

Influence of N₂ background pressure on the incorporation of arsenic during molecular beam epitaxy growth of GaAs

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(Received 28 October 2003; accepted 5 April 2004; published 9 June 2004)

The influence of a high N₂ background pressure on the molecular beam epitaxy growth of GaAs has been investigated. Measurements to determine the minimum As₄ pressure necessary to maintain stoichiometric growth at different substrate temperatures with and without a high N₂ background pressure were performed. The As₄ pressures required for cases when a high N₂ background was present were systematically above those required without a N₂ background. The GaAs growth process has been modeled using kinetic rate equations and by including surface site blocking terms the model accounts for the data taken by the authors. The model also agrees well with GaAs growth kinetic data published by several other authors. © 2004 American Vacuum Society.

[DOI: 10.1116/1.1755712]

I. INTRODUCTION

MBE growth of the dilute III–V nitride compound semiconductor GaAsN on GaAs(100) substrates often uses rf or microwave plasmas as sources of atomic nitrogen.^{1–3} The nitrogen plasma source used in our MBE system produces a N₂ background pressure during normal operation of the source in the range of $1–6 \times 10^{-5}$ Torr since only a small fraction of the beam emitted from the source is in the form of atomic nitrogen.

Many studies of GaAsN have found that the optimal growth temperature is around 480 °C, much lower than the 600 °C typically used for the growth of GaAs, at which temperature the minimum As₄ flux required to maintain stoichiometric growth is expected to be reduced due to lower As desorption losses. However, we have found that at 480 °C the minimum As₄ flux required to maintain stoichiometric growth, although reduced, is significantly higher for the growth of GaAsN than for GaAs grown at the same substrate temperature.

To investigate this effect we have first measured the minimum As₄ flux required to maintain stoichiometric growth of GaAs as a function of substrate temperature in the absence of significant N₂ in the chamber. Further measurements were performed to determine the minimum As₄ flux required in the presence of N₂ at a background level of 6×10^{-5} Torr. During these measurements the plasma was not ignited, thus GaAs was grown with the only influence of the plasma source being the high background level of N₂ it added to the growth environment.

II. EXPERIMENT

All GaAs(001) substrates were chemically degreased, lightly etched and indium soldered onto molybdenum substrate holders before being loaded into the growth system, which was a highly modified Varian MBE-360 system pumped by a 500 l/s ion pump giving a residual gas pressure

of $< 10^{-9}$ Torr. The substrate temperature was measured by a thermocouple positioned close to the back of the substrate holder and was calibrated against the transition between the $c(4 \times 4)$ and (2×4) reconstructions at a temperature of 530 °C. The temperature was therefore accurate to ± 5 °C with a short term stability of ± 0.2 °C. This transition temperature is weakly dependent on the As₄ flux and since our experiments required different As₄ background pressures this calibration was performed each time the As source temperature was changed. The N₂ flux was adjusted by a leak valve and controlled by monitoring the mass 28 peak using a mass spectrometer. The nitrogen plasma source power supply was turned off during all of the experiments to prevent any cracking of the N₂.

Following oxide removal from all samples, a 0.25 μm GaAs buffer layer was grown at a substrate temperature of 600 °C and a growth rate of 1 $\mu\text{m}/\text{h}$ with an As₄ overpressure. The samples were then set to the required substrate temperature for the experiment and the As₄ source temperature was set to achieve the desired flux. The As₄ pressure was determined using a monitoring ion gauge in the sample position. The Ga shutter was then opened for 10 min allowing the growth of a GaAs layer at the required substrate temperature and As₄ flux. The Ga flux was calibrated to a growth rate of 1 ML/s at 600 °C as determined by GaAs reflection high-energy electron diffraction (RHEED) oscillations. At the end of this growth period the Ga shutter was closed and the substrate heater turned off.

Once the indicated temperature reached 100 °C the sample was inspected visually through one of the view ports in the MBE chamber. If the surface still appeared mirror smooth, the sample was heated back up to the required substrate temperature and the procedure was repeated with a lower As₄ flux. Once a dull surface exhibiting obvious non-specular reflection was found, it was clear that the As₄ flux must have dropped beneath the minimum necessary causing the formation of Ga balls. The experiment was then terminated and the next sample was loaded. The minimum necessary As₄ flux was taken to be the average of the last growth

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that did not and the first growth that did produce a dull surface.

