Realization of In-Situ Sub Two-Dimensional Quantum Structures by Strained Layer Growth Phenomena in the $In_xGa_{1-x}As$/GaAs System

J. PAMULAPATI,* P.K. BHATTACHARYA, J. SINGH, and P.R. BERGER†

Solid State Electronics Laboratory, Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, MI 48109-2122

C.W. SNYDER‡ and B.G. ORR

Harrison M. Randall Laboratory of Physics, University of Michigan, Ann Arbor, MI 48109-1120

R.L. TOBER

U.S. Army Laboratory Command, Army Research Laboratories, 2800 Powder Mill Road, Adelphi, MD 20783-1197

We confirm that as the misfit strain in pseudomorphic epitaxial layer increases, surface thermodynamics controlled growth modes can change from a layer-by-layer to a three-dimensional (3-D) island mode. Both in-situ reflection high energy electron diffraction studies and in-situ scanning tunneling microscopy studies are utilized to demonstrate this transition to 3-D growth. This concept allows one to grow GaAs/$In_xGa_{1-x}As$/GaAs heterostructures where the electrons in $In_xGa_{1-x}As$ are possibly confined in lower dimensions.

Key words: InGaAs/GaAs, reflection high energy electron diffraction (RHEED), scanning tunneling microscopy (STM), strained layer growth

INTRODUCTION

Epitaxial growth techniques such as molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD) routinely provide two-dimensional (2-D) electronic systems by allowing fabrication of quantum wells. The realization of quantum wells is possible because of the layer-by-layer growth mode that is achievable under certain growth conditions.[1,2] Based on thermodynamic considerations, the growth modes of an epilayer growing on a substrate can be classified as:

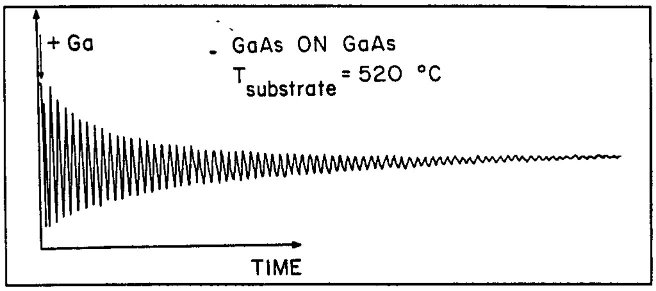
- Frank-van der Merwe (layer-by-layer);
- Vollmer-Weber (three dimensional islands [3-D]); and
- Stranski-Krastanov (where growth changes from layer-by-layer to 3-D island mode).[3]

The layer-by-layer growth mode has proven to be ideal for the growth of heterostructure systems.

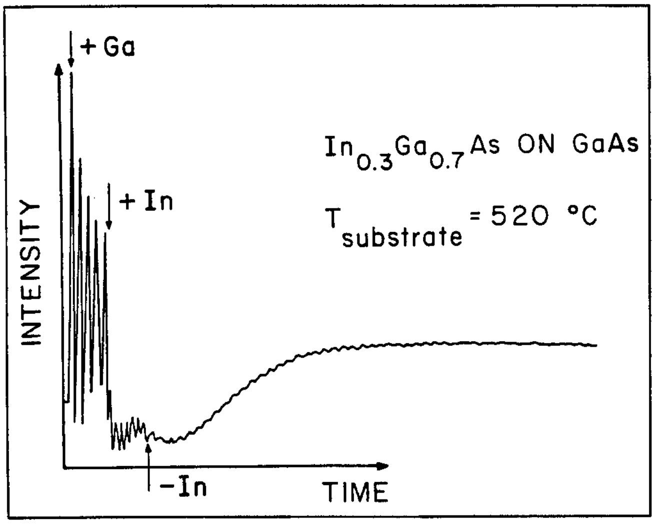
Attempts to realize lower dimensional electronic systems are increasing, partly fueled by theoretical calculations of the superior electronic and optical properties of such structures.[4,5] In this study, we have examined the potential of exploiting molecular beam epitaxy, under conditions where free energy considerations require a three-dimensional island surface structure, to realize "quantum boxes". In lattice matched systems, it is possible to achieve 3-D island growth, but this surface structure is random and leads to broadening or scattering rather than uniform sub 2-D confinement. For this reason, we use the strained $In_xGa_{1-x}As$/GaAs system. Since InAs and GaAs have a 7% misfit, we expect that as In composition in the $In_xGa_{1-x}As$ epilayer is increased, strain

*Present address: U.S. Army Electronics Technology and Device Laboratory, Fort Monmouth, NJ 07703-500.
†Present address: Department of Electrical Engineering, University of Delaware, Newark, DE 19716-3130.
‡Present address: AT &T Bell Laboratories, Murray Hill, NJ 07974.

