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Incorporation mechanism of excess arsenic in MBE growth of GaAs at low temperature

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GaAs layers grown by MBE(molecular beam epitaxy) in a temperature range between 200 ℃ and 300 ℃ (LT-GaAs) contain excess As up to about 2 at.%, but possess high crystalline quality. According to reported equilibrium phase diagrams of GaAs, the solid solubility limit of excess As in GaAs is only about 0.1 at.% at most even near the melting point[1]. The high concentration of excess As in LT-GaAs, therefore, is considered to result from a unique surface process of the MBE growth. Because of a high concentration of excess As in a LT-GaAs layer, this material shows novel electrical and properties, and, hence, attracts considerable attention from a point of view of device applications, stimulating a large number of studies on this material in recent years[2,3]. In this thesis, a study aimed at clarifying the unique surface process of the MBE growth, through which a high concentration of excess As is incorporated into an epilayer. On the basis of the results reported in the thesis, one cannot only make precise control of the excess As concentration in LT-GaAs, but also obtain important insight of basic surface processes of the MBE growth.

LT-GaAs layers were grown in a temperature range between 200 and 310 ℃ with different As_4 fluxes at a growth rate of 0.9μm/h to a thickness of approximately 6000 Å. Concentrations of excess As in LT-GaAs layers were estimated from changes of a lattice spacing which were measured by using the X-ray diffraction technique[4]. In order to avoid a large error of substrate temperatures measured by a thermocouple placed at the back of a substrate holder, surface temperatures of epilayers were measured by using an infrared pyrometer through a quartz rod brought close to the substrate surface. Arsenic atom fluxes, which were supplied to a growth surface, were measured by using RHEED intensity oscillations at 320 ℃ where exactly a half of As atoms in As_4 molecules arriving at the growth surface are known to be incorporated into the growth layer according to a recent study[5]. In Fig. 1, As atom fluxes arriving at the growth surface, which were measured with the RHEED intensity oscillations, are plotted as a function of As_4 fluxes in BEP for both 320 ℃ and 530 ℃.

Fig. 2 shows changes of lattice spacings of LT-GaAs layers, $\Delta d/d$, as a function of the ratio of the As atom flux to the Ga atom flux for four growth temperatures. There is two main experiment results. The first one is that, at all employed growth temperatures,

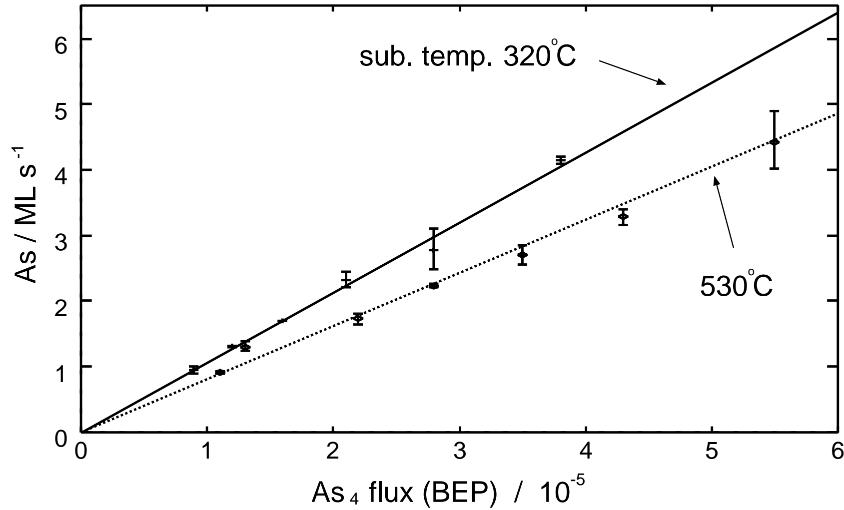

Fig. 1. Relationship of As atom fluxes which are incorporated into grown layers with As_4 fluxes in BEP. Data on the solid line and dotted line were obtained by the measurements of RHEED intensity oscillations at 320 °C and 530 °C, respectively.