This procedure was repeated for substrate temperatures of 400, 480 and 600 °C and all experiments were performed once with the plasma source N₂ leak valve closed (background N₂ pressure < 1 × 10⁻⁹ Torr) and once with a N₂ background pressure of 6 × 10⁻⁵ Torr as determined by the mass spectrometer. Secondary ion mass spectrometry (SIMS) measurements were performed on samples grown under similar conditions as described above and no evidence of nitrogen incorporation was found.

III. RESULTS

The experimental data in Fig. 2 show a systematic decrease of the necessary minimum As₄ pressure with decreasing temperature as well as a systematically higher necessary As₄ pressure for the case when N₂ is present in the growth chamber. This behavior cannot be explained by interference of the N₂ beam with the As₄ beam as it passes from source to substrate since the influence of the higher N₂ background pressure on the mean free path of the As₄ is not significant. This effect, however, can be explained in terms of physisorbed N₂ occupying surface sites which reduces the available sites for As₄. To test this hypothesis a rate equation based model for GaAs growth has been developed which accounts for the blocking effect of a high residual background gas pressure.

IV. GaAs GROWTH MODEL

The aim of this model was not only to produce a rate equation model that would explain our findings but also to agree with GaAs growth measurements published by other authors (e.g., Refs. 4–6).

A. Definition of states

Foxon and Joyce⁷ suggested that the chemisorption of As₄ involves a physisorbed state wherein the As₄ molecules are first weakly attached to the surface before a dissociative chemisorption process takes place. In the physisorbed state no electron transfer takes place and the molecules maintain their identity.⁸ In this article, we include a physisorbed state not only for the As₄ molecules but also for the Ga atoms, since they are believed to be a fast diffusing species during the growth of GaAs. If the Ga were to chemisorb directly from the vacuum, the chemical bonds formed with the substrate would slow this diffusion process considerably.

In the chemisorbed state molecular bonds are broken and the atoms attach via chemical bonds to the surface. The bonds of the chemisorbed atoms, however, are not in the same configuration as in the bulk material since they terminate the surface. It is well known in MBE that at higher temperatures or if the Ga flux exceeds the available As₄ flux, Ga balls or clusters are formed on the surface.⁹ For this reason, and since we are observing the threshold As₄ pressure to avoid such clusters, a chemisorbed Ga cluster state is included in this model as well as an As chemisorbed state. The atoms in the Ga cluster state will form clusters or balls and

destroy the sample if the population in this state exceeds a critical value, beyond which the sample cannot be recovered by exposing it to an As₄ overpressure for an extended period of time.

As or Ga atoms that are chemisorbed onto the surface (and are not in the cluster state) are considered to have been incorporated if they are subsequently covered with an additional layer of the alternate atomic species and therefore have the same bonding configuration as in bulk material.

B. Blocking of sites

Molecules or atoms in the physisorbed state diffuse on the surface before either desorbing or encountering a suitable site for chemisorption. However, if another species (such as N₂ in our case) is also in a physisorbed state, these atoms or molecules will occupy a certain fraction of the suitable sites thereby decreasing the probability of chemisorption of any diffusing species by a factor (1 - ϑ_i) where ϑ_i is the fractional coverage of each *i* species occupying the physisorbed state. Xie *et al.*¹⁰ used a similar concept to describe the blocking effects of hydrogen surfactant atoms. Since N₂ will not crack on the surface and therefore will not chemisorb, the blocking due to N₂ takes place in the physisorbed state.

C. Physisorbed As₄ state

As₄ molecules arrive into this state from the vapor at the rate F_{As_4} [in monolayers per second (ML/s)]. They can then either re-evaporate into the vapor or be chemisorbed onto the surface. The rate of desorption of As₄ molecules depends on the population in the physisorbed state (ϑ_{As_4} in monolayers) and the desorption time constant of the As₄ which is assumed to be of the form

$$\tau_{des,As_4} = f_0^{-1} \exp(E_{des,As_4}/kT), \quad (1)$$

where f_0 is the attempt frequency which is taken to be the Debye frequency, k the Boltzmann constant, T the temperature in degrees Kelvin and E_{des,As_4} the energy barrier to desorption from the physisorbed state.

