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Peculiarities of strain relaxation in linearly graded $In_xGa_{1-x}As/GaAs(001)$ metamorphic buffer layers grown by molecular beam epitaxy

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A R T I C L E I N F O

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A B S T R A C T

This paper presents a comprehensive study of structural, optical and electrical properties of heterostructures with linearly graded $In_xGa_{1-x}As$ metamorphic buffer layers (MBLs) grown by molecular beam epitaxy on GaAs (001) substrates. The low density of threading dislocations (well below 10^6 cm^{-2}) in 1-µm-thick $In_{0.3}Ga_{0.7}As$ layers grown atop of the linearly graded $In_xGa_{1-x}As/GaAs$ MBLs has been confirmed by using transmission electron microscopy (TEM). X-ray diffraction (XRD) data demonstrate good agreement between the experimentally measured In step-back and its calculations in the frames of existing models. Combining the XRD reciprocal space maps (RSM) of the structures and the spatially-resolved selective area electron diffraction measurements by cross-sectional TEM in depth-profiled RSM diagrams allowed direct visualization of the strain relaxation dynamics during the MBL growth. Strong effect of the azimuth angle and the value of an unintentional initial miscut of nominally (001) oriented GaAs substrate on the strain relaxation dynamics was observed.

1. Introduction

Heteroepitaxy of semiconductor alloys has opened up new opportunities for designing various device heterostructures containing lattice-mismatched strained layers. To achieve the best structural quality of a heterostructure the epitaxial growth should proceed on substrates lattice-matched to the main body of the heterostructure. Because of the discreteness of the lattice constants (a) of the III–V binaries (minimum $\Delta a/a$ step is ~3.3–3.5%) commonly used as substrates, the studies aimed at creating low-defect-density III–V templates with intermediate a values have been attracting great attention for many years. The employing of a metamorphic growth concept using compositionally graded alloy layers [1–3] allowed one to improve significantly characteristics of heterostructures grown on lattice-mismatched substrates as well as extend the functionality of semiconductor devices grown on GaAs substrates by molecular beam epitaxy (MBE) [4–6].

The compositionally graded layers are widely used in heteroepitaxial systems since late 60 s of the last century [7], and the $In_xGa_{1-x}As/GaAs$ system has been the most extensively studied one. To reduce the density of threading dislocations (TDs) after the metamorphic buffer layer (MBL), being a key parameter characterizing the MBL structures, different designs of the $In_xGa_{1-x}As/GaAs$ MBLs (different grading profiles and grading rates) were employed, as well as various MBE growth conditions (low [8] and high growth temperatures [9,10], using of different dopants during MBL growth [8,11,12], utilizing of As_2 and As_4 fluxes [13] etc.) were explored. Based on both experimental data and theoretical concepts, several models of strain relaxation in compositionally graded MBLs have been developed [13–20]. However, since the dislocation dynamics as well as the processes of stress relaxation in these structures have not been completely understood yet, while new factors affecting them appear, MBE growth of the MBLs is still attributed to a category of art.

X-ray diffraction (XRD) reciprocal space mapping (RSM) is commonly used for characterization of MBLs see e.g. [21,22]. Due to the difficulty in correlating the RMS angular positions to a certain depth of MBL Chauveau et al. [23] used TEM to check the MBL thicknesses. Based on the TEM thickness and RSM data they calculated and plotted depth profiles of residual strain in both [110] and [1–10] directions in the linearly graded InAlAs MBLs with a step of ~50 nm. The depth profiles of lattice mismatch, residual strain, crystallographic tilt, and the rocking curve full-width-at-half-maximum (FWHM) for metamorphic $In_xAl_{1-x}As$ buffer layers grown on GaAs substrates were also obtained from the XRD RSM [24]. The tilt angle was directly obtained from the 004 RSM data as a function of composition by assuming the monotonous composition variation across the sample thickness.

