# Observation of lattice relaxation at the GaAsP/GaAs interface beyond the critical thickness by transmission electron microscopy

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The lattice relaxation at the $GaAs_{1-x}P_x$/GaAs interface is observed using thickness fringe images in transmission electron microscopy. The bending of the equal thickness fringes observed near the interface is explained, assuming that crystal planes are inclined near the interface and that the inclination has a maximum at the interface. The magnitudes of inclination and the thickness of the strained region are estimated for various phosphorous composition and the GaAsP thickness. The lattice relaxation mechanisms for GaAsP on GaAs is described. It is indicated from the thickness fringe observation that the lattice relaxation occurs gradually beyond the critical thickness.

## I. INTRODUCTION

The lattice-mismatched heteroexpitaxial growth of III-V compounds or alloys has been investigated for device applications. Homoepitaxial growth is preferable so as to avoid the generation of new dislocations at the interface. On the other hand, from the device application standpoint, heteroepitaxial growth is very attractive for producing new devices, such as high efficiency tandem solar cells, optoelectrical integrated circuits, and three-dimensional integrated circuits. The main problems with heteroepitaxial growth are the generation of threading dislocations at the interface and the strain caused by the lattice mismatch and/or the thermal expansion mismatch. The heteroepitaxial growth technology has been improved by using strained layer superlattice intermediate layers,[1] thermal cycle annealing,[2] etc. It is very important to control the dislocations and the strain furthermore in order to improve heteroepitaxial layer quality.

It is known that the lattice constant parallel to the interface coincides with that of the substrate with the tensile stress until the critical thickness in the case that the lattice constant of the epitaxial layer is smaller than that of the substrate.[3] The stress is relaxed, forming misfit dislocations beyond the critical thickness. There are some reports on the theoretical and the experimental critical thickness.[4–10] However, the lattice relaxation process beyond the critical thickness has not yet been investigated very much. This is due to the difficulty in observing the lattice relaxation feature near the interface.

Usually, the strain in the heteroepitaxial layer is measured as an average value for the whole layer using x-ray diffraction, photoluminescence, etc. However, the strain distribution is not expected to be uniform in the layer. The conditions at the top and the bottom layer are different because the top of the layer is exposed to the air and the bottom of the layer is attached to the substrate. The strain should change near the interface or the surface. It is also useful to study the strain distribution in the heteroepitaxial layer in order to clarify the strain relaxation mechanisms in the layer.

Kakibayashi and Nagata reported the compositional analysis of AlGaAs/GaAs multilayer structures by the thickness fringe method in the transmission electron microscopy (TEM).[11] Recently, they observed the strain distribution in InP/InGaP heterostructures by a similar method.[12] The presented results were limited to thin layers without misfit dislocations.

In this paper, we show that the bending of the equal thickness fringe is caused by the inclination of the crystal plane. Next, we describe the direct observation of lattice relaxation in GaAsP/GaAs heterostructures beyond the critical thickness by thickness fringes in TEM images varying phosphorous composition and the thickness.

## II. EXPERIMENT

GaAsP layers were grown by conventional atmospheric pressure metalorganic chemical vapor deposition (MOCVD). The source materials for Ga, As, and P were trimethylgallium, $AsH_3$, and $PH_3$, respectively. The substrate was $n$-type (001) GaAs. The growth temperature and the growth rate were 750 °C and 50 nm/min, respectively. The phosphorous composition ($x$) was varied from 0.06 to 0.25. Under these conditions, the lattice mismatch of the epitaxial layer and the substrate changed from 0.21% to 0.89%. The thickness was varied from 1 to 3 $\mu$m. Four samples were evaluated in this study. The GaAsP thickness $t$ and the phosphorous composition $x$ of the samples are shown in Fig. 1. The critical thick-

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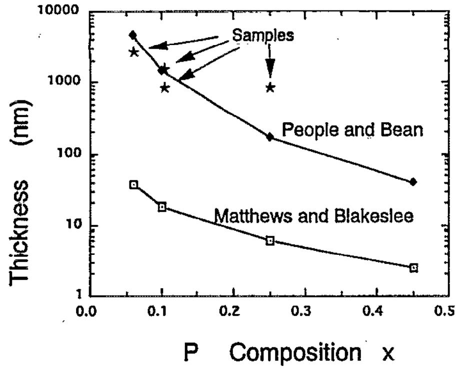



FIG. 1. Thickness and phosphorous composition of samples. The critical thickness is also shown in this figure.

