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Examination of GaAs and InAs Langmuir evaporation by simulation

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Abstract. The process of GaAs and InAs substrates high-temperature annealing under the Langmuir evaporation conditions is studied by Monte Carlo simulation. The temperature range of gallium arsenide and indium arsenide congruent and incongruent evaporation are determined. It was demonstrated that the congruent evaporation temperature T_c is sensitive to the terrace width of vicinal surface and surface defects concentration. The terrace width decrease or the point defects concentration increase results in the congruent evaporation temperature decrease. The Ga and In diffusion length at congruent temperatures are estimated. The surface morphology transformation kinetics during high-temperature annealing is analyzed. The surface point defects influence on the metal droplet formation is demonstrated.

1. Introduction

In the process of high-temperature annealing of $A^{III}B^{V}$ semiconductor in vacuum (the Langmuir evaporation), due to the different volatilities of the material components, the surface relief develops and metal droplets form. An important parameter of the annealing process is the congruent evaporation temperature T_c, the temperature above which the V group elements evaporate more intensively than the elements of group III. A large number of experimental studies are devoted to the Langmuir evaporation of GaAs [1–3]. There is much less information on the high-temperature annealing of InAs [4]. It was experimentally shown that the GaAs congruent evaporation temperature depends on the surface orientation [2]. Literary data indicate a spread in the T_c values, even on the GaAs surfaces with a certain surface orientation.

In this work, the factors affecting the GaAs and InAs substrates congruent evaporation temperature are analyzed using Monte Carlo (MC) simulation. The model value T_c and the evaporation activation energy values E_{des} for different GaAs surface orientations are compared to the available experimental data. Similar dependences for InAs are obtained for the first time.

2. Monte Carlo model

The simulations were carried out using the program package SilSim3D on the base of kinetic lattice MC model. The elementary events included in the model are: desorption from the surface Me(s), Me(l) and As_2, As_2 dissociation and formation, diffusion of all components over the surface, metal phase transformations (melting, dissolution, crystallization), As dissolution and diffusion in a liquid metal. Here Me(s) and Me(l) are metals in solid and liquid states, and the metals are gallium or indium. The MC model realization detailes can be found elsewhere [5]. The choice of the model energy parameters was carried out according to the same procedure, as described in [5, 6].

3. Results and discussions

The simulations are carried out on the vicinal GaAs and InAs substrates in the temperature range of 800 – 1100 K for gallium arsenide and 600 – 900 K for indium arsenide. The terrace width L of vicinal surfaces varied from 20 nm to 360 nm. The dependences of As_2 and Ga or In evaporation rates on temperature during GaAs and InAs substrates high-temperature annealing are shown in Fig. 1. At $T > T_c$, the arsenic evaporation rate becomes higher than that of Ga or In evaporation rates. At these temperatures, metal droplets appear on the surface. At $T < T_c$, the arsenic evaporation rate is equal to gallium (indium) evaporation rate. From the plots of Fig. 1, the T_c values of GaAs and InAs are estimated for two substrate surface orientations and L = 20 nm: T_c (GaAs(111)A) = 930 K, T_c (InAs(111)A) = 650 K, T_c (GaAs(111)B) = 890 K, T_c (InAs(111)B) = 620 K. The congruent evaporation temperature on the (111)B surface is lower than that on the (111)A surface. This fact is confirmed by experiments on gallium arsenide [2]. For InAs the T_c value was experimentally determined only for the (111)B surface: T_c $(InAs)_{exp}$ = 630 K [4]. This value is in agreement with the model one. The Ga and In desorption activation energies are determined from Fig. 1. For the (111)A surface orientation they are: E_{des_Ga} = 2.5 eV, E_{des_In} = 2.3 eV.

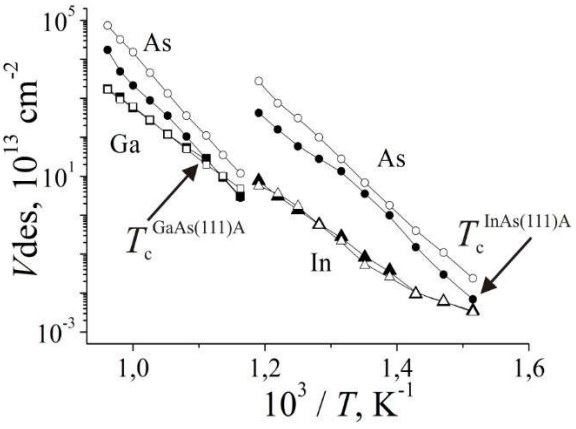

Fig. 1. *The temperature dependence of GaAs and InAs substrate evaporation rate for W = 40 nm and L = 20 nm; solid symbols correspond to the (111)A surface, open symbols – to the (111)B surface, Ga – squares, In – triangles, As_2 – circles.*

