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Crystal Growth of GaInP on (111)B GaAs by Metalorganic Chemical Vapor Deposition

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The structure and the mechanism of the roughening of lattice-matched GaInP crystal grown on an exactly oriented (111)B and a misoriented (111)B GaAs substrate by metalorganic chemical vapor deposition were studied. A multiple twin structure was revealed to form at the periphery of a primary twin, which was found to result in the rough surface of the exact (111)B GaInP. The suppression of a roughening by growth on a misoriented substrate surface would be responsible for the suppression of the primary twinning and further multiple twinning by the preferential growth at steps, which follow the stacking sequence.

1. Introduction

GaInP crystal is now used as an active layer in visible laser diodes with an emission wavelength around 670 nm. The ordered structure in GaInP epitaxially grown on a (001) GaAs substrate and the non-ordered crystal formation[1,2] in GaInP grown on a (111)B GaAs substrate have been reported. It has also been reported[3,4] that growth of AlGaAs on an exactly oriented (111)B surface gives a roughened surface, and that an epitaxial single crystal with a specular surface is obtained by tilting the substrate surface from the exact (111)B plane. The mechanism of the epitaxial growth on a (111)B surface, however, has not been clarified.

In this paper, the structure of GaInP crystals grown on an exactly (111)B-oriented GaAs substrate and a 2^{o} off (111)B GaAs substrate at 680^{o}C by atmospheric pressure metalorganic chemical vapor deposition (MOCVD) was studied by transmission electron microscopy, and the mechanism of the surface roughening by growth on exactly oriented (111)B surface and the drastic suppression of the roughening by growth on misoriented (111)B surface are discussed.

2. Experimental

Non-doped and lattice-matched $Ga_{0.5}In_{0.5}P$ crystals were grown on an exact (111)B GaAs substrate at 680^{o}C by MOCVD. And a (111)B-oriented GaAs substrate with the surface misoriented 2^{o} around the <1$\bar{1}$0> axis towards the <11$\bar{2}$> direction was also used. (110) cross sectional thin specimens of the GaInP/GaAs crystals were prepared using sputter-etching (fast Ar-atom bombardment) and were investigated by transmission electron microscopy.

3. Results and discussion

The surface of the GaInP layer grown on an exactly oriented (111)B GaAs substrate is roughened and cloudy, as can be seen in the Nomarski interference contrast optical micrograph of Figure 1. A number of protrusions or retractions less than 3.5 um in diameter are seen. A cross-section electron micrograph of the GaInP layer is shown in Fig.2(a), in which the retracted

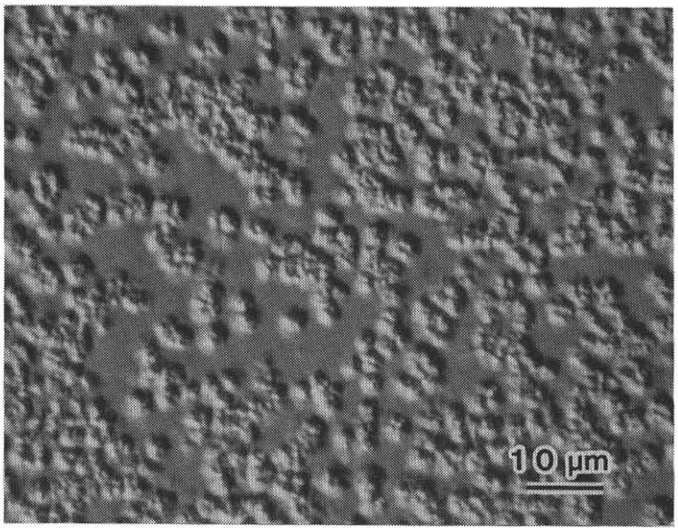

Fig.1 Nomarski interference contrst optical micrograph of the GaInP surface grown on an exact (111)B GaAs.