(Received May 4, 1995; revised November 15, 1995)



a



b

Fig. 1. RHEED oscillation data for MBE growth of (a) GaAs on GaAs, and (b) pseudomorphic $In_{0.3}Ga_{0.7}As$ on GaAs.

energy will dominate the growth modes in the pseudomorphic (no dislocation) regime. We have earlier discussed the possibility of a transition from a free energy minimum state which is atomically flat to one which has 3-D surface islands.[6–9] In Fig. 1, we show in-situ reflection high energy electron diffraction (RHEED) oscillation data for lattice-matched and strained epitaxy on GaAs substrates. As can be seen in Fig. 1a, the oscillations persist due to the layer-by-layer growth mode; while during strained epitaxy, there is a much weaker specular reflection and a much weaker oscillation amplitude in which the surface smoothness does not recover, suggesting a 3-D growth mode. According to the free energy minimization criteria for (100) surface used by us,[6] we have shown that an island height of n (in terms of monolayers) given by

$$n^3 \cong 2\frac{W_1}{W_2}\left(\frac{R_0}{d_c}\right) \tag{1}$$

should occur, where d_c is the critical thickness and W_1 and W_2 are the nearest and second neighbor bond energies. In the InGaAs-on-GaAs systems, we expect a 2-D to 3-D surface transition when the In content exceeds ~20%. To provide direct evidence of the growth modes, we have carried out in-situ scanning tunneling microscopy (STM) studies of the strained growth of InGaAs on GaAs substrates. We have also made photoreflectance measurements for the confined structure which show high quality signals, signifying the fairly ordered nature of the 3-D growth.

MOLECULAR BEAM EPITAXIAL GROWTH

In this section, we describe the growth conditions for carrying out in-situ RHEED and STM measurements and ex-situ optical studies. For the STM measurements, MBE growth was carried out in an ultra high vacuum (UHV) system equipped with in-situ STM and RHEED. This system allowed us to obtain reproducible STM images of the various phases of InGaAs on GaAs (100) growth. Starting with a p^+ GaAs (100) substrate, a 400 nm nominally undoped GaAs buffer layer was grown at 570°C under arsenic-rich conditions. The growth was then interrupted, and the substrate temperature was lowered to 520°C and a few more monolayers of GaAs were grown. The growth of these layers was terminated by shuttering the Ga source, turning off the As furnace, and then slowly cooling the sample to room temperature under a decreasing As flux. The quality of the (2×4) RHEED pattern was used to manually control the rate of cooling. After 20–40 min of cooling, the pressure in the MBE chamber reached the 10^{-10} Torr range and the sample was transferred to the STM chamber. This growth procedure produced a buffer layer surface that was ordered and atomically flat, with only an occasional kink or step, for areas up to 100 nm × 100 nm. Having grown the buffer layer in this manner, we then grew a four monolayer $In_{0.35}Ga_{0.65}As$ film at a substrate temperature of 520°C, starting with a RHEED maximum. The growth was monitored by RHEED. Before the indium shutter was opened to grow the ternary compound, we observed strong specular oscillations and a sharp, streaked (2 × 4) pattern. Upon opening the In shutter, the pattern changed immediately to a streaked, diffuse (1 × 1) pattern[10] and much weaker oscillations were observed. Simultaneous measurement of the integral order streak spacing showed a gradual 0.7% increase in the surface lattice constant up to the thickness of four monolayers at which point the growth was interrupted. A shuttering of the In and Ga sources, followed by (within 0.5 s) a reduction of the substrate temperature to 425°C, and enclosing the sample in a liquid nitrogen cooled shroud enabled us to preserve the (1×1) pattern and the lattice relaxation.