stoichiometric GaAs layers are obtained under the flux condition where a number of Ga atom arriving at the growth surface is exactly equal to a number of As atoms resulting from a dissociating reaction of As_4 molecules at a GaAs surface. The other main result is a tendency of saturation of the concentration of excess As at high As_4 fluxes, and monotonous decrease of the saturation value with the rise in growth temperature. On the basis of these experimental results, a model, where chemisorbed As atoms on a As-terminated GaAs surface serve as precursor of excess As in LT-GaAs layer, is proposed. In this model, almost all chemisorbed As atoms desorb when Ga atoms newly arrive at the surface, with few As atoms incorporated into the crystal. The existence of the chemisorbed As atoms are suggested by the atomic structure of the c(4×4) surface reconstruction which occurs at a relatively low temperature during the MBE growth. In addition, in the temperature range above 270 °C, as expected from this model, the concentration of excess As is found to vary with an excess part of the As flux for the growth of stoichiometric GaAs in a manner similar to the Langmuir adsorption isotherm. This model provides an answer for the fundamental question of the MBE growth of LT-GaAs: why an extremely high concentration of excess As can be incorporated into an epilayer.

In order to examine compositions of growth layers and the occupation position of excess As atoms, we measured Raman scattering spectra of LT-GaAs layers grown in the above experiments. As a higher concentration of excess As is contained, a larger deviation from the selection rule is observed. This deviation depends only on the concentration of excess as in LT-GaAs Layers but does not depend on their growth temperatures. The result of the Raman scattering spectra suggest that excess As atoms do not simply occupy antisites. Stoichiometric LT-GaAs layers also exhibit the selection rule violation of, which was found to be caused by lattice defects in LT-GaAs layer by transmission electron microscopy.

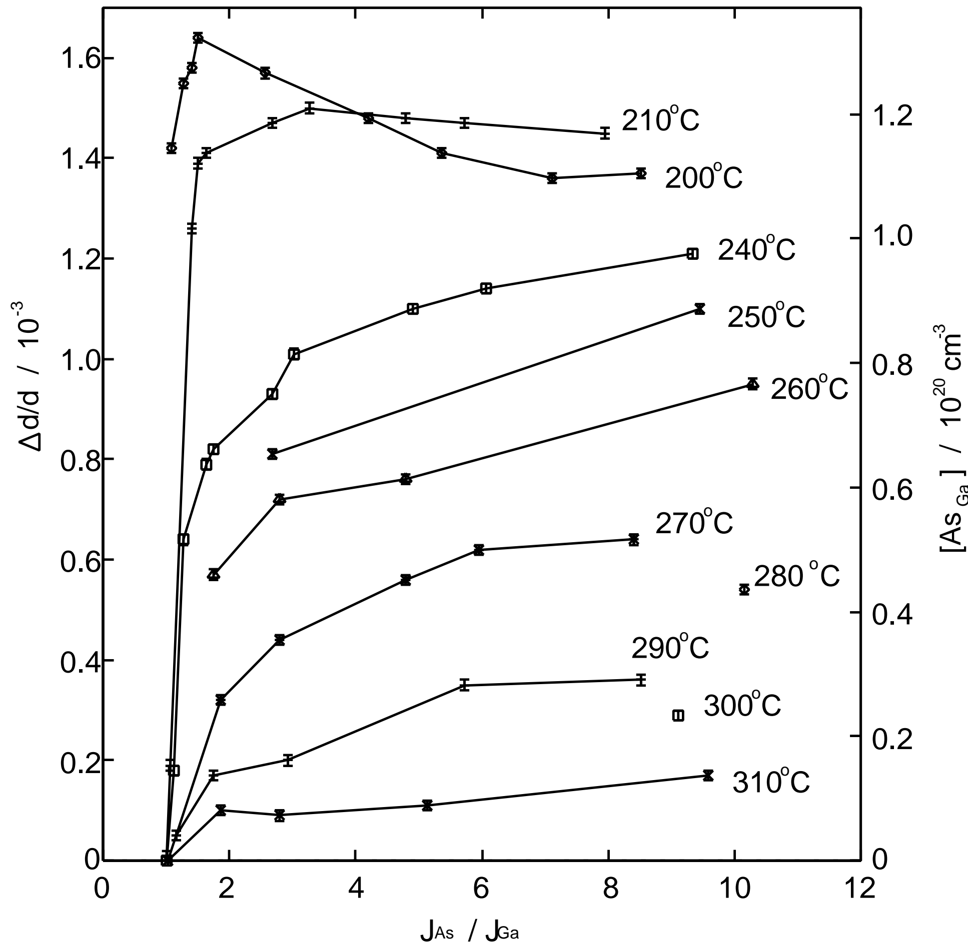

Fig. 2.Changes of lattice spacings of LT-GaAs layers as a function of the ratio of As and Ga atom fluxes. Concentrations of antisite As corresponding to changes of lattice spacings [4] are also shown in the figure.

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