SCANNING TUNNELING MICROSCOPY FOR MORPHOLOGICAL CHARACTERIZATION OF InN THIN FILMS

R. Fainchtein, D. Dayan*, W.A. Bryden, J.C. Murphy and T.O. Poehler

Applied Physics Laboratory
Johns Hopkins University
Laurel, Maryland 20707 U.S.A.

*On leave from: Nuclear Research Center Negev
P.O. Box 9001, Beer Sheva, Israel

INTRODUCTION

InN is a promising semiconductor material because of its wide energy band gap (~ 2 eV). This characteristic makes the material, in concert with analogous group III metal nitrides, suitable for the production of electromagnetic sources and detectors tuned to cover the visible part of the optical spectrum. In order to make the appropriate electronic devices thin films of InN of good quality material have to be produced. This is so far the issue preventing the utilization of this material for device applications. Several techniques of film deposition have been and continue to be investigated in order to obtain device quality material. Among them, reactive magnetron sputtering offers to be a promising deposition technique. Crystalline films produced by reactive magnetron sputtering and other deposition techniques exhibit columnar microstructure. The structure consists of a network of low density material or voids that surrounds an array of parallel rod-shaped columnar regions of higher density. The formation of those columns are known to depend on the deposition parameters. Among them are the nature of the substrate [1], the rate of deposition [2], the pressure and composition of the gas phase [3], the film thickness and the ratio between the substrate temperature Ts and the film material melting point Tm [4,5]. The presence of microscopic voids within the grains contributes to the degradation of the surface flatness and to the formation of surface porosity [6].

High resolution direct studies of thin film morphology and structure have typically been done by transmission electron microscopy, either by direct imaging [7], or by replica observations [8]. The use of the transmission electron microscope technique demands that either the deposited film be detached from its substrate for examination or that the substrate itself be sufficiently thin to allow the composite substrate-specimen to be examined by electron transmission. Traditionally semiconductors have been thinned by chemical methods, which are generally rapid and simple. However, chemical methods are inadequate for multicomponent semiconductors. Their main limitation is the selective etching of phases and almost total lack of control over the thinning of specific areas of the specimen. Ion milling, which corresponds to sputtering with relative low energy ions (3 - 6 KV) provides a means of circumventing these problems, although it is not without its own difficulties. The most crucial limitation is the radiation damage or ion damage, i.e. the production of lattice defects, which can give rise to structural features that are not representative of the bulk material [9].

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This paper reports on a new technique to study the microstructure of the air sample interface of thin layer films. Compared to other methods the STM is non-destructive and gives high resolution lateral in-plane and perpendicular to the plane images of the surface. The surface morphology of InN thin films grown by radio frequency reactive magnetron sputtering (RFRMS) on different substrates and at different temperatures was investigated using a scanning tunneling microscope.

EXPERIMENT

Sample Preparation

Thin films of InN were deposited by RFRMS onto different substrates at different temperatures. The deposition technique employs radio frequency excitation at 13.56 MHz, a magnetron sputtering source and a chemically reactive gas. The magnetron sputtering system which has been described in detail elsewhere [10], is based on a liquid-nitrogen-trapped diffusion-pumped vacuum station achieving vacuum base pressures of the order of 3×10^{-8} Torr. Sputter deposition was carried out in a pure N_2 atmosphere at pressures of 5 mTorr. The sputtering target used was made of 99.999% pure In metal. The films were deposited on a circular area of ~ 7 mm diameter. Substrates consisted of; oriented (0001) sapphire single crystals, randomly oriented sapphire single crystals, high-purity fused quartz and Si (111) single crystals. After a degreasing and cleaning process, four substrates were mounted in a supporting metallic plate backed by a tungsten wire resistive heater and inserted into the vacuum system. The system was pumped down to its base pressure and preheated to a temperature of ~ 900 °C. For each deposition the sputtering parameters were kept constant and were closely monitored and recorded. The substrate temperature was set to different value between 50 and 600 °C for each of the different runs. The temperature was preset before sputtering and a constant electrical power was maintained to the heater during the run. This resulted in a slow temperature increase of ~ 50 °C from beginning to end of the deposition. The power supplied to the target during deposition was maintained constant for all the films. Therefore, it is reasonable to assume that besides the variations in yield due to changes in the sticking coefficient with temperature, and the variations in yield due to desorption, the deposition rate was maintained constant. Increases in the temperature led to thickness variations which went from 0.6 μm for the low temperature films to 1.3 μm for the high temperature films. All the deposited films were characterized electrically and by X-ray diffraction.

