Feedback Control of Surface Roughness of $GaAs$ (001) Thin Films Using Kinetic Monte-Carlo Models*

Yiming Lou and Panagiotis D. Christofides†

Abstract— **In this study, we follow the methodology presented in [7] to study estimation and control of surface roughness of gallium arsenide ($GaAs$) (001) thin films during deposition in a horizontal-flow quartz reactor using triisobutylgallium ($TIBGa$) and tertiarybutylarsine ($TBAs$) as precursors and H_2 as the carrier gas. The adsorption of $TIBGa$ onto the surface and the migration of Ga atoms on the surface are considered as the two rate-limiting steps in the film growth and are explicitly modelled within a kinetic Monte-Carlo simulation framework. The energy barriers and the pre-exponential factor of the migration rates of Ga atoms on the surface used in the simulations are initially determined by fitting the simulation results to experimental data reported in [6]. Then, a roughness estimator is constructed that allows computing estimates of the surface roughness of the $GaAs$ thin films at a time-scale comparable to the real-time evolution of the process using discrete on-line roughness measurements. The roughness estimates are fed to a proportional-integral (PI) feedback controller which is used to control the surface roughness to a desired level by manipulating the substrate temperature. Application of the proposed estimator/controller structure to the process model based on a large-lattice kinetic Monte-Carlo simulator demonstrates successful regulation of the surface roughness to the desired level. The proposed approach is shown to be superior to PI control with direct use of the discrete roughness measurements. The reason is that the available measurement techniques do not provide measurements at a frequency that is comparable to the time-scale of evolution of the dominant film growth dynamics.**

I. INTRODUCTION

Deposition of thin films from gas phase precursors has great industrial importance. The modern integrated circuit technology depends strongly on the uniformity and microstructure of thin films of advanced materials. Due to the increasingly stringent requirements on the quality of such films including uniformity, composition and microstructure, real-time feedback control of thin film deposition becomes increasingly important.

While deposition uniformity control can be accomplished on the basis of continuum type distributed models (see the book [1] for results and references on this problem), precise control of film properties requires models that predict how the film state (microscopic scale) is affected by changes in the controllable process parameters (macroscopic scale). From a microscopic point of view, the rates of surface micro-processes (e.g., adsorption, desorption, migration and reaction) are key factors that determine thin film microstructure and composition. These rates depend strongly on macroscopic process parameters like precursor concentration and substrate temperature to name a few. Kinetic Monte-Carlo (MC) simulation [4] provides a framework for modeling the effect of macroscopic process variables on thin film micro-structure.

The accuracy of solutions from one kinetic MC simulation run depends on the size of the lattice used in the simulation which, in turn, determines the computational requirements of the simulation. Specifically, the larger the lattice, the smaller the fluctuations contained in the simulation results. However, the computational requirements of kinetic MC simulators, based on large lattice models, make their direct use in an on-line feedback control scheme impossible. Motivated by this, recent research efforts have focused on the construction of estimators and controllers, which can be implemented in real-time with reasonable computing power, for thin film surface roughness and growth rate regulation based on kinetic MC simulators using multiple small-lattice models [7], [8]. Other approaches have also been developed to: (a) identify linear models from outputs of kinetic Monte-Carlo simulators and perform controller design by using linear optimal control theory [11], and (b) construct reduced-order approximations of the master equation [3].

Gallium arsenide ($GaAs$) is an important compound semiconductor that has many applications including light-emitting diodes, microwave devices, broadband communications, and space solar cells. Extensive research has been carried out to study the $GaAs$ thin film growth by using kinetic Monte-Carlo simulations. Monte-Carlo simulation for $GaAs$ thin film growth by MBE was first performed in [10]. By using Monte-Carlo models to simulate the formation of $GaAs$ thin films, phenomena such as atomic nucleation, growth, island formation and structural transformation can be studied.

