and Gilmer[70] but this is unlikely since this occurs at temperatures well above those used in MBE. It could more reasonably relate to the roughening transition proposed in the M-C simulations[34] but this has not been firmly established.

The precise temperatures quoted in that study are probably incorrect since more recent work in our laboratory has shown that similar SLs can be grown at significantly higher temperatures. Fig 6 shows a typical example of a SL consisting of alternate layers of GaAs 3ML and AlAs 3ML (3+3) grown using As_2 at 650°C. One interesting feature seen in this micrograph and generally observed in such samples is the tendency for the quality of the interfaces to improve through the structure. The initial layers grown on the macroscopically rough alloy follow its texture but they become progressively smoother as growth proceeds.

High resolution lattice plane images of SLs have also been obtained using TEM[71]. There is insufficient contrast for GaAs-AlGaAs interfaces to accurately locate the position of the heterojunction but it is just

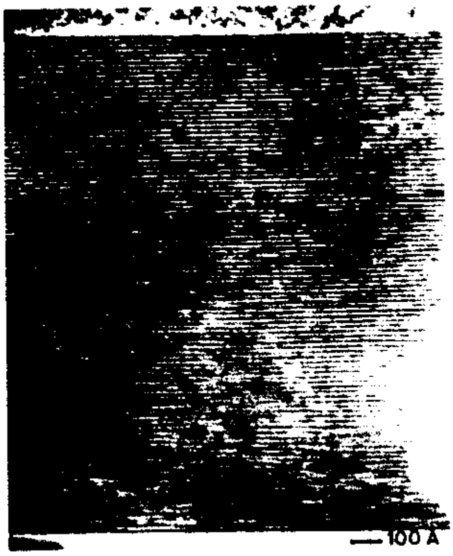

Fig. 6. A cross-section TEM photograph (J.P. Gowers) of a GaAs (3ML) AlAs (3ML) superlattice grown (D. Hilton) by MBE using As_2 at a substrate temperature of 650°C. Note that the initially rough interfaces become progressively smoother as growth proceeds.

possible to resolve the boundary for GaAs-AlAs structures. In both cases it is clear that no crystallographic disorder is present at the interface since no dislocations, stacking faults or twin boundaries are observed.

Two different X-ray techniques have been used to study MQWs and SLs. The small angle scattering method relies on the difference in refractive index between GaAs and AlAs. Below a critical angle X-rays are totally reflected by the surface but beyond this point interference maxima are observed from which the periodic nature of the structure can be deduced[72-73] and in addition the individual thicknesses of GaAs and AlAs can be estimated.

More recently high angle X-ray diffractometry has been applied to similar structures[73-77]. Using the difference in the positions of Bragg reflections from the substrate and the MQW or SL an average aluminum content can be obtained. In making this calculation it is of course essential to take into account the tetragonal distortion of the AlAs or AlGaAs layers. Additional satellite peaks are observed around the main Bragg reflections of the MQW or SL layers due to the periodic modulation of the atomic scattering factor and lattice parameter. The periodicity of the MQW or SL can be deduced from the spacing of the satellite features. Finally by modelling the integrated intensities of the satellite reflections it is possible to deduce both the individual layer thicknesses and to estimate the interface roughness[77]. From such studies estimates of interface roughness from 1 to 4 ML have been proposed, somewhat better interfaces being obtained for AlAs/GaAs structures than equivalent AlGaAs/GaAs samples.

The optical properties of MQWs and SLs have been extensively studied using photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy at low temperatures. Since this subject will be discussed in detail elsewhere I will confine my remarks largely to those relevant to the study of the properties of the interface. In the case of MQWs, contrary to the situation in bulk GaAs, luminescence is observed due to the recombination of free or weakly bound excitons.

