Surface segregation of InGaAs films by the evolution of reflection high-energy electron diffraction patterns*

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Surface segregation is studied via the evolution of reflection high-energy electron diffraction (RHEED) patterns under different values of As_4 BEP for InGaAs films. When the As_4 BEP is set to be zero, the RHEED pattern keeps a $4\times3/(n\times3)$ structure with increasing temperature, and surface segregation takes place until 470 °C. The RHEED pattern develops into a metal-rich (4×2) structure as temperature increases to 495 °C. The reason for this is that surface segregation makes the In inside the InGaAs film climb to its surface. With the temperature increasing up to 515 °C, the RHEED pattern turns into a GaAs(2×4) structure due to In desorption. While the As_4 BEP comes up to a specific value (1.33×10^{-4} Pa–1.33×10^{-3} Pa), the surface temperature can delay the segregation and desorption. We find that As_4 BEP has a big influence on surface desorption, while surface segregation is more strongly dependent on temperature than surface desorption.

Keywords: reflection high-energy electron diffraction, InGaAs films, surface segregation, surface desorption

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1. Introduction

Indium gallium arsenide (InGaAs) is an important compound semiconductor material. As its carrier mobility is 10 times greater than that of silicon, it has now become a candidate material for high-speed microelectronics devices such as dual-channel high-frequency devices and high electron mobility transistors.[1]

So far, high-quality heterostructure thin films of $In_xGa_{1-x}As$[2] have been fabricated by many research teams through molecular beam epitaxy (MBE), and their surface morphologies and surface structures have also been investigated. The surface roughness and surface structure of thin films are extremely important for physical research and practical applications because the preparation of semiconductor optoelectronic devices is greatly influenced by surface segregation. Since In and Ga have different chemical potentials and different binding energies, the In segregation of $In_xGa_{1-x}As$ films is inevitable under certain conditions. Although In surface segregation has been studied by many research teams, there is still considerable controversy about the internal mechanism of surface segregation.[3−5]

Sears *et al.*[6] used X-ray photoelectron spectroscopy (XPS) to analyse the variation of In composition in annealing $In_xGa_{1-x}As$ films, and they thought the variation was caused by In surface segregation. Millunchick *et al.*[7] also studied In surface segregation in $In_xGa_{1-x}As$ films by *in situ* Auger electron spectrometry (AES).

In this paper, reflection high-energy electron diffraction (RHEED) is used to study In surface segre-

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gation in-depth through an investigation of RHEED pattern evolution, along with the variations in As_4 BEP and the substrate temperature, and a series of experimental results with directive significance is obtained.

2. Experiment

The experiment was carried out in an MBE vacuum chamber of ultrahigh vacuum (4×10^{-9} Pa–8×10^{-9} Pa). The substrate is an epi-ready n-GaAs (001) wafer, and the Si doping concentration is N_D is $1.49\times10^{18}/cm^3$. The substrate temperature (thermocouple reading) was calibrated by different substrate temperatures in a variety of GaAs (001) surface reconstruction phases and transition temperatures under As_4 BEP.[8] The detailed deposition process of $In_xGa_{1-x}As$ heterostructure thin films and the method of determining the composition of thin films has been published previously,[9,10] so those details will not be mentioned here. In this paper, we elucidate an effective method to research In surface segregation at different substrate temperatures and As_4 BEP by RHEED pattern evolution.

3. Results and analysis

After annealing, the samples were quenched to room temperature. The RHEED patterns are shown in Fig. 1. The [110] direction has a 2× with a weak 4× structure. In the $[1\bar{1}0]$ direction, a 3× pattern is clearly visible. The sample shows a typical $4\times3/(n\times3)$ surface structure, and the pattern in the [100] direction is a 1× pattern, which is consistent with the results given by other research groups.[11] The evolution of RHEED patterns indicates that the change in the [100] direction was rather small. When the sample temperature was gradually raised to 450 °C, the RHEED patterns always maintained a $4\times3/(n\times3)$ surface structure under any value of As_4 BEP, which is denoted as state A. Thus we speculate that the In segregation did not occur inside the $In_xGa_{1-x}As$ film at a temperature lower than 450 °C. Here, we will analyse the In segregation at high temperature in the following two cases.