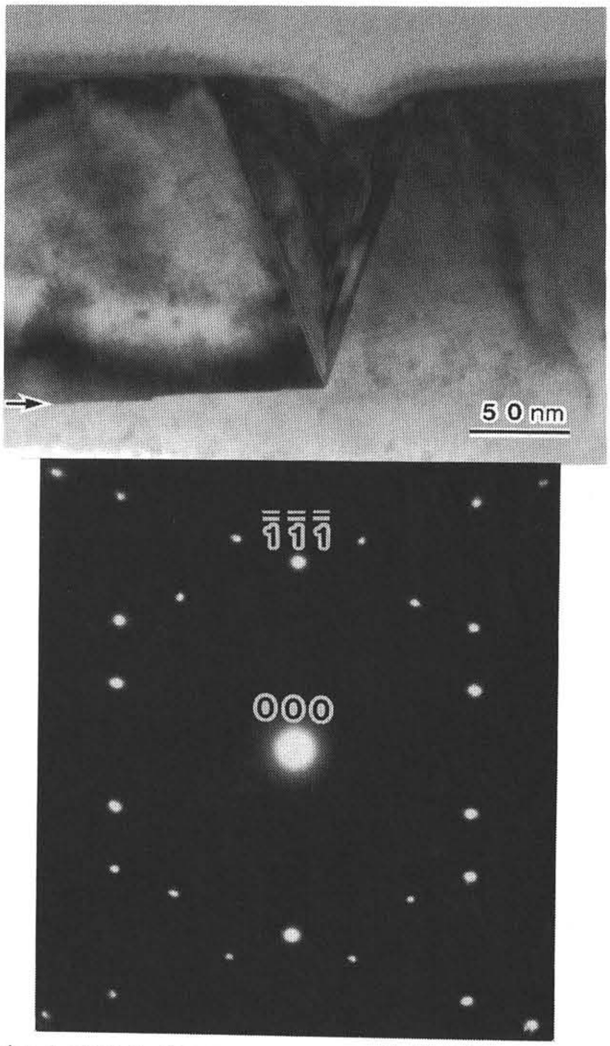

Fig.2 GaInP grown on an exact (111)B GaAs.
(a) cross-section transmission electron micrograph.
(b) electron diffraction pattern.

region 0.05 um wide at the surface may be part of a retraction or a protrusion in Fig.1. The corresponding electron diffraction pattern is complicated, as is shown in Fig.2(b). All of the spots are due to the sphalerite crystal structure, and no ordered structure spots are seen.

The pattern in Fig.2(b) was expalined to be caused by three kinds of twinned crystals and the parent crystal. One of them, which we denote the 'primary twin', is the twin with the $(\bar{1}\bar{1}\bar{1})$ twin plane parallel to the (111)B substrate surface. The others are the twins with twin planes inclined to the substrate surface. Because diffraction patterns with their twin spots were frequently observed at the rough regions, the protrusions or retractions in the optical micrograph must be related to the multiple twinning.

The primary twin boundary, as is indicated by an arrow in Fig.2(a), was frequently observed in the GaInP layer but not at the GaInP/GaAs interface. The anomalous V-shaped structure, whose surface is rough, was often observed at the end of the primary twin in the GaInP layer, as is shown in Fig.2(a). The V-shaped region forming a triangular domain shows a retracted surface.

A high-resolution transmission electron micrograph of a V-shaped region is shown in Fig.3. The V-shaped region contains four crystal domains. The lower region is the parent GaInP crystal, which has the same orientation as the (111)B GaAs substrate. The left region is the primary twin with the $(\bar{1}\bar{1}\bar{1})$ twin plane parallel to the (111)B substrate surface, and the region inside V-shaped region consists of two types of twins with the $(\bar{1}\bar{1}1)$ twin planes 70.5^{o} inclined to the (111)B surface. One is the secondary twin which has a twin-relation with the primary twin. The electron diffraction

