Simulation in thin film technology

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ABSTRACT

Simulation and modeling find more and more their way into thin film technology. Beside theoretical models for layer design, pre-production design analysis, and real time process control, atomistic simulation techniques gain of importance. Here, especially classical procedures such as Direct Simulation and Particle-in-Cell Monte Carlo (DSMC/PIC-MC), kinetic Monte Carlo (kMC) and Molecular Dynamics (MD) as well as quantum mechanical techniques based on Density Functional Theory (DFT) have to be mentioned. These methods are applied in order to investigate the material transport inside the coating facilities, the thin film growth in dependence of characteristic process conditions, and the optical and electronic thin film properties. By combination of these atomistic techniques in a suitable manner, a multiple scale simulation model can be realized for investigating the influence of specific process conditions on the resulting layer properties. The further extension of this “virtual coater” concept with respect to rate equation models enables the possibility to investigate also the interaction of laser irradiation with the modeled thin film structures.

Keywords: Modeling, Thin films, Multiple scale model, Direct Simulation Monte Carlo, Particle-in-Cell Monte Carlo, Molecular Dynamics, Density Functional Theory, Optical properties

1. INTRODUCTION

In general, the modeling of physical processes has been established in various fields of applications. For example, in the framework of machine construction, simulations based on FEM or FDM are necessary to meet the state of the art demands. Additionally, for complex physical processes specific theoretical models and algorithms are developed, which are only applicable to the corresponding problem. The thin film technology is a prominent example, where isolated solutions are developed in order to solve specific mechanisms and processes. Here, the simulation tools and theoretical models are in particular applied to clarify three essential issues. The first one is dedicated to investigate the fundamental limits of the material properties. An important example is the transmittance range of an optical material without any inclusions and defects. Furthermore, the available variation range of the index of refraction for a selected coating material is of main interest. It is well known that the film density is directly linked to the refractive index. The second issue is the analysis of the simulated system dynamics, which is equal to the determination of the response function of the system to a variable set of control parameters. Thereby, the resulting layer properties, which react sensible on the process parameters, can be understood in detail on the basis of theoretical studies. In this case, the input parameters for the simulation can be adjusted with highest numerical accuracy suppressing every experimental fluctuation. In the course of the thin film design synthesis, the influence of the controlled variation of the material and layer thicknesses on the transfer function is used in order to achieve an optimized target design. The third case is the application of these simulation techniques to optimize the coating process and to improve the resulting layer properties.

However, recent progresses in thin film technology resulted in an increase in purity, a reduction of particle contamination, and a precision in layer thickness on the atomic scale. The state of the art technology allows to manufacture optical coatings with losses in the range of few ppm in the near infrared wavelength range and components with high power handling capability. With respect to the demands on optical coatings, a multiplicity of coating techniques has been developed, but even when considering only one selected coating method, a large variety of technical implementations and modifications can be found in the production field. Nevertheless, the fundamental processes of the thin film nucleation are not understood in detail, and the optimization of coating processes is still based on empirical

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experience regarding the correlation between thin film properties and process parameters. In the course of the rapid development of modern photonics the quality requirements on optical coatings have reached a level, which will not be achievable by the present empirical optimization strategies in near future. This indicates the need for more sophisticated optimization concepts based on modelling of deposition processes on a fundamental theoretical level. Such novel simulation approaches offer the possibility to investigate in the influence of the process parameters on the resulting film properties. Consequently, a significant narrowing of the process parameter space may be achieved prior to the implementation of a specific deposition process to facilitate coating production with improved flexibility and economy.

In order to support the experimental endeavors to realize high quality optical coatings, a large number of different simulation techniques are applied in thin film technology. These methods are often related to the design of complex layer systems for specific applications and to optical monitoring concepts, where pre- and post-processing simulations gain of importance. Furthermore, the different processes taking place during the coating manufacturing are also covered by numerous simulation techniques. For instance, the material transport within the vacuum chamber is investigated by plasma and flux simulations, the thin film growth is typically described by classical atomistic simulation techniques, and the optical properties of the coating materials can be modeled using ab-initio quantum mechanical calculations. Not least considering high power laser applications, the interaction of the laser irradiation with dielectric materials can be determined by solving rate equation models, and absorption as well as the laser damage threshold can be predicted by numerical approaches.

