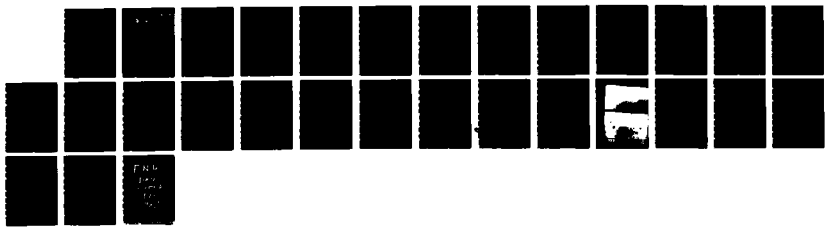
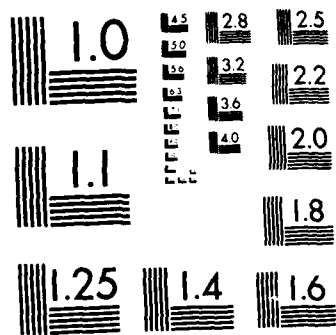


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CALIFORNIA LOS ANGELES DEPT OF MATERIA A MADHUKAR
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Status Report

on

**Some Investigations of Molecular Beam Epitaxial Growth
of III-V Semiconductor Films via Monte-Carlo Computer Simulations,
Carrier Tunnelling and Spectroscopic Ellipsometry**

AFOSR Contract #F49620-83-C-0074

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Submitted by

Assistant of
A. Madhukar
Dept. of Materials Science
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Introduction for Year 1:

The research sponsored under AFOSR contract #F49620-83-C-0074 began on April 15, 1983. The anticipated plan of the work during the first budget year involved primarily

- (a) Spectroscopic Ellipsometry studies of MBE grown GaAs/Al_xGa_{1-x}As(100) and GaAs/In_xGa_{1-x}As(100) structures in the visible to UV range.
- (b) Extending the ellipsometry wave length region from 8000Å to 2.2 μm (i.e. covering the visible to near IR range).
- (c) Carrying out C-V and tunnelling measurements on the GaAs/Al_xGa_{1-x}As/GaAs structures at the JPL Electrical Measurements Laboratory while establishing the same capabilities at USC.
- (d) Studying the phenomena of surface orientation induced "miscibility gap" in the MBE growth of Al_xGa_{1-x}As alloys on GaAs(110).
- (e) Carrying out Monte-Carlo computer simulations of GaAs/Al_xGa_{1-x}As(100) and (110) MBE growth.

The GaAs/Al_xGa_{1-x}As MBE samples, appropriate for the experimental investigations noted under (a), (c) and (d) were to be grown under the supervision of Prof. M. Gershenson. The GaAs/In_xGa_{1-x}As(100) systems were to be grown at the JPL MBE facility as a collaboration between the principal investigator (A. Madhukar) and Dr. F. J. Grunthaner of JPL.

Progress during Year 1:**(1) MBE Growth of GaAs/Al_xGa_{1-x}As**

From the time of the inception of this work, it became clear at a relatively early stage that the USC MBE facility required major effort and investment to be able to grow reliable samples. In an effort to achieve this aim, the principal investigator (A. Madhukar) was forced to take responsibility of the MBE growth as well - a situation not originally anticipated. Accordingly, from July, 1983 until December 1983, major effort was spent making the USC MBE machine operational and putting in place basic support facilities (such as substrate cleaning and preparation). The situation with regard to the MBE machine thus, unfortunately, deprived us of appropriate GaAs/Al_xGa_{1-x}As samples to be able to proceed with the experiments noted under (a), (c) and (d). We did, however, grow

a few GaAs/Al_xGa_{1-x}As/GaAs tunnelling structures around April/May 1983, had them fabricated into actual tunnel structures, and carried out Fowler-Nordheim resonance tunnelling experiments at JPL. The results indicated that the interfacial quality of these structures were rather poor. Indeed, these findings were at the base of the principal investigator taking on the additional responsibility of implementing the MBE growth program as well.

The following major items have to-date been accomplished on the MBE machine:

1. Detecting and fixing leaks.
2. Fixing cryopump, quadrupole mass analyzer, RHEED gun and screen.
3. Redesigning source-oven thermocouple arrangement to achieve stable temperature control and the attendant control on flux.
4. Redesign the pumping system and configuration on the growth chamber. Implementation of the design to be effected over the next few months.

We have, during the past month, grown GaAs samples with a view towards calibration of fluxes, substrate temperature (via usage of eutectic alloys and IR pyrometer) and to obtain an idea of the unintentional background doping type and level. Hall measurements indicate the samples to be p-type GaAs with doping levels between $1 - 5 \times 10^{15}/\text{cm}^3$.

We are now preparing to begin growth of Al_xGa_{1-x}As single interface structures on both GaAs(100) and GaAs(110) surfaces so as to begin the experimental studies noted under (a), (c) and (d).

(2) Spectroscopic Ellipsometry

(a) Preliminary investigations of GaAs/In_xGa_{1-x}As single alloy layers, grown on the JPL MBE system, have been carried out. Some results have also been obtained on GaAs/InAs strained layer superlattices with individual layer thicknesses corresponding to 4 and 8 atomic layers each. Systematic investigations of the comparison of the alloy and superlattice optical behaviour are in progress.

(b) The ellipsometry instrumentation was extended into the near IR regime and testing of the system begun.

(3) Surface Orientation Induced Miscibility Gap: The GaAs/Al_xGa_{1-x}As(110) System

In the absence of reliable MBE samples we spent time fruitfully developing a theoretical model which postulates a possible mechanism for the reported quasi-periodic fluctuations in the Al concentration along the growth direction. The basic physical idea is that a lattice-mismatch induced strain dependent exchange reaction between Al and Ga, coupled with the strain-memory effect, can give rise to long range periodic variations in the Al concentration. This is a totally kinetic and new mechanism which does not involve bulk diffusion - the process responsible for the spinodal decomposition mechanism of phase-separation. Our theory makes specific predictions for the behaviour of the alloy as a function of concentration, etc. which can be tested by the planned experiments.

(4) Electrical Measurements

Setting up C-V and tunnelling measurements capability at USC is underway and expected to be finished in the next few months. The Hall mobility measurements facility has been in operation and several measurements as a function of temperature down to 4.2K have been made.

AFOSR Contract #F49620-83-C-0074

Status Report for Years 2 and 3:

We have conducted systematic studies¹⁻⁴ of the RHEED intensity behavior of both static and dynamic GaAs/AlGaAs(100) surfaces using a Physical Electronics (ϕ) Model 400 MBE system. The objective of these studies has been to examine surface kinetic processes and their consequences for surface morphology critical for formation of high quality interfaces between thin as well as thick layers. A number of fundamental features and their pragmatic consequences for defining "optimal" growth conditions have been determined. These have led to new ideas¹⁻⁹ and variations on the customary practices in MBE growth. A variety of GaAs/Al_xGa_{1-x}As^{2,11} and GaAs/In_xGa_{1-x}As^{6,10} quantum well structures employing these new ideas and optimal growth conditions have been grown. The latter have been examined^{6,10} with high resolution cross sectional transmission electron microscopy (XTEM) using a Philips 420T Analytical Microscope (with a line to line resolution of 2.2°A and a point to point resolution of 3.2°A) available at the USC Center for Electron Microscopy and Microanalysis (CEMMA). During the same period facilities for optical and transport studies were established within this author's laboratory with funds provided under the DoD-URIP program. Single and multiple well structures of GaAs/Al_{0.33}Ga_{0.67}As with GaAs well thicknesses $\leq 20\text{ML}(56.6\text{\AA})$, grown under our RHEED determined optimized growth conditions, have been examined via photoluminescence spectroscopy and reveal the narrowest line widths reported to date.¹¹ The quality of both the normal and inverted interfaces is shown to be kinetically controlled and better than $\pm 1\text{ML}$ fluctuation in the

well width. Since many of the findings of our RHEED and TEM studies have not yet appeared in published literature (though some of these have been presented at various recent conferences), we provide here a brief description of some of these findings.

In the typical behavior of the specular beam RHEED intensity oscillation shown in Fig. 1, these are six basic features which are of significance:

1. The intensity, I_0 , of the starting static (i.e. stabilized, no growth) surface.
2. The oscillation period, τ_p .
3. The rate of damping of the oscillation maxima and the behavior (rise or decay) of the oscillation minima.
4. The intensity, I_g , of the steady state growth (i.e. when the oscillations, at least on the level of detection, have died out)
5. The rate of recovery of the intensity upon termination of growth
6. The intensity, I_R , of the recovered, stabilized surface.

Each of these features as a function of various combinations of the substrate temperature, As_4 pressure and growth rate (i.e. group III flux) provides considerable information on the role of kinetic processes as well as pragmatic considerations for growth optimization, depending upon the nature of the quantum well structure and the desired objective. In the following we discuss each of these features individually.

A. STATIC SURFACES

The chemical cleanliness, surface stoichiometry and surface morphology of the surfaces on which interfaces are formed is of utmost importance to realization of high quality quantum well structures. A study⁷ of the behaviour of the static surface specular beam intensity, I_0 , as a function of the substrate temperature (T_s) and As_4 pressure (P_{As_4}) was undertaken to examine the morphological quality of the starting GaAs(100), As-stabilized (2x4) surfaces. In Fig. 2 we show the behaviour of I_0 as a function of T_s at a fixed P_{As_4} .