It has been shown that exciton linewidths become broader as the well width decreases. Whilst it is clear that factors other than structural imperfections, for example impurities, can cause broadening and it is also evident that the conditions used for the optical experiments are crucial, particularly the excitation density used, a comparison of line widths taken on a series of samples where only the well width has been intentionally varied can give information on the structure of the interfaces. In MQWs variations in exciton linewidth could arise from either inter-well thickness variations due to changes in growth rate

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through the structure or to variations in width within each individual well whilst maintaining a constant average layer thickness (intra-well fluctuations). Variations in well width will in general give rise to changes in the confinement energies of electrons and holes, together with somewhat smaller changes in the binding energy of the excitons; both factors will give rise to the line broadening observed. A simple estimate of the influence of a given degree of variation in well width can be obtained from considering the confinement energy, E, for a particle of effective mass m, in an infinite potential well of width W.

$$E = h^2/(8m\ W^2)\ .$$

Assuming a fluctuation of dW in well width the resulting change in energy dE will be given by

$$dE = -[h^2/(4m\ W^3)]\ dW\ .$$

The confinement energies of electrons and holes can be described more correctly using the envelope function approximation for confined states taking into account non-parabolicity in the conduction band and correcting for the binding energy of the excitons[78]. From this more appropriate calculation it can be shown that the expected variation at small well widths is significantly different from that given above but the general trend to broader lines with decreasing W is confirmed as shown in Fig. 7.

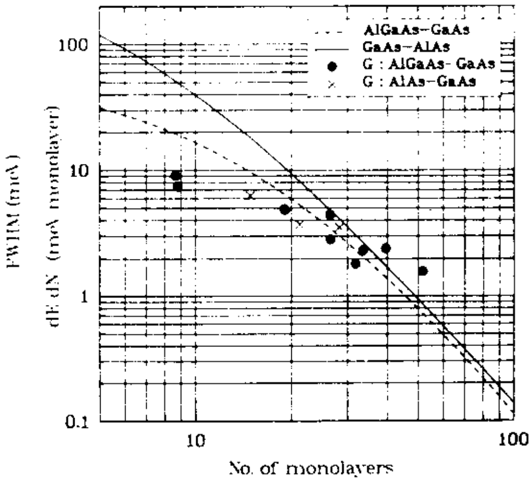

Fig. 7. The rate of change of energy per monolayer for the lowest energy electron to heavy hole transition observed in $Al_{0.33}Ga_{0.67}As$-GaAs and AlAs-GaAs MQWs (G. Duggan). Also shown are measured peak widths (P. Dawson and K.J. Moore) for 60 period MQWs grown by MBE (C.T. Foxon and D. Hilton) in a Varian GEN-II at 630°C using the minimum possible As flux.

Three physically different situations can be envisaged which determine how variations in well width will influence the measured line widths of excitons in PL and PLE spectra.

If monolayer steps occur on a lateral scale much smaller than the diameter of an exciton (~300Å in bulk GaAs), at both interfaces (type A), the energies of given peaks in PL and PLE should be identical but will not correspond to those expected for an integer number of monolayers.

If the lateral scale of monolayer steps becomes comparable to the diameter of an exciton then a so-called "Stokes shift" is expected. In this (type B) situation excitons will be created corresponding to all well widths and hence energies with equal probability, but there will be a tendency for them to thermalize to lower energy states in the wider parts of the well. This will result in a small difference in peak positions in PL and PLE for given transitions. Under such conditions the excitons may be trapped at steps (localized) which will modify the observed energy of the transitions giving rise to the observed "Stokes shift". The magnitude of this effect has been estimated to be at most a few meV and to depend upon the lateral size of the defect[79].

The third physical possibility (type C) arises when the terrace length is much longer than an exciton diameter. In this case two or three discrete peaks with energies corresponding to an integer number of monolayers should be seen in PLE. Even if PL is observed from the narrower regions of a given well it should show significantly lower intensity and the intensity ratios should change with temperature. If on the other hand discrete peaks arise from inter-well fluctuations no change in relative intensities is expected with increasing temperature.

Since the two interfaces ("normal and inverted") are not equivalent it is possible in MQWs to have combinations of two of the types of interface discussed above. The resulting linewidth and number of peaks observed will depend upon the particular combination.

Weisbuch et al[80-81] first showed that for samples grown under optimum conditions the measured linewidths corresponded to interfacial roughening of no more than 1ML. There was also evidently a trend towards better interfaces in samples with narrower well widths. They later demonstrated that the choice of optimum substrate temperature (660-690°C) was the crucial factor in determining the line width[82]. At 570°C the interface roughness was about 5-6ML (15Å) decreasing with increasing temperature, at the same time they observed a corresponding increase in photoluminescence efficiency. Above 690°C the linewidths increased but no corresponding decrease in efficiency was observed. In this study there appears to be a very small "Stokes shift" present which indicates that one of the interfaces is probably of type B and since only single peaks were observed the other interface is probably of type A.