The present contribution is focused on atomistic simulation techniques used in thin film technology and their respective application areas. In section 2, the benefits of applying plasma simulation techniques such as Direct Simulation and Particle-in-Cell Monte Carlo (DSMC/PIC-MC) for investigating the material flux during the coating process are described. In the following, the usage of classical Molecular Dynamics (MD) to model thin film growth in dependence of characteristic process conditions is presented in section 3. In section 4, Density Functional Theory (DFT) calculations are introduced in order to evaluate optical and electronic properties of dielectric material. In addition, a concept is described that combines plasma and thin film growth models to a "virtual PVD coater" model (section 5). This approach is discussed with regard to the underlying target to correlate specific production parameters with the resulting structural, optical and electronic film properties of the coatings. Finally, a possibility is depicted to link the virtual coater concept to rate equation models (section 6) for investigating laser material interactions.

2. TRANSPORT SIMULATIONS APPLYING DSMC/PICMC

While computational material modelling is already successfully applied for identifying novel functional materials, it cannot be neglected that there is a strong interaction of intrinsic material properties with conditions imposed by mechanisms of the deposition process. For TiO_2 prepared by reactive magnetron sputtering and evaporation, it is shown that the phase compositions of the layers after deposition and after post-deposition annealing strongly depend on process conditions such as total pressure, reactive gas fraction and magnetic field [1]. Further, the electric power mode in magnetron sputtering influences significantly the ion energy distribution at the substrate [2], which in turn can have a tremendous effect on the morphology and phase composition of the deposited layer [3].

For that reason, a comprehensive model of thin film deposition needs to include the dynamics of thin film growth as well as the transport mechanisms on reactor scale. For the description of the gas and material transport within the deposition chamber it is important to consider the characteristics of the transport flow. Processes operating at pressures of several mbar or higher can be usually well described by using a continuum approximation for the gas flow e.g. based on Navier-Stokes equations. For this purpose, a large variety of commercial and open source finite element continuum solvers are available. However, for deposition methods such as magnetron sputtering or ion beam sputtering applied for precision optical coatings, typical gas pressures in the coating chamber are in the 10^{-2} – 10^{-4} mbar range corresponding to a mean free path of gas molecules within 1-100 cm. If the mean free path comes close to characteristic geometric dimensions of the reactor, the continuum approach holds no longer, and characteristic deviations occur between computational fluid dynamic and kinetic solutions based on the more general Boltzmann equation [4].

The Boltzmann equation describes the time development of a six dimensional phase space, which would be very computationally expensive via a finite element approach. Instead, probabilistic approaches based on tracking the trajectories and collisions of representative test particles are a feasible way of solving the Boltzmann equation via statistical means. For description of the movement of neutral particles, the "Direct Simulation Monte Carlo" (DSMC) –

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method [5] has been established, while the more general case of neutral and charged particles including self-consistent computation of the electro-magnetic field is referred to as “Particle-in-Cell Monte Carlo” (PIC-MC) – method [6].

In the DSMC method particle-particle collisions are treated statistically within simulation cells. For ensuring numerically and physically accurate results, the cell spacing shall be less than the mean free path of particles, and the iteration time step shall be chosen in a way that most particles do not pass more than one cell boundary per iteration. Therefore, in three-dimensional DSMC simulation, the number of simulation cells scales as a function of pressure p with p^3, and the number of time steps scales linearly in p, respectively. While the Boltzmann equation is principally valid for any pressure, the computational load of the DSMC drastically increases with pressure and therefore, for typical reactor dimensions it is practically limited to total pressures well below 1 mbar. For gas dynamics in magnetron sputtering or ion beam sputtering and by taking advantage of massive parallelization [7] it is even feasible to use DSMC within three-dimensional industrial scale reactor geometries. Typical applications of DSMC in magnetron sputtering are the computation of gas dynamics in in-line sputter coaters with consideration of substrate movement [8]. Also, the flux of sputtered particles towards the substrate can be computed and shielding orifices can be designed in order to optimize the film thickness homogeneity [9].