Images

STM images of the front surface of the InN films were collected in air with a commercial STM [11]. The STM was operated in a constant current mode (with fast and tight feedback response with respect to the scanning speed). The tunneling current was held constant at 1nA for most of the scans and at 0.5 nA on the scans of the 500 °C and higher temperature films. A bias voltage of 100mV with the tip positive with respect to the substrate was maintained in the scans of the low temperature films (from 50 to 400 °C). For films deposited at temperatures higher than 400 °C the bias voltage was increased to 1V, again with the tip being kept positive with respect to the substrate. No variations in the structures revealed by the images were observed with changes in the bias voltage (even if the polarity was reversed) or, changes in the tunneling current. The only noticeable effects of these variations, were the image resolution and the tunneling gap stability (image noise), which were more apparent in small area scans (100 x 100 nm). The scans were made at a rate of 5 Hz per line. An image consists of 200 lines with 200 resolved elements per line. Typical images covered an area of 625 x 625 nm, closeups of 200 x 200 nm and expansions of 1460 x 1460 nm were made on most of the images. On samples showing very large features coverages were increased to areas as high as 14 μm x 14 μm. The tunneling tips were made of Pt with 20% Ir. Tips were either mechanically polished [11] or AC electrochemically polished in a 20% solution of KCN in water [12]. Better image resolution was obtained with the electropolished tips.

RESULTS

Figures 1-2 are height encoded three dimensional plots of the STM topographs of different InN films. The gray scale used is linear, going from 0 for black to full scale for white, given by the scale in the perpendicular to the plane axis. The figures show typical images of the InN films at four different temperatures and on four different substrates. The images reveal very clearly the presence and shape of the grains and voids in these polycrystalline samples. The grains have typical dimensions of few tens of Angstroms on films deposited at low substrate temperatures to thousands of angstroms on films deposited at high substrate temperatures. The trend of increased grain growth as a function of the substrate temperature is clearly visible in the sequence of images of Figure 1.

Grain sizes were determined by applying the lineal analysis techniques used in metallography [13]. By counting the total number of boundaries of a given length of line a good estimate of the grain diameter can be obtained. Since not all the grains were equiaxed, line scans along the two orthogonal diagonals of an image scan were used and their resulting values averaged. The average grain diameter for the InN films deposited onto different substrates at different temperatures is presented in Fig. 3 as a function of substrate temperature. The results show that at low substrate temperatures (from 50 to 300 °C) there is a monotonic small increase in grain dimensions with increasing substrate temperature until an exponential growth regime becomes apparent beyond 400 °C. The films deposited at 350 °C show an anomalous behavior with respect of the two regimes giving a clear indication of a transition region.