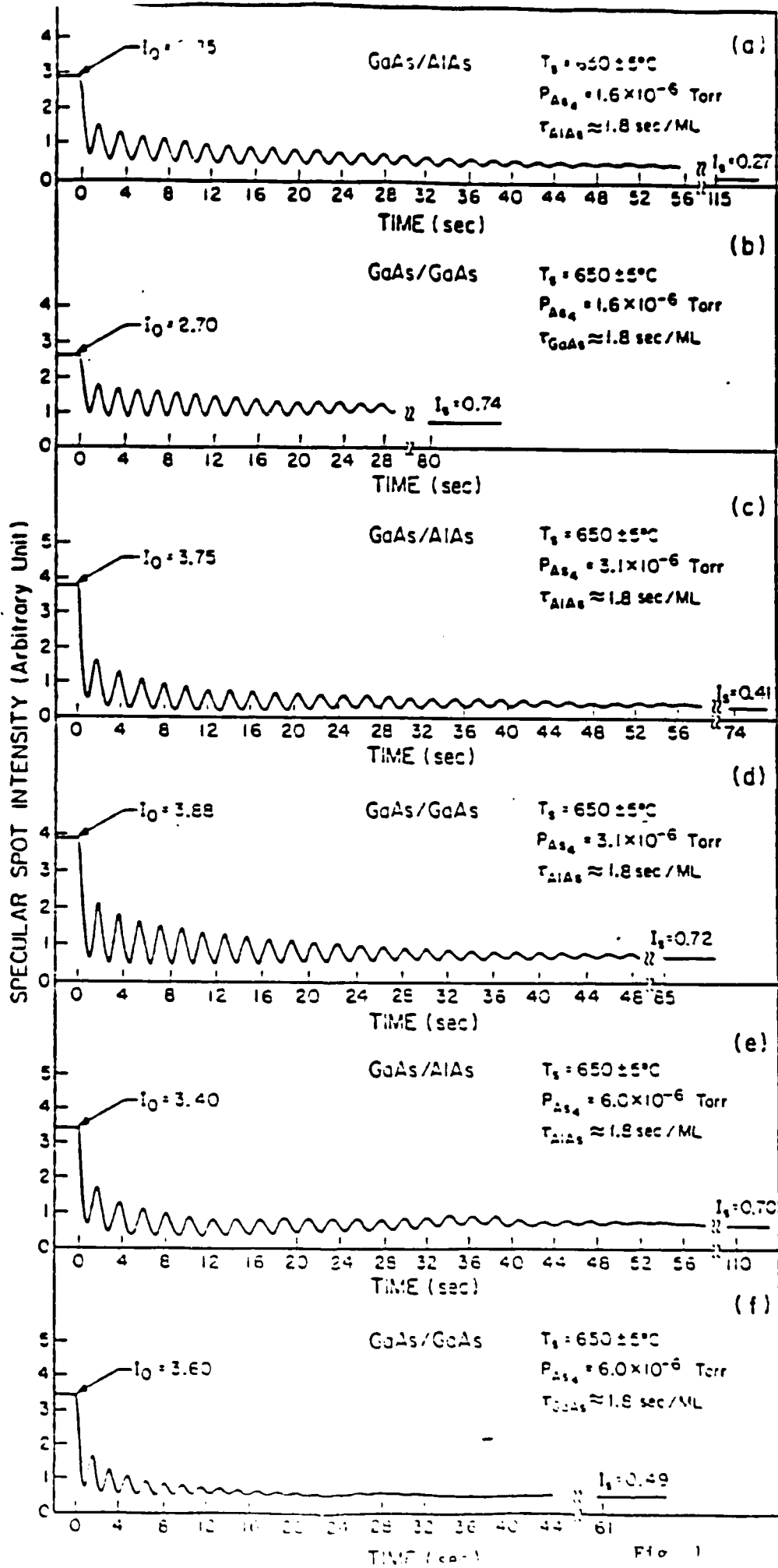


Fig. 1

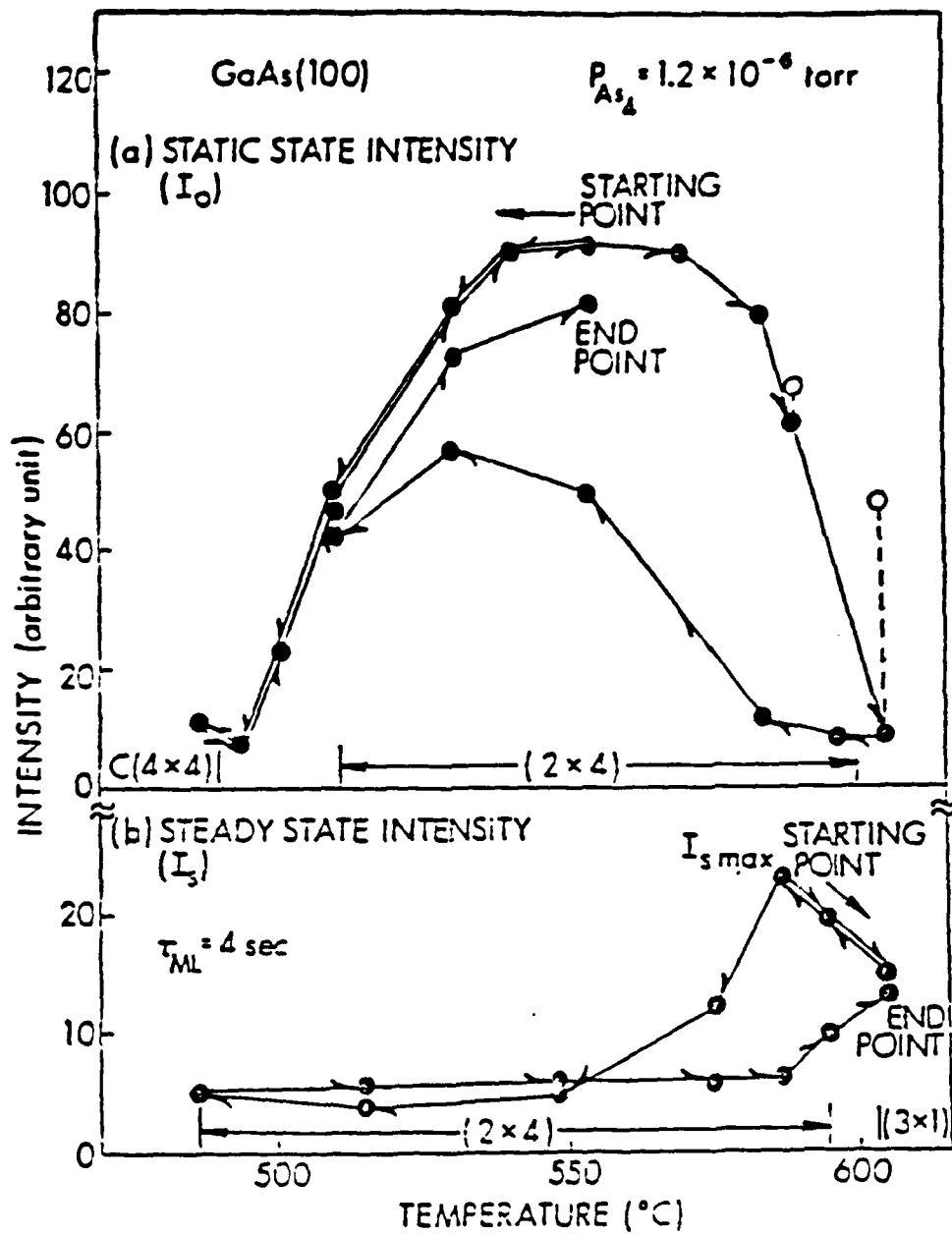


Fig. 2

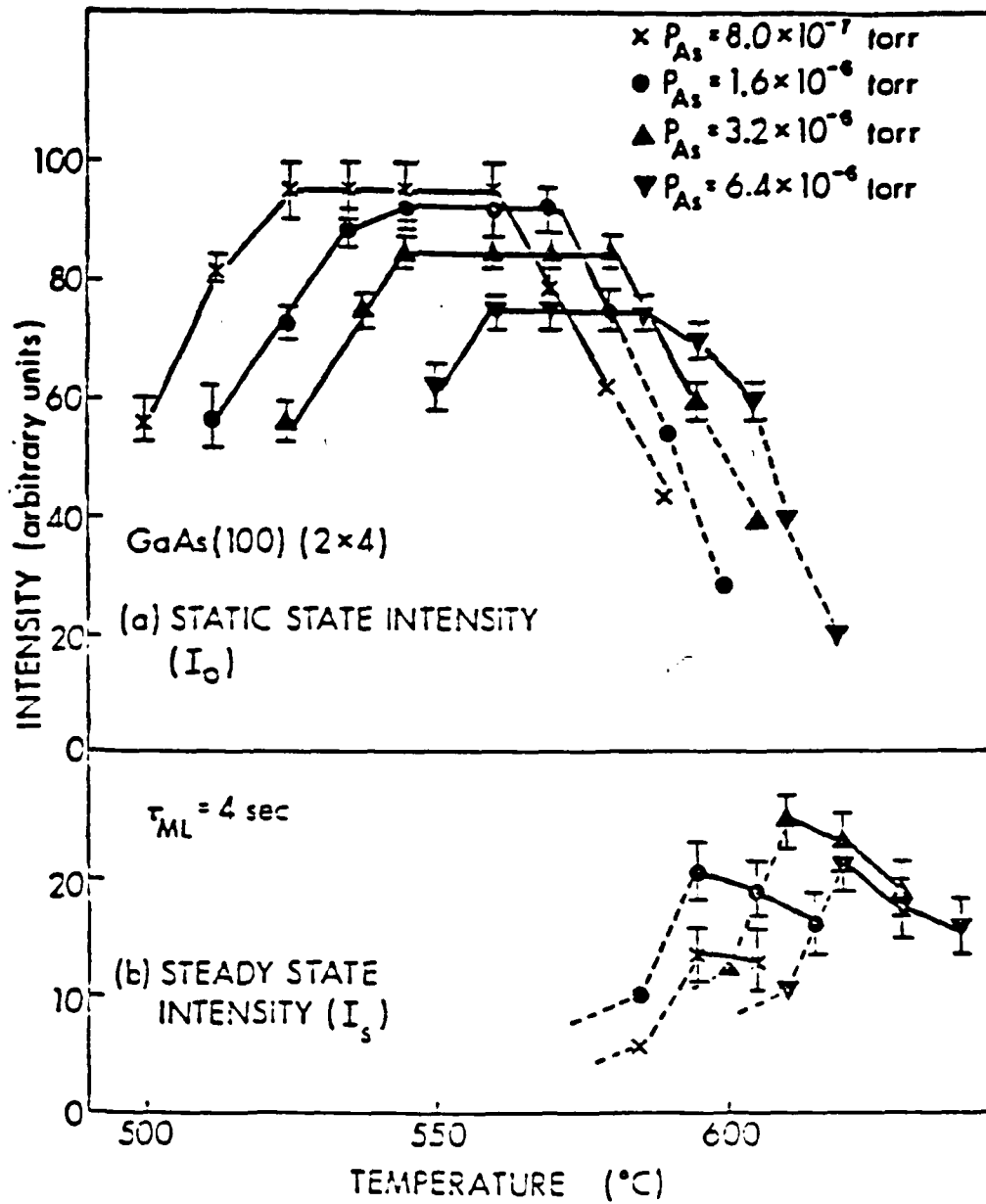


Fig. 3

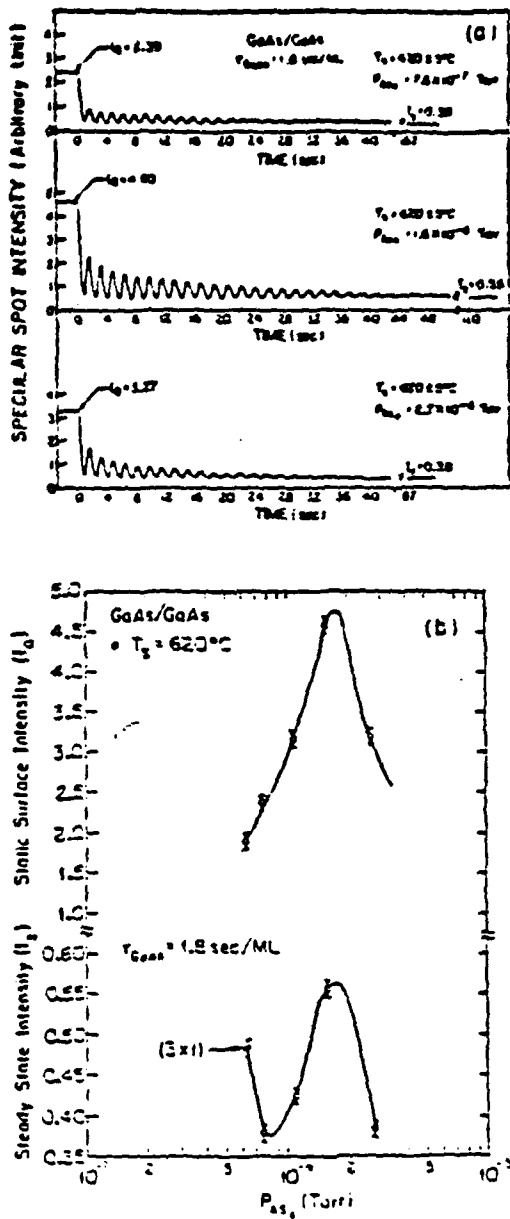


Fig. 4

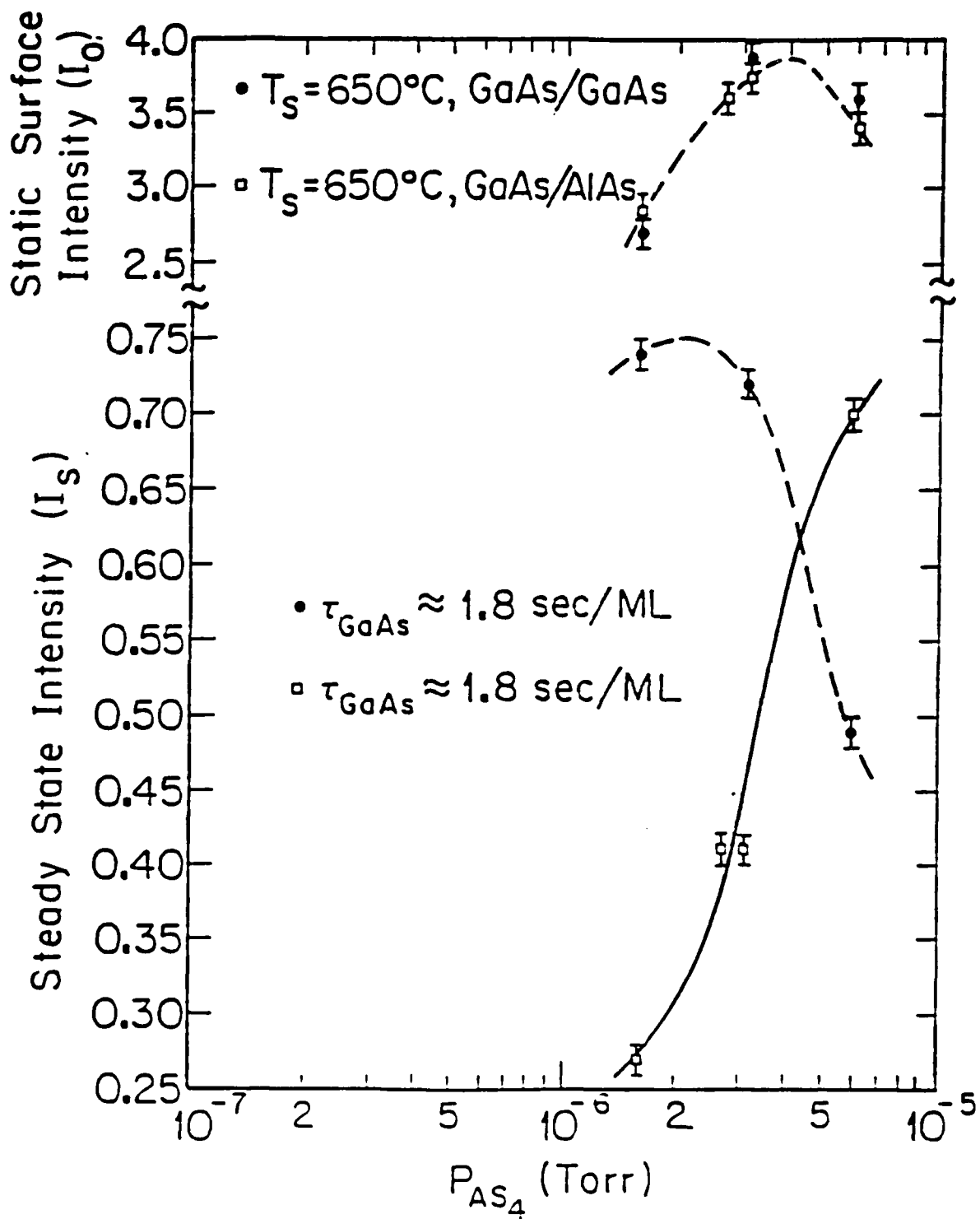


Fig. 5

An important feature, not reported in previous literature, is the existence of irreversible behaviour on the high T_s side, near the GaAs congruent temperature. The deterioration of I_0 with increasing T_s near the congruent temperature, as well as the lower values of I_0 upon subsequent decrease in T_s (i.e. lack of recovery of surface smoothness) can both be understood within the kinetics embodied in the CDRI model. The obvious pragmatic implication of the existence of bi-stability in the step density for growth of quantum well structures is that care has to be taken to ensure that growth is commenced on the smoother step density distribution. In Fig. 3 is shown the behaviour of I_0 as a function of T_s for various P_{As_4} . Note that with increasing P_{As_4} , the plateau region of the overall cap-like behaviour shifts to higher T_s and becomes narrower in range, consistent with the CDRI model. In Fig. 4 and fig. 5 we show examples of I_0 variation with P_{As_4} at two fixed T_s values--620°C and 650°C. The former is near the GaAs congruent temperature and the latter at higher T_s value. Note also that generally the highest electron mobilities have been reported for modulation doped GaAs/ $Al_{0.3}Ga_{0.7}As$ heterojunction grown near 620°C whereas, the highest photoluminescence efficiencies are reported for significantly higher temperature growth (650°C to 700°C). One notices the existence of an optimum As_4 pressure for realization of the smoothest static surface at a given T_s . Such surfaces are best suited for formation of highest quality GaAs/AlGaAs interfaces.⁴