In this study, we follow the methodology presented in [7] to study estimation and control of surface roughness of $GaAs$ (001) thin films during deposition in a horizontal-flow quartz reactor using triisobutylgallium ($TIBGa$) and tertiarybutylarsine ($TBAs$) as precursors and H_2 as the carrier gas. The adsorption of $TIBGa$ onto the surface and the migration of Ga atoms on the surface are considered as the two rate-limiting steps in the film growth and are explicitly modelled within a kinetic Monte-Carlo simulation framework. The energy barriers and the pre-exponential factor of the migration rates of Ga atoms on the

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surface used in the simulations are initially determined by fitting the simulation results to experimental data reported in [6]. Then, a roughness estimator is constructed that allows computing estimates of the surface roughness of the $GaAs$ thin films at a time-scale comparable to the real-time evolution of the process using discrete on-line roughness measurements. The roughness estimates are fed to a proportional-integral (PI) feedback controller which is used to control the surface roughness to a desired level by manipulating the substrate temperature. Application of the proposed estimator/controller structure to the process model based on a large-lattice kinetic Monte-Carlo simulator demonstrates successful regulation of the surface roughness to the desired level. The proposed approach is shown to be superior to PI control with direct use of the discrete roughness measurements. The reason is that the available measurement techniques do not provide measurements at a frequency that is comparable to the time-scale of evolution of the dominant film growth dynamics.

II. SURFACE MICRO-STRUCTURE MODEL FOR $GaAs$ THIN FILM GROWTH

In this study, we consider the MOCVD growth of $GaAs$ in a horizontal-flow quartz reactor [6]. The precursors are triisobutylgallium ($TIBGa$) and tertiarybutylarsine ($TBAs$) and the carrier gas is H_2. The pressures of the precursors and of the carrier gas are: 0.25 mTorr for $TIBGa$, 25 mTorr for $TBAs$ and 20 Torr for H_2. Therefore, the growth occurs in an As-rich environment. Under these precursor pressures, the growth rate is $0.5\mu m/hr$; this growth rate is independent of the substrate temperature when the substrate temperature varies from $825K$ to $900K$. Due to the fact that the pressure of $TBAs$ is much higher than that of $TIBGa$ (the As/Ga ratio is 100), the diffusing species controlling the epitaxial growth is the Ga atoms. The rate-limiting steps are the adsorption of $TIBGa$ and the migration of Ga atoms. Because the decomposition of $TIBGa$ is very fast, the adsorption of $TIBGa$ onto the surface can be simply modelled by the adsorption of Ga atoms onto the surface. As-related kinetics can be incorporated into the model by using "effective" energy barriers to model the surface migration rates of Ga atoms [10].

The formation of $GaAs$ thin films by adsorption and migration of Ga atoms is a stochastic process because: (a) the exact time and location of the occurrence of one specific surface micro-process (adsorption or migration) are unknown, and (b) the probability with which each surface micro-process may occur is only available. Therefore, the surface evolution model should be established based on probability theory. The evolution of the probability that the thin film is at a certain micro-configuration is described by the master equation [12]. Due to the extremely large number of possible states for most systems of realistic size, direct solution of the master equation for any system of meaningful size, using numerical methods for integration of ordinary differential equations (e.g., Runge-Kutta) impossible. Monte-Carlo techniques provide a way to obtain unbiased realizations of a stochastic process, which is consistent to the master equation. Following the methodology developed in [4], we have shown that the algorithm of kinetic Monte-Carlo simulation used in this study is consistent to the master equation in the sense that each Monte-Carlo event in simulations is picked following a probability density function which is derived based on the same assumptions employed in the derivation of the master equation. A detailed discussion of this consistency is omitted due to space limitations; it is included in the journal version of this study [9].

We use the Monte-Carlo model presented in [10] to model the rate-limiting surface micro-processes during the growth of $GaAs$ thin films. Because of the high As/Ga ratio, it is assumed that all the surface sites are available for adsorption of Ga atoms at all times and the adsorption rate of Ga atoms is treated as site independent. Furthermore, when the growth rate is fixed, the adsorption rate on each surface site is a constant, i.e.,

$$w_a = F \tag{1}$$

The migration rate of each surface Ga atom depends on its local environment. Under the consideration of only first nearest neighbor interactions, the migration rate of a surface Ga atom from a surface site with n first nearest neighbors is:

$$w_m(n) = \nu_0 \exp(-\frac{E_s + nE_n}{k_B T}) \tag{2}$$

where E_s is the energy barrier associated with migration due to surface effects, E_n is the energy barrier associated with migration due to nearest neighbor interactions, k_B is the Boltzmann's constant, and ν_0 is the pre-exponential factor.

When the rates of two events are determined based on the corresponding rate expressions, a kinetic MC simulation is executed following the algorithm reported in [5]. Upon an executed event, a real-time increment Δt is computed by:

$$\Delta t = \frac{-\ln \xi}{W_a + W_m} \tag{3}$$

where ξ is a random number in the $(0, 1)$ interval, W_a is the total rate of adsorption and W_m is the total rate of migration.