We previously showed that T_c depends on the vicinal surface terrace width [7]. This fact illustrated in Fig. 2. The smaller the terrace width L is, the lower the congruent evaporation temperature is. The step edges on the vicinal surface supply As and Ga (In) adatoms to the terrace. Two As adatoms can create an As_2 molecule. Gallium (indium)

atoms can evaporate from the surface; arsenic evaporates only in the molecular form. T_c corresponds to the temperature when metal droplets are formed on the surface. On ideal singular surfaces the adatom sources are vacancy islands (islands edges) formed on the surface during a high-temperature annealing. The vacancy island density is determined by the temperature. The average distance between the islands on the (111)A surface is equal to the double Ga (In) diffusion length before evaporation $\lambda_{dif\_Ga(In)}$. In Fig. 2 the maximal T_c corresponds to the congruent evaporation temperature on the singular surface. When the terrace width L becomes smaller than $2\cdot\lambda_{dif\_Ga(In)}$, T_c decreases, since step edges supply additional Ga (In) atoms on to the terrace and droplets can be nucleated at lower temperatures. So, in the plots of Fig. 2 $\lambda_{dif\_Ga(In)}$ can be estimated: $\lambda_{dif\_Ga} \sim 60$ nm, $\lambda_{dif\_In} \sim 130$ nm. The adatom diffusion length before evaporation can be estimated using a standard expression $\lambda_{dif} \sim a_0 \cdot \exp((E_{des}-E_{dif})/(2kT))$, where a_0 is the distance between the crystal surface atoms. Through this, the estimate at $T = 1000$ K $\lambda_{dif\_Ga} \sim 80$ nm and at $T = 870$ K $\lambda_{dif\_In} \sim 200$ nm was carried out. Both estimates show that, at the congruent evaporation temperature, λ_{dif_In} is approximately equal to twice λ_{dif_Ga}.

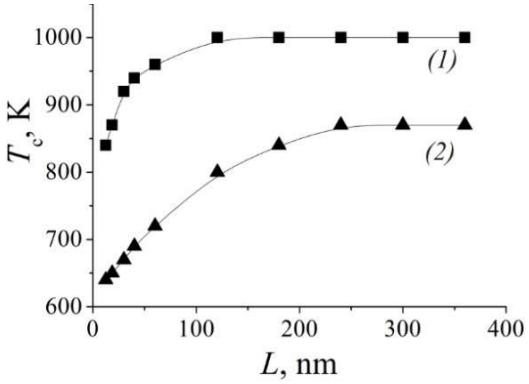

Fig. 2. *The congruent evaporation temperature of GaAs (1) and InAs (2) versus terrace length L for (111)A surface orientations.*

The congruent evaporation temperature on the (111)A substrate is sensitive to any surface defects, which initiate surface decomposition during the annealing. We analyzed the influence of point defects on the T_c value. With the increasing defect concentration, T_c decreases, but, after some critical concentration, it becomes constant. Again, it happens when the distance between the defects becomes smaller than $2\cdot\lambda_{dif\_Ga(In)}$. In the example of GaAs(111)A substrates annealing, we examined the metal droplets formation on an ideal singular surface and on the surface containing point defects. The top views of these surfaces after annealing at different temperatures are shown in Fig. 3.

The annealing duration corresponds to the initial stage of surface decomposition (less than one bilayer is evaporated). So, the annealing time decreases with the temperature rise. To achieve the same surface decomposition level, a different time is required for ideal and defected surfaces. There is some delay time τ_d for the vacancy island formation on the ideal surface. So, in Fig. 3, the ideal surface annealing time is longer than the defected one by τ_d. It is seen that for the ideal surface $T_c > 960$ K and for the surface with $n_{def} = 1.65\cdot10^{11}$ cm^{-2}, $T_c < 960$ K. Not every point defect transforms into a vacancy island, since the distance between the defects is shorter than λ_{dif_Ga}. The

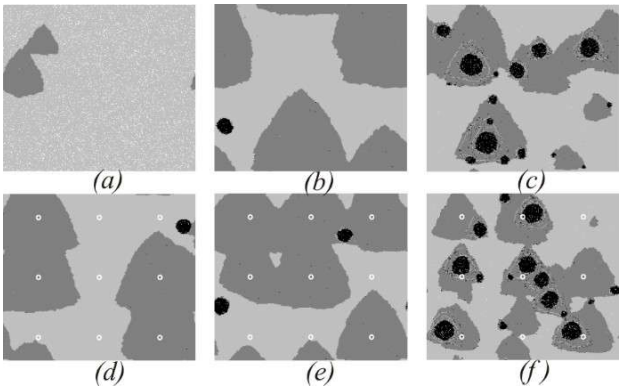

Fig. 3. *Top views of an ideal singular surface 120×120 nm (a-c) and surface with 9 point defects (d-f) at different temperatures: 960 K (a, d), 980 K (b, e), 1050 K (c, f).*

Ga atom, released from one point defect instead of evaporating, can embed in the closest point defect or returns to the original source. At high temperatures the droplet concentration does not depend on the initial defect concentration. At these temperatures, the vacancy islands expanded rapidly due to high gallium and arsenic evaporation rates. Since arsenic evaporates more intensively than gallium, several Ga droplets can be formed inside one vacancy island. The metal droplets are nucleated in the high-temperature annealing process and are usually located at the angles of vacancy islands. Moreover, one large Ga drop can be divided into several "daughter" drops. A similar fact was observed experimentally [3].

4. Conclusions

The GaAs and InAs Langmuir evaporation characteristics were analyzed using Monte Carlo simulation. The congruent evaporation temperature T_c for both materials with (111)A and (111)B surface orientations are obtained. The dependence of the T_c value on the terrace width of the vicinal surface is demonstrated. The Ga and In diffusion lengths, before the evaporation at congruent temperatures, are estimated: $\lambda_{dif\_In} \sim 2\cdot\lambda_{dif\_Ga}$. It is shown that the T_c value depends on the surface perfection; the presence of defects lowers the congruent evaporation temperature on the surfaces with the (111)A orientation. The gallium and indium desorption activation energies E_{des} of GaAs and InAs Langmuir evaporation are determined. The calculated T_c and E_{des} values obtained for GaAs are in agreement with the experimental data, and, for InAs, they are predictions of the model.

Acknowledgements

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