For an optical study of growth uniformity and for the observation of excitonic states, in these pseudomorphic layers, MBE growth of the heterostructures was carried out on (100) semi-insulating or n^+ (silicon doped) GaAs substrates. Growth was initiated with a 30 period undoped GaAs (20Å) /$Al_{0.30}Ga_{0.70}As$ (20Å) superlattice buffer followed by 2000Å of GaAs. These layers were grown at 620°C. This was followed by the growth of $In_xGa_{1-x}As$ ($0.1 \le x \le 0.3$) at 520°C, at a rate ≤1.0 μm/h. This temperature was chosen since our studies show that under these conditions, the system should be able to reach surface thermodynamic equilibrium. The thickness of the $In_xGa_{1-x}As$ layer was varied from 20 to 55Å, keeping the total thickness below the critical thickness, in order to consistently compare with STM results. The substrate temperature was further lowered to 400°C and a 1500Å GaAs layer was grown. The low substrate temperature was chosen to reduce the surface mobility of the In adatoms and reduce In interdiffusion. The top GaAs layer was also grown to provide quantum confinement heterostructures and to prevent surface depletion of the pseudomorphic layers.

EXPERIMENTAL RESULTS

Scanning Tunneling Microscopy Studies

Shown in Fig. 2a is a typical STM image of buffer layer surface. The dark rows running diagonally across the image in the [$\bar{1}$10] direction are spaced 1.6 nm apart, corresponding to the 4x spacing. These are the rows of missing As dimers. Higher resolution, smaller scale images show the expected coexistence of the (2 × 4) and c(2 × 8) reconstructions.[11,12]

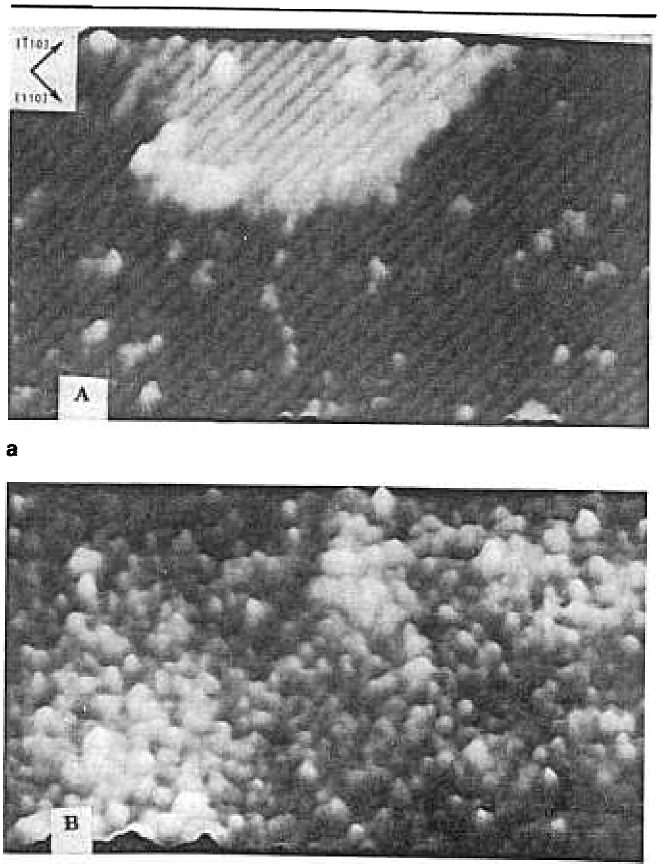


Fig. 2. (a) STM image of the surface of a GaAs (100) buffer layer. The tunneling voltage was 2.8 V, applied to the sample, and the tunneling current was 70 pA. The As termined surface shows rows of missing As dimers along the [$\bar{1}$10] direction that are spaced 1.6 nm apart, corresponding to the 4X spacing. The corrugation is 0.2 nm, and (b) 90 nm × 50 nm STM scan of the surface after four monolayers of $In_{0.35}Ga_{0.65}As$ have been deposited. The crystal axes are the same as in (a). The tunneling voltage was 3.0 V, applied to the sample, and the tunneling current was 30 pA. The surface is covered with islands roughly 3.2 nm in diameter and with an average corrugation of 0.6 nm. Examination with RHEED shows this surface to have a diffuse (1 × 1) pattern and a 0.7% relaxation of the surface lattice constant.