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This paper presents the results of MBE growth and comprehensive study of structural, optical and electrical properties of heterostructures with linearly graded $In_xGa_{1-x}As$ MBLs grown on GaAs (001) substrates. New sophisticated approach, combining XRD RSM technique at different reflections and selective area electron diffraction (SAED) measurements by TEM made along the growth axis at the structure cross-section, was applied to follow the details of stress relaxation process in the InGaAs MBL. An effect of the initial substrate miscut (within 1°) on the stress relaxation dynamics has been found. To characterize electrical and optical properties of the linearly graded $In_xGa_{1-x}As$/GaAs MBLs as well as the thick $In_{0.3}Ga_{0.7}As$ layer grown atop the Hall effect and optical transmission (OT) measurements were employed.

2. Designing the $In_xGa_{1-x}As$/GaAs MBL structures

It is well known that MBLs with monotonically changed composition (x) exhibit some residual strain near the surface. This residual strain produces a dislocation-free strained top region (or, more correctly, the region with low TD density) [14], while the rest of the graded buffer layer is nearly completely relaxed through the formation of misfit dislocations (MDs). The residual strain at the MBL surface (ε_s) should be taken into account during growth of the following structure by means of matching the equilibrium lattice constant of the subsequent (top) layer a_l to the in-plane lattice constant on the MBL surface ($a^{\parallel}_{MBL}$). In other words, it is necessary to reduce the composition (x) of the top $In_xGa_{1-x}As$ layer by the so called In step-back (Δx_0) for $a^{\parallel}_{MBL} = a_l$.

The linear and parabolic composition profiles in the $In_xGa_{1-x}As$/GaAs MBLs appear to represent the best compromise between reducing the TD density and preserving the small roughness of the MBL surface, in case of using optimum MBE growth conditions [13]. However, Romanato et al. believe that the parabolic profile is more preferable because it provides lower ε_s value, and a large part of this strain is frozen by the work hardening. So, this makes parabolic profile more stable to possible stresses induced by the following heterostructure layers. However, the parabolic profile is rather difficult to realize in MBE using standard thermo-controllers for effusion cells. On the other hand, the parabolic profile in $In_xGa_{1-x}As$/GaAs MBLs does not ensure precise calculations of ε_s (see, e.g. Table II in Ref. [13]) in contrast to the linear profile allowing the estimation of Δx_0 with a much higher accuracy.

According to the Dunstan model [19,20], the ε_s value of the linearly graded MBL does not depend on both the thickness of the graded layer and the maximum composition reached. The strained region near the MBL surface may be accurately quantified since the integrated strain-thickness product has the same value as for single layers, i. e. equal to the relaxation constant which is found to be $K = 0.8 \pm 0.1$nm for the $In_xGa_{1-x}As$/GaAs system [20] (0.83 nm [25]). Thus, if the product of the average residual strain in the linear grade ($\varepsilon_{av} = \varepsilon_s/2$) and the thickness of the strained region ($d_f = \varepsilon_s/\nu$) is equal to relaxation constant K ($d_f \cdot \varepsilon_{av} = K$), one can easily estimate both $\varepsilon_s = [2K\nu]^{1/2}$ and $d_f = [2K/\nu]^{1/2}$, where ν is the composition grading rate expressed as an equivalent strain per unit thickness. Taking account of $\varepsilon_s = \Delta\varepsilon \approx 0.07\Delta x_0$ (the lattice mismatch between InAs and GaAs is $\Delta a/a$~0.07), the optimum In step-back for $In_xGa_{1-x}As$ layer grown atop of the linearly graded $In_xGa_{1-x}As$/GaAs MBL should be equal to $\Delta x_0 = 0.083$ for the grading rate of 30 mol% In/μm corresponding to $\nu = 2.1\times10^{-5}$ nm^{-1} [25]. It should be noted that despite the relatively high scatter in the data reported on $In_xGa_{1-x}As$/GaAs MBL samples, Dunstan pointed out that majority of them being essentially perfect fell into the distribution with $K = 0.8 \pm 0.1$nm on the strain-thickness space [20].