B. THE STEADY STATE GROWTH FRONT

In figures 2, 4 and 5 is also shown the behaviour of the steady state intensity, I_s , corresponding to the static surfaces represented by I_0 in these figures. Let us first consider I_s behaviour as a function of T_s at a fixed As_4 pressure shown in fig. 2. One notices that the maximum in I_s occurs in the regime where I_0 has started dropping and is irreversible. An irreversible behaviour of I_s is also found, but unlike I_0 , this is found to occur in the low temperature regime where I_0 is reversible. The implication thus is that even though the starting static GaAs surface may not be of the highest smoothness near

the congruent temperature, the steady state growth front step density distribution is the smoothest in this temperature range. Thus if interfaces are formed by deposition of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ on a growing GaAs layer, as has been the customary practice in MBE, then the optimum temperature, for a fixed As_4 pressure, is the value at which maximum in I_s is found at the growth rate to be employed.

Correspondingly, the behaviour of I_s as a function of P_{As_4} at fixed T_s shown in figs. 4 and 5 reveals the existence of an optimum As_4 pressure. Note that, remarkably, while at 620°C and for a growth rate of 0.56 ML/sec. the optimum pressure for both static (i.e. I_0) and steady state (i.e. I_s) intensities occurs at essentially the same As_4 pressure (of about 1.8×10^{-6} Torr in our MBE machine), increasing T_s by 30°C to 650°C while maintaining the same growth rate of 0.56 ML/sec., gives rise to two different optimum As_4 pressures (fig. 5) for I_0 and I_s . Indeed, this is the more common occurrence, the coincidence of I_0 and I_s being true only for certain unique combinations of T_s , and growth rates. The underlying atomistic kinetic reasons for these behaviour is, once again, shown³ to be fully consistent with the CDRI model, though we do not discuss it here.

In Fig. 5 is also shown the I_s behaviour of AIAs grown on GaAs at the same growth rate of 0.56 ML/sec. as used for GaAs. Note that the optimum As_4 pressure for AIAs steady state growth front is somewhere beyond (but in our estimation near) 6×10^{-6} Torr. For a variety of practical reasons we cannot raise the As_4 pressure in our MBE system beyond about 6×10^{-6} Torr. It is thus clear that for the formation of GaAs on AlGaAs interfaces (the so-called inverted interface) under the customary practice of growth of one layer on the dynamic growth front of the previous layer, the optimal As_4 pressure during growth of AlGaAs is different from that for optimal static and steady state GaAs surfaces. However, there is no report of GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ structures grown with usage of such optimal and different As_4 pressures to achieve the highest quality normal and inverted interfaces. We

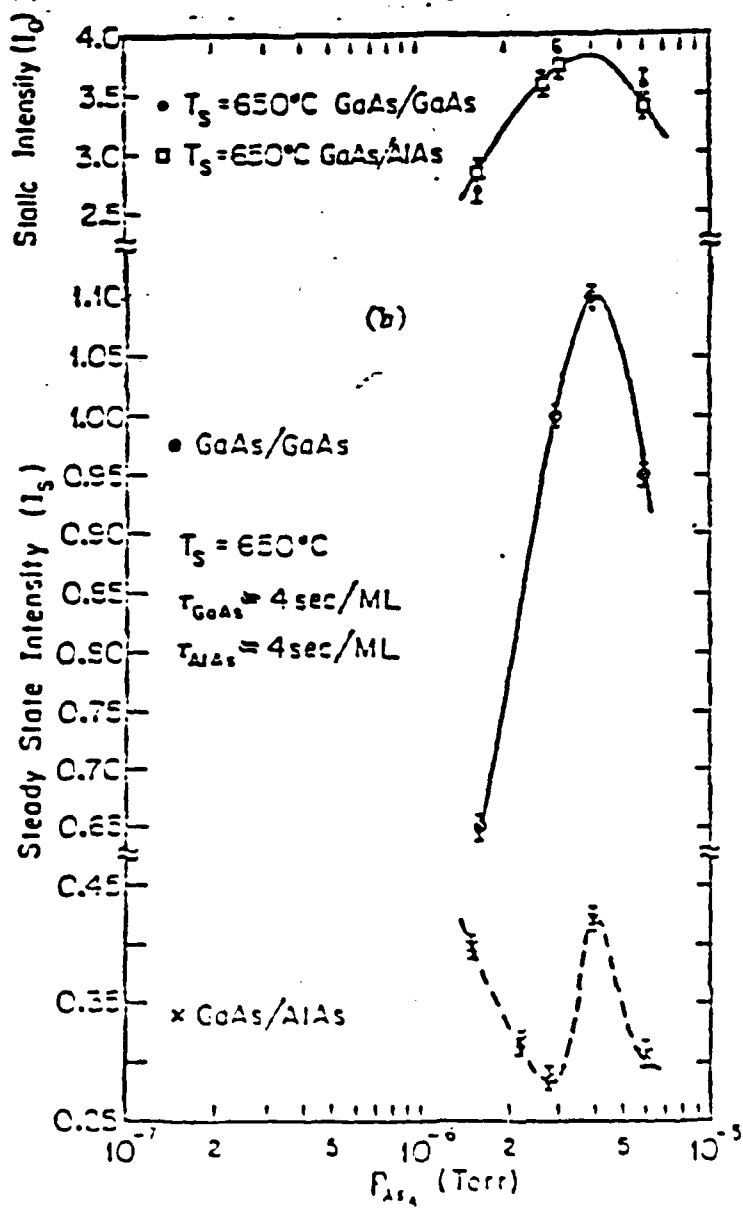
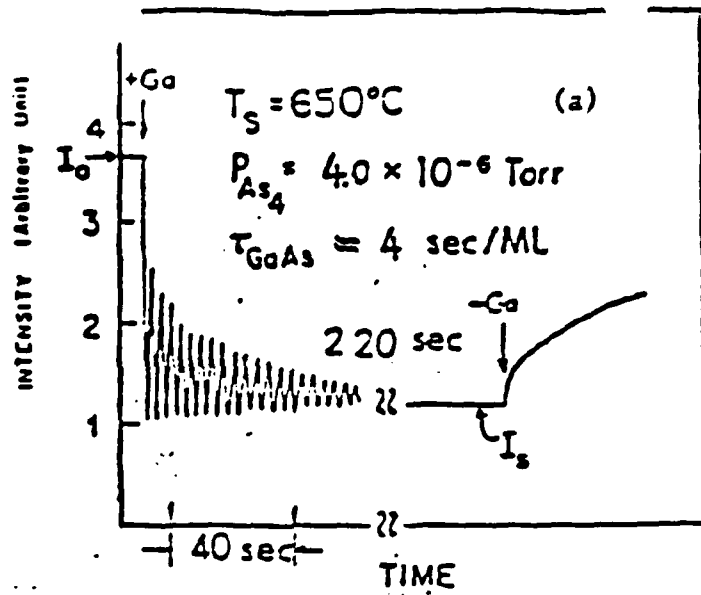


Fig. 6

have grown structures using two different As_4 pressures to achieve the highest quality normal and inverted interfaces. We have grown structures using two different As_4 pressures optimized for GaAs and AlAs growth during growth of GaAs/ Al_xGa_{1-x} As quantum well structures following the customary practice of formation of interfaces on dynamic growth fronts. To study optical and transport characteristics of such structures is our immediate objective and it is for the realization of this objective that funds are sought under this proposal.

Finally, in fig. 6 we show an example of a very unique set of growth conditions for which the maximum in I_0 of GaAs (i.e. static GaAs surface) and I_s of both GaAs and AlAs occur at the same pressure of 4×10^{-6} Torr. Note that the substrate temperature is $650^\circ C$, same as before, the only difference being that the growth rate is reduced to 0.26ML/sec., just about half of before. Such a growth condition would appear to be uniquely optimum for growth of GaAs/AlAs superlattice structures.

