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Toward controlled ultra-high vacuum chemical vapor deposition processes

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Chapter 1

Introduction

"Therefore I would not have it unknown to Your Holiness, that the only thing which induced me to look for another way of reckoning the movements of the heavenly bodies was that I knew that mathematicians by no means agree in their investigation thereof. For, in the first place, they are so much in doubt concerning the motion of the sun and the moon, that they can not even demonstrate and prove by observation the constant length of a complete year; and in the second place, in determining the motions both of these and of the five other planets, they fail to employ, consistently one set of first principles and hypotheses, but use methods of proof based only upon the apparent revolutions and motions."

– Nicolaus Copernicus, 1543.

The publication of "De revolutionibus orbium coelestium" (On the Revolutions of the Heavenly Spheres) in 1543 by Nicolaus Copernicus is considered to have marked the beginning of the scientific revolution. The third updated and annotated edition of this manuscript was interestingly published in 1616 in Amsterdam by Nicolas Mulerius, professor of medicine and mathematics at the University of Groningen. The cover page and one of the models is shown in Fig. 1.1. The preface of Copernicus' work considers the justification of his research and methods, addressed to Pope Paul III and, indirectly, to the physics and mathematics communities. His work was highly unconventional, as his theory had to be understood through mathematics instead of physics. Now, 475 years later, we can safely say that the field of physics can no longer be understood without at least a basic understanding of mathematics. This thesis considers the application of mathematical system and control engineering principles to a physical ultra-high vacuum chemical vapor deposition (UHVCVD) process, used for thin film deposition. As Copernicus did in his methods, we aim to develop mathematical models and laws (or hypotheses) in order to test them with observations from the physical system. However, before doing so, we will introduce the physical process that we are working with and motivate our contributions. Let us accordingly begin this thesis by providing background information on thin films in Section 1.1. Following this, we will discuss vacuum evaporation principles and some relevant techniques in Section 1.2. Subsequently, we will motivate the research directions in Section 1.4. This is followed by an outline of the remaining thesis chapters in Section 1.5. Lastly, we relate the chapters and their content to the original publications in Section 1.6.
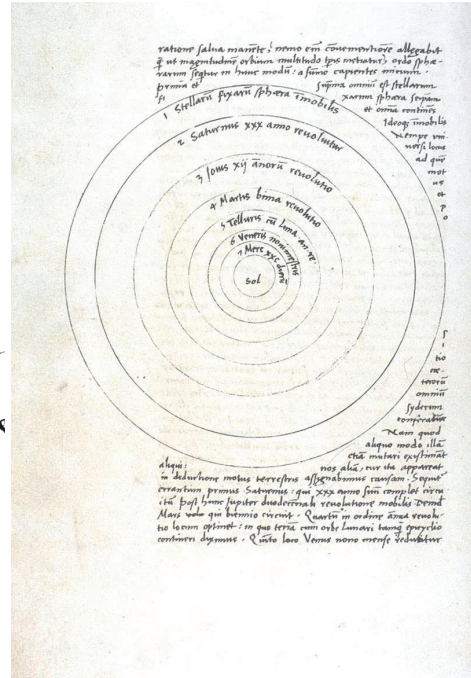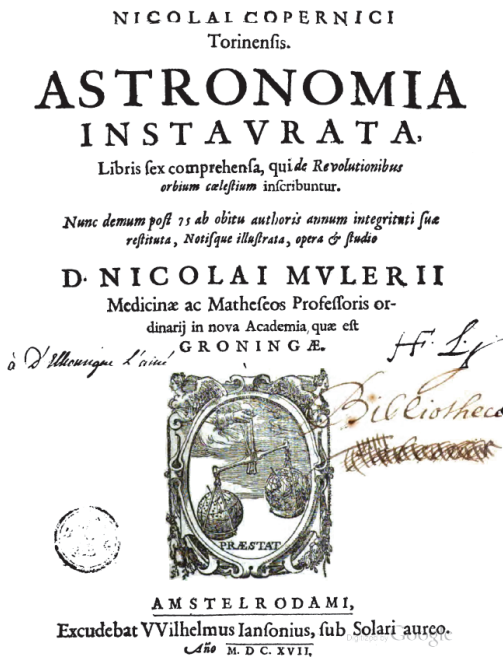
Figure 1.1: The third edition cover and the heliocentric model from Copernicus

1.1   Thin films

The name thin film suggests that thin films are specified by their thickness. The small film thickness is however a characteristic of the thin film but no specific bounds can be given. The reason for this is that we consider a body to be a thin film when certain physical anomalies appear. Accordingly, the physics branch of thin films deals with systems which have the common property that one dimension is very small, while other characteristics and the methods of investigating them may differ completely between the films (Eckertová 1977). Outside this branch, we are typically concerned with characterization of three dimensional bodies. For such bodies, we assume that their characteristics are volume independent. This assumption is however only valid for a small surface-to-volume ratio. For bodies that are very small in one dimension, like thin films, this surface-to-body ratio increases drastically and this assumption therefore becomes invalid. Let us shortly motivate why this is the case. In a bulk material, there are always forces acting upon given molecules or atoms. These forces are from all directions. They can be very structured or periodical, as is the case for crystalline materials, or they can lack such properties, as is typically the case for amorphous materials. However, when considering a surface area, some of these forces are cut off. The molecules or atoms at a surface are accordingly in a differ-

