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Application and Modelling of Synchronization of Nucleation by Means of Intermittent Radiant Heating

Mats I. LARSSON and Göran V. HANSSON

Department of Physics and Measurement Technology, Linköping University, S-581 83 Linköping, Sweden

The method of synchronization of nucleation (SN) by means of periodically altering the substrate temperature has been studied experimentally using RHEED. Results for SN applied to growth of Si and $Si_{1-x}Ge_x$ on Si(111) are presented. SN has also been studied theoretically by using a simple Monte Carlo simulation model and by applying a realistic temperature function. We predict that correctly applied SN should also improve the 2D growth character for Si on Si(100) epitaxy as was observed for growth on Si(111).

1. INTRODUCTION

We report studies of SN applied by intermittent radiant heating of 3" Si wafers during phase locked molecular beam epitaxy (MBE) of Si and $Si_{1-x}Ge_x$ on Si(111)-substrates [1]. SN by means of modulated resistive heating has previously been shown to improve the layer-by-layer growth for both Si and Ge [2]. We obtained generally an improvement in the detectable number of reflection high energy electron diffraction (RHEED) oscillations when applying SN compared to growth at constant substrate temperature (T_s) for both homo- and hetero-epitaxial growth. The experimental results are supported by Monte Carlo simulation results.

The experiments have been carried out in a Vacuum Generators V-80 Si-MBE-system. The cleaning procedure of the 3" Si wafers has been documented previously [3,4]. Both Si and Ge were evaporated by magnetically deflected electron beam evaporators. We used a 25 kV RHEED-gun, a diffraction geometry with an incident angle of ≈ 0.7° (kinematical in-phase conditions) and an azimuth ≈ 10° off-rotated from the <110>-azimuth. At this position the (0,1) beam in the zeroth Laue zone is exceptionally large and bright. By probing this reflex we could continuously monitor the RHEED oscillations while operating the magnetically driven shutters which are shifting the RHEED-pattern ≲ 1 mm. The RHEED-intensity oscillations were monitored with the data acquisition system LOCUS [1]. The base pressure was below $5 \cdot 10^{-10}$ Torr and the pressure during growth was typically on the low 10^{-9} Torr scale. All experiments have been performed on Si(111)-substrates and the substrate temperature was 450°C.

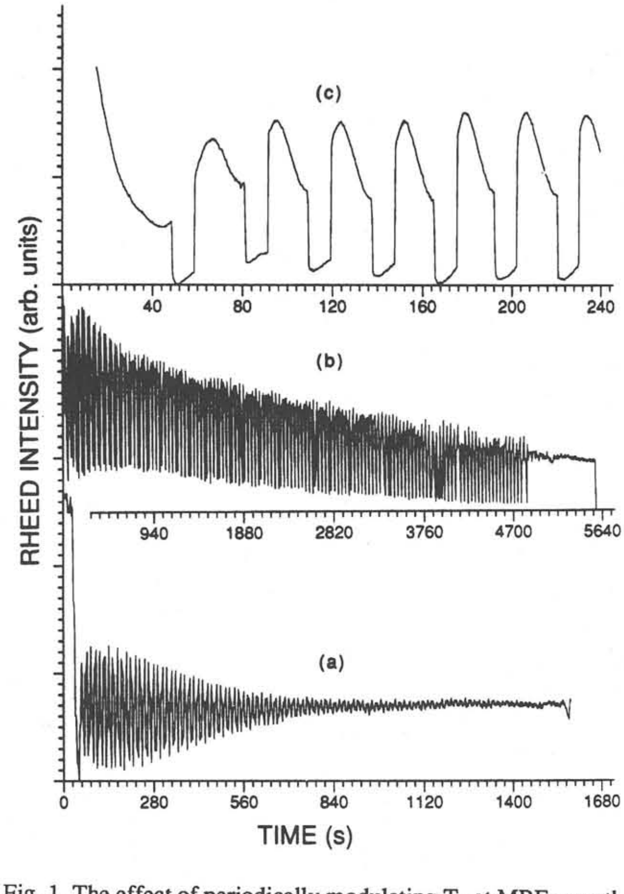

Fig. 1. The effect of periodically modulating T_s at MBE growth of Si on Si(111). (a) Approximately 100 RHEED-intensity oscillations obtained with constant T_s = 580 °C.
(b) Approximately 150 RHEED-intensity oscillations obtained with T_s varied between 530 °C and 570 °C. (c) The beginning of the SN experiment. The RHEED-beam is shifted when temperature modulation pulses are applied.

2. RESULTS AND DISCUSSION

Examples of RHEED oscillations from experiments with and without SN for growth of Si on Si(111) are shown in Fig. 1. Approximately 150 periods were acquired with and ≈100 without SN.