(i) Under zero As_4 BEP, in which case when the sample temperature was continually raised to 470 °C, the 3× RHEED pattern in the $[1\bar{1}0]$ direction began to blur and the line in the middle of the main bright spot gradually dimmed, as shown in Fig. 2. We denote the state as B. This is because, with the sample temperature climbing, the surface segregation of In started to occur. Meanwhile, when the In content was gradually increased on the surface, the surface structure was changed gradually and the surface roughness was increased, the background of the RHEED patterns became gradually brighter. When the sample temperature continued to rise, the In segregation continued to strengthen, the In composition was increased on the surface, and the surface structure underwent a dramatic change. The 3× RHEED patterns in the $[1\bar{1}0]$ direction became more and more blurry, until they disappeared.

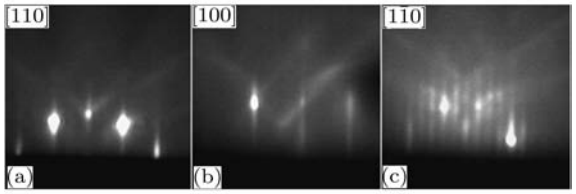


Fig. 1. RHEED patterns after growing $In_xGa_{1-x}As$ in directions: (a) [110], (b) 100, and (c) $[1\bar{1}0]$.

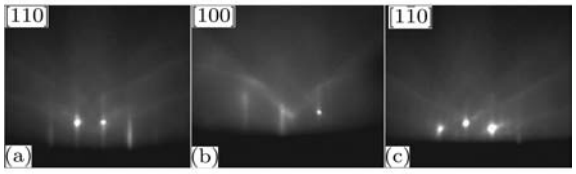


Fig. 2. RHEED patterns at the onset of In surface segregation in the directions: (a) [110], (b) [100], and (c) $[1\bar{1}0]$.

When the sample temperature reached 495 °C, the RHEED image in the $[1\bar{1}0]$ direction gradually showed 2× patterns, and a 4× pattern in the [110] direction was clear. The whole sample surface was a (4×2) structure phase, as shown in Fig. 3, which is denoted as state C. Accordingly, we speculate that the In surface segregation continued to proceed as the sample temperature increased. A large amount of In segregated out onto the surface from the inside of the sample. Under zero As pressure, a large number of In atoms gathered toward the surface. A (4×2) structure phase of a metal-rich III–V compound semiconductor appeared on the surface of the sample.[12]

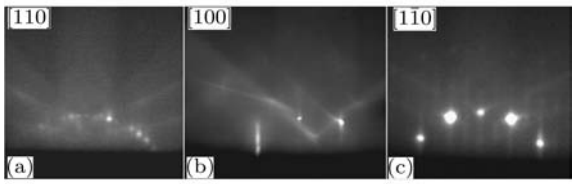


Fig. 3. RHEED patterns at T_{sample} = 495 °C and 0 As pressure in directions: (a) [110], (b) [100], and (c) $[1\bar{1}0]$.

When the sample temperature was increased to 515 °C, the 2× pattern in the [1$\bar{1}$0] direction gradually turned into a 4× structure, and the 4× pattern also changed into 2× in the [110] direction. The sample presented the GaAs (2×4) surface structure as shown in Fig. 4, which is denoted as state D. For this phenomenon, we believe that after the temperature exceeds 495 °C, the In desorption on the sample surface becomes significant. So we think that 495 °C is the critical point at which In segregation is finished and In desorption begins to take place. When the temperature increases to 515 °C, little In is left on the surface, only GaAs. This result is consistent with the AES[13,14] and XPS[15,16] results given by many research groups.

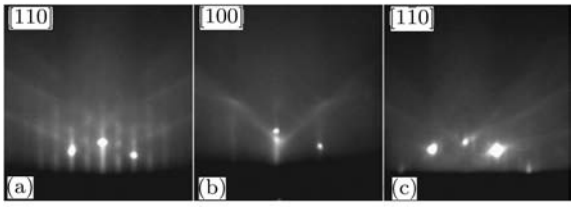


Fig. 4. RHEED patterns at $T_{\rm sample}$ = 515 °C and 0 As pressure in directions: (a) [110], (b) [100], and (c) [1$\bar{1}$0].