Samples with somewhat narrower linewidths were later obtained by a number of groups, Devaud et al[83] reviewed this work and showed that for "better" samples peak splitting was observed with a spacing corresponding to an integral number of MLs with no evidence for a "Stokes shift". Their samples were grown at about 695°C using the minimum amount of As required to give As stable growth. These samples probably contain one interface of type A (GaAs grown on AlGaAs) followed by a second interface of type C (AlGaAs grown on GaAs).

In our laboratory we have also prepared samples of this type grown under rather different conditions, the substrate temperature was 630°C and again minimal As flux was used. Growth rates of 1ML s^{-1} were used for GaAs with correspondingly higher rates for the AlGaAs barriers. The low temperature linewidths are shown in Fig. 7. The interface roughness is less than 1ML and the trend observed earlier to "better" interfaces in narrower AlGaAs samples is observed[80-81]. Electrical measurements (see below) have established that the purity of our samples is among the best available to date and it is perhaps significant that good interface properties have been obtained at lower substrate temperature than reported above.

Our samples fall into several categories. For relatively wide MQWs (similar to those discussed above) double peaks are observed with a separation corresponding to 1ML difference in well width and no "Stokes shift" (<1meV) is observed. Studies of the temperature dependence of the PL from these samples indicate that the variations arise from intra-well fluctuations and not variations in growth rate. It is probable therefore that these MQWs have type A, at the "inverted", and type C, at the "normal", interfaces. The roughness produced by growing the alloy film has been sufficiently smoothed by the growth of the binary well to give a type C "normal" heterojunction.

For our narrowest AlGaAs MQWs (9ML) a quite different behaviour is seen, a single line with a half width of about 9 meV is observed in PL corresponding to a well width variation of about 1/2 ML. From a comparison of PL and PLE a "Stokes shift" of between 6 and 8 meV is observed depending upon the detection energy used for PLE. In this sample therefore the interfaces are of type A and type B and insufficient GaAs has been grown to reach the steady state step distribution needed for a type C interface.

Also included in Fig. 7 are measured linewidths for AlAs-GaAs MQWs grown under identical conditions. Because the expected energy shift per monolayer is higher for any given well width in MQWs with AlAs barriers, as shown in Fig. 7, for equivalent interface roughness one would expect broader lines. In fact the measured linewidths are almost identical in wider well samples to those observed in AlGaAs MQWs, it follows that this must mean that the interfaces obtained in the all binary structures are "better" than those observed in the AlGaAs-GaAs samples. For the narrow well samples they appear to be of comparable quality.

Much narrower line widths than those described above have been reported by Reynolds et al[84] for a series of AlGaAs-GaAs MQWs with well widths of around 100Å (36ML). The exact growth conditions were not specified in this paper but a reference to earlier work suggests that they may be the samples grown at 700°C exhibiting multiple peaks[85]. The spacing of peaks corresponded to well width fluctuations of about 1/2 ML which is physically unreasonable for an intra-well variation and the same authors later suggested[86] that they arose from inter-well variations. To see such small inter-well variations suggests that the interfaces seen by the exciton are indeed much "better" than those observed before but this result could also be explained if both interfaces are of type A, that is, with extremely small terrace widths relative to the exciton diameter[85].

In summary for conventional growth by MBE the so-called "inverted" interface produced when growing GaAs on AlGaAs or AlAs seems probably to be of type A, that is, with monolayer steps on lateral scale very short compared to an exciton diameter. The "normal" interface obtained when growing AlGaAs on GaAs appears to be of type B or C. Type B interfaces are in general obtained during growth at lower substrate temperatures or in samples where only thin layers of GaAs (<50Å or 20ML) are grown after an Al containing layer. Type C interfaces will be obtained by growth at high temperatures of a GaAs layer sufficiently thick to reach the steady state step distribution corresponding to the particular growth conditions. Finally the degradation in exciton line width observed when growth takes place above an optimum temperature may arise as a result of increasing the terrace length on the Al containing surface, converting a type A interface into type B which appears to be less perfect in PL or PLE.