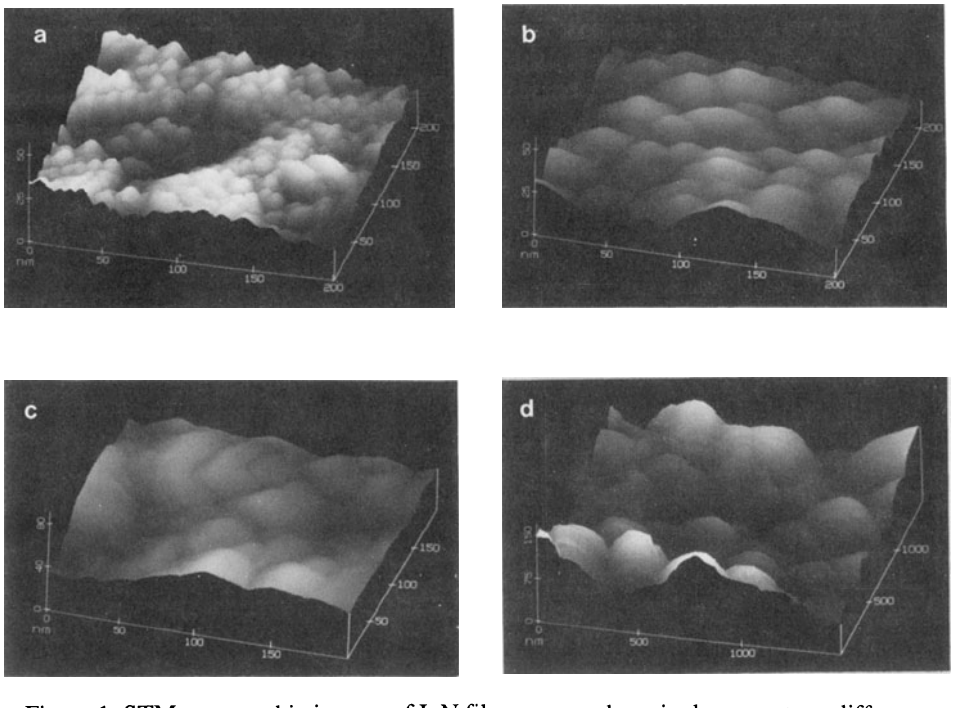

Figure 1. STM topographic images of InN films sputter-deposited on quartz at different temperatures. (a) 50 °C; (b) 300 °C; (c) 500 °C; (d) 600 °C.

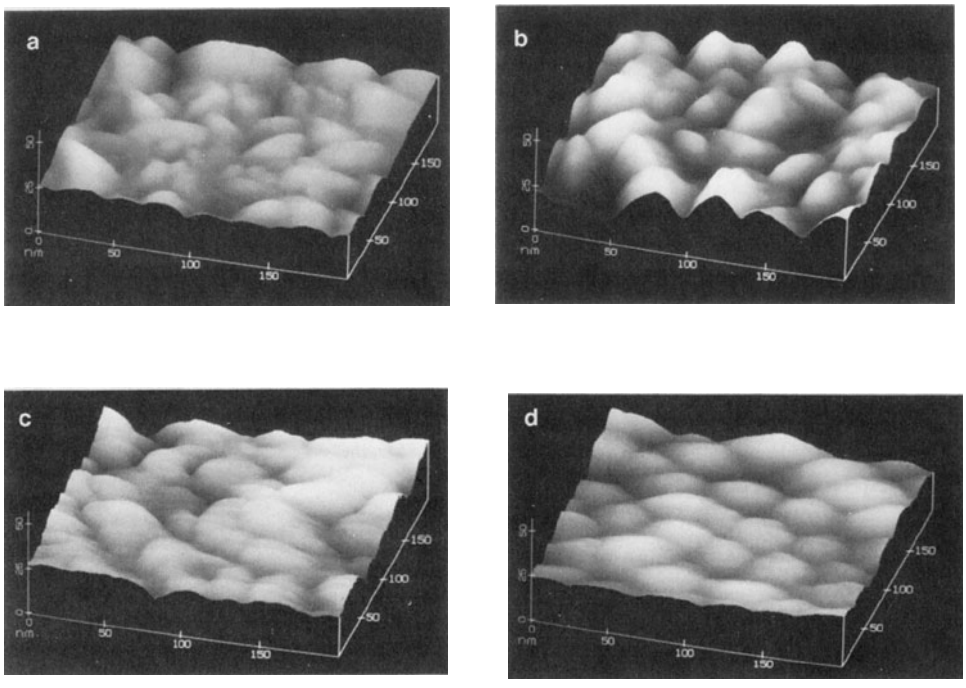

Figure 2. STM topographic images of InN films sputter-deposited at 350 °C on different substrates: (a) quartz; (b) silicon; (c) randomly oriented single crystal sapphire; (d) (0001) oriented single crystal sapphire.

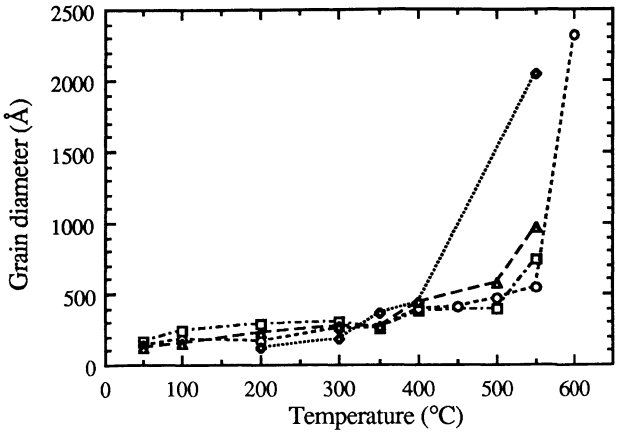

Figure 3. Average grain diameter of InN polycrystalline films deposited on different substrates at different temperatures. Film substrates consist of: (○) quartz; (□) silicon; (△) randomly oriented single crystal sapphire; (◇) (0001) oriented single crystal sapphire.