III. COMPUTATION OF KINETIC MONTE-CARLO MODEL PARAMETERS USING EXPERIMENTAL DATA

The predictions of the Monte-Carlo simulation depend on the rates of adsorption and migration used in the calculations. When the film growth rate is fixed (which is the case in our process), the rate of adsorption can be directly calculated based on the growth rate. However, there are three parameters in the expression of the migration rate, ν_0, E_s and E_n, in Eq.(2) that need to be computed. In this study, the values for these parameters for $GaAs$

thin film growth by MOCVD will be computed based on the experimental data reported in [6]. Specifically, we determine the parameters for migration rate so that the model predictions of the saturated surface roughness value match the experimental results (see [9] for the explicit mathematical expressions for the saturated surface roughness and a detailed discussion on the reason why we use the saturated surface roughness values to fit the migration parameters). To this end, we use the parameters reported in [10] for $GaAs$ thin film growth by MBE as our initial guess for the parameters of Eq.(2) and run MC simulations for the evolution of the surface roughness of $GaAs$ (001) surface. MC simulations are performed on an 80×80 lattice under four different substrate temperatures (T=713K, 750K, 775K and 800K). Because in the experimental work of [6], the $GaAs$ thin film surface roughness is measured by STM after cooling down the film to room temperature at a rate of $2K/s$, the cooling down process is also simulated. Fig.1 shows the evolution of surface roughness when (a) $T = 713K$, (b) $T = 750K$, (c) $T = 775K$ and (d) $T = 800K$, respectively. The experimental results of the saturated surface roughness after the growth of a $0.5\mu m$-thick $GaAs$ film are: 2.8Å when $T = 825K$ and $\sim$ 1.3Å when $T = 850K$, $875K$ and $900K$ [6].

Comparing the simulation results of the saturated surface roughness to the experimental results. We find that the saturated roughness obtained from MC simulation when $T = 713K$ is very close to the experimental results of saturated roughness when $T = 825K$, and the saturated roughness obtained from MC simulation when $T = 775K$ and $T = 800K$ is very close to the experimental results of saturated roughness when $T = 900K$. Therefore, we compute the parameters of migration rates in $GaAs$ thin film growth by MOCVD such that the migration rates of Ga atoms in $GaAs$ by MOCVD at T=825K and T=900K are equal to those in $GaAs$ by MBE at T=713K and T=788K (T = 788K is the average of T = 775K and T = 800K), respectively (details of this approach are included in [9]).

The values of the migration rate parameters, obtained by following the above method and used in all the simulations of the present work, are: $\nu_0 = 5.8 \times 10^{13} s^{-1}$, $E_s = 1.82eV$ and $E_n = 0.27eV$. Fig.2 shows the evolution of surface roughness obtained from kinetic MC simulations using the above computed migration parameters when the substrate temperatures are $T = 825K$, $850K$, $875K$ and $900K$, respectively. The values of the saturated roughness from all the simulations are very close to the experimental results reported in [6]. Thus, we have obtained a kinetic MC model for the surface roughness of the MOCVD of $GaAs$ thin films whose predictions are consistent to the experimental measurements.

IV. REAL-TIME ESTIMATION OF THIN FILM SURFACE ROUGHNESS

To obtain accurate real-time estimates of the surface roughness, we follow the methodology proposed in [7] to combine the kinetic MC simulator, based on multiple small lattice models, with an adaptive filter, to reject the stochastic fluctuations on the surface roughness profile, and a measurement error compensator to improve the estimates of surface roughness using on-line measurements. Specifically, the adaptive filter is a second-order dynamical system with the following state-space representation:

$$\frac{d\hat{y}_r}{d\tau} = y_1; \quad \frac{dy_1}{d\tau} = \frac{K}{\tau_I}(y_r - \hat{y}_r) - \frac{1}{\tau_I}y_1 \qquad (4)$$

where y_r is the output of the kinetic MC simulation based on multiple small lattice models, $\hat{y}_r$ is the filter output, K is the filter gain and τ_I is the filter time constant. To accelerate the response of the filter and avoid large overshoot, $\tau_I = 0.5/K$. To achieve both fast tracking of the dynamics of the outputs and efficient noise rejection, the gain of the filter is

