

Behaviour of RHEED Oscillation during LT-GaAs Growth

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Abstract: The behaviour of decay constant of RHEED oscillation during MBE growth on GaAs (001) surface at low temperature growth conditions is studied in this work. The dependence of decay constant on As-to-Ga ratio, substrate temperature and the excess of As content in the layer are examined here.

Keywords: MBE, RHEED, LT-GaAs

1 Introduction and Experimental Preliminaries

Nowadays, molecular-beam-epitaxial (MBE) growth of GaAs at low temperature (LT) – around 200 °C – has become an expanding important method since it provides highly insulating films and contributes to the synthesis of magnetic semiconductors [1]. It was shown that LT growth leads to incorporation of excess As in the crystal up to 1.5% depending on the growth parameters [2, 3]. The high concentration of excess As in LT-GaAs results in many new properties. As-grown and annealed LT-GaAs layers exhibit extremely high electrical resistivity and very short lifetimes of photoexcited carriers [4]. Their electrical parameters can be interpreted using the combined band and hopping conduction model [5, 6]. The majority of excess As is in antisite position, while the remaining As excess originate from interstitial As or Ga vacancies [7, 8]. The uniqueness of LT-GaAs is its high density of midgap states resulting from excess As, while the structure of the matrix remains perfect [9].

The use of reflection high-energy electron diffraction (RHEED) to control the growth of LT-GaAs has been reported in [10-12]. It is not easy to observe RHEED oscillations at LT growth. The RHEED oscillations are very strongly influenced by the growth parameters, such as deposition temperature, As-to-Ga ratio, etc.

RHEED oscillations were observed in two regions of As-to-Ga ratio at LT growth. One of these regions is close and another is far from the unity of As-to-Ga ratio. The strongest oscillations were observed when this ratio was nearly unity [10, 11]. Oscillations were also found if the ratio was larger than hundred [12].

The RHEED and its intensity oscillations at LT-GaAs growth exhibit certain particular behaviours. The intensity, phase and decay constant of oscillations depend on the As-to-Ga ratio, excess As content and substrate temperature, too. We investigate here the decay constant of oscillation during the growth of LT-GaAs. The deposition temperature and the range of the As-to-Ga ratio are 200 °C and 0.9-1.3, respectively. This investigation is based on the measurement and the observed intensity oscillations of RHEED which are described in the literature [9-11].

2 Results and Discussion

The temporal evaluations of RHEED specular intensity during the LT-GaAs growth – where the As-to-Ga ratio is close to unity – are shown in Figs. 1 and 2 of Refs. [10] and [11], respectively. It can be observed in these figures that when the As-to-Ga ratio moves off from unity, then the decay time of oscillations becomes small. If the ratio is 1.3 then the oscillation intensity becomes weak so its evaluation is difficult. The decay constant of the oscillations was determined as described in our previous work [13]. The amplitude decay of oscillations was investigated peak to peak. The peak to peak series are determined with the subtraction of the background. After the subtraction, an exponential function is fitted to determine the decay of intensity with the help of least-squares method. The exponential approximation of the time dependence of intensity is $I(t) = B_0 \exp(-t/\tau_d)$, where τ_d is the decay time constant and B_0 is a scaling factor. The extracted decay time constants vs. As-to-Ga ratio are shown in Fig. 1.

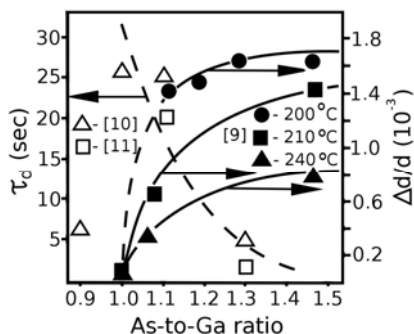


Figure 1

left: The decay constant vs As-to-Ga ratio at 200°C, *right:* The lattice spacing vs As-to-Ga ratio at 200, 210 and 240°C. The lines serves guide the eye only

It is known that the effect of the strain in the grown layer can be observed also with the help of RHEED oscillations. If the strain is large in the grown structure then the decay time constant is small. If the strain is small or absent in the structure then the decay time constant is large. This effect is demonstrated and described in the case of InGaAs/GaAs heterostructures – as a good model system – in our previous work [13, 14]. We can observe very strong change in the decay time constant depending on the As-to-Ga ratio at 200 °C (see Fig. 1). Depending on the growth parameters these LT-GaAs layers may contain large amount of excess As atoms. The majority of excess As is in the antisite position. The lattice spacing of LT-GaAs becomes greater than in the stoichiometric case. The relative increase of lattice spacing ($\Delta d/d$) of the non-stoichiometric LT-GaAs was determined in Ref [9]. The functions of $\Delta d/d$ vs As-to-Ga ratio are depicted also in Fig. 1.

