AD-A156 325 INVESTIGATIONS OF THE MOLECULAR BEAM EPITAXIAL GROWTH AND CHARACTERISTICS..(U) UNIVERSITY OF SOUTHERN CALIFORNIA LOS ANGELES DEPT OF MATERIA.. A MADHUKAR 1/1

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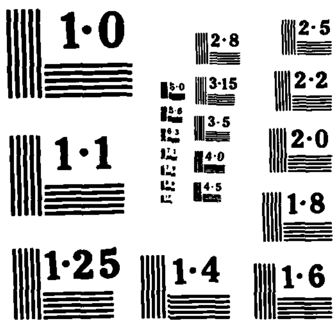

NATIONAL BUREAU OF STANDARDS
MICROCOPY RESOLUTION TEST CHART

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Principal Investigator, A. Madhukar

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Molecular Beam Epitaxial Growth [illegible] Characteristics of External Excitation Induced Non-Equilibrium Phases of Immiscible III-V Compound Semi[illegible]

19. ABSTRACT *(Continue on reverse if necessary and identify by block number)*

This annual report describes progress made during the per[illegible] through Oct. 31,1984 on theoretical and experimental studies of the kinetics and [illegible] molecular beam epitaxial (MBE) growth of III-V semiconductor structures. Spe[illegible] gress is reported on (i) Monte-Carlo computer simulations of MBE growth and [illegible] dynamics of reflection-high-energy-electron-diffraction (RHEED) intensities [illegible] laser-induced-desorption (iii) Experimental studies of the RHEED intens[illegible] GaAs $In_xGa_{1-x}As$(100) MBE growth (iv) the first realization of GaAs InAs strained [illegible] structures with [illegible] lattice mismatch) and transmission electron microscopy studies show[illegible] interfaces, and (v) establishment of a facility for magneto-absorpt[illegible] studies

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ANNUAL REPORT

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REPORTING PERIOD: Nov. 1, 1983 -- Oct. 31, 1984

Investigations of the Molecular Beam Epitaxial Growth and Characteristics of External Excitation Induced Non-Equilbrium Phases of Immiscible III-V Compound Semiconductors

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Description of Work:

This program undertakes experimental and theoretical investigations of the role of growth kinetics and mechanism(s) in molecular beam epitaxial growth of III-V semiconductors and its possible control via laser excitation to effect metastable structures and phases. The experimental work on laser induced MBE growth is collaboratively carried out, under the principal responsibility of Dr. F. J. Grunthaner at the Jet Propulsion Laboratory, Cal Tech, through a subcontract from USC.

Progress: Several significant steps on both the theoretical and experimental fronts were accomplished during this reporting period. The status of the latter has now been brought to the stage where the very first experiments on the influence of photons (using a CO_2 laser) on the growth front during GaAs(100) homoepitaxy are about to begin. In the following we provide a brief description of the highlights.

A. THEORETICAL WORK

A.1 Computer Simulations of MBE Growth:

Since the acquisition of the VAX 11/750 computer system under the present contract, we have been able to bring to fruition many of our ideas relating to the MBE growth kinetics via large scale and realistic computer simulations employing the Monte-Carlo methods.

Deriving from the limited information on the group V adsorption kinetics provided by the very early work of Arthur[1] and of Foxon and Joyce,[2] we were able to significantly improve upon the physics and chemistry of earlier computer simulations[3,4] and identify the competition between the group III migration kinetics and the surface molecular reaction kinetics of the group V molecules as being the essential kinetics controlling the structural and chemical nature of the MBE growth front, and consequently of the interfaces formed between materials under the usual growth conditions and customary practices. This work has led to the identification of the III-V MBE growth as a configuration-dependent-reactive-incorporation[5,6] (CRDI) growth process which is quite distinct from the conventional notions of either 2-dimensional nucleation and growth or continuous growth invoked so far to understand MBE growth. Under appropriate growth conditions (which are not the ones normally employed during III-V MBE) the CDRI growth mechanism reduces the the conventional nucleation or continuous growth mechanisms.