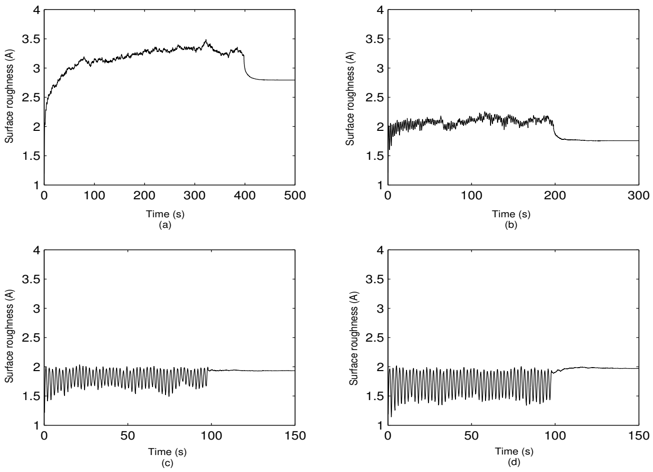

Fig. 1. Surface roughness under different substrate temperatures when the parameters of migration rate are the same to those for $GaAs$ thin film growth by MBE reported in [10]. (a) T=713K, (b) T=750K, (c) T=775K and (d)=800K.

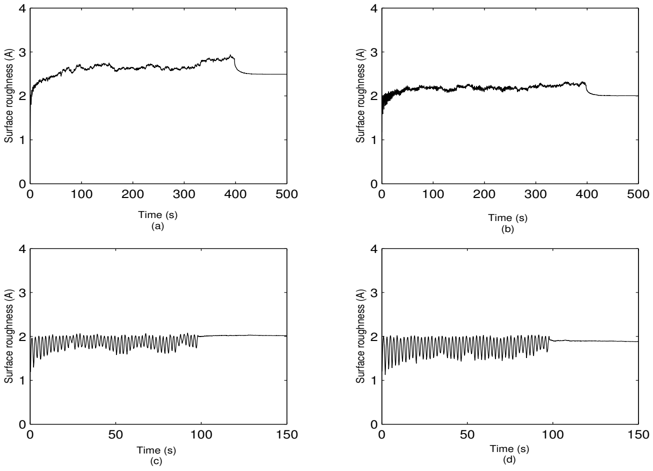

Fig. 2. Surface roughness under different substrate temperatures when the parameters of migration rate are adjusted to match the experimental results reported in [6]. (a) T=825K, (b) T=850K, (c) T=875K and (d)=900K.

adaptively adjusted according to the following law:

$$K(\tau) = K_0 \frac{|\int_{\tau-\Delta\tau}^{\tau} y_r(t)dt - \int_{\tau-2\Delta\tau}^{\tau-\Delta\tau} y_r(t)dt|}{\Delta\tau^2} + K_s \quad (5)$$

where K_0 is a constant, K_s is the steady-state gain for the adaptive filter and $\Delta\tau$ is the time interval between two updates of K. Although a better tracking performance is expected when a small $\Delta\tau$ is used, a very small $\Delta\tau$ will not significantly reduce the effect of fluctuations on the filter output and should be avoided.

The measurement error compensator uses the available on-line measurements (in the numerical simulations the values of the surface roughness are obtained from the large lattice kinetic Monte-Carlo simulator) to produce improved estimates of the surface roughness. The state-space representation of the measurement error compensator is:

$$\begin{aligned} &\frac{de}{d\tau} = K_e(y_h(\tau_{m_i}) - \hat{y}(\tau_{m_i})); \\ &for \quad \tau_{m_i} < \tau \leq \tau_{m_{i+1}}; \quad i = 1, 2, \cdots \\ &\hat{y} = \hat{y}_r + e \end{aligned} \quad (6)$$

where K_e is the compensator gain, e is the estimated model error, which is used to compensate the model output, $\hat{y}$ is the estimate, $\hat{y}_r$ is the filtered output from the kinetic MC simulator using multiple small lattice models and y_h is the output of the large lattice model (in an experimental set-up y_h is obtained from the measurement sensor at discrete time instants $[\tau_{m_1}, \tau_{m_2}, \cdots]$).

The combination of the kinetic MC simulator based on multiple small-lattice models, the adaptive filters and the measurement error compensators functions as an estimator, which is capable to estimate the evolution of surface roughness, while the solution time needed to run the estimator is comparable to real-time process evolution (see [9] for simulation results that demonstrate the effectiveness of the estimator).