C. GROWTH INTERRUPTION AND INTENSITY RECOVERY

The intensity recovery behaviour upon termination of GaAs growth (on GaAs(100) $As(2 \times 4)$ surface) in the steady state was first reported¹² by Neave et al. who found it to be well fit by a sum of two exponentials--one with a short time constant (τ_1) representing an initial fast recovery and another with a long time constant (τ_2) representing subsequent slow recovery. A systematic study of this recovery behaviour for growth termination at various integral and non-integral monolayer depositions as well as in the steady state was first taken up by Lewis et al.⁸ In addition, the As_4 pressure and substrate temperature dependence of the recovery behaviour for growth termination at various stages was examined in these collaborative studies^{8,9} with our group. The sum-of-two-exponential behaviour was found to be a reasonable description for most cases. The behaviour of short and long time constants as a function of amount of material deposited at the time

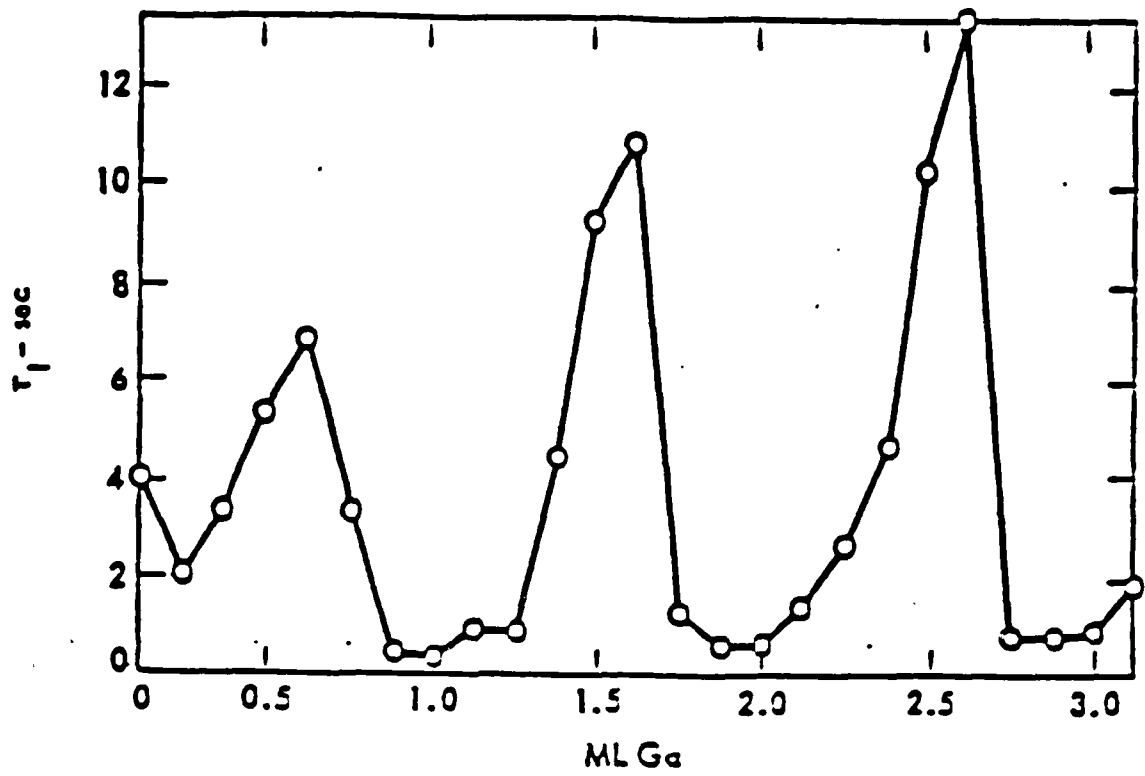
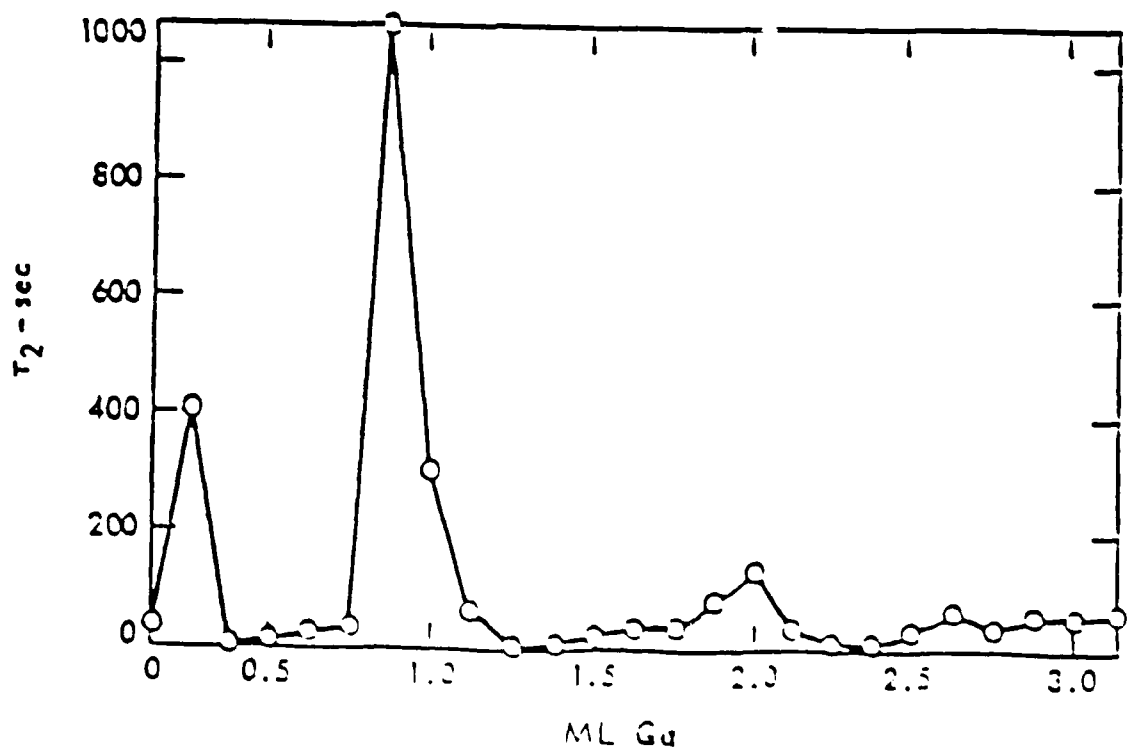


Fig. 7



of growth termination is shown in fig. 7. The temperature, P_{As_4} and growth rate dependence of τ_1 and τ_2 , when coupled with the CDRI model, provides considerable insight into the kinetic processes operative during the relaxation of the growth front step density towards the smoother step density of the static surface (the intensity during growth is generally found to be lower than that for the starting static surface for well prepared surfaces). It has been suggested^{8,12} that the initial fast recovery is primarily a manifestation of the high mobility of isolated or low-coordination Ga atoms at the time of growth interruption whereas the subsequent slower recovery stage reflects the slower kinetics of coordinated movement of several atoms involved in rearrangement of the step density distribution. The matter in our view¹³ is more involved than simply relying upon the migration kinetics of group III atoms or group of atoms, but we do not discuss this issue any further here.

The pragmatic consequence of the growth interruption and intensity recovery behaviour is related to the formation of interfaces. It is clear that interruption of growth allows the growth front to relax towards a smoother step density configuration and interfaces formed on such relaxing or fully relaxed (i.e. static) surfaces are likely to show higher structural and chemical perfection^{1,11} as compared to the customary practice of interface formation on dynamic growth fronts (an example of this customary practice is shown in fig. 8). An example of such a behaviour is shown in fig. 9 for the formation of inverted GaAs/ $Al_xGa_{1-x}As$ interface. Curve (a) of fig. 9 shows the behaviour when the $Al_{0.2}Ga_{0.8}As$ layer is allowed to recover fully (i.e. a static $Al_{0.2}Ga_{0.8}As$ surface has been achieved) before commencement of GaAs growth. Note that the substrate temperature used is $550^\circ C$, somewhat on the low side, and the growth rate is $0.25 ML/sec.$ considerably slower than the customary $ML/sec.$ The low T_s value of $550^\circ C$ leads to the fairly long recovery time of some 190 secs. (at the As_4 pressure employed). Upon commencement of GaAs growth, the relative intensities of the first couple of maxima are