The effect of modified growth procedures on interfaces: The idea that interrupting growth might lead to smoother interfaces was implicit in the model proposed by Neave et al[28] to explain observed RHEED oscillations and subsequent recovery and was explicitly proposed as a result of the M-C studies of film growth[40]. The procedure adopted is to close the shutters in front of the group III sources and allow the surface to anneal under an As flux for a period ranging from a few seconds to several minutes. During this time it is supposed that the number of monolayer steps on the surface will decrease as a result of migration from small to large islands of the group III elements.

The first direct evidence for effect of interrupts on the quality of the interfaces came from studies of the PL linewidth in samples grown using this modified procedure[87-89]. For all of these studies a low substrate temperature was used and only the GaAs surface could change during the growth interrupt. There is no evidence from RHEED recovery studies that this is expected to occur for AlGaAs or AlAs surfaces. This was later confirmed in an elegant series of experiments by Tanaka et al[90-91]. They studied the effects of interrupts at each of the two interfaces and compared the resulting PL with samples grown conventionally and with interrupts at both interfaces for the complete range of AlGaAs alloys. For Al fractions of >0.5 they propose that the "inverted" interface will be of type A with atomic steps of 40A spacing and that these will not change on annealing. For the GaAs "normal" interface, however, a 200Å step length can be increased by the interrupt resulting in a change from a type B to a type C interface. RHEED data taken in the same equipment seems to confirm this picture. For AlGaAs alloys with an Al content of <0.3 the behaviour is similar to GaAs.

A somewhat similar picture was proposed by Koteles et al[92] who used both PL and PLE to confirm the interpretation of their data. They observed multiple peaks corresponding to monolayer differences in well width but in addition observed peaks associated with bound excitons in samples where growth had been interrupted. They stressed that multiple peaks observed in PL did not prove the existence of monolayer steps.

There are several major problems in accepting this data at face value. First the linewidths obtained using the normal growth process[87-01] are far worse than those reported previously[80-83] and those illustrated in Fig. ,. Only by using the modified procedure were linewidths comparable to those obtained conventionally achieved. The second key point is that completely conflicting data for samples grown under similar conditions has been presented which seems to be equally valid[93-94]. In one of the above reports discrete peaks were again ascribed to monolayer fluctuations but a careful examination of the data suggests that the energy differences are too small and additionally a significant "Stokes shift" is observed suggesting the presence of bound rather than free excitons.

One odd fact which seems to have been overlooked is that having the As flux present during growth interrupts may be detrimental since it has been established that this can limit the diffusion length of the group

III atoms on the surface[44]. It may be that a modified procedure where both group III and group V fluxes are interrupted perhaps for different times may prove a useful alternative technique.

One other point which has received little attention so far is the role of impurities reaching the surface during the growth interrupt. This may directly influence the subsequent growth by pinning steps which might otherwise propagate in a two-dimensional mode. It may also lead to reduced optical efficiency and increased carbon incorporation[94-95].

At present we can be sure that interrupting growth is changing the nature of the interface but far from certain exactly what effect this perturbation produces. The simplified model proposed by most authors is apt to be misleading and studies may at best relate to what happens in a particular MBE machine under the conditions used and should not be taken as proof that identical results will be obtained under different conditions. Much more careful work is needed to correctly identify those conditions which will lead to genuinely smoother interfaces of type C.

3. GROWTH OF HIGH PURITY STRUCTURES

Many of the more demanding applications of MBE require very high purity layers with free carrier concentrations below 10^{14} cm^{-3}. The detailed techniques we use to obtain material of this quality have been reported elsewhere and will be briefly outlined below[96]. The MBE sources are outgassed to high temperature in the preparation chamber and loaded with the best possible sources of material. The whole MBE equipment is then baked for 2 weeks resulting in a base pressure of $<1\times10^{-10}$ Torr. Before each growth the sources, with the exception of As, are carefully outgassed at least 50°C above their operating point. Samples are loaded using high purity In onto Mo blocks which are then outgassed to remove water vapour before transfer to the growth chamber, impurities evolved during the removal of the native oxide are allowed to pump away before growth. Finally growth rates of 1 ML s^{-1} are used for GaAs with the As flux adjusted to a minimum required to give a 2-fold RHEED pattern on the (100) surface in the [110] azimuth. Sample rotation rates and growth times for individual layers have been adjusted to give complete numbers of monolayers.