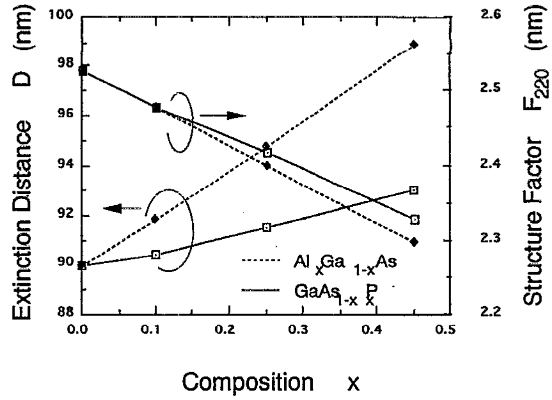



FIG. 3. Extinction distance $D$ and structure factor $F_{220}$ as a function of the composition $x$ for $GaAs_{1-x}P_x$ and $Al_xGa_{1-x}As$.

nesses calculated by People and Bean[7] and Matthews and Blakeslee[3] are also shown in this figure. In this system, the difference in the thermal expansion coefficient between GaAsP and GaAs is negligible.

A schematic illustration of the characterization method is shown in Fig. 2. The sample is first polished down to 150 $\mu$m, followed by cleavage to a size of 0.5×0.5 mm in the (110) and (1–10) planes. The sample is set on the TEM sample holder so that the electron beam is parallel to [100] direction. The small chip is slightly tilted around the [001] axis so as to keep the GaAsP/GaAs heterointerface parallel to the electron beam. The TEM operation voltage is 200 kV.

## III. CALCULATION OF THICKNESS FRINGE DISTANCE

First, we derive the calculated thickness fringe distance of $GaAs_{1-x}P_x$ for various $x$ by using the two-wave approximation of electron diffraction. The calculation is performed without taking account of the strain or deformation. The lattice constant of GaAsP is calculated using Vegard's law. The structure factor ($F_{hkl}$) and the extinction distance ($D$) for $GaAs_{1-x}P_x$ are expressed as follows:[13,14]

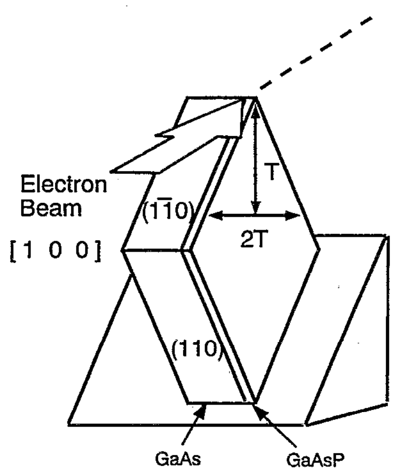



FIG. 2. Schematic illustration of samples used in the TEM observation.

$$F_{hkl}=4\{f^{\mathrm{Ga}}+i^{(h+k+l)}[(1-x)f^{\mathrm{As}}+xf^{\mathrm{P}}]\} \tag{1}$$

and

$$D=\frac{\pi m_0 V_0 v \cos\phi}{hF_{hkl}}\ , \tag{2}$$

where $f$ is the scattered amplitude of the individual atom, $m_0$ is the static electron mass, $V_0$ is the volume of the unit cell, $v$ is the electron velocity, $f$ is the Bragg angle, and $h$ is Planck's constant. The thickness fringe distance is calculated by $0.5\times D$. Figure 3 shows the calculated extinction distance and the structure factor $F_{220}$ of $GaAs_{1-x}P_x$ for various $x$ using a 220 wave. The calculated $F_{220}$ and $D$ of $Al_xGa_{1-x}As$ for various $x$ values are also shown in this figure. $D$ increases gradually with increasing phosphorous or aluminum composition. It has been reported that the abruptness of the AlGaAs/GaAs interface can be evaluated precisely by using the equal thickness fringe distance.[11] However, the equal thickness fringe distance for GaAsP is less sensitive to the P composition since the increase of $D$ with $x$ for $GaAs_{1-x}P_x$ is smaller than that for $Al_xGa_{1-x}As$. In the case of GaAsP grown on a GaAs substrate, the misfit strain increases with increasing P composition. Therefore, the thickness fringes of GaAsP are affected by the strain or deformation rather than the composition. The characterization results are described in the next section.