The CDRI growth process based computer simulations[5,6] permitted not only a direct examination of the role of various competing surface kinetics in defining the time evolution of the growth front morphology (step density distribution, terrace width distribution, etc.) but also allowed a direct calculation of the time-dependent behaviour of the various reflected electron beam intensities as measured in the reflection-high-energy-electron-diffraction (RHEED) experiments (see Sec. B) during MBE growth. This ability of the computer simulations to predict the growth condition dependence of a real-time, in-situ measureable quantity allows RHEED intensity dynamics to be exploited as both, a basic research tool providing direct information on atomic kinetic processes at the surface, as well s a pragmatic tool for optimising the growth conditions for the realization of high quality interfaces in device structures. The most significant predictions[5-7] of the CDRI growth based simulations (whose experimental confirmation is discussed in Sec. B.) during this reporting period are;

1. the striking sensitivity of the growth front morphology to the group V overpressure employed during MBE growth -- a fact hitherto unappreciated, but of great significance to the quality of the interfaces and thin films.

2. The clear delineation of the role of kinetics versus thermodynamics in controlling interfacial characteristics as exemplified by the notion of "growth interruption" that follows from the CDRI model.

3. The role of thermodynamic accomodation of lattice-mismatch, ($\Delta a/a$), induced strain energy in a homogeneous fashion (i.e. without generation of misfit dislocations) via growth interruption during heteroepitaxy of highly lattice mismatched systems (such as GaAs/InAs with $(\Delta a/a) \sim 7.4\%$ to be discussed in Section B).

4. Predictions of the growth conditions desirable for achieving high room temperature photoluminescence efficiency in systems such as $GaAs/Al_xGa_{1-x}As(100)$.

(A.2) Laser Induced Desorption and Surface Migration:

A theory of laser induced desorption has been developed employing a classical stochastic approach. The objective is to develop a theoretical formalism capable of describing the influence of a laser, coupled to the vibrational modes of the adsorbate-surface bond, in enhancing the desorption rate of the adsorbate over and above that corresponding to the substrate temperature employed. The physical basis of the formalism derives from the classical, activation-over-the-barrier nature of desorption and surface migration processes, and consequently it employs a description of the motion of the adsorbate normal to the surface in terms of a starting Langevin equation. The Langevin equation incorporates the influence of the surface, modelled as a thermal bath, through a random force acting on the adsorbate-surface atom(s) bond(s), a friction term representing the dissipation of the adsorbate energy back to the surface (i.e. the thermal bath) and a coherent force representing the coupling of the laser to the dipole moment of the adsorbate-surface atom(s) bond(s). The explicit dependence of the motion of the adsorbate on the spatial coordinates of the surface atom(s) to which it is considered directly bonded is then eliminated, leading to a Fokker-Planck equation whose solution gives the probability, $W(t,z,u)$, of finding the adsorbate a given normal distance z away from the surface with a velocity u at a time t. Defining a criteria for escape of the adsorbate in terms of $z>z_c$, and $u>u_c$ where z_c and u_c are critical distance and velocity, respectively, we calculate the average desorption rate over a period of oscillation of the laser electric field.

The results, in the absence of the laser, can be obtained analytically and reduce to the well known Arrhenius form derived previously.[8] In the presence of the laser, the complicated nature of the mathematics has not allowed us to find an exact, closed form, expression for the desorption rate, even though certain approximate analytic expressions can be extracted in certain limiting cases. We have, therefore, obtained the results via numerical solutions to obtain a reliable view of the role of the laser characteristics, in relationship to the surface-adsorbate bond(s), in influencing the desorption rate. While these results are to be published, in Fig. 1 we show an illustrative example for the case where the laser frequency is near resonance with the adsorbate-surface bond vibrational frequency. A variety of other cases are presently under investigation. An attempt to develop a theory for laser induced surface migration along similar lines is underway.