Using this technique we have grown several thick intentionally doped GaAs films with electron mobilities at 77K of $>1\times10^{5}$ and a best value of 1.33×10^{5} cm^{2} V^{-1} s^{-1}, free electron concentrations were about 2×10^{14}

cm^{-3}. This suggests the background acceptor concentration in our material is below $2x10^{14}$ cm^{-3}. Recently we have reported an undoped GaAs film, grown using a superlattice prelayer, which at low temperature has luminescence dominated by free exciton emission[97].

For several years the best reported mobilities for 2DEG samples were approximately $2x10^{6}$ cm^{-2} V^{-1} s^{-1} at a sheet carrier density of about $5x10^{11}$ cm^{-2} [99-100]. In our studies we were able to obtain similar mobility material at somewhat lower electron densities, as shown in Fig. 8[101]. We reasoned that the ionized Si atoms close to the surface could contribute significantly to the total ionized impurity scattering and by further optimizing the structure managed to obtain mobilities of $3x10^{6}$ and $4.4x10^{6}$ cm^{2} V^{-1} s^{-1} at 4.2 and <2K respectively[102-103]. The important point first discussed by Weimann and Schlapp[100] is that for very wide spacer layers (>400Å in our case) the mobility is limited mostly by the quality of the undoped GaAs and nature of the interface. Using an MQW prelayer to improve the background scattering in the GaAs and a wide lightly doped region of AlGaAs to avoid the scattering by ionized impurities near the surface has recently enabled English et al to grow a 2DEG with a mobility at <2K of $5x10^{6}$ cm^{2} V^{-1} s^{-1} at a sheet carrier density of $1.6x10^{11}$ cm^{-2} [104].

The mobility of the 2DEG formed at the GaAs-AlGaAs interface depends upon the order in which the layers are grown. For the "normal" structure, as outlined above, extremely high mobilities are obtained, but for the "inverted" heterostructure the best results reported so far are more than a decade lower[105]. Three reasons have been suggested for the inferior performance: interface roughness (as discussed above), impurity build-up at the interface and Si migration from the underlying doped AlGaAs due to segregation. We have recently studied this problem in modulation doped quantum wells (QWs) thin enough for the electrons to sample both interfaces. We have observed that in as-grown and annealed samples the low electron mobility is associated with electron localization at the inferior inverted interface[106]. The localization can be removed by replacing the bulk AlGaAs layers by a short period SL of equivalent band gap. In such QWs the 2DEG mobility is, for samples with narrow undoped spacer layers, equivalent to that obtained in a conventional 2DEG formed at the "normal" interface.

Radulescu et al[107] also studied transport in QWs and found that an anisotropic mobility exists for samples grown with thick AlGaAs barriers. This was removed by replacing the first confinement layer with either a short period SL or AlGaAs grown at higher substrate temperatures (690°C). Again this points to the "inverted interface" being degraded by an intrinsic morphology problem rather than other extrinsic factors.

For our work at Philips on understanding the growth and properties of SLs we have grown four sets of samples: the first set were MQWs with well widths from 9 to 54ML with direct bandgap barriers thick enough (55ML) to provide isolated QWs, the second set had 27ML AlAs barriers with GaAs well widths from 2ML to 27ML, the third set had GaAs well widths of 9ML with AlAs barriers from 2 to 27ML and the final set are SLs with equal GaAs and AlAs thicknesses from 1 to 6ML. In each series control samples of known properties were grown to check the state of the machine.