Low roughness of the surface is a necessary characteristic for device quality thin films. The statistical surface roughness defined as the RMS value of the surface height variations can be obtained from the STM topographs. The surface roughness for the films deposited on different substrates are plotted in Fig. 4 as a function of substrate temperature. The values were computed on images covering a scanned area of 625 x 625 nm^2. The results show at first an increase of roughness with temperature on all substrates going from 50 to 200 °C. From 200 °C to 350 °C the roughness decreases reaching an overall minima at 350 °C. The parameter subsequently increases very sharply as the temperature increases. In two of the substrates (silicon and randomly oriented single crystal sapphire) the roughness decreases on going up in substrate temperatures from 450 °C to 500 °C.

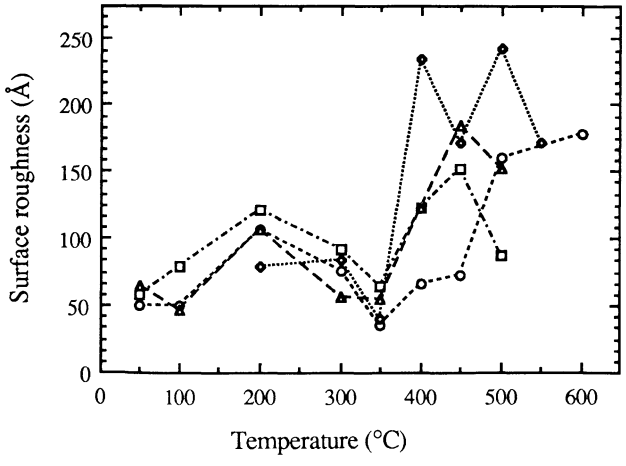

Figure 4. Surface roughness of InN polycrystalline films deposited on different substrates at different temperatures. Film substrates consist of: (○) quartz; (□) silicon; (△) randomly oriented single crystal sapphire; (◇) (0001) oriented single crystal sapphire.

DISCUSSION

Reported studies of thin film deposition have established that the substrate temperature is the most influential parameter governing the structure of vapor deposited films under no additional ion bombardment [14-15]. Films deposited under similar conditions at different temperatures will show four different structural characteristics depending on T/T_m. The relevant processes governing the structure are atomic shadowing and diffusion. At low substrate temperatures bulk diffusion has little effect on the surface morphology which appears to be dominated by substrate roughness and atomic shadowing. As the temperature of the substrate is increased bulk diffusion becomes more and more important up to the point in which it completely dominates the structure of the resulting film.

Four morphological regions have been identified in a structure vs temperature and gas pressure diagram; normally refered as Zone 1, Zone T, Zone 2, and Zone 3 [14-15]. In this study the pressure of the nitrogen which acted as the momentum transfer and reaction gas was maintained constant throughout the depositions. Evidence of zone transitions is present in the results of Figures 3 and 4. In the case of InN it is very hard to relate the temperature parameter to T_m in order to directly compare the structural results to the Thornton model, since the material decomposes before melting.

However, the results of Figures 3 and 4 do provide a quantitative way to identify the deposition and growth characteristics of the films on an absolute temperature scale. At low temperatures; from 50 °C to 200 °C, the grain dimensions remain fairly constant, the films first become slightly rougher as temperature increases before they become smoother. These conditions correlate with the Zone 1 description made by Thornton, namely the structure of the crystals is not well defined and the structures are in general larger than the crystallographic grain size. As the temperature is increased the films become smoother; which is a clear indication of Zone T. Maximum smoothness of the films is achieved at 350 °C. From 400 ° C on to 500 °C the film structure falls into the classification of Zone 2. The structure is dominated by adatom surface diffusion, the grains grow with increased temperature and their structure is columnar. Beyond 500 °C the structure corresponds to that of Zone 3. The grain diameter approaches the dimensions of the film thickness, the growth is dominated by bulk diffusion and the grains tend to become equiaxed.

Besides the topographic results, the STM also provides information on the electronic condition of the sample, all of the InN films studied were imaged at bias voltages below the band gap voltage of intrinsic InN. This condition indicates that the surface is that of a heavily doped semiconductor, which could arise from non-stochiometric composition of the semiconductor and/or the oxidized material present at the surface.

CONCLUSIONS

In this study we intended to use the STM to resolve grain structures as small as 10 nm, no atomic resolution is attempted. We take advantage of the large dynamic imaging range of the instrument to resolve morphological features of polycrystalline thin films. We have been able to demonstrate that with relative ease this technique can yield structural information of thin films nondestructively with higher resolution than other electron microscopy techniques.

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