Figure 1.2: Thin film examples

Two examples of thin films are shown. In the left figure, a bell of soap shows different colors. The bell consists of a thin water film encapsulated by two soap films. Light is reflected from both layers of soap in the same direction. The thickness (or thinness) of the layer of water determines which wavelength of light results from the interference. The figure on the right shows camera lenses. These lenses are covered with anti-reflective coatings, which are thin films. A complex lens can have over 30 air-to-glass interfaces and the anti-reflective coating is essential in reducing the accompanied light loss caused by these transitions. Without these coatings, combining such a large number of lenses would result in unacceptable accumulated loss of light. (left: ©Brocken Inaglory, CC BY-SA 3.0 / right: ©Bill Ebbesen, CC BY 3.0)

ent state than the molecules or atoms in the bulk material. We consider this to be a surface state and the associated energy can differ substantially from the bulk state energy. For a body that is very small in one dimension, the two surfaces are so close to each other that they can have a profound influence on the internal characteristics of the body. This thin body can therefore differ substantially from a thicker version of itself. In fact, this difference can be so profound, that completely new phenomena can arise. This accordingly justifies why an entire branch of physics has been dedicated to thin films and the related technological branches.

Optical phenomena are some of the most apparent phenomena associated with thin films. An example is that of interference colors, which can be seen when water is covered by a thin layer of oil or with the bell of soap shown in Fig. 1.2. These phenomena have attracted attention since the second half of the seventeenth century and have resulted in modern applications of anti-reflective and decorative coatings (see Fig. 1.2 for the application to camera lenses). Electronic properties have been studied from the beginning of the twentieth century. Phenomena that received particular interest are super- and semiconductivity. The development of electronics during and since the second world war has resulted in continuously decreasing electronic dimensions. This has been further promoted by the light-weight requirements of the space and avionic industries, as well as the development of medical electronics, which can be placed on or in the body of a patient. One of the main contributors

to this advance in recent years is the consumer electronics industry, e.g. computers, smartphones, tablets, watches, cars and many other applications. This miniaturization of electronic elements like tubes, resistors and capacitors has invoked the use of semiconductor elements, like diodes and transistors. This was followed by the introduction of small ceramic plates on which the elements are prefabricated, mostly in the form of thin films.

Associated thin film production technologies can be considered as a further subdivision of the physics of thin films branch. In most of the processes, the thin film is deposited on a substrate. These depositions can occur through a chemical reaction (chemisorption), through physical forces of attraction (physisorption) or a combination of both. Accordingly, the deposition processes are typically categorized as chemical vapor deposition (CVD) or physical vapor deposition (PVD) processes. Further categorization of the processes is difficult because the processes, but also the application of each process, may vary in many ways. What further complicates the matter is that the number of processes is vast. In (Vossen and Kern 1978) 33 different processes are listed and a subset is often considered in the literature.

In this thesis, we will focus on a specific realization of thin film deposition through vacuum evaporation. We will use the next section to go further into detail on this deposition technology.

1.2   Thin film deposition through vacuum evaporation processes

Deposition through vacuum evaporation (Berry et al. 1968, Eckertová 1977, Vossen and Kern 1978, Ohring 2001), sometimes referred to as thermal evaporation, has been one of the most popular thin film deposition methods due to its relative simplicity and its potential to produce films of extreme high purity. The actual deposition process consists of several steps:

1. Transformation of the materials to be deposited (precursors) to the vapor phase;

2. Mass transfer of precursors from the evaporation source to the substrate;

3. Deposition of atoms or molecules on the substrate.

Vacuum evaporation deposition is generally considered to be a PVD process, since the principle mechanism binding the vapors to the substrate (or any surface) is physisorption. However, physical, chemical and hybrid binding mechanisms are all possible. There are situations where a direct gas insertion is desired instead of an evaporation or sublimation process, but we will not cover these situations in this thesis. Instead, we will assume in the sequel that evaporation sources releases precursor atoms, as this is typically the case. Evaporating atoms directly furthermore

allows for the formation of films of the highest purity, which is one of the main benefits of the vacuum evaporation process that plays a central role in this thesis.

The depositions in a vacuum evaporation deposition process typically occur inside a dedicated chamber, called the deposition chamber. Such a chamber allows for easy cleaning after a process run. The deposition chamber is located in a vacuum system, which is an integrated part of the vacuum evaporation reactor. Before commencing depositions, the vacuum system holding the deposition chamber needs to decrease the pressure many orders of magnitude from atmospheric condition so that: (i) undesired atoms or molecules are (mostly) removed from the deposition chamber, (ii) deposition chamber pressure is below the associated vapor pressure of precursors and (iii) the mean free path of precursors becomes far larger than the deposition chamber dimensions. Evaporation (or sublimation) of the precursors is achieved by either resistance heating, radio-frequency induction or electron bombardment. Mass transfer subsequently occurs through the free molecular flow (FMF, also known as Knudsen flow) transport regime. The evaporation is typically performed in line-of-sight of the substrate, since desorption of the precursors from surfaces other than the evaporation source typically occurs in minimalistic degree, caused by relatively low surface temperatures and low vapor pressures of the precursor elements.