## IV. RESULTS

Figure 4 show the typical thickness fringe image for GaAsP grown on GaAs substrate. The phosphorous composition is 0.1 and the thickness is 1.5 $\mu$m. Misfit dislocations are observed at the interface since the thickness is over the critical thickness. The equal thickness fringes in the GaAsP layer and the GaAs substrate are bent near the interface. It is evident from Fig. 3 that the bending is too large to be explained by the fluctuation of the phosphorous composition during growth. Therefore other effects should be taken into account to explain the bending.

For further discussion, parameters such as $v_e$, $v_s$, $h_s$, and $h_s$ are defined. They are shown in Fig. 5 with the illustration of the thickness fringe image, $v$ corresponds to the

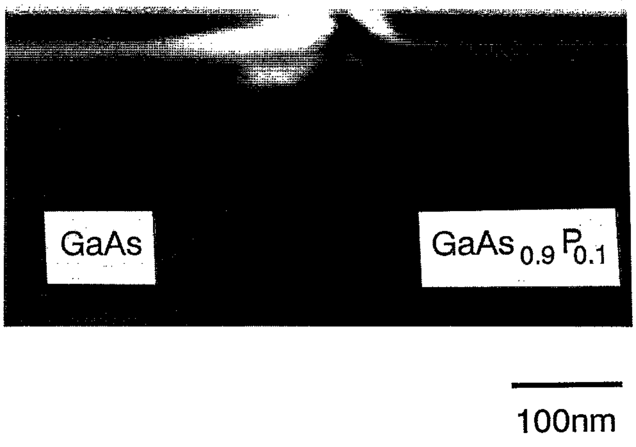



FIG. 4. Typical TEM image of GaAsP on GaAs. The thickness is 1.5 $\mu$m and the phosphorous composition is 0.1.

thickness of the region where the fringe is bended, and $h$ corresponds to the magnitude of the bending at the interface. Subscript $e$ means the epitaxial layer and $s$ means the substrate. The third fringe is analyzed in this experiment since it is more sensitive to the strain or the composition than the first or second one.

Table I shows $v$ and $h$ for various samples. The critical thickness $h_c$ was calculated by the equation obtained by People and Bean.[7] When $x=0.06$, $h_e$ and $h_s$ is small compared with other samples. $v_e$ and $v_s$ decrease and $h_e$ and $h_s$ increase with the increase of thickness from 1 to 1.5 $\mu$m or with the increase of $x$ from $x=0.1$ to 0.25 at 1 $\mu$m.

## V. DISCUSSION

First, the origin of the thickness fringe bending is discussed. When the lattice constants of the epitaxial layer and the substrate is different, the misfit strain is relaxed above the critical thickness by introducing misfit dislocations. If the

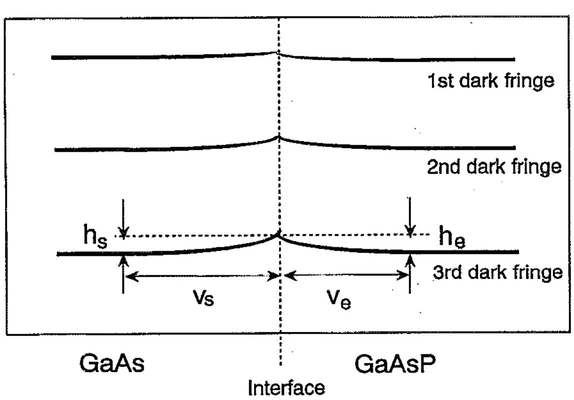



FIG. 5. Schematic illustration of the TEM thickness fringe image.

TABLE I. Summary of results obtained from the thickness fringe analysis.

| $x$ | $t$ ($\mu$m) | $t/h_c$ | $v_e$ (nm) | $v_s$ (nm) | $h_e$ (nm) | $h_s$ (nm) |
|---|---|---|---|---|---|---|
| 0.06 | 3 | 0.67 | 184 | 167 | 128 | 25 |
| 0.1 | 1 | 0.67 | 205 | 195 | 47 | 55 |
| 0.1 | 1.5 | 1 | 189 | 172 | 72 | 74 |
| 0.25 | 1 | 5.9 | 190 | 124 | 74 | 89 |

lattice mismatch is relaxed completely and the lattice is cubic even near the interface, the bending of the thickness fringe should not be observed. There are two possible ways to explain the bending near the interface. The ways are the difference in the stress variation near the interface applied to the GaAsP layer and the GaAs substrate (the variation of $V_0$ near the interface) and the variation of $\phi$ near the interface.