For the chemisorption of As₄ we assume the process suggested by Foxon and Joyce.⁷ In this case two As₄ molecules meet in the physisorbed state in the vicinity of four sites which are available for the chemisorption of As. The As₄ molecules then break up with four of the eight atoms chemisorbed onto the surface while the other four form an As₄ molecule that desorbs into the vapor.

The rate of chemisorption will therefore depend first on the probability of two As₄ molecules meeting, which is proportional to the square of their populations, multiplied by the diffusion constant,¹¹ for which the time constant is labeled τ_{chem,As_4} with a definition similar to Eq. (1) but using the energy E_{chem,As_4} . Second, the rate of chemisorption will depend on the number of available sites and, since four sites are necessary for this process, it is proportional to the probability of finding four adjacent free sites, which is (1 - Θ_{As})⁴ where Θ_{As} is the population of As in monolayers in the chemisorbed state. Third, since as described in Sec. IV B the path

to chemisorption can be blocked by physisorbed Ga or N₂, the factor $(1 - \vartheta_{\text{Ga}})(1 - \vartheta_{\text{N}_2})$ must also be included, where ϑ_{Ga} and ϑ_{N_2} are the physisorbed populations of Ga and N₂, respectively. For GaAs it has been found¹² that above ≈ 300 °C there is no reverse pathway for As₄ to desorb from the GaAs surface and so it is assumed that no As₄ can return from the chemisorbed state into the physisorbed state under standard growth temperatures. Therefore the rate equation controlling the As₄ population in the physisorbed state can be expressed as

$$\dot{\vartheta}_{\text{As}_4} = F_{\text{As}_4} - \vartheta_{\text{As}_4} \frac{1}{\tau_{\text{des,As}_4}} - 2D_{\text{As}_4} \vartheta_{\text{As}_4}^2 \times (1 - \Theta_{\text{As}})^4 \frac{1}{\tau_{\text{chem,As}_4}} (1 - \vartheta_{\text{N}_2})(1 - \vartheta_{\text{Ga}}). \quad (2)$$

The factor of 2 in the chemisorption term represents the fact that two As₄ molecules are lost for each chemisorption event.

D. Physisorbed Ga state

To formulate the rate equation describing the physisorbed Ga state we follow the same considerations as above by having an incoming flux from the vacuum (F_{Ga}) and assuming the Ga can leave by overcoming a desorption energy barrier $E_{1,\text{des,Ga}}$. To be chemisorbed the Ga must find an As covered site and this process is therefore proportional to the As surface coverage Θ_{As} . To enter the cluster state the Ga must chemisorb on top of another Ga and this process is therefore proportional to $(1 - \Theta_{\text{As}})$. The Ga can desorb from either of these two chemisorbed states and the desorption fluxes will depend on the populations in the states, $(1 - \Theta_{\text{As}})$ for Ga chemisorbed onto a “normal” lattice site and Θ_{Ga} for the cluster state, respectively. The associated desorption energies are $E_{2,\text{des,Ga}}$ and $E_{3,\text{des,Ga}}$ for the normal lattice site and the cluster state, respectively. The rate equation controlling the Ga population in the physisorbed state is therefore

$$\dot{\vartheta}_{\text{Ga}} = F_{\text{Ga}} - \vartheta_{\text{Ga}} \frac{1}{\tau_{1,\text{des,Ga}}} - D_{\text{Ga}} \vartheta_{\text{Ga}} \Theta_{\text{As}} \frac{1}{\tau_{\text{chem,Ga}}} (1 - \vartheta_{\text{N}_2}) \times (1 - \vartheta_{\text{As}_4}) + (1 - \Theta_{\text{As}}) \frac{1}{\tau_{2,\text{des,Ga}}} - D_{\text{Ga}} \vartheta_{\text{Ga}} (1 - \Theta_{\text{As}}) \times \frac{1}{\tau_{\text{clus,Ga}}} (1 - \vartheta_{\text{N}_2})(1 - \vartheta_{\text{As}_4}) + \Theta_{\text{Ga}} \frac{1}{\tau_{3,\text{des,Ga}}}. \quad (3)$$

E. Physisorbed N₂ state

Since SIMS measurements show that no N₂ is incorporated into the lattice when growing GaAs with a high N₂ background pressure, the rate equation controlling the nitrogen physisorbed population will only contain the incoming flux F_{N_2} and the desorbing flux, which depends on the desorption energy $E_{\text{des,N}_2}$ and is of the form

$$\dot{\vartheta}_{\text{N}_2} = F_{\text{N}_2} (1 - \vartheta_{\text{N}_2}) - \vartheta_{\text{N}_2} \frac{1}{\tau_{\text{des,N}_2}}. \quad (4)$$

The reflection term $(1 - \vartheta_{\text{N}_2})$ regulates the incoming flux so that the physisorbed N₂ population cannot exceed 1 ML.