For PIC-MC the movement of charged particles within self-consistently computed electro-magnetic fields is even more numerically challenging and requires more numerical constraints to be considered. In typical plasma simulation cases, the most severe constraint is that, in order to prevent *numerical heating*, the cell spacing has to be less than $k \times L_D$, where L_D is the electron Debye length and k is a factor in the order of three [10]. For realistic plasma densities e.g. in the range of $10^{17}/m^3$ in magnetron sputtering, the cell spacing would need to be in the order of 100 µm or less, which is hardly feasible in realistic three-dimensional geometries. As a compromise, plasma simulation at substantial lower power density compared to the experimental conditions can still yield significant insight into the governing mechanisms. As an example it is possible to realistically describe the so called *cross corner effect* in magnetron sputtering, which is an enhanced erosion of linear sputter targets close to the corner region, by low-density PIC-MC simulation as shown in [11]. While PIC-MC simulations of magnetron discharges at higher plasma density become feasible in two dimensions [12] it is shown that three-dimensional magnetron discharges are characterized by propagating ionization waves which cannot be described within two dimensional models by definition [13]. Such ionization waves can also be experimentally observed either at very low discharge currents [14] or also under high plasma density conditions [15], and they can have an impact on the energy distribution of positive ions [16].

In order to model high density, three-dimensional discharges, there is still much development work needed for PIC-MC. An important step could be the introduction of self-adaptive gridding schemes, where the high cell resolution is only applied within hot spots of the discharge, where the Debye length constraint is most critical. With respect to a holistic model of film growth, the role of DSMC and PIC-MC simulation is to provide detailed data about the growth conditions at the substrate. These include the particle flux density as well as the energy and angular distribution functions. For ion beam sputtering, a coupled simulation model consisting of a DSMC transport simulation and subsequent Molecular Dynamics simulation of TiO_2 is demonstrated in [22].

3. THIN FILM GROWTH SIMULATIONS APPLYING CLASSICAL MD

Thin film growth takes place on nanometer and sub-nanometer scale and therefore, an appropriate simulation technique for modeling layer formation has to represent the important growth mechanisms on atomic level. Essentially in literature, two classical atomistic simulation techniques are described to investigate thin film growth related problems, namely kinetic Monte Carlo (kMC) and classical Molecular Dynamics. Due to the underlying calculation procedure, kMC is able to model long term effects such as thermal surface diffusion, which has constants in the range between 10^{-8} and 10^{-3} cm^2s^{-1} [17]. In contrast, the time resolution in MD is in the range of 1 fs to resolve atomic vibration. Consequently, surface diffusion cannot be investigated in the framework of thin film growth modeling. Nevertheless, it has been shown [18] that thermal activated surface diffusion can be neglected when modeling the deposition of high energetic atoms applying similar conditions to that of sputter processes. Generally, the simulation techniques have to meet the requirements of the physical problem to be investigated. Therefore, it is not necessary to decide, which is the most appropriate simulation procedure in general but which is the best suitable procedure to model the physical process of interest. Since thin film growth in modern ion sputter processes is characterized by high energetic coating atoms and amorphous film growth, this contribution is focused on classical MD.

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Molecular Dynamics is based on solving the Newton´s equation of motion for a given atomic system. Therefore, the most essential input is a classical potential describing well the interactions between all involved particles. In the simplest case, the potential depends only on the distances between the species. For the time integration different boundary conditions can be applied, referred to as ensemble, since they satisfy the condition of a specific thermodynamic state. The most important ensembles for modeling thin film growth are the microcanonical ensemble (NVE), where number of particles N, volume V, and system energy E are held fix and the canonical ensemble (NVT), where N, V, and system temperature T are kept constant. In order to investigate layer formation a ground structure has to be prepared in advance, which is used as substrate for the growth simulation. The deposition of coating material is modeled for each atom in a single MD simulation (see for example [19,20]). In most cases the simulation is divided in two subsequent parts, applying first the NVE ensemble and second the NVT ensemble. This procedure ensures in the first step the physical correct modeling of energy input into the already existing film and in the second step the adaption of the system to a given substrate temperature.