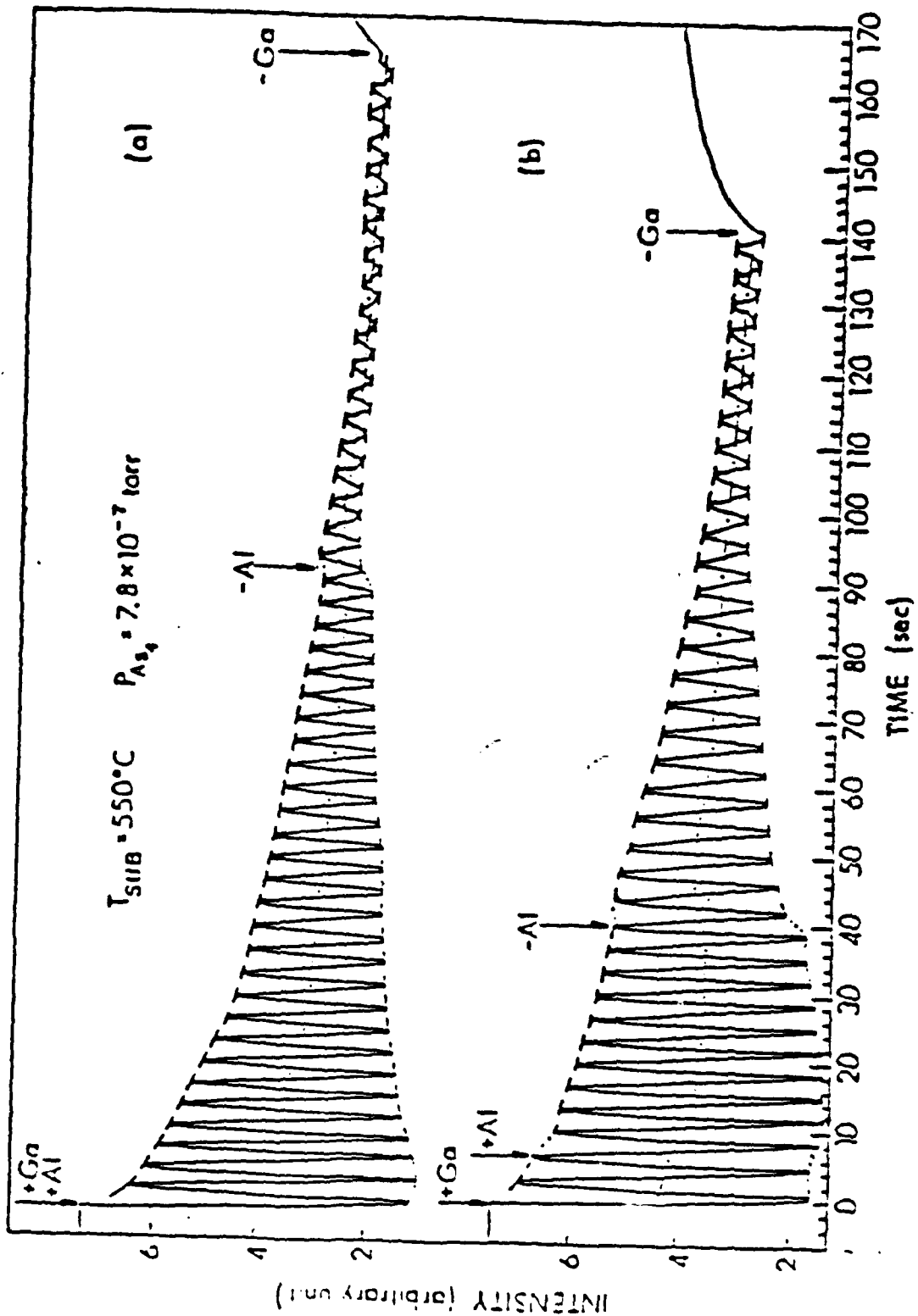


FIG. 8

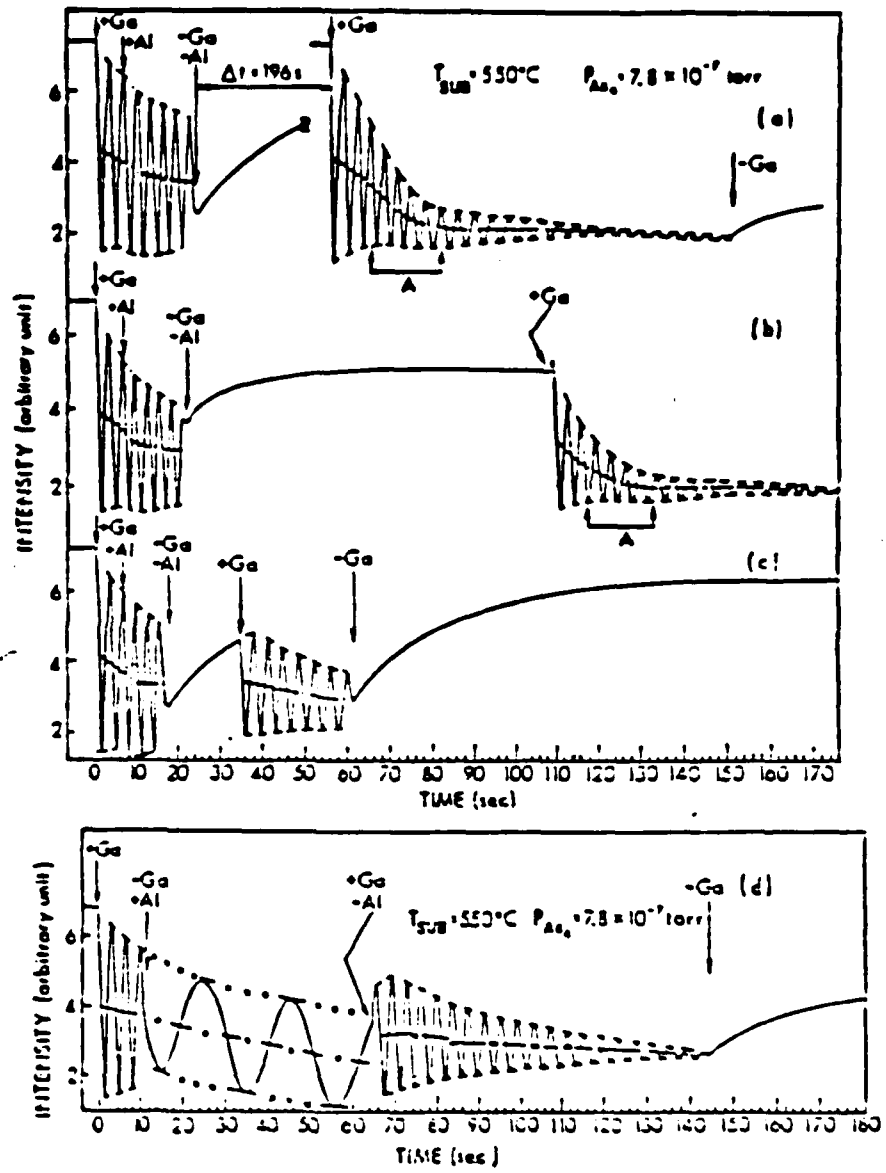


Fig. 9

GaAs/AlGaAs GROWTH AT 640°C
— RHEED OSCILLATION BEHAVIOR —

$$P_{As} = 3 \times 10^{-6} \text{ Torr}$$
$$\tau_{ML}(\text{GaAs}) = 4.7 \text{ sec.} \quad \tau_{ML}(\text{AlGaAs}) = 3.7 \text{ sec.}$$
$$x = 20\%$$