F. Chemisorbed As state

The rate equation for the chemisorbed As state describes the fraction of surface sites that are covered by As. This fraction is increased by As arriving from the physisorbed As state and by Ga atoms leaving the chemisorbed state. It is decreased by Ga atoms chemisorbing from the Ga physisorbed state and by As leaving the surface. Since Foxon and Joyce⁷ did not detect As₄ leaving the surface above 300 °C the desorbing As flux is assumed to be entirely in the form of As₂. Foxon and Joyce¹² established that if there is a physisorbed state for As₂ its lifetime would have to be $< 10^{-5}$ s and it is therefore assumed that the As₂ desorbs directly from the surface into the vacuum. The rate equation describing the chemisorbed As population can then be formulated as

$$\dot{\Theta}_{\text{As}} = 4D_{\text{As}_4} \vartheta_{\text{As}_4}^2 (1 - \Theta_{\text{As}})^4 \frac{1}{\tau_{\text{chem,As}_4}} (1 - \vartheta_{\text{N}_2})(1 - \vartheta_{\text{Ga}}) - 2\Theta_{\text{As}}^2 \frac{1}{\tau_{\text{des,As}_2}} - D_{\text{Ga}} \vartheta_{\text{Ga}} \Theta_{\text{As}} \frac{1}{\tau_{\text{chem,Ga}}} (1 - \vartheta_{\text{N}_2}) \times (1 - \vartheta_{\text{Ga}}) + (1 - \Theta_{\text{As}}) \frac{1}{\tau_{2,\text{des,Ga}}}. \quad (5)$$

The factor 4 in the first term on the right hand side takes into account that for every As₄ that is chemisorbed four As surface sites are covered and the factor 2 in the second term accounts for the fact that for each desorbing As₂ two chemisorbed As atoms are lost.

G. Ga cluster state

In Sec. IV A a Ga cluster state was introduced since there must be a pathway for Ga atoms to form clusters on top of a Ga terminated surface. The energy barrier for chemisorption into the cluster state is $E_{\text{clus,Ga}}$ and the barrier to desorption from this state is $E_{3,\text{des,Ga}}$. The rate equation describing the Ga population in this state can therefore be formulated as

$$\dot{\Theta}_{\text{Ga}} = -\Theta_{\text{Ga}} \frac{1}{\tau_{3,\text{des,Ga}}} + D_{\text{Ga}} \vartheta_{\text{Ga}} (1 - \Theta_{\text{As}}) \frac{1}{\tau_{\text{clus,Ga}}} (1 - \vartheta_{\text{N}_2}) \times (1 - \vartheta_{\text{As}_4}). \quad (6)$$

Under standard growth conditions, the Ga cluster state will not be significantly occupied ($\Theta_{\text{Ga}} \ll 1$) and therefore its influence on the chemisorbed As state can be neglected.

V. FITTING AND MODELING PROCESS

The five coupled, first order differential equations [Eqs. (2)–(6)] were solved numerically using the Simulink® package in Matlab®. The growth process was modeled for 10 min real time at each temperature step to simulate our experi-

ments. For simulations in the absence of an impinging Ga flux the physisorbed (ϑ_i) and chemisorbed (Θ_i) populations after 10 min of the last temperature step were used as the initial conditions for the next. For cases including a Ga flux the simulation was first performed without Ga for 10 min real time, with the Ga being added afterwards using the initial conditions found by the simulation without the Ga.

To find values for the energy barriers associated with the model it had to be compared with existing GaAs growth data. The data presented by Brennan, Tsao, and Hammons⁴ as well as the growth rate data published by Spring Thorpe and co-workers^{5,6} was used to obtain the energy parameters. All curves comparing the model to the data presented by Brennan and co-workers⁴ are normalized to the incoming flux and the desorbing and incoming fluxes are expressed in units of atoms/s.