As input parameters for the MD thin film growth simulation characteristic process parameters can be applied, e.g. deposition energy and angle of incidence of the coating atoms at the substrate, deposition ratio of involved species, and the substrate temperature. According to these values, thin film growth simulations can be performed and the influence on the resulting layer properties can be investigated. The output of the MD growth simulation methods are in particular the structural layer properties, such as layer density, surface roughness, and stoichiometry. But also the near field characteristic is available in terms of radial distribution function and nearest neighbor statistics. By application of suitable approximations, the total number of atoms in the growth structure can be increased drastically. These approximations depend essentially on the increase of the deposition ratio [21] or the reduction of considered atoms in the deposition event modeling. Applying these kinds of approximations, structure sizes including 10^5-10^6 atoms can be achieved [19,22] and hence, thin film thicknesses of few tens of nm can be modeled depending on the used ground structure area.

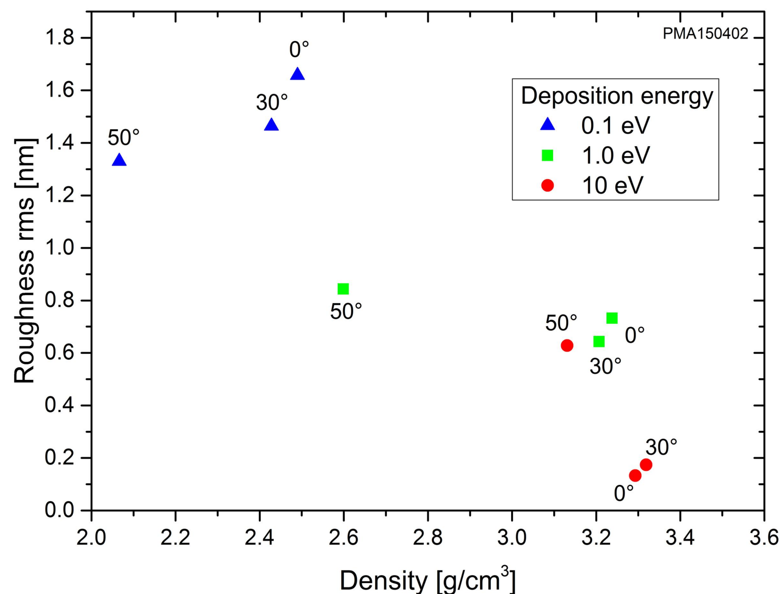

Figure 1. Roughness versus density of Al_2O_3 thin film growth for different discrete parameter sets of angle of incidence (labels) and deposition energy.

In figure 1, the resulting layer density and surface roughness is shown for modeled Al_2O_3 thin films applying different sets of discrete deposition energies and angles of incidence (AOI). For the simulations, 10^5 coating atoms were deposited on a substrate area of 56 nm^2. The resulting layer thickness is approximately 18 nm. Obviously, a strong dependence of the structural properties on the deposition energy and the AOI can be stated. Generally, an increase in the deposition

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energy results in an increase of film density and a decrease of surface roughness. For the angular distribution, the overall trend is a reduction of density and an increase in roughness for larger AOI. This effect can be explained by the formation of voids and columns in the layer structure due to shadowing effects. By the choice of the input parameters the influence of different coating processes and process geometries on layer growth can be investigated.

Another important point in thin film technology is the interface problematic in the transition region between two layers of different materials or between different phases, e.g. crystalline and amorphous, of the same material. Here, studies on fundamental basis are needed, which bring new insights or can support empirically found dependencies. This issue can be investigated by applying thin film growth models based on classical MD. The necessary requirement is a classical potential, which can describe well the interaction between the different coating materials or the configuration of different material phases, respectively. Exemplary, figure 2 (left) shows the growth of TiO_2 on a rutile substrate structure. The transition between the crystalline ground structure (bottom) and the amorphous layer (top) is clearly visible, accompanied by a transition area of approximately 1-2 nm thickness.