Under zero As pressure, the above evolution of the RHEED patterns showed the whole process of structure phase transition (A→B→C→D) with the sample temperature increasing as shown in Fig. 5.

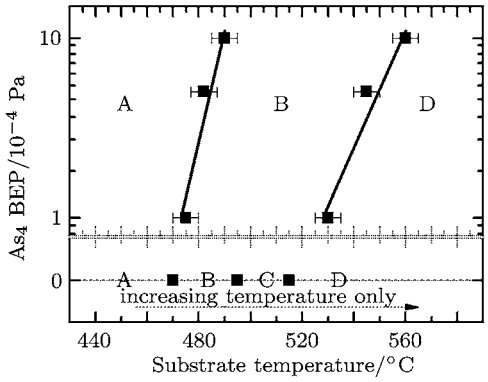


Fig. 5. RHEED-derived transition pressure (As_4 BEP) and temperature for the $In_xGa_{1-x}As$ surface.

(ii) Under a certain As pressure, in which case the In segregation in the $In_xGa_{1-x}As$ film is quite different from the above results, the most obvious difference is that the structure phase transited directly from state B to state D, and state C disappeared. The transition process became A→B→D, because the metal-rich surface would not appear under a certain As pressure. We also find that the transition temperatures from state A to state B were slightly different under different As pressures.

The transition temperature $T_{\rm c(A\to B)}$ was 475 °C for 1.33×10^{-4} Pa As_4 BEP, while $T_{\rm c\,(A\to B)}$ increased to 490 °C for 1.33×10^{-3} Pa As BEP. This suggests that As_4 BEP delayed the In segregation in $In_xGa_{1-x}As$ films. In the same way, the temperature $T_{\rm c(B\to D)}$ increased from 530 °C to 560 °C for 1.33×10^{-4} Pa and 1.33×10^{-3} Pa As_4 BEP, as shown in Fig. 5. This is because of the existence of As_4 BEP, where a large number of As atoms combine with the remaining In atoms when the In–As bond is broken. Under a given sample temperature and As_4 BEP, a balance between the breaking and the re-forming of the In–As bond can be reached. Only if the sample temperature continues to increase so that the balance is broken, will the In surface desorption be able to take place. At the same time, we also find that compared with the In surface segregation, In desorption depends strongly on As_4 BEP. The As_4 BEP affects the transition temperature strongly when desorption happens, surface segregation is more sensitive to temperature than surface desorption, and the segregation is less affected by As_4 BEP. This is probably due to the fact that In surface segregation is a process in which the In atoms gather to the surface from the inside of the sample, so it is more closely related to sample temperature; while surface desorption is a process in which In escapes from the sample surface after the In–As bond has broken, so it is affected more strongly by As_4 BEP.

4. Conclusions

In this paper, the surface segregation of $In_xGa_{1-x}As$ films is studied by the evolution of RHEED patterns under different values of As_4 BEP. When the As_4 BEP is set to be zero, with increasing temperature, RHEED patterns begin to evolve from the 4×3/(n×3) surface structure phase gradually, and surface segregation starts to take place at 470 °C.

The RHEED patterns develop into metal-rich (4×2) structures when the temperature reaches 495 °C, while when surface segregation is ended, surface desorption begins. At a temperature of 515 °C, the RHEED patterns turn into a GaAs (2×4) structure after In desorption has taken place. While the As_4 BEP is set to be a specific value, surface temperature can delay the segregation, and desorption will

be delayed. We find that As_4 BEP has a great influence on surface desorption, while surface segregation is more strongly dependent on temperature than surface desorption. This may be related to the physical mechanism within them. In this study, we use RHEED patterns to study the In segregation in an $In_xGa_{1-x}As$ film, which has a high directive experimental guiding significance and real application value for the preparation of high-quality $In_xGa_{1-x}As$ thin films and relevant $In_xGa_{1-x}As$ quantum-dot devices.

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