Figure 2b shows a typical STM image of the surface of this InGaAs film. Note the 3-D character of the surface even in the pseudomorphic phase of growth. We note that Fig. 2b represents a typical growth front observed under high strain (In content >20%) conditions, which is independent of the phase of RHEED oscillations at the start of InGaAs growth. It is important to observe that although there is some disorder in the sizes of the InGaAs islands, it appears that they have a fairly narrow size distribution. Theoretically, we expect the free energy minimum conditions to lead to islands with uniform sizes if entropy considerations are ignored. Although STM can only examine fairly small regions of the substrate, within these regions the root mean square (RMS) height of the islands is ~0.6 nm, an on the average their lateral extent is ~3.2 ± 0.5 nm. The uniformity of these island sizes on a larger scale is confirmed by photoreflectance (PR) and piezoreflectance (PzR) studies to be described later.

Optical Studies

Initial investigation of the optical properties consisted of low temperature photoluminescence (PL) measurements. Since $In_xGa_{1-x}As$ has a lower bandgap than GaAs, we expect confined states in this region. Shown in Fig. 3 is the PL spectra of the GaAs/$In_{0.30}Ga_{0.70}As$/GaAs heterostructure with a 3.5 nm pseudomorphic well. A peak with a linewidth of ~13 meV is observed. The energy position of the emission confirms that it originates from the $In_xGa_{1-x}As$ layer

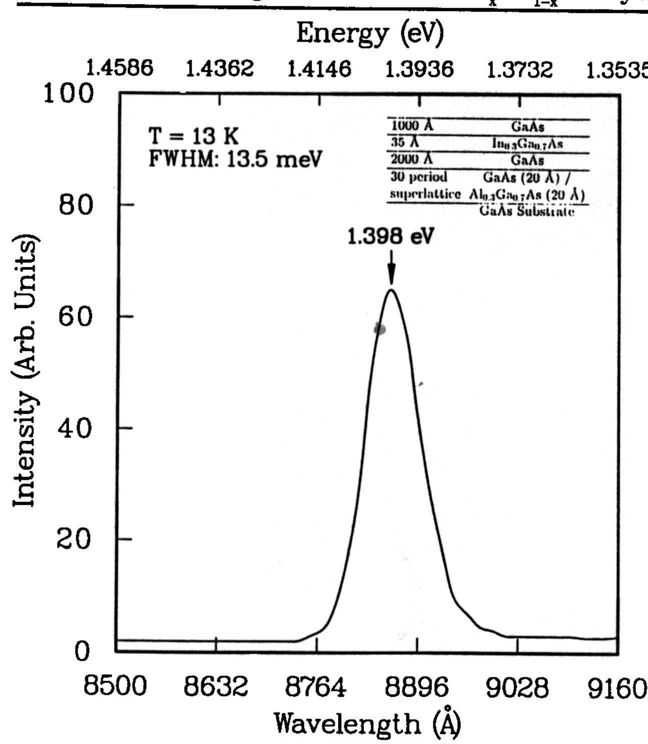
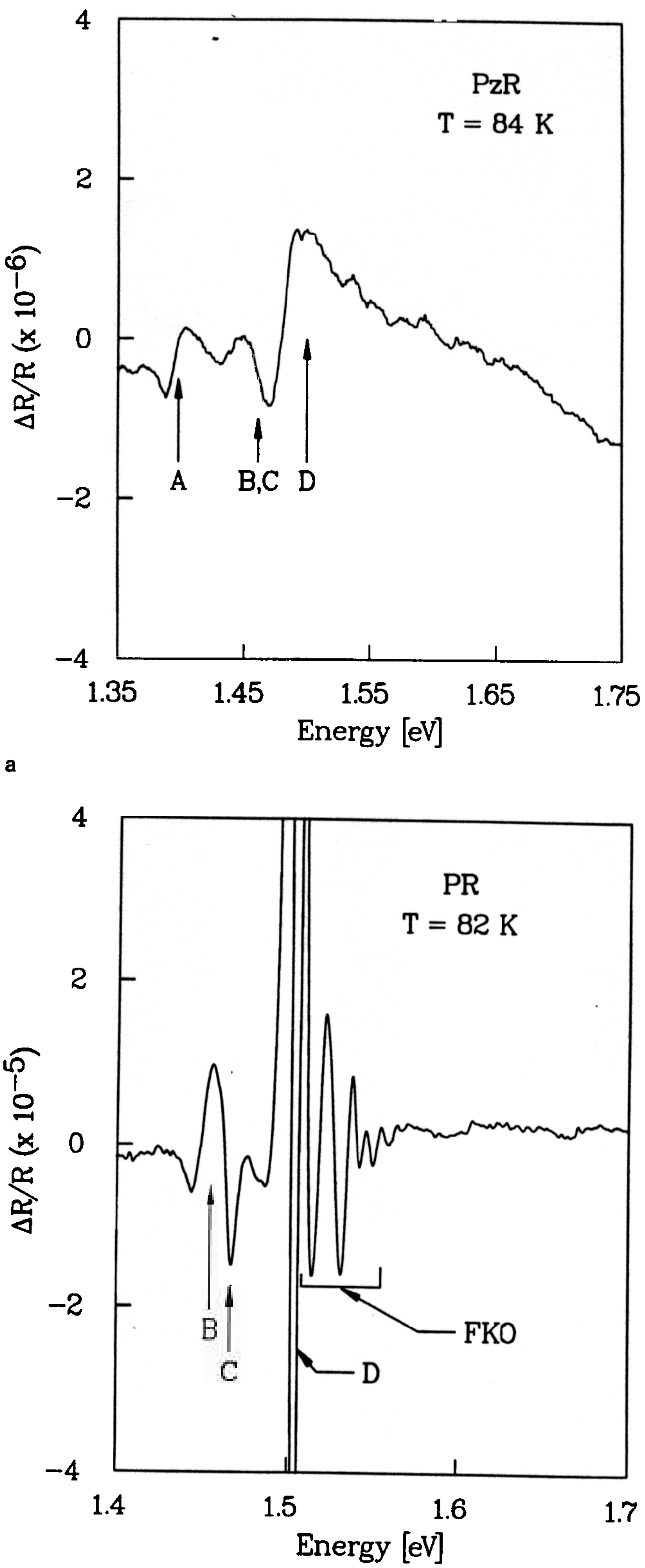