In addition, a specific problem in thin film technology is often summarized under the topic “defects”. Here, especially the influence of contaminants on the layer growth and the resulting properties is of interest. Again, suitable models based on MD can be applied to investigate this topic in detail (see for example [23]). A basic approach for investigating the influence of defects on layer growth can be realized applying structured substrates as shown in figure 2 (right). In this case, the structured system has been prepared by addition of a 3x3x2 nm^3 cube on the top of the flat substrate, representing the defect. In the following, TiO_2 layer growth has been modeled using perpendicular angle of incidence and thermal deposition energy (0.1 eV). Figure 2 (right) shows that the defect acts as seed for the observed columnar structure. The voids that are visible beside the central column are caused by shadowing effects. The same simulation has been repeated using a deposition energy of 10 eV. In this case, the defect has been completely enclosed by the coating material and no voids could be observed. It has to be mentioned, that typical defect diameters are in range of some μm and can be therefore not modeled applying the state of the art computational power. Nevertheless, the investigations for smaller defect sizes can give important hints and will help to understand the underlying mechanisms.

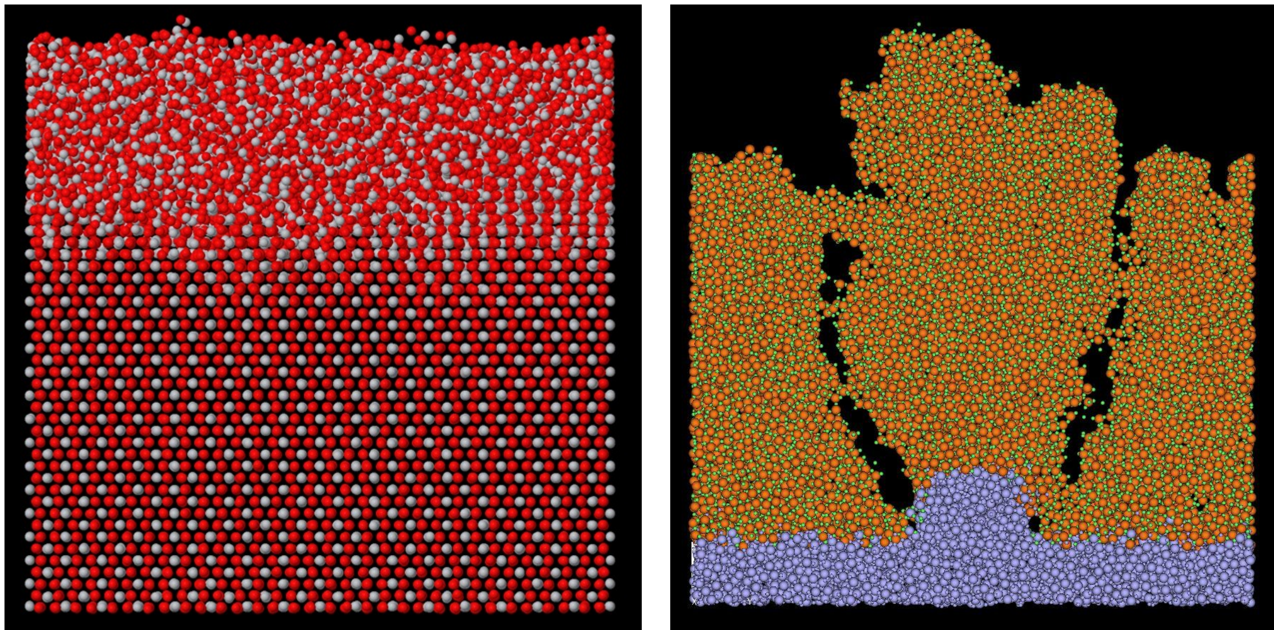

Figure 2. TiO_2 thin film growth on a rutile substrate using 10 eV deposition energy and 0° angle of incidence (left), base area ~ 7.3x7.3 nm^2; TiO_2 thin film growth on a structured substrate applying 0.1 eV deposition energy and 0° angle of incidence (right), base area ~14.0x18.2 nm^2.

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4. CALCULATION OF OPTICAL PROPERTIES APPLYING DFT

The most accurate atomistic simulation procedures are based on quantum mechanics. In chemistry and physics, a common method to investigate the electronic structure of atomic systems is the Density Functional Theory (DFT). By applying appropriate approximations it became possible to get a solution of the Schrödinger equations of a many body system. The higher accuracy of DFT in comparison to the classical atomistic techniques demands a clear increase in computation power. Therefore, typical structure sizes applied to DFT calculations are in the range of few hundreds of atoms. This limits the area of application, since for the already discussed problems concerning film growth behavior larger structure sizes and hence, classical techniques have to be used. Nevertheless, for the precise calculation of optical as well as electronic properties for specified materials and compounds, quantum mechanical simulation procedures are indispensable.