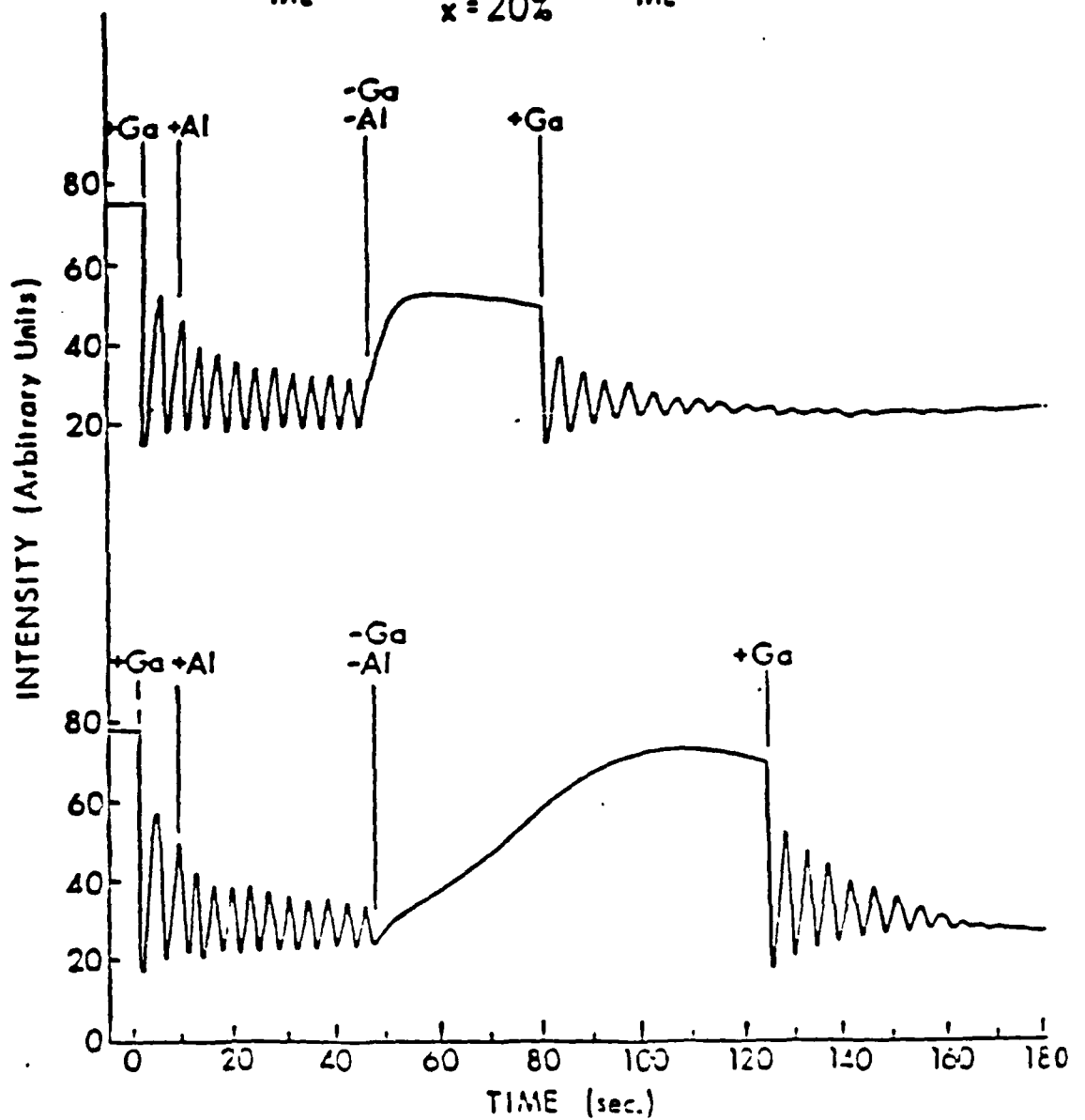


Fig. 10

comparable to that for $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ growth on GaAs (i.e. the normal interface) indicating that the inverted interface can be comparably good to the normal interface. One notes, however, that the rate of decay of the GaAs oscillation maxima is much faster than that for $\text{Al}_{0.2}\text{Ga}_{0.3}\text{As}$ deposition on GaAs (or GaAs deposition on GaAs). The steady state GaAs value, nevertheless, is essentially the same as for GaAs deposition on GaAs. We believe both these features to be explicable in terms of impurity incorporation on the $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ surface during the time of growth interruption. Impurities gathered by Al can act as heterogeneous nucleation centers, thus creating a rougher surface morphology upon commencement of GaAs growth and cause rapid decay in the oscillations. However, after 10-15 ML deposition the impurities buried at the interface have no significant influence on the morphology of continued GaAs growth on GaAs and thus the steady state intensity reaches the same value it does for no growth interruption.

The danger of background impurity incorporation during interruption of growth to achieve higher structural and chemical integrity of the interface thus suggests the need for optimizing the growth interruption time. One may simply shorten the growth interruption time, as in curve (b) of fig. 9 or achieve the same effect by employing higher substrate temperature to hasten the recovery kinetics, as shown in fig.10.

D. IMPLICATIONS FOR GROWTH OF MULTIPLE QUANTUM WELLS

The implications of even the few results of our findings of the RHEED intensity dynamics presented here are numerous, some of which we have noted in the preceding. Here we merely note two examples of the notion of growth interruption which was motivated by our CDRI model and the observation that I_s is generally lower than I_o .

The first example is also the first usage⁶ of the growth interruption idea and relates to the GaAs/ $\text{In}_x\text{Ga}_{1-x}\text{As}$ strained layer system. Based upon considerations of optimization between the kinetic rates embodied in the CDRI model and the desirability of allowing as

Fig. 11(a)

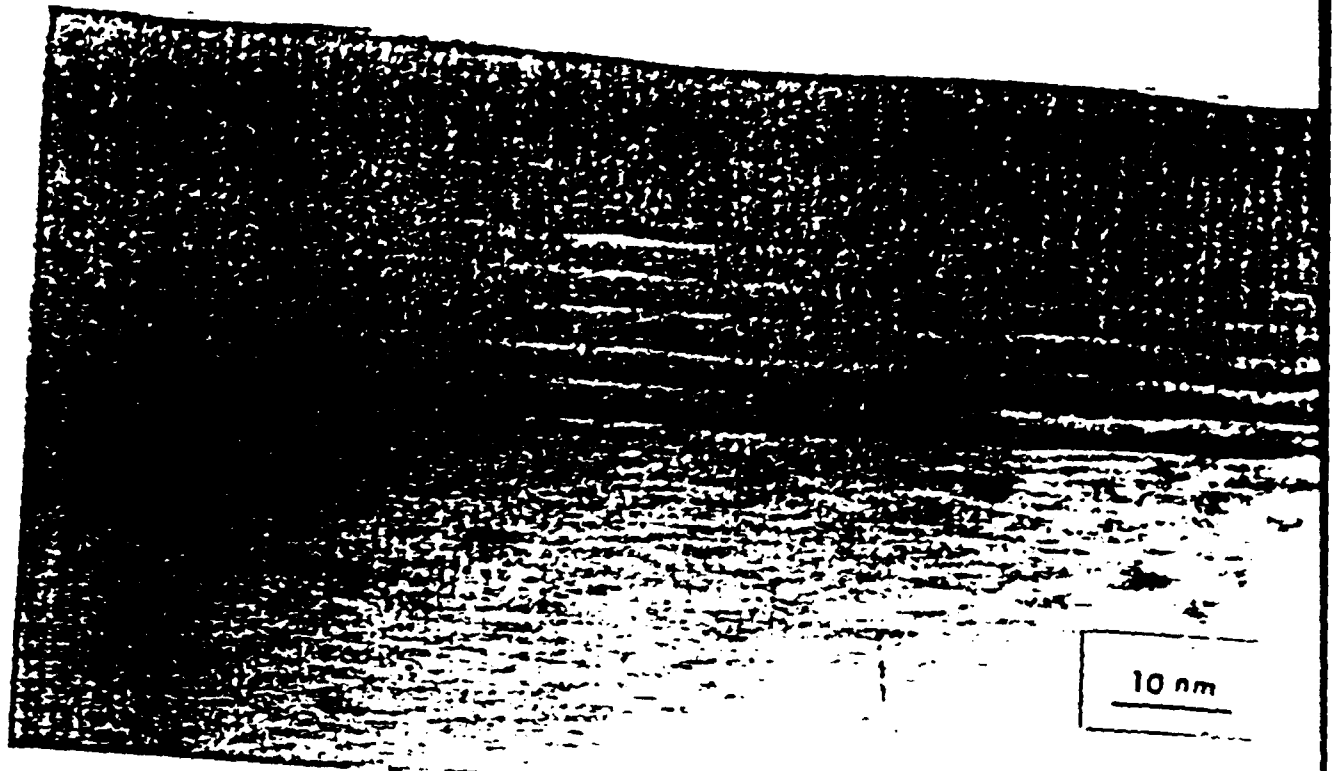
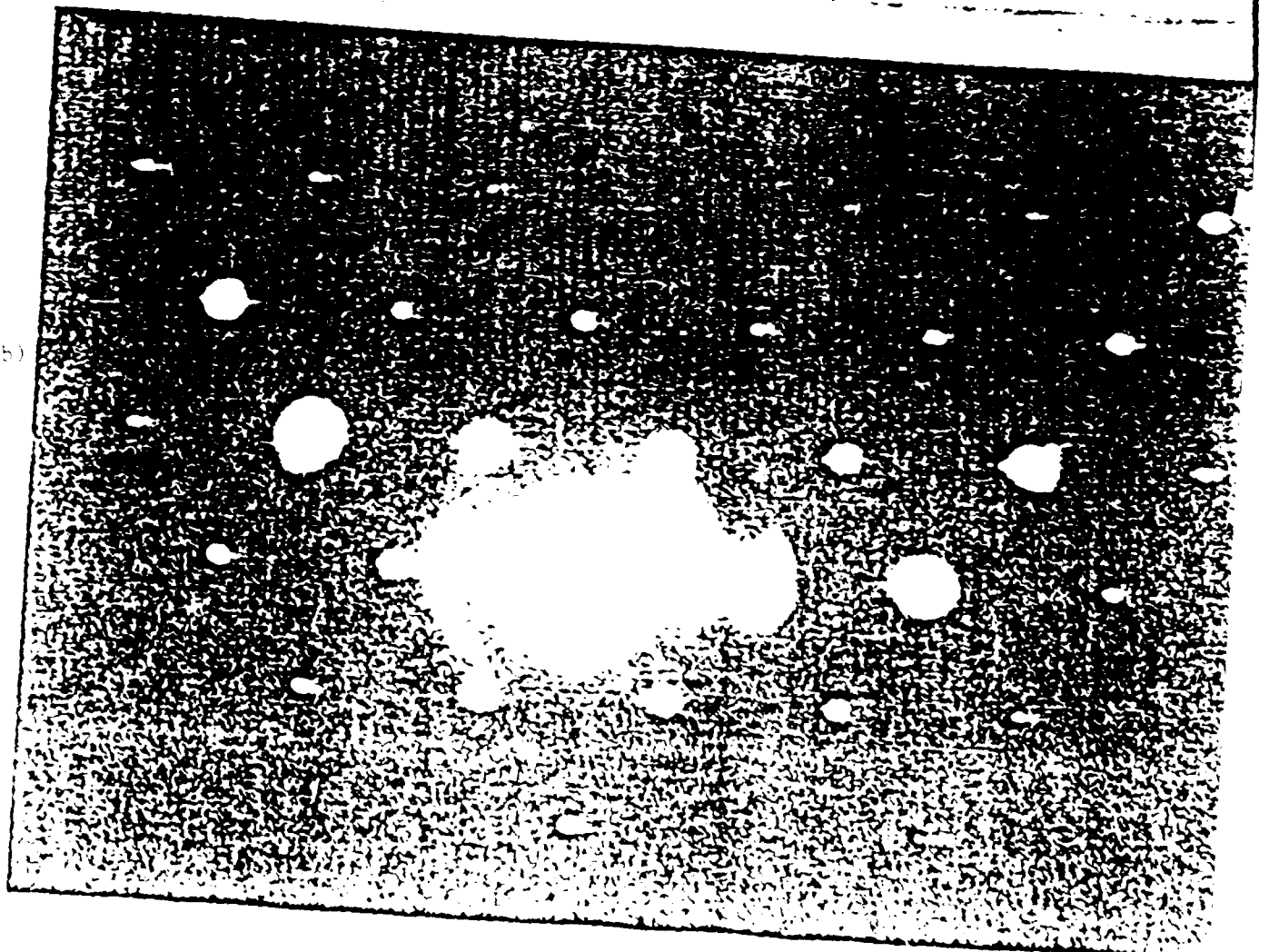


Fig. 11(b)



complete a thermodynamic relaxation of the lattice strain arising from the lattice mismatch as possible, we grew GaAs/InAs multiple quantum well structures involving 7% lattice mismatch directly on GaAs substrate employing growth interruption. The results of this first sample grown in October 1983 are shown in the XTEM image contrast photograph shown in fig. 11a, along with the diffraction condition employed (fig. 11b). The imaging was performed on the USC Philips 420T microscope using the (0,0,0) and (2,0,0) two-beam conditions. Note the remarkably defect free nature of the interfaces in the MQW. Details of XTEM results may be found in references (6) and (10). A variety of GaAs/InAs MQW and superlattice structures have been grown in collaboration with the JPL group on their Riber1000-2 MBE system and XTEM studies carried out at USC. Some of these results may be found in reference (14).

