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GROWTH PROCESSES OF ZnTe EPILAYERS DEPOSITED BY MBE ON GaAs(100) VICINAL SURFACES — STUDIES BY STATIC AND DYNAMIC RHEED

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Static and dynamic reflection high energy electron diffraction (RHEED) has been applied for studying the initial growth processes of ZnTe crystallized by molecular beam epitaxy (MBE) on vicinal surfaces of GaAs(100) substrates. Atomically smooth ZnTe epilayers have been grown by MBE when *in situ* thermal desorption of the substrate protecting oxide layer was performed in the ultra high vacuum environment of the vacuum growth chamber just before the growth of ZnTe started. By gradual increasing of the substrate temperature of the crystallized ZnTe epilayers from 300°C to 420°C, when recording the RHEED intensity oscillations at these and eleven intermittent temperatures, it has been shown that the transition from the 2D-nucleation growth mechanism to the step-flow growth mechanism of ZnTe occurs at 410°C. Measuring periods of RHEED intensity oscillations recorded during the MBE growth processes it has been demonstrated that the growth rate of ZnTe at constant fluxes of the constituent elements decreases with increasing temperature from 0.37 ML/s at 300°C to 0.22 ML/s at 400°C.

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1. Introduction

It is well known [1] that atomically smooth epilayers deposited by MBE on off-oriented GaAs(100) substrates (exhibiting steps and plane parallel terraces, called vicinal surfaces) grow by two different mechanisms. The first one is defined by 2D-nucleation process occurring on the vicinal surfaces, while the second is defined by surface migration of adsorbed constituent atoms of the epilayer towards the steps. When the migrating atoms reach the step, they become incorporated

(at the step) into the crystal lattice of the epilayer. This is the step-flow growth mechanism. The occurrence of the definite growth mechanism depends on the temperature of the substrate and the structural parameters of the vicinal surfaces. We have used static and dynamic reflection high energy electron diffraction (RHEED) for studies of the initial growth processes [2] of ZnTe on vicinal surfaces of GaAs(100) substrates. The following two problems have been investigated in details: (i) control over transition from 2D-nucleation growth mechanism to step-flow growth mechanism, (ii) growth rates of ZnTe epilayers in dependence on temperature.

2. Substrate preparation procedure

The pre-growth preparation procedure of the substrate surface plays a crucial role in controlling the initial growth process of epilayers in heteroepitaxy. In our experiments with ZnTe growth by MBE three different procedures of pre-growth preparation of the 2° off-oriented GaAs(100) substrate surface have been investigated. As a qualification criterion for the preparation procedure, the static RHEED diffractograms have been used. It is well known [2] that atomically smooth surfaces are characterized by a streaky RHEED pattern, preferably with pronounced surface reconstruction features. On the other side, when the RHEED pattern characteristic for the investigated surface is spotty, the surface is rough on atomic scale.

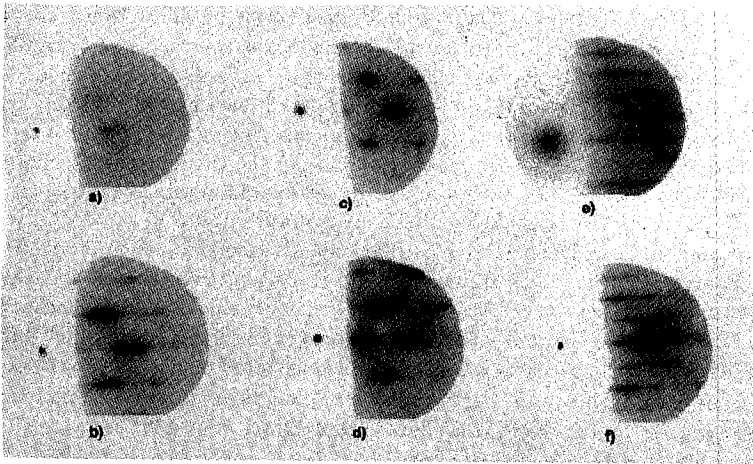


Fig. 1. RHEED diffractograms taken from (a)–(d) 2° off-oriented GaAs(100) substrates, pre-growth prepared according to different procedures; (e) 2000 nm thick ZnTe epilayer grown on the substrate (d); (f) 2000 nm thick ZnTe epilayer grown on the substrate (b).