For the determination of the optical properties, at first the frequency dependent dielectric function must be calculated on the basis of DFT. By application of the Kramers-Kronig relation also the real part is accessible. Having both, the real and the imaginary part of the dielectric function, all other optical parameters, e.g. reflectivity, index of refraction, extinction coefficient, and absorption coefficient can be derived. The basic requirement for the calculation of the optical properties is a prepared supercell, containing the atomic system of interest and being in the most cases periodic in all directions. Therefore, the density and the stoichiometry of the system have to be defined in advance. This in turn implies detailed information about the used model material.

In [24], the index of refraction was calculated for several TiO_2 supercells, having densities in the range between 3.21 g/cm^3 and 4.14 g/cm^3. It could be shown that the refractive index rises with increasing film density and has values between 2.12 and 2.32 at a wavelength of 1000 nm. This result was in agreement with the experimental data of several TiO_2 single layers, which were prepared applying different coating techniques.

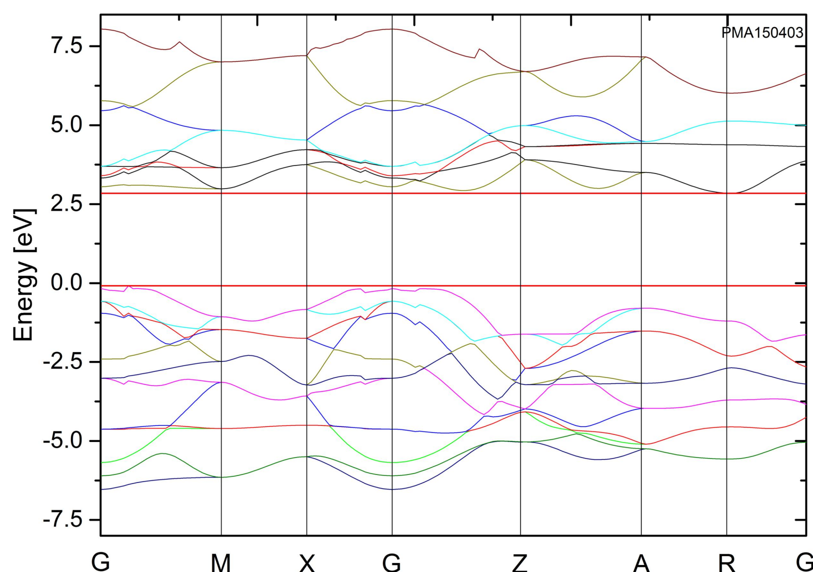

Figure 3. Calculated band structure of rutile applying the HSE06 functional. Indirect band gap value is 2.93 eV.

If the bandgap of the investigated material is directly calculated from the eigenvalues of the Kohn-Sham equations, the choice of the functional to treat the exchange and correlation energy is quite important. For example, the band gap for rutile TiO_2 is underestimated applying a pure DFT functional by approximately 1.5 eV [25]. This situation can be improved using a hybrid functional (HF), which is a linear combination of a density functional and the exact Hartree-Fock exchange functional. A very common HF is the HSE06 functional according to Heyd, Scuseria and Ernzerhof (see for example [26]). In figure 3, the calculated band structure for rutile is shown applying the HSE06 functional. The resulting bandgap of 2.93 eV is very close to the experimental band gap for rutile of 3.03 eV [27]. Even better agreement to experimental data is achieved by using many body perturbation theory based on the theory of the Green´s functions

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and in combination with the GW approximation [28]. The drawback of the GW method is its high computational cost, which is usually an order of magnitude higher than that of a typical DFT calculation [29].

With respect to the experiment, it is also possible to obtain the bandgap of amorphous materials from the energy dispersive absorption spectrum, which is also available from the DFT calculations. By application of e.g. Tauc- or Urbach-plot, the size of the bandgap can be determined. In [24], the calculated bandgaps of several TiO_2 structures having different densities and the experimental bandgaps of various single layers are evaluated applying the Tauc-plot. The comparison shows a very good agreement of both data sets.