We can observe (see Fig. 1) that the decay time constant of oscillations τ_d decreases rapidly during the LT-GaAs growth with increasing of As-to-Ga ratio, that is, also with increasing of the excess As content. The decay of oscillations can have several causes. The excess As gives rise to lattice mismatch, so also to strain, in the grown layer. This strain can influence the decay of intensity oscillations. At first, we investigate this effect because the strain effects are known and the influence of growth phenomena are unknown. From the given parameters (see Fig. 1) the mismatch dependence of the oscillation decay can be determined. The variation of decay time constant vs. $\Delta d/d$ is shown in the insert of Fig. 2. It is clear, that not only the mismatch is responsible for the decay but the growth conditions, too. Changes of the excess As modify not only the mismatch but the growth conditions, e.g. growth rate, too [15]. So, both the mismatch and the growth parameters influence the behaviour of the oscillation decay. We approximate this decay with an exponential function. Furthermore, we suppose that the both effects such as the mismatch and the growth influence can be separated from each other. In this way the decay phenomenon can be described by two time constants, as follows $I(t) = B_0 \exp[-(t/\tau_G) + (-t/\tau_M)] = B \exp(-t/\tau_M)$, where τ_G and τ_M are the assumed time constants of the separated influences, such as growth and mismatch, respectively. B and B_0 are the scaling factors which depend on the excess As and also on the $\Delta d/d$. The decay originating from the mismatch can be expressed as follows:

$$\frac{1}{\tau_M(\Delta d/d)} = \frac{1}{\tau_d(\Delta d/d)} - \ln\left(\frac{B_0}{B(\Delta d/d)}\right) \frac{1}{t} = \frac{1}{\tau_d(\Delta d/d)} - e(\Delta d/d)$$

where the factors are functions of $\Delta d/d$ and also of the As:Ga ratio and the term $e(\Delta d/d)$ serves as an operational aid only. In the case of stoichiometric LT-GaAs growth ($\Delta d/d=0$) there is no decay due to mismatch. This means, that for $\Delta d/d=0$ the reciprocal value of the decay time constant originates fully from the crystal growth phenomenon ($\tau_d(0)=\tau_G(0)=\tau_{Go}$), that is the value of $1/\tau_M(0)$ is zero. The

value of τ_{Go} is constant. The other component of τ_G , τ_{G1} depends on As-to-Ga ratio (or $\Delta d/d$), where the whole τ_G is $\tau_G = \tau_{G1}(As-to-Ga) + \tau_{Go}$. So the second term of the $1/\tau_M(\Delta d/d)$ expression $e(\Delta d/d)$ has also an independent and dependent part on As-to-Ga ratio (or $\Delta d/d$). The interchange between As-to-Ga ratio and $\Delta d/d$ may be only in the case of the narrow range of growth parameters where these ratios are proportional with each other.