Figure 1 shows RHEED diffractograms taken from GaAs(100) off-oriented substrate surfaces prepared according to the following procedures: (a) epiready surface (Sumitomo), covered with a protection oxide layer (at room temperature), (b) epiready surface (Sumitomo) after *in situ* thermally desorbed oxide at 580°C ,

(c) substrate produced by the Institute of Electronic Materials Technology, Warsaw (ITME), etched according to the following procedure: etching in $\text{NH}_4\text{OH} + \text{H}_2\text{O}_2 + \text{H}_2\text{O}$, 20:7:973 for 3 min, rinsing in deionized water for 3 min, etching in $\text{NH}_4\text{OH} + \text{H}_2\text{O}$, 1:10 for 15 s drying in pure nitrogen; RHEED after pre-growth annealing in ultra high vacuum (UHV) for 15 min at temperature increasing until 580°C , and (d) etched ITME substrate after annealing for 30 min at 580°C in Te flux (Te_2 beam equivalent pressure was equal to 10^{-7} hPa). Two ZnTe epilayers 2000 nm thick, have been grown on the two substrate surfaces, structurally best according to diffractograms shown in Fig. 1 (*in situ* deoxidized Sumitomo (Fig. 1b) and etched ITME with subsequent annealing in Te (Fig. 1d)). In both cases the MBE growth has been performed at 300°C when delivering the constituent elements from two effusion cells charged with solid Te and Zn, respectively. RHEED diffractograms taken from the epilayers are shown in Fig. 1e and Fig. 1f. Analyzing the RHEED diffractograms one may conclude that the pre-growth preparation procedure that allows one to obtain the ZnTe epilayers of the best quality on GaAs 2° off-oriented substrates is this one, in which the *in situ* thermal desorption of the protecting oxide has been employed. Here (Fig. 1f), a clear (2×2) reconstruction of the Zn-stabilised ZnTe surface may be recognised.

3. Growth mechanism dependence on the growth temperature

It is possible to explore the growth dynamics of MBE by monitoring temporal variations in the intensity of various features in the RHEED patterns (dynamic RHEED). Neave et al. [3] examined RHEED oscillations occurring during the growth of GaAs(100) substrates at increase of the substrate temperatures. The principal finding of this work was the progressing reduction in the intensity of the RHEED oscillations until a critical temperature when there are no oscillations. This effect has been interpreted as a transition from the low-temperature growth by 2D-nucleation on the vicinal surfaces to a high-temperature mode where the increased mobility of the adsorbed constituents of the grown compound (Ga and As) leads to direct incorporation of migrating adatoms into the growth front at the terrace edges. Thus, in a high temperature regime, the growth proceeds by step propagation (step-flow mode), for which the mean surface step density remains approximately constant, in contrast to the 2D-growth mode in which the step density varies as on a flat surface of an exactly oriented GaAs(100) substrate. The recorded sequence of measured intensity oscillations of the half-order feature of the RHEED pattern, occurring during MBE growth of ZnTe on Sumitomo epi-ready deoxidized GaAs substrates at increasing temperatures is shown in Fig. 2. One may clearly see from the presented oscillograms that the relevant transition temperature for ZnTe was equal to 410°C . To our best knowledge this is the first ever published experimental result concerning the MBE growth mode transition for ZnTe grown on vicinal surfaces of GaAs(100) substrates.

Having recorded RHEED intensity oscillations during the MBE growth, one may estimate the growth rate of the epilayer. It has been originally shown by Neave et al. [4] that the period of RHEED intensity oscillations exactly corresponds to the growth rate of a single molecular (Ga + As) monolayer of MBE grown GaAs.

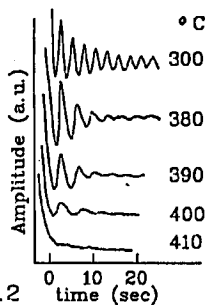


Fig. 2

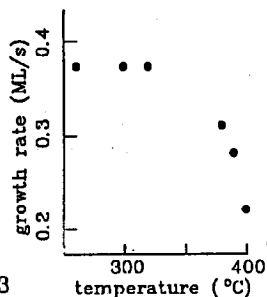


Fig. 3

Fig. 2. Oscillations of the 1/2 order RHEED feature recorded during MBE growth of ZnTe epilayers.

Fig. 3. Dependence of the growth rate of MBE grown ZnTe on the substrate temperature.

This principle holds also for other material systems [2]. We have found from data presented in Fig. 2 that the growth rate of ZnTe crystallized on vicinal surfaces of GaAs(100) substrates shown in Fig. 3 decreases with increasing temperature from 0.37 ML/s at 300°C to 0.22 ML/s at 400°C.

4. Conclusions

Growing by MBE ZnTe on substrate surfaces prepared by *in situ* thermal deoxidation we have obtained atomically smooth heterolayers with (2×2) surface reconstruction (Zn-stabilized surface). ZnTe heteroepilayers grow on vicinal GaAs substrates by the 2D-nucleation mechanism at the substrate temperatures below 410°C, while over this temperature the growth occurs according to the step-flow mechanism. Analysing the temperature dependence of periods of the RHEED intensity oscillations occurring during ZnTe growth, the growth rate decrease with increase in the substrate temperature, caused by desorption processes more intense at the higher substrate temperature, has been evidenced.

Acknowledgments

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