5. CONCEPT OF VIRTUAL COATER

The ultimate goal is to develop a theoretical model, which is able to simulate thin film growth in dependence of the characteristic process parameters and the underlying coating technique, and which can be used to improve the optical properties of the prepared layer systems. As already pointed out, each mentioned simulation technique is appropriate to model specific issues in thin film technologies, taking place on well-defined length scales. Hence, there is no single simulation technique being able to model the full coating process from coating material generation to the optical film properties. Consequently, several simulation techniques have to be combined to a multiple scale model, whereby each method within the simulation chain is dedicated to a specified problem. A multiple scale thin film growth model has been realized in the concept of the "virtual coater". Here, the coating material generation and the material transport to the substrates are simulated applying DSMC/PICMC techniques. In particular, the coating chamber geometry and the characteristic parameters of the ion sources or magnetrons are used as input parameters. The output of the calculations is the energy and the angular distributions of the coating material at the substrates. These distributions are in turn used as input parameters for the thin film growth calculations applying classical simulation techniques (MD, kMC). Afterwards, the structural parameters are extracted and used to prepare a small representative supercell. For this cell, the electronic and optical properties are subsequently calculated on the basis of DFT. Applying the concept of the virtual coater, a simulation of the full coating process becomes possible. Further details of the applied software interfaces between the different techniques and the transferred data sets can be found in [22]. The schematical representation of the virtual coater concept is displayed in figure 4.

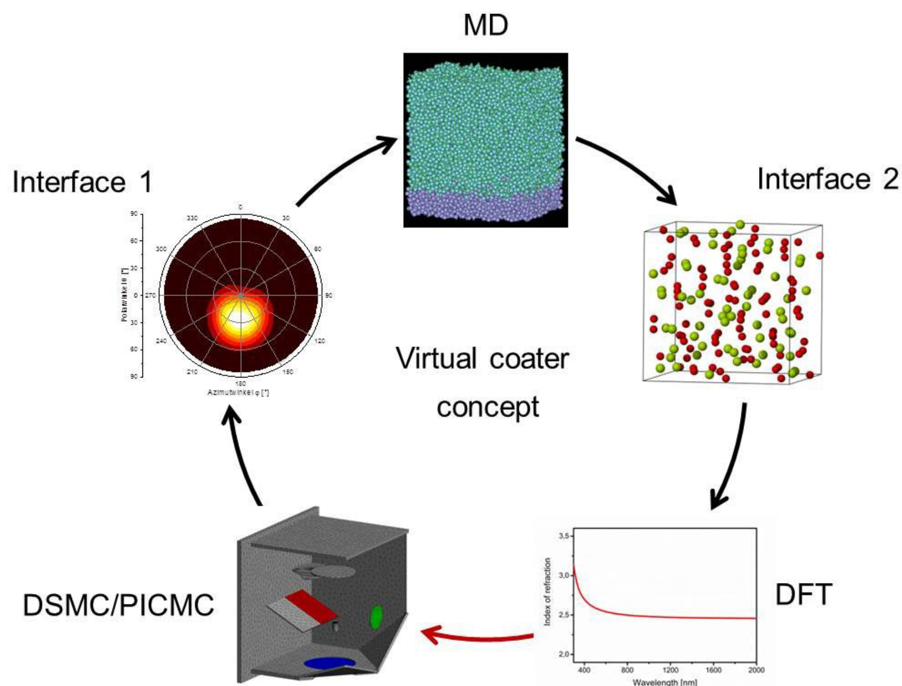

Figure 4. Schematical representation of the virtual coater concept.

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The discussed concept can be applied to various coating plants and techniques, because the geometry and the characteristic process parameters are integrated into the simulation. In [22], the multiple scale model has been applied to an existing IBS coating facility to investigate the growth of TiO_2 single layers. The final comparison of optical data shows a good agreement between experimental and calculated index of refraction.

In addition, the concept can be used for the improvement of optical thin films. This is adumbrated in figure 4 with the red arrow, which closes the circle. By changing the starting parameter (geometry/process parameter), the influence on the final results can be investigated and therefore, an improvement procedure can be achieved.