V. FEEDBACK CONTROL OF SURFACE ROUGHNESS

The production of high-quality thin films requires that the surface roughness is maintained at a desired level. In this study, we consider feedback control of surface roughness of $GaAs$ thin films by manipulating the substrate temperature, which is assumed to change only with respect to time. This is a reasonable formulation for the manipulated input and is practically feasible for many experimental and industrial deposition processes.

The fact that the model that describes the evolution of the thin film growth process is not available in closed-form (we only have available a kinetic MC model) motivates the use of a proportional-integral (PI) feedback controller to regulate the surface roughness. Furthermore, from simulation results shown in Fig.2, we can see that even when the substrate temperature is fixed, the surface roughness oscillates around a fixed value. This oscillatory behavior is an intrinsic characteristic of the film growth process considered in the present work and it is not our control objective to eliminate this oscillation; rather, we will control the surface roughness at a desired level (range). To eliminate unnecessary control actions, which may lead to poor closed-loop performance, the control objective is to stabilize the surface roughness value close to a desired level (specified by a certain tolerance ϵ). For this purpose, a PI feedback control algorithm is used of the following form:

$$u(\tau) = K_c[\hat{e} + \frac{1}{\tau_c}\int_0^{\tau} \hat{e}(t) \cdot dt] \quad (7)$$

$$\hat{e}(t) \quad = \quad \begin{cases} \hat{y} - y_{set}, & for \quad |\hat{y} - y_{set}| > \epsilon \\ 0, & for \quad |\hat{y} - y_{set}| \leq \epsilon \end{cases} \quad (8)$$

where y_{set} is the desired level of surface roughness, $\hat{y}$ is the output of the roughness estimator, K_c is the proportional gain and τ_c is the integral time constant.

The PI controller is coupled with the roughness estimator presented in the previous section. A diagram of the closed-loop system under the developed estimator/controller structure is shown in Fig.3. Several closed-loop simulation runs were performed to evaluate the effectiveness of the developed estimator/controller structure shown in Fig.3. In our simulations, the outputs from six kinetic MC simulators running on 30×30 lattice models are averaged within the estimator. A 150×150 lattice MC model is used to describe the evolution of the process, which corresponds to a 600Å×600Å $GaAs$ (001) surface. The desired roughness is 1.5Å and the tolerance is 0.1Å. The time interval between two available measurements is taken to be $3.0s$; this specification is made based on the fact that high-speed scanning tunnelling microscopy (STM) reported in [2] is able to measure the morphology of a 600Å×600Å surface with an acquisition time of $3s$ and the fact that it is feasible to perform STM measurement during epitaxial growth of $GaAs$ layers [13]. The parameters for the roughness estimator and the PI controller used in the simulations are shown in Table I. Initially, the $GaAs$ thin film grows on a perfect surface at $T = 800K$ and the roughness increases.

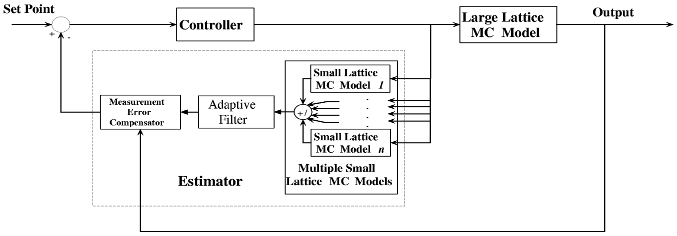

Fig. 3. Diagram of the closed-loop system under the developed estimator/controller structure.

TABLE I

ROUGHNESS ESTIMATOR AND CONTROLLER PARAMETERS.

K_0	0.05	s/Å	K_s	0.1		K_e	0.08	s^{-1}
K_c	15	K/Å	τ_c	0.2	s	ϵ	0.1	Å

Then, the controller is activated when the roughness reaches 2.3Å. Fig.4 shows the evolution of the surface roughness and the substrate temperature under feedback control. The results clearly show that the developed estimator/controller structure can successfully drive the surface roughness to the desired level.

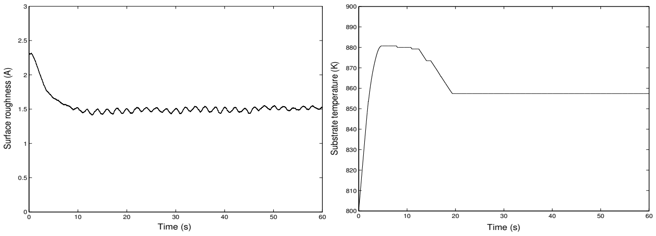

Fig. 4. Evolution of the surface roughness (left plot) and the substrate temperature (right plot) under feedback control based on the roughness estimator.