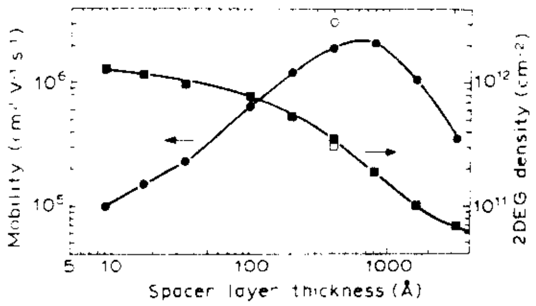

Fig. 8. Variation in carrier density and mobility of 2DEG samples at 4K as a function of the undoped spacer layer thickness. The results plotted are for samples illuminated to saturate the persistent photo-conductivity effect. Two thicknesses of doped AlGaAs were used, 400Å (solid symbols) and 500Å (open symbols).

From the first set of samples we were able to compare quantitatively the measurements of well and barrier thickness determined by RHEED, X-ray, TEM and PLE measurements with satisfactory agreement[108]. Samples grown for this study also showed for the first time clearly resolved features for the 2S state of the light and heavy hole excitons in PL and PLE[109-110] from which accurate estimates of the exciton binding energy were obtained. In addition transitions to the confined hole state of the split-off band were identified[111].

In the second set of samples we see a type I emission process from electrons and holes confined in GaAs wells >11ML thick, for narrower GaAs wells type II luminescence from electrons confined in the AlAs barriers[112] recombining with holes in the GaAs is also observed. Fig. 9 shows an example of low temperature PL from two samples of this type. Features relating to both type I and type II recombination can be identified in the spectra together with phonon replicas associated with the type II emission process.

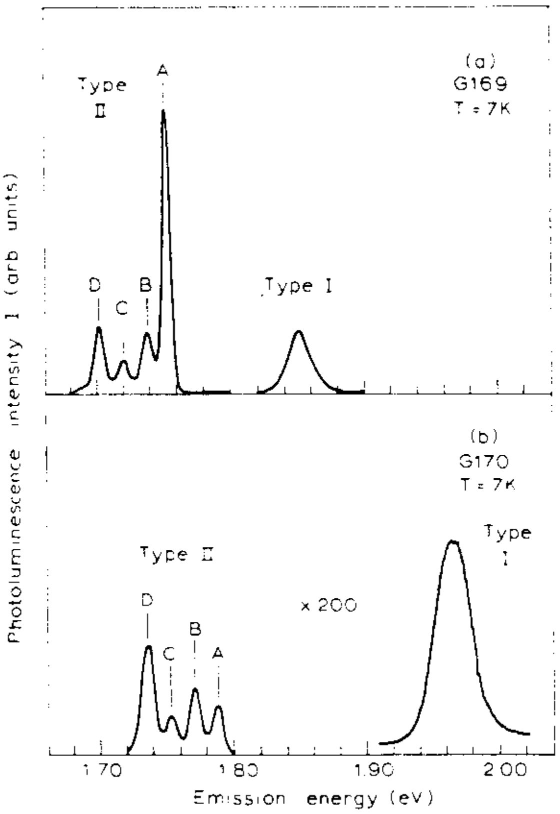

Fig. 9. Low temperature (7K) photoluminescence spectra of two GaAs-AlAs MQWs showing emission from type I recombination of electrons and holes localized in the GaAs and type II luminescence from electrons at the X minimum of the AlAs recombining with holes in the GaAs.

Standard 9ML GaAs, 27ML AlAs control samples were grown during the course of this study under a variety of growth conditions. The spectra obtained are extremely sensitive to the choice of growth rates and substrate temperature. Spectra like those shown in Fig. 9 are only obtained under optimum conditions. Much broader, less well resolved features are observed when poorer interfaces were obtained.

From the third set of samples we were able to show a gradual increase in confinement energy at the X point in AlAs and a decrease in confinement energy at the Γ point in GaAs (due to coupling of electron states in the GaAs) as the AlAs barrier width decreased. This data is shown in Fig. 10[113]. This results in a reversion to type I emission for samples with a GaAs well width of less than 4-5ML. PL and PLE studies from the SL samples are not yet complete.