<u>(B) EXPERIMENTAL WORK:</u>

(B.1) RHEED Intensity Dynamics Studies:

In an effort to monitor the influence of lasers on the surface kinetic processes, we undertook first a systematic study of the RHEED pattern and intensity behaviour during growth of GaAs/$In_xGa_{1-x}As(100)$ interfaces. Such studies, on one hand, provide the reference information needed to compare with the behaviour under laser excitation, and on the other, form the basis of a reasonable atomistic understanding of the MBE growth kinetics when combined with the CDRI model based computer simulations discussed in Section (A.1). Note that the $x \neq o$ heteroepitaxy studies, when contrasted with the $x=o$ GaAs homoepitaxy studies, provide a means of investigating the role of lattice mismatch induced strain in influencing the growth kinetics and mechanism(s) controlling the creation of high-interfacial quality metastable structures between GaAs and $In_xGa_{1-x}As(100)$.

Amongst a variety of RHEED intensity studies undertaken during this reporting period, the highlights are;

(i) A systematic study of the As_4 pressure dependence of the GaAs(100) growth front morphology at various substrate temperatures, on the 2-fold versus 4-fold axis of the usually employed (2 X 4) As-stabilized surface reconstruction, at different Bragg and out-of-phase diffraction conditions, and for different growth rates. The observations have been found to be consistent with the predictions of the CDRI growth model based computer simulations and provide an important data base for analysing in detail the nature of the surface kinetic processes. As an example, our previously reported[9] two-time-scale exponential behaviour of the specular beam intensity recovery upon termination of growth is shown by these results to involve an initial fast recovery time which increases and then decreases with increasing As_4 pressure while the subsequent slower recovery time increases monotonically. The former, suggested to reflect atomic migration, thus appears to behave in a manner similar[7,10] to the damping of the oscillation amplitude during growth which requires an optimum pressure for the best oscillations, whereas the latter, suggested to represent step rearrangements, does not. While at face value this observation is qualitatively consistent with the CDRI model, it presents new challenging information for the computer simulations, as was hoped for in this interactive theory-experiment investigations of the role of kinetic processes in MBE growth.

(ii) Observation[11] of As_4 pressure controlled RHEED intensity oscillation period and amplitude under conditions where a few monolayers (3 to 30) of Ga are deposited first in the absence of the As_4 and then As_4 flux is turned on to react with the Ga metallic layer. The RHEED oscillation period is found to be inversely proportional to the As_4 pressure and provides information on the rate of As_4 incorporation. The very existence of the oscillations indicates an essentially layer-by-layer mechanism of the As_4 reaction with the

Ga layer to form GaAs. A variety of data reflecting different substrate temperatures, etc. have been obtained and are presently being analyzed. Preliminary examination suggests that the data might be capable of providing an activation energy for the As_4 reaction rate with Ga.

(iii) As_4 induced RHEED intensity oscillations on In layers deposited upon GaAs(100). In contrast to the Ga overlayer case, upon opening of the As_4 flux an island growth is suggested by the resulting RHEED pattern during formation of InAs. These observations suggest that the surface tension of the In and Ga films as deposited on the GaAs(100) surface strongly affect the subsequent growth. Invoking the essential role of surface kinetic processes at the base of the CDRI model, the competition between surface migration, reactive incorporation and island formation for strain relief would suggest that a high effective temperature of the surface atoms, together with a lower substrate temperature might be required for such growth of metastable structures -- a situation one might potentially achieve via the use of appropriate laser energy.

(B.2) <u>Laser-Induced MBE:</u>

From the point of view of the work on laser influence on MBE growth, the first half of this reporting period has unfortunately been a frustrating one. During the installation phase of the laser additions to the MBE system several difficulties with both the CO_2 and the excimer systems were encountered which may be traced to damage during the shipment to JPL from the vendor, Tachisto Laser Systems. Resolution of the difficulties took up the better part of the first half of this reporting period. By the end of May 1984, the repaired laser systems were delivered and tested to meet the specifications. Mounting table geometries were designed, the necessary window materials were delivered and the window mounting system tested. Considerable effort followed on the issue of coupling laser light in and safely out of the MBE system. During this process we were, regrettably, led to conclude that two cell ports must be used for this purpose so that excess laser energy can be safely dumped by specular reflection. We have now reached the stage where the first calibration experiments involving a CO_2 laser are about to begin.