Both the physical and chemical binding mechanisms are highly sensitive to temperature. As a consequence, choosing appropriate operating temperatures of reactor components is very important. There are three temperature domains of interest: (i) evaporation source temperatures, (ii) reactor surface temperatures and (iii) substrate temperature. The evaporation source temperatures (i) are determined by the characteristics of the evaporation source and the required precursors. When considering the reactor surface temperatures (ii), we can distinguish between hot-wall and cold-wall processes. In cold-wall processes, the reactor is not heated (only indirectly through the evaporation) or some parts may even be cooled, this will therefore cause vapor pressures to be very low. In hot-wall processes, the reactor is heated and this promotes further precursor migration. The latter can be desirable when the precursors are not evaporated in line-of-sight of the substrate. Placing the evaporation sources out of the line-of-sight of the substrate can promote purity and uniformity by decreasing the chance that some contaminants reach the substrate and by causing the angle of incidence of arriving precursors to be distributed more uniformly. The choice between a hot-wall or a cold-wall process is hence dependent on the required precursors and on purity and uniformity demands. The substrate temperature (iii) influences characteristics of the layer that grows on the substrate. Requirements on the substrate temperature are therefore often dictated by the application of the thin film.

There are three particular realizations of vacuum evaporation deposition process that we highlight. The first is Ultra-High Vacuum Chemical Vapor Deposition (UHVCVD) (Meyerson 1986, Greve and Racanelli 1991, Meyerson 1992, Adam et al. 2010) and is shown in Fig. 1.3a. This realization is the main subject of this
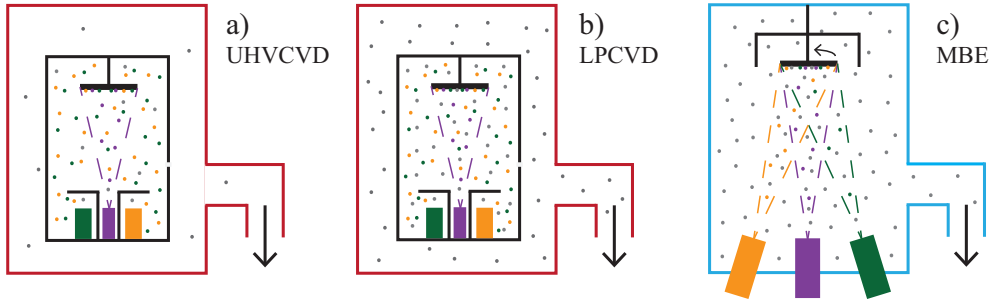
Figure 1.3: Comparison of vacuum evaporation processes

A comparison between ultra-high vacuum chemical vapor deposition (UHVCVD), low pressure chemical vapor deposition (LPCVD) and molecular beam epitaxy (MBE) is shown. UHVCVD and LPCVD are hot wall processes, which is shown in red here, while MBE is a cold wall process, which is shown in blue. UHVCVD and LPCVD furthermore have an additional chamber inside the reactor, which is the deposition chamber. This chamber is replaced by a shield in the MBE process. The deposition chamber serves to reduce leakage to the pump so that some precursor partial pressures can build-up. There are three precursors, which are yellow, purple and green. The blocks in these colors are the associated evaporation sources. The purple precursor is evaporated in line-of-sight of the substrate in all three processes. In UHVCVD and LPCVD, the yellow and and green precursors are not evaporated in line-of-sight of the precursors. The green, yellow and purple dots are precursor atoms and the grey dots are residual molecules or atoms. The UHVCVD process has significantly less residual molecules or atoms in the process due to the UHV conditions. The downward pointing arrows indicate the system exhaust, this is where the vacuum pumps are connected.

thesis. The second is Low Pressure Chemical Vapor Deposition (LPCVD) (Fossum et al. 1985, Rausch and Burte 1993) and is shown in Fig. 1.3b. This realization is closely related to UHVCVD but operates in less strict pressure regions. The third is Molecular Beam Epitaxy (MBE) (Cho and Arthur 1975, Schlom et al. 2008) and is shown in Fig. 1.3c. This realization is the Physical vapor deposition (PVD) cousin to LPCVD and UHVCVD and can be considered as the state-of-the-art in vacuum evaporation deposition. We will use the remainder of this section to provide some background information on these processes and to compare them to each other.