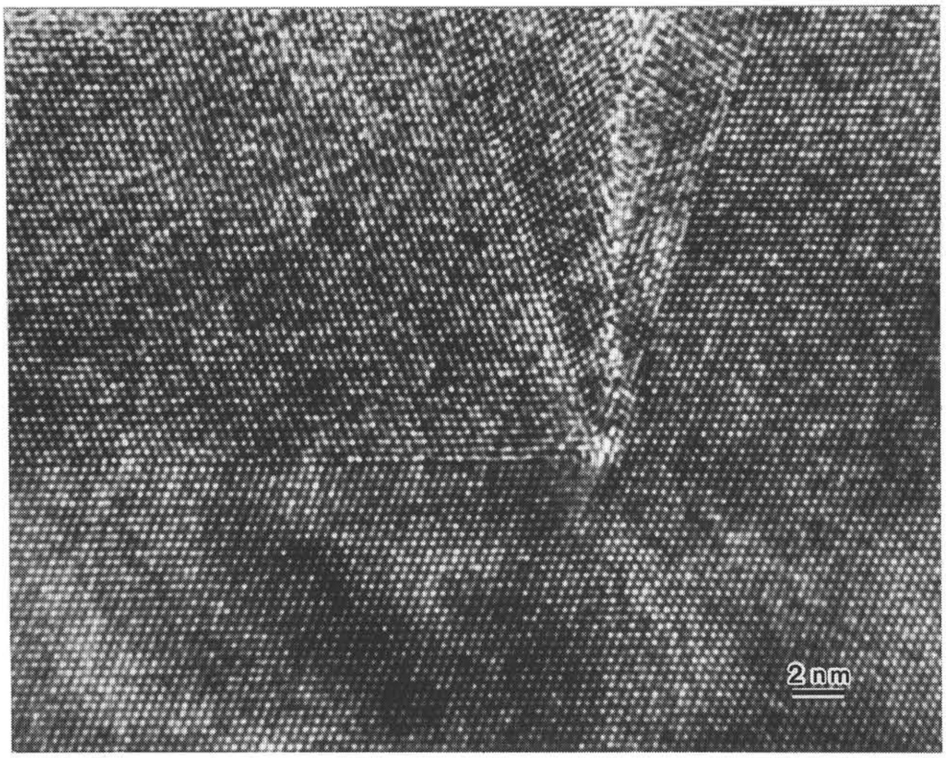

Fig.3 High-resolution transmission electron micrograph of the multiple-twinned region viewed along the <110> direction.

pattern expected from this multiple twin structure is the same as that in Fig.2(b).

The crystal planes inside V-shaped region parallel to the (111)B substrate is the (115) plane. Since the growth rate of the (115) plane is different from that of the {111} surfaces of the parent crystal and the primary twin, the surface would have the protrusion or the retraction. The size (3.5 um) observed in the optical micrograph, however, is much larger than the size of the V-shaped region. To understand this difference, the size of the primary twin must be considered. The V-shaped multiple twin is running in the $<1\bar{1}0>$ direction perpendicular to the cross-section micrograph. Since the equivalent directions to the $<1\bar{1}0>$ direction are $<\bar{1}01>$ and $<01\bar{1}>$ on the (111)B surface, the primary twin must be bounded by the V-shaped multiple twins along these three $<1\bar{1}0>$ directions, forming a triangular primary twin under plan-view observation. Triangular protrusions were often observed in optical micrographs near the periphery of the substrate wafer. The triangular region is presumed to be the primary twin bounded by the V-shaped twin. Therefore the size of the roughened regions is larger than the V-shaped twins and would be close to the size of the primary twin.

The growth was carried out on the misoriented (111)B substrate. A GaInP layer of a single crystal with a textureless specular surface could be successfully obtained.