A. Desorbing As₄ flux

The desorbing As₄ flux, which is the sum of 50% of the chemisorbed As₄ and the desorption process as described by Eq. (2), was fitted to the temperature dependent data presented by Brennan and co-workers⁴ for an incoming As₄ flux of 0.14 ML/s. Figure 1(a) shows the data points with the fitted curve and as can be seen, the curve fits the data reasonably well.

B. Desorbing As₂ flux

The desorbing As₂ flux is fitted to the data from Ref. 4 and the result can be seen in Fig. 1(b). There is a good agreement for lower temperatures although at temperature above 620 °C the fit deviates, possibly due to out-diffusion of As from the bulk which is not included in this model.

C. Desorbing Ga flux

The desorbing Ga flux can be seen in Fig. 1(c) together with the data from Ref. 4 and the model fit is in excellent agreement with the data.

D. Growth rate

The growth rate was simulated assuming an incoming Ga flux of 1 ML/s and an As₄ flux of 0.7 ML/s (in As₄ molecules). The Ga flux corresponds with the data of Spring Thorpe and co-workers^{5,6} and the fitted curves are in excellent agreement with the data.

E. Minimum necessary As₄ flux

Figure 2 shows the minimum necessary As₄ flux required to maintain a Ga cluster population of less than 10 ML as a function of temperature together with the data points determined by our experiments for the case with no N₂ and with 6×10^{-5} Torr N₂ pressure in the growth chamber. The experimental points were normalized to the model output at 600 °C for the no N₂ case. It has been found in the literature that for As-induced RHEED oscillations, up to 15 ML of Ga can be deposited and the sample still recovered¹³ if the As₄

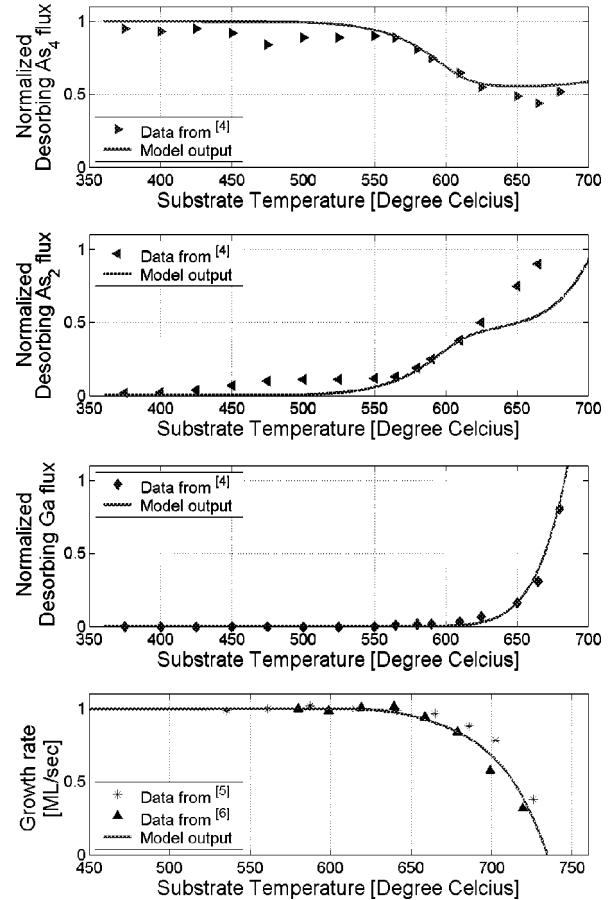


FIG. 1. Fitted model outputs. (a) Desorbing As₄ flux normalized to the incoming As₄ flux vs substrate temperature. (b) Desorbing As₂ flux normalized to the incoming As₄ flux vs substrate temperature. (c) Desorbing Ga flux normalized to the incoming As₄ flux vs substrate temperature. (d) Growth rate vs substrate temperature. The data points for Figs. 1(a)–1(c) are taken from Ref. 4 and for Fig. 1(d) from Refs. 5 and 6.

flux is restarted at an elevated growth temperature. Since our samples were immediately cooled following the 10 min growth the opportunity for Ga balls to be consumed is reduced and so the threshold for the Ga cluster population was taken to be 10 ML.