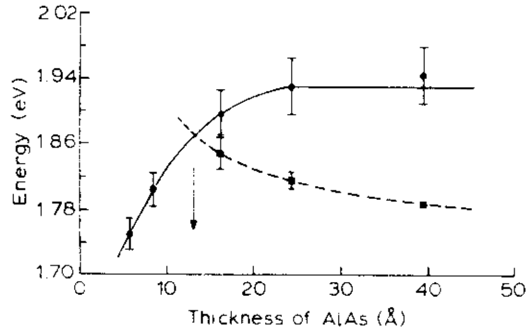

Fig. 10. The variation in energy of the type I and type II transitions in GaAs-AlAs MQWs as a function of AlAs thickness for a constant GaAs well width of 9ML. A transition from type I to type II behaviour is observed at AlAs thicknesses between 4 and 5ML.

4. DEVICES BASED UPON LOW-DIMENSIONAL STRUCTURES GROWN BY MBE

Many potential high speed device structures have been proposed which rely on the use of low-dimensional structures for their operation. Of these only the high electron mobility transistor has demonstrated improved performance over conventional devices for both microwave and digital applications. The growth, properties and performance limitations of this device have recently been discussed in detail by Drummond et al[114] and only key points relating to the MBE growth process will be outlined here.

The high electron mobility transistor (HEMT), two-dimensional electron gas transistor (TEGFET) or modulation doped field effect transistor (MODFET) is based on a narrow spacer 2DEG. The current between a pair of source drain contacts is modulated by the potential applied to a gate electrode to provide transistor action similar to that observed in a conventional GaAs MESFET. The high frequency performance of this new type of device, in particular the noise performance, is better than that of the conventional FET for equivalent device geometries. Very approximately, the performance of 0.5 micron HEMT is equivalent to that of a 0.25 micron MESFET. The improved performance arises in part from the higher electron velocity achieved in 2DEGs even under high injection conditions and partly from the electron confinement to the narrow region at the heterojunction interface.

For low noise applications a 2DEG density of about $1\text{x}10^{12}$ cm^{-2} is required to reduce the source resistance to an acceptably low level. This can readily be obtained using a narrow undoped spacer (0-20Å) and a relatively highly doped AlGaAs region ($3\text{-}4\text{x}10^{18}$ cm^{-3}). Parallel conduction is avoided by using a recessed gate geometry and high frequency performance can be aided by using thick heavily doped regions of GaAs in the source and drain regions of the device. With such a structure transconductances of 300 mS/mm are readily achieved in the AlGaAs system using a 0.5 micron gate length.

For digital integrated circuit applications the main problems encountered relate to the defect density observed on the surface after growth. For example in a 4 Kb static random access memory 27000 transistors are integrated each of which must perform to specification. To achieve even a reasonable yield defect densities of <500 cm^{-2} must be obtained. On typical MBE material defects arise from poor substrate preparation or mounting, from oxidation of the group III sources and from particulates within the MBE equipment. A variety of methods has been proposed to improve these factors but this still remains a major problem. Gas source MBE does seem to offer some significant advantages in this respect over conventional MBE and may be attractive in this application.

Finally it is appropriate to point out that other materials systems such as AlInAs-GaInAs may offer some significant advantages over the AlGaAs-GaAs devices currently being produced[115].

The properties of both conventional and MQW lasers grown by MBE have been reviewed by Tsang[10] who made a major contribution in this area. He discovered that AlGaAs with high optical efficiency could be obtained by growth at high temperatures (680-700°). There are two possible reasons for the improved properties, either reduction in the number of intrinsic defects or a reduced incorporation of oxygen which is known to affect both the electrical and optical properties of AlGaAs[115]. At present no clear evidence exists to distinguish between the two possibilities.

The use of MQW structures has resulted in devices with improved threshold current, arising from higher net optical gain, and improved threshold-temperature dependence[10] compared with conventional double heterostructure lasers. It has also proved possible to achieve lasing at wavelengths as low as 710 nm using MQWs with GaAs well widths of only 4-5 MLs[116].

5. CONCLUSIONS

MBE has emerged as a powerful growth technique for the preparation of structures based mainly on AlGaAs-GaAs lattice matched alloys. Using the RHEED oscillation technique it can provide complex samples with individual layers down to a monolayer scale of known thickness and composition. Such well characterized samples are essential to study the physical properties of low-dimensional structures. A detailed picture of the AlGaAs-GaAs interface has emerged from X-ray, TEM, PL and PLE studies and novel devices such as HEMTs and MQW lasers based on low-dimensional principles have shown improved performance compared with conventional structures.

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