A growth model on (111)B surfaces is proposed as follows. The adatoms supplied from the vapor phase migrate on the atomic terraces of the substrate surface and are incorporated at the surface steps, leading to the propagation of steps and crystal growth. Two-dimensional nucleation occurs on the terraces between steps when the diffusion length of the adatoms at the growth temperature is shorter than the inter-step distance, as is shown in Fig.4(a). The faulted (twinned) overlayer might accidentally happen on an exactly oriented (111)B surface with no step, since the difference of formation energy between two-dimensional twinned (Fig.4(b)) and untwinned (Fig.4(a)) nucleations would not be large. The faulted overlayer (primary twin) grows laterally and would meet the normal layer, as is shown in Fig.4(c). At the boundary the V-shaped multiple-twinning would occur, as shown in Fig.3. The surface of the primary twin and surrounding V-shaped multiple twins would be rough. On the misoriented (111)B surface, no two-dimensional island is formed because of enough steps to incorporate the supplied adatoms. With step-flow growth, the arrangement of atoms is restricted by the upper step structure, and therefore the structure of epitaxial overlayer coincides with the sequence of the parent crystal, as is shown in Fig.4(d). The advantage of growth on misoriented (111)B substrates is that it suppresses primary twinning and the subsequent multiple twinning, providing a single crystal with a specular surface.

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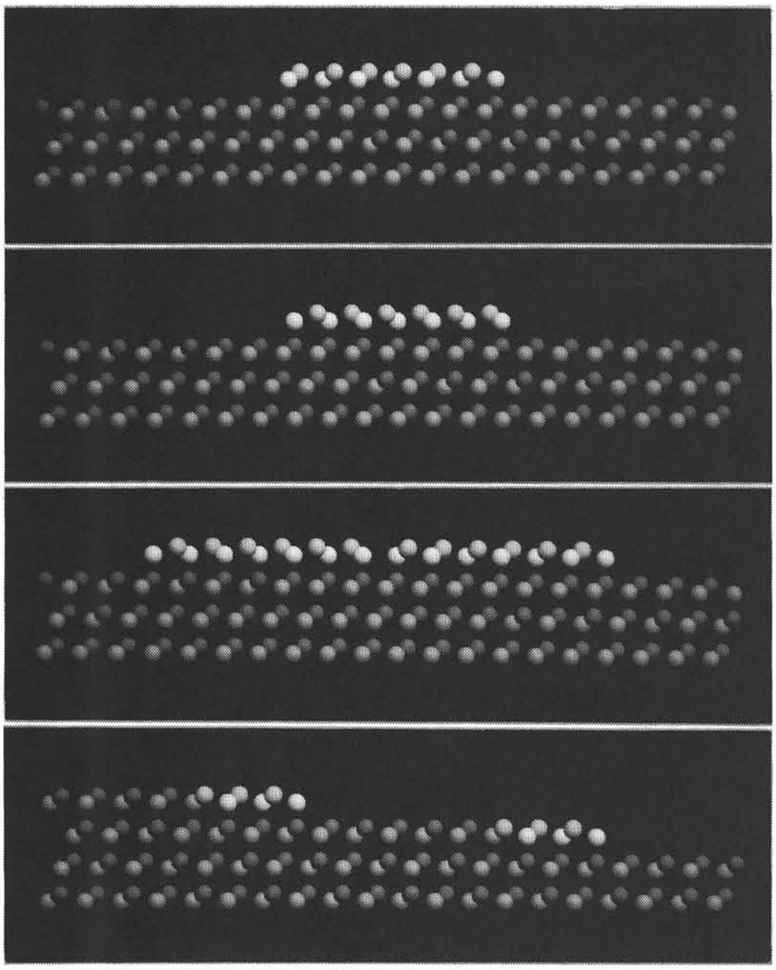

Fig.4 Graphical illustration of 2-dimensional nucleation formation and growth on (111)B surfaces. (a) Overgrowth with a normal stacking sequence, (b) overgrowth with relationship of twinning, (c)inital stage of multiple twin structure on an exact (111)B surface, and (d) lateral growth from the steps on a misoriented (111)B surface.