1.2.1   Ultra-high vacuum chemical vapor deposition

The first application of UHVCVD as described by (Meyerson 1986) considers the chemisorption of a Germanium vapor to a Silicon substrate. This application utilizes purely chemical binding, but there is often a combination of physical and chemical binding mechanisms in modern applications. Such a process can include line-of-sight evaporators such as effusion cells (also known as Knudsen cells), but also evaporators that are not placed in line-of-sight of the substrate. Precursors evaporating from the latter migrate to the substrate via multiple deposition chamber surface

collisions. To promote repeated desorption of the precursors from these surfaces, a hot-wall process is desired. This heating of the deposition chamber surfaces then causes physisorptions to be reduced. Accordingly, binding of these precursors to the substrate requires either: (i) a chemical binding to the substrate, (ii) a chemical change in the precursor (by binding with another precursor) or (iii) a substrate that has a reduced temperature in comparison to the deposition chamber walls. Notice that (i) relies on chemisorption, (iii) on physisorption and (ii) on both. Combining this insight with the observation that application of (iii) is not directly visible in the literature then justifies the general association to CVD processes. The indirect migration of precursors to the substrate causes a pressure buildup in the deposition chamber. This is exacerbated with increasing surface temperatures and with decreasing deposition chamber leakage to the vacuum pumps. UHVCVD is a particularly suitable processes when precursors with high vapor pressures are involved, because these precursors can be promoted to migrate with a limited heating of the deposition chamber surfaces. The indirect migration of the precursor to the substrate can have three favorable effects. The first is that any undesired elements escaping from the evaporation source can easily become stuck to the deposition chamber surfaces when their vapor pressure is significantly lower than the vapor pressure of the precursor. The second is that the indirect migration of the precursors to the substrate surface allows for increased film uniformity, due to the more uniform angle of incidence in comparison with a line-of-sight evaporation. The third is that there is potentially a high degree of controllability of this process because, in particular when (ii) is utilized, chemical reactions can be activated. This in turn allows for time to settle on desired precursor pressures and temperatures before activation. UHV-CVD is accordingly an interesting candidate technology for applications that require extreme high purity of the deposited film. However, the technology is lacking in terms of controllability and repeatability. There are three main causes: Run-to-run variations in the substrate (on the nanoscale), run-to-run variations in evaporation sources and *in-situ* sensor placement restrictions. It is difficult to detect and correct for the variations during processing due to *in-situ* sensor placement restrictions. The *in-situ* sensor placement restrictions are a direct consequence of a necessary reactor bake-out, which is instrumental to reaching UHV conditions.

1.2.2   Low pressure chemical vapor deposition

The main difference between UHVCVD and LPCVD lies in the vacuum level. The LPCVD vacuum conditions can be achieved without the aforementioned bake-out of the reactor. This in turn causes LPCVD to have less restrictions on sensor placement inside the vacuum system. For example, the well-known quartz crystal microbalance and some mass spectrometers can be used for certain applications in LPCVD, while this is not possible in UHVCVD. There is accordingly more flexibility to deal with run-to-run variations in LPCVD than there is in UHVCVD. The price to pay is

that the lower vacuum contains more residual atoms or molecules, which in turn can cause impurities in the deposited thin film.

1.2.3   Molecular beam epitaxy

MBE is strictly a line-of-sight evaporation process and belongs to the PVD processes. The precursors are typically evaporated in effusion cells. These cells facilitate temperature control and a shutter, which allows for a high degree of outgoing precursor flux control. When the cells are heated sufficiently and the shutter is open, the precursors are transported to the substrate, where they are bound through physisorption and where they can chemically react with other precursors. The substrate is typically rotating around its central axis to promote deposition uniformity. MBE processes are cold wall processes, as it is desired to have the Knudsen cells dictate all precursor fluxes to the substrate. The deposition chamber is furthermore replaced by a shield, intended to catch any precursor atoms that do not hit the substrate. This shield can be cooled to further promote physisorption of the precursors. MBE operates under similar conditions as the LPCVD process, and therefore offers more flexibility in sensor placement than a UHVCVD process. There are little restrictions on substrate and evaporator temperature. MBE processes are popular due to their relatively high degree of controllability in comparison with other thin film deposition techniques. The process does however face two major disadvantages: (i) fluxes and temperatures need to be controlled very precisely since the depositions are almost instantaneous as a consequence of the direct physisorption that occurs after evaporation and (ii) impurities in the deposited film can occur due to contaminants in the evaporation source and residual gasses in the system, as a result of the process not being performed in UHV and evaporation being line-of-sight.

1.3   UHVCVD operation recipe

Let us shortly describe a typical UHVCVD operation recipe before we continue to the motivation for the contributions in this thesis. UHVCVD operation recipes are application dependent, but a controlled batch deposition process roughly consist of the following processing steps:

1. Pre-process substrate, reactant sources and deposition chamber.

2. Build-in components and create UHV conditions.

3. Regulate to desired background pressures and temperatures.

4. Enable chemical reactions at substrate.

5. Iterate the steps 3 & 4.

6. Cool down reactor.

The pre-processing of components in step 1 is typically a cleaning and mounting process. Step 2 involves preparing the reactor for deposition. The reactor needs to be heated so that residual molecules evaporate and can be pumped out of the system, in order to achieve the UHVCVD conditions. This step can furthermore involve pre-heating of the precursor evaporation sources to remove contaminants. In step 3 the reactor is prepared for the first deposition cycle. The reactor is regulated so that desired background pressures and temperatures are attained. At this stage undesired reactions are generally avoided by carefully choosing which precursors are available and/or by temperature control. The reactions are subsequently activated in step 4. This activation can be performed by either adding a previously missing precursor (preferably as a molecular beam) and/or through the control of surface temperatures (in particular the substrate temperature). We remark that activation of the reactions is not essential, but it does employ a high degree of control over the depositions that occur. Step 5 involves iterating steps 3 & 4, in order to build up layer thickness. Iterations should be small enough to allow for the desired chemical equilibrium to be maintained. Continuous operation is achieved when the iteration time is infinitesimally small. Lastly, when the desired thickness is achieved, the reactor is gradually cooled down in step 6.

1.4 Motivation

With this thesis, we aim to contribute to the development of control for UHVCVD reactors. To this end, we contribute to: (i) development of the physical UHVCVD process in combination with a mathematical process model and controller design and (ii) development of mathematical control algorithms. This results in the thesis being split in two parts:

- Part I: Modeling of free molecular flow dynamics, partial pressure measurement and controller implementation.

- Part II: Controller design for deterministic systems with stochastic initial conditions.

We use the remainder of this section to motivate our interest in the two directions listed above.

1.4.1 Modeling of free molecular flow dynamics, partial pressure measurement and controller implementation

In the first part of the thesis we provide the first steps in facilitating controller implementation for UHVCVD processes. We have motivated in Section 1.2.1 why UHV-

CVD can be the deposition technology of choice for some applications, in particular when extreme high purity of the deposited thin films is required. However, we also have discussed that the controllability and reproducibility are still open issues, caused by run-to-run variations in the evaporation sources and substrates. An effective method of dealing with such issues by implementing run-to-run and real-time feedback control (Edgar et al. 2000).

A conceptual control diagram for UHVCVD processes is shown in Fig. 1.4. The run-to-run controller aims to optimize the runs with knowledge on previous runs. Such a controller can consist of (a collection of) algorithms and a reference manager. The reference manager sets desired (future) values for the real-time controllers. The run-to-run controller can use both (previous) real-time and *ex-situ* data for its purpose. The real-time controllers aim to steer relevant temperatures and partial pressures to the desired values as dictated by the reference manager. Such controllers therefore rely on real-time (and past) measurements of these states and the availability of such measurements is thus essential.

The relevant states to measure for UHVCVD reactor controller design are: (i) relevant temperatures, (ii) film characteristics (like thickness) and (iii) precursor partial pressures (or fluxes). The states (i) and (iii) together affect the chemical reactions that are the depositions, which in turn affects (ii), this view is in accordance with modeling for UHVCVD as in (Greve and Racanelli 1991). Measuring temperatures (i) can be done through contact (thermocouple) and optically (pyrometer). Both of these methods are well established and compatible with UHV. Temperature control is typically performed by placing PID-controlled heating or cooling elements at locations of interest, such as the substrate. Such a form of control is rudimentary but works well, in particular in combination with a reference manager. Measuring layer characteristics (ii) can be done optically through Ellipsometry, reflection or transmission measurements (as for example implemented in (Middlebrooks and Rawlings 2007)). The techniques are also well established but require application dependent models to relate measured variables to film characteristics. Also, any control action based on a real-time layer characteristic measurement will demand a change in pressure or temperature (through the reference manager, for example), since these are the variables that affect the chemical reactions that are the depositions. We therefore consider such a controller to be of a higher hierarchical level than the partial pressure or temperature controllers in such a process. This causes its effectiveness to be dependent on the performance of the lower level pressure and temperature controllers. Developing such a controller is accordingly interesting once good temperature and partial pressure controllers are available. In contrast to the above, a method for measuring the precursor partial pressures (iii) is not clearly visible in the literature. The exposition above motivates our interest in the development of a real-time precursor partial pressure controller for a UHVCVD reactor. Such a controller can accordingly reduce the effects of the run-to-run variations in the evaporation sources.