The N₂ flux was estimated from the N₂ pressure as determined by the flux monitoring ion gauge relative to the Ga beam equivalent pressure and was approximately 100 times the incoming Ga flux. It was also assumed approximately 5

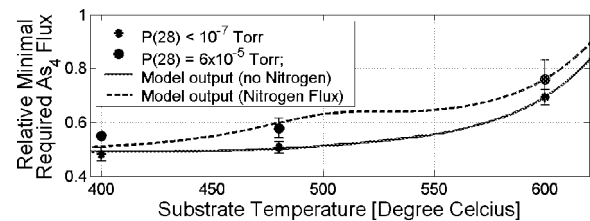


FIG. 2. Experimental data for the minimum required As₄ flux together with the simulation results. The solid line and the *'s represent the no nitrogen case and the dashed line with the ●'s represent the case of 6×10^{-5} Torr of N₂ in the chamber. All the data points are normalized by matching the data point at 600 °C to the model output at 600 °C for the no N₂ case.

N₂ molecules can occupy one lattice site and these considerations lead to an incoming N₂ flux (F_{N_2}) of 20 ML/s. It can be seen that the simulated curve in the absence of N₂ follows the data points very closely while there is some deviation for the 400 °C case when N₂ is present.

VI. DISCUSSION

Following the fitting process described in the previous section the values for the energy barriers associated with all the transitions in the model were found to be: $E_{\text{des,As}_4} = 1.66$ eV; $E_{\text{chem,As}_4} = 0.20$ eV; $E_{\text{des,As}_2} = 2.41$ eV; $E_{1,\text{des,Ga}} = 0.97$ eV; $E_{\text{chem,Ga}} = 0.00$ eV; $E_{2,\text{des,Ga}} = 1.84$ eV; $E_{\text{clus,Ga}} = 0.14$ eV; $E_{3,\text{des,Ga}} = 2.40$ eV; $E_{\text{des,N}_2} = 2.06$ eV. The energy barrier to the chemisorption of As₄ molecules is in good agreement with the value found by Foxon and Joyce⁷ while the desorption energy differs significantly. This may be caused by the fact that the desorption energy was determined by Foxon and Joyce at substrate temperatures below 150 °C where the chemisorbed arsenic population may be well above 1 ML¹⁴ and our model does not allow for an As population above one ML. Furthermore, the rates of As₄ chemisorption and desorption depend strongly on the two energies involved in these processes. Since the population in the physisorbed state is limited and the chemisorption process depends on the square of the population, the energy barrier for desorption must be much higher than the energy barrier to chemisorption, which suggests that the chemisorption process might involve a more complicated process. The energy barrier to the chemisorption of Ga from the physisorbed state was found to be close to 0 eV, which agrees with the observation that Ga is chemisorbed from the vacuum without any energy barrier. The desorption energy found for As₂ is close to the desorption energy found by Pristovsek *et al.*¹⁵ and is similar to the energy found by Ito and Shiraishi¹⁶ using Monte Carlo simulations. It differs from the value of 3.9 eV found, for example, by Karpov and Maiorov.¹⁷ Karpov, however, used a substantial higher attempt frequency than the Debye frequency. The relatively high values for the desorption energies of N₂ and Ga from the physisorbed state might suggest that processes, not included in this model, may take place during the growth of GaAs.

VII. CONCLUSIONS

The minimum As₄ flux required to maintain stoichiometric growth of GaAs in the presence of a high background

N₂ pressure has been measured for substrate temperatures between 400 and 600 °C and has been found to be higher than is required in the absence of a N₂ background. A kinetic growth model has been presented which accounts for these observations and is consistent with GaAs growth data published by other authors.

The model introduces the concept of site blocking which reduces the probability of chemisorption of As₄. Furthermore, the model suggests that a Ga cluster (or ball) state may be occupied at growth temperatures commonly employed for GaAs and AlGaAs and this would have implications for efforts to produce smooth interfaces between these and other III/V compound semiconductors.

ACKNOWLEDGMENTS

The authors wish to thank Ashland Chemicals for their ongoing support. Financial support by La Trobe University, the Australian Government, the “Marianne and Dr. Fritz Walter Fischer Stiftung” as well as the German Academic Exchange Service (DAAD) is gratefully acknowledged.

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