Because the lattice constant of GaAsP is smaller than that of GaAs, the tensile stress is applied to the GaAsP layer and the compressive stress is applied to the GaAs substrate. Therefore, assuming that the bending is caused by the stress, the equal thickness fringe should bend towards the different direction. In the case of compressive stress, $V_0$ is decreased, and in the case of tensile stress, $V_0$ is increased. In our case, however, the thickness fringes of GaAs and GaAsP are bended towards the same direction (towards the edge). Therefore, the stress is not a large factor to determine the thickness fringe bending.

Therefore, it is reasonable to suppose that the crystal planes are inclined near the interface. The inclination of the crystal plane at the interface is expressed by $\theta$. Since the electron beam is parallel to the [100] direction, $\theta$ can be regarded as $\phi$ in Eq. (2). When $\theta$ is increased, $D$ decreases, resulting in the bending of the thickness fringe. The cubic lattice of the epitaxial layer is deformed and the crystal planes become inclined on approaching the interface. The inclination has a maximum at the interface. $v_e$ and $v_s$ are the strained region of the GaAsP layer and GaAs substrate near the interface, respectively, and $h_e$ and $h_s$ correspond to the magnitude of crystal plane inclination for GaAsP an GaAs, respectively, at the interface. The quantitative discussion of $v$ is possible. However, the quantitative estimation of $\theta$ is difficult at present since the Bragg condition changes when $\theta$ varies. Only the quantitative comparison is possible. The determination of the crystal plane inclination $\theta$ for InGaP/InP heterostructures without dislocations has been reported with the use of the computer simulation of the thickness fringe image.[12] The similar simulation is necessary in order to determine $\theta$ of GaAsP/GaAs heterostructure.

The experimental results are summarized as follows:

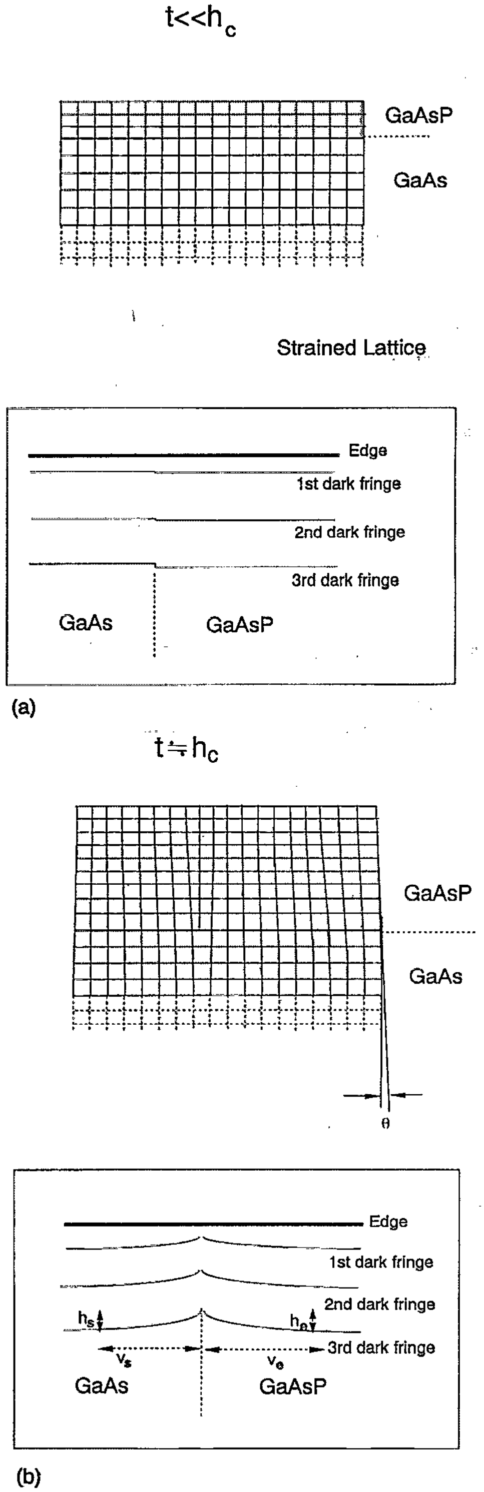
(1) $h_e$ and $h_s$ increase with the lattice mismatch.

(2) $v_e$ and $v_s$ decrease and $h_e$ and $h_s$ increase with increasing epitaxial layer thickness.

(3) $v_e$ and $v_s$ decrease with increasing lattice mismatch.