(B.3) <u>Growth of GaAs/InAs(100) strained layer superlattices:</u>

Guided by the role of growth kinetics as exemplified in the CDRI model based computer simulations and the RHEED intensity observations on deposition of submonolayers and integral monolayers of InAs on GaAs(100), we were motivated to employ the notion of growth interruption as a means of allowing the growing overlayer sufficient opportunity to homogeneously accomodate the strain induced by the 7.4% lattice mismatch between GaAs and InAs. A variety of multiple quantum wells (MQW) and superlattice structures were grown allowing growth interruption up to one and two minutes after each sequential deposition of InAs and GaAs, respectively. Note that these MQW structures are grown directly on a GaAs buffer layer, and not on an $In_xGa_{1-x}As$ alloy buffer layer at a composition corresponding to the average lattice constant of the layered structure as has been attempted in the past. In the present structures then, the strain alternates between compressive and tensile for InAs and GaAs, respectively. It is perhaps also worth noting that the successful realization of strained layer structures reported to date is restricted to a lattice mismatch less than 1.5%, corresponding to about 30% In in InGaAs.

(B.4) **Transmission Electron Microscopy:**

The GaAs/InAs(100) multilayered structures grown with the InAs and GaAs individual layer thickness up to 30 monolayers (i.e. ~80°A) were examined *via* high resolution cross sectional transmission electron microscopy employing a Philips 420 T analytical microscope at the USC Center for Electron Microscopy and Microanalysis (CEMMA). The microscope was operated at 120 keV with a line resolution of 2°A. The (0,0,0) and (2,0,0) beams were employed to obtain bright and dark field transmission electron image contrast of the GaAs and InAs layers. The corresponding diffraction patterns were taken along the <110> direction to obtain satellite spots as an independent means of checking the integrity of the MQW structures.

The TEM results revealed remarkably well defined interfaces for a system involving 7.4% lattice mismatch. The unit cell thickness (i.e. the sum of a GaAs and InAs layer thickness) extracted from the image contrast studies on one of the samples was 35.2°A while the corresponding value extracted from the satellite spots in the diffraction pattern was 34.3°A, in good agreement. This sample was grown with 4 monolayers of InAs alternating with 8 monolayers of GaAs. The experimentally determined unit cell thickness is then found to be in remarkable agreement with a thickness of 35.59°A estimated on the basis of the intended growth thickness combined with an estimate of the tetragonal distortion along the growth direction induced by the lattice mismatch. Surprisingly, the estimate of the elongation of the InAs lattice constant in the growth direction is based upon a continuum theory and the elastic constants of bulk InAs and yet gives such good agreement with the experimental values.

The interfaces exhibit remarkably low misfit dislocation density. A certain waviness, estimated to be on the scale of a monolayer, is however, clearly present and may, to some extent, be a consequence of a 10% type of uncertainty in the delivery on In and Ga monolayers inherently present due to the typical 0.1 second opening and closing times of the cell shutters. The compositional quality of the interfaces is difficult to assess in electron microscopy since image contrast is very sensitive to imaging conditions, TEM specimen thickness and surfrace uniformity, contamination from oxide formation, etc. We are, however, presently looking at ways of obtaining some information on this aspect.