Fig. 3. Low temperature photoluminescence spectra of GaAs/$In_{0.3}Ga_{0.7}As$/GaAs pseudomorphic heterostructure. Inset shows the structure of the grown layer.



b

Fig. 4. Piezoreflectance (a) and photoreflectance (b) data measured at 84 and 82K, respectively, for the GaAs/$In_{0.3}Ga_{0.7}As$/GaAs pseudomorphic heterostructure.

and we believe it is excitonic in nature.

To shed further light on the spectra, we used modulation spectroscopy techniques to probe the electronic structure in such a way as to reject all contributions to reflectance except those originating at high symmetry points in the Brillouin zone.[13] Figure 4 shows PzR and PR spectra recorded at a sample temperature of 82K. From measurements on several calibrations, samples consisting of single layer GaAs, GaAs/AlGaAs quantum well (QW) and InGaAs/GaAs QW, we believe that the peaks labeled A and B/C originate from heavy hole (HH) and light hole (LH) transitions, respectively. The peak labeled D originates from GaAs excitons and the features labeled E are Franz-Keldysh oscillations (FKO). The sharpness of the features labeled A and B confirm the uniformity of the island size in the 3-D mode of growth.

DISCUSSION

In 2-D lattice matched quantum wells, there is a lifting of degeneracy between the heavy hole $\left(HH - |\frac{3}{2} \pm \frac{3}{2}>\right)$ states and the light hole $\left(LH - |\frac{3}{2} \pm \frac{1}{2}>\right)$ states due to quantum confinement. In presence of biaxial strain produced in a 2-D quantum well, additional splitting is produced by the shear component of strain.[14] The net separation is then given by

$$\Delta E_{tot}\,(HH - LH) = \Delta E_{QC}(HH - LH) + \Delta E_{sh} \qquad (2)$$

where $\Delta E_{sh} = 5.96\,\varepsilon$ eV, and $\varepsilon = 0.07x$. Here ΔE_{QC} is the splitting due to quantum confinement and ε is the misfit. The factor 5.96 appears due to the various deformation potentials and force constants for the $In_xGa_{1-x}As$ system. In the InGaAs/GaAs system, if we assume that the band discontinuities $\Delta E_c : \Delta E_\upsilon$ = 60:40, the lowest light hole state is in the GaAs region and the HH-LH separation can be shown to be $\Delta E_{HH-LH} \sim 0.3_x$ (eV) if the strain is biaxial.[15] If on the other hand, the InGaAs was surrounded by GaAs as would occur if the InGaAs regions were growing in 3-D islands which were then surrounded by GaAs, the strain on the InGaAs would primarily be of hydrostatic nature. Since hydrostatic pressure does not lift the HH-LH degeneracy, we expect that the splitting of the HH-LH peaks could be used as a parameter to distinguish the 2-D quantum well from a lower dimensional well, as in a quantum box.