We have developed our Monte Carlo simulation model [5] in order to simulate SN by means of intermittent heating during Si MBE. The model is described in detail elsewhere [5], so the basic ideas are just summarized in this paper. The first step in the simulations is to choose a deposition site for the atom that is arriving at the surface. The newly-arrived atoms undergo a search process analogous to that described by Clarke and Vvedensky [6] in their study of aspects of low-temperature growth on Si(100). This search process is instantaneous in that it is completed before the deposition of the next atom.

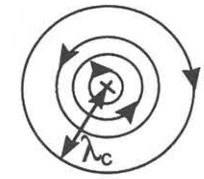

Fig. 2. The search for a change in the surface height is performed in circles with a maximum radius λ_c. A new starting-point and the direction of the trajectory are generated for circles of gradually increasing size.

In our model, the search is performed over a circular area $\pi\lambda_c^2$, where λ_c is the maximum adatom diffusion length see Fig. 2, which is given according to Irisawa et al. [7] by

$$\lambda_c = (D/J)^{1/4},$$

$$D = a^2 \nu \exp(-\frac{E_s}{kT_s})$$

and

$$J = RN, \qquad (1)$$

where D = surface diffusion constant, J = incoming atomic flux, a = nearest neighbor distance (3.84 Å), ν = atomic vibration frequency ($5 \cdot 10^{13}$ s^{-1}), E_s = activation energy for surface diffusion (1.5 eV), k = Boltzmann's constant, R = growth rate and N = number density ($5 \cdot 10^{22}$ cm^{-3}). There is unity sticking probability for diffusing adatoms at any height variation in the search area. When a height variation has been encountered, the adatom will be captured at the energetically most favorable site in the closest surface unit cell. Basically two selection rules are employed in order to do this. Formation of dimers has the highest priority and aligning the dimers into well ordered dimer rows has the second highest priority. Our computer representation of the surface lattice is similar to the one used by Barnett and Scott [8]. In all simulations a 160x80 matrix was used, with a one ML high step in the middle separating one 2x1 reconstructed terrace from one 1x2 reconstructed terrace of equal size. Periodic boundary conditions were employed at the boundaries of the matrix.

The simulated temperature pulse was activated on a predefined phase in the reversed step density oscillations. The correspondence between reversed step density oscillations and RHEED oscillations has been shown previously [9]. In the simulations we are using a temperature function based on Stefan-Boltzmann's radiation law which closely describes the experimental situation with a substrate in a two face geometry [10]. The most persistent simulated reversed step density oscillations are shown in Fig. 3a which are obtained by applying the increase in the temperature function in Fig. 3d phase locked to minima in the reversed step density curve.

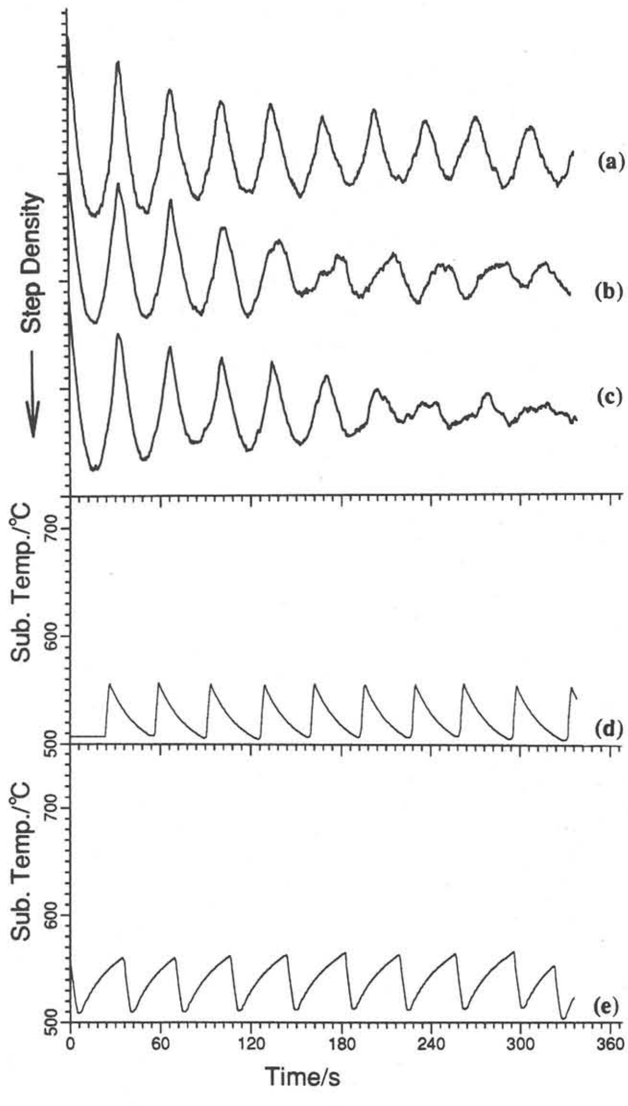

Fig. 3. The dependence of the simulated step density oscillations on applied T_S. (a) Step density oscillations simulated with modulated T_S according to (d). (b) Step density oscillations simulated with modulated T_S according to (e). (c) Step density oscillations simulated with constant T_S = 533 °C. (d) T_S describes a fast rise followed by a slow decrease. (e) T_S describes a fast fall followed by a slow increase.