To test the robustness of the proposed estimator/controller structure, we considered controlling the $GaAs$ thin film growth process in the presence of 10% uncertainty in the energy associated with a single bond on the surface (i.e., the E_s used in the roughness estimator is $1.82eV$ but the E_s used in the kinetic MC model based on the large lattice is $2.0eV$). Fig.5 shows the corresponding output and input profiles, respectively. The controller exhibits very good robustness properties.

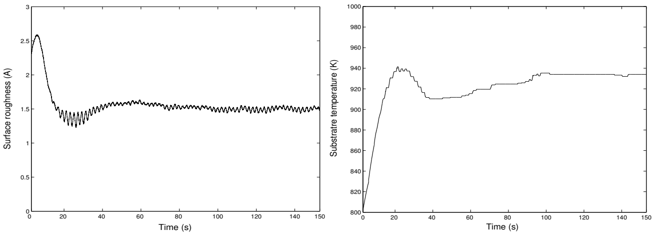

Fig. 5. Evolution of the surface roughness (left plot) and the substrate temperature (right plot) under feedback control based on the roughness estimator - closed-loop system simulation under uncertainty.

To show the importance of using the roughness estimator for feedback control and not relying exclusively on the roughness measurements (which are obtained every 3.0 seconds), we applied the PI controller (with the same parameters as in Table I) to the kinetic MC model assuming that new roughness measurements are fed into the controller every 3.0 seconds (this is consistent with our previous simulations). Also, to prevent the substrate temperature from obtaining unreasonably high or low values, the substrate temperature is constrained to be $750K \leq T \leq 950K$. Note, that when the roughness is controlled using the proposed controller/estimator structure, the substrate temperature is always within $750K \leq T \leq 950K$. Fig.6 shows the evolution of the surface roughness and the substrate temperature, respectively. We can see that the PI controller, based on discrete roughness measurements, is not able to control the surface roughness to the desired level; this simulation shows the usefulness of the proposed estimator/controller structure.

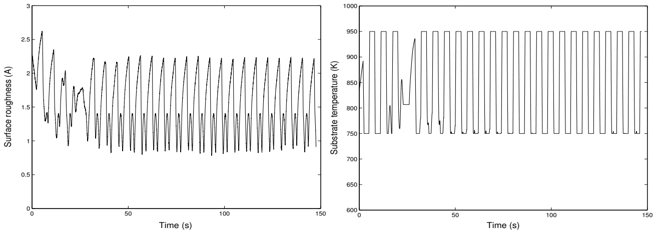

Fig. 6. Evolution of the surface roughness (left plot) and the substrate temperature (right plot) under feedback control without roughness estimator.

Note, that by tuning the PI controller, the surface roughness could be controlled to the desired level by using only the on-line roughness measurements. However, further controller tuning can not achieve a closed-loop performance as good as that achieved under feedback control using the roughness estimator. To show this, we applied the PI controller with a new set of parameters ($K_c = 5K/\text{Å}$, $\tau_c = 1.0s$) to the same kinetic MC model of the $GaAs$ thin film growth process assuming that new roughness measurements are fed into the controller every 3.0 seconds. The parameters of the PI controller are tuned to make the controller able to drive the surface roughness to the desired level. Fig.7 shows the profiles of surface roughness and substrate temperature. With the new controller parameters, the surface roughness is eventually controlled to the desired level, but significant oscillations can be observed in the closed-loop roughness profile and the process takes significantly longer time to reach the desired level (compare Fig.4 and Fig.7; in Fig.4, the surface roughness in the closed-loop simulation reaches the desired level at about $t = 10s$, but in Fig.7, the surface roughness in the closed-loop simulation reaches the desired level at about $t = 30s$). Note, that we have also tried many other sets of tuning parameters for the PI controller but it turns out that it is hard to simultaneously achieve short transient time and reduced oscillation when control of the surface roughness is performed using only the discrete roughness measurements.