Controller design is typically done in four steps. The first considers system de-
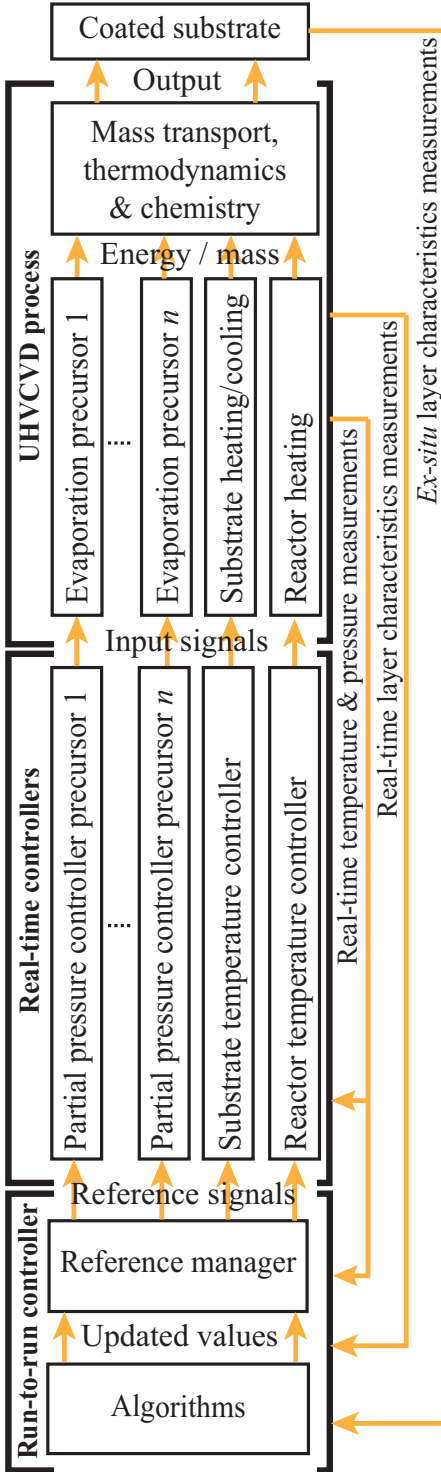
Figure 1.4: Conceptual control diagram for UHVCVD processes

A conceptual control diagram for UHVCVD processes is shown. The diagram contains two controller layers and UHVCVD process dynamics. The process dynamics are subject to heating, cooling and evaporations through the system inputs, supplied by the real-time controllers. These changes in energy and mass cause depositions to occur through mass transport, thermodynamics and chemistry and therefore result in a coated substrate. The evaporation, heating and cooling processes can be controlled individually by the real-time controllers, which in turn aim to follow a reference trajectory supplied by the reference manager. This reference manager is part of the run-to-run controller and is updated/directed through algorithms. The process can accordingly improve performance between runs through this mechanism. The real-time controllers require past and real-time measurements of the variables that they control, so that these variables can be steered to the supplied reference value. The run-to-run controller can use both the real-time data and ex-situ data for learning purposes.

sign so that we obtain a system with actuation and measurements that are required for both the modeling of the system and the controller design. The second step considers the modeling of the systems dynamics, so that we obtain a sufficiently accurate model that relates the reactor input to the (indirectly) measured reactor outputs. The third step considers the model-based controller design. Such a controller design is typically model based so that tuning can be performed analytically and offline, having a model furthermore allows for usage of controllers that rely on predictions from the model online. The last step is the integration of the previously mentioned components, to obtain the controlled system.

In part I of this thesis, we will present contributions to UHVCVD reactor design, modeling of free molecular flow (FMF) dynamics and controller design for partial pressure control in UHV conditions. Our contributions to the UHVCVD reactor design concerns implementation and calibration of an atomic absorption spectroscopy (AAS) based measurement that can measure precursor partial pressures with excellent accuracy and in very low pressure regions. We contribute to the modeling of FMF dynamics by proposing a flux dynamical model. This model allows us to consider the spatial dependence of fluxes and sorption explicitly and can therefore provide further insights in these phenomena. We validate both our AAS-based measurement and the fluxes model by comparing their performances with a lumped (benchmark) model and the theoretical partial pressure in the vapor pressure regime. Our contribution to the controller design for partial pressure control utilizes the AAS-based partial pressure measurement but not the fluxes model. We instead implement two simple controllers that require minimal identification using input-output data. The first controller is a proportional integral controller with a feedforward component. The second controller is a model-free proportional integral controller (based on results from (Fliess and Join 2013)). We show that we can achieve good performance with both controllers and compare their performances.

1.4.2   Controller design for deterministic systems with stochastic initial conditions

In Part II of this thesis we provide controller design (methods) that can deal with uncertainties in initial conditions of the states of a deterministic dynamical system. Let us first motivate why such a controller design can be useful for UHVCVD processes. From there, we will move on a more general introduction of the subject, as we will contribute to this control problem in a general setting.

Controlling variations in UHVCVD processes

UHVCVD is a process type that can potentially benefit from controlling variations that are inherited from the initial conditions. Our contributions in Part I facilitate improved understanding of process dynamics through modeling and real-time par-

tial pressure measurement implementation. The application of such an optical sensor can furthermore provide highly accurate feedback usable for control over essential reactor conditions like the considered partial pressures. The principle goal is to achieve sufficient understanding of dynamics and measurements so that we can operate accurately in millipascal pressure regions to deposit thin films with nanometer precision. At this point, we expect to find remaining and very apparent influence from (small) run-to-run process variations, which are easily significant when working on the considered scales. Specific sources of such variations are potentially: (i) the substrates and evaporation sources that are replaced between runs and (ii) temperatures that are in transient upon initialization due to the absence of significant convections in the vacuum. Significant effort is typically put in reducing the variations in (i), but they remain significant in (budget constrained) best practise. A logical step is accordingly to focus effort on reducing the effect of such variations during processing, using the high precision measurements available. For (i) we can consider most of the variations through the states, in particular when they have dynamics influenced by the process. Indeed, the evaporation sources might vary in amount of precursor material, resistance or specific heat, which will all change with evaporations. The substrate surface might vary in roughness, transparency and chemical composition, which will all change with depositions. For (ii) we have that initial temperature states can be dependent on the time of initialization and on spatial temperature uncertainties. The latter refers to the temperatures of all bodies that are not measured directly. Such an initial state can accordingly be considered a random variable and control design for such a system can explicitly consider knowledge on this random variable to shape the transient performance. However, before we exacerbate on this problem setting, let us now first provide a general introduction to the topic. This will allow us to formulate this problem in a general setting, which in turn allows for further development and other applications.