6. FROM ATOMISTIC TO RATE EQUATION MODELS

Finally, an example for a further extension of the virtual coater concept can be realized with regard to the power handling capabilities. For the theoretical modeling, rate equation models are applied to investigate the interaction of high power laser irradiation with dielectric materials. In the framework of thin film technology adapted rate equations are used to calculate the free electron density during irradiation with short laser pulses to model for example the laser induced damage threshold [30] and the linear as well as the nonlinear material absorption [31]. Since the optical and electronic properties in terms of the band gap and the refractive index represent an essential part of the needed input parameters for the rate equation calculations, it is quite desirable to use the concept of the virtual coater in order to calculate these values. In this way it is possible to connect the theoretical modeling of the thin film preparation and the behavior of the thin films in the respective application area.

In figure 5 (right) the development of the free electron density is shown applying the Keldysh theory [32] for a rutile layer with a band gap of 2.93 eV and an effective mass between 0.62 to 0.64 $m_{electron}$ [33] (theoretically determined value) during laser irradiation with 180 fs pulse duration at 800 nm laser wavelength. It can be observed that a pulse energy density of ~0.237 J/cm^2 is enough to reach the critical electron density level of ~10^{21} 1/cm^3 for optical breakdown. It has to be mentioned that the rate equation model delivers a very good agreement to the experimental findings, but the exact measurement of the band gap as well as the effective mass of the electrons in the conduction band achieving the necessary accuracy is not possible. By the application of the virtual coater concept the properties of the deposited layer are reproduced on a theoretical level, and consequently, the necessary values can be extracted directly form the simulated structures.

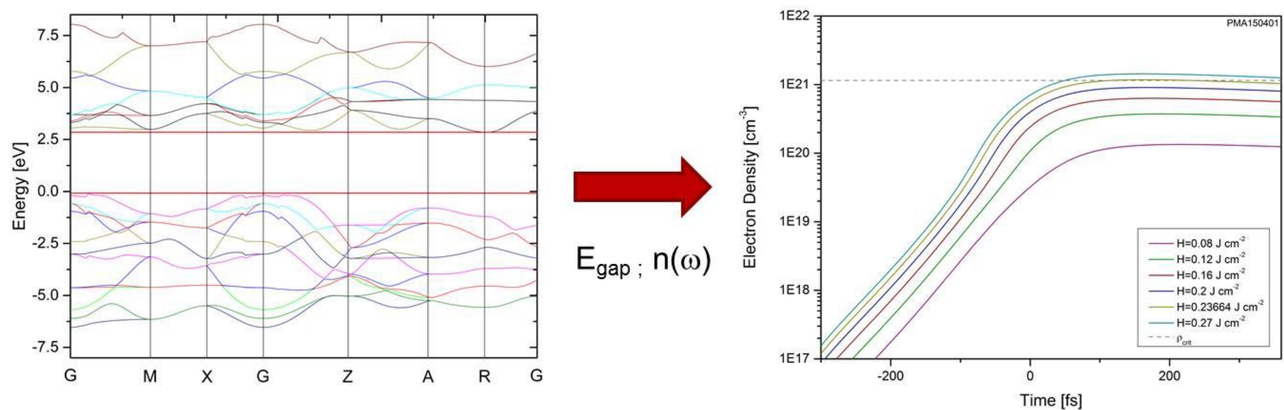

Figure 5. Schematical representation of the interface between DFT calculation of optical properties and rate equation models. The right plot shows the development of the free electron density of rutile (bandgap 2.93 eV) for an ultra-short pulse with 180 fs pulse length and 800 nm wavelength.

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7. SUMMARY

The present contribution gives an overview concerning the application of atomistic simulation techniques in thin film and coating process related problems. The different techniques are described according to the fundamental calculation procedure and with respect to specific tasks, e.g. material transport simulation, thin film growth modeling, and calculation of optical properties. Furthermore, an approach is presented, which combines the discussed techniques to a virtual coater concept that is able to model the full coating process. Finally, an extension of the virtual coater concept is presented, which links the determined optical properties with rate equation models to investigate the laser material interaction.

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