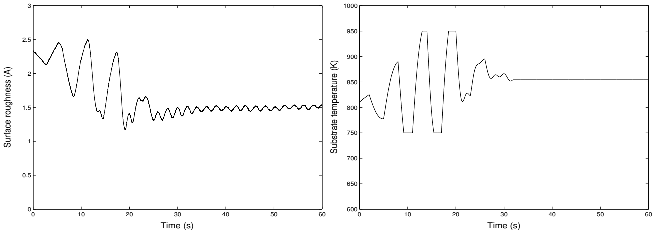

Fig. 7. Evolution of the surface roughness (left plot) and the substrate temperature (right plot) under feedback control without roughness estimator.

We also studied the effect of time delays in the measurements of surface roughness on the closed-loop system performance under the developed estimator/controller structure. To this end, we applied the estimator/controller structure (the parameters of the roughness estimator and the PI controller are the same to those shown in Table I) to the process model based on a 150×150-lattice kinetic Monte-Carlo simulator assuming that the new roughness measurements are fed into the estimator every 3.0 seconds with a time-delay of $t_d = 3.0s$. The resulting profiles of surface roughness and substrate temperature are shown in Fig. 8. The developed estimator/controller structure successfully drives the surface roughness to the desired level in the presence of a time-delay in the roughness measurements.

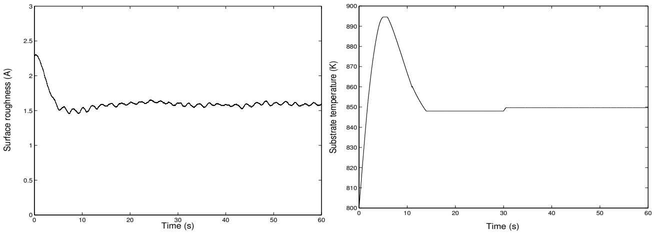

Fig. 8. Evolution of the surface roughness (left plot) and the substrate temperature (right plot) under feedback control based on the roughness estimator - delayed roughness measurements.

To show that the estimator/controller structure is able to control the surface roughness independently of the frequency at which the roughness measurements are available, we implemented the developed estimator/controller structure without using roughness measurements, i.e. the controller determines the substrate temperature based only on the output of the kinetic MC simulator which uses six small-lattice models and the adaptive filter. Fig.9 shows the resulting profiles of surface roughness and substrate temperature. Our simulation results show that this open-loop implementation of the controller/estimator structure (with same parameters as those shown in Table I) successfully drives the surface roughness to the desired level.

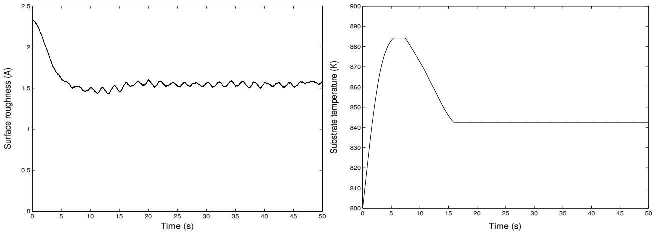

Fig. 9. Evolution of the surface roughness (left plot) and the substrate temperature (right plot) under open-loop implementation of the estimator/controller structure.

VI. CONCLUSIONS

In this study, we followed the methodology presented in [7] to study estimation and control of surface roughness of $GaAs$ (001) thin films during deposition in a horizontal-flow quartz reactor using $TIBGa$ and $TBAs$ as precursors and H_2 as the carrier gas. The adsorption of $TIBGa$ onto the surface and the migration of Ga atoms on the surface were considered as the two rate-limiting steps in the film growth and were explicitly modelled within a kinetic Monte-Carlo simulation framework. The energy barriers and the pre-exponential factor of the migration rates of Ga atoms on the surface used in the simulations were initially determined by fitting the simulation results to experimental data reported in [6]. Then, a roughness estimator was constructed that allows computing estimates of the surface roughness of the $GaAs$ thin films at a time-scale comparable to the real-time evolution of the process using discrete on-line roughness measurements. The roughness estimates are fed to a PI feedback controller which is used to control the surface roughness to a desired level by manipulating the substrate temperature. Application of the proposed estimator/controller structure to the process model based on a large-lattice kinetic Monte-Carlo simulator demonstrates successful regulation of the surface roughness to the desired level. The proposed approach was shown to be superior to PI control with direct use of the discrete roughness measurements. The reason is that the available measurement techniques do not provide measurements at a frequency that is comparable to the time-scale of evolution of the dominant film growth dynamics.

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