The second example pertains to GaAs/AlAs superlattices involving ultrathin layers grown under the uniquely optimum conditions indicated by the behaviour of static GaAs and steady state GaAs and AlAs intensities shown in Fig. (6) and discussed in section (C). In fig. 12 we show the behaviour of the specular beam intensity during growth of $(\text{GaAs})_2/(\text{AlAs})_2$ and $(\text{GaAs})_6/(\text{AlAs})_6$ superlattices.² Curves (a) and (c) correspond to the customary practice of growth on dynamic growth fronts, whereas curves (b) and (d) show the effect of interruption of growth after AlAs and GaAs layer depositions. Several remarkable features are to be noted.

(i) for the $(\text{GaAs})_2/(\text{AlAs})_2$ superlattice with "dynamic" normal and inverted interfaces (curve a), the inverted interfaces are seen to be of poorer quality than the normal interfaces--a consequence of the slower migration of Al compared to Ga.

(ii) With growth interruption, both AlAs and GaAs growth fronts recover (the latter to the starting intensity of the static GaAs substrate surface) and consequently the normal and

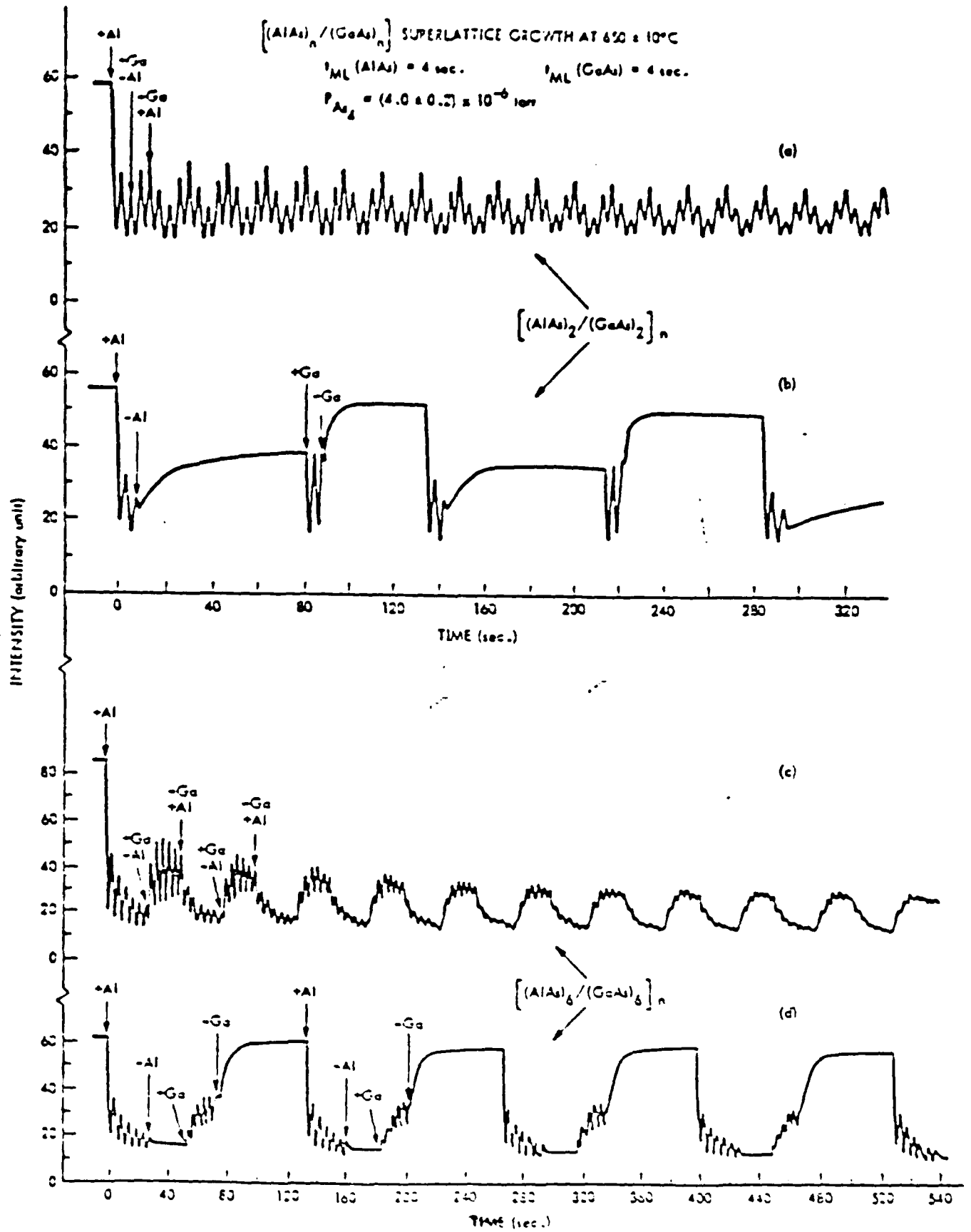


Fig. 12

inverted interfaces are significantly better. Indeed, given the optimum conditions employed, they are expected to be the best for growth at 650°C with a growth rate of 0.26ML/sec. Note that the recovery of the AIAs surface is slower, once again manifesting the slower kinetics of Al migration as well as As₄ adsorption-desorption kinetics of AIAs.

(iii) For the dynamic growth of (GaAs)₆/(AIAs)₆ (curve C), in contrast to the (GaAs)₂/(AIAs)₂ (curve a), the overall amplitude of oscillations continues to decay. Thus, the quality of both normal and inverted dynamic interfaces gets progressively worse with growth of each subsequent quantum well. This, in spite of the optimum growth conditions. It is a consequence of the rougher growth front of the very first 6 ML AIAs layer propagating its adverse influence to the formation of each subsequent interface, until a steady state in which the effective migration length, λ_{eff} (Ga or Al), larger than the average terrace width, $W(\text{GaAs or AIAs})$, is reached.

The net consequence is that, even for such thin individual layers grown under optimum conditions, the overall optical and transport properties of the superlattice are going to be dominated by the majority of the fairly degraded quantum wells, rather than the first few which may be of acceptable quality. It is thus to be recognized that optical properties of a structure involving a very few quantum wells are generally going to be better than of another grown under identical conditions but involving growth to a much larger total superlattice thickness.

(iv) Finally, in curve (d) it is seen that growth interruption at a 6ML AIAs thickness does not show any detectable intensity recovery on a similar time scale. It is more difficult for Al with slow migration rate to smoothen the rougher growth front left behind by a 6ML thick AlAs layer than for the 2ML thick layer. AIAs growth interruption for a reasonable and practicable duration thus does not help improve the inverted interface, in spite of the

optimum growth conditions, even for such thin layers. By contrast, interruption of GaAs growth nevertheless allows even the 6ML growth front step density to recover rapidly to the smoothest starting GaAs surface--a consequence of the fast Ga migration and As_4 desorption kinetics. The net consequence is that the 6ML thick GaAs is able to heal even the rough growth front of 6ML AIAs, as if it were a buffer layer, resulting in the formation of normal interfaces of the same high quality as the first interface. Equally, each subsequent inverted interface then maintains the same quality as the first one, even though it is not as good as the normal interface. Thus, though limited, but nevertheless significant advantages accrue from GaAs growth interruption in that it prevents propagation of the adverse influence of the rougher growth front of AIAs, thereby preventing progressive deterioration of the quality of each subsequent quantum well in a superlattice of reasonable thickness.

A variety of new and significant findings on both the fundamental kinetics of MBE growth and its consequences for optimization of growth conditions have emerged from our work. It is hoped that the few examples provided here have served sufficiently well, the intended purpose--to illustrate the richness of the subject, the potential for using such information for growth of quantum well structures and the need for study of the optical and transport properties of such structures. Indeed some very interesting innovations on the MBE growth technique itself can be combined with the RHEED studies to create not only improved quality structures of systems such as GaAs/ $Al_xGa_{1-x}As$, but even create new and novel structures made of new metastable phases of materials. One example of such an investigation currently underway in the author's group, is the use of coherent and incoherent light to controllably influence the MBE growth kinetics with a view towards realizing this objective.

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