We have separated the supposed strain effect from the effects due to growth which can be responsible for the decay of oscillation. In the case of InGaAs growth, we have supposed that the effect of growth remains constant in low In content region, because the one of the most important growth parameters, the deposition temperature, remained the same during the experiment. With this supposition we have obtained good agreement between the theoretical critical layer thickness and the threshold thickness, which is derived from the τ_M decay constant [14]. In the case of InGaAs In substitutes Ga in the lattice. Both elements establish similarly strong sp^3 type bonding in the lattice because the similar bonding structure. The situation in the case of LT-GaAs is quite different. The excess As which substitutes Ga in the lattice has different and weaker bonding than sp^3 hybrid. This fact modifies locally the probability of chemisorbtion of As atoms so also the probability of the incorporation of the further excess As atoms in the crystal [16]. The concentration of excess As can be determined from the chemisorbtion rate of As atoms. As atoms that are chemisorbed on the arsenic-terminated GaAs (001) surface serve as precursors of excess As, and hence, the concentration of excess As depends directly on the steady-state coverage of the chemisorbed As atoms [9]. The presented excess atoms As perturbs the bonding behaviour in the crystal, that is, the energy distribution along the surface. We use a simple description for the changing of the unperturbed surface layer by layer. At the first step, the unperturbed area A^* can be written as follows: $A_1^* = Ab - Aa$, where A is the whole area of the investigated surface. The factors b and a , which are less than one, give the areas on the surface which can be covered by chemisorbed As and which can be incorporate excess As, respectively. The second step can be described as follows: $A_2^* = (Ab - Aa)b - Aa$. The n^{th} layer we can get by follow-up the former given algorithm. The size of the perturbed area depends also on the number of the grown layers. This dependence can be negligible if the number of the layers is not large [16]. Among the surface reconstructions of the GaAs (001) surface, the $c(4 \times 4)$ reconstruction occurs at LTs under high As flux [17-20]. The value of b can be estimated because the maximum coverage of chemisorbed As atoms is 0.75 monolayers like in the case of this reconstruction. The value of a can be estimated by the maximum excess As content which is 0.015 [9]. It can be seen that the factor b is larger than a , so we can get, after arrangement of the expression A^* and neglecting small terms, the following simple power function for n^{th} step: $A_n^* = Ab^n$. We suppose that the intensity of RHEED is proportional to the size of the smooth part of surface. A continuous description by replacing of n by rt , yields

$I(t) = cA^*(t) = cb^{rt}A$, where r is the growth rate, t is the growth time and c is a constant characterizing the diffraction power. This can be written in the following form: $I(t) = cA\exp(-t/\tau_{GI})$, where τ_{GI} is the decay time constant originated from growth phenomena, which depends on the As-to-Ga ratio, this is, $\tau_{GI} = -1/r\ln b$. The τ_{Go} and τ_{GI} dependence on b are depicted in the insert of Fig. 2.

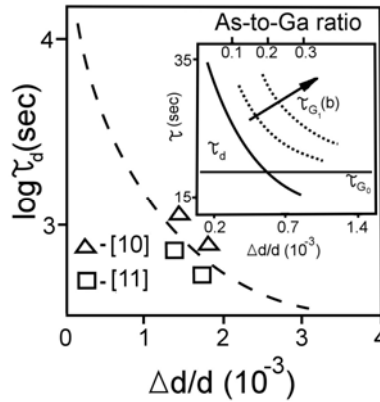


Figure 2

The function of τ_M vs $\Delta d/d$ which is derived from the high temperature InGaAs growth. The calculated data originated from the LT-GaAs growth. *insert*: The decay time constant vs lattice spacing derived from Fig. 1.

To justify our analysis we compare the values of τ_M extracted from the oscillation decay of LT-GaAs growth and the material independent decay constant, which is calculated from the effect of mismatch. The variation of $\tau_M(\Delta d/d)$ should be determined as follows: $1/\tau_M \propto 1/\tau_d - 1/\tau_{Go} - 1/\tau_{GI}$, similarly as described in Ref [14]. The strain dependent decay time constant vs composition in the case of InGaAs is given [14]. The composition independent variation of τ_M vs $\Delta d/d$ can be derived from the above mentioned dependence with the help of the modified Vegard's law [21, 22]. The material independent variation is shown in Fig. 2. The calculated τ_M data from LT-GaAs are depicted in this figure where the fitting parameter of b was 0.63. The value one of As-to-Ga ratio serves as a reference point for the calculation of τ_M . In this calculation we have taken into consideration also the As-to-Ga ratio of 1.3. The τ_M determined from LT growth corresponds to the calculated dependence, but we have to note here that the ratio of 1.3 is very difficult to evaluate. We can establish that the separation of the growth and mismatch effect on the decay of RHEED oscillations can describe the LT growth only in a narrow range. The intensity oscillation at the As-to-Ga ratio of 1.3 is very uncertain to evaluate because the weak intensity. This drastical intensity damage can result not only from the mismatch joined with the reduction of unperturbed area but it can be also explained the change of the sticking coefficient of the deposited species.

Conclusion

The decay and absence of the RHEED intensity oscillations at LT-GaAs growth can origin from several effects e.g. change of sticking coefficients, change of the morphology of the grown surface and change of the mechanical strain in the layer. Here was found, that the separation of growth and strain influence on the RHEED oscillation decay in the case of LT-GaAs is possible in a narrow region of As-to-Ga ratio.

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