(B.5) Optical and Magneto-Optical Absorption:

Although the intent of the original proposal was to establish a far from infra-red magneto-optical absorption capability at JPL in the third year of the contract, it was decided that given the immediate need for information on the electron structure of the grown GaAs/In_xGa_{1-x}As MQW structures combined with the limited man power at JPL it was necessary to set up such a facility at USC. Consequently, during this reporting period considerable additional effort had to be mounted at USC towards the [preparation work necessary for the] establishment of such a capability. This included a six-month rigorous training period of a graduate student and appropriate survey of experimental literature for considerations of instrument design, including the requirements imposed by the desired physics of the single and multiple interface structures. Fortunately, some capital equipment money at USC could be made available from sources other than the present contract (which provides for monies for an optics/magneto-optics system during the present budget year, Dec. 1, 1984--Nov. 30, 1985) so that the initial phase of procurement of a limited system for optical absorption could be begun during the second year (i.e. this reporting period). The basic and immediate need for such a system was demanded by the structures grown even in the absence of laser excitation, such as the GaAs/InAs multiple quantum well structures on which TEM structural studies were done (in Section B.4). Considerable progress has been made towards this end and we hope to have a working optical absorption system in the visible range by April 1985. It is our intent to proceed with the procurement of the appropriate cryostat, magnet, IR source and detection systems for the magneto-optics capability as soon as the monies year-marked for this purpose under the present contract become available.

References:

1. J. R. Arthur, Surf. Sc. 43, 449 (1974)

2. C. T. Foxon & B. A. Joyce, Surf. Sc. 50, 435 (1975); ibid 64, 298 (1977)

3. A. Madhukar, Surf. Sc. 132, 344 (1983)

4. J. Singh & A. Madhukar, Jour. Vac. Sc. Tech. B1, 305 (1983)

5. S. V. Ghaisas & A. Madhukar, Jour. Vac. Sc. Tech. B (To Appear)

6. S. V. Ghaisas & A. Madhukar, Phys. Rev. Letts. (Submitted)

7. A. Madhukar & S. V. Ghaisas (To be Published)

8. Y. Zeri, A. Redondo and W. A. Goddard III, Surf. Sc. 131, 221 (1983)

9. B. F. Lewis, T. C. Lee, F. J. Grunthaner, A. Madhukar, R. Fernandez and J. Maserjian, Jour. Vac. Sc. Tech. B2, 419 (1984)

10. B. F. Lewis, F. J. Grunthaner, A. Madhukar, T. C. Lee & R. Fernandez, Jour. Vac. Sc. Tech. B Submitted

11. B. F. Lewis, F. J. Grunthaner, A. Madhukar & R. Fernandez, (To be Published)

PUBLICATIONS (Cumulative List)

83-K-0050-1. B. F. Lewis, T. C. Lee, F. J. Grunthaner, A. Madhukar, R. Fernandez and J. Maserjian, **"RHEED Oscillation Studies of MBE Growth Kinetics and Lattice Mismatch Strain Induced Effects During InGaAs Growth on GaAs(100)"**, Jour. Vac. Sc. Tech. B2, 419 (1984)

83-K-0050-2. S. V. Ghaisas & A. Madhukar, **"Monte-Carlo Simulations of MBE Growth of III-V Semiconductors: The Growth Kinetics, Mechanism and Consequences for the Dynamics of RHEED Intensity"**, Jour. Vac. Sc. Tech. B (Sumbitted)

83-K-0050-3. S. V. Ghaisas & A. Madhukar, **"Role of Surface Molecular Reactions in Influencing the Growth Mechanisms and the Nature of Non-Equilibrium Surfaces: A Monte-Carlo Study of Molecular Beam Epitaxy"**, Phys. Rev. Letts. (Submitted)

83-K-0050-4. B. F. Lewis, F. J. Grunthaner, A. Madhukar, T. C. Lee and R. Fernandez, **"RHEED Intensity Behaviour During *Homoepitaxial* MBE Growth Kinetics and Mechanisms"**, Jour. Vac. Sc. Tech. B (Submitted)

83-K-0050-5. F. J. Grunthaner, M. Y. Yen, R. Fernandez, T. C. Lee, A. Madhukar and B. F. Lewis, **"Molecular Beam Epitaxial Growth and Transmission Electron Microscopy Studies of Thin GaAs/InAs(100) Multiple Quantum Well Structures"**, App. Phys. Letts. (Submitted)

GRADUATE STUDENTS/POST DOCTORAL:

1. Mr. T. C. Lee (Graduate Student)
2. Dr. Marshall Thomsen (Post Doctoral Fellow)

MONEY SPENT ON EQUIPMENT:

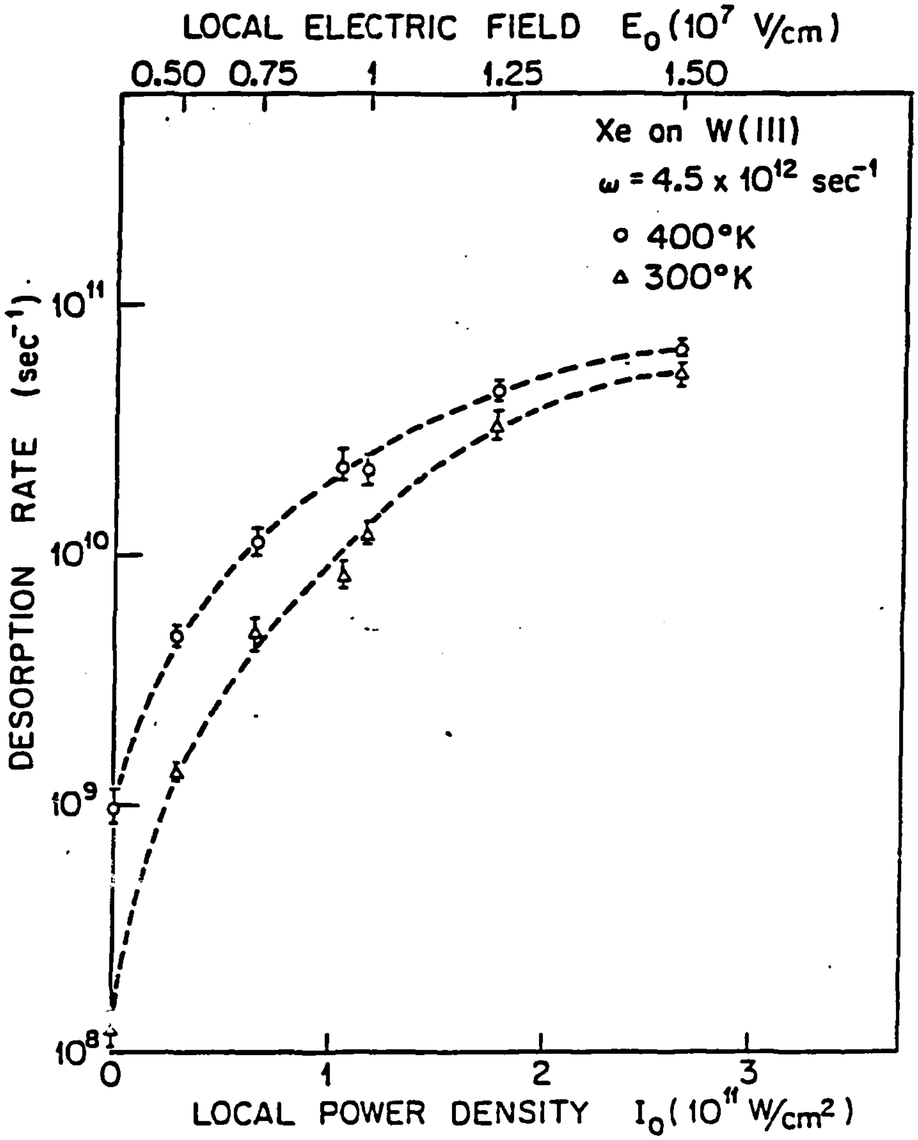
I. UNIVERSITY OF SOUTHERN CALIFORNIA

1.	VAX 11/750 Computer System	$ 92,346.00
2.	Printer and Terminals	$ 13,836.00
3.	Magnetic Tape Drive and Control	$ 5,014.00

4.	Memory Extension to 4MB	$ 5,014.00
5.	Disks	$ 1,120.00
6.	Tektronix Oscillioscope	$ 1,400.00
7.	Scanner Main Frame	$ 2,935.00
8.	Apple II+	$ 4,712.00
9.	Digital Plotter	$ 1,269.00
	TOTAL EQUIPMENT EXPENDITURES	**$128,106.00**

II. JET PROPULSION LABORATORY (Subcontract)

We have requested the Jet Propulsion Laboratory to provide us the information on capital equipment expenditures on the subcontract. We would be pleased to forward this information as soon as we receive it.