Since the tendency of $v_e$ and $h_e$ with increasing the thickness and the composition is similar to that of $v_s$ and $h_s$, respectively, we discuss about $v$ and $h$.

From the above mentioned results, the following lattice relaxation process is obtained as shown in Figs. 6(a)–6(c).



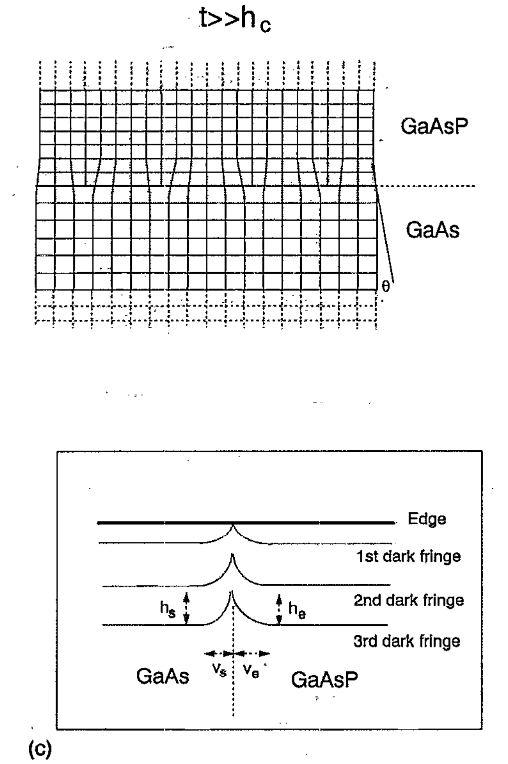




FIG. 6. TEM image and corresponding lattice relaxation of GaAsP/GaAs heterostructures. (a) $t \ll h_c$, (b) $t = h_c$, and (c) $t \gg h_c$.

When the epitaxial layer thickness is sufficiently thinner than the critical thickness [Fig. 6(a)], the lattice constant of GaAsP parallel to the interface coincides with that of the GaAs substrate. In this case, the bending of the thickness fringe is not observed and the equal thickness fringe distances change abruptly at the interface, which is the case of AlGaAs on GaAs.[11] With the increase of lattice mismatch or thickness, the misfit dislocations are generated at the interface to relax the lattice mismatch. The lattice is deformed at the interface, resulting in the inclination of the lattice plane. If the thickness is comparable to the critical thickness obtained by People and Bean, the strained region ($v$) is large and the inclination of the lattice plane is small [Fig. 6(b)]. This means that the misfit strain is not completely relaxed by the misfit dislocations. With the increase of the thickness, the strain region is reduced and the inclination of the lattice plane is increased with the sufficient number of misfit dislocations [Fig. 6(c)].

The relaxation of the lattice mismatch is classified as the elastic relaxation and the plastic relaxation. The elastic relaxation is caused mainly by the lattice distortion, and the plastic relaxation is caused by the generation of misfit dislocations. If sufficient misfit dislocations are generated to relax the lattice mismatch completely, i.e., the plastic relaxation is dominant, $v$ should have a minimum and $\theta$ has a maximum [Fig. 6(c)]. On the other hand, if the sufficient misfit dislocations are not generated enough to relax the lattice mismatch, i.e., the elastic relaxation component is increased, $\theta$ is reduced, and $v$ is increased [Fig. 6(b)].

The lattice relaxation features of the lattice mismatch vary for different lattice mismatch and thickness. It is indicated that the lattice mismatch is not completely relaxed by the misfit dislocations at $t=1$ $\mu$m and $x=0.1$ because $h$ increases and $v$ decreases with increasing thickness from 1 to 1.5 $\mu$m. It proves that the lattice relaxation occurs not abruptly but gradually beyond the critical thickness. $v$ also decreases and $h$ increases with increasing the composition from 0.1 to 0.25. This is due to the increase of the lattice mismatch. The small value of $h$ is obtained in the case of $x=0.06$.

## VI. CONCLUSION

The lattice relaxation for GaAsP grown on GaAs substrate was characterized by thickness fringe analysis in TEM. The careful measurement of thickness fringes shows that the crystal plane is inclined with approaching the interface and the inclination has a maximum at the interface. The strained region of the GaAsP layer grown on GaAs was estimated for various phosphorous composition and thickness. A lattice relaxation model for a GaAsP/GaAs heterostructure was proposed. The lattice relaxation occurs gradually beyond the critical thickness.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