It is important to note that for biaxial strain, regardless of whether the LH state is confined in InGaAs or GaAs, the HH-LH separation must increase with excess In content. We find that for compositions of 15 and 30% in the well, the measured separation is 35 and 30 meV. If the LH is confined in InGaAs, the separation should be 60 and 120 meV. If the LH is confined in GaAs, the separation should be 40 and 30 meV, respectively. This additional evidence suggests the 3-D nature of the growth at higher In compositions. Furthermore, it is of interest to note that by virtue of the island formation during InGaAs growth and the overgrown GaAs layer, in-situ quantum boxes can be formed by this technique.

Therefore, both from STM and reflectance measurements, we see direct evidence that the growth of pseudomorphic InGaAs tends toward a three-dimensional island mode from the layer-by-layer mode, as the strain increases. The STM shows that after depositing only four monolayers of $In_{0.35}Ga_{0.65}As$ on a buffer layer, the growth no longer proceeds in a layer-by-layer mode. In addition, the reflectance measurements indicate a good degree of correlation in the island widths and heights, as evidenced by the existence and definition of peaks in the modulated spectra. It may be noted that although the thickness of the InGaAs are different for the STM and the optical measurements, a valid comparison can be made since both are in the below-critical-thickness regime.

CONCLUSION

In conclusion, we present a method of realizing in-situ lower dimensional quantum structures by the introduction of compressive strain into the well. This is clearly evidenced by the STM studies of the pseudomorphic layers on a small scale and the PL and reflectance studies on a larger scale. It may be feasible to use this self-organization to grow arrays of light-emitters with quantum box active regions.

ACKNOWLEDGMENTS

The work was supported by the U.S. Office of Naval Research under contract N00014-89-J1519 and the Army Research Office under contracts DAAL03-87-K0007 and DAAL03-92-G0109.

REFERENCES

1. J. Singh and K.K. Bajaj, *Superlattices and Microstructures*, 2, 185 (1986).
2. J.H. Neave, P.J. Dobson, B.W. Joyce and J. Zhang, *Appl. Phys. Lett.* 47, 100 (1985).
3. For a review, see M. Wortis, *Fundamental Problems in Statistical Mechanics*, Vol. 6, ed. E.G.D. Cohen (Amsterdam: North-Holland, 1985), p. 87.
4. A. Gossard, J.H. English, P.M. Petroff, J. Cibert, G.J. Dolan and S.J. Pearton, *J. Cryst. Growth* 81, 101 (1987).
5. J.P. Harbison, *Gallium Arsenide and Related Compounds 1988*, Institute of Phys. Conf. Series Number 96, (Bristol: IOP Publishing), p. 1.
6. P.R. Berger, K. Chang, P. Bhattacharya, J. Singh and K.K. Bajaj, *Appl. Phys. Lett.* 53, 684 (1988).
7. D.J. Eaglesham and M. Cerullo, *Phys. Rev. Lett.* 64, 1943 (1990).
8. D.J. Srolovitz, *Acta Metall.* 37, 621 (1989).
9. C.W. Snyder, B.G. Orr, D. Kessler and L.M. Sander, *Phys. Rev. Lett.* 66, 3032 (1991).
10. H. Nakao and T. Yao, *Jpn. J. Appl. Phys.* 28, L352 (1989).
11. M.D. Pashley, K.W. Haberen, W. Friday, J.M. Woodall and P.D. Kirchner, *Phys. Rev. Lett.* 60, 2176 (1988).
12. D.K. Beigelsen, L.E. Swartz and R.D. Bringans, *J. Vac. Sci. Technol A* 8, 280 (1990).
13. See, for example, D.E. Aspnes, *Handbook on Semiconductors*, ed. T.S. Moss (New York: North-Holland, 1980), p. 109.
14. H. Kato, N. Iguchi, S. Chika, M. Nakayama and N. Sano, *J. Appl. Phys.* 52, 588 (1986).
15. H. Kato, N. Iguchi, S. Chika, M. Nakayama and N. Sano, *Jpn. J. Appl. Phys.* 25, 1327 (1986).