Shows the calculated desorption rate as a function of the laser power density incident on the surface - adsorbate bond. The parameters employed are appropriate for Xe adsorbed on W(111) and the desorption rate in the absence of the laser corresponds to the experimentally determined values.

OFFICE OF NAVAL RESEARCH

PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS REPORT

for

1 October 1983 through 30 September 1984

for

Contract N00014-83-K-0050

Task No. NR__________

Investigations of the Molecular Beam Epitaxial Growth and Characteristics of External Escitation Induced Non-Equilibrium Phases of Immiscible III-V Compound Semiconductors

Anupam Madhukar

University of Southern California
Materials Science Department
Los Angeles, CA 90089-0241

(a) Papers Submitted to Refereed Journals

1. S. V. Ghaisas and A. Madhukar, "Monte-Carlo Simulations of MBE Growth of III-V Semiconductors: The Growth Kinetics, Mechanisms and Consequences for the Dynamics of RHEED Intensity", Jour. Vac. Sc. Tech. B; AFOSR

2. B. F. Lewis, F. J. Grunthaner, A. Madhukar, T. C. Lee and R. Fernandez, "RHEED Intensity Behaviour During Homoepitaxial Growth of GaAs(100) and Implications for Growth Kinetics and Mechanisms"; Jour. Vac. Sc. Tech. B; NASA

(b) Papers Published in Refereed Journals

1. B. F. Lewis, T. C. Lee, F. J. Grunthaner, A. Madhukar, R. Fernandez and J. Maserjian, "RHEED Oscillation Studies of MBE Growth Kinetics and Lattice Mismatch Strain-Induced Effects During InGaAs Growth on GaAs(100)", Jour. Vac. Sc. Tech. 33, 419 (1984); NASA and AFOSR

(c) Books (and sections thereof) Submitted for Publication

None

(d) Books (and sections thereof) Published

None

(e) Patents Filed

None

(f) Patents Granted

None

(g) Invited Presentations at Topical or Scientific/Technical Society Conferences

1. F. J. Grunthaner, A. Madhukar, M. Y. Yen, B. F. Lewis, T. C. Lee and R. Fernandez, "Control of Interface Morphology in MBE Growth of InAs/GaAs Superlattices by RHEED Intensity Measurement", International Conference on Superlattices, Microstructures and Devices", Champaign, IL, August 1984, NASA and AFOSR

(h) Contributed Presentations at Topical or Scientific/Technical Society Conferences

1. B. F. Lewis, T. C. Lee, F. J. Grunthaner, A. Madhukar, R. Fernandez and J. Maserjian, "RHEED Oscillation Studies of MBE Growth Kinetics and Lattice Mismatch Strain-Induced Effects During InGaAs Growth on GaAs(100)", 11th Annual PCSI Conference, Pinehurst, NC, January 30--February 3, 1984; NASA

2. B. F. Lewis, F. J. Grunthaner, A. Madhukar, T. C. Lee and R. Fernandez, "RHEED Intensity Behaviour During MBE Growth of $In_xGa_{1-x}As$ on GaAs(100) and Implications for Growth Kinetics and Mechanisms", 3rd International Conference on Molecular Beam Epitaxy, San Francisco, August 1--3, 1984; NASA

3. S. V. Ghaisas and A. Madhukar, "Monte-Carlo Simulations of MBE Growth of III-V Semiconductors: The Growth Kinetics, Mechanisms and Consequences for the Dynamics of RHEED Intensity", 11th Annual PCSI Conference, Pinehurst, NC, January 30--February 3, 1984

(i) Honors/Awards/Prizes

None

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