General introduction

One of the major focuses for many control engineering applications is the control of variations in the states of a process. Such variations are present in all aspects of all processes, but the magnitude of these variations either forces us to consider them explicitly, or allows us to ignore their effect. A stochastic noise, if present, can be a particular difficult type of perturbation to deal with. In this case, we generally consider a control system to be 'good' if the effect of this perturbation remains within acceptable bounds, which can be determined based on the system characteristics. A general underlying assumption is that the perturbation influences the vector field of the systems' dynamics. Such problems are considered in the field of stochastic control (Aström 1970, Bertsekas 1976) and one of the ways to express knowledge on the state is through a probability density function (pdf) that changes over time (Kárný 1996, Sun 2006). This function then maps values of the state space to proba-

bilities of occurrence. The Fokker-Planck equation or forward Kolmogorov equation (Risken 1989, Gardiner 1985) is a well-known result related to this approach, commonly specified for an Itô process (driven by a Wiener process), such as Brownian motion (Itô et al. 2012).

Efforts to improve control system design, combined with ongoing advances in sensor and actuator technologies, are increasingly successful in minimizing the perturbation effects on the vector field. We can hence turn our attention to other sources of variation. A subject that has received little attention in the literature, is the variations in the initial conditions. This will be the focus in Part II of this thesis. An example of an application where such variations are relevant is a high-precision and high-frequency manufacturing processes where natural and uncontrollable variation is present in the input materials. The presence of such undesirable *ab initio* conditions can decrease the end-products quality when it is not taken into account explicitly in the control design. In other words, although the use of high-precision systems can minimize product variations due to measurement noises and lack of accuracy in the actuator systems, it cannot preempt variability in the initial conditions.

In Part II of this thesis, one of the main assumptions on the system is that the initial conditions are random variables where we have apriori knowledge of its pdf. Notice that such a formalism encompasses also the standard deterministic setting by taking a Dirac pdf. When both the initial time and initial state are random variables (which can represent time-varying initial conditions), our results are still applicable by considering the marginal distribution of the joint distribution on the initial state random variable, which leads to conservative results. For such systems with stochastic initial conditions, designing a control law with a particular asymptotic behavior as the main criteria does not reveal extra information since all possible trajectories converge to the desired operating point or trajectory, independent of the initial state pdf. On the other hand, since the closed-loop systems' transient behaviour is highly dependent on the initial state, we investigate control design methods where we take into account the evolution of the state pdf in the control design problems.

In our first main result, we consider a containment control problem (CCP). Here, in addition to achieving desired asymptotic behaviour, the controller needs to guarantee that the cumulative distribution of the trajectories over a prescribed set at a given transient time reaches a prescribed value. Such a problem formulation can be related to the funnel control problem for deterministic systems (Ilchmann et al. 2002, Ilchmann et al. 2007). In these papers, the control problem is to design control laws that guarantee the state trajectories from *all* initial conditions remain in a desired funnel, which contracts to the desired state. As the funnel must contain all initial conditions at the initial time, the results are conservative and applicable for linear systems with known relative degree, minimum phase with positive definite high-frequency gain matrix. In this case, our CCP case can be interpreted as a modification to the funnel control whose initial funnel need to cover only a set of initial states with the desired cumulative distribution at the initial time. Our setting is how-

ever more general than the funnel one and it admits a general class of linear systems, namely all controllable linear systems.

In our second result, we consider a different transient performance criterion where, in addition to the asymptotic behaviour requirement, the controller must ensure that the evolution of state pdf at a given transient time is close to a desired pdf. We will refer to this problem as the shape control problem (SCP). The SCP requires particularly the guarantee that a distance between the two pdfs is less than a given prescribed level. A recent work related to the shaping control of pdf has also appeared in Buehler et al. (Buehler et al. 2016). In (Buehler et al. 2016), a generic control framework using stochastic MPC is proposed for stochastic nonlinear systems, where the initial condition is a random variable and the disturbance is a stochastic process. In the present work, a stochastic MPC problem is proposed where the distance of evolving pdf to a desired one must be minimized. In contrast to the result presented in (Buehler et al. 2016) which does not yield an analytical solution, we restrict our problem only to the random initial condition case that has allowed us to construct simple control laws with a guaranteed level of performance and to provide rigorous analysis of the method.