Romanato et al. [13] argued that the Dunstan model overestimates the degree of strain relaxation, and for the correct estimation of ε_s (and, hence, the In step-back $\Delta\varepsilon = \varepsilon_s \approx 0.07\Delta x_0$) in $In_xGa_{1-x}As$/GaAs MBLs one should use a power law dependence (with n=2) in the strain relaxation equation ($h\cdot\varepsilon_s^2 = K_2$), the relaxation constant K_2 being equal to $K_2 = 3.7\times10^{-3} \pm 0.0007$ nm. The choice of the different integer powers in this equation reflects different interpretations of the mechanisms for generation of new MDs [13,20]. According to ref. [13], for the same grading rate of 30 mol.% In/μm, the optimum In step-back value should be slightly higher: $\Delta x_0 = 0.095$. It should be noted that the difference in the calculated In step-back values between the two models exceeds slightly the accuracy of MBE process, determined by the flux variations from one growth run to another (~0.5÷1 mol% of In content in $In_xGa_{1-x}As$). The true value for specific growth conditions can be obtained experimentally by using the above estimation as a first-order approximation.

3. Experiment

The linearly graded undoped $In_xGa_{1-x}As$/GaAs MBLs were grown by MBE on initially well-cut GaAs(001) ± 0.1° substrates, using a two-chamber MBE setup (SemiTEq, Russia). The general MBE growth conditions were chosen after Tångring et al. [8,11,12]: the substrate temperature (T_S) of 380–400 °C, the grading rate of 30 mol.% In/μm, the growth rate of (0.6÷0.8) μm/h, the V/III flux ratio of As/(In+Ga) ~1.5÷2. Both the "pure" (not overgrown) MBL layers with the linear composition profile and the structures comprising the 1-μm-thick $In_xGa_{1-x}As$ bulk layers on top of the linearly graded $In_xGa_{1-x}As$ MBL with different In step-back values $\Delta x \le \Delta x_0$ were grown and studied (see Table 1).

To implement the linear composition profile $x(d)$ (where d is the layer thickness) in $In_xGa_{1-x}As$/GaAs MBL we determined experimentally the dependence of an InAs growth rate on the temperature of the indium cell (T_{In}). This dependence at a fixed Ga flux defines both the local In content (x) and the growth rate $\upsilon_{InGaAs}=f(x)$ at any thickness of the MBE grown $In_xGa_{1-x}As$ MBL (Fig. 1a). To eliminate the non-linearity of $x(d)$ which occurs if simple linear T_{In} grading is used (so called LTG profile) (Fig. 1b, dashed line), the whole range of T_{In} variation was divided into several (up to five) unequal sections, and a specific T_{In} linear grading rate was set for each section..

XRD measurements were carried out by using multifunctional triple-crystal diffractometer D8 Discover (Bruker-AXS, Germany) in a parallel X-ray beam geometry. The tube with a rotating copper anode (λ=0.15406 nm) was used as a 6 kW X-Ray source. XRD ω–2θ diffraction curves measured around a (004) GaAs reflection as well as RSM around both symmetric (004) and non-symmetric (224) reflections were used for the analysis of crystalline properties of the $In_xGa_{1-x}As$/GaAs MBL structures. The (224) RSM maps were measured in a grazing incidence geometry.

The integral structural properties of the samples were studied also by TEM in a cross-sectional geometry by using a Philips EM-420 microscope. Electrical properties (the concentration and mobility of the charge carriers) of the $In_{0.3}Ga_{0.7}As$ layers grown atop the MBL were determined by using Hall effect measurements. The measurements of the OT spectra were performed in a standard geometry of passing the collimated light of a halogen lamp through the structure and comparing the transmission spectrum of the structure with that of the source lamp.

The SAED technique using a TEM setup Jeol JEM-2100F with an effective aperture diameter of ~100 nm was employed for depth profiling of both lattice mismatch and epilayer tilt along the cross-section of the $In_xGa_{1-x}As$/GaAs MBL structures. For that the SAED images were taken sequentially from the structure surface down to the GaAs buffer layer at the points spaced by ~30 nm, provided the appropriate sample orientation: a <110> axis is parallel to the electron beam. The spacing between (002) and (00-2) reflections corresponding to the planes parallel to the growth surface as well as that between (220) and (−2-20) reflections corresponding to the planes parallel to the growth axis were used for defining either vertical ($a^{\perp}$) or