The shape and phase of the temperature function is in this case similar to the experimental conditions under which the RHEED oscillations in Fig. 1b and 1c were acquired. The reversed step density curve (Fig. 3b) corresponding to decreasing T_s (Fig. 3e) phase locked to maxima in the reversed step density has faster decay of the oscillations. Finally, the curve presented in Fig. 3c is simulated for a constant $T_s = (T_{max} + T_{min})/2$ (calculated from the temperature function in Fig. 3d) shows the fastest oscillation decay. The effective T_s which can be deduced from the relation between the maximum diffusion length and T_s is roughly given by

$$\lambda_c \sim \exp(-E/nkT_s) \quad (2)$$

where n = 4 [7] or 6 [11] for moderate T_s. If T_s is a periodic function of t an effective maximum diffusion length can be expressed by

$$\lambda_c \sim \ln\{ \frac{1}{\tau} \int_0^\tau \exp(-E/nkT_s(t))dt\} = \exp(-E/nkT_{s,eff}) \quad (3)$$

where τ is the period time.

$$T_{s,eff} = - \frac{E}{nk} / \ln\{ \frac{1}{\tau} \int_0^\tau \exp(-E/nkT_s(t))dt\}. \quad (4)$$

can easily be derived from Eq. 3. By applying Eq. 4 to the temperature function in Fig. 3d $T_{s,eff}$ can be shown to be in close proximity to the arithmetic mean $<T_s>$. $T_{s,eff} = <T_s> \pm 1°C$ for $1 \leq n \leq 10$, where $<T_s> =$ 527°C.

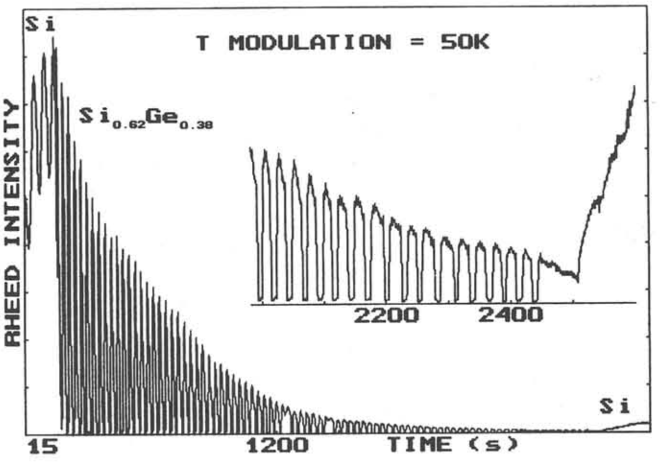

Fig. 4. RHEED-intensity oscillations acquired during the growth of a 277 Å thick $Si_{0.62}Ge_{0.38}$-layer with applied SN by means of modulating T_s.

Experiments were also performed to investigate how SN affects epitaxial growth of $Si_{1-x}Ge_x$-alloys on Si(111), see Fig. 4. The structure was grown on a Si(111)7x7 surface starting with a few biatomic layers Si, then 87 biatomic layers $Si_{0.62}Ge_{0.38}$ (277 Å) was grown followed by some additional layers of pure Si. A characteristic recovery of the RHEED intensity after the $Si_{1-x}Ge_x$ growth can be observed. 1000 W was applied to the front heater during 10 s, starting at each local minima in the RHEED-intensity, which gives the type of temperature function shown in Fig. 3d. The average T_s is then gradually increased to a higher level, due to the effect of combined heating by both the back and the front heater. The stabilized T_s varies between 520 and 570 °C during the temperature modulated part of the RHEED oscillations.

Our experiments support the idea that temperature modulation improves the 2D character of heteroepitaxial growth, since without SN we have never maintained the RHEED oscillations for growth of thicknesses exceeding very much the equilibrium critical thickness h_c [12] (h_c = 110Å for x = 38%). However, T_s is 70 °C to 120 °C higher when applying SN than in the case of growth at constant T_s. How this difference in T_s affects the damping of the RHEED oscillations has not been clarified. Nevertheless, we have observed that SN applied at approx. the same temperature range during homoepitaxial growth of Si on Si(111)7x7 was generally giving a slower damping of the RHEED oscillations, compared with growth at the corresponding constant $T_{s,eff}$ [2].

3. SUMMARY AND ACKNOWLEDGEMENTS

To conclude, this study predicts that SN should improve the layer-by-layer growth of Si on Si(100) if the method is applied similarly as was done for Si and $Si_{1-x}Ge_x$ growth on Si(111).

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