1.5   Outline & contributions

The remainder of the thesis is structured as follows. Part I contains the Chapters 2, 3 & 4. This is the part of the thesis containing the contributions that are directly related and implemented for the UHVCVD process. Part II contains the chapters 5 & 6. This is the part of the thesis that contributes to the control literature in a more general sense, with possible (future) application to UHVCVD. Chapter 7 rounds up with conclusions and an outlook on future research.

In Chapter 2 we present our contributions to the modeling for control of FMF dynamics. Throughout the chapter, we explain some of the fundamental dynamics that need to be described and how we incorporate them in our dynamical formulation. We validate a part of these dynamics by comparing them with results from the literature. We furthermore present a state-of-the-art model-based controller design for such a process and provide numerical simulation results for this controller design, as an academic example.

We use Chapter 3 to present our contributions on the AAS-based sensor design and on the validation of this sensor design and part of the FMF model, that is relevant for dynamics in the vapor pressure regime. To this end, we start by providing background information on the AAS measurement principle. This is followed by a description of the design of our experimental setup that features the AAS-based measurements. We then exacerbate on how we apply the FMF model and how we process the AAS signal for our experimental study. Lastly, we present and discuss the experimental results.

Chapter 4 is the final chapter in Part I. Here, we present the two controller designs for partial pressures that require minimal identification using input-output data. We implement these controllers in our experimental setup to evaluate their performance. To achieve this, we make a small adjustment to the experimental setup presented in Chapter 3 and this will be presented first. We then present a short analysis of the evaporation system dynamics, so that we can motivate some of the controller design choices that we make. We follow by presenting the two controller designs with first results, which show their efficacy. Lastly, we compare the controller performances and provide some concluding remarks.

We start Part II of the thesis with Chapter 5. We shift our attention to the problem case for deterministic systems with stochastic initial conditions. This chapter considers the CCP and we present solutions for both linear and nonlinear systems. We start by defining the CCP and provide a simple example to show the nontriviality of the control problem. This is followed by the result for linear systems. After this, we present the result for nonlinear systems, which is based on results in contraction-based control methods. Lastly, we numerically simulate the nonlinear result for a robot manipulator example.

The second and last chapter of Part II, Chapter 6, contains the results related to the SCP. We present solutions for linear systems, having initial and desired pdfs that are either linearly or nonlinearly matching. We again start by defining the SCP. Following this, we present the result for two linearly matching pdfs. Subsequently, we present the results for nonlinearly matching pdfs. We round up with a numerical simulation for a linear system with linearly matching initial and desired pdfs.

We conclude this thesis with Chapter 7. Here, we specify the conclusions for each part of the thesis and we provide recommendations for future research.

1.6   Origins of the chapters

The work in this thesis is related to papers that have been published and presented at conferences and papers that have been submitted to system and control journals. These papers are under review at time of writing. The list of papers, in order of appearance in this thesis, is accordingly:

[1] Dresscher, M., Jawawardhana, B., Barradas-Berglind, J.J. & Scherpen, J.M.A.: 2017, A modeling framework and flux controller for free molecular flow deposition processes, *American Control Conference*, pp. 2164–2170.

[2] Dresscher, M., Jawawardhana, B., Kooi, B.J. & Scherpen, J.M.A.: 2018, Toward Controlled UHVCVD: Modeling of flow dynamics and AAS partial pressure measurement implementation, under review for a journal.

[3] Dresscher, M. & Jawawardhana, B.: 2017, Precribing transient and asymptotic behaviour of LTI systems with stochastic initial conditions, *IFAC-PapersOnLine*

50(1),1822-1827. 20th IFAC World Congress.

[4] Dresscher, M. & Jawawardhana, B.: 2017, Precribing transient and asymptotic behaviour of non-linear systems with stochastic initial conditions, *IEEE 56th Annual Conference on Decision and Control (CDC)*, 1957-1962.

[5] Dresscher, M. & Jayawardhana, B.: 2018, Prescribing Transient and Asymptotic Behavior to Deterministic Systems with Stochastic Initial Conditions, under review for a journal.

Let us now shortly list how these papers are connected to the individual chapters of the thesis. Chapter 2 presents contributions from [1] & [2]. Specifically, we use the modeling framework as presented in [2], while we present the validation of dynamics, the controller design and numerical simulation results from [1]. Chapter 3 is based solely on [2]. This chapters contains the main experimental contributions from this paper. Chapter 4 contains results that have not been published or prepared for submission at the time of writing. Chapter 5 is based on [3], [4] & [5]. The first solution to the CCP has been presented in [3] for the linear case. This result has been generalized further in [5]. The solution to the CCP for nonlinear systems was first presented in [4]. As for the linear case, a more general solution was presented in [5]. Lastly, Chapter 6 contains contributions from [3] & [5]. The SCP was first solved in [3] for first order systems. In order to generalize the results, we have presented a new approach for the analysis and control design in [5]. The latter are the results presented in this thesis.

1.7 Applied units

All units that are applied in this thesis are in accordance with the international system of units (SI).

Part I

Modeling of Free Molecular Flow Dynamics, Partial